United States
Environmental Protection
Agency

Clean Air Interstate Rule, Acid Rain Program, and Former NO_X Budget Trading Program

2012
PROGRESS REPORT

Environmental and Health Results

Photo credit: S.J. Nelson

At a Glance:

Environmental and Health Results in 2012

By reducing SO_2 and NO_x emissions (precursors to $PM_{2.5}$ and ozone formation) the ARP, NBP, and CAIR significantly benefit human health and welfare.

Air Quality: Between 1989 to 1991 and 2010 to 2012, average ambient particulate sulfate concentrations have decreased by 59 percent in the Mid-Atlantic, 57 percent in the Midwest, 63 percent in the Northeast, and 56 percent in the Southeast.

In CAIR states, 99th percentile average 1-hour ozone concentrations decreased by 18 percent between 2000 to 2002 and 2010 to 2012.

Acid Deposition: Between 1989 to 1991 and 2010 to 2012, wet deposition of sulfate decreased by 59 percent across the Eastern United States.

Surface Water Chemistry: Levels of Acid Neutralizing Capacity (ANC) have increased significantly from 1990 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.

Program Basics

The Clean Air Interstate Rule (CAIR) and the Acid Rain Program (ARP) are both cap and trade programs designed to reduce emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) from power plants.

The ARP, established under Title IV of the 1990 Clean Air Act (CAA) Amendments, requires power plants to make major emission reductions of SO_2 and NO_x, the primary precursors of acid rain. The SO_2 program sets a permanent cap on the total amount of SO_2 that may be emitted by electric generating units (EGUs) in the contiguous United States. The program is phased in, with the final 2010 SO_2 cap set at 8.95 million tons, a level of about one-half of the emissions from 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 traditional, rate-based regulatory system. Since the program began in 1995, the ARP has achieved significant emission reductions. For more information on the ARP, please visit the ARP website at <www.epa.gov/airmarkets/progsregs/arp/index.html>.

The NO_x Budget Trading Program (NBP) operated from 2003 to 2008. The NBP was a cap and trade program that required NO_x emission reductions from power plants and industrial units in the eastern U.S. during the summer months. For more information on the NBP, please visit the NO_x Budget Trading Program/NO_x SIP Call website at <www.epa.gov/airmarkets/progsregs/nox/index.html>.

CAIR addresses regional interstate transport of soot (fine particulate matter) and smog (ozone) pollution. CAIR requires certain eastern states to limit annual emissions of SO_2 and NO_x, which contribute to the formation of fine particulate matter. It also requires certain states to limit ozone season NO_x emissions, which contribute to the formation of ozone during the summer ozone season (May through September). CAIR includes three separate cap and trade programs to achieve the required reductions: the CAIR SO_2 annual trading program, the CAIR NO_x annual trading program, and the CAIR NOx ozone season trading program. The CAIR SO_2 annual program began in 2010, while the CAIR NO_x annual and ozone season programs began in 2009. The reduction in fine particulate matter ($PM_{2.5}$) and ozone formation resulting from CAIR implementation provides health benefits as well as improved visibility in national parks and improvements in freshwater aquatic ecosystems in the eastern U.S. For more information on CAIR, please visit the CAIR website at <www.epa.gov/airmarkets/progsregs/cair/index.html>.

Health Benefits of the ARP, NBP, and CAIR

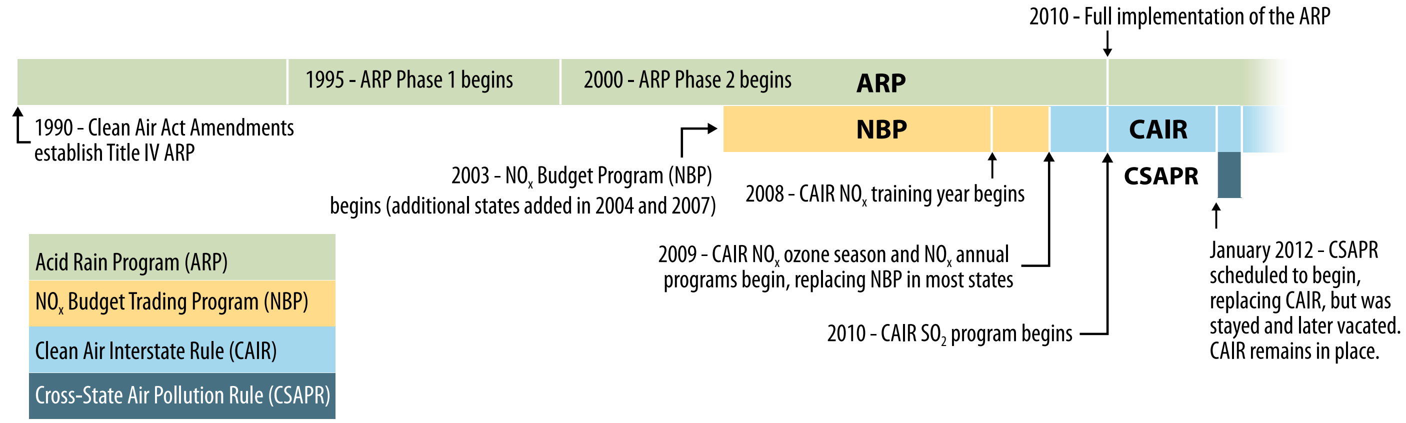
By reducing precursors (SO_2 and NO_x) to $PM_{2.5}$ formation and a precursor (NO_x) to ground-level ozone formation, emission reductions achieved by the ARP, NBP, and CAIR significantly benefit human health and welfare.[1,2,3] Exposure to $PM_{2.5}$ and ozone is linked to premature death, as well as a variety of non-fatal effects including heart attacks, hospital and emergency department visits for respiratory and cardiovascular symptoms, acute bronchitis, aggravated asthma, and days when people miss work or school.[4,5]

Cross-State Air Pollution Rule and Litigation

EPA issued the Cross-State Air Pollution Rule (CSAPR) in July 2011, requiring 28 states in the eastern half of the U.S. to significantly improve air quality by reducing power plant emissions that cross state lines and contribute to fine particle and ozone pollution in other states. CSAPR includes three separate cap and trade programs to achieve these reductions: the CSAPR SO_2 annual and NO_x annual trading programs to address fine particle pollution and the CSAPR NO_x ozone season trading program to address ozone pollution. The CSAPR trading programs were scheduled to replace the CAIR trading programs starting on January 1, 2012. However, on December 30, 2011, the U.S. Court of Appeals for the District of Columbia Circuit (D.C. Circuit) stayed CSAPR pending judicial review and on August 21, 2012 the court issued a decision vacating the rule.[6] On April 29, 2014, the U.S. Supreme Court reversed the D.C. Circuit decision.[7] EPA is reviewing the Supreme Court opinion and will determine any next steps once that review is complete. At this time, CAIR remains in place and no immediate action from States or affected sources is expected. For more information on CSAPR, please visit the CSAPR website at <www.epa.gov/airtransport/CSAPR/index.html>.

Figure 1 contains important milestones for ARP, NBP, CAIR, and CSAPR.

Figure 1: History of ARP, NBP, CAIR, and CSAPR



Source: EPA, 2013

Next Steps in Addressing Interstate Air Pollution Transport

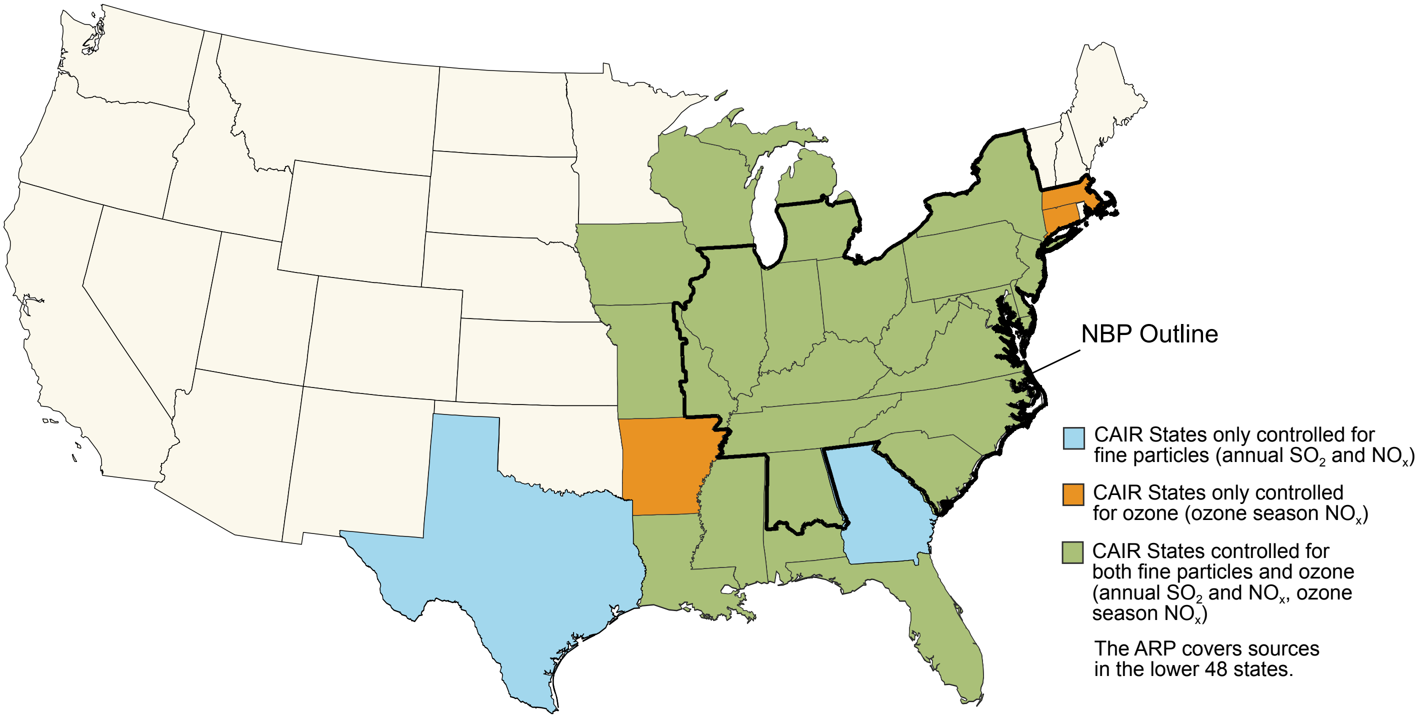
EPA is working with state partners on next steps to address air pollution that crosses state boundaries—particularly with respect to recently promulgated health-based air quality standards.

After considering input from states and other stakeholders, EPA is on a path to define upwind states' obligations under the 2008 ozone standards to address transported air pollution affecting the eastern half of the U.S. This path will lead to a proposed rulemaking in late summer 2014. In addition, EPA will continue supporting efforts across the U.S. that reduce SO_2 and NO_x emissions by implementing existing programs, finalizing pending rules, and working with regional, state, and local air quality planners to evaluate the need for additional clean air actions.

ARP, NBP, and CAIR Affected States

The ARP is a nationwide program affecting large fossil fuel-fired power plants across the country. CAIR covers 27 eastern states and the District of Columbia (D.C.) and requires reductions in annual emissions of SO_2 and NO_x from 24 states and D.C. (to achieve improvements in fine particle pollution in downwind areas) and emission reductions of NO_x during the ozone season from 25 states and D.C. (to achieve improvements in ozone pollution in downwind areas). The former NBP affected 20 eastern states and D.C. State coverage for CAIR, ARP, and NBP is shown in Figure 2.

Figure 2: ARP, NBP, and CAIR States



Source: EPA, 2013

2012 Progress Reports

Each year EPA releases reports summarizing progress under both CAIR and the ARP. In the 2012 reports, EPA presents emissions and compliance data for both CAIR and the ARP to show reductions in power sector emissions of SO_2 and NO_x and the effect of these programs on air quality and the environment. While several other programs contribute to NO_x and SO_2 emission reductions and improved air quality (e.g., mobile source emission control programs), this series of reports focuses on achievements related to emission reductions at power sector sources under CAIR, the ARP, and the former NBP.

The first report in this series, the *CAIR, ARP, and Former NBP 2012 SO_2 and NO_x Emissions, Compliance, and Market Analyses Report*, was released in December 2013 and presented 2012 data on combined emission reductions and compliance results for CAIR and the ARP. It also showed some historic NBP emissions data and evaluated shared progress under these programs in 2012 by analyzing emission reductions and market activity. The *Environmental and Health Results Report* provides further 2012 trends analysis by comparing changes in emissions to changes in air quality, acid deposition, and surface water chemistry.

Air Quality

To evaluate the impact of emission reductions on the environment, scientists and policymakers use data collected from long-term national air quality and deposition monitoring networks (see text box). 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 over time and in different areas.

Sulfur Dioxide

SO_2 is one of a group of highly reactive gases known as “oxides of sulfur.” The largest sources of SO_2 emissions are fossil fuel combustion at power plants (60 percent) and other industrial facilities (19 percent). Smaller sources of SO_2 emissions include industrial processes such as extracting metal from ore, and the burning of high sulfur containing fuels by locomotives, large ships, and non-road equipment. In addition to contributing to the formation of fine particle pollution ($PM_{2.5}$), SO_2 is linked with a number of adverse health effects on the respiratory system. See the “Particulate Matter” section on page 14 for more information on the health effects of $PM_{2.5}$ pollution.

Data collected from monitoring networks show that the decline in SO_2 emissions from the power sector has improved air quality. Based on EPA’s latest air trends data, the national composite average of SO_2 annual mean ambient concentrations decreased 85 percent between 1980 and 2012, as shown in Figure 3 (based on state, local, and EPA monitoring sites located primarily in urban areas). The two largest single-year reductions (over 20 percent reduction each) occurred in the first year of the ARP, between 1994 and 1995, and recently between 2008 and 2009, just prior to the start of the CAIR SO_2 program. These trends are consistent with the regional ambient air quality trends observed in the Clean Air Status and Trends Network (CASTNET).

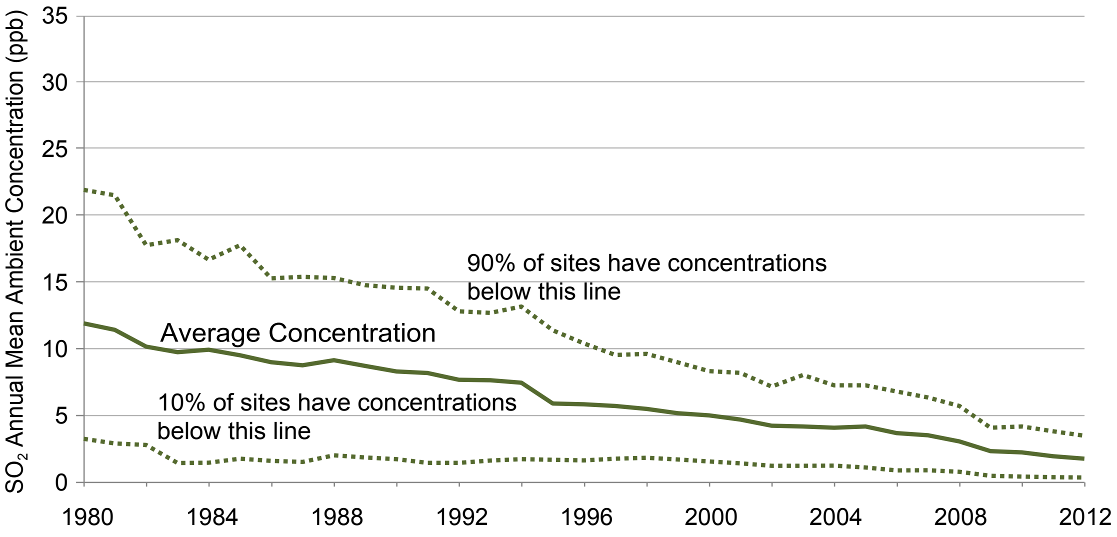
About Long-term Ambient and Deposition Monitoring Networks

The Clean Air Status and Trends Network (CASTNET) provides long-term monitoring of air quality in rural areas to determine trends in regional atmospheric nitrogen, sulfur, and ozone concentrations and deposition fluxes (the rate of particles and gases being deposited to a surface) of sulfur and nitrogen pollutants in order to evaluate the effectiveness of national and regional air pollution control programs. CASTNET now operates 90 regional sites throughout the contiguous United States, Alaska, and Canada. Sites are located in areas where urban influences are minimal. Information and data from CASTNET are available on the CASTNET website at <www.epa.gov/castnet>.

The Air Quality System (AQS) is a database that contains ambient air pollution data collected by EPA, state, local, and tribal air pollution control agencies from thousands of monitoring stations. EPA hosts information and data from AQS, available at the Air Quality System website at <www.epa.gov/ttn/airs/airsaqs>.

The National Atmospheric Deposition Program/National Trends Network (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. NADP/NTN has grown 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 at <nadp.isws.illinois.edu/ntn>.

Figure 3: National SO_2 Air Quality



Source: EPA, 2013

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 CASTNET 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 (before implementation of the ARP) and 2010 to 2012 (most recent available data) in Table 1, on page 6.

Average ambient SO_2 concentrations declined in all regions following implementation of the ARP and other emission reduction programs. The most dramatic decline was along the Ohio River Valley and in western Pennsylvania.

Like SO_2 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 (μg/m^3).

Ambient particulate sulfate concentrations have also decreased since the ARP was implemented, with average concentrations decreasing by 56 to 63 percent in observed regions. Both the magnitude and spatial extent of the highest concentrations have dramatically declined, with the largest decreases observed along the Ohio River Valley.

Table 1: Regional Changes in Air Quality and Deposition of Sulfur and Nitrogen Compounds, 1989–1991 versus 2010–2012, from Rural Monitoring Networks

Measurement	Region	Annual Average, 1989–1991	Annual Average, 2010–2012	Percent Change	Number of Sites
Ambient SO_2 Concentration (micrograms per cubic meter, $\mu g/m^3$)	Mid-Atlantic	***13.0***	***3.0***	***-77***	12
	Midwest	***11.0***	***2.7***	***-75***	10
	Northeast	5.6	0.9	-84	3
	Southeast	***5.1***	***1.1***	***-78***	8
Ambient Particulate Sulfate Concentration ($\mu g/m^3$)	Mid-Atlantic	***6.3***	***2.6***	***-59***	12
	Midwest	***5.8***	***2.5***	***-57***	10
	Northeast	3.5	1.3	-63	3
	Southeast	***5.5***	***2.4***	***-56***	8
Ambient Total Nitrate Concentration (Nitrate + Nitric Acid) ($\mu g/m^3$)	Mid-Atlantic	***3.3***	***1.7***	***-48***	12
	Midwest	***4.6***	***3.0***	***-35***	10
	Northeast	1.8	0.8	-56	3
	Southeast	***2.2***	***1.3***	***-41***	8
Dry Inorganic Nitrogen Deposition (kilograms nitrogen per hectare, kg-N/ha)	Mid-Atlantic	***2.5***	***1.2***	***-52***	12
	Midwest	***2.5***	***1.5***	***-40***	10
	Northeast	1.3	0.5	-62	3
	Southeast	***1.7***	***1.0***	***-41***	8
Total Inorganic Nitrogen Deposition (kg-N/ha)	Mid-Atlantic	***8.8***	***4.2***	***-52***	12
	Midwest	***8.8***	***5.2***	***-41***	10
	Northeast	6.6	2.9	-56	3
	Southeast	***6.4***	***3.5***	***-45***	8
Dry Sulfur Deposition (kilograms sulfur per hectare, kg-S/ha)	Mid-Atlantic	***7.0***	***1.6***	***-77***	12
	Midwest	***6.6***	***1.8***	***-73***	10
	Northeast	2.6	0.6	-77	3
	Southeast	***3.1***	***0.9***	***-71***	8
Total Sulfur Deposition (kg-S/ha)	Mid-Atlantic	***16.0***	***4.0***	***-75***	12
	Midwest	***15.0***	***4.0***	***-73***	10
	Northeast	9.5	2.3	-76	3
	Southeast	***10.4***	***2.9***	***-72***	8
Wet Nitrogen Deposition from Inorganic Nitrogen (kg-N/ha)	Mid-Atlantic	***6.2***	***4.5***	***-27***	11
	Midwest	***5.8***	***5.0***	***-14***	27
	Northeast	***5.6***	***3.7***	***-34***	17
	Southeast	***4.4***	***3.5***	***-20***	23
Wet Sulfur Deposition from Sulfate (kg-S/ha)	Mid-Atlantic	***9.2***	***3.6***	***-61***	11
	Midwest	***7.1***	***3.2***	***-55***	27
	Northeast	***7.5***	***2.8***	***-63***	17
	Southeast	***6.1***	***2.7***	***-56***	23

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 to 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 italics 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, 2013

Nitrogen Oxides

NO_x is a group of highly reactive gases including nitrogen dioxide, nitrous acid, and nitric acid. In addition to contributing to the formation of ground-level ozone and $PM_{2.5}$, NO_x is linked with a number of adverse health effects on the respiratory system. See the "Ozone" section on page 9 for more information on the health effects of ozone pollution.

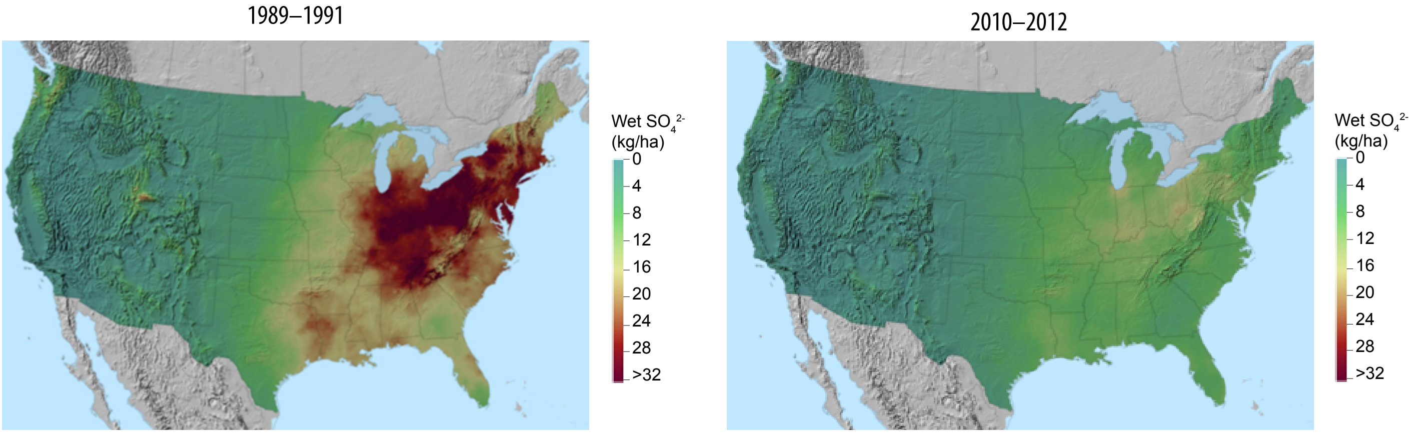
From 2010 to 2012, reductions in NO_x emissions during the ozone season from power plants under the NO_x SIP Call, ARP, and CAIR have continued to contribute to significant regional improvements in ambient total nitrate (NO_3^- plus HNO_3) concentrations. For instance, annual mean ambient total nitrate concentrations for 2010 to 2012 in the Mid-Atlantic region were 48 percent less than the annual mean concentration in 1989 to 1991 (see Table 1).

Although the ARP and CAIR NO_x programs have contributed to significant NO_x reductions, 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.

Acid Deposition

As SO_2 and NO_x gases react in the atmosphere with water, oxygen, and other chemicals, they form various acidic compounds that get deposited to the ground in the form of wet and dry acid deposition. Trends in National Atmospheric Deposition Program/National Trends Network (NADP/NTN) monitoring network 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 2010 to 2012 observation periods, decreases in wet deposition of sulfate averaged more than 59 percent for the eastern United States (see Table 1 and Figure 4).

Figure 4: Three-Year Mean Wet Sulfate Deposition



1989–1991

2010–2012

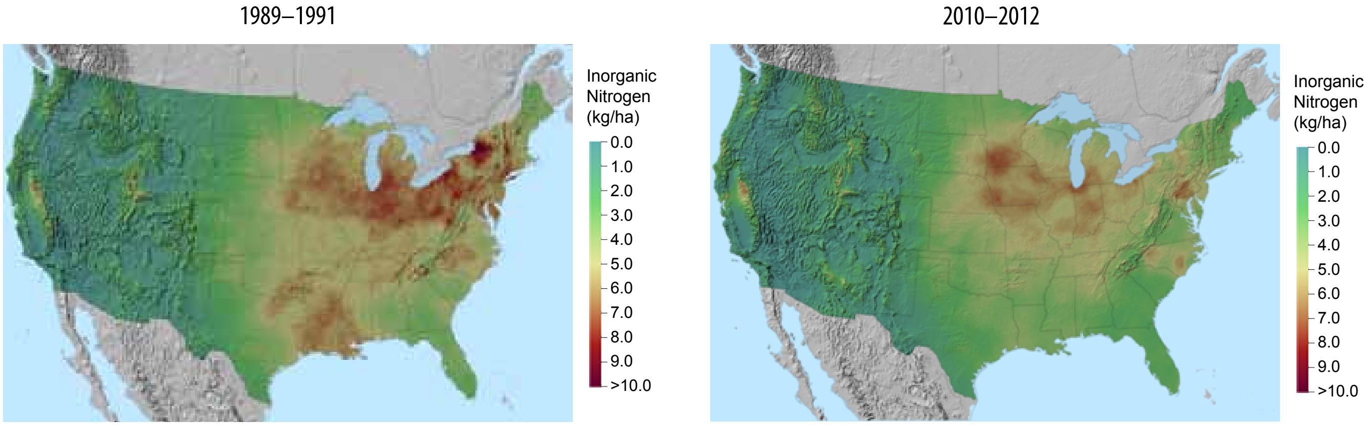
Source: EPA, 2013

Along with wet sulfate deposition, wet sulfate concentrations have also decreased by similar percentages. A strong correlation between large-scale SO_2 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 of similar magnitude as that of wet deposition in the Mid-Atlantic and Midwest, with reductions of 75 and 73 percent, respectively (see Table 1). Because continuous data records are available from only a few sites in the Northeast, it is unclear if the observed reductions in total deposition are representative for that region.

A principal reason for reduced sulfate deposition in the Northeast is a decrease 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 lowered SO_2 emissions 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 nearly 70 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 changes from source categories other than ARP and CAIR sources contribute to changes in air concentrations and deposition of nitrogen. Inorganic nitrogen in wet deposition decreased comparably in the Mid-Atlantic and Northeast (see Figure 5). Decreases in dry and total inorganic nitrogen deposition at CASTNET sites have generally been greater than that of wet deposition, with a 52 and 41 percent decrease in total nitrogen deposition for the Mid-Atlantic and Midwest, respectively (see Table 1).

Figure 5: Three-Year Mean Wet Inorganic Nitrogen Deposition



Source: EPA, 2013

Ozone

Ozone pollution forms when NO_x and volatile organic compounds (VOCs) react in the presence of sunlight. Major sources of NO_x and VOC emissions include motor vehicles, solvents, industrial facilities, and electric power plants. Meteorology plays a significant role in ozone formation and hot, sunny days are most favorable for ozone production. In general, ozone concentrations increase during the daylight hours, peak in the afternoon, and drop in the evening. For ozone, EPA and states typically regulate NO_x emissions in the summer months when sunlight intensity and temperatures are highest. Under CAIR, the NO_x ozone season is from May 1 to September 30.

Exposure to ozone has been linked to a variety of health effects including respiratory diseases such as asthma, emphysema, and bronchitis, and increased susceptibility to respiratory infections. More serious effects include emergency room visits, hospital admissions, and premature mortality. For more information on the health and environmental effects of ground-level ozone, visit EPA's Ground-level Ozone website at <www.epa.gov/ozonepollution>.

Ozone Standards

The CAA requires EPA to set National Ambient Air Quality Standards (NAAQS) for ground level ozone and five other criteria pollutants. In 1979, EPA established the NAAQS for 1-hour ozone at 0.12 ppm, and in 1997, a new, more stringent 8-hour ozone standard of 0.08 ppm was promulgated, revising the 1979 standard. In March 2008, EPA changed the 8-hour ozone standard to 0.075 ppm. CAIR was promulgated to help downwind states in the eastern U.S. achieve the 1997 ozone NAAQS, and therefore analyses in this report focus on that standard. EPA is currently working with states to address transported air pollution with respect to the 2008 ozone NAAQS.

Measuring and Evaluating Changes in Ozone

To better understand how the CAIR, NBP, and ARP NO_x programs affected ozone formation in the atmosphere, this report examines changes in ozone concentrations before and after implementation of the NBP and CAIR. The report also compares regional and geographic trends in ozone levels to changes in NO_x emissions from CAIR sources.

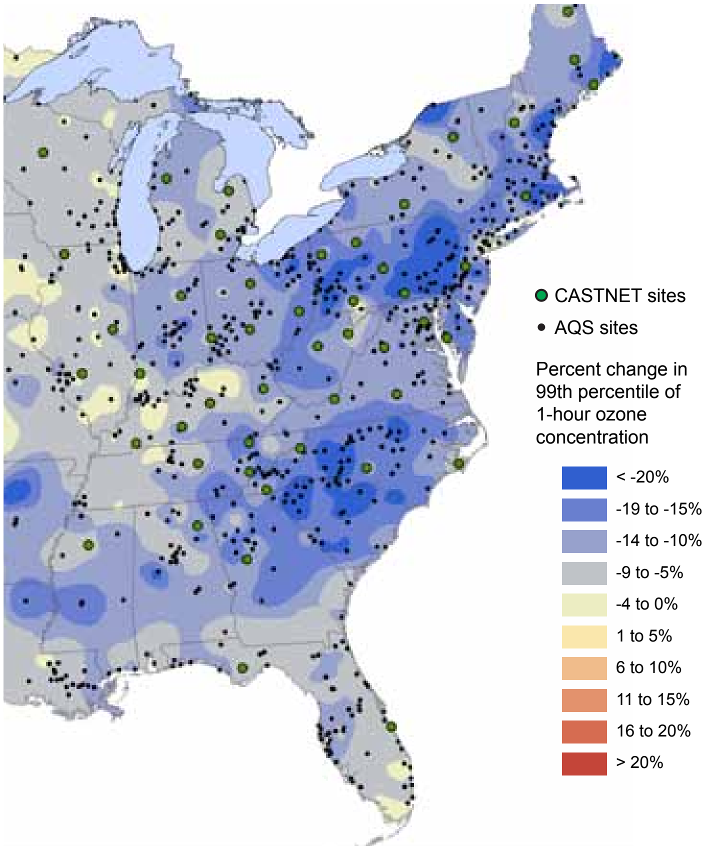
Two long-term monitoring networks, the State and Local Air Monitoring Stations (SLAMS) and CASTNET, measure ozone levels as well as meteorological and other air quality data throughout the United States. SLAMS monitoring sites are used for regulatory compliance and are located mainly in urban areas and report data to EPA's Air Quality System (AQS). CASTNET sites measure trends in ozone at rural sites and these data are also used for regulatory compliance and reported to AQS. The changes in eastern ozone concentrations presented in this report depict data from AQS and CASTNET monitoring sites located within both CAIR and adjacent states (states within 200 km of a CAIR NO_x ozone season state's borders). These analyses show a range of ozone reductions based on the metric used and the years examined.

Rural ozone measurements are useful in assessing the impacts on air quality resulting from regional NO_x emission reductions because these monitoring sites are typically less affected by local sources of NO_x (e.g., industrial, mobile, and power generation sources) than urban measurements. Consequently, the formation of ozone in these areas is particularly sensitive to changes in levels of regional NO_x emissions. The majority of reductions in rural ozone concentrations can therefore be attributed to reductions in regional NO_x emissions and transported ozone. EPA investigated trends in both 1-hour and rolling 8-hour ozone concentrations as measured at CASTNET monitoring sites within the CAIR NO_x ozone season region and in adjacent states.

Changes in 1-Hour Ozone Concentrations in the East

EPA examined changes in unadjusted regional 1-hour ozone concentrations, as measured at urban (AQS) and rural (CASTNET) sites. Results demonstrate how NO_x emission reduction policies have affected ozone concentrations in the eastern United States. Figure 6 shows changes in the 99th percentile of 1-hour ozone concentrations between 2000–2002 (before implementation of the NBP) and 2010–2012 (under the CAIR NO_x ozone season program). Using this metric, an overall regional reduction in ozone levels was observed between these two time periods, with an average reduction in ozone concentrations in CAIR states of 18 percent.

Figure 6: Percent Change in 1-Hour Ozone Concentrations during the Ozone Season, 2000–2002 versus 2010–2012

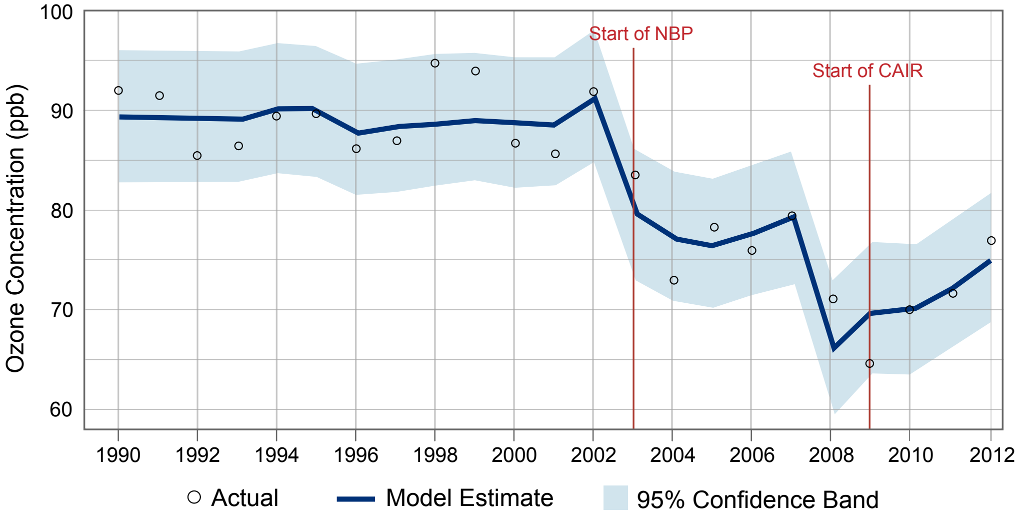


Note: Data are from AQS and CASTNET monitoring sites with two or more years of data within each three-year monitoring period.
Source: EPA, 2013

Regional Trends in Ozone

An Autoregressive Integrated Moving Average (ARIMA) model was used to determine the trend in regional ozone concentrations since implementation of various programs geared towards reducing NO_x emissions. The ARIMA model is an advanced statistical analysis tool that can evaluate trends and changes in response to events over time (time series analysis). The average of the 99th percentile of the rolling 8-hour daily maximum ozone concentrations (the highest daily levels of ozone) measured at CASTNET sites during the CAIR NO_x ozone season was modeled (Figure 7).

Figure 7: Shift in 99th Percentile 8-hour Seasonal Rural Ozone Concentrations in the CAIR NO_X Ozone Season Region, 1990–2012



Note: Ozone concentration data are from CASTNET sites that met completeness criteria and are located in and adjacent to the CAIR NO_x region.
Source: EPA, 2013

Overall, regional ozone concentrations have dropped 15 ppb (16 percent) since 1990. Furthermore, results show a significant decrease (11 ppb) in the modeled trend of regional 99th percentile ozone concentrations in the 2003 and 2004 seasons following the implementation of the NBP in 2003. That event was followed by an additional 10 percent (8 ppb) reduction in the modeled trend of regional ozone concentrations just prior to the start of CAIR on May 1, 2009. This decrease in the modeled trend may be linked to actions taken for CAIR compliance, however other factors may include meteorology and changes in energy demand.

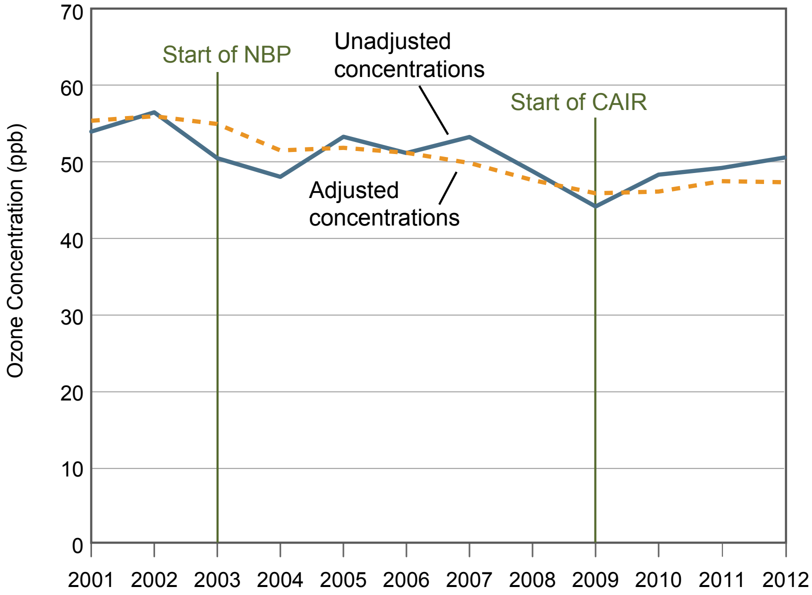
There have been substantial reductions in NO_x emissions, including from the power sector and mobile sources, and great improvements in ozone air quality. Despite this progress, ozone concentrations have started to increase again in recent years. Meteorologically-adjusted seasonal averages of ozone concentrations at EPA monitoring sites in the CAIR region (see Figure 8 on page 12) suggest that meteorology (hotter summers) explains a significant portion of the recent uptick in the 99th percentile data. This is supported by observations that the 2009 weekly averages of daily maximum temperatures at CASTNET sites in the CAIR region were typically cooler than the preceding three year average while the 2012 temperatures were typically warmer.[8] This indicates that it is important to account for meteorological effects when assessing any trends in air pollution after CAIR was implemented.

Changes in 8-hour Ozone Concentrations

Daily maximum 8-hour ozone concentration data from EPA and daily meteorology data from the National Weather Service were retrieved for 82 urban areas and 36 rural CASTNET monitoring sites located in the CAIR NO_x ozone season program region for May to September of 2001 to 2012. As noted earlier, weather plays an important role in ozone formation. Accordingly, EPA uses a statistical model to describe the relationship between daily maximum 8-hour ozone concentrations and several meteorological parameters.[9] The model accounts for the influence of weather on seasonal average ozone concentrations by adjusting the observed ozone trend in each area or monitoring site to account for year-to-year variability in meteorological conditions. The most influential meteorological parameters, with respect to ozone formation, in this model are the daily maximum 1-hour temperature and the mid-day (10 a.m. to 4 p.m.) relative humidity. Meteorologically-adjusted ozone trends can provide additional insight on the influence of CAIR NO_x ozone season program emission reductions on regional air quality.

Figure 8 shows the trends in the May to September average daily maximum 8-hour ozone concentrations averaged across the 82 urban areas and 36 CASTNET sites in the CAIR NO_x ozone season region before and after adjusting for the influence of weather.[10] The average reduction in the unadjusted concentrations measured in the CAIR NO_x region from the 2001–2003 period to the 2010–2012 period was about 8 percent. Meteorologically-adjusted data showed a 15 percent reduction over the same region and time frame. As noted in the previous section ("Regional Trends in Ozone"), ozone concentrations have increased in recent years relative to low values in 2009. Figure 8 shows that the majority of this increase is likely due to meteorological conditions.

Figure 8: Seasonal Average of 8-hour Ozone Concentrations in CAIR Sites Unadjusted and Adjusted for Weather



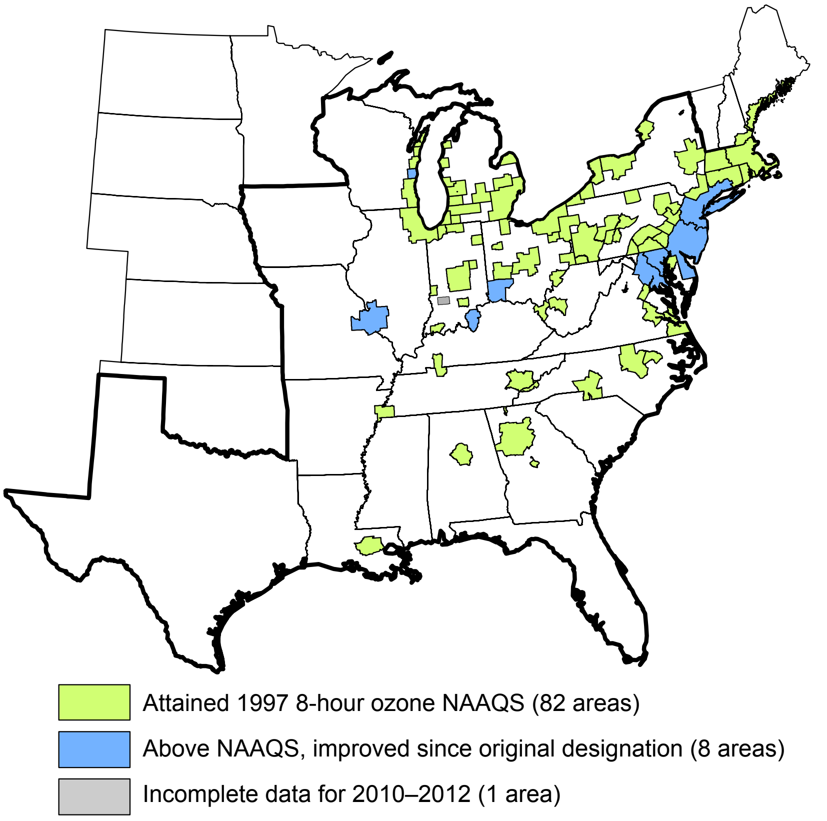
Note: For a monitor or urban area to be included in this trends analysis, it had to provide complete and valid data for 75 percent of the days in the May to September period, for each of the years from 2001 to 2012. In addition, urban areas often include more than one monitoring site. In these cases, the highest observed ozone concentration in the area was used for each day.
Source: EPA, 2013

Changes in Ozone Nonattainment Areas

EPA designated 113 areas as nonattainment for the 8-hour ozone standard (0.08 ppm) adopted in 1997 using data from 2001 to 2003.[11] Of those areas, 91 are in the East (as shown in Figure 9) and are home to about 109 million people.[12] Based on data gathered from 2010 to 2012, 90 percent (82 areas) of the eastern ozone nonattainment areas show concentrations below the level of the 1997 standard while eight areas show concentrations above the 1997 standard. Compared with the 2001 to 2003 period, these eight eastern ozone nonattainment areas all showed improvement in the 2010 to 2012 period toward meeting the standard.

Given that the majority of power sector NO_x emission reductions occurring after 2003 are attributable to the NBP and CAIR, and power sector emissions are an important component of the NO_x emission inventory, it is reasonable to conclude that these NO_x reduction programs have been a significant contributor to these improvements in ozone air quality. However, because areas continue to be out of attainment for both the 1997 and 2008 ozone NAAQS,[13,14] additional NO_x emission reductions are still needed to attain EPA's health based air quality standards. As part of an effort to help states achieve some of these reductions, EPA is working to define upwind states' obligations under the 2008 ozone standards to address transported air pollution affecting the eastern half of the U.S.

Figure 9: Changes in Ozone Nonattainment Areas in the CAIR Region, 2001–2003 (Original Designations) versus 2010–2012



CAIR States controlled for PM and/or ozone are outlined.

Source: EPA, 2013

Particulate Matter

Particulate matter—also known as particle pollution or PM—is a complex mixture of extremely small particles and liquid droplets. Particle pollution is made up of a number of components, including acid-forming nitrate and sulfate compounds, organic chemicals, metals, and soil or dust particles. Fine particles ($PM_{2.5}$) can form when gases emitted from power plants, industrial sources, automobiles, and other sources react in the air.

Particle pollution—especially fine particles—contains microscopic solids or liquid droplets so small that they can get deep into the lungs and cause serious health problems. Numerous scientific studies have linked particle pollution exposure to a variety of problems, including: increased respiratory symptoms, such as irritation of the airways, coughing, or difficulty breathing; decreased lung function; aggravated asthma; development of chronic bronchitis; irregular heartbeat; nonfatal heart attacks; and premature death in people with heart or lung disease.

For more information on the health and environmental effects of particulate matter, visit EPA's Particulate Matter website at <www.epa.gov/air/particlepollution>.

Particulate Matter Standards

The CAA requires EPA to set NAAQS for particle pollution. The first PM standard for fine particles was set by EPA in 1997 at 65 micrograms per cubic meter ($\mu g/m^3$) measured as the three year average of the 98th percentile for 24-hour exposure, and at 15 $\mu g/m^3$ for annual exposure measured as the three-year annual mean. EPA revised the air quality standards for particle pollution in 2006, tightening the 24-hour fine particle standard to 35 $\mu g/m^3$ and retaining the annual fine particle standard at 15 $\mu g/m^3$. Recently, in December 2012, EPA strengthened the annual fine particle standard to 12 $\mu g/m^3$. CAIR was promulgated to help downwind states in the eastern U.S. achieve the 1997 annual average $PM_{2.5}$ NAAQS, and, therefore, analyses in this report focus on that standard.

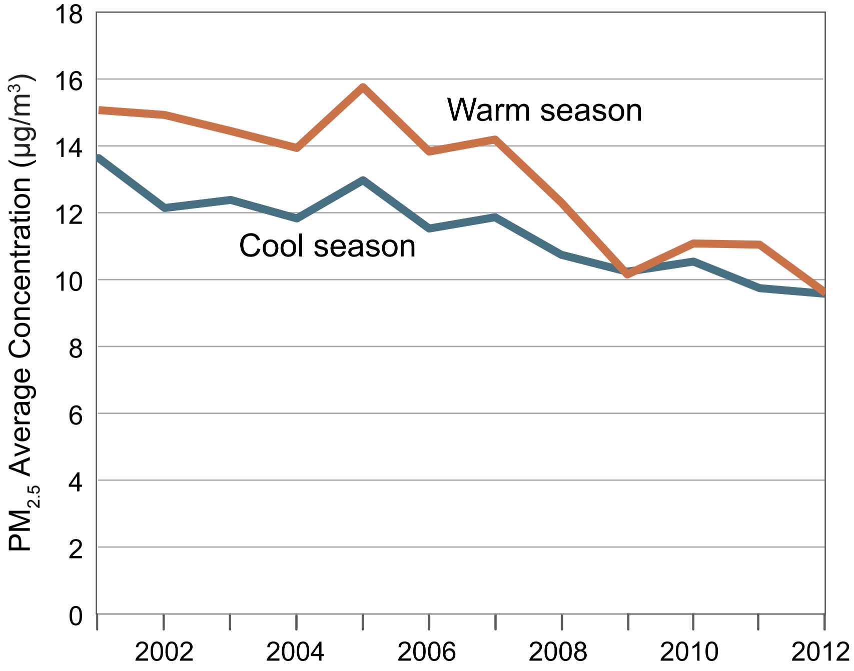
Annual Emission Reduction Programs and $PM_{2.5}$

The CAIR NO_x annual program and CAIR SO_2 programs were established in 2009 and 2010 to address the interstate transport of $PM_{2.5}$ pollution throughout the year and help eastern U.S. counties attain the $PM_{2.5}$ annual standard. To better understand how emission reductions under CAIR and the ARP affected the formation of $PM_{2.5}$, this report presents regional and geographic trends in $PM_{2.5}$ levels prior to implementation of any of the CAIR annual programs, and for 2012.

Trends in $PM_{2.5}$ Concentrations

Average $PM_{2.5}$ concentration data were assessed from 431 urban AQS areas located in the CAIR NO_x and SO_2 annual program region. Figure 10 shows separate trends in $PM_{2.5}$ concentrations in the CAIR NO_x and SO_2 annual program region for the warm months (April to September) and cool months (October to March) unadjusted for the influence of weather. The annual average $PM_{2.5}$ concentration has decreased by about 36 percent in the warm season and about 29 percent in the cool season between 2001 and 2012. However, as noted earlier, weather plays an important role in the formation of PM. Meteorologically adjusted data, currently unavailable, would provide additional insight on the influence of annual CAIR SO_2 and NO_x program emission reductions on regional air quality.

Figure 10: $PM_{2.5}$ Seasonal Trends



Notes:

- For a $PM_{2.5}$ monitoring site to be included in the trends analysis, it had to meet all of the following criteria: 1) each site-year quarterly mean concentration value had to encompass at least 11 or more samples, 2) all four quarterly mean values had to be valid for a given year (i.e., meet criterion #1), and 3) all 12 years of site-level seasonal means had to be valid for the given site (i.e. meet criteria #1 and #2).
- Annual "cool" season mean values for each site-year were computed as the average of the first and fourth quarterly mean values. Annual "warm" season mean values for each site-year were computed as the average of the second and third quarterly mean values. For a given year, all of the seasonal mean values for the monitoring sites located in the CAIR Region were then averaged together to obtain a single year (composite) seasonal mean value.

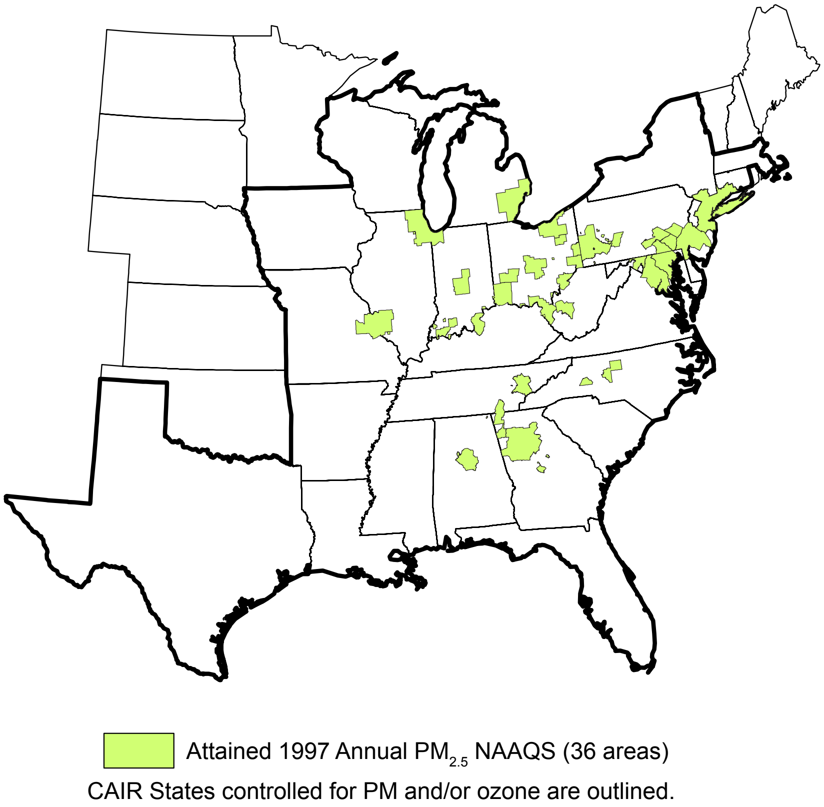
Source: EPA, 2013

Changes in $PM_{2.5}$ Nonattainment Areas

In January 2005, EPA designated 39 areas as nonattainment for the 1997 annual average $PM_{2.5}$ standard, one of which was also designated nonattainment for the 24-hour average $PM_{2.5}$ standard.[15] These designations were made using data from 2001 to 2003. Of those areas, 36 are in the East (as shown in Figure 11) and are home to about 75 million people.[16] Based on data gathered from 2010 to 2012, all of these original eastern areas show concentrations below the level of the 1997 $PM_{2.5}$ standard (15.0 $\mu g/m^3$), indicating improvements in $PM_{2.5}$ air quality.

Given that the majority of power sector NO_x and SO_2 emission reductions occurring after 2003 are attributable to the ARP, NBP, and CAIR, it is reasonable to conclude that these emission reduction programs have been a significant contributor to these improvements in $PM_{2.5}$ air quality.

Figure 11: Changes in PM Nonattainment Areas in the CAIR Region, 2001–2003 (Original Designations) versus 2010–2012


🟩 Attained 1997 Annual $PM_{2.5}$ NAAQS (36 areas)

CAIR States controlled for PM and/or ozone are outlined.

Source: EPA, 2013

Ecosystems

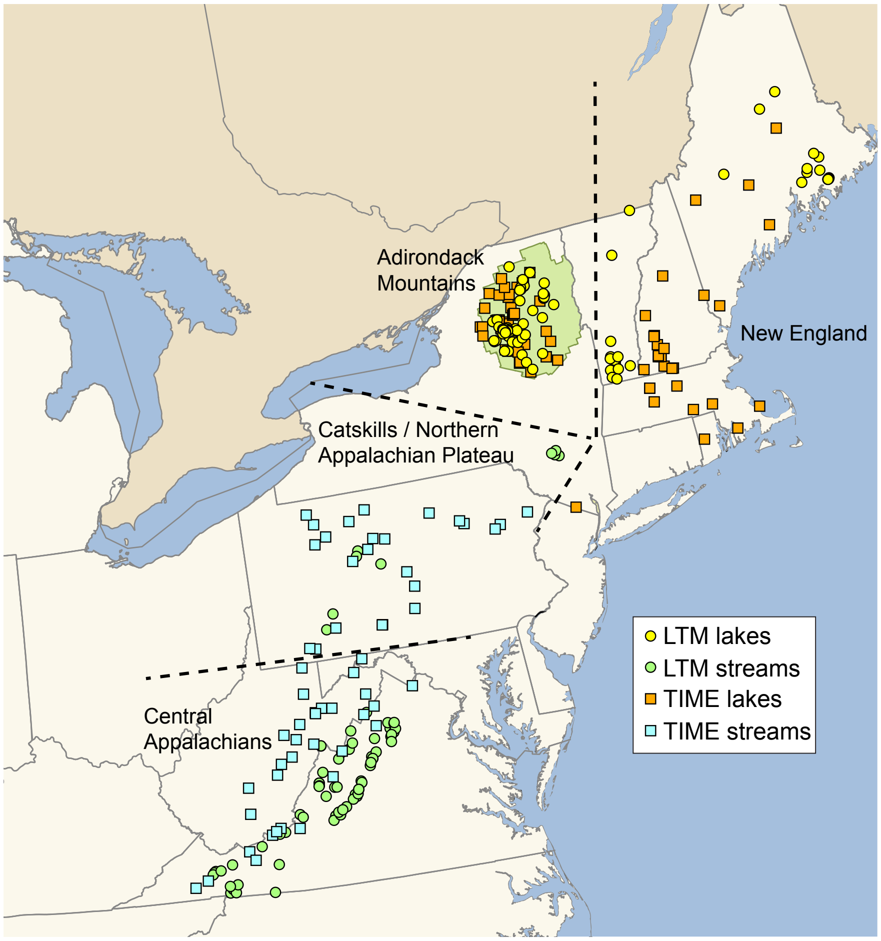
Acid deposition (i.e., acid rain) resulting from SO_2 and NO_x emissions negatively affects the health of lakes, streams, and other ecosystems in the United States. The *2011 National Acid Precipitation Assessment Program (NAPAP) Report to Congress* noted that the ARP has been successful in reducing emissions of SO_2 and NO_x from power generation. The report also notes that acid deposition has decreased to the extent that some acid-sensitive lakes and streams are beginning to recover from acidification.[17]

Improvements in Surface Water Chemistry

Surface water chemistry provides direct indications of the potential effects of acidic deposition on the overall health of aquatic ecosystems. In collaboration with other federal and state agencies and universities, EPA administers two monitoring programs that provide information on the impacts of acidic deposition on otherwise pristine aquatic systems: the Temporally Integrated Monitoring of Ecosystems (TIME) and the Long-term Monitoring (LTM) programs. These programs are designed to track changes in surface water chemistry in the four regions sensitive to acid rain shown in Figure 12: New England, the Adirondack Mountains, the Northern Appalachian Plateau, and the central Appalachians (the Valley and Ridge and Blue Ridge Provinces).

For more information about these programs, visit EPA's Surface Water Monitoring website at <www.epa.gov/airmarkets/assessments/surfacewater.html>.

Figure 12: Long Term Monitoring Program Sites



Source: EPA, 2013

Table 2: Regional Trends in Sulfate, Nitrate, ANC, Base Cations, and DOC at Long-term Monitoring Sites, 1990–2012

Region	Water Bodies Covered	% of Sites with Improving Sulfate Trend	% of Sites with Improving Nitrate Trend	% of Sites with Improving ANC Trend	% of Sites with Improving Base Cations Trend	% of Sites with Increasing DOC Trend
Adirondack Mountains	50 lakes in NY	100%	54%	76%	88%	62% (29 sites)
New England	26 lakes in ME and VT	100%	18%	43%	74%	39% (13 sites)
Catskills/N. Appalachian Plateau*	9 streams in NY and PA	80%	40%	58%	90%	0% (9 sites)
Central Appalachians	66 streams in VA	15%	58%	15%	14%	N/A

* Data for PA streams in N. Appalachian Plateau is only available through 2011.

Notes:

- Trends are significant at the 95 percent confidence interval ($p < 0.05$).
- Base cations are calculated as the sum of calcium, magnesium, potassium, and sodium ions.
- Trends are determined by multivariate Mann-Kendall tests.
- DOC was only examined in low-ANC waterbodies (ANC less than 25 μeq/L).
- DOC is not currently measured in Central Appalachian streams.

Source: EPA, 2013

Acidified drainage water mobilizes toxic forms of aluminum from soils and clays, harming fish and wildlife. Five chemical indicators of aquatic ecosystem response to emission changes are presented: trends in sulfate and nitrate anions, sum of base cations, acid neutralizing capacity (ANC), and dissolved organic carbon (DOC). These indicators provide information regarding the surface water sensitivity to acidification and the degree of impact on the aquatic ecosystem. Trends in these measured chemical indicators in drainage waters allow for the determination of whether the water bodies are improving and heading towards recovery or if they are still acidifying. The following is a description of each indicator:

Sulfate is the primary anion in most acid-sensitive waters and has the potential to acidify drainage waters and leach base cations and toxic forms of aluminum from the soils.

Nitrate has the same potential as sulfate to acidify drainage waters. However, nitrogen is an important nutrient for plant and algae growth and a large portion of nitrogen inputs from deposition are quickly taken up by plants, leaving less leaching of nitrate into surface waters.

Base cations are the positively charged ions in soils and surface waters that buffer both sulfate and nitrate anions, thereby preventing surface water acidification. Base cation availability is largely a function of underlying geology and soil age, such that young soils of cation-rich bedrock will tend to have a greater buffering capacity.

Acid Neutralizing Capacity (ANC) is a measure of overall buffering capacity against acidification, and indicates the ability to neutralize strong acids that enter aquatic systems. When ANC is low, and especially when it is negative, stream water pH is also low (less than pH 6, commonly less than pH 5), and may be harmful to fish and other aquatic organisms essential for a healthy aquatic ecosystem. Figure 13 on page 19 shows how waterbody acidification is categorized by ANC concentration. Recovery of an aquatic ecosystem is indicated by increasing trends in ANC and base cations and decreasing trends in sulfate and nitrate concentrations.

Dissolved organic carbon (DOC) is essentially dissolved organic material that is an important part of the acid-base chemistry of most freshwater systems (particular low-ANC waterbodies) because it can assist in neutralizing strong acids. A host of factors control DOC concentrations in surface waters and increases can be indicative of reduced acidification and/or a sign of increased decomposition of organic matter in the watershed.

For more information on the negative effects of acid deposition, see EPA's Acid Rain page at <www.epa.gov/acidrain/effects/surface_water.html>.

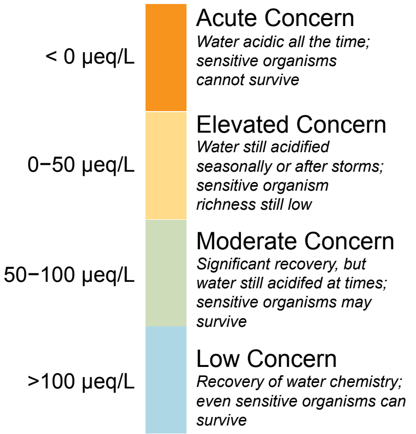
Table 2 on page 18 shows regional trends in indicators of acidified surface waters from 1990 (before implementation of the ARP) to 2012 in lakes and streams through the LTM program. Over this time frame, significant improving trends in sulfate concentrations are found at nearly all LTM monitoring sites in New England, the Adirondacks, and the Catskill mountains/Northern Appalachian Plateau. However, in the Central Appalachians only 15 percent of monitored streams have a decreasing sulfate trend, while 20 percent of monitored streams had increasing sulfate concentrations. This is due to the highly weathered soils of the Central Appalachians which are able to store deposited sulfate such that the decrease in acidic deposition has not yet resulted in lower sulfate concentrations in most streams. However, as long-term sulfate deposition exhausts the soil's ability to store more 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 some streams in this region are not changing or are still increasing despite reduced sulfate deposition.

Nitrate concentration trends are variable across the LTM site network, with improving trends measured at approximately half of all the monitored sites. This improvement in nitrate trend may only be partially explained by decreasing deposition. Ecosystem factors, such as vegetation disturbances, increased uptake by vegetation, and soil retention are also known to affect surface water nitrate concentrations.

Improving ANC trends are likely the result of reductions in sulfate deposition. Recovery in ANC, however, often lags behind declining surface water sulfate and nitrate concentrations. Dynamics in surface water chemistry are complicated and highly dependent on local factors such as watershed size, geology and hydrology, which can influence the availability of base cations and DOC, thereby inhibiting ANC recovery. From 1990 to 2012, ANC concentration increased markedly at LTM monitoring sites in the Adirondacks (76 percent), in the Catskills/northern Appalachian Plateau (58 percent), and in New England (43 percent). In contrast, only 15 percent of LTM streams in the Central Appalachians had improving ANC trends, likely due to decreasing base cation levels and the still increasing sulfate concentrations at some sites.

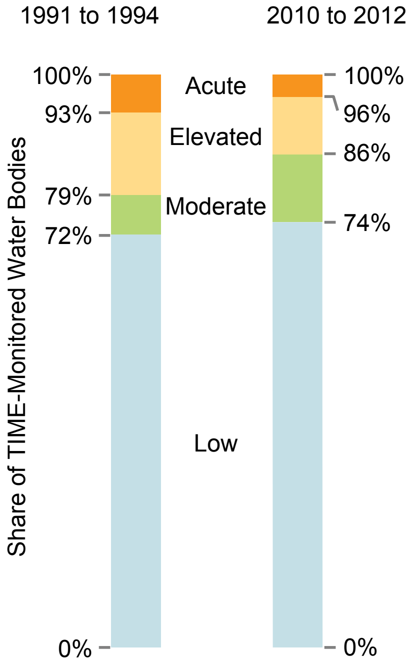
Figure 14 presents a comparison of the average ANC value of the 580 lakes in the Northeast monitored and modeled under the TIME program for the 1991–1994 and 2010–2012 time periods. Seven percent of lakes in the 1991–1994 time period had mean ANC levels below 0 microequivalents per liter (μeq/L) and were categorized as acute concern, but less than 4 percent of lakes were categorized as acute concern in the 2010–2012 time frame and the percentage of lakes in the elevated concern category dropped from 14 percent to 10 percent over the same time frame. Meanwhile, the net percentage of lakes in the moderate concern category increased from 7 percent to 12 percent. These results point to a decrease in acidity, particularly for the subset of TIME lakes in the acute and elevated concern categories.

Figure 13: Acid Neutralizing Capacity and Aquatic Ecosystem Concern Levels



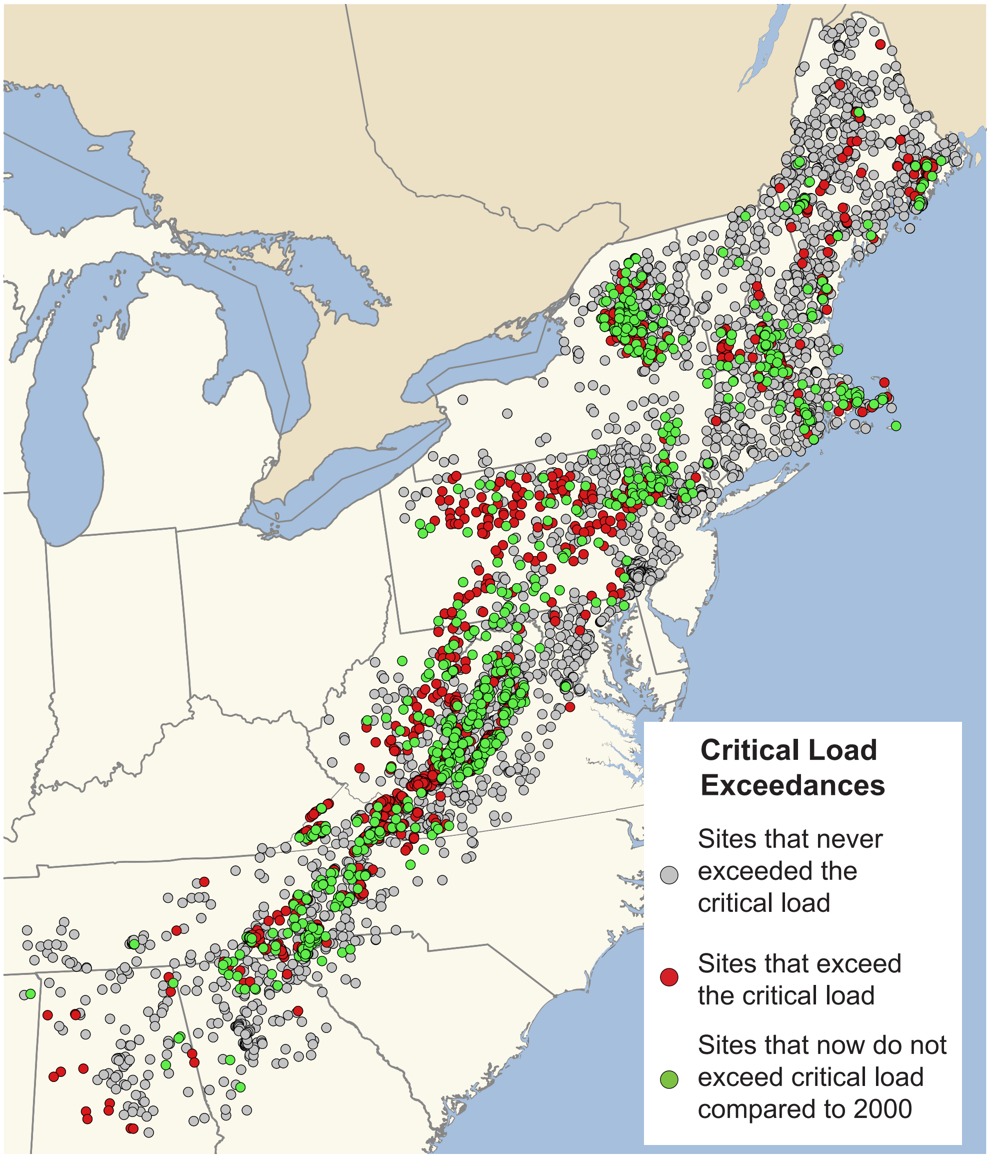
Source: EPA, 2013

Figure 14: TIME Lakes by ANC Category, 1991–1994 vs. 2010–2012



Source: EPA, 2013

Figure 15: Lake and Stream Exceedances of Estimated Critical Loads for Total Nitrogen and Sulfur Deposition for the Period between 2000–2002 and 2010–2012



Notes:

- Surface water samples from the represented waterbodies collected through the National Surface Water Survey (NSWS), Environmental Monitoring and Assessment Program (EMAP), Wadeable Stream Assessment (WSA), National Lake Assessment (NLA), Temporally Integrated Monitoring of Ecosystems (TIME), Long Term Monitoring (LTM), and other water quality programs.
- Steady state exceedances calculated in units of $meq/m^2/yr$

Source: EPA, 2013

Critical Loads and Exceedances

Although significant reductions in SO_2 and NO_x emissions and acid deposition have occurred since inception of the emission reduction programs, current emission levels may not be sufficient to allow full recovery of acid-sensitive ecosystems. A critical load is an assessment approach used to provide a quantitative estimate of whether deposition levels resulting from reduction in SO_2 and NO_x emissions and acid deposition are protective of aquatic resources. This approach provides a useful lens through which to help understand the potential aquatic ecological benefits that have resulted from emission reduction programs such as the ARP and CAIR.

Estimations of critical load exceedances serve as a measurement for determining if present acid deposition levels will provide sufficient reductions to allow the systems to recover over time or if they will never recover under present loading scenarios. If acidic deposition is less than the calculated critical load, harmful ecological effects (e.g., reduced reproductive success, stunted growth, loss of biological diversity) are not anticipated, and ecosystems damaged by past exposure are expected to eventually recover.[18] Lake and stream waters having an ANC value greater than 50 μeq/L are classified as having a moderately healthy aquatic community; therefore, this ANC value is often used as a goal for ecological protection of drainage waters affected by acid deposition.

Figure 15 shows a comparison of the estimated critical load exceedances for waterbodies for the period between 2000–2002 and 2010–2012. For this analysis, the critical load represents the annual deposition load of sulfur and nitrogen to which a lake or stream could be subjected and still support a moderately healthy ecosystem (i.e., having an ANC greater than 50 μeq/L). Surface water samples from 4,886 lakes and streams along acid sensitive regions of the Appalachian Mountains and some adjoining northern coastal plain regions were collected through a number of water quality monitoring programs. Critical load exceedances for those waterbodies were calculated using the Steady-State Water Chemistry model.[19, 20]

For the period from 2010–2012, 27 percent of all the represented waterbodies were shown to still receive levels of combined total sulfur and nitrogen deposition in excess of their critical load, a 37 percent improvement over the 2000–2002 period when 42 percent were in exceedance. Regional differences in critical load exceedances were examined for New England, the Adirondack Mountains, the Northern Mid-Atlantic, the Southern Mid-Atlantic, and the Southern Appalachian Mountains, summarized in Table 3.

Table 3: Critical Load Exceedances by Region

		Water Bodies in Exceedance of Critical Load				
		2000–2002		**2010–2012**		
Region	**Number of Water Bodies Modeled**	**Number of Sites**	**Percent of Sites**	**Number of Sites**	**Percent of Sites**	**Percent Reduction**
New England (ME, NH, VT, RI, CT)	1,298	273	21%	147	11%	46%
Adirondack Mountains (NY)	341	160	47%	70	21%	56%
Northern Mid-Atlantic (PA, NY, NJ)	784	263	34%	155	20%	41%
Southern Mid-Atlantic (VA, WV, MD)	1,690	1,070	63%	745	44%	30%
Southern Appalachian Mountains (NC, TN, SC, GA, AL)	773	308	40%	192	25%	38%
Total	**4,886**	**2,074**	**42%**	**1,309**	**27%**	**37%**

Source: EPA, 2013

This analysis suggests that emission reductions achieved since 2000 are anticipated to contribute to broad surface water improvements and increased aquatic ecosystem protection across the five regions along the Appalachian Mountains. This result is consistent with the water quality monitoring findings (see Table 2), except that the anticipated improvements (e.g., reduction of exceedances) based on the critical load analysis are much larger. This is expected as water quality improvements often lag behind the reduction in acidic deposition while critical loads represent the equilibrium conditions between deposition and water quality. Based on this critical load analysis, current acidic deposition loadings still fall short for recovery of many modeled waterbodies, which indicates additional emission reductions would be necessary for acid-sensitive aquatic ecosystems along the Appalachian Mountains to recover and be protected from acid deposition.

Online Resources

The availability and transparency of data, from emission measurement to allowance trading to deposition monitoring, is a cornerstone of effective emission reduction programs. EPA develops and manages programs for collecting these data and assessing the effectiveness of emission reduction programs, including the ARP, NBP, and CAIR. These data are then made available to the public in readily usable and interactive formats.

Figure 16 shows EPA's newly redesigned Power Plant Emission Trends page at < www.epa.gov/airmarkets/progress/datatrends/index.html> which presents current and historical emissions data in an easy-to-understand and visually appealing way. For each pollutant, emissions data for SO_2, NO_x and CO_2 are easily tracked and downloaded using bar charts, Google Earth mapping plug ins, and motion charts, with the supporting data available for download.

Figure 16: Power Plant Emission Trends Page

Power Plant Emission Trends

EPA collects detailed sulfur dioxide (SO_2), nitrogen oxides (NO_x), and carbon dioxide (CO_2) emission data and other information from coal-fired power plants across the country. Collected as part of the Acid Rain Program (ARP) and the Clean Air Interstate Rule (CAIR), these data are available online, making it easy for the public to understand emissions from these sources and to see when and where emission changes have occurred.

See our Emissions Tracking Highlights page for more detailed information on the power sector including quarterly emissions, unit characteristics, and emission controls. For more detailed information on the results of these programs, including emissions compliance, market analysis, and health and environmental results, see our progress reports.

Source: EPA, 2014

Notes

1. U.S. Environmental Protection Agency (U.S. EPA). 2009. *Acid Rain and Related Programs: 2008 Emission, Compliance, and Market Analyses Progress Report* (pdf, 1.2 MB). September.

2. U.S. Environmental Protection Agency (U.S. EPA). 2009. *The NO_x Budget Trading Program: 2008 Environmental Results Progress Report* (pdf, 5.5 MB). September.

3. U.S. Environmental Protection Agency (U.S. EPA). 2011. *EPA's NO_x Reduction Program and Clean Air Interstate Rule 2009 Environmental and Health Results Progress Report* (pdf, 2.8 MB). January.

4. U.S. Environmental Protection Agency (U.S. EPA). 2009. *Integrated Science Assessment for Particulate Matter (Final Report)*. EPA-600-R-08-139F. National Center for Environmental Assessment – RTP Division. December.

5. U.S. Environmental Protection Agency (U.S. EPA). 2013. *Integrated Science Assessment for Ozone and Related Photochemical Oxidants*. EPA/600/R-10/076F. Research Triangle Park, NC: U.S. EPA. February. Available on the Internet at http://oaspub.epa.gov/eims/eimscomm.getfile?p_download_id=511347

6. EME Homer City Generation, L.P. v. EPA, 696 F.3d 7, 37-38 (D.C. Cir. 2012).

7. EPA v. EME Homer City Generation, L.P., No. 12-1182, slip op. at 32 (U.S. Apr. 29, 2014).

8. U.S. Environmental Protection Agency (U.S. EPA). 2011. *EPA's NO_x Reduction Program and Clean Air Interstate Rule 2009 Environmental and Health Results Progress Report* (pdf, 2.8 MB). January.

9. Cox, W. M. & Chu, S.H. 1996. Assessment of interannual ozone variation in urban areas from a climatological perspective. *Atmospheric Environment*. 30:16, 2615–2625.

Camalier, L., Cox, W.M. & Dolwick, P. 2007. The effects of meteorology on ozone in urban areas and their use in assessing ozone trends. *Atmospheric Environment*. 41:33, 7127–7137.

10. The seasonal average ozone concentration is calculated as the average of the daily maximum 8-hour ozone levels during the ozone season. These results provide a combined seasonal average for CAIR ozone season states and do not show variations in ozone concentrations for specific urban or rural areas.

11. 40 CFR Part 81. Designation of Areas for Air Quality Planning Purposes.

12. U.S. Census. 2010.

13. Nonattainment and maintenance areas in the U.S. for the 1997 8-hour ozone NAAQS: <www.epa.gov/oaqps001/greenbk/map8hrnm.html>.

14. Nonattainment areas in the U.S. for the 2008 8-hour ozone NAAQS: http://www.epa.gov/oaqps001/greenbk/map8hr_2008.html

15. 40 CFR Part 81. Designation of Areas for Air Quality Planning Purposes.

16. U.S. Census. 2010.

17. Burns, D.A., Lynch, J.A., Cosby, B.J., Fenn, M.E., Baron, J.S., US EPA Clean Air Markets Div., 2011, *National Acid Precipitation Assessment Program Report to Congress 2011: An Integrated Assessment*, National Science and Technology Council, Washington, DC, 114 p.

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

19. Sullivan T.J., B.J. Cosby, J.R. Webb, R.L. Dennis, A.J. Bulger, and F.A. Deviney, Jr. 2007. Streamwater acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National Park, Virginia. *Environmental Monitoring and Assessment* 137:85–99

20. Nilsson, J. & Grennfelt, P. (Eds) (1988) Critical loads for sulphur and nitrogen. UNECE/Nordic Council workshop report, Skokloster, Sweden. March 1988. Nordic Council of Ministers: Copenhagen.