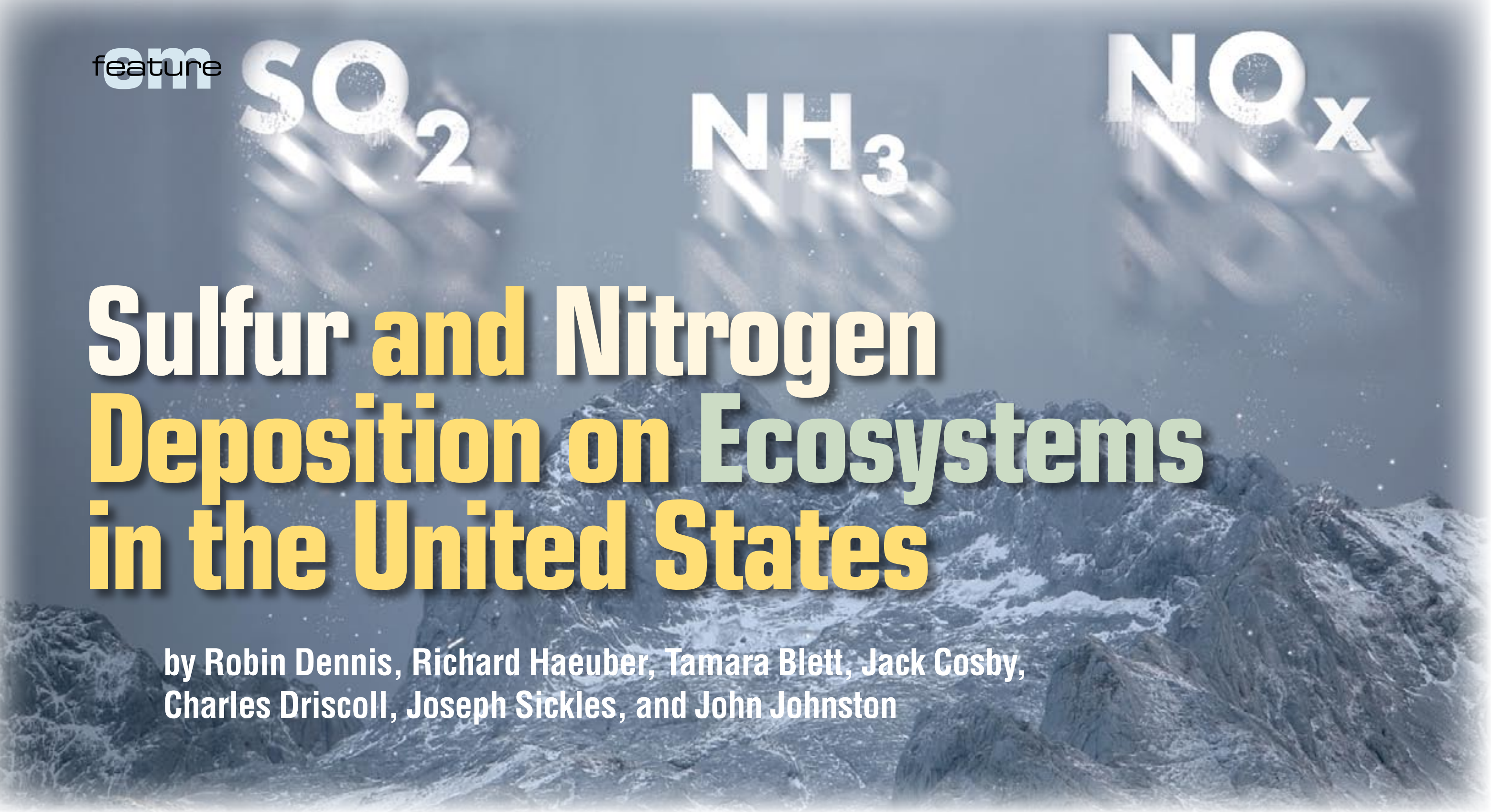

Sulfur and Nitrogen Deposition on Ecosystems in the United States

by Robin Dennis, Richard Haeuber, Tamara Blett, Jack Cosby, Charles Driscoll, Joseph Sickles, and John Johnston

The ecological impacts of atmospheric sulfur and nitrogen deposition first gained attention in the United States in the early 1970s with reports of "acid rain" falling to Earth, causing lakes and streams to become acidic and resulting in conditions that were unsuitable for reproduction and survival of fish in those waters. Many years of research in the United States, Canada, and Europe have since confirmed the link between acidic deposition and ecosystem health. Today, there is a much greater understanding of the complex interactions between sulfur and nitrogen deposition and the natural environment.[1] The impacts of these pollutants are not limited to acid rain alone, but are also related to issues as diverse as elevated ozone concentrations in the lower atmosphere, fish mercury levels, and the trophic status of downstream coastal estuaries.[2,3]

Robin Dennis is with the Atmospheric Sciences Modeling Division, National Oceanic and Atmospheric Administration, in partnership with the U.S. Environmental Protection Agency (EPA); Richard Haeuber, is with EPA's Clean Air Markets Division, Office of Atmospheric Programs; Tamara Blett is with the Air Resources Division, National Park Service; Jack Cosby is with the Department of Environmental Sciences, University of Virginia; Charles Driscoll is with the Department of Civil and Environmental Engineering, Syracuse University; Joseph Sickles is with EPA's Environmental Sciences Division, National Exposure Research Laboratory; and John M. Johnston is with EPA's Ecosystem Research Division, NERL. E-mail: dennis.robin@epa.gov.

> **Many of the largest emissions reductions will be in the eastern United States.**

Atmospheric deposition of sulfur and nitrogen occurs when emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) and the compounds resulting from their chemical reactions in air are deposited in either a wet form (e.g., as rain, snow, and fog) or a dry form (e.g., gases and particles). Prevailing winds can transport the compounds hundreds of kilometers from their source regions, often across state and national borders. Their deposition affects terrestrial, freshwater, and coastal ecosystems in various ways and at different spatial and temporal scales across the country. Documented effects in the United States include

- acidification of lakes and streams with reductions of species diversity and loss of aquatic biota (e.g., fish) (eastern United States);
- acidification of forest soils causing the depletion of available calcium and magnesium and the release of aluminum that is toxic to plant roots and fish (eastern United States);
- nitrogen saturation of watersheds where more nitrogen is deposited than plants can use, causing nitrogen to accumulate in soils, lakes, and streams (eastern and western United States);
- modification of forest chemistry, growth, and regeneration increasing vulnerability to threats such as pests and disease (eastern and western United States);

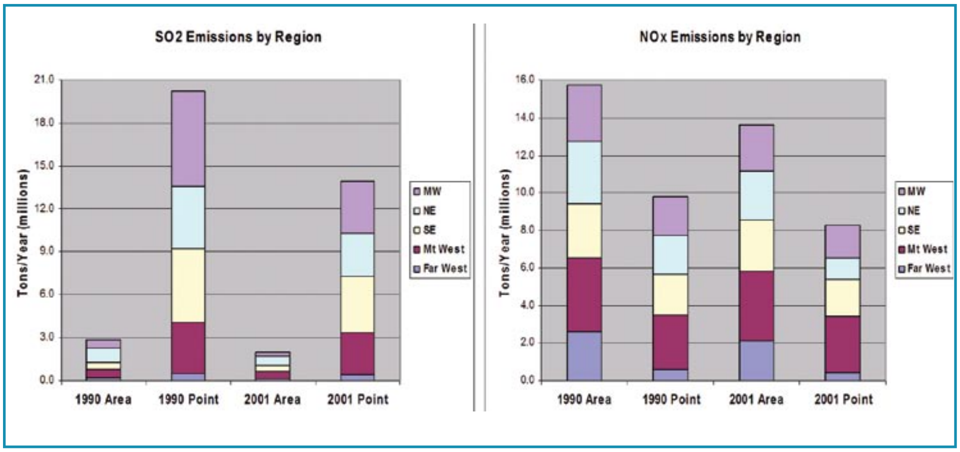

Figure 1. Point and area emissions of SO_2 and NO_x for 1990 and 2001 by region.

Notes: MW = EPA Region 5, NE = EPA Regions 1, 2, and 3, SE = EPA Region 4, Mountain West = EPA Regions 6, 7, and 8, and Far West = EPA Regions 9 and 10.
Source: www.epa.gov/air/data/geosel.html.

- stimulation of species shifts in nutrient poor communities, such as those occurring in some high elevation lakes, alpine tundra, coastal sage communities, serpentine grasslands, and lichen (western United States);
- alteration of species richness and diversity of soil flora (western United States); and
- contribution to the eutrophication of coastal waters, including loss of submerged aquatic vegetation and increased frequency of hypoxic conditions (eastern United States).

Perhaps the most thoroughly investigated effect is the acidification of ecosystems as a result of inputs of strong acid anions (acid deposition). Many studies on acidification and its effects have been conducted in the eastern United States, particularly in the Northeast. Acid deposition is comprised of sulfuric acid, nitric acid, and ammonia resulting from emissions of SO_2, NO_x, and ammonia (NH_3). NH_3 is included under acid deposition because it can be converted to oxidized nitrogen in soils, contributing to the acidification of soil and surface waters. Moreover, both ammonium and nitrate have fertilization effects. Thus, it is actually total nitrogen deposition that results in ecosystem effects.

In the western United States, where atmospheric pollutant loads (particularly sulfur deposition) are lower than in the eastern United States, recent research has documented the effects of excess nitrogen to nutrient-limited ecosystems.[4] The effects of excess nitrogen on ecosystems can begin prior to effects due to acidification. These eutrophication effects include changes in soil and plant chemistry, as well as shifts in species richness and diversity of aquatic and terrestrial ecosystems. As with acidification, it is total nitrogen that contributes to ecosystem impacts, and both ammonium and nitrate play important roles. Because ammonium is taken up preferentially by microorganisms, in some ecosystems it can be an early contributor to eutrophication. Once in terrestrial and aquatic ecosystems, most of the forms of nitrogen are converted to nitrate, which is used by plants, and the excess is flushed into surface waters.

RESPONSIBLE EMISSION SOURCES

SO_2 emissions are largely from point sources, mostly power plants, in the Midwest and Northeast (U.S. Environmental Protection Agency [EPA] Regions 1, 2, 3, and 5), with the Southeast (EPA Region 4) also strongly contributing (see Figure 1). NO_x emissions are from both area (mostly transportation) and point sources, with the proportional contribution from area sources increasing as the contribution from large stationary sources (i.e., power plants) declines. NO_x emissions are widely distributed throughout the United States, but the Far West (EPA Region 9) has relatively smaller point source NO_x emissions. NH_3 emissions are primarily from agricultural sources.

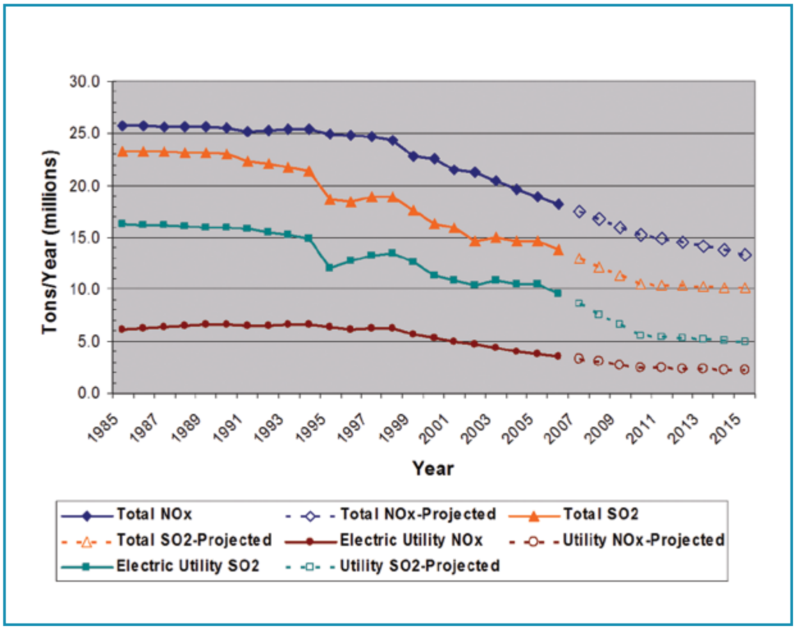

Figure 2. Trends in national and utility-only NO_x and SO_2 emissions, from 1985 to 2006 and projected to 2015.

Notes: There are only very small decreases projected from 2015 to 2020.
Source: EPA National Emissions Inventory for 1985–2006 trends (www.epa.gov/ttn/chief/trends/index.html#tables); EPA for projections to 2020 (www.epa.gov/airmarkets/progsregs/epa-ipm/index.html#results).

ACTIONS TO REDUCE EMISSIONS

Starting with the 1990 Clean Air Act Amendments (CAAA), several regulatory programs have been implemented to control SO_2 and NO_x emission sources, continuing with recent rules set to begin implementation in 2009 and 2010. These emissions reduction programs will help address both human health and ecological impacts (Figure 2 shows past and projected emission trends).

The initial focus of large-scale emissions reduction programs (e.g., Title IV) was to reduce acidic deposition, primarily as a result of SO_2 emissions and sulfur deposition. As knowledge has increased regarding the ecological effects of nitrogen deposition, the ecosystem protection focus has broadened to include both sulfur and nitrogen (see "Major Emissions Reduction Programs").

ACTIONS TO TRACK PROGRESS AND PROGRAM EFFECTIVENESS

A forward-looking system of monitoring was established to track progress, assess ecosystem response to reductions in acid deposition emissions, and inform future policy development, including continuous emissions monitors (CEMs) at the stack for power sector sources; deposition and air quality monitoring through the National Atmospheric Deposition Program (NADP) and Clean Air Status and Trends Network (CASTNET) networks; and long-term surface water quality monitoring to assess the response of lake and stream water chemistry through the Temporally Integrated Monitoring of Ecosystems (TIME) and Long-Term Monitoring (LTM) programs, providing a means of tracking ecological response to changes in emissions.

Fifteen-plus years of monitoring and assessment show that the first three emissions reduction programs listed (and other programs) have been an effective and efficient means of meeting the emissions reduction goals set under the Clean Air Act. As of 2006, these programs have achieved substantial reductions in SO_2 emissions and NO_x emissions from 1990 levels (see Figure 2) and monitoring efforts show clear signs of environmental response.

MAJOR EMISSIONS REDUCTION PROGRAMS

- **Title IV (Acid Rain Program), 1990 CAAA** — designed to mitigate impacts of acid deposition across the eastern United States by controlling SO_2 and NO_x emissions from power generation sources.[5]
- **NO_x Budget Trading Program (NBP)** — initiated in 2003 to implement the 1998 NO_x State Implementation Plan Call, the NBP is focused on helping eastern states attain the National Ambient Air Quality Standards (NAAQS) for ozone.[6]
- **Mobile Source Programs** — addressing on-road and nonroad engines, as well as fuel content.
- **Clean Air Interstate Rule (CAIR)** — a March 2005 regulation mandating additional reductions in power generation SO_2 and NO_x emissions across the eastern United States to help attain the ozone and fine particulate matter ($PM_{2.5}$) NAAQS.

Deposition Reductions

Wet deposition is fairly well characterized by monitoring at approximately 270 NADP sites in the United States (http://nadp.sws.uiuc.edu). Substantial decreases in sulfate wet deposition have been measured (see Figure 3). Dry deposition is tracked through CASTNET, with 87 sites in the United States (www.epa.gov/castnet). Dry sulfur deposition declined significantly between 1990 and 2005 at 34 eastern CASTNET reference sites (see Figure 4).

Ecosystem Response

EPA currently funds two surface water programs as part of the Environmental Monitoring and Assessment Program: the LTM and the TIME programs. Together, these two monitoring programs (sometimes referred to as TIME/LTM) provide important information on changes in water chemistry in

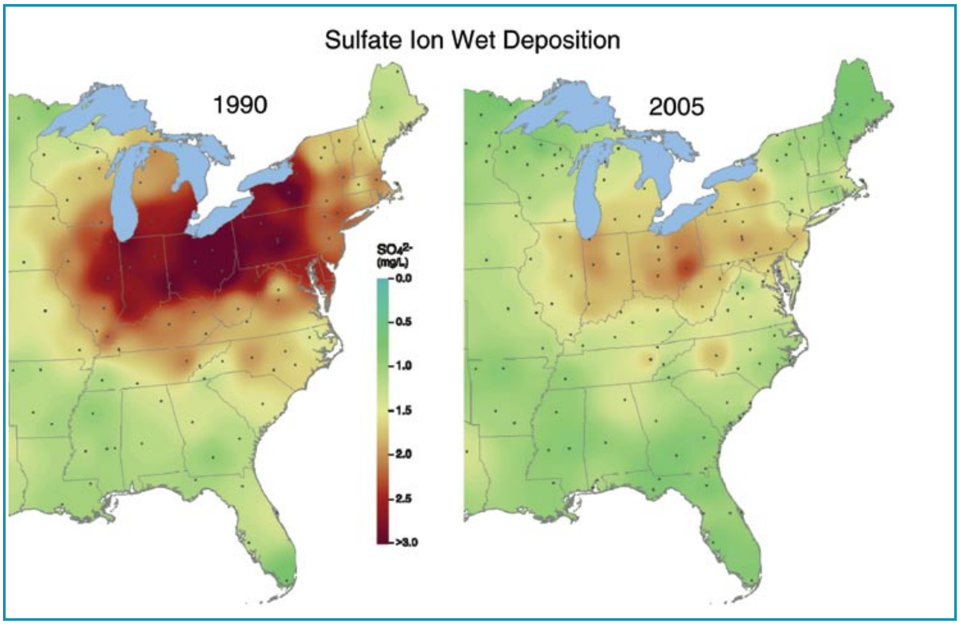

Figure 3. Three-year averages of sulfate wet deposition centered on 1990 and 2002, based on NADP/NTN wet deposition data.[5]

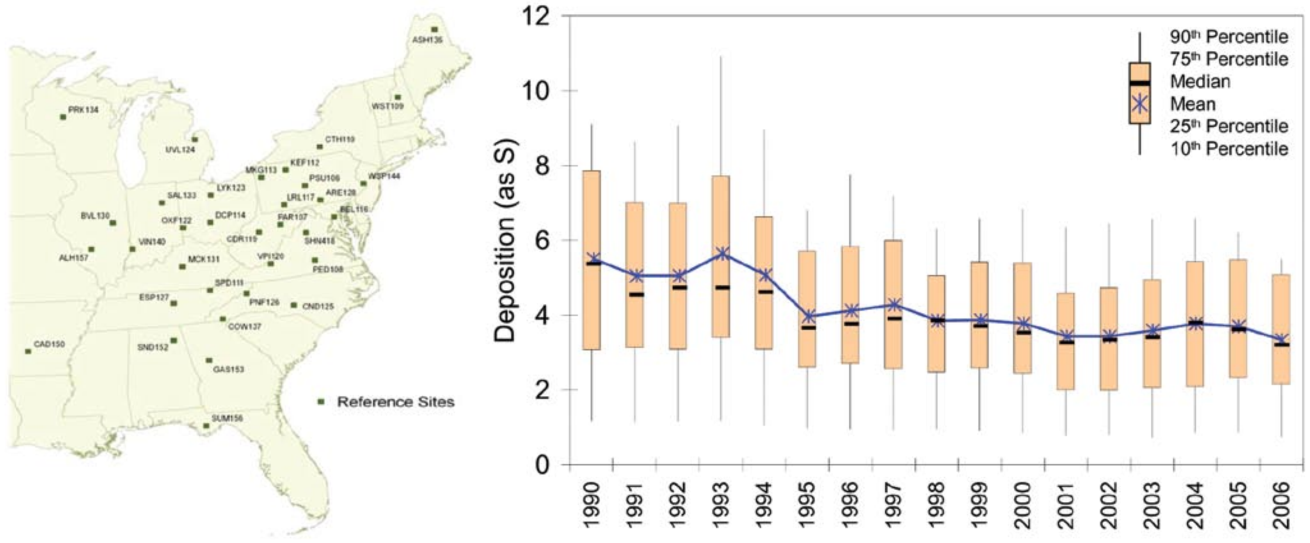

Figure 4. Trends in dry sulfur deposition (as S; kg/ha/yr) at 34 CASTNET eastern reference sites.[16]

response to the deposition of air pollutants (see Figure 5). The TIME/LTM sites constitute the most geographically extensive network tracking changes in water chemistry to assess whether sensitive ecosystems are recovering from decreases in acidic deposition.

Ecological response to emissions reductions is a long-term, incremental process that occurs in stages.[1] In the case of acid rain, decreases in acidic deposition facilitate an initial phase of chemical recovery. Recovery times vary widely across ecosystems. The potential for the second phase of ecosystem recovery—biological recovery—is greatly enhanced as chemical conditions in lakes and streams improve. Quantification of the biological recovery phase, however, is monitored poorly and consequently not well understood.[1,7]

It is important to note that little is known about tree response to changes in acid deposition.

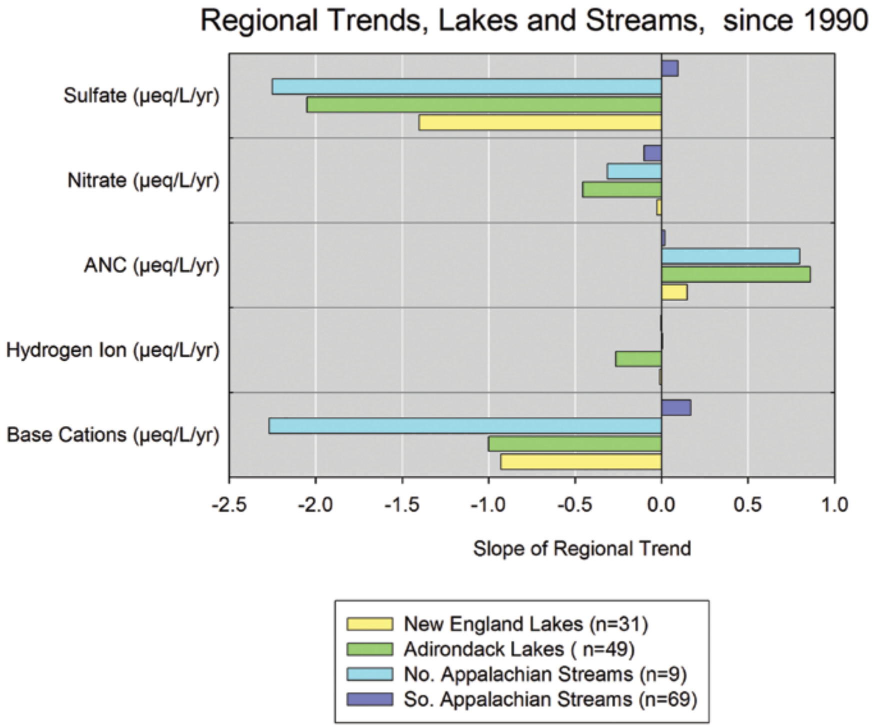

Figure 5. Regional trends in lake- and stream-water chemistry to show degree of recovery in the eastern United States.[5]

ECOLOGICAL RESPONSE IN THE EAST

Rigorous monitoring and program assessment of chemical response indicators have demonstrated program effectiveness and provided important scientific insights regarding ecological response and recovery in some areas in the northeastern United States. Monitoring data from the TIME/LTM programs show a mix of surface water chemistry response as a result of CAA emissions reduction programs (Figure 5).[8] Signs of recovery in New England lakes, Adirondack Mountain lakes, and Northern Appalachian streams include reductions in sulfate and aluminum concentrations, as well as decreases in acidity. In Southern Appalachian streams, sulfur deposition has accumulated in forest soils and the slow release of these stored elements has delayed the recovery of streams, even though emissions and deposition have decreased.[5,8,9]

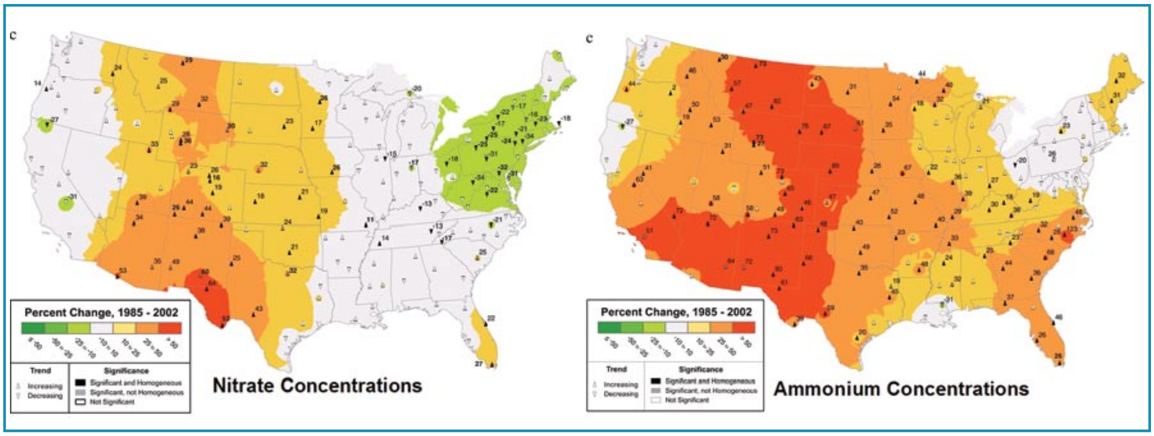

Figure 6. National, linear trends in wet concentrations of nitrate and ammonium for data at NADP/NTN sites, from 1985 to 2002.[17]

Decreases in SO_2 and NO_x emissions, and consequent reductions in acidic deposition, are consistent with the trends originally expected under 1990 CAAA programs. The Title IV Acid Rain Program, including a cap-and-trade mechanism, has been effective in achieving emissions reductions. Still, ecosystems are responding slowly, with recovery more evident in some areas than others. Thus, the emissions reduction levels mandated under the 1990 CAAA may not have been sufficient to allow full recovery of affected ecosystems, or sufficient ecosystem protection in the future.

Models are key research tools for synthesizing the complexity of the various processes involved in acidic deposition. Atmospheric models like the Community Multiscale Air Quality (CMAQ) model are used to simulate transport, chemical transformations, and deposition of air pollutants at a regional or national scale in response to changes in emissions. Biogeochemical models such as MAGIC and PnET-BGC are used to evaluate ecosystem response to historical changes and future scenarios of atmospheric deposition. Model calculations suggest that substantial additional reductions in SO_2 and NO_x emissions are needed to facilitate additional chemical recovery, leading to additional biological recovery. Recent studies conclude that recovery of acid-sensitive ecosystems will require 40–80% further reductions in utility SO_2 emissions beyond those anticipated with full implementation of Title IV of the 1990 CAAA.[1,10]

Efforts to assist states in the eastern United States in attaining the health-based NAAQS for ozone and $PM_{2.5}$ resulted in recent rules, including CAIR, that will achieve further SO_2 and NO_x emissions reductions. While not specifically designed to address ecological impacts, these emissions reductions will contribute to significant additional ecological recovery. As shown in Figure 2, a 50% reduction in SO_2 emissions beyond the Title IV program is anticipated from CAIR implementation in the eastern United States. However, because chemical and biological processes mediating the sulfur and nitrogen cycles respond over decadal and longer time spans, even these further emissions reductions might not result in full chemical and biological recovery by 2050.[10] For nutrient deposition, the anticipated reductions in NO_x emissions, coupled with anticipated increases in NH_3 emissions, could result in total nitrogen deposition being dominated by total ammonia deposition by 2020.

ECOLOGICAL RESPONSE IN THE WEST

While ecosystems are responding to decreasing emissions and deposition in the eastern United States, the situation may be somewhat different in the western United States. Although levels of atmospheric nitrogen deposition are low in the West compared to the East, some areas downwind of large metropolitan centers, large point sources, or large agricultural operations have elevated nitrogen deposition levels.[4] In addition, many western ecosystems are sensitive to elevated nitrogen deposition because they evolved under low-nitrogen conditions. Over the past 20 years, nitrate wet deposition and concentrations have been increasing across much of the Western United States (see Figure 6, left panel) and ammonium deposition is increasing at an even faster rate (see Figure 6, right panel). Emissions sources are changing as well. For example, oil and gas production has been increasing rapidly across the western United States. Emissions may increase at a higher rate if current production leases and coal-fired power plants awaiting permits (new sources) are developed. Finally, NH_3 emissions from agriculture (both crops and livestock) may be significant contributors to nitrogen deposition in the West.

Various studies in western North America demonstrate that some aquatic and terrestrial plant and microbial communities may be altered significantly by nitrogen deposition.[4,11] For example, recent evidence has led some researchers to conclude that high-altitude watersheds in the Colorado Front Range show symptoms of ecological impacts at relatively low nitrogen deposition levels occurring currently[12,13] and even as far back as the 1950 and 1960s.[14] Farther West, levels of nitrate in streams and groundwater in the San Gabriel and San Bernardino Mountains have been found to be linked strongly to nitrogen deposition. Nitrate concentrations in streams of Devil's Canyon in the San Bernardino Mountains and in chaparral watersheds with high smog exposure in the San Gabriel Mountains are the highest in North America for forested watersheds.[4] Various other studies in the western United States have documented nitrogen-related species shifts in desert grass species, coastal sage communities, soil biota, and lichens.[4] Ecosystem changes induced by nitrogen deposition in the

FUTURE NEEDS

Monitoring

- **Emissions.** Continue CEMs on utilities; better quantify mobile source NO_x; improve NH_3 emissions monitoring and tracking.
- **Deposition.** Continue NADP; expand CASTNET to include monitoring of NH_3 and NO_x (fill major N-deposition budget gaps); improve spatial coverage.
- **Water Chemistry.** Improve spatial and temporal monitoring of water chemistry.
- **Soils.** Develop better soil monitoring protocols and link data to stream and forest health.
- **Biotic Responses.** Monitor response to changing sulfur and nitrogen deposition.

Modeling

- Enhance model capabilities regarding terrestrial and aquatic ecosystem responses.
- Advance atmospheric models to consider bi-directional exchange of NH_3.
- Create continuous deposition time series by combining model outputs and observations.
- Link atmospheric emissions-transport-deposition models (e.g., CMAQ) with watershed transport-effects models (e.g., MAGIC, PnET-BGC).
- Identify critical thresholds and discontinuities in biological dynamics and system function to inputs of sulfur and nitrogen deposition for use in forecasting.

Decision Support

- Develop common metrics or approaches to determine whether programs are protective of sensitive biological systems, at local to regional scales, and provide indicators for broad tracking and assessment (e.g., critical loads).
- Develop modeling approaches to address multiple stressors and receptors across multiple scales.

western United States suggest that reductions in nitrogen deposition are necessary before additional impacts occur that may be difficult to reverse.

FUTURE DIRECTIONS AND NEEDS

Clean Air Act programs have achieved significant reductions of SO_2 and NO_x emissions, decreasing deposition of ecologically harmful pollutants in many areas. The forthcoming implementation of mobile source regulations and CAIR will result in substantial additional reductions of SO_2 and NO_x emissions, providing further potential for ecosystem recovery and future protection. As described above, ecosystems are responding slowly, with recovery more evident in some areas than others. Moreover, many of the largest emissions reductions will be in the eastern United States. As studies described above indicate, the consequent emissions levels due to these various regulatory programs may not be sufficient to allow full recovery of affected ecosystems in all areas of the country or to provide sufficient ecosystem protection in the future.

Given current ecological impacts of sulfur and nitrogen deposition, and the potential of continued impacts in the foreseeable future, it is important to maintain and enhance our capacity to track and assess program implementation and effectiveness (see "Future Needs"). Recent judgments of expert bodies, such as the National Research Council, emphasize the importance of program accountability as essential for an effective air quality management system in the United States.[15] Preserving and strengthening the chain of accountability is necessary if we are to determine whether ecosystems are recovering from past injury and ensure they will be sufficiently protected in the future through informed policy development and program implementation. **em**

REFERENCES

1. Driscoll, C.T.; Lawrence, G.B.; Bulger, A.J.; Butler, T.J.; Cronan, C.S.; Eagar, C.; Lambert, K.F.; Likens, G.E.; Stoddard, J.L.; Weathers, K.C. Acidic Deposition in the Northeastern United States: Sources and Inputs, Ecosystem Effects, and Management Strategies; *BioScience* **2001**, *51* (3), 180-198.
2. Driscoll, C.T.; Whitall, D.; Aber, J.; Boyer, E.; Castro, M.; Cronan, C.; Goodale, C.L.; Groffman, P.; Hopkinson, C.; Lambert, K.; Lawrence, G.; Ollinger, S. Nitrogen Pollution in the Northeastern United States: Sources, Effects, and Management Options; *BioScience* **2003**, *53* (4), 357-374.
3. Driscoll, C.T.; Han, Y.-J.; Chen, C.Y.; Evers, D.C.; Lambert, K.F.; Holsen, T.M.; Kamman, N.C.; Munson, R.K. Mercury Contamination in Forest and Freshwater Ecosystems in the Northeastern United States; *BioScience* **2007**, *57* (1), 17-28.
4. Fenn, M.E.; Baron, J.S.; Allen, E.B.; Rueth, H.M.; Nydick, K.R.; Geiser, L.; Bowman, W.D.; Sickman, J.O.; Meixner, T.; Johnson, D.W.; Neitlich, P. Ecological Effects of Nitrogen Deposition in the Western United States; *BioScience* **2003**, *53* (4), 404-420.
5. *Acid Rain Program 2006 Progress Report*; EPA-430-R-07-009; Office of Air and Radiation, U.S. Environmental Protection Agency, Washington, DC, 2007a.
6. *NO_x Budget Trading Program: 2007 Program Compliance and Environmental Results*; EPA-430-R-07-011; Office of Air and Radiation, U.S. Environmental Protection Agency, Washington, DC, 2007b.
7. Baldigo, B.P.; Lawrence, G.B. Effects of Stream Acidification and Habitat on Fish Populations of a North American River; *Aquatic Sciences* **2001**, *63*, 196-222.
8. Kahl, J.S.; Stoddard, J.L.; Haeuber, R.; Paulsen, S.G.; Birnbaum, R.; Deviney, F.A.; Webb, J.R.; DeWalle, D.R.; Sharpe, W.; Driscoll, C.T.; Herlihy, A.; Kellogg, J.H.; Murdoch, P.S.; Roy, K.; Webster, K.E.; Urquhart, N.S. Have U.S. Surface Waters Responded to the 1990 Clean Air Act Amendments?; *Environ. Sci. Technol.* **2004**, *38* (24), 484A-490A.
9. Webb, J.R.; Cosby, B.J.; Deviney, F.A., Jr.; Galloway, J.N.; Maben, S.W.; Bulger, A.J. Are Brook Trout Streams in Western Virginia and Shenandoah National Park Recovering from Acidification?; *Environ. Sci. Technol.* **2004**, *38* (15), 4091-4096.
10. *National Acid Precipitation Assessment Report to Congress: An Integrated Assessment*; Executive Office of the President, National Science and Technology Council, Washington, DC, 2005.
11. Fenn, M.E.; Poth, M.A.; Aber, J.D.; Baron, J.S.; Bormann, B.T.; Johnson, D.W.; Lemly, A.D.; McNulty, S.G.; Ryan, D.F.; Stottlemyer, R. Nitrogen Excess in North American Ecosystems: Predisposing Factors, Ecosystem Responses, and Management Strategies; *Ecolog. Appl.* **1998**, *8* (3), 706-733.
12. Baron, J.S.; Rueth, H.M.; Wolfe, A.M.; Nydick, K.R.; Allstott, E.J.; Minear, J.T.; Moraska, B. Ecosystem Responses to Nitrogen Deposition in the Colorado Front Range; *Ecosystems* **2000**, *3*, 352-368.
13. Bowman, W.D.; Gartner, J.R.; Holland, K.; Wiedermann, M. Nitrogen Critical Loads for Alpine Vegetation and Terrestrial Ecosystem Response: Are We There Yet?; *Ecolog. Appl.* **2006**, *16* (3), 1183-1192.
14. Baron, J.S. Hindcasting Nitrogen Deposition to Determine an Ecological Critical Load; *Ecolog. Appl.* **2006**, *16* (2), 433-439.
15. *Air Quality Management in the United States*; National Research Council, The National Academies Press, Washington, DC, 2004.
16. *Clean Air Status and Trends Network (CASTNET): 2005 Annual Report*; Office of Air and Radiation, U.S. Environmental Protection Agency, Washington, DC, 2006.
17. Figure first published in Lehmann, C.M.B., Bowersox, V.C.; Larson, S.M. Spatial and Temporal Trends of Precipitation Chemistry in the United States, 1985–2002; *Environ. Pollut.* **2005**, *135* (3), 347-361.