

Acid Rain and Related Programs: 2008 Environmental Results



The Acid Rain Program (ARP), established under Title IV of the 1990 Clean Air Act Amendments, requires major emission reductions of sulfur dioxide (SO₂) and nitrogen oxide (NO_x), the primary precursors of acid rain, from the electric power industry. The SO₂ program sets a permanent cap on the total amount of SO₂ that may be emitted by electric generating units (EGUs) in the contiguous United States, and includes provisions for trading and banking allowances. The program is phased in, with the final 2010 SO₂ cap set at 8.95 million tons, a level of about one-half of the emissions from the power sector in 1980. NO_x reductions under the ARP are achieved through a program that applies to a subset of coal-fired EGUs and is closer to a more traditional, rate-based regulatory system.

The emission reductions achieved under the ARP have led to important environmental and public health benefits. These include improvements in air quality with significant benefits to human health; reductions in acid deposition; the beginnings of recovery from acidification in fresh water lakes and streams; improvements in visibility; and reduced risk to forests, materials, and structures. Table 1 on page 2 shows the regional changes in key air quality and atmospheric deposition measurements linked to the ARP's SO₂ and NO_x emission reductions.

During 2009, EPA is releasing a series of reports summarizing progress under the ARP. This third report compares changes in emissions to changes in air quality, acid deposition, and surface water chemistry. For more information on the ARP, please visit: <http://www.epa.gov/airmarkets/progsregs/arp/index.html>.

Air Quality Sulfur Dioxide

Data collected from monitoring networks show that the decline in SO₂ emissions from the power industry has improved air quality. Based on EPA's latest air emission trends data located at www.epa.gov/airtrends/index.html, the national composite average of SO₂ annual mean ambient concentrations decreased 71 percent between 1980 and 2008, as shown in Figure 1 on page 3 (based on

At a Glance: ARP Results in 2008

Air Quality: Emission reductions achieved under the ARP have led to improvements in air quality with significant benefits to human health.

- Between 1989-1991 and 2006-2008 average ambient sulfate concentrations have decreased by 38 percent in the Mid-Atlantic, 44 percent in the Midwest, 43 percent in the Northeast, and 28 percent in the Southeast.

Acid Deposition: Monitoring data show significant improvements in the primary acid deposition indicator.

- Between the 1989 to 1991 and 2006 to 2008 observation periods, average decreases in wet deposition of sulfate averaged more than 30 percent for the eastern United States.
- Reduction of total sulfur deposition (wet plus dry deposition) during the 1989 to 1991 and 2006 to 2008 observation periods has been even more dramatic, with average reductions of about 40 percent.

Surface Water Chemistry: Long-term surface water monitoring programs indicate trends toward recovery from acidification.

- Levels of Acid Neutralizing Capacity (ANC), the ability of a water body to neutralize acid deposition, have increased significantly from 1990 to 2007 in lake and stream long-term monitoring sites in New England, the Adirondack Mountains, and the Northern Appalachian Plateau.
- Although water quality has improved, many lakes and streams still have acidic conditions harmful to their biota and further emission reductions are needed for full ecosystem protection and recovery of sensitive aquatic systems.

state, local, and EPA monitoring sites located primarily in urban areas). The largest single-year reduction (20 percent) occurred in the first year of the ARP, between 1994 and 1995. These trends are consistent with the regional ambient air quality trends observed in the Clean Air Status and Trends Network (CASTNET).

Acid Rain and Related Programs: 2008 Environmental Results

Table 1: Regional Changes in Air Quality and Deposition of Sulfur and Nitrogen Compounds, 1989-1991 versus 2006-2008, from Rural Monitoring Networks

Measurement	Region	Average, 1989-1991	Average, 2006-2008	Percent Change	Number of Sites
Ambient SO ₂ Concentration (µg/m ³)	Mid-Atlantic	13	6	-54	12
	Midwest	11	5	-55	10
	Northeast	5.5	2.1	-62	3
	Southeast	5.3	2.9	-45	9
Ambient Sulfate Concentration (µg/m ³)	Mid-Atlantic	6.4	4	-38	12
	Midwest	5.9	3.3	-44	10
	Northeast	3.5	2	-43	3
	Southeast	5.3	3.8	-28	9
Wet Sulfate Deposition (kg-S/ha)	Mid-Atlantic	9.2	6.3	-32	11
	Midwest	7.1	4.5	-37	27
	Northeast	7.5	5	-33	17
	Southeast	6.1	3.9	-36	23
Dry Sulfur Deposition (kg-S/ha)	Mid-Atlantic	6.3	3.3	-48	8
	Midwest	7	3.4	-51	9
	Northeast	4	1.5	-63	2
	Southeast	1.2	0.8	-33	2
Total Sulfur Deposition (kg-S/ha)	Mid-Atlantic	16	10	-38	8
	Midwest	16	9	-44	9
	Northeast	11	6	-45	2
	Southeast	8	5.3	-34	2
Total Ambient Nitrate Concentration (Nitrate + Nitric Acid) (µg/m ³)	Mid-Atlantic	3.2	2.2	-31	12
	Midwest	4.6	3.3	-28	10
	Northeast	1.7	1	-41	3
	Southeast	2.2	1.7	-23	9
Wet Inorganic Nitrogen Deposition (kg-N/ha)	Mid-Atlantic	6.2	4.9	-21	11
	Midwest	5.8	5.2	-10	27
	Northeast	5.6	4.4	-21	17
	Southeast	4.4	3.5	-20	23
Dry Inorganic Nitrogen Deposition (kg-N/ha)	Mid-Atlantic	2.4	1.6	-33	9
	Midwest	2.7	2	-26	9
	Northeast	1.8	0.9	-50	2
	Southeast	0.88	0.96	9	2
Total Inorganic Nitrogen Deposition (kg-N/ha)	Mid-Atlantic	8.5	6.4	-25	9
	Midwest	9.3	7.5	-19	9
	Northeast	6.6	4.8	-27	2
	Southeast	5.9	4.9	-17	2

Notes:

- Averages are the arithmetic mean of all sites in a region that were present and met the completeness criteria in both averaging periods. Thus, average concentrations for 1989-1991 may differ from past reports.
- Total deposition is estimated from raw measurement data, not rounded, and may not equal the sum of dry and wet deposition.
- Percent change in bold indicates that differences were statistically significant at the 95 percent confidence level. Changes that are not statistically significant may be unduly influenced by measurements at only a few locations or large variability in measurements.

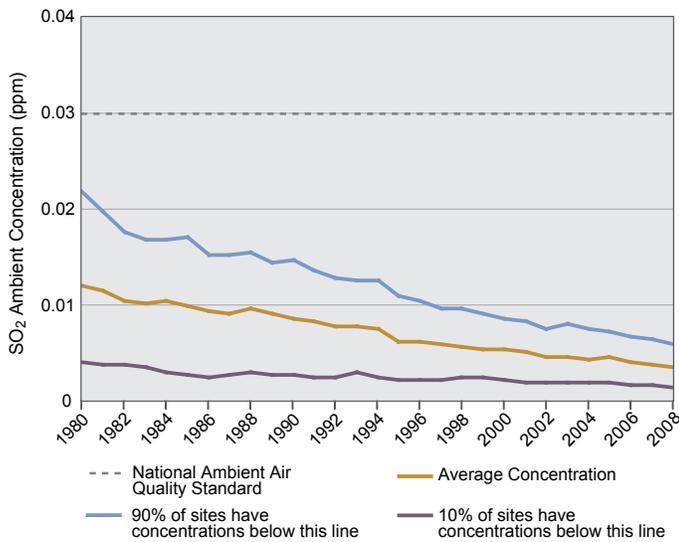
Source: EPA, 2009

During the late 1990s, following implementation of Phase I of the ARP, dramatic regional improvements in SO₂ and ambient sulfate concentrations were observed at CASTNET sites throughout the eastern United States, and these improvements continue today. Analyses of regional monitoring data from CASTNET show the geographic pattern of SO₂ and airborne sulfate in the eastern United States. Three-year mean annual concentrations of SO₂ and sulfate from CASTNET long-term monitoring sites are compared from 1989 to 1991 and 2006 to 2008 in both tabular form and graphically in maps (see Table 1 on page 2, Figure 2 on page 3, and Figure 3 on page 4).

The maps in Figure 2 show that from 1989 to 1991, prior to implementation of Phase I of the ARP, the highest annual ambient concentrations of SO₂ in the East were observed in western Pennsylvania and along the Ohio River Valley. The maps indicate a significant decline in those concentrations in nearly all affected areas after implementation of the ARP and other programs.

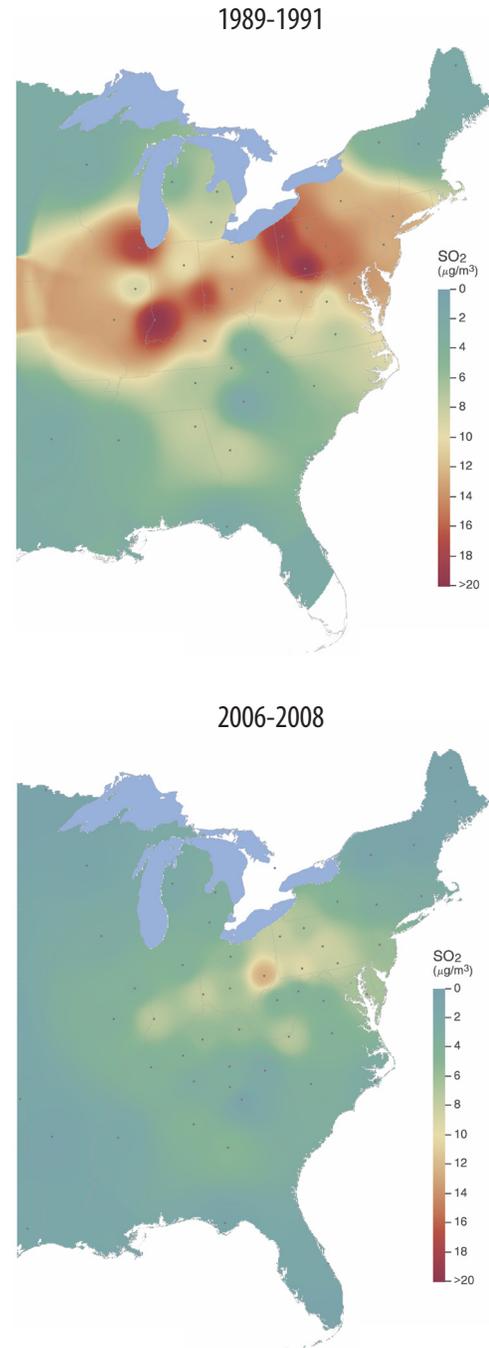
Before the ARP, in 1989-1991, the highest annual ambient sulfate concentrations were observed in western Pennsylvania, along the Ohio River Valley, and in northern Alabama at levels greater than 11 micrograms per cubic meter (µg/m³). Most of the eastern United States experienced annual ambient sulfate concentrations greater than 5 µg/m³.

Figure 1: National SO₂ Air Quality, 1980-2008



Source: EPA, 2009

Figure 2: Annual Mean Ambient SO₂ Concentration



Notes:

- For maps depicting these trends for the entire continental United States, visit <www.epa.gov/castnet>.
- Dots on all maps represent monitoring sites. Lack of shading for southern Florida indicates lack of monitoring coverage in the 1989-1991 period.

Source: CASTNET, 2009

Figure 3: Annual Mean Ambient Sulfate Concentration

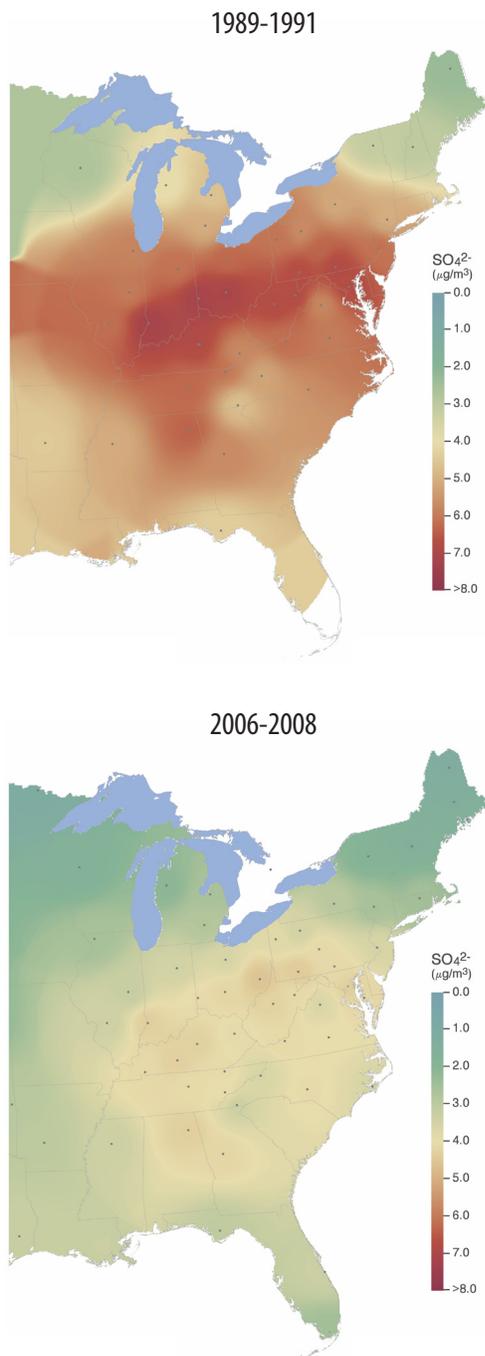
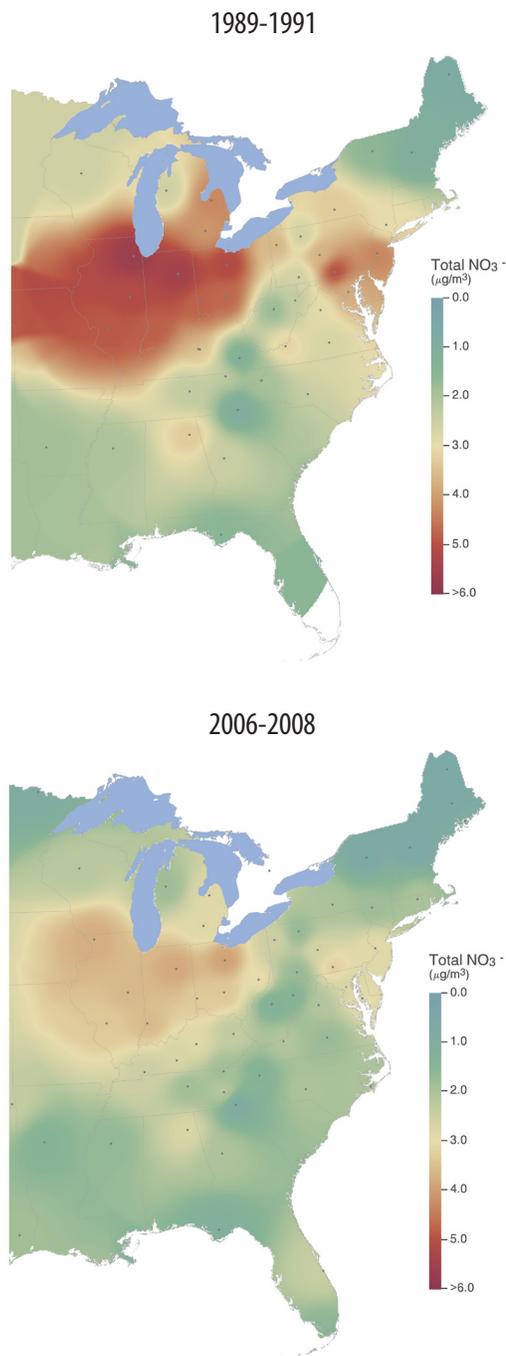


Figure 4: Annual Mean Ambient Total Nitrate Concentration



Notes:

- For maps depicting these trends for the entire continental United States, visit <www.epa.gov/castnet>.
- Dots on all maps represent monitoring sites. Lack of shading for southern Florida indicates lack of monitoring coverage in the 1989-1991 period.

Source: CASTNET, 2009

Like SO₂ concentrations, ambient sulfate concentrations have decreased since the program was implemented, with average concentrations decreasing from 28 to 44 percent in regions of the East (see Table 1 on page 2). Both the magnitude and spatial extent of the highest concentrations have dramatically declined, with the largest decreases observed along the Ohio River Valley (see Figure 3 on page 4).

Nitrogen Oxides

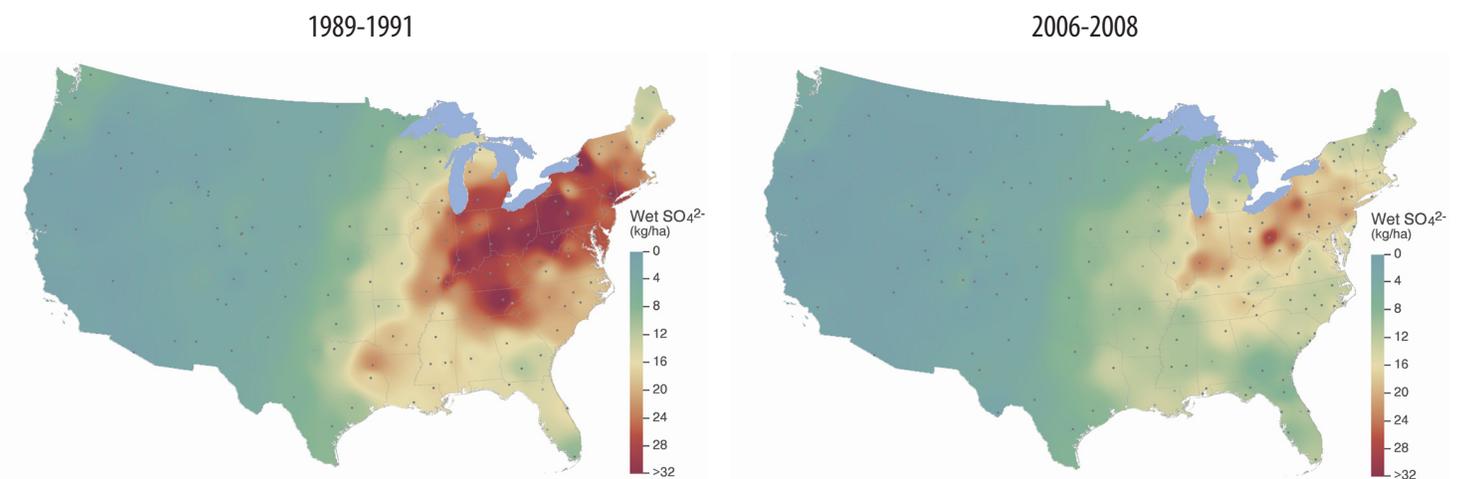
Although the ARP has met its NO_x emission reduction targets, emissions from other sources (such as motor vehicles and agriculture) contribute to ambient nitrate concentrations in many areas. NO_x levels can also be affected by emissions transported via air currents over wide regions.

From 2006 to 2008, reductions in NO_x emissions during the ozone season from power plants under the NO_x SIP Call have continued to result in significant region-specific improvements in ambient total nitrate (NO₃⁻ plus HNO₃) concentrations. For instance, annual mean ambient total nitrate concentrations for 2006 to 2008 in the Mid-Atlantic region were 31 percent less than the annual mean concentration in 1989 to 1991 (see Table 1 on page 2 and Figure 4 on page 4). While these improvements might be partly attributed to added NO_x controls installed for compliance with the NO_x SIP Call, the findings at this time are not conclusive.

Acid Deposition

National Atmospheric Deposition Program/National Trends Network (NADP/NTN) monitoring data show significant improvements in the primary acid deposition indicators. For example, wet sulfate deposition (sulfate that falls to the earth through rain, snow, and fog) has decreased since the implementation of the ARP in much of the Ohio River Valley and northeastern United States. Some of the greatest reductions have occurred in the mid-Appalachian region, including Maryland, New York, West Virginia, Virginia, and most of Pennsylvania. Other less dramatic reductions have been observed across much of New England, portions of the southern Appalachian Mountains, and some areas of the Midwest. Between the 1989 to 1991 and 2006 to 2008 observation periods, average decreases in wet deposition of sulfate averaged more than 30 percent for the eastern United States (see Table 1 on page 2 and Figure 5). Along with wet sulfate deposition, wet sulfate concentrations have also decreased by similar percentages. A strong correlation between large-scale SO₂ emission reductions and large reductions in sulfate concentrations in precipitation has been noted in the Northeast, one of the areas most affected by acid deposition. The reduction in dry and total sulfur deposition (wet plus dry) has been even more dramatic than that of wet deposition in the Mid-Atlantic and Midwest, with reductions of 38 and 44 percent, respec-

Figure 5: Annual Mean Wet Sulfate Deposition



Source: NADP, 2009

About Long-term Ambient and Deposition Monitoring Networks

To evaluate the impact of emission reductions on the environment, scientists and policymakers use data collected from long-term national monitoring networks such as CASTNET and the NADP/NTN. These complementary, long-term monitoring networks provide information on a variety of indicators necessary for tracking temporal and spatial trends in regional air quality and acid deposition (see Table 2).

CASTNET provides atmospheric data on the dry deposition component of total acid deposition, ground-level ozone, and other forms of atmospheric pollution. Established in 1987, CASTNET now consists of more than 80 sites across the United States. EPA's Office of Air and Radiation operates 50 of the monitoring stations; the National Park Service (NPS) funds and operates approximately 30 stations in cooperation with EPA. Many CASTNET sites have a continuous 20-year data record, reflecting EPA's commitment to long-term environmen-

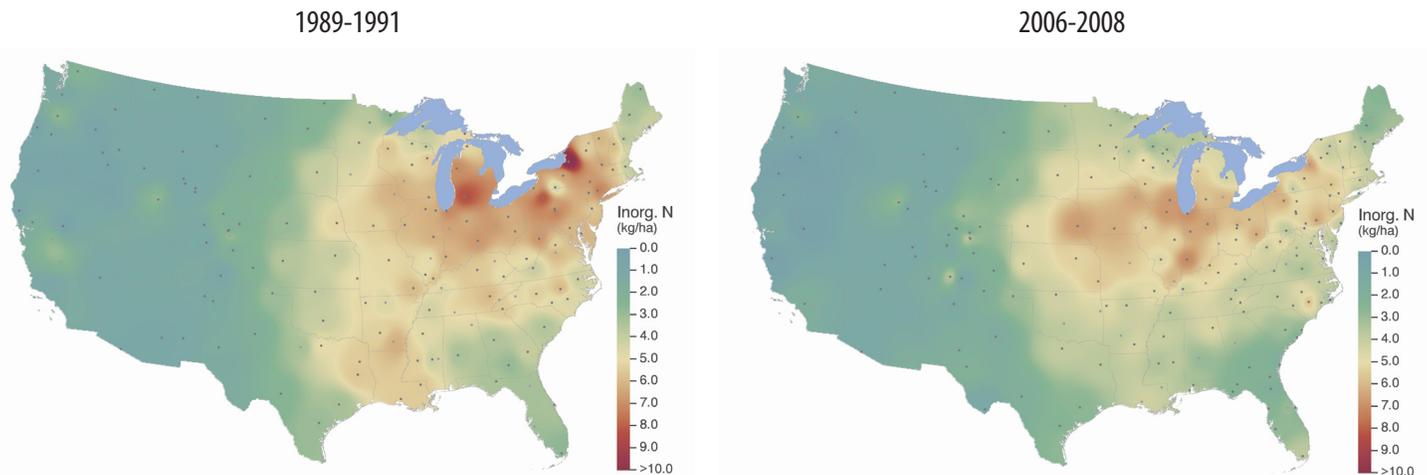
tal monitoring. Information and data from CASTNET are available at www.epa.gov/castnet.

NADP/NTN is a nationwide, long-term network tracking the chemistry of precipitation. NADP/NTN provides concentration and wet deposition data on hydrogen ion (acidity as pH), sulfate, nitrate, ammonium, chloride, and base cations. The network is a cooperative effort involving many groups, including the State Agricultural Experiment Stations, U.S. Geological Survey (USGS), U.S. Department of Agriculture, EPA, NPS, the National Oceanic and Atmospheric Administration (NOAA), and other governmental and private entities. NADP/NTN has grown from 22 stations at the end of 1978 to more than 250 sites spanning the continental United States, Alaska, Puerto Rico, and the Virgin Islands. Information and data from NADP/NTN are available at nadp.sws.uiuc.edu.

Table 2: Air Quality and Acid Deposition Measures

Chemical Name	Chemical Symbol	Measured in:		Why are these measured by the networks?
		Ambient Air	Wet Deposition	
Sulfur Dioxide	SO ₂	X		Primary precursor of wet and dry acid deposition; primary precursor to fine particles in many regions.
Sulfate Ion	SO ₄ ²⁻	X	X	Major contributor to wet acid deposition; major component of fine particles in the Midwest and East; can be transported over large distances; formed from reaction of SO ₂ in the atmosphere.
Nitrate Ion	NO ₃ ⁻	X	X	Contributor to acid and nitrogen wet deposition; major component of fine particles in urban areas; formed from reaction of NO _x in the atmosphere.
Nitric Acid	HNO ₃	X		Strong acid and major component of dry nitrogen deposition; formed as a secondary product from NO _x in the atmosphere.
Ammonium Ion	NH ₄ ⁺	X	X	Contributor to wet and dry nitrogen deposition; major component of fine particles; provides neutralizing role for acidic compounds; formed from ammonia gas in the atmosphere.
Ionic Hydrogen	H ⁺		X	Indicator of acidity in precipitation; formed from the reaction of sulfate and nitrate in water.
Calcium	Ca ₂ ⁺	X	X	These base cations neutralize acidic compounds in precipitation and the environment; also play a major role in plant nutrition and soil productivity.
Magnesium	Mg ₂ ⁺	X	X	
Potassium	K ⁺	X	X	
Sodium	Na ⁺	X	X	

Source: EPA, 2009

Figure 6: Annual Mean Wet Inorganic Nitrogen Deposition

Source: NADP, 2009

tively (see Table 1 on page 2). Because continuous data records are available from only a few sites in the Northeast and Southeast, it is unclear if the observed reductions in total deposition are representative for those regions.

A principal reason for reduced sulfate deposition in the Northeast is a reduction in the long-range transport of sulfate from emission sources located in the Ohio River Valley. The reductions in sulfate documented in the Northeast, particularly across New England and portions of New York, were also affected by SO₂ emission reductions in eastern Canada. NADP data indicate that similar reductions in precipitation acidity, expressed as hydrogen ion (H⁺) concentrations, occurred concurrently with sulfate reductions, with reductions of 30 to 40 percent over much of the East.

Reductions in nitrogen deposition recorded since the early 1990s have been less pronounced than those for sulfur. As noted earlier, emission trends from source categories other than ARP sources significantly affect air concentrations and deposition of nitrogen. Inorganic nitrogen in wet deposition decreased commensurately in the Mid-Atlantic and Northeast (see Figure 6). Decreases in dry and total inorganic nitrogen deposition at CASTNET sites have generally been greater than that of wet deposition, with a 25 and 19 percent decrease in total nitrogen deposition for the Mid-Atlantic and Midwest, respectively (see Table 1 on page 2). Other source sectors and pollutants, particularly agriculture and ammonium respectively, affect nitrogen transport and deposition.

Ambient Mercury Monitoring

The NADP membership of federal agencies, states, tribes, academic institutions, industry, and other organizations have established a new network to measure atmospheric concentrations of mercury throughout the U.S. The focus of this effort is to develop national capacity to monitor the three ambient mercury species—gaseous oxidized mercury (GOM), particulate-bound mercury (PBM_{2.5}), and gaseous elemental mercury (GEM). Datasets generated from this network are used to estimate mercury dry deposition, assess mercury source/receptor relationships, evaluate atmospheric models, and determine long-term trends. Currently 20 sites provide high-resolution, high-quality atmospheric data. In 2009, seven monitoring sites were added to the network, some of which were co-sponsored by EPA, including sites operated by the New Jersey Department of Environmental Protection, University of Utah, University of New Hampshire, and others. High quality speciated data are available through the NADP Atmospheric Mercury Network webpage. NADP also began supporting a site liaison to oversee quality assurance of network data by manually examining raw instrument data and performing annual site visits. NADP worked with site operators and the broader mercury scientific community to create a field standard operating procedure (SOP) for monitoring atmospheric mercury species in a network mode to ensure cross-network data comparability. Additionally, NADP has developed a

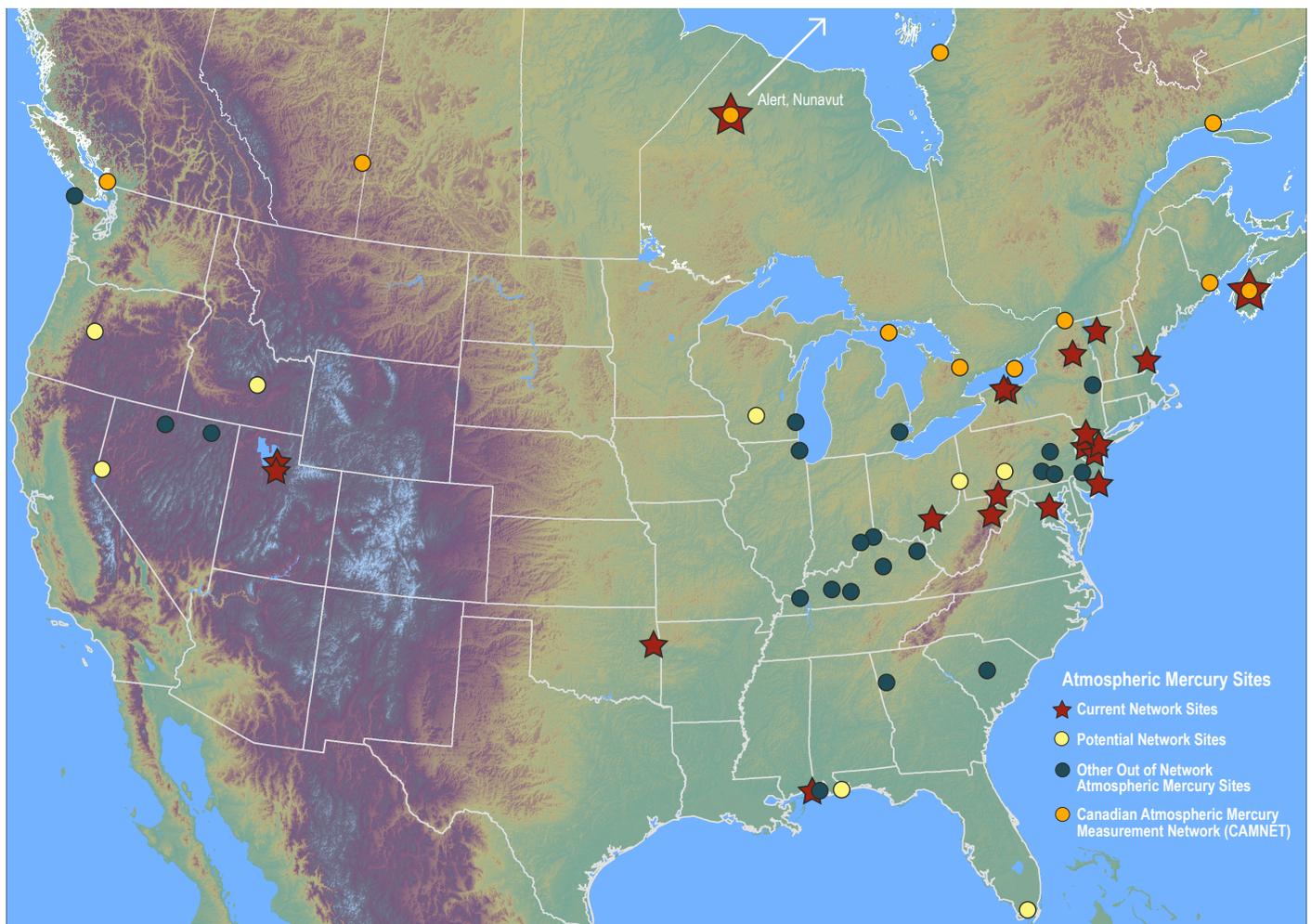
data management SOP to provide routine automated quality assured data and display network data products on the web.

The NADP Atmospheric Mercury Network is one component of a broader effort to establish a more comprehensive, integrated mercury monitoring network called MercNet. A May 2008 National Mercury Monitoring Workshop was an important step in building further broad support for MercNet. The workshop included participants from federal agencies (EPA, USGS, NOAA, U.S. Fish and Wildlife Service, NPS), state and tribal agencies, the NADP, industry, and academic and private research institutions. Workshop participants agreed on the overall goal of a mercury monitoring network: “To establish an integrated, national network to systematically monitor, assess, and report on policy-relevant indicators of atmospheric mercury

concentrations and deposition, and mercury levels in land, water, and biota in terrestrial, freshwater, and coastal ecosystems in response to changing mercury emissions over time.” Workshop scientists considered the conceptual framework for MercNet to include national distribution of sites to understand the sources, consequences, and changes in U.S. mercury pollution. The design elements of this effort include a national distribution of sites; monitoring mercury concentrations within air, water, fish, sediments and wildlife at each site; and a network that builds on existing monitoring efforts, where possible, to maximize information, benefits, coordination, and efficiency. Collaboration and partnerships among existing mercury science and monitoring programs are integral to MercNet.

For more information, visit the NADP MercNet website: nadp.sws.uiuc.edu/mercnet/.

Figure 7: Ambient Mercury Monitoring Locations



Source: NADP, 2009

For a copy of the 2008 MercNet National Mercury Monitoring Workshop Report, visit: nadp.sws.uiuc.edu/mercnet/MercNetFinalReport.pdf.

Improvements in Surface Water Chemistry

Acid rain, resulting from SO₂ and NO_x emissions, is one of many large-scale anthropogenic effects that negatively affect the health of 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. Long-term surface water monitoring networks provide information on the chemistry of lakes and streams and on how water bodies are responding to changes in emissions. 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, of those areas (see Figures 8-10 on pages 9 and 10).

Two EPA-administered monitoring programs provide information on the effects of acid rain on aquatic systems: 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 Clean Air Act (CAA) Amendments in reducing the acidity of surface waters in four regions: New England, the Adirondack Mountains, the Northern Appalachian Plateau, and the Ridge and Blue Ridge Province. 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.

The data presented here show regional trends in acidification from 1990 to 2007 in lakes and streams sampled through the LTM program (see Figures 8-10 on pages 9 and 10). Only sites that have a complete data record for the time period are represented. Three indicators of acidity in surface waters are presented—measured ions of sulfate and nitrate and acid neutralizing capacity (ANC). These indicators provide information regarding both sensitivity to surface water acidification and the level of acidification that has occurred today and in the past. Trends in these chemical receptors allow for the determination of whether the conditions of the water bodies are improving and heading towards recovery or if the conditions are degrading. Significant trends are statistically significant at the 95% confidence interval ($p < 0.05$).

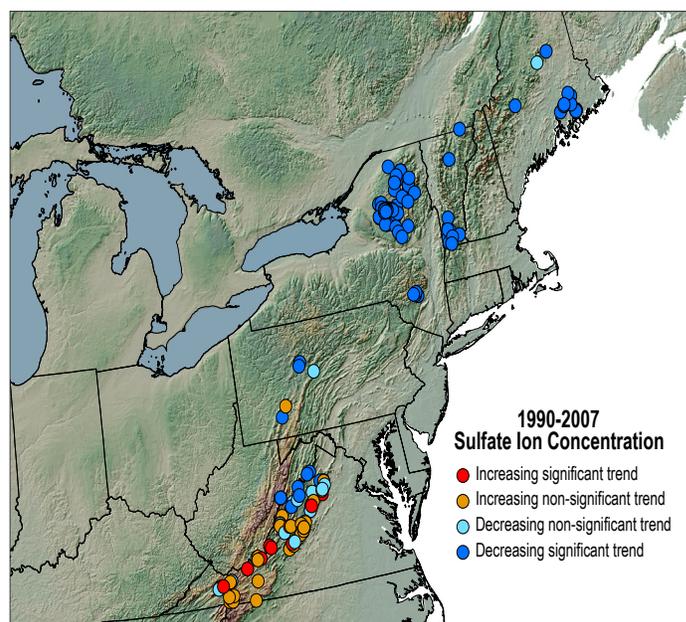
Measurements of sulfate ion concentrations in surface waters provide important information on the extent of cation leaching in soils and how sulfate concentrations relate to deposition and to the levels of ambient atmospheric sulfur.

In June 2009, the regional TIME and LTM cooperators met for a workshop at Penn State University. The goals of the workshop were to exchange science between regions; to discuss ways to improve program management, relevance, and visibility; to clarify current goals and objectives of the programs; and to envision future goals and operations. For more information on the TIME and LTM programs, visit: www.epa.gov/airmarkets/assessments/TIMELTM.html.

Assessments of acidic deposition effects dating from the 1980s to the present have shown sulfate to be the primary negatively charged ion in most acid-sensitive waters.¹ Nitrate has the same potential as sulfate to acidify drainage waters and leach acidic aluminum cations from watershed soils. In most watersheds, however, nitrogen is a limiting nutrient for plant growth, and therefore most nitrogen inputs through deposition are quickly incorporated into biomass as organic nitrogen with little leaching of nitrate into surface waters.

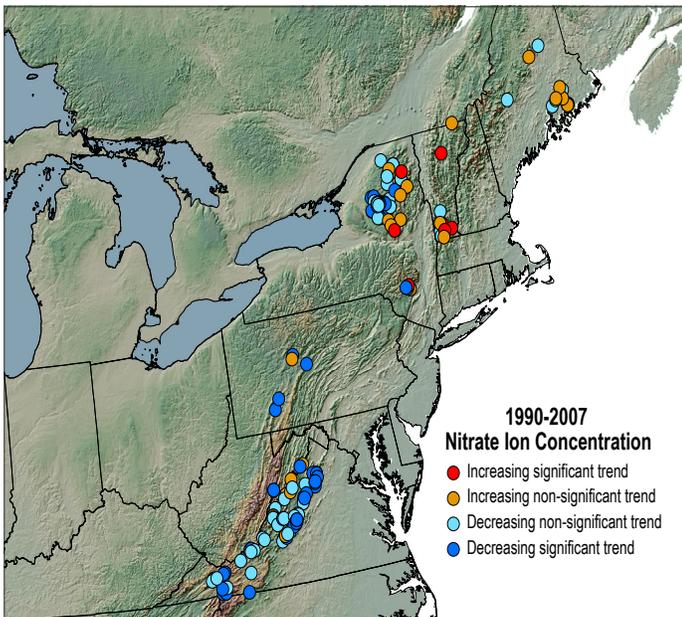
ANC is an important measure of the sensitivity and the degree of surface water acidification or recovery that occurs

Figure 8: Trends in Lake and Stream Water Chemistry at LTM Sites, 1990-2007, Sulfate Ion Concentration (µeq/L/yr)



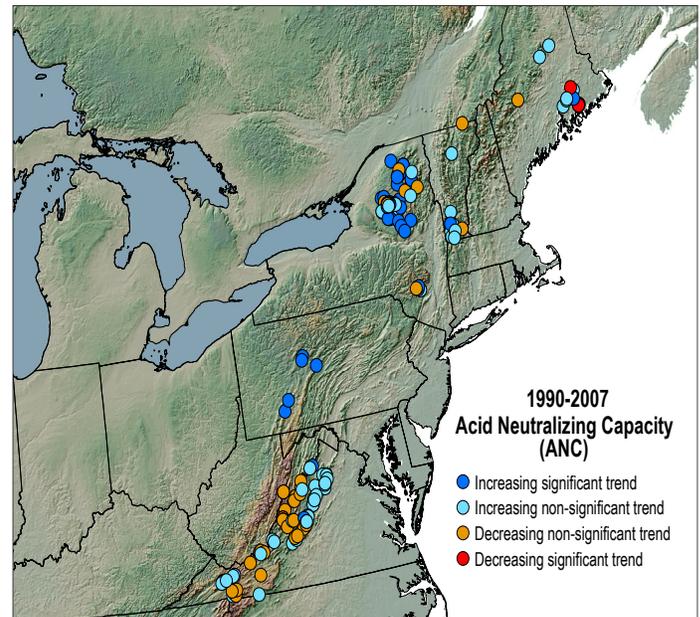
Source: EPA, 2009

Figure 9: Trends in Lake and Stream Water Chemistry at LTM Sites, 1990-2007, Nitrate Ion Concentration ($\mu\text{eq/L/yr}$)



Source: EPA, 2009

Figure 10: Trends in Lake and Stream Water Chemistry at LTM Sites, 1990-2007, ANC Levels ($\mu\text{eq/L/yr}$)



Source: EPA, 2009

over time. Acidification results in the diminishing ability of water in the lake or stream to neutralize strong acids that enter aquatic ecosystems. Water bodies with ANC values defined as less than or equal to 0 microequivalents² per liter ($\mu\text{eq/L}$) are acidic. Lakes and streams having spring-time ANC values less than 50 $\mu\text{eq/L}$ are generally considered “sensitive” to acidification. Lakes and streams with ANC higher than 50 $\mu\text{eq/L}$ are generally considered less sensitive or insensitive to acidification. When ANC is low, and especially when it is negative, stream water pH is also low (less than 6), and there may be adverse impacts on fish and other animals essential for a healthy aquatic ecosystem. Movement toward recovery of an aquatic ecosystem is indicated by positive trends in ANC and negative trends in sulfate and nitrate.

Table 3 presents the aggregate sulfate, nitrate, and ANC trends ($\mu\text{eq/L/yr}$) represented by the LTM sites shown in Figures 8-10 for four acid sensitive regions of the eastern U.S. The maps and summary results indicate that:

- Sulfate concentrations are declining at almost all sites in the Northeast (New England, Adirondacks/Catskills and Pennsylvania [Northern Appalachians]). However, in the Southern Blue Ridge (Central Appalachians), sulfate concentrations in many streams are increasing. This region has highly weathered soils that can store

Table 3: Trend Slopes for LTM Sites in Four Eastern U.S. Regions, 1990-2007

Region	Sulfate Slope	Nitrate Slope	ANC Slope
Adirondack Mountains	-2.225	-0.179	0.765
N. Appalachian Plateau	-2.396	-0.206	0.706
New England	-1.58	0.009	0.198
Ridge and Blue Ridge Province	0.07	-0.141	0.107

Notes:

- Bold values indicate significance at 95% confidence interval ($p < 0.05$). Confidence levels are used to express the reliability and significance of the estimate.
- The slope or trend in a simple linear regression (SLR) model corresponds to the change in the chemical variable over time. A negative or positive slope indicates whether the chemical variable in the regional distribution of water bodies is decreasing or increasing, respectively.
- Adirondack 2007 data is only for a partial year from January through May 1, 2007.
- The table of values represents the average trend for all the sites in each of the four regions.

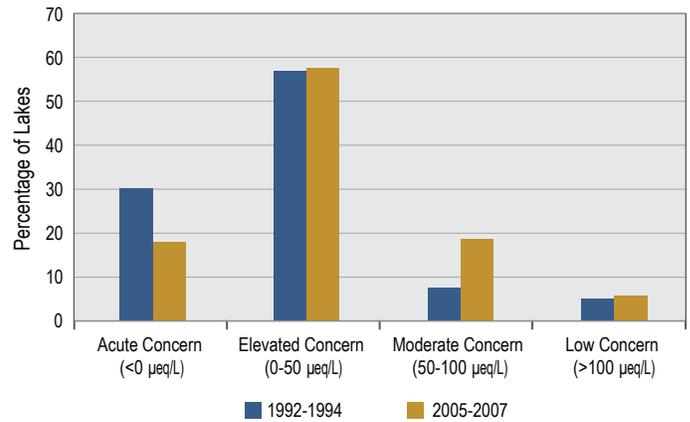
Source: EPA, 2009

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. Thus sulfate concentrations in surface waters, mainly streams in this region, are increasing despite reduced sulfate deposition.

- Nitrate concentrations are decreasing in three of the four regions, but several lakes and streams indicate flat or slightly increasing nitrate trends. This trend does not appear to reflect changes in emissions or deposition in these areas and is likely a result of ecosystem factors.
- ANC, as measured in surface waters, is on average increasing in three of the four regions, which in part can be attributed to declining sulfate deposition. The site trends also indicate variation within each region. Only two sites indicate a significant downward trend in ANC.

The ANC of northeastern U.S. lakes monitored under the TIME and LTM programs was also evaluated for 1992 to 1994 and 2004 to 2007 to assess the impacts of ARP implementation. The analysis in Figure 11 compares average ANC levels for the northeastern lakes that had data in each time period. Thirty percent of lakes in 1992 to 1994 had three-year mean ANC levels below 0 µeq/L. These lakes are categorized as “acute concern,” in which a near complete loss of fish populations is expected, and planktonic communities have low diversity and are dominated by acid-tolerant forms (see Table 4). The percentage of lakes in this category dropped to 18 percent in 2004 to 2007 (see Figure 11). As a result, the three other categories (elevated, moderate, or low concern) experienced slight increases. These results point to a decrease in acidity, particularly for the subset of lakes with low ANC.

Figure 11: Northeastern Lakes by ANC Status Category, 1992-1994 versus 2005-2007



Notes:

- Based on 156 TIME/LTM monitored sites.
- Adirondack 2007 data is only for a partial year from January through May 1, 2007.
- See Table 4 for descriptions of level of concern categories.
- It is important to note that the wide range of ANC values within these categories makes it likely that substantial improvements in ANC may occur without changing the categorization of a given lake.

Source: EPA, 2009

Western Adirondack Stream Survey

The Adirondack Mountain region of New York has long been a focal point for environmental concern over acid deposition. Poor buffering capability of the soils in the Adirondack region makes the surface waters particularly susceptible to acidification. The effects of acid deposition and recent improvements in surface water acidity have been well documented for lakes in the region largely due to

Table 4: Aquatic Ecosystem Status Categories for the Adirondack Mountains

Category Label	ANC Level	Expected Ecological Effects
Acute Concern	< 0 micro equivalent per Liter (µeq/L)	Near complete loss of fish populations is expected. Planktonic communities have extremely low diversity and are dominated by acidophilic forms. The numbers of individuals in plankton species that are present are greatly reduced.
Elevated Concern	0-50 µeq/L	Fish species richness is greatly reduced (more than half of expected species are missing). On average, brook trout populations experience sub-lethal 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 declines.
Moderate Concern	50-100 µeq/L	Fish species richness begins to decline (sensitive species are lost from lakes). Brook trout populations are sensitive and variable, with possible sub-lethal effects. Diversity and distribution of zooplankton communities begin to decline as species that are sensitive to acid deposition are affected.
Low Concern	> 100 µeq/L	Fish species richness may be unaffected. Reproducing brook trout populations are expected where habitat is suitable. Zooplankton communities are unaffected and exhibit expected diversity and distribution.

Source: EPA, 2009

comprehensive lake monitoring and assessment programs.³ In contrast, only a few stream surveys have been conducted since the 1980s, which has provided little data to assess the current status of Adirondack streams.

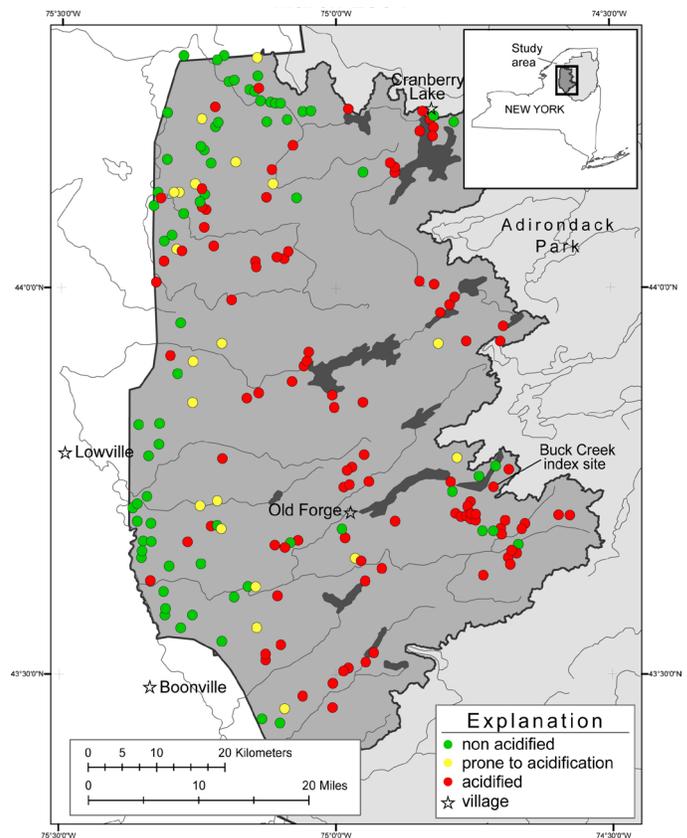
Although regional lake and stream chemistry are similar, streams are often more prone to acidification than lakes because they can receive much of their water from shallow soils that are often ineffective at neutralizing acidity. Therefore, observed improvement in lake acidity over the past two decades may not be occurring in streams. For these reasons, the New York State Energy Research and Development Authority (NYSERDA) sponsored the Western Adirondack Stream Survey (WASS) from 2003 to 2005 to assess the current chemical and biological condition of streams in the western section of the Adirondacks.⁴⁻⁵

Researchers from U.S. Geological Survey, New York State Department of Environmental Conservation, Adirondack Lakes Survey Corporation and the University of Texas at Arlington assessed 565 streams representing 825 km² of the Black River and Oswegatchie River drainages within the boundaries of the Adirondack Park (Figure 12). Streams were sampled during base-flow, snowmelt and fall storm events for water chemistry to determine if they were episodically or chronically acidic.⁶ The health of the biological community (diatoms and macroinvertebrates) was also measured in a subset of streams.

The extent of stream acidification in the western Adirondack Mountain region remains high. The streams sampled in this study showed that 66 percent or 718 km of streams are prone to acidification and likely have levels of acidity harmful to their biota. Of the 66 percent of streams found to be prone to acidification, about 50 percent were likely to be chronically acidified, with the other 50 percent episodically acidified.

The impacts of acidification on the health of the aquatic communities also remain pronounced. The percentage of streams determined to be moderately to severely impacted on the basis of their diatom community ranged from 66 to 80 percent over the different surveys. WASS showed clear evidence that over half of the assessed streams had macroinvertebrate communities that were moderately to severely impacted.⁷ The survey also showed that two thirds of streams sampled have poor water quality during some point during the year that can be toxic to fish such as brook trout.

Figure 12: Acidification Categories of Sites Sampled in the March 2004 Survey



Note: Non acidified sites had base cation surplus (BCS) values > 25 eq L-1; sites prone to acidification had BCS values > 0 eq L-1 but < 25 eq L-1; acidified sites had BCS values < 0 eq L-1.

Source: NYSERDA, 2008

These results also indicate that the recovery from acidification in these western Adirondack streams has been minimal. Comparison with historical water chemistry data available for 12 streams showed that less than half of the streams had higher acid neutralizing capacity (ANC) in 2003 to 2005 than in the early 1980s. The overall increase for these streams was 13 µeq/L over the 23 year period. Further reductions in acid deposition in the Adirondacks are necessary for greater recovery of these sensitive aquatic systems.

Critical Loads Case Study: Central Appalachian Mountain Streams

The central Appalachian Mountain region of Virginia and West Virginia that includes the Shenandoah National Park is known to be sensitive to acidic deposition. Poor soils and low weathering rates of the bedrock beneath the mountain terrain in the central Appalachian Mountain region make the streams particularly susceptible to acidification. As a result, acid deposition has impacted many miles of streams in the region, greatly reducing the diversity of aquatic life including important recreational fish species such as brook trout.

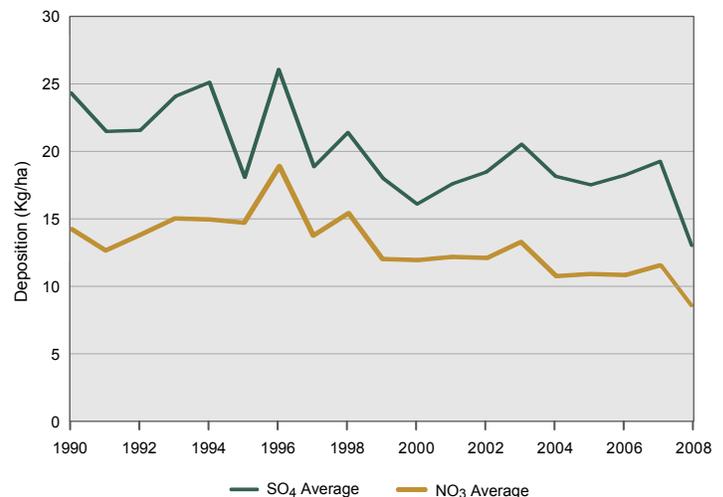
In this case study, critical loads of deposition were calculated for 92 streams in the central Appalachian Mountain region. The critical load is the maximum exposure to pollutant deposition below which significant harmful effects to ecological health of the environment do not occur according to present knowledge. If the actual pollutant deposition to a lake or stream is greater than its critical load—if the critical load is exceeded—then that water body is at risk for continuing ecological damage. If pollutant deposition is less than the critical load, adverse ecological effects are not anticipated (and if the water body has been damaged by past deposition, recovery is expected).

There are numerous peer-reviewed scientific methods and models that can be used to calculate critical loads for surface water acidity for streams. Drawing on recent scientific studies⁸ in the eastern United States, this case study uses the Steady-State Water Chemistry (SSWC) and the Model of Acidification of Groundwater In Catchment (MAGIC) models to calculate the critical load. The analysis uses water chemistry data from the TIME/LTM, Virginia Trout Stream Sensitivity Study (VTSSS), and other programs that are part of the Environmental Monitoring and Assessment Program (see discussion of surface water trends on pages 9-11). The focus of this case study is on the combined load of sulfur and nitrogen deposition below which the ANC level would still support healthy aquatic ecosystems. Research studies have shown that surface water with ANC values greater than 50 $\mu\text{eq/L}$ tends to protect most fish (e.g., brook trout, others) and other aquatic organisms (Table 4 on page 11). In this case, the critical load represents the combined deposition load of sulfur and nitrogen to which a stream and its watershed could be subjected and still have a surface water concentration ANC of 50 $\mu\text{eq/L}$ on an annual basis. Critical loads of combined total sulfur and nitrogen are expressed in terms of ionic charge balance as milliequivalent⁹ per square meter per year ($\text{meq/m}^2/\text{yr}$).

In the United States, the critical loads approach is not an officially accepted approach to ecosystem protection. For example, language specifically requiring a critical loads approach does not exist in the Clean Air Act. However, recent activities within federal and state agencies, as well as the research community, indicate that critical loads are emerging as a useful ecosystem protection and program assessment tool.

In June 2008, a report was released by the Nature Conservancy and the Cary Institute of Ecosystem Studies that called on Congress, federal and state agencies, conservation groups, and scientists to work together to establish critical loads to protect sensitive ecosystems. The report, *Threats from Above: Air Pollution Impacts on Ecosystems and Biological Diversity in the Eastern United States*,¹⁰ is based on the results of an expert workshop to evaluate air pollution effects in the Northeast and Mid-Atlantic and identify conservation implications. The report recommended that Congress direct the EPA to develop and implement a deposition-based air quality standard, like a critical load, for sulfur and nitrogen pollution in sensitive ecosystems that receive high deposition levels.

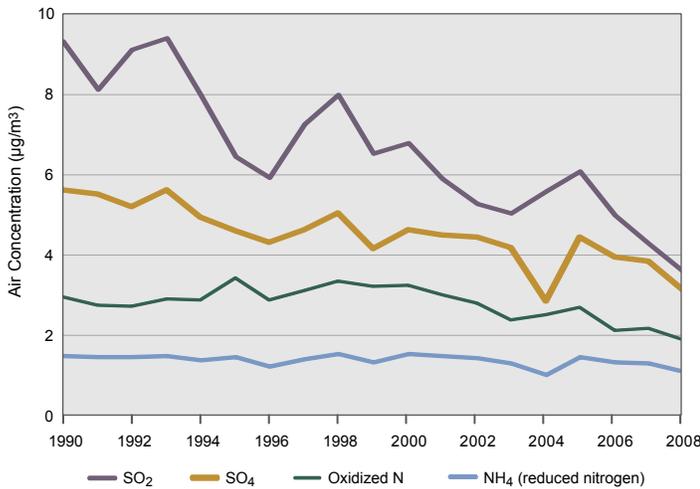
Figure 13: Annual Average Wet Deposition of Sulfate and Nitrate in the Shenandoah National Park and Surrounding Areas, 1990-2008



Note: NADP Sites: VA28, VA27, VA00, VA99, WV04, WV18

Source: NADP, 2009

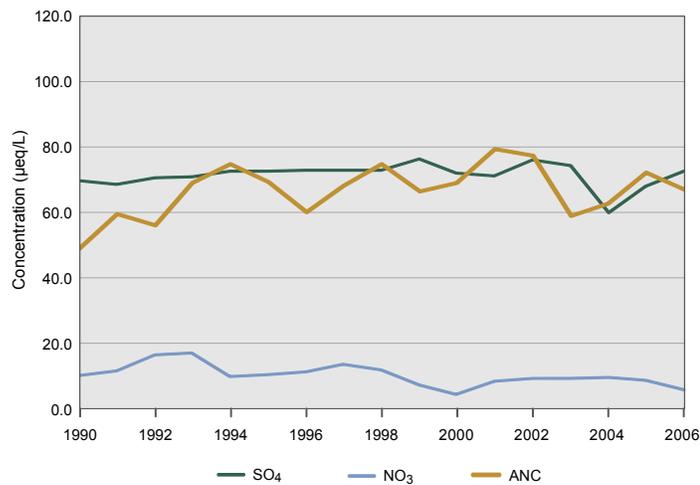
Figure 14: Annual Average Air Concentration of SO₂, Oxidized Nitrogen, SO₄, and Reduced N in the Shenandoah National Park and Surrounding Areas, 1990-2008



Note: CASTNET Site: SHN418

Source: CASTNET, 2009

Figure 15: Annual Average Surface Water Sulfate and Nitrate Concentration in the Central Appalachian Mountain Region, 1990-2006, Compared with ANC



Source: EPA, 2009

Based on environmental monitoring data collected since the 1990s in the central Appalachian Mountain region, emission reductions from the ARP and other programs have resulted in substantial decreases in atmospheric concentration and deposition of sulfur and nitrogen. Between

1990 and 2008, annual average wet deposition of sulfur and nitrogen has decreased by approximately 47 percent and 41 percent, respectively, as shown in Figure 13 on page 13. These deposition reductions parallel the decreases in the air concentrations of these pollutants over the same period as shown in Figure 14.

The declines in air concentrations and deposition of sulfur and nitrogen since the 1990s have resulted in only slight signs of recovery from acid rain in the streams of the central Appalachian Mountain region. Figure 15 shows trends in sulfate, nitrate, and ANC for central Appalachian Mountain streams monitored through the LTM program. Sulfate concentrations in these streams have remained level, nitrate concentrations have dropped slightly, and the resulting overall trend for ANC is a slight increase. These observed trends indicate an important first step towards ecological recovery.

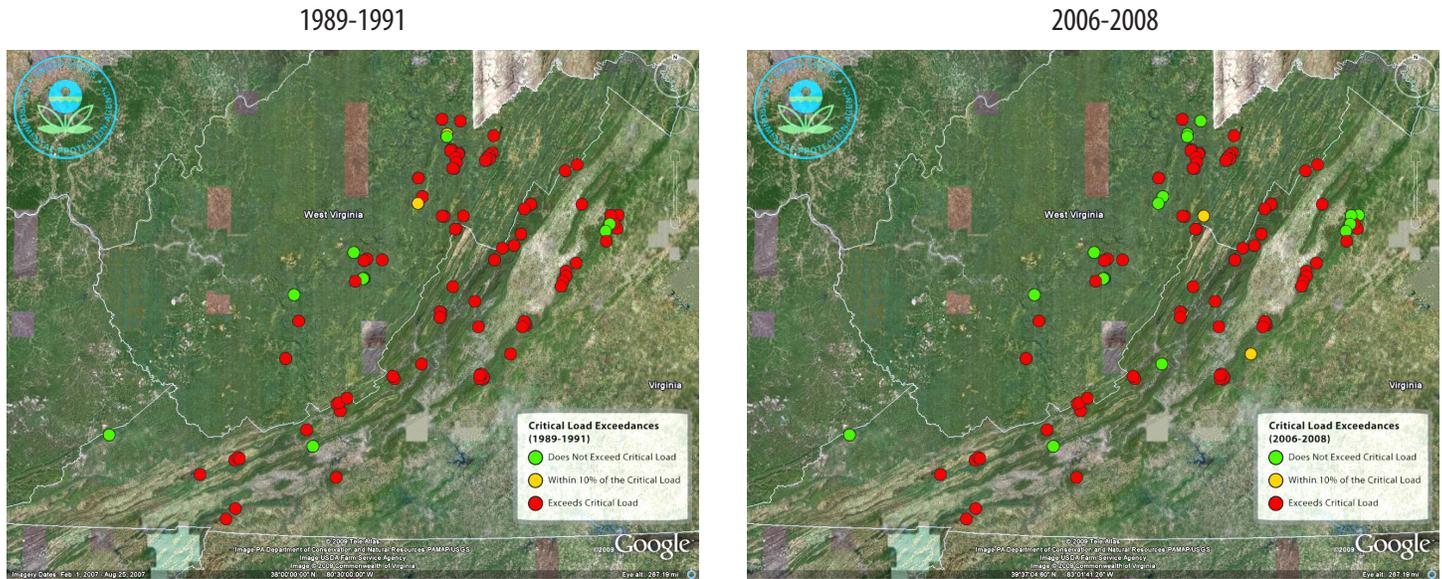
It is difficult to determine whether central Appalachian Mountain region aquatic ecosystems will recover and be sufficiently protected from acid deposition based on environmental monitoring data alone. The critical load provides a benchmark to gauge whether deposition has decreased enough to protect the ecological health of a lake or stream. In Figure 16 on page 15, a critical load exceedance indicates that the combined sulfur and nitrogen deposition was greater than a stream could sustain and still maintain the ANC level of 50 µeq/L or above. Exceedances were calculated from deposition for the period before implementation of the ARP (1989–1991) and for a recent period after ARP implementation (2006–2008).

For the period before ARP implementation, 90 percent of streams received levels of combined sulfur and nitrogen deposition that exceeded the critical load and could not be neutralized by the environment. For the period from 2006 to 2008, 82 percent of the streams examined continued to receive greater acid deposition than could be neutralized, only an eight percent improvement from before ARP implementation.

Figure 16 on page 15 also shows streams where deposition was within 10 percent of the critical load. These streams illustrate areas where ecosystem health has improved only slightly over time.

This critical load analysis shows that emission reductions achieved by the ARP have resulted in some improvement in environmental conditions and increased ecosystem protection in the central Appalachian Mountain region. The analysis also demonstrates that the central Appalachian Mountain region remains at risk to acidification due to

Figure 16: Comparison of Critical Load Exceedances in Central Appalachian Mountain Streams before and after Implementation of the Acid Rain Program



Source: EPA, 2009

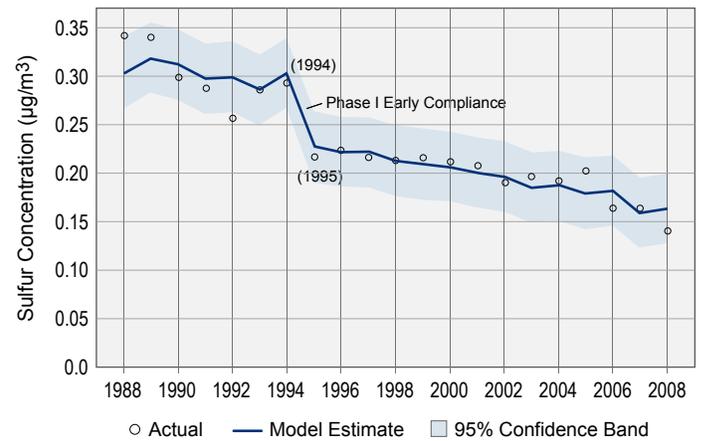
current acidic deposition levels and deeper reductions are necessary for recovery of these sensitive aquatic systems and full ecosystem protection.

Trends in Atmospheric Sulfur Concentrations

ARIMA Model

To help assess the trend in atmospheric sulfur concentrations since inception of the ARP, an Autoregressive Integrated Moving Average (ARIMA) model¹¹ was used to plot average sulfur concentrations captured by the 12 long-term CASTNET sites in Ohio, Pennsylvania and West Virginia. The ARIMA model, an advanced statistical analysis tool that can evaluate trends over time, shows that from 1994 to 1995 there was a step decrease in average atmospheric sulfur concentrations that is statistically significant at the 99.9 percent level. This step decrease is the result of ARP pre-compliance and the start of phase I of the ARP (Figure 17). From 1996 to 2008 the ARIMA model shows a steady downward trend (a negative slope) in atmospheric sulfur concentrations that is also statistically significant at the 99.9 percent confidence level. The results of this ARIMA assessment demonstrate the substantial impact of the ARP on atmospheric sulfur concentrations over time.

Figure 17: Ambient Sulfur Concentrations at CASTNET Sites in Ohio, Pennsylvania, and West Virginia



Note: Sulfur concentrations include ambient SO₂ and sulfate.

Source: EPA, 2009

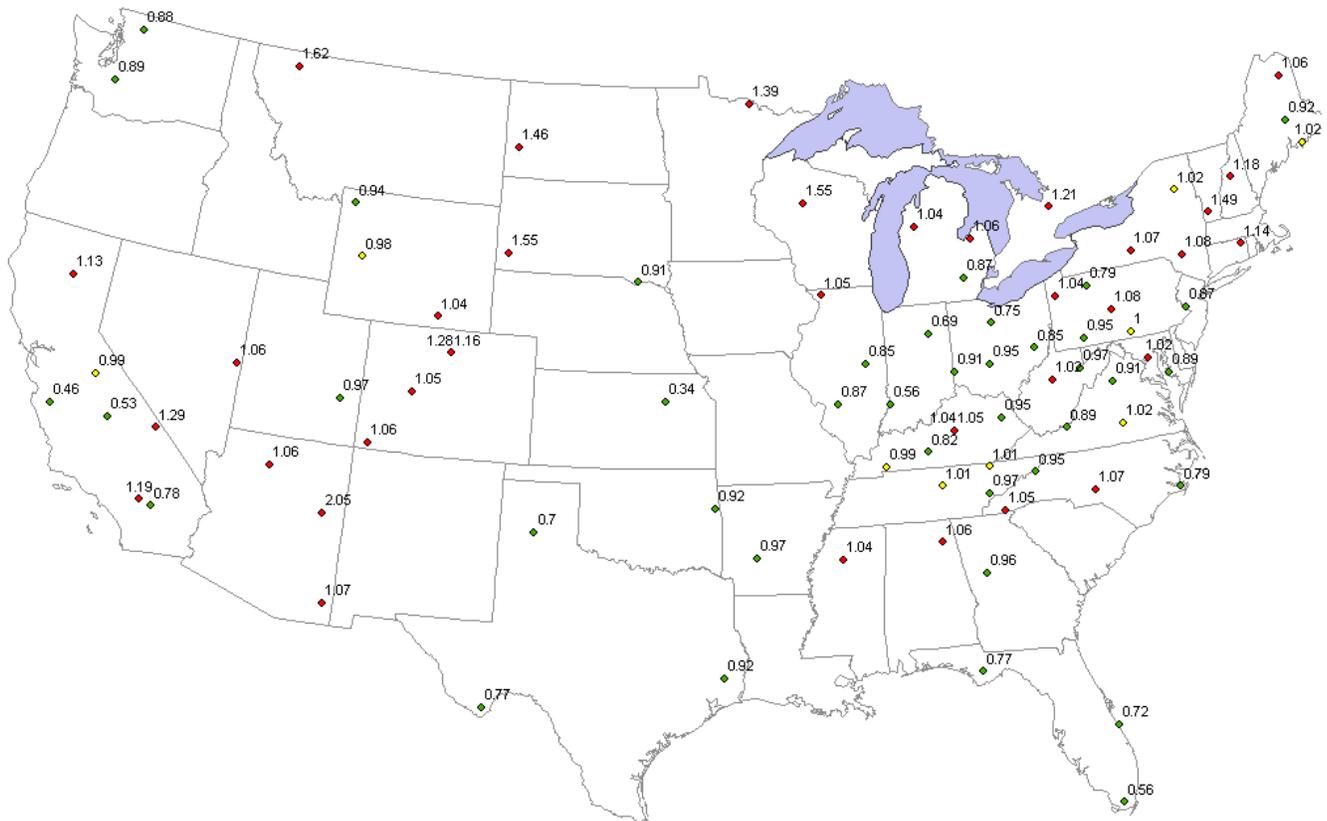
Ammonium Sulfate

SO₂ and NO_x emissions can react in the atmosphere to form fine particles, which are harmful to humans and sensitive ecosystems. Sulfate particles are formed after gaseous SO₂ is emitted and oxidized. When the oxidative potential of the atmosphere is high, a large fraction of atmospheric sulfur in the form of gaseous SO₂ is converted into particulate sulfate.

Some of the common particulate sulfate compounds formed in the atmosphere include ammonium sulfate ((NH₄)₂SO₄), ammonium bisulfate (NH₄HSO₄), and letovicite ((NH₄)₃H(SO₄)₂). Ammonium sulfate is a particularly stable atmospheric compound, meaning that once it is formed it will be transported and deposited in that form. Ammonium sulfate makes up a significant fraction of fine particulate matter (PM_{2.5}) in the Northeast.¹² When at least two ammonium ions are present per one sulfate ion in the atmosphere, ammonium sulfate is the dominant sulfate particulate compound formed.

Particulate sulfate, including ammonium sulfate, is collected by the CASTNET Teflon® filter. The relative average annual 2007 proportions of ammonium and sulfate were determined at each CASTNET site (Figure 18). When exactly twice the concentration of ammonium to sulfate is measured, it is implied that all of the particulate sulfur is in the form of ammonium sulfate. This is signified by a ratio of 1 in Figure 18 (0.5 moles of ammonium divided by moles of sulfate) at the given CASTNET sites. Ratios larger than one suggest that proportionally, at least two times more ammonium ions are present than sulfate on the particulate filter. Conversely, ratios less than one suggest proportionally more sulfate is present than ammonium. For locations with ratios equal to or greater than one, the chemical state of the atmosphere is recognized to be “sulfur limited” with respect to sulfate PM formation, meaning that the amount of sulfate available in the atmosphere is controlling the formation of ammonium sulfate. Thus it is expected that decreases in sulfur emissions in these areas would likely lead to a more pronounced decrease in particulate sulfate for-

Figure 18: Ratios of Ammonium versus Sulfate, as Measured at CASTNET Locations in 2007



Note: Ratios near or larger than 1.00 suggest that most of the sulfate is in the form of (NH₄)₂SO₄.

Source: EPA, 2009

mation and potentially a decrease in $PM_{2.5}$ in areas where ammonium sulfate is a significant fraction of the PM.

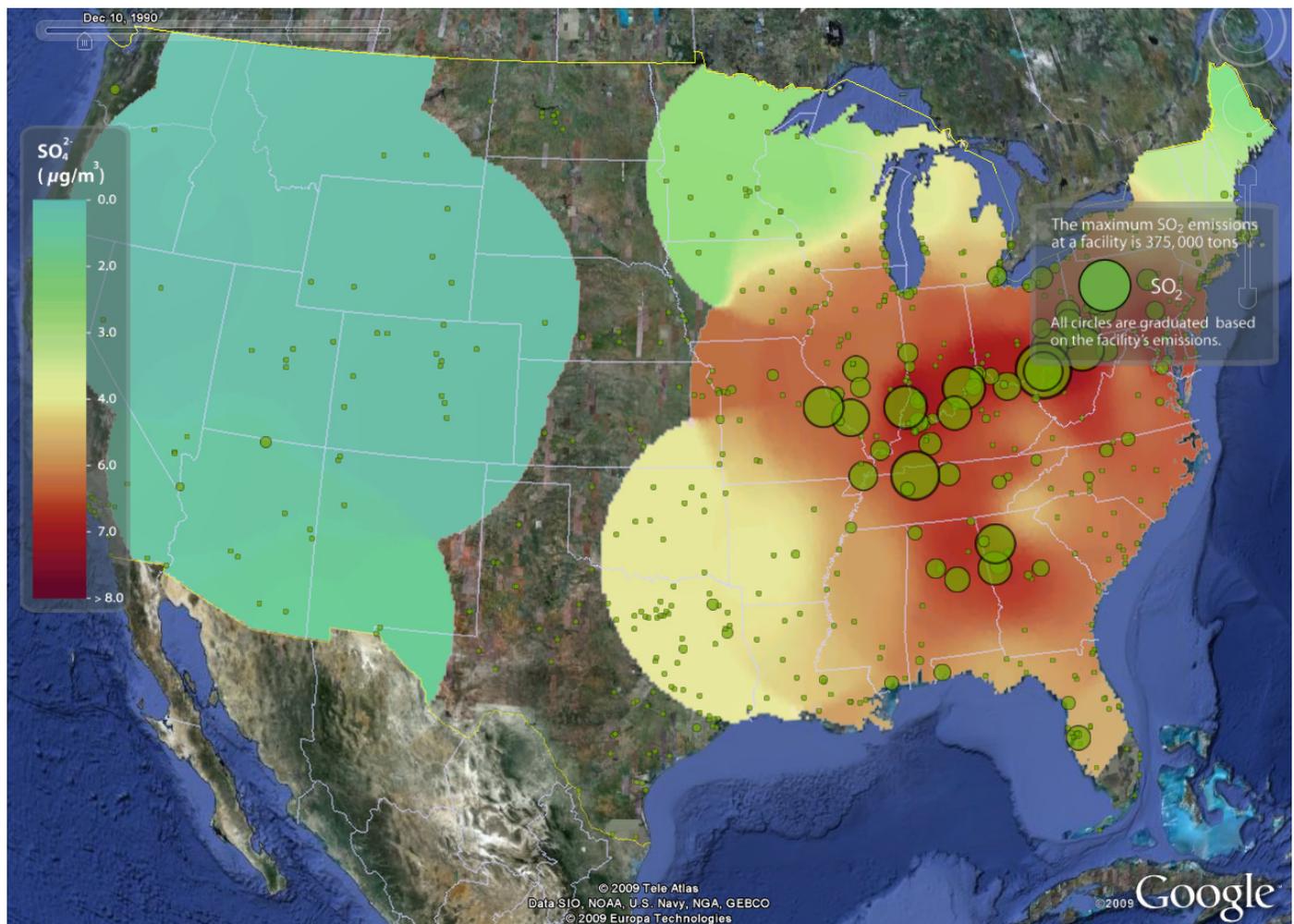
Online Information, Data, and Resources

The availability and transparency of data, from emission measurement to allowance trading to deposition monitoring, is a cornerstone of effective cap and trade programs. CAMD, in the Office of Air and Radiation's Office of Atmospheric Programs, develops and manages programs for collecting these data and assessing the effectiveness of cap and trade programs, including the ARP.

The CAMD Web site provides a public resource for general information on how market-based programs work and what they have accomplished, along with the processes, information, and tools necessary to participate in any of these market-based programs. For information about EPA's air emission trading programs, see <www.epa.gov/airmarkets>. For information about the ARP, see <www.epa.gov/airmarkets/progsregs/arp/index.html>.

To increase data transparency, EPA has created supplementary maps that allow the user to display air market program data geospatially on an interactive

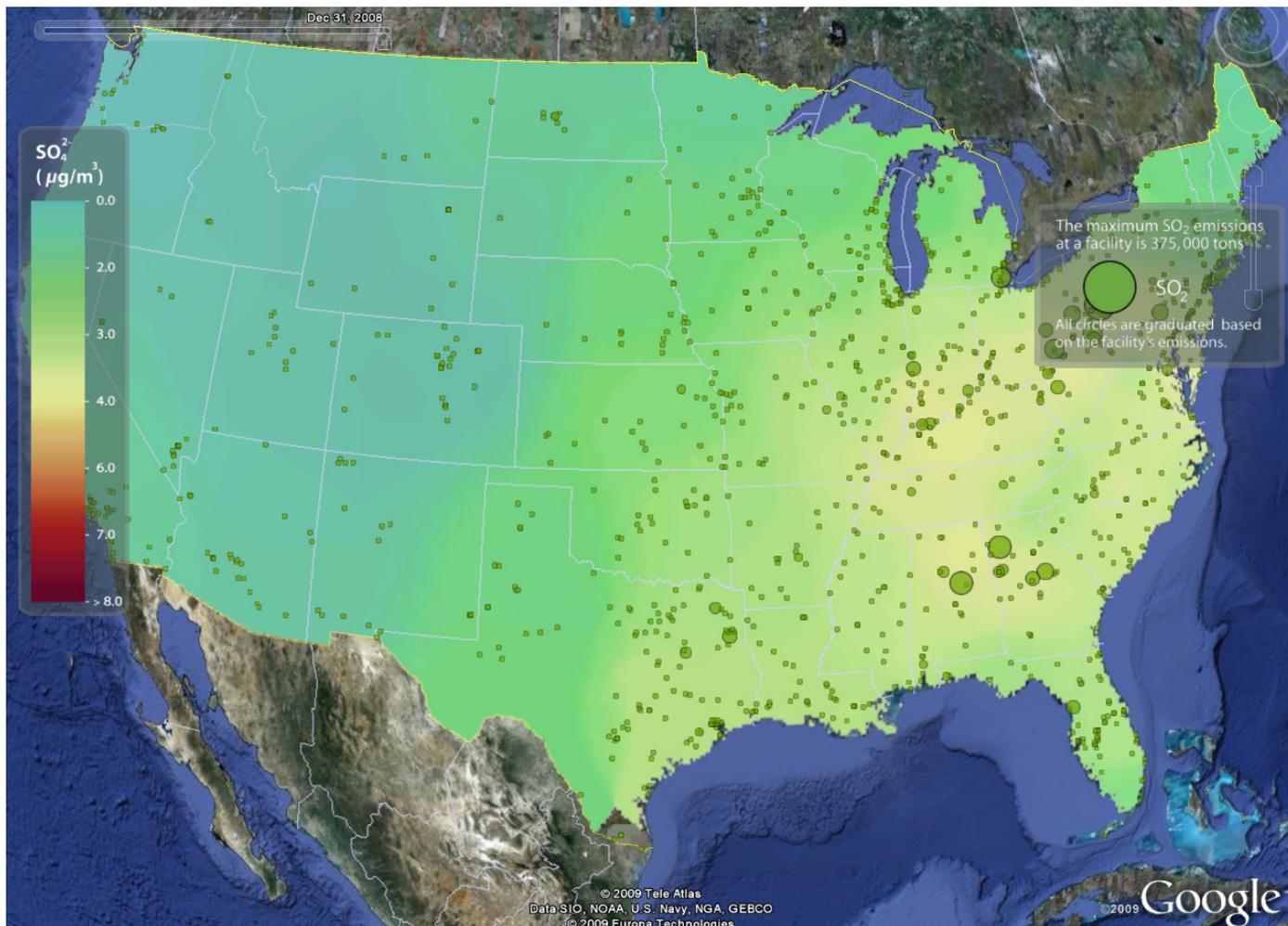
Figure 19: U.S. Sulfur Dioxide Emissions from ARP Sources and Ambient Sulfate Concentrations, 1990



Note: This example depicts 1990 SO_2 emissions from ARP sources along with 1990 sulfate concentration data as measured by the CASTNET monitoring program.

Source: EPA, 2009

Figure 20: U.S. Sulfur Dioxide Emissions from ARP Sources and Ambient Sulfate Concentrations, 2008



Note: This example depicts 2008 SO₂ emissions from ARP sources along with 2007 sulfate concentration data as measured by the CASTNET monitoring program.

Source: EPA, 2009

3D platform. Figures 19 and 20 are examples of these interactive maps. The maps come in the form of a KMZ file (a compressed KML file) that is downloaded directly to the user's computer. Data can be explored in new and meaningful ways by turning different layers on and off, overlaying data points and satellite imagery, and using navigation tools to change the view of the Earth's surface. KMZ/KML files are supported by programs such as Google

Earth, ESRI Arc Explorer, and NASA WorldWind View. These interactive mapping applications provide a unique way to identify environmental trends and track the progress of various EPA programs, such as the ARP.

For more information or to utilize the program, visit the Web site at epa.gov/airmarkets/progress/interactivemapping.html.

Endnotes

- ¹ Driscoll, C.T., Lawrence, G., Bulger, A., Butler, T., Cronan, C., Eagar, C., Lambert, K.F., Likens, G.E., Stoddard, J., and Weathers, K. 2001. Acid Deposition in the Northeastern U.S.: Sources and Inputs, Ecosystem Effects, and Management Strategies. *Bioscience*, 51:180-198.
- ² An equivalent is a measure of a substance's ability to combine with other substances. The equivalent is formally defined as the amount of a substance, in moles, that will react with one mole of electrons. A microequivalent is one millionth of an equivalent.
- ³ Temporally Integrated Monitoring of Ecosystems (TIME) program, Long Term Monitoring (LTM) program, and Adirondack Long-Term Monitoring (ALTM) program.
- ⁴ Results from the *2003-2005 Western Adirondack Stream Survey Final Report*: 08-22 New York State Energy Research and Development Authority 2008.
- ⁵ Lawrence, G. B., Roy, K. M., Baldigo, B. P., Simonin, H. A., Capone, S. B., Sutherland, J. W., Nierzwicki-Bauer, S. A., Boylen, C. W. 2008. Chronic and Episodic Acidification of Adirondack Streams from Acid Rain in 2003-2005. *Journal of Environmental Quality* 37: 2264-2274.
- ⁶ Acidification categories of streams: Nonacidified streams had base cation surplus (BCS) values > 25 µeq/L, sites prone to acidification had BCS values between 0-25 µeq/L, and acidified sites had BCS values < 0 µeq/L. Base-cation surplus (BCS) is an adjusted measure of Acid Neutralizing Capacity (ANC) that includes the concentration of strong organic anions.
- ⁷ Baldigo, P. B., Lawrence G. B., Bode R. W., Simonin H. A., Roy K. M., Smith A. J., 2009. Impacts of acidification on macroinvertebrate communities in streams of the western Adirondack Mountains, New York, USA, *Ecological Indicators*, Volume 9(2), Pages 226-239.
- ⁸ These studies reflect research completed by E&S Environmental and the University of Virginia in partnerships with US EPA, NPS, and USDA Forest Service:
 Sullivan, T. J., B. J. Cosby, J. R. Webb, R. L. Dennis, A. J. Bulger, F. A. Deviney Jr. 2008. Streamwater acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National Park, Virginia. *Environmental Monitoring and Assessment* (2008) 137:85-99.
 Dupont, J., Clair, T.A., Gagnon, C., Jeffries, D.S, Kahl, J.S., Nelson, S.J., and Peckenham, J.M. 2005. Estimation of Critical Loads of Acidity for Lakes in Northeastern United States and Eastern Canada, *Environmental Monitoring and Assessment*, 109:275-291.
- ⁹ A milliequivalent is one thousandth of an equivalent.
- ¹⁰ Lovett, G.M., and T.H. Tear. 2008. Threats from Above: Air Pollution Impacts on Ecosystems and Biological Diversity in the Eastern United States. The Nature Conservancy and the Cary Institute of Ecosystem Studies.
- ¹¹ Statistical Analysis with SAS/STAT® Software.
- ¹² Review of the National Ambient Air Quality Standards for Particulate Matter: Policy Assessment of Scientific and Technical Information, OAQPS Staff Paper. <www.epa.gov/ttn/naaqs/standards/pm/s_pm_cr_sp.html>