National Acid Precipitation Assessment Program Report to Congress 2011:

An Integrated Assessment

2011

December 28, 2011

Members of Congress:

I am pleased to transmit the *National Acid Precipitation Assessment Program Report to Congress: An Integrated Assessment*. The National Acid Precipitation Assessment Program (NAPAP) is a Federal interagency program that coordinates acid rain research and reports to Congress on the effects of acid precipitation on sensitive ecosystems. Specifically, this report presents the latest scientific information and analysis concerning the costs, benefits, and environmental effectiveness of the Acid Rain Program (ARP)—a bipartisan mandate under Title IV of the 1990 Clean Air Act Amendments signed into law by President George H. W. Bush to reduce sulfur dioxide (SO_2) and nitrogen oxides (NO_X) emissions from electric generating sources. Notably, the SO_2 program includes the use of a creative emissions cap-and-trade program that combines the best of American science, government, and market-driven innovation.

This NAPAP Report to Congress focuses on emission reductions from power plants, summarizes changes in deposition rates and environmental impacts, and evaluates the ecological effects expected to accompany future reductions in SO_2 and NO_X emissions. The report concludes that Title IV has successfully reduced emissions of SO_2 and NO_X from power generation (i.e., the sources covered by the ARP), including: SO_2 emissions to 5.7 million tons in 2009, 64% lower than 1990 emissions and below the 2010 statutory cap of 8.95 million tons; and NO_X emissions to 2 million tons in 2009, 67% lower than 1995 emissions and substantially exceeding the Title IV goal. The emission reductions achieved under the ARP have contributed to measurable improvements in air quality; decreases in acid deposition; the beginnings of recovery of acid-sensitive lakes and streams in some areas; and improvements in visibility.

This report also estimates the benefits and costs of complying with Title IV. The human health benefits of improved air quality are estimated to be in the range of $170 billion to $430 billion in 2010 alone. Substantial additional benefits result from improved visibility and improved ecological conditions. These benefits greatly exceed the cost of complying with Title IV, which are estimated at about $3 billion per year—less than half the initial estimates of compliance with Title IV's tightening of the SO_2 emission cap in the year 2000, which now applies to fully 96 percent of the Nation's total electricity generation from fossil fuels. The use of a cap-and-trade mechanism significantly lowered costs by giving utilities flexibility in achieving SO_2 emissions reductions.

Despite the environmental improvements reported here, research over the past few years indicates that recovery from the effects of acidification is not likely for many sensitive forests and aquatic ecosystems without additional decreases in acid deposition. Scientific publications and acid deposition models show that, for these sensitive regions, the SO_2 and NO_X emission reductions achieved under Title IV from power plants are insufficient to achieve full recovery or to prevent further acidification.

Sincerely,
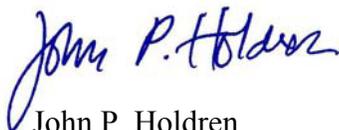
John P. Holdren
Assistant to the President for Science and Technology
Director, Office of Science and Technology Policy

Acknowledgments

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, Natural Resources, and Sustainability (CENRS), of the National Science and Technology Council (NSTC). 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 CENRS.

Contents

List of Tables

List of Figures

Acronyms and Abbreviations

AEL	alternative emission limit
AHM	Alpine Hydrochemical Model
Al^{2+}, Al^{3+}	aluminum cations
AMS	Allowance Management System
AIRMoN	Atmospheric Integrated Research Monitoring Network
ANC	acid neutralizing capacity
ARP	Acid Rain Program
BenMAP	Benefits Mapping and Analysis Program
BTU	British thermal unit
CAAA	Clean Air Act Amendments
CAIR	Clean Air Interstate Rule
CAMD	Clean Air Markets Division
CASTNET	Clean Air Status and Trends Network
CAVR	Clean Air Visibility Rule
CEMS	continuous emission monitoring system
CENRS	Committee on Environment, Natural Resources, and Sustainability
CFR	Code of Federal Regulations
CMAQ	Community Multi-Scale Air Quality
CMAS	Community Modeling and Analysis System
CO_2	carbon dioxide
CPI	consumer price index
CSAPR	Cross-State Air Pollution Rule

DOC	dissolved organic carbon
DOE	U.S. Department of Energy
DOI	U.S. Department of the Interior
dv	deciview
ECMPS	Emissions Collection and Monitoring Plan System
EGUs	electric generating units
EMAP	Environmental Monitoring and Assessment Program
EPA	U.S. Environmental Protection Agency
GAO	U.S. General Accounting Office
HNO_3	nitric acid
ILWAS	Integrated Lake-Watershed Acidification Study
IMPROVE	Interagency Monitoring of Protected Visual Environments
IPM	Integrated Planning Model
kg/ha/yr	kilograms per hectare per year
lb	pound
LTM	Long-Term Monitoring
MACT	Maximum Achievable Control Technology
MAGIC	Model of Acidification of Groundwater in Catchments
meq/m^2/yr	milliequivalent per square meter per year
mmBTU	million British thermal units
MPI	market price index
MW	megawatt
N_2O	nitrous oxide
NAAQS	National Ambient Air Quality Standards
NADP	National Atmospheric Deposition Program
NAMS	National Air Monitoring Stations
NAPAP	National Acid Precipitation Assessment Program

NASA	National Aeronautics and Space Administration
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
PnET-BGC	photosynthetic evapotranspiration-biogeochemical
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
SO_4^{2-}	sulfate
SSWC	steady-state water chemistry
TIME	Temporally Integrated Monitoring of Ecosystems
USDA	U.S. Department of Agriculture
USGS	U.S. Geological Survey
XML	Extensible Markup Language
μeq/L	microequivalents per liter
μg/m^3	micrograms per cubic meter

EXECUTIVE SUMMARY

The National Acid Precipitation Assessment Program Report to Congress 2011

Acid deposition, more commonly known as acid rain, occurs when emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) react in the atmosphere with water, oxygen, and oxidants to form various acidic compounds. Prevailing winds transport the acidic compounds hundreds of miles, often across state and national borders. These acidic compounds then fall to earth in either a wet form (rain, snow, and fog) or a dry form (gases, aerosols, and particles). At certain levels, the acidic compounds, including small particles such as sulfates and nitrates, can cause many negative human health and environmental effects.

What Are the Effects of Acid Rain?

Ecosystems and human health are subject to many stresses, including acid rain. Scientific research has shown that SO_2 and NO_x air pollutants and the acid rain formed by these pollutants can

- Degrade air quality,

- Impair visibility,

- Negatively impact human health,

- Acidify lakes and streams,

- Harm sensitive forests,

- Harm sensitive coastal ecosystems, and

- Accelerate the decay of building materials, paints, and cultural artifacts, such as buildings, statues, and sculptures.

Why Is This Report Being Sent to Congress, and What Is the Role of the National Acid Precipitation Assessment Program in the Report?

In 1990, Congress enacted Title IV as part of the Clean Air Act Amendments (CAAA). The Acid Rain Program (ARP) created under Title IV requires significant decreases in the emissions of SO_2 and NO_x from fossil fuel–burning power plants to improve air quality and protect ecosystems that have suffered damage from acid deposition. Under Title IX of the 1990 CAAA, the National Acid Precipitation Assessment Program (NAPAP) was asked to periodically assess and report to Congress on (1) implementation of the ARP, (2) the most recent scientific information related to acid deposition and its effects, and (3) additional decreases in acid deposition necessary to prevent adverse ecological effects. This 2011 NAPAP Report to Congress (RTC) focuses on emission reductions from power plants, summarizes changes in deposition rates and environmental

impacts, and projects the ecological effects of additional reductions in SO_2 and NO_x emissions.

What Are the Results of Implementing Title IV of the 1990 Clean Air Act Amendments?

Implementation of Title IV has successfully and substantially reduced emissions of SO_2 and NO_x from power generation (i.e., the sources covered by the ARP), including the following.

- In 2009, SO_2 emissions were 5.7 million tons, 64% lower than 1990 emissions and 67% lower than 1980 emissions, a level below the 2010 Title IV statutory cap of 8.95 million tons of SO_2 emissions.

- In 2009, NO_x emissions were 2 million tons, 67% lower than 1995 emissions, substantially exceeding the Title IV goal of a 2-million-ton reduction in NO_x emissions from projected 2000 levels without the ARP, as required by the 1990 CAAA.

- In addition, SO_2 emissions from all sources, including those sources not covered by the ARP, have decreased by 59% since 1990, and emissions of NO_x from all sources have decreased by 40% since 1990.

Water samples being collected on the Laurel Prong fork of the Rapidan River in Virginia (photo courtesy of U.S. EPA).

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

- SO_2 concentrations in the atmosphere, a precursor to fine particles and acid deposition, have decreased since 1990. Average annual SO_2 concentrations in 2007–2009 were 57% to 63% lower than in 1989–1991 in the Midwest and eastern United States.

- Sulfate concentrations in the atmosphere, a major component of fine particles, especially in the eastern United States, have decreased since 1990. Average annual sulfate concentrations in 2007–2009 were 41% to 49% lower than in 1989–1991 in the Midwest and eastern United States.

- Wet sulfate deposition, a major component of acid rain, has decreased since 1990. Average annual sulfate deposition in the Northeast and Southeast in 2007–2009 was 43% lower than in 1989–1991, deposition in the Mid-Atlantic was 42% lower, and the Midwest was 44% lower.

- Wet inorganic nitrogen deposition has decreased regionally from historical levels. However, decreases were less than those of wet sulfate deposition because of the continuing large contribution from other sources of NO_x, such as on-road vehicles and non-road vehicles. Still, average annual wet inorganic nitrogen deposition in 2007–2009 was 16% to 27% lower than in 1989–1991 in the Midwest and eastern United States.

- Levels of acid neutralizing capacity (ANC), an indicator of the ability of a waterbody to neutralize acid deposition, have shown improvement from 1990 to 2008 at many lake and stream long-term monitoring sites in the eastern United States, including New England and the Adirondack Mountains. Many lakes and streams still have acidic conditions harmful to their biota even though the increases in ANC indicate that some recovery from acidification is occurring in sensitive aquatic ecosystems.

- Data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network show no statistically significant trends in visibility at most monitoring sites over the 10-year period from 1999 to 2008. The Northeast, however, has sites with improving visibility on both the best and worst visibility days, principally due to regionally decreased sulfate particulate concentrations.

Further, because emission reductions result in fewer fine particles and lower ozone concentrations in the air, thousands fewer premature deaths, hospital admissions, and emergency room visits are projected in 2010. The value of the resulting human health benefits from emission reductions range from $170 to $430 billion per year (2008$).

Emission reductions from the existing ARP have been augmented by additional programs, such as the Clean Air Interstate Rule (CAIR) and the Clean Air Visibility Rule (CAVR), which have increased reductions beyond initial ARP levels.

What Is the Role of Cap and Trade in the Success of the Acid Rain Program?

The success of the SO_2 emission-reduction program is due to the combined use of an overall emission cap for SO_2, which ensures that these reductions are achieved and maintained, and a trading system that facilitates lowest-cost emission reductions. Together, this is known as cap and trade. NO_x reductions under the ARP are achieved through a program that applies to a subset of coal-fired electric generating units (EGUs) and is closer to a more traditional, rate-based regulatory system. Sources controlled in both the SO_2 and NO_x components of the ARP have demonstrated very high levels of compliance, averaging 99% annual compliance since the beginning of the program. The inherent flexibility for sources to choose how to control their SO_2 emissions in the cap-and-trade approach for SO_2 has been successful at reducing compliance costs to a fraction of the cost estimated in 1990. Several factors are responsible for the relatively low costs of SO_2 reductions realized under Title IV, including the widespread availability of low

sulfur coal and technical innovations that facilitated use of that coal, lower than anticipated scrubber costs, the opportunity to bank allowances, and development of an efficient, high-volume market for allowances. Although the costs are low, the ARP achieves substantial health and environmental benefits through air quality improvements.

What Is the Future of Current Clean Air Rules?

Emissions of SO_2 and NO_x are expected to decline further as additional programs are implemented to control emissions from fossil fuel–burning power plants. In March 2005, the U.S. Environmental Protection Agency (EPA) promulgated 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 attain the National Ambient Air Quality Standards (NAAQS) for fine particulate matter ($PM_{2.5}$) by reducing and capping SO_2 and NO_x emissions in 28 eastern states and the District of Columbia. 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, leaving CAIR in place until EPA issued new rules to replace CAIR. On July 6th, 2011, EPA replaced CAIR with the final Cross-State Air Pollution Rule (CSAPR), which will control SO_2 and NO_x emissions from fossil fuel-burning power plants in 28 states in the eastern half of the country. EPA is also working on a set of additional rules covering SO_2 and NO_x emissions from the electric power sector to address transport of pollution contributing to ozone and fine particle problems, as well as Maximum Achievable Control Technology (MACT) for the power sector and other industries. These rules could lower stationary source emissions for these same pollutants in the next 5 years.

Are Ecosystems Recovering from the Effects of Acid Rain?

Despite the environmental improvements reported here, research over the past few years indicates that recovery from the effects of acidification is not likely

for many sensitive areas without additional decreases in acid deposition. Many published articles, as well as the modeling presented in this report, show that the SO_2 and NO_x emission reductions achieved under Title IV from power plants are now recognized as insufficient to achieve full recovery or to prevent further acidification in some regions. Additional SO_2 and NO_x emission reductions from power plants and other source sectors are needed to improve air quality, reduce deposition, and further reduce the number of acidic lakes and streams in many regions of the United States. Some of these additional emissions reductions may be achieved through implementation of existing or future regulations to address transport of ozone and fine particles, including the CSAPR in the eastern United States, and other rules affecting mobile sources, SIPs, NAAQS for ozone and $PM_{2.5}$, as well as future rules to reduce air toxics and other pollutants from power plants.

What Is the Importance of Long-term Environmental Monitoring in Understanding the Effects of Acid Rain?

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

What Acid Rain–related Topics Are Currently at the Forefront of Scientific Knowledge?

Previous NAPAP RTCs have reported on the state of the science and emerging issues related to acid deposition. This report also summarizes recent science published since the 2005 NAPAP RTC. Topics covered in this science summary relate to ecosystem responses to emission

controls and acid deposition reductions that are receiving increasing attention in the scientific literature, including the following:

- Scientists have observed delays in ecosystem recovery in the eastern United States, despite decreases in emissions and deposition over the past 30 years;

- A growing body of literature documents impacts of nitrogen deposition on western ecosystems;

- "Critical loads" are increasingly used by scientists as a tool for quantifying the sulfur and nitrogen deposition levels at which ecosystems are impacted in order to better inform air quality policies;

- Recent literature identifies linkages between a changing climate, an altered carbon cycle and ecosystem response to acid deposition as an important emerging area of scientific investigation; and

- Scientists identify multi-pollutant interactions, including reactions among SO_2, NO_x, ozone, and mercury, as an under-studied area of ecosystem impacts, which, if better understood, could have implications for future air pollutant policy.

Sugar maple is a tree species sensitive to acid rain (photo courtesy of U.S. EPA).

INTRODUCTION

The 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 (DOE), the U.S. Department of Agriculture (USDA), the U.S. Department of the Interior (DOI)/U.S. Geological Survey (USGS), the DOI/National Park Service (NPS), the National Aeronautics and Space Administration (NASA), and the National Oceanic and Atmospheric Administration (NOAA). The Acid Rain Program (ARP), authorized under Title IV of the CAAA, regulates the emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) from electric generating units (EGUs) that use fossil fuel (e.g., coal, gas, oil). These emissions contribute to acid deposition and the formation of fine particulate matter ($PM_{2.5}$) and ozone, which ultimately leads to a wide range of environmental impacts, including harm to human health and visibility impairment. The NAPAP Reports to Congress (RTCs) provide an assessment of the implementation of the ARP, including the impacts and benefits of the sulfur and nitrogen emission reductions achieved by the program. This NAPAP RTC is written to effectively and fully communicate the results of the assessment to decision makers. Congress has asked NAPAP to assess all available data and information to answer two questions.

1. What are the costs, benefits, and effectiveness of Title IV? This question addresses the costs and economic impacts of complying with the ARP, as well as the benefit analyses associated with various human health and welfare effects, including reduced visibility and effects on ecosystems.

2. What reductions in deposition rates are needed to prevent adverse ecological effects? This is a complex question addressing how much deposition can occur before harmful environmental effects take place.

Impacts of SO_2 and NO_x Emissions

SO_2 and NO_x emissions are the primary precursors involved in the formation of acid rain, also known as acid deposition. The ARP was created under the 1990 CAAA to reduce the adverse effects of acid deposition through reductions in annual emissions of SO_2 and NO_x. These emissions may

be transported up to hundreds of miles away from their emitting source, and thus have the potential to impact large areas and populations. They may cause such impacts as diminished air quality, damage to human health, acidification of lakes and streams, harm to sensitive forests and coastal ecosystems, degradation of visibility, and the acceleration of the decay of building materials. Certain acid-sensitive ecosystems and regions may be more susceptible to the impacts of acid deposition. Additionally, some groups of people—such as children, the elderly, and people with pre-existing conditions—may be more sensitive to the related health impacts caused by diminished air quality due to SO_2 and NO_x emissions. The ARP regulates SO_2 and NO_x emissions from EGUs, but does not regulate vehicular or agricultural sources, both of which emit nitrogen and, to a lesser extent, sulfur compounds. In some areas, emissions from these other sources can be significant.

Emissions have declined since the initiation of the ARP, and these reductions have led to increased visibility in some locations, reduced incidences of health impacts, improved conditions in some acidified lakes and streams, and reduced nitrogen deposition to some sensitive ecosystems. Ecological recovery has been identified in some areas, but this remains a complex process because recovery often lags behind declines in emissions. Current research looks to address the issue of delayed ecological response, as well as how climatic changes and multi-pollutant interactions may affect recovery.

Structure of the Report

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

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

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

- Chapter 3 covers advances in the state of the science since the last NAPAP RTC regarding atmospheric deposition and the impacts of acid deposition on aquatic and terrestrial ecosystems, including ecosystems in the Rocky Mountains, the far western United States, and coastal areas. Chapter 3 also reports on research on the level of pollutant deposition below which ecosystem damage does not occur (i.e., critical loads), the interactions between atmospheric deposition and climate change, and multi-pollutant interactions.

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

Visibility is good for hikers on Rescue Creek Trail in Yellowstone National Park (photo courtesy of NPS).

CHAPTER 1
Acid Rain Program Elements and Implementation

Established under Title IV of the 1990 CAAA, the ARP requires major emission reductions of SO_2 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 and includes descriptions of the program and its sources, program compliance, the allowance market, ARP benefits and costs, and the tools used to assess the progress of the program.

Overview of Emission Reductions

The implementation of Title IV has successfully reduced emissions of SO_2 and NO_x from EGUs. Under Title IV of the 1990 CAAA, Congress established a permanent cap on the total amount of SO_2 that may be emitted by EGUs in the conterminous United States. This cap has been phased in, with the final 2010 SO_2 cap set at 8.95 million tons—a level of about one-half of the emissions from EGUs in 1980. In 2009, 3,572 EGUs were subject to the SO_2 provisions of Title IV. By 2009, the sources (i.e., EGUs) covered by the ARP had reduced their combined SO_2 emissions by approximately 67% from 1980 levels and 64% 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 2009, the 960 sources subject to ARP NO_x regulations emitted 67% fewer emissions than in 1995.[1] Heat input and electricity generation increased by approximately 30% over this same period, and the average retail price of electricity was about the same in 2009 as it was in 1990 (Figure 1-1).

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

At a Glance: ARP Results in 2009

- **SO_2 emissions:** 5.7 million tons
- **SO_2 compliance:** 100%
- **SO_2 allowances:** Allowance bank increased by almost 4 million allowances from 2008 levels
- **SO_2 allowance prices:** : In 2009, allowance prices fell from $187 per ton to $61 per ton
- **NO_x emissions:** 2.0 million tons
- **NO_x compliance:** 100%

---

[1] As described in Chapter 2 of this report, other programs—such as the Ozone Transport Commission (OTC); the NO_x Budget Trading Program (NBP) 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 2009.
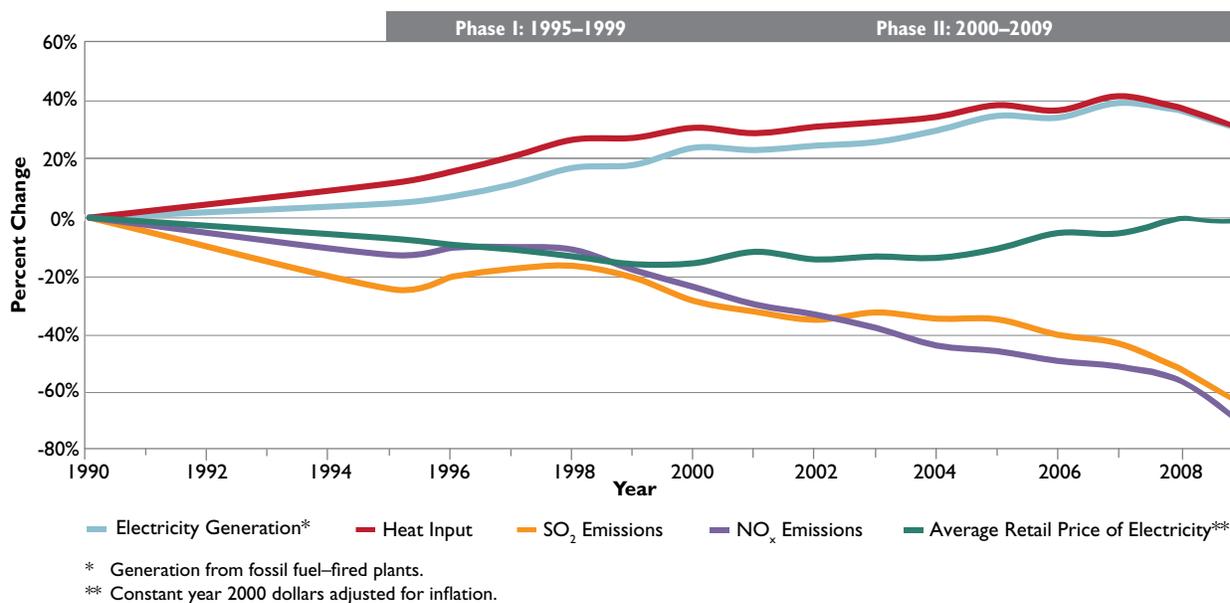
Figure 1–1. Trends in electricity generation, fossil fuel energy use, prices, and emissions from power plants affected by the ARP, 1990–2009 (U.S. EPA, 2010b).

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 install pollution controls, implement efficiency measures, change to lower sulfur coal, or sell or purchase allowances, 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.

Acid Rain Program Design

SO₂ Program

The SO_2 emission-reduction program created under Title IV represents a substantial change from traditional command and control regulatory approaches that establish source-specific emission limitations. Instead, the program combines an overall emission cap for SO_2, which ensures that emission reductions are achieved and maintained, with a trading system that facilitates lowest-cost emission reductions. The ARP features tradable SO_2 emission allowances, where one allowance is a limited authorization to emit one ton of SO_2. A fixed number of allowances is issued by the government, and these allowances may be bought, sold, or banked for future use by EGUs or other parties (e.g., utilities, brokers, or anyone else interested in holding allowances). Existing sources are allocated allowances each year. New sources do not receive allowances and instead must buy them; however, the required purchase of allowances has not been a barrier to market entry (i.e., new sources have been able to acquire

the allowances needed to compete effectively in the market). At the end of each year, all affected sources are obliged to surrender to EPA the number of allowances that correspond to their annual SO_2 emissions (one allowance for each ton of SO_2).

Title IV establishes a two-phased tightening of the SO_2 emission cap, adjusting the allocation of SO_2 allowances to fossil fuel–fired EGUs to reach the permanent cap on the number of allowances of 8.95 million tons annually in 2010. Once the bank of unused allowances is depleted, the limit on allowance allocations constrains emissions to the level of the cap.

Phase I of the ARP (1995–1999) affected 263 of the larger (>100 megawatt [MW]), higher-emitting EGUs, which are located primarily in the central and eastern United States. Phase I SO_2 allowance allocations were distributed to each source based on the following formula: the product of an emission rate of 2.5 pounds (lb) SO_2/million British thermal units (mmBTU) of heat input and its average heat input for 1985–1987. Some Phase II sources chose to "opt-in" to Phase I and comply early, bringing the total number of units participating in Phase I to more than 400.

Phase II began in 2000 and extended to all existing EGUs serving generators larger than 25 MW and all new fossil fuel–fired generation units throughout the country. In 2009, the total number of units covered by the SO_2 criteria was 3,572 sources, which represented 96% of total electricity generation from fossil fuels. In Phase II, all Phase I and Phase II SO_2 affected sources are allocated allowances equivalent to an amount no greater than the product of 1.2 lb SO_2/mmBTU and their average heat input for 1985–1987.

Title IV requires that sources monitor emissions continuously and report their emissions quarterly. Failure to surrender sufficient allowances results in two significant automatic penalties. Any source that fails to hold enough allowances to match its SO_2 emissions for the previous year must pay to EPA, by July 1, an automatic penalty of $2,000 (inflation-adjusted to $3,517 for 2009) per ton of emissions in excess of allowances held. The source must also immediately surrender to EPA an amount of allowances, issued for the year

the payment is due, equaling the tons of excess emissions. A source may sell or bank for future use any remaining SO_2 allowances not needed for compliance during a year. Sources may use these banked allowances as needed to comply with the program in future years until the bank is depleted.

Title IV mandates that a limited number of allowances allocable to existing sources be withheld and auctioned, with revenues from the auction returned pro rata to existing sources. The annual SO_2 auction provides an opportunity for sources to buy and sell allowances. The auctions help ensure that new sources have an opportunity to obtain allowances beyond those allocated initially to existing EGUs. Complete results of the annual SO_2 allowance auction are available at http://www.epa.gov/airmarkets/trading/auction.html.

In addition to the Title IV SO_2 program, recent programs such as the Clean Air Interstate Rule (CAIR) and the Clean Air Visibility Rule (CAVR) also have achieved SO_2 reductions. Additional rules covering SO_2 emissions from the electric power sector, as well as Maximum Achievable Control Technology (MACT) for the power sector and other industries, are being developed by EPA and may lead to further SO_2 reductions from stationary sources.

Rampart Reservoir in Pike National Forest and the surrounding forests are examples of western habitats that may be impacted by acid precipitation (photo courtesy of U.S. EPA).

NO$_x$ Program

In contrast to the system established for SO$_2$ emissions, the ARP does not establish tradable emission allowances for NO$_x$ emission reductions. Instead, sources control how much NO$_x$ is emitted from coal-fired boilers based on the use of cost-effective control technologies for each unit of fuel consumed (lb NO$_x$ per mmBTU). There are two phases of the NO$_x$ component: Phase I began in 1996 (delayed 1 year because of litigation), and Phase II began in 2000. During Phase I, which applied to specific coal-fired boilers statutorily affected by Phase I SO$_2$ requirements, the NO$_x$ emission rate was set at 0.50 lb NO$_x$/mmBTU for dry-bottom, wall-fired units and 0.45 lb NO$_x$/BTU for tangentially fired units. Beginning in 2000, Phase II plants were required to meet emission rates between 0.40 lb NO$_x$/mmBTU and 0.86 lb NO$_x$/mmBTU, depending on the type of boiler. In 2009, 960 units were subject to ARP NOx program requirements, which represented 63% of total electricity generation from fossil fuels.

Although the ARP does not include NO$_x$ emission trading, sources are provided a degree of flexibility through emission-averaging provisions, whereby a company can meet its NO$_x$ emission limitations by averaging the emission rates of two or more boilers (see text box). This enables sources to reduce their NO$_x$ emissions at lower cost by allowing them to over-control at EGUs where it is technically easier to control emissions. At the end of the year, sources must demonstrate compliance with NO$_x$ emission requirements by achieving an annual emission rate at or below mandated levels, as outlined in their EPA-approved compliance plans. As with the SO$_2$ program, it is important to note that a number of other programs have contributed to NO$_x$ emission reductions from ARP sources (see Chapter 2 for a description of these programs). Current programs, such as CAIR and CAVR, have achieved annual NO$_x$ emission reductions beyond those achieved by the Title IV NO$_x$ program. Further reductions in NOx emissions from stationary sources will also come from the Cross-State Air Pollution Rule (CSAPR), which replaces CAIR to address the transport of pollution from the electric power sector contributing to ozone and fine particle problems, as well as MACT for the power sector and other industries.

Sources Achieved 100% NO$_x$ Compliance in 2009 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 (297 units used this option in 2009).

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 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 (three units used this option in 2009).

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 (660 units used this option in 2009).

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

Title IV Affected Sources

SO₂ Sources

Sources that are subject to the SO_2 component include boilers or combustion turbines that burn fossil fuel, serve generators with a design capacity greater than 25 MW, and produce electricity for sale. Several types of units meeting these criteria are not affected by the ARP. These include simple combustion turbines that began to produce electricity for sale before November 15, 1990; cogeneration units whose annual electricity sales remain below the threshold established by regulation; and specific qualifying facilities and independent power producers that are contractually bound to sell electricity at a price that was established before November 15, 1990. Despite these exceptions, almost all non-cogeneration units that have total design capacity greater than 25 MW and that produce electricity for sale now must participate in the ARP.

NOₓ Sources

Some of the sources subject to the SO_2 requirements of Title IV are also subject to the Title IV NO_x requirements. All units where coal accounted for more than 50% of heat input for at least 1 year during the 1990 through 1995 time period and that are configured for a specific type of boiler (i.e., cell burner, cyclone, dry-bottom wall-fired, tangentially fired, vertically fired, or wet-bottom) are affected by the Title IV NO_x criteria.

Table 1-1 lists the sources affected by Title IV NO_x emission components in 2009. For more details on ARP applicability criteria, see 40 Code of Federal Regulations (CFR) 72.6, which provides ARP applicability regulations established under Title IV.

Compliance

SO₂ Compliance

The Title IV SO_2 program has achieved a near-perfect compliance record since the program took effect in 1995. In 2009, as in each year since 2005, the program achieved 100% compliance, and all ARP facilities complied with the requirement to hold enough allowances to cover SO_2 emissions. EPA allocated 9.5 million SO_2 allowances under the ARP for 2009. Together with the 8.5 million unused allowances carried over (or banked) from prior years, 18.0 million allowances were available for use in 2009 (see Figure 1-2). ARP sources emitted approximately 5.7 million tons of SO_2 in 2009, less than the allowances allocated for the year, and far less than the total allowances available. As a result, the bank increased between 2008 and 2009 by nearly 4 million allowances to 12.3 million, a 45% increase. The bank includes the unused allowances from previous years, plus the unused allowances allocated in 2009 (i.e., all of the allowances above the yellow line in Figure 1-2). In 2010, the total number of Title IV SO_2 allowances allocated annually dropped to 8.95 million and will remain statutorily fixed at that annual level.

Table 1-1. Title IV NOₓ Affected Units by Boiler Type and NOₓ Emission Limit		
Coal-Fired Boiler Type	Title IV Standard NOₓ Emission Limits (lb/mmBTU)	Number of Units
Phase I, Group 1—Tangentially Fired	0.45	132
Phase I, Group 1—Dry-Bottom, Wall-fired	0.50	107
Phase II, Group 1—Tangentially Fired	0.40	296
Phase II, Group 1—Dry-Bottom, Wall-fired	0.46	290
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		960

Source: U.S. EPA, 2009c

The existence of the allowance market has given some sources the incentive to reduce their SO_2 emissions below the level of their allowance allocation in order to sell their allowances to other sources or to "bank" them for use in future years. Other sources have been able to postpone or reduce expenditures for pollution control by purchasing allowances from sources that controlled emissions beyond their allowance allocation level. As shown in Figure 1-2, the bank or store of unused allowances grew throughout Phase I (1995–1999) as sources reduced emissions more than required. These early reductions reduced the amount of fine particles and acid deposition in the early years of Title IV implementation, increasing the human health and ecological benefits of the program in those years. Beginning in 2000, with Phase II of the program, the set of sources covered by the program expanded and the tighter Phase II emission cap took effect. As Figure 1-2 shows, sources began to use previously banked allowances in addition to allocations from the current year, to comply with Title IV. As a result, emission levels for 2000–2005 were greater than annual allocations as sources used banked allowances for compliance.

NO_x Compliance

Affected sources can comply by either meeting a unit-specific NO_x emission rate or including two or more units in an emission rate averaging plan. As with the SO_2 program, the NO_x program has had a high rate of compliance. Since 2000, there have been 4 years in which a single unit was out of compliance. In 2009, all 960 units that were subject to ARP NO_x criteria achieved compliance.

2009 SO_2 Allowance Market

The number of allowances (authorizations to emit SO_2) allocated to each source in any given year is determined by the Clean Air Act. A recent review (Burtraw and Szambelan, 2009) of emission trading generally concluded that the SO_2 allowance market has "been liquid and active, and according to most observers, [has] worked well in achieving the emission caps at less cost than would have been achieved with traditional approaches to regulation."

Figure 1-3 shows the cumulative volume of SO_2 allowances transferred under the ARP. The cumulative volume exceeded 400 million allowances in 2009, with the majority of these allowances transferred in private transactions. The figure differentiates between allowances transferred in private transactions and those annually allocated and transferred to source accounts by EPA.
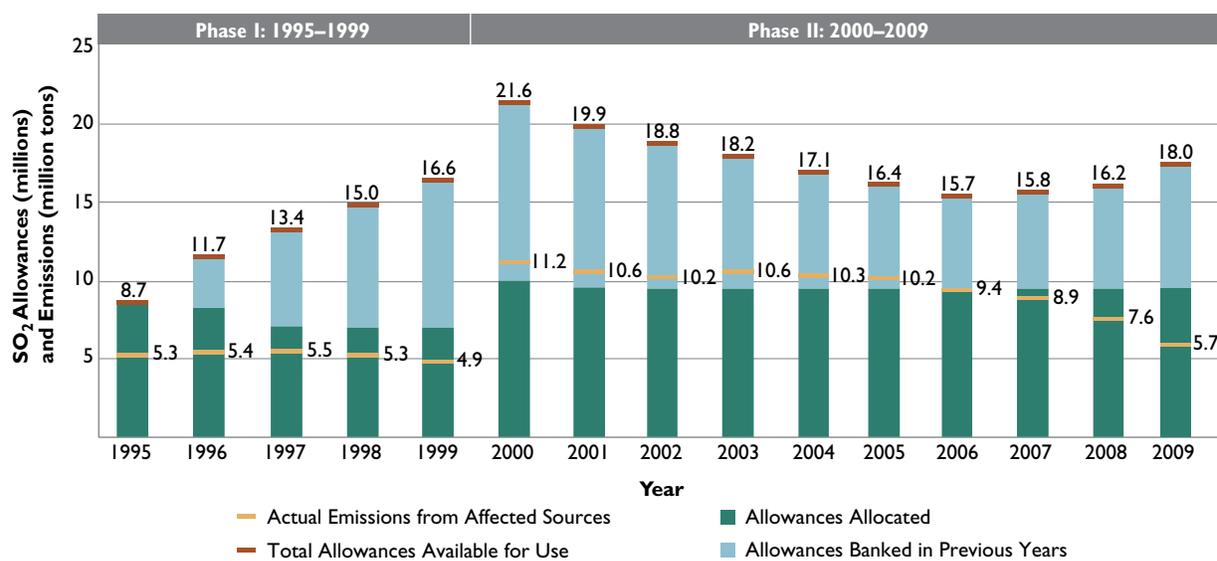
Figure 1-2. SO_2 emissions and the allowance bank, 1995–2009 (U.S. EPA, 2010b).

Private transactions are indicative of both market interest and the use of SO_2 allowances as a compliance strategy. Of the nearly 406 million allowances transferred since 1994, about 67% were traded in private transactions. In December 2001, parties began to use a system developed by EPA to allow online SO_2 allowance transfers. By 2008, account holders registered over 99% of all private allowance transfers through EPA's online transfer system. Allowance transfers are posted and updated daily on http://www.epa.gov/airmarkets.

In 2009, 2,716 private SO_2 allowance transfers involving approximately 15.1 million allowances of past, current, and future vintages were recorded in EPA's Allowance Management System (AMS). About 4 million SO_2 allowances (26%) were transferred in economically significant transactions (i.e., between economically unrelated parties). Transfers between economically unrelated parties are "arm's length" transactions and are considered a better indicator of an active, functioning market than are transactions among the various facility and general accounts associated with a given company. In the majority of all private transfers, allowances were acquired by power companies. Figure 1-4 shows that the annual volume of SO_2 allowances transferred under the ARP, excluding allocations,

retirements (i.e., used allowances surrendered), and other transfers by EPA, has fluctuated since official recording of transfers began in 1994.

SO$_2$ Allowance Market in Brief (close of 2009)

Total Value of the SO$_2$ Allowance Market: $1.1 billion*

- Average Nominal Price: $61 per ton
- Total Allowance Volume (allowable emissions): 18,017,192

2009 Private Transactions

- 2,716 transactions moving 15.1 million allowances
- 26% of allowances transferred between economically unrelated parties

* Total value of allowance market is a snapshot based on the average nominal price as of December 2009 ($61/ton) and the total allowance volume available for 2009 compliance.

Source: U.S. EPA, 2010b
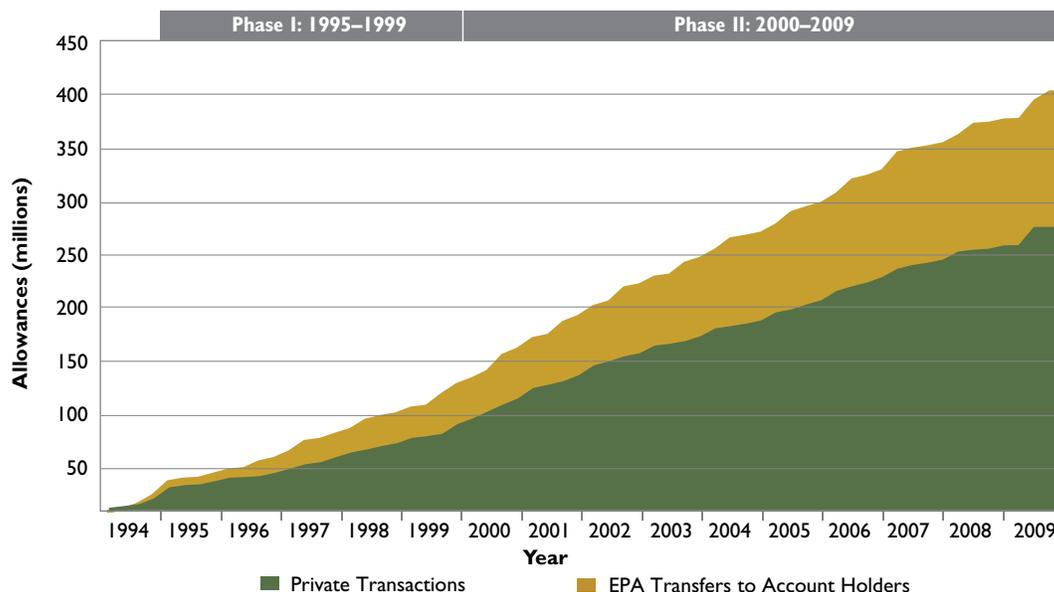
Figure 1-3. Cumulative SO_2 allowances transferred under the ARP, 1994–2009 (U.S. EPA, 2010b).

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 http://www.environmentalmarkets.org). EPA provides a list of brokers and environmental groups that may be interested in facilitating trades or helping parties retire allowances voluntarily (see 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 http://www.epa.gov/airmarkets/business/transfer.html).
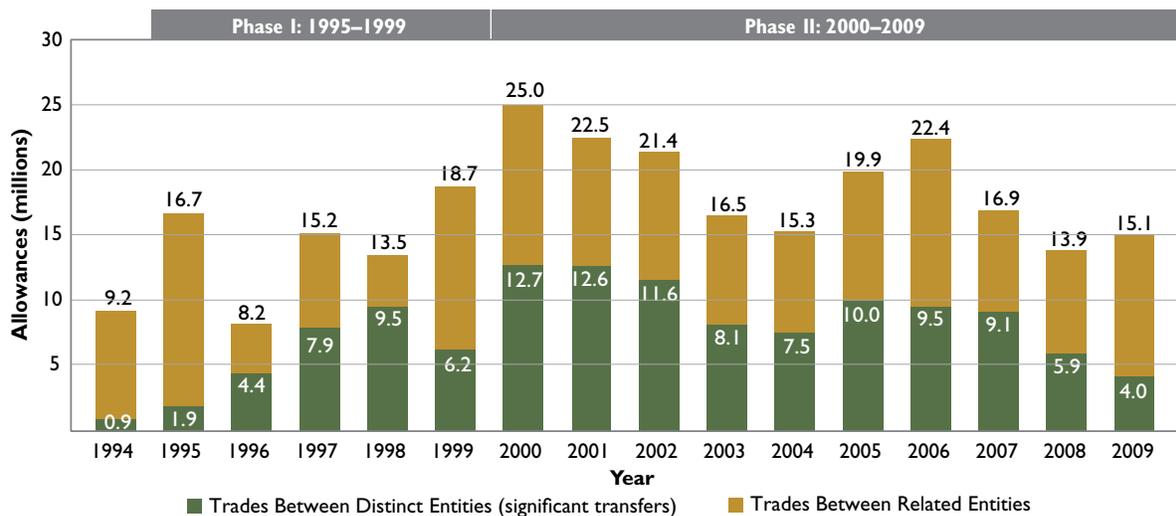
Figure 1-4. SO$_2$ allowances transferred under the ARP (U.S. EPA, 2010b).

Over the first decade of the ARP, SO_2 allowance prices were stable and significantly lower than projected. Just prior to the beginning of the program in 1995, SO_2 allowance prices on the spot market were close to $150 per ton. The cost of allowances was initially projected to be between $250 and $500 per ton during Phase I (1995 to 1999) and $500 to $1,000 per ton in Phase II (beyond 2000). Actual allowance prices in Phase I were in the $100 to $200 range, with a low of $65 in 1996. Allowance prices did display some variability (as would commodities in any market), but it was within this very limited range and tended to be concentrated around times of regulatory change or uncertainty, such as the beginning of Phase I in 1995 and the transition to Phase II in 2000. Even as the more stringent Phase II requirements became effective in 2000, however, prices remained generally below the $200 mark until they started to rise at the end of 2003 with the proposal of CAIR (see Figure 1-5).

When CAIR was proposed in late 2003, allowance prices were influenced by the more stringent CAIR SO_2 cap and new compliance deadline. After CAIR was finalized in March 2005, SO_2 allowance prices continued to trend upward. CAIR was the most significant driver of the price adjustment that began in 2004 and culminated with prices around $1,600 per ton for a short time in December

2005. The ARP SO_2 market essentially became the CAIR SO_2 market. The July 2008 court decision to vacate CAIR (subsequently revised to a remand in December of the same year) caused a dramatic drop in allowance prices as it created an uncertainty regarding the ongoing use of those allowances for compliance with the CAIR SO_2 trading program requirements. During 2009, the value of the SO_2 allowance market experienced a 67% price decline; the monthly average allowance price fell from $187 per ton in January to $61 per ton by December (based on the market price index [MPI]) (see Figure 1-5). That decline continued in 2010, with the SO_2 allowance price falling to an average of $40 per ton by May 2010. Together with the price decline, the volume of significant transactions fell sharply in 2009. Looking forward, it is estimated that allowance prices will remain low. Programs such as CAIR have led to significant reductions in SO_2 emissions levels beyond the Title IV cap level. In addition, recent court decisions prohibit use of Title IV allowances for compliance in current or future programs, such as CAIR or the CSAPR, which replaces CAIR starting in 2012. Thus, the Title IV SO_2 trading program is likely to continue to be characterized by low volume and prices in the future. The program has met its environmental goals, and EPA expects that new emerging programs are likely to further lower power sector SO_2 and NO_x emissions.
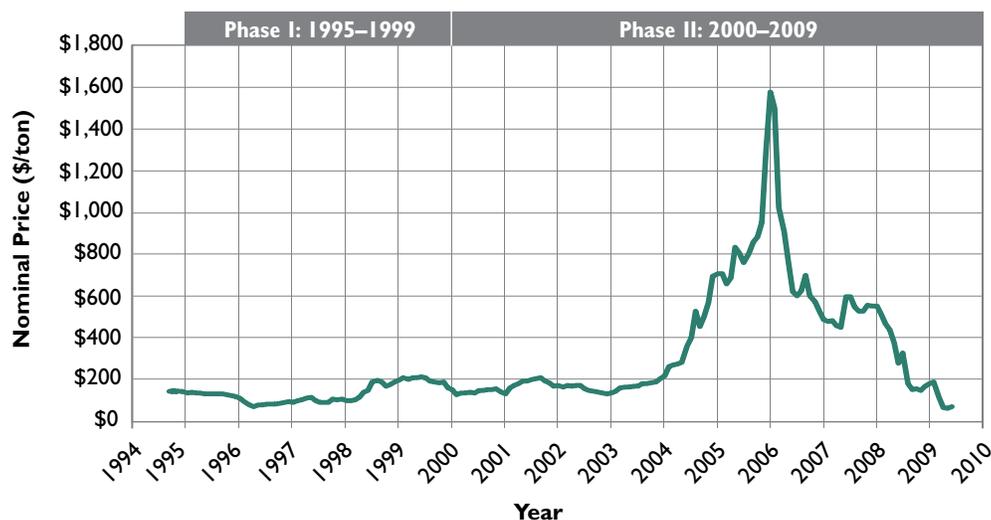
Source: CantorCO2e, 2009

Figure 1-5. Average monthly SO_2 allowance price, August 1994–May 2009 (U.S. EPA, 2009c).

Market Changes

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/marketassessmnt.pdf.

Program Benefits and Costs

Benefits

Emissions of SO_2 and NO_x result in a variety of air pollutants, including not only the strong acids that impact ecosystems through atmospheric deposition but also atmospheric concentrations of particulate matter and ground-level ozone. These multiple air pollutants have important impacts on human health and a wide range of ecological and environmental resources. Due to the multi-pollutant nature of these environmental impacts, the emission reductions achieved by the ARP result in many societal benefits, including the following:

- **Health Benefits.** These include avoided premature mortality and avoided morbidity associated with reduced human exposures to $PM_{2.5}$ and ozone, which are secondary air pollutants that form as a result of SO_2 and NO_x emissions.

- **Visibility Benefits.** Reductions in air pollutants, particularly in $PM_{2.5}$, improve visibility, which leads to physical and economic benefits in both recreational and residential settings.

The Clean Air Interstate Rule

In March 2005, EPA promulated 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 SO_2 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 SO_2 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 program, and started in 2010 for the SO_2 annual program. The rule also establishes a second phase for all three programs, beginning in 2015. CAIR gave affected states SO_2 and NO_x emission budgets and the flexibility in their SIPs to reduce emissions using a strategy that best suits their circumstances, including, as one option, an EPA-administered, regional cap-and-trade program. 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, 2011, EPA finalized the Cross-State Air Pollution Rule (CSAPR), which replaces CAIR and requires eastern, southern, and central states to significantly improve air quality by reducing power plant emissions that cross state lines and contribute to ground-level ozone and fine particle pollution in other states.

- **Agricultural and Forest Productivity Benefits.** Ground-level ozone inhibits plant growth. As a result, reductions in ozone concentrations yield physical and economic benefits in the form of enhanced agricultural and forest productivity.

- **Ecological Benefits.** A wide range of ecological resources are susceptible to damage when exposed to ambient air pollution or deposition of pollutants to terrestrial or aquatic environments. For a small portion of these effects, it is possible to quantify and estimate the economic value of avoided pollutant exposure.

- **Materials Damage Benefits.** Some materials are susceptible to accelerated deterioration when exposed to air pollution; as a result, reduction in air pollution can extend the life of these materials, yielding physical and economic benefits.

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

Still, it is important to recognize that benefits beyond human health improvements result from emission reductions such as those achieved by the ARP. For example, a recent report (U.S. EPA, 2011) fulfilling CAAA Section 812 requirements quantified the overall benefits of implementing the 1990 CAAA and estimated that all CAAA programs taken together will result in $40 billion

Reductions in emissions of SO_2 and NO_x result in improved visibility (photo courtesy of NPS).

in benefits due to improvements in recreational and residential visibility in 2010. There also have been recent advances in efforts to quantify the ecological benefits of emission reductions. For example, Banzhaf et al. (2006) used a contingent valuation study to estimate the total economic value of reducing ecological impacts of acid deposition in New York's Adirondack Park from air quality policies that reduced SO_2 and NO_x emissions. The study estimated total statewide benefits ranging from $336 million to $749 million annually. That total increased to $1.1 billion when alternative assumptions were used regarding ecological change. Significant future analytical work and basic ecological and economic research is needed to build a sufficient base of knowledge and data to support an adequate assessment of ecological benefits. For the current analysis, this incomplete coverage of effects represents a significant source of uncertainty in assessing the benefits of ARP implementation, which make up a significant portion of total CAAA benefits.

Since publication of the last NAPAP RTC (NSTC, 2005), there have been some efforts to quantify the benefits of implementing the ARP. In 2005, a peer-reviewed journal article assessed the human health and welfare benefits of ARP implementation for the prospective year, 2010 (Chestnut and Mills, 2005). The benefits were estimated using the modeled emission reductions and ambient air quality expected to be achieved in 2010 under the ARP. The majority of the monetized benefits of ARP implementation reported in the study were from the prevention of health-related impacts (e.g., premature death) due to reductions in ambient concentrations of $PM_{2.5}$ and ground-level ozone. A concentration response function developed by Pope et al. (2002) was used to estimate incidences of adult premature mortality as a result of $PM_{2.5}$ exposure. EPA currently uses the Pope estimate to represent the lower end of the range of potential human health benefits. The Chestnut and Mills (2005) study estimated the $PM_{2.5}$ and ozone health-related benefits of the ARP to be $134 billion and $5.5 billion (2008$) annually, respectively; they also estimated benefits from visibility improvements in national parks and wilderness areas in California, the Colorado plateau, and the southeastern United States at about $2.5 billion (2008$) annually.

Since publication of the Chestnut and Mills results, the assumptions used to develop human health effects estimates have changed. For example, EPA now also includes concentration–response functions derived from a study by Laden et al. (2006) and an expert elicitation to estimate the range of incidences of adult premature mortality as a result of $PM_{2.5}$ exposure. The Laden et al. estimate is currently used to represent the upper end of the range of potential human health benefits. Additionally, many underlying modeling assumptions have been updated, including population forecasts and baseline incidence rates. A majority of these updated assumptions are discussed in detail in the recent $PM_{2.5}$ Regulatory Impact Analysis (U.S. EPA, 2006b).

The benefits analysis included here updates the estimates found in the Chestnut and Mills (2005) study. This analysis was performed using EPA's Benefits Mapping and Analysis Program (BenMAP) (U.S. EPA, 2008a). BenMAP is a tool that estimates the impacts of a change in air pollution on human health. Specifically, for this analysis, BenMAP was used to estimate the human health benefits from a reduction in $PM_{2.5}$ and ground-level ozone pollution due to implementation of ARP emission reductions. The analysis relied on modeled air quality data representing expected air quality in 2010, both in the absence of the ARP and with ARP implementation. BenMAP used these geographically distributed estimates of 2010 air quality to calculate a reduction in $PM_{2.5}$ and ground-level ozone concentrations attributable specifically to ARP implementation. BenMAP combined this pollution reduction with geographically specific population data and baseline incidence data and entered this information into epidemiological functions to estimate health benefits. The epidemiological functions used for this analysis were the most recent sets of functions used by EPA for health effects assessments and employed in the $PM_{2.5}$ Regulatory Impact Analysis (U.S. EPA, 2006b).

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

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 the sum of individual rows due to rounding.
Source: U.S. EPA, 2009b

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
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 Day	910,000	$87
Acute Respiratory Symptoms	2,600,000	$170
Total Value Range		**$4,100–$17,000**

Note: Totals may not reflect the sum of individual rows due to rounding.
Source: U.S. EPA, 2009b

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; however, the sources 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 shut down 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.

Factors Responsible for the Low Cost of SO$_2$ 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.

While this flexible emission-reduction framework is designed to maximize efficiency and minimize costs, some inefficiencies do exist. A recent analysis 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 (Burtraw and Szambelan, 2009). Still, multiple studies on Title IV implementation have found that the program has had lower-than-expected costs, particularly for Phase II, 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 $940 million (Carlson et al., 2000) to $814 million (Ellerman, 2003)—the cost of emission reductions was found to be approximately 50% lower than similar abatement under a command and control program (all estimates in 2000$). While assessments of Phase I costs show the cost effectiveness of reductions, an assessment of Phase II cost estimates shows that costs were much lower than initially projected.

One of the first projections of Phase II costs from the Edison Electric Institute shows an expected cost of $7.5 billion annually in the year 2010. Similarly, the first EPA projection (1990) for annual Phase II costs was approximately $6 billion annually. However, as the approach of Phase I neared, projections for Phase II costs declined significantly. In 1994, the U.S. General Accounting Office (GAO) estimated Phase II costs to be approximately $2.5 billion per year in 2010 (2000$) (U.S. GAO, 1994). Following that assessment, estimates provided by Ellerman (2003) 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 SO$_2$ component to be between $1.1 and $1.8 billion (2000$) (NSTC,

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 RTC (NSTC, 2005). Chestnut and Mills (2005) estimated the total annual costs for reducing SO_2 at approximately $2 billion (2000$) per year, with NO_x emission reductions costing an additional $1 billion annually— still a fraction of the initial cost estimates.

Additionally, a report (U.S. EPA, 2011) under Section 812 of the CAAA analyzing the costs and benefits of implementing CAAA programs (Titles I through IV) estimates that the direct compliance costs in the year 2000 of implementing all programs affecting the utility sector was $1.37 billion (2006$), or $1.17 billion when deflated to year 2000 (using the consumer price index [CPI] inflation calculator). Given that a substantial portion of the utility sector emission reductions under CAAA programs between 1990 and 2000 resulted from Title IV implementation, this is a reasonable qualitative estimate of implementing Title IV through 2000. This estimate corroborates the estimates discussed above and included in the 2005 NAPAP RTC (NSTC, 2005). The same Section 812 report (U.S. EPA, 2011) estimates costs of CAAA implementation in 2010 and 2020. Those estimates show that the cost of CAAA program implementation increases in each of those years due to the implementation of

CAIR and other emission-reduction programs.

The costs to the government of administering the Title IV SO_2 component are also less than in conventional regulatory programs. For example, the Title IV performance-based approach eliminates the need to devise source-specific emission limits, review control technologies, and prepare and approve detailed compliance schedules and permits. Because the regulating authority does not need to approve each source's compliance choices, the focus is on ensuring that each source has at least one allowance for each unit of pollution emitted, which entails less administrative resources and expenditure.

Program Assessment Tools

Emission Monitoring and Reporting

The ARP requires regulated sources to measure, record, and report emissions using continuous emission monitoring systems (CEMS) or an approved alternative measurement method. The vast majority of emissions are monitored with CEMS; however, alternatives are used at some facilities to provide an efficient means of monitoring emissions from the large universe of EGUs with lower overall mass emissions. Table 1-4 shows the number of units with and without SO_2 CEMS for various fuel types in 2009, as well as the amount of SO_2 emissions monitored using CEMS. Although only 31% of units use CEMS, 99% of all SO_2 emissions from ARP sources are monitored in this fashion.

Table 1-4. EGUs and SO_2 Emissions Covered by Monitoring Method for the ARP, 2009				
EGU Type	Type of Monitoring System	Number of EGUs Monitored	Percentage of EGUs Monitored	Percentage of SO_2 Emissions Monitored
Coal-Fired	CEMS	1,044	29.4	98.8
Gas-Fired	CEMS	14	0.4	0.03
	Non-CEMS	2,277	64.1	0.06
Oil-Fired	CEMS	43	1.2	0.16
	Non-CEMS	158	4.5	0.8
Other	CEMS	13	0.4	0.15
	Non-CEMS	1	<0.1	<0.01

Note: The table excludes affected units that did not operate in 2009. "Other fuel units" include units that in 2009 combusted primarily wood, waste, or other nonfossil fuels.
Source: U.S. EPA, 2010b

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

Air Quality, Deposition, and Ecological Monitoring

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

CASTNET and NADP/NTN monitoring site in Gunnison County, CO (photo courtesy of U.S. EPA).

Wet Deposition Monitoring

The National Atmospheric Deposition Program/ National Trends Network (NADP/NTN) is a nationwide network of predominantly rural precipitation monitoring stations. Operating since 1978, the network collects weekly wet acid deposition data to determine geographic patterns and temporal long-term trends. NADP/NTN is responsible for measuring the wet deposition component of total pollution loads across the United States. NADP/NTN is a collaborative effort between many different organizations and consists of 250 monitoring stations spanning the continental United States, Alaska, Canada, Hawaii, Puerto Rico, and the Virgin Islands (Figure 1-6). Additionally, the Atmospheric Integrated Research Monitoring Network (AIRMoN) is a NADP wet deposition subnetwork designed to quantify the extent to which changes in emissions affect air quality and atmospheric deposition at selected locations. AIRMoN-wet focuses on daily measurements of acids, nutrients, and base cations, including ammonium, in precipitation at seven sites. Quality assured data for all NADP networks are available from the NADP Web site at http://nadp.sws.uiuc.edu/.

Air Quality and Dry Deposition Monitoring

The Clean Air Status and Trends Network (CASTNET) is a regional, long-term environmental monitoring program administered and operated by EPA and NPS. Developed from the existing National Dry Deposition Network (NDDN), CASTNET was established in 1991 under the CAAA. The regional monitoring network was formed to assess trends in acidic deposition due to emission-reduction programs, such as the ARP. CASTNET has since become the nation's primary monitoring network for measuring concentrations of air pollutants that form the dry component of acidic deposition and affect regional ecosystems

and rural ambient ozone levels. CASTNET is able to provide the data needed to assess and report on geographic patterns and long-term temporal trends in ambient air pollution and dry atmospheric deposition. Presently, a total of 86 operational CASTNET sites are located in or near rural areas and sensitive ecosystems and collect data on ambient levels of pollutants where urban influences are minimal (Figure 1-6). As part of an interagency agreement, NPS sponsors 27 of the CASTNET sites, which are located in national parks and other Class-I areas designated as deserving special protection from air pollution. Quality assured data are available at http://www.epa.gov/CASTNET/.
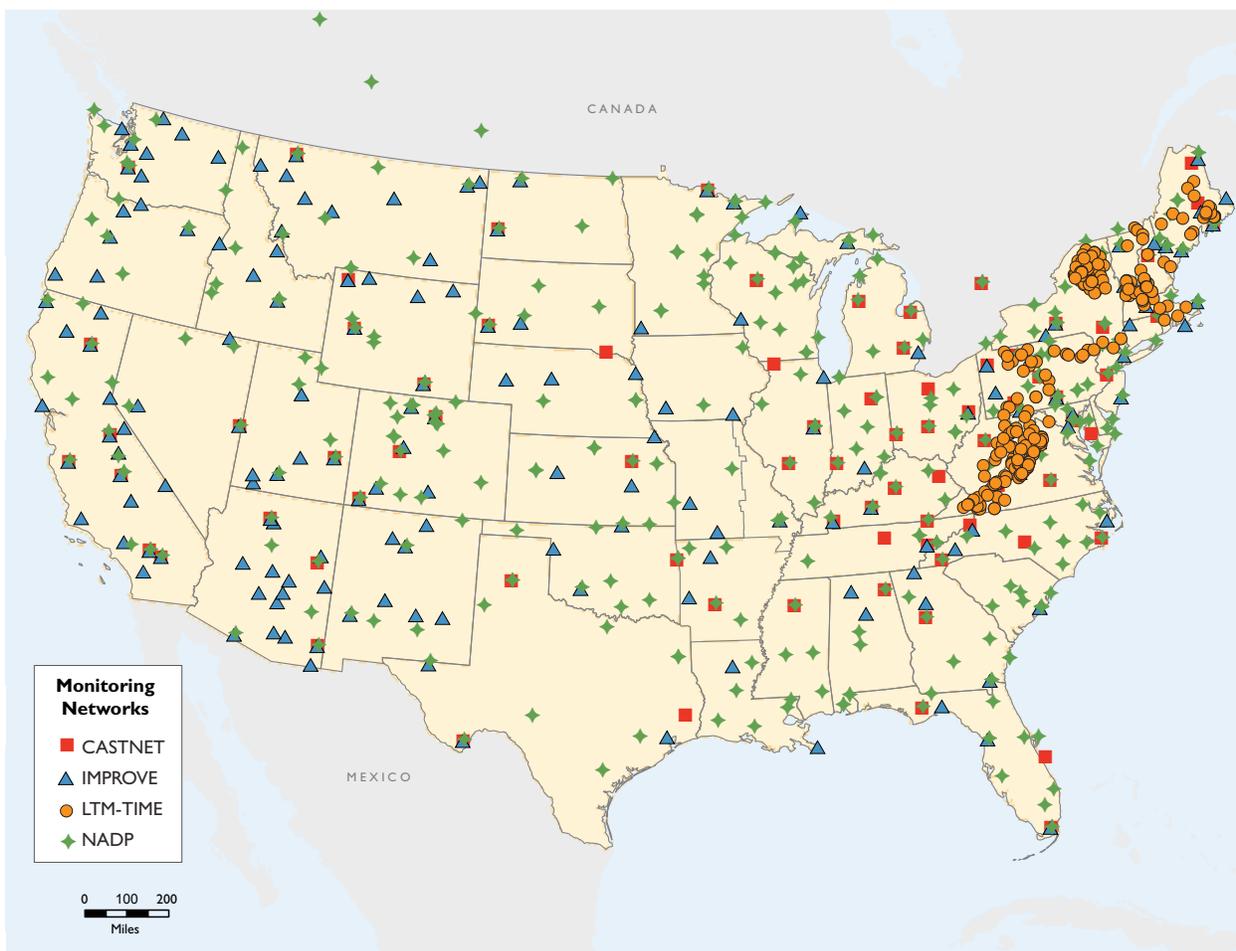
Figure 1-6. Air quality, deposition, and water quality monitoring networks (prepared by U.S. EPA).

Visibility Monitoring

Interagency Monitoring of Protected Visual Environments (IMPROVE) is a long-term monitoring network that measures current visibility conditions, tracks changes in visibility, and determines the causes of visibility impairment in national parks and wilderness areas. IMPROVE was established in 1985 to aid the development of Federal and state implementation plans to protect visibility in Class I areas, as stipulated in the 1977 CAAA. IMPROVE began collecting data in 1988 at 20 Class I areas. The network expanded to monitor the impacts of the Regional Haze Rule and now consists of 167 sites nationwide (Figure 1-6). IMPROVE is a collaborative monitoring effort. Data and additional information about IMPROVE are available at http://vista.cira.colostate.edu/improve.

Visibility varies at the Look Rock Air Quality Monitoring Station in the Great Smoky Mountains National Park. On a good visibility day (top photo), one can see for 100 miles. On a bad visibility day (bottom photo), the visual range shrinks to 15 miles (photo courtesy of NPS).

Ecological Monitoring – Lakes and Streams

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

CHAPTER 2

Results of the Acid Rain Program: Status and Trends of Emissions, Deposition, Air Quality, Surface Water, and Visibility, 1990 to 2009

Since its inception in 1995, the ARP has made significant progress in reducing emissions. Starting in 2007, emissions were below the 2010 SO_2 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.

Emissions

SO₂ Emissions

As shown in Figure 2-1, SO_2 emissions in the United States declined between 1980 and 2009 (see data available at http://www.epa.gov/ttn/chief/trends). In 2009, ARP sources had reduced annual SO_2 emissions by 67%, compared with 1980 levels, and 64%, compared with 1990 levels. ARP sources emitted 5.7 million tons of SO_2 in 2009, which was well below the 2009 annual emission cap of 9.5 million tons and already below the annual cap of 8.95 million tons established

> The SO_2 requirements under the ARP apply to EGUs (i.e., fossil fuel–fired combustors) that serve a generator that provides electricity for sale. The vast majority of ARP SO_2 emissions result from coal-fired EGUs, although the program also applies to oil and gas units.

by Title IV as the level for full implementation of the ARP in 2010. In addition, national SO_2 emissions from all sources (including those not covered by the ARP) also have fallen by approximately 63%, from nearly 26 million tons in 1980 to about 9.5 million tons in 2009. The declines in SO_2 emissions from all sources in the United States likely result from a combination of several factors, including the following:

- Increased use of emission-control technologies, especially flue-gas desulfurization (or scrubbers);

- Reduced heat input (a measure of the amount of fuel used) at ARP sources; and

- Fuel switching.

The states with the highest-emitting sources in 1990 (i.e., Illinois, Indiana, Kentucky, Missouri, Ohio, Pennsylvania, Tennessee, and West Virginia) have generally seen the greatest SO_2 reductions under the ARP (see Figure 2-2).

Most of these states are upwind of the areas the ARP was designed to protect, and reductions have resulted in important environmental and health benefits over a large region.
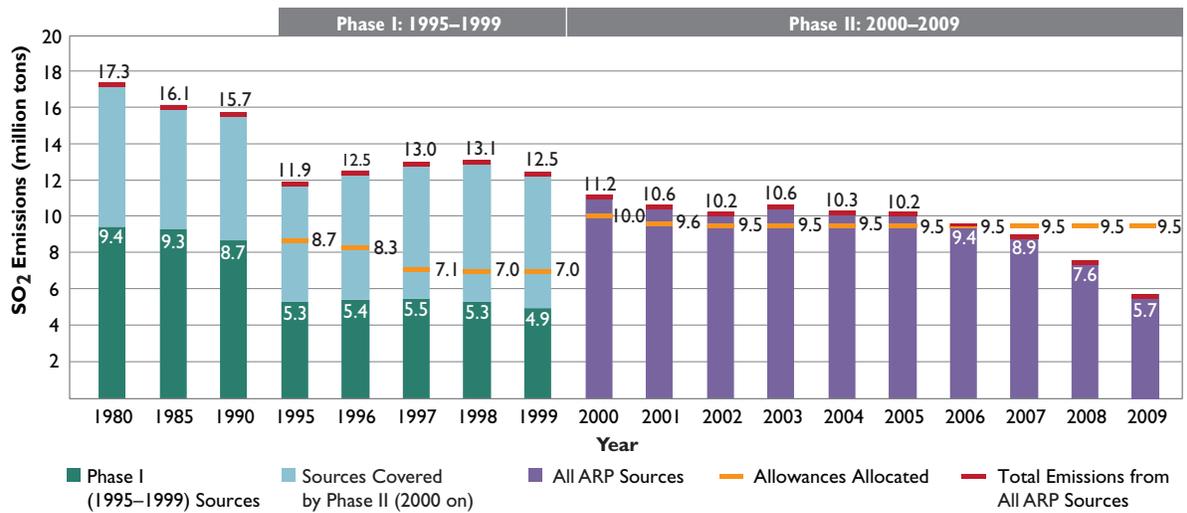
Figure 2-1. SO_2 emissions from ARP sources, 1980 to 2009 (U.S. EPA, 2010b).
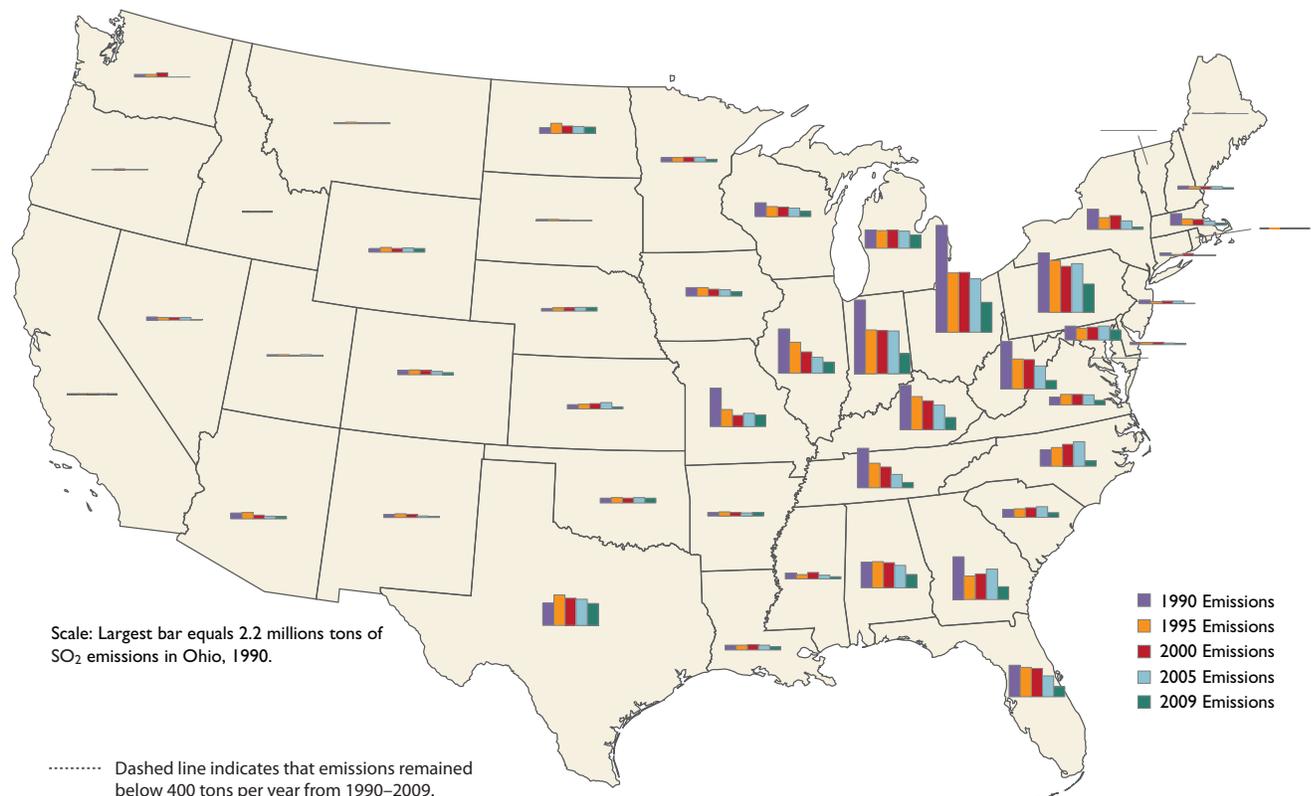
Figure 2-2. Changes in annual SO_2 emission levels by state from 1990 to 2009 for ARP sources (U.S. EPA, 2010b).

From 1990 to 2009, annual SO_2 emissions in 41 states and the District of Columbia fell by a total of approximately 10 million tons. In contrast, annual SO_2 emissions increased by a total of 39,521 tons in seven states (Idaho, Montana, Nebraska, North Dakota, Oregon, Rhode Island, and Vermont) from 1990 to 2009. The seven states with the greatest reductions in annual emissions since 1990 include Ohio, which decreased emissions by more than 1.5 million tons, and Illinois, Indiana, Kentucky, Pennsylvania, Tennessee, and West Virginia, each of which reduced total emissions during this time period by more than 500,000 tons. To view emission data in an interactive format using Google Earth or a similar three-dimensional platform, go to http://www.epa.gov/airmarkets/progress/interactivemapping.html.

NO_x Emissions

NO_x emissions from all ARP sources were 2.0 million tons in 2009, and emissions have decreased 4.1 million tons since 1995 (see Figure 2-3). The goal of the ARP NO_x program is to limit NO_x emissions from the affected coal-fired boilers so that their emissions are at least 2 million tons less than the projected level for the year 2000 without implementation of Title IV. The 2009 emission level of 2.0 million tons is 6.1 million tons less than the projected level of annual NO_x emissions in 2000 without the ARP, or more than triple the Title IV NO_x emission-reduction objective. While the ARP was responsible for a large portion of these annual NO_x reductions, other programs—such as the NO_x Budget Trading Program (NBP) under EPA's NO_x State Implementation Plan (SIP) Call and other regional and state NO_x emission-control programs—also contributed significantly to NO_x reductions.

As noted, from 1995 to 2009, annual NOx emissions from ARP sources dropped by 4.1 million tons, a net decrease of 67%. During this period, 44 states and the District of Columbia reduced NO_x emissions, while 6 other states accounted for only about 5,542 tons of increased NO_x emissions (see Figure 2-4).

Other Programs Contributing to NO_x Emission Reductions

Ozone Transport Commission (OTC)—Established under the 1990 CAAA, this commission consists of states primarily located in the Northeast and mid-Atlantic region. The OTC developed the OTC NO_x Budget Program, which operated from 1999–2002. 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 SO_2 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 SO_2 program. In 2009, CAIR was remanded, but remained in place while EPA developed a new rule (U.S. EPA, 2009e).

Cross-State Air Pollution Rule (CSAPR)—On July 6, 2011, EPA finalized the CSAPR, which replaces CAIR and requires eastern, southern, and central states to significantly improve air quality by reducing power plant emissions that cross state lines and contribute to ground-level ozone and fine particle pollution in other states. The rule reduces annual SO_2, annual NO_x, and ozone-season NO_x emissions, while providing sources with flexibility in how to comply with the program. The rule also allows air quality-assured trading that uses an allowance market infrastructure based on existing, successful allowance trading programs. At the same time, the trading ensures the elimination, in each state, of the emissions that contribute to downwind air quality problems.
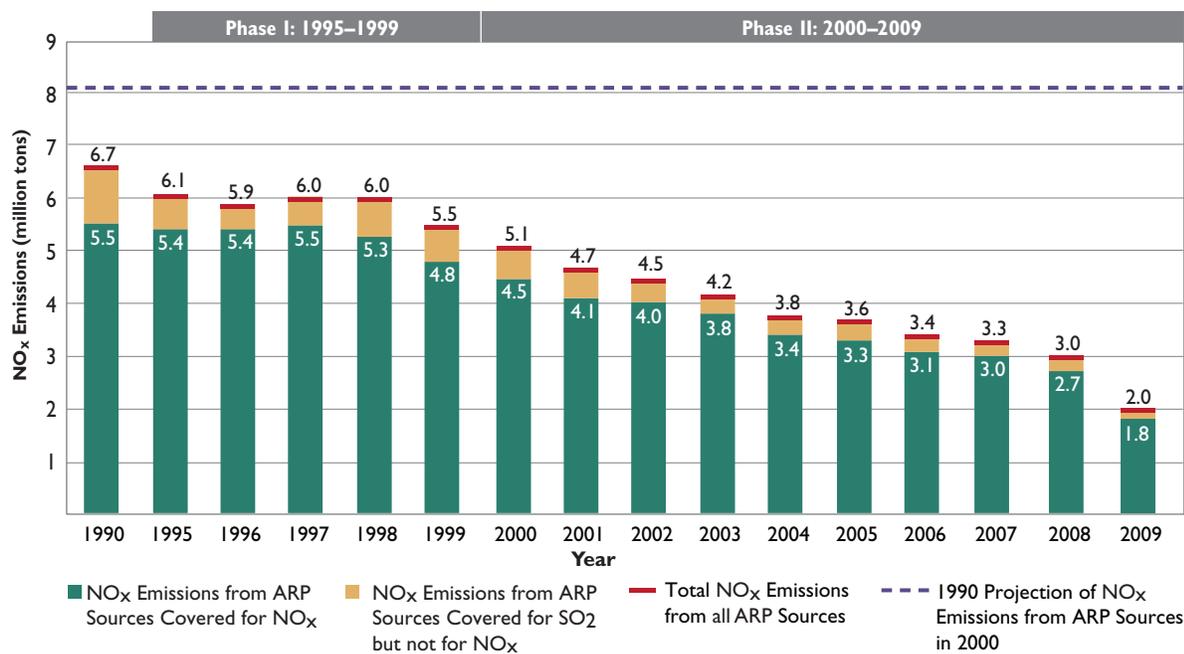
Figure 2-3. NO$_x$ emission levels for all ARP sources, 1990 to 2009 (U.S. EPA, 2010b).

Scale: Largest bar equals 500,000 tons of NO$_x$ emissions in Ohio, 1990.

Note: Darker area represents the NO$_x$ Budget Trading Program geographic region.

.... Dashed line indicates that emissions remained below 400 tons per year from 1990–2009.
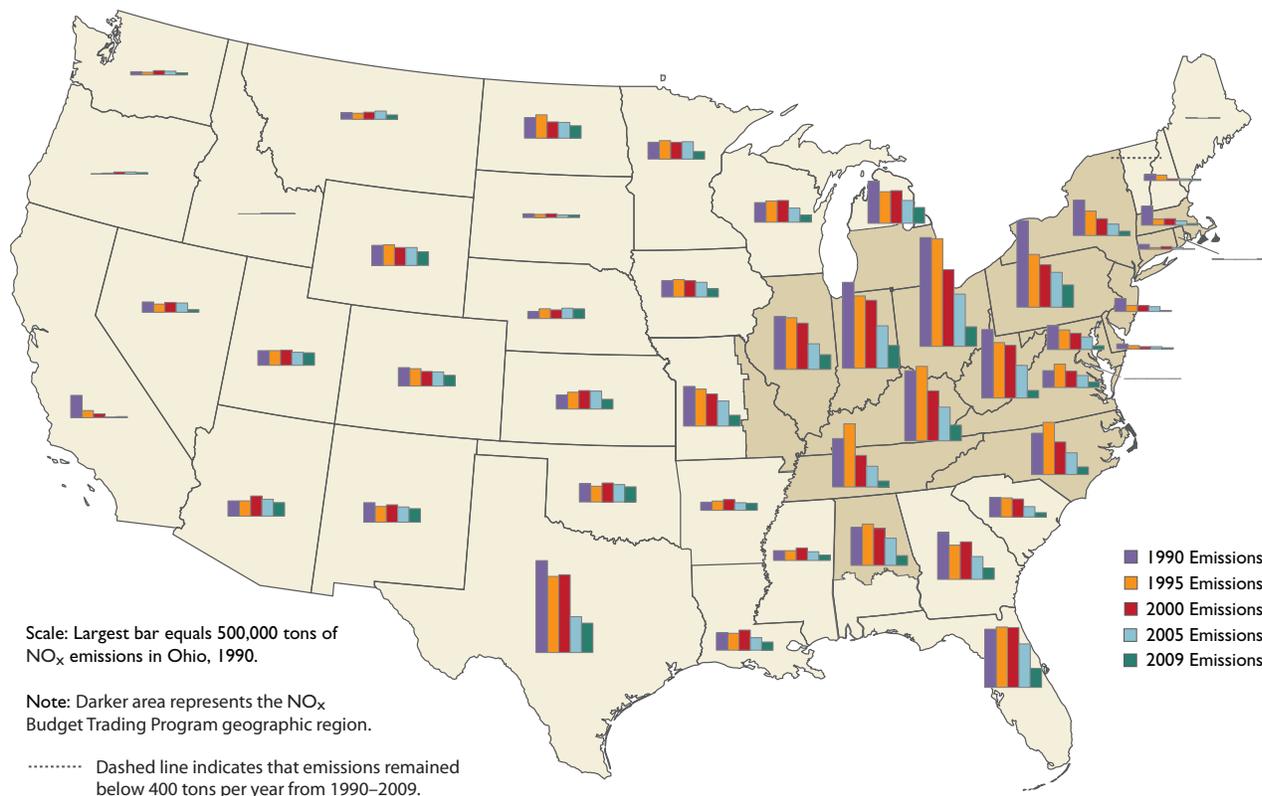
Figure 2-4. Changes in annual NO$_x$ emission levels by state from 1990 to 2009 for ARP sources (U.S. EPA, 2010b).

The states subject to EPA's 1998 NO_x SIP Call have achieved significant reductions in ozone-season NO_x emissions since the baseline years of 1990 and 2000. All of these states have achieved reductions since 1990 as a result of programs implemented under the 1990 CAAA, with many states reducing their emissions by more than half since 1990. A significant portion of these decreases in NO_x emissions has been achieved since 2000, largely as a result of decreases under ozone-season NO_x trading programs implemented by the Ozone Transport Commission (OTC) from 1999 to 2002, and under the NO_x SIP Call from 2003 to 2009. For reports about these programs, go to http://www.epa.gov/airmarkets/progress/progress-reports.html.

Air Quality

Emission reductions achieved under the ARP have led to improvements in air quality, with significant benefits to human health. Since the early 1990s, improvements in ambient SO_2, sulfate, and nitrate concentrations have varied regionally in the eastern United States (see Table 2-1).

SO_2 and Sulfate

Data collected from monitoring networks show that the decline in SO_2 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 SO_2 annual mean ambient concentrations decreased 76% between 1980 and 2009, 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 1980–2009, the largest single-year reduction (20%) occurred in the first year of the ARP, between 1994 and 1995. The second-largest single-year reduction (16%) occurred most recently between 2008 and 2009. These trends are consistent with the regional ambient air quality trends observed in CASTNET.

During the late 1990s, dramatic regional improvements in SO_2 and ambient sulfate concentrations were observed at CASTNET sites throughout the eastern United States following

Table 2-1. Regional Changes in Air Quality from the 1989 to 1991 and 2007 to 2009 Observation Periods				
Region	Average Concentration (µg/m³), 1989–1991[a]	Average Concentration (µg/m³), 2007–2009[a]	Percentage Change[b]	Number of Sites
Ambient SO₂				
Northeast[c]	5.5	1.7	-69	3
Mid-Atlantic	13.0	5.0	-62	12
Southeast	5.1	2.2	-57	9
Midwest	11.0	4.1	-63	10
Ambient Sulfate				
Northeast[c]	3.5	1.8	-49	3
Mid-Atlantic	6.3	3.5	-44	12
Southeast	5.4	3.2	-41	8
Midwest	5.8	3.1	-47	10
Total Ambient Nitrate				
Northeast[c]	1.8	1.0	-44	3
Mid-Atlantic	3.3	2.0	-39	12
Southeast	2.2	1.5	-32	8
Midwest	4.6	3.2	-30	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, 2010c

| Phase I: 1995–1999 | Phase II: 2000–2009 |
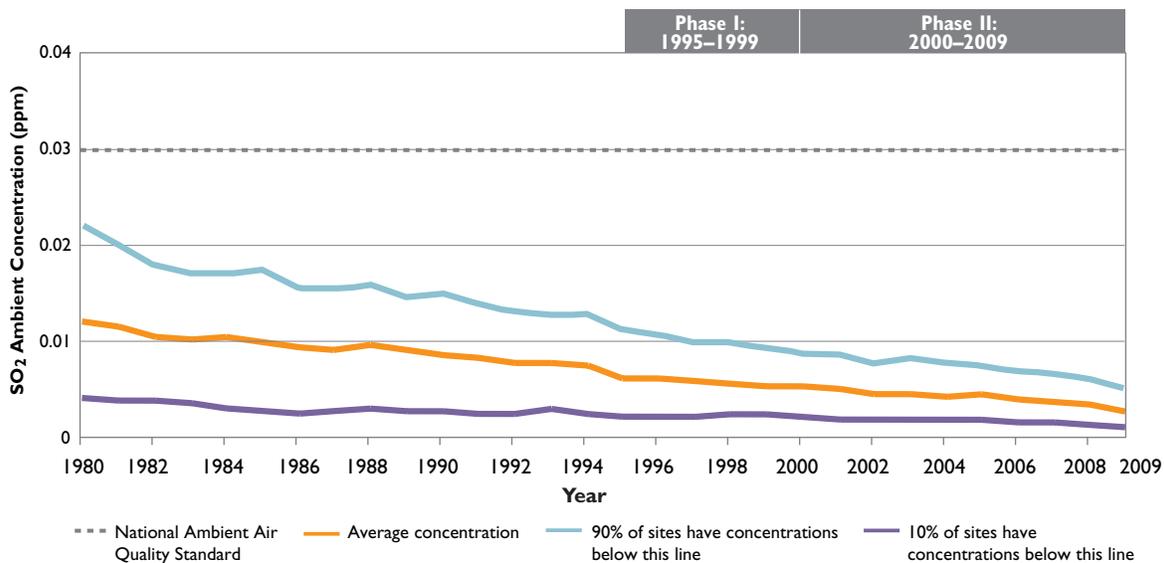
Figure 2-5. National SO$_2$ air quality, 1980 to 2009 (U.S. EPA, 2010c).

implementation of Phase I of the ARP. These improvements continue today. Analyses of regional monitoring data from CASTNET show the geographic pattern of SO$_2$ and airborne sulfate in the eastern United States. Three-year mean annual concentrations of SO$_2$ and sulfate from CASTNET long-term monitoring sites in the eastern United States are compared for the 1989 to 1991 and 2007 to 2009 observation periods (see Figures 2-6 and 2-7). For the 1989 to 1991 observation period, few data on the ambient concentrations of SO$_2$ and sulfate exist from CASTNET sites in the western United States; therefore, changes in ambient concentrations in this region could not be assessed for these two periods.

Figure 2-6 shows that from 1989 to 1991, prior to implementation of Phase I of the ARP, the highest annual ambient concentrations of SO$_2$ in the eastern United States were observed in western Pennsylvania and along the Ohio River Valley. In comparison, the map for the 2007 to 2009 observation period indicates a significant decline in ambient SO$_2$ concentrations in nearly all affected areas after implementation of the ARP and other programs.

Concentration—A measure of how much of a given substance (e.g., nitric acid) is mixed with another substance (e.g., water), expressed as a quantity or mass per unit volume.

Deposition—A measure of the amount of chemical that is transferred to the earth's surface—typically by rainfall, snow, hail, or sleet—expressed as the amount or mass transferred to a specific area of ground.

Two scenarios

Deposition is a combination of both concentration and the amount of rain. For example, the West has a higher atmospheric pollutant concentration, but less rainfall; the East has a lower concentration, but much more rain and therefore more deposition.

WEST

High concentration 20 mg/L **X** Low precipitation 11 inches **=** Deposition 5.5 g/m^2/year 55 kg/hectare/year

EAST

Low concentration 10 mg/L **X** High precipitation 40 inches **=** Deposition 11.0 g/m^2/year 110 kg/hectare/year
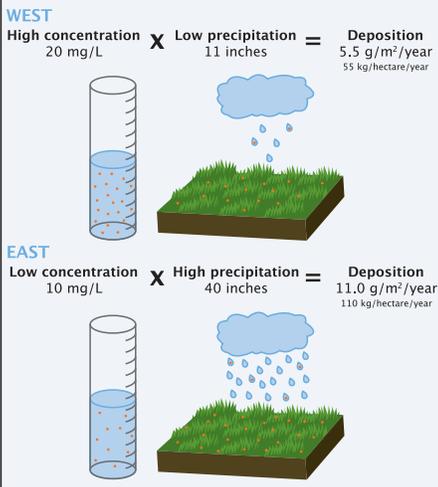
Note: Example is only for wet deposition (courtesy of NADP).
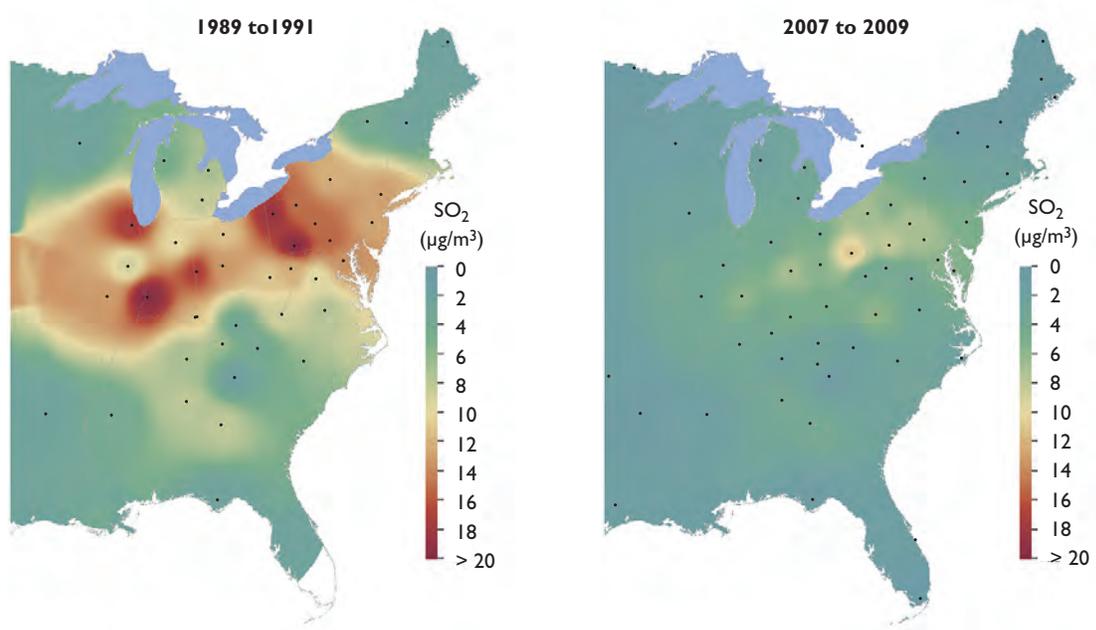
Note: For maps depicting these trends for the entire conterminous United States, visit http://www.epa.gov/castnet.
Dots on all maps represent monitoring sites. Lack of shading for southern Florida on the map on the left indicates lack
of monitoring coverage in the 1989 to 1991 time period.

Figure 2-6. Annual mean ambient SO_2 concentrations in the eastern United States for the 1989–1991 and 2007–2009 observation periods (U.S. EPA, 2010c).
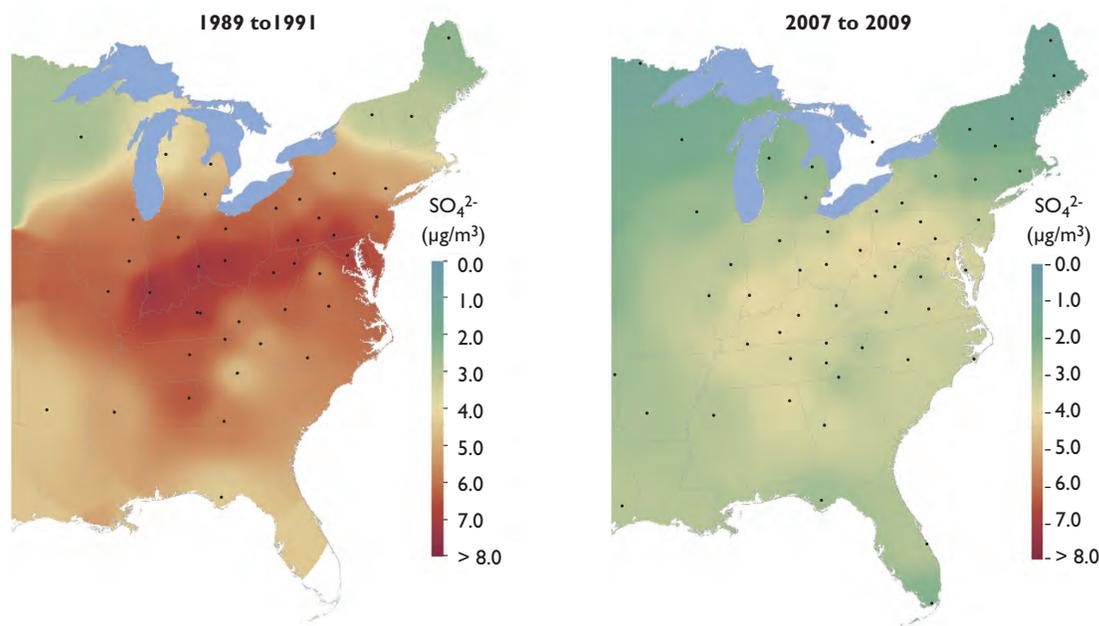
Note: For maps depicting CASTNET data for the conterminous United States, visit http://www.epa.gov/castnet.
Dots on all maps represent monitoring sites. Lack of shading for southern Florida on the map on the left indicates lack
of monitoring coverage in the 1989 to 1991 time period.

Figure 2-7. Annual mean ambient sulfate concentrations in the eastern United States for the 1989–1991 and 2007–2009 observation periods (U.S. EPA. 2010c).

What is the Difference Between SO_2, sulfate, and total sulfur deposition?

Sulfur is an element that exists in several different forms. SO_2 and sulfate are the forms of sulfur examined in this report.

SO_2—When sulfur-containing substances, such as coal, are burned, the sulfur is primarily converted to SO_2. Emissions from ARP sources and ambient air quality are discussed in terms of SO_2 concentrations.

Sulfate—SO_2 is oxidized in the atmosphere to form sulfate (SO_4^{2-}). 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 of sulfate (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.

Like SO_2 concentrations, ambient sulfate concentrations have decreased since the ARP was implemented, with average values decreasing by 41% to 49% 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 ($\mu g/m^3$). Most of the eastern United States experienced annual ambient sulfate concentrations greater than 5$\mu g/m^3$. 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).

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.

NO_x and Nitrate

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 also can be affected by emissions transported via air currents over wide regions.

From 2007 to 2009, reductions in NO_x emissions during the ozone season from power plants under the NO_x SIP Call and CAIR have continued to result in significant region-specific improvements in ambient total nitrate (i.e., nitrate [NO_3^-] plus nitric acid [HNO_3]) concentrations. For instance, annual mean ambient total nitrate concentrations for the 2007 to 2009 observation period in the mid-Atlantic region were 39% less than the annual mean concentration in the 1989 to 1991 period (see Figure 2-8). Although these improvements might be partly attributed to added NO_x controls installed for compliance with the NO_x SIP Call and CAIR, the findings at this time are not conclusive.
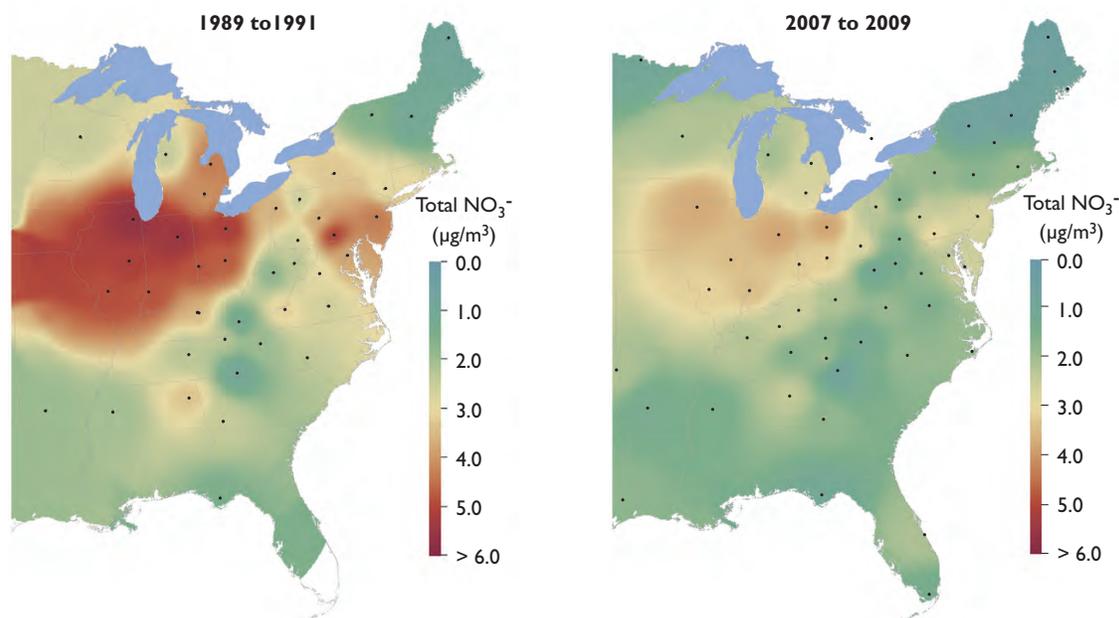
1989 to1991

2007 to 2009

Total NO$_3^-$
(µg/m^3)

0.0

1.0

2.0

3.0

4.0

5.0

> 6.0

Total NO$_3^-$
(µg/m^3)

0.0

1.0

2.0

3.0

4.0

5.0

> 6.0

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 2-8. Annual mean ambient total nitrate concentrations in the eastern United States for the 1989–1991 and 2007–2009 observation periods (U.S. EPA, 2010c).

What is the difference between NO$_x$, nitrate, and total nitrogen deposition?

Nitrogen 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 (NO$_3^-$). Similar to SO$_2$, 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.

Acid Deposition

NADP/NTN monitoring data show significant improvements across the eastern United States in the primary acid deposition indicators—sulfur and nitrogen. The sulfur indicator is assessed using the atmospheric deposition levels of sulfate (wet deposition) and sulfur (dry and total deposition). The nitrogen indicator is measured using levels of wet inorganic nitrogen deposition (combined deposition of inorganic nitrate and ammonium ions in wet deposition) and total inorganic nitrogen deposition (combined deposition of inorganic nitrate and ammonium deposition in precipitation, dry particulate deposition, and gaseous nitric acid). Table 2-2 provides an overview of changes in atmospheric deposition in regions of the eastern United States from the 1989 to 1991 and 2007 to 2009 observation periods.

Table 2-2. Regional Changes in Atmospheric Deposition from the 1989 to 1991and 2007 to 2009 Observation Periods				
Region	Average Deposition (kg/ha), 1989–1991	Average Deposition (kg/ha), 2007–2009	Percent Change[a]	Number of Sites
Wet Sulfate				
Northeast	7.5	4.3	-43	17
Mid-Atlantic	9.2	5.3	-42	11
Southeast	6.1	3.5	-43	23
Midwest	7.1	4.0	-44	27
Dry Sulfur				
Northeast[b]	2.9	1.0	-66	3
Mid-Atlantic	6.7	2.9	-57	11
Southeast[b]	1.2	0.7	-42	2
Midwest	6.5	2.8	-57	10
Total Sulfur[c]				
Northeast[b]	9.8	4.7	-52	3
Mid-Atlantic	16	8.0	-50	11
Southeast[b]	8.0	4.6	-43	2
Midwest	15.0	7.0	-53	10
Wet Inorganic Nitrogen				
Northeast	5.6	4.1	-27	17
Mid-Atlantic	6.2	4.5	-27	11
Southeast	4.4	3.4	-23	23
Midwest	5.8	4.9	-16	27
Dry Inorganic Nitrogen				
Northeast[b]	1.4	0.6	-57	3
Mid-Atlantic	2.5	1.5	-40	11
Southeast[b]	0.9	0.8	-11	2
Midwest	2.5	1.8	-28	10
Total Inorganic Nitrogen[c]				
Northeast[b]	6.5	4.4	-32	3
Mid-Atlantic	8.7	6.0	-31	11
Southeast[b]	5.9	4.8	-19	2
Midwest	9.0	6.9	-23	10

[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, 2010c

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

Sulfur Deposition

Between the 1989 to 1991 and 2007 to 2009 observation periods, decreases in wet deposition of sulfate averaged more than 40% for the eastern United States. Some of the greatest decreases have occurred in the mid-Appalachian region, including Maryland, New York, West Virginia, Virginia, and most of Pennsylvania (see Figure 2-9). Less dramatic reductions have been observed across much of New England, portions of the southern Appalachian Mountains, and some areas of the Midwest. A principal reason for decreased sulfate deposition levels in the Northeast is a reduction in the long-range transport of sulfate from emission sources located in the Ohio River Valley. The decreases in sulfate documented in the Northeast, particularly across New England and portions of New York, were also affected by SO_2 emission reductions in eastern Canada.

Portions of the eastern United States have also experienced significant decreases in dry and total sulfur deposition since the initiation of the ARP. Dry sulfur deposition levels in the mid-Atlantic region and the Midwest have decreased by 57% between the 1989 to 1991 and 2007 to 2009 observation periods. Decreases in total sulfur deposition for the same areas were 50% and 53%, respectively, for the same period. Continuous data records for dry and

1989 to1991   **2007 to 2009**
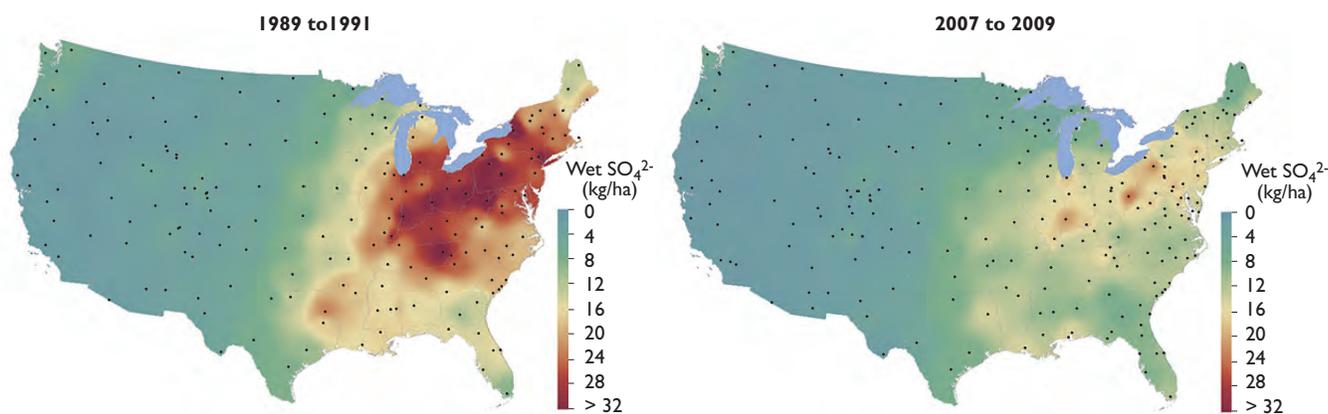
Note: Dots on all maps represent NADP monitoring sites. Data source: NADP

Figure 2-9. Annual mean wet sulfate deposition in the United States for the 1989–1991 and 2007–2009 observation periods (U.S. EPA, 2010c).

total sulfur deposition are available from only a few sites in the Northeast and Southeast; therefore, the observed decreases in deposition may not be representative of these regions.

Other studies reported in the scientific literature indicate decreases in sulfate concentrations in precipitation since the 1990s; these decreases are similar to those reported in this report. Historical data show that decreases in sulfate concentrations in precipitation are approaching 50% since the 1980s for many monitoring sites in the eastern United States (Lehmann et al., 2005; Kvale and Pryor, 2006). Between the 1989 to 1991 and 2007 to 2009 observation periods, sulfate concentrations in precipitation have decreased by approximately 40% throughout the eastern United States. A strong correlation between large-scale SO_2 emission reductions and large decreases in sulfate concentrations in precipitation has been noted in the Northeast and mid-Atlantic regions (Lehmann et al., 2005), which are among the areas most affected by acid deposition.

Total Reactive Nitrogen

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 (NH_3), ammonium ion (NH_4^+), nitric oxide (NO), nitrite (NO_2^-), nitric acid (HNO_3), nitrous oxide (N_2O), nitrate (NO_3^-), and organic compounds (e.g., urea, amines, nucleic acids). Sources of reactive nitrogen include agricultural practices, vehicle exhaust, and EGUs.

Nitrogen Deposition (Nitrate and Ammonium)

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

- NO_x emission declines have been less than the declines in SO_2 emissions.

- The proportion of nationwide NO_x emissions that originates from EGUs is much lower (about 20%) than the proportion of SO_2 emissions that originates from these same units (about 70%) (U.S. EPA, 2006a).

- NO_x emissions originate from a number of non-ARP sources, including motor vehicles.

- The contribution of ammonia (NH_3) emissions to overall nitrogen deposition originates largely from agricultural sources and is not regulated by the Clean Air Act (Stephen and Aneja, 2008).

- A large number of chemical species of nitrogen contribute to total nitrogen deposition, but are not well measured by existing monitoring networks (Sparks et al., 2008).

Nitrogen deposition levels across the United States vary by region and by the type of deposition. Regional differences in these trends are apparent. The mid-Atlantic and northeastern states generally show the greatest downward trends in nitrogen deposition, and parts of the Southeast and West show the smallest decreases (Lehmann et al., 2005). In some regions (e.g., the Front Range of Colorado, Iowa), nitrogen deposition has increased in recent years (Burns, 2003; Ingersoll et al., 2008). Between the 1989 to 1991 and 2007 to 2009 observation periods, decreases in wet inorganic nitrogen (nitrate and ammonium) deposition levels in the eastern United States ranged from 16% in the Midwest to 27% in the mid-Atlantic region and the Northeast. Decreases in dry and total inorganic

1989 to1991

2007 to 2009

Inorganic
Nitrogen
(kg/ha)
0.0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
9.0
> 10.0

Inorganic
Nitrogen
(kg/ha)
0.0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
9.0
> 10.0
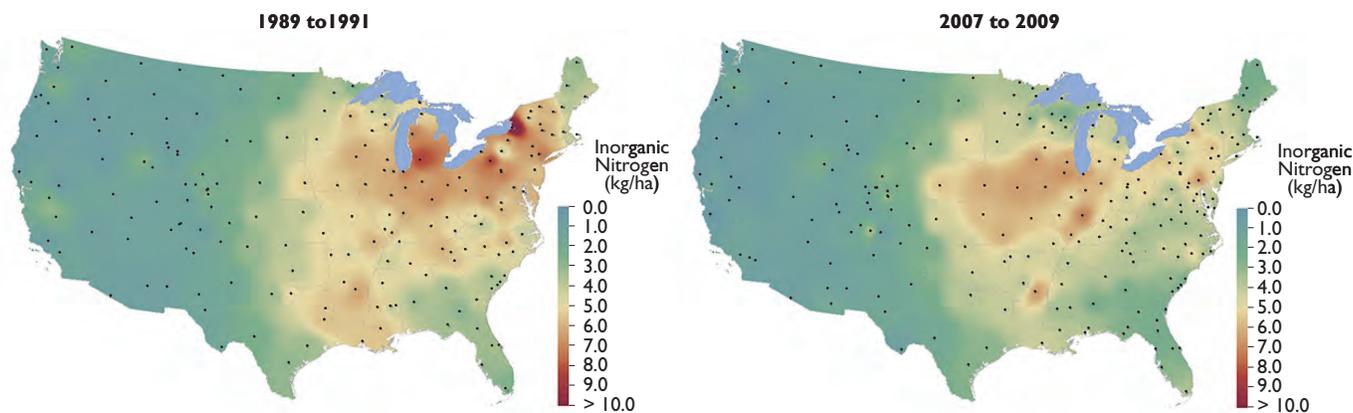
Note: Dots on all maps represent NADP monitoring sites. Data source: NADP

Figure 2-10. Annual mean wet inorganic nitrogen deposition in the United States for the 1989–1991 and 2007–2009 observation periods. (U.S. EPA, 2010c).

nitrogen deposition generally have been greater than those of wet deposition, with a 23% and 31% decrease in total nitrogen deposition for the Midwest and the mid-Atlantic region, respectively.

Surface Water Monitoring

Acid rain, resulting from SO_2 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.

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 (SO_4^{2-}) and nitrate (NO_3^-) 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

Waters of Acute Concern
Water acidic all the time; sensitive organisms cannot survive

Waters of Elevated Concern
Water still acidified seasonally or after storms; sensitive organism richness still low

Waters of Moderate Concern
Significant recovery, but water still acidified at times; sensitive organisms may survive

Waters of Low Concern
Recovery of water chemistry; even sensitive organisms can survive

Note: See Chapter 3 for further discussion of the ecological effects of acidification.
Waters of acute concern are defined as having ANC less than 0 µeq/L.
Waters of elevated concern are defined as having ANC from 0 to less than 50 µeq/L.
Waters of moderate concern are defined as having ANC from 50 to less than 100 µeq/L.
Waters of low concern are defined as having ANC greater than or equal to 100 µeq/L.

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

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

Monitoring the trends in these indicators enables determination of whether conditions in acid-sensitive lakes and streams are improving and heading toward chemical recovery or if conditions are degrading. Chemical recovery is a prerequisite for the subsequent recovery of aquatic plants and animals. Movement toward recovery of a lake or stream ecosystem is indicated by increases in ANC levels and decreases in sulfate and nitrate concentrations. Table 2-3 presents the percentage of monitoring sites in four acid-sensitive regions (i.e., New England, Adirondack Mountains, Northern Appalachian Plateau, and Central Appalachians) of the eastern United States (Figure 2-12) with improving sulfate, nitrate, and ANC trends. The sites represent LTM lakes and streams monitored from 1990 to 2008. These regional trends were calculated using data from all the sites located within the region that had a complete data record for the time period considered. Trends are considered statistically significant if they exceed the 95% confidence interval ($p < 0.05$).

As seen in Table 2-3 and Figure 2-13, significant improving trends in sulfate concentrations from 1990 to 2008 are found regionally and 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 show degrading trends, 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.

Surface Water Monitoring Networks

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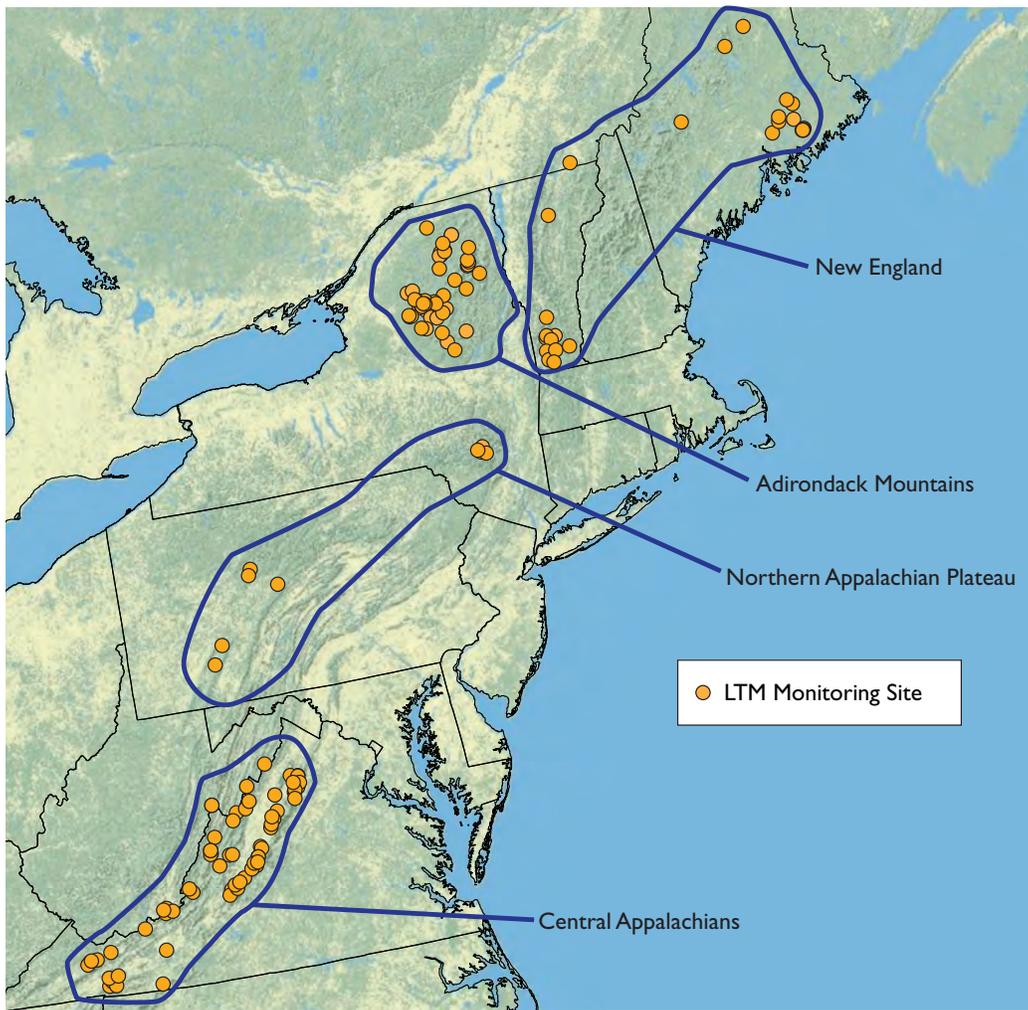
Figure 2-12. LTM site locations and regional groupings (prepared by U.S. EPA).

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 N = 50	90	32	58
Northern Appalachian Plateau	Streams in NY, PA N = 9	78	33	56
New England	Lakes in ME, VT N = 26	96	31	12
Central Appalachians	Streams in VA N = 60	12	45	12

[a] Trends determined by *multivariate Mann-Kendall tests.*
Data source: U.S. EPA
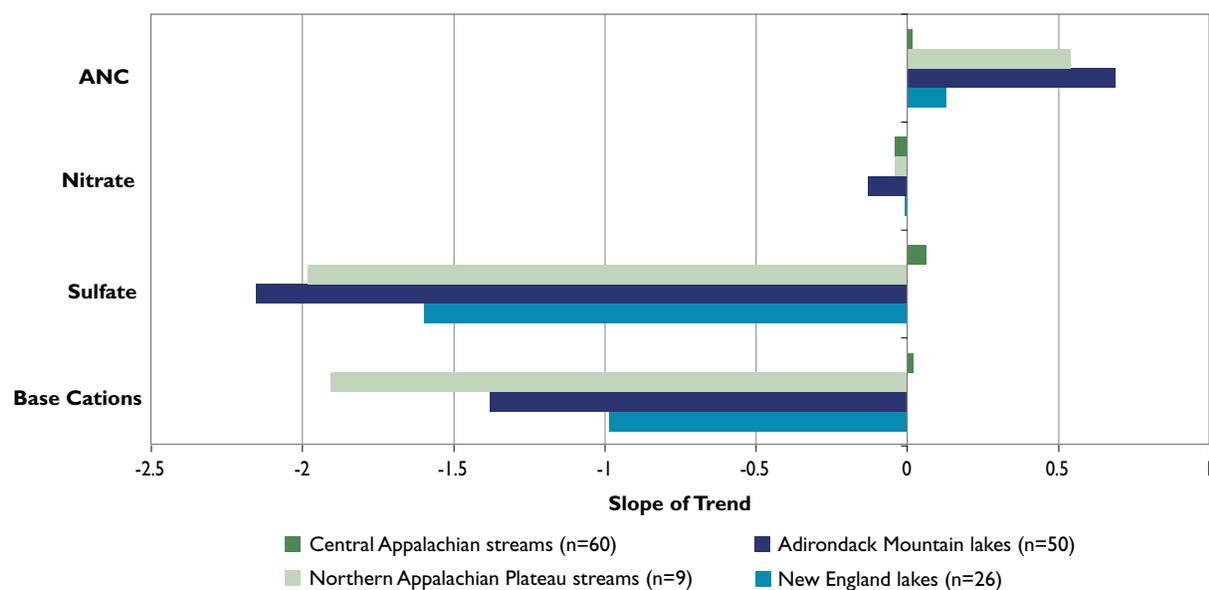
Figure 2-13. Trends in acidification of LTM lakes and streams, 1990–2008 (prepared by U.S. EPA).

Trends in surface water nitrate concentrations are also mixed (Figure 2-13) and indicate flat or slightly degrading nitrate trends at several individual LTM lake and stream sites (Table 2-3). 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 such as soil retention, denitrification rates, and vegetation disturbance.

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

significant trends observed at individual LTM sites showed increasing ANC levels (improving trend) in surface waters. Overall, only seven sites indicate a statistically significant degrading trend in ANC.

> 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.

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

2006 to 2008, while the percentage of lakes in the moderate concern category increased from 6.4% to 13.5%. These results point to a decrease in acidity, particularly for the subset of lakes with low ANC.

Case Study—Critical Loads for Lakes and Streams

The critical loads approach (see text box) is an assessment tool that can be used to determine the degree to which air pollution may be affecting ecological health. This approach provides a useful lens through which to assess the results of

current policies and programs and to evaluate the potential value of proposed policy options in terms of ecosystem protection. The critical loads approach has been employed routinely as an assessment tool for many years in the countries of the European Union and Canada. This case study will explore the extent to which lakes in the Adirondack Mountains and streams in the central Appalachians are protected from acidifying nitrogen and sulfur deposition as a result of Title IV emission reductions. Further discussion of critical loads can also be found in Chapter 3 of this report.

Acid Neutralizing Capacity

Lakes and streams with ANC values defined as less than or equal to 0 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 "moder-

ate 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	Less than 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 to less than 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 to less than 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	Greater than or equal to 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.
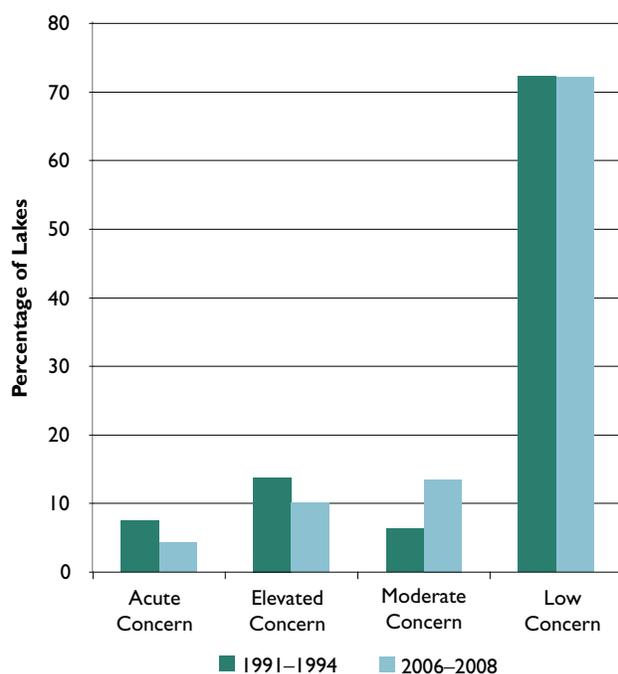
Figure 2-14. Northeastern TIME Lakes by ANC Category, 1991-1994, vs. 2006-2008. Acute Concern— less than 0 μeq/L, Elevated Concern—0 μeq/L to less than 50 μeq/L, Moderate Concern—50 μeq/L to less than 100 μeq/L, Low Concern—greater or equal to 100 μeq/L (prepared by U.S. EPA).

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

For these particular analyses, the critical load represents the combined deposition loads of sulfur and nitrogen to which a lake or stream could be subjected and still have an ANC of 50 μeq/L or higher. While a critical load can be calculated for any ANC level, 50 μeq/L was chosen because it provides a moderate level of protection for most fish and other aquatic organisms, although systems can still become episodically acidic and some sensitive species may be lost. Critical loads of combined total sulfur and nitrogen deposition are expressed in terms of ionic charge balance as milliequivalents per square meter per year (meq/m^2/yr). When actual measured deposition of nitrogen and sulfur is greater than the critical load, the critical load is "exceeded," meaning that combined sulfur and nitrogen deposition was greater than a lake or stream could sustain and still maintain the ANC level of 50 μeq/L or above. In some cases, negative critical loads may occur when the supply of base cations is naturally low. In these cases, the lake or stream would have a natural, pre-

Critical Loads

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 the pollutant exposure is higher than, or "exceeds," the critical load and the ecosystem continues to be exposed to damaging levels of pollutants.

acidification ANC level below 50 μeq/L. An ANC of 50 μeq/L or greater would not be achievable regardless of changes in acidic deposition levels. In order to assess the extent to which regional lake and stream ecosystems are protected by the emission reductions achieved by Title IV, these case studies compare the amount of deposition systems can receive—the critical load—to measured deposition for the period before implementation of the ARP (1989–1991) and for a recent period after ARP implementation (2006–2008).

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

Adirondack Mountain Lakes

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

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

Adirondack mountain lakes are sensitive to acidic deposition (photo courtesy of NASA).
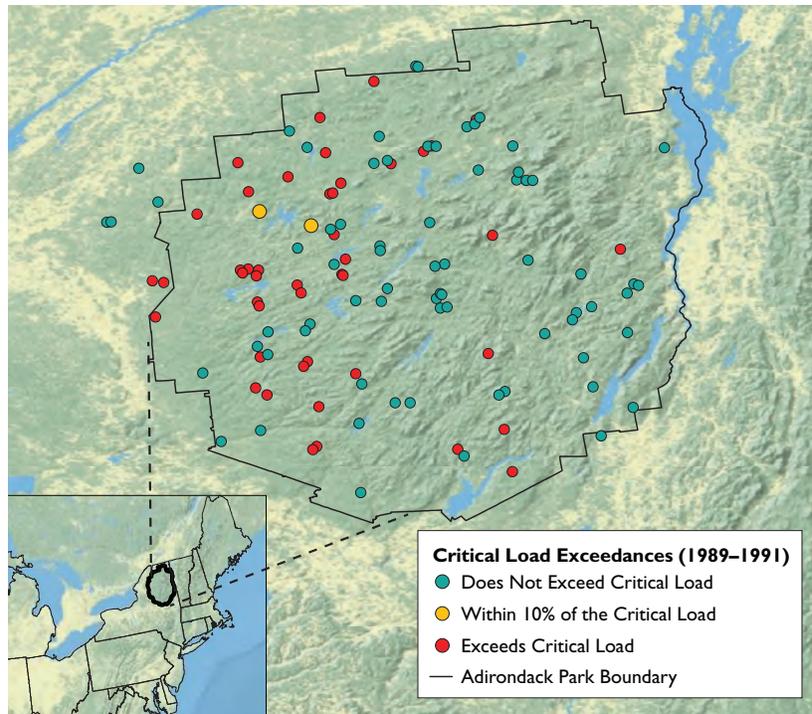
Figure 2-15. Critical load exceedances in Adirondack Mountain TIME lakes, 1989 to 1991 (No. lakes = 117) (prepared by U.S. EPA).
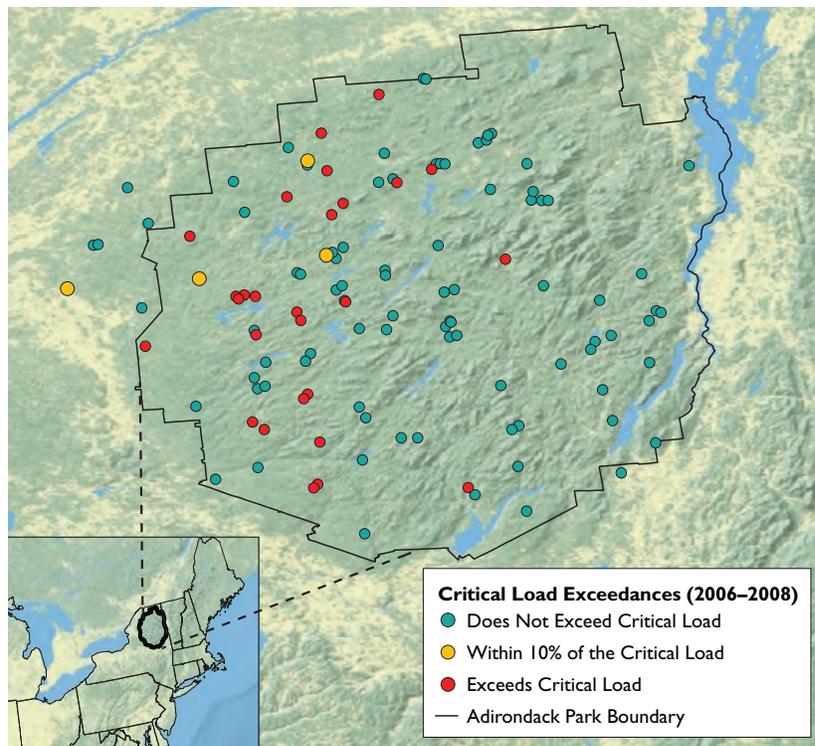
Figure 2-16. Critical load exceedances in Adirondack Mountain TIME lakes, 2006 to 2008 (No. lakes = 117) (prepared by U.S. EPA).

Central Appalachian Streams

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

On average, the calculated critical load of sulfur and nitrogen for streams in the central Appalachian region is 370 meq/m²/yr. For the period before ARP implementation (1989 to 1991), 41% of streams in the central Appalachian region received levels of combined sulfur and nitrogen deposition that exceeded the critical load, and therefore, could not be adequately neutralized by the environment (Figure 2-17). For the period from 2006 to 2008, 31% of the streams examined continued to receive acid deposition levels that exceeded the critical load (Figure 2-18). Thus, only an additional 10% of streams in the central Appalachian region are protected from ecological damage from acidic deposition, as compared to the period before ARP implementation. Although some streams in the central Appalachians likely have improved, approximately 35,000 km of streams still exceed their calculated critical load of sulfur and nitrogen based on the statistically representative population of lakes determined by EMAP.
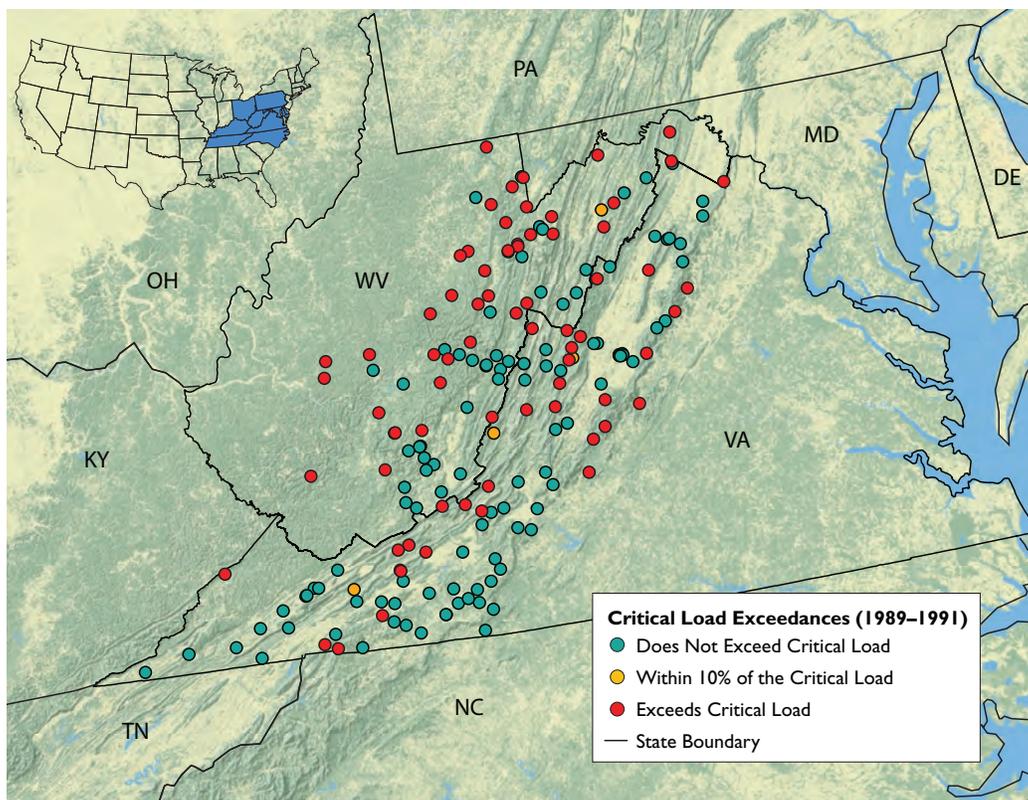
Figure 2-17. Critical load exceedances in central Appalachian streams, 1989 to 1991 (No. streams = 184) (prepared by U.S. EPA).

Visibility

In the atmosphere, SO_2 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 Measures

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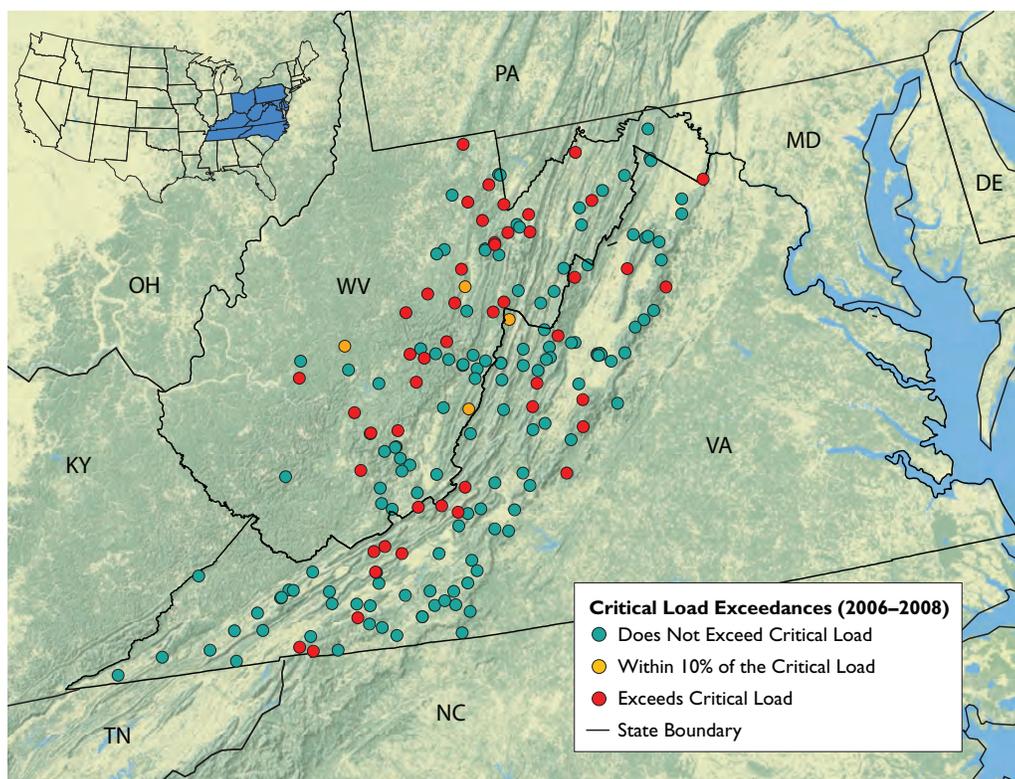
Figure 2-18. Critical load exceedances in central Appalachian streams, 2006 to 2008 (No. streams = 184) (prepared by U.S. EPA).

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 significant trends for either the best or worst 20% of visibility conditions, most have improving visibility. From 1999 through 2008, for the best visibility conditions, 45 sites show significant improvements and no sites show significant decline; for the worst visibility conditions, 10 sites show significant improvement and 5 sites show significant decline (all trends are at the $p < 0.05$ significance level).The Northeast has sites with improving visibility on both best and worst visibility days, principally due to regionally decreased sulfate particulate concentrations. Improvement for these

> As dv measurements increase, visibility decreases.

northeastern sites is about 5 dv for worst days and 3 dv for best days over the 10-year period. In the Pacific Northwest, a significant improving trend of about 3 dv to 5 dv per decade for worst haze periods is found at monitoring sites near and/or downwind from Seattle and Portland and may reflect changing impacts of these urban/ industrial areas. A better understanding of the causes of individual site trends requires additional analysis. Trends for each individual site shown on the map in Figures 2-19 and 2-20 can be viewed at http://views.cira.colostate.edu/web/Trends.
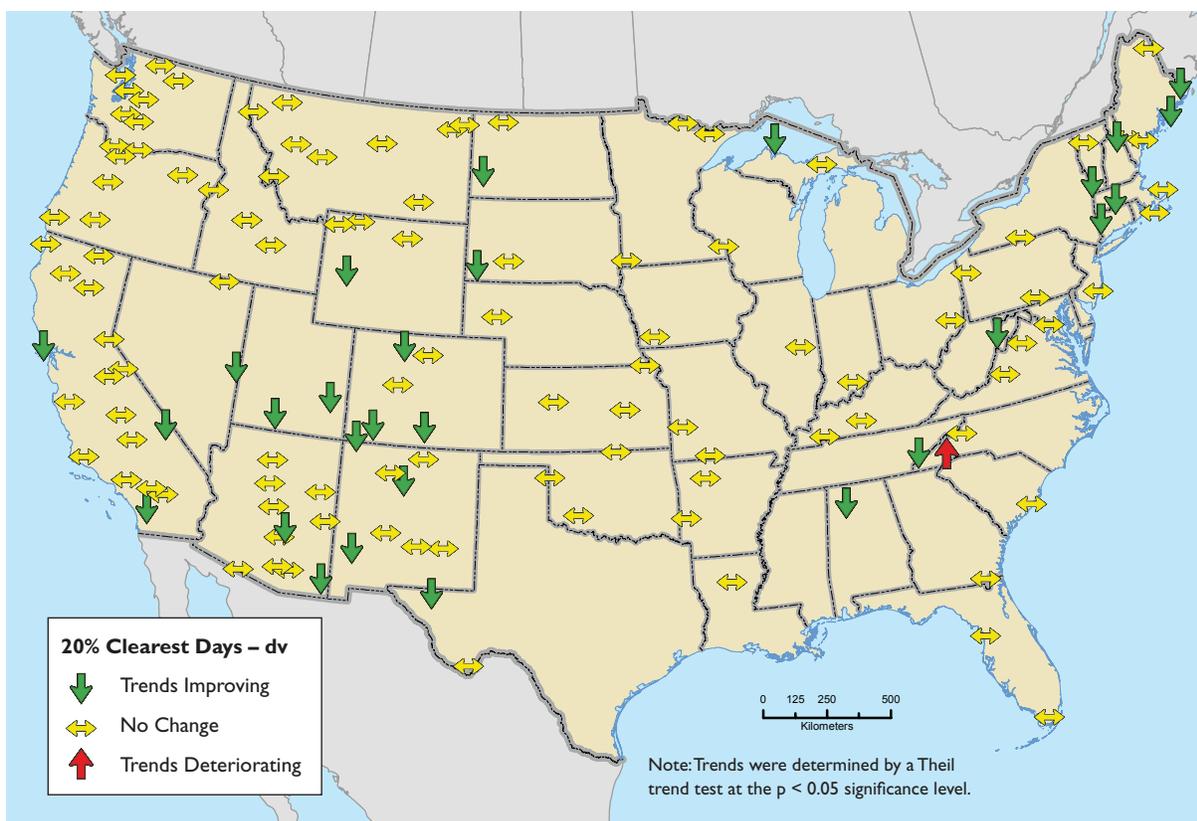
Figure 2-19. 1999 to 2008 trends for the change in visibility on the clearest days (best 20% of visibility) as measured at 140 IMPROVE monitoring stations (prepared by NPS).
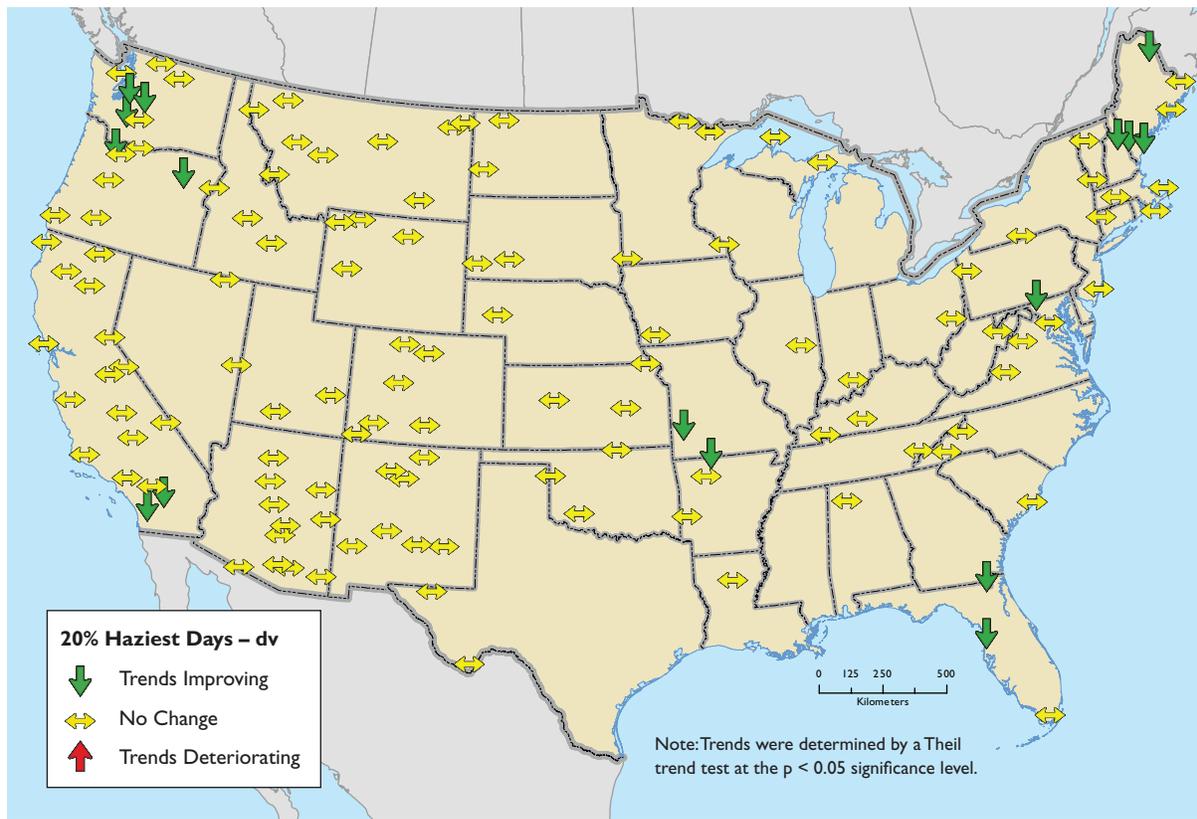
Figure 2-20. 1999 to 2008 trends for the change in visibility on the haziest days (worst 20% of visibility) as measured at 140 IMPROVE monitoring stations (prepared by NPS).

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

Chapter 2 focused on the environmental results of the ARP, presenting data from national monitoring networks on SO_2 and NO_x emissions, air quality, atmospheric deposition, surface 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

State of the Science at a Glance

- Scientists have observed delays in ecosystem recovery in the eastern United States, despite decreases in emissions and deposition over the past 30 years.

- A growing body of literature documents impacts of nitrogen deposition on western ecosystems.

- "Critical loads" are increasingly used by scientists as a tool for quantifying the sulfur and nitrogen deposition levels at which ecosystems are impacted in order to better inform air quality policies.

- Recent literature identifies linkages between a changing climate, an altered carbon cycle, and ecosystem response to acid deposition as an important emerging area of scientific investigation.

- Scientists identify multi-pollutant interactions, including reactions among SO_2, NO_x, ozone, and mercury, as an under-studied area of ecosystem impacts, which if better understood, would likely have implications for future air pollutant policies.

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 serves as an update to 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.

Effects of Atmospheric Deposition on Aquatic and Terrestrial Ecosystems

This section expands on the surface 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.

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 following sections 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.

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

Nitrate Concentration Trends

Trends in nitrate concentrations in headwater 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 as are those of sulfur. Nonetheless, over the long term, decreases in NO_x emissions from the power generation sector are expected to assist in efforts to reduce nitrate concentrations in surface waters.

> 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 sulfate concentrations have been the sharpest, generally showing decreases of about 2 to 3 µeq/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 monitoring program 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., 2006; 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).

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 hinge in part on the acidity of surface waters declining to a level at which minimal episodic acidification occurs because the survival of acid-sensitive aquatic species is dependent on both the magnitude and duration of episodic acidification (Baldigo and Murdoch, 1997). Although low-flow ANC values in the range of 30 to 50 µeq/L have been suggested to provide protection against acidification and biological harm during high-flow periods (Driscoll et al., 2001; Kahl et al., 2004), other studies have shown that episodic acidification occurs in streams with low-flow ANC values as high as 100 to 150 µeq/L (Lawrence, 2002). Some evaluations of ecosystem recovery (e.g., Kahl et al., 2004) provide percent-improvement values for various metrics of stream acidity and the extent of acidic waters in a region. These evaluations are based largely on low-flow conditions or chronic acidification scenarios and do not fully consider episodic acidification. One study in Maryland

The Laurel Prong fork of the Rapidan River in Virginia (photo courtesy of U.S. EPA).

evaluated changes in episodic stream acidification from the early 1990s to the late 1990s and found little change in the magnitude of acidification during storms. However, the study also found a shift towards a diminished role for sulfate in episodic acidification, suggesting a decreasing contribution of acid deposition to this phenomenon in parallel with declining sulfate deposition (Kline et al., 2007). Studies by Rice et al. (2006) and Lawrence et al. (2008) compared the percent of streams or watershed area that is chronically acidified with the percent that is episodically acidified in the Shenandoah National Park, VA, and the western Adirondack Mountains, NY, respectively. In each case, the total percentage of acidified streams or watershed area increased substantially when episodic acidification was considered (see Table 3-1).

One noteworthy exception to the generalizations above about geographic patterns of improvement in surface water chemistry is watersheds in the Southeast (e.g., the central Appalachian region of Virginia) where stream ANC values have generally not improved and sulfate concentrations have shown little change since the early 1990s (Kahl et al., 2004; Webb et al., 2004; U.S. EPA, 2009a; Chapter 2 of this report). The lack of chemical recovery in the streams of the central Appalachian region is believed to be due in part to the old and highly weathered condition of non-glaciated southern soils. These soils contain an abundance of adsorbed sulfate that can be slowly released as sulfuric acid deposition decreases, slowing ecosystem recovery (Webb et

al., 2004). In these soils, decades of sulfate release and decreased deposition levels may be necessary to shrink the pool of sulfate in the soils to the point where acidification of streams no longer occurs. Ecosystem recovery of acidified watersheds in the highly weathered soils of the Southeast will likely continue to lag behind that of the glaciated Northeast by several years to several decades.

Recent research has shown that decreases in base cations (e.g., calcium, magnesium, potassium, sodium) in soils may also be slowing the recovery of aquatic and terrestrial ecosystems even though they are receiving reduced loads of acid deposition (Bailey et al., 2005; Sullivan et al., 2006b; Johnson et al., 2008; Warby et al., 2009). The base cations calcium, magnesium, and potassium are important nutrients for the growth and health of forest vegetation and are readily accessed from the pool of cations held on the surfaces of soil particles. These base cations can be displaced from the surfaces of soil particles by hydrogen and aluminum ions and leached into local waters, effectively removing these nutrients from soils and lowering the levels that are available to the plants growing in the

> 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.

Table 3-1. Comparison of Chronically and Episodically Acidified Streams in a Watershed				
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 μeq/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 μeq/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 μeq/L during summer, and episodic acidification is defined by the same threshold value during snowmelt conditions.

soils. Acid deposition accelerates this as part of the acidification process, which initially leads to increases in base cation concentrations in surface waters, but eventually acidifies the waters as well. Base cation depletion refers to this accelerated loss of these cations from soils. Depletion of calcium is of particular concern because this cation is in high demand by sugar maple (St. Clair et al., 2008). Although a variety of mechanisms (e.g., maturation of forest vegetation) contribute to base cation depletion, acid deposition and the consequent mobilization of aluminum in soils have been identified as important drivers of depletion (Lawrence et al., 1995). Acid deposition and aluminum mobilization have contributed to dieback and decline of sugar maple (Bailey et al., 2004; Hallet et al., 2006; St. Clair et al., 2008) and to freezing injury in red spruce (DeHayes et al., 1999; Hawley et al., 2006). Recent work has found that declining growth rates can begin in apparently healthy sugar maple trees up to two decades before a strong decline in tree health becomes visually evident (Long et al., 2009). Other studies have found linkages between acid deposition, soil calcium availability, and the reproductive success of bird species (Graveland, 1998; Hames et al., 2002).

The ANC value of a waterbody reflects the sum of the base cation concentrations (calcium, magnesium, potassium, sodium) minus the sum of the acid anion concentrations (primarily sulfate and nitrate). Depletion of base cations from soils over time will provide lower concentrations of these base cations to waters and result in a lower ANC value than would otherwise be predicted based on the acid anion concentrations alone. In this way, losses of exchangeable soil base cations can slow the recovery of aquatic ecosystems (Likens et al., 1996; Lawrence et al., 1999). The scientific consensus is that base cation depletion is slowing the rate of chemical recovery of lakes and streams and, therefore, also the rate of biological recovery in many of the sensitive aquatic ecosystems targeted by Title IV of the CAAA (Driscoll et al., 2001). Re-supply of these base cations generally occurs when chemical breakdown (i.e., weathering) of minerals in bedrock begins to release cations to soils and waters at a rate that is faster than the

Sugar maple showing brilliant fall colors (photo courtesy of Jacquie Tinkler, NPS).

leaching loss caused by acid deposition. Acid deposition must likely decrease below a critical level that will vary among ecosystem locations before large increases in the ANC of acidified waters can occur. The role of base cation depletion is even greater when considering episodic acidification because soil chemistry plays an even greater role in stream chemistry during high-flow conditions than during low-flow conditions (Lawrence, 2002).

Important complications to this simplified conceptualization of the link between acid deposition and base cation depletion arise from the uptake of base cations by vegetation and from other factors. For example, the cation balance is also controlled in part by forest age, health, and root-mining ability, as well as harvesting practices (Blum et al., 2002; Hamburg et al., 2003; Grigal and

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

Ohmann, 2005). The exact rate of acid deposition that will allow recovery of base cation–depleted soils is not well known because the rate of mineral weathering is not well quantified in most soils, can be highly variable among locations, and is difficult to measure in the field (Bailey et al., 2003). Current models (such as the Model of Acidification of Groundwater in Catchments [MAGIC]) that are commonly applied to make quantitative predictions about acidification and critical loads in sensitive ecosystems can be, and have been, used to examine the role of base-cation depletion; however, they have not yet been applied widely for this purpose in the United States. Base-cation depletion is a high-priority area for future acidification research and is of great relevance to the development of future air quality policies designed to protect ecosystems.

Recovery of Biota

Aquatic Recovery

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

Acidification results in changes not only to the constituent or habitat that may be targeted as a threshold indicator but also to a wide variety of other chemical constituents and physical habitats. As a result, a complex set of changes in the ecological community may occur, including a shift in the members of the community from acid-sensitive to acid-tolerant species. Acid-

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.

tolerant communities that develop in response to acidification may then resist change as conditions become less acidic, limiting the re-establishment of the acid-sensitive species that were originally impacted by acidification (Frost et al., 2006). Other factors may also slow this process of biological recovery, including physical limitations on dispersal, the proximity of species that may have historically resided in the water prior to acidification (Yan et al., 2003; Langford et al., 2009), and calcium concentrations that tend to be lower relative to ANC during ecosystem recovery than during acidification (Jeziorski et al., 2008; Cairns and Yan, 2009). These factors act to delay biological recovery relative to chemical recovery (Snucins, 2003), and some scientists have suggested that active ecosystem restoration (e.g., reintroduction of previously lost species) may be needed to meet the goal of a "pre-acidification" biological community (Snucins and Gunn, 2003; Binks et al., 2005).

Recreational fishing in South Rolly Lake in Alaska. (photo courtesy U.S. Fish and Wildlife Service).

When a different pathway of change is followed during recovery than was followed during acidification, this behavior is called "hysteresis." A conceptual representation of hypothetical acidification and recovery patterns in a waterbody is presented in Figure 3-1. Point 1 represents the pre-acidification status of water quality and the biological community. Point 2 shows the status when conditions were most acidic, and Point 3 represents current conditions where hysteretic behavior is exhibited because the rate of recovery does not match the rate of acidification. At this point, the water quality has shown some recovery, but a pre-acidification biological community has not yet been achieved. The two dashed lines represent possible future recovery trajectories for the biological community as future acid deposition levels continue to decrease. These recovery patterns may or may not be linear as shown, and some evidence suggests that "threshold" behavior marked by a sharp change in slope with time may occur in biological communities recovering from acidification (Monteith et al., 2005). One trajectory ends at Point 4, where water quality has fully recovered, but the biological community has only recovered slightly. The other trajectory returns to Point 1, representing a full recovery of water quality and the biological community. Some studies

indicate that even with full recovery of water quality (less acidic stream on the right side of the figure), the biological community will likely differ from what existed prior to acidification (Frost et al., 2006; Cairns and Yan, 2009), suggesting that recovery may ultimately be closer to Point 4 than Point 1.

Despite the concerns raised about limitations on recovery from acidification due to competition, dispersal, and other ecological factors that must be considered in addition to chemical thresholds, several studies in Europe and Canada have demonstrated biological recovery in aquatic communities that have become less acidic in response to decreases in acid deposition (Tipping et al., 2002; Findlay, 2003; Halvorsen et al., 2003). Additionally, studies in which lime has been added to waterbodies or soils to hasten recovery from acidification have shown trends towards a pre-acidification aquatic community (Raddum and Fjellheim, 2003), but the success of these efforts has sometimes required active restoration of some species (Driscoll et al., 1996; Clair and Hindar, 2005).

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.
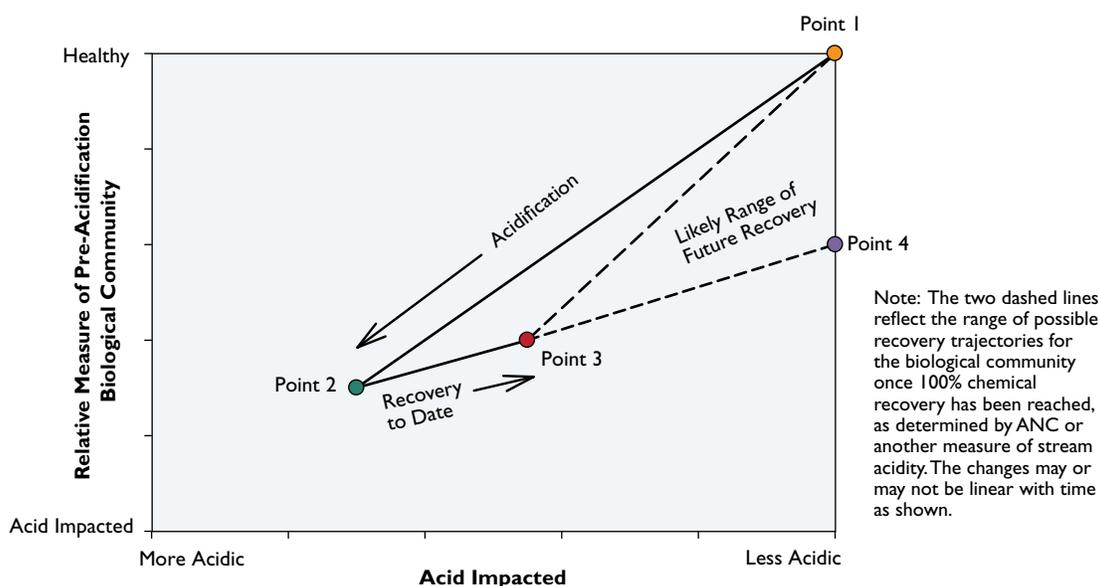
Figure 3-1. Conceptual figure showing hypothetical acidification and recovery patterns in a waterbody that has been acidified by atmospheric deposition (prepared by USGS).

monitoring networks regularly provide such evaluations and because most available data originate from ad-hoc studies. Nonetheless, a few studies have evaluated aquatic communities by sampling waters over a period when acidity was declining, and the general conclusion is that there is little evidence to date of widespread recovery in these communities (Simonin et al., 2005; Burns et al., 2008a; Warren et al., 2008).

Terrestrial Recovery

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

As described previously, several studies have shown the link between acid deposition and losses of base cations from soil. Many of these studies have included data from re-sampled soil locations, generally documenting a lack of improvement or continued degradation of soil base cation status in the Adirondack Mountains and Allegheny Plateau of Pennsylvania (Bailey et al., 2005; Sullivan et al., 2006b; Johnson et al., 2008; Warby et al., 2009). Furthermore, a clear link between the base cation status of soils and the health of sugar maple has been established in recent years (Bailey et al., 2004; Hallet et al., 2006; Long et al., 2009). The role of acidification in red spruce freezing injury was established by the early 1990s (McLaughlin et al., 1993). Recent data have largely confirmed the findings of earlier studies on red spruce (Hawley et al., 2006) and have also found potential effects of acid deposition on other species of forest vegetation (Jenkins et al., 2007; Zaccherio and Finzi, 2007). Currently, no published studies can document an improvement in the status of terrestrial ecosystems

> 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.

in the eastern United States relative to acidification effects due to decreased emissions and deposition. This conclusion likely results from three factors: (1) few studies have evaluated terrestrial ecosystem health relative to acidification effects over time, (2) soils in the most acid-sensitive regions continue to acidify, and (3) the potential for lags in response in long-lived species such as forest trees.

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

As shown in Chapter 2, the Rocky Mountain region of Colorado and adjoining states receives atmospheric deposition levels of sulfur and nitrogen that are generally less than half the deposition levels measured in mountainous parts of the eastern United States. Yet, the Colorado Rockies landscape is generally more sensitive to nutrient over-enrichment from atmospheric nitrogen deposition than landscapes in the eastern United

Central Appalachian basic boulderfield forest, Shenandoah National Park (photo courtesy of Gary P. Fleming, Virginia Department of Conservation and Recreation).

States. This sensitivity is due to the steep slopes, thin or non-existent soil cover, short growing season, and less vegetation per unit landscape area (Clow and Sueker, 2000). Several studies have measured ANC values near or below zero during snowmelt in headwater streams, small headwater lakes, and ephemeral pools, and downward trends in ANC values were evident in some waters through the 1990s (Caine, 1995; Williams and Tonnessen, 2000; Campbell et al., 2004). However, the extent of waters that become episodically acidified in the Colorado Rockies is generally less than in the eastern United States, and no deleterious effects of acidification on fish have been documented.

> 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.

Atmospheric deposition of nitrogen has been of interest in the Rocky Mountains because both aquatic and terrestrial ecosystems in this region were historically nitrogen limited and have shown species shifts and growth responses at nitrogen deposition levels as low as 1.5 kg/ha/yr (Wolfe et al., 2003; Baron, 2006). Several factors distinguish nitrogen deposition patterns in the Colorado Rockies from those in the eastern United States. First, the character of nitrogen deposition is different in the Colorado Rockies than in the eastern United States. Ammonia nitrogen (i.e., NH_3-N) deposition, which generates greater acidity than does nitrate, generally exceeds that of nitrate nitrogen (i.e., NO_3-N), especially at sites east of the Continental Divide in the Front Range of the Rocky Mountains (Heuer et al., 2000; Burns, 2003; Beem et al., 2010). This pattern is believed to result from the deposition of emissions carried by upslope winds to the Front Range from agricultural sources that are located to the east of the mountains (Baron et al., 2004). As a result, atmospheric nitrogen deposition at many monitoring sites in the Colorado Rockies is not decreasing with decreases in power plant emissions in the western United States. Instead nitrogen loads are still
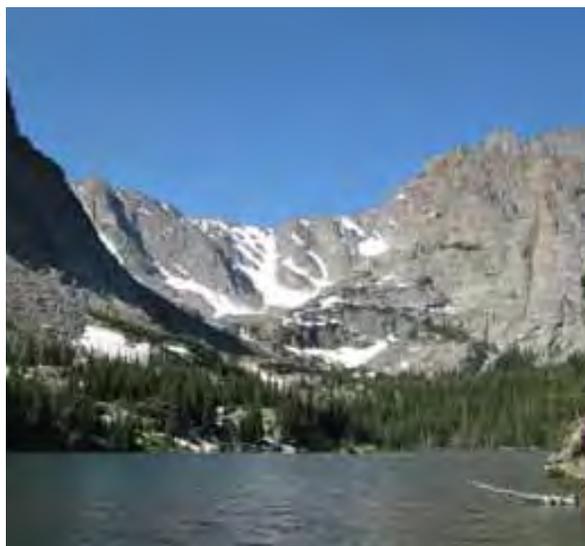
Loch Vale in Rocky Mountain National Park receives among the highest loads of atmospheric nitrogen deposition in the Front Range of the Colorado Rockies (photo courtesy USGS).

increasing in some western locations. This pattern is especially driven by a tendency towards increasing trends in ammonia deposition (Burns, 2003).

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

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

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

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

Much of the Grand Teton National Park is managed as a Class 1 wilderness area (photo courtesy of NPS).

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

In many areas of the far western United States (lands to the west of the Rocky Mountains, especially the West Coast states), nitrogen loads to terrestrial systems have historically been low compared with loads to terrestrial systems in the eastern United States. The native plant communities (e.g., coastal sage scrub, desert scrub, annual grasslands) are adapted to thrive in low-nitrogen environments. When nitrogen loads to these habitats increase, the native plant and soil communities have difficulty competing with invasive species that are better adapted to more nitrogen-rich environments. In California, nitrogen deposition levels as low as 5–8 kg/ha/yr have major effects on epiphytic lichens (i.e., lichens that grow on trees and shrubs) and contribute to changes in native plant communities by enhancing invasion by exotic annual grasses (Fenn et al., 2003a; Weiss, 2006). In coastal sage scrub, desert scrub, and annual grasslands in California, nitrogen deposition favors non-native grass invasions, which can lead to altered plant communities. In southern California chaparral (Egerton-Warburton et al., 2001) and coastal sage scrub ecosystems (Egerton-Warburton and Allen, 2000), the diversity of the mycorrhizal fungi communities that grow in association with roots also is negatively impacted by nitrogen deposition. Shifts in these plant communities are of additional concern because the invasive vegetation that results

from increased nitrogen loads also may increase the risk of wildfires due to increased fuel availability. In the desert scrub ecosystems of Joshua Tree National Park, invasive grasses can provide sufficient fuel load to carry fire, particularly in a wet year. This increased fuel load puts the Park's native vegetation at risk, including its namesake, the Joshua tree. Fire occurrence in the desert is a rare event in the absence of the invasive grasses (Allen et al., 2009).

Increased nitrogen deposition also can impact forests in the far western United States. Nitrogen deposition effects, in combination with the physiological effects of ozone, decrease the amount of roots and increase susceptibility of ponderosa pine to bark beetle attack and mortality (Jones et al., 2004). Forest sustainability is thus threatened, and fire risk is increased (Grulke et al., 2009). Studies have been conducted to assess various methods of reducing nitrogen saturation symptoms. Using fire as a management technique to reduce excess nitrogen was found to be ineffective in chaparral ecosystems, presumably because of the limited capacity of fire to reduce the large nitrogen stores in the mineral soil of these ecosystems (Meixner et al., 2006). Similarly, in forests affected by excess nitrogen, a combination of periodic nitrogen release by fire and significant reductions in nitrogen deposition are needed to mitigate the effects of nitrogen accumulated from long-term nitrogen deposition (Gimeno et al., 2009).

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

In the streams and lakes of the far western United States, increased nitrogen deposition can alter lake and stream chemistry. At moderate nitrogen

Desert scrub ecosystem in Joshua Tree National Park may be impacted by nitrogen deposition (photo courtesy of USGS).

deposition levels of 10–14 kg/ha/yr for chaparral catchments and of 17 kg/ha/yr for forested catchments in the southwestern Sierra Nevada and in the Transverse Ranges near Los Angeles, high concentrations of nitrate are found in stream water. In chaparral, forested, and high-elevation wilderness catchments in the Los Angeles air basin, up to 40% of the nitrate exported in stream water was directly deposited from the atmosphere and transported through the system without biological assimilation (Michalski et al., 2004), clearly indicating that the source of this excess nitrogen was air pollutant deposition in these watersheds.

High-elevation watersheds in the Sierra Nevada appear to export nitrate during early snowmelt, even with relatively low atmospheric nitrogen deposition similar to the high tendency to export nitrate discussed above for the Colorado Rockies. Although the role of nitrogen deposition on nitrate leaching in the high-elevation Sierra Nevada is not entirely clear, nitrogen deposition does not appear to have as severe an impact on lake and stream water chemistry, particularly in Sierra Nevada catchments, where soils tend to be thicker than those of the Colorado Front Range. Differing responses between the two regions may be due to higher

nitrogen deposition in the Colorado Rockies or to environmental differences. For example, the greater extent of soil freezing in the Colorado Rockies may reduce the nitrogen retention capacity of these watersheds (Sickman et al., 2002). However, watersheds in the Sierra Nevada are expected to be sensitive to increases in nitrogen deposition in the seasonal snowpack. This expectation is based on the large pulses of nitrate that are flushed to surface waters during snowmelt-driven spring runoff (Sickman et al., 2003); a short growing season; the limited soil and vegetation cover; and the limited biological nitrogen demand during the early spring runoff period (Fenn et al., 2003b).

Effects of Atmospheric Nitrogen Deposition on Coastal Estuaries

Eutrophication and associated hypoxia is widespread in estuaries of the United States (Figure 3-2),including Long Island Sound, the Chesapeake Bay, and the Gulf of Mexico near the Mississippi River Delta (Diaz, 2001; Kemp et al., 2005). Nitrogen and phosphorus are the nutrients that commonly limit algal growth in coastal ecosystems. The role of nitrogen in estuarine eutrophication and hypoxia is often attributed to a combination of the delivery of excess nitrogen by rivers and the direct atmospheric deposition of nitrogen onto the water surface. These same mechanisms also deliver nitrogen to the open ocean, and atmospheric nitrogen deposition has been shown to affect the growth of marine plankton (Krishnamurthy et al., 2010). The potential sources of nitrogen to estuaries include runoff from fertilizer and animal waste applied to agricultural landscapes, human waste in urban/suburban landscapes, and atmospheric nitrogen deposition. The relative role of atmospheric nitrogen deposition to total nitrogen loads to coastal estuaries differs widely, from about 15% to 25% in the Mississippi Delta region of the Gulf of Mexico, to 20% to 50% in Chesapeake Bay and Tampa Bay, and as much as 60% to 80% in some coastal river estuaries of northern New England (McIsaac et al., 2001; Boyer et al., 2002; Poor et al., 2006; Whitall et al., 2007; Alexander et al., 2008; Howarth, 2008). The relative contributions of nitrate and ammonia as sources of atmospheric nitrogen to estuaries also vary widely across coastal regions of

Figure 3-2. Map of major estuaries in the United States (prepared by U.S. EPA).

the United States. Nitrate is generally the dominant form of nitrogen in atmospheric deposition across the United States; however, ammonia can be the dominant atmospheric species of nitrogen in parts of estuarine watersheds, particularly those with abundant agricultural land (Lawrence et al., 2000; Whitall et al., 2003). The most recently available data indicate that the majority of U.S. estuaries show signs of eutrophication (Bricker et al., 2008), and the number of impacted estuaries in the United States and globally has increased greatly since the 1960s (Diaz and Rosenberg, 2008). Although most U.S. coastal waters have not shown changes in nutrient loads or trophic status since the 1990s (Alexander and Smith, 2006), about 20% to 25% of these coastal waters show improved trophic status in recent decades (Alexander and Smith, 2006; Bricker et al., 2008), most notably Tampa Bay. The role of decreased loads of atmospheric nitrogen deposition to observed decreases in estuarine eutrophication is likely only minor, but is not well quantified (Greening and Janicki, 2006).

The contribution of headwater streams to the total nitrogen loads in rivers can be substantial. For example, small streams account for more than two-thirds of the water volume and approximately two-thirds of the nitrogen delivered to large rivers in the Northeast (Alexander et al., 2007). Despite a higher removal rate (denitrification) of nitrate from headwater reaches than from large river channels during transport (Alexander et al., 2008), the colder temperatures at the higher elevations of the headwaters act to slow this rate of loss (Schaefer and Alber, 2007). Therefore, a substantial

amount of the nitrogen deposited on upland forested areas from the atmosphere may later be exported to larger rivers and estuaries. These studies emphasize the connection of upland ecosystems to coastal ecosystems and indicate that the effects of atmospheric nitrogen deposition extend beyond acidification in sensitive upland landscapes.

The issue of ocean acidification has emerged in recent years, with the observation that rising levels of carbon dioxide (CO_2) in the atmosphere are causing decreases in ocean pH and alkalinity, with numerous implications for the health of marine ecosystems (Caldeira and Wickett, 2003). Some recent studies have explored the role of atmospheric nitrogen and sulfur deposition on ocean acidification and concluded that acid deposition contributed no more than a few percent to ocean acidification trends in recent years (Bates and Peters, 2007). However, the contribution of acid deposition to marine pH declines is likely greater in coastal regions closer to continents (Doney et al., 2007). Further scrutiny of the role of acid deposition on the pH and alkalinity of coastal ecosystems may be needed in the future given the serious consequences of ocean acidification that are currently forecast.

Eutrophication and Hypoxia

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, they are decomposed by bacteria that use the dissolved oxygen in the water. "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, crabs, shrimp, and other organisms. Benthic (bottom-dwelling) organisms such as oysters that are unable to move to more oxygenated waters are particularly susceptible.

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

Critical Loads

Critical loads, which were introduced in Chapter 2, can be viewed as providing a synthesis of scientific knowledge about thresholds of harm to or alteration of ecosystems that can inform the development, implementation, and assessment of air quality policy and the management of sensitive ecosystems. Critical loads have been widely used for large-scale applications and policy purposes in Europe and Canada since the 1980s (Jeffries and Ouimet, 2005; Spranger et al., 2008), but historically have not been widely used for these purposes in the United States (Porter and Johnson, 2007). For example, critical load maps for soils and waters are produced by several European countries that are parties to the *Convention on Long-Range Transboundary Air Pollution*. Although a signatory to this Convention, the United States did not commit to the provisions regarding critical loads contained in the Convention (see recent discussion in Spranger et al., 2008). Academic critical load studies in the United States also date to the 1980s (Henriksen and Brakke, 1988), and many studies continue to be published (Baron, 2006; Fenn et al., 2008; Sullivan et al., 2008); however, most past work has been of limited geographic scope. Recently published studies provide examples of larger-scale assessments of critical loads in the United States, including nationwide critical load maps for forest soils (McNulty et al., 2007) and a review of published studies throughout the United States for empirical critical loads of nitrogen (Pardo et al., 2011).

Critical loads can be calculated for any species of concern or for a sensitive biological community within an aquatic or terrestrial ecosystem if a threshold of atmospheric deposition can be identified below which negative biological effects on the species or alteration of the community are not expected to occur. Because various species in an ecosystem may have differing sensitivities to air pollutant loads, a large number of species-specific or community-specific critical loads are likely to exist for a given location (Sullivan et al., 2008).

Critical loads are often linked to thresholds at which the organism experiences a certain biological effect, which may range from stunted growth to reduced reproductive levels to increased mortality. Critical loads are usually calculated in reference to a chemical quantity believed to provide an index of acidification or nutrient over-enrichment. Potential indices include ANC levels or nitrate concentrations in a waterbody, or the base cation saturation level of a soil. Multiple critical loads and ecological effects end points provide policy makers or land managers with multiple options to determine the level of protection that is warranted based on the species or communities of greatest concern at a given location. This is part of the process of establishing a target load, which may differ from the strictly science-based critical load. The target load reflects inclusion of economic, social, cultural, or other considerations to establish the protection level of the ecosystem in question, as well as the load that is deemed "achievable" by managers or regulators (Porter et al., 2005; Porter and Johnson, 2007).

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

Despite the lack of a long history of applying critical loads to inform air pollutant policies in the United States, interest in the use of critical loads in the United States has increased in recent years with the advent of a Critical Load Science Committee within NADP (NADP, 2009), several recent workshops and meetings on this topic, and several publications exploring greater use of critical loads as a policy-relevant environmental assessment tool (Porter et al., 2005; Fisher et al., 2007; Burns et al., 2008b; Glavich and Geiser, 2008). Recently published studies have generally found that for many regions well known for their sensitivity to either acidification or excess nitrogen deposition (e.g., the Adirondack Mountains of New York, the central Appalachian region of Virginia, the Front Range of Colorado), the current levels of atmospheric deposition generally exceed the critical loads for many species of concern (Baron, 2006; McNulty et al., 2007; Sullivan et al., 2008). Case studies exploring the application of the critical load approach for the Adirondack Mountains and the central Appalachian region are also included in Chapter 2 and Chapter 4 of this report. Finally, consideration of two key factors is warranted when using scientific information on critical loads to inform public policy implementation: (1) critical loads can have high uncertainty that stems from model assumptions and data, as well as uncertainty in the threshold itself (Skeffington, 2006; Li and McNulty, 2007),

and (2) other variables such as forest harvesting and climate change will affect critical loads in addition to atmospheric deposition (Watmough et al., 2003). When critical load calculations are being used to inform air pollutant policies, scientific uncertainty must be properly considered, along with the role of other mitigating variables.

Atmospheric Deposition and Ecosystem Services

In addition to the health benefits of the ARP discussed in Chapter 1 of this report, benefits to ecosystem services also might be expected, and these benefits can be evaluated by considering the varied material and intangible goods and services by which ecosystems support human well-being (PCAST, 2011). Ecosystem services can be defined as the sum total of benefits that humans receive from the resources and processes of natural ecosystems (Corvalan et al., 2005). These benefits range from extractive services or provisioning (e.g., wood products, minerals, water) to regulating services (e.g., climate regulation, nutrient cycling) to cultural services (e.g., outdoor recreation). The ability of ecosystems to provide these services over the long term is impacted by disturbances (Beier et al., 2008), including atmospheric deposition. The response of ecosystems to disturbances, such as acid deposition, depends on the resiliency of the ecosystem in question. Over the past two decades, researchers have begun to establish the value of these various ecosystem services (Costanza et al., 1997) and to develop approaches for valuating both the direct use-related benefits, as well as the non-use benefits of ecosystems; most studies indicate these non-use benefits exceed those of the more traditional market-based benefits derived from resource extraction (Walsh et al., 1990).

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—These models were simple and used 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 (see the figure on the following page). The dynamic models listed 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.

- Integrated Lake-Watershed Acidification Study (ILWAS) model (Gherini et al., 1985)

- Birkenes model (Christophersen et al., 1982)

- Model of Acidification of Groundwater in Catchments (MAGIC) (Cosby et al., 1985c)

- Photosynthetic evapotranspiration-biogeochemical (PnET-BGC) (Gbondo-Tugbawa et al., 2001).

All of the dynamic models can be combined with atmospheric models, such as the Community Multiscale Air Quality model (CMAQ), that estimate atmospheric deposition to landscapes from present emission levels or future emission scenarios (Schwede et al., 2009).

(continued)

Models Used to Investigate the Ecosystem Effects of Acid Deposition (continued)
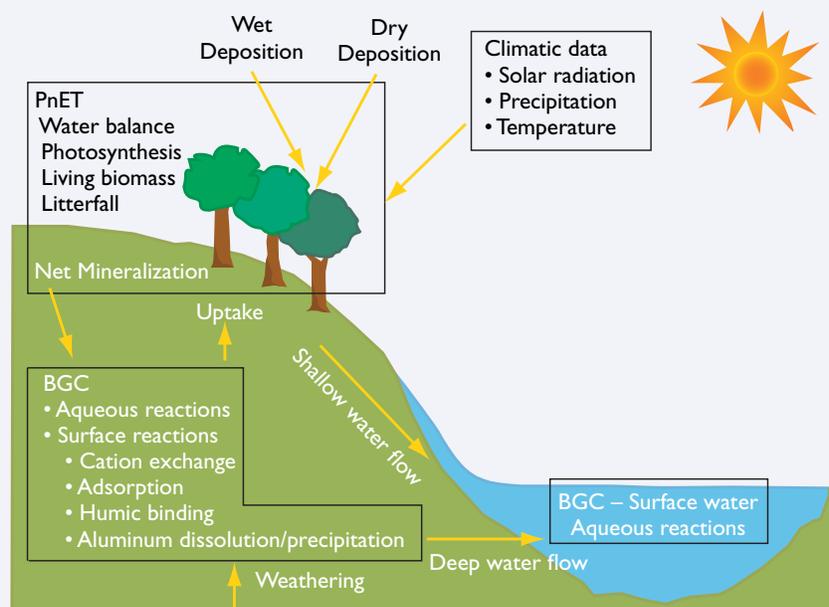
Conceptual diagram of the biogeochemical processes and chemical stores of the PnET-BGC model (figure courtesy of Charles Driscoll, Syracuse University).

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

- DayCent/PHREEQC (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). Steady-state models, such as those listed below, 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, 2005a; Chen and Driscoll, 2005b; 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.

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

The greatest challenge in developing specific data on the economic benefits of changes in acid deposition due to emission reductions lies with the availability of comprehensive scientific evidence that defines the extent and magnitude of the adverse effects that can be directly attributed to acid deposition from among multiple ecosystem stressors (Krupnik and Siikimaki, 2008). In one notable study, Banzhaf et al. (2006) estimated ecological benefits of a policy that would substantially decrease adverse effects of acid deposition in the Adirondack Park of New

Forest harvesting is an example of an ecosystem service that provides monetary benefit to land owners (photo courtesy of Douglas Burns, USGS).

York. These benefits are in the range of about $300 million to $1.1 billion annually for the Adirondack Park only and do not consider ecological benefits in other regions of the United States from these deposition reductions. As described in Evans et al. (2008), researchers are applying economic methods to show the total value people place on ecological improvements to important resources, such as the Adirondack Park, expected from further reductions in acid deposition. However, as discussed in Chapter 1 of this report, no studies have comprehensively evaluated the monetary benefits to ecosystem services of the ARP across all ecosystem types that would be sufficient to make comparisons to overall health benefits and costs of the ARP. Significant future analytical work and basic ecological and economic research is needed to build a sufficient base of knowledge and data to support an adequate assessment of ecological benefits.

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

The Earth has warmed by an average of 0.74° C between 1906 and 2005, and there is a high likelihood that recent warming over the past several decades has been driven in large part by increases in atmospheric concentrations of CO_2 and other greenhouse gases (IPCC, 2007). Under a range of likely future scenarios of greenhouse gas emissions and a range of climate model predictions, the Earth is likely to warm by another 1.4°–5.8° C during the twenty-first century (IPCC, 2007).

Ongoing and future predicted changes in the climate and the carbon cycle have numerous implications for the response of ecosystems to atmospheric deposition of sulfur and nitrogen. In addition to warming air temperatures, a wide array of other environmental variables are predicted to change with the climate, including water availability and the rates of many nutrient cycling processes. These additional environmental changes and their effects are likely to vary by region (Boisvenue and Running, 2006). In short, 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 (McLaughlin and Percy, 1999). For example, although the effects of an average increase in air temperature are likely to induce widespread changes in ecosystems, an increase in the magnitude and frequency of a wide variety of climatic events (e.g., floods, droughts, fires) also is expected. Some have suggested that these events are likely to affect ecosystems to a greater extent than long-term average changes in temperature (Dale et al., 2001; Jentsch et al., 2007). Despite these complexities, a brief assessment of likely interactions between ecosystems, acid deposition, and climate change is warranted in this current assessment given the potential magnitude and multi-faceted impacts of the projected changes. This section focuses on ecosystem effects and does not discuss the interactions between climate change and greenhouse gases or how these interactions may affect the transport, atmospheric interactions, and atmospheric deposition of sulfur and nitrogen. These atmospheric changes have been discussed elsewhere (Tagaris et al., 2008) and are beyond the scope of this assessment.

> 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.

Implications of Climatically Driven Changes in the Species Composition of Ecosystems

Perhaps the greatest impact of climate change is likely to be wholesale changes predicted for the biological communities found in many ecosystems sensitive to acid deposition effects, particularly those at high elevations (Ibanez et al., 2007). For example, a migration northward and to higher elevations is predicted for many tree species under most current climate change forecasts as they are replaced by species better adapted to a warmer climate (McKenney et al., 2007; Lenoir

et al., 2008). Evidence indicates that migration of lower elevation forests to higher elevations is already occurring in Vermont, suggesting this shift is ongoing in mountainous areas that have warmed in recent decades (Beckage et al., 2008). Similar cold-to-warm water species shifts are also predicted for aquatic ecosystems (Mohseni et al., 2003; Durance and Ormerod, 2007). The rate at which these ecosystem changes will occur is not well known and is likely to reflect interactions with other contemporaneous changes, such as those in water availability, light, insect defoliation, land use, and air pollutant deposition. Overall, the evidence suggests that many species may reach an out-of-equilibrium state with respect to a future warmer climate (Higgins and Harte, 2006; Mohan et al., 2009), and this state may result in increased sensitivity of many terrestrial and aquatic ecosystems to the effects of other disturbances, such as sulfur and nitrogen deposition. A European study (Alcamo et al., 2002) concluded that climate change is likely to have little effect on the sensitivity of forested ecosystems to air pollutant deposition. However, the weight of evidence from other studies indicates that, despite some uncertainty in the effects of climate change on ecosystems (Aber et al., 2001), ongoing and likely future climate change will provide an added stressor to many ecosystems in the United States. Furthermore, several studies indicate that the combination of atmospheric deposition of sulfur and nitrogen with high ozone concentrations will act as additional ecosystem stressors in many regions, such as the eastern United States (McLaughlin and Percy, 1999; Mohan et al., 2009). The increases in extreme events, such as droughts and fires, which are predicted with future climate change, will likely act as additional stressors to many of these ecosystems, with greater effects expected in the western United States (Dale et al., 2001; Knapp et al., 2008).

Climate Warming Will Affect Many Temperature-Sensitive Biogeochemical Processes in Ecosystems

Many biogeochemical processes affect the cycling of atmospherically deposited sulfur and nitrogen in ecosystems and play a pivotal role in the

acidification of ecosystems, including mineral weathering, nitrification, and sulfur and nitrogen mineralization. The rates of nearly all of these processes are likely to be affected by changes in climate. For example, the rates of mineral weathering and nitrification increase with increasing temperature if all other factors are held constant (White et al., 1999; Dalias et al., 2002). Studies have shown widely varying biogeochemical responses to past and predicted climate warming in the United States, and these varying responses reflect interactions with precipitation, snow and ice, and seasonal variations. For example, Campbell et al. (2009) predict higher nitrate concentrations in soil water and streams at Hubbard Brook, NH, due to warming-induced increases in nitrogen mineralization and nitrification, yet they also predict decreased rates of mineral weathering due to decreases in soil moisture. At the Rocky Mountain National Park in the western United States, stream nitrate concentrations have been increasing in recent years due to the melting of glaciers and snow fields by increased warming in summer through fall and the leaching of nitrate that occurs when the underlying soils are exposed to microbial processes (Baron et al., 2009). These

Climate change studies in Loch Vale have documented increasing air temperatures, earlier snowmelt, and earlier runoff. These changes may have important impacts on water availability, water quality, and ecosystem function (photo courtesy of USGS).

results highlight the sensitivity of long-term stream water quality to changes in snowfall, snowmelt, and the mass of glaciers in high-elevation watersheds of the western United States. Data from the European Alps show a recent trend towards increasing solute concentrations in high-elevation lakes, consistent with accelerated mineral weathering rates driven by warmer air temperatures and less-extensive snow cover that lasts for a shorter duration (Mosello et al., 2002; Rogora et al., 2003). Increased mineral weathering rates would likely result in a greater ability of soils to neutralize acid deposition and might accelerate recovery in sensitive ecosystems when accompanied by decreasing atmospheric deposition rates of sulfur and nitrogen. However, a recent study in the Colorado Rockies showed that regions underlain by sulfide minerals such as pyrite may experience greater acidification with warmer air temperatures due to increased mineral weathering rates and the subsequent formation of sulfuric acid (Mast et al., 2010).

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

Implications of Changes in Carbon Dioxide Concentrations in the Atmosphere

One of the fundamental factors that affects the rates of photosynthesis and terrestrial ecosystem growth is the availability of CO_2. Global atmospheric CO_2 concentrations have increased by nearly 40% since the mid-eighteenth century and are expected to double during the twenty-first century (IPCC, 2007). The ecosystem "CO_2 fertilization effect" has been much discussed in the literature (Tans et al., 1990; Nowak et al., 2004). Indeed, measurements and satellite observations of forested ecosystems, as well as CO_2 manipulation experiments, have generally reported increasing net ecosystem growth (primarily in forests) as CO_2 concentrations have increased in temperate regions of the world, although there have been fewer monitoring-based studies in the United States than in Europe (Norby et al., 2005; Boisvenue and Running, 2006). The ecological responses have not been consistent, however, in all studies and across all regions. A wide variety of factors (e.g., disturbance history, water availability, solar radiation), in addition to experimentally altered CO_2 concentrations, can contribute to these diverse patterns in net terrestrial ecosystem growth trends (Nemani et al., 2003; Boisvenue and Running, 2006; Potter et al., 2006).

Research at the Forest-Atmosphere Carbon Transfer and Storage (FACTS-I) facility in the Duke Forest has shown that nitrogen deposition can affect the uptake of atmospheric carbon dioxide as forests grow (photo courtesy of U.S. Department of Energy's Brookhaven National Laboratory).

There are several challenges in trying to quantify the role of CO_2 in forest growth. Many factors generally co-vary across sites, and experiments necessary to understand the interacting effects of these multiple factors have not been undertaken (Norby and Luo, 2004; Rustad, 2008). Also, the nearly impossible task of designing experiments that adequately consider variations in the numerous factors that influence ecosystem growth has been noted (Aber et al., 2001; Boisvenue and Running, 2006). When variations in atmospheric sulfur and, especially, nitrogen deposition are added to this mix of factors, the difficulty in attributing cause and effect to observed changes in ecosystems increases further (Bytnerowicz et al., 2007), and some argue, can only be interpreted through the use of models (Aber et al., 2001). Nonetheless, results from a CO_2 enrichment experiment that simulated likely twenty-first century concentrations showed increased soil acidification and increased rates of mineral weathering, suggesting that important interactions with terrestrial ecosystem effects of atmospheric deposition may occur in the future (Andrews and Schlesinger, 2001).

Changes in the Water Cycle are Key to Understanding Ecosystem Effects

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

Earlier snowmelt has also been noted in upland watersheds in the Northeast (Burns et al., 2007). Model predictions with likely future climate scenarios indicate continued diminishing snowfall, less snowpack, and less of a role for snowmelt in the hydrologic cycle of temperate regions of the Northern Hemisphere (IPCC, 2007).

These patterns may affect the cycling of sulfur and nitrogen pollutants through ecosystems and watersheds by impacting episodic acidification, soil freezing, and the flows and temperatures in surface waters. When snow melts, surface-water chemistry is often at its most acidic and has its highest nitrate concentrations; therefore, a diminished snowmelt may lessen the episodic acidification of aquatic ecosystems (Moore et al., 1997).

However, these predicted trends of diminishing snowmelt are uncertain, and an increase in mid-winter rain has been noted in the eastern United States (Hodgkins et al., 2003). This increase in winter rain will also likely trigger strong episodic acidification, even when the rain falls on a diminished snowpack. Additionally, increases in large rainfall events have been observed and are predicted to increase with climate change in the twenty-first century (Murdoch et al., 2000). Increased episodic acidification from these large rainfall events may offset the predicted decreasing trend in snowmelt. In addition, diminished snow cover may change soil freezing patterns in winter, which are dependent on the magnitude of warming. Frequent freeze-thaw behavior in soils can increase nitrate leaching and raise nitrate concentrations in surface waters (Fitzhugh et al., 2003).

Finally, earlier and diminished snowmelt may result in lower stream flows and warmer water temperatures in the later spring and summer. Warmer temperatures can result in increased growth rates. Increased aquatic growth can lower nitrate concentrations through biological uptake (Sommaruga-Wogarth et al., 1997); however, increased growth also can have negative impacts (e.g., hypoxia; see the section *Effects of Atmospheric Nitrogen Deposition on Coastal Estuaries* in this chapter for further discussion). Warmer temperatures and lower flows in streams may also have deleterious effects on cold-water fish species (Mohseni et al., 2003).

A nearly universal prediction from models of future climate is that droughts will increase in severity and duration, even in areas such as the eastern United States where overall increases in precipitation are predicted (IPCC, 2007). This forecast has strong implications for the impacts of acidification on aquatic ecosystems. When soils are dry, oxidized nitrogen and sulfur species accumulate and are released and flushed into local waterbodies upon re-wetting. Several studies have shown increased concentrations of nitrate and sulfate and decreased ANC values in surface waters when wet conditions return following drought periods. These impacts are especially apparent in wetland-influenced watersheds (Tipping et al., 2003; Watmough et al., 2004; Schiff et al., 2005).

This pattern of nitrogen and sulfur storage and release has further implications for the episodic acidification of waterbodies. Soils affected by acid deposition often contain levels of stored nitrogen and sulfur that are equivalent to decades of atmospheric deposition. Therefore, the aquatic ecosystem effects of severe, post-drought episodic acidification in lakes and streams and other surface waters may persist for decades after deposition levels have declined below critical loads (Tipping et al., 2003; Eimers et al., 2007).

Rapid shifts from periods of dry to wet conditions are also expected to increase in the future. Increases in acidification driven by such climatic shifts have been shown to directly affect aquatic biological communities such as diatom algae (Faulkenham et al., 2003) and to alter physico-chemical variables that affect aquatic life, such as the penetration of ultraviolet radiation (Yan et al., 1996).

Nitrogen Availability has Important Implications for the Response of Ecosystems to Climate Change

One of the most uncertain and important ecosystem-related issues regarding the interaction of the atmospheric deposition of pollutants with changes in climate and CO_2 levels concerns the role of nitrogen availability and its impact on growth and carbon sequestration. Growth in the majority of U.S. and global ecosystems is limited by nitrogen availability, suggesting that atmospheric nitrogen

deposition may be acting to increase growth and carbon sequestration in many ecosystems across the United States. However, in terrestrial ecosystems, excess atmospheric deposition of nitrogen also contributes to ecosystem acidification, calcium depletion in soils, loss of diversity, and excess runoff of nitrate, which can, in turn, lead to over-enrichment in freshwater and estuarine ecosystems. Therefore, when considering atmospheric deposition of nitrogen, the potential "benefit" of increased growth and carbon sequestration must be weighed against the potential "costs" of resulting nutrient over-enrichment, decreased ecosystem services (e.g., biodiversity), and the promotion of invasive species (Fenn et al., 2003b). These issues are brought into sharp focus by studies that seek to determine the role of nitrogen in the present and future growth of terrestrial ecosystems (particularly forests) and the sequestration of carbon as climate and atmospheric CO_2 concentrations continue to change.

One general conclusion is that net ecosystem growth in temperate and boreal forests has increased coincident with climate change in the United States, across Europe, and in parts of Asia over the past few decades (Goodale et al., 2002; Nemani et al., 2003; Boisvenue and Running, 2006; Magnani et al., 2007). However, many local and regional exceptions to this generalization can be found (Korner, 2003). Another general conclusion is that atmospheric nitrogen deposition acts to increase net ecosystem growth and carbon sequestration in mid-latitude forests (Reich et al., 2006; Magnani et al., 2007), although the magnitude of this nitrogen-driven carbon sink is likely much less than originally estimated by some studies (Sutton et al., 2008). Recent estimates of this sink are approximately 3% to 21% of annual CO_2 emitted globally from fossil fuel combustion (Churkina et al., 2009; Thomas et al., 2009; Zaehle et al., 2010). The interactions of carbon and nitrogen are critical in controlling the magnitude of the terrestrial sequestration of a large amount of the anthropogenic (i.e., man-made) CO_2 that would otherwise reside in the atmosphere and further alter the global climate. As CO_2 in the atmosphere increases, terrestrial carbon sinks (especially forests) have been shown to increase; however, a point may be reached when nitrogen and other nutrients become limiting (Johnson, 2006; van Groenigen

et al., 2006). The role of atmospheric nitrogen deposition in the global carbon cycle highlights the complex linkages among biogeochemical cycles and the important link between air pollutant deposition and global climate change.

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).

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); and

- Chronic inputs of atmospheric nitrogen deposition (Findlay, 2005).

CASTNET monitoring site in Rocky Mountain National Park, Larimer County, CO (photo courtesy of U.S. EPA).

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 CO_2 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).

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

> Terrestrial ecosystems store large amounts of carbon and the potential for these ecosystems to sequester or release additional carbon will likely contribute to future climate change. Research indicates that atmospheric nitrogen deposition affects the ability of terrestrial ecosystems to sequester atmospheric carbon dioxide.

Summary of Climate Change— Atmospheric Deposition Linkages

The results summarized here provide a strong scientific basis for joint consideration of climate change and air quality policies affecting atmospheric nitrogen and sulfur deposition. Topics considered under the umbrella of global climate change include past, ongoing, and future expected changes in a wide range of factors that affect ecosystems, such as warmer air temperatures, water availability, and the frequency of large magnitude events (e.g., droughts, floods, fires). The combination of patterns in the atmospheric deposition of sulfur and nitrogen with these global climate change factors has implications for the growth of ecosystems, expected shifts of species, and the acid/base chemistry of surface waters. Untangling the interactions among multiple biogeochemical cycles and the resulting ecosystem responses to changes in these cycles is complex and uncertain. For example, global climate change generally provides an additional set of interacting stressors in ecosystems that are negatively impacted by acid deposition (see Figure 3-3). However, climate change also may increase rates of chemical weathering in some locations and, therefore,

ecosystems may be able to sustain greater amounts of sulfur or nitrogen deposition before experiencing negative effects. Conversely, atmospheric nitrogen deposition may currently be limiting global warming through enhanced ecosystem carbon sequestration in U.S. regions that are only lightly to moderately impacted by air pollutant deposition.

Currently, the degree of uncertainty regarding many of the predicted effects of global climate change on ecosystems is high and often not well quantified. Environmental monitoring programs are helpful in understanding climate-change effects on ecosystems, but often are not adequate in geographic scope or in the number of parameters measured to fully understand these effects. For example, the leading programs that measure atmospheric deposition (NADP and CASTNET) and surface-water chemistry (LTM and TIME) generally do not measure many climatically relevant variables, such as snow-water equivalents and air and water temperature. Additionally, several of the papers reviewed here highlighted the critical need for more multi-factorial and interdisciplinary experiments to test the effects of varying air/soil/water temperature, CO_2 concentrations, and
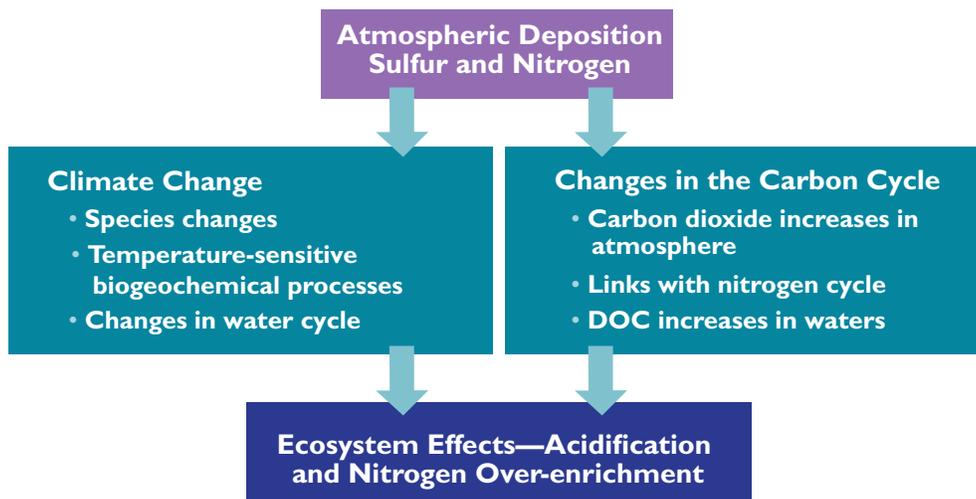
Note: Although the rate of atmospheric deposition of sulfur and nitrogen are the principal drivers of the ecosystem effects discussed in this chapter, ongoing and future changes in climate and the carbon cycle are expected to interact with these ecosystem effects in ways that are currently difficult to predict. Ongoing research efforts are helping to unravel the role of climate change on ecosystems so that more accurate model forecasts can better constrain the role of climate and carbon.

Figure 3-3. Climate change, acid deposition, and other stressors (prepared by USGS).

pollutant deposition levels on ecosystems. There is also a need for consideration of climate-change effects in acidification and nitrogen effects models, such as MAGIC and PnET-BGC, as they are used to provide predictions of the expected impacts of future air pollutant deposition scenarios. Work of this type is ongoing, but not yet widely available in the published literature. Evaluations of the ability of these models to incorporate changing climatic effects would also be helpful in providing feedback to discussions about future policies. Finally, past and future disturbances that are not necessarily climate-change driven (e.g., land disturbance, land use changes, invasive species, fire) can have ecosystem effects that are as great or greater than those derived from climate change and are also worthy of consideration in future scenario modeling.

Multi-Pollutant Interactions in Ecosystems

Throughout this chapter, the interacting effects of nitrogen and sulfur deposition have been discussed in reference to the acidification and nutrient over-enrichment of ecosystems. In the previous section, the interacting effects of acid deposition and changes in the carbon cycle and climate change were discussed. There are also important interactions between acid deposition and the ecosystem effects of ozone and mercury, and these interactions will be discussed in this section. In recent years, interest has been increasing in development of air quality policies that target multiple pollutants (NRC, 2004; Brook et al., 2009; NARSTO, 2011). This section focuses on the ecosystem aspects of two pollutants (i.e., ozone and mercury) for which clear links have been demonstrated.

Ozone Interactions with Atmospheric Sulfur and Nitrogen Deposition

Ozone is taken up by plants and, in high concentrations, can directly damage plant cell membranes, reduce the rate of photosynthesis and growth, and cause species shifts in forests (Karnosky et al., 2007). In short, ozone is yet another pollutant stressor that can interact with sulfur and nitrogen deposition and changes in climate and CO_2 concentrations to affect

ecosystem health (McLaughlin and Percy, 1999). Ozone concentrations and trends are highly variable across the globe and the United States, but leveling or slight decreases in concentrations in North America have been noted in recent years (Vingarzan, 2004; Oltmans et al., 2006). Trends in ozone concentrations are affected by trends in (1) the sources of ozone, such as fossil fuel use, (2) the principle atmospheric precursors of ozone (i.e., NO_x, volatile organic compounds, and carbon monoxide), and (3) climatic factors, particularly summer stagnation episodes that favor the highest concentrations. Results from models that simulate likely twenty-first century climate, combined with chemical transport model outputs, indicate a likelihood of increased future ozone concentrations in the United States (Jacob and Winner, 2009). Additionally, tropospheric ozone (ozone in the lowest portion of the Earth's atmosphere) provides the third-strongest warming influence of all greenhouse gases (Mickley et al., 1999), and recent work suggests that ozone also contributes indirectly to warming by slowing growth, and subsequently CO_2 uptake, in terrestrial ecosystems (Sitch et al., 2007).

Few studies have simultaneously considered the ecosystem effects of ozone combined with sulfur and nitrogen deposition. The added complication of ongoing changes in the climate and the carbon cycle provide additional challenges for understanding patterns and predicting likely ecosystem impacts of ozone, sulfur, and nitrogen loading. The inability of current models to adequately consider the simultaneous effects of these pollutants has been noted (Aber et al., 2001; Dewar et al., 2009). Despite the lack of adequate models and sufficient experiments to understand multi-factorial variations of these pollutants, evidence supports a joint, interacting role of ozone and nitrogen deposition in a variety of ecosystem effects, including sensitivity to insects and pathogens; frost sensitivity; drought; and fire (Bytnerowicz et al., 2007). Thresholds of nitrogen deposition and ozone concentration likely exist, below which ecosystem effects cannot be detected and at which benefits such as increased ecosystem growth and carbon sequestration are likely. However, the exact levels of these thresholds are not well known, nor are the tipping points above

which negative ecosystem effects sharply increase (Aber et al., 2001). Better identification of such patterns in ecosystem effects could provide vital information for use in future air quality policies.

Mercury Interactions with Atmospheric Sulfur and Nitrogen Deposition

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

Great Smoky Mountains National Park protects one of the last wild trout habitats in the eastern United States (photo courtesy of the NPS).

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

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

CHAPTER 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.

Adverse impacts to ecological processes or ecosystem components include the results of reductions in ANC and 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; effects in alpine lakes; coastal eutrophication as a result of atmospheric deposition; reductions in biodiversity;

At a Glance: Modeling Results

- Additional emission reductions are still needed for ecological recovery, especially in acid-sensitive areas.

- Reduction of SO_2 and NO_x to below 1.10 and 0.44 million tons/year from the power sector, respectively, would dramatically reduce the number of lakes classified as waters of acute concern.

- Reduction of SO_2 to 0.44 million tons/year from the power sector would result in nearly full protection of lakes in the Northeast and the Adirondack Mountains.

- Some lag between emission reductions and ecological recovery is expected in certain areas, especially streams in the southeastern United States.

| Waters of Acute Concern Water acidic all the time; sensitive organisms cannot survive | Waters of Elevated Concern Water still acidified seasonally or after storms; sensitive organism richness still low | Waters of Moderate Concern Significant recovery, but water still acidified at times; sensitive organisms may survive | Waters of Low Concern Recovery of water chemistry; even sensitive organisms can survive |

Note: See text box titled "Acid Neutralizing Capacity" in Chapter 2 for a full discussion of the ecological effects of acidification.
Waters of acute concern are defined as having ANC less than 0 µeq/L.
Waters of elevated concern are defined as having ANC from 0 to less than 50 µeq/L.
Waters of moderate concern are defined as having ANC from 50 to less than 100 µeq/L.
Waters of low concern are defined as having ANC greater than or equal to 100 µeq/L.

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

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 because 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., 2008; U.S. EPA, 2008b, 2009f). 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 categories of acidification concerns, and these categories 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.

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, 2008b, 2009f). 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 SO_2 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., 2006; Driscoll et al., 2007; Sullivan et al., 2008; 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 England, and the Northern Appalachian Plateau. However, many lakes and streams still remain impacted by acid deposition under current deposition levels (U.S. EPA, 2009a, 2009d). For example, the extent of stream acidification in the western Adirondack Mountains remains high. The streams sampled in a recent study showed that 66%, or 718 km, of streams are prone to acidification and likely have levels of acidity harmful to their biota. Of the 66% of streams found to be prone to acidification, about half were likely to be chronically acidified (i.e., base cation surplus < 0 µg/L), with

the other 50% episodically acidified (i.e., base cation surplus > 25 µg/L) (Lawrence et al., 2008).

A major assessment of acid deposition and its effects in the Adirondack Mountains found that full implementation of the 1990 CAAA will result in only modest recovery in northeastern lakes and streams impacted by acid deposition (Sullivan et al., 2006a). However, based on model projections, this study also concluded that aggressive reductions of SO_2 and NO_x emissions from power generation of up to 70% and 50%, respectively, beyond 2001 levels, would allow for chemical recovery (i.e., become nonacidic) of most lakes in the Adirondack Mountains.

Researchers in the southern Appalachian Mountains concluded that streams in this region are still threatened by acid deposition (Sullivan et al., 2008). In particular, many brook trout streams in Virginia are still vulnerable to acidification at current deposition levels (Cosby et al., 2006). Based on model projections from two recent studies (Cosby et al., 2006; Sullivan et al., 2008), researchers concluded that further reductions of sulfate deposition beyond levels achieved by the Title IV SO_2 emission reductions are necessary to prevent further acidification of southern Appalachian Mountain streams and to allow currently impacted streams to recover in the region. Cosby et al. (2006) concluded that a moderate reduction of sulfur and nitrogen deposition of 50% and 22%, respectively, beyond Title IV produced a small improvement in stream water quality over the long term (by 2100) relative to current conditions for brook trout streams in Virginia. A more stringent scenario of 62% reduction for sulfur and 30% reduction for nitrogen deposition produced additional improvements in stream water quality over the long term (by 2100), although these reductions still did not return stream water quality to preacidification conditions (Cosby et al., 2006). In sensitive streams in North Carolina, Tennessee, and South Carolina, Sullivan et al. (2008) modeled similar emission reductions, as did Cosby et al. (2006), and found little future improvement in stream conditions resulting from moderate and aggressive emission controls (Sullivan et al., 2008).

Analysis of the Environmental Impact of Further Air Emission Reductions

In response to the CAAA Title IX statutory requirement to identify deposition rates that would prevent adverse ecological effects, this chapter analyzes several emission-reduction scenarios that broadly bound the range of reductions presented in the literature discussed above and are similar to scenarios modeled in past NAPAP reports. These results provide an indication of the environmental improvements that would be expected from additional emission reductions from sources affected by Title IV and other emission-reduction programs. These environmental improvements do not constitute full recovery in all areas of acid-sensitive forests, lakes, or streams that have been impacted by acid deposition. They do provide, however, an indication of the scope and magnitude of the impact of emission reductions on deposition levels and on acid-sensitive ecosystems (Figure 4-2). Acid-sensitive ecosystems are generally characterized by a low ability to buffer against acidification due to hydrologic, geologic, and soil characteristics. Other sensitive ecosystems, such as estuaries and western U.S. alpine lakes, would also benefit from reductions in nitrogen deposition, but effects on those ecosystems are not analyzed here. In addition, climate change may alter the response of lake and stream water quality to declining acidic deposition (see Chapter 3 for a discussion of climatic change and surface-water recovery). However, this modeling analysis, like most previous scientific studies of surface-water recovery from acidification, assumes a constant influence of climate over the simulation period. While the information presented in this chapter is useful to inform future actions, the analysis presented here focuses solely on ecological recovery. Other aspects of implementing these emission-reduction scenarios, such as the costs and additional benefits (e.g., human health), were not analyzed and are beyond the scope of this assessment. Additional information would be important to inform future actions, including the costs and other impacts of emission reductions from the power sector and other sectors and the value that the public places on further improvements to the environment and human health.

Scenarios Analyzed

This modeling effort analyzed several scenarios representing emission changes from stationary sources within the power sector for the purpose of evaluating ecological recovery. Results are presented for total sulfur and total nitrogen deposition because these pollutants are the primary components of acid deposition and the cause of anthropogenic surface-water acidification. This analysis focuses on reductions from the power generation sector (i.e., EGUs); however, many other sources also emit sulfur and nitrogen pollutants that contribute to acid deposition. For example, sources outside of the power generation sector are projected to emit approximately 52% of the SO_2 and 84% of the NO_x emitted in 2020 under the Base Case scenario described below (U.S. EPA, 2006c).

The following analysis compares emission and deposition levels under a Base Case scenario with three other sensitivity scenarios of various levels of additional reductions. The Base Case scenario consists of currently implemented programs and programs that were finalized as of spring 2005. These programs include Title IV, CAIR, and Tier II and Heavy Duty Diesel SO_2 and NO_x reductions. The analysis employs models and analytical tools that are widely used and peer reviewed. EPA's Response Surface Model (RSM) was used to estimate future deposition loads (U.S. EPA, 2006c). RSM is based on an air quality modeling approach known as meta-modeling that aggregates numerous individual Community Multiscale Air Quality model (CMAQ) simulations into a multidimensional air quality response surface. The Integrated Planning Model (IPM) was used to

Acid-Sensitive Areas

- Western United States
- New England
- Northeastern Coastal Plain
- Ouachita Mountains
- Upper Midwest
- Southern Appalachians
- Florida Highlands
- Central Appalachians and Northern Appalachian Plateau
- Adirondack Mountains
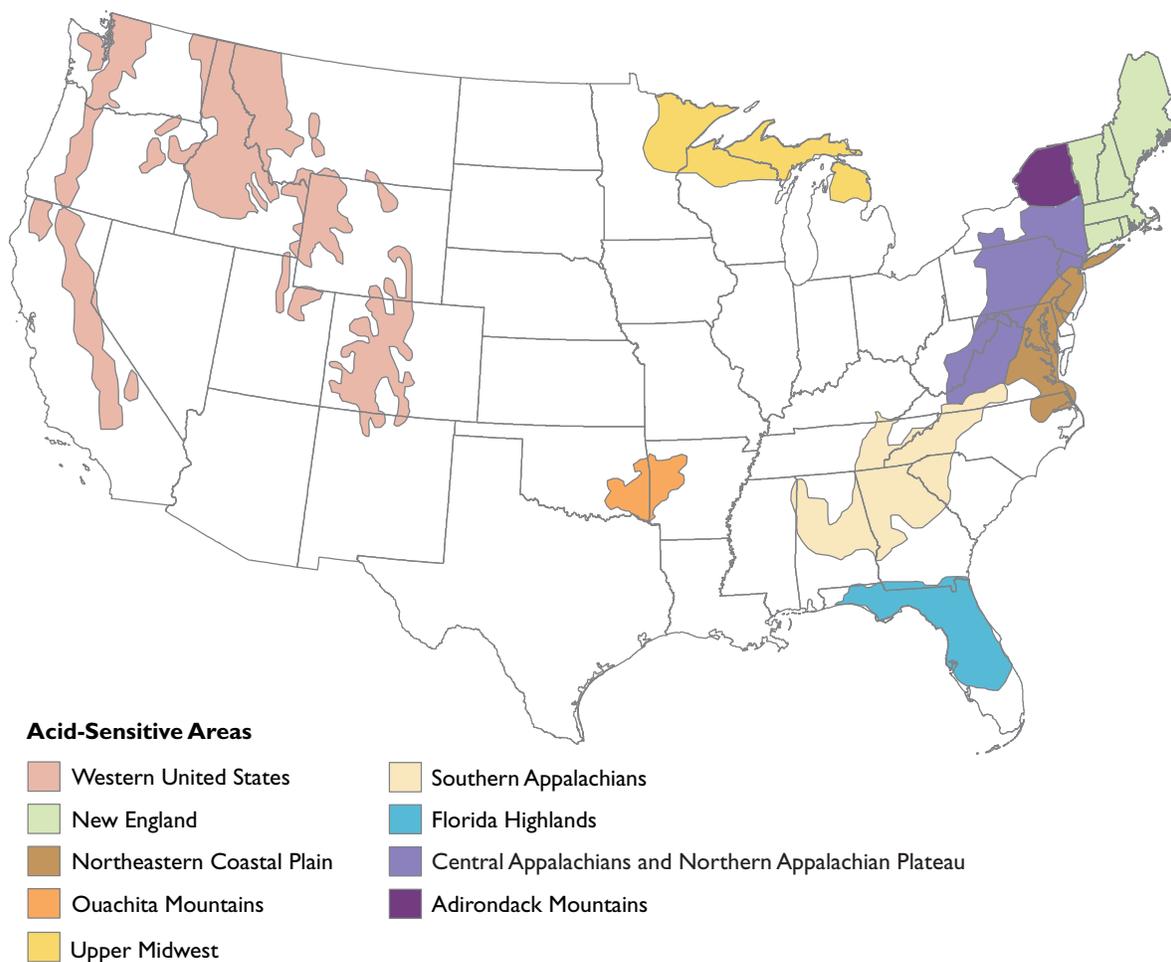
Figure 4-2. Map of acid-sensitive ecosystems in the United States (prepared by U.S. EPA).

estimate the power sector emissions from EGUs. As with any detailed analysis of complex scenarios, the results presented in this report are subject to uncertainties concerning emission estimates, air quality modeling, deposition projections, and the impact of emission reductions on ecological systems. See the text box titled *Modeling Tools Used in this Analysis* for a more 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 SO_2 and NO_x emission reductions beyond levels achieved by Title IV alone, the levels of SO_2 and NO_x modeled for the other future scenarios include additional reductions from the CAIR SO_2 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, 2006c). No controls were applied to the Canadian, Mexican, or offshore emission inventories. Additional details on the scenarios are summarized in Figure 4-3 and 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 Clean Air Act 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 SO_2 emissions from the power generation sector beyond the Base Case scenario, resulting in an annual SO_2 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 SO_2 emissions from the power generation sector beyond the Base Case scenario, equaling annual SO_2 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 SO_2 emissions from the power generation sector. It includes an additional 90% reduction in SO_2 emissions from the power generation sector beyond the Base Case scenario, resulting in 0.44 million tons SO_2/year in 2020, and a 50% reduction in SO_2 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.
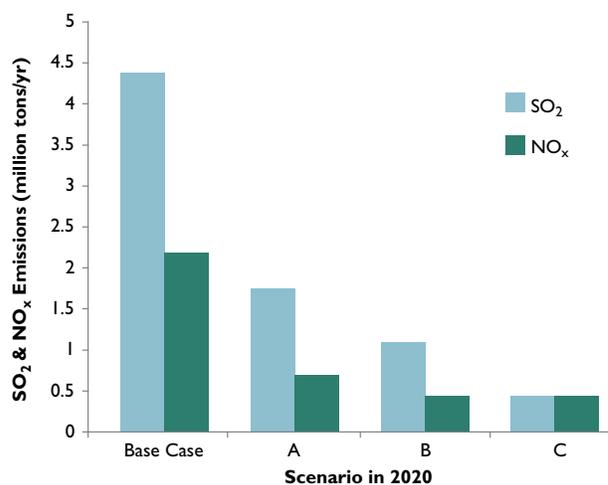

Figure 4-3. Projected annual SO_2 and NO_x emission levels in 2020 for the Base Case scenario and Scenarios A through C. Emission levels are for the continental United States (prepared by U.S. EPA).

Modeling Tools Used in this Analysis

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 SO_2, NO_x, CO_2, and mercury from the electric power sector. IPM provides the EGU emission inventory used as an input to CMAQ.

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 and 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 peer-reviewed, state-of-the-science model supported by the Community Modeling and Analysis System (CMAS) Center (http://www.cmascenter.org).

- 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, 2006b).

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, 2006c). 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.

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).
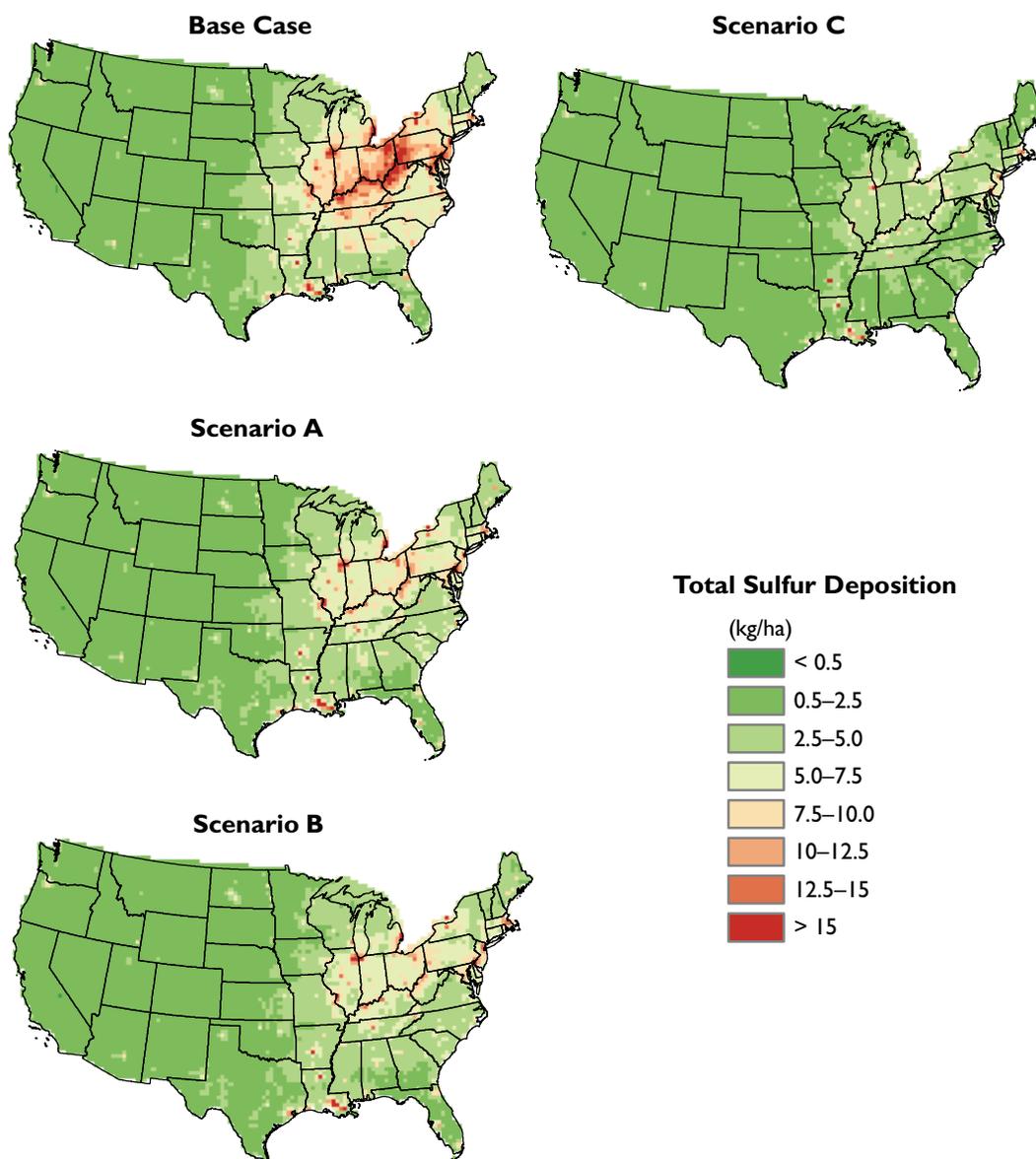

Figure 4-4. Projected change in annual total sulfur deposition from Base Case in 2020 with additional SO$_2$ and NO$_x$ emission reductions from the power generation sector and industrial boilers (prepared by U.S. EPA).

Scenario B extends the area of largest reduction in sulfur deposition southward and westward. For example, under Scenario B, the central Appalachians, Northern Appalachian Plateau, southern Appalachians, and Piedmont regions would all experience reductions in sulfur deposition beyond levels achieved by the 2020 Base Case scenario. Scenario C would extend the area with the greatest reductions in sulfur deposition beyond the 2020 Base Case scenario northward to Maine and westward through the Midwest across the Plains states to the Rocky Mountains (Figure 4-5), with some areas in the eastern United States experiencing reductions up to 60%. These reductions are expected to provide ecological benefits to these acid-sensitive regions, but will not be sufficient to allow full recovery. Many of the most sensitive sites within these acid-sensitive regions will not likely gain full recovery within the timeframe modeled in this analysis.

Scenario A

Scenario B

Total Sulfur Deposition
(Percent change)

🟥	10 to 20
🟧	0 to 10
⬜	-10 to 0
🟨	-20 to -10
🟩	-30 to -20
🟦	< -30

Scenario C
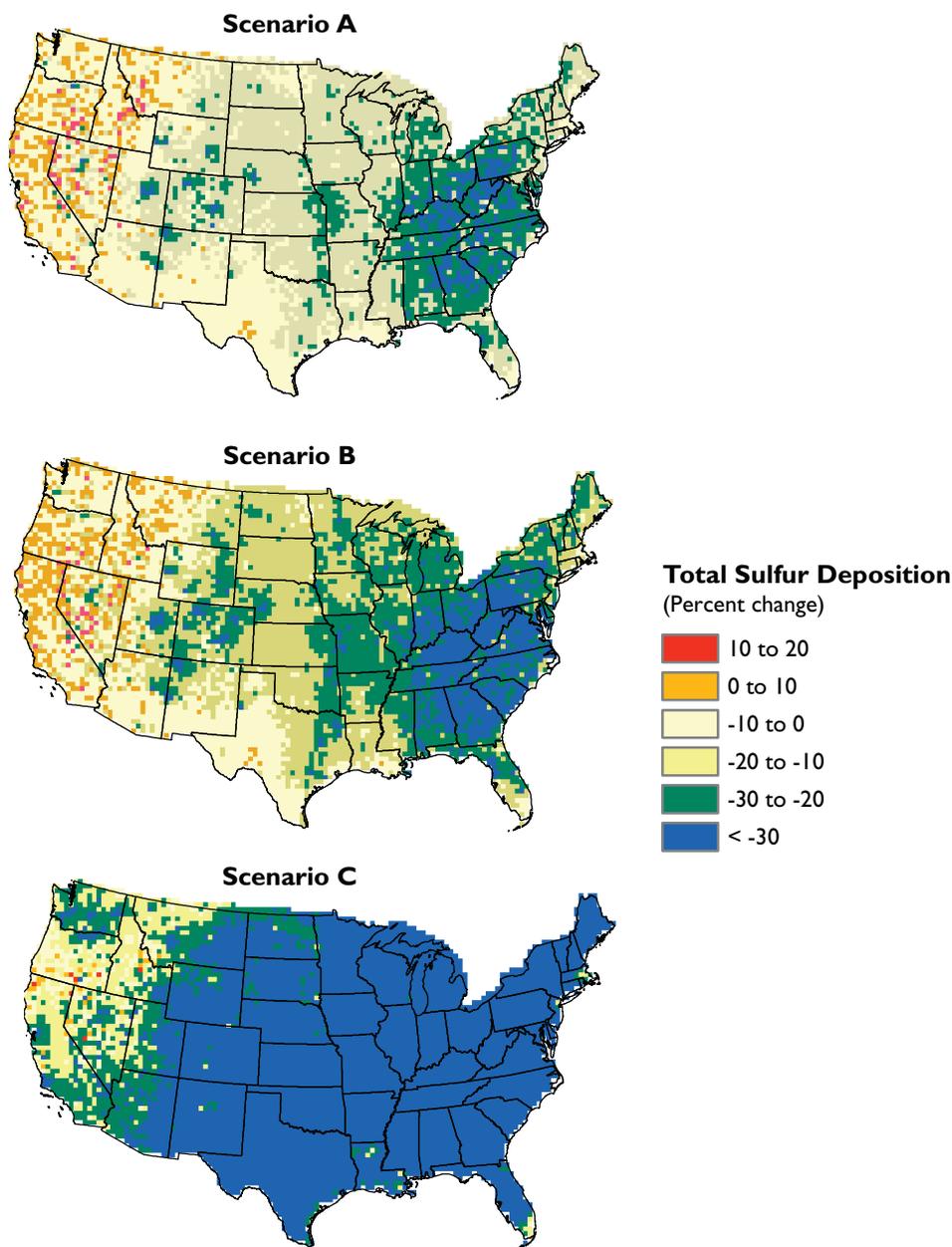
Figure 4-5. Projected percentage change in annual total sulfur deposition from Base Case in 2020 with additional SO_2 and NO_x emission reductions from the power generation sector and industrial boilers (prepared by U.S. EPA).

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.
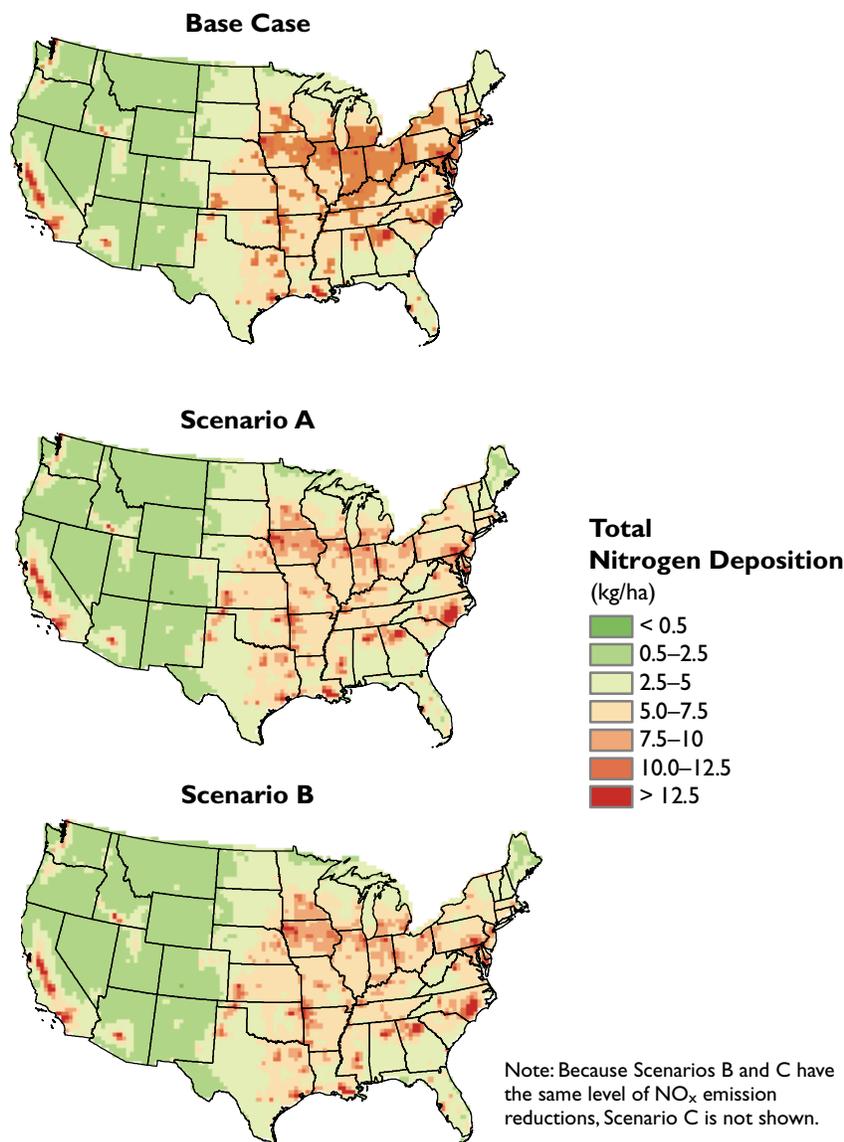

Figure 4-6. Projected change in annual total nitrogen deposition from Base Case in 2020 with additional SO_2 and NO_x emission reductions from the power generation sector (prepared by U.S. EPA).

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 4-7), as a result of higher NO_x emissions form other sources. 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.
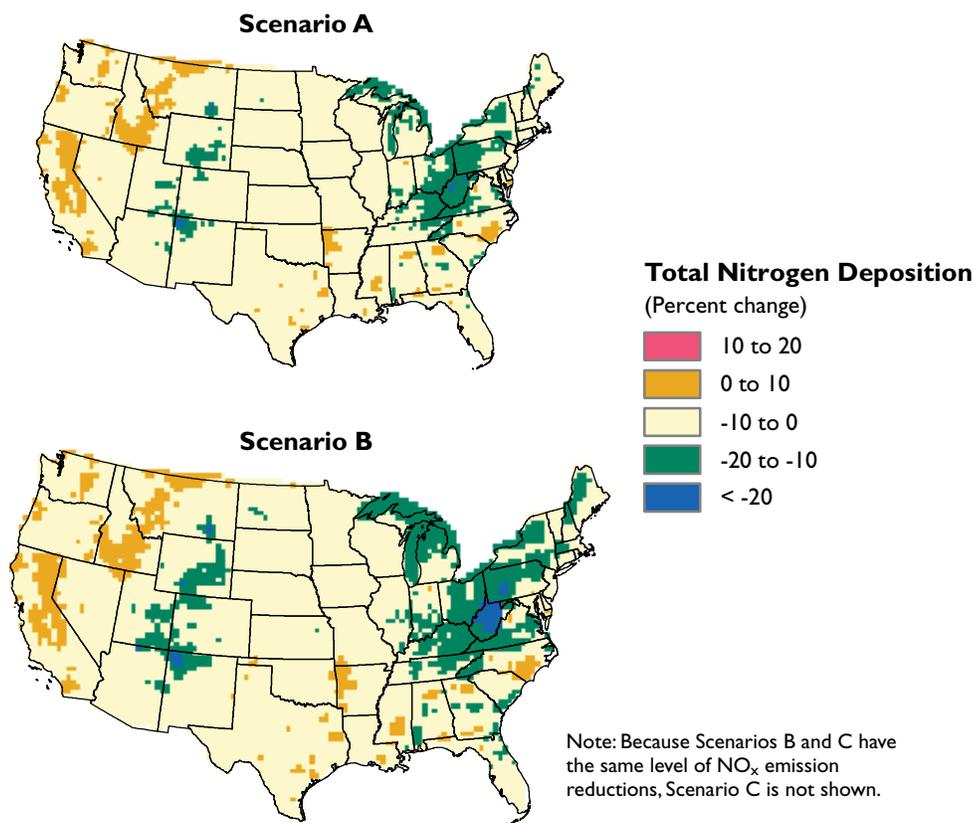

Figure 4-7. Projected percentage change in annual total nitrogen deposition from Base Case in 2020 with additional SO_2 and NO_x emission reductions from the power generation sector (prepared by U.S. EPA).

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. 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 from the power generation sector 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 from the power generation sector in 2020, with reductions over 30% in some areas, under Scenarios B and C as compared to the 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.

Regional Ecosystem Implications of Changes in Sulfur and Nitrogen Deposition

The projected sulfur and nitrogen deposition displayed in Figure 4-4 and 4-6 represents the amount of total sulfur and nitrogen deposition a given region will receive in 2020. As described in Chapter 3 of this report, sulfur and nitrogen deposition has the potential to cause ecological harm, including acidification, to sensitive ecosystems. Not all deposition loads will cause ecological harm, as some ecosystems have the ability to neutralize acids that are deposited from the atmosphere. However, some ecosystems have a limited ability to neutralize even low levels of acidic deposition to surface waters and soils. When these ecosystems exceed their ability to neutralize acidic deposition, surface waters and soils acidify to the point where aquatic and terrestrial life is harmed. The degree to which an ecosystem will be harmed is controlled by the level of ecosystem sensitivity and deposition load. Moreover, patterns of ecosystem

sensitivity vary by region of the country. Typically, in the eastern United States when deposition loads of sulfur and nitrogen are greater than 12 kg/ha/yr and 8 kg/ha/yr, respectively, widespread effects of acidification occur in surface waters. When deposition loads are below 5 kg/ha/yr for both sulfur and nitrogen, the most sensitive ecosystem elements are generally protected. While any level of deposition below 12 kg/ha/yr of sulfur and 8 kg/ha/yr of nitrogen provides additional protection, the most sensitive ecosystems are likely still strongly impacted (Aber et al., 2003; Brookshire et al., 2007). In special cases, particularly in western states, considerably lower levels of combined sulfur and nitrogen deposition may acidify headwater streams. In the Rocky Mountains, headwater streams can become episodically acidified during snowmelt when deposition loads are as low as 2 kg/ha/yr for combined sulfur and nitrogen.

Nitrogen deposition also contributes to nutrient over-enrichment of ecosystems. Nitrogen is a nutrient required by living systems, and so the level of nitrogen in an ecosystem often limits the system's productivity (i.e. the growth rate of plants and other organisms in the system). Because nitrogen is an important nutrient, the majority of nitrogen deposition is used or stored by ecosystems to allow for increased productivity. Nitrogen over-enrichment may cause eutrophication and lead to changes in species composition. The degree to which an ecosystem will be over enriched, and potentially harmed, is controlled by the ability of the ecosystem to use the additional nitrogen deposition, but still maintain a healthy and diverse ecological community.

- For many Rocky Mountain ecosystems, nitrogen deposition levels as low as 1.5 kg/ha/yr correspond to shifts in aquatic species composition, such as lake plankton, and levels as low as 4 kg/ha/yr can cause similar shifts in alpine plant communities (Baron, 2006; Bowman et al., 2006).

- In many areas of the far western United States, native plant communities such as coastal sage scrub, desert scrub, and annual grasslands are adapted to low-nitrogen environments. When nitrogen loads to these habitats

exceed 5–8 kg/ha/yr, the native plant and soil communities can be replaced by invasive species that are better adapted to more nitrogen-rich environments (Fenn et al., 2003a).

- For eastern and northern forests, the amount of nitrogen deposition generally harmful to forest ecosystems is less clear. Studies have shown that nitrogen deposition exceeding 8–26 kg/ha/yr led to increased surface water nitrate leaching and declines in growth and survivorship of sensitive tree species (Aber et al., 2003; McNulty et al., 2005). However, other studies based on modeling find that negative effects could occur in eastern and northern forests when nitrogen deposition is as low as 3–7 kg/ha/yr (Pardo and Duarte, 2007).

Changes in the Ecological Condition of Lakes and Streams

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

The Northeast, the Adirondack Mountains, and the Southeast were chosen for this modeling assessment because they are among the most acid-sensitive ecosystems and are located downwind of many of the emission sources affected by Title IV (see Figure 4-8). The best-available environmental data were used to calibrate MAGIC (e.g., water quality, soil, deposition). In addition, MAGIC was calibrated using data collected on acidification in lakes and streams by the National Surface Water Survey (NSWS) and other programs, such as EMAP and the TIME and LTM programs. Both the NSWS and EMAP used statistical methods to sample

a representative number of lakes and streams characteristic of the water quality and condition for all lakes or streams in each study region. For example, the NSWS approach represents approximately 7,100 lakes in the Northeast (Kaufmann et al., 1988) and 73,500 stream km in the southeast United States (U.S. EPA, 1988). Therefore, within the limitations of these surveys (only lakes larger than 4 hectares are included), the MAGIC results are indicative of the level of acidification in lakes and streams throughout these regions. For the analysis included in this report the MAGIC results represent approximately 1450 lakes in the Adirondack Mountains, 1200 lakes in the Northeast, and 47,500 km of streams in the Southeast. 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,

Lake Tear of the Clouds in the Adirondack Mountains (photo courtesy of New York State Department of Environmental Conservation).

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 (A, B, and C) were modeled in this analysis. The implementation of all scenarios is expected to significantly reduce the remaining percentage of lakes and streams in the acute and elevated concern categories in all three areas beyond what would occur with implementation of current emission-reduction programs included in the Base Case scenario. Although the effects of emission reductions

beyond the Base Case scenario differ by region, the amount of reductions results in a modest change in the number of waterbodies of acute concern in all regions. The greater the emission reductions, the larger the number of lakes and streams of elevated concern that improve and move into the moderate or low concern categories (see Figure 4-1 for an explanation of the stages of recovery from acidification).

Lakes in the Northeast and the Adirondack Mountains change relatively quickly in response to changes in deposition, resulting in shorter recovery times from acidification than streams in
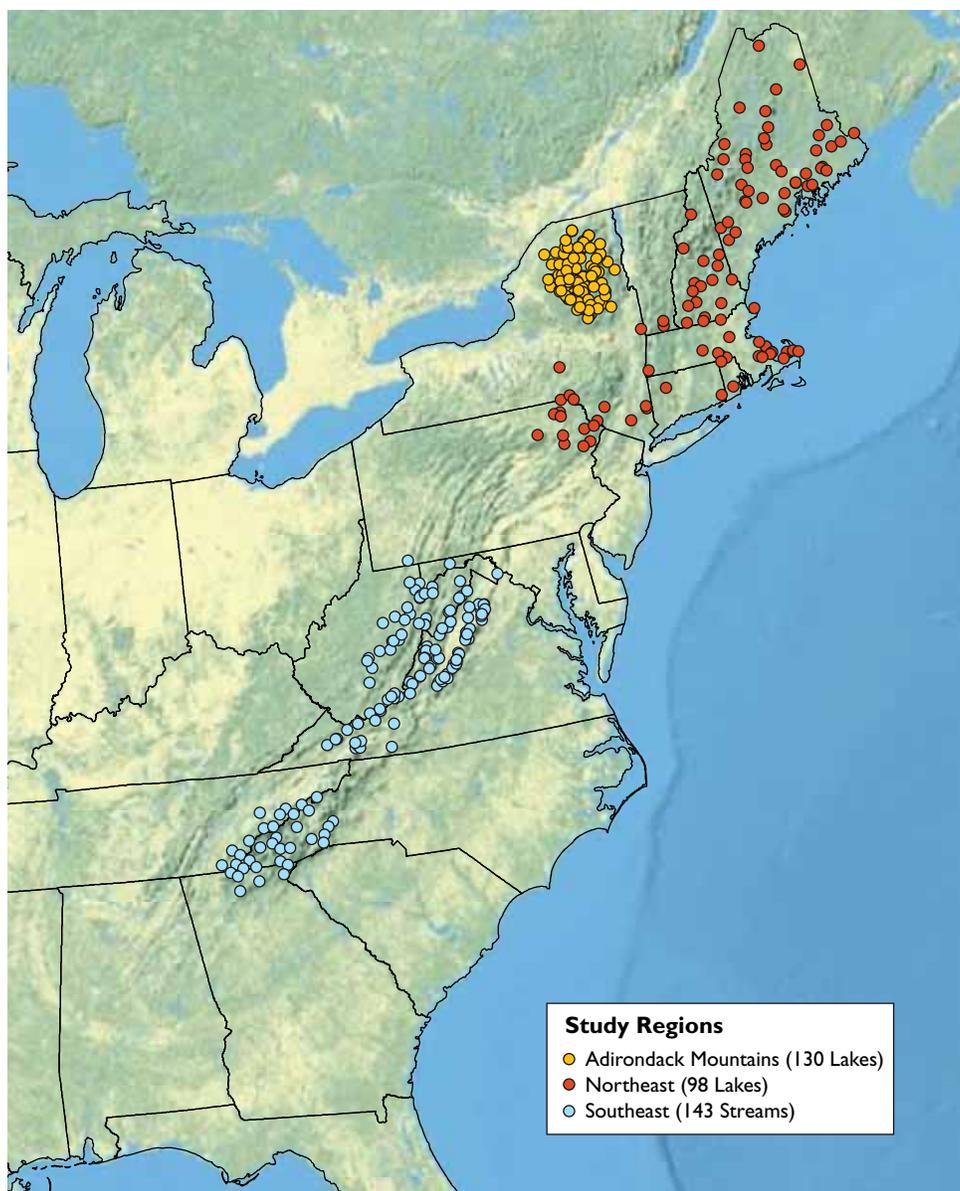

Figure 4-8. Regions and waterbodies modeled using MAGIC (prepared by U.S. EPA).

the Southeast. Modeling results indicate that the Base Case scenario would result in 6% and 8% of lakes remaining in the acute concern class in the Northeast and the Adirondack Mountains, respectively, by 2050 (Figure 4-9). Although these results would improve the health of fish populations and other acid-sensitive species, 40% and 14% of modeled lakes in the Adirondack Mountains and the Northeast, respectively, still would remain in the elevated concern class and experience episodic acidification (Figure 4-10) under the Base Case scenario. With implementation of Scenario A, water quality conditions improve in the Northeast and the Adirondack Mountains, but remain similar to those in the Base Case scenario. The percentage of lakes of acute concern would decrease to 5% for the Northeast and 4% for the Adirondack Mountains from the levels in the Base Case scenario, while 11% and 37% of modeled lakes would remain of elevated concern in these regions, respectively. A near elimination of lakes of acute concern in the Northeast and the Adirondack Mountains by 2050 only occurs given the stringent emission-reduction scenarios of Scenarios B and C. Under Scenarios B and C, the percentage of lakes in the elevated concern class would also decrease, which is expected

to improve the health of fish populations and allow other acid-sensitive species to return. With the implementation of Scenario C, the proportion of the lakes of elevated concern would decrease from 40% to 27% in the Adirondack Mountains and from 14% to 10% in the Northeast. Given that some lakes are naturally acidic because of organic acids, it is not expected that all lakes in a region will have an ANC level greater than 50 µeq/L. MAGIC also can estimate the water quality condition before anthropogenic acidification started, providing a target "natural" ANC level for assessing whether a population of lakes or streams has achieved full recovery. Under Scenario C, the percentage of lakes in the elevated concern class in 2050 is closer to modeled preacidification levels than under any other scenario, particularly for lakes in the Northeast. This suggests the scale of emission reductions under Scenario C would approach full protection of lakes in the Northeast and Adirondack Mountains from acid deposition (Figures 4-9 and 4-10).

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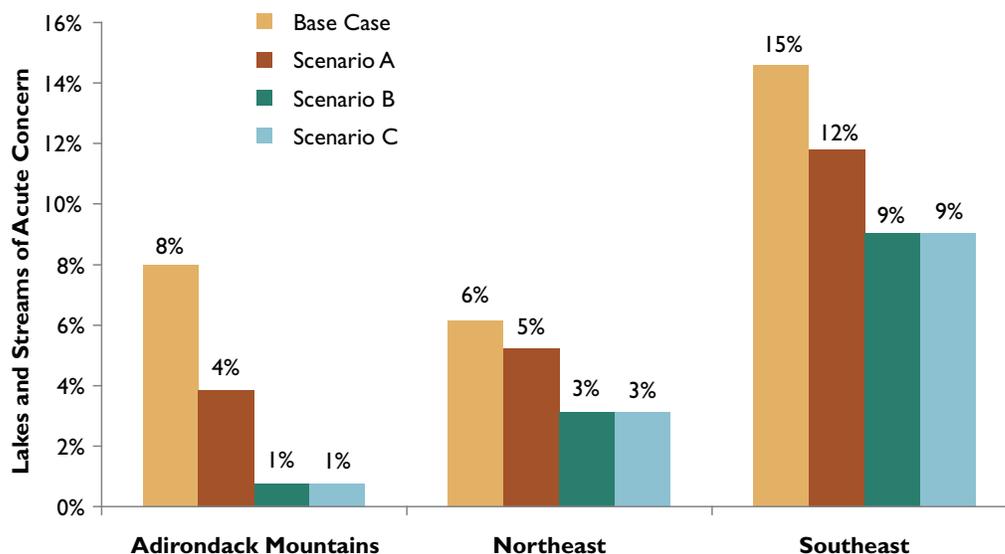
Notes: The area of the Northeast modeled by MAGIC includes Maine, New Hampshire, Vermont, Massachusetts, Rhode Island, Connecticut, New York (not including the Adirondacks), and northeastern Pennsylvania. The area of the Southeast includes Virginia, West Virginia, western North Carolina, eastern Tennessee,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

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

Figure 4-9. Projected changes in the number of lakes and streams of acute concern (ANC < 0 µeq/L) in 2050 with additional SO$_2$ and NO$_x$ emission reductions (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, western North Carolina, eastern Tennessee, northern Georgia, and northwestern South Carolina.

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 = 0–50 µeq/L) in 2050 with additional SO_2 and NO_x emission reductions (prepared by U.S. EPA).

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 SO_2 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.

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). Some lakes have naturally low acidic

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

Figure 4-11 shows lakes where deposition exceeds, does not exceed, and would be within 10% of the critical load. For Adirondack Mountain lakes under the Base Case scenario, 13% of lakes received levels of combined sulfur and nitrogen deposition that exceeded the critical load and could not be neutralized by the environment. For Scenarios A through C, 6%, 5%, and 1%, respectively, of the lakes would continue to receive acid deposition levels that exceeded the critical load. These results indicate that additional emission reductions lead to further ecosystem protection of lakes in the Adirondack Mountain region.
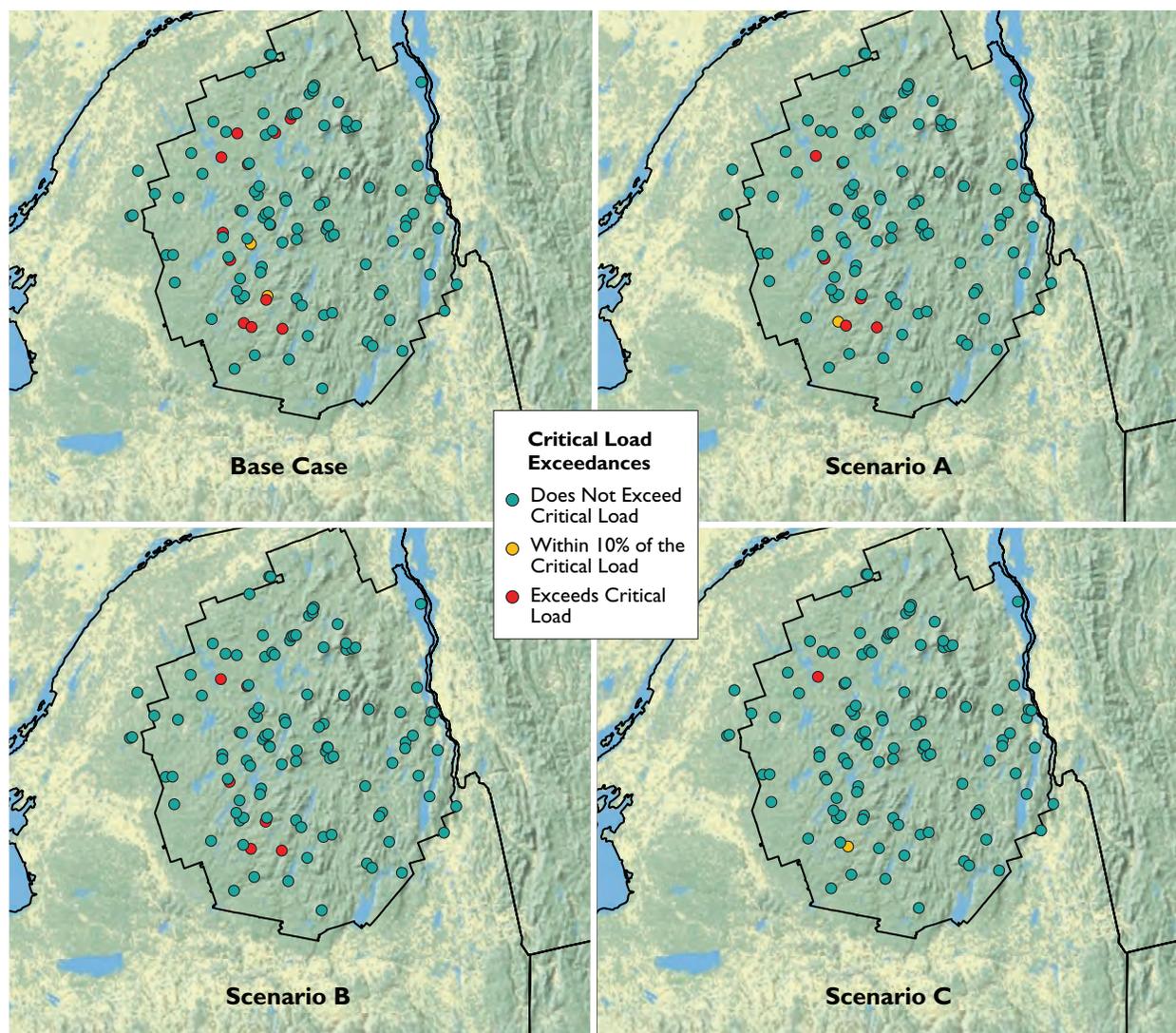
Note: Critical load exceedances for sulfur and nitrogen for 111 lakes in the Adirondack Mountains under Base Case scenario and Scenarios A through C.

Figure 4-11. Critical load exceedances in the Adirondack Mountains (prepared by U.S. EPA).

SUMMARY

Title IV has been successful in reducing emissions of SO_2 and NO_x from power generation to the levels set by Congress. In fact, by 2009, SO_2 emissions from power plants were already 3.25 million tons lower than the final 2010 cap level of 8.95 million tons, and NO_x emissions were 6.1 million tons less than the projected level in 2000 without the ARP, or more than triple the Title IV NO_x emission-reduction objective. As a result of these emission reductions, air quality has improved, providing significant human health benefits, and acid deposition has decreased to the extent that some acid-sensitive areas are beginning to show signs of recovery. Current emission reductions and the passage of time, which is needed for affected ecosystems to respond to the new environmental conditions, are expected to allow more acid-sensitive areas to recover. However, current emission-reduction levels (rules finalized as of spring 2005) are not sufficient to allow full recovery of acid-sensitive ecosystems. Estimates from modeling presented in this report show that additional emission reductions are necessary in order to protect acid-sensitive ecosystems.

This report analyzes a range of SO_2 and NO_x emission-reduction scenarios to evaluate the extent to which further reductions could achieve additional environmental recovery and minimize the adverse ecological effects associated with acid rain. The results of the modeling presented in this report indicate that broader recovery is not predicted without additional emission reductions. To fully protect acid-sensitive ecosystems in all regions affected by acid deposition will require reductions in acid-yielding emissions from source categories beyond power plant SO_2 emissions. The information presented in this report provides part of what is needed to determine appropriate future action; other information that is needed includes the costs and other impacts of emission reductions, the role of climate change and multiple pollutant interaction, and the value the public places on further improvements to the environment and human health.

REFERENCES

Aber, J., R.P. Neilson, S. McNulty, J.M. Lenihan, D. Bachelet, and R.J. Drapek. 2001. Forest processes and global environmental change: Predicting the effects of individual and multiple stressors. *BioScience 51*:735–751.

Aber, J.D., R.T. Holmes, R.J. Freuder, C.L. Goodale, S.V. Ollinger, C.T. Driscoll, and G.E. Likens. 2002. Inorganic nitrogen losses from a forested ecosystem in response to physical, chemical, biotic, and climatic perturbations. *Ecosystems 5*:648–658.

Aber, J.D., C.L. Goodale, S.V. Ollinger, M.L. Smith, A.H. Magill, M.E. Martin, R.A. Hallett, and J.L. Stoddard. 2003. Is nitrogen deposition altering the nitrogen status of northeastern forests? *BioScience 53*:375–389.

Alcamo, J., P. Mayerhofer, R. Guardans, T. van Harmelen, J. van Minnend, J. Onigkeit, M. Posch, and B. de Vries. 2002. An integrated assessment of regional air pollution and climate change in Europe: Findings of the AIR-CLIM Project. *Environmental Science & Policy 5*:257–272.

Alexander, R.B., and R.A. Smith. 2006. Trends in the nutrient enrichment of U.S. rivers during the late 20th century and their relation to changes in probable trophic conditions. *Limnology & Oceanography 51*:639–654.

Alexander, R.B., E.W. Boyer, R.A. Smith, G.E. Schwarz, and R.B. Moore. 2007. The role of headwater streams in downstream water quality. *Journal of the American Water Resources Association 43*:41–59.

Alexander, R.B., R.A. Smith, G.E. Schwarz, E.W. Boyer, J.V. Nolan, and J.W. Brakebill. 2008. Differences in phosphorus and nitrogen delivery to the Gulf of Mexico from the Mississippi River. *Environmental Science & Technology 42*:822–830.

Allen, E.B., L.E. Rao, R.J. Steers, A. Bytnerowicz, and M.E. Fenn. 2009. Impacts of atmospheric nitrogen deposition on vegetation and soils in Joshua Tree National Park. Pp. 78–100 *in* R.H. Webb, L.F. Fenstermaker, J.S. Heaton, D.L. Hughson, E.V. McDonald, and D.M. Miller, editors. *The Mojave Desert: Ecosystem Processes and Sustainability*. Las Vegas, NV: University of Nevada Press.

Andrews, J.A., and W.H. Schlesinger. 2001. Soil CO_2 dynamics, acidification, and chemical weathering in a temperate forest with experimental CO_2 enrichment. *Global Biogeochemical Cycles 15*:149–162.

Bailey, S.W., D.C. Buso, and G.E. Likens. 2003. Implications of sodium mass balance for interpreting the calcium cycle of a forested ecosystem. *Ecology 84*:471–484.

Bailey, S.W., S.B. Horsley, R.P. Long, and R.A. Hallett. 2004. Influence of edaphic factors on sugar maple nutrition and health on the Allegheny Plateau. *Soil Science Society America Journal 68*:243–252.

Bailey, S.W., S.B. Horsley, and R.P. Long. 2005. Thirty years of change in forest soils of the Allegheny Plateau, Pennsylvania. *Soil Science Society of America Journal 69*:681–690.

Baker, J.P., and S.W. Christensen. 1991. Effects of acidification on biological communities in aquatic ecosystems. Pp. 83–106 *in* D.F. Charles, editor. *Acidic Deposition and Aquatic Ecosystems*. New York: Springer-Verlag.

Baldigo, B.P., and P.S. Murdoch. 1997. Effect of stream acidification and inorganic aluminum on mortality of brook trout (*Salvelinus fontinalis*) in the Catskill Mountains, New York. *Canadian Journal of Fisheries and Aquatic Sciences 54*:603–615.

Banzhaf, H.S., D. Burtraw, D. Evans, and A. Krupnick. 2006. Valuation of natural resource improvements in the Adirondacks. *Land Economics 82*:445–464.

Baron, J.S. 2006. Hindcasting nitrogen deposition to determine an ecological critical load. *Ecological Applications 16*:433–439.

Baron, J.S., S. Del Grosso, D.S. Ojima, D.M. Theobald, and W.J. Parton. 2004. Nitrogen emissions along the Colorado Front Range: Response to population growth, land and water use change, and agriculture. Pp. 117-127 *in* R. DeFries, G. Asner, and R. Houghton, editors. *Ecosystems and Land Use Change*. Washington, DC: American Geophysical Union.

Baron, J.S., T.M. Schmidt, and M.D. Hartman. 2009. Climate-induced changes in high elevation stream nitrate dynamics. *Global Change Biology 15*:1777–1789.

Bates, N.R., and A.J. Peters. 2007. The contribution of atmospheric acid deposition to ocean acidification in the subtropical North Atlantic Ocean. *Marine Chemistry 107*:547–558.

Beckage, B., B. Osborne, D.G. Gavin, C. Pucko, T. Siccama, and T. Perkins. 2008. A rapid upward shift of a forest ecotone during 40 years of warming in the Green Mountains of Vermont. *Proceedings of the National Academy of Sciences 105*:4197–4202.

Beem, K.B., S. Raja, F.M. Schwandner, C. Taylor, T. Lee, A.P. Sullivan, C.M. Carrico, G.R. McMeeking, D. Day, E. Levin, J. Hand, S.M. Kreidenweis, B. Schichtel, W.C. Malm, and J.L. Collett, Jr. 2010. Deposition of reactive nitrogen during the Rocky Mountain Airborne Nitrogen and Sulfur (RoMANS) study. *Environmental Pollution 158*:862–872.

Beier, C.M., T.M. Patterson, and F.S. Chapin III. 2008. Ecosystem services and emergent vulnerability in managed ecosystems: A geospatial decision-support tool. *Ecosystems 11*:923–938.

Bell, M.L., A. McDermott, S.L. Zeger, J.M. Samet, and F. Dominici. 2004. Ozone and short-term mortality in 95 U.S. urban communities, 1987–2000. *Journal of the American Medical Association 292*:2372–8.

Bell, M.L., F. Dominici, and J.M. Samet. 2005. A meta-analysis of time-series studies of ozone and mortality with comparison to the national morbidity, mortality, and air pollution study. *Epidemiology 16*:436–45.

Bergstrom, A.-K., and M. Jansson. 2006. Atmospheric nitrogen deposition has caused nitrogen enrichment and eutrophication of lakes in the northern hemisphere. *Global Change Biology 12*:635–643.

Binks, J.A., S.E. Arnott, and W.G. Spules. 2005. Local factors and colonist dispersal influence crustacean zooplankton recovery from cultural acidification. *Ecological Applications 15*:2025–2036.

Blum, J.D., C.E. Johnson, T.G. Siccama, C. Eagar, T.J. Fahey, G.E. Likens, A. Klaue, C.A. Nezat, and C.T. Driscoll. 2002. Mycorrhizal weathering of apatite as an important calcium source in base-poor forest ecosystems. *Nature 417*:729–731.

Boisvenue, C., and S.W. Running. 2006. Impacts of climate change on natural forest productivity—evidence since the middle of the 20th century. *Global Change Biology 12*:862–882.

Bowman, W.D., T.A. Theodose, and M.C. Fisk. 1995. Physiological and production responses of plant growth forms to increases in limiting resources in alpine tundra: Implications for differential community response to environmental change. *Oecologia 101*:217–227.

Bowman, W.D., J.R. Gartner, K. Holland, and M. Wiedermann. 2006. Nitrogen critical loads for alpine vegetation and terrestrial ecosystem response: Are we there yet? *Ecological Applications 16*:1183–1193.

Boyer, E.W., C.L. Goodale, N.A. Jaworski, and R.W. Howarth. 2002. Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern U.S.A. *Biogeochemistry 57/58*:137–169.

Branfireun, B.A., N.T. Roulet, C.A. Kelly, and J.W.M. Rudd. 1999. In situ sulfate stimulation of mercury methylation in a boreal peatland: Toward a link between acid rain and methylmercury contamination in remote environments. *Global Biogeochemical Cycles 13*:743–750.

Bricker, S., B. Longstaff, W. Dennison, A. Jones, K. Boicourt, C. Wicks, and J. Woerner. 2008. Effects of nutrient enrichment in the nation's estuaries: A decade of change. *Harmful Algae 8*:21–32.

Brook, J.R., K.L. Demerjian, G. Hidy, L.T. Molina, W.T. Pennell, and R. Scheffe. 2009. New directions: Results-oriented multi-pollutant air quality management. *Atmospheric Environment 43*:2091–2093.

Brookshire, E.N.J., H.M. Valett, S.A. Thomas, and J.R. Webster. 2007. Atmospheric N deposition increases N loss from temperate forests. *Ecosystems 10*:252-262.

Bulger, A.J., B.J. Cosby, C.A. Dolloff, K.N. Eshleman, J.R. Webb, and J.N. Galloway. 1999. *SNP:FISH, Shenandoah National Park: Fish in Sensitive Habitats, Volumes through IV*. Project final report. Charlottesville, VA: University of Virginia, Department of Environmental Sciences. Project Completion Report to the National Park Service. Cooperative Agreement CA-4000-2-1007, Supplemental Agreement #2.

Burns, D.A. 2003. Atmospheric nitrogen deposition in the Rocky Mountains of Colorado and southern Wyoming—a review and new analysis of past study results. *Atmospheric Environment 37*:921–932.

Burns, D.A., C.T. Driscoll, G.M. Lovett, K.C. Weathers, M.J. Mitchell, and K.M. Roy. 2005. An assessment of recovery and key processes affecting the response of surface waters to reduced levels of acid precipitation in the Adirondack and Catskill Mountains. *New York Energy Research and Development Authority Report 05-03*, Albany, NY.

Burns, D.A., M.R. McHale, C.T. Driscoll, and K.M. Roy. 2006. Response of surface water chemistry to reduced levels of acid precipitation: Comparison of trends in two regions of New York. *Hydrological Processes 20*:1611–1627.

Burns, D.A., J. Klaus, and M.R. McHale. 2007. Recent climate trends and implications for water resources in the Catskill Mountain region, New York, USA. *Journal of Hydrology 336*:155–170.

Burns, D.A., K. R. Murray, R.W. Bode, and S.I. Passy. 2008a. Changes in stream chemistry and biology in response to reduced levels of acid deposition during 1987–2003 in the Neversink River Basin, Catskill Mountains. *Ecological Indicators 8*:191–203.

Burns, D.A., T. Blett, R. Haeuber, and L.H. Pardo. 2008b. Critical loads as a policy tool for protecting ecosystems from the effects of air pollutants. *Frontiers in Ecology & the Environment 6*:156–159.

Burtraw, D., and S.J. Szambelan. 2009. *U.S. Emissions Trading Markets for SO$_2$ and NO$_x$*. Discussion Paper RFF DP 09-40. Resources for the Future, Washington, DC.

Burtraw, D., D.A. Evans, A. Krupnick, K. Palmer, and R. Toth. 2005. Economics of pollution trading for SO$_2$ and NO$_x$. *Annual Review of Environment and Resources 30*:253–89.

Bytnerowicz, A., K. Omasa, and E. Paoletti. 2007. Integrated effects of air pollution and climate change on forests: A northern hemisphere perspective. *Environmental Pollution 147*:438–445.

Caine, N. 1995. Temporal trends in the quality of streamwater in an alpine environment: Green Lakes Valley, Colorado Front Range, U.S.A. *Geografiska Annaler. Series A, Physical Geography 77A*:207–220.

Cairns, A., and N. Yan. 2009. A review of the influence of low ambient calcium concentrations on freshwater daphniids, gammerids, and crayfish. *Environmental Reviews 17*:67–79.

Caldeira, K., and M.E. Wickett. 2003. Anthropogenic carbon and ocean pH. *Nature 425*:365.

Campbell, D.H., E. Muths, J.T. Turk, and P.S. Corn. 2004. Sensitivity to acidification of subalpine ponds and lakes in north-western Colorado. *Hydrological Processes 18*:2817–2834.

Campbell, J.L., L.E. Rustad, E.W. Boyer, S.F. Christopher, C.T. Driscoll, I.J. Fernandez, P.M. Groffman, D. Houle, J. Kiekbusch, A.H. Magill, M.J. Mitchell, and S.V. Ollinger. 2009. Consequences of climate change for biogeochemical cycling in forests of northeastern North America. *Canadian Journal of Forest Research 39*:264–284.

CANTORCO2e. 2009. *Market Price Index*. Online Information. CANTORCO2e, San Francisco, CA. Available at http://www.cantorco2e.com/Environment/?page=USAComp_MarketData-MPI (accessed 2009).

Carlson, C., D. Burtraw, M. Cooper, and K.L. Palmer. 2000. Sulfur dioxide control by electric utilities: What are the gains from trade? *Journal of Political Economy 108*(6):1292–1326.

Chen, L.M., and C.T. Driscoll. 2004. Modeling the response of soil and surface waters in the Adirondack and Catskill regions of New York to changes in atmospheric deposition and historical land disturbance. *Atmospheric Environment 38*:4099–4109.

Chen, L.M., and C.T. Driscoll. 2005a. Regional application of an integrated biogeochemical model to northern New England and Maine. *Ecological Applications 15*:1783–1797.

Chen, L.M., and C.T. Driscoll. 2005b. Regional assessment of the response of the acid-base status of lake watersheds in the Adirondack region of New York to changes in atmospheric deposition using PnET-BGC. *Environmental Science & Technology 39*:787–794.

Chestnut, L.G., and D.M. Mills. 2005. A fresh look at the benefits and costs of the U.S. Acid Rain Program. *Journal of Environmental Management 77*(3):252–266.

Chow, A.T., K.K. Tanji, and S. Gao. 2003. Production of dissolved organic carbon (DOC) and trihalomethane (THM) precursor from peat soils. *Water Research 37*:4475–4485.

Christophersen, N., H. Seip, and R. Wright. 1982. A model for streamwater chemistry at Birkenes, Norway. *Water Resources Research 18*:977-996.

Churkina, G., V. Brovkin, W. von Bloh, K. Trusilova, M. Jung, and F. Dentener. 2009. Synergy of rising nitrogen depositions and atmospheric CO_2 on land carbon uptake moderately offsets global warming. *Global Biogeochemical Cycles 23*, GB4027, doi:10.1029/2008GB03291.

Civerolo, K.L., C. Hogrefe, B. Lynn, C. Rosenzweig, R. Goldberg, J. Rosenthal, K. Knowlton, and P.L. Kinney. 2008. Simulated effects of climate change on summertime nitrogen deposition in the eastern U.S. *Atmospheric Environment 42*:2074–2082.

Clair, T.A., and J.M. Ehrmann. 1998. The influence of changing seasonal climates in modifying discharge, dissolved organic carbon and nitrogen export in wetland influenced rivers: A neural network approach. *Water Resources Research 34*:447–455.

Clair, T.A., and A. Hindar. 2005. Liming for the mitigation of acid rain effects in freshwaters: A review of recent results. *Environmental Reviews 13*:91–128.

Clair, T.A., I.F. Dennis, R. Vet, and H. Laudon. 2008. Long-term trends in catchment dissolved organic carbon and nitrogen from three acidified catchments in Nova Scotia, Canada. *Biogeochemistry 87*:83–97.

Clark, J.M., P.J. Chapman, A.L. Heathwaite, and J.K. Adamson. 2006. Suppression of dissolved organic carbon by sulfate induced acidification during simulated droughts. *Environmental Science & Technology 40*:1776–1783.

Clark, J.M., S.H. Bottrell, C.D. Evans, D.T. Monteith, R. Bartlett, R. Rose, R.J. Newton, and P.J. Chapman. 2010. The importance of the relationship between scale and process in understanding long-term DOC dynamics. *Science of the Total Environment 408*:2768–2775.

Clow, D.W., and J.K. Sueker. 2000. Relations between basin characteristics and stream water chemistry in alpine/subalpine basins in Rocky Mountain National Park, Colorado. *Water Resources Research 36*:49–61.

Clow, D.W., J.O. Sickman, R.G. Striegl, D.P. Krabbenhoft, J.G. Elliott, M. Domblaser, D.A. Roth, and D.H. Campbell. 2003. Changes in the chemistry of lakes and precipitation in high-elevation national parks in the western United States, 1985–1999. *Water Resources Research 39*, 1171, doi:10.1029/2008WR001533.

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 Organization Press.

Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985a. Modeling the effects of acid deposition: Assessment of a lumped parameter model of soil water and streamwater chemistry. *Water Resources Research 21*:51–63.

Cosby, B.J., R.F. Wright, G.M. Hornberger, and J.N. Galloway. 1985b. Modeling the effects of acid deposition: Estimation of a long term water quality responses in a small forested catchment. *Water Resources Research 21*:1591–1601.

Cosby, B.J., G.M. Hornberger, J.N. Galloway, and R.F. Wright. 1985c. Time scales of catchment acidification: A quantitative model for estimating freshwater acidification. *Environmental Science & Technology 19*:1144–1149.

Cosby, B.J., J.R. Webb, J.N. Galloway, and F.A. Deviney. 2006. *Acidic deposition Impacts on Natural Resources in Shenandoah National Park.* Technical Report NPS/NER/NRTR—2006/066. U.S. Department of the Interior, National Park Service, Northeast Region, Philadelphia, PA.

Costanza, R., R. d'Arge, R. de Groot, S. Farber, M. Grasso, B. Hannon, K. Limburg, S. Naeem, R.V. O'Neill, J. Paruelo, R.G. Raskin, P. Sutton, and M. van den Belt. 1997. The value of the world's ecosystem services and natural capital. *Nature 387*:253–260.

Dale, V.H., L.A. Joyce, S. McNulty, R.P. Neilson, M.P. Ayres, M.D. Flannigan, P.J. Hanson, L.C. Irland, A.E. Lugo, C.J. Peterson, D. Simberloff, F.J. Swanson, B.J. Stocks, and B.M. Wotton. 2001. Climate change and forest disturbances. *BioScience 51*:723–734.

Dalias, P., J.M. Anderson, P. Bottner, and M.M. Coûteaux. 2002. Temperature responses of net nitrogen mineralization and nitrification in conifer forest soils incubated under standard laboratory conditions. *Soil Biology & Biochemistry 34*:691–701.

DeHayes, D.H., P.G. Schaberg, G.L. Hawley, and G.R. Strimbeck. 1999. Acid rain impacts calcium nutrition and forest health. *BioScience 49*:787–800.

Deviney, F.A., K.C. Rice, and G.M. Hornberger. 2006. Time series and recurrence interval models to predict the vulnerability of streams to episodic acidification in Shenandoah National Park, Virginia. *Water Resources Research 42,* WO945, doi: 10.1029/2005WR004740.

Dewar, R.C., O. Franklin, A. Makela, R.E. McMurtrie, and H.T. Valentine. 2009. Optimal function explains forest responses to global change. *BioScience 59*:127–139.

Diaz, R.J. 2001. Overview of hypoxia around the world. *Journal of Environmental Quality 30*:275–281.

Diaz, R.J., and R. Rosenberg, 2008. Spreading dead zones and consequences for marine ecosystems. *Science 321*:926-929.

Dillon, P.J., and M.L. Molot. 1997. Dissolved organic and inorganic carbon mass balance in central Ontario lakes. *Biogeochemistry 36*:29–42.

Doney, S.C., N. Mahowald, I. Lima, R.A. Feely, F.T. Mackenzie, J.-F. Lamarque, and P.J. Rasch. 2007. Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and the inorganic carbon system. *Proceedings of the National Academy of Sciences 104*:14580–14585.

Drevnick, P.E., D.E. Canfield, P.R. Gorski, A.L.C. Shinneman, D.R. Engstrom, D.C.G. Muir, G.R. Smith, P.J. Garrison, L.B. Cleckner, J.P. Hurley, R.B. Noble, R.R. Otter, and J.T. Oris. 2007. Deposition and cycling of sulfur controls mercury accumulation in Isle Royale fish. *Environmental Science & Technology 41*:7266–7272.

Driscoll, C.T., R.M. Newton, C.P. Gubala, J.P. Baker, and S.W. Christensen. 1991. Adirondack mountains. Pp. 133–202 *in* D.F. Charles, editor. *Acidic Deposition and Aquatic Ecosystems: Regional Case Studies*. New York, NY: Springer-Verlag.

Driscoll, C.T., C.P. Cirmo, T.J. Fahey, V.L. Blette, P.A. Bukaveckas, D.A. Burns, C.P. Gubala, D.J. Leopold, R.M. Newton, D.J. Raynal, C.L. Schofield, J.B. Yavitt, and D.B. Porcella. 1996. The Experimental Watershed Liming Study: Comparison of lake and watershed neutralization strategies. *Biogeochemistry 32*:143–174.

Driscoll, C.T., G.B. Lawrence, A.J. Bulger, T.J. Butler, C.S. Cronan, C. Eagar, K.F. Lambert, G.E. Likens, J.L. Stoddard, and K.C. Weathers. 2001. Acidic deposition in the Northeastern United States: Sources and inputs, ecosystem effects, and management strategies. *BioScience 51*:180–198.

Driscoll, C.T., K.M. Driscoll, M.J. Mitchell, and D.J. Raynal. 2003. Effects of acidic deposition on forest and aquatic ecosystems in New York State. *Environmental Pollution 123*:327–336.

Driscoll C.T., K.M. Driscoll, K.M. Roy, and J. Dukett. 2007. Changes in the chemistry of lakes in the Adirondack region of New York following declines in acidic deposition. *Applied Geochemistry 22*:1181–1188.

Dupont, J., T.A. Clair, C. Gagnon, D.S. Jeffries, J.S. Kahl, S.J. Nelson, and J.M. Peckenham. 2005. Estimation of critical loads of acidity for lakes in the northeastern United States and eastern Canada. *Environmental Monitoring & Assessment 109*:275–291.

Durance, I., and S.J. Ormerod. 2007. Climate change effects on upland stream macroinvertebrates over a 25-year period. *Global Change Biology 13*:942–957.

Egerton-Warburton, L.M., and E.B. Allen. 2000. Shifts in arbuscular mycorrhizal communities along an anthropogenic nitrogen deposition gradient. *Ecological Applications 10*:484–496.

Egerton-Warburton, L.M., R.C. Graham, E.B. Allen, and M.F. Allen. 2001. Reconstruction of the historical changes in mycorrhizal fungal communities under anthropogenic nitrogen deposition. *Proceedings of the Royal Society of London B 268*:2479–2484.

Eimers, M.C., S.A. Watmough, J.M. Buttle, and P.J. Dillon. 2007. Drought-induced sulphate release from a wetland in south-central Ontario. *Environmental Monitoring & Assessment 127*:399–407.

Ellerman, A.D. 2003. *Ex Post Evaluation of Tradable Permits: The U.S. SO₂ Cap-and-Trade Program*. Massachusetts Institute of Technology, Center for Energy and Environmental Policy Research, Cambridge, MA.

EPRI (Electric Power Research Institute). 1993. *Integrated Analysis of Fuel, Technology and Emission Allowance Markets: Electric Utility Responses to the Clean Air Act Amendments of 1990*. Electric Power Research Institute, Palo Alto, CA.

Escobar, I.C., A.A. Randall, and J.S. Taylor. 2001. Bacterial growth in distribution systems: Effect of assimilable organic carbon and biodegradable dissolved organic carbon. *Environmental Science & Technology 35*:3442–3447.

Eshleman, K.N., B.E. McNeil, and P.A. Townsend. 2009. Validation of a remote sensing based index of forest disturbance using streamwater nitrogen data. *Ecological Indicators 9*:476-484.

Evans, C.D., P.J. Chapman, J.M. Clark, D.T. Monteith, and M.S. Cresser. 2006. Alternative explanations for rising dissolved organic carbon export from organic soils. *Global Change Biology 12*:2044–2053.

Evans, D.A., H.S. Banzhaf, D. Burtraw, A.J. Krupnick, and J. Siikimaki. 2008. Valuing benefits from ecosystem improvements using stated preference methods: An example from reducing acidification in the Adirondacks Park. Pp. 101–117, Chapter 9, *in* R.A. Askins, G.D. Dreyer, G.R. Visgilio, and D.M. Whitelaw, editors. *Saving Biological Diversity*. New York: Springer-Verlag.

Faulkenham, S.E., R.I. Hall, P.J. Dillon, and T. Karst-Riddoch. 2003. Effects of drought-induced acidification on diatom communities in acid-sensitive Ontario lakes. *Limnology & Oceanography 48*:1662–1673.

Fenn, M.E., J.S. Baron, E.B. Allen, H.M. Rueth, K.R. Nydick, L. Geiser, W.D. Bowmann, J.O. Sickman, T. Meixner, D.W. Johnson, and P. Neitlich. 2003a. Ecological effects of nitrogen deposition in the western United States. *BioScience 53*:404–420.

Fenn, M.E., M.A. Poth, A. Bytnerowicz, J.O. Sickman, and B.K. Takemoto. 2003b. Effects of ozone, nitrogen deposition, and other stressors on montane ecosystems in the Sierra Nevada. Pp. 111–155 *in* A. Bytnerowicz, M.J. Arbaugh, and R. Alonso, editors. *Developments in Environmental Science, Volume 2: Ozone Air Pollution in the Sierra Nevada: Distribution and Effects on Forests*. Amsterdam: Elsevier.

Fenn, M.E., L. Geiser, R. Bachman, T.J. Blubaugh, and A. Bytnerowicz. 2007. Atmospheric deposition inputs and effects on lichen chemistry and indicator species in the Columbia River Gorge, USA. *Environmental Pollution 146*:77–91.

Fenn, M.E., S. Jovan, F. Yuan, L. Geiser, T. Meixner, and B.S. Gimeno. 2008. Empirical and simulated critical loads for nitrogen deposition in California mixed conifer forests. *Environmental Pollution 155*:492–511.

Findlay, D.L. 2003. Response of phytoplankton communities to acidification and recovery in Killarney Park and the experimental lakes area, Ontario. *Ambio 32*:190–195.

Findlay, S.E.G. 2005. Increased carbon transport in the Hudson River: Unexpected consequence of nitrogen deposition? *Frontiers in Ecology & the Environment 3*:133–137.

Fisher, L.S., P.A. Mays, and C.L. Wylie. 2007. An overview of nitrogen critical loads for policy makers, stakeholders, and industries in the United States. *Water, Air, & Soil Pollution 179*:3–18.

Fitzhugh, R.D., G.E. Likens, C.T. Driscoll, M.J. Mitchell, P.M. Groffman, T.J. Fahey, and J.P. Hardy. 2003. Role of soil freezing events in interannual patterns of stream chemistry at the Hubbard Brook Experimental Forest, New Hampshire. *Environmental Science & Technology 37*:1575–1580.

Frey, K.E., and L.C. Smith. 2005. Amplified carbon release from vast west Siberian peatlands by 2100. *Geophysical Research Letters 32*, LO9401, doi:10.1029/2004GL022025.

Frost, T.M., J.M. Fischer, J.L. Klug, S.E. Arnott, and P.K. Montz. 2006. Trajectories of zooplankton recovery in the Little Rock Lake whole-lake acidification experiment. *Ecological Applications 16*:353–367.

Gbondo-Tugbawa, S.S., C.T. Driscoll, J.D. Aver, and G.E. Likens. 2001. Evaluation of an integrated model (PnET-BGC) at a northern hardwood ecosystem. *Water Resources Research 37*:1057–1070.

Geiser, L.H., and P.N. Neitlich. 2007. Pollution and climate gradients in western Oregon and Washington indicated by epiphytic macrolichens. *Environmental Pollution 145*:203–218.

Gherini, S.A., L. Mok, R.J.M. Hudson, G.F. Davis, C.W. Chen, and R.A. Goldstein. 1985. The ILWAS model: Formulation and application. *Water, Air, & Soil Pollution 26*:425–459.

Giardina, C.P., and M.G. Ryan. 2000. Evidence that decomposition rates of organic carbon in mineral soil do not vary with temperature. *Nature 404*:858–861.

Gilmour, C.C., E.A. Henry, and R. Mitchell. 1992. Sulfate stimulation of mercury methylation in freshwater sediments. *Environmental Science & Technology 26*:2281–2287.

Gimeno, B.S., F. Yuan, M.E. Fenn, and T. Meixner. 2009. Management options for mitigating nitrogen (N) losses from N saturated mixed conifer forests in California. Pp. 425–455 *in* A. Bytnerowicz, M.J. Arbaugh, A.R. Riebau, and C. Andersen, editors. *Wildland Fires & Air Pollution. Developments in Environmental Science, Volume 8*. Amsterdam: Elsevier.

Glavich, D.A., and L.H. Geiser. 2008. Potential approaches to developing lichen-based critical loads and levels for nitrogen, sulfur and metal-containing atmospheric pollutants in North America. *The Bryologist 111*:638–649.

Goodale, C.L., M.J. Apps, R.A. Birdsey, C.B. Field, L.S. Heath, R.A. Houghton, J.C. Jenkins, G.H. Kohlmaier, W. Kurz, S. Liu, G.-J. Nabuurs, S. Nilsson, and A.Z. Shvidenko. 2002. Forest carbon sinks in the Northern Hemisphere. *Ecological Applications 12*:891–899.

Graveland, J. 1998. Effects of acid rain on bird populations. *Environmental Reviews 6*:41–54.

Greening, H., and A. Janicki. 2006. Toward reversal of eutrophic conditions in a subtropical estuary: Water quality and seagrass response to nitrogen loading reductions in Tampa Bay, Florida, USA. *Environmental Management 38*:163–178.

Grigal, D.F., and P.R. Ohmann. 2005. Calcium and forest systems: Diffusion from deep sources. *Soil Science 170*:129–136.

Grimm, J.W., and J.A. Lynch. 2004. Enhanced wet deposition estimates using modeled precipitation inputs. *Environmental Monitoring & Assessment 90*:243–268.

Grulke, N.E., R.A. Minnich, T.D. Paine, S.J. Seybold, D.J. Chavez, M.E. Fenn, P.J. Riggan, and A. Dunn. 2009. Air pollution increases forest susceptibility to wildfires: A case study in the San Bernardino Mountains in southern California. Pp. 365–403 *in* A. Bytnerowicz, M.J. Arbaugh, A.R. Riebau, and C. Andersen, editors. *Wildland Fires and Air Pollution. Developments in Environmental Science, Volume 8*. Amsterdam: Elsevier.

Hallet, R.A., S.W. Bailey, S.B. Horsley, and R.P. Long. 2006. Influence of nutrition and stress on sugar maple at a regional scale. *Canadian Journal of Forest Research 36*:2235–2246.

Halvorsen, G.A., E. Heegaard, A. Fjellheim, and G.G. Raddum. 2003. Tracing recovery from acidification in the western Norwegian Nausta watershed. *Ambio 32*:235–239.

Hamburg, S.P., R.D. Yanai, M.A. Arthur, J.D. Blum, and T.G. Siccama. 2003. Biotic control of calcium cycling in northern hardwood forests: Acid rain and aging forests. *Ecosystems 6*:399–406.

Hames, R.S., K.V. Rosenberg, J.D. Lowe, S.E. Barker, and A.A. Dhondt. 2002. Adverse effects of acid rain on the distribution of the wood thrush *Hylocichla mustelina* in North America. *Proceedings of the National Academy of Sciences 99*:11235–11240.

Hartman, M.D., J.S. Baron, and D.S. Ojima. 2007. Application of a coupled ecosystem-chemical equilibrium model, DayCent-Chem, to stream and soil chemistry in a Rocky Mountain watershed. *Ecological Modelling 200*:493–510.

Hawley, G.J., P.G. Schaberg, C. Eager, and C.H. Borer. 2006. Calcium addition at the Hubbard Brook Experimental Forest reduced winter injury to red spruce in a high-injury year. *Canadian Journal of Forest Research 36*:2544–2549.

Held, I.M., and B.J. Soden. 2006. Robust responses of the hydrological cycle to global warming. *Journal of Climate 19*:5686–5699.

Henriksen, A. 1979. A simple approach for identifying and measuring acidification of freshwater. *Nature 278*:542–545.

Henriksen, A., and D.F. Brakke. 1988. Sulfate deposition to surface waters. *Environmental Science & Technology 22*:8–14.

Henriksen, A., and M. Posch. 2001. Steady-state models for calculating critical loads of acidity for surface waters. *Water, Air, & Soil Pollution: Focus 1*:375–398.

Heuer, K., K.A. Tonnessen, and G.P. Ingersoll. 2000. Comparison of precipitation chemistry in the central Rocky Mountains, Colorado, USA. *Atmospheric Environment 34*:1713–1722.

Higgins, P.A.T., and J. Harte. 2006. Biophysical and biogeochemical responses to climate change depend on dispersal and migration. *BioScience 56*:407–417.

Hodgkins, G.A., R.W. Dudley, and T.G. Huntington. 2003. Changes in the timing of high river flow in New England over the 20th century. *Journal of Hydrology 278*:244–252.

Howarth, R.W. 2008. Estimating atmospheric nitrogen deposition in the northeastern United States: Relevance to Narragansett Bay. Pp. 47–65, Chapter 3, *in* A. Desbonnet and B. A. Costa-Pierce, editors. *Science for Ecosystem-based Management: Narragansett Bay in the 21stCentury.* Springer Series on Environmental Management. New York: Springer.

Hrabik, T.R., and C.J. Watras. 2002. Recent declines in mercury concentration in a freshwater fishery: Isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little Rock Lake. *Science of the Total Environment 297*:229–237.

Huang, Y., F. Dominici, and M.L. Bell. 2005. Bayesian hierarchical distributed lag models for summer ozone exposure and cardio-respiratory mortality. *Environmetrics 16*:547–562.

Hudson, J.J., P.J. Dillon, and K.M. Somers. 2003. Long-term patterns in dissolved organic carbon in boreal lakes: The role of incident radiation, precipitation, air temperature, southern oscillation, and acid deposition. *Hydrology & Earth System Sciences 7*:390–398.

Ibanez, I., J.S. Clark, S. Ladeau, and J.H.R. Lambers. 2007. Exploiting temporal variability to understand tree recruitment response to climate change. *Ecological Monographs 77*:163–177.

ICF (ICF International, Inc.). 1989. *Economic Analysis of Title IV (Acid Rain Provisions) of the Administration's Proposed Clean Air Act Amendments (H.R. 3030/S. 1490).* ICF International, Inc., Fairfax, VA.

Ingersoll, G.P., M.A. Mast, D.H. Campbell, D.W. Clow, L. Nanus, and J.T. Turk. 2008. Trends in snowpack chemistry and comparison to National Atmospheric Deposition Program results for the Rocky Mountains, U.S., 1993–2004. *Atmospheric Environment 42*:6098–6113.

Interlandi, S.J., and S.S. Kilham. 2001. Limiting resources and the regulation of diversity in phytoplankton communities. *Ecology 82*:1270–1282.

IPCC (Intergovernmental Panel on Climate Change). 2007. *Climate Change 2007*: *Synthesis Report.* Intergovernmental Panel on Climate Change, Geneva, Switzerland.

Ito, K., S.F. De Leon, and M. Lippmann. 2005. Associations between ozone and daily mortality: Analysis and meta-analysis. *Epidemiology 16*(4):446–57.

Jacob, D.J., and D.A. Winner. 2009. Effect of climate change on air quality. *Atmospheric Environment 43*:51–63.

Jeffries, D., and R. Ouimet. 2005. Critical loads—Are they being exceeded? Pp. 341–368, Chapter 8, *in* D. Jeffries and R. Ouimet, editors. *2004 Canadian Acid Deposition Science Assessment.* Ottawa, Ontario: Environment Canada.

Jenkins, M.A., S. Jose, and P.S. White. 2007. Impacts of an exotic disease and vegetation change on foliar calcium cycling in Appalachian forests. *Ecological Applications 17*:869–881.

Jentsch, A., J. Kreyling, and C. Beierkuhlein. 2007. A new generation of climate-change experiments: Events, not trends. *Frontiers in Ecology & the Environment* 5:365–374.

Jeremiason, J.D., D.R. Engstrom, E.B. Swain, E.A. Nater, B.M. Johnson, J.E. Almendinger, B.A. Monson, and R.K. Kolka. 2006. Sulfate addition increases methylmercury production in an experimental wetland. *Environmental Science & Technology* 40:3800–3806.

Jeziorski, A., N.D. Yan, A.M. Paterson, A.M. DeSellas, M.A. Turner, D.S. Jeffries, B. Keller, R.C. Weeber, D.K. McNicol, M.E. Palmer, K. McIver, K. Arseneau, B.K. Ginn, B.F. Cumming, and J.P. Smol. 2008. The widespread threat of calcium decline in fresh waters. *Science 322*:1374–1377.

Johnson, A.H., A.J. Moyer, J.E. Bedison, S.L. Richter, and S.A. Willig. 2008. Seven decades of calcium depletion in organic horizons of Adirondack forest soils. *Soil Science Society of America Journal 72*:1824–1830.

Johnson, D.W. 2006. Progressive N limitation in forests: Review and implications for long-term responses to elevated CO_2. *Ecology 87*:64–75.

Jones, M.E., T.D. Paine, M.E. Fenn, and M.A. Poth. 2004. Influence of ozone and nitrogen deposition on bark beetle activity under drought conditions. *Forest Ecology & Management 200*:67–76.

Jovan, S. 2008. Lichen bioindication of biodiversity, air quality, and climate: Baseline results from monitoring in Washington, Oregon, and California. *General Technical Report PNW-GTR-737*. U.S. Department of Agriculture, Forest Service, Pacific Northwest Research Station, Portland, OR.

Jovan, S., and B. McCune. 2005. Air-quality bioindication in the greater central valley of California, with epiphytic macrolichen communities. *Ecological Applications 15*:1712–1726.

Kahl, J.S., J.L. Stoddard, R. Haeuber, S.G. Paulsen, R. Birnbaum, F.A. Deviney, J.R. Webb, D.R. DeWalle, W. Sharpe, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg, P.S. Murdoch, K. Roy, K.E. Webster, and N.S. Urquhart. 2004. Have U.S. surface waters responded to the 1990 Clean Air Act Amendments? *Environmental Science & Technology 38*:484A–490A.

Kamman, N.C., P.M. Lorey, C.T. Driscoll, R. Estabrook, A. Major, B. Pientka, and E. Glassford. 2004. Assessment of mercury in waters, sediments, and biota of New Hampshire and Vermont lakes, USA, sampled using a geographically randomized design. *Environmental Toxicology & Chemistry 22*:1172–1186.

Karnosky, D.F., J.M. Skelly, K.E. Percy, and A.H. Chappelka. 2007. Perspectives regarding 50 years of research on effects of tropospheric ozone air pollution on U.S. forests. *Environmental Pollution 147*:489–506.

Kaufmann, P.R, A.T. Herlihy, J.W. Elmwood, M.E. Mitch, W.S. Overton, M.J. Sale, J.J. Messer, K.A. Cougan, D.V. Peck, K.H. Reckhow, A.J. Kinney, S.J. Christie, D.D. Brown, C.A. Hagley, and H.I. Jager. 1988. *Chemical Characteristics of Streams in the Mid-Atlantic and Southeastern United States. Volume 1: Population Descriptions and Physico-Chemical Relationships.* EPA/600/3-88/021a. U.S. Environmental Protection Agency, Washington, D.C.

Kemp, W.M., W.R. Boynton, J.E. Adolf, D.F. Boesch, W.C. Boicourt, G. Brush, J.C. Cornwell, T.R. Fisher, P.M. Glibert, J.D. Hagy, L.W. Harding, E.D. Houde, D.G. Kimmel, W.D. Miller, R.I.E. Newell, M.R. Roman, E.M. Smith, and J.C. Stevenson. 2005. Eutrophication of Chesapeake Bay: Historical trends and ecological interactions. *Marine Ecology Progress Series 303*:1–29.

Kline, K.M., K.N. Eshleman, R.P. Morgan II, and N.M. Castro. 2007. Analysis of trends in episodic acidification of streams in western Maryland. *Environmental Science & Technology 41*:5601–5607.

Knapp, A.K., B. Smith, S.D. Smith, D.D. Briske, P.A. Fay, S.W. Leavitt, M.D. Smith, Y. Luo, C. Beier, M. Reichstein, J.L. Heisler, A.T. Classen, J.E. Bell, R. Sherry, and E. Weng. 2008. Consequences of more extreme precipitation regimes for terrestrial ecosystems. *BioScience 58*:811–821.

Korner, C. 2003. Slow in, rapid out—carbon flux studies and Kyoto targets. *Science 300*:1242–1243.

Kowalik, R.A., D.M. Cooper, C.D. Evans, and S.J. Ormerod. 2007. Acidic episodes retard the biological recovery of upland British streams from chronic acidification. *Global Change Biology 13*:2439–2452.

Kramer, R.A., T.P. Holmes, and M. Haefele. 2003. Contingent valuation of forest ecosystem protection. Pp. 303–320, Chapter 17, *in* E.O. Sills and K.L. Abt, editors. *Forests in a Market Economy.* Kluwer: Dordrecht.

Krishnamurthy, A., J.K. Moore, N. Mahowald, C. Luo, and C.S. Zender. 2010. Impacts of atmospheric nutrient inputs on marine biogeochemistry. *Journal of Geophysical Research 115*, GO1006, doi: 10.1029/2009JG001115.

Krupnick, A., and J. Siikimaki. 2008. Valuation of the environmental effects of N and S (non-materials). Annex F *in Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Ecological Criteria.* Report EPA/600/R-08/082F. U.S. Environmental Protection Agency, Research Triangle Park, NC.

Kvale, K.F., and S.C. Pryor. 2006. Precipitation composition in the Ohio River Valley: Spatial variability and temporal trends. *Water, Air, & Soil Pollution 170*:143–160.

Laden, F., J. Schwartz, F.E. Speizer, and D.W. Dockery. 2006. Reduction in fine particulate air pollution and mortality: Extended follow-up of the Harvard six cities study. *American Journal of Respiratory and Critical Care Medicine 173*:667–672.

Langford, T.E.L., P.J. Shaw, A.J.D. Ferguson, and S.R. Howard. 2009. Long-term recovery of macroinvertebrate biota in grossly polluted streams: Re-colonisation as a constraint to ecological quality. *Ecological Indicators 9*:1064–1077.

Lawler, J.J., J. Rubin, B.J. Cosby, I.J. Fernandez, J.S. Kahl, and S.A. Norton. 2005. Predicting recovery from acidic deposition: Applying a modified TAF (Tracking and Analysis Framework) model to Maine (USA) high-elevation lakes. *Water, Air, & Soil Pollution 164*:383–399.

Lawrence, G.B. 2002. Persistent episodic acidification of streams linked to acid rain effects on soil. *Atmospheric Environment 36*:1589–1598

Lawrence, G.B., M.B. David, and W.C. Shortle. 1995. A new mechanism for calcium loss in forest-floor soils. *Nature 378*:162–165.

Lawrence, G.B., M.B. David, G.M. Lovett, P.S. Murdoch, D.A. Burns, J.L. Stoddard, B.P. Baldigo, J.H. Porter, and A.W. Thompson. 1999. Soil calcium status and the response of stream chemistry to changing acid deposition rates. *Ecological Applications 9*:1059–1072.

Lawrence, G.B., D.A. Goolsby, W.A. Battaglin, and G.J. Stensland. 2000. Atmospheric nitrogen in the Mississippi River basin—emissions, deposition, and transport. *Science of the Total Environment 248*:87–99.

Lawrence, G.B, K.M. Roy, B.P. Baldigo, H.A. Simonin, S.B. Capone, J.W. Sutherland, S.A. Nierzwicki-Bauer, and C.W. Boylen. 2008. Chronic and episodic acidification of Adirondack streams from acid rain in 2003–2005. *Journal of Environmental Quality 37*:2264–2274.

Lehmann, C.M.B., V.C. Bowersox, and S.M. Larson. 2005. Spatial and temporal trends of precipitation chemistry in the United States, 1985–2002. *Environmental Pollution 135*:347–361.

Lenoir, J., J.C. Gegout, P.A. Marquet, P. de Ruffray, and H. Brisse. 2008. A significant shift in plant species optimum elevations during the 20th century. *Science 320*:1768–1771.

Levy, J.I., S.M. Chemerynski, and J.A. Sarnat. 2005. Ozone exposure and mortality: An empiric Bayes metaregression analysis. *Epidemiology 16*:458–68.

Li, H., and S.G. McNulty. 2007. Uncertainty analysis on simple mass balance model to calculate critical loads for soil acidity. *Environmental Pollution 149*:315–326.

Likens, G.E., C.T. Driscoll, and D.C. Buso. 1996. Long-term effects of acid rain: Response and recovery of a forest ecosystem. *Science 272*:244–246.

Long, R.P., S.B. Horsley, R.A. Hallett, and S.W. Bailey. 2009. Sugar maple growth in relation to nutrition and stress in the northeastern United States. *Ecological Applications 19*:1454–1466.

Magnani, F., M. Mencuccini, M. Borghetti, P. Berbigier, F. Berninger, S. Delzon, A. Grelle, P. Hari, P.G. Jarvis, P. Kolari, A.S. Kowalski, H. Lankreijer, B.E. Law, A. Lindroth, D. Loustau, G. Manca, J.B. Moncrieff, M. Rayment, V. Tedeschi, R. Valentini, and J. Grace. 2007. The human footprint in the carbon cycle of temperate and boreal forests. *Nature 447*:848–850.

Mast, M.A., J.T. Turk, D.W. Clow, and D.H. Campbell. 2010. Response of lake chemistry to changes in atmospheric deposition and climate in three high-elevation wilderness areas of Colorado. *Biogeochemistry.* doi:10.1007/s10533-010-9443-4.

McIsaac, G.F., M.B. David, G.Z. Gertner, and D.A. Goolsby. 2001. Eutrophication: Nitrate flux in the Mississippi River. *Nature 414*:166–167.

McKenney, D.W., J.H. Pedlar, K. Lawrence, K. Campbell, and M.F. Hutchinson. 2007. Potential impacts of climate change on the distribution of North American trees. *BioScience 57*:939–948.

McLaughlin, S., and K. Percy. 1999. Forest health in North America: Some perspectives on actual and potential roles of climate and air pollution. *Water, Air, & Soil Pollution 116*:151–197.

McLaughlin, S.B., M.G. Tjoelker, and W.K. Roy. 1993. Acid deposition alters red spruce physiology: Laboratory studies support field observations. *Canadian Journal of Forest Research 23*:380–386.

McNulty, S.G., J. Boggs, J.D. Aber, L. Rustad, and A. Magill. 2005. Red spruce ecosystem level changes following 14 years of chronic N fertilization. Forest *Ecology & Management 219*:279-291.

McNulty, S.G., E.C. Cohen, J.A. Moore Myers, T.J. Sullivan, and H. Li. 2007. Estimates of critical acid loads and exceedances for forest soils across the conterminous United States. *Environmental Pollution 149*:281–292.

Meixner, T., C. Gutmann, R. Bales, A. Leydecker, J. Sickman, J. Melack, and J. McConnell. 2004. Multidecadal hydrochemical response of a Sierra Nevada watershed: Sensitivity to weathering rate and changes in deposition. *Journal of Hydrology 285*:272–285.

Meixner, T., M.E. Fenn, P. Wohlgemuth, M. Oxford, and P. Riggan. 2006. N saturation symptoms in chaparral catchments are not reversed by prescribed fire. *Environmental Science & Technology 40*:2887–2894.

Michalski, G., T. Meixner, M. Fenn, L. Hernandez, A. Sirulnik, E. Allen, and M. Thiemens. 2004. Tracing atmospheric nitrate deposition in a complex semiarid ecosystem using Δ^{17}O. *Environmental Science & Technology 38*:2175–2181.

Mickley, L.J., P.P. Murti, D.J. Jacob, J.A. Logan, D.M. Koch, and D. Rind. 1999. Radiative forcing from tropospheric ozone calculated with a unified chemistry-climate model. *Journal of Geophysical Research 104*:30153–30172.

Milly, P.C.D., K.A. Dunne, and A.V. Vecchia. 2005. Global pattern of trends in streamflow and water availability in a changing climate. *Nature 438*:347–350.

Mohan, J.E., R.M. Cox, and L.R. Inverson. 2009. Composition and carbon dynamics of forests in northeastern North America in a future, warmer world. *Canadian Journal of Forest Research 39*:213–230.

Mohseni, O., H.G. Stefan, and J.G. Eaton. 2003. Global warming and potential changes in fish habitat in U.S. streams. *Climatic Change 59*:389–409.

Monteith, D.T., A.G. Hildrew, R.J. Flower, P.J. Raven, W.R.B. Beaumont, P. Collen, A.M. Kreiser, E.M. Shilland, and J.H. Winterbottom. 2005. Biological responses to the chemical recovery of acidified fresh waters in the UK. *Environmental Pollution 137*:83–101.

Monteith, D.T., J.L. Stoddard, C.D. Evans, H.A. de Wit, M. Forsius, T. Høgåsen, A. Wilander, B.L. Skjelkvåle, D.S. Jeffries, J. Vuorenmaa, B. Keller, J. Kopácek, and J. Vesely. 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. *Nature 450*:537–540.

Moore, M.V., M.L. Pace, J.R. Mather, P.S. Murdoch, R.W. Howarth, C.L. Folt, C.Y. Chen, H.F. Hemond, P.A. Flebbe, and C.T. Driscoll. 1997. Potential effects of climate change on freshwater ecosystems of the New England/Mid-Atlantic region. *Hydrological Processes 11*:925–947.

Morgan, C., and N. Owens. 2001. Benefits of water quality policies: The Chesapeake Bay. *Ecological Economics 39*:271–284.

Mosello, R., A. Lami, A. Marchetto, M. Rogora, B. Wathne, L. Lien, J. Catalan, L. Camarero, M. Ventura, R. Psenner, K. Koinig, H. Thies, S. Sommaruga-Wögrath, U. Nickus, D. Tait, B. Thaler, A. Barbieri, and R. Harriman. 2002. Trends in the water chemistry of high altitude lakes in Europe. *Water, Air, & Soil Pollution: Focus 2*:75–89.

Mote, P.W., A.F. Hamlet, M.P. Clark, and D.P. Lettenmaier. 2005. Declining mountain snowpack in western North America. *Bulletin of the American Meteorological Society 86*:39–49.

Munthe, J., R.A. Bodaly, B.A. Branfireun, C.T. Driscoll, C.C. Gilmour, R. Harris, M. Horvat, M. Lucotte, and O. Malm. 2007. Recovery of mercury-contaminated fisheries. *Ambio 36*:33–44.

Murdoch, P.S., J.S. Baron, and T.L. Miller. 2000. Potential effects of climate change on surface-water quality in North America. *Journal of the American Water Resources Association 36*:347–366.

Murphy, K.J., P. Alpert, and D. Cosentino. 1999. Local impacts of a rural coal-burning generating station on lichen abundance in a New England forest. *Environmental Pollution 105*:349-354.

NADP (National Atmospheric Deposition Program). 2009. *CLAD – Critical Loads of Atmospheric Deposition Science Committee*. Online information. National Atmospheric Deposition Program, Washington, D.C. Available at http://nadp.sws.uiuc.edu/clad/ (accessed August 25, 2009).

Nanus, L., D.H. Campbell, G.P. Ingersoll, D.W. Clow, and M.A. Mast. 2003. Atmospheric deposition maps for the Rocky Mountains. *Atmospheric Environment 37*:4881–4892.

NARSTO. 2011. *Technical Challenges of Risk and Results-Based Multipollutant Air Quality Management*. Edited by G. Hidy, J.R., Brook, K.L., Demerjian, L.T., Molina, W.T., Pennell, and R. Scheffe, Dordrecht, Netherlands: Springer-Verlag.

Nemani, R.R., C.D. Keeling, H. Hashimoto, W.M. Jolly, S.C. Piper, C.J. Tucker, R.B. Myneni, and S.W. Running. 2003. Climate-driven increases in global terrestrial primary production from 1982 to 1999. *Science 300*:1560–1563.

Nilsson, J., and P. Grennfelt (editors). 1988. *Critical Loads for Sulphur and Nitrogen*. UNECE/Nordic Council Workshop Report, Skokloster, Sweden. Nordic Council of Ministers, Copenhagen.

Norby, R.J., and Y. Luo. 2004. Evaluating ecosystem responses to rising atmospheric CO_2 and global warming in a multi-factor world. *New Phytologist 162*:281–293.

Norby, R.J., E.H. DeLucia, B. Gielen, C. Calfapietra, C.P. Giardina, J.S. King, J. Ledford, H.R. McCarthy, D.J.P. Moore, R. Ceulemans, P. De Angelis, A.C. Finzi, D.F. Karnosky, M.E. Kubiske, M. Lukac, K.S. Pregitzer, G.E. Scarascia-Mugnozza, W.H. Schlesinger, and R. Oren. 2005. Forest response to elevated CO_2 is conserved across a broad range of productivity. *Proceedings of the National Academy of Sciences 102*:18052–18056.

Nowak, R.S., D.S. Ellsworth, and S.D. Smith. 2004. Functional responses of plants to elevated CO_2—do photosynthetic and productivity data from FACE experiments support early predictions? *New Phytologist 162*:253–280.

NRC (National Research Council). 2004. *Air Quality Management in the United States*. The National Academies Press, Washington, DC.

NSTC (National Science and Technology Council). 1998. *NAPAP Biennial Report to Congress: An Integrated Assessment*. Executive Office of the President, National Science and Technology Council, National Acid Precipitation Assessment Program, Silver Spring, MD. Available at http://www.p2pays.org/ref/11/10588/.

NSTC (National Science and Technology Council). 2005. *National Acid Precipitation Assessment Program Report to Congress: An Integrated Assessment*. Executive Office of the President, National Science and Technology Council, Washington, DC. Available at http://www.esrl.noaa.gov/csd/aqrs/reports/napapreport05.pdf.

Oltmans, S.J., A.S. Lefohn, J.M. Harris, I. Galbally, H.E. Scheel, G. Bodeker, E. Brunke, H. Claude, D. Tarasick, B.J. Johnson, P. Simmonds, D. Shadwick, K. Anlauf, K. Hayden, F. Schmidlin, T. Fujimoto, K. Akagi, C. Meyer, S. Nichol, J. Davies, A. Redondas, and E. Cuevas. 2006. Long-term changes in tropospheric ozone. *Atmospheric Environment 40*:3156–3173.

Pardo, L.H., and N. Duarte. 2007. *Assessment of Effects of Acidic Deposition on Forested Ecosystems in Great Smoky Mountains National Park Using Critical Loads for Sulfur and Nitrogen*. U.S. Department of Agriculture Forest Service, South Burlington, VT.

Pardo, L.H., M.J. Robin-Abbott, and C.T. Driscoll (editors). 2011. *Assessment of Nitrogen Deposition Effects and Empirical Critical Loads for Nitrogen for Ecoregions of the United States*. General Technical Report NRS-80. U.S. Department of Agriculture, U.S. Forest Service, Newtown Square, PA.

Passy, S., I. Ciugulea, and G.B. Lawrence. 2006. Diatom diversity in chronically versus episodically acidified Adirondack streams. *International Review of Hydrobiology 91*:594–608.

PCAST (President's Council of Advisors on Science and Technology). 2011. *Sustaining Environmental Capital: Protecting Society and the Economy.* Executive Office of the President, Office of Science and Technology Policy, Washington, DC. Available at http://www.whitehouse.gov/sites/default/files/microsites/ostp/pcast_sustaining_environmental_capital_report.pdf

Poor, N., C. Pollman, P. Tate, M. Begum, M. Evans, and S. Campbell. 2006. Nature and magnitude of total inorganic nitrogen and other inorganic species to the Tampa Bay watershed, FL, USA. *Water, Air, & Soil Pollution 170*:267–83.

Pope, C.A., R.T. Burnett, M.J. Thun, E.E. Calle, D. Krewski, K. Ito, and G.D. Thurston. 2002. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *Journal of the American Medical Association 287*:1132–1141.

Porter, E., and S. Johnson. 2007. Translating science into policy: Using ecosystem thresholds to protect resources in Rocky Mountain National Park. *Environmental Pollution 149*:268–280.

Porter, E., T. Blett, D.U. Potter, and C. Huber. 2005. Protecting resources on Federal lands: Implications of critical loads for atmospheric deposition of nitrogen and sulfur. *BioScience 55*:603–612.

Posch, M. 2002. Impacts of climate change on critical loads and their exceedances in Europe. *Environmental Science & Policy 5*:307–317.

Potter, C., S. Klooster, R.R. Nemani, V. Genovese, S. Hiatt, M. Fladeland, and P. Gross. 2006. Estimating carbon budgets for U.S. ecosystems. *Eos, Transactions of the American Geophysical Union 87*:85, 90.

Raddum, G.G., and A. Fjellheim. 2003. Liming of River Audna, southern Norway: A large-scale experiment of benthic invertebrate recovery. *Ambio 32*:230–234.

Reich, P.B., B.A. Hungate, and Y. Luo. 2006. Carbon-nitrogen interactions in terrestrial ecosystems in response to rising atmospheric carbon dioxide. *Annual Review of Ecology, Evolution, & Systematics 37*:611–636

Rice, K.C., F.A. Deviney Jr., G.M. Hornberger, and J.R. Webb. 2006. Predicting the vulnerability of streams to episodic acidification and potential effects on aquatic biota in Shenandoah National Park, Virginia. *U.S. Geological Survey Scientific Investigations Report 2005–5259*, U.S. Geological Survey, Reston, VA.

Robinson, B., T.W. Barnett, G.R. Harwell, S.E. Moore, M. Kulp, and J.S. Schwartz. 2008. pH and acid anion time trends in different elevation ranges in the Great Smoky Mountains National Park. *Journal of Environmental Engineering 134*:800–808.

Rogora, M., R. Mosello, and S. Arisci. 2003. The effect of climate warming on the hydrochemistry of alpine lakes. *Water, Air, & Soil Pollution 148*:347–361.

Rustad, L.E. 2008. The responses of terrestrial ecosystems to global climate change: Towards an integrated approach. *Science of the Total Environment 404*:222–235.

Saros, J.E., T.J. Michel, S.J. Interlandi, and A.P. Wolfe. 2005. Resource requirements of *Asterionella formosa* and *Fragilaria crotonensis* in oligotrophic alpine lakes: Implications for recent phytoplankton community reorganizations. *Canadian Journal of Fisheries & Aquatic Sciences 62*:1681–1689.

Schaefer, S.C., and M. Alber. 2007. Temperature controls a latitudinal gradient in the proportion of watershed nitrogen exported to coastal ecosystems. *Biogeochemistry 85*:333–346.

Schiff, S.L., J. Spoelstra, R.G. Semkin, and D.S. Jeffries. 2005. Drought induced pulses of SO_4^{2-} from a Canadian shield wetland: Use of $\delta^{34}S$ and $\delta^{18}O$ in SO_4^{2-} to determine sources of sulfur. *Applied Geochemistry 20*:691–700.

Schindler, D.W. 1998. A dim future for boreal waters and landscapes: Cumulative effects of climatic warming stratospheric ozone depletion, acid precipitation, and other human activities. *BioScience 48*:157–164.

Schofield, C.L. 1976. Acid precipitation: Effects on fish. *Ambio 5*:228–230.

Schwartz, J. 2005. How sensitive is the association between ozone and daily deaths to control for temperature? *American Journal of Respiratory and Critical Care Medicine 171*:627–31.

Schwede, D.B., R.L. Dennis, and M.A. Bitz. 2009. The watershed deposition tool: A tool for incorporating atmospheric deposition in water-quality analyses. *Journal of the American Water Resources Association 45*:973–985.

Scudder, B.C., L.C. Chasar, D.A. Wentz, N.J. Bauch, M.E. Brigham, P.W. Moran, and D.P. Krabbenhoft. 2009. Mercury in fish, bed sediment, and water from streams across the United States, 1998–2005. *U.S. Geological Survey Scientific Investigations Report 2009–5109*, U.S. Geological Survey, Reston, VA.

Seastedt, T.R., and L. Vaccaro. 2001. Plant species richness, productivity, and nitrogen and phosphorus limitations across a snowpack gradient in alpine tundra, Colorado, U.S.A. *Arctic, Antarctic, & Alpine Research 33*:100–106.

Sickman, J.O., J.M. Melack, and J.L. Stoddard. 2002. Regional analysis of nitrogen yield and retention in high-elevation ecosystems of the Sierra Nevada and Rocky Mountains. *Biogeochemistry 57*:341–374.

Sickman, J.O., A. Leydecker, C.C.Y. Chang, C. Kendall, J.M. Melack, D.M. Lucero, and J. Schimel. 2003. Mechanisms underlying export of N from high-elevation catchments during seasonal transitions. *Biogeochemistry 64*:1–24.

Simonin, H.A., J.R. Colquhoun, E.A. Paul, J. Symula, and H.J. Dean. 2005. Have Adirondack stream fish populations changed in response to decreases in sulfate deposition? *Transactions of the American Fisheries Society 134*:338–345.

Sitch, S., P.M. Cox, W.J. Collins, and C. Huntingford. 2007. Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. *Nature 448*:791–794.

Skeffington, R.A. 2006. Quantifying uncertainty in critical loads: (A) literature review. *Water, Air, & Soil Pollution 169*:3–24.

Snucins, E. 2003. Recolonization of acid-damaged lakes by the benthic invertebrates *Stenacron interpunctatum*, *Stenonema femoratum*, and *Hyalella azteca*. *Ambio 32*:225–229.

Snucins, E., and J.M. Gunn. 2000. Interannual variation in the thermal structure of clear and colored lakes. *Limnology & Oceanography 45*:1647–1654.

Snucins, E., and J. Gunn. 2003. Use of rehabilitation experiments to understand the recovery dynamics of acid-stressed fish populations. *Ambio 32*:240–243.

Sommaruga-Wogarth, S., K.A. Koinig, R. Schmidt, R. Sommaruga, R. Tessadri, and R. Psenner. 1997. Temperature effects on the acidity of alpine lakes. *Nature 387*:64–67.

Sparks, J.P., J. Walker, A. Turnipseed, and A. Guenther. 2008. Dry nitrogen deposition estimates over a forest experiencing free air CO_2 enrichment. *Global Change Biology 14*:768–781.

Spranger, T., J.-P. Hettelingh, J. Slootweg, and M. Posch. 2008. Modelling and mapping long-term risks due to reactive nitrogen effects: An overview of LRTAP convention activities. *Environmental Pollution 154*:482–487.

St. Clair, S.B., W.E. Sharpe, and J.P. Lynch. 2008. Key interactions between nutrient limitation and climatic factors in temperate forests: A synthesis of the sugar maple literature. *Canadian Journal of Forest Research 38*:401–414.

Stephen, K., and V.P. Aneja. 2008. Trends in agricultural ammonia emissions and ammonium concentrations in precipitation over the Southeast and Midwest United States. *Atmospheric Environment 42*:3238–3252.

Stoddard, J.L., J.S. Kahl, F.A. Deviney, D.R. DeWalle, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg, P.S. Murdoch, J.R. Webb, and K.E. Webster. 2003. *Response of Surface Water Chemistry to the Clean Air Act Amendments of 1990*. EPA-620-R-03-001, U.S. Environmental Protection Agency, Washington, DC.

Striegl, R.G., G.R. Aiken, M.M. Dornblaser, P.A. Raymond, and K.P. Wikland. 2005. A decrease in discharge-normalized DOC export by the Yukon River during summer through autumn. *Geophysical Research Letters 32*, L21413, doi: 10.1029/2005GL024413.

Sullivan, T.J., B.J. Cosby, A.T. Herlihy, J.R. Webb, A.J. Bulger, K.U. Snyder, P.F. Brewer, E.H. Gilbert, and D.L. Moore. 2004. Regional model projections of future effects of sulfur and nitrogen deposition on streams in the southern Appalachian Mountains. *Water Resources Research 40*, W02101, doi: 10.1029/2003WR001998.

Sullivan, T.J., I.J. Fernandez, A.T. Herlihy, C.T. Driscoll, T.C. McDonnell, N.A. Nowicki, K.U. Snyder, and J.W. Sutherland. 2006a. Acid-base characteristics of soils in the Adirondack Mountains, New York. *Soil Science Society of America Journal 70*:141–152.

Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger, K.U. Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell, and N.A. Nowicki. 2006b. *Assessment of the Extent to which Intensively-Studied Lakes are Representative of the Adirondack Mountain Region*. Final Report. New York State Energy Research and Development Authority (NYSERDA), Albany, NY. Available at http://nysl.nysed.gov/uhtbin/cgisirsi/ Qcwd6NzFby/NYSL/138650099/8/4298474 (accessed November 1, 2007).

Sullivan T.J., B.J. Cosby, J.R. Webb, R.L. Dennis, A.J. Bulger, and F.A. Deviney, Jr. 2008. Streamwater acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National Park, Virginia. *Environmental Monitoring & Assessment 137*:85–99.

Sutton, M.A., D. Simpson, P.E. Levy, R.I. Smith, S. Ris, M. van Oijen, and W. deVries. 2008. Uncertainties in the relationship between atmospheric nitrogen deposition and carbon sequestration. *Global Change Biology 14*:2057–2063.

Sverdrup, H., and W. deVries. 1994. Calculating critical loads for acidity with the simple mass balance method. *Water, Air, & Soil Pollution 72*:143–162.

Tagaris, E., K.-J. Liao, K. Manomaiphiboon, J.-K. Woo, S. He, P. Amar, and A.G. Russell. 2008. Impacts of future climate change and emissions reductions on nitrogen and sulfur deposition over the United States. *Geophysical Research Letters 35*, L08811, doi:10.1029/2008GL033477.

Tans, P.P., I.Y. Fung, and T. Takahashi. 1990. Observational constraints on the global atmospheric CO_2 budget. *Science 247*:1431–1438.

Thomas, R.Q., C.D. Canham, K.C. Weathers, and C.L. Goodale. 2009. Increased tree carbon storage in response to nitrogen deposition in the US. *Nature Geoscience 3*:13–17.

Tipping, E., J.A.B. Bass, and D. Hardie. 2002. Biological responses to the reversal of acidification in surface waters of the English Lake District. *Environmental Pollution 116*:137–146.

Tipping, E., E.J. Smith, A.J. Lawlor, S. Hughes, and P.A. Stevens. 2003. Predicting the release of metals from ombrotrophic peat due to drought-induced acidification. *Environmental Pollution 123*:239–253.

Todorova, S.G., C.T. Driscoll Jr., D.A. Matthews, S.W. Effler, M.E. Hines, and E.A. Henry. 2009. Evidence for regulation of monomethyl mercury by nitrate in a seasonally stratified, eutrophic lake. *Environmental Science & Technology 43*:6572–6578.

U.S. EPA (Environmental Protection Agency). 1988. *Eastern Lake Survey Phase I*. EPA/600/4-88/032. U.S. Environmental Protection Agency, Office of Research and Development, Washington D.C.

U.S. EPA (Environmental Protection Agency). 2001. *Visibility in Mandatory Federal Class I Areas (1994–1998)*: *A Report to Congress*. EPA-452-R-01-008. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.

U.S. EPA (Environmental Protection Agency). 2003. *Guidance for Tracking Progress Under the Regional Haze Rule*. EPA-454/B-03-004. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC. Available at http://www.epa.gov/ttnamti1/files/ambient/visible/tracking.pdf.

U.S. EPA (Environmental Protection Agency). 2006a. *Acid Rain and Related Programs: 2006 Progress Report*. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. Available at http://www.epa.gov/airmarkets/progress/docs/2006-ARP-Report.pdf.

U.S. EPA (Environmental Protection Agency). 2006b. *Regulatory Impact Analysis for the Review of the Particulate Matter National Ambient Air Quality Standards*. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.

U.S. EPA (Environmental Protection Agency). 2006c. *Technical Support Document for the Proposed PM NAAQS Rule: Response Surface Modeling*. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.

U.S. EPA (Environmental Protection Agency). 2007. *2005/2006 National Listing of Fish Advisories*. U.S. Environmental Protection Agency, Washington, DC.

U.S. EPA (Environmental Protection Agency). 2008a. *BenMAP 3.0 Beta*. Software. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Health and Environmental Impact Division, Air Benefit-Cost Group, Research Triangle Park, NC.

U.S. EPA (Environmental Protection Agency). 2008b. *Integrated Science Assessment for Oxides of Nitrogen and Sulfur Review*. EPA/600/R-08/082F. U.S. Environmental Protection Agency, National Center for Environmental Assessment-RTP Division, Office of Research and Development, Research Triangle Park, NC. Available at http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485.

U.S. EPA (Environmental Protection Agency). 2009a. *Acid Rain and Related Programs: 2007 Progress Report*. EPA-430-K-08-010. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. January. Available at http://www.epa.gov/airmarkt/progress/docs/2007ARPReport.pdf.

U.S. EPA (Environmental Protection Agency). 2009b. *Acid Rain and Related Programs: 2008 Emission, Compliance, and Market Analyses*. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. September. Available at http://www.epa.gov/airmarkt/progress/ARP_2.html.

U.S. EPA (Environmental Protection Agency). 2009c. *Acid Rain and Related Programs: 2008 Emission, Compliance, and Market Data*. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. July. Available at http://www.epa.gov/airmarkt/progress/ARP_1.html.

U.S. EPA (Environmental Protection Agency). 2009d. *Acid Rain and Related Programs: 2008 Highlights*. EPA-430-R-09-031. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. December. Available at http://www.epa.gov/airmarkt/progress/ARP_2008_Highlights.pdf.

U.S. EPA (Environmental Protection Agency). 2009e. I. *NOx Budget Trading Program – Basic Information*. U.S. Environmental Protection Agency, Washington, DC. Available at http://www.epa.gov/airmarkt/progsregs/nox/sipbasic.html.

U.S. EPA (Environmental Protection Agency). 2009f. *Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur*. EPA-452/R-09-008a. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Health and Environmental Impacts Division, Research Triangle Park, NC. Available at http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr_rea.html.

U.S. EPA (Environmental Protection Agency). 2010a. *Acid Rain and Related Programs: 2009 Emissions and Compliance*. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. August. Available at http://www.epa.gov/airmarkets/progress/ARP09_1.html.

U.S. EPA (Environmental Protection Agency). 2010b. *Acid Rain and Related Programs: 2009 Emission, Compliance, and Market Analyses*. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. September. Available at http://www.epa.gov/airmarkets/progress/ARP09_2.html.

U.S. EPA (Environmental Protection Agency). 2010c. *Acid Rain and Related Programs: 2009 Environmental Results*. U.S. Environmental Protection Agency, Office of Air and Radiation, Clean Air Markets Division, Washington, DC. October. Available at http://www.epa.gov/airmarkets/progress/ARP09_3.html.

U.S. EPA (Environmental Protection Agency). 2011. *Benefits and Costs of the Clean Air Act 1990–2020.* Final Report. U.S. Environmental Protection Agency, Office of Air and Radiation, Washington D.C. March. Available at http://www.epa.gov/air/sect812/feb11/fullreport.pdf.

U.S. GAO (Government Accounting Office). 1994. *Allowance Trading Offers an Opportunity to Reduce Emissions at Less Cost*. U.S. General Accounting Office, Washington, DC.

van Groenigen, K.-J., J. Six, B.A. Hungate, M.-A. de Graff, N. van Breemen, and C. van Kessel. 2006. Element interactions limit soil carbon storage. *Proceedings of the National Academy of Sciences 103*:6571–6574.

Vingarzan, R. 2004. A review of surface ozone background levels and trends. *Atmospheric Environment 38*:3431–3442.

Walsh, R.G., R.D. Bjonback, R.A. Aiken, and D.H. Rosenthal. 1990. Estimating the public benefits of protecting forest quality. *Journal of Environmental Management 30*:175–189.

Warby, R.A.F., C.E., Johnson, and C.T. Driscoll. 2005. Chemical recovery of surface waters across the northeastern United States from reduced inputs of acidic deposition: 1984–2001. *Environmental Science & Technology 39*:6548–6554.

Warby, R.A.F., C.E. Johnson, and C.T. Driscoll. 2009. Continuing acidification of organic soils across the northeastern USA: 1984–2001. *Soil Science Society of America Journal 73*:274–284.

Warren, D.R., G.E. Likens, D.C. Buso, and C.E. Kraft. 2008. Status and distribution of fish in an acid-impacted watershed of the Northeastern United States (Hubbard Brook, NH). *Northeast Naturalist 15*:375–390.

Watmough, S.A., J. Aherne, and P.J. Dillon. 2003. Potential impact of forest harvesting on lake chemistry in south-central Ontario at current levels of acid deposition. *Canadian Journal of Fisheries & Aquatic Sciences 60*:1095–1103.

Watmough, S.A., M.C. Eimers, J. Aherne, and P.J. Dillon. 2004. Climate effects on stream nitrate at 16 forested catchments in south central Ontario. *Environmental Science & Technology 38*:2383–2388.

Weathers, K.C., S.M. Simkin, G.M. Lovett, and S.E. Lindberg. 2006. Empirical modeling of atmospheric deposition in mountainous landscapes. *Ecological Applications 16*:1590–1607.

Webb, J.R., B.J. Cosby, F.A. Deviney, J.N. Galloway, S.W. Maben, and A.J. Bulger. 2004. Are brook trout streams in western Virginia and Shenandoah National Park recovering from acidification? *Environmental Science & Technology 38*:4091–4096.

Weiss, S.B. 2006. *Impacts of Nitrogen Deposition on California Ecosystems and Biodiversity*. CEC-500-2005-165. California Energy Commission, PIER Energy-Related Environmental Research, Sacramento, CA.

Wentz, F.J., L. Ricciardulli, K. Hilburn, and C. Mears. 2007. How much more rain will global warming bring? *Science 317*:233–235.

Whitall, D., B. Hendrickson, and H. Paerl. 2003. Importance of atmospherically deposited nitrogen to the annual nitrogen budget of the Neuse River estuary, North Carolina. *Environment International 29*:393–399.

Whitall, D., S. Bricker, J. Ferreira, A.M. Nobre, T. Simas, and M. Silva. 2007. Assessment of eutrophication in estuaries: Pressure-state-response and nitrogen source apportionment. *Environmental Management 40*:678–690.

White, A.F., A.E. Blum, T.D. Bullen, D.V. Vivit, M. Schultz, and J. Fitzpatrick. 1999. The effect of temperature on experimental and natural chemical weathering rates in granitoid rocks. *Geochimica et Cosmochimica Acta 63*:3277–3291.

Wiener, J.G., B.C. Knights, M.B. Sandheinrich, J.D. Jeremiason, M.E. Brigham, D.R. Engstrom, L.G. Woodruff, W.F. Cannon and S.J. Balogh. 2006. Mercury in soils, lakes, and fish in Voyageurs National Park (Minnesota): Importance of atmospheric deposition and ecosystem factors. *Environmental Science & Technology 40*:6261–6281.

Williams, M.W., and K.A. Tonnessen. 2000. Critical loads for inorganic nitrogen deposition in the Colorado Front Range, USA. *Ecological Applications 10*:1648–1665.

Wolfe, A.P., A.C. Van Gorp, and J.S. Baron. 2003. Recent ecological and biogeochemical changes in alpine lakes of Rocky Mountain National Park (Colorado, USA): A response to anthropogenic nitrogen deposition. *Geobiology 1*:153–168.

Woodruff, T.J., J.D. Parker, and K.C. Schoendorf. 2006. Fine particulate matter ($PM_{2.5}$) air pollution and selected causes of postneonatal infant mortality in California. *Environmental Health Perspectives 114*:786–790.

Worall, F., and T.P. Burt. 2007. Flux of dissolved organic carbon from UK rivers. *Global Biogeochemical Cycles 21*, GB1013, doi: 10.1029/2006GB002709

Worrall, F., T. Burt, and R. Shedden. 2003. Long-term records of riverine dissolved organic matter. *Biogeochemistry 64:*165–178.

Wright, R.F., J. Aherne, K. Bishop, L. Camarero, B.J. Cosby, M. Erlandsson, C.D. Evans, M. Forsius, D.W. Hardekopf, R. Helliwell, J. Hruška, A. Jenkins, J. Kopáček, F. Moldan, M. Posch, and M. Rogora. 2006. Modelling the effect of climate change on recovery of acidified freshwaters: Relative sensitivity of individual processes in the MAGIC model. *Science of the Total Environment 365*:154–166.

Wu, W., and C.T. Driscoll. 2010. Impact of climate change on three-dimensional dynamic critical load functions. *Environmental Science & Technology 44*:720–726.

Yan, N.D., W. Keller, N.M. Scully, D.R.S. Lean, and P.J. Dillon. 1996. Increased UV-B penetration in a lake owing to drought-induced acidification. *Water, Air, & Soil Pollution 182*:57-71.

Yan, N.D., B. Leung, W. Keller, S.E. Arnott, J.M. Gunn, and G.G. Raddum. 2003. Developing conceptual frameworks for the recovery of aquatic biota from acidification. *Ambio 32*:165–169.

Zaccherio, M.T., and A.C. Finzi. 2007. Atmospheric deposition may affect northern hardwood forest composition by altering soil nutrient supply. *Ecological Applications 17*:1929–1941.

Zaehle, S., A.D. Friend, P. Friedlingstein, F. Dentener, P. Peylin, and M. Schulz. 2010. Carbon and nitrogen cycle dynamics in the O-CN land surface model: 2. Role of the nitrogen cycle in the historical terrestrial carbon balance. *Global Biogeochemical Cycles 24*:GB1006,doi:10.1029/2009GB003522.

Zhang, X., F.W. Zwiers, G.C. Hegerl, F.H. Lambert, N.P. Gillett, S. Solomon, P.A. Stott, and T. Nozawa. 2007. Detection of human influence on twentieth-century precipitation trends. *Nature 448*:461–465.

To contact the NAPAP organization or to request copies of this report,
contact National Acid Precipitation Assessment Program

USGS
425 Jordan Rd.
Troy, NY 12180

e-mail: daburns@usgs.gov
Telephone: 518-285-5662

Copy of the report is available at: http://ny.water.usgs.gov/