

National Acid Precipitation Assessment Program Report to Congress:

An Integrated Assessment



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About the National Acid Precipitation Assessment Program

NAPAP is a cooperative federal program first authorized in 1980 to coordinate acid rain research and report the findings to Congress. The research, monitoring, and assessment efforts by NAPAP and others in the 1980s culminated in Title IV of the 1990 Clean Air Act Amendments (CAAA), also known as the Acid Deposition Control Program. In a bold new approach to environmental protection, Title IV includes a market-based program that provides economic incentives for controlling emissions of sulfur dioxide from electricity generating facilities. Title IX of the CAAA reauthorized NAPAP to conduct acid rain research and monitoring and to periodically assess the costs, benefits, and effectiveness of Title IV. The NAPAP member agencies are the U.S. Environmental Protection Agency, the U.S. Department of Energy, the U.S. Department of Agriculture, the U.S. Department of Interior, the National Aeronautics and Space Administration, and the National Oceanic and Atmospheric Administration. This report is the fourth published by NAPAP since 1990 assessing Title IV.

In 1997 NAPAP began to operate under the auspices of the Committee on Environment and Natural Resources (CENR) of the National Science and Technology Council. NAPAP's goal continues to be providing credible technical findings on acid deposition and its effects to inform the public decision-making process. To ensure that this goal is met, NAPAP coordinates its activities through the Air Quality Research Subcommittee of the CENR.

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Acronyms

AEL	Alternative Emission Limit
AHM	Alpine Hydrochemical Model
Al ²⁺ , Al ³⁺	aluminum cations
AMS	Allowance Management System
ANC	acid neutralizing capacity
ARP	Acid Rain Program
BenMAP	Benefits Mapping and Analysis Program
BTU	British thermal unit
CAA	Clean Air Act
CAAA	Clean Air Act Amendments
CAIR	Clean Air Interstate Rule
CAMD	Clean Air Markets Division
CASTNET	Clean Air Status and Trends Network
CEMS	continuous emission monitoring system
CENR	Committee on Environment and Natural Resources Research
CFR	Code of Federal Regulations
CMAQ	Community Multi-Scale Air Quality
CMAS	Community Modeling and Analysis
CO ₂	carbon dioxide
CPI	cost performance index
DOC	dissolved organic carbon
DOI	U.S. Department of the Interior
dv	deciview
ECMPS	Emissions Collection and Monitoring Plan System
EGU	electric generating units
EMAP	Environmental Monitoring and Assessment Program
EPA	U.S. Environmental Protection Agency
GAO	General Accounting Office
HNO ₃	nitric acid
ILWAS	Integrated Lake-Watershed Acidification Study
IMPROVE	Interagency Monitoring of Protected Visual Environment
IPM	Integrated Planning Model
kg/ha/yr	kilograms/hectare/year
lb	pound
LTM	Long-Term Monitoring
MAGIC	Model of Acidification of Groundwater in Catchments

meq/L/yr	milliequivalents per liter per year
meq/m ² /yr	milliequivalent per square meter per year
meq/m ² /yr	milliequivalents per square meter per year
MMBTU	million British thermal units
MPI	market price index
MW	megawatt
N ₂ O	nitrous oxide
NAAQS	National Ambient Air Quality Standards
NADP	National Atmospheric Deposition Program
NAMS	National Air Monitoring Stations
NAPAP	National Acid Precipitation Assessment Program
NBP	NO _x Budget Trading Program
NDDN	National Dry Deposition Network
NH ₃	ammonia
NH ₃ -N	ammonia nitrogen
NH ₄ ⁺	ammonium ion
NO	nitric oxide
NO ₂ ⁻	nitrite
NO ₃ ⁻	nitrate
NO ₃ ⁻ plus HNO ₃	ambient total nitrate
NO ₃ -N	nitrate nitrogen
NOAA	National Oceanic and Atmospheric Administration
NO _x	nitrogen oxides
NO _x SIP Call	NO _x State Implementation Plan Call
NPS	National Park Service
NSTC	National Science and Technology Council
NSWS	National Surface Water Survey
NTN	National Trends Network
O ₃	ozone
OMB	Office of Management and Budget
OTC	Ozone Transport Commission
PM _{2.5}	fine particulate matter
RIA	Regulatory Impact Assessment
RSM	Response Surface Model
RTC	Report to Congress
SIP	state implementation plan
SLAMS	State and Local Air Monitoring Stations
SMBE	simple mass balance equation

SO ₂	sulfur dioxide
SSWC	steady-state water chemistry
TIME	Temporally Integrated Monitoring of Ecosystems
USGS	U.S. Geological Survey
XML	Extensible Markup Language
µeq/L	microequivalents per liter
µg/m ³	micrograms per cubic meter

1 **Executive Summary**

2 ***What is acid rain?***

3 Acid deposition, more commonly known as acid rain, occurs when emissions of sulfur dioxide (SO₂) and
4 nitrogen oxides (NO_x) react in the atmosphere (with water, oxygen, and oxidants) to form various acidic
5 compounds. Prevailing winds transport the acidic compounds hundreds of miles, often across state and
6 national borders. These acidic compounds then fall to earth in either a wet form (rain, snow, and fog) or a
7 dry form (gases, aerosols, and particles). At certain levels the acidic compounds, including small particles
8 such as sulfates and nitrates, can cause many negative human health and environmental effects.

9 ***What are the effects of acid rain?***

10 Ecosystems and human health are subject to many stresses, including acid rain. Scientific research has
11 shown that SO₂ and NO_x air pollutants and the acid rain formed by these pollutants can

- 12 ▪ Degrade air quality
- 13 ▪ Impair visibility
- 14 ▪ Negatively impact public health
- 15 ▪ Acidify lakes and streams
- 16 ▪ Harm sensitive forests
- 17 ▪ Harm sensitive coastal ecosystems
- 18 ▪ Accelerate the decay of building materials, paints, and cultural artifacts, such as buildings,
19 statues, and sculptures.

20 ***Why is this report being sent to Congress, and what is the role of the National 21 Acid Precipitation Assessment Program (NAPAP) in the report?***

22 In 1990, Congress enacted Title IV as part of the Clean Air Act Amendments (CAAA). The Acid Rain
23 Program (ARP) created under Title IV requires significant decreases in the emissions of SO₂ and NO_x
24 from fossil fuel-burning power plants to improve air quality and protect ecosystems that have suffered
25 damage from acid deposition. Under Title IX of the 1990 CAAA, the National Acid Precipitation
26 Assessment Program (NAPAP) was asked to periodically assess and report to Congress on
27 (1) implementation of the ARP, (2) the most recent scientific information related to acid deposition and
28 its effects, and (3) additional decreases in acid deposition necessary to prevent adverse ecological effects.
29 This NAPAP Report to Congress (RTC) focuses on emission reductions from power plants, summarizes
30 changes in deposition rates and environmental impacts, and projects the ecological effects of additional
31 reductions in SO₂ and NO_x emissions.

32 ***What are the results of implementing Title IV of the 1990 Clean Air Act 33 Amendments?***

34 Implementation of Title IV has successfully and substantially reduced emissions of SO₂ and NO_x from
35 power generation (i.e., the sources covered by the ARP), including the following:

- 36 ▪ In 2008, SO₂ emissions were 7.6 million tons, 52% lower than 1990 emissions and 56% lower
37 than 1980 emissions, a level below the 2010 Title IV statutory cap on SO₂ emissions.¹

¹ 2008 was the latest year for which emissions data were available when this report was written and reviewed. More recent emissions data are available on the EPA Web site at <http://www.epa.gov/airmarkets/emissions>.

- 1 ▪ In 2008, NO_x emissions were 3 million tons, 55% lower than 1990 emissions, exceeding the Title
2 IV goal of a 2 million ton reduction in NO_x emissions from projected 2000 levels without the
3 ARP, as required by the 1990 Clean Air Act (CAA).
- 4 ▪ In addition, SO₂ emissions from all sources, including those sources not covered by the ARP
5 (e.g., automobiles, volcanoes), have decreased by 50% since 1990, and emissions of NO_x from all
6 sources have decreased by 36% since 1990.

7 The emission reductions achieved under the ARP have contributed to measurable improvements in air
8 quality; decreases in acid deposition; the beginnings of recovery of acid-sensitive lakes and streams in
9 some areas; and improvements in visibility, as exhibited by the following:

- 10 ▪ SO₂ concentrations in the atmosphere, a precursor to fine particles and acid deposition, have
11 decreased since 1990. Average annual SO₂ concentrations in 2006–2008 were 45% to 55% lower
12 than in 1989–1991 in the Midwest and eastern United States.
- 13 ▪ Sulfate concentrations in the atmosphere, a major component of fine particles, especially in the
14 eastern United States, have decreased since 1990. Average annual sulfate concentrations in 2006–
15 2008 were 28% to 44% lower than in 1989–1991 in the Midwest and eastern United States.
- 16 ▪ Wet sulfate deposition, a major component of acid rain, has decreased since 1990. Average
17 annual sulfate deposition in the Northeast in 2006–2008 was 33% lower than in 1989–1991,
18 deposition in the Mid-Atlantic was 32% lower, the Midwest was 37% lower, and deposition in
19 the Southeast was 36% lower.
- 20 ▪ Wet inorganic nitrogen deposition has decreased regionally from historical levels. However,
21 decreases were less than those of wet sulfate deposition because of the relatively moderate NO_x
22 reduction from power plants and the continuing large contribution (over 82% of total 2008 NO_x
23 emissions) from other sources of NO_x, such as on-road vehicles and non-road vehicles. Still,
24 average annual wet nitrate deposition in 2006–2008 was 10% to 21% lower than in 1989–1991 in
25 the Midwest and eastern United States.
- 26 ▪ Levels of acid neutralizing capacity (ANC), an indicator of the ability of a waterbody to
27 neutralize acid deposition, have shown improvement from 1990 to 2008 at many lake and stream
28 long-term monitoring sites in the eastern United States, including New England and the
29 Adirondack Mountains. Many lakes and streams still have acidic conditions harmful to their biota
30 even though the increases in ANC indicate that some recovery from acidification is occurring in
31 sensitive aquatic ecosystems.
- 32 ▪ Data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network
33 show no statistically significant trends in visibility at most monitoring sites over the 10-year
34 period from 1999 to 2008. The Northeast, however, has sites with improving visibility on both the
35 best and worst visibility days, principally due to regionally decreased sulfate particulate
36 concentrations.

37 Further, because emission reductions result in fewer fine particles and lower ozone concentrations in the
38 air, thousands fewer premature deaths, hospital admissions, and emergency room visits are projected in
39 2010. The value of the resulting public health benefits range from \$175 to \$430 billion per year (2008\$).

40 ***What is the role of cap and trade in the success of the Acid Rain Program?***

41 The success of the SO₂ emission reduction program is due to the combined use of an overall emission cap
42 for SO₂, which ensures that these reductions are achieved and maintained, and a trading system that
43 facilitates lowest-cost emission reductions. Together, this is known as cap and trade. NO_x reductions
44 under the ARP are achieved through a program that applies to a subset of coal-fired electric generating
45 units (EGUs) and is closer to a more traditional, rate-based regulatory system. Sources controlled in both

1 the SO₂ and NO_x components of the ARP have demonstrated very high levels of compliance, averaging
2 99% annual compliance since the beginning of the program. The inherent flexibility for sources to choose
3 how to control their SO₂ emissions in the cap-and-trade approach for SO₂ has been successful at reducing
4 compliance costs to a fraction of the cost estimated in 1990. Several factors are responsible for the
5 relatively low costs of SO₂ reductions realized under Title IV, including the widespread availability of
6 low sulfur coal and technical innovations that facilitated use of that coal, lower than anticipated scrubber
7 costs, the opportunity to bank allowances, and development of an efficient, high-volume market for
8 allowances. Although the costs are low, the ARP achieves substantial health and environmental benefits
9 through air quality improvements.

10 ***What is the future of current clean air rules?***

11 Emissions of SO₂ and NO_x are expected to decline further as additional programs are implemented to
12 control emissions from fossil fuel-burning power plants. In March 2005, the U.S. Environmental
13 Protection Agency (EPA) promulgated the Clean Air Interstate Rule (CAIR) to achieve further emission
14 reductions beyond levels reached under the ARP and other programs, such as the NO_x State
15 Implementation Plan (SIP) Call. CAIR was designed to help states in the eastern United States attain the
16 National Ambient Air Quality Standards (NAAQS) for fine particulate matter (PM_{2.5}) by reducing and
17 capping SO₂ and NO_x emissions in 28 eastern states and the District of Columbia. On July 11, 2008, the
18 U.S. Court of Appeals for the D.C. Circuit issued a ruling vacating CAIR in its entirety. The Court
19 subsequently remanded CAIR to EPA, leaving CAIR in place until EPA issued new rules to replace
20 CAIR. On July 6, 2010, EPA proposed to replace CAIR with a transport rule that will further control SO₂
21 and NO_x emissions from fossil fuel-burning power plants in 31 states and the District of Columbia.

22 ***Are ecosystems recovering from the effects of acid rain?***

23 Despite the environmental improvements reported here, research over the past few years indicates that
24 recovery from the effects of acidification is not likely for many sensitive areas without additional
25 decreases in acid deposition. Many published articles, as well as the modeling presented in this report,
26 show that the SO₂ and NO_x emission reductions achieved under Title IV from power plants are now
27 recognized as insufficient to achieve recovery or to prevent further acidification in some regions.
28 Additional SO₂ and NO_x emission reductions from power plants and other source sectors are needed to
29 improve air quality, reduce deposition, and further reduce the number of acidic lakes and streams in many
30 regions of the United States. Some of these additional emission reductions may be achieved through
31 implementation of existing or future regulations to address transport of ozone and fine particles, including
32 CAIR and its replacement rules in the eastern United States, rules affecting mobile sources, SIPs to
33 achieve the ozone and NAAQS for ozone and PM_{2.5}, and future rules to reduce air toxics and other
34 pollutants from power plants.

35 ***What is the importance of long-term environmental monitoring in understanding*** 36 ***the effects of acid rain?***

37 Emissions, air quality, deposition, and ecological monitoring are critical components of implementing
38 environmental programs, such as the Title IV ARP. These monitoring efforts allow researchers and
39 policymakers to assess the effectiveness of Title IV and other air quality programs. Emissions monitoring
40 is conducted by affected sources; additional types of monitoring are conducted by a wide variety of
41 federal and state agencies, universities, and other organizations. The agencies of NAPAP continue to have
42 a strong commitment to the research and monitoring that makes assessments like this NAPAP RTC
43 possible.

1 **What acid rain–related topics are currently at the forefront of scientific**
2 **knowledge?**

3 This report also describes several issues pertinent to ecosystem response to emissions controls and acid
4 deposition that are receiving increasing attention in the scientific literature, including the following:

- 5 ▪ An observed delay in ecosystem recovery in the eastern United States, even with decreases in
6 emissions and deposition over the past 30 years
- 7 ▪ Emerging ecosystem impacts of nitrogen deposition in the west
- 8 ▪ The application of critical deposition loads as a tool for scientists to better inform air quality
9 policies
- 10 ▪ The role of changes in climate and the carbon cycle as factors that affect the response of
11 ecosystems to acid deposition
- 12 ▪ The interaction of multiple pollutants in ecosystems.

1 Introduction

National Acid Precipitation Assessment Program

The National Acid Precipitation Assessment Program (NAPAP), a cooperative federal program, was first authorized in 1980 and re-authorized under Title IX of the 1990 Clean Air Act Amendments (CAAA) to coordinate acid rain research and monitoring and to periodically report to Congress. NAPAP is comprised of the U.S. Environmental Protection Agency (EPA), the U.S. Department of Energy, the U.S. Department of the Interior (DOI)/U.S. Geological Survey (USGS), the DOI/National Park Service (NPS), and the National Oceanic and Atmospheric Administration (NOAA). The Acid Rain Program (ARP) is authorized under Title IV of the CAAA and regulates the emissions of sulfur dioxide (SO₂) and nitrogen oxides (NO_x) from electric generating units (EGUs) that use fossil fuel (e.g., coal, gas, oil).

This NAPAP Report to Congress (RTC) provides an update on implementation of the ARP and the status and trends in emissions, deposition, air quality, surface water chemistry, ecosystem effects, and visibility. The report assesses the state of the science and discusses the expected effects of further emission reductions.

2

3 The National Acid Precipitation Assessment Program (NAPAP) Reports to Congress (RTCs) provide an
4 assessment of the results of Title IV of the Clean Air Act Amendments (CAAA). Through the Acid Rain
5 Program (ARP), Title IV requires significant reductions in the emissions of sulfur dioxide (SO₂) and
6 nitrogen oxides (NO_x) from power generation sources. These emissions contribute to acid deposition and
7 the formation of fine particulate matter (PM_{2.5}) and ozone, which ultimately lead to a wide range of
8 environmental impacts, including harm to human health and visibility impairment.

9 NAPAP assesses the implementation of the ARP, including the impacts and benefits of the sulfur and
10 nitrogen emission reductions achieved by the program. This NAPAP RTC is written to effectively and
11 fully communicate the results of the assessment to decision makers. Congress has asked NAPAP to assess
12 all available data and information to answer two questions:

- 13 1. *What are the costs, benefits, and effectiveness of Title IV?* This question addresses the costs and
14 economic impacts of complying with the ARP, as well as the benefit analyses associated with
15 various human health and welfare effects, including reduced visibility and effects on ecosystems.
- 16 2. *What reductions in deposition rates are needed to prevent adverse ecological effects?* This is a
17 complex question addressing how much deposition can occur before negative environmental
18 effects take place.

19 Accomplishments

20 The ARP has accomplished much over the years to enhance both human welfare and the environment
21 through reducing SO₂ and NO_x emissions. However, it is important to note that although the ARP plays a
22 significant role in these emission reductions, the program does not control all sources of nitrogen and
23 sulfur. For example, the ARP does not regulate vehicular or agricultural sources, both of which emit
24 nitrogen and, to a lesser extent, sulfur compounds. In some areas, emissions from these other sources can
25 be significant.

26 Achievements of the ARP include the following:

- 27 ■ Reduced SO₂ and NO_x emissions
- 28 ■ Lower-than-predicted implementation costs

- 1 ▪ Reduced incidences of health impacts such as asthma, bronchitis, and premature mortality due to
- 2 PM_{2.5} and ozone, leading to very substantial public health benefits
- 3 ▪ Improved visibility at some sites
- 4 ▪ Improved conditions in some acidified lakes and streams so that they can once again support fish
- 5 and other aquatic life
- 6 ▪ Reduced nitrogen deposition to sensitive forests, such as those along the Appalachian Mountains
- 7 ▪ Reduced nitrogen deposition to nitrogen-sensitive coastal waters along the East Coast.

8 A few of the specific accomplishments that have resulted from controls and actions of the ARP are
 9 highlighted below.

10 **Reduced SO₂ and NO_x Emissions**

11 Title IV of the 1990 CAAA requires significant reductions of
 12 SO₂ and NO_x emissions from specified electric generating
 13 units (EGUs). Between 1980 and 2008, ARP sources (i.e.,
 14 EGUs) reduced their SO₂ emissions 56%, and between 1990
 15 and 2008, the units reduced their SO₂ and NO_x emissions by
 16 52% and 55% respectively (see **Figure I-1**). Starting in 2007,
 17 SO₂ emissions were below the final SO₂ cap of 8.95 million
 18 tons set for compliance in 2010.

19 **Significant Health Benefits**

20 Fine particles formed from SO₂ and NO_x emissions and ozone
 21 formed from NO_x emissions can cause respiratory and
 22 cardiovascular health problems in humans, especially to more
 23 sensitive groups such as children, the elderly, and individuals
 24 with pre-existing conditions. One of the results of reduced
 25 emissions is cleaner air with fewer fine particles and less
 26 ozone. Recent analyses (see Chapter 1) translate improved
 27 human health into tangible economic benefits. In 2010, the
 28 public health benefits of having fewer particles in the air range
 29 from \$171 billion to \$413 billion. The public health benefits
 30 related to reduced ozone are \$4.1 to \$17.0 billion annually.

31 **Improved Aquatic Ecosystem Condition**

32 Lakes and streams have been monitored in acid-sensitive regions of the eastern United States to provide
 33 information on the effects of acid rain and the response of these waterbodies to emission reductions.
 34 Many lakes and streams previously demonstrated to have been acidified by atmospheric deposition, such
 35 as those in the Appalachian Mountains in Pennsylvania, the Adirondack Mountains of New York, and the
 36 mountainous regions of New England, are showing improved conditions. In these regions, the
 37 concentration of sulfate in many lakes and streams has decreased as SO₂ emissions have decreased. In
 38 addition, many lakes and streams are less acidic, providing conditions for improved ecosystem health.

39 **Looking Forward**

40 Ecological recovery is a complex process. Although NAPAP has identified improving conditions in some
 41 places, these trends are not evident at all monitored sites, nor are positive trends evident nationwide. In
 42 the southern Appalachian Mountains in the eastern United States; the Front Range of the Rocky
 43 Mountains; and the far western states, the condition of lakes and streams is not improving. In sensitive
 44 mountainous areas of the western United States, where even small levels of nitrogen deposition may have

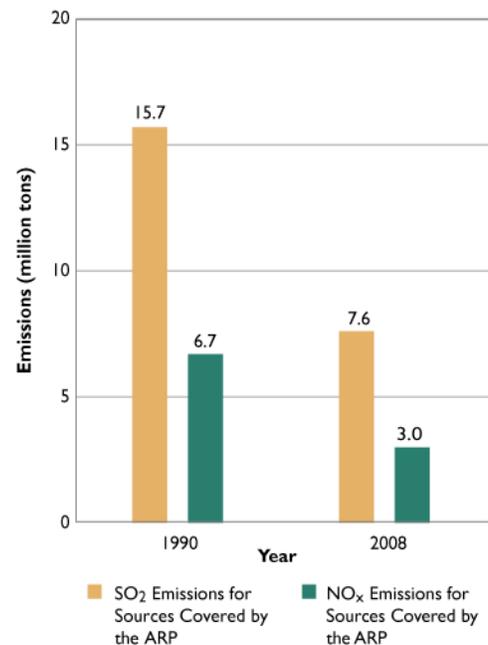


Figure I-1. Emissions from ARP sources in 1990 and 2008 (Prepared by U.S. EPA).

1 negative impacts, ecosystem health may be continuing to decline. Investigations are currently under way
2 to ascertain the ecosystem condition and trends in these locations.

3 **Structure of the Report**

4 This NAPAP RTC is directed to Congress, but it provides valuable economic and scientific information
5 to all public officials who are responsible for determining or evaluating air quality policy. The goal of this
6 report is to present highly technical information pertinent to current public policy issues in a format that
7 can be understood by the nonscientific reader. Where more scientific or economic detail is desired,
8 references are noted in the text and provided at the end of the chapters. Chapter 1, 2, and 3 address the
9 first question posed to NAPAP by Congress, and Chapter 4 focuses on the Congress's second question.

10 Chapter 1 of this report presents the status of implementation of Title IV, including information on ARP
11 design, compliance, costs, and allowance transactions.

12 Chapter 2 presents an analysis of the observed changes, both past and present, in emissions of acid rain
13 precursors, air pollutant concentrations, deposition of acidic species, and the measured effects of
14 acidifying deposition on surface water quality and visibility.

15 Chapter 3 covers advances in the state of the science since the last NAPAP RTC regarding atmospheric
16 deposition and the impacts of acid deposition on aquatic and terrestrial ecosystems, including ecosystems
17 in the Rocky Mountains, the far western United States, and coastal areas. Chapter 3 also reports on
18 research on critical loads, the interactions between atmospheric deposition and climate change, and multi-
19 pollutant interactions.

20 Chapter 4 addresses the question posed by Congress in Title IX of the 1990 CAAA regarding the
21 ecological impacts of further emission reductions and the uncertainties in the estimated relationships
22 between emission reductions and the resulting ecological effects. Several scenarios representing a range
23 of additional emission reductions are used to investigate the effects on acid-sensitive ecosystems in the
24 eastern United States.

1. Acid Rain Program Elements and Implementation

Established under Title IV of the 1990 CAAA, the ARP requires major emission reductions of SO₂ and NO_x, the primary precursors of acid rain, from the electric power industry. Since its implementation in 1995, the ARP has achieved significant emission reductions as electricity generation has increased. This chapter focuses on the ARP, including descriptions of the program and its sources, program compliance, the allowance market, ARP benefits and costs, and tools used to assess the progress of the program.

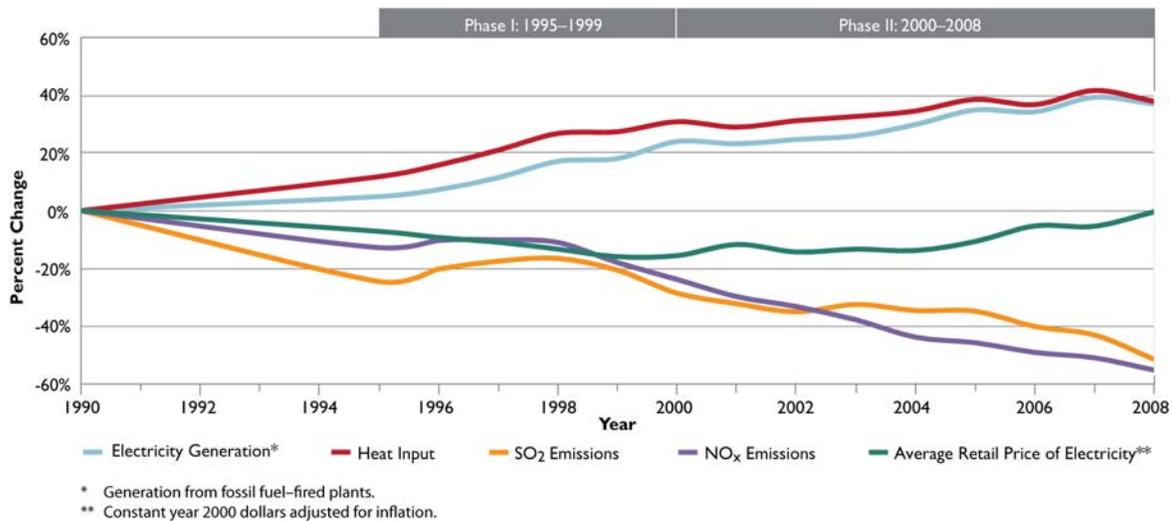
1.1 Overview of Emission Reductions

The implementation of Title IV has successfully reduced emissions of SO₂ and NO_x from EGUs. Under Title IV of the 1990 CAAA, Congress established a permanent cap on the total amount of SO₂ that may be emitted by EGUs in the conterminous United States. This cap has been phased in, with the final 2010 SO₂ cap set at 8.95 million tons, a level of about one-half of the emissions from EGUs in 1980. In 2008, approximately 3,572 EGUs were subject to the SO₂ provisions of Title IV. By 2008, the sources (i.e., EGUs) covered by the ARP had reduced their combined SO₂ emissions by approximately 56% from 1980 levels and 52% from 1990 levels. NO_x reductions under the ARP apply to a subset of coal-fired EGUs and are regulated in a manner that is closer to a more traditional, rate-based regulatory system. In 2008, the 969 sources subject to ARP NO_x regulations emitted 55% fewer emissions than in 1990¹. Heat input and electricity generation increased by approximately 40% over this same period, and the average retail price of electricity was about the same in 2008 as it was in 1990 (**Figure 1-1**).

At a Glance: ARP Results in 2008

- SO₂ emissions: 7.6 million tons
- SO₂ compliance: 100%
- SO₂ allowances: Allowance bank increased by almost 2 million allowances from 2007 levels
- SO₂ allowance prices: Since July 2008, allowance prices have fallen sharply, with a monthly average nominal price in May 2009 of \$71/ton
- NO_x emissions: 3.0 million tons
- NO_x compliance: 100%

The emission reductions achieved under Title IV are discussed in more detail in Chapter 2 of this report.



Source: Energy Information Administration (electricity generation, retail price); U.S. EPA (heat input and emissions, representing all affected ARP units), 2009a.

Figure 1-1. Trends in electricity generation, fossil fuel energy use, prices, and emissions from power plants affected by the ARP, 1990–2008 (U.S. EPA, 2009a).

¹ As described in Chapter 2 of this report, other programs—such as the Ozone Transport Commission (OTC), the NO_x Budget Program under EPA’s NO_x State Implementation Plan (SIP) Call, and other regional and state NO_x emission-control programs—also contributed significantly to the NO_x reductions achieved by ARP sources in 2008.

1 1.2 Acid Rain Program Design

What Is Cap and Trade?

Cap and trade is a policy tool for protecting human health and the environment by controlling large amounts of pollution from a group of sources. A cap-and-trade program first sets a cap, or maximum limit, on pollution emissions. The cap is chosen in order to achieve a desired environmental effect. Sources covered by the program then receive authorizations to emit in the form of emission allowances, with the total amount of allowances limited by the cap. Each source can design its own compliance strategy to meet the overall reduction requirement. For example, under the ARP, sources can sell or purchase allowances, install pollution controls, and implement efficiency measures, among other options. Individual control requirements are not specified under a cap-and-trade program, but each emission source must surrender allowances equal to its actual emissions in order to comply. To guarantee that the overall cap is achieved, sources must completely and accurately measure and report all emissions.

2

3 1.2.1 SO₂ Program

4 The SO₂ emission reduction program created under Title IV represents a substantial change from
5 traditional command and control regulatory approaches that establish source-specific emission
6 limitations. Instead, the program combines an overall emission cap for SO₂, which ensures that emission
7 reductions are achieved and maintained with a trading system that facilitates lowest-cost emission
8 reductions. The ARP features tradable SO₂ emission allowances, where one allowance is a limited
9 authorization to emit one ton of SO₂. A fixed number of allowances is issued by the government, and
10 these allowances may be bought, sold, or banked for future use by EGUs or other parties (e.g., utilities,
11 brokers, or anyone else interested in holding allowances). Existing sources are allocated allowances each
12 year. New sources do not receive allowances and instead must buy them; however, the required purchase
13 of allowances has not been a barrier to market entry (i.e., new sources have been able to acquire the
14 allowances needed to compete effectively in the market). At the end of each year, all affected sources are
15 obliged to surrender to the U.S. Environmental Protection Agency (EPA) the number of allowances that
16 correspond to their annual SO₂ emissions (one allowance for each ton of SO₂).

17 Title IV establishes a two-phased tightening of the SO₂ emissions cap, adjusting the allocation of SO₂
18 allowances to fossil fuel-fired EGUs to reach the permanent cap on the number of allowances of 8.95
19 million tons annually in 2010. Once the bank of unused allowances is depleted, the limit on allowance
20 allocations constrains emissions to the level of the cap.

21 Phase I of the ARP (1995–1999) affected 263 of the larger (>100 megawatt [MW]), higher-emitting
22 EGUs, which are located primarily in the central and eastern United States. In 2008, Phase I EGUs
23 represented 18% of generation of total electricity generation from fossil fuels. Phase I SO₂ allowance
24 allocations were distributed to each source based on the following formula: the product of an emission
25 rate of 2.5 pounds (lb) SO₂/million British thermal units (MMBTU) of heat input and its average heat
26 input for 1985–1987. Some Phase II sources chose to “opt-in” to Phase I and comply early, bringing the
27 total number of units participating in Phase I to more than 400.

28 Phase II began in 2000 and extended to all existing EGUs serving generators larger than 25 MW and all
29 new fossil fuel-fired generation units throughout the country. In 2008, the total number of units covered
30 by the SO₂ criteria was 3,572 sources, which represented 95% of total electricity generation from fossil
31 fuels. In Phase II, all Phase I and Phase II SO₂ affected sources are allocated allowances equivalent to an
32 amount no greater than the product of 1.2 lb SO₂/MMBTU and their average heat input for 1985–1987.

33 Title IV requires that sources monitor emissions continuously and report their emissions quarterly. Failure
34 to surrender sufficient allowances results in two significant automatic penalties. Any source that fails to

1 hold enough allowances to match its SO₂ emissions for the previous year must pay to EPA, by July 1, an
2 automatic penalty of \$2,000 (inflation-adjusted to \$3,337 for 2008) per ton of emissions in excess of
3 allowances held. The source must also immediately surrender to EPA an amount of allowances, issued for
4 the year the payment is due, equaling the tons of excess emissions. A source may sell or bank for future
5 use any remaining SO₂ allowances not needed for compliance during a year. Sources may use these
6 banked allowances as needed to comply with the program in future years until the bank is depleted.

7 Title IV mandates that a limited number of allowances allocable to existing sources be withheld and
8 auctioned, with revenues from the auction returned pro rata to existing sources. The annual SO₂ auction
9 provides an opportunity for sources to buy and sell allowances. The auctions help ensure that new sources
10 have an opportunity to obtain allowances beyond those allocated initially to existing EGUs. Complete
11 results of the annual SO₂ Allowance Auction are available at
12 <http://www.epa.gov/airmarkets/trading/auction.html>.

The Clean Air Interstate Rule

In March 2005, EPA promulgated the Clean Air Interstate Rule (CAIR) to achieve further emission reductions beyond levels reached under the ARP and other programs, such as the NO_x State Implementation Plan (SIP) Call. CAIR was designed to help states in the eastern United States address ozone nonattainment and attain the National Ambient Air Quality Standards (NAAQS) for PM_{2.5} by reducing and capping SO₂ and NO_x emissions in 28 eastern states and the District of Columbia. CAIR created three separate compliance programs: an annual NO_x program, an ozone season NO_x program, and an annual SO₂ program. Each of the three programs uses a two-phased approach, with declining emission caps in each phase. The first phase began in 2009 for the NO_x annual and NO_x ozone season programs, and started in 2010 for the SO₂ annual program. The rule also establishes a second phase for all three programs beginning in 2015. CAIR gave affected states SO₂ and NO_x emission budgets and the flexibility in their state implementation plans (SIPs) to reduce emissions using a strategy that best suits their circumstances, including EPA-administered, regional cap-and-trade programs as one option. On July 11, 2008, the U.S. Court of Appeals for the D.C. Circuit issued a ruling vacating CAIR in its entirety. The Court subsequently remanded CAIR to EPA on December 23, 2008, leaving CAIR in place until EPA issues new rules to replace CAIR. On July 6, 2010, EPA released a proposed Transport Rule to replace CAIR.

13

14 1.2.2 NO_x Program

15 In contrast to the system established for SO₂ emissions, the ARP does not establish tradable emission
16 allowances for NO_x emission reductions. Instead, sources control how much NO_x is emitted from coal-
17 fired boilers based on the use of cost-effective control technologies for each unit of fuel consumed (pound
18 of NO_x [lb NO_x] per MMBTU). There are two phases of the NO_x component: Phase I began in 1996
19 (delayed 1 year because of litigation), and Phase II began in 2000. During Phase I, which applied to
20 specific coal-fired boilers statutorily affected by Phase I SO₂ requirements, the NO_x emission rate was set
21 at 0.50 lb NO_x/MMBTU for dry-bottom, wall-fired units and 0.45 lb NO_x/MMBTU for tangentially fired
22 units. Beginning in 2000, Phase II plants were required to meet emission rates between 0.40 lb
23 NO_x/MMBTU and 0.86 lb NO_x/MMBTU, depending on the type of boiler. In 2008, 969 units were
24 subject to ARP NO_x program requirements.

25 Although the ARP does not include NO_x emission trading, sources are provided a degree of flexibility
26 through emission-averaging provisions, whereby a company can meet its NO_x emission limitations by
27 averaging the emission rates of two or more boilers. This enables sources to reduce their NO_x emissions at
28 lower cost by allowing them to over-control at EGUs where it is technically easier to control emissions.
29 At the end of the year, sources must demonstrate compliance with NO_x emission requirements by

1 achieving an annual emission rate at or below mandated levels, as outlined in their EPA-approved
 2 compliance plans. It is important to note that a number of other programs have contributed to NO_x
 3 emission reductions from ARP sources (see Chapter 2 for a description of these programs).

4 **1.3 Title IV Affected Sources**

5 **1.3.1 SO₂ Sources**

6 Sources that are subject to the SO₂ component include boilers or combustion turbines that burn fossil fuel,
 7 serve generators with a design capacity greater than 25 MW, and produce electricity for sale. Several
 8 types of units meeting these criteria are not affected by the ARP. These include simple combustion
 9 turbines that began to produce electricity for sale before November 15, 1990; cogeneration units whose
 10 annual electricity sales remain below the threshold established by regulation; and specific qualifying
 11 facilities and independent power producers that are contractually bound to sell electricity at a price that
 12 was established before November 15, 1990. Despite these exceptions, almost all non-cogeneration units
 13 that have total design capacity greater than 25 MW and that produce electricity for sale now must
 14 participate in the ARP.

15 **1.3.2 NO_x Sources**

16 Some of the sources subject to the SO₂ requirements of Title IV are also subject to the Title IV NO_x
 17 requirements. All units where coal accounted for more than 50% of heat input for at least 1 year during
 18 the 1990 through 1995 time period and that are configured for a specific type of boiler (i.e., cell burner,
 19 cyclone, dry-bottom wall-fired, tangentially fired, vertically fired, or wet bottom) are affected by the Title
 20 IV NO_x criteria.

21 **Table 1-1** lists the sources affected by Title IV NO_x emission components in 2008. For more details on
 22 ARP applicability criteria, see 40 Code of Federal Regulations (CFR) 72.6, which provides ARP
 23 applicability regulations established under Title IV.

24 **Table 1-1. Title IV NO_x Affected Units by Boiler Type and NO_x Emission Limit**

Coal-Fired Boiler Type	Title IV Standard NO _x Emission Limits (lb/MMBTU)	Number of Units
Phase I Group 1 Tangentially Fired	0.45	133
Phase I Group 1 Dry-Bottom, Wall-fired	0.50	107
Phase II Group 1 Tangentially Fired	0.40	300
Phase II Group 1 Dry-Bottom, Wall-fired	0.46	294
Cell Burners	0.68	37
Cyclones >155 MW	0.86	54
Wet-Bottom > 65 MW	0.84	20
Vertically Fired	0.80	24
Total All Units		969

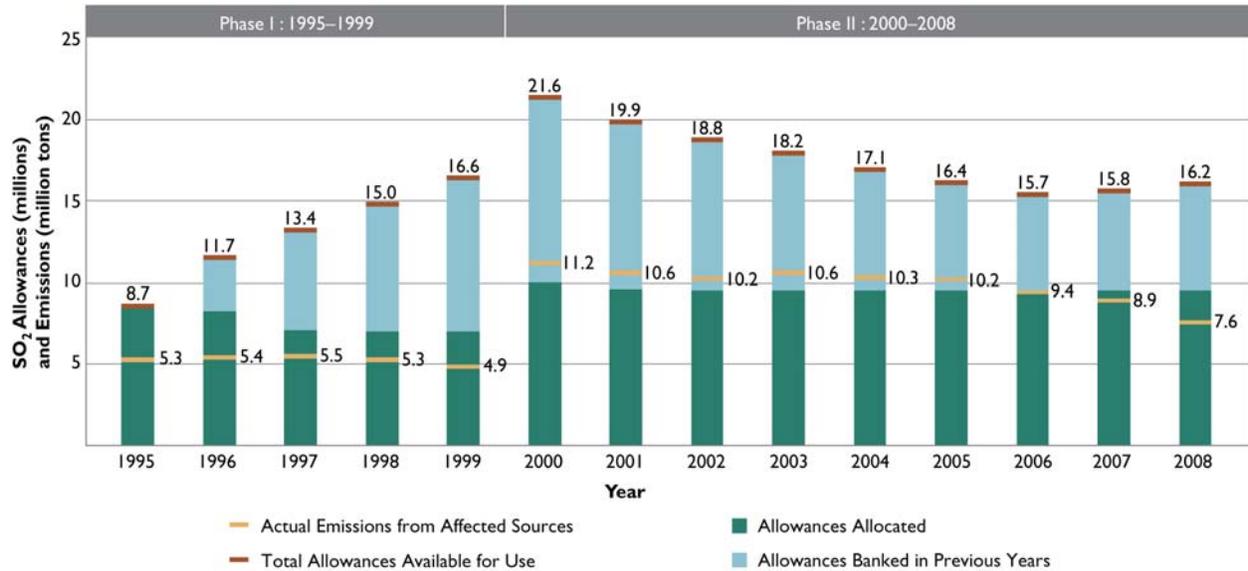
Source: U.S. EPA, 2009b

25 **1.4 Compliance**

26 **1.4.1 SO₂ Compliance**

27 The Title IV SO₂ program has achieved a near-perfect compliance record since the program took effect in
 28 1995. In 2008, as in each year since 2005, the program achieved 100% compliance, and all ARP facilities
 29 complied with the requirement to hold enough allowances to cover SO₂ emissions. EPA allocated 9.5
 30 million SO₂ allowances under the ARP for 2008. Together with the 6.7 million unused allowances carried

1 over (or banked) from prior years, 16.2 million allowances were available for use in 2008 (see
 2 **Figure 1-2**). ARP sources emitted approximately 7.6 million tons of SO₂ in 2008, less than the
 3 allowances allocated for the year, and far less than the total allowances available. As a result, the bank
 4 increased between 2007 and 2008 by nearly 2 million allowances to 8.6 million, a 28% increase. The
 5 bank includes the unused allowances from previous years, plus the unused allowances allocated in 2008
 6 (i.e., all of the allowances above the yellow line in Figure 1-2). In 2010, the total number of Title IV SO₂
 7 allowances allocated annually will drop to 8.95 million and remain statutorily fixed at that annual level.



8 **Figure 1-2. SO₂ emissions and the allowance bank, 1995–2008 (U.S. EPA, 2009a).**

10 The existence of the allowance market has given some sources the incentive to reduce their SO₂ emissions
 11 below the level of their allowance allocation in order to sell their allowances to other sources or bank
 12 them for use in future years. Other sources have been able to postpone or reduce expenditures for
 13 pollution control by purchasing allowances from sources that controlled beyond their allowance allocation
 14 level. As shown in Figure 1-2, the “bank” or store of unused allowances grew throughout Phase I (1995–
 15 1999) as sources reduced emissions more than required. These “early reductions” reduced the amount of
 16 fine particles and acid deposition in the early years of Title IV implementation, increasing the human
 17 health and ecological benefits of the program in those years. Beginning in 2000, with Phase II of the
 18 program, the set of sources covered by the program expanded, and the tighter Phase II emission cap took
 19 effect. As Figure 1-2 shows, sources began to use previously banked allowances in addition to allocations
 20 from the current year to comply with Title IV. As a result, emission levels for 2000–2005 were greater
 21 than annual allocations as sources used banked allowances for compliance.

22 **1.4.2 NO_x Compliance**

23 Affected sources can comply by either meeting a unit-specific NO_x emission rate or including two or
 24 more units in an emission rate averaging plan (see text box). As with the SO₂ program, the NO_x program
 25 has had a high rate of compliance. Since 2002, there have only been 2 years in which a single unit was out
 26 of compliance. In 2008, all 969 units that were subject to ARP NO_x criteria achieved compliance.

Sources Achieved 100% NO_x Compliance in 2008 Using a Variety of NO_x Compliance Plan Options

Standard Limitation—A unit with a standard limit meets the applicable individual NO_x limit prescribed for its boiler type under 40 CFR Parts 76.5, 76.6, or 76.7 (290 units used this option in 2008).

Alternative Emission Limit (AEL)—A utility can petition for a less-stringent AEL if it properly installs and operates the NO_x emission-reduction technology prescribed for that boiler, but it is still unable to meet its standard limit. EPA determines whether an AEL is warranted based on analyses of emission data and information about the NO_x control equipment (six units used this option in 2008).

Emissions Averaging—Many companies meet their NO_x emission-reduction requirements by choosing to become subject to a group NO_x limit, rather than by meeting individual NO_x limits for each unit. The group limit is established at the end of each calendar year. The group rate must be less than or equal to the British thermal unit (BTU)-weighted group rate that the units would have had if each had emitted at their standard limit rate (673 units used this option in 2008).

Note: Unit counts do not include those with a retired unit exemption.

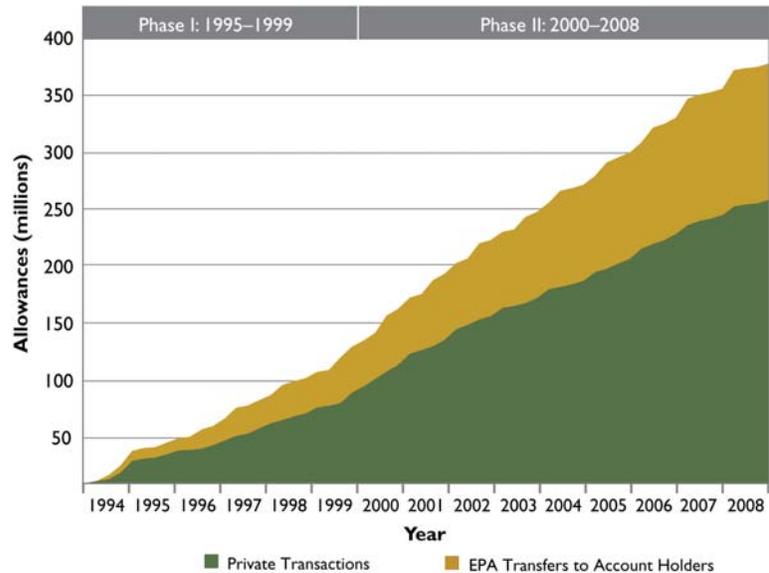
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2 **1.5 2008 SO₂ Allowance Market**

3 The number of allowances (authorizations to emit SO₂) allocated to each source in any given year is
 4 determined by the CAA. A recent review (Burtraw and Szambelan, 2009) of emission trading generally
 5 concluded that the SO₂ allowance market has “been liquid and active, and according to most observers,
 6 [has] worked well in achieving the emission caps at less cost than would have been achieved with
 7 traditional approaches to
 8 regulation.”

9 **Figure 1-3** shows the cumulative
 10 volume of SO₂ allowances
 11 transferred under the ARP. The
 12 figure differentiates between
 13 allowances transferred in private
 14 transactions and those annually
 15 allocated and transferred to source
 16 accounts by EPA.

17 Private transactions are indicative
 18 of both market interest and use of
 19 SO₂ allowances as a compliance
 20 strategy. Of the nearly 379 million
 21 allowances transferred since 1994,
 22 about 68% were traded in private
 23 transactions. In December 2001,
 24 parties began to use a system
 25 developed by EPA to allow online
 26 SO₂ allowance transfers. In 2008,
 27 account holders registered over
 28 99% of all private allowance
 29 transfers through EPA’s online
 30 transfer system. Allowance
 31 transfers are posted and updated
 32 daily on <http://www.epa.gov/airmarkets>.



Note: EPA transfers include allocations to sources; transactions at auction; and conservation, renewable energy, and Phase I extension reserves.

Figure 1-3. Cumulative SO₂ allowances transferred under the ARP, 1994-2008 (U.S. EPA, 2009a).

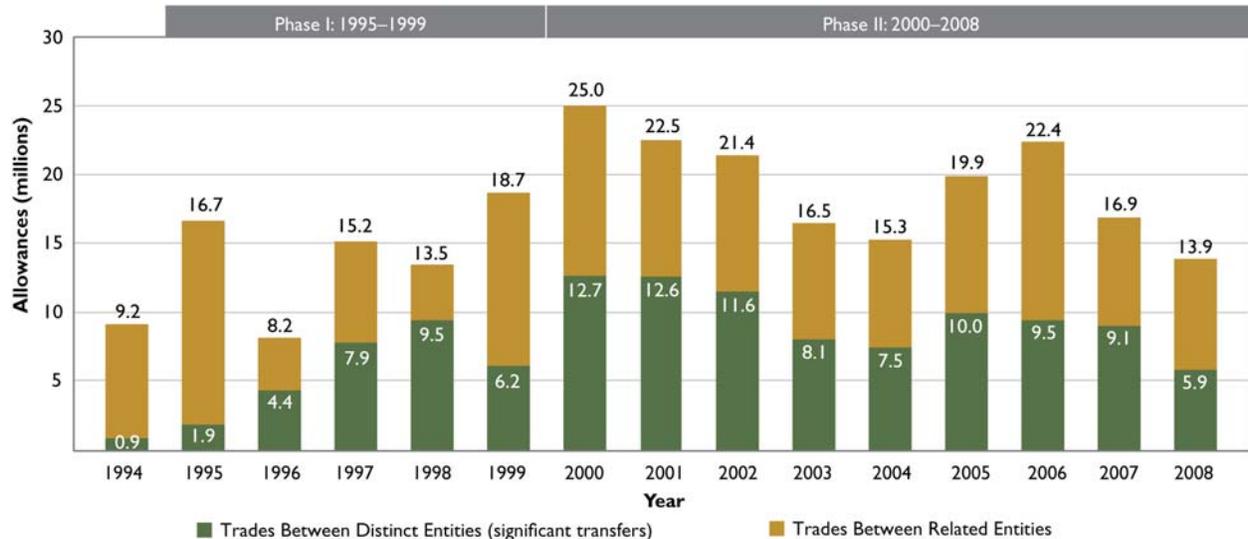
How Are Allowances Traded and Tracked?

Once allowances have been auctioned and allocated, utilities can buy, sell, trade, or save them to meet their compliance needs. Along with the utilities that hold allowances for compliance purposes, other parties, such as brokers, environmental groups, and private citizens, maintain accounts in EPA’s Allowance Management System (AMS). The AMS database records account balances and transaction records and allows public access to the trading history of each allowance until it is finally retired. EPA does not maintain any sensitive business data, such as the price associated with allowance transfers. Allowance brokers and other market participants generally maintain a market price index (MPI) to track trends in prices over time and provide market signals similar to other commodity markets.

Most allowance transactions take place in the over-the-counter market, where prices are determined by each day’s bids and offers, and immediate settlement cash trades are enacted bilaterally or through brokers. Once trading parties agree on a price, they generally complete the transaction using standard contracts developed by trade associations or other market players (see, for example, the sample contract available at [Hhttp://www.environmentalmarkets.org](http://www.environmentalmarkets.org)). EPA provides a list of brokers and environmental groups that may be interested in facilitating trades or in helping parties retire allowances voluntarily (see [Hhttp://www.epa.gov/airmarkets/trading/buying.html](http://www.epa.gov/airmarkets/trading/buying.html)). At some point after a transaction is complete, the account representative of the transferring or selling party will usually register the transfer of allowances with EPA. The representative can submit a paper form or transfer the allowances online using the Clean Air Markets Division (CAMD) Business System (see [Hhttp://www.epa.gov/airmarkets/business/transfer.html](http://www.epa.gov/airmarkets/business/transfer.html)).

1

2 In 2008, 3,236 private SO₂ allowance transfers involving approximately 13.9 million allowances of past,
 3 current, and future vintages were recorded in EPA’s Allowance Management System (AMS). About 5.9
 4 million SO₂ allowances (42%) were transferred in economically significant transactions (i.e., between
 5 economically unrelated parties). Transfers between economically unrelated parties are “arm’s length”
 6 transactions and are considered a better indicator of an active, functioning market than are transactions
 7 among the various facility and general accounts associated with a given company. In the majority of all
 8 private transfers, allowances were acquired by power companies. **Figure 1-4** shows the annual volume of
 9 SO₂ allowances transferred under the ARP (excluding allocations, retirements [i.e., used allowances
 10 surrendered], and other transfers by EPA) since official recording of transfers began in 1994.



11
 12
 13

Figure 1-4. SO₂ allowances transferred under the ARP (U.S. EPA, 2009a).

1 Over the first decade of the ARP, SO₂
 2 allowance prices were stable and significantly
 3 lower than projected. Just prior to the
 4 beginning of the program in 1995, SO₂
 5 allowance prices on the spot market were close
 6 to \$150 per ton. The cost of allowances was
 7 initially projected to be between \$250 and
 8 \$500 per ton during Phase I (1995 to 1999)
 9 and \$500 to \$1,000 per ton in Phase II (beyond
 10 2000). Actual allowance prices in Phase I were
 11 in the \$100 to \$200 range, with a low of \$65 in
 12 1996. Allowance prices did display some
 13 variability (as would commodities in any
 14 market), but it was within this very limited
 15 range and tended to be concentrated around
 16 times of regulatory change or uncertainty, such
 17 as the beginning of Phase I in 1995 and the
 18 transition to Phase II in 2000. Even as the more stringent Phase II requirements became effective in 2000,
 19 however, prices remained generally below the \$200 mark until they started to rise at the end of 2003 with
 20 the proposal of the Clean Air Interstate Rule (CAIR).

SO₂ Allowance Market in Brief (close of 2008)

Total Value of the SO₂ Allowance Market: \$2.9 billion*

- Average Nominal Price: \$179 per ton
- Total Allowance Volume (allowable emissions): 16,227,082

2008 Private Transactions

- 3,236 transactions moving 13.9 million allowances
- 42% of allowances transferred between economically unrelated parties

* Total value of allowance market is a snapshot based on the average nominal price as of December 2008 (\$179/ton) and total allowance volume available for 2008 compliance.
 Source: U.S. EPA, 2009a

21 When CAIR was proposed in late 2003, allowance prices were influenced by the more stringent CAIR
 22 SO₂ cap and new compliance deadline. After CAIR was finalized in March 2005, SO₂ allowance prices
 23 continued to trend upward. CAIR was the most significant driver of the price adjustment that began in
 24 2004 and culminated with prices around \$1,600 per ton for a short time in December 2005. The ARP SO₂
 25 market essentially became the CAIR SO₂ market. In 2008, the value of the SO₂ allowance market
 26 experienced a 65% price decline; the monthly average allowance price fell from \$509 per ton in January
 27 to \$179 per ton by December (based on the MPI). That decline continued in 2009, with the SO₂ allowance
 28 price falling to an average of \$71 per ton by May 2009 (see **Figure 1-5**). Together with the price decline,
 29 the volume of significant transactions fell sharply in 2008.



Source: CantorCO2e, 2009

Figure 1-5. Average monthly SO₂ allowance price, August 1994–May 2009 (U.S. EPA, 2009b).

Market observers should not confuse temporary high prices in the market response to major regulatory changes (i.e., more or less regulation), where buyers and sellers are searching for a new equilibrium based on available information they have from consultants and various services, with price volatility. EPA and market analysts have identified these regulatory forces—the CAIR emission caps and compliance deadlines, followed by the rule changes resulting from the July 2008 CAIR court decision—as the primary factors affecting current market conditions in the period 2004–2008 and not inherent volatility in cap-and-trade programs due to shifts in other variables that influence the market. For further analysis, see <http://www.epa.gov/airmarkets/resource/docs/marketassessment.pdf>.

1

2 1.6 Program Benefits and Costs

3 1.6.1 Benefits

4 Emissions of SO₂ and NO_x result in a variety of air pollutants, including not only the strong acids that
5 impact ecosystems through atmospheric deposition but also the atmospheric concentrations of particulate
6 matter and ground-level ozone. These multiple air pollutants have important impacts on human health and
7 a wide range of ecological and environmental resources. Due to the multi-pollutant nature of these
8 environmental impacts, the emission reductions achieved by the ARP result in many societal benefits,
9 including the following:

- 10 ■ **Health Benefits.** These include avoided premature mortality and avoided morbidity associated
11 with reduced human exposures to air pollutants, such as PM_{2.5} and ozone, which are secondary air
12 pollutants that form as a result of SO₂ and NO_x emissions.
- 13 ■ **Visibility Benefits.** Reductions in air pollutants, particularly in PM_{2.5}, improve visibility, which
14 leads to physical and economic benefits in both recreational and residential settings.
- 15 ■ **Agricultural and Forest Productivity Benefits.** Ground-level ozone inhibits plant growth; as a
16 result, reductions in ozone concentrations yield physical and economic benefits in the form of
17 enhanced agricultural and forest productivity.
- 18 ■ **Ecological Benefits.** A wide range of ecological resources are susceptible to damage when
19 exposed to ambient air pollution or deposition of pollutants to terrestrial or aquatic environments.
20 For a small portion of these effects, it is possible to quantify and estimate the economic value of
21 avoided pollutant exposure.
- 22 ■ **Materials Damage Benefits.** Some materials are susceptible to accelerated deterioration when
23 exposed to air pollution; as a result, reduction in air pollution can extend the life of these
24 materials, yielding physical and economic benefits.

25 A comprehensive assessment of the benefits of ARP implementation would evaluate the entire suite of
26 human health and environmental benefits resulting from ARP emission reductions. However, because
27 human health impacts are more readily quantified, air pollution benefits analyses traditionally have
28 focused on human health rather than on ecological health, aesthetic effects, or natural resource
29 productivity. For example, the science and economics of human health benefits assessment and valuation
30 is much better developed than the corresponding science in support of assessing the effects of emission
31 reductions on ecological systems. Moreover, the monetized human health benefits of reducing air
32 pollution generally significantly outweigh monetary benefits in other categories, such as improvements in
33 visibility or ecosystem condition. For these reasons, this report focuses on human health improvements in
34 assessing the monetary benefits of ARP implementation.

35 Still, it is important to recognize that benefits beyond human health improvements result from emission
36 reductions such as those achieved by the ARP. For example, a recent draft report (U.S. EPA, 2010b)

1 fulfilling CAAA Section 812 requirements quantified the overall benefits of implementing the 1990
2 CAAA and estimated that all CAAA programs taken together will result in \$40 billion in benefits due to
3 improvements in recreational and residential visibility in 2010. There have also been recent advances in
4 efforts to quantify the ecological benefits of emission reductions. For example, Banzhaf et al. (2006) used
5 a contingent valuation study to estimate the total economic value of reducing ecological impacts of acid
6 deposition in New York's Adirondack Park from air quality policies that reduced SO₂ and NO_x emissions.
7 The study estimated total statewide benefits ranging from \$336 million to \$749 million annually, but that
8 total increased to \$1.1 billion when alternative assumptions were used regarding ecological change.
9 Significant future analytical work and basic ecological and economic research is needed to build a
10 sufficient base of knowledge and data to support an adequate assessment of ecological benefits. For the
11 current analysis, this incomplete coverage of effects represents a significant source of uncertainty in
12 assessing the benefits of ARP implementation.

13 Since publication of the last NAPAP RTC (NSTC, 2005), there have been some efforts to quantify the
14 benefits of implementing the ARP. In 2005, a peer-reviewed journal article assessed the human health and
15 welfare benefits of ARP implementation for the prospective year 2010 (Chestnut and Mills, 2005). The
16 benefits were estimated using modeled emission reductions and ambient air quality expected to be
17 achieved in 2010 under the ARP. The majority of the monetized benefits of ARP implementation reported
18 in the study were from the prevention of health-related impacts (e.g., premature death) due to reductions
19 in ambient concentrations of PM_{2.5} and ground-level ozone. A concentration response function developed
20 by Pope et al. (2002) was used to estimate incidences of adult premature mortality as a result of PM_{2.5}
21 exposure. The Chestnut and Mills (2005) study estimated the PM_{2.5} and ozone health-related benefits of
22 the ARP to be \$134 billion and \$5.5 billion annually, respectively; they also estimated benefits from
23 visibility improvements in national parks and wilderness areas in California, the Colorado plateau, and the
24 Southeast at about \$2.5 billion annually.²

25 Since publication of this article (Chestnut and Mills, 2005), the assumptions used to develop human
26 health effects estimates have changed. For example, EPA now also includes concentration-response
27 functions derived from a study by Laden et al. (2006) and an expert elicitation to estimate incidences of
28 adult premature mortality as a result of PM_{2.5} exposure. Additionally, many underlying modeling
29 assumptions have been updated, including population forecasts and baseline incidence rates. A majority
30 of these updated assumptions are discussed in detail in the recent PM_{2.5} Regulatory Impact Analysis (U.S.
31 EPA, 2010a).

32 The benefits analysis included here updates the estimates found in the Chestnut and Mills (2005) study.
33 The analysis included here was performed using EPA's Benefits Mapping and Analysis Program
34 (BenMAP) (U.S. EPA, 2008c). BenMAP is a tool that estimates the impacts of a change in air pollution
35 on human health. Specifically, for this analysis, BenMAP was used to estimate the human-health benefits
36 from a reduction in PM_{2.5} and ground-level ozone pollution due to implementation of ARP emission
37 reductions. The analysis relied on modeled air quality data³ representing expected air quality in 2010,
38 both in the absence of the ARP and with ARP implementation. BenMAP processed these geographically
39 distributed estimates of 2010 air quality to calculate a reduction in PM_{2.5} and ground-level ozone
40 concentrations attributable specifically to ARP implementation. BenMAP combined this pollution
41 reduction with geographically specific population data and baseline incidence data and entered this
42 information into epidemiological functions to estimate health benefits. The epidemiological functions

² Year 2008\$ inflated from \$108 billion, \$4.384 billion, and \$2 billion 2000\$ using Consumer Price Index (CPI) inflation calculator.

³ The original air quality estimates were based on a regulatory air quality modeling platform that has been substantially updated since 2005. The updates include (1) major changes to base year and future case emissions and meteorological inputs, (2) a new air quality model with improved chemistry and other scientific features, and (3) new methods for projecting future air quality relative to current measured data. The effects of these improvements on the benefits from the ARP have not been quantified.

1 used for this analysis were the most recent sets of functions used by EPA for health effects assessments
2 and employed in the PM_{2.5} Regulatory Impact Analysis (U.S. EPA, 2010a).

3 The results of the revised assessment show an increase in the estimated value of PM_{2.5} and ozone health
4 benefits expected from ARP implementation in 2010. Depending on which of two studies is used (Pope et
5 al., 2002 or Laden et al., 2006) as the primary estimate of incidences of adult mortality avoided, the
6 monetized PM_{2.5} benefit increase ranges from 25% to 204% more than was estimated by Chestnut and
7 Mills (2005) (see **Table 1-2**). Using updated methods to assess ground-level ozone benefits results in total
8 benefits ranging from 75% to 319% of those estimated by Chestnut and Mills (2005) (see **Table 1-3**). As
9 mentioned above, these updated benefits do not include human welfare benefits due to improved visibility
10 or changed ecological conditions, such as reduced acidification of lakes and streams.

11 **Table 1-2. Estimated PM_{2.5} Health Benefits due to ARP Implementation in 2010**

Health Effect	Incidences Avoided	Monetized Value (millions; 2008\$)
Adult Mortality^a from PM_{2.5}		
Pope et al., 2002	20,000	\$160,000
Laden et al., 2006	50,000	\$400,000
Range of Expert Elicitation	7,000 to 66,000	\$58,000 to \$520,000
Infant Mortality from PM_{2.5}		
Woodruff et al., 2006	82	\$710
Morbidity from PM_{2.5}		
Acute Bronchitis	28,000	\$2.2
Acute Myocardial Infarction	30,000	\$3,500
Acute Respiratory Symptoms	12,000,000	\$790
Asthma Exacerbation	280,000	\$15
Chronic Bronchitis	12,000	\$5,800
Emergency Room Visits; Respiratory	18,000	\$7.2
Hospital Admissions; Cardiovascular	10,000	\$300
Hospital Admissions; Respiratory	4,800	\$72
Lower Respiratory Symptoms	290,000	\$5.6
Upper Respiratory Symptoms	220,000	\$6.8
Work Loss Days	2,500,000	\$640
Total Value		
Pope et al., 2002		\$170,000
Laden et al., 2006		\$410,000

^a Valuation includes a 3% discount rate for future incidences of premature mortality avoided.

Note: Totals may not reflect individual rows from rounding.

Source: U.S. EPA, 2009a

12 **Table 1-3. Estimated Ozone Health Benefits due to ARP Implementation in 2010**

Health Effect	Incidences Avoided	Monetized Value (millions; 2008\$)
Mortality		
Mortality, Non-Accidental (Ito et al., 2005)	1,900	\$17,000
Mortality, Non-Accidental (Schwartz, 2005)	660	\$5,700
Mortality, Non-Accidental (Bell et al., 2004)	430	\$3,700
Mortality, All Cause (Levy et al., 2005)	2,000	\$17,000

Health Effect	Incidences Avoided	Monetized Value (millions; 2008\$)
Mortality, All Cause (Bell et al., 2005)	1,400	\$12,000
Mortality, Cardiopulmonary (Huang et al., 2005)	720	\$6,200
Morbidity		
Hospital Admissions, Respiratory (age 65 and up)	3,000	\$75
Hospital Admissions, Respiratory (age 0–2)	2,500	\$26
Emergency Room Visits, Respiratory	1,900	\$0.74
School Loss Days	910,000	\$87
Acute Respiratory Symptoms	2,600,000	\$170
Total Value Range		\$4,100–\$17,000

Note: Totals may not reflect individual rows from rounding.

Source: U.S. EPA, 2009a

1.6.2 Costs

In addition to the environmental benefits described above, significant economic benefits also have resulted from using the cap-and-trade mechanism employed by Title IV. Cap-and-trade programs provide sources with flexibility in how they achieve their emission target. The cap establishes the emission level for emission sources; the sources, however, are provided with the flexibility of choosing how to abate their emissions. Each source can choose to invest in abatement equipment or energy efficiency measures, switch to fuel sources with no or reduced emissions, or shutdown or reduce output from higher emitting sources. Cap-and-trade programs also allow sources to trade allowances, providing an additional option for complying with the emission target. Sources that have high marginal abatement costs (i.e., the cost of reducing the next unit of emissions) can purchase additional allowances from sources that have low marginal abatement costs. In this way, both buyers and sellers of allowances can benefit. Sources with low costs can reduce their emissions below their allowance holdings and earn revenues from selling their excess allowances—a reward for better environmental performance. Sources with high costs can purchase additional allowances at a price that is lower than the cost to reduce a unit of pollution at their facility.

A recent analysis (Burtraw and Szambelan, 2009) of Title IV implementation concluded that the program did not fully achieve least-cost and some opportunities for additional costs savings were unrealized, at least during the first several years of the program. Still, multiple studies on Title IV implementation have found the program has had lower-than-expected costs, as well as cost savings compared to conventional regulatory approaches (Burtraw et al., 2005). In its 2005 RTC (NSTC, 2005), NAPAP reported on various cost estimates of Title IV implementation, including how the estimates had changed over time. Early projections of annual Phase I compliance costs ranged from just under \$678 million (ICF, 1989) to \$1.5 billion (EPRI, 1993); later studies estimated that Phase I costs ranged from \$814 million (Ellerman, 2002) to \$940 million (Carlson et al., 2000) (all estimates in 2000\$). The first EPA estimate (1990) for annual Phase II costs was approximately \$6 billion. As the approach of Phase I neared, estimates for Phase II costs declined, with the U.S. General Accounting Office (GAO) estimating Phase II costs of approximately \$2.5 billion per year in 2010 (2000\$) (U.S. GAO, 1994).

The 2005 NAPAP RTC (NSTC, 2005) also provided estimates of the cost of full implementation of Title IV SO₂ emission reductions that were substantially less than predicted in 1990. Estimates provided by Ellerman (2002) and by Carlson et al. (2000) were \$1.3 to \$1.5 billion per year (2000\$) and \$1.1 billion per year (2000\$) by 2010, respectively. An Office of Management and Budget (OMB) analysis presented in the 2005 NAPAP report estimated costs of the SO₂ component between \$1.1 and \$1.8 billion (2000\$) (National Science and Technology Council, 2005). In the most recent study evaluating the cost of implementing only Title IV, Chestnut and Mills (2005) estimated total annualized costs at a slightly higher level than studies reported in the 2005 NAPAP report (NSTC, 2005). Chestnut and Mills (2005)

1 estimated the total annual costs for reducing SO₂ at approximately \$2 billion (2000\$) per year, with NO_x
2 emission reductions costing an additional \$1 billion annually.

3 However, as additional regulations (e.g., CAIR) are developed to control SO₂ and NO_x emissions from
4 power-generation sources, it becomes increasingly difficult to assess the cost of implementing Title IV
5 alone. For example, a report (U.S. EPA, 2010b) under Section 812 of the CAAA analyzing costs and
6 benefits of implementing CAAA programs (Titles I through IV) estimates that the direct compliance costs
7 in the year 2000 of implementing all programs affecting the utility sector was \$1.37 billion (\$2006), or
8 \$1.17 billion when deflated to year \$2000 (using the CPI inflation calculator). Given that a substantial
9 portion of the utility sector emission reductions under CAAA programs between 1990 and 2000 resulted
10 from Title IV implementation, this is a reasonable qualitative estimate of implementing Title IV through
11 2000, and this estimate is generally consistent with the estimates discussed above and included in the
12 2005 NAPAP report (NSTC, 2005). The same Section 812 report (U.S. EPA, 2010b) estimates costs of
13 CAAA implementation in 2010 and 2020, and shows that the cost of CAAA program implementation
14 increases in each of those out years due to the implementation of CAIR and other emission-reduction
15 programs.

16 The costs to the government of administering the Title IV SO₂ component are also less than in
17 conventional regulatory programs. For example, the Title IV performance-based approach eliminates the
18 need to devise source-specific emission limits, review control technologies, and prepare and approve
19 detailed compliance schedules and permits. Because the regulating authority does not need to approve
20 each source's compliance choices, the focus is on ensuring that each source has at least one allowance for
21 each unit of pollution emitted, which entails less administrative resources and expenditure.

Factors Responsible for the Low Cost of SO₂ Reductions

- **Switching to Low-sulfur Fuel.** Low-sulfur coal became less costly to transport at the time when demand increased.
- **Low Scrubber Costs.** Costs were lower than expected.
- **Technological Innovation.** Technological improvements that allowed switching coals emerged quickly.
- **Efficient Allowance Market.** An efficient, high-volume market emerged.
- **Banking of Unused Allowances.** The program offers the flexibility to bank allowances for future use.
- **Low Administrative Costs.** The costs to the government of administering the Title IV SO₂ program are also less than in conventional regulatory programs. For example, Title IV's performance-based approach eliminates the need to devise source-specific emission limits, review control technologies, and prepare and approve detailed compliance schedules and permits.

22

23 1.7 Program Assessment Tools

24 1.7.1 Emission Monitoring and Reporting

25 The ARP requires regulated sources to measure, record, and report emissions using continuous emission
26 monitoring systems (CEMS) or an approved alternative measurement method. The vast majority of
27 emissions are monitored with CEMS; however, alternatives are used at some facilities to provide an
28 efficient means of monitoring emissions from the large universe of EGUs with lower overall mass
29 emissions. **Table 1-4** shows the number of units with and without SO₂ CEMS for various fuel types in
30 2008, as well as the amount of SO₂ emissions monitored using CEMS. Although only 32% of units use
31 CEMS, 99% of all SO₂ emissions from ARP sources are monitored in this fashion.

1 CEMS and approved alternatives are a cornerstone of the
 2 ARP's accountability and transparency. Since the program's
 3 inception in 1995, affected sources have met stringent
 4 monitoring quality assurance and control requirements and
 5 have reported hourly emission data in quarterly electronic
 6 reports to EPA. Using automated software audits, EPA
 7 rigorously checks the completeness, quality, and integrity of these data. All emission data are available to
 8 the public on the Data and Maps Web site maintained by EPA's Clean Air Markets Division (CAMD) at
 9 <http://camddataandmaps.epa.gov/gdm/>. Another CAMD Web site (<http://www.epa.gov/airmarkets/>)
 10 provides access to other data associated with emission trading programs, including reports, maps, charts,
 11 and file downloads that cover source information, emissions, allowances, program compliance,
 12 atmospheric deposition and air quality, and aquatic ecosystem response.

The emission monitoring requirements for the ARP are found in 40 CFR Part 75. Compliance with these provisions is also required for sources participating in the CAIR programs.

13 **Table 1-4. EGUs and SO₂ Emissions Covered by Monitoring Method for the ARP, 2008**

EGU Type	Type of Monitoring System	Number of EGUs Monitored	Percentage of EGUs Monitored	Percentage of SO ₂ Emissions Monitored
Coal-Fired	CEMS	1,055	29.74	98.68
Gas-Fired	CEMS	19	0.54	0.03
	Non-CEMS	2,259	63.69	0.06
Oil-Fired	CEMS	42	1.18	0.21
	Non-CEMS	159	4.48	0.88
Other	CEMS	12	0.34	0.13
	Non-CEMS	1	0.03	0.00

Note: "Other fuel units" include units that combusted primarily wood, waste, or other nonfossil fuel in 2008. The total number of units in the table excludes the 25 affected units that did not operate in 2008.

Source: U.S. EPA, 2009a

14 1.7.2 Air Quality, Deposition, and Ecological Monitoring

15 Air quality, deposition, and ecological monitoring are also important components of the overall
 16 implementation of Title IV. This section presents information about the monitoring networks that are used
 17 to assess the progress of the ARP; other monitoring networks (e.g., National Air Monitoring Stations
 18 [NAMS]) exist, but are not discussed here. Several monitoring networks (Figure 1-6) designed to measure
 19 changes in air quality and acid deposition as a result of emission reductions are currently in operation and
 20 used by the ARP. In addition, a surface water monitoring network in acid-sensitive areas of the eastern
 21 United States measures changes in lake and stream chemistry in response to changes in emissions and
 22 atmospheric deposition. Together, this information allows policymakers to accurately assess the impact of
 23 Title IV and other air quality policies and to determine if the environmental goals are being achieved.
 24 Recent results of the ARP are presented in Chapter 2. Additionally, researchers continue to study the
 25 impacts of emission reductions on lakes, streams, forests, and coastal ecosystems (Chapter 3).

26 1.7.2.1 Wet Deposition Monitoring

27 The National Atmospheric Deposition Program/National Trends Network (NADP/NTN) is a nationwide
 28 network of predominantly rural precipitation monitoring stations. Operating since 1978, the network
 29 collects weekly wet acid deposition data to determine geographic patterns and temporal long-term trends.
 30 NADP/NTN is responsible for measuring the wet deposition component of total pollution loads across the
 31 United States. The network is a collaborative effort between many different organizations and consists of
 32 250 monitoring stations spanning the continental United States, Alaska, Canada, Hawaii, Puerto Rico, and

1 the Virgin Islands. Quality assured data are available from the NADP Web site at
2 <http://nadp.sws.uiuc.edu/>.

3 **1.7.2.2 Air Quality and Dry Deposition Monitoring**

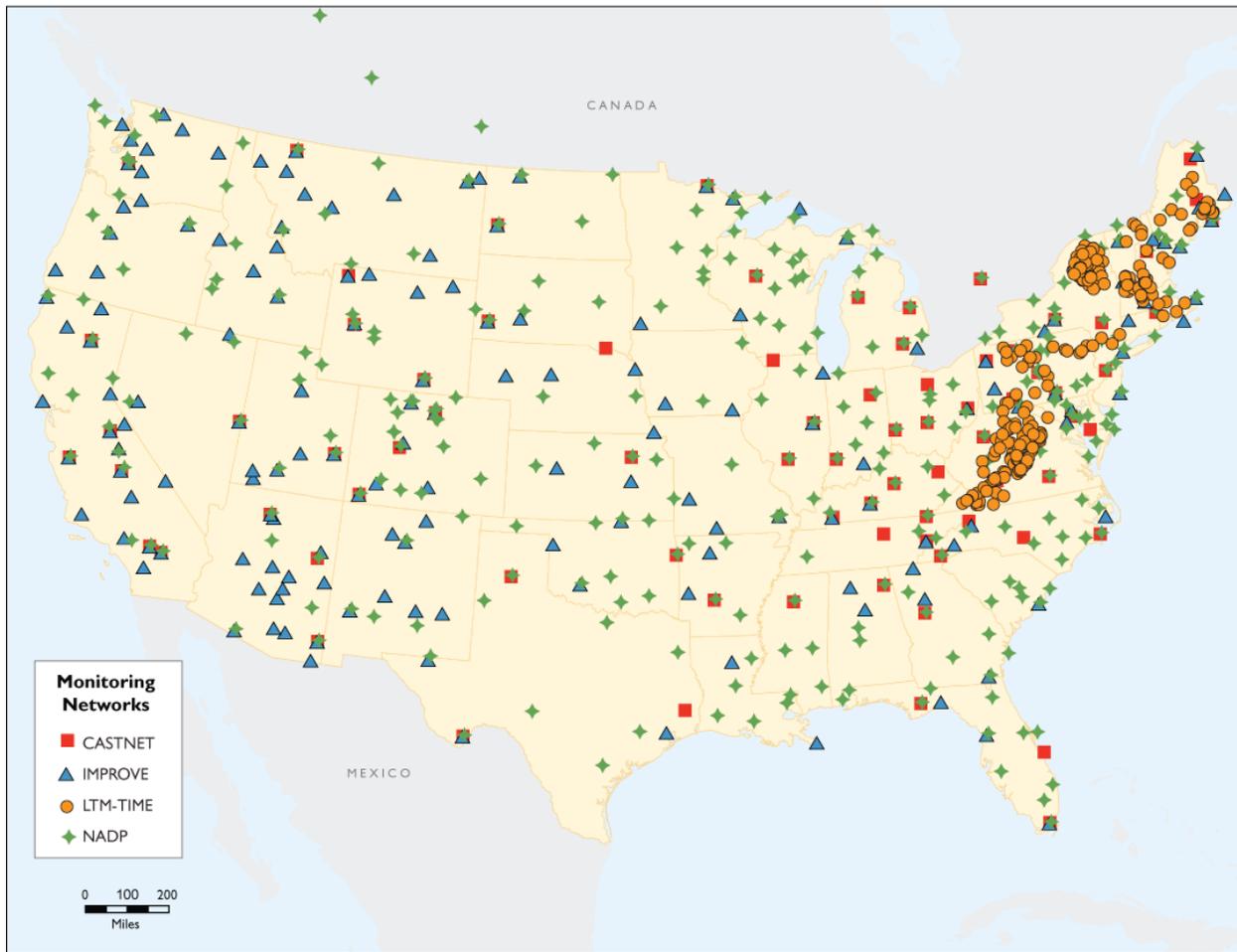
4 The Clean Air Status and Trends Network (CASTNET) is a regional, long-term environmental
5 monitoring program administered and operated by EPA and the National Park Service (NPS). Developed
6 from the existing National Dry Deposition Network (NDDN), CASTNET was established in 1991 under
7 the CAAA. The regional monitoring network was formed to assess trends in acidic deposition due to
8 emission-reduction programs, such as the ARP. CASTNET has since become the nation's primary
9 monitoring network for measuring concentrations of air pollutants that form the dry component of acidic
10 deposition and affect regional ecosystems and rural ambient ozone levels. CASTNET is able to provide
11 the data needed to assess and report on geographic patterns and long-term temporal trends in ambient air
12 pollution and dry atmospheric deposition. CASTNET can also be used to track changes in measurements
13 associated with climate change (e.g., temperature, precipitation). Presently, a total of 86 operational
14 CASTNET sites are located in or near rural areas and sensitive ecosystems and collect data on ambient
15 levels of pollutants where urban influences are minimal. As part of an interagency agreement, the NPS
16 sponsors 27 of the CASTNET sites, which are located in national parks and other Class-I areas designated
17 as deserving special protection from air pollution. Quality assured data are available at
18 <http://www.epa.gov/CASTNET/data.html>.

19 **1.7.2.3 Visibility Monitoring**

20 The Interagency Monitoring of Protected Visual Environments (IMPROVE) is a long-term monitoring
21 network that measures current visibility conditions, tracks changes in visibility, and determines the causes
22 of visibility impairment in national parks and wilderness areas. IMPROVE was established in 1985 to aid
23 the development of federal and state implementation plans to protect visibility in Class I areas as
24 stipulated in the 1977 amendments to the CAA. IMPROVE began collecting data in 1988 at 20 Class I
25 areas. The network expanded to monitor the impacts of the Regional Haze Rule and now consists of 167
26 sites nationwide. IMPROVE is a collaborative monitoring effort. Data and additional information about
27 IMPROVE are available at <http://vista.cira.colostate.edu/improve/>.

28 **1.7.2.4 Ecological Monitoring – Lakes and Streams**

29 The Temporally Integrated Monitoring of Ecosystems (TIME) and the Long-Term Monitoring (LTM)
30 programs are complementary monitoring networks that provide information on a variety of indicators
31 necessary for tracking temporal and spatial trends in environmental response to changes in regional air
32 quality and acid deposition in ecosystems sensitive to acid rain in the eastern United States. TIME was
33 developed as a special study within EPA's Environmental Monitoring and Assessment Program (EMAP)
34 to track trends in acid-relevant chemistry of particular classes of acid-sensitive lakes in the Northeast and
35 streams in the central Appalachians. Because TIME sites were selected through a rigorous statistical
36 sampling effort, measurements from these sites are used to extrapolate from a small number of regionally-
37 representative sampling sites to a much larger number of lakes and streams. In contrast, the primary
38 objective of LTM is to detect long-term trends in the acid-base status of sensitive lakes and streams
39 across a gradient of acidic deposition. The LTM network consists of a subset of lakes and streams that are
40 particularly sensitive to acidity, with most site records extending back to the early 1980s. TIME and LTM
41 monitoring sites are located in New England, the Adirondack Mountains, the Northern Appalachian
42 Plateau, and the central Appalachians. Data are used to characterize how the most sensitive of aquatic
43 systems in each region are responding to changing deposition and to provide information on seasonal
44 chemistry and episodic acidification.



1
2

Figure 1-6. Air quality, deposition, and air quality monitoring networks (Prepared by U.S. EPA).

2. Results of the Acid Rain Program: Status and Trends of Emissions, Deposition, Air Quality, Surface Water, and Visibility, 1990 to 2008

Since its inception in 1995, the ARP has made significant progress in reducing emissions. In fact, 2007 and 2008 emissions were below the 2010 SO₂ cap set at 8.95 million tons, a level of about one-half of the emissions from EGUs in 1980. These emission reductions have led to important environmental benefits, including improvements in air quality, reductions in acid deposition, the beginnings of recovery from acidification in freshwater lakes and streams, and improvements in visibility.

2.1 Emissions

2.1.1 SO₂ Emissions

As shown in **Figure 2-1**, SO₂ emissions in the United States declined between 1980 and 2008 (see data available at <http://www.epa.gov/ttn/chieftrends>). In 2008, ARP sources had reduced annual SO₂ emissions by 56%, compared with 1980 levels, and 52%, compared with 1990 levels. ARP sources emitted 7.6 million tons of SO₂ in 2008, which was well below the 2008 annual emission cap of 9.5 million tons and already below the annual cap of 8.95 million tons established by Title IV as the level for full implementation of the ARP in 2010. In addition, national SO₂ emissions from all sources (including those not covered by the ARP) also have fallen by approximately 56%, from nearly 26 million tons in 1980 to about 11.4 million tons in 2008. The declines in SO₂ emissions from all sources in the United States likely result from a combination of several factors, including the following:

The SO₂ requirements under the ARP apply to EGUs, fossil fuel-fired combustors that serve a generator that provides electricity for sale. The vast majority of ARP SO₂ emissions result from coal-fired EGUs, although the program also applies to oil and gas units.

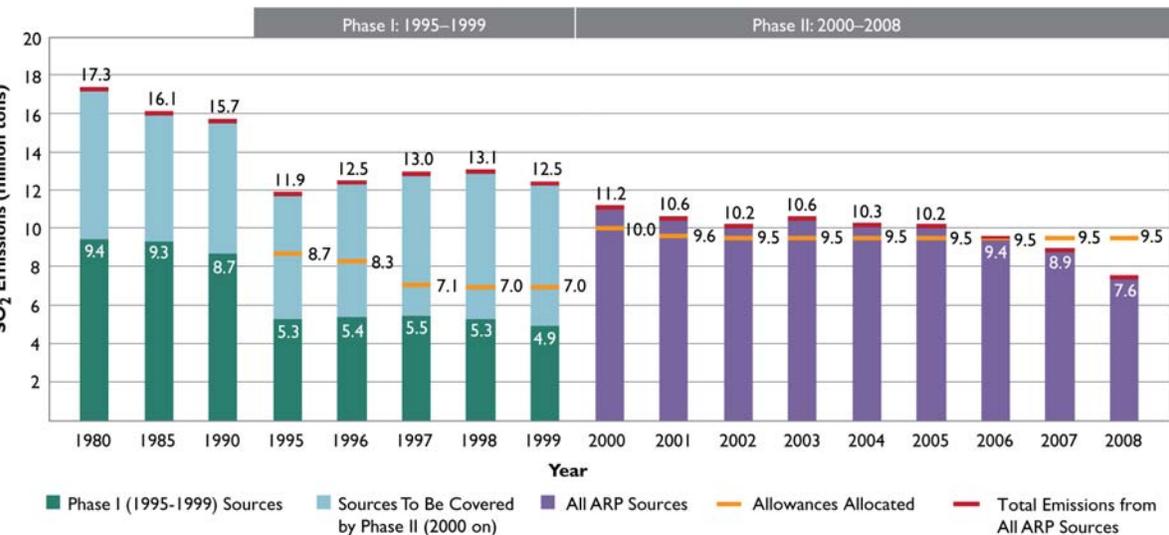
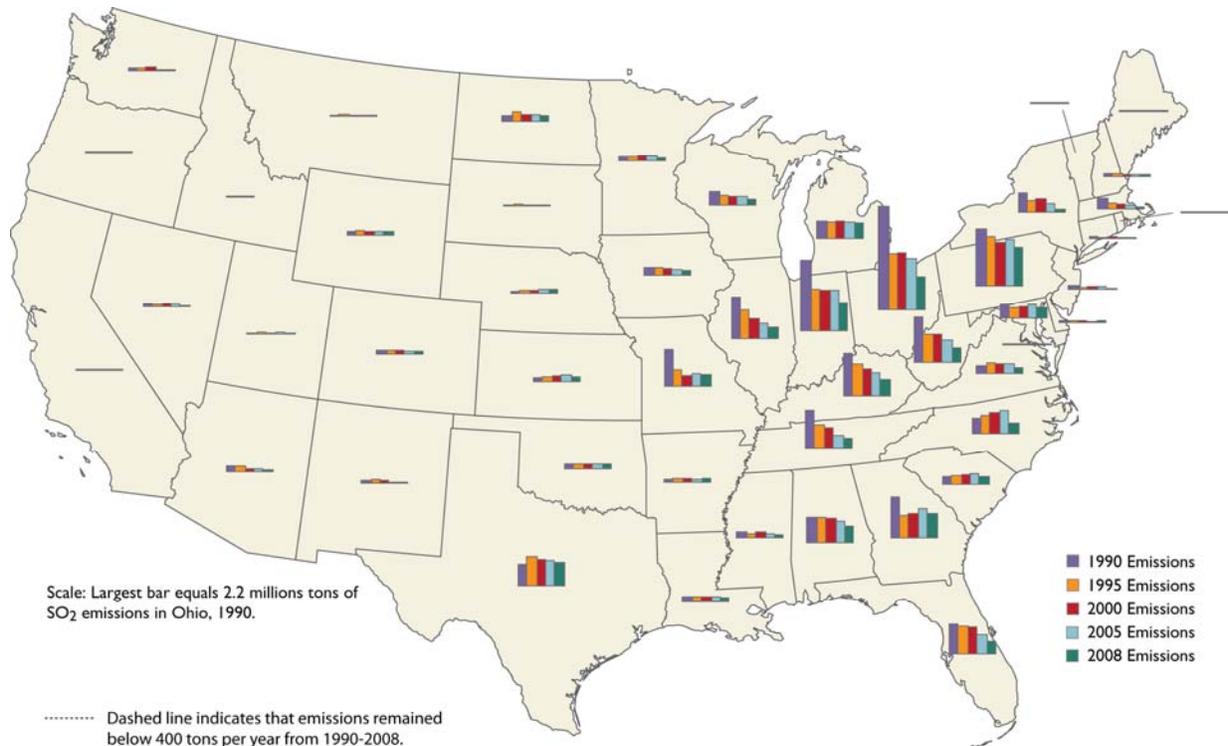


Figure 2-1. SO₂ emissions from ARP sources, 1980 to 2008 (U.S. EPA, 2009a).

1 The states with the highest-emitting sources in 1990 (i.e., Illinois, Indiana, Kentucky, Missouri, Ohio,
 2 Pennsylvania, Tennessee, and West Virginia) have generally seen the greatest SO₂ reductions under the
 3 ARP (see **Figure 2-2**). Most of these states are upwind of the areas the ARP was designed to protect, and
 4 reductions have resulted in important environmental and health benefits over a large region.



5 **Figure 2-2. Changes in SO₂ emission levels by state from 1990 to 2008 for ARP sources**
 6 **(U.S. EPA, 2009a).**
 7

8 From 1990 to 2008, annual SO₂ emissions in 38 states and the District of Columbia fell by a total of
 9 approximately 8.2 million tons. In contrast, annual SO₂ emissions increased by a total of 79,309 tons in
 10 10 states from 1990 to 2008. The 7 states with the greatest reductions in annual emissions since 1990
 11 include Ohio, which decreased emissions by more than 1.5 million tons, and Illinois, Indiana, Kentucky,
 12 Missouri, Tennessee, and West Virginia, each of which reduced total emissions during this time period by
 13 more than 500,000 tons. To view emission data in an interactive format using Google Earth or a similar
 14 three-dimensional platform, go to <http://www.epa.gov/airmarkets/progress/interactivemapping.html>.

15 2.1.2 NO_x Emissions

16 NO_x emissions from all ARP sources were 3.0 million tons in 2008, and emissions have decreased 3.7
 17 million tons since 1990 (see **Figure 2-3**). The goal of the ARP NO_x program is to limit NO_x emissions
 18 from the affected coal-fired boilers so that their emissions are at least 2 million tons less than the
 19 projected level for the year 2000 without implementation of Title IV. The 2008 emission level of 3.0
 20 million tons is 5.1 million tons less than the projected level in 2000 without the ARP, or more than double
 21 the Title IV NO_x emission-reduction objective. While the ARP was responsible for a large portion of
 22 these annual NO_x reductions, other programs—such as the NO_x Budget Trading Program (NBP) under
 23 EPA’s NO_x State Implementation Plan (SIP) Call and other regional and state NO_x emission-control
 24 programs—also contributed significantly to the NO_x reductions.

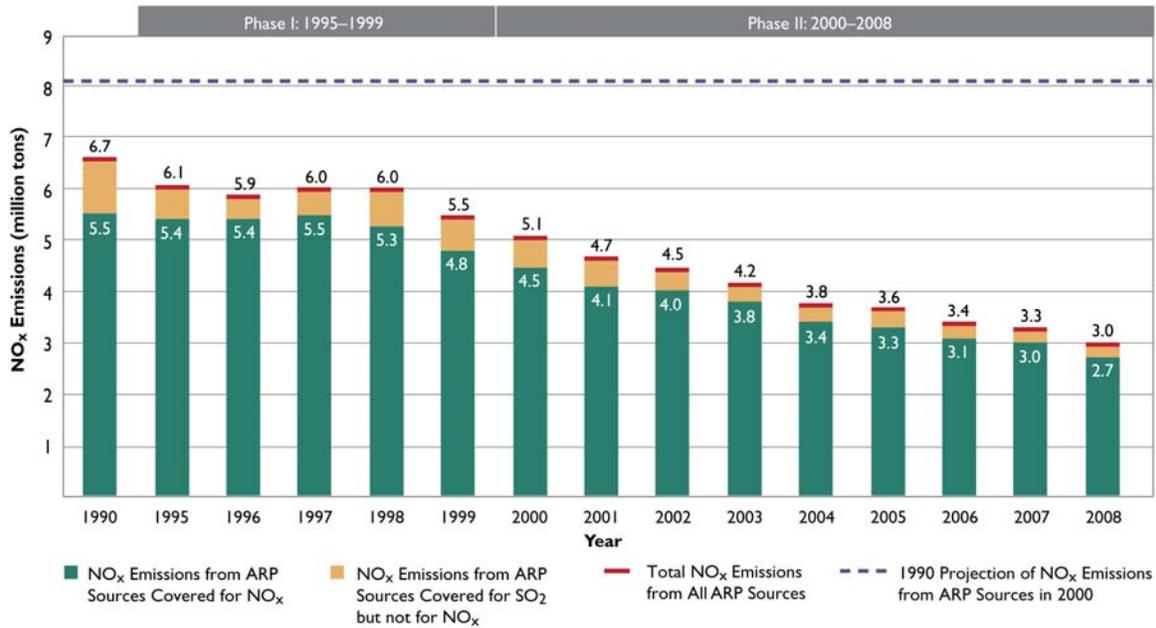


Figure 2-3. NO_x emission levels for all ARP sources, 1990 to 2008 (U.S. EPA, 2009a).

From 1990 to 2008, annual NO_x emissions from ARP sources dropped by about 3.7 million tons, a net decrease of 55%. During this period, 42 states and the District of Columbia reduced NO_x emissions, while 6 other states accounted for only about 15,600 tons of increased NO_x emissions (see **Figure 2-4**).

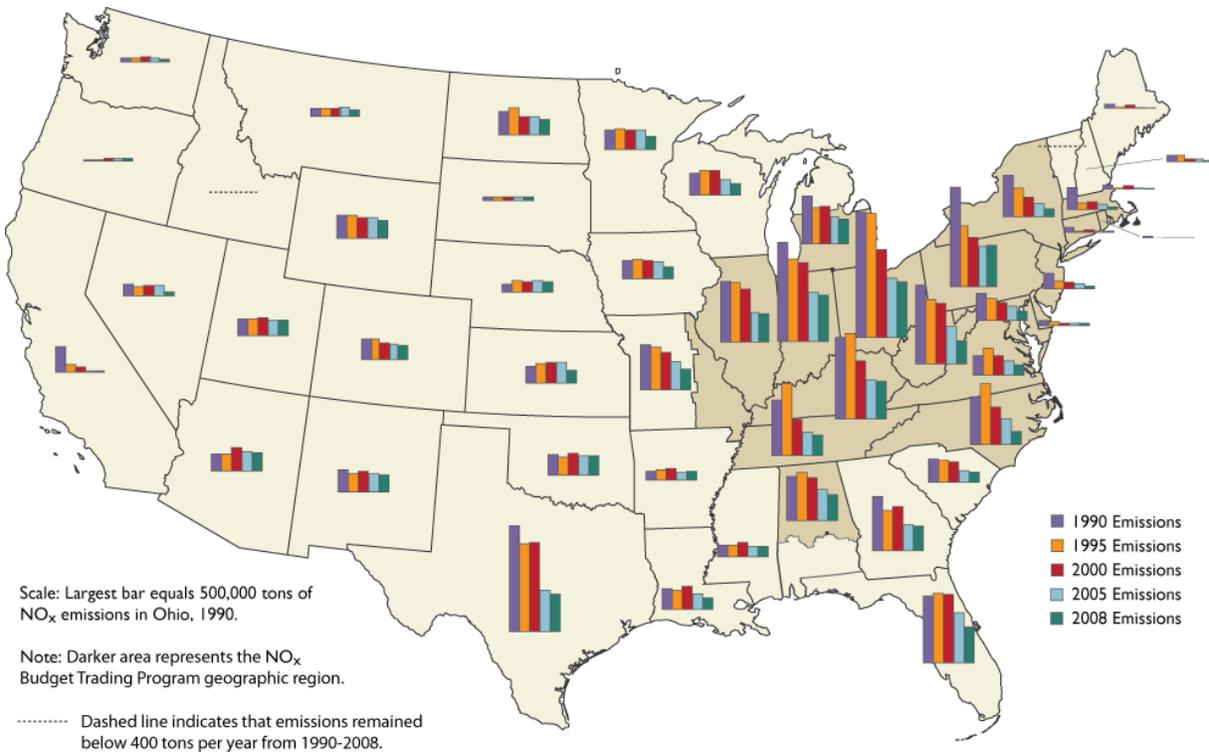


Figure 2-4. Changes in NO_x emission levels by state from 1990 to 2008 for ARP sources (U.S. EPA, 2009a).

1 The states subject to EPA's 1998 NO_x SIP Call have achieved significant reductions in ozone-season NO_x
 2 emissions since the baseline years of 1990 and 2000. All of these states have achieved reductions since
 3 1990 as a result of programs implemented under the 1990 CAAA, with many states reducing their
 4 emissions by more than half since 1990. A significant portion of these decreases in NO_x emissions has
 5 been achieved since 2000, largely as a result of decreases under ozone-season NO_x trading programs
 6 implemented by the Ozone Transport Commission (OTC) from 1999 to 2002, and under the NO_x SIP Call
 7 from 2003 to 2008. For reports about these programs, go to
 8 <http://www.epa.gov/airmarkets/progress/progress-reports.html>.

A variety of other programs have contributed to NO_x emission reductions:

Ozone Transport Commission (OTC)—Established under the 1990 CAAA, this organization consists of states primarily located in the Northeast and mid-Atlantic region and developed the OTC NO_x Budget Program, which operated from 1995–2002. As part of this program, 11 states and the District of Columbia entered into a memorandum of understanding to achieve regional emission reductions of NO_x through the use of control technologies and an ozone-season cap-and-trade program.

NO_x SIP Call—Issued in 1998 to reduce the regional transport of ground-level ozone, the NO_x SIP Call required states to reduce ozone-season NO_x emissions by meeting emission budgets.

NO_x Budget Trading Program (NBP)—This market-based cap-and-trade program was developed under the NO_x SIP Call and replaced the OTC NO_x Budget Program in 2003. The NBP was created to reduce NO_x emissions from power plants and other large combustion sources in the eastern United States.

Clear Air Interstate Rule (CAIR)—Promulgated in 2005, CAIR was designed to reduce emissions of SO₂ and NO_x and replaced the NBP in 2009. This rule created three separate trading programs: an annual NO_x program, an ozone-season NO_x program, and an annual SO₂ program. In 2008, CAIR was remanded, but remains in place while EPA develops a new rule (U.S. EPA, 2009f).

9

10 2.2 Air Quality

11 Emission reductions achieved under the ARP have led to improvements in air quality, with significant
 12 benefits to human health. Since the early 1990s, improvements in ambient SO₂, sulfate, and nitrate
 13 concentrations have varied regionally in the eastern United States (see **Table 2-1**).

14 **Table 2-1. Regional Changes in Air Quality from the 1989 to 1991**
 15 **and 2006 to 2008 Observation Periods**

Region	Average Concentration (µg/m ³), 1989–1991 ^a	Average Concentration (µg/m ³), 2006–2008 ^a	Percentage Change ^b	Number of Sites
Ambient SO₂				
Northeast ^c	5.5	2.1	–62	3
Mid-Atlantic	13	6	–54	12
Southeast	5.3	2.9	–45	9
Midwest	11	5	–55	10

Region	Average Concentration ($\mu\text{g}/\text{m}^3$), 1989–1991 ^a	Average Concentration ($\mu\text{g}/\text{m}^3$), 2006–2008 ^a	Percentage Change ^b	Number of Sites
Ambient Sulfate				
Northeast ^c	3.5	2	–43	3
Mid-Atlantic	6.4	4	–38	12
Southeast	5.3	3.8	–28	9
Midwest	5.9	3.3	–44	10
Total Ambient Nitrate				
Northeast ^c	1.7	1	–41	3
Mid-Atlantic	3.2	2.2	–31	12
Southeast	2.2	1.7	–23	9
Midwest	4.6	3.3	–28	10

^a Averages are the arithmetic mean of all sites in a region that were present and met the completeness criteria in both averaging periods. Thus, the average concentrations may differ from past reports.

^b All values are statistically significant at the 95% confidence level unless otherwise indicated.

^c Percentage change in this region was not tested for statistical significance because too few monitoring sites are available.

Source: U.S. EPA, 2009c

2.2.1 SO₂ and Sulfate

What is the difference between SO₂, sulfate, and total sulfur deposition?

Sulfur is an element that exists in several different forms. SO₂ and sulfate are the forms of sulfur examined in this report.

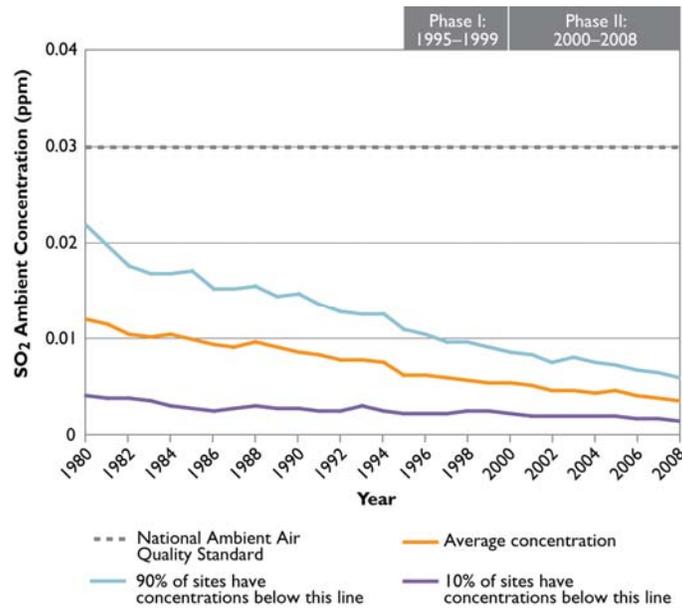
SO₂—When sulfur-containing substances, such as coal, are burned, the sulfur is primarily converted to SO₂. Emissions from ARP sources and ambient air quality are discussed in terms of SO₂ concentrations.

Sulfate—SO₂ is oxidized in the atmosphere to form sulfate (SO₄²⁻). Acid produced during the oxidation process is a major contributor to wet acidifying deposition. In this report, ambient air quality and levels of wet deposition are discussed in terms of sulfate concentrations. Also, sulfate concentrations in water are an indicator (along with base cation and nitrate concentrations) of lake and stream acidification where there are no easily weathered soil or bedrock sources (e.g., gypsum).

Total Sulfur—Total sulfur represents the sum of all sulfur species. In this report, total deposition is discussed in terms of the levels of sulfur deposited, representing the sum of wet and dry deposition.

Data collected from monitoring networks show that the decline in SO₂ emissions from the power industry has improved air quality. Based on EPA's latest air emission trends data (see <http://www.epa.gov/airtrends/index.html>), the national composite average of SO₂ annual mean ambient concentrations decreased 71% between 1980 and 2008, as shown in **Figure 2-5** (based on state, local, and EPA monitoring sites located primarily in urban areas). Although **Figure 2-5** shows a steady declining trend from 1980–2008, the largest single-year reduction (20%) occurred in the first year of the ARP,

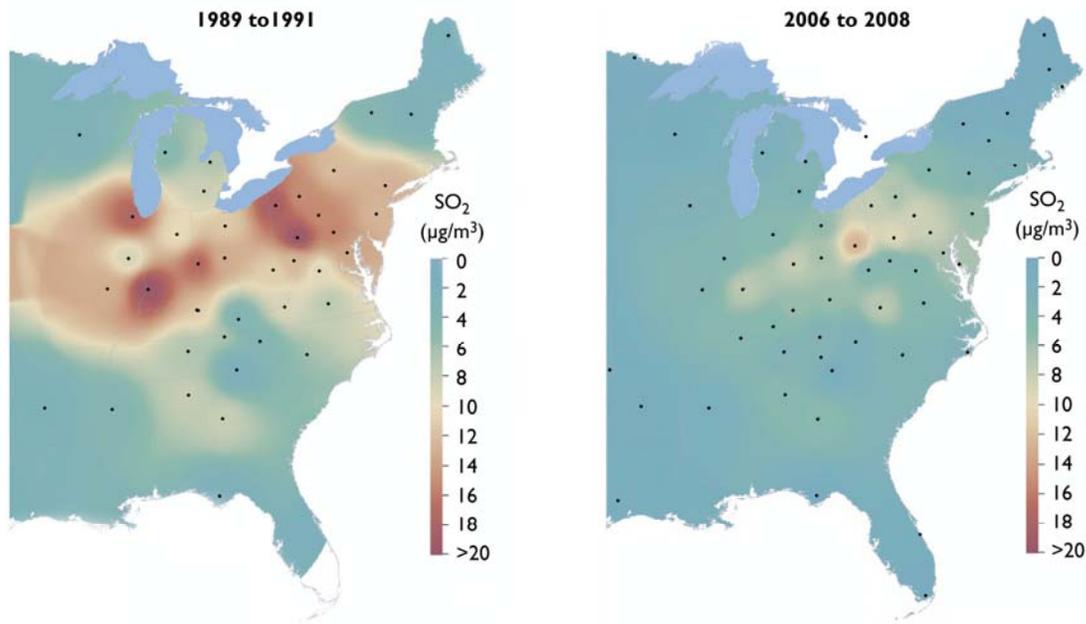
1 between 1994 and 1995. These trends are consistent with the regional ambient air quality trends observed
 2 in CASTNET.



3
 4

Figure 2-5. National SO₂ air quality, 1980 to 2008 (U.S. EPA, 2009c).

5 During the late 1990s, dramatic regional improvements in SO₂ and ambient sulfate concentrations were
 6 observed at CASTNET sites throughout the eastern United States following implementation of Phase I of
 7 the ARP. These improvements continue today. Analyses of regional monitoring data from CASTNET
 8 show the geographic pattern of SO₂ and airborne sulfate in the eastern United States. Three-year mean
 9 annual concentrations of SO₂ and sulfate from CASTNET long-term monitoring sites in the eastern
 10 United States are compared for the 1989 to 1991 and 2006 to 2008 observation periods (see **Figures 2-6**
 11 **and 2-7**, below). For the 1989 to 1991 observation period, few data on the ambient concentrations of SO₂
 12 and sulfate exist from CASTNET sites in the western United States; therefore, changes in ambient
 13 concentrations in this region could not be assessed.



Note: For maps depicting these trends for the entire conterminous United States, visit <http://www.epa.gov/castnet>. Dots on all maps represent monitoring sites. Lack of shading for southern Florida on the map on the left indicates lack of monitoring coverage in the 1989 to 1991 time period.

Figure 2-6. Annual mean ambient SO₂ concentrations in the eastern United States for the 1989 to 1991 and 2006 to 2008 observation periods (U.S. EPA, 2009c).

Figure 2-6 shows that from 1989 to 1991, prior to implementation of Phase I of the ARP, the highest annual ambient concentrations of SO₂ in the eastern United States were observed in western Pennsylvania and along the Ohio River Valley. In comparison, the map for the 2006 to 2008 observation period indicates a significant decline in ambient SO₂ concentrations in nearly all affected areas after implementation of the ARP and other programs.

Like SO₂ concentrations, ambient sulfate concentrations have decreased since the ARP was implemented, with average values decreasing by 28% to 44% throughout the eastern United States. During the 1989 to 1991 observation period, the highest annual ambient sulfate concentrations were observed in western Pennsylvania, along the Ohio River Valley, and in northern Alabama at levels greater than 11 micrograms per cubic meter (µg/m³). Most of the eastern United States experienced annual ambient sulfate concentrations greater than 5 µg/m³. Since the ARP was implemented, both the magnitude and spatial extent of the highest ambient sulfate concentrations have dramatically declined, with the largest decreases observed along the Ohio River Valley (see Figure 2-7).

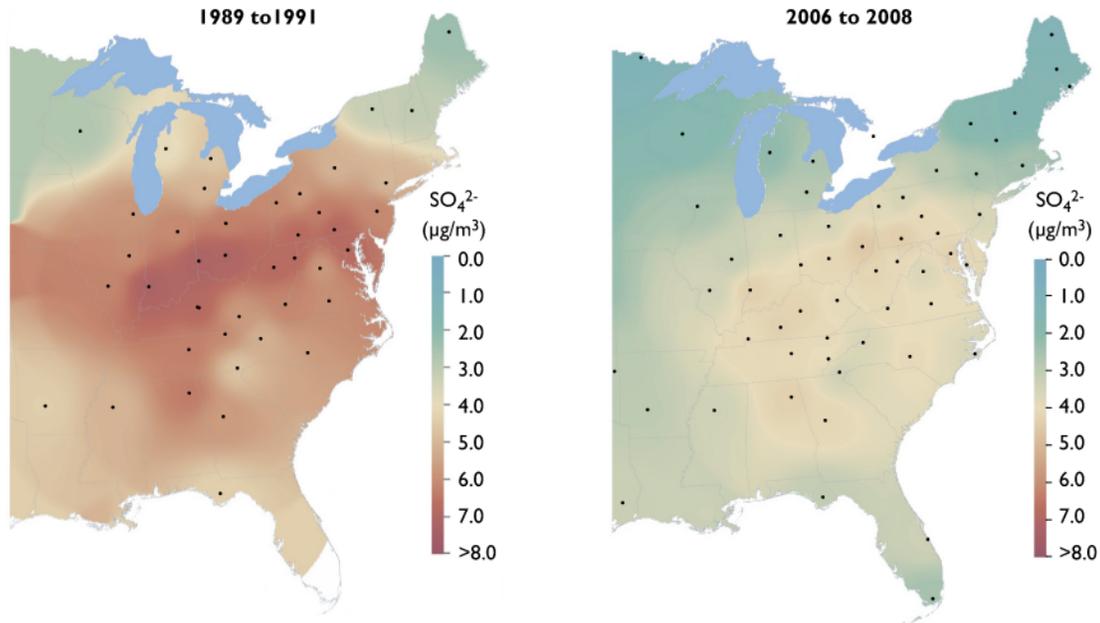


Figure 2-7. Annual mean ambient sulfate concentrations in the eastern United States for the 1989 to 1991 and 2006 to 2008 observation periods (U.S. EPA. 2009c).

2.2.2 NO_x and Nitrate

What is the difference between NO_x, nitrate, and total nitrogen deposition?

Nitrogen is an element and exists in several different forms that are of interest in this report, including NO_x and nitrate.

NO_x—During combustion, nitrogen in fuel (e.g., coal) and the atmosphere combines with oxygen at high temperatures and pressure to form NO_x. Emissions from ARP sources are discussed in terms of NO_x concentrations.

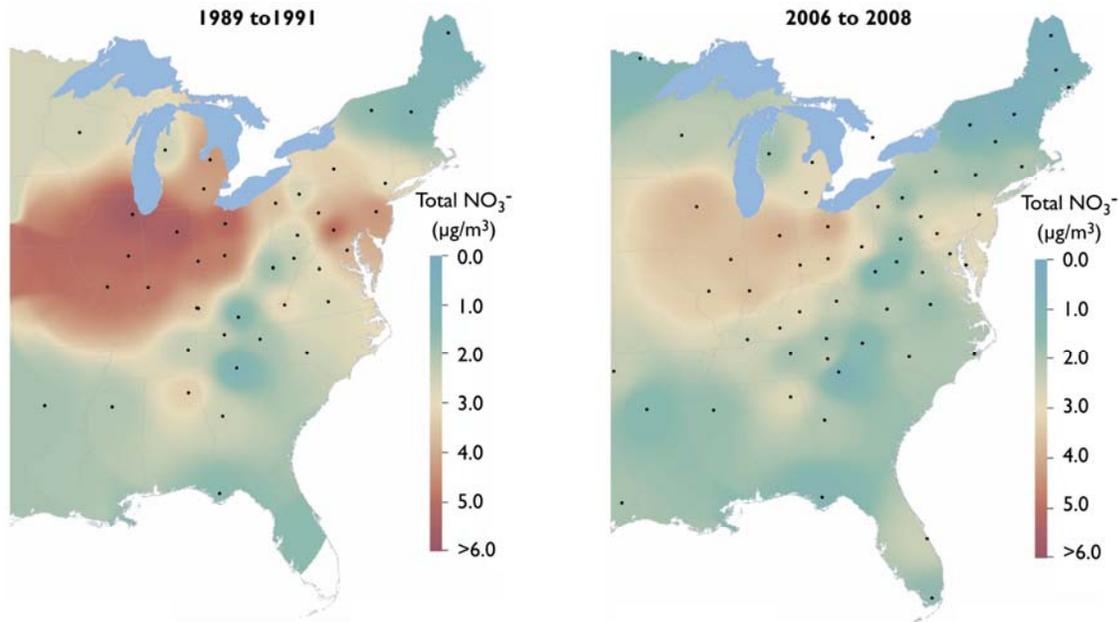
Nitrate—NO_x is oxidized in the atmosphere to form nitrate (NO₃⁻). Similar to SO₂, acid produced during the oxidation process is a contributor to acidifying deposition. In this report, ambient air quality and wet deposition are discussed in terms of nitrate concentrations. Total ambient nitrate concentrations reported here consist of the sum of nitric acid, ionic nitrates, and particulate nitrates. Also, nitrate concentration in water is an indicator (along with base cation and sulfate concentrations) of lake and stream acidification.

Total Nitrogen—Total nitrogen represents the sum of all nitrogen species. In this report, total nitrogen deposition is discussed in terms of the levels of nitrogen deposited, representing the sum of wet and dry deposition.

Although the ARP has met its NO_x emission-reduction targets, emissions from other sources (e.g., mobile sources) contribute to ambient nitrate concentrations in many areas. NO_x levels can also be affected by emissions transported via air currents over wide regions.

From 2006 to 2008, reductions in NO_x emissions during the ozone season from power plants under the NO_x SIP Call have continued to result in significant region-specific improvements in ambient total nitrate (i.e., nitrate [NO₃⁻] plus nitric acid [HNO₃]) concentrations. For instance, annual mean ambient total

1 nitrate concentrations for the 2006 to 2008 observation period in the mid-Atlantic region were 31% less
 2 than the annual mean concentration in the 1989 to 1991 period (see **Figure 2-8**). Although these
 3 improvements might be partly attributed to added NO_x controls installed for compliance with the NO_x SIP
 4 Call, this has not been studied to date.



Note: For maps depicting these trends for the entire conterminous United States, visit <http://www.epa.gov/castnet>.
 Dots on all maps represent monitoring sites. Lack of shading for southern Florida on the map on the left indicates lack of monitoring coverage in the 1989 to 1991 time period.

5
 6 **Figure 2-8. Annual mean ambient total nitrate concentrations in the eastern United States for the**
 7 **1989 to 1991 and 2006 to 2008 observation periods (U.S. EPA, 2009c).**

8 **2.3 Acid Deposition**

9 NADP/NTN monitoring data show significant improvements
 10 across the eastern United States in the primary acid
 11 deposition indicators—sulfur and nitrogen. The sulfur
 12 indicator is assessed using the atmospheric deposition levels
 13 of sulfate (wet deposition) and sulfur (dry and total
 14 deposition), as well as the concentration of sulfate in
 15 precipitation. The nitrogen indicator is measured using levels
 16 of wet inorganic nitrogen deposition (combined deposition of
 17 inorganic nitrate and ammonium ions in wet deposition) and
 18 total inorganic nitrogen deposition (combined deposition of
 19 inorganic nitrate and ammonium deposition in precipitation, dry particulate deposition, and gaseous nitric
 20 acid). **Table 2-2** provides an overview of changes in atmospheric deposition in regions of the eastern
 21 United States from the 1989 to 1991 and 2006 to 2008 observation periods.

Atmospheric deposition occurs as wet deposition, which falls to the earth through rain, snow, and fog, and as dry deposition, which falls to the earth as gas and particles in the absence of precipitation or fog. Total atmospheric deposition is the sum of wet and dry deposition.

1
2

Table 2-2. Regional Changes in Atmospheric Deposition from the 1989 to 1991 and 2006 to 2008 Observation Periods

Region	Average Deposition (kg/ha), 1989–1991	Average Deposition (kg/ha), 2006–2008	Percent Change ^a	Number of Sites
Wet Sulfate				
Northeast	7.5	5	-33	17
Mid-Atlantic	9.2	6.3	-32	11
Southeast	6.1	3.9	-36	23
Midwest	7.1	4.5	-37	27
Dry Sulfur				
Northeast ^b	4	1.5	-63	2
Mid-Atlantic	6.3	3.3	-48	8
Southeast ^b	1.2	0.8	-33	2
Midwest	7	3.4	-51	9
Total Sulfur^c				
Northeast ^a	11	6	-45	2
Mid-Atlantic	16	10	-38	8
Southeast ^a	8	5.3	-34	2
Midwest	16	9	-44	9
Wet Inorganic Nitrogen				
Northeast	5.6	4.4	-21	17
Mid-Atlantic	6.2	4.9	-21	11
Southeast	4.4	3.5	-20	23
Midwest	5.8	5.2	-10	27
Dry Inorganic Nitrogen				
Northeast ^b	1.8	0.9	-50	2
Mid-Atlantic	2.4	1.6	-33	9
Southeast ^b	0.88	0.96	9	2
Midwest	2.7	2	-26	9
Total Inorganic Nitrogen^c				
Northeast ^b	6.6	4.8	-27	2
Mid-Atlantic	8.5	6.4	-25	9
Southeast ^b	5.9	4.9	-17	2
Midwest	9.3	7.5	-19	9

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^a All values are statistically significant at the 95% confidence level unless otherwise indicated.

^b Percentage change in this region was not tested for statistical significance because too few monitoring sites are available.

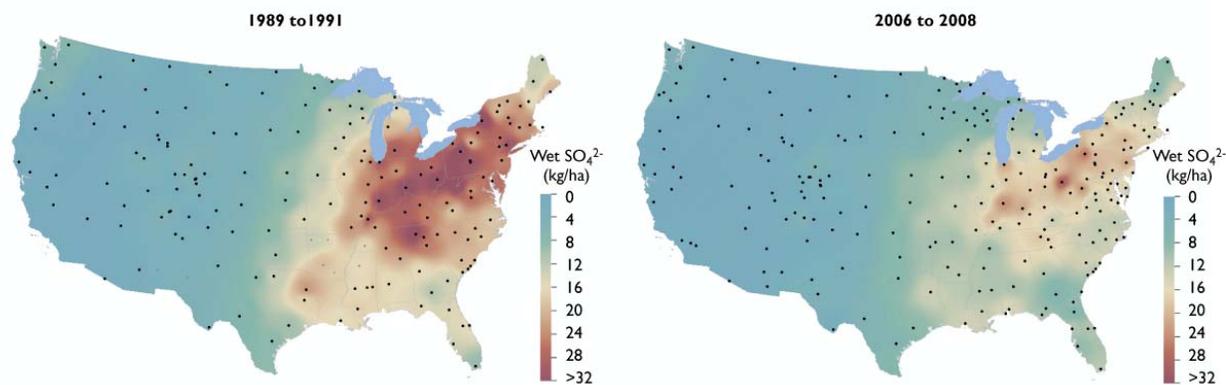
^c Total deposition is estimated from raw measurement data, not rounded, and may not equal the sum of dry and wet deposition.

Source: U.S. EPA, 2009c

1 One recent scientific advancement pertinent to obtaining more accurate estimates of atmospheric
 2 deposition is the increasing use of statistical and empirical models supported by detailed spatial
 3 measurements that more accurately incorporate the enhancement of precipitation by orographic lift (i.e.,
 4 where moist air is forced to a higher elevation by mountainous or other rising terrain, causing fog and/or
 5 precipitation) and other factors (Nanus et al., 2003; Grimm and Lynch, 2004; Weathers et al., 2006).
 6 Using such approaches to obtain better estimates of atmospheric deposition is important because most
 7 deposition monitoring sites tend to be located at the lowest elevations within sensitive ecosystems and,
 8 therefore, tend to underestimate deposition relative to a model that incorporates terrain and mountain
 9 effects into deposition estimates. For example, Weathers et al. (2006) estimated that total nitrogen and
 10 sulfur deposition levels for the Acadia National Park in Maine and the Great Smoky Mountain National
 11 Park in Tennessee and North Carolina were about 70% greater than indicated by data collected from
 12 nearby wet and dry monitoring stations. These modeling results highlight that atmospheric deposition to
 13 sensitive high-elevation ecosystems is generally greater than indicated by data from most precipitation
 14 sampling sites, which tend to be in easily accessible locations at lower elevations.

15 2.3.1 Sulfur Deposition

16 Between the 1989 to 1991 and 2006 to 2008 observation periods, decreases in wet deposition of sulfate
 17 averaged more than 30% for the eastern United States. Some of the greatest decreases have occurred in
 18 the mid-Appalachian region, including Maryland, New York, West Virginia, Virginia, and most of
 19 Pennsylvania (see **Figure 2-9**). Other, less dramatic reductions have been observed across much of New
 20 England, portions of the southern Appalachian Mountains, and some areas of the Midwest. A principal
 21 reason for decreased sulfate deposition levels in the Northeast is a reduction in the long-range transport of
 22 sulfate from emission sources located in the Ohio River Valley. The decreases in sulfate documented in
 23 the Northeast, particularly across New England and portions of New York, were also affected by SO₂
 24 emission reductions in eastern Canada.



25 Note: Dots on all maps represent NADP monitoring sites. Data source: NADP

26 **Figure 2-9. Annual mean wet sulfate deposition in the United States**
 27 **for the 1989 to 1991 and 2006 to 2008 observation periods (U.S. EPA, 2009c).**

28 Portions of the eastern United States have also experienced significant decreases in dry and total sulfur
 29 deposition since the initiation of the ARP. Dry sulfur deposition levels in the mid-Atlantic region and the
 30 Midwest have decreased by 48% and 51%, respectively, between the 1989 to 1991 and 2006 to 2008
 31 observation periods. Decreases in total sulfur deposition for the same areas were 38% and 44%,
 32 respectively, for the same period. Continuous data records for dry and total sulfur deposition are available
 33 from only a few sites in the Northeast and Southeast; therefore, the observed decreases in deposition may
 34 not be representative of these regions.

1 Other studies reported in the scientific literature indicate decreases in sulfate concentrations in
2 precipitation since the 1990s; these decreases are similar to those reported here. Historical data show that
3 decreases in sulfate concentrations in precipitation are approaching 50% since the 1980s for many
4 monitoring sites in the eastern United States (Lehmann et al., 2005; Kvale and Pryor, 2006). Between the
5 1989 to 1991 and 2006 to 2008 observation periods, sulfate concentrations in precipitation have decreased
6 by approximately 30% throughout the eastern United States. A strong correlation between large-scale SO₂
7 emission reductions and large decreases in sulfate concentrations in precipitation has been noted in the
8 Northeast and mid-Atlantic regions (Lehmann et al., 2005), which are among the areas most affected by
9 acid deposition.

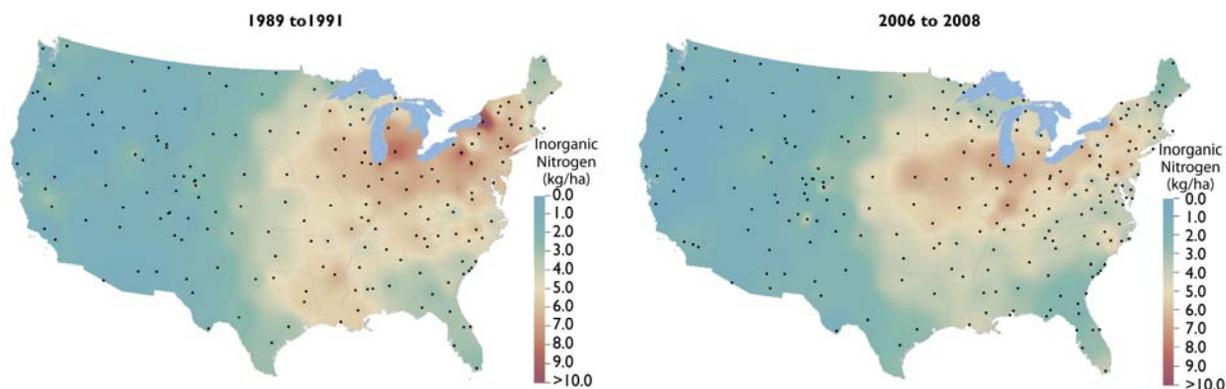
10 **2.3.2 Nitrogen Deposition (NO₃⁻ and Ammonium [NH₄⁺])**

11 Analyses of data from existing networks, such as NADP/NTN and CASTNET, indicate that nitrogen
12 deposition has decreased since the early 1990s (see **Figure 2-10**); however, this decline was more
13 significant after 2000. Decreases in nitrogen deposition in the United States have been less pronounced,
14 show greater spatial variation in temporal trends, and are less certain than those of sulfur deposition for a
15 variety of reasons, including the following:

- 16 ■ NO_x emission declines have been less than the declines in SO₂ emissions
- 17 ■ The proportion of nationwide NO_x emissions that originates from EGUs is much lower (about
18 20%) than the proportion of SO₂ emissions that originates from these same units (about 70%)
19 (U.S. EPA, 2009d)
- 20 ■ NO_x emissions originate from a number of non-ARP sources, including motor vehicles
- 21 ■ The contribution of ammonia (NH₃) emissions to overall nitrogen deposition originates largely
22 from agricultural sources and is not regulated by the CAA (Stephen and Aneja, 2008)
- 23 ■ A large number of chemical species of nitrogen contribute to total nitrogen deposition, but are not
24 well measured by existing monitoring networks (Sparks et al., 2008).

25 Nitrogen deposition levels across the United States
26 vary by region and by the type of deposition.
27 Regional differences in these trends are apparent.
28 The mid-Atlantic and northeastern states generally
29 show the greatest downward trends in nitrogen
30 deposition, and parts of the Southeast and West
31 show the smallest decreases (Lehmann et al.,
32 2005). In some regions (e.g., the Front Range of
33 Colorado, Iowa), nitrogen deposition has been
34 increasing in recent years (Burns, 2003; Ingersoll
35 et al., 2008). Between the 1989 to 1991 and 2006
36 to 2008 observation periods, decreases in wet
37 inorganic nitrogen (nitrate and ammonium)
38 deposition levels in the eastern United States
39 ranged from 10% in the Midwest to 21% in the
40 mid-Atlantic region and the Northeast. Decreases
41 in dry and total inorganic nitrogen deposition
42 generally have been greater than those of wet deposition, with a 19% and 25% decrease in total nitrogen
43 deposition for the Midwest and the mid-Atlantic region, respectively.

Historically, scientists have focused their studies of nitrogen deposition on the wet deposition of nitrate. Scientists recently expanded their focus to other facets of nitrogen deposition and increasingly assess nitrogen sources and deposition in terms of total reactive nitrogen. Total reactive nitrogen considers all biologically, chemically, and radiatively active nitrogen compounds in both wet and dry deposition, such as ammonia gas (NH₃), ammonium ion (NH₄⁺), nitric oxide (NO), nitrite (NO₂⁻), nitric acid (HNO₃), nitrous oxide (N₂O), nitrate (NO₃⁻), and organic compounds (e.g., urea, amines, nucleic acids). Sources of reactive nitrogen include agricultural practices, vehicle exhaust, and EGUs.

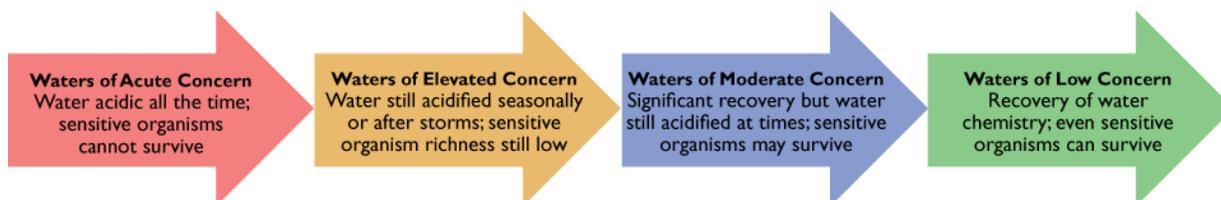


Note: Dots on all maps represent NADP monitoring sites. Data source: NADP

Figure 2-10. Annual mean wet inorganic nitrogen deposition in the United States (U.S. EPA, 2009c).

2.4 Surface Water Monitoring

Acid rain, resulting from SO₂ and NO_x emissions, is one of many large-scale anthropogenic effects that negatively impact the health of biota in lakes and streams in the United States. Surface water chemistry provides direct indicators of the potential effects of acidic deposition on the overall health of aquatic ecosystems. Since the implementation of the ARP, scientists have measured changes in some lakes and streams in the eastern United States and found signs of recovery in many, but not all, regions and waterbodies. **Figure 2-11** provides an overview of the stages of recovery in acidic lakes and streams used in this report. As described in Chapter 3 of this report, organisms may survive in recovered lakes and streams, but may differ from the species that originally inhabited those waterbodies.



Note: Waters of acute concern are defined as having ANC < 0 µeq/L.
 Waters of elevated concern are defined as having ANC between 0-50 µeq/L.
 Waters of moderate concern are defined as having ANC between 50 and 100 µeq/L.
 Waters of low concern are defined as having ANC >100 µeq/L.

Figure 2-11. Stages of recovery for acidic lakes and streams (Prepared by U.S. EPA).

Three indicators of acidity in surface waters are used to provide information regarding both sensitivity to surface water acidification and the level of current and past acidification. These indicators are concentrations of sulfate (SO₄²⁻) and nitrate (NO₃⁻) ions, and acid neutralizing capacity (ANC). Sulfate and nitrate are negatively charged ions with the potential to acidify waters and leach cations, including acidic aluminum cations, from watershed soils. Aluminum and H⁺ cations are known to be toxic to aquatic life, given high enough concentrations. Base cations, the by-products of weathering reactions that neutralize acids in watersheds, may also be indicators of changes in acidification. Assessments of acidic deposition effects, dating from the 1970s to the present, have shown that sulfate is the primary negatively charged ion in most acid-sensitive waters (Driscoll et al., 2001). Sulfate ion concentrations in surface waters provide important information on the extent of base cation (i.e., calcium, magnesium, potassium, sodium) leaching in soils and provide insight on how sulfate concentrations relate to the levels of atmospheric sulfur concentrations and atmospheric sulfur deposition. Nitrogen is an important nutrient for

1 plant growth; therefore, nitrogen inputs through deposition are incorporated into forest biomass and soil
 2 organic matter, with only a fraction of the input leaching as nitrate into lakes and streams. However, as
 3 atmospheric nitrogen deposition increases to levels in excess of ecosystem needs, there is greater potential
 4 for increased leaching of nitrate into lakes and streams, leading to increased acidification. ANC is a
 5 measure of the acid-buffering capacity of water and an important indicator of the sensitivity and the
 6 degree of surface water acidification or recovery that occurs over time. Acidification results in a
 7 diminished ability of water in a lake or stream to neutralize strong acids that enter aquatic ecosystems.

Long-term surface water monitoring networks provide information on the chemistry of lakes and streams and on how waterbodies are responding to changes in emissions. Two EPA-administered monitoring programs provide information on the effects of acid rain on lakes and streams: the Temporally Integrated Monitoring of Ecosystems (TIME) program and the Long-Term Monitoring (LTM) program. These programs were designed to track the effect of the 1990 CAAA in reducing the acidity of lakes and streams in four acid-sensitive regions of the eastern United States. The surface water chemistry trend data in the four regions monitored by the TIME and LTM programs are essential for tracking the ecological response to ARP emission reductions. Additional information about these programs is available in Chapter 1.

9 Monitoring the trends in these indicators enables determination of whether conditions in acid-sensitive
 10 lakes and streams are improving and heading toward chemical recovery or if conditions are degrading.
 11 Chemical recovery is a prerequisite for the subsequent recovery of aquatic plants and animals. Movement
 12 toward recovery of a lake
 13 or stream ecosystem is

14 indicated by increases in
 15 ANC levels and decreases
 16 in sulfate and nitrate
 17 concentrations. **Table 2-3**
 18 presents the percentage of
 19 monitoring sites in four
 20 acid-sensitive regions of
 21 the eastern United States
 22 (**Figure 2-12**) with
 23 improving sulfate, nitrate,
 24 and ANC trends. The
 25 sites represent LTM lakes
 26 and streams monitored
 27 from 1990 to 2008. These
 28 regional trends were
 29 calculated using data
 30 from all the sites located
 31 within the region that had
 32 a complete data record
 33 for the time period
 34 considered. Trends are
 35 considered statistically
 36 significant if they exceed
 37 the 95% confidence
 38 interval ($p < 0.05$).

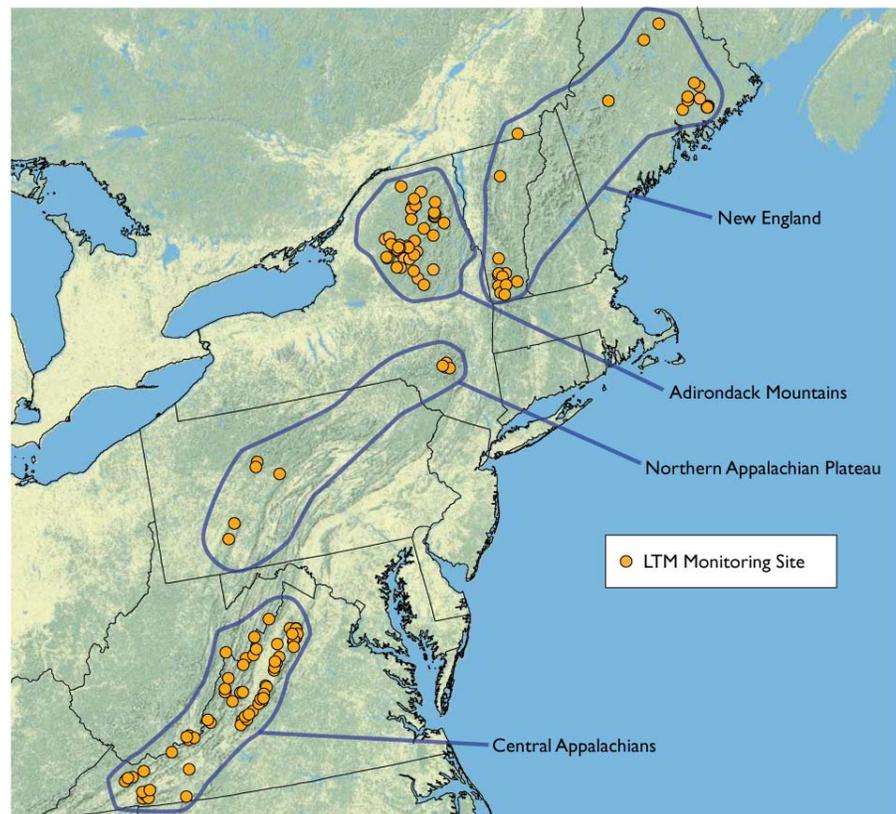


Figure 2-12. LTM site locations and regional groupings
 (Prepared by U.S. EPA).

Table 2-3. Regional Trends in Sulfate, Nitrate, and ANC at LTM Sites from 1990 to 2008

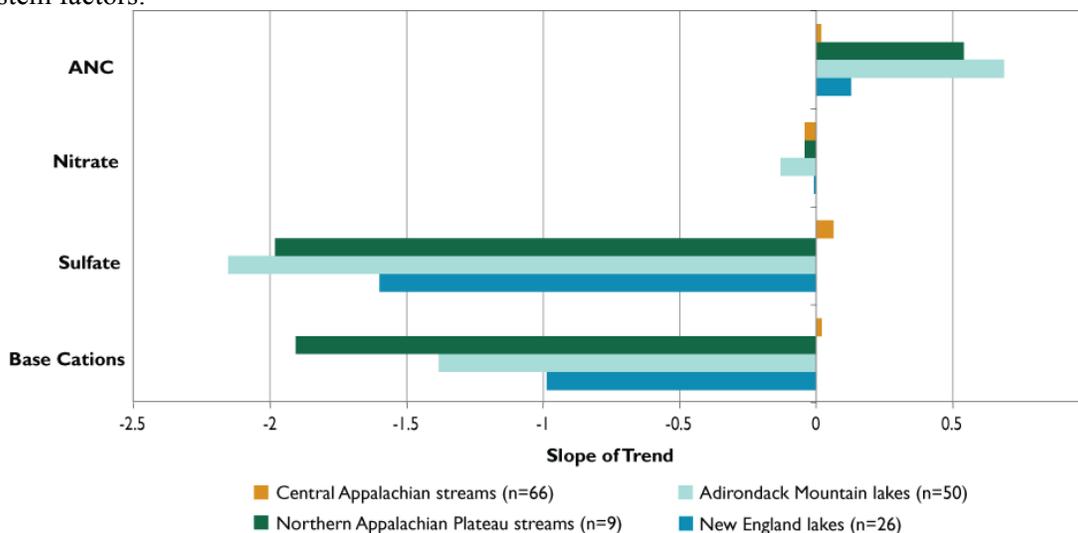
Region	Waterbodies Covered	% of Sites with Improving Sulfate Trend ^a	% of Sites with Improving Nitrate Trend ^a	% of Sites with Improving ANC Trend ^a
Adirondack Mountains	Lakes in NY No = 50	90%	32%	58%
Northern Appalachian Plateau	Streams in NY, PA No = 9	78%	33%	56%
New England	Lakes in ME, VT No = 26	96%	31%	12%
Central Appalachians	Streams in VA No = 66	12%	45%	12%

^a Trends determined by *multivariate Mann-Kendall tests*.

Note: Data source: U.S. EPA.

As seen in **Table 2-3** and **Figure 2-1**, significant improving trends in sulfate concentrations from 1990 to 2008 are found at nearly all monitoring sites in New England, the Adirondack Mountains, and the Northern Appalachian Plateau. However, in the central Appalachians, sulfate concentrations in 21% of individual streams monitored by the LTM program (**Figure 2-12**) show degrading trends (**Figure 2-13**), and improving trends were noted at only 12% of sites. The central Appalachians have highly weathered soils that can store large amounts of deposited sulfate. As long-term sulfate deposition exhausts the soil’s ability to store sulfate, a decreasing proportion of the deposited sulfate is retained in the soil, and an increasing proportion is exported to surface waters. In addition, sulfate stored from decades of sulfur deposition leaches out from the soils over time. Thus, sulfate concentrations in streams in this region are increasing despite reduced levels of sulfate deposition.

Trends in surface water nitrate concentrations are also mixed (**Table 2-3**). Trends at several individual LTM lake and stream sites indicate flat or slightly degrading nitrate trends (**Figure 2-13**). Improving trends for nitrate concentrations were noted at only 31% to 45% of sites in the four regions. These trends do not appear to reflect changes in emissions or deposition in these areas and are likely a result of ecosystem factors.



Note: Base cations were calculated from the sum of calcium, magnesium, and potassium ions.

Slope of trend is the median slope value for all sites in the region.

ANC, nitrate, sulfate, and base cations were measured in µg/L.

Figure 2-13. Trends in acidification of LTM lakes and streams, 1990–2008 (Prepared by U.S. EPA).

Improving trends are increases in ANC levels and decreases in sulfate and nitrate concentrations. Degrading trends show decreases in ANC levels and increases in sulfate and nitrate concentrations.

1
2 Declines in sulfate deposition levels likely result in many of the improving trends for ANC, but ANC
3 levels still lag behind improving sulfate trends at many of the monitoring sites. From 1990 to 2008,
4 monitoring sites that showed a majority of improving ANC trends were in the Adirondack Mountains
5 (58%) and Northern Appalachian Plateau (56%). Streams in the central Appalachians and in New
6 England showed only a few statistically significant trends of improving ANC (12%) (**Table 2-3**). The
7 declining trends of base cations (**Figure 2-13**) in lakes and streams may delay the onset of recovery.
8 Decreasing base cation levels can balance out declining levels of sulfate and nitrate, preventing ANC
9 from increasing. The observed improving trends for ANC can, in part, be attributed to declines in sulfate
10 deposition levels. From 1990 to 2008, many of the statistically significant trends observed at individual
11 LTM sites showed increasing ANC levels (improving trend) in surface waters. Overall, only seven sites
12 indicate a statistically significant degrading trend in ANC.

Lakes and streams with ANC values defined as less than or equal to 0 microequivalents per liter (µeq/L) are acidic and are considered to be of “acute concern.” Lakes and streams having springtime ANC values less than 50 µeq/L are generally considered “sensitive” to acidification and are of “elevated concern.” Lakes and streams with ANC higher than 50 µeq/L are generally considered less sensitive or insensitive to acidification and may be considered of “moderate concern” (i.e., ANC levels between 50 and 100 µeq/L) or “low concern” (i.e., ANC levels greater than 100 µeq/L). When ANC is low, and especially when it is negative, stream water pH is also low (certainly less than 6 and commonly less than 5), and there may be adverse impacts on fish and other animals essential for a healthy aquatic ecosystem (see table below).

Aquatic Ecosystem Concern Levels and Ecological Effects

Category Label	ANC Level (µeq/L)	Expected Ecological Effects
Acute Concern	< 0	Near complete loss of fish populations is expected. Planktonic communities have extremely low diversity and are dominated by acid-tolerant forms. The numbers of individuals in plankton species that are present are greatly reduced.
Elevated Concern	0–50	Fish species richness is greatly reduced (e.g., more than half of expected species are missing). On average, brook trout populations experience sublethal effects, including loss of health and reproduction (fitness). During episodes of high acid deposition, brook trout populations may experience lethal effects. Diversity and distribution of zooplankton communities decline.
Moderate Concern	50–100	Fish species richness begins to decline (e.g., sensitive species are lost from lakes). Brook trout populations are sensitive and variable, with possible sublethal effects. Diversity and distribution of zooplankton communities begin to decline as species that are sensitive to acid deposition are affected.
Low Concern	> 100	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.

13
14 In order to assess the impacts of the implementation of the ARP, the average ANC levels of lakes in the
15 Northeast monitored under the TIME program were evaluated and compared for the 1991 to 1994 and
16 2006 to 2008 time periods (see **Figure 2-14**). Seven and a half percent of lakes in the 1991 to 1994 time
17 frame had 3-year mean ANC levels below 0 microequivalents per liter (µeq/L) and were categorized as
18 being of acute concern (see text box). The percentage of lakes in this category dropped to 4.3% in the
19 2006 to 2008 time frame. Additionally, the percentage of lakes in the elevated concern category dropped
20 from 13.8% for the 1991 to 1994 time period to 10.1 percent from 2006 to 2008, while the percentage of

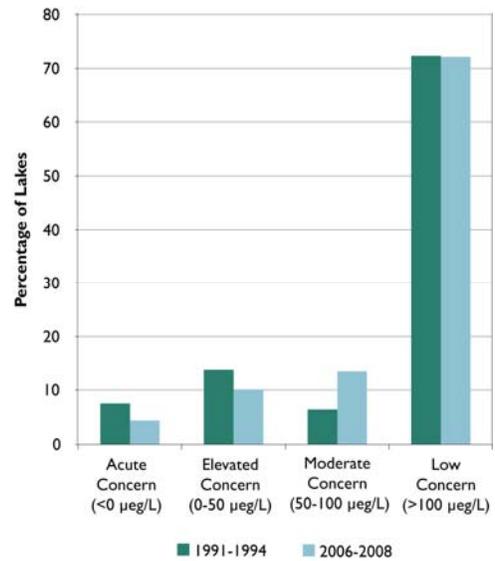
1 lakes in the moderate concern category increased from
 2 6.4% to 13.5%. These results point to a decrease in acidity,
 3 particularly for the subset of lakes with low ANC.

4 **2.4.1 Case Study—Critical Loads for Lakes and**
 5 **Streams**

6 The critical loads approach (see text box) is an assessment
 7 tool that can be used to determine the degree to which air
 8 pollution may be affecting ecological health. This
 9 approach provides a useful lens through which to assess
 10 the results of current policies and programs and to evaluate
 11 the potential value of proposed policy options in terms of
 12 ecosystem protection. The critical loads approach has been
 13 employed routinely as an assessment tool for many years
 14 in the countries of the European Union and Canada. This
 15 case study will explore the extent to which lakes in the
 16 Adirondack Mountains and streams in the central
 17 Appalachians are protected from acidifying nitrogen and
 18 sulfur deposition as a result of Title IV emission
 19 reductions. Further discussion of critical loads can also be
 20 found in Chapter 3 of this report.

21 The critical load for a lake or stream provides a benchmark
 22 against which to assess the extent that a waterbody is
 23 potentially at risk due to current deposition levels. The
 24 analysis focuses on the combined load of sulfur and
 25 nitrogen deposition below which the ANC level would still
 26 support healthy aquatic ecosystems. There are numerous
 27 methods and models that can be used to calculate critical
 28 loads for acidity. Drawing on the peer-reviewed scientific
 29 literature (e.g., Dupont et al., 2005), both case studies use the
 30 Steady-State Water Chemistry (SSWC) model to calculate
 31 the critical load. Critical loads were calculated for 117 lakes
 32 in the Adirondack Mountains and 184 streams in the central
 33 Appalachian region. The modeled lakes and streams in both
 34 case studies are a statistical population of waterbodies that
 35 represents 1,830 lakes of various sizes found throughout the
 36 Adirondack Mountains with surface areas ≥ 4 ha and 70,000
 37 km of streams in the central Appalachians. Both statistical
 38 populations were determined by EMAP.

39 For these particular analyses, the critical load represents the
 40 combined deposition loads of sulfur and nitrogen to which a
 41 lake or stream could be subjected and still have an ANC of
 42 50 $\mu\text{eq/L}$ or higher. While a critical load can be calculated for
 43 any ANC level, 50 $\mu\text{eq/L}$ was chosen because it tends to
 44 protect most fish and other aquatic organisms, although
 45 systems can still become episodically acidic and some
 46 sensitive species may be lost. Critical loads of combined total
 47 sulfur and nitrogen deposition are expressed in terms of ionic
 48 charge balance as milliequivalents per square meter per year



Notes:
 Based on 305 TIME monitoring sites in the Northeast.
 See text box for descriptions of level of concern
 categories.

Figure 2-14. Northeastern Lakes by ANC Category, 1991–1994 vs. 2006–2008 (Prepared by U.S. EPA).

*A **critical load** is a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specific sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt, 1988). If pollutant exposure is less than the critical load, adverse ecological effects (e.g., reduced reproductive success, stunted growth, loss of biological diversity) are not anticipated, and recovery is expected over time if an ecosystem has been damaged by past exposure. A **critical load exceedance** is the measure of pollutant exposure above the critical load. This means pollutant exposure is higher than, or “exceeds,” the critical load and the ecosystem continues to be exposed to damaging levels of pollutants.*

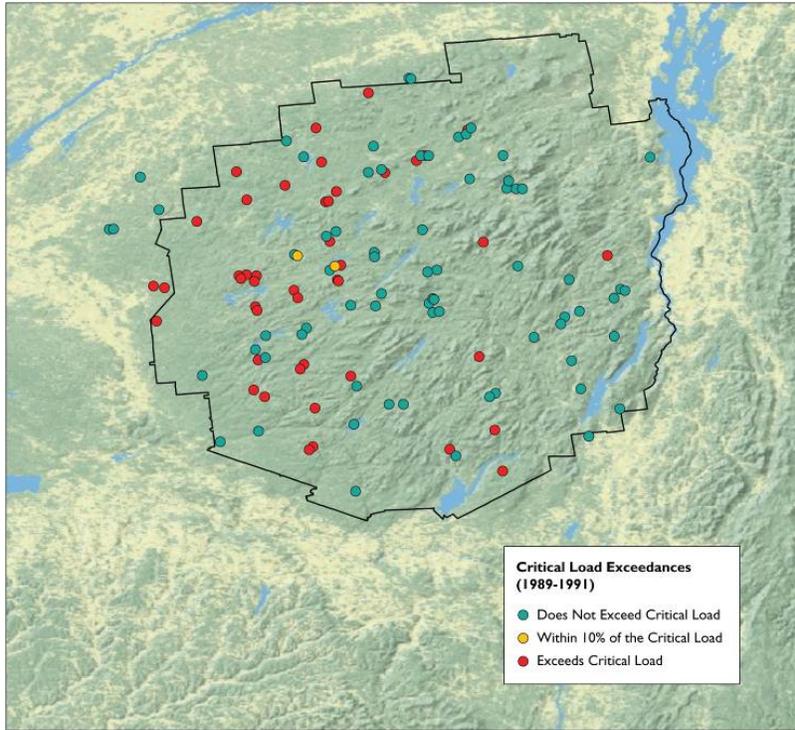
1 (meq/m²/yr). When actual measured deposition of nitrogen and sulfur is greater than the critical load, the
2 critical load is “exceeded,” meaning that combined sulfur and nitrogen deposition was greater than a lake
3 or stream could sustain and still maintain the ANC level of 50 µeq/L or above. In order to assess the
4 extent to which regional lake and stream ecosystems are protected by the emission reductions achieved by
5 Title IV, these case studies compare the amount of deposition systems can receive—the critical load—to
6 measured deposition for the period before implementation of the ARP (1989–1991) and for a recent
7 period after ARP implementation (2006–2008).

8 Overall, these critical load analyses show that emission reductions achieved by the ARP have resulted in
9 improved environmental conditions and increased ecosystem protection in both the Adirondack
10 Mountains and the central Appalachian region. However, the analyses also show that, despite some
11 ecological recovery, lakes and streams in these regions remain at risk due to current acid deposition
12 levels. Thus, further emission reductions are necessary for recovery of these sensitive aquatic systems and
13 ecosystem protection. In Chapter 4 of this report, a modeling analysis examines the degree of ecosystem
14 recovery and protection that would be achieved through further emission reductions.

15 **2.4.1.1 Adirondack Mountain Lakes**

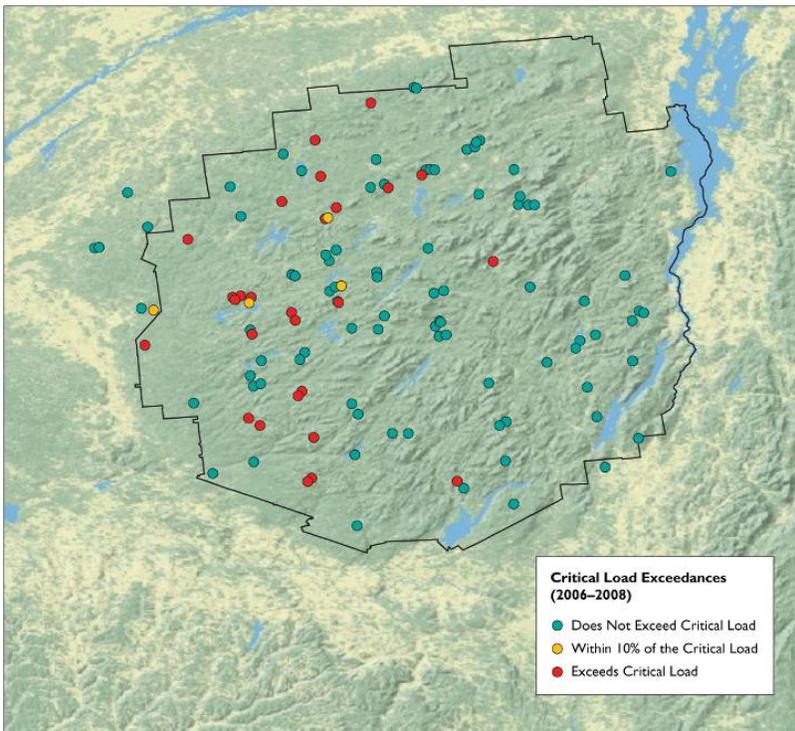
16 The Adirondack Mountains are located in northern New York. Many lakes in this region are sensitive to
17 acidic deposition because of the abundance of shallow soils with low levels of base cations (i.e., calcium,
18 magnesium, sodium, potassium) and the underlying igneous bedrock with low weathering rates and
19 limited ability to buffer acidity (Driscoll et al., 1991; Sullivan et al., 2006a). Decreases in regional sulfur
20 deposition have resulted in decreased sulfate concentrations in Adirondack Mountain lakes, which is an
21 important sign of ecological recovery (see **Figure 2-13**). While inter-annual variability in ANC levels and
22 nitrate concentrations is evident in this region, the overall trend in these lakes is a slight increase in ANC
23 levels and modestly decreasing nitrate concentrations.

24 On average, the calculated critical load of sulfur and nitrogen for lakes in the Adirondack Mountains is
25 162 meq/m²/yr. For the period from 1989 to 1991, before ARP implementation, 45% of the lakes
26 examined received levels of combined sulfur and nitrogen deposition that exceeded the critical load
27 (**Figure 2-15**). For the 2006 to 2008 period, 30% of lakes exceeded the critical load (**Figure 2-16**). Thus,
28 during the 2006 to 2008 period, 15% fewer of the Adirondack Mountain lakes examined were receiving
29 sulfur and nitrogen deposition loads that threaten the health of these ecosystems. Although lakes in the
30 Adirondack Mountains have improved, over 550 lakes still exceed their calculated critical load of sulfur
31 and nitrogen based on the statistically representative population of lakes determined by EMAP.



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Figure 2-15. Critical load exceedances in Adirondack Mountain TIME lakes, 1989 to 1991 (No. Lakes = 117) (Prepared by U.S. EPA).



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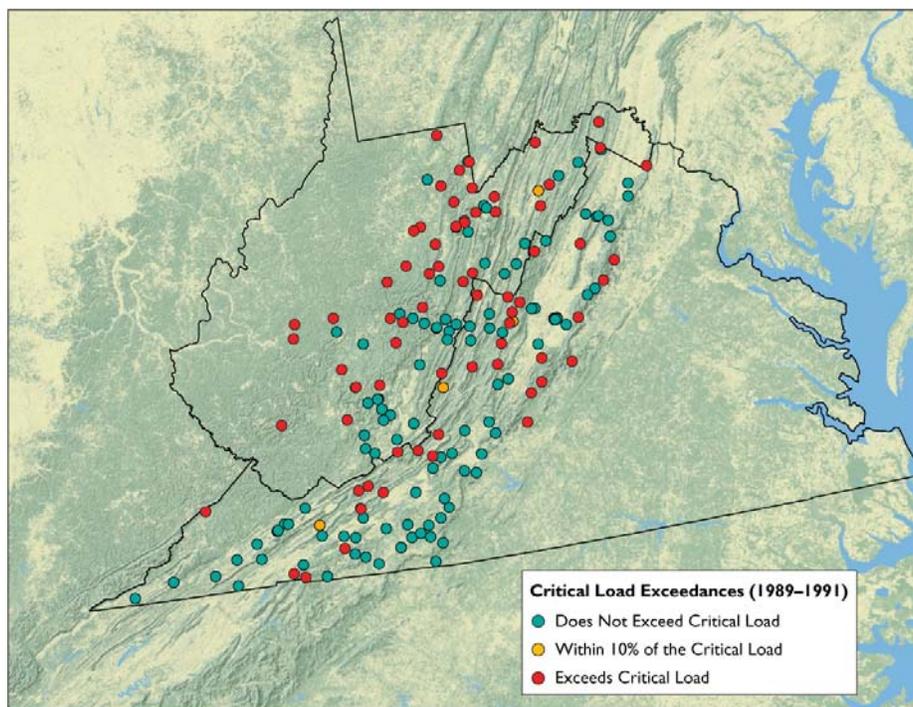
Figure 2-16. Critical load exceedances in Adirondack Mountain TIME lakes, 2006 to 2008 (No. Lakes = 117) (Prepared by U.S. EPA).

1 **2.4.1.2 Central Appalachian Streams**

2 The central Appalachian region of Virginia and West Virginia, including Shenandoah National Park, is
 3 known to be sensitive to acidic deposition. Base-poor soils and low weathering rates of the bedrock that
 4 underlies this region result in streams that are particularly susceptible to acidification. As a result, many
 5 miles of streams in the region have been impacted by acid deposition, with greatly reduced diversity of
 6 aquatic life, including important recreational fish species, such as brook trout. The declines in deposition
 7 of sulfur and nitrogen since the 1990s have resulted in only slight signs of recovery from acid rain in the
 8 streams of this region. **Figure 2-13** shows monitored trends in sulfate and nitrate concentrations and ANC
 9 levels for central Appalachian streams. Sulfate
 10 concentrations in these streams have remained level,
 11 while nitrate concentrations and ANC levels have
 12 improved slightly.

13 On average, the calculated critical load of sulfur and
 14 nitrogen for streams in the central Appalachian
 15 region is 370 meq/m²/yr. For the period before ARP
 16 implementation (1989 to 1991), 41% of streams in
 17 the central Appalachian region received levels of
 18 combined sulfur and nitrogen deposition that exceeded the critical load, and therefore, could not be
 19 adequately neutralized by the environment (**Figure 2-17**). For the period from 2006 to 2008, 31% of the
 20 streams examined continued to receive acid deposition levels that exceeded the critical load
 21 (**Figure 2-18**). Thus, only an additional 10% of streams in the central Appalachian region are protected
 22 from ecological damage from acidic deposition, as compared to the period before ARP implementation.
 23 Although some streams in the central Appalachians likely have improved, approximately 35,000 km of
 24 streams still exceed their calculated critical load of sulfur and nitrogen based on the statistically
 25 representative population of lakes determined by EMAP.

Negative critical loads occur when the supply of base cations is naturally low. In these cases, the lake or stream would have a natural, pre-acidification acid neutralizing capacity (ANC) level below 50 µeq/L, and an ANC of 50 µeq/L or greater would not be achievable regardless of changes in acidic deposition levels.



26
 27 **Figure 2-17. Critical load exceedances in central Appalachian streams, 1989 to 1991**
 28 **(No. Streams = 184) (Prepared by U.S. EPA).**

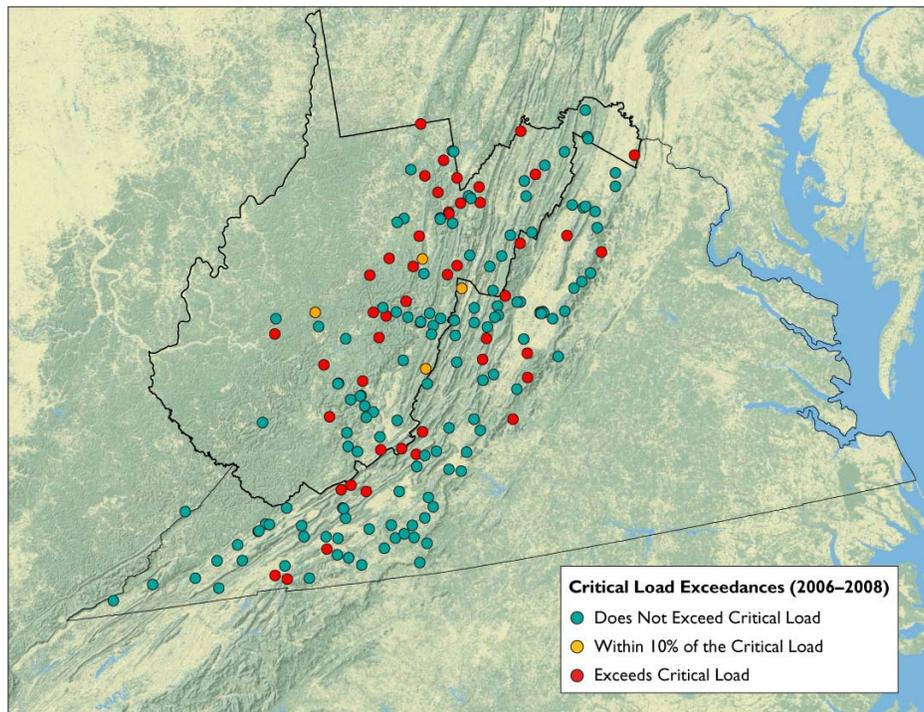


Figure 2-18. Critical load exceedances in central Appalachian streams, 2006 to 2008 (No. Streams = 184) (Prepared by U.S. EPA).

2.5 Visibility

In the atmosphere, SO₂ and NO_x gases are transformed into fine particles of sulfate and nitrate. Sulfate and nitrate particles scatter and absorb light energy, impairing visibility and contributing to regional haze. Visibility refers to the clarity with which distant objects are perceived (U.S. EPA, 2001). Sulfate is generally the dominant contributor to visibility impairment in the eastern United States and a large contributor in the western United States. Nitrate is a significant contributor to visibility impairment in parts of California and in the Midwest during the winter. Changes in visibility conditions are measured in deciview (dv) units, which are approximately proportional to the perceived change in haze levels and increase as visibility decreases. A one dv change is a small but noticeable change under many viewing conditions. Average, naturally occurring conditions (without pollution) are about 7 dv to 8 dv in the eastern United States and 4.5 dv to 5.5 dv in the western United States. For the worst haze days, visibility conditions exceed natural levels by 10 dv to 18 dv in the eastern United States and by about 3 dv to 15 dv in the western United States.

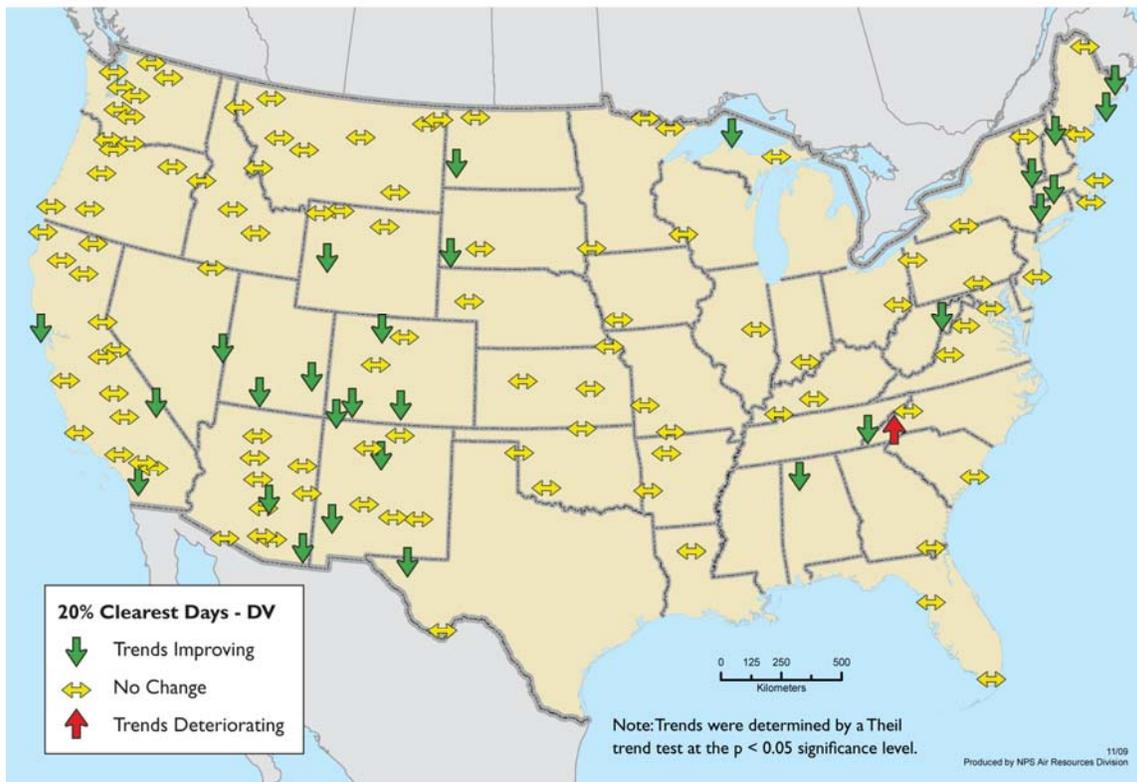
Visibility is measured in “deciviews”, which is a way of characterizing the relationship between air pollution and perception of visibility. The clearest days are those that demonstrate the highest daily visibility (i.e., lowest deciview [dv] values) for a given year. Trends among the clearest days are assessed by comparing the clearest 20% of days every year (i.e., best 20% of visibility).

The haziest days are those that demonstrate the lowest daily visibility (i.e., highest dv values) for a given year. Trends among the haziest days are assessed by comparing the haziest 20% of days every year (i.e., worst 20% of visibility) (U.S. EPA, 2003).

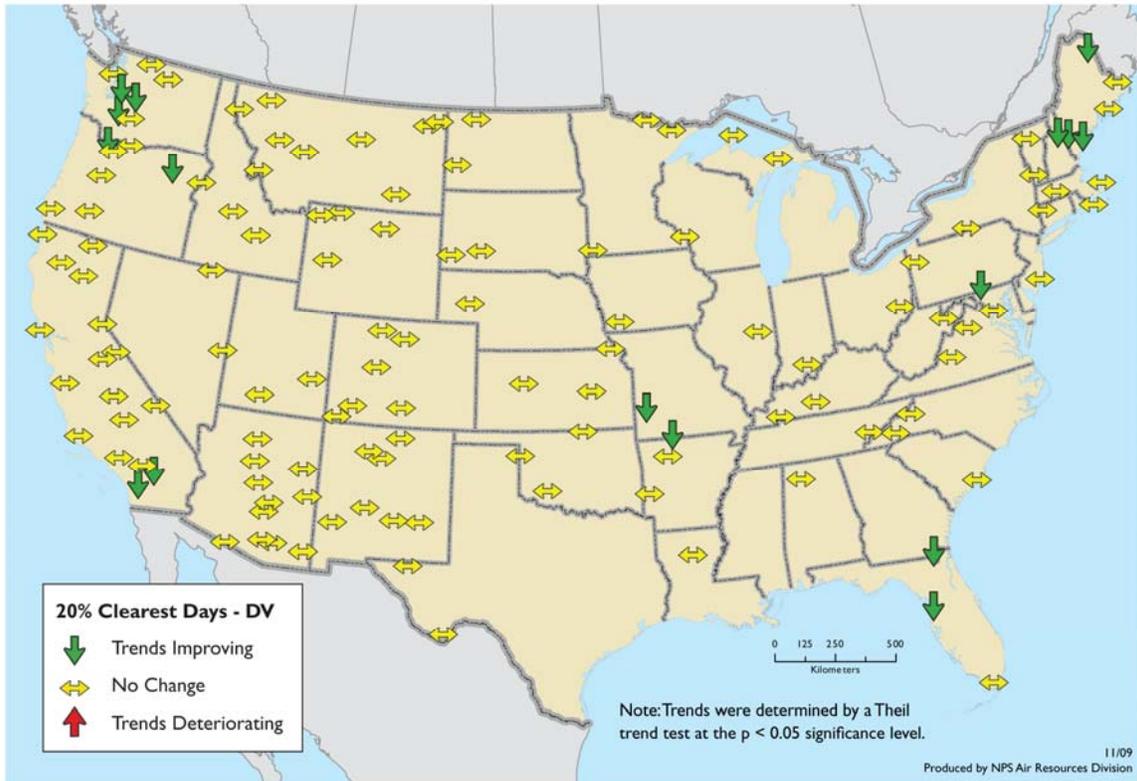
Data from the IMPROVE network, which monitors visibility in and near national parks and wilderness areas, show no statistically significant trends in visibility at most of the 128 sites monitored over the 10-year period from 1999 to 2008 (see **Figures 2-19 and 2-20**). Of the sites with statistically

As dv measurements increase, visibility decreases.

1 significant trends for either the best or worst 20% of visibility conditions, most have improving visibility.
 2 From 1999 through 2008, for the best visibility conditions, 45 sites show significant improvements and no
 3 sites show significant decline; for the worst visibility conditions, 10 sites show significant improvement
 4 and 5 sites show significant decline (all trends are at the $p < 0.05$ significance level). The Northeast has
 5 sites with improving visibility on both best and worst visibility days, principally due to regionally
 6 decreased sulfate particulate concentrations. The rate in improvement for these northeastern sites is about
 7 5 dv for worst days and 3 dv for best days over the 10-year period. In the Pacific Northwest, a significant
 8 improving trend of about 3 dv to 5 dv per decade for worst haze periods is found at monitoring sites near
 9 and/or downwind from Seattle and Portland and may reflect changing impacts of these urban/industrial
 10 areas. A better understanding of the causes of individual site trends requires additional analysis. Trends
 11 for each individual site shown on the maps (Figures 2-19 and 2-20) can be viewed at
 12 <http://views.cira.colostate.edu/web/Trends>.



13
 14 **Figure 2-19. 1999 to 2008 trends for the change in visibility on the clearest days**
 15 **(best 20% of visibility) as measured at 140 IMPROVE monitoring stations (Prepared by NPS).**



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Figure 2-20. 1999 to 2008 trends for the change in visibility on the 20% haziest days (worst 20% of visibility) as measured at 140 IMPROVE monitoring stations (Prepared by NPS).

3. Effects of Acid Deposition on Ecosystems: Advances in the State-of-the Science

3.1 Introduction

Chapter 2 focused on the environmental results of the ARP, presenting data from national monitoring networks on SO₂ and NO_x emissions, air quality, atmospheric deposition, stream water chemistry, and visibility. This chapter expands on this information by examining the most recent research into how ecosystems respond to acid deposition, especially the processes that control the recovery of ecosystems as acid deposition decreases.

In Chapter 2, two general trends were discussed regarding the current recovery status of affected ecosystems: (1) these ecosystems are trending generally towards recovery, but improvements in ecosystem condition shown by surface water chemistry monitoring data thus far have been less than the improvements in deposition; and (2) ecosystem impacts and trends vary widely by geographic region, but the evidence of improvement is strongest and most evident in the Northeast. These trends are not uniform across the United States, however, and in some regions (e.g., central Appalachian Mountain region), trends in improved water quality are generally not evident.

Despite the strong link in many areas between reduced emissions and reduced acidity of atmospheric deposition, the link is less clear between reduced acidity and recovery of the biological communities that live in aquatic and terrestrial ecosystems that have experienced deleterious effects from acid deposition. The recovery of these communities is proceeding at a slower pace than, for example, the improvements in stream and lake ANC would indicate. The goal of this chapter is to synthesize the science in a weight-of-evidence manner to provide policy makers with tangible evidence and likely causative factors regarding ecosystem status and recovery patterns to date. This chapter updates the 2005 NAPAP RTC (NSTC, 2005), with an emphasis on scientific studies and monitoring since 2003, which was the last year for consideration of research results in the 2005 report. Several issues pertinent to ecosystem response to emission controls and acid deposition are receiving increasing attention in the scientific literature and will be discussed in this chapter, including the (1) observed delay in ecosystem recovery in the eastern United States, even with decreases in emissions and deposition over the past 30 years; (2) emerging ecosystem impacts of nitrogen deposition in the western United States, (3) the application of critical deposition loads as a tool for scientists to better inform air quality policies; (4) the role of changes in climate and the carbon cycle as factors that affect the response of ecosystems to acid deposition; and (5) the interaction of multiple pollutants in ecosystems. Throughout this chapter, the value of long-term environmental monitoring data in informing air quality policy will be highlighted, including the limitations of assessing the current status of some ecosystem indicators for which continuous, long-term data are lacking.

3.2 Effects of Atmospheric Deposition on Aquatic and Terrestrial Ecosystems

This section expands on the stream water chemistry section of Chapter 2 and describes the latest studies on the spatial and temporal patterns of recovery in a variety of ecosystems that are sensitive to acid deposition. Factors that control ecosystem sensitivity to deposition are also discussed, with an emphasis on the links between terrestrial and aquatic ecosystems. This section first considers the eastern United States, then the Rocky Mountain region, the far western United States, and finally the coastal areas of the United States. This is a logical division because the effects of atmospheric deposition and issues discussed by scientists and policy makers are somewhat different across these regions and ecosystems.

3.2.1 Ecosystem Recovery in Aquatic and Terrestrial Ecosystems of the Eastern United States

The regions of the United States with an abundance of acid-sensitive waters and ecosystems were well recognized by the end of the 1980s during the decade of intensive research performed under the original NAPAP. These acid-sensitive ecosystems are generally located in upland, mountainous terrain in the eastern United States and are underlain by bedrock that is resistant to weathering, such as granite or quartzite sandstone. The sections below examine trends in surface water chemistry, the impacts of episodic acidification on surface waters, the effects of stored sulfate in the soils of the Southeast, base cation depletion in soils, and how aquatic and terrestrial ecosystems recover from acidifying deposition.

3.2.1.1 Patterns in Surface Water Chemistry and Soils

Monitoring of lakes and stream water chemistry in the eastern United States has shown the continuation of widespread water quality improvements that are consistent with a trend towards recovery in these aquatic ecosystems (Kahl et al., 2004; Driscoll et al., 2007). Decreasing trends in sulfate concentrations have been the sharpest, generally showing decreases of about 2 to 3 $\mu\text{eq/L/yr}$ since the early 1990s in waters in the Northeast and the Northern Appalachian Plateau. These trends are consistent with the large decreases in SO_2 emissions and sulfur deposition and are paralleled by some improvement in pH, ANC, and aluminum (i.e., Al^{+2} and Al^{+3}) concentrations. However, the improvements in pH, ANC, and aluminum concentrations have generally been much smaller than the decreases in sulfate concentrations. Nitrate concentrations in these surface waters have also shown smaller decreases that have been less

persistent and less consistent than those of sulfate concentrations (Kahl et al., 2004; Burns et al., 2005; Driscoll et al., 2007). The most recent trends in surface water chemistry from the EPA LTM/TIME monitoring programs are reported in Chapter 2 and shown in Table 2-3. These trends are generally consistent with those reported in the

studies cited here, indicating sharp decreases in sulfate concentrations, some modest decreases in nitrate concentrations, and increases in ANC in the eastern United States. Streams in the central Appalachian region show the weakest trends in recovery. At least over periods of a decade or more, trends in nitrate concentrations in surface waters are not as clearly linked to trends in nitrogen deposition as trends in sulfate concentrations in surface waters are linked to trends in sulfur deposition (Aber et al., 2002; Burns et al., 2006b; Eshleman et al., 2009). Nonetheless, if temporal trends follow observed spatial patterns of nitrogen deposition and surface water nitrate concentrations over the long term, then decreases in these nitrate concentrations are expected to eventually follow decreases in nitrogen deposition (Aber et al., 2003).

Aluminum (i.e., Al^{+2} and Al^{+3}) levels in surface water are monitored because acidifying deposition releases aluminum from soils and high concentrations of aluminum are toxic to many aquatic biota.

Trends in nitrate concentrations in surface waters are influenced by natural disturbances such as insect defoliation and ice storms, as well as human disturbances such as forest harvesting, and are therefore not as clearly linked to trends in nitrogen deposition. Nonetheless, over the long term, decreases in nitrogen deposition are expected to lead to decreases in nitrate concentrations in surface waters.

Surface water chemistry monitoring programs that document the effects of acid deposition tend to collect samples at regular time intervals, resulting in data that primarily reflect baseflow or non-storm conditions (although there are exceptions). The shortcoming of such a sampling strategy is that surface waters, especially streams and small, shallow lakes, tend to acidify during rainstorms and snowmelt. This phenomenon is termed episodic acidification and has been well documented (Lawrence, 2002; Deviney et al., 2006). Additionally, episodic acidification has been shown to affect surface water biota and to slow ecosystem recovery in waters that otherwise appear to be improving based on data collected at low-flow conditions (Passy et al., 2006; Kowalik et al., 2007). These findings suggest that biological recovery will

1 hinge in part on the acidity of surface waters declining to a level at which minimal episodic acidification
 2 occurs because the survival of acid-sensitive aquatic species is dependent on both the magnitude and
 3 duration of episodic acidification (Baldigo and Murdoch, 1997). Although low-flow ANC values in the
 4 range of 30 to 50 $\mu\text{eq/L}$ have been suggested to provide protection against acidification and biological
 5 harm during high-flow periods (Driscoll et al., 2001; Kahl et al., 2004), other studies have shown episodic
 6 acidification occurs in streams with low-flow ANC values as high as 100 to 150 $\mu\text{eq/L}$ (Lawrence, 2002).
 7 Some evaluations of ecosystem recovery (e.g., Kahl et al., 2004) provide percent improvement values for
 8 various metrics of stream acidity and the extent of acidic waters in a region. These evaluations are based
 9 largely on low-flow conditions or chronic acidification scenarios and do not fully consider episodic
 10 acidification. One study in Maryland evaluated changes in episodic stream acidification from the early
 11 1990s to the late 1990s and found little change in the magnitude of acidification during storms. However,
 12 the study also found a shift towards a diminished role for sulfate in episodic acidification, suggesting a
 13 decreasing contribution of acid deposition to this phenomenon in parallel with declining sulfate
 14 deposition (Kline et al., 2007). Studies by Rice et al. (2006) and Lawrence et al. (2008) compared the
 15 percent of streams or watershed area that is chronically acidified with the percent that is episodically
 16 acidified in the Shenandoah National Park, VA, and the western Adirondack Mountains, NY,
 17 respectively. In each case, the total percentage of acidified streams or watershed area increased
 18 substantially when episodic acidification was considered (see **Table 3-1**).

19 **Table 3-1. Comparison of the Percent of Acidified Streams or Watershed Area Considered**
 20 **Chronically Acidic with Values Obtained when Both Chronic and Episodic**
 21 **Acidification Are Considered**

Location	Percent of streams or watershed area that is chronically acidic	Percent of streams or watershed area that is episodically acidic	Total percent of streams or watershed area that is chronically and episodically acidic	Reference
Shenandoah National Park, VA ^a	8.7	22.5	31.2	Rice et al., 2006
western Adirondack Mountains, NY ^b	35	30	65	Lawrence et al., 2008

^a Chronic acidification is defined as the percent of watershed area with ANC <0 $\mu\text{eq/L}$ for at least 168 successive hours at a 4-year recurrence interval, and episodic acidification is defined as the percent of watershed area with ANC < 20 $\mu\text{eq/L}$ for at least 72 successive hours at a 4-year recurrence interval.

^b Chronic acidification is defined as percent of streams with a base cation surplus (a surrogate for ANC that eliminates the contribution from dissolved organic carbon [DOC]) value < 25 $\mu\text{eq/L}$ during summer, and episodic acidification is defined by the same threshold value during snowmelt conditions.

22 One noteworthy exception to the generalizations above about geographic patterns of improvement in
 23 surface water chemistry is watersheds in the Southeast (e.g., the central Appalachian region of Virginia)
 24 where stream ANC values have generally not improved and sulfate concentrations have shown little
 25 change since the early 1990s (Kahl et al., 2004; Webb et al., 2004; U.S. EPA, 2009e; Chapter 2 of this
 26 report). The lack of chemical recovery in the streams of the central Appalachian region is believed to be
 27 due in part to the old and highly weathered condition of non-glaciated southern soils. These soils contain
 28 an abundance of adsorbed sulfate that can be slowly released as sulfuric acid deposition decreases,
 29 slowing ecosystem recovery (Webb et al., 2004).
 30 In these soils, decades of sulfate release and
 31 decreased deposition levels may be necessary to
 32 shrink the pool of sulfate in the soils to the point
 33 where acidification of streams no longer occurs.

Ecosystem recovery of acidified watersheds in highly weathered soils of the Southeast will likely continue to lag behind that of the Northeast by several years or more likely, decades.

1 Ecosystem recovery of acidified watersheds in the
 2 highly weathered soils of the Southeast will likely
 3 continue to lag behind that of the glaciated Northeast by
 4 several years to several decades.

5 Recent research has shown that decreases in base
 6 cations (calcium, magnesium, potassium, sodium) in
 7 soils may also be slowing the recovery of aquatic and
 8 terrestrial ecosystems even though they are receiving
 9 reduced loads of acid deposition (Bailey et al., 2005;
 10 Sullivan et al., 2006b; Johnson et al., 2008; Warby et
 11 al., 2009). The base cations calcium, magnesium, and
 12 potassium are important nutrients for the growth and
 13 health of forest vegetation and are readily accessed
 14 from the pool of cations held on the surfaces of soil
 15 particles. These base cations can be displaced from the
 16 surfaces of soil particles by hydrogen and aluminum
 17 ions and leached into local waters, effectively removing
 18 these nutrients from soils and lowering the levels that
 19 are available to the plants growing in the soils. Acid
 20 deposition accelerates this as part of the acidification
 21 process, which initially leads to increases in base cation
 22 concentrations in surface waters, but eventually
 23 acidifies the waters as well. Base cation depletion refers
 24 to this accelerated loss of these cations from soils.
 25 Depletion of calcium is of particular concern because
 26 this cation is in high demand by sugar maple (St. Clair
 27 et al., 2008). Although a variety of mechanisms (e.g.,
 28 maturation of forest vegetation) contribute to base
 29 cation depletion, acid deposition and the consequent
 30 mobilization of aluminum in soils have been identified
 31 as important drivers of depletion (Lawrence et al.,
 32 1995). Acid deposition and aluminum mobilization
 33 have contributed to dieback and decline of sugar maple
 34 (Bailey et al., 2004; Hallet et al., 2006; St. Clair et al.,
 35 2008) and to freezing injury in red spruce (DeHayes et
 36 al., 1999; Hawley et al., 2006). Recent work has found
 37 that declining growth rates can begin in apparently
 38 healthy sugar maple trees up to two decades before a
 39 strong decline in tree health becomes visually evident
 40 (Long et al., 2009). Other studies have found linkages between acid deposition, soil calcium availability,
 41 and the reproductive success of bird species (Graveland, 1998; Hames et al., 2002).

42 The ANC value of a waterbody reflects the sum of the base cation concentrations (calcium, magnesium,
 43 potassium, sodium) minus the sum of the acid anion concentrations (primarily sulfate and nitrate).
 44 Depletion of base cations from soils over time will provide lower concentrations of these base cations to
 45 waters and result in a lower ANC value than would otherwise be predicted based on the acid anion
 46 concentrations alone. In this way, losses of exchangeable soil base cations can slow the recovery of
 47 aquatic ecosystems (Likens et al., 1996; Lawrence et al., 1999). The scientific consensus is that base
 48 cation depletion is slowing the rate of chemical recovery of lakes and streams and, therefore, also the rate
 49 of biological recovery in many of the sensitive aquatic ecosystems targeted by Title IV of the CAAA



Sugar maple showing brilliant fall colors. Studies have shown that the health and growth rates of sugar maple can decline due to low availability of calcium and magnesium ions in soils and that these cations can become depleted from soils due to acid deposition. A recent study indicates that sugar maple growth rates can decrease for many years before visual signs of a decline in tree health become evident (Long et al., 2009).

Base cation depletion is a high-priority area for future acidification research of high relevance to the development of future air quality policies designed to protect ecosystems.

1 (Driscoll et al., 2001). Re-supply of these base cations generally occurs when chemical breakdown (i.e.,
2 weathering) of minerals in bedrock begins to release cations to soils and waters at a rate that is faster than
3 the leaching loss caused by acid deposition. Acid deposition must likely decrease below a critical level
4 that will vary among ecosystem locations before large increases in the ANC of acidified waters can occur.
5 The role of base cation depletion is even greater when considering episodic acidification because soil
6 chemistry plays an even greater role in stream chemistry during high-flow conditions than during low-
7 flow conditions (Lawrence, 2002).

8 Important complications to this simplified conceptualization of the link between acid deposition and base
9 cation depletion arise from the uptake of base cations by vegetation and from other factors. For example,
10 the cation balance is also controlled in part by forest age, health, and root-mining ability, as well as
11 harvesting practices (Blum et al., 2002; Hamburg et al., 2003; Grigal and Ohmann, 2005). The exact rate
12 of acid deposition that will allow recovery of base cation-depleted soils is not well known because the
13 rate of mineral weathering is not well quantified in most soils, can be highly variable among locations,
14 and is difficult to measure in the field (Bailey et al., 2003). Current models (such as the Model of
15 Acidification of Groundwater in Catchments [MAGIC]) that are commonly applied to make quantitative
16 predictions about acidification and critical loads in sensitive ecosystems can be and have been used to
17 examine the role of base-cation depletion; however, they have not yet been applied widely for this
18 purpose in the United States. Base-cation depletion is a high-priority area for future acidification research
19 and is of great relevance to the development of future air quality policies designed to protect ecosystems.

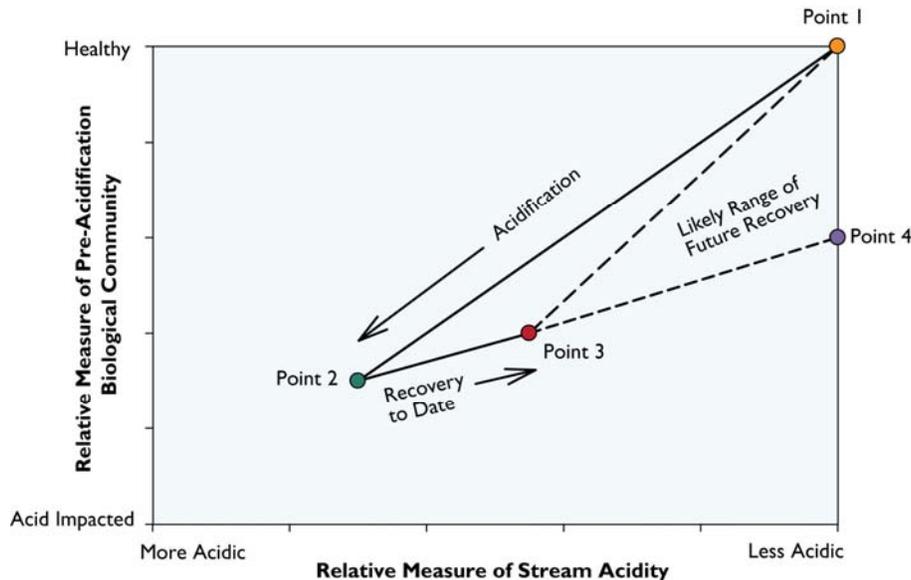
20 **3.2.1.2 Recovery of Biota**

21 **3.2.1.2.1 Aquatic Recovery**

22 Surface water chemistry data commonly have been favored as an ecosystem indicator of acidification
23 because these data are less expensive and more straightforward to obtain than data on aquatic biological
24 populations. In addition, many studies have shown thresholds that link water chemistry values to the
25 survival and reproductive success of organisms, such as brook trout, mayfly, and many others (Schofield,
26 1976; Baker and Christensen, 1991). Using surface water chemistry data alone to assess ecosystem
27 acidification assumes that when water chemistry degrades below a certain threshold, acid-sensitive
28 organisms will be replaced by acid-tolerant organisms, and that when the water chemistry improves to a
29 chemical threshold capable of successfully supporting more acid-sensitive organisms, then the organisms
30 will soon return to the waterbody. However, this is an oversimplified view of how ecosystems will
31 respond to changes in lake or stream water quality.

32 Acidification results in changes not only to the constituent or habitat that may be targeted as a threshold
33 indicator but also to a wide variety of other chemical constituents and physical habitats. As a result, a
34 complex set of changes in the ecological community may occur, including a shift in the members of the
35 community from acid-sensitive to acid-tolerant species. Acid-tolerant communities that develop in
36 response to acidification may then resist change as conditions become less acidic, limiting the re-
37 establishment of the acid-sensitive species that were originally impacted by acidification (Frost et al.,
38 2006). Other factors may also slow this process of biological recovery, including physical limitations on
39 dispersal, the proximity of species that may have historically resided in the water prior to acidification
40 (Yan et al., 2003; Langford et al., 2009), and calcium concentrations that tend to be lower relative to ANC
41 during ecosystem recovery than during acidification (Jeziorski et al., 2008; Cairns and Yan, 2009). These
42 factors act to delay biological recovery relative to chemical recovery (Snucins, 2003), and some scientists
43 have suggested that active ecosystem restoration (e.g., reintroduction of previously lost species) may be
44 needed to meet the goal of a “pre-acidification” biological community (Snucins and Gunn, 2003; Binks et
45 al., 2005).

1 When a different pathway of change is followed during recovery than was followed during acidification,
 2 this behavior is called “hysteresis.” A conceptual representation of hypothetical acidification and recovery
 3 patterns in a waterbody is presented in **Figure 3-1**. Point 1 represents the pre-acidification status of water
 4 quality and the biological community. Point 2 shows the status when conditions were most acidic, and
 5 Point 3 represents current conditions where water quality has shown some recovery, but a pre-
 6 acidification biological community has not yet been achieved. The two dashed lines represent possible
 7 future recovery trajectories for the biological community as future acid deposition levels continue to
 8 decrease. These recovery patterns may or may not be linear as shown, and some evidence suggests that
 9 “threshold” behavior marked by a sharp change in slope with time may occur in biological communities
 10 recovering from acidification (Monteith et al., 2005). One trajectory ends at Point 4, where water quality
 11 has fully recovered, but the biological community has only recovered slightly. The other trajectory returns
 12 to Point 1, representing a full recovery of water quality and the biological community. Some studies
 13 indicate that even with full recovery of water quality (less acidic stream on the right side of the figure),
 14 the biological community will likely differ from what existed prior to acidification (Frost et al., 2006;
 15 Cairns and Yan, 2009) suggesting that recovery may ultimately be closer to Point 4 than Point 1. Note
 16 that both of the hypothetical recovery trajectories follow a different path than did the acidification phase,
 17 indicating that acidified ecosystems show hysteretic behavior.



18
 19 **Figure 3-1. Conceptual figure showing hypothetical acidification and recovery patterns in a**
 20 **waterbody that has been acidified by atmospheric deposition. The two dashed lines reflect the**
 21 **range of possible recovery trajectories for the biological community once 100% chemical recovery**
 22 **has been reached, as determined by ANC or another measure of stream acidity. The changes may**
 23 **or may not be linear with time as shown (Prepared by USGS).**

24 Despite the concerns raised about limitations on
 25 recovery from acidification due to competition,
 26 dispersal, and other ecological factors that must be
 27 considered in addition to chemical thresholds,
 28 several studies in Europe and Canada have
 29 demonstrated biological recovery in aquatic
 30 communities that have become less acidic in
 31 response to decreases in acid deposition (Tipping
 32 et al., 2002; Findlay, 2003; Halvorsen et al., 2003).

There have been few studies in the United States on the recovery of aquatic biota over a period of gradually improving acidification status; this is largely because none of the existing U.S. monitoring networks regularly provide such evaluations and because most available data originate from ad-hoc studies.

1 Additionally, studies in which lime has been added to waterbodies or soils to hasten recovery from
2 acidification have shown trends towards a pre-acidification aquatic community (Raddum and Fjellheim,
3 2003), but the success of these efforts has sometimes required active restoration of some species (Driscoll
4 et al., 1996; Clair and Hindar, 2005).

5 There have been few studies in the United States on the recovery of aquatic biota over a period of
6 gradually improving acidification status; this is largely because none of the existing U.S. monitoring
7 networks regularly provide such evaluations and because most available data originate from ad-hoc
8 studies. Nonetheless, a few studies have evaluated aquatic communities by sampling waters over a period
9 when acidity was declining, and the general conclusion is that there is little evidence to date of
10 widespread recovery in these communities (Simonin et al., 2005; Burns et al., 2008a; Warren et al.,
11 2008).

12 3.2.1.2.2 Terrestrial Recovery

13 Few studies have tracked the status and potential
14 recovery of terrestrial ecosystems during the period
15 of decreasing acid deposition since the 1980s. This is
16 due in part to the historical focus on aquatic
17 acidification and a lack of understanding of the
18 terrestrial impacts of acidification at the time that the
19 CAA was amended in 1990. Since the early 1990s,
20 scientific understanding of terrestrial ecosystem effects of acid deposition has increased greatly, and
21 studies indicate continued degradation of soil base status (calcium and magnesium), as well as continued
22 deleterious effects on sensitive species such as sugar maple and red spruce in the most acid-sensitive
23 regions.

Currently, no published studies can document an improvement in the status of terrestrial ecosystems in the eastern United States relative to acidification effects due to decreased emissions and deposition.

24 As described previously, several studies have shown the link between acid deposition and losses of base
25 cations from soil. Many of these studies have included data from re-sampled soil locations, generally
26 documenting a lack of improvement or continued degradation of soil base cation status in the Adirondack
27 Mountains and Allegheny Plateau of Pennsylvania (Bailey et al., 2005; Sullivan et al., 2006b; Johnson et
28 al., 2008; Warby et al., 2009). Furthermore, a clear link between the base cation status of soils and the
29 health of sugar maple has been established in recent years (Bailey et al., 2004; Hallet et al., 2006; Long et
30 al., 2009). The role of acidification in red spruce freezing injury was established by the early 1990s
31 (McLaughlin et al., 1993). Recent data have largely confirmed the findings of earlier studies on red
32 spruce (Hawley et al., 2006) and have also found potential effects of acid deposition on other species of
33 forest vegetation (Jenkins et al., 2007; Zaccherio and Finzi, 2007). Currently, no published studies can
34 document an improvement in the status of terrestrial ecosystems in the eastern United States relative to
35 acidification effects due to decreased emissions and deposition. This conclusion likely results from three
36 factors: (1) there are few studies that have evaluated terrestrial ecosystem health relative to acidification
37 effects over time, (2) continued acidification of soils in the most acid-sensitive regions, and (3) lags in
38 response that would be expected in long-lived species such as forest trees.

39 3.2.2 Current Effects of Atmospheric Deposition on Aquatic and Terrestrial Ecosystems 40 in the Rocky Mountain Region of the United States

41 As shown in Chapter 2, the Rocky Mountain region of
42 Colorado and adjoining states receives atmospheric
43 deposition levels of sulfur and nitrogen that are generally
44 less than half the deposition levels measured in
45 mountainous parts of the eastern United States. Yet, the
46 Colorado Rockies landscape is generally more sensitive

The character of nitrogen deposition is different in the Colorado Rockies than in the East, with ammonia nitrogen deposition generally exceeding that of nitrate nitrogen.

1 to nutrient over-enrichment from
2 atmospheric nitrogen deposition than
3 landscapes in the eastern United
4 States. This sensitivity is due to the
5 steep slopes, thin or non-existent soil
6 cover, short growing season, and less
7 vegetation per unit landscape area
8 (Clow and Sueker, 2000). Several
9 studies have measured ANC values
10 near or below zero during snowmelt
11 in headwater streams, small
12 headwater lakes, and ephemeral
13 pools, and downward trends in ANC
14 values were evident in some waters
15 through the 1990s (Caine, 1995;
16 Williams and Tonnessen, 2000;
17 Campbell et al., 2004). However, the
18 extent of waters that become
19 episodically acidified in the Colorado
20 Rockies is generally less than in the
21 eastern United States, and no
22 deleterious effects of acidification on
23 fish have been documented.



Loch Vale in Rocky Mountain National Park receives among the highest loads of atmospheric nitrogen deposition in the Front Range of the Colorado Rockies. This watershed has been the site of long-term research into the effects of air pollutants on alpine and sub-alpine ecosystems (photo courtesy of Jill Baron, USGS).

24 Atmospheric deposition of nitrogen has been of interest in the Rocky Mountains because both aquatic and
25 terrestrial ecosystems in this region were historically nitrogen limited and have shown species shifts and
26 growth responses at nitrogen deposition levels as low as 1.5 kg/ha/yr (Wolfe et al., 2003; Baron, 2006).
27 Several factors distinguish nitrogen deposition patterns in the Colorado Rockies from those in the eastern
28 United States. First, the character of nitrogen deposition is different in the Colorado Rockies than in the
29 eastern United States. Ammonia nitrogen (i.e., $\text{NH}_3\text{-N}$) deposition, which generates greater acidity than
30 does nitrate, generally exceeds that of nitrate nitrogen (i.e., $\text{NO}_3\text{-N}$), especially at sites east of the
31 Continental Divide in the Front Range of the Rocky Mountains (Heuer et al., 2000; Burns, 2003; Beem et
32 al., 2010). This pattern is believed to result from the deposition of emissions carried by upslope winds to
33 the Front Range from agricultural sources that are located to the east of the mountains (Baron et al.,
34 2004). As a result, atmospheric nitrogen deposition at many monitoring sites in the Colorado Rockies is
35 not decreasing with decreases in power plant emissions in the western United States. Instead nitrogen
36 loads are still increasing in some western locations. This pattern is especially driven by a tendency
37 towards increasing trends in ammonia deposition (Burns, 2003).

38 Many effects on terrestrial and aquatic ecosystems from current levels of atmospheric nitrogen deposition
39 have been demonstrated in the Rocky Mountain region. Over the past two to three decades, increases in
40 nitrate concentrations and inorganic nitrogen loads in high-elevation streams and lakes have been
41 documented in a few areas of the Colorado Rockies (Williams and Tonnessen, 2000; Clow et al., 2003).
42 However, throughout a wider range of high-elevation waters in the western United States, trends have
43 been slight enough that other factors such as recent rainfall (Clow et al., 2003) and the melting of glaciers
44 (Baron et al., 2009) can greatly affect nitrate trends. Studies in which historical atmospheric deposition
45 levels were indirectly evaluated through measurements of nitrogen levels in lake sediment cores have
46 concluded that nitrogen deposition over the Colorado Rockies and many western mountain ranges began
47 to increase greatly after the 1950s. This trend is marked by shifts in lake plankton communities from low
48 nutrient species to those indicative of an increased nutrient supply (Wolfe et al., 2003; Saros et al., 2005).
49 The general conclusion across high-elevation waters of the western United States is that as atmospheric

1 nitrogen deposition increases from low levels of < 2 kg/ha/yr to higher levels, plankton biodiversity tends
2 to decrease and growth tends to increase as the aquatic-limiting nutrient shifts from nitrogen to
3 phosphorus (Interlandi and Kilham, 2001; Bergstrom and Jansson, 2006). These shifts and changes in
4 plankton communities are assumed to occur at a given location as atmospheric nitrogen deposition
5 increases over time and spatially from regions of low atmospheric nitrogen deposition to regions of high
6 atmospheric nitrogen deposition.

7 The changes described above for aquatic ecosystems are also paralleled by observed changes in terrestrial
8 plant communities, especially those in high-elevation alpine climatic zones of the Colorado Rockies.
9 Increases in atmospheric nitrogen deposition tend to increase growth and decrease species diversity in
10 alpine plant communities, with sedge and grass communities generally favored over shrubs (Bowman et
11 al., 1995; Seastedt and Vaccaro, 2001). Changes in vegetation nitrogen content of individual species can
12 occur at atmospheric nitrogen deposition levels as low as 4 kg/ha/yr, and whole community changes were
13 observed at values of 10 kg/ha/yr, as determined by nitrogen addition experiments (Bowman et al., 2006).

14 Together, the results of studies highlight the high sensitivity of aquatic and terrestrial ecosystems in the
15 Rocky Mountain region to atmospheric nitrogen deposition. Deleterious effects are generally observed at
16 deposition levels that are significantly less than those commonly measured in high-elevation ecosystems
17 of the eastern United States. These results are pertinent to the management of ecosystems in this region
18 because of the large number of National Parks (e.g., Rocky Mountain, Grand Teton, Yellowstone,
19 Glacier) that are managed as Class 1 Wilderness areas under CAA provisions to prevent significant
20 deterioration of ecosystems (Porter and Johnson, 2007). Thus, a more active federal role in land
21 management and ecosystem protection from air pollutant deposition may be necessary in the large tracts
22 of Class 1 wilderness land that are present in the west.

23 **3.2.3 Current Effects of Atmospheric Nitrogen Deposition on Terrestrial and Aquatic** 24 **Ecosystems in the Far Western United States**

25 In many areas of the far western United States (lands to the west of the Rocky Mountains, especially the
26 west coast states), nitrogen loads to terrestrial systems have historically been low compared with loads to
27 terrestrial systems in the eastern United States. The native plant communities (e.g., coastal sage scrub,
28 desert scrub, annual grasslands) are adapted to thrive in low-nitrogen environments. When nitrogen loads
29 to these habitats increase, the native plant and soil communities have difficulty competing with invasive
30 species that are better adapted to more nitrogen-rich environments. In California, nitrogen deposition
31 levels as low as 5–8 kg/ ha/yr have major effects on epiphytic lichens (i.e., lichens that grow on trees and
32 shrubs) and contribute to changes in native plant communities by enhancing invasion by exotic annual
33 grasses (Fenn et al., 2003a; Weiss, 2006). In coastal sage scrub, desert scrub, and annual grasslands in
34 California, nitrogen deposition favors non-native grass invasions, which can lead to altered plant
35 communities. In southern California chaparral (Egerton-Warburton et al., 2001) and coastal sage scrub
36 ecosystems (Egerton-Warburton and Allen, 2000), the diversity of the mycorrhizal fungi communities
37 that grow in association with roots is also negatively impacted by nitrogen deposition. Shifts in these
38 plant communities are of additional concern because the invasive vegetation that results from increased
39 nitrogen loads may also increase the risk of wildfires due to increased fuel availability. In the desert scrub
40 ecosystems of Joshua Tree National Park, invasive grasses can provide sufficient fuel load to carry fire,
41 particularly in a wet year. This increased fuel load puts the Park's native vegetation at risk, including its
42 namesake, the Joshua tree. Fire occurrence in the desert is a rare event in the absence of the invasive
43 grasses (Allen et al., 2009).

44 Increased nitrogen deposition can also impact forests in the far western United States. Nitrogen deposition
45 effects, in combination with the physiological effects of ozone, decrease the amount of roots, and increase
46 susceptibility of ponderosa pine to bark beetle attack and mortality (Jones et al., 2004). Forest
47 sustainability is thus threatened, and fire risk is increased (Grulke et al., 2009). Studies have been

1 conducted to assess various methods of reducing nitrogen saturation symptoms. Using fire as a
2 management technique to reduce excess nitrogen was found to be ineffective in chaparral ecosystems,
3 presumably because of the limited capacity of fire to reduce the large nitrogen stores in the mineral soil of
4 these ecosystems (Meixner et al., 2006). Similarly, in forests affected by excess nitrogen, a combination
5 of periodic nitrogen release by fire and significant reductions in nitrogen deposition are needed to
6 mitigate the effects of nitrogen accumulated from long-term nitrogen deposition (Gimeno et al., 2009).

7 Nitrogen deposition effects on the lichen community are pronounced and widespread in the forests, oak
8 woodlands, and chaparral of California, as evidenced by major shifts from dominance of lichen species
9 that thrive in low-acid, low-nitrogen environments to communities dominated by lichen species that thrive
10 in acidic, high-nitrogen environments (Fenn et al., 2008; Jovan, 2008; Jovan and McCune, 2005). Similar
11 effects of nitrogen in lichen communities are well documented in forests near urban and agricultural
12 emissions source regions in Washington and Oregon and in the Columbia River Gorge corridor (Fenn et
13 al., 2007; Geiser and Neitlich, 2007).

14 In the streams and lakes of the far western United States, increased nitrogen deposition can alter lake and
15 stream chemistry. At moderate nitrogen deposition levels of 10–14 kg/ha/yr for chaparral catchments and
16 of 17 kg/ha/yr for forested catchments in the southwestern Sierra Nevada and in the Transverse Ranges
17 near Los Angeles, high concentrations of nitrate are found in stream water. In chaparral, forested, and
18 high-elevation wilderness catchments in the Los Angeles air basin, up to 40% of the nitrate exported in
19 stream water was directly deposited from the atmosphere and transported through the system without
20 biological assimilation (Michalski et al., 2004), clearly indicating that the source of this excess nitrogen
21 was air pollutant deposition in these watersheds.

22 High-elevation watersheds in the Sierra Nevada appear to export nitrate during early snowmelt, even with
23 relatively low atmospheric nitrogen deposition similar to the high tendency to export nitrate discussed
24 above for the Colorado Rockies. Although the role of nitrogen deposition on nitrate leaching in the high-
25 elevation Sierra Nevada is not entirely clear, nitrogen deposition does not appear to have as severe an
26 impact on lake and stream water chemistry, particularly in Sierra Nevada catchments, where soils tend to
27 be thicker than those of the Colorado Front Range. Differing responses between the two regions may be
28 due to higher nitrogen deposition in the Colorado Rockies or to environmental differences. For example,
29 the greater extent of soil freezing in the Colorado Rockies may reduce the nitrogen retention capacity of
30 these watersheds (Sickman et al., 2002). However, watersheds in the Sierra Nevada are expected to be
31 sensitive to increases in nitrogen deposition in the seasonal snowpack. This expectation is based on the
32 large pulses of nitrate that are flushed to surface waters during snowmelt-driven spring runoff, (Sickman
33 et al., 2003), a short growing season, the limited soil and vegetation cover, and the limited biological
34 nitrogen demand during the early spring runoff period (Fenn et al., 2003b).

35 **3.3 Effects of Atmospheric Nitrogen Deposition on Coastal Estuaries**

36 Eutrophication and associated hypoxia is widespread in estuaries of the United States, including Long
37 Island Sound, the Chesapeake Bay, and the Gulf of Mexico near the Mississippi River Delta (Diaz, 2001;
38 Kemp et al., 2005). Nitrogen and phosphorus are the nutrients that commonly limit algal growth in coastal
39 ecosystems. The role of nitrogen in estuarine eutrophication and hypoxia is often attributed to a
40 combination of the delivery of excess nitrogen by rivers and the direct atmospheric deposition of nitrogen
41 onto the water surface. These same mechanisms also deliver nitrogen to the open ocean, and atmospheric
42 nitrogen deposition has been shown to affect the growth of marine plankton (Krishnamurthy et al., 2010).
43 The potential sources of nitrogen to estuaries include runoff from fertilizer and animal waste applied to
44 agricultural landscapes, human waste in urban/suburban landscapes, and atmospheric nitrogen deposition.
45 The relative role of atmospheric nitrogen deposition to total nitrogen loads to coastal estuaries differs
46 widely, from about 15% to 25% in the Mississippi Delta region of the Gulf of Mexico, to 20% to 50% in
47 Chesapeake Bay and Tampa Bay, and as much as 60% to 80% in some coastal river estuaries of northern

1 New England (McIsaac et al., 2001; Boyer et al.,
2 2002; Poor et al., 2006; Whitall et al., 2007;
3 Howarth, 2008; Alexander et al., 2008). The
4 relative contributions of nitrate and ammonia as
5 sources of atmospheric nitrogen to estuaries also
6 vary widely across coastal regions of the United
7 States. Nitrate is generally the dominant form of
8 nitrogen in atmospheric deposition across the
9 United States; however, ammonia can be the
10 dominant atmospheric species of nitrogen in parts
11 of estuarine watersheds, particularly those with
12 abundant agricultural land (Lawrence et al., 2000;
13 Whitall et al., 2003). The most recently available
14 data indicate that the majority of U.S. estuaries
15 show signs of eutrophication (Bricker et al., 2008).
16 Although most of these coastal waters have not shown changes in nutrient loads or trophic status since the
17 1990s (Alexander and Smith, 2006), about 20% to 25% of U.S. coastal waters show improved trophic
18 status in recent decades (Alexander and Smith, 2006; Bricker et al., 2008), most notably Tampa Bay. The
19 role of decreased loads of atmospheric nitrogen deposition to observed decreases in estuarine
20 eutrophication are likely only minor, but are not well quantified (Greening and Janicki, 2006).

Eutrophic waters are those with an overabundance of nutrients, such as nitrogen or phosphorus. As nutrient levels in the water increase, the growth of algae and plankton increases. When these organisms die and are decomposed by bacteria, dissolved oxygen in the water is consumed. “Dead zones” characterized by hypoxia (i.e., low levels of dissolved oxygen in the water) or anoxia (i.e., no dissolved oxygen in the water) may result, harming or killing fish, crustaceans, and other organisms. Benthic (bottom-dwelling) organisms that are unable to move to more oxygenated waters are particularly susceptible.

21 The contribution of headwater streams to the total nitrogen loads in rivers can be substantial. For
22 example, small streams account for more than two-thirds of the water volume and approximately two-
23 thirds of the nitrogen delivered to large rivers in the Northeast (Alexander et al., 2007). Despite a higher
24 removal rate (denitrification) of nitrate from headwater reaches than from large river channels during
25 transport (Alexander et al., 2008), the colder temperatures at the higher elevations of the headwaters act to
26 slow this rate of loss (Schaefer and Alber, 2007). Therefore, a substantial amount of the nitrogen
27 deposited on upland forested areas from the atmosphere may later be exported to larger rivers and
28 estuaries. These studies emphasize the connection of upland ecosystems to coastal ecosystems and
29 indicate that the effects of atmospheric nitrogen deposition extend beyond acidification in sensitive
30 upland landscapes.

31 The issue of ocean acidification has emerged in recent years, with the observation that rising levels of
32 carbon dioxide (CO₂) in the atmosphere are causing decreases in ocean pH and alkalinity, with numerous
33 implications for the health of marine ecosystems (Caldeira and Wickett, 2003). Some recent studies have
34 explored the role of atmospheric nitrogen and sulfur deposition on ocean acidification and concluded that
35 acid deposition contributed no more than a few percent to ocean acidification trends in recent years (Bates
36 and Peters, 2007). However, the contribution of acid deposition to marine pH declines is likely greater in
37 coastal regions closer to continents (Doney et al., 2007). Further scrutiny of the role of acid deposition on
38 the pH and alkalinity of coastal ecosystems may be needed in the future given the serious consequences
39 of ocean acidification that are currently forecast.

40 **3.4 Emerging Issues at the Interface of Science and Policy in Managing the** 41 **Effects of Acid Deposition on Ecosystems**

42 **3.4.1 Critical Loads**

43 Critical loads, which were introduced in Chapter 2, can be viewed as providing a synthesis of scientific
44 knowledge about thresholds of harm to or alteration of ecosystems that can inform the development,
45 implementation, and assessment of air quality policy and the management of sensitive ecosystems.
46 Critical loads have been widely used for large-scale applications and policy purposes in Europe and
47 Canada since the 1980s (Jeffries and Ouimet, 2005; Spranger et al., 2008), but historically have not been

1 widely used for these purposes in the United States (Porter and Johnson, 2007). For example, critical load
2 maps for soils and waters are produced by several European countries that are parties to the Convention
3 on Long-range Transboundary Air Pollution. Although a signatory to this Convention, the United States
4 did not commit to the provisions regarding critical loads contained in the Convention (see recent
5 discussion in Spranger et al., 2008). Academic critical load studies in the United States also date to the
6 1980s (Henriksen and Brakke, 1988), and many studies continue to be published (Baron, 2006; Fenn et
7 al., 2008; Sullivan et al., 2008); however, most past work has been of limited geographic scope. Recently
8 published studies provide examples of larger-scale assessments of critical loads in the United States,
9 including nationwide critical load maps for forest soils (McNulty et al., 2007) and a review of published
10 studies throughout the United States for empirical critical loads of nitrogen (Pardo et al., in press).

11 Critical loads can be calculated for any species of concern or for a sensitive biological community within
12 an aquatic or terrestrial ecosystem if a threshold of atmospheric deposition can be identified above which
13 negative biological effects on the species or alteration of the community is expected. Because various
14 species in an ecosystem may have differing sensitivities to air pollutant loads, a large number of species-
15 specific or community-specific critical loads are likely to exist for a given location (Sullivan et al., 2008).
16 Critical loads are often linked to thresholds at which the organism experiences a certain biological effect,
17 which may range from stunted growth to reduced reproductive levels to increased mortality. Critical loads
18 are usually calculated in reference to a chemical quantity believed to provide an index of acidification or
19 nutrient over-enrichment. Potential indices include ANC levels or nitrate concentrations in a waterbody,
20 or the base cation saturation level of a soil. Multiple critical loads and ecological effects end points
21 provide policy makers or land managers with multiple options to determine the level of protection that is
22 warranted based on the species or communities of greatest concern at a given location. This is part of the
23 process of establishing a target load, which may differ from the strictly science-based critical load. The
24 target load reflects inclusion of economic, social, cultural, or other considerations to establish the
25 protection level of the ecosystem in question, as well as the load that is deemed “achievable” by managers
26 or regulators (Porter et al., 2005; Porter and Johnson, 2007).

27 In addition to a variety of critical loads applicable for each ecosystem based on the species and biological
28 effects being considered, there are different types of critical loads and different methods for establishing
29 critical loads. Critical loads can be determined by an empirical approach based on field- or laboratory-
30 based studies that define a chemical threshold of sensitivity for a species or community. A recent example
31 is the establishment of a critical load of 1.5 kg/ha/yr of atmospheric nitrogen deposition to protect against
32 eutrophication (nutrient over-enrichment) in lakes in the Rocky Mountain National Park. This load was
33 established based on evidence from lake sediment cores that showed a shift in phytoplankton species as
34 nitrogen deposition values rose above 1.5 kg/ha/yr in the 1960s (Porter and Johnson, 2007). Critical loads
35 may also be determined by a mathematical expression, such as the Steady-State Mass Balance model for
36 soils. This model compares the concentrations of neutralizing base cations (e.g., calcium, magnesium) to
37 those of potentially toxic acidifying cations (e.g., aluminum) in soils (Sverdrup and deVries, 1994). Both
38 the empirical and mathematical expression approaches develop steady-state critical loads based on an
39 assessment of current levels of atmospheric deposition relative to some threshold of harm or change.
40 Critical loads can also be calculated using a dynamic approach that involves more sophisticated modeling
41 of time-varying patterns of emissions, deposition, soil chemistry, and lake or stream chemistry. This
42 approach often uses some of the acidification models (e.g., MAGIC or PnET-BGC) described in the text
43 box on page 3-13 and can provide information on the time expected to reach a threshold based on a given
44 emissions policy as it is implemented over time (Sullivan et al., 2008). The dynamic critical load
45 approach is especially valuable for evaluating the success of air quality policies and programs as they are
46 implemented over multiple years.

47 Despite the lack of a long history of applying critical loads to inform air pollutant policies in the United
48 States, interest in the use of critical loads in the United States has increased in recent years with the

1 advent of a Critical Load science committee within the NADP (NADP, 2009), several recent workshops
2 and meetings on this topic, and several publications exploring greater use of critical loads as a policy-
3 relevant environmental assessment tool (Porter et al., 2005; Fisher et al., 2007; Burns et al., 2008b;
4 Glavich and Geiser, 2008). Recently published studies have generally found that for many regions well
5 known for their sensitivity to either acidification or excess nitrogen deposition (e.g., the Adirondack
6 Mountains of New York, the central Appalachian region of Virginia, the Front Range of Colorado), the
7 current levels of atmospheric deposition generally exceed the critical loads for many species of concern
8 (Baron, 2006; McNulty et al., 2007; Sullivan et al., 2008). Case studies exploring the application of the
9 critical load approach in for the Adirondack Mountains and the central Appalachian region are also
10 included in Chapter 2 and Chapter 4 of this report. Finally, consideration of two key factors is warranted
11 when using scientific information on critical loads to inform public policy implementation: (1) critical
12 loads can have high uncertainty that stems from model assumptions and data, as well as uncertainty in the
13 threshold itself (Skeffington, 2006; Li and McNulty, 2007), and (2) other variables such as forest
14 harvesting and climate change will affect critical loads in addition to atmospheric deposition (Watmough
15 et al., 2003). When critical load calculations are being used to inform air pollutant policies, scientific
16 uncertainty must be properly considered, along with the role of other mitigating variables.

17

Models Used to Investigate the Ecosystem Effects of Acid Deposition

Models play a pivotal role in understanding the ecosystem effects of acid deposition, as well as helping to inform air quality policy. Below are the principal models that have been used to study ecosystem effects.

Initial models were simple, using an analysis of surface water chemistry based on charge balance or titration by strong acids (Henriksen, 1979).

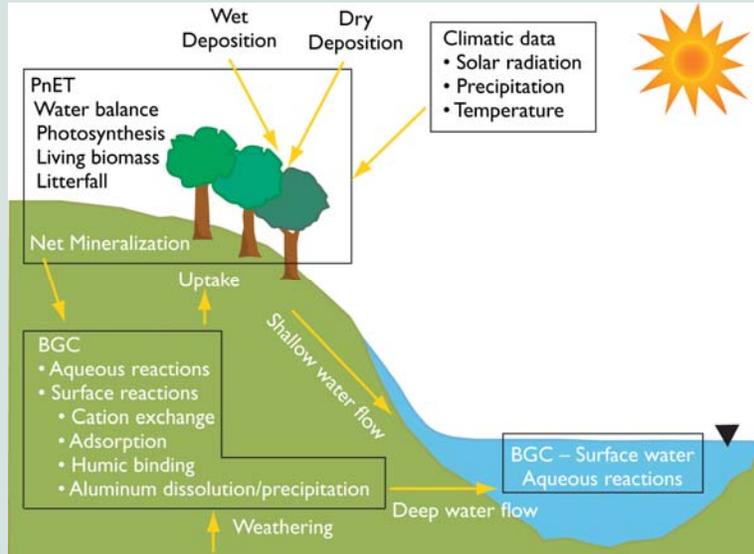
Dynamic Models – Later, more formal computer models were developed that simulate biogeochemical processes and hydrology occurring in catchments (e.g., see the accompanying figure). The dynamic models listed below all share an ability to make predictions about future concentrations and loads of chemical constituents (e.g., pH, ANC) in lakes and streams given the chemistry of atmospheric deposition. All of them can be combined with atmospheric models such as CMAQ (Community Multi-Scale Air Quality model) that estimate atmospheric deposition to landscapes from present emission levels or future emission scenarios (Schwede et al., 2009):

- Integrated Lake-Watershed Acidification Study (ILWAS) (Gherini et al., 1985)
- Birkenes model (Chrisopherson et al., 1982)
- Model of Acidification of Groundwater in Catchments (MAGIC) (Cosby et al., 1985d)
- PnET-BGC (Gbondon-Tugbawa et al., 2001)

(continued)

18

Models Used to Investigate the Ecosystem Effects of Acid Deposition (continued)



Conceptual diagram of the biogeochemical processes and chemical stores of the pNET-BGC model (figure courtesy of Charles Driscoll, Syracuse University).

Other dynamic models have been applied more commonly in ecosystems of the western United States:

- DayCent/PHREECQ (DayCent-Chem) (Hartman et al., 2007)
- Alpine Hydrochemical Model (AHM) (Meixner et al., 2004).

Steady State Models – These are another class of models that focus only on a steady-state approximation of either water or soil chemistry (Henriksen and Posch, 2001). These models are computationally simpler and demand less input data to run:

- Steady-State Water Chemistry (SSWC)
- Simple Mass Balance Equation (SMBE).

Several recent surface water chemistry modeling studies have explored likely future ecosystem responses to existing air pollutant policies (e.g., Title IV), along with some future emissions scenarios (Chen and Driscoll, 2004; Sullivan et al., 2004; Chen and Driscoll, 2005b; Chen and Driscoll, 2005a; Lawler et al., 2005; Sullivan et al., 2008). These modeling studies not only generally confirm the success of Title IV at improving the acid-base status of surface waters, but also highlight regional differences and point out that additional emission reductions will be needed to realize the goal of full ecosystem recovery to conditions resembling pre-acidification conditions. Chapter 4 of this report includes a modeling analysis in which MAGIC was used to assess aquatic ecosystem recovery in several regions of the eastern United States as a result of implementing current policies and programs in comparison to results achieved as a result of implementing three alternative future emission-reduction scenarios.

1

2 3.4.2 Atmospheric Deposition and Ecosystem Services

3 In addition to the health benefits of the ARP discussed in Chapter 1 of this report, benefits to ecosystem
 4 services might be expected as well, and these benefits can be evaluated by considering the varied material
 5 and intangible goods and services by which ecosystems support human well-being. Ecosystem services
 6 can be defined as the sum total of benefits that humans receive from the resources and processes of
 7 natural ecosystems (Corvalan et al., 2005). These benefits range from extractive services or provisioning

1 (e.g., wood products, minerals, water) to regulating services (e.g., climate regulation and nutrient cycling)
2 to cultural services (e.g., outdoor recreation). The ability of ecosystems to provide these services over the
3 long term is impacted by disturbances (Beier et al., 2008), including atmospheric deposition. The
4 response of ecosystems to disturbances, such as acid deposition, depends on the resiliency of the
5 ecosystem in question. Over the past two decades, researchers have begun to establish the value of these
6 various ecosystem services (Costanza et al., 1997) and to develop approaches for valuating both the direct
7 use-related benefits, as well as the non-use benefits of ecosystems; most studies indicate these non-use
8 benefits exceed those of the more traditional market-based benefits derived from resource extraction
9 (Walsh et al., 1990).

10 A recent review discusses and summarizes studies on estimating the monetary value of the effects of
11 sulfur and nitrogen emissions and deposition on ecosystems (Krupnik and Siikimaki, 2008). The studies
12 cited can be divided into those that estimate the monetary value of terrestrial, aquatic, and transitional
13 (mainly wetland) ecosystems. Several studies have generally estimated the value of adverse ecosystem
14 effects, but many fewer studies have attempted to relate specific adverse effects that can be directly
15 attributed to sulfur and nitrogen deposition. Some studies have estimated the value of benefits from
16 minimizing or eliminating ecosystem disturbances such as insect defoliation of forests (Kramer et al.,
17 2003) and estuarine eutrophication (Morgan and Owens, 2001), in which acid deposition is clearly one
18 contributing factor among many. However, the monetary role of acid deposition cannot be estimated in
19 most of these studies.

20 The greatest challenge in developing specific data on the economic benefits of changes in acid deposition
21 due to emission reductions lies with the availability of comprehensive scientific evidence that defines the
22 extent and magnitude of the adverse effects that can be directly attributed to acid deposition from among
23 multiple ecosystem stressors (Krupnik and Siikimaki, 2008). In one notable study, Banzhaf et al. (2006)
24 estimated ecological benefits of a policy that would substantially decrease adverse effects of acid
25 deposition in the Adirondack Park of New York. These benefits are in the range of about \$300 million to
26 \$1.1 billion for the Adirondack Park only and do not consider ecological benefits in other regions of the
27 United States from these deposition reductions. As described in Evans et al (2008), researchers are
28 applying economic methods to show the total value people place on ecological improvements to
29 important resources, such as the Adirondack Park, expected from further reductions in acid deposition.
30 However, as discussed in Chapter 1 of this report, no studies have comprehensively evaluated the
31 monetary benefits to ecosystem services of the ARP across all ecosystem types that would be sufficient to
32 make comparisons to overall health benefits and costs of the ARP. Significant future analytical work and
33 basic ecological and economic research is needed to build a sufficient base of knowledge and data to
34 support an adequate assessment of ecological benefits.

35 **3.5 What are the Implications of Ongoing and Future Changes in Climate and** 36 **the Carbon Cycle for the Effects of Acid Deposition on Ecosystems?**

37 The Earth has warmed by an average of 0.74° C
38 between 1906 and 2005, and there is a high
39 likelihood that recent warming over the past several
40 decades has been driven in large part by increases
41 in atmospheric concentrations of CO₂ and other
42 greenhouse gases (IPCC, 2007). Under a range of
43 likely future scenarios of greenhouse gas emissions
44 and a range of climate model predictions, the Earth is likely to warm by another 1.4°–5.8° C during the
45 twenty-first century (IPCC, 2007).

Ongoing and future predicted climate change is likely to greatly affect the interactions of sulfur and nitrogen deposition with ecosystems in a complex manner that does not easily lend itself to simple summaries and has high uncertainty.

46 Ongoing and future predicted changes in the climate and the carbon cycle have numerous implications for
47 the response of ecosystems to atmospheric deposition of sulfur and nitrogen. In addition to warming air

1 temperatures, a wide array of other environmental variables are predicted to change with the climate,
2 including water availability and the rates of many nutrient cycling processes. These additional
3 environmental changes and their effects are likely to vary by region (Boisvenue and Running, 2006). In
4 short, ongoing and future predicted climate change is likely to greatly affect the interactions of sulfur and
5 nitrogen deposition with ecosystems in a complex manner that does not easily lend itself to simple
6 summaries and has high uncertainty (McLaughlin and Percy, 1999). For example, although the effects of
7 an average increase in air temperature are likely to induce widespread changes in ecosystems, an increase
8 in the magnitude and frequency of a wide variety of climatic events (e.g., floods, droughts, fires) also is
9 expected. Some have suggested that these events are likely to affect ecosystems to a greater extent than
10 long-term average changes in temperature (Jentsch et al., 2007; Dale et al., 2001). Despite these
11 complexities, a brief assessment of likely interactions between ecosystems, acid deposition, and climate
12 change is warranted in this current assessment given the potential magnitude and multi-faceted impacts of
13 the projected changes. This section focuses on ecosystem effects and does not discuss the interactions
14 between climate change and greenhouse gases or how these interactions may affect the transport,
15 atmospheric interactions, and atmospheric deposition of sulfur and nitrogen. These atmospheric changes
16 have been discussed elsewhere (Tagaris et al., 2008) and are beyond the scope of this assessment.

17 **3.5.1 Implications of Climatically Driven Changes in the Species Composition of** 18 **Ecosystems**

19 Perhaps the greatest impact of climate change is likely to be wholesale changes predicted for the
20 biological communities found in many ecosystems sensitive to acid deposition effects, particularly those
21 at high elevations (Ibanez et al., 2007). For example, a migration northward and to higher elevations is
22 predicted for many tree species under most current climate change forecasts as they are replaced by
23 species better adapted to a warmer climate (McKenney et al., 2007; Lenoir et al., 2008). Evidence
24 indicates that migration of lower elevation forests to higher elevations is already occurring in Vermont,
25 suggesting this shift is ongoing in mountainous areas that have warmed in recent decades (Beckage et al.,
26 2008). Similar cold-to-warm water species shifts are also predicted for aquatic ecosystems (Mohseni et
27 al., 2003; Durance and Ormerod, 2007). The rate at which these ecosystem changes will occur is not well
28 known and is likely to reflect interactions with other contemporaneous changes, such as those in water
29 availability, light, insect defoliation, land use, and air pollutant deposition. Overall, the evidence suggests
30 that many species may reach an out-of-equilibrium state with respect to a future warmer climate (Higgins
31 and Harte, 2006; Mohan et al., 2009), and this state may result in increased sensitivity of many terrestrial
32 and aquatic ecosystems to the effects of other disturbances, such as sulfur and nitrogen deposition. A
33 European study (Alcamo et al., 2002) concluded that climate change is likely to have little effect on the
34 sensitivity of forested ecosystems to air pollutant deposition. However, the weight of evidence from other
35 studies indicates that, despite some uncertainty in the effects of climate change on ecosystems (Aber et
36 al., 2001), ongoing and likely future climate change will provide an added stressor to many ecosystems in
37 the United States. Furthermore, several studies indicate that the combination of atmospheric deposition of
38 sulfur and nitrogen with high ozone concentrations will act as additional ecosystem stressors in many
39 regions, such as the eastern United States (McLaughlin and Percy, 1999; Mohan et al., 2009). The
40 increases in extreme events, such as droughts and fires, which are predicted with future climate change,
41 will likely act as additional stressors to many of these ecosystems, with greater effects expected in the
42 western United States (Dale et al., 2001; Knapp et al., 2008).

43 **3.5.2 Climate Warming Will Affect Many Temperature-Sensitive Biogeochemical** 44 **Processes in Ecosystems**

45 Many biogeochemical processes affect the cycling of atmospherically deposited sulfur and nitrogen in
46 ecosystems and play a pivotal role in the acidification of ecosystems, including mineral weathering,
47 nitrification, and sulfur and nitrogen mineralization. The rates of nearly all of these processes are likely to
48 be affected by changes in climate. For example, the rates of mineral weathering and nitrification increase

1 with increasing temperature if all other factors are held constant (White et al., 1999; Dalias et al., 2002).
2 Studies have shown widely varying biogeochemical responses to past and predicted climate warming in
3 the United States, and these varying responses reflect interactions with precipitation, snow and ice, and
4 seasonal variations. For example, Campbell et al. (2009) predict higher nitrate concentrations in soil water
5 and streams at Hubbard Brook, NH, due to warming-induced increases in nitrogen mineralization and
6 nitrification, yet they also predict decreased rates of mineral weathering due to decreases in soil moisture.
7 At the Rocky Mountain National Park in the western United States, stream nitrate concentrations have
8 been increasing in recent years due to the melting of glaciers and snow fields by increased warming in
9 summer through fall and the leaching of nitrate that occurs when the underlying soils are exposed to
10 microbial processes (Baron et al., 2009). These results highlight the sensitivity of long-term stream water
11 quality to changes in snowfall, snowmelt, and the mass of glaciers in high-elevation watersheds of the
12 western United States. Data from the European Alps show a recent trend towards increasing solute
13 concentrations in high-elevation lakes, consistent with accelerated mineral weathering rates driven by
14 warmer air temperatures and less extensive snow cover that lasts for a shorter duration (Mosello et al.,
15 2002; Rogora et al., 2003). Increased mineral weathering rates would likely result in a greater ability of
16 soils to neutralize acid deposition and might accelerate recovery in sensitive ecosystems when
17 accompanied by decreasing atmospheric deposition rates of sulfur and nitrogen. However, a recent study
18 in the Colorado Rockies showed that regions underlain by sulfide minerals such as pyrite may experience
19 greater acidification with warmer air temperatures due to increased mineral weathering rates and the
20 subsequent formation of sulfuric acid (Mast et al., in press).

21 For a range of likely twenty-first century climate change scenarios in Europe, Posch (2002) concluded
22 that ecosystems will generally become less sensitive to acid deposition, primarily due to increasing rates
23 of mineral weathering in soils driven by warmer temperatures. The amount of land in Europe subjected to
24 sulfur and nitrogen deposition loads in excess of the critical load for acidification is likely to decrease
25 with climate change, except in areas predicted to become more arid. Decreases in the exceedance of
26 critical loads for nitrogen-driven nutrient over-enrichment are also predicted with future climate change in
27 Europe (Posch, 2002), but these decreases were less than those modeled for decreases in critical load
28 exceedances for acidification. In contrast, existing studies in the United States predict higher nitrate
29 concentrations (which should lead to greater critical load exceedances for nutrient over-enrichment) with
30 future climate change (Campbell et al., 2009; Wu and Driscoll, 2010). These opposing forecasts for
31 expected future nitrate chemistry in surface waters indicate the current high uncertainty about the role of
32 climate change and points to a need for additional research and improved models to explore this important
33 issue (Spranger et al., 2008).

34 **3.5.3 Implications of Changes in Carbon Dioxide Concentrations in the Atmosphere**

35 One of the fundamental factors that affect the rates of photosynthesis and terrestrial ecosystem growth is
36 the availability of CO₂. Global atmospheric CO₂ concentrations have increased by nearly 40% since the
37 mid-eighteenth century and are expected to double during the twenty-first century (IPCC, 2007). The
38 ecosystem “CO₂ fertilization effect” has been much discussed in the literature (Tans et al., 1990; Nowak
39 et al., 2004). Indeed, measurements and satellite observations of forested ecosystems, as well as CO₂
40 manipulation experiments, have generally reported increasing net ecosystem growth (primarily in forests)
41 as CO₂ concentrations have increased in temperate regions of the world, although there have been fewer
42 monitoring-based studies in the United States than in Europe (Norby et al., 2005; Boisvenue and
43 Running, 2006). The ecological responses have not been consistent, however, in all studies and across all
44 regions. A wide variety of factors (e.g., disturbance history, water availability, solar radiation), in addition
45 to experimentally altered CO₂ concentrations, can contribute to these diverse patterns in net terrestrial
46 ecosystem growth trends (Nemani et al., 2003; Boisvenue and Running, 2006; Potter et al., 2006). There
47 are several challenges in trying to quantify the role of CO₂ in forest growth. Many factors generally co-
48 vary across sites, and experiments necessary to understand the interacting effects of these multiple factors
49 have not been undertaken (Norby and Luo, 2004; Rustad, 2008). Also, the nearly impossible task of

1 designing experiments that adequately consider variations in the numerous factors that influence
2 ecosystem growth has been noted (Aber et al., 2001; Boisvenue and Running, 2006). When variations in
3 atmospheric sulfur and, especially, nitrogen deposition are added to this mix of factors, the difficulty in
4 attributing cause and effect to observed changes in ecosystems increases further (Bynterwicz et al., 2007),
5 and some argue, can only be interpreted through the use of models (Aber et al., 2001). Nonetheless,
6 results from a CO₂ enrichment experiment that simulated likely twenty-first century concentrations
7 showed increased soil acidification and increased rates of mineral weathering, suggesting that important
8 interactions with terrestrial ecosystem effects of atmospheric deposition may occur in the future (Andrews
9 and Schlesinger, 2001).

10 **3.5.4 Changes in the Water Cycle are Key to Understanding Ecosystem Effects**

11 Under various likely twenty-first century climate-change scenarios, most climate models predict an
12 overall increase in global precipitation of about 1–3% per degree of temperature increase (Held and
13 Soden, 2006). Recent observations and satellite measurements generally support an even greater increase
14 in precipitation of about 5–10% per degree of warming (Wentz et al., 2007; Zhang et al., 2007). Such a
15 change is expected to increase the wet deposition of sulfur and nitrogen in the eastern United States if all
16 other factors remain the same (Civerolo et al., 2008). However, the prediction of climate “wetting” is less
17 certain than the prediction of climate warming, and changes in precipitation patterns are predicted to vary
18 widely over the United States, with forecasts for a wetter climate in the eastern United States and for a
19 drier climate in large parts of the western United States (Milly et al., 2005). These drier climate patterns
20 are already evident in the western United States, which is exhibiting smaller winter snowpacks and earlier
21 snowmelt in mountainous areas (Mote et al., 2005).

22 Earlier snowmelt has also been noted in upland watersheds in the Northeast (Burns et al., 2007). Model
23 predictions with likely future climate scenarios indicate continued diminishing snowfall, less snowpack,
24 and less of a role for snowmelt in the hydrologic cycle of temperate regions of the Northern Hemisphere
25 (IPCC, 2007). These patterns may affect the cycling of sulfur and nitrogen pollutants through ecosystems
26 and watersheds by impacting episodic acidification, soil freezing, and the flows and temperatures in
27 surface waters. When snow melts, surface-water chemistry is often at its most acidic and has its highest
28 nitrate concentrations; therefore, a diminished snowmelt may lessen the episodic acidification of aquatic
29 ecosystems (Moore et al., 1997). However, these predicted trends of diminishing snowmelt are uncertain,
30 and an increase in mid-winter rain has been noted in the eastern United States (Hodgkins et al., 2003).
31 This increase in winter rain will also likely trigger strong episodic acidification, even when the rain falls
32 on a diminished snowpack. Additionally, increases in large rainfall events have been observed and are
33 predicted to increase with climate change in the twenty-first century (Murdoch et al., 2000). Increased
34 episodic acidification from these large rainfall events may offset the predicted decreasing trend in
35 snowmelt. In addition, diminished snow cover may change soil freezing patterns in winter, which are
36 dependent on the magnitude of warming. Frequent freeze-thaw behavior in soils can increase nitrate
37 leaching and raise nitrate concentrations in surface waters (Fitzhugh et al., 2003). Finally, earlier and
38 diminished snowmelt may result in lower stream flows and warmer water temperatures in the later spring
39 and summer. Warmer temperatures can result in increased growth rates. Increased aquatic growth can
40 lower nitrate concentrations through biological uptake (Sommaruga-Wogarth et al., 1997); however,
41 increased growth also can have negative impacts (e.g., hypoxia; see the section *Effects of Atmospheric*
42 *Nitrogen Deposition on Coastal Estuaries* in this chapter for further discussion). Warmer temperatures
43 and lower flows in streams may also have deleterious effects on cold water fish species (Mohseni et al.,
44 2003).

45 A nearly universal prediction from models of future climate is that droughts will increase in severity and
46 duration, even in areas such as the eastern United States, where overall increases in precipitation are
47 predicted (IPCC, 2007). This forecast has strong implications for the impacts of acidification on aquatic
48 ecosystems. When soils are dry, oxidized nitrogen and sulfur species accumulate and are released and

1 flushed into local waterbodies upon re-wetting. Several studies have shown increased concentrations of
2 nitrate and sulfate and decreased ANC values in surface waters when wet conditions return following
3 drought periods. These impacts are especially apparent in wetland-influenced watersheds (Tipping et al.,
4 2003; Watmough et al., 2004; Schiff et al., 2005). This pattern of nitrogen and sulfur storage and release
5 has further implications for the episodic acidification of waterbodies. Soils affected by acid deposition
6 often contain levels of stored nitrogen and sulfur that are equivalent to decades of atmospheric deposition.
7 Therefore, the aquatic ecosystems effects of severe, post-drought episodic acidification in lakes and
8 streams and other surface waters may persist for decades after deposition levels have declined below
9 critical loads (Tipping et al., 2003; Eimers et al., 2007). Rapid shifts from periods of dry to wet conditions
10 are also expected to increase in the future. Increases in acidification driven by such climatic shifts have
11 been shown to directly affect aquatic biological communities such as diatom algae (Faulkenham et al.,
12 2003) and to alter physico-chemical variables that affect aquatic life, such as the penetration of ultraviolet
13 radiation (Yan et al., 1996).

14 **3.5.5 Nitrogen Availability has Important Implications for the Response of Ecosystems** 15 **to Climate Change**

16 One of the most uncertain and important ecosystem-related issues regarding the interaction of the
17 atmospheric deposition of pollutants with changes in climate and CO₂ levels concerns the role of nitrogen
18 availability and its impact on growth and carbon sequestration. Growth in the majority of U.S. and global
19 ecosystems is limited by nitrogen availability, suggesting that atmospheric nitrogen deposition may be
20 acting to increase growth and carbon sequestration in many ecosystems across the United States.
21 However, in terrestrial ecosystems, excess atmospheric deposition of nitrogen also contributes to
22 ecosystem acidification, calcium depletion in soils, loss of diversity, and excess runoff of nitrate, which
23 can, in turn, lead to over-enrichment in freshwater and estuarine ecosystems. Therefore, when considering
24 atmospheric deposition of nitrogen, the potential “benefit” of increased growth and carbon sequestration
25 must be weighed against the potential “costs” of resulting nutrient over-enrichment, decreased ecosystem
26 services (e.g., biodiversity), and the promotion of invasive species (Fenn et al., 2003b). These issues are
27 brought into sharp focus by studies that seek to determine the role of nitrogen in the present and future
28 growth of terrestrial ecosystems (particularly forests) and the sequestration of carbon as climate and
29 atmospheric CO₂ concentrations continue to change.

30 One general conclusion is that net ecosystem growth in temperate and boreal forests has increased
31 coincident with climate change in the United States, across Europe, and in parts of Asia over the past few
32 decades (Goodale et al., 2002; Nemani et al. 2003; Boisvenue and Running, 2006; Magnani et al., 2007).
33 However, many local and regional exceptions to this generalization can be found (Korner, 2003). Another
34 general conclusion is that atmospheric nitrogen deposition acts to increase net ecosystem growth and
35 carbon sequestration in mid-latitude forests (Reich et al., 2006; Magnani et al., 2007), although the
36 magnitude of this nitrogen-driven carbon sink is likely much less than originally estimated by some
37 studies (Sutton et al., 2008). Recent estimates of this sink are approximately 3% to 21% of annual CO₂
38 emitted globally from fossil fuel combustion (Churkina et al., 2009; Thomas et al., 2009; Zaehle et al.,
39 2010). The interactions of carbon and nitrogen are critical in controlling the magnitude of the terrestrial
40 sequestration of a large amount of the anthropogenic (i.e., man-made) CO₂ that would otherwise reside in
41 the atmosphere and further alter the global climate. As CO₂ in the atmosphere increases, terrestrial carbon
42 sinks (especially forests) have been shown to increase; however, a point may be reached when nitrogen
43 and other nutrients become limiting (Johnson, 2006; van Groenigen et al., 2006). The role of atmospheric
44 nitrogen deposition in the global carbon cycle highlights the complex linkages among biogeochemical
45 cycles and the important link between air pollutant deposition and global climate change.

3.5.6 Dissolved Organic Carbon - Acid Deposition Interactions: A Case Study of Climate Change

Widespread increases in the concentrations and loads of dissolved organic carbon (DOC) in surface waters have been reported in the United States and Europe (Driscoll et al., 2003; Evans et al., 2006; Monteith et al., 2007). DOC is formed as organic matter decomposes and dissolves in water. DOC levels in surface waters are important because of the demonstrated link between atmospheric nitrogen and sulfur deposition and DOC loads in these waters (Clark et al., 2010). DOC plays a pivotal role in human health concerns related to water supply and distribution through the formation of disinfection by-products (Escobar et al., 2001; Chow et al., 2003). Additionally, DOC plays an important role in a wide array of aquatic ecosystem effects and interactions, including light penetration, water temperature, thermal stratification, plankton growth, pH and acidification, and the transport of trace metals (Snucins and Gunn, 2000).

The terrestrial carbon cycle plays an important role in modulating increases in atmospheric carbon dioxide concentrations and climate change.

A variety of causes have been offered to explain why DOC concentrations are changing in remote fresh waterbodies that are not greatly influenced by human land use; these causes include the following:

- Decreasing atmospheric sulfur deposition (Evans et al., 2006; Monteith et al., 2007)
- Climate warming (Worall and Burt, 2007; Clair et al., 2008)
- Changes in precipitation amount (Hudson et al., 2003; Worrall et al., 2003)
- Changes in incident solar radiation (Hudson et al., 2003)
- Decreases in sea salt deposition (Monteith et al., 2007)
- Chronic inputs of atmospheric nitrogen deposition (Findlay, 2005).

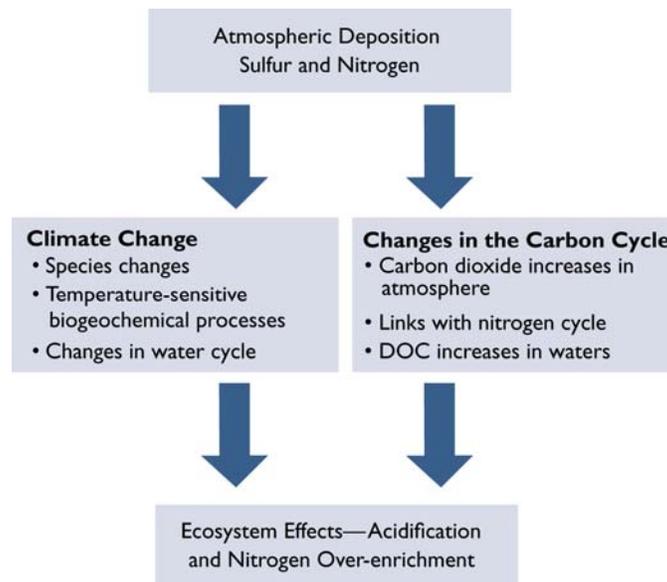
Reduced levels of sulfur deposition appear to be playing an important role in increased DOC concentrations in many surface waters in North America (Monteith et al., 2007), and the strength of this association is likely to be greatest where atmospheric deposition loads are greatest (Clark et al., 2010). Decreases in atmospheric sulfur deposition over the past 10–20 years have resulted in increasing pH and decreasing ionic strength in many waters, both of which should increase the solubility of organic matter in soils and sediment and, therefore, the DOC levels in surface waters. These observed increases in DOC concentrations may simply reflect ecosystem recovery and a return to the “natural” levels of DOC that were present before widespread acid deposition acidified aquatic ecosystems. Also, some observed DOC increases that appear to be driven by drought occurrence may actually result from the drought conditions favoring oxidation of previously reduced sulfur compounds that often originate from atmospheric deposition (Clark et al., 2006).

Climate variation also probably partly explains increasing surface-water DOC trends in some regions (Hudson et al., 2003; Worrall et al., 2003). Fundamentally, warmer soil and sediment temperatures should increase the decomposition rate of organic matter and DOC levels in waters. However, variations in moisture, nutrient availability, the availability of dissolved oxygen, and other variables would be expected to mask a simple DOC to temperature relationship (Giardina and Ryan, 2000). For example, the Arctic regions of North America have experienced some of the greatest warming trends on Earth. Climate warming is expected to significantly increase DOC concentrations, and fluxes in Arctic surface waters (Clair and Ehrmann, 1998; Frey and Smith, 2005). However, data from the Yukon basin supports decreased DOC export, which may result from the conversion of soil carbon to CO₂ in the active soil permafrost layer (Striegl et al., 2005). Also, research conducted in Canadian lakes has highlighted the important role of other climate-related factors (e.g., drought-wetting cycles, variations in solar radiation) on surface water DOC concentrations, (Dillon and Molot, 1997; Schindler, 1998; Hudson et al., 2003).

1 Regardless of the relative roles of climate change and changes in atmospheric deposition of sulfur and
 2 nitrogen in driving these recent patterns of increased DOC concentrations and loads in surface waters, this
 3 issue highlights the interplay of acid deposition and climate change. Increases in DOC concentrations
 4 generally result in decreases in ANC; therefore, these increased DOC concentrations are likely slowing
 5 the rate of recovery of aquatic ecosystems as inferred from measurements of ANC (Monteith et al., 2007).
 6 Because of the widespread importance of DOC for aquatic ecosystems, future monitoring and modeling
 7 efforts should continue to scrutinize and consider the interplay of climate and atmospheric deposition.
 8 Historically, the recognition and study of DOC trends was not much discussed or considered until data
 9 from long-term monitoring networks, such as the LTM and TIME programs in the United States and other
 10 programs across the world, showed the magnitude and widespread geographic nature of these trends.

11 3.5.7 Summary of Climate Change – Atmospheric Deposition Linkages

12 The results summarized here provide a strong scientific basis for joint consideration of climate change
 13 and air quality policies affecting atmospheric nitrogen and sulfur deposition. Topics considered here
 14 under the umbrella of global climate change include past, ongoing, and future expected changes in a wide
 15 range of factors that affect ecosystems, such as warmer air temperatures, water availability, and the
 16 frequency of large magnitude events (e.g., droughts, floods, fires). The combination of patterns in the
 17 atmospheric deposition of sulfur and nitrogen with these global climate change factors has implications
 18 for the growth of ecosystems, expected shifts of species, and the acid/base chemistry of surface waters.
 19 Untangling the interactions among multiple biogeochemical cycles and the resulting ecosystem responses
 20 to changes in these cycles is complex and uncertain. For example, global climate change generally
 21 provides an additional set of interacting stressors in ecosystems that are negatively impacted by acid
 22 deposition (see **Figure 3-2**). However, climate change also may increase rates of chemical weathering in
 23 some locations and, therefore, ecosystems may be able to sustain greater amounts of sulfur or nitrogen
 24 deposition before experiencing negative effects. Conversely, atmospheric nitrogen deposition may
 25 currently be limiting global warming through enhanced ecosystem carbon sequestration in U.S. regions
 26 that are only lightly to moderately impacted by air pollutant deposition.



27
 28 **Figure 3-2. Although the rate of atmospheric deposition of sulfur and nitrogen are the principal**
 29 **drivers of the ecosystem effects discussed in this chapter, ongoing and future changes in climate**
 30 **and the carbon cycle are expected to interact with these ecosystem effects in ways that are**
 31 **currently difficult to predict. Ongoing research efforts are helping to unravel the role of climate**
 32 **change on ecosystems so that more accurate model forecasts can better constrain the role of**
 33 **climate and carbon (Prepared by USGS).**

1 Currently, the degree of uncertainty regarding many of the predicted effects of global climate change on
2 ecosystems is high and often not well quantified. Environmental monitoring programs are helpful in
3 understanding climate-change effects on ecosystems, but often are not adequate in geographic scope or in
4 the number of parameters measured to fully understand these effects. For example, the leading programs
5 that measure atmospheric deposition (NADP and CASTNET) and surface-water chemistry (LTM and
6 TIME) generally do not measure many climatically relevant variables, such as snow-water equivalents
7 and air and water temperature. Additionally, several of the papers reviewed here highlighted the critical
8 need for more multi-factorial and interdisciplinary experiments to test the effects of varying air/soil/water
9 temperature, CO₂ concentrations, and pollutant deposition levels on ecosystems. There is also a need for
10 consideration of climate-change effects in acidification and nitrogen effects models, such as MAGIC and
11 PnET-BGC, as they are used to provide predictions of the expected impacts of future air pollutant
12 deposition scenarios. Work of this type is ongoing, but not yet widely available in the published literature.
13 Evaluations of the ability of these models to incorporate changing climatic effects would also be helpful
14 in providing feedback to discussions about future policies. Finally, past and future disturbances that are
15 not necessarily climate-change driven (e.g., land disturbance, land use changes, invasive species, fire) can
16 have ecosystem effects that are as great or greater than those derived from climate change and are also
17 worthy of consideration in future scenario modeling.

18 **3.6 Multi-Pollutant Interactions in Ecosystems**

19 Throughout this chapter, the interacting effects of nitrogen and sulfur deposition have been discussed in
20 reference to the acidification and nutrient over-enrichment of ecosystems. In the previous section, the
21 interacting effects of acid deposition and changes in the carbon cycle and climate change were discussed.
22 There are also important interactions between acid deposition and the ecosystem effects of ozone and
23 mercury, and these interactions will be discussed in this section. In recent years, interest has been
24 increasing in development of air quality policies that target multiple pollutants (NRC, 2004; Brook et al.,
25 2009; NARSTO, 2010). This section focuses on the ecosystem aspects of two pollutants (i.e., ozone and
26 mercury) for which clear links have been demonstrated.

27 **3.6.1 Ozone Interactions with Atmospheric Sulfur and Nitrogen Deposition**

28 Ozone is taken up by plants and, in high concentrations, can directly damage plant cell membranes,
29 reduce the rate of photosynthesis and growth, and cause species shifts in forests (Karnosky et al., 2007).
30 In short, ozone is yet another pollutant stressor that can interact with sulfur and nitrogen deposition and
31 changes in climate and CO₂ concentrations to affect ecosystem health (McLaughlin and Percy, 1999).
32 Ozone concentrations and trends are highly variable across the globe and the United States, but leveling
33 or slight decreases in concentrations in North America have been noted in recent years (Vingarzan, 2004;
34 Oltmans et al., 2006). Trends in ozone concentrations are affected by trends in (1) the sources of ozone,
35 such as fossil fuel use, (2) the principle atmospheric precursors of ozone (i.e., NO_x, volatile organic
36 compounds, and carbon monoxide), and (3) climatic factors, particularly summer stagnation episodes that
37 favor the highest concentrations. Results from models that simulate likely twenty-first century climate,
38 combined with chemical transport model outputs, indicate a likelihood of increased future ozone
39 concentrations in the United States (Jacob and Winner, 2009). Additionally, tropospheric ozone (ozone in
40 lowest portion of the Earth's atmosphere) provides the third-strongest warming influence of all
41 greenhouse gases (Mickley et al., 1999), and recent work suggests that ozone also contributes indirectly to
42 warming by slowing growth, and subsequently CO₂ uptake, in terrestrial ecosystems (Sitch et al., 2007).

43 Few studies have simultaneously considered the ecosystem effects of ozone combined with sulfur and
44 nitrogen deposition. The added complication of ongoing changes in the climate and the carbon cycle
45 provide additional challenges for understanding patterns and predicting likely ecosystem impacts of
46 ozone, sulfur, and nitrogen loading. The inability of current models to adequately consider the
47 simultaneous effects of these pollutants has been noted (Aber et al., 2001; Dewar et al., 2009). Despite the

1 lack of adequate models and sufficient experiments to understand multi-factorial variations of these
2 pollutants, evidence supports a joint, interacting role of ozone and nitrogen deposition in a variety of
3 ecosystem effects, including sensitivity to insects and pathogens; frost sensitivity; drought; and fire
4 (Bytnerowicz et al., 2007). Thresholds of nitrogen deposition and ozone concentration likely exist, below
5 which ecosystem effects cannot be detected and at which benefits such as increased ecosystem growth
6 and carbon sequestration are likely, although the interactive effects of these pollutants on ecosystems are
7 not well known (Aber et al., 2001). However, the exact levels of these thresholds are not well known, nor
8 are the tipping points above which negative ecosystem effects sharply increase. Better identification of
9 such patterns in ecosystem effects could provide vital information for use in future air quality policies.

10 **3.6.2 Mercury Interactions with Atmospheric Sulfur and Nitrogen Deposition**

11 Although most surface waters have very low concentrations, mercury is of environmental concern
12 because it accumulates in living cells and is biomagnified in aquatic and terrestrial food webs to the
13 extent that elevated mercury concentrations are present in fish throughout the United States (Scudder et
14 al., 2009). Mercury is a potent neurotoxin, and human health warnings that suggest limiting the
15 consumption of certain fish due to high mercury levels are widespread in the United States (U.S. EPA,
16 2007a). Atmospheric sulfur and nitrogen are closely linked to mercury because emissions from coal
17 burning are a major source of all three of these pollutants. Moreover, atmospheric sulfur and nitrogen
18 deposition can affect the cycling and bioaccumulation of mercury in ecosystems, primarily because of the
19 interactions of mercury and sulfur.

20 The mercury found in the tissue of biota at high ecosystem trophic levels (e.g., predator fish and birds) is
21 dominantly in the methyl form (methylmercury), and this is the dominant form found in humans as well.
22 Methylation is the biogeochemical process by which mercury is converted to methylmercury, and most
23 methylation in ecosystems is believed to be carried out by the same bacteria that convert sulfate to sulfide
24 forms. This process tends to proceed in environments (e.g., wetlands, lake-bottom sediments) where
25 sulfate is introduced in runoff and oxygen is absent. Several studies have shown that additions of sulfate
26 increase methylmercury concentrations in waters, and by inference, would be expected to also increase
27 mercury levels in biota (Branfireun et al., 1999; Gilmour et al., 1992; Jeremiason et al., 2006). Similar
28 studies have shown that decreases in sulfate concentrations can likewise decrease methylmercury
29 concentrations in waters and fish tissue, even in the absence of any changes in atmospheric mercury
30 deposition rates (Hrabik and Watras, 2002; Drevnick et al., 2007).

31 The findings from these studies suggest that improvements in ecosystem health, with respect to mercury
32 bioaccumulation levels, might be achieved by simultaneously decreasing sulfur and mercury deposition
33 levels. These findings also suggest that a multi-pollutant policy, considering both mercury and sulfur,
34 could be used strategically to reduce mercury bioaccumulation in many environmental settings. In
35 addition to direct links between methylmercury and sulfate, studies have found that pH and mercury
36 levels in fish tissue are inversely related in many regions. This suggests that ecosystem acidification and
37 mercury bioaccumulation are also linked, and a variety of mechanisms have been suggested to explain
38 this relationship (Kamman et al., 2004; Munthe et al., 2007; Wiener et al., 2006; Scudder et al., 2009).
39 Another recent study shows that high nitrate concentrations in waters may act to suppress the formation of
40 methylmercury when environmental conditions might otherwise favor high rates of methylation
41 (Todorova et al., 2009), but this type of interaction has not yet been demonstrated in a natural ecosystem
42 where atmospheric deposition is the principal source of nitrogen and sulfur.

4. Beyond Title IV – Ecological Impacts of Further Emission Reductions

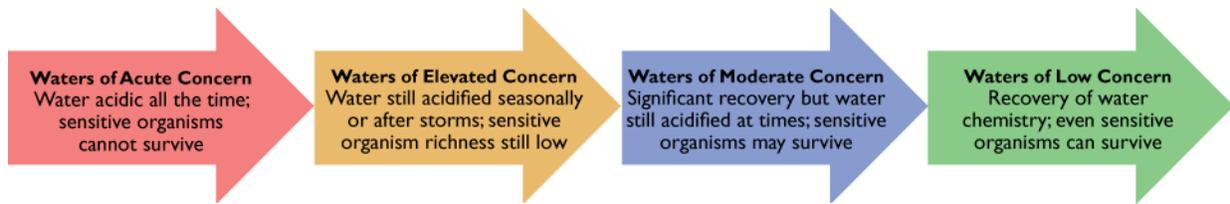
Title IX of the 1990 CAAA requires NAPAP to report quadrennially on “the reduction in deposition rates that must be achieved in order to prevent adverse ecological effects” (Public Law 101-549-Nov. 15, 1990). NAPAP presented a working definition of “adverse ecological effects” in the 1996 NAPAP RTC (NSTC, 1998) based on the intent of Congress, as expressed in the 1990 CAAA and shaped by other relevant environmental statutes (i.e., Comprehensive Environmental Response, Compensation and Liability Act, and the Clean Water Act) and associated regulations. The definition is as follows:

Adverse ecological effects: any injury (i.e., loss of chemical or physical quality or viability), to any ecological or ecosystem component, up to and including at the regional level, over both long- and short-terms.

A broad consensus of scientists studying acid deposition and ecosystem recovery have published reports since publication of the last NAPAP report (NSTC, 2005) indicating that further emission reductions beyond those achieved by Title IV are necessary to allow sensitive forests and aquatic ecosystems to recover from acidification. The SO₂ and NO_x emission reductions achieved under Title IV from the power sector are now recognized as significant, but not enough to achieve full recovery or to prevent further acidification in the eastern United States. Modeling analyses demonstrate that further reductions in SO₂ and NO_x emissions from the power generation sector are needed to reduce the amount of acidic deposition in sensitive ecosystems and result in healthier forests and fewer acidic lakes and streams.

Adverse impacts to ecological processes or ecosystem components include the results of reductions in ANC, pH, and increases in aluminum concentrations in a lake or stream; loss of fish and other biota; loss of important nutrients, such as calcium, from forest soils; and increased susceptibility of trees to pests, disease, and winter temperatures. These, in turn, lead to decreasing forest growth and forest dieback. Adverse ecological impacts also include the effects of nitrogen saturation in forests, alpine lakes, and coastal eutrophication as a result of atmospheric deposition, reductions in biodiversity, fire regime shifts, and injuries to plants as a result of ozone exposure. Other effects of impaired air quality addressed by NAPAP (e.g., materials, visibility, human health) are not considered here as this chapter focuses on ecological effects.

Deposition levels that correlate with a “threshold” of adversity are scientifically complex and can be difficult to establish because most biological responses to changes in acid-base chemistry occur along a continuum with no single value or set of chemical concentrations that represents a threshold for “significant adverse biological effects” (Bulger et al., 1999; Sullivan et al., 2006; U.S. EPA, 2008a, 2009g). Therefore, this report describes ecosystem responses along a continuum, enabling decision makers to determine levels of acceptable risk. The analysis in this chapter summarizes the results into classes of acidification concerns, and these classes describe the path that lakes and streams follow in recovery from acidic precipitation (**Figure 4-1**) as indicated by changes in water chemistry and inferred biological response.



Note: See Chapters 2 and 3 for a full discussion of the ecological effects of acidification.
 Waters of acute concern are defined as having ANC < 0 µeq/L.
 Waters of elevated concern are defined as having ANC between 0-50 µeq/L.
 Waters of moderate concern are defined as having ANC between 50 and 100 µeq/L.
 Waters of low concern are defined as having ANC >100 µeq/L.

Figure 4-1. Stages of recovery for acidic lakes and streams (Prepared by U.S. EPA).

NAPAP presented a working definition of “adverse ecological effects” in the 1996 NAPAP RTC (NAPAP, 1998) based on the intent of Congress, as expressed in the 1990 CAAA and shaped by other relevant environmental statutes (Comprehensive Environmental Response, Compensation and Liability Act, and the Clean Water Act) and associated regulations. The definition is as follows:

Adverse ecological effects: any injury (i.e., loss of chemical or physical quality or viability), to any ecological or ecosystem component, up to and including at the regional level, over both long- and short-terms.

Adverse impacts to ecological processes or ecosystem components include the results of reductions in ANC, pH, and an increase in aluminum concentrations in a lake or stream: loss of fish and other biota; loss of important nutrients, such as calcium, from forest soils; and increased susceptibility of trees to pests, disease, and winter temperatures. These, in turn, lead to decreasing forest growth and forest dieback. Adverse ecological impacts also include the effects of nitrogen saturation in forests, alpine lakes, and coastal eutrophication as a result of atmospheric deposition, reductions in biodiversity, fire regime shifts, and injuries to plants as a result of ozone exposure. Other areas addressed by NAPAP (i.e., materials, visibility, human health) would follow the same definition, but are not considered here as ecological effects. This chapter is based on the above definition of adverse ecological effects and uses the same approach of investigating ecosystem responses along a continuum. This chapter focuses on the associated effects and recovery from acidic deposition, as indicated by changes in water chemistry and inferred biological response.

4.1 Recent Assessments

Although the definition of a “threshold” is complex, a significant amount of research in the past decade indicates that ecosystems continue to be affected by acid deposition. Two recent syntheses of the science found that, for the near term, sulfur deposition still is the primary source of acidification in most sensitive areas of eastern North America (U.S. EPA, 2008a, 2009g). However, these syntheses also found that nitrogen deposition, particularly ammonia, is playing a greater role in both short- and long-term acidification of lakes and streams as ammonia is nitrified to nitrate, which can acidify soils and surface water when the amount exceeds biological uptake. Under certain deposition levels, sulfur and nitrogen could have approximately equal roles in surface water acidification. Decreases in emissions of SO₂ and increases in the level of nitrogen saturation of forest soils have contributed to the increasing role of nitrogen in surface-water acidification in the eastern United States.

In the period since the 2005 NAPAP RTC, various studies (Warby et al., 2005; Burns et al., 2006a; Driscoll et al., 2007; Sullivan et al., 2007; Lawrence et al., 2008; Robinson et al., 2008) concluded that current acid deposition loads are causing ecological damage in sensitive lakes and streams in the eastern United States. The results presented in Chapter 2 of this report, as well as studies focused on specific regions of the country, demonstrate improving water quality in most of the Adirondack Mountains, New

1 England, and the Northern Appalachian Plateau. However, many lakes and streams still remain impacted
2 by acid deposition under current deposition levels (U.S. EPA, 2007b, 2008b). For example, the extent of
3 stream acidification in the western Adirondack Mountains remains high. The streams sampled in a recent
4 study showed that 66%, or 718 km, of streams are prone to acidification and likely have levels of acidity
5 harmful to their biota. Of the 66% of streams found to be prone to acidification, about half were likely to
6 be chronically acidified (i.e., base cation surplus < 0 µg/L), with the other 50% episodically acidified (i.e.,
7 base cation surplus > 25 µg/L) (Lawrence et al., 2008).

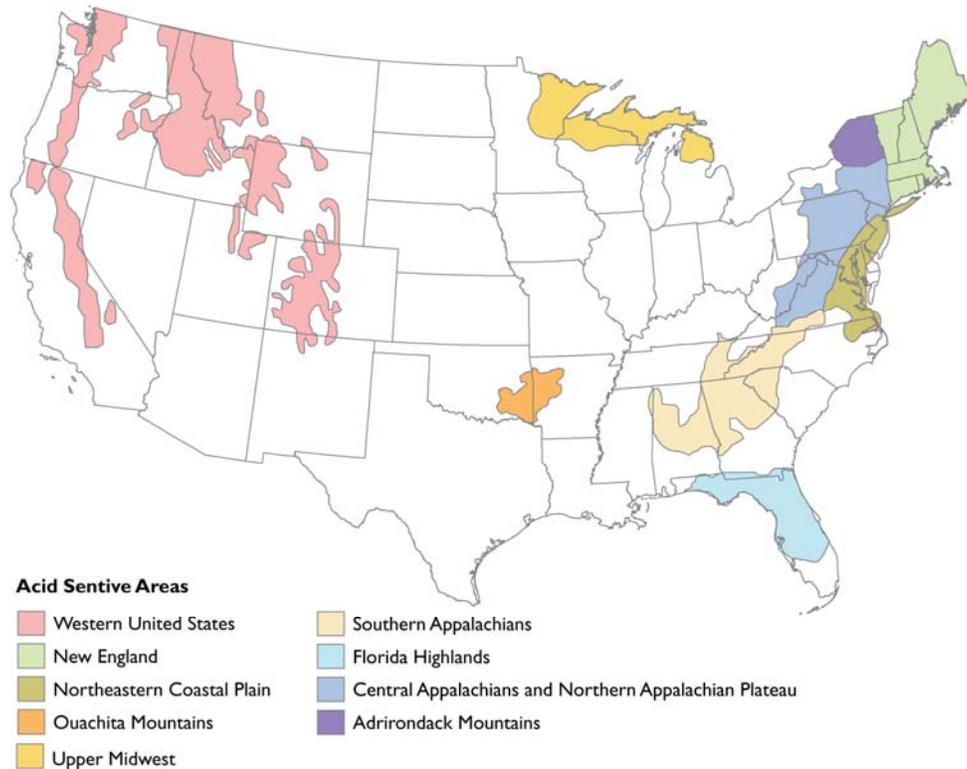
8 A major assessment of acid deposition and its effects in the Adirondack Mountains found that full
9 implementation of the 1990 CAAA will result in only modest recovery in northeastern lakes and streams
10 impacted by acid deposition (Sullivan et al., 2006). However, based on model projections, this study also
11 concluded that aggressive reductions of SO₂ and NO_x emissions from power generation of up to 70% and
12 50%, respectively, beyond 2001 levels, would allow for chemical recovery (i.e., become nonacidic) of
13 most lakes in the Adirondack Mountains.

14 Researchers in the southern Appalachian Mountains concluded that streams in this region are still
15 threatened by acid deposition (Sullivan et al., 2007). In particular, many brook trout streams in Virginia
16 are still vulnerable to acidification at current deposition levels (Cosby et al., 2006). Based on model
17 projections from two recent studies (Cosby et al., 2006; Sullivan et al., 2007), researchers concluded that
18 further reductions of sulfate deposition beyond levels achieved by the Title IV SO₂ emission reductions
19 are necessary to prevent further acidification of southern Appalachian Mountain streams and to allow
20 currently impacted streams to recover in the region. Cosby et al. (2006) concluded that a moderate
21 reduction of sulfur and nitrogen deposition of 50% and 22%, respectively, beyond Title IV produced a
22 small improvement in stream water quality over the long term (by 2100) relative to current conditions for
23 brook trout streams in Virginia. A more stringent scenario of 62% reduction for sulfur and 30% reduction
24 for nitrogen deposition produced additional improvements in stream water quality over the long term (by
25 2100), although these reductions still did not return stream water quality to preacidification conditions
26 (Cosby et al., 2006). In sensitive streams in North Carolina, Tennessee, and South Carolina, Sullivan et
27 al., (2007) modeled similar emission reductions, as did Cosby et al. (2006) and found little future
28 improvement in stream conditions resulting from moderate and aggressive emission controls (Sullivan et
29 al., 2007).

30 **4.2 Analysis of the Environmental Impact of Further Air Emission Reductions**

31 In response to the CAAA Title IX statutory requirement to identify deposition rates that would prevent
32 adverse ecological effects, this chapter analyzes several emission-reduction scenarios that broadly bound
33 the range of reductions presented in the literature discussed above and are similar to scenarios modeled in
34 past NAPAP reports. These results provide an indication of the environmental improvements that would
35 be expected from additional emission reductions from sources affected by Title IV and other emission-
36 reduction programs. These environmental improvements do not constitute full recovery in all areas of
37 acid-sensitive forests, lakes, or streams that have been impacted by acid deposition. They do provide,
38 however, an indication of the scope and magnitude of the impact of emission reductions on deposition
39 levels and on acid-sensitive ecosystems (**Figure 4-2**). Other sensitive ecosystems, such as estuaries and
40 western U.S. alpine lakes, would also benefit from reductions in nitrogen deposition, but effects on those
41 ecosystems are not analyzed here. In addition, climate change may alter the response of lake and stream
42 water quality to declining acidic deposition (see Chapter 3 for a discussion of climatic change and
43 surface-water recovery). However, this modeling analysis, like most previous scientific studies of surface-
44 water recovery from acidification, assumes a constant influence of climate over the simulation period.
45 While the information presented in this chapter is inform future actions, the analysis presented here
46 focuses solely on ecological recovery. Other aspects of implementing these emission reduction scenarios,
47 such as the costs and additional benefits (e.g., human health), were not analyzed and are beyond the scope
48 of this assessment. Additional information would be important to inform future actions, including the

1 costs and other impacts of emission reductions from the power sector and other sectors and the value that
 2 the public places on further improvements to the environment and human health.



3
 4 **Figure 4-2. Map of acid-sensitive ecosystems in the United States (Prepared by U.S. EPA).**

5 **4.2.1 Scenarios Analyzed**

6 This modeling effort analyzed several scenarios representing emission changes from stationary sources
 7 within the power sector for the purpose of evaluating ecological recovery. Results are presented for total
 8 sulfur and total nitrogen deposition because these pollutants are the primary components of acid
 9 deposition and the cause of anthropogenic surface-water acidification. This analysis focuses on reductions
 10 from the power generation sector (i.e., EGUs); however, many other sources also emit sulfur and nitrogen
 11 pollutants that contribute to acid deposition. For example, sources outside of the power generation sector
 12 are projected to emit approximately 52% of the SO₂ and 84% of the NO_x emitted in 2020 under the Base
 13 Case scenario described below (U.S. EPA, 2006b).

14 The following analysis compares emission and deposition levels under a Base Case scenario with three
 15 other sensitivity scenarios of various levels of additional reductions. The Base Case scenario consists of
 16 currently implemented programs and programs that were finalized as of spring 2005. These programs
 17 include Title IV, CAIR, and Tier II and Heavy Duty Diesel SO₂ and NO_x reductions (projected to take
 18 effect by 2010). The analysis employs models and analytical tools that are widely used and peer reviewed.
 19 EPA's Response Surface Model (RSM) was used to estimate future deposition loads (U.S. EPA, 2006b).
 20 RSM is based on an air quality modeling approach known as meta-modeling that aggregates numerous
 21 individual air quality modeling simulations into a multidimensional air quality response surface. The
 22 Integrated Planning Model (IPM) was used to estimate the power sector emissions from EGUs. As with
 23 any detailed analysis of complex scenarios, the results presented in this report are subject to uncertainties
 24 concerning emission estimates, air quality modeling, deposition projections, and the impact of emission
 25 reductions on ecological systems. See the text box titled *Modeling Tools Used in this Analysis* for a more
 26 detailed description of the air quality and emission models used in this analysis.

The scenarios described below represent emission levels for each pollutant in 2020. Because the Base Case scenario includes implementation of CAIR, resulting in significant SO₂ and NO_x emission reductions beyond levels achieved by Title IV alone, the levels of SO₂ and NO_x modeled for the other future scenarios include additional reductions from the CAIR SO₂ and NO_x cap levels. The projected emissions used for the air quality modeling in 2020 are somewhat higher than the cap levels for all scenarios as a result of the early reductions and allowance banking predicted by the emission model (i.e., IPM). For the mobile sectors, the growth and controls are calculated together by the MOBILE6 and NONROAD models (U.S. EPA, 2006b). No controls were applied to the Canadian, Mexican, or offshore emission inventories. Additional details on the scenarios are presented below.

- **Base Case Scenario (2020).** This scenario includes rules that were finalized as of spring 2005, including CAIR and the Non-Road Diesel Rule. It does not include new or anticipated actions under the CAA or other emission reductions that would be necessary to attain and maintain the fine particle and ozone NAAQS for which states are required to submit SIPs or to achieve regional haze-reduction goals.
- **Scenario A.** This scenario includes an additional 60% reduction in SO₂ emissions from the power generation sector beyond the Base Case scenario, resulting in an annual SO₂ emission level of 1.75 million tons/year in 2020. It also includes national annual NO_x emission reductions from the power generation sector of 68% beyond the Base Case scenario, or an annual emission level of 0.7 million tons/year of NO_x in 2020.
- **Scenario B.** This scenario includes an additional 75% reduction in SO₂ emissions from the power generation sector beyond the Base Case scenario, equaling annual SO₂ emissions of 1.10 million tons/year in 2020. It also includes national annual NO_x emission reductions from the power generation sector of 80% beyond the Base Case scenario, or annual NO_x emissions of 0.44 million tons/year in 2020.
- **Scenario C.** This scenario is roughly equivalent to elimination of SO₂ emissions from the power generation sector. It includes an additional 90% reduction in SO₂ emissions from the power generation sector beyond the Base Case scenario, resulting in 0.44 million tons SO₂/year in 2020, and a 50% reduction in SO₂ emissions from nonpower generation sources (e.g., industrial boilers). The scenario also includes national annual NO_x emission reductions of approximately 80% beyond the Base Case scenario for the power sector, resulting in annual NO_x emissions of 0.44 million tons/year in 2020 (see **Figure 4-3**).

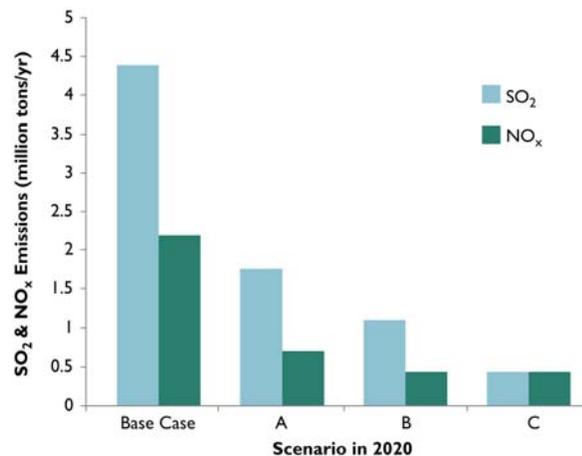


Figure 4-3. Projected annual SO₂ and NO_x emission levels in 2020 for the Base Case scenario and Scenarios A through C. Emission levels are for the continental United States (Prepared by U.S. EPA).

Modeling Tools Used in this Analysis

Air Quality and Deposition Models—Air quality models use mathematical and numerical techniques to simulate the physical and chemical processes that affect air pollutants as they disperse and react in the atmosphere. Using inputs of meteorological data and source information such as emission rates, these models are designed to characterize primary pollutants that are emitted directly into the atmosphere and, in some cases, secondary pollutants that are formed as a result of complex chemical reactions within the atmosphere.

- The **Community Multi-scale Air Quality (CMAQ)** model is a three-dimensional, regional grid-based air quality model designed to simulate air quality deposition over the contiguous United States for a period of 1 year. The CMAQ model includes state-of-the-science capabilities for conducting urban- to regional-scale simulations of multiple air quality issues, including tropospheric ozone, fine particles, toxics, acid deposition, and visibility degradation. The CMAQ model is a publicly available (supported by the Community Modeling and Analysis System [CMAS] Center; <http://www.cmascenter.org>), peer-reviewed, state-of-the-science model consisting of a number of science attributes that are critical for simulating the oxidant precursors and nonlinear organic and inorganic chemical relationships associated with the formation of sulfate, nitrate, and organic aerosols.
- The **Response Surface Model (RSM)** is based on an approach known as air quality meta-modeling that statistically links pollution emissions and air quality/deposition derived from other models, such as the CMAQ model. The RSM aggregates numerous individual air quality modeling simulations from the CMAQ model to produce a multidimensional air quality response surface, which can be used to predict how emission changes affect air quality. The RSM approach allows for the evaluation of how emission-control scenarios improve air quality across the United States. The RSM model was used in support of the Regulatory Impact Analysis (RIA) for the proposed NAAQS for PM_{2.5} (U.S. EPA, 2006a).

The **Integrated Planning Model (IPM)** is used to analyze the projected impact of environmental policies on the electric power sector in the 48 contiguous states and the District of Columbia. IPM is a multiregional, dynamic, deterministic linear programming model of the U.S. electric power sector. It provides forecasts of least-cost capacity expansion, electricity dispatch, and emission-control strategies for meeting energy demands and environmental, transmission, dispatch, and reliability constraints. IPM can be used to evaluate the cost and emission impacts of proposed policies to limit emissions of SO₂, NO_x, CO₂, and mercury from the electric power sector.

Water/Watershed Modeling—The Model of Acidification of Groundwater in Catchments (MAGIC) was developed to estimate acidification of lakes and streams in response to sulfur and nitrogen deposition (Cosby et al., 1985a,b,c; U.S. EPA, 2006b). MAGIC was the principal model used by NAPAP to estimate future damage and recovery to lakes and streams in the eastern United States in the 1998 and 2005 integrated assessments (NSTC, 1998 and 2005). The model simulates soil solution chemistry and lake and stream chemistry to predict the monthly and annual average concentrations of the major ions in these waters. MAGIC consists of (1) a submodel in which the concentrations of major ions are assumed to be governed by simultaneous reactions involving sulfate adsorption, cation exchange, dissolution-precipitation-speciation of aluminum, and dissolution-speciation of inorganic carbon; and (2) a mass balance submodel in which the flux of major ions to and from the soil is assumed to be controlled by atmospheric inputs, chemical weathering, net uptake and loss in biomass, and losses to runoff. Nitrogen retention and loss is modeled in two ways. . The simpler method (used for the majority of the analysis presented in this report) assumes that the percentage of nitrogen deposition retained by the soil remains constant over time. The more complex approach simulates nitrogen saturation and links net immobilization of nitrogen to the carbon/nitrogen ratio of the soil organic matter pool. Both require specification of net nitrogen uptake in vegetation, rate of denitrification, and nitrogen fixation. At the heart of MAGIC is the size of the pool of exchangeable base cations in the soil. As the fluxes to and from the pool change over time because of changes in atmospheric deposition, the chemical equilibria between soil and soil solution shift to give changes in surface water chemistry. MAGIC provides a widely accepted tool for modeling the response of lake and stream chemistry to sulfur and nitrogen deposition.

4.2.2 Changes in Sulfur Deposition

All future control scenarios modeled in this analysis are projected to lead to significant regional reductions in sulfur deposition as compared to projected conditions under the Base Case scenario in 2020 (**Figure 4-4**). Modeling of the Base Case scenario indicates that implementation of current rules in 2020 is expected to reduce sulfur deposition from 2010 levels by greater than 5 to 10 kilograms/hectare/year (kg/ha/yr) for much of the eastern United States. Under Scenario A in 2020, reductions in sulfur deposition of 10% to 20% beyond 2020 Base Case levels would be found in much of the Plains states and Northeast, with reductions of 30% to 45% in the Midwest, Northern Appalachian Plateau, central Appalachians, and southern Appalachians stretching from Indiana, Ohio, and Pennsylvania to Georgia and Alabama. The western states, from the Rocky Mountains west, would see a mixture of reductions and increases in deposition of about 10% under Scenario A (**Figure 4-5**).

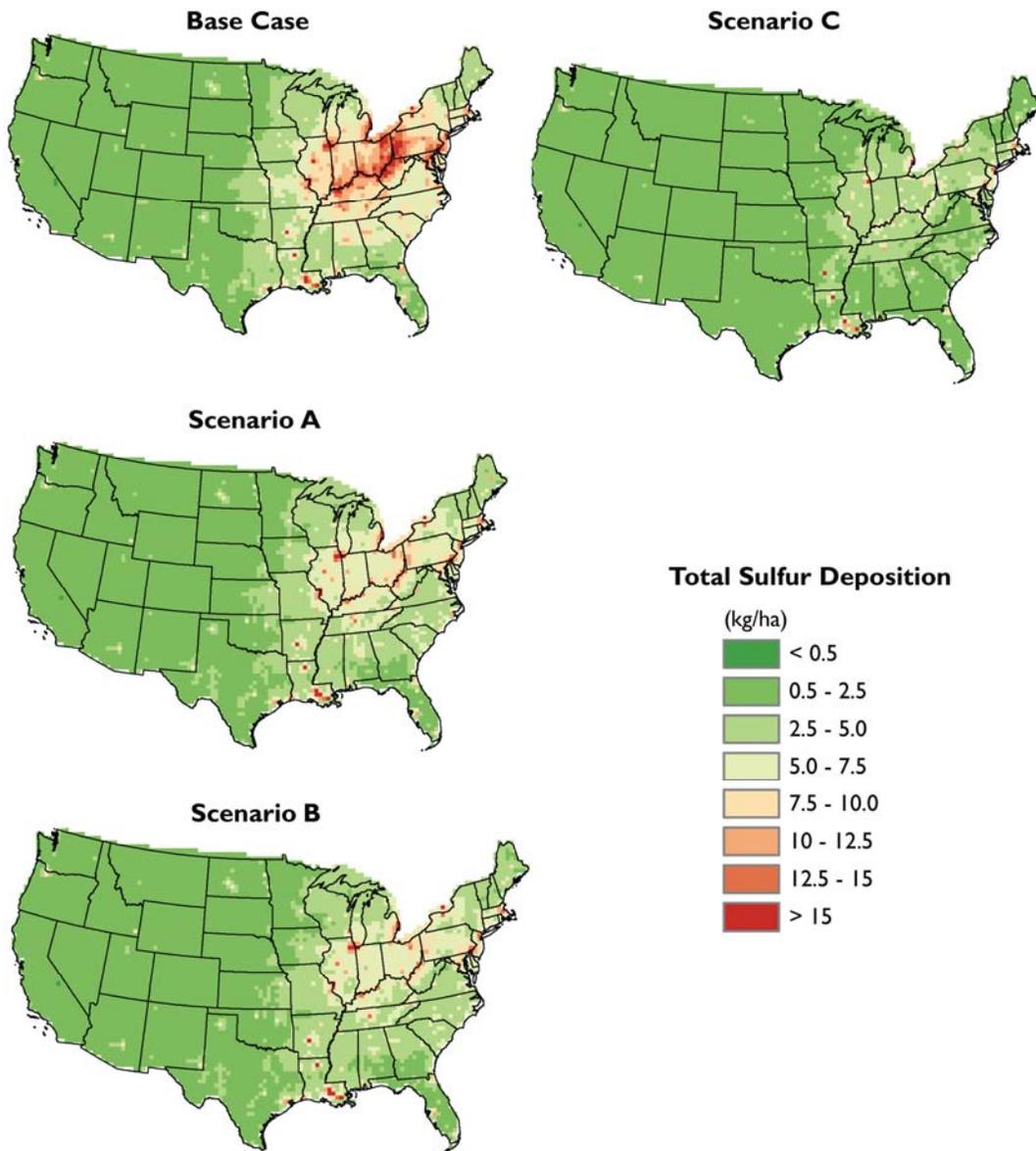
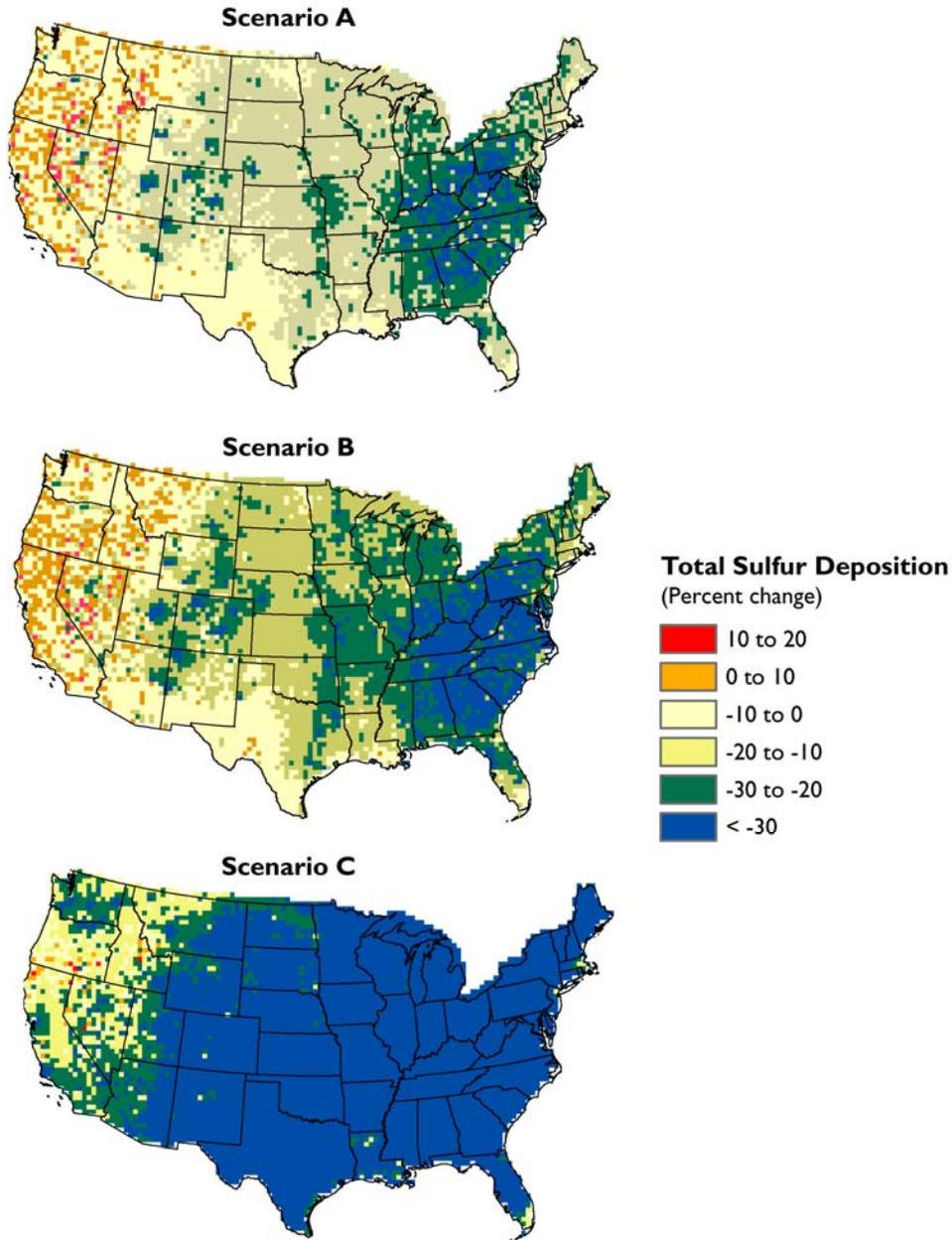


Figure 4-4. Projected annual total sulfur deposition for the Base Case scenario in 2020 and Scenarios A through C in 2020, with additional SO₂ and NO_x emission reductions. Deposition is reported in kg/ha/yr (Prepared by U.S. EPA).



1
 2 **Figure 4-5. Projected percentage changes in annual total sulfur deposition from the Base Case**
 3 **scenario in 2020, with the additional SO₂ and NO_x emission reductions in Scenarios A through C**
 4 **(Prepared by U.S. EPA).**

5 Scenario B extends the area of largest reduction in sulfur deposition southward and westward. For
 6 example, under Scenario B, the central Appalachians, Northern Appalachian Plateau, southern
 7 Appalachians, and Piedmont regions would all experience reductions in sulfur deposition beyond levels
 8 achieved by the 2020 Base Case scenario. Scenario C would extend the area with the greatest reductions
 9 in sulfur deposition beyond the 2020 Base Case scenario northward to Maine and westward through the
 10 Midwest across the Plains states to the Rocky Mountains (**Figure 4-5**), with some areas in the eastern
 11 United States experiencing reductions up to 60%. These reductions are expected to provide ecological
 12 benefits to these acid-sensitive regions, but may not solve all ecological problems related to acid
 13 deposition in those areas.

4.2.3 Changes in Nitrogen Deposition

Modeling of the 2020 Base Case scenario indicates that, in 2020, rules currently in effect are expected to achieve reductions in nitrogen deposition of 2.5 to 5 kg/ha/yr beyond 2010 levels across the United States (**Figure 4-6**). Under Scenario A, additional reductions in deposition of 10% to 20% beyond the levels seen under the 2020 Base Case scenario would occur in much of the central Appalachians and the eastern Ohio River Valley stretching from northern Pennsylvania to southern Virginia (**Figure 4-7**). Similar levels of deposition reductions would be found in the Four Corners region (i.e., Arizona, Colorado, New Mexico, Utah) of the western United States and along the Front Range of the Rocky Mountains. The model results suggest a slight increase in deposition in east Texas, western Arkansas, and eastern North Carolina. In addition, west of the Rocky Mountains, some areas would experience nitrogen deposition increases of up to 10%, including areas in Montana and Idaho and the Sierra Nevada and San Bernardino mountains of California. Increases in these regions are a result of higher NO_x emissions from other sources, such as non-EGU industrial sources. The rest of the country would see additional reductions in deposition of up to 10% beyond the levels seen under the 2020 Base Case scenario.

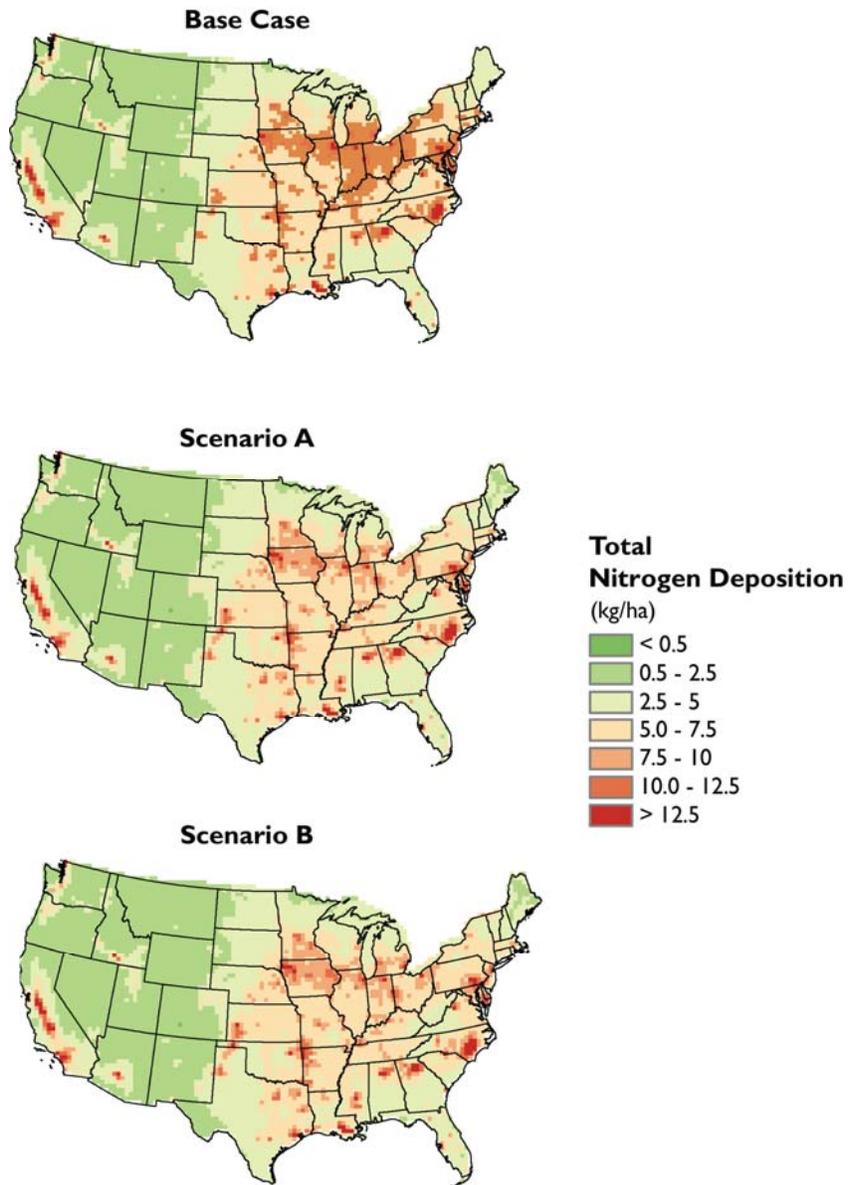
Scenario B extends the area of 10% to 20% additional reductions in nitrogen deposition to the upper Midwest and parts of the Northeast, along the Colorado Front Range, and in the Four Corners region, with regions in West Virginia experiencing up to 30% reductions. Large portions of the United States, including the Plains states, Midwest, and Western states, will experience minimal reductions in nitrogen deposition of 0% to 10%. However, areas in Montana, Idaho, and California will continue to have increases in deposition of up to 10% (**Figure 4-5**). Emission levels in Scenario B would also lead to significant additional reductions in nitrogen deposition compared to the 2020 Base Case scenario in sensitive ecosystems still experiencing water quality or forest health problems as a result of acidification. For example, the Adirondack Mountains would receive a 20% reduction in deposition under Scenario B as compared to the 2020 Base Case scenario.

NO_x emissions are the same in Scenarios B and C. Although particulate chemistry would indicate some change in nitrogen deposition based on changes in sulfur emissions, Scenario C showed no significant differences in nitrogen deposition compared to Scenario B.

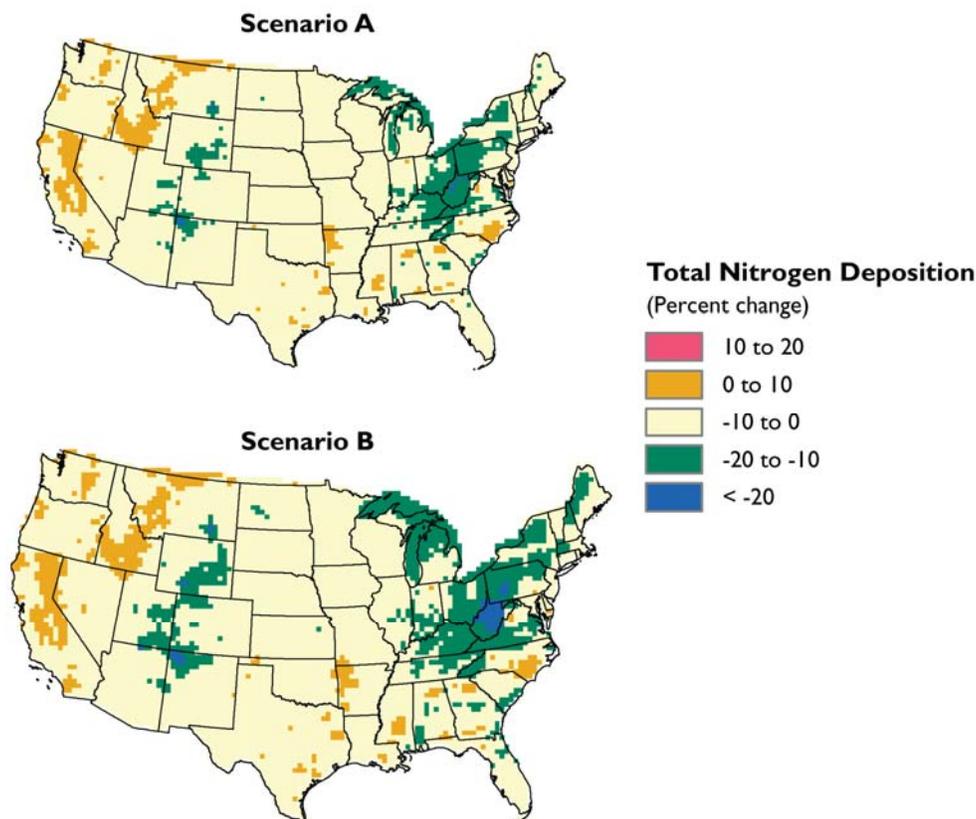
Additional Ecological Impacts of Decreases in Nitrogen Deposition

In addition to acidification, nitrogen deposition can contribute to the eutrophication of estuaries and nutrient enrichment of forests. The nitrogen emission-reduction scenarios presented here are expected to provide ecological benefits to sensitive ecosystems affected by excess nitrogen loading. For example, based on the results of the Response Surface Model (RSM), the Chesapeake Bay Estuary would receive an approximate 10% to 20% reduction in nitrogen deposition under Scenarios B and C as compared to the 2020 Base Case scenario. Additionally, forests in the Front Range region of Colorado would receive approximately a 20% reduction in nitrogen deposition in 2020, with reductions over 30% in some areas, under Scenarios B and C as compared to the Base Case scenario.

28



1
 2 **Figure 4-6. Projected annual total nitrogen deposition for the Base Case scenario in 2020 and**
 3 **Scenarios A and B in 2020 with additional SO₂ and NO_x emission reductions. Because Scenarios B**
 4 **and C have the same level of NO_x emission reductions, Scenario C is not shown.**
 5 **Deposition is reported in kg/ha/yr (Prepared by U.S. EPA).**



1
2 **Figure 4-7. Projected percentage changes in annual total nitrogen deposition from the Base Case**
3 **scenario in 2020 with additional SO₂ and NO_x emission reductions. Because Scenarios B and C**
4 **have the same level of NO_x emission reductions, Scenario C is not shown (Prepared by U.S. EPA).**

5 **4.2.4 Changes in the Ecological Condition of Lakes and Streams**

6 The magnitude of emission reductions influences both the amount of recovery from acidification and the
7 rate at which recovery occurs. The rate of recovery is also influenced by the geological and ecological
8 characteristics of the lakes or streams in the area. The path of ecological recovery from acidification in
9 lakes and streams is displayed in **Figure 4-1**. In this analysis, the ecological response to
10 emission reductions of lakes in the Adirondacks Mountains and in the Northeast and streams in the
11 Southeast (central and southern Appalachian Mountains) were modeled using MAGIC, which estimates
12 acidification of lakes and streams in response to sulfur and nitrogen deposition (Wright et al., 2006) (see
13 the *Modeling Tools Used in this Analysis* text box for a description of MAGIC).

14 The Northeast, the Adirondacks Mountains, and the Southeast were chosen for this modeling assessment
15 because they are among the most acid-sensitive ecosystems and are located downwind of many of the
16 emission sources affected by Title IV (see **Figure 4-8**). The best-available environmental data were used
17 to calibrate MAGIC (e.g., water quality, soil, deposition). In addition, MAGIC was calibrated using data
18 collected on acidification in lakes and streams by the National Surface Water Survey (NSWS) and other
19 programs, such as EMAP and the TIME and LTM programs. Both the NSWS and EMAP used statistical
20 methods to sample a representative number of lakes and streams characteristic of the water quality and
21 condition for all lakes or streams in each study region. For example, the NSWS approach represents
22 approximately 28,000 lakes in the Adirondack Mountains and the Northeast and 56,000 stream reaches in
23 the southeast United States (Herlihy et al., 1993; Stoddard et al., 2003). Therefore, within the limitations
24 of these surveys (only lakes larger than 4 hectares are included), the MAGIC results are indicative of the



Figure 4-8. Regions and waterbodies modeled using MAGIC (Prepared by U.S. EPA).

29
30

level of acidification in lakes and streams throughout these regions. Although there are some uncertainties with regard to the model, particularly concerning watershed nitrogen dynamics, MAGIC provides a generally accurate, well-tested, and widely accepted tool for modeling the response of surface water chemistry to sulfur and nitrogen deposition.

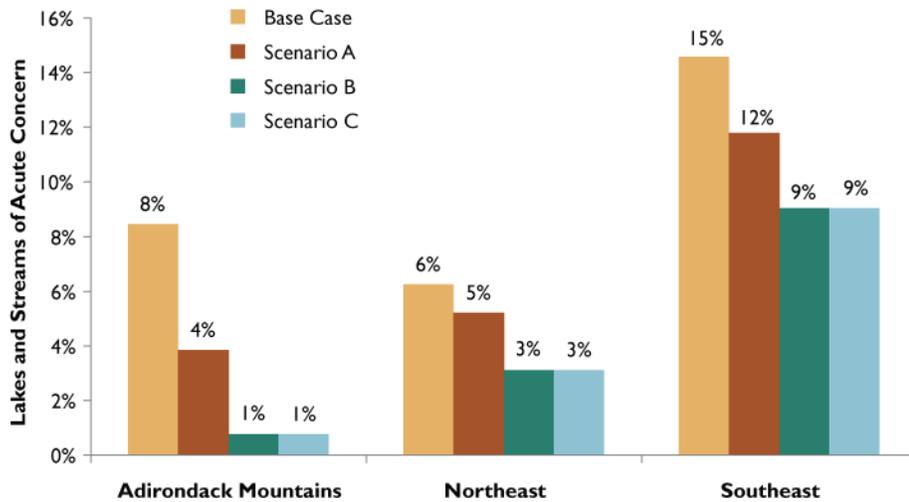
Climate conditions and emissions under the 2020 Base Case scenario and future control Scenarios A, B, and C were assumed to be held constant after 2020. Because of time lags inherent in ecological response to changes in pollutant emissions and deposition, lake and stream water quality in 2020 would represent only a small portion of the recovery expected as a result of emission reductions included in the scenarios analyzed. Therefore, lake and stream conditions are presented for the year 2050, allowing the emission reductions to take effect more fully. Regional forest ecosystem responses to changes in deposition were not modeled in this assessment.

Changes in lake and stream water quality for the three reduction scenarios

31 (A, B, and C) were modeled in this analysis. The implementation of all scenarios is expected to
 32 significantly reduce the remaining percentage of lakes and streams in the acute and elevated concern
 33 categories in all three areas beyond what would occur with implementation of current emission-reduction
 34 programs included in the Base Case scenario. Although the effects of emission reductions beyond the
 35 Base Case scenario differ by region, the amount of reductions results in a modest change in the number of
 36 waterbodies of acute concern in all regions. The greater the emission reductions, the larger the number of
 37 lakes and streams of elevated concern that improve and move into the moderate or low concern categories
 38 (see **Figure 4-1** for an explanation of the stages of recovery from acidification).

39 Lakes in the Northeast and the Adirondack Mountains change relatively quickly in response to changes in
 40 deposition, resulting in shorter recovery times from acidification than streams in the Southeast. Modeling
 41 results indicate that the Base Case scenario would result in 6% and 8% of lakes remaining in the acute
 42 concern class in the Northeast and the Adirondack Mountains, respectively, by 2050 (**Figure 4-9**).
 43 Although these results would improve the health of fish populations and other acid-sensitive species, 40%
 44 and 14% of modeled lakes in the Adirondack Mountains and the Northeast, respectively, still would
 45 remain in the elevated concern class and experience episodic acidification (**Figure 4-10**) under the Base
 46 Case scenario. With implementation of Scenario A, water quality conditions improve in the Northeast and
 47 the Adirondack Mountains, but remain similar to those in the Base Case scenario. The percentage of lakes
 48 of acute concern would decrease to 5% for the Northeast and 4% for the Adirondack Mountains from the

1 levels in the Base Case scenario, while 11% and 37% of modeled lakes would remain of elevated concern
 2 in these regions, respectively. A near elimination of lakes of acute concern in the Northeast and the
 3 Adirondack Mountains by 2050 only occurs given the stringent emission-reduction scenarios of Scenarios
 4 B and C. Under Scenarios B and C, the percentage of lakes in the elevated concern class would also
 5 decrease, which is expected to improve the health of fish populations and allow other acid-sensitive
 6 species to return. With the implementation of Scenario C, the proportion of the lakes of elevated concern
 7 would decrease from 40% to 27% in the Adirondack Mountains and from 14% to 10% in the Northeast.
 8 Given that some lakes are naturally acidic because of organic acids, it is not expected that all lakes or
 9 streams in a region will have an ANC level greater than 50 $\mu\text{eq/L}$. MAGIC also can estimate the water
 10 quality condition before anthropogenic acidification started, providing a target “natural” ANC level for
 11 assessing whether a population of lakes or streams has achieved full recovery. Under Scenario C, the
 12 percentage of lakes in the elevated concern class in 2050 is closer to modeled preacidification levels than
 13 under any other scenario, particularly for lakes in the Northeast. This suggests the scale of emission
 14 reductions under Scenario C would approach full protection of lakes in the Northeast and Adirondack
 15 Mountains from acid deposition (Figures 4-9 and 4-10).

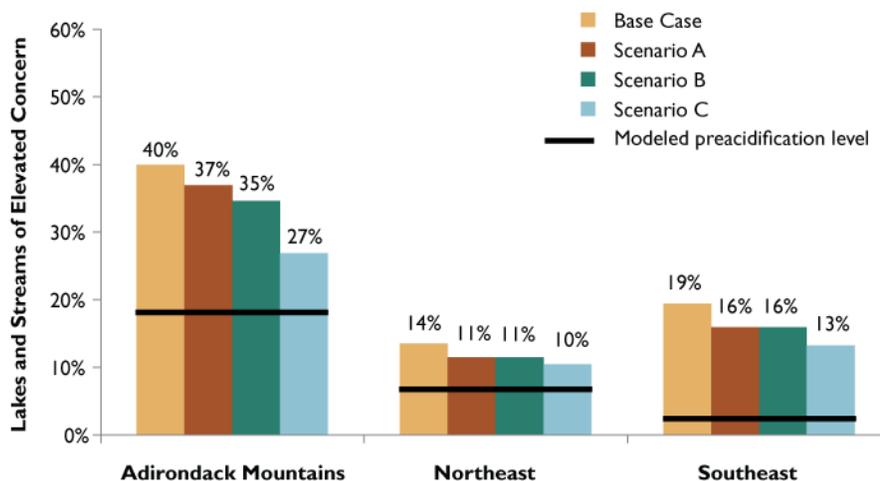


Notes: The area of the Northeast modeled by MAGIC includes Maine, New Hampshire, Vermont, Massachusetts, Rhode Island, Connecticut, New York (not including the Adirondacks), and northeastern Pennsylvania. The area of the Southeast includes Virginia, West Virginia, North Carolina, eastern Tennessee, northern Georgia, and northwestern South Carolina.

The amount of acidification projected in the Base Case scenario includes emission reductions as a result of CAIR and other mobile source regulations finalized after 2005.

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Figure 4-9. Projected changes in the number of lakes and streams of acute concern (ANC < 0 $\mu\text{eq/L}$) in 2050 with additional SO_2 and NO_x emission reductions. Under preacidification conditions, none of these lakes or streams would be in the acute concern class (Prepared by U.S. EPA).



Notes: The area of the Northeast modeled by MAGIC includes Maine, New Hampshire, Vermont, Massachusetts, Rhode Island, Connecticut, New York (not including the Adirondacks), and northeastern Pennsylvania. The area of the Southeast includes Virginia, West Virginia, North Carolina, eastern Tennessee, northern Georgia, and northwestern South Carolina.

The amount of acidification projected in the Base Case scenario includes emission reductions as a result of CAIR and other mobile source regulations finalized after 2005.

Figure 4-10. Projected changes in the number of lakes and streams of elevated concern (ANC is 0-50 µeq/L) in 2050 with additional SO₂ and NO_x emission reductions. The black line represents the percent of lakes or streams that would naturally occur in the elevated concern class as projected with MAGIC modeling (Prepared by U.S. EPA).

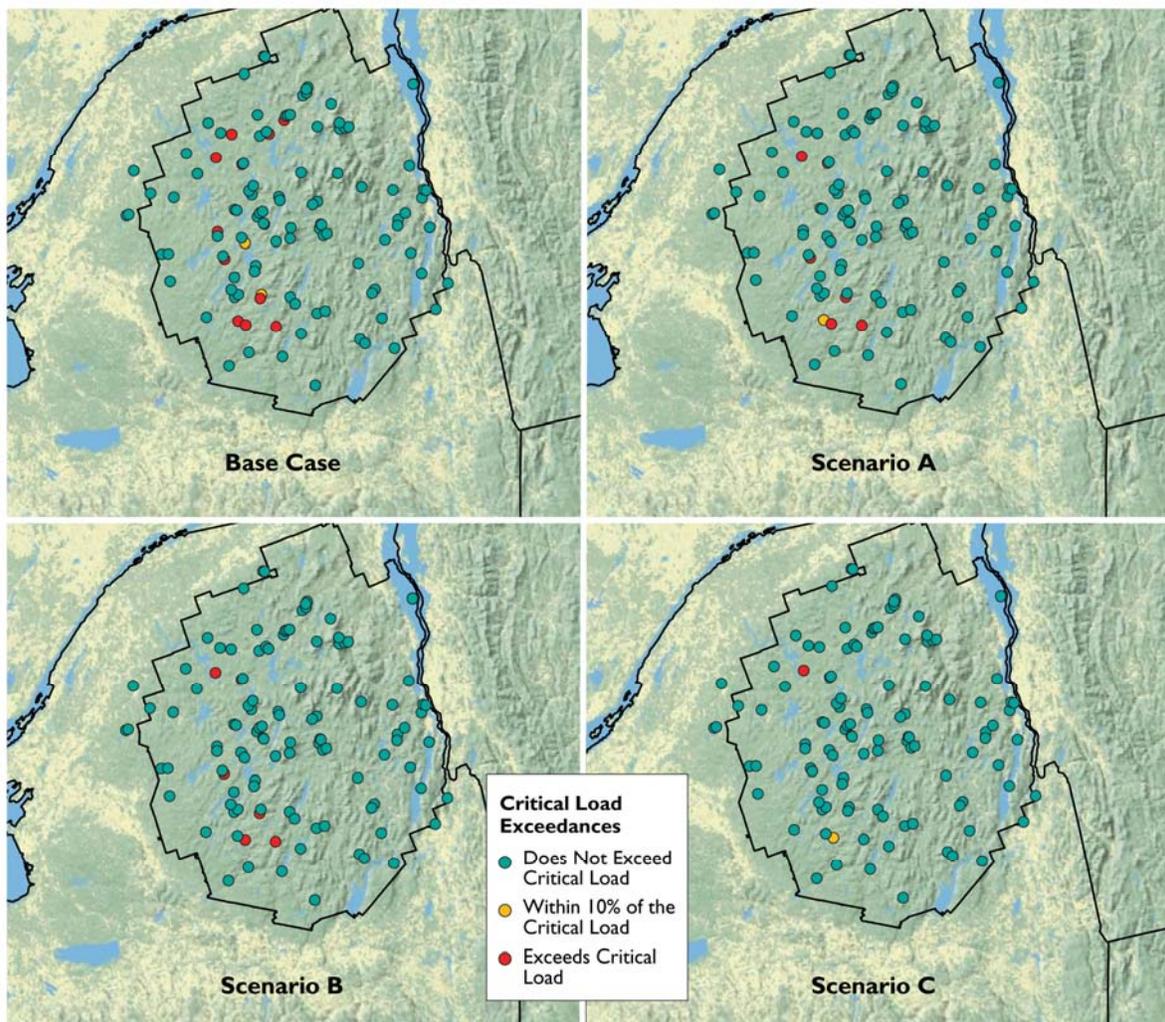
The story is somewhat different for the Southeast (**Figures 4-9 and 4-10**). Because of the unique watershed characteristics of the area, southeastern streams manifest changes over a longer period of time in response to emission reductions. Sulfur retention in the clay-rich soils of the Southeast and the eventual release of sulfur back into the surface water is an important reason for the delayed ecosystem response in this region. As a result, modeling indicates that 15% of streams would remain of acute concern in 2050 in response to emission levels under the Base Case scenario. With implementation of Scenario A, water quality conditions improve in the Southeast with the percentage of streams of acute concern decreasing to 12% of modeled streams by 2050. Under reductions of 75% and 90% in SO₂ emissions from power generation sources (i.e., Scenarios B and C) beyond the levels in the Base Case scenario, additional recovery is expected, and streams in the acute concern class drop to 9% of modeled streams by 2050. Under Scenarios A, B, and C, the percentage of streams in the elevated concern class also would begin to decrease, showing a move toward ecosystem recovery. However, even under the large emission reductions included in Scenario C, 13% of modeled streams would remain of elevated concern in 2050 (**Figure 4-10**). This lag in the recovery of streams of acute concern in the Southeast is due primarily to the large amount of sulfur that has been adsorbed by southeastern soils from decades of acid deposition. This adsorbed sulfur is predicted to be slowly released into streams over time, slowing the rate of stream recovery. For this reason, the response of streams in the Southeast is expected to lag emission reductions to a greater extent than lakes in the Northeast and the Adirondack Mountains, and the emission reductions in Scenarios A, B, and C will take longer to yield results in this region.

4.3 Critical Loads

Critical loads were calculated using a long-term steady-state model for the EMAP lakes in the Adirondack Mountains of New York under the Base Case scenario and Scenarios A through C (see *Adirondack Mountains Critical Load Case Study* in Chapter 2 for more details on the lakes modeled here). The analysis focuses on the combined load of sulfur and nitrogen deposition to which a lake could be subject and still support a moderately healthy aquatic ecosystem (i.e., ANC greater than 50 µeq/L).

1 Some lakes have naturally low acidic conditions. The preacidification ANC levels of the lakes were
 2 estimated using the MAGIC model. Of the 117 EMAP lakes modeled, six lakes had preacidification ANC
 3 levels below an ANC of 50 µeq/L. Because of their natural acidity levels, it is unlikely that these six lakes
 4 would reach an ANC of 50 µeq/L or greater. These lakes were removed from this critical load analysis.
 5 Overall, this analysis shows that the future emission reductions (i.e., Scenarios A through C) would result
 6 in significant ecological recovery, as defined by reaching an ANC level greater than 50 µeq/L, and would
 7 significantly increase ecosystem protection of lakes in the Adirondack Mountains from acidic deposition.

8 **Figure 4-11** shows lakes where deposition exceeds, does not exceed, and would be within 10% of the
 9 critical load. For Adirondack Mountain lakes under the Base Case scenario, 13% of lakes received levels
 10 of combined sulfur and nitrogen deposition that exceeded the critical load and could not be neutralized by
 11 the environment. For Scenarios A through C, 6%, 5%, and 1%, respectively, of the lakes would continue
 12 to receive acid deposition levels that exceeded the critical load. These results indicate that additional
 13 emission reductions lead to further ecosystem protection of lakes in the Adirondack Mountain region.



14 **Figure 4-11. Critical load exceedances for sulfur and nitrogen for 111 lakes in the Adirondack**
 15 **Mountains under the Base Case scenario and Scenarios A through C (Prepared by U.S. EPA).**
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- 1 Aber, J., R.P. Neilson, S. McNulty, J.M. Lenihan, D. Bachelet, and R.J. Drapek. 2001. Forest processes
2 and global environmental change: predicting the effects of individual and multiple stressors.
3 *BioScience* 51:735–751.
- 4 Aber, J.D., C.L. Goodale, S.V. Ollinger, M.L. Smith, A.H. Magill, M.E. Martin, R.A. Hallett, and J.L.
5 Stoddard. 2003. Is nitrogen deposition altering the nitrogen status of northeastern forests?
6 *BioScience* 53:375–389.
- 7 Aber, J.D., R.T. Holmes, R.J. Freuder, C.L. Goodale, S.V. Ollinger, C.T. Driscoll, and G.E. Likens. 2002.
8 Inorganic nitrogen losses from a forested ecosystem in response to physical, chemical, biotic, and
9 climatic perturbations. *Ecosystems* 5:648–658.
- 10 Alcamo, J., P. Mayerhofer, R. Guardans, T. van Harmelen, J. van Minnen, J. Onigkeit, M. Posch, and B.
11 de Vries. 2002. An integrated assessment of regional air pollution and climate change in Europe:
12 Findings of the AIR-CLIM Project. *Environmental Science & Policy* 5:257–272.
- 13 Alexander, R.B., and R.A. Smith. 2006. Trends in the nutrient enrichment of U.S. rivers during the late
14 20th century and their relation to changes in probable trophic conditions. *Limnology &
15 Oceanography* 51:639–654.
- 16 Alexander, R.B., E.W. Boyer, R.A. Smith, G.E. Schwarz, and R.B. Moore. 2007. The role of headwater
17 streams in downstream water quality. *Journal of the American Water Resources Association*
18 43:41–59.
- 19 Alexander, R.B., R.A. Smith, G.E. Schwarz, E.W. Boyer, J.V. Nolan, and J.W. Brakebill. 2008.
20 Differences in phosphorus and nitrogen delivery to the Gulf of Mexico from the Mississippi
21 River. *Environmental Science & Technology* 42:822–830.
- 22 Allen, E.B., L.E. Rao, R.J. Steers, A. Bytnerowicz, and M.E. Fenn. 2009. Impacts of atmospheric
23 nitrogen deposition on vegetation and soils in Joshua Tree National Park. Pp. 78–100 in R.H.
24 Webb, L.F. Fenstermaker, J.S. Heaton, D.L. Hughson, E.V. McDonald, and D.M. Miller, editors.
25 *The Mojave Desert: Ecosystem Processes & Sustainability*. Las Vegas, NV: University of Nevada
26 Press.
- 27 Anderson, J.B., R.E. Baumgardner, Jr., and S.E. Grenville. 2006. Trends in cloud water sulfate and nitrate
28 as measured at two mountain sites in the Eastern United States: Regional contributions and
29 temporal changes compared with regional changes in emissions, 1986–1999. *Atmospheric
30 Environment* 40(23):4423–4437.
- 31 Andrews, J.A., and W.H. Schlesinger. 2001. Soil CO₂ dynamics, acidification, and chemical weathering
32 in a temperate forest with experimental CO₂ enrichment. *Global Biogeochemical Cycles* 15:149–
33 162.
- 34 Bailey, S.W., D.C. Buso, and G.E. Likens. 2003. Implications of sodium mass balance for interpreting the
35 calcium cycle of a forested ecosystem. *Ecology* 84:471–484.
- 36 Bailey, S.W., S.B. Horsley, and R.P. Long. 2005. Thirty years of change in forest soils of the Allegheny
37 Plateau, Pennsylvania. *Soil Science Society of America Journal* 69:681–690.
- 38 Bailey, S.W., S.B. Horsley, R.P. Long, and R.A. Hallett. 2004. Influence of edaphic factors on sugar
39 maple nutrition and health on the Allegheny Plateau. *Soil Science Society of America Journal*
40 68:243–252.

- 1 Baker, J.P., and S.W. Christensen. 1991. Effects of acidification on biological communities in aquatic
2 ecosystems. Pp. 83–106 in D.F. Charles , editor. *Acidic Deposition & Aquatic Ecosystems*. New
3 York: Springer-Verlag.
- 4 Baldigo, B.P., and P.S. Murdoch. 1997. Effect of stream acidification and inorganic aluminum on
5 mortality of brook trout (*Salvelinus fontinalis*) in the Catskill Mountains, New York. *Canadian*
6 *Journal of Fisheries and Aquatic Sciences* 54:603–615.
- 7 Banzhaf, H.S., D. Burtraw, D. Evans, and A. Krupnick. 2006. Valuation of natural resource
8 improvements in the Adirondacks. *Land Economics* 82(3):445–464.
- 9 Baron, J.S., S. Del Grosso, D.S. Ojima, D.M. Theobald, and W.J. Parton. 2004. Nitrogen emissions along
10 the Colorado Front Range: response to population growth, land and water use change, and
11 agriculture. Pp. 117-127 in R. DeFries, G. Asner, and R. Houghton, editors. *Ecosystem*
12 *Interactions & Land Use Change*. Washington, DC: American Geophysical Union.
- 13 Baron, J.S., T.M. Schmidt, and M.D. Hartman. 2009. Climate-induced changes in high elevation stream
14 nitrate dynamics. *Global Change Biology* 15:1777–1789.
- 15 Baron, J.S. 2006. Hindcasting nitrogen deposition to determine an ecological critical load. *Ecological*
16 *Applications* 16:433–439.
- 17 Bates, N.R., and A.J. Peters. 2007. The contribution of atmospheric acid deposition to ocean acidification
18 in the subtropical North Atlantic Ocean. *Marine Chemistry* 107:547–558.
- 19 Beckage, B., B. Osborne, D.G. Gavin, C. Pucko, T. Siccama, and T. Perkins. 2008. A rapid upward shift
20 of a forest ecotone during 40 years of warming in the Green Mountains of Vermont. *Proceedings*
21 *of the National Academy of Sciences* 105:4197–4202.
- 22 Beem, K.B., S. Raja, F.M. Schwnadner, C. Taylor, T. Lee, A.P. Sullivan, C.M. Carrico, G.R.
23 McMeeking, D. Day, E. Levin, J. Hand, S.M. Kreidenweis, B. Schichtel, W.C. Malm, and J.L.
24 Collett, Jr. 2010. Deposition of reactive nitrogen during the Rocky Mountain Airborne Nitrogen
25 and Sulfur (RoMANS) study. *Environmental Pollution* 158:862–872.
- 26 Beier, C.M., T.M. Patterson, and F.S. Chapin III. 2008. Ecosystem services and emergent vulnerability in
27 managed ecosystems: a geospatial decision-support tool. *Ecosystems* 11:923–938.
- 28 Bell, M.L., A. McDermott, S.L. Zeger, J.M. Samet, and F. Dominici. 2004. Ozone and short-term
29 mortality in 95 U.S. urban communities, 1987-2000. *Journal of the American Medical*
30 *Association* 292(19):2372–8.
- 31 Bell, M.L., F. Dominici, and J.M. Samet. 2005. A meta-analysis of time-series studies of ozone and
32 mortality with comparison to the national morbidity, mortality, and air pollution study.
33 *Epidemiology* 16(4):436–45.
- 34 Bergstrom, A.-K., and M. Jansson. 2006. Atmospheric nitrogen deposition has caused nitrogen
35 enrichment and eutrophication of lakes in the northern hemisphere. *Global Change Biology*
36 12:635–643.
- 37 Binks, J.A., S.E. Arnott, and W.G. Spules. 2005. Local factors and colonist dispersal influence crustacean
38 zooplankton recovery from cultural acidification. *Ecological Applications* 15:2025–2036.

- 1 Blum, J.D., C.E. Johnson, T.G. Siccama, C. Eagar, T.J. Fahey, G.E. Likens, A. Klaue, C.A. Nezat, and
2 C.T. Driscoll. 2002. Mycorrhizal weathering of apatite as an important calcium source in base-
3 poor forest ecosystems. *Nature* 417:729–731.
- 4 Boisvenue, C., and S.W. Running. 2006. Impacts of climate change on natural forest productivity -
5 evidence since the middle of the 20th century. *Global Change Biology* 12:862–882.
- 6 Bowman, W.D., J.R. Gartner, K. Holland, and M. Wiedermann. 2006. Nitrogen critical loads for alpine
7 vegetation and terrestrial ecosystem response: Are we there yet? *Ecological Applications*
8 16:1183–1193.
- 9 Bowman, W.D., T.A. Theodose, and M.C. Fisk. 1995. Physiological and production responses of plant
10 growth forms to increases in limiting resources in alpine tundra: Implications for differential
11 community response to environmental change. *Oecologia* 101:217–227.
- 12 Boyer, E.W., C.L. Goodale, N.A. Jaworski, and R.W. Howarth. 2002. Anthropogenic nitrogen sources
13 and relationships to riverine nitrogen export in the northeastern U.S.A. *Biogeochemistry*
14 57/58:137–169.
- 15 Branfireun, B.A., N.T. Roulet, C.A. Kelly, and J.W.M. Rudd. 1999. In situ sulfate stimulation of mercury
16 methylation in a boreal peatland: toward a link between acid rain and methylmercury
17 contamination in remote environments. *Global Biogeochemical Cycles* 13:743–750.
- 18 Bricker, S., B. Longstaff, W. Dennison, A. Jones, K. Boicourt, C. Wicks, and J. Woerner. 2008. Effects of
19 nutrient enrichment in the nation’s estuaries: A decade of change. *Harmful Algae* 8:21–32.
- 20 Brook, J.R., K.L. Demerjian, G. Hidy, L.T. Molina, W.T. Pennell, and R. Scheffe. 2009. New Directions:
21 Results-oriented multi-pollutant air quality management. *Atmospheric Environment* 43:2091–
22 2093.
- 23 Bulger, A.J., B.J. Cosby, C.A. Dolloff, K.N. Eshleman, J.R. Webb, and J.N. Galloway. 1999. *SNP:FISH,*
24 *Shenandoah National Park: Fish in Sensitive Habitats, Volumes through IV.* Project final report.
25 Charlottesville, VA: University of Virginia, Department of Environmental Sciences. Project
26 Completion Report to the National Park Service. Cooperative Agreement CA-4000-2-1007,
27 Supplemental Agreement #2.
- 28 Burns, D.A., C.T. Driscoll, G.M. Lovett, K.C. Weathers, M.J. Mitchell, and K.M. Roy. 2005. An
29 assessment of recovery and key processes affecting the response of surface waters to reduced
30 levels of acid precipitation in the Adirondack and Catskill Mountains. *New York Energy Research*
31 *and Development Authority Report 05-03*, Albany, NY.
- 32 Burns, D.A., J. Klaus, and M.R. McHale. 2007. Recent climate trends and implications for water
33 resources in the Catskill Mountain region, New York, USA. *Journal of Hydrology* 336:155–170.
- 34 Burns, D.A., K. R. Murray, R.W. Bode, and S.I. Passy. 2008a. Changes in stream chemistry and biology
35 in response to reduced levels of acid deposition during 1987–2003 in the Neversink River Basin,
36 Catskill Mountains. *Ecological Indicators* 8:191–203.
- 37 Burns, D.A., M.R. McHale, C.T. Driscoll, and K.M. Roy. 2006b. Response of surface water chemistry to
38 reduced levels of acid precipitation: comparison of trends in two regions of New York.
39 *Hydrological Processes* 20:1611–1627.

- 1 Burns, D.A., T. Blett, R. Haeuber, and L.H. Pardo. 2008b. Critical loads as a policy tool for protecting
2 ecosystems from the effects of air pollutants. *Frontiers in Ecology & the Environment* 6:156–159.
- 3 Burns, D.A. 2003. Atmospheric nitrogen deposition in the Rocky Mountains of Colorado and southern
4 Wyoming—A review and new analysis of past study results. *Atmospheric Environment* 37:921–
5 932.
- 6 Burns D.A., M.R. McHale, C.T. Driscoll, and K.M. Roy. 2006a. Response of surface water chemistry to
7 reduced levels of acid precipitation: comparison of trends in two regions of New York, USA.
8 *Hydrological Processes* 20:1611–1627.
- 9 Burtraw, D., and S.J. Szambelan. 2009. *U.S. Emissions Trading Markets for SO₂ and NO_x*. Discussion
10 Paper RFF DP 09-40. Resources for the Future, Washington, DC.
- 11 Burtraw, D., D.A. Evans, A. Krupnick, K. Palmer, and R. Toth. 2005. Economics of pollution trading for
12 SO₂ and NO_x. *Annual Review of Environment and Resources* 30:253–89.
- 13 Butler, T.J., G.E. Likens, F.M. Vermeulen, and B.J.B. Stundler. 2003. The relation between NO_x
14 emissions and precipitation NO₃ in the eastern USA. *Atmospheric Environment* 37:2093–2104.
- 15 Bytnerowicz, A., K. Omasa, and E. Paoletti. 2007. Integrated effects of air pollution and climate change
16 on forests: a northern hemisphere perspective. *Environmental Pollution* 147:438–445.
- 17 Caine, N. 1995. Temporal trends in the quality of streamwater in an alpine environment: Green Lakes
18 Valley, Colorado Front Range, U.S.A. *Geografiska Annaler. Series A, Physical Geography*
19 77A:207–220.
- 20 Cairns, A., and N. Yan. 2009. A review of the influence of low ambient calcium concentrations on
21 freshwater daphniids, gammerids, and crayfish. *Environmental Reviews* 17:67–79.
- 22 Caldeira, K., and M.E. Wickett. 2003. Anthropogenic carbon and ocean pH. *Nature* 425:365.
- 23 Campbell, D.H., E. Muths, J.T. Turk, and P.S. Corn. 2004. Sensitivity to acidification of subalpine ponds
24 and lakes in north- western Colorado. *Hydrological Processes* 18:2817–2834.
- 25 Campbell, J. L., L.E. Rustad, E.W. Boyer, S.F. Christopher, C.T. Driscoll, I.J. Fernandez, P.M. Groffman,
26 D. Houle, J. Kiebusch, A.H. Magill, M.J. Mitchell, and S.V. Ollinger. 2009. Consequences of
27 climate change for biogeochemical cycling in forest of northeastern North America. *Canadian*
28 *Journal of Forest Research* 39:264–284.
- 29 CANTORCO₂e. 2009. *Market Price Index*. Online Information. CANTORCO₂e, San Francisco, CA.
30 Available at http://www.cantorco2e.com/Environment/?page=USAComp_MarketData-MPI
31 (accessed 2009).
- 32 Carlson, C., D. Burtraw, M. Cooper, and K.L. Palmer. 2000. Sulfur dioxide control by electric utilities:
33 What are the gains from trade? *Journal of Political Economy* 108(6):1292–1326.
- 34 Chen, L.M., and C.T. Driscoll. 2004. Modeling the response of soil and surface waters in the Adirondack
35 and Catskill regions of New York to changes in atmospheric deposition and historical land
36 disturbance. *Atmospheric Environment* 38:4099–4109.

- 1 Chen, L.M., and C.T. Driscoll. 2005a. Regional application of an integrated biogeochemical model to
2 northern New England and Maine. *Ecological Applications* 15:1783–1797.
- 3 Chen, L.M., and C.T. Driscoll. 2005b. Regional assessment of the response of the acid-base status of lake
4 watersheds in the Adirondack region of New York to changes in atmospheric deposition using
5 PnET-BGC. *Environmental Science & Technology* 39:787–794.
- 6 Chestnut, L.G., and D.M. Mills. 2005. A fresh look at the benefits and costs of the U.S. Acid Rain
7 Program. *Journal of Environmental Management* 77(3):252–266.
- 8 Chow, A.T., K.K. Tanji, and S. Gao. 2003. Production of dissolved organic carbon (DOC) and
9 trihalomethane (THM) precursor from peat soils. *Water Research* 37:4475–4485.
- 10 Christoperson, N., H.M. Seip, and R.F. Wright. 1982. A model for streamwater chemistry at Birkenes,
11 Norway. *Water Resources Research* 18:977–986.
- 12 Churkina, G., V. Brovkin, W. von Bloh, K. Trusilova,, M. Jung, and F. Dentener. 2009. Synergy of rising
13 nitrogen depositions and atmospheric CO₂ on land carbon uptake moderately offsets global
14 warming. *Global Biogeochemical Cycles* 23:23, GB4027, doi:10.1029/2008GB03291.
- 15 Civerolo, K.L., C. Hogrefe, B. Lynn, C. Rosenzweig, R. Goldberg, J. Rosenthal, K. Knowlton, and P.L.
16 Kinney. 2008. Simulated effects of climate change on summertime nitrogen deposition in the
17 eastern US. *Atmospheric Environment* 42:2074–2082.
- 18 Clair, T.A., and A. Hindar. 2005. Liming for the mitigation of acid rain effects in freshwaters: a review of
19 recent results. *Environmental Reviews* 13:91–128.
- 20 Clair, T.A., and J.M. Ehrmann. 1998. The influence of changing seasonal climates in modifying
21 discharge, dissolved organic carbon and nitrogen export in wetland influenced rivers: a neural
22 network approach. *Water Resources Research* 34:447–455.
- 23 Clair, T.A., I.F. Dennis, R. Vet, and H. Laudon. 2008. Long-term trends in catchment dissolved organic
24 carbon and nitrogen from three acidified catchments in Nova Scotia, Canada. *Biogeochemistry*
25 87:83–97.
- 26 Clark, J.M., P.J. Chapman, A.L. Heathwaite, and J.K. Adamson. 2006. Suppression of dissolved organic
27 carbon by sulfate induced acidification during simulated droughts. *Environmental Science &*
28 *Technology* 40:1776–1783.
- 29 Clark, J.M., S.H. Bottrell, C.D. Evans, D.T. Monteith, R. Bartlett, R. Rose, R.J. Newton, P.J. Chapman.
30 2010. The importance of the relationship between scale and process in understanding long-term
31 DOC dynamics. *Science of the Total Environment* 408:2768-2775.
- 32 Clow, D.W., and J.K. Sueker. 2000. Relations between basin characteristics and stream water chemistry
33 in alpine/subalpine basins in Rocky Mountain National Park, Colorado. *Water Resources*
34 *Research* 36:49–61.
- 35 Clow, D.W., J.O. Sickman, R.G. Striegl, D.P. Krabbenhoft, J.G. Elliott, M. Domblaser, D.A. Roth, and
36 D.H. Campbell. 2003. Changes in the chemistry of lakes and precipitation in high-elevation
37 national parks in the western United States, 1985-1999. *Water Resources Research* 39(6), 1171,
38 doi:10.1029/2008WR001533.

- 1 Corvalan, C., S. Hales, and A. McMichael. 2005. *Ecosystems and Human Well-Being: Health Synthesis: A Report of the Millenium Ecosystem Assessment*. Geneva, Switzerland: World Health
2 Organization Press.
3
- 4 Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985a. Modeling the effect of acid
5 deposition: assessment of a lumped parameter model of soil water and streamwater chemistry.
6 *Water Resources Research* 21:51–63.
- 7 Cosby, B.J., R.F. Wright, G.M. Hornberger, and J.N. Galloway. 1985b. Modeling the effect of acid
8 deposition: estimation of a long term water quality responses in a small forested catchment.
9 *Water Resources Research* 21:1591–1603.
- 10 Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985c. Timescales of catchment
11 acidification. *Environmental Science and Technology* 19:1144–1149.
- 12 Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985d. Time scales of catchment
13 acidification: A quantitative model for estimating freshwater acidification. *Environmental Science
14 & Technology* 19:1144–1149.
- 15 Cosby, B.J., J.R. Webb, J.N. Galloway, and F.A. Deviney. 2006. *Acidic deposition impacts on natural
16 resources in Shenandoah National Park*. Technical Report NPS/NER/NRTR—2006/066. U.S.
17 Department of the Interior, National Park Service, Northeast Region, Philadelphia, PA.
- 18 Costanza, R., R. d’Arge, R. de Groot, S. Farber, M. Grasso, B. Hannon, K. Limburg, S. Naeem, R.V.
19 O’Neill, J. Paruelo, R.G. Raskin, P. Sutton, and M. van den Belt. 1997. The value of the world’s
20 ecosystem services and natural capital. *Nature* 387:253–260.
- 21 Dale, V.H., L.A. Joyce, S. McNulty, R.P. Neilson, M.P. Ayres, M.D. Flannigan, P.J. Hanson, L.C. Irland,
22 A.E. Lugo, C.J. Peterson, D. Simberloff, F.J. Swanson, B.J. Stocks, and B.M. Wotton. 2001.
23 Climate change and forest disturbances. *BioScience* 51:723–734.
- 24 Dalias, P., J.M. Anderson, P. Bottner, and M.M. Coûteaux. 2002. Temperature responses of net nitrogen
25 mineralization and nitrification in conifer forest soils incubated under standard laboratory
26 conditions. *Soil Biology & Biochemistry* 34:691–701.
- 27 DeHayes, D.H., P.G. Schaberg, G.L. Hawley, and G.R. Strimbeck. 1999. Acid rain impacts calcium
28 nutrition and forest health. *BioScience* 49:787–800.
- 29 Deviney, F.A., K.C. Rice, and G.M. Hornberger. 2006. Time series and recurrence interval models to
30 predict the vulnerability of streams to episodic acidification in Shenandoah National Park,
31 Virginia. *Water Resources Research* 42.
- 32 Dewar, R.C., O. Franklin, A. Makela, R.E. McMurtrie, and H.T. Valentine. 2009. Optimal function
33 explains forest responses to global change. *BioScience* 59:127–139.
- 34 Diaz, R.J. 2001. Overview of hypoxia around the world. *Journal of Environmental Quality* 30:275–281.
- 35 Dillon, P.J., and M.L. Molot. 1997. Dissolved organic and inorganic carbon mass balance in central
36 Ontario lakes. *Biogeochemistry* 36:29–42.

- 1 Doney, S.C., N. Mahowald, I. Lima, R.A. Feely, F.T. Mackenzie, J.-F. Lamarque, and P.J. Rasch. 2007.
2 Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and
3 the inorganic carbon system. *Proceedings of the National Academy of Sciences* 104:14580–
4 14585.
- 5 Drevnick, P.E., P.E. Drevnick, D.E. Canfield, P.R. Gorski, A.L.C. Shinneman, D.R. Engstrom, D.C.G.
6 Muir, G.R. Smith, P.J. Garrison, L.B. Cleckner, J.P. Hurley, R.B. Noble, R.R. Otter, and J.T.
7 Oris. 2007. Deposition and cycling of sulfur controls mercury accumulation in Isle Royale fish.
8 *Environmental Science & Technology* 41:7266–7272.
- 9 Driscoll, C.T., C.P. Cirno, T.J. Fahey, V.L. Blette, P.A. Bukaveckas, D.A. Burns, C.P. Gubala, D.J.
10 Leopold, R.M. Newton, D.J. Raynal, C.L. Schofield, J.B. Yavitt, and D.B. Porcella. 1996. The
11 Experimental Watershed Liming Study: Comparison of lake and watershed neutralization
12 strategies. *Biogeochemistry* 32:143–174.
- 13 Driscoll, C.T., G.B. Lawrence, A.J. Bulger, T.J. Butler, C.S. Cronan, C. Eagar, K.F. Lambert, G.E.
14 Likens, J.L. Stoddard, and K.C. Weathers. 2001. Acidic Deposition in the Northeastern United
15 States: Sources and Inputs, Ecosystem Effects, and Management Strategies. *BioScience* 51:180–
16 198.
- 17 Driscoll, C.T., K.M. Driscoll, M.J. Mitchell, and D.J. Raynal. 2003. Effects of acidic deposition on forest
18 and aquatic ecosystems in New York State. *Environmental Pollution* 123:327–336.
- 19 Driscoll, C.T., R.M. Newton, C.P. Gubala, J.P. Baker, and S.W. Christensen. 1991. Adirondack
20 mountains. Pp. 133–202 in D.F. Charles, editor. *Acidic Deposition and Aquatic Ecosystems:*
21 *Regional Case Studies*. New York, NY: Springer-Verlag.
- 22 Driscoll C.T., K.M. Driscoll, K.M. Roy, and J. Dukett. 2007. Changes in the chemistry of lakes in the
23 Adirondack region of New York following declines in acidic deposition. *Applied Geochemistry*
24 22(6):1181–1188.
- 25 Dupont, J., T.A. Clair, C. Gagnon, D.S. Jeffries, J.S. Kahl, S.J. Nelson, and J.M. Peckenham. 2005.
26 Estimation of critical loads of acidity for lakes in the northeastern United States and eastern
27 Canada. *Environmental Monitoring and Assessment* 109:275–291.
- 28 Durance, I., and S.J. Ormerod. 2007. Climate change effects on upland stream macroinvertebrates over a
29 25- year period. *Global Change Biology* 13:942–957.
- 30 Egerton-Warburton, L.M., and E.B. Allen. 2000. Shifts in arbuscular mycorrhizal communities along an
31 anthropogenic nitrogen deposition gradient. *Ecological Applications* 10:484–496.
- 32 Egerton-Warburton, L.M., R.C. Graham, E.B. Allen, and M.F. Allen. 2001. Reconstruction of the
33 historical changes in mycorrhizal fungal communities under anthropogenic nitrogen deposition.
34 *Proceedings of the Royal Society of London B* 268:2479–2484.
- 35 Eimers, M.C., S.A. Watmough, J.M. Buttle, and P.J. Dillon. 2007. Drought-induced sulphate release from
36 a wetland in south-central Ontario. *Environmental Monitoring & Assessment* 127:399–407.
- 37 Ellerman, A.D. 2003. *Ex Post Evaluation of Tradable Permits: The U.S. SO₂ Cap-and-Trade Program*.
38 Massachusetts Institute of Technology, Center for Energy and Environmental Policy Research,
39 Cambridge, MA.

- 1 EPRI (Electric Power Research Institute). 1993. *Integrated Analysis of Fuel, Technology and Emission*
2 *Allowance Markets: Electric Utility Responses to the Clean Air Act Amendments of 1990*. Electric
3 Power Research Institute, Palo Alto, CA.
- 4 Escobar, I.C., A.A. Randall, and J.S. Taylor. 2001. Bacterial growth in distribution systems: effect of
5 assimilable organic carbon and biodegradable dissolved organic carbon. *Environmental Science*
6 *& Technology* 35:3442–3447.
- 7 Eshleman, K.N., B.E. McNeil, and P.A. Townsend. 2009. Validation of a remote sensing based index of
8 forest disturbance using streamwater nitrogen data. *Ecological Indicators* 9:476-484.
- 9 Evans, C.D., P.J. Chapman, J.M. Clark, D.T. Monteith, and M.S. Cresser. 2006. Alternative explanations
10 for rising dissolved organic carbon export from organic soils. *Global Change Biology* 12:2044–
11 2053.
- 12 Evans, D.A., H.S. Banzhaf, D. Burtraw, A.J. Krupnick, and J. Siikimaki. 2008. Valuing benefits from
13 ecosystem improvements using stated preference methods: an example from reducing
14 acidification in the Adirondacks Park. Pp. 101–117, Chapter 9, in R.A. Askins, G.D. Dreyer, G.R.
15 Visgilio, and D.M. Whitelaw, editors. *Saving Biological Diversity*. Springer: New York.
- 16 Faulkenham, S.E., R.I. Hall, P.J. Dillon, and T. Karst-Riddoch. 2003. Effects of drought-induced
17 acidification on diatom communities in acid-sensitive Ontario lakes. *Limnology & Oceanography*
18 48:1662–1673.
- 19 Fenn, M.E., J.S. Baron, E.B. Allen, H.M. Rueth, K.R. Nydick, L. Geiser, W.D. Bowman, J.O. Sickman,
20 T. Meixner, D.W. Johnson, and P. Neitlich. 2003a. Ecological effects of nitrogen deposition in
21 the western United States. *BioScience* 53:404–420.
- 22 Fenn, M.E., M.A. Poth, A. Bytnerowicz, J.O. Sickman, and B.K. Takemoto, 2003b. Effects of ozone,
23 nitrogen deposition, and other stressors on montane ecosystems in the Sierra Nevada. Pp. 111–
24 155 in A. Bytnerowicz, M.J. Arbaugh, and R. Alonso, editors. *Developments in Environmental*
25 *Science, Volume 2: Ozone Air Pollution in the Sierra Nevada: Distribution and Effects on*
26 *Forests*. Amsterdam: Elsevier.
- 27 Fenn, M.E., L. Geiser, R. Bachman, T.J. Blubaugh, A. Bytnerowicz. 2007. Atmospheric deposition inputs
28 and effects on lichen chemistry and indicator species in the Columbia River Gorge, USA.
29 *Environmental Pollution* 146:77–91.
- 30 Fenn, M.E., M.A. Poth, J.D. Aber, J.S. Baron, B.T. Bormann, D.W. Johnson, A.D. Lemly, S.G. McNulty,
31 D.F. Ryan, and R. Stottlemeyer. 1998. Nitrogen excess in North American ecosystems:
32 predisposing factors, ecosystem responses, and management strategies. *Ecological Applications*
33 8:706–733.
- 34 Fenn, M.E., S. Jovan, F. Yuan, L. Geiser, T. Meixner, and B.S. Gimeno. 2008. Empirical and simulated
35 critical loads for nitrogen deposition in California mixed conifer forests. *Environmental Pollution*
36 155:492–511.
- 37 Findlay, D.L. 2003. Response of phytoplankton communities to acidification and recovery in Killarney
38 Park and the experimental lakes area, Ontario. *Ambio* 32:190–195.

- 1 Findlay, S.E.G. 2005. Increased carbon transport in the Hudson River: Unexpected consequence of
2 nitrogen deposition? *Frontiers in Ecology & the Environment* 3:133–137.
- 3 Fisher, L.S., P.A. Mays, and C.L. Wylie. 2007. An overview of nitrogen critical loads for policy makers,
4 stakeholders, and industries in the United States. *Water, Air, & Soil Pollution* 179:3–18.
- 5 Fitzhugh, R.D., G.E. Likens, C.T. Driscoll, M.J. Mitchell, P.M. Groffman, T.J. Fahey, and J.P. Hardy.
6 2003. Role of soil freezing events in interannual patterns of stream chemistry at the Hubbard
7 Brook Experimental Forest, New Hampshire. *Environmental Science & Technology* 37:1575–
8 1580.
- 9 Frey, K.E., and L.C. Smith. 2005. Amplified carbon release from vast west Siberian peatlands by 2100.
10 *Geophysical Research Letters* 32.
- 11 Frost, T.M., J.M. Fischer, J.L. Klug, S.E. Arnott, and P.K. Montz. 2006. Trajectories of zooplankton
12 recovery in the Little Rock Lake whole-lake acidification experiment. *Ecological Applications*
13 16:353–367.
- 14 Gbondo-Tugbawa, S.S., C.T. Driscoll, J.D. Aver, and G.E. Likens. 2001. Evaluation of an integrated
15 model (PnET-BGC) at a northern hardwood ecosystem. *Water Resources Research* 37:1057-
16 1070.
- 17 Geiser, L.H., and P.N. Neitlich. 2007. Pollution and climate gradients in western Oregon and Washington
18 indicated by epiphytic macrolichens. *Environmental Pollution* 145:203–218.
- 19 Gherini, S.A., L. Mok, R.J.M. Hudson, G.F. Davis, C.W. Chen, and R.A. Goldstein. 1985. The ILWAS
20 model: Formulation and application. *Water, Air, & Soil Pollution* 26:425–459.
- 21 Giardina, C.P., and M.G. Ryan. 2000. Evidence that decomposition rates of organic carbon in mineral soil
22 do not vary with temperature. *Nature* 404:858–861.
- 23 Gilmour, C.C., E.A. Henry, and R. Mitchell. 1992. Sulfate stimulation of mercury methylation in
24 freshwater sediments. *Environmental Science & Technology* 26:2281–2287.
- 25 Gimeno, B.S., F. Yuan, M.E. Fenn, and T. Meixner. 2009. Management options for mitigating nitrogen
26 (N) losses from N saturated mixed conifer forests in California. Pp. 425–455, in A. Bytnerowicz,
27 M.J. Arbaugh, A.R. Riebau, and C. Andersen, editors. *Wildland Fires & Air Pollution.*
28 *Developments in Environmental Science, Volume 8.* Amsterdam: Elsevier.
- 29 Glavich, D.A., and L.H. Geiser. 2008. Potential approaches to developing lichen-based critical loads and
30 levels for nitrogen, sulfur and metal-containing atmospheric pollutants in North America. *The*
31 *Bryologist* 111:638–649.
- 32 Goodale, C.L., M.J. Apps, R.A. Birdsey, C.B. Field, L.S. Heath, R.A. Houghton, J.C. Jenkins, G.H.
33 Kohlmaier, W. Kurz, S. Liu, G.-J. Nabuurs, S. Nilsson, and A.Z. Shvidenko. 2002. Forest
34 carbon sinks in the Northern Hemisphere. *Ecological Applications* 12:891–899.
- 35 Graveland, J. 1998. Effects of acid rain on bird populations. *Environmental Reviews* 6:41–54.

- 1 Greening, H., and A. Janicki. 2006. Toward reversal of eutrophic conditions in a subtropical estuary:
2 Water quality and seagrass response to nitrogen loading reductions in Tampa Bay, Florida, USA.
3 *Environmental Management* 38:163–178.
- 4 Grigal, D.F., and P.R. Ohmann. 2005. Calcium and forest systems: diffusion from deep sources. *Soil*
5 *Science* 170:129–136.
- 6 Grimm, J.W., and J.A. Lynch. 2004. Enhanced wet deposition estimates using modeled precipitation
7 inputs. *Environmental Monitoring and Assessment* 90:243–268.
- 8 Grulke, N.E., R.A. Minnich, T.D. Paine, S.J. Seybold, D.J. Chavez, M.E. Fenn, P.J. Riggan, and A. Dunn.
9 2009. Air pollution increases forest susceptibility to wildfires: a case study in the San Bernardino
10 Mountains in southern California. Pp. 365–403 in A. Bytnerowicz, M.J. Arbaugh, A.R. Riebau,
11 and C. Andersen, editors. *Wildland Fires & Air Pollution. Developments in Environmental*
12 *Science, Volume 8*. Amsterdam: Elsevier.
- 13 Hallet, R.A., S.W. Bailey, S.B. Horsley, and R.P. Long. 2006. Influence of nutrition and stress on sugar
14 maple at a regional scale. *Canadian Journal of Forest Research* 36:2235–2246.
- 15 Halvorsen, G.A., E. Heegaard, A. Fjellheim, and G.G. Raddum. 2003. Tracing recovery from
16 acidification in the western Norwegian Nausta watershed. *Ambio* 32:235–239.
- 17 Hamburg, S.P., R.D. Yanai, M.A. Arthur, J.D. Blum, and T.G. Siccama. 2003. Biotic control of calcium
18 cycling in northern hardwood forests: Acid rain and aging forests. *Ecosystems* 6:399–406.
- 19 Hames, R.S., K.V. Rosenberg, J.D. Lowe, S.E. Barker, and A.A. Dhondt. 2002. Adverse effects of acid
20 rain on the distribution of the wood thrush *Hylocichla mustelina* in North America. *Proceedings*
21 *of the National Academy of Sciences* 99:11235–11240.
- 22 Hartman, M.D., J.S. Baron, and D.S. Ojima. 2007. Application of a coupled ecosystem-chemical
23 equilibrium model, DayCent-Chem, to stream and soil chemistry in a Rocky Mountain watershed.
24 *Ecological Modelling* 200:493–510.
- 25 Hawley, G.J., P.G. Schaberg, C. Eager, and C.H. Borer. 2006. Calcium addition at the Hubbard Brook
26 Experimental Forest reduced winter injury to red spruce in a high-injury year. *Canadian Journal*
27 *of Forest Research* 36:2544–2549.
- 28 Held, I.M., and B.J. Soden. 2006. Robust responses of the hydrological cycle to global warming. *Journal*
29 *of Climate* 19:5686–5699.
- 30 Henriksen, A., and D.F. Brakke. 1988. Sulfate deposition to surface waters. *Environmental Science &*
31 *Technology* 22:8–14.
- 32 Henriksen, A., and M. Posch. 2001. Steady-state models for calculating critical loads of acidity for
33 surface waters. *Water, Air & Soil Pollution Focus* 1:375–398.
- 34 Henriksen, A. 1979. A simple approach for identifying and measuring acidification of freshwater. *Nature*
35 278:542–545.

- 1 Herlihy, A.T., P.R. Kaufman, M.R. Church, P.J. Wigington, Jr., J.R. Webb, and M.J. Sale. 1993. The
2 effects of acidic deposition on streams in the Appalachian Mountain and Piedmont region of the
3 mid-Atlantic United States. *Water Resources Research* 29(8):2687–2703.
- 4 Heuer, K., K.A. Tonnessen, and G.P. Ingersoll. 2000. Comparison of precipitation chemistry in the
5 central Rocky Mountains, Colorado, USA. *Atmospheric Environment* 34:1713–1722.
- 6 Higgins, P.A.T., and J. Harte. 2006. Biophysical and biogeochemical responses to climate change depend
7 on dispersal and migration. *BioScience* 56:407–417.
- 8 Hodgkins, G.A., R.W. Dudley, and T.G. Huntington. 2003. Changes in the timing of high river flow in
9 New England over the 20th century. *Journal of Hydrology* 278:244–252.
- 10 Howarth, R.W. 2008. Estimating atmospheric nitrogen deposition in the northeastern United States:
11 Relevance to Narragansett Bay. Pp. 47–65, Chapter 3, in A. Desbonnet and B. A. Costa-Pierce,
12 editors. *Science for ecosystem-based management: Narragansett Bay in the 21st century*. Springer
13 Series on Environmental Management. New York: Springer.
- 14 Hrabik, T.R., and C.J. Watras. 2002. Recent declines in mercury concentration in a freshwater fishery:
15 isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little
16 Rock Lake. *Science of the Total Environment* 297:229–237.
- 17 Huang, Y., F. Dominici, and M.L. Bell. 2005. Bayesian hierarchical distributed lag models for summer
18 ozone exposure and cardio-respiratory mortality. *Environmetrics* 16:547–562.
- 19 Hudson, J.J., P.J. Dillon, and K.M. Somers. 2003. Long-term patterns in dissolved organic carbon in
20 boreal lakes: the role of incident radiation, precipitation, air temperature, southern oscillation and
21 acid deposition. *Hydrology & Earth System Science* 7:390–398.
- 22 Ibanez, I., J.S. Clark, S. Ladeau, and J.H.R. Lambers. 2007. Exploiting temporal variability to understand
23 tree recruitment response to climate change. *Ecological Monographs* 77:163–177.
- 24 ICF (ICF International, Inc.). 1989. *Economic Analysis of Title V (Acid Rain Provisions) of the*
25 *Administration's Proposed Clean Air Act Amendments (H.R. 3030/S. 1490)*. ICF International,
26 Inc., Fairfax, VA.
- 27 Ingersoll, G.P., M.A. Mast, D.H. Campbell, D.W. Clow, L. Nanus, and J.T. Turk. 2008. Trends in
28 snowpack chemistry and comparison to National Atmospheric Deposition Program results for the
29 Rocky Mountains, US, 1993–2004. *Atmospheric Environment* 42:6098–6113.
- 30 Interlandi, S.J., and S.S. Kilham. 2001. Limiting resources and the regulation of diversity in
31 phytoplankton communities. *Ecology* 82:1270–1282.
- 32 IPCC (Intergovernmental Panel on Climate Change). 2007. *Climate Change 2007: Synthesis Report*.
33 Intergovernmental Panel on Climate Change, Geneva, Switzerland.
- 34 Ito, K., S.F. De Leon, and M. Lippmann. 2005. Associations between ozone and daily mortality: Analysis
35 and meta-analysis. *Epidemiology* 16(4):446–57.
- 36 Jacob, D.J., and D.A. Winner. 2009. Effect of climate change on air quality. *Atmospheric Environment*
37 43:51–63.

- 1 Jeffries, D., and R. Ouimet. 2005. Critical loads – are they being exceeded? Pp. 341–368, Chapter 8, in
2 D. Jeffries and R. Ouimet, editors. *2004 Canadian Acid Deposition Science Assessment*.
3 Environment Canada, Ottawa, Ontario.
- 4 Jenkins, M.A., S. Jose, and P.S. White. 2007. Impacts of an exotic disease and vegetation change on foliar
5 calcium cycling in Appalachian forests. *Ecological Applications* 17:869–881.
- 6 Jentsch, A., J. Kreyling, and C. Beierkuhlein. 2007. A new generation of climate-change experiments:
7 Events, not trends. *Frontiers in Ecology & the Environment* 5:365–374.
- 8 Jeremiason, J.D., D.R. Engstrom, E.B. Swain, E.A. Nater, B.M. Johnson, J.E. Almendinger, B.A.
9 Monson, and R.K. Kolka. 2006. Sulfate addition increases methylmercury production in an
10 experimental wetland. *Environmental Science & Technology* 40:3800–3806.
- 11 Jeziorski, A., N.D. Yan, A.M. Paterson, A.M. DeSellas, M.A. Turner, D.S. Jeffries, B. Keller, R.C.
12 Weeber, D.K. McNicol, M.E. Palmer, K. McIver, K. Arseneau, B.K. Ginn, B.F. Cumming, and
13 J.P. Smol, 2008. The widespread threat of calcium decline in fresh waters. *Science* 322:1374–
14 1377.
- 15 Johnson, A.H., A.J. Moyer, J.E. Bedison, S.L. Richter, and S.A. Willig. 2008. Seven decades of calcium
16 depletion in organic horizons of Adirondack forest soils. *Soil Science Society of America Journal*
17 72:1824–1830.
- 18 Johnson, D.W. 2006. Progressive N limitation in forests: review and implications for long-term responses
19 to elevated CO₂. *Ecology* 87:64–75.
- 20 Jones, M.E., T.D. Paine, M.E. Fenn, and M.A. Poth. 2004. Influence of ozone and nitrogen deposition on
21 bark beetle activity under drought conditions. *Forest Ecology & Management* 200:67–76.
- 22 Jovan, S., and B. McCune, 2005. Air-quality bioindication in the greater central valley of California, with
23 epiphytic macrolichen communities. *Ecological Applications* 15:1712–1726.
- 24 Jovan, S. 2008. Lichen bioindication of biodiversity, air quality, and climate: Baseline results from
25 monitoring in Washington, Oregon, and California. *General Technical Report PNW-GTR-737*.
26 U.S. Department of Agriculture, Forest Service, Pacific Northwest Research Station, Portland,
27 OR.
- 28 Kahl, J.S., J.L. Stoddard, R. Haeuber, S.G. Paulsen, R. Birnbaum, F.A. Deviney, J.R. Webb, D.R.
29 DeWalle, W. Sharpe, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg, P.S. Murdoch, K. Roy, K.E.
30 Webster, and N.S. Urquhart. 2004. Have US surface waters responded to the 1990 Clean Air Act
31 Amendments? *Environmental Science & Technology* 38:484A–490A.
- 32 Kamman, N.C., P.M. Lorey, C.T. Driscoll, R. Estabrook, A. Major, B. Pientka, and E. Glassford. 2004.
33 Assessment of mercury in waters, sediments, and biota of New Hampshire and Vermont lakes,
34 USA, sampled using a geographically randomized design. *Environmental Toxicology &*
35 *Chemistry* 22:1172–1186.
- 36 Karnosky, D.F., J.M. Skelly, K.E. Percy, and A.H. Chappelka. 2007. Perspectives regarding 50 years of
37 research on effects of tropospheric ozone air pollution on U.S. forests. *Environmental Pollution*
38 147:489–506.

- 1 Kemp, W.M., W.R. Boynton, J.E. Adolf, D.F. Boesch, W.C. Boicourt, G. Brush, J.C. Cornwell, T.R.
2 Fisher, P.M. Glibert, J.D. Hagy, L.W. Harding, E.D. Houde, D.G. Kimmel, W.D. Miller, R.I.E.
3 Newell, M.R. Roman, E.M. Smith, and J.C. Stevenson. 2005. Eutrophication of Chesapeake Bay:
4 Historical trends and ecological interactions. *Marine Ecology Progress Series* 303:1–29.
- 5 Kline, K.M., K.N. Eshleman, R.P. Morgan II, and N.M. Castro. 2007. Analysis of trends in episodic
6 acidification of streams in western Maryland. *Environmental Science & Technology* 41:5601–
7 5607.
- 8 Knapp, A.K., B. Smith, S.D. Smith, D.D. Briske, P.A. Fay, S.W. Leavitt, M.D. Smith, Y. Luo, C. Beier,
9 M. Reichstein, J.L. Heisler, A.T. Classen, J.E. Bell, R. Sherry, and E. Weng. 2008. Consequences
10 of more extreme precipitation regimes for terrestrial ecosystems. *BioScience* 58:811–821.
- 11 Korner, C. 2003. Slow in, rapid out--carbon flux studies and Kyoto targets. *Science* 300:1242–1243.
- 12 Kowalik, R.A., D.M. Cooper, C.D. Evans, and S.J. Ormerod. 2007. Acidic episodes retard the biological
13 recovery of upland British streams from chronic acidification. *Global Change Biology* 13:2439–
14 2452.
- 15 Kramer, R.A., T.P. Holmes, and M. Haefele. 2003. Contingent valuation of forest ecosystem protection.
16 Pp. 303–320, Chapter 17, in E.O. Sills and K.L. Abt, editors. *Forests in a Market Economy*.
17 Kluwer:Dordrecht.
- 18 Krishnamurthy, A., J.K. Moore, N. Mahowald, C. Luo, and C.S. Zender. 2010. Impacts of atmospheric
19 nutrient inputs on marine biogeochemistry. *Journal of Geophysical Research* 115, GO1006, doi:
20 10.1029/2009JG001115.
- 21 Kruger, J., and M. Dean. 1997. Looking back on SO₂ trading: What’s good for the environment is good
22 for the market. *Public Utilities Reports* 135(15).
- 23 Krupnick, A., and J. Siikimaki. 2008. Valuation of the environmental effects of N and S (non-materials).
24 Annex F in *Integrated science assessment for oxides of nitrogen and sulfur – ecological criteria*.
25 Report EPA/600/R-08/082F. U.S. Environmental Protection Agency. Research Triangle Park,
26 NC.
- 27 Kvale, K.F., and S.C. Pryor. 2006. Precipitation composition in the Ohio River Valley: Spatial variability
28 and temporal trends. *Water, Air, & Soil Pollution* 170:143–160.
- 29 Laden, F., J. Schwartz, F.E. Speizer, and D.W. Dockery. 2006. Reduction in fine particulate air pollution
30 and mortality: Extended follow-up of the Harvard six cities study. *American Journal of*
31 *Respiratory and Critical Care Medicine* 173:667–672.
- 32 Langford, T.E.L., P.J. Shaw, A.J.D. Ferguson, and S.R. Howard. 2009. Long-term recovery of
33 macroinvertebrate biota in grossly polluted streams: Re-colonisation as a constraint to ecological
34 quality. *Ecological Indicators* 9:1064–1077.
- 35 Lawler, J.J., J. Rubin, B.J. Cosby, I.J. Fernandez, J.S. Kahl, and S.A. Norton. 2005. Predicting recovery
36 from acidic deposition: applying a modified TAF (Tracking and Analysis Framework) model to
37 Maine (USA) high elevation lakes. *Water, Air, & Soil Pollution* 164:383–399.

- 1 Lawrence, G.B., K.M. Roy, B.P. Baldigo, H.A. Simonin, S.B. Capone, J.W. Sutherland, S.A. Nierzwicki-
2 Bauer, and C.W. Boylen. 2008. Chronic and episodic acidification of Adirondack streams from
3 acid rain in 2003–2005. *Journal of Environmental Quality* 37:2264–2274.
- 4 Lawrence, G.B., D.A. Goolsby, W.A. Battaglin, and G.J. Stensland. 2000. Atmospheric nitrogen in the
5 Mississippi River basin—emissions, deposition, and transport. *Science of the Total Environment*
6 248:87–99.
- 7 Lawrence, G.B., M.B. David, and W.C. Shortle. 1995. A new mechanism for calcium loss in forest-floor
8 soils. *Nature* 378:162–165.
- 9 Lawrence, G.B., M.B. David, G.M. Lovett, P.S. Murdoch, D.A. Burns, J.L. Stoddard, B.P. Baldigo, J.H.
10 Porter, and A.W. Thompson. 1999. Soil calcium status and the response of stream chemistry to
11 changing acid deposition rates. *Ecological Applications* 9:1059–1072.
- 12 Lawrence, G.B. 2002. Persistent episodic acidification of streams linked to acid rain effects on soil.
13 *Atmospheric Environment* 36(10):1589–1598
- 14 Lehmann, C.M.B., V.C. Bowersox, and S.M. Larson. 2005. Spatial and temporal trends of precipitation
15 chemistry in the United States, 1985–2002. *Environmental Pollution* 135:347–361.
- 16 Lenoir, J., J.C. Gegout, P.A. Marquet, P. de Ruffray, and H. Brisse. 2008. A significant shift in plant
17 species optimum elevations during the 20th century. *Science* 320:1768–1771.
- 18 Levy, J.I., S.M. Chemerynski, and J.A. Sarnat. 2005. Ozone exposure and mortality: An empiric bayes
19 metaregression analysis. *Epidemiology* 16(4):458–68.
- 20 Li, H., and S.G. McNulty. 2007. Uncertainty analysis on simple mass balance model to calculate critical
21 loads for soil acidity. *Environmental Pollution* 149:315–326.
- 22 Likens, G.E., C.T. Driscoll, and D.C. Buso. 1996. Long-term effects of acid rain: response and recovery
23 of a forest ecosystem. *Science* 272:244–246.
- 24 Long, R.P., S.B. Horsley, R.A. Hallett, and S.W. Bailey. 2009. Sugar maple growth in relation to
25 nutrition and stress in the northeastern United States. *Ecological Applications* 19:1454–1466.
- 26 Magnani, F., M. Mencuccini, M. Borghetti, P. Berbigier, F. Berninger, S. Delzon, A. Grelle, P. Hari, P.G.
27 Jarvis, P. Kolari, A.S. Kowalski, H. Lankreijer, B.E. Law, A. Lindroth, D. Loustau, G. Manca,
28 J.B. Moncrieff, M. Rayment, V. Tedeschi, R. Valentini, and J. Grace. 2007. The human footprint
29 in the carbon cycle of temperate and boreal forests. *Nature* 447:848–850.
- 30 Mast, M.A., J.T. Turk, D.W. Clow, and D.H. Campbell. In press. Response of lake chemistry to changes
31 in atmospheric deposition and climate in three high-elevation wilderness areas of Colorado. *Earth*
32 *and Environmental Science*.
- 33 McIsaac, G.F., M.B. David, G.Z. Gertner, and D.A. Goolsby. 2001. Eutrophication: nitrate flux in the
34 Mississippi River. *Nature* 414:166–167.
- 35 McKenney, D.W., J.H. Pedlar, K. Lawrence, K. Campbell, and M.F. Hutchinson. 2007. Potential impacts
36 of climate change on the distribution of North American trees. *BioScience* 57:939–948.

- 1 McLaughlin, S., and K. Percy. 1999. Forest health in North America: some perspectives on actual and
2 potential roles of climate and air pollution. *Water, Air, & Soil Pollution* 116:151–197.
- 3 McLaughlin, S.B., M.G. Tjoelker, and W.K. Roy. 1993. Acid deposition alters red spruce physiology:
4 Laboratory studies support field observations. *Canadian Journal of Forest Research* 23:380–386.
- 5 McLean, B.J. 1996. *The Sulfur Dioxide (SO₂) Allowance Trading Program: The First Five Years*. U.S.
6 Environmental Protection Agency, Washington, DC.
- 7 McNulty, S.G., E.C. Cohen, J.A. Moore Myers, T.J. Sullivan, and H. Li. 2007. Estimates of critical acid
8 loads and exceedances for forest soils across the conterminous United States. *Environmental*
9 *Pollution* 149:281–292.
- 10 Meixner, T., C. Gutmann, R. Bales, A. Leydecker, J. Sickman, J. Melack, and J. McConnell. 2004.
11 Multidecadal hydrochemical response of a Sierra Nevada watershed: sensitivity to weathering
12 rate and changes in deposition. *Journal of Hydrology* 285:272–285.
- 13 Meixner, T., M.E. Fenn, P. Wohlgemuth, M. Oxford, and P. Riggan. 2006. N saturation symptoms in
14 chaparral catchments are not reversed by prescribed fire. *Environmental Science & Technology*
15 40:2887–2894.
- 16 Michalski, G., T. Meixner, M. Fenn, L. Hernandez, A. Sirulnik, E. Allen, and M. Thiemens. 2004.
17 Tracing atmospheric nitrate deposition in a complex semiarid ecosystem using $\Delta^{17}\text{O}$.
18 *Environmental Science & Technology* 38:2175–2181.
- 19 Mickley, L.J., P.P. Murti, D.J. Jacob, J.A. Logan, D.M. Koch, and D. Rind. 1999. Radiative forcing from
20 tropospheric ozone calculated with a unified chemistry-climate model. *Journal of Geophysical*
21 *Research* 104:30153–30172.
- 22 Milly, P.C.D., K.A. Dunne, and A.V. Vecchia. 2005. Global pattern of trends in streamflow and water
23 availability in a changing climate. *Nature* 438:347–350.
- 24 Mohan, J.E., R.M. Cox, and L.R. Inverson. 2009. Composition and carbon dynamics of forests in
25 northeastern North America in a future, warmer world. *Canadian Journal of Forest Research*
26 39:213–230.
- 27 Mohseni, O., H.G. Stefan, and J.G. Eaton. 2003. Global warming and potential changes in fish habitat in
28 U.S. streams. *Climatic Change* 59:389–409.
- 29 Monteith, D.T., A.G. Hildrew, R.J. Flower, P.J. Raven, W.R.B. Beaumont, P. Collen, A.M. Kreiser, E.M.
30 Shilland, and J.H. Winterbottom. 2005. Biological responses to the chemical recovery of acidified
31 fresh waters in the UK. *Environmental Pollution* 137:83–101.
- 32 Monteith, D.T., J.L. Stoddard, C.D. Evans, H.A. de Wit, M. Forsius, T. Høgåsen, A. Wilander, B.L.
33 Skjelkvåle, D.S. Jeffries, J. Vuorenmaa, B. Keller, J. Kopáček, and J. Vesely. 2007. Dissolved
34 organic carbon trends resulting from changes in atmospheric deposition chemistry. *Nature*
35 450:537–540.
- 36 Moore, M.V., M.L. Pace, J.R. Mather, P.S. Murdoch, R.W. Howarth, C.L. Folt, C.Y. Chen, H.F.
37 Hemond, P.A. Flebbe, and C.T. Driscoll. 1997. Potential effects of climate change on freshwater
38 ecosystems of the New England/Mid-Atlantic region. *Hydrological Processes* 11:925–947.

- 1 Morgan, C., and N. Owens. 2001. Benefits of water quality policies: the Chesapeake Bay. *Ecological*
2 *Economics* 39:271–284.
- 3 Mosello, R., A. Lami, A. Marchetto, M. Rogora, B. Wathne, L. Lien, J. Catalan, L. Camarero, M.
4 Ventura, R. Psenner, K. Koinig, H. Thies, S. Sommaruga-Wögrath, U. Nickus, D. Tait, B. Thaler,
5 A. Barbieri, and R. Harriman. 2002. Trends in the water chemistry of high altitude lakes in
6 Europe. *Water, Air, & Soil Pollution Focus* 2:75–89.
- 7 Mote, P.W., A.F. Hamlet, M.P. Clark, and D.P. Lettenmaier. 2005. Declining mountain snowpack in
8 western North America. *Bulletin of the American Meteorological Society* 86:39–49.
- 9 Munthe, J., R.A. Bodaly, B.A. Branfireun, C.T. Driscoll, C.C. Gilmour, R. Harris, M. Horvat, M. Lucotte,
10 and O. Malm. 2007. Recovery of Mercury-Contaminated Fisheries. *Ambio* 36:33–44.
- 11 Murdoch, P.S., J.S. Baron, and T.L. Miller. 2000. Potential effects of climate change on surface-water
12 quality in North America. *Journal of the American Water Resources Association* 36:347–366.
- 13 NADP (National Atmospheric Deposition Program). 2009. *CLAD – Critical Loads Ad Hoc*
14 *Subcommittee*. Online information. National Atmospheric Deposition Program, Washington, D.C.
15 Available at: <http://nadp.sws.uiuc.edu/clad/> (accessed August 25, 2009).
- 16 Nanus, L., D.H. Campbell, G.P. Ingersoll, D.W. Clow, and M.A. Mast. 2003. Atmospheric deposition
17 maps for the Rocky Mountains. *Atmospheric Environment* 37:4881–4892.
- 18 NARSTO. 2010. *Technical Challenges of Risk – and Results-Based Multipollutant Air Quality*
19 *Management*. Edited by G. Hidy, J.R., Brook, K.L., Demerjian, L.T., Molina, W.T., Pennell, and
20 R. Scheffe, R., Springer, Dordrecht, Netherlands.
- 21 Nemani, R.R., C.D. Keeling, H. Hashimoto, W.M. Jolly, S.C. Piper, C.J. Tucker, R.B. Myneni, and S.W.
22 Running. 2003. Climate-driven increases in global terrestrial primary production from 1982 to
23 1999. *Science* 300:1560–1563.
- 24 Nilsson, J., and P. Grennfelt (eds). 1988. *Critical Loads for Sulphur and Nitrogen*. UNECE/Nordic
25 Council Workshop Report, Skokloster, Sweden, March . Nordic Council of Ministers,
26 Copenhagen.
- 27 Norby, R.J., and Y. Luo. 2004. Evaluating ecosystem responses to rising atmospheric CO₂ and global
28 warming in a multi-factor world. *New Phytologist* 162:281–293.
- 29 Norby, R.J., E.H. DeLucia, B. Gielen, C. Calfapietra, C.P. Giardina, J.S. King, J. Ledford, H.R.
30 McCarthy, D.J.P. Moore, R. Ceulemans, P. De Angelis, A.C. Finzi, D.F. Karnosky, M.E.
31 Kubiske, M. Lukac, K.S. Pregitzer, G.E. Scarascia-Mugnozza, W.H. Schlesinger, and R. Oren.
32 2005. Forest response to elevated CO₂ is conserved across a broad range of productivity.
33 *Proceedings of the National Academy of Sciences* 102:18052–18056.
- 34 Nowak, R.S., D.S. Ellsworth, and S.D. Smith. 2004. Functional responses of plants to elevated CO₂ – do
35 photosynthetic and productivity data from FACE experiments support early predictions? *New*
36 *Phytologist* 162:253–280.
- 37 NRC (National Research Council). 2004. *Air Quality Management in the United States*. The National
38 Academies Press, Washington, DC.

- 1 NSTC (National Science and Technology Council). 1998. *NAPAP Biennial Report to Congress: An*
2 *Integrated Assessment*. Executive Office of the President, National Science and Technology
3 Council, National Acid Precipitation Assessment Program, Silver Spring, MD. Available at
4 <http://www.p2pays.org/ref/11/10588/>.
- 5 NSTC (National Science and Technology Council). 2005. *National Acid Precipitation Assessment*
6 *Program Report to Congress: An Integrated Assessment*. Executive Office of the President,
7 National Science and Technology Council, Washington, DC. Available at
8 <http://www.esrl.noaa.gov/csd/aqrs/reports/napapreport05.pdf>.
- 9 Oltmans, S.J., A.S. Lefohn, J.M. Harris, I. Galbally, H.E. Scheel, G. Bodeker, E. Brunke, H. Claude, D.
10 Tarasick, B.J. Johnson, P. Simmonds, D. Shadwick, K. Anlauf, K. Hayden, F. Schmidlin, T.
11 Fujimoto, K. Akagi, C. Meyer, S. Nichol, J. Davies, A. Redondas, and E. Cuevas. 2006. Long-
12 term changes in tropospheric ozone. *Atmospheric Environment* 40:3156–3173.
- 13 Pardo, L.H., M.J. Robin-Abbott, and C.T. Driscoll (editors). In press. *Assessment of Effects of Nitrogen*
14 *Deposition Effects and Empirical Critical Loads for Nitrogen for Ecoregions of the United*
15 *States*. USDA Forest Service General Technical Report.
- 16 Passy, S., I. Ciugulea, and G.B. Lawrence. 2006. Diatom diversity in chronically versus episodically
17 acidified Adirondack streams. *International Review of Hydrobiology* 91:594–608.
- 18 Poor, N., C. Pollman, P. Tate, M. Begum, M. Evans, and S. Campbell. 2006. Nature and magnitude of
19 total inorganic nitrogen and other inorganic species to the Tampa Bay watershed, FL, USA.
20 *Water, Air, & Soil Pollution* 170:267–83.
- 21 Pope, C.A., R.T. Burnett, M.J. Thun, E.E. Calle, D. Krewski, K. Ito, and G.D. Thurston. 2002. Lung
22 cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution.
23 *Journal of the American Medical Association* 287(9):1132–1141.
- 24 Porter, E., and S. Johnson. 2007. Translating science into policy: Using ecosystem thresholds to protect
25 resources in Rocky Mountain National Park. *Environmental Pollution* 149:268–280. (ch3)
- 26 Porter, E., T. Blett, D.U. Potter, and C. Huber. 2005. Protecting resources on federal lands: Implications
27 of critical loads for atmospheric deposition of nitrogen and sulfur. *BioScience* 55:603–612.
- 28 Posch, M. 2002. Impacts of climate change on critical loads and their exceedances in Europe.
29 *Environmental Science & Policy* 5:307–317.
- 30 Potter, C., S. Klooster, R.R. Nemani, V. Genovese, S. Hiatt, M. Fladeland, and P. Gross. 2006. Estimating
31 carbon budgets for U.S. ecosystems. *Eos, Transactions of the American Geophysical Union*
32 87:85, 90.
- 33 Raddum, G.G., and A. Fjellheim. 2003. Liming of River Audna, southern Norway: A large-scale
34 experiment of benthic invertebrate recovery. *Ambio* 32: 230–234.
- 35 Reich, P.B., B.A. Hungate, and Y. Luo. 2006. Carbon-nitrogen interactions in terrestrial ecosystems in
36 response to rising atmospheric carbon dioxide. *Annual Review of Ecology, Evolution, &*
37 *Systematics* 37:611–636

- 1 Rice, K.C., F.A. Deviney Jr., G.M. Hornberger, and J.R. Webb. 2006. Predicting the vulnerability of
2 streams to episodic acidification and potential effects on aquatic biota in Shenandoah National
3 Park, Virginia. *U.S. Geological Survey Scientific Investigations Report 2005-5259*, U.S.
4 Geological Survey, Reston, VA.
- 5 Robinson, B., T.W. Barnett, G.R. Harwell, S.E. Moore, M. Kulp, and J.S. Schwartz. 2008. pH and acid
6 anion time trends in different elevation ranges in the Great Smoky Mountains National Park
7 *Journal of Environmental Engineering* 134(9):800–808.
- 8 Rogora, M., R. Mosello, and S. Arisci. 2003. The effect of climate warming on the hydrochemistry of
9 alpine lakes. *Water, Air, & Soil Pollution* 148:347–361.
- 10 Rustad, L.E. 2008. The responses of terrestrial ecosystems to global climate change: Towards and
11 integrated approach. *Science of the Total Environment* 404:222–235.
- 12 Saros, J.E., T.J. Michel, S.J. Interlandi, and A.P. Wolfe. 2005. Resource requirements of *Asterionella*
13 *formosa* and *Fragilaria crotonensis* in oligotrophic alpine lakes: Implications for recent
14 phytoplankton community reorganizations. *Canadian Journal of Fisheries & Aquatic Sciences*
15 62:1681–1689.
- 16 Schaefer, S.C., and M. Alber. 2007. Temperature controls a latitudinal gradient in the proportion of
17 watershed nitrogen exported to coastal ecosystems. *Biogeochemistry* 85:333–346.
- 18 Schiff, S.L., J. Spoelstra, R.G. Semkin, and D.S. Jeffries. 2005. Drought induced pulses of SO_4^{2-} from a
19 Canadian shield wetland: Use of $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ in SO_4^{2-} to determine sources of sulfur. *Applied*
20 *Geochemistry* 20:691–700.
- 21 Schindler, D.W. 1998. A dim future for boreal waters and landscapes: cumulative effects of climatic
22 warming stratospheric ozone depletion, acid precipitation, and other human activities. *BioScience*
23 48:157–164.
- 24 Schofield, C.L. 1976. Acid precipitation: Effects on fish. *Ambio* 5:228–230.
- 25 Schwartz, J. 2005. How sensitive is the association between ozone and daily deaths to control for
26 temperature? *American Journal of Respiratory and Critical Care Medicine* 171(6):627–31.
- 27 Schwede, D.B., R.L. Dennis, and M.A. Bitz. 2009. The watershed deposition tool: a tool for incorporating
28 atmospheric deposition in water-quality analyses. *Journal of the American Water Resources*
29 *Association* 45:973–985.
- 30 Scudder, B.C., L.C. Chasar, D.A. Wentz, N.J. Bauch, M.E. Brigham, P.W. Moran, and D.P. Krabbenhoft.
31 2009. Mercury in fish, bed sediment, and water from streams across the United States, 1998–
32 2005. *U.S. Geological Survey Scientific Investigations Report 2009-5109*, Reston, VA.
- 33 Seastedt, T.R., and L. Vaccaro. 2001. Plant species richness, productivity, and nitrogen and phosphorus
34 limitations across a snowpack gradient in alpine tundra, Colorado, U.S.A. *Arctic, Antarctic, &*
35 *Alpine Research* 33:100–106.
- 36 Sickman, J.O., A. Leydecker, C.C.Y. Chang, C. Kendall, J.M. Melack, D.M. Lucero, D.M., and J.
37 Schimel. 2003. Mechanisms underlying export of N from high-elevation catchments during
38 seasonal transitions. *Biogeochemistry* 64:1–24.

- 1 Sickman, J.O., J.M. Melack, J.L. Stoddard. 2002. Regional analysis of nitrogen yield and retention in
2 high-elevation ecosystems of the Sierra Nevada and Rocky Mountains. *Biogeochemistry* 57:341–
3 374.
- 4 Simonin, H.A., J.R. Colquhoun, E.A. Paul, J. Symula, and H.J. Dean. 2005. Have Adirondack stream fish
5 populations changed in response to decreases in sulfate deposition? *Transactions of the American*
6 *Fisheries Society* 134:338–345.
- 7 Sitch, S., P.M. Cox, W.J. Collins, and C. Huntingford. 2007. Indirect radiative forcing of climate change
8 through ozone effects on the land-carbon sink. *Nature* 448:791–794.
- 9 Skeffington, R.A. 2006. Quantifying uncertainty in critical loads: (A) literature review. *Water, Air, & Soil*
10 *Pollution* 169:3–24.
- 11 Snucins, E., and J. Gunn. 2003. Use of rehabilitation experiments to understand the recovery dynamics of
12 acid-stressed fish populations. *Ambio* 32:240–243.
- 13 Snucins, E., and J.M. Gunn. 2000. Interannual variation in the thermal structure of clear and colored
14 lakes. *Limnology & Oceanography* 45:1647–1654.
- 15 Snucins, E. 2003. Recolonization of acid-damaged lakes by the benthic invertebrates *Stenacron*
16 *interpunctatum*, *Stenonema femoratum* and *Hyalella azteca*. *Ambio* 32:225–229.
- 17 Sommaruga-Wogarth, S., K.A. Koinig, R. Schmidt, R. Sommaruga, R. Tessadri, and R. Psenner. 1997.
18 Temperature effects on the acidity of alpine lakes. *Nature* 387:64–67.
- 19 Sparks, J.P., J. Walker, A. Turnipseed, and A. Guenther. 2008. Dry nitrogen deposition estimates over a
20 forest experiencing free air CO₂ enrichment. *Global Change Biology* 14:768–781.
- 21 Spranger, T., J.-P. Hettelingh, J. Slootweg, and M. Posch. 2008. Modelling and mapping long-term risks
22 due to reactive nitrogen effects: an overview of LRTAP convention activities. *Environmental*
23 *Pollution* 154:482–487.
- 24 St. Clair, S.B., W.E. Sharpe, and J.P. Lynch. 2008. Key interactions between nutrient limitation and
25 climatic factors in temperate forests: a synthesis of the sugar maple literature. *Canadian Journal*
26 *of Forest Research* 38:401–414.
- 27 Striegl, R.G., G.R. Aiken, M.M. Dornblaser, P.A. Raymond, and K.P. Wikland. 2005. A decrease in
28 discharge-normalized DOC export by the Yukon River during summer through autumn.
29 *Geophysical Research Letters* 32:L21413, doi: 10.1029/2005GL024413.
- 30 Stephen, K., and V.P. Aneja. 2008. Trends in agricultural ammonia emissions and ammonium
31 concentrations in precipitation over the Southeast and Midwest United States. *Atmospheric*
32 *Environment* 42:3238–3252.
- 33 Stoddard, J.L., J.S. Kahl, F.A. Deviney, D.R. DeWalle, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg, P.S.
34 Murdoch, J.R. Webb, and K.E. Webster. 2003. *Response of surface water chemistry to the Clean*
35 *Air Act Amendments of 1990*. EPA-620-R-03-001, U.S. Environmental Protection Agency,
36 Washington, DC.

- 1 Striegl, R.G., G.R. Aiken, M.M. Dornblaser, P.A. Raymond, and K.P. Wikland. 2005. A decrease in
2 discharge-normalized DOC export by the Yukon River during summer through autumn.
3 *Geophysical Research Letters* 32:L21413.
- 4 Sullivan, T.J., B.J. Cosby, A.T. Herlihy, J.R. Webb, A.J. Bulger, K.U. Snyder, P.F. Brewer, E.H. Gilbert,
5 and D.L. Moore. 2004. Regional model projections of future effects of sulfur and nitrogen
6 deposition on streams in the southern Appalachian Mountains. *Water Resources Research*
7 40:W02101.
- 8 Sullivan, T.J., B.J. Cosby, J.R. Webb, R.L. Dennis, A.J. Bulger, and F.A. Deviney, Jr. 2008. Streamwater
9 acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National
10 Park, Virginia. *Environmental Monitoring & Assessment* 137:85–99.
- 11 Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger, K.U.
12 Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell, and N.A.
13 Nowicki. 2006a. *Assessment of the Extent to which Intensively-Studied Lakes are Representative*
14 *of the Adirondack Mountain Region*. Final Report. New York State Energy Research and
15 Development Authority (NYSERDA), Albany, NY. Available at
16 <http://nysl.nysed.gov/uhtbin/cgisirsi/Qcwd6NzFby/NYSL/138650099/8/4298474> (accessed
17 November 1, 2007).
- 18 Sullivan, T.J., I.J. Fernandez, A.T. Herlihy, C.T. Driscoll, T.C. McDonnell, N.A. Nowicki, K.U. Snyder,
19 and J.W. Sutherland. 2006b. Acid-base characteristics of soils in the Adirondack Mountains, New
20 York. *Soil Science Society of America Journal* 70:141–152.
- 21 Sullivan T.J., B.J. Cosby, J.R. Webb, R.L. Dennis, A.J. Bulger, and F.A. Deviney, Jr. 2007. Streamwater
22 acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National
23 Park, Virginia. *Environmental Monitoring and Assessment* 137:85–99.
- 24 Sutton, M.A., D. Simpson, P.E. Levy, R.I. Smith, S. Ris, M. van Oijen, and W. deVries. 2008.
25 Uncertainties in the relationship between atmospheric nitrogen deposition and carbon
26 sequestration. *Global Change Biology* 14: 2057–2063.
- 27 Sverdrup, H., and W. deVries. 1994. Calculating critical loads for acidity with the simple mass balance
28 method. *Water, Air, & Soil Pollution* 72:143–162.
- 29 Tagaris, E., K.-J. Liao, K. Manomaiphiboon, J.-K. Woo, S. He, P. Amar, and A.G. Russell. 2008. Impacts
30 of future climate change and emissions reductions on nitrogen and sulfur deposition over the
31 United States. *Geophysical Research Letters* 35, L08811, doi:10.1029/2008GL033477.
- 32 Tans, P.P., I.Y. Fung, and T. Takahashi. 1990. Observational constraints on the global atmospheric CO₂
33 budget. *Science* 247:1431–1438.
- 34 Thomas, R.Q. Canham, C.D., Weathers, K.C., and C.L. Goodale. 2009. Increased tree carbon storage in
35 response to nitrogen deposition in the US. *Nature Geoscience* 3:13–17.
- 36 Tipping, E., E.J. Smith, A.J. Lawlor, S. Hughes, and P.A. Stevens. 2003. Predicting the release of metals
37 from ombrotrophic peat due to drought-induced acidification. *Environmental Pollution* 123:239–
38 253.

- 1 Tipping, E., J.A. B. Bass, and D. Hardie. 2002. Biological responses to the reversal of acidification in
2 surface waters of the English Lake District. *Environmental Pollution* 116:137–146.
- 3 Todorova, S.G., C.T. Driscoll Jr., D.A. Matthews, S.W. Effler, M.E. Hines, and E.A. Henry. 2009.
4 Evidence for regulation of monomethyl mercury by nitrate in a seasonally stratified, eutrophic
5 lake. *Environmental Science & Technology* 43:6572–6578.
- 6 U.S. EPA (Environmental Protection Agency). 2001. *Visibility in Mandatory federal Class I Areas (1994-
7 1998): A Report to Congress*. EPA-452-R-01-008. U.S. Environmental Protection Agency, Office
8 of Air Quality Planning and Standards, Research Triangle Park, NC.
- 9 U.S. EPA (Environmental Protection Agency). 2003. *Guidance for Tracking Progress Under the
10 Regional Haze Rule*. EPA-454/B-03-004. U.S. Environmental Protection Agency, Office of Air
11 Quality Planning and Standards, Research Triangle Park, NC. Available at
12 <http://www.epa.gov/ttnamti1/files/ambient/visible/tracking.pdf>.
- 13 U.S. EPA (Environmental Protection Agency). 2006a. *Regulatory Impact Analysis for the Review of the
14 Particulate Matter National Ambient Air Quality Standards*. U.S. Environmental Protection
15 Agency, Washington, DC.
- 16 U.S. EPA (Environmental Protection Agency). 2006b. *Technical Support Document for the Proposed PM
17 NAAQA Rule: Response Surface Modeling*. U.S. Environmental Protection Agency, Office of Air
18 Quality Planning and Standards, Research Triangle Park, NC.
- 19 U.S. EPA (Environmental Protection Agency). 2007a. *2005/2006 National Listing of Fish Advisories*.
20 U.S. Environmental Protection Agency, Washington, DC.
- 21 U.S. EPA (Environmental Protection Agency). 2009a. *Acid Rain and Related Programs: 2007 Highlights*.
22 EPA-430-K-08-010. U.S. Environmental Protection Agency, Washington, DC.
23 <http://www.epa.gov/airmarkt/progress/docs/2007ARPreport.pdf>
- 24 U.S. EPA (Environmental Protection Agency). 2008a. *Integrated Science Assessment for Oxides of
25 Nitrogen and Sulfur Review*. EPA/600/R-08/082F. U.S. Environmental Protection Agency,
26 National Center for Environmental Assessment-RTP Division, Office of Research and
27 Development, Research Triangle Park, NC. Available at
28 <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.
- 29 U.S. EPA (Environmental Protection Agency). 2008b. *Acid Rain and Related Programs: 2008
30 Highlights*. EPA-430-R-09-031. U.S. Environmental Protection Agency, Office of Air and
31 Radiation, Clean Air Markets, Washington, DC. Available at
32 http://www.epa.gov/airmarkt/progress/ARP_2008_Highlights.pdf.
- 33 U.S. EPA (Environmental Protection Agency). 2008c. *BenMAP 3.0 Beta*. Software. U.S. Environmental
34 Protection Agency, Office of Air Quality Planning and Standards, Health and Environmental
35 Impact Division, Air Benefit-Cost Group, Research Triangle Park, NC.
- 36 U.S. EPA (Environmental Protection Agency). 2009a. *Acid Rain and Related Programs: 2008 Emission,
37 Compliance, and Market Analyses*. U.S. Environmental Protection Agency, Washington, DC.
38 September. Available at http://www.epa.gov/airmarkt/progress/ARP_2.html.

- 1 U.S. EPA (Environmental Protection Agency). 2009b. *Acid Rain and Related Programs: 2008 Emission,*
2 *Compliance and Market Data.* U.S. Environmental Protection Agency, Washington, DC. July.
3 Available at http://www.epa.gov/airmarkt/progress/ARP_1.html.
- 4 U.S. EPA (Environmental Protection Agency). 2009c. *Acid Rain and Related Programs: 2008*
5 *Environmental Results.* U.S. Environmental Protection Agency, Washington, DC. Available at
6 http://www.epa.gov/airmarkt/progress/ARP_3.html
- 7 U.S. EPA (Environmental Protection Agency). 2009d. *Acid Rain and Related Programs: 2007*
8 *Highlights.* EPA-430-K-08-010. U.S. Environmental Protection Agency, Washington, DC.
9 Available at http://www.epa.gov/airmarkt/progress/ARP_2008_Highlights.pdf.
- 10 U.S. EPA (Environmental Protection Agency). 2009e. *Acid Rain and Related Programs: 2007 Progress*
11 *Report.* U.S. Environmental Protection Agency, Washington, DC. Available at
12 <http://www.epa.gov/airmarkt/progress/arp07.html>.
- 13 U.S. EPA (Environmental Protection Agency). 2009f. *I. NO_x Budget Trading Program – Basic*
14 *Information.* U.S. Environmental Protection Agency, Washington, DC. Available at
15 <http://www.epa.gov/airmarkt/progsregs/nox/sipbasic.html>.
- 16 U.S. EPA (Environmental Protection Agency). 2009g. *Risk and Exposure Assessment for Review of the*
17 *Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur.*
18 EPA-452/R-09-008a. U.S. Environmental Protection Agency, Office of Air Quality Planning and
19 Standards, Health and Environmental Impacts Division, Research Triangle Park, NC. Available at
20 http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr_rea.html.
- 21 U.S. EPA (Environmental Protection Agency). 2010a. *Final Regulatory Impact Analysis (RIA) for the*
22 *SO₂ National Ambient Air Quality Standards (NAAQS).* U.S. Environmental Protection Agency,
23 Office of Air Quality Planning and Standards, Health and Environmental Impact Division, Air
24 Benefit-Cost Group, Research Triangle Park, NC.
- 25 U.S. EPA (Environmental Protection Agency). 2010b. *The Benefits and Costs of the Clean Air Act: 1990*
26 *to 2020.* Preliminary Draft Report. U.S. Environmental Protection Agency, Office of Air and
27 Radiation, Washington, DC. Available at <http://www.epa.gov/air/sect812/>.
- 28 U.S. GAO (Government Accounting Office). 1994. *Allowance Trading Offers an Opportunity to Reduce*
29 *Emissions at Less Cost.* U.S. General Accounting Office, Washington, DC.
- 30 van Groenigen, K.-J., J. Six, B.A. Hungate, M.-A. de Graff, N. van Breemen, and C. van Kessel. 2006.
31 Element interactions limit soil carbon storage. *Proceedings of the National Academy of Sciences*
32 *103:6571–6574.*
- 33 Vingarzan, R. 2004. A review of surface ozone background levels and trends. *Atmospheric Environment*
34 *38:3431–3442.*
- 35 Walsh, R.G., R.D. Bjornback, R.A. Aiken, and D.H. Rosenthal. 1990. Estimating the public benefits of
36 protecting forest quality. *Journal of Environmental Management* *30:175–189.*
- 37 Warby, R.A.F., C.E., Johnson, and C.T. Driscoll. 2005. Chemical recovery of surface waters across the
38 northeastern United States from reduced inputs of acidic deposition: 1984–2001. *Environmental*
39 *Science and Technology* *39:6548–6554.*

- 1 Warby, R.A.F., C.E. Johnson, and C.T. Driscoll. 2009. Continuing acidification of organic soils across
2 the northeastern USA: 1984 – 2001. *Soil Science Society of America Journal* 73:274–284.
- 3 Warren, D.R., G.E. Likens, D.C. Buso, and C.E. Kraft. 2008. Status and distribution of fish in an acid-
4 impacted watershed of the Northeastern United States (Hubbard Brook, NH). *Northeast*
5 *Naturalist* 15:375–390.
- 6 Watmough, S.A., J. Aherne, and P.J. Dillon. 2003. Potential impact of forest harvesting on lake chemistry
7 in south-central Ontario at current levels of acid deposition. *Canadian Journal of Fisheries and*
8 *Aquatic Sciences* 60:1095–1103.
- 9 Watmough, S.A., M.C. Eimers, J. Aherne, and P.J. Dillon. 2004. Climate effects on stream nitrate at 16
10 forested catchments in south central Ontario. *Environmental Science & Technology* 38:2383–
11 2388.
- 12 Weathers, K.C., S.M. Simkin, G.M. Lovett, and S.E. Lindberg. 2006. Empirical modeling of atmospheric
13 deposition in mountainous landscapes. *Ecological Applications* 16:1590–1607.
- 14 Webb, J.R., B.J. Cosby, F.A. Deviney, J.N. Galloway, S.W. Maben, and A.J. Bulger. 2004. Are brook
15 trout streams in western Virginia and Shenandoah National Park recovering from acidification?
16 *Environmental Science & Technology* 38:4091–4096.
- 17 Weiss, S.B. 2006. *Impacts of nitrogen deposition on California ecosystems and biodiversity*. CEC-500-
18 2005-165. California Energy Commission, PIER Energy-Related Environmental Research,
19 Sacramento, CA.
- 20 Wentz, F.J., L. Ricciardulli, K. Hilburn, and C. Mears. 2007. How much more rain will global warming
21 bring? *Science* 317:233–235.
- 22 Whitall, D., B. Hendrickson, and H. Paerl. 2003. Importance of atmospherically deposited nitrogen to the
23 annual nitrogen budget of the Neuse River estuary, North Carolina. *Environment International*
24 29:393–399.
- 25 Whitall, D., S. Bricker, J. Ferreira, A.M. Nobre, T. Simas, and M. Silva. 2007. Assessment of
26 eutrophication in estuaries: Pressure-state-response and nitrogen source apportionment.
27 *Environmental Management* 40:678–690.
- 28 White, A.F., A.E. Blum, T.D. Bullen, D.V. Vivit, M. Schultz, and J. Fitzpatrick. 1999. The effect of
29 temperature on experimental and natural chemical weathering rates in granitoid rocks.
30 *Geochimica et Cosmochimica Acta* 63:3277–3291.
- 31 Wiener, J.G., B.C. Knights, M.B. Sandheinrich, J.D. Jeremiason, M.E. Brigham, D.R. Engstrom, L.G.
32 Woodruff, W.F. Cannon and S.J. Balogh. 2006 Mercury in Soils, Lakes, and Fish in Voyageurs
33 National Park (Minnesota): importance of Atmospheric Deposition and Ecosystem Factors.
34 *Environmental Science & Technology* 40:6261–6281.
- 35 Williams, M.W., and K.A. Tonnessen. 2000. Critical loads for inorganic nitrogen deposition in the
36 Colorado Front Range, USA. *Ecological Applications* 10:1648–1665.

- 1 Wolfe, A.P., A.C. Van Gorp, and J.S. Baron. 2003. Recent ecological and biogeochemical changes in
2 alpine lakes of Rocky Mountain National Park (Colorado, USA): a response to anthropogenic
3 nitrogen deposition. *Geobiology* 1:153–168.
- 4 Wolfe, A.P., A.C. Van Gorp, and J.S. Baron. 2003. Recent ecological and biogeochemical changes in
5 alpine lakes of Rocky Mountain National Park (Colorado, USA): A response to anthropogenic
6 nitrogen deposition. *Geobiology* 1:153–168.
- 7 Woodruff, T.J., J.D. Parker, and K.C. Schoendorf. 2006. Fine particulate matter (PM_{2.5}) air pollution and
8 selected causes of postneonatal infant mortality in California. *Environmental Health Perspectives*
9 114(5):786–790.
- 10 Worall, F., and T.P. Burt. 2007. Flux of dissolved organic carbon from U.K. rivers. *Global*
11 *Biogeochemical Cycles* 21, GB1013, doi: 10.1029/2006GB002709
- 12 Worrall, F., T. Burt, and R. Shedden. 2003. Long term records of riverine dissolved organic matter.
13 *Biogeochemistry* 64:165–178.
- 14 Wright, R.F., J. Aherne, K. Bishop, L. Camarero, B.J. Cosby, M. Erlandsson, C.D. Evans, M. Forsius,
15 D.W. Hardekopf, R. Helliwell, J. Hruška, A. Jenkins, J. Kopáček, F. Moldan, M. Posch, M.
16 Rogora. 2006. Modelling the effect of climate change on recovery of acidified freshwaters:
17 Relative sensitivity of individual processes in the MAGIC model. *Science of the Total*
18 *Environment* 365:154–166.
- 19 Wu, W., and C.T. Driscoll. 2010. Impact of climate change on three-dimensional dynamic critical load
20 functions. *Environmental Science & Technology* 44:720–726.
- 21 Yan, N.D., B. Leung, W. Keller, S.E. Arnott, J.M. Gunn, and G.G. Raddum. 2003. Developing
22 conceptual frameworks for the recovery of aquatic biota from acidification. *Ambio* 32: 165–169.
- 23 Yan, N.D., B. Leung, W. Keller, S.E. Arnott, J.M. Gunn, and G.G. Raddum. 2003. Developing
24 conceptual frameworks for the recovery of aquatic biota from acidification. *Ambio* 32:165-169.
- 25 Zaccherio, M.T., and A.C. Finzi. 2007. Atmospheric deposition may affect northern hardwood forest
26 composition by altering soil nutrient supply. *Ecological Applications* 17:1929–1941.
- 27 Zaehle, S., A.D. Friend, P. Friedlingstein F. Dentener, P. Peylin, and M. Schulz. 2010. Carbon and
28 nitrogen cycle dynamics in the O-CN land surface model: 2. Role of the nitrogen cycle in the
29 historical terrestrial carbon balance. *Global Biogeochemical Cycles* 24:GB1006.
- 30 Zhang, X., F.W. Zwiers, G.C. Hegerl, F.H. Lambert, N.P. Gillett, S. Solomon, P.A. Stott, and T. Nozawa.
31 2007. Detection of human influence on twentieth-century precipitation trends. *Nature* 448:461–
32 465.