







# Acid Rain and Related Programs 2009 Environmental Results

The Acid Rain Program (ARP), established under Title IV of the 1990 Clean Air Act (CAA) Amendments, requires major emission reductions of sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>), the primary precursors of acid rain, from the electric power industry. The SO<sub>2</sub> program sets a permanent cap on the total amount of SO<sub>2</sub> that may be emitted by electric generating units (EGUs) in the contiguous United States. The program is phased in, with the final 2010 SO<sub>2</sub> cap set at 8.95 million tons, a level of about one-half of the emissions from the power sector in 1980. NO<sub>x</sub> reductions under the ARP are achieved through a program that applies to a subset of coal-fired EGUs and is closer to a 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 the following page shows the regional changes in key air quality and atmospheric deposition measurements linked to the ARP's SO<sub>2</sub> and NO<sub>x</sub> emission reductions.

During 2010, 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 <www.epa.gov/airmarkets/progsregs/ arp/index.html>.

# **Air Quality**

### **Sulfur Dioxide**

Data collected from monitoring networks show that the decline in  $SO_2$  emissions from the power industry has improved air quality. Based on data from EPA's latest air emission trends report, the national composite average of  $SO_2$  annual mean ambient concentrations decreased 76 percent between 1980 and 2009, as shown in Figure 1 (based on state, local, and EPA monitoring sites located primarily

## At a Glance: ARP Results in 2009

**Air Quality:** Between 1989–1991 and 2007–2009, average ambient sulfate concentrations have decreased by 44 percent in the Mid-Atlantic, 47 percent in the Midwest, 49 percent in the Northeast, and 41 percent in the Southeast.

Acid Deposition: Between the 1989–1991 and 2007–2009 observation periods, regional decreases in wet deposition of sulfate across the Eastern United States averaged 43 percent.

**Surface Water Chemistry:** Levels of Acid Neutralizing Capacity (ANC), the ability of a water body to neutralize acid deposition, have increased significantly from 1990 to 2008 in lake and stream long-term monitoring sites in the Adirondack Mountains and the Northern Appalachian Plateau. These increasing ANC levels indicate trends toward recovery from acidification.

in urban areas). The largest single-year reduction (20 percent) occurred in the first year of the ARP, between 1994 and 1995. The second largest single-year reduction (16 percent) occurred most recently between 2008 and 2009. These trends are consistent with the regional ambient air quality trends observed in the Clean Air Status and Trends Network (CASTNET).

### Figure 1: National SO<sub>2</sub> Air Quality, 1980–2009





# Table 1: Regional Changes in Air Quality and Deposition of Sulfur and Nitrogen Compounds, 1989–1991 versus 2007–2009, from Rural Monitoring Networks

Measurement	Region	Average, 1989–1991	Average, 2007–2009	Percent Change	Number of Sites
Ambient SO <sub>2</sub> Concentra-	Mid-Atlantic	13	5	-62	12
tion (μg/m³)	Midwest	11	4.1	-63	10
	Northeast	5.5	1.7	-69	3
	Southeast	5.1	2.2	-57	8
Ambient Sulfate Concen-	Mid-Atlantic	6.3	3.5	-44	12
tration (µg/m³)	Midwest	5.8	3.1	-47	10
	Northeast	3.5	1.8	-49	3
	Southeast	5.4	3.2	-41	8
Wet Sulfate Deposition	Mid-Atlantic	9.2	5.3	-42	11
(kg⁻S/ha)	Midwest	7.1	4	-44	27
	Northeast	7.5	4.3	-43	17
	Southeast	6.1	3.5	-43	23
Dry Sulfur Deposition	Mid-Atlantic	6.7	2.9	-57	11
(kg⁻S/ha)	Midwest	6.5	2.8	-57	10
	Northeast	2.9	1	-66	3
	Southeast	1.2	0.7	-42	2
Total Sulfur Deposition	Mid-Atlantic	16	8	-50	11
(kg-S/ha)	Midwest	15	7	-53	10
	Northeast	9.8	4.7	-52	3
	Southeast	8	4.6	-43	2
Total Ambient Nitrate	Mid-Atlantic	3.3	2	-39	12
Concentration (Nitrate + Nitric Acid) (µg/m³)	Midwest	4.6	3.2	-30	10
	Northeast	1.8	1	-44	3
	Southeast	2.2	1.5	-32	8
Wet Inorganic Nitrogen	Mid-Atlantic	6.2	4.5	-27	11
Deposition (kg-N/ha)	Midwest	5.8	4.9	-16	27
	Northeast	5.6	4.1	-27	17
	Southeast	4.4	3.4	-23	23
Dry Inorganic Nitrogen Deposition (kg-N/ha)	Mid-Atlantic	2.5	1.5	-40	11
	Midwest	2.5	1.8	-28	10
	Northeast	1.4	0.6	-57	3
	Southeast	0.9	0.8	-11	2
Total Inorganic Nitrogen	Mid-Atlantic	8.7	6	-31	11
Deposition (kg <sup>-</sup> N/ha)	Midwest	9	6.9	-23	10
	Northeast	6.5	4.4	-32	3
	Southeast	5.9	4.8	-19	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 and values 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.



#### Figure 2: Annual Mean Ambient SO<sub>2</sub> Concentration

- 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, 2010

Dramatic regional improvements in  $SO_2$  and ambient sulfate concentrations were observed following implementation of Phase I of the ARP during the late 1990s at CAST-NET sites throughout the eastern United States, and 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 are compared

from 1989 to 1991 and 2007 to 2009 in both tabular form and graphically in maps (see Table 1 and Figures 2 and 3).

The maps in Figure 2 show that the average annual ambient concentrations of  $SO_2$  from 1989 to 1991 were highest 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.



#### 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, 2010

Like SO<sub>2</sub> concentrations, the highest average annual ambient sulfate concentrations from 1989 to 1991 were observed in western Pennsylvania and along the Ohio River Valley. Most of the eastern United States experienced annual ambient sulfate concentrations greater than 5 micrograms per cubic meter ( $\mu$ g/m<sup>3</sup>).

Ambient sulfate concentrations have also decreased since the program was implemented, with average concentrations decreasing from 41 to 49 percent in regions of the East (see Table 1). 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).

#### **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. Ambient nitrate levels can also 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 Clean Air Interstate Rule (CAIR) have continued to result in significant region-specific improvements in ambient total nitrate ( $NO_3$ - plus HNO<sub>3</sub>) concentrations. For instance, annual mean ambient total nitrate concentrations for 2007 to 2009 in the Mid-Atlantic region were 39 percent less than the annual mean concentration in 1989 to 1991 (see Table 1 and Figure 4). While 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.

## **Acid Deposition**

National Atmospheric Deposition Program/National Deposition 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 2007 to 2009 observation periods, average decreases in wet deposition of sulfate averaged more than

### Figure 5: Annual Mean Wet Sulfate Deposition



Source: NADP, 2010

Figure 6: Annual Mean Wet Inorganic Nitrogen Deposition





43 percent for the eastern United States (see Table 1 and Figure 5). Along with wet sulfate deposition, wet sulfate concentrations have also decreased by similar percentages. A strong correlation between large-scale SO<sub>2</sub> 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 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 50 and 53 percent, respectively (see Table 1). 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  $SO_2$  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 31 and 23 percent decrease in total nitrogen deposition for the Mid-Atlantic and Midwest, respectively (see Table 1).

## **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 59 of the monitoring stations; the National Park Service (NPS) funds and operates approximately 25 stations in cooperation with EPA. Many CASTNET sites have a continuous 20-year data record, reflecting EPA's commitment to long-term environmental monitoring.

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 (USDA), 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 the NADP's website.

NADP is running a pilot study to determine the feasibility of operating a long-term passive ammonia (NH<sub>3</sub>) monitoring network. The pilot network (AMoN) has been measuring 2-week samples of ambient NH<sub>3</sub> for over two years at more than 20 sites. It will be the first nationwide network to measure NH<sub>3</sub> routinely. More information on AMoN can be found on the NADP website.

	Chemical Symbol	Measured in:				
Chemical Name		Ambient Air	Wet Deposition	Why are these measured by the networks?		
Sulfur Dioxide	SO <sub>2</sub>	Х		Primary precursor of wet and dry deposition; primary precursor of fine particles ( $PM_{2.5}$ ).		
Sulfate Ion	S042-	Х	Х	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	NO3-	Х	Х	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 <sub>3</sub>	Х		Strong acid and major component of dry nitrogen deposition; formed as a secondary		
				product from NO <sub>x</sub> in the atmosphere.		
Ammonium Ion	NH <sub>4</sub> +	Х	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.		
lonic Hydrogen	H+		Х	Indicator of acidity in precipitation; formed from the reaction of sulfate and nitrate in water.		
Calcium	Ca <sub>2</sub> +	Х	Х	These base cations neutralize acidic compounds in precipitation and the environment;		
Magnesium	Mg <sub>2</sub> +	Х	Х	also play a major role in plant nutrition and soil productivity.		
Potassium	K+	Х	Х			
C 11	Na+	X	X			

#### **Table 2: Air Quality and Acid Deposition Measures**

## **Ambient Mercury Monitoring**

In addition to SO<sub>2</sub> and NO<sub>x</sub>, coal-fired power plants release mercury into the atmosphere where it can be transported and deposited locally, regionally, and globally. NADP recently launched the Atmospheric Mercury Network (AMNet) for monitoring three atmospheric mercury species: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM<sub>2.5</sub>). Data sets 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 in North America (as shown in the map in Figure 7) participate in the network, generating high-resolution, high-quality speciated atmospheric data. In 2010, the University of California - Santa Cruz was the most recent partner to join AMNet, establishing a new network site in Elkhorn Slough, California.

#### **The AMNet Database**

The AMNet database has received extensive quality assurance (QA)/quality control (QC) review by a team of data quality experts from EPA, USGS, NADP, and other institutions. Quality control flags to determine whether datum records are valid or invalid were developed based on peerreviewed criteria. The AMNet QA/QC review of data is a robust, tiered approach, including: automatic screening of each raw datum and quality control flagging; additional rigorous automatic screening, accounting for calibration data and other checks (e.g., extensive 24-hour trap bias, etc.) and quality control flagging; and NADP site liaison manual review of data, where automatically screened data are reviewed against corresponding monthly site operator field report notes (e.g., glassware changes, site shelter power failure, etc.).



Source: NADP/AMNet, 2010

## **Figure 7: Ambient Mercury Monitoring Locations**

At present, more than 56 site years of data have been quality assured. NADP now offers mercury speciation data products available through the AMNet website (nadp.sws. uiuc.edu/amn/), including bi-hourly graphical plots and data tables for each site.

#### Beltsville, Maryland Case Study

Beltsville, MD is home to NADP/AMNet (site ID: MD99), CASTNET (site ID: BEL116), NADP/Mercury Deposition Network (site ID: MD99), and other monitoring sites where mercury, meteorology data, and other ancillary data are recorded. The data plot shown below represents an important example of the value of maintaining complementary, collocated monitoring network sites. In this example, collocated AMNet bi-hourly gaseous oxidized mercury (GOM) and CASTNet hourly SO2 values (Figure 8) from Beltsville are plotted over the same time period. GOM is measured in picograms per cubic meter ( $pg/m^3$ ). SO<sub>2</sub> is a trace gas that is often used as a signature of coal-fired power plants. The plot shows both GOM and SO<sub>2</sub> following a very similar pattern over the same sample time period, which warrants further analysis to determine if there is a relationship between the ambient data and emissions. The measurements collected at AMNet can also be used with source-receptor modeling and other tools to provide useful information on source attributions and dry/total deposition estimates for sensitive ecosystems.

#### Figure 8: Bi-Hourly Gaseous Oxidized Mercury Concentrations and Hourly CASTNET SO<sub>2</sub> Gas Concentrations at the AMNet Site ID MD99/CASTNET Site ID BEL116



Source: NADP/AMNet, 2010

## **Improvements in Surface Water Chemistry**

Acid rain resulting from  $SO_2$  and  $NO_x$  emissions is one of many large-scale anthropogenic impacts 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 9–12).

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 CAA Amendments in reducing the acidity of surface waters in four regions: New England, the Adirondack Mountains, the Northern Appalachian Plateau, and the Central Appalachians (the Ridge and Valley and Blue Ridge Provinces). 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 (see Figure 9).

The data presented here show regional trends in acidification from 1990 to 2008 in lakes and streams sampled through the LTM program (see Figures 10–12). Only sites that have a complete data record for the time period are represented. Three indicators of acidity in surface waters







Figure 10: Trends in Lake and Stream Water Chemistry at LTM Sites, 1990–2008 — Sulfate Ion Concentration (µeq/L/yr)

Source: EPA, 2010

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.

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.<sup>1</sup> 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 from 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 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 microequivalents<sup>2</sup> per liFigure 11: Trends in Lake and Stream Water Chemistry at LTM Sites, 1990–2008 — Nitrate Ion Concentration (µeq/L/yr)



Source: EPA, 2010



Figure 12: Trends in Lake and Stream Water Chemistry at LTM

Sites, 1990–2008 — ANC Levels (µeq/L/yr)

Source: EPA, 2010

ter ( $\mu$ eq/L) are of acute concern for acidification. Lakes and streams having springtime ANC values less than 50  $\mu$ eq/L are generally considered of elevated concern for acidification. Lakes and streams with ANC higher than 50  $\mu$ eq/L are generally considered of moderate to low concern for 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 increasing trends in ANC and decreasing trends in sulfate and nitrate.

Region	Waterbodies Covered	% of Sites with Improving Sulfate Trend	% of Sites with Improving Nitrate Trend	% of Sites with Improving ANC Trend	% of Sites with Improving DOC Trend
Adirondack Mountains	50 lakes in NY	90%	32%	58%	42% (26 sites)
Catskills / N. Appalachian Plateau	9 streams in NY and PA	78%	33%	56%	29% (7 sites)
New England	26 lakes in ME and VT	96%	31%	12%	20% (10 sites)
Central Appalachians	66 streams in VA	12%	45%	12%	NA

#### Table 3: Regional Trends in Sulfate, Nitrate, ANC, and DOC at Long-term Monitoring Sites, 1990–2008

Notes:

- Trends are determined by multivariate Mann-Kendall tests.
- DOC was only examined in low-ANC waterbodies (ANC less than 25  $\mu eq/L).$
- DOC is not currently measured in Central Appalachian streams. Source: EPA, 2010

Dissolved organic carbon (DOC), essentially organic material, is derived from many sources, some of which include: atmospheric deposition, decaying leaf litter, soil organic matter, aquatic sediments, and aquatic organisms. DOC is an important part of the acid-base chemistry of most lowalkalinity freshwater systems. A host of factors control DOC in surface water including the inputs from acidifying deposition, discharge, temperature, and nutrient enrichment. Recently, scientists have suggested that increased concentrations of DOC are likely due to declining sulfate content from atmospheric deposition, increasing seasonal temperatures, or a combination of both.<sup>3,4</sup> With increasing loading of acid deposition, soils release lower quantities of organic acids, thereby causing DOC to decrease in surface water. This means that as surface waters recover from acidic deposition, DOC concentrations are returning toward pre-industrial levels. On the other hand, as temperatures warm, more organic acids in the soil break down and are released to surface waters and increase DOC concentrations. These increasing DOC concentrations could be a possible sign of climate change. Another mechanism that could cause increases in DOC is a soil microbial response to nitrogen deposition that results in greater export of humic material to surface waters.<sup>5</sup>

DOC is an important water chemistry parameter and may be affected by acidification. Table 3 presents the aggregate sulfate, nitrate, ANC, and DOC trends represented by the LTM sites shown in Figures 10–12 for four acid sensitive regions of the eastern United States, as well as DOC trends for low ANC (ANC<25  $\mu$ eq/L) waterbodies.

## **Acidification of Soils**

Soils are also affected by acidic deposition. As acidic deposition enters the soil, it can cause base cations, such as calcium ( $Ca^{2+}$ ), magnesium ( $Mg^+$ ), or potassium (K<sup>+</sup>), to exchange with H<sup>+</sup> and Al<sup>3+</sup> ions. This causes base cations to be lost from the soil through transport to surface water, which may lead to declines in soil pH. As acidic deposition continues, more base cations exchange, causing a further decline in base cation adsorption to the soil surface (i.e., percent base saturation). This is important because base cations are important plant nutrients and their loss is a potential threat to forest productivity and health.<sup>10</sup> Additionally, the immobilization of Al<sup>3+</sup> by acidic deposition can also be toxic to plants. While surface waters have experienced some chemical recovery (Table 3), soils are still likely acidifying.<sup>11</sup> In 2009, researchers at Syracuse University published results from a 2001 soil survey for soils in 139 watersheds across the northeastern United States. Many of these watersheds had previously been sampled as part of the EPA's Direct/Delayed Response Project in 1984. This comparative study showed that over the 17-yr interval, median base saturation in the Oa-horizon (organic surface soil layer) exhibited a statistically significant decrease from 56% in 1984 to 33% in 2001. Soil pH also decreased from 3.05 to 2.95 pH units over the same time period. These results are consistent with other research, showing continued soil acidification for this region.<sup>12</sup> Soil acidification is likely to continue until acidic deposition inputs to soils decline to the point where soil base cation pools are sufficient to neutralize them.

The maps and summary results indicate that:

- Sulfate concentrations are declining at most<sup>6</sup> sites in the Northeast (New England, Adirondacks, Catskills/ Northern Appalachian Plateau). However, in the Central Appalachians, sulfate concentrations in some streams (21%) are increasing. This region has 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. Thus sulfate concentrations in surface waters, mainly streams in this region, are increasing despite reduced sulfate deposition.
- Nitrate concentrations are decreasing in some of the sites in all 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. In 2008, 45% of the Central Appalachian streams had a decreasing trend in nitrate, compared to 24% in 2007. This increase in the number of sites with a decreasing nitrate trend may be due to continued recovery following gypsy moth defoliation in the early 1990s. Gypsy moth defoliation has been shown to increase nitrate export from affected forests to surface waters by as much as 50 times.<sup>7</sup> While defoliation from gypsy moths may only occur over several months, impacts on nitrate transport and in-stream concentrations may be seen for many years.8
- ANC, as measured in surface waters, is increasing in many of the sites in the Adirondack and Catskills/Northern Appalachian Plateau regions, which in part can be attributed to declining sulfate deposition. The site trends also indicate variation within each region. Only 12% of sites in New England and the Central Appalachians have improving ANC trends, but overall, only seven sites in all regions have a significant downward trend in ANC.
- DOC is increasing in only about 20% to 42% of the low ANC lakes and streams of the Adirondack Mountains, Catskills/Northern Appalachian Plateau, and New England. The Adirondack Mountains have the highest percentage (42%) of lakes with an increasing DOC trend. These results suggest that the change of DOC in the LTM catchments is complex, with the majority of low ANC waterbodies not changing over the past 18 years. Of the lakes and streams with increasing DOC, no single envi-

ronmental factor is likely for the cause of the increase. Declines in sulfate deposition (Figure 5) and warmer seasonal and annual temperatures may have contributed to the rise in surface DOC.<sup>9</sup>

The ANC of northeastern U.S. lakes monitored under the TIME program was also evaluated for the 1991–1994 and 2006-2008 periods to assess the impacts of ARP implementation. The analysis in Figure 13 compares average ANC levels for the northeastern lakes that had data in each time period. From 1991 to 1994, 7.5 percent of lakes 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 acidtolerant forms (see Table 4). The percentage of lakes in this category dropped to 4.3 percent in 2006 to 2008 (see Figure 13). Additionally, the percentage of elevated concern lakes dropped from 13.8 percent for the 1991-1994 time period to 10.1 percent from 2006-2008, while the percentage of lakes with in the moderate concern category increased from 6.4 to 13.5 percent. These results point to a decrease in acidity, particularly for the subset of lakes with low ANC.

# Figure 13: Northeastern Lakes by ANC Status Category, 1991–1994 versus 2006–2008



Notes:

- Based on 305 EMAP/TIME monitoring sites.
- 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.

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 vari-
		able, 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.

### **Table 4: Aquatic Ecosystem Status Categories for Northeastern Lakes**

Source: EPA, 2010

# **Critical Loads and Exceedances**

The Northeast and Mid-Appalachian Highlands in the eastern United States has been strongly affected by acidic deposition. Many of the small forested watersheds, particularly along the Appalachian Mountain spine, have soils and surface waters that are unable to buffer the acidity from acidic deposition, causing the ecosystem to acidify. As a result the health of some tree species and aquatic biota have declined or species, such as brook trout, are no longer present in the ecosystem.

Since the early 1980s, acidic deposition has acidified many lakes and many miles of streams in the Northeast and Mid-Appalachian Highlands.<sup>13</sup> However, with the implementation of Title IV and other emission reduction programs, acidic deposition has declined throughout the eastern United States as emissions of NO<sub>x</sub> and SO<sub>2</sub> have declined (see Acid Rain and Related Programs: 2009 Emission, Compliance, and Market Analyses report available at < www. epa.gov/airmarkets/progress/ARP09\_2.html>). Surface waters across the region have also shown signs of recovery as indicated by declining sulfate concentrations and increasing ANC levels (see Table 3). In the 2007 and 2008 Acid Rain Progress Reports, critical loads were used to gauge the extent to which the acid sensitive areas of the Adirondack Mountains and Central Appalachian Mountains have recovered from acid deposition. Results from these regions indicate that 10-15% of monitored lakes and streams now receive levels of acid deposition that are generally low enough that aquatic ecosystems are protected in comparison to deposition levels in the early 1990s.

The critical load approach is an assessment tool that can be used to determine the degree to which air pollution may be affecting ecological health. 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.<sup>14</sup> 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. Building on past critical load studies, this analysis explores the extent to which lakes in the Northeast and streams in the Mid-Appalachian Highlands in Virginia and West Virginia are protected from acidifying nitrogen and sulfur deposition as a result of recent emission reductions.

The critical load for a lake or stream provides a benchmark against which to assess the extent to which a waterbody is potentially at risk due to current acidic 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. If pollutant exposure is less than the critical load, adverse ecological effects (e.g., reduced reproductive success, stunted growth, loss of biological diversity) are not anticipated, and recovery is expected over time if an ecosystem has been damaged by past exposure. A critical load exceedance is the measure of pollutant exposure above the critical load. This means pollutant exposure is higher than, or "exceeds," the critical load and the ecosystem continues to be exposed to damaging levels of pollutants.

The scientific research community has recently completed and published many peer-reviewed scientific articles that advance the tools for calculating critical loads in the United States. Drawing on the methods from the peer-reviewed scientific literature,<sup>15</sup> critical loads were calculated for over 1,300 lakes and streams using the Steady-State WaFigure 14: Lake and Stream Exceedences of Estimated Critical Loads (Sulfur + Nitrogen) for Total Nitrogen and Sulfur Deposition for the Period 1989–1991

#### Critical Load Exceedances, 1989-1991

- Does Not Exceed Critical Load
- Within 10% of the Critical Load
- Exceeds Critical Load



Source: EPA, 2010

ter Chemistry (SSWC) model and the Model of Acidification of Groundwater In Catchments (MAGIC) model. These critical load estimates represent only lakes and streams where surface water samples have been collected and do not represent all types of lakes and streams in the study region. Water quality chemistry include data collected by EPA-administered surface water monitoring and assessment programs, such as the National Surface Water Survey (NSWS), Environmental Monitoring and Assessment Program (EMAP), the TIME program, and the LTM program. The lakes and streams associated with these programs consist of a subset of lakes and streams that are located in areas most affected by acid deposition and many sites provide long term records of surface acidification. The NSWS and EMAP programs employ probability sampling; each monitoring site was chosen statistically from a predefined target population. In New England and the Mid-Appalachian Highlands, the target populations include lakes and streams likely to be responsive to changes in acidic deposition.

Figure 15: Lake and Stream Exceedences of Estimated Critical Loads (Sulfur + Nitrogen) for Total Nitrogen and Sulfur Deposition for the Period 2007–2009

#### Critical Load Exceedances, 2007-2009

- Does Not Exceed Critical Load
- Within 10% of the Critical Load
- Exceeds Critical Load



Source: EPA, 2010

For this particular analysis, the critical load represents the combined deposition loads of sulfur and nitrogen to which a lake or stream could be subjected and still have a calculated ANC of 50  $\mu$ eq/L or higher. While a critical load can be calculated for any ANC level, this level was chosen because it tends to protect most fish and other aquatic organisms, although systems can 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 calculated ANC level of 50  $\mu$ eq/L or above. In order to assess the extent to which regional lake and stream ecosystems are protected by the emission reductions achieved by Title IV, this case study compares the amount of deposition systems can receivethe critical load—to measured deposition for the period before implementation of the ARP (1989 to 1991) and for a recent period after ARP implementation (2007 to 2009).

Overall, this critical load analysis shows that emission reductions achieved by the ARP have resulted in improved environmental conditions and increased ecosystem protection in the Northeast and Mid-Appalachian Highlands. For the period from 2007 to 2009, 26% of the waterbodies examined received levels of combined sulfur and nitrogen deposition that exceeded the critical load (Figure 15). This is an improvement when compared to the 1989-1991 period before implementation of Title IV, during which 42% of waterbodies exceeded the critical load (Figure 14). Thus, during the 2007 to 2009 period, 37% of those waterbodies exceeding their critical load in the previous period were no longer receiving sulfur and nitrogen deposition loads that threaten the health of these ecosystems. Areas with the largest concentration of lakes where acid deposition currently is greater than-or exceeds-estimated critical loads include the Adirondack mountain region in New York, southern New Hampshire and Vermont, northern Massachusetts, northeast Pennsylvania, and the central Appalachian Mountains of Virginia and West Virginia (Figure 15).

Reductions in acidic deposition have occurred over the past decade, as demonstrated by the deposition maps in Figures 5 and 6 on page 5. However, this comparison of past and current total deposition estimates with critical loads estimates indicates that acid-sensitive ecosystems in the Eastern United States are still at risk of acidification at current deposition levels. As a result, additional reductions in acidic deposition from current levels might be necessary to fully protect these ecosystems.

## **Ammonium Deposition in the Eastern United States**

Ammonium (NH<sub>4</sub><sup>+</sup>) forms when ammonia gas (NH<sub>3</sub>) reacts with sulfur, nitrogen and other acidic compounds in the atmosphere. Ammonium is of concern because it contributes to the formation of fine particles, which have negative human health effects and can cause reduced visibility. Wet deposition of NH4<sup>+</sup> is measured at NTN sites across the US while dry deposition of NH<sub>4</sub><sup>+</sup> is estimated from weekly filter pack concentrations measured at CASTNET sites and estimated deposition velocities. While total nitrate deposition fluxes have decreased 24% in the eastern US, wet deposition has only decreased 4% since 1990. The 36 CAST-NET sites used for this analysis are shown in Figure 16. Figure 17 shows the wet and dry NH<sub>4</sub><sup>+</sup> deposition flux from 36 CASTNET sites between 1990 and 2009. The three year average for wet and dry NH<sub>4</sub><sup>+</sup> flux between 1990–1992 is 2.34 and 0.42 kg-N/ha, respectively. The most recent three year average, 2007–2009, the wet  $NH_4^+$  flux is 2.24 and the dry flux is 0.31 kg-N/ha. While there is a small decrease

in dry deposition (0.11 kg-N/ha), the 3 year average total nitrogen deposition decreased from 7.62 to 5.80 kg-N/ha (1.82 kg-N/ha), a larger reduction likely attributed to the large reductions we have seen in particulate nitrate (NO<sub>3</sub><sup>-</sup>) deposition fluxes. The NH<sub>4</sub><sup>+</sup> ion is the basic component of PM<sub>2.5</sub> formation, causing negative health effects and reduced visibility. In addition, NH<sub>4</sub><sup>+</sup> leads to eutrophication of ecosystems. While NO<sub>x</sub> emissions under the Acid Rain Program, the NO<sub>x</sub> Budget Trading Program, and CAIR have resulted in significant improvements in air quality, gaseous NH<sub>3</sub> emissions have been increasing. This will provide a

# Figure 16: Eastern U.S. CASTNET Monitoring Locations Analyzed for Ammonium Deposition



Source: EPA, 2010

# Figure 17: Measured Wet and Estimated Dry Deposition of Ammonium at Eastern CASTNET sites, 1990–2009





challenge in the future for understanding changes in atmospheric chemistry and nitrogen deposition to forests, lakes, streams and other sensitive areas. Therefore, it is important to continue monitoring long-term trends in air quality and deposition to further understand pollutants which contribute to poor air quality.

# **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 Acid Rain Program. CAMD then makes these data available to the public in readily usable and interactive formats. The CAMD website 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.

To increase data transparency, EPA has created supplementary maps that allow the user to display air market program data geospatially on an interactive 3D platform. Figures 18 and 19 are examples of these 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 this tool, visit the website at <www.epa.gov/airmarkets/progress/interactivemapping.html>.



Note: This example depicts  $1990 \text{ SO}_2$  emissions from ARP sources along with 1990 sulfate concentration data as measured by the CASTNET monitoring program.

In another effort to increase data transparency, EPA regularly posts updates of quarterly  $SO_2$  and  $NO_x$  emissions data from coal-fired power plants controlled under the ARP and other programs to make it easy for the public to track changes in emissions from these sources (available at: <www.epa.gov/airmarkets/quarterlytracking.html>). The data presented on the quarterly emissions tracking website compare emissions, emission rates, and heat input from power plant units in the ARP. These files graphically and spatially compare quarterly emission data from the most recent completed quarter of 2010 with data for the same quarter from 2008.

Interactive motion charts are a key feature on the quarterly tracking website. Figure 20 on page 17 shows examples of motion charts created to show changes in ARP SO<sub>2</sub> emissions and SO<sub>2</sub> emission rates over time (from 1990 to

2009). These motion charts show, historically, how coalfired power plants have responded to the ARP. Each circle on the motion chart represents a facility in the ARP with one or more units that burn coal to create electricity. The size and color of these circles tell us something about the facility. To the right of the motion chart you'll find two legends. The color spectrum at the top represents the emissions generated per unit of fuel (also known as the SO<sub>2</sub> emission rate), with warmer colors (yellow through red) representing a high emission rate and cooler colors (green through blue) representing a low emission rate. The size of the circle on the chart is proportional to the emissions from that plant. On the interactive mapping website, the user can watch this data move through time by clicking the play button.

For more information or to utilize these and other tools, visit the website at < www.epa.gov/airmarkets/>.



Note: This example depicts 2009 SO<sub>2</sub> emissions from ARP sources along with 2009 sulfate concentration data as measured by the CASTNET monitoring program.

Source: EPA, 2010

# Figure 19: US SO<sub>2</sub> Emissions and Sulfate Concentrations, 2009

#### Figure 20: Motion Charts of Annual ARP Coal-Fired Emissions, SO<sub>2</sub> Emission Rates and Heat Input over Time, 1990 and 2009



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Source: EPA, 2010

## Notes

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<sup>2</sup> 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.

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 $^6$  "Most", "many", "some", "few" improving indicate greater than 75%, from 50–75%, from 25–50%, and less than 25% of lakes or streams with a statistically significant trend at the 95% confidence level respectively.

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