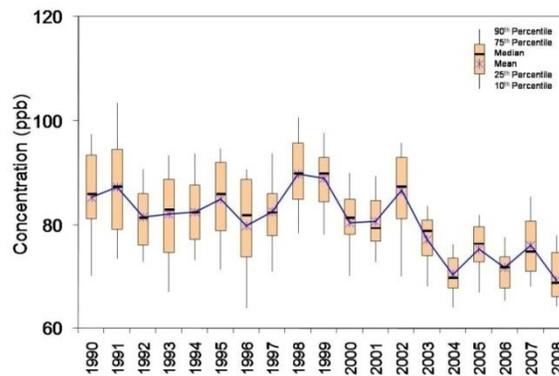




Fourth Highest Daily Maximum 8-Hour Ozone Concentrations (ppb) for 2008



Clean Air Status and Trends Network (CASTNET) 2008 Annual Report



Trend in Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) – Eastern United States

Clean Air Status and Trends Network (CASTNET)

2008 Annual Report

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Executive Summary

This report summarizes Clean Air Status and Trends Network (CASTNET) data collected during 2008 and examines trends in air quality since 1990. The report presents maps of the geographic distribution of pollutants measured in 2008. The report also discusses significant network events and presents an analysis of data quality. CASTNET began collecting measurements in 1991 with the incorporation of 50 sites from the National Dry Deposition Network, which had been in operation since 1987. CASTNET measures rural, regionally representative concentrations of sulfur and nitrogen species and ozone in order to evaluate the effectiveness of national and regional air pollution control programs.

Key Results through 2008

- ◆ The air quality, as represented by CASTNET pollutant measurements, was the best in the history of the network.
- ◆ Mean annual sulfur dioxide and particulate sulfate concentrations declined significantly in the eastern United States over the 19-year period 1990 through 2008. Sulfur dioxide levels declined 48 percent while sulfate concentrations declined 26 percent.
- ◆ Total (dry + wet) sulfur deposition declined by 38 percent from 1990 through 2008.
- ◆ Mean annual concentrations of total nitrate (nitric acid plus particulate nitrate) declined 25 percent in the eastern United States over the 19-year period.
- ◆ Total nitrogen deposition declined by 19 percent from 1990 through 2008.
- ◆ Ozone levels measured at CASTNET sites in 2008 continue to show a downward trend that began after a peak in 2002. The mean fourth highest daily maximum 8-hour average ozone concentration (69 ppb) for 2008 was the lowest in the history of the network. Only two eastern and four California sites recorded exceedances of the 8-hour standard of 0.08 ppm during the most recent 3-year period (2006–2008). During 2008, no eastern site recorded a fourth highest daily maximum 8-hour ozone concentration above 85 ppb. The recent declines in rural ozone and nitrate levels have been attributed to the documented decline in nitrogen oxide emissions in the eastern United States.
- ◆ Measurements taken during 2008 and historical data collected over the period 1990–2007 were analyzed relative to data quality objectives and their associated metrics. These analyses demonstrate that CASTNET data can be used with confidence and that CASTNET continues to produce information of the highest quality.

Chapter I: CASTNET Overview

CASTNET is a national air quality monitoring network designed to provide data to assess trends in air quality, atmospheric deposition, and ecological effects due to changes in air pollutant emissions. CASTNET began operation in 1991 to provide long-term monitoring of air quality in rural areas and to determine trends in regional atmospheric nitrogen, sulfur, and ozone concentrations and deposition fluxes of sulfur and nitrogen pollutants. CASTNET was preceded by the National Dry Deposition Network, which began in 1987. The primary sponsors of CASTNET are the Environmental Protection Agency and the National Park Service. As of December 2008, the network operated 84 monitoring stations throughout the contiguous United States, Alaska, and Canada. Air quality measurements collected in 2008 show the continuing decline in pollution levels throughout the network. Mean annual sulfur dioxide concentrations and mean fourth highest daily maximum 8-hour average ozone concentrations were the lowest in the history of CASTNET.

Background

The U.S. Congress, under a provision in the 1990 Clean Air Act Amendments (CAAA), directed the Environmental Protection Agency (EPA) to establish the Clean Air Status and Trends Network (CASTNET) to assess the effectiveness of the Acid Rain Program (ARP), which was promulgated to reduce emissions of sulfur dioxide (SO₂) and nitrogen oxides (NO_x) from electric generating units (EGUs). Congress recognized the need to assess and track real-world environmental results as the ARP was implemented, and emission reductions became effective. Congress mandated that CASTNET provide consistent, long-term measurements for determining relationships between changes in emissions and any subsequent changes in air quality, atmospheric deposition, and ecological effects.

The ARP has produced significant reductions in SO₂ and NO_x emissions from EGUs since 1995. More recent NO_x emission control programs also produced substantive declines in NO_x emissions in the eastern United States. These programs include the Ozone Transport Commission (OTC) NO_x Budget (1999–2002) and the NO_x State Implementation Plan (SIP) Call/NO_x Budget Trading Program (NBP), which operated from 2003 through 2008. The NBP placed a cap on total NO_x emissions from EGUs in the eastern United States during the ozone season (May 1 through September 30) when the potential for ozone formation is high. The Clean Air Interstate Rule (CAIR), which was issued in March 2005, aimed to permanently lower SO₂ and NO_x emissions in the eastern United States. CAIR, as promulgated, established three compliance programs: an annual NO_x program, an ozone season NO_x program, and an annual

SO₂ program. Although CAIR was remanded back to EPA in 2008, these programs remain in effect while EPA works to develop a replacement rule. The first phase of the annual and ozone season NO_x requirements began in 2009. The SO₂ requirements will begin in 2010. EPA relies on CASTNET and other long-term monitoring networks to provide the air quality measurements used to assess the effectiveness of emission control programs.

CASTNET started as the National Dry Deposition Network (NDDN), which began operation in 1987. The 50 NDDN sites were transferred to CASTNET in 1991. The network, as of December 2008, included 84 monitoring stations at 82 site locations throughout the contiguous United States, Alaska, and Canada. CASTNET is sponsored by EPA and the National Park Service (NPS). NPS began its participation in CASTNET in 1994 under an agreement with EPA. With the participation of NPS, the network became a national, rather than a primarily eastern, network. NPS is responsible for the protection and enhancement of air quality and related values in national parks and wilderness areas. The CASTNET sites sponsored by NPS as of December 2008 numbered 25. Additional information on the NPS Air Monitoring program can be found on the Web site: <http://www.nature.nps.gov/air/monitoring/>.

CASTNET stations measure rural, regionally representative concentrations of sulfur and nitrogen pollutants and ozone in order to detect and quantify trends, define the geographic distribution of rural pollutants, and estimate dry deposition of pollutants. Dry deposition is calculated from CASTNET concentration measurements and modeled deposition velocities using the Multi-Layer Model (MLM). The model requires several meteorological parameters and information on vegetation and land use specific to each site. CASTNET data are also used for atmospheric model evaluation, e.g., the Community Multiscale Air Quality (CMAQ) modeling system, and to provide input to ecological models.

This report summarizes CASTNET/NDDN monitoring activities and the resulting concentration and deposition data collected over the 19-year period from 1990 through 2008. Additional information, previous annual reports, and other CASTNET documents can be found on the EPA Web site, <http://www.epa.gov/castnet/>. The CASTNET database is also available to the public by accessing the “Download Data” link on EPA’s CASTNET Web page. The Web site provides a complete archive of concentration and deposition data for all EPA and NPS-sponsored CASTNET sites. Fully validated data are available approximately 10 months following collection. NPS data collected during 2008 from the NPS-sponsored CASTNET sites and the NPS Gaseous Pollutant Monitoring Program are available in the NPS 2008 Annual Data Summary Report (Ray, 2009) and the NPS 2008 Data Quality Assurance Report (ARS, 2009).

Cooperating Networks

In addition to EPA and NPS, the principal sponsors, CASTNET monitors air quality and deposition in cooperation with other national networks.

- ♦ **National Atmospheric Deposition Program (NADP)** operates the **National Trends Network (NTN)**, which includes monitoring stations with wet deposition samplers to measure the concentrations and deposition rates of air pollutants removed from the atmosphere by precipitation. NADP/NTN operates wet deposition samplers at or near virtually every CASTNET site. NADP operates the **Mercury Deposition Network (MDN)**, which operates samplers to measure mercury in precipitation. MDN samplers are operated at several CASTNET sites. Additionally, NADP operates the **Ammonia Monitoring Network (AMoN)**, which operates triplicate passive ammonia (NH₃) samplers at about 20 NTN locations. AMoN has been operating for two years as a NADP initiative. AMoN provides information on 2-week average NH₃ concentrations. Additional information on NADP can be found on its Web site: <http://nadp.sws.uiuc.edu/>.
- ♦ **Canadian Air and Precipitation Monitoring Network (CAPMoN)** operates 28 measurement sites throughout Canada and one in the United States. CASTNET and CAPMoN both operate samplers at monitoring stations in Ontario, Canada and at Pennsylvania State University.
- ♦ **Interagency Monitoring of Protected Visual Environments (IMPROVE)** measures aerosol pollutants near more than 30 CASTNET sites. IMPROVE measures particulate air pollutants that affect visibility.

EPA relies on CASTNET and these other long-term national networks to generate the data needed to assess the effectiveness of promulgated emission control programs under several different mandates including the Government Performance and Results Act, the National Acid Precipitation Assessment Program (NAPAP), Title IX of the CAAA, and the United States – Canada Air Quality Agreement.

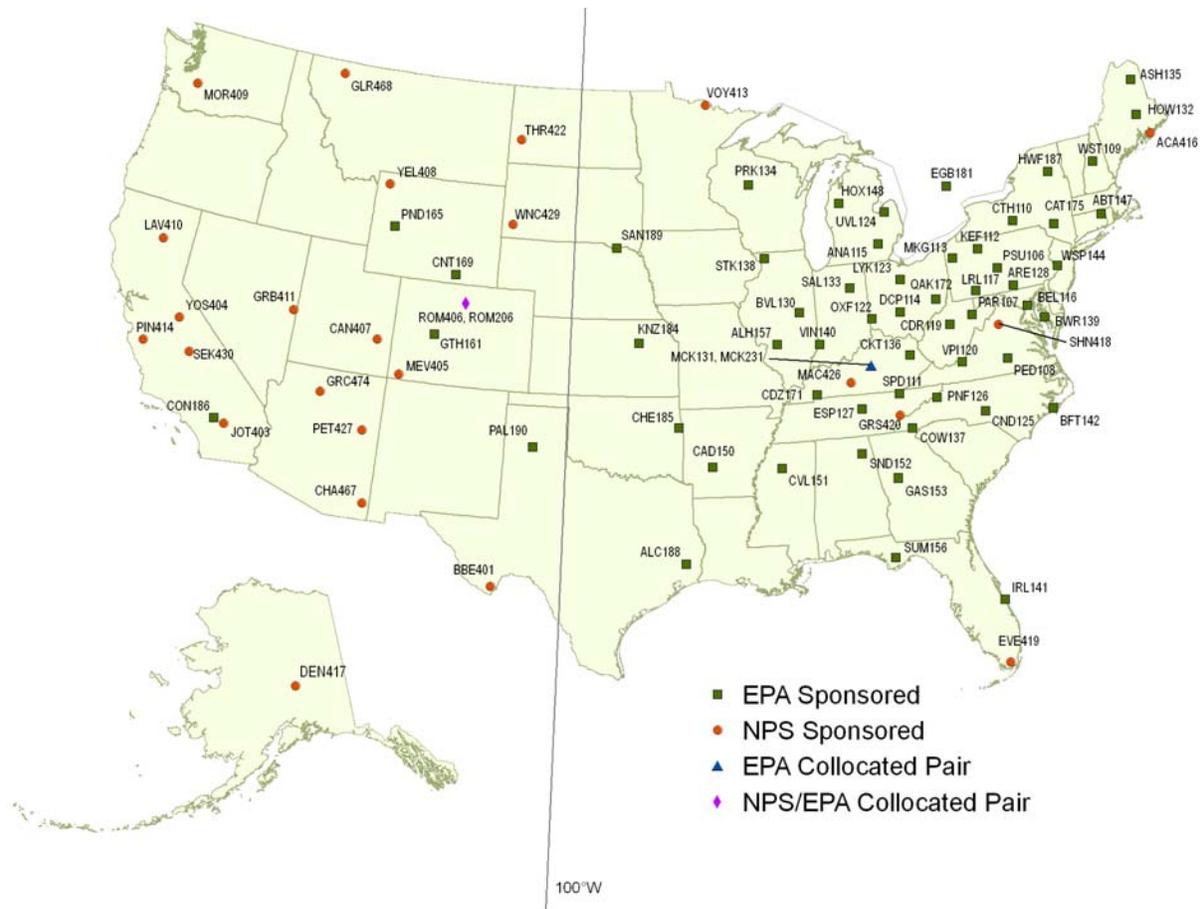
Although EPA and NPS are the primary sponsors of CASTNET, other organizations, Native American tribes, universities, and government agencies play a role in sponsoring individual CASTNET sites. These co-sponsors provide in-kind services that support the overall operation of a site, including site operation, land use, or both. All of the sites added during recent expansion of the network (since 2001) have an associated co-sponsor. Three CASTNET monitoring sites are located on tribal lands: Cherokee Nation in eastern Oklahoma (CHE185),

Alabama-Coushatta in eastern Texas (ALC188), and Santee Sioux in northern Nebraska (SAN189). State agencies also operate special purpose air pollutant measurement devices at some CASTNET sites.

Locations of Monitoring Sites

Figure I-1 shows the locations of CASTNET monitoring sites as of December 2008. Eighty-four sites were operational at 82 distinct locations. Most CASTNET sites are located in rural or remote locations away from pollutant emission sources and heavily populated areas. Appendix A provides the location of each site by state and includes information on start date, latitude, longitude, elevation, and the parameters measured. For the purposes of this report, CASTNET sites are called “western” or “eastern” depending on whether they are west or east of 100 degrees west longitude. In general, sample flow rates are set to 1.5 liters per minute (lpm) in the east and at a higher rate of 3.0 lpm in the west due to the lower pollutant concentrations generally found in the western United States.

Figure I-1 CASTNET Sites as of December 2008



CASTNET Reference Sites

Chapters 2 through 4 present maps illustrating the magnitude of pollutant concentrations and deposition fluxes across the United States. In addition, measurements from 34 CASTNET eastern reference sites (Figure I-2) were analyzed for each pollutant in order to determine trends in concentrations and trends in rates of dry, wet, and total deposition. These 34 sites have been reporting CASTNET measurements since at least 1990. The reference sites were selected using criteria similar to those used by EPA in its National Air Quality and Emissions Trends Report (2000). The criteria include site longevity and data completeness. EPA's procedures to interpolate and extrapolate quarterly mean data were also used to replace missing or invalid data. Trends in pollutant concentrations measured at CASTNET western reference sites (Figure I-3) are also presented. The 17 western reference sites have been operating since at least 1996.

In chapters 2 through 4, the data from the 34 eastern reference sites were aggregated and then presented using box plots for the period 1990 through 2008. Data from the 17 western reference sites were aggregated and then presented using box plots for the period 1996 through 2008.

Figure I-2 CASTNET Eastern Reference Sites



Figure I-3 CASTNET Western Reference Sites



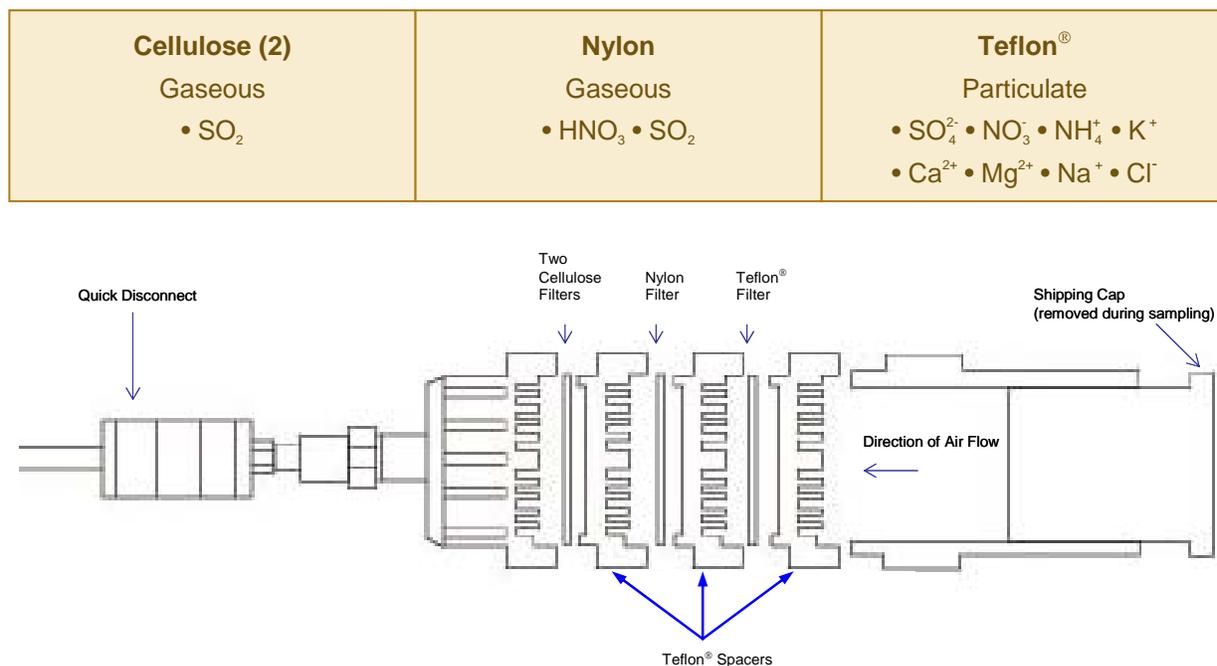
Measurements Collected at CASTNET Sites

CASTNET was designed primarily to measure trends in seasonal and annual average concentrations and to model depositions over many years. Consequently, measurement of weekly average (Tuesday to Tuesday) concentrations was selected as the basic sampling strategy. Over the course of the week, air is drawn at a controlled flow rate through an open-face 3-stage filter pack (Figure I-4) mounted atop a 10-meter tower to collect air pollutants in the form of gases and particles. The first stage of the filter pack encloses a Teflon filter, the second a nylon filter, and the third holds two potassium carbonate (K_2CO_3)-impregnated cellulose filters. The filter pack is changed out each Tuesday and shipped to the analytical chemistry laboratory for analysis.

CASTNET Ambient Measurements

- ◆ Sulfur dioxide (SO_2)
- ◆ Particulate sulfate (SO_4^{2-})
- ◆ Particulate nitrate (NO_3^-)
- ◆ Nitric acid (HNO_3)
- ◆ Particulate ammonium (NH_4^+)
- ◆ Particulate calcium (Ca^{2+})
- ◆ Particulate sodium (Na^+)
- ◆ Particulate magnesium (Mg^{2+})
- ◆ Particulate potassium (K^+)
- ◆ Particulate chloride (Cl^-)
- ◆ Ozone (O_3)
- ◆ Meteorological variables and information on land use and vegetation

Figure I-4 Three-Stage Filter Pack



The filter packs are prepared, loaded, shipped, received, extracted, and analyzed at the MACTEC Gainesville, FL laboratory. Following receipt from the field, exposed Teflon filters and blanks are extracted and then analyzed for sulfate (SO_4^{2-}), nitrate (NO_3^-), and concentrations of chloride (Cl^-) by micromembrane-suppressed ion chromatography (IC). Teflon filter extracts are also analyzed for ammonium (NH_4^+) by the automated indophenol method with the

Bran+Luebbe AutoAnalyzer 3. The Teflon filter extracts are additionally analyzed for calcium (Ca^{2+}), sodium (Na^+), magnesium (Mg^{2+}), and potassium (K^+) by inductively coupled plasma-atomic emission spectrometry (ICP-AES) using a Perkin Elmer Optima 3000 Dual View spectrometer. The cellulose filter extracts are analyzed for SO_2 as sulfate using IC. In November 2008, the analytical laboratory began using the Dionex AS22 analytical column/AG22 guard column for the impregnated cellulose filter analyses. The nylon filter extracts are analyzed via IC for nitric acid (HNO_3) and for SO_2 . The SO_2 concentrations from the cellulose and nylon filters are summed to obtain total SO_2 .

CASTNET also measures hourly ozone (O_3) concentrations, one of the major components of smog. In addition to the air pollutants, CASTNET sites collect hourly measurements of temperature, solar radiation, relative humidity, precipitation, wind speed and direction, standard deviation of wind direction (sigma theta), and surface wetness. These meteorological measurements are used to gauge the transport of air pollutants and as input to the MLM, a numerical model used for estimating dry deposition to ecosystems in the atmospheric boundary layer. The ozone and meteorological measurements are recorded continuously and archived as hourly averages.

Delivery of CASTNET Ozone Measurements to the EPA Air Quality System

CASTNET was not designed to operate as a network for demonstration of compliance with National Ambient Air Quality Standards (NAAQS) for O_3 . Instead, CASTNET was designed to provide information on geographic patterns in regional, rural O_3 levels. However, because of interest in rural O_3 concentrations by the regulatory and scientific communities, EPA has directed MACTEC to prepare EPA-sponsored sites to meet 40 CFR Part 58 monitoring requirements and to ultimately deliver CASTNET O_3 measurements to the EPA Air Quality System (AQS). AQS is EPA's repository of ambient air quality monitoring data from more than 10,000 monitors. State, local, and tribal agencies and other organizations collect data and submit them to AQS on a periodic basis. AQS data are flagged with status codes to indicate the type of data (e.g., regulatory or non-regulatory) submitted.

CASTNET monitoring equipment has been upgraded as one step toward meeting Part 58 criteria. Upgrades include new ozone analyzers, data loggers with improved data communication, long-term manufacturer support for analyzers and loggers, and internal shelter temperature sensors. These instrument upgrades support conversion to regulatory-compliant operating and reporting procedures. The new data loggers and

analyzers allow more efficient tracking and recording of shelter temperature and provide system monitoring data that enable remote troubleshooting and optimization of operations.

The data logger is programmed to control daily zero/span/precision (z/s/p) checks for the O₃ analyzer. The O₃ concentration needed for the precision (90 parts per billion [ppb]) and span (400 ppb) checks is produced internally by the onboard ozonator. Zero air is used for all quality control (QC) checks, including daily zero checks, and as the dilution medium for the ozonator precision and span checks. Instrument span and precision checks must be within ± 7 percent of reference value and zero checks must be ± 15 ppb for O₃ data to be considered valid.

All CASTNET continuous measurements, including the O₃ data and supporting parameters, are collected via the data logger, polled remotely, archived at the CASTNET Data Management Center (DMC), and uploaded to AIRNow every hour. For sites to meet Part 58 monitoring criteria, data will be collected in accordance with EPA Quality Assurance (QA) Handbook Volume II (EPA, 2008b). Multi-point calibrations, which will be performed every six months, will also be in compliance with the EPA QA handbook. Ozone transfer standards will be used in accordance with EPA Technical Assistance Document, “Transfer Standards for the Calibration of Ambient Air Monitoring Analyzers for Ozone” (EPA, 2009d).

Data will be uploaded to the AQS data submittal Web application in batch format using text files of raw ozone data and measurement accuracy and precision data. The AQS data submittal Web application is accessed at <http://www.epa.gov/ttn/airs/airsaqs/aqsweb/>.

AQS is divided into two parts:

1. CDX – Central Data Exchange: A “Windows Explorer” for AQS. This component transfers and manages files to and from AQS. Batch data files are uploaded here and become available in the AQS application for processing.
2. AQS Application: This application is used to interact with, process, and create reports of data.

Calculating Dry Deposition

Dry deposition processes are modeled as resistances to deposition. The original network design was based on the assumption that dry deposition or flux could be estimated as the linear product of measured pollutant concentration (C) and modeled deposition velocity (V_d). The equation to estimate flux is shown in Figure I-5. Measured atmospheric concentrations are calculated based on the mass of each analyte in each filter extract and the volume of air

sampled. The rate of deposition of a pollutant, also known as deposition velocity, is influenced by meteorological conditions, vegetation, and atmospheric and plant chemistry. The deposition velocity values for each site are calculated for each hour of each year using the MLM. The MLM is described by Meyers *et al.* (1998) and Finkelstein *et al.* (2000). The data used in the MLM to estimate dry deposition are derived from meteorological measurements and pollutant concentrations taken at the site together with an estimation of the vegetation leaf-out and leaf area index (LAI).

The schematic of the MLM in Figure I-5 shows the relationships among the various resistances and illustrates the meteorological and other data that are required as model input. An improved version of the MLM (Schwede, 2006) includes changes to the soil moisture factor, which affects the stomatal and soil resistances, and to the radiation algorithm, which also affects the stomatal resistance. The deposition velocities and fluxes presented in Chapter 3 were calculated using the updated model.

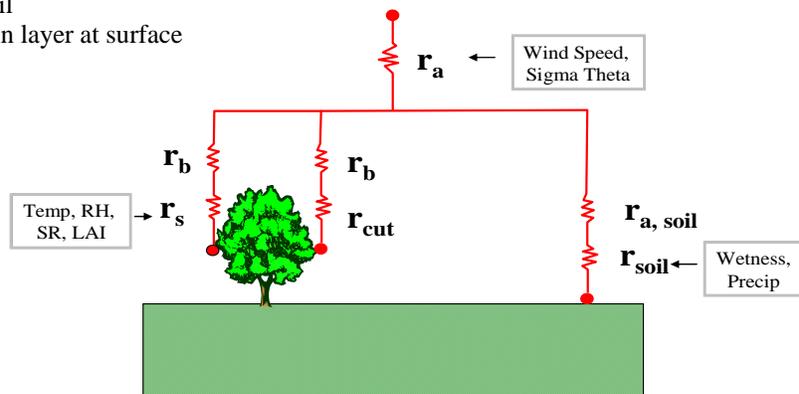
Figure I-5 Multi-Layer Model

$$\text{Flux} = \bar{C} \times \bar{V}_d$$

$$1/V_d = \frac{1}{\frac{1}{r_s + r_b} + \frac{1}{r_{cut} + r_b} + \frac{1}{r_{a, soil} + r_{soil}}} + r_a$$

Resistances

- r_a = aerodynamic
- $r_{a, soil}$ = aerodynamic near soil
- r_b = boundary layer in thin layer at surface
- r_{cut} = cuticular
- r_s = stomatal
- r_{soil} = soil



Sulfur Dioxide and Nitrogen Oxides Emissions

The ARP, established under Title IV of the 1990 CAAA, was designed to reduce the adverse effects of acid deposition by requiring major reductions of SO₂ and NO_x emissions from the electric power industry. EPA recently assessed the effects of the ARP and related programs in

its 2008 progress report (EPA, 2009a). The report summarizes 2008 compliance with the ARP and reported progress toward achieving environmental goals.

Under the ARP, SO₂ reductions are achieved using a cap and trade program that sets a permanent cap on the total amount of SO₂ that may be emitted by all regulated EGUs in the contiguous United States. The program began in 1995 and is underway with the final 2010 SO₂ cap set at 8.95 million tons, a level equal to about one-half of the emissions from the power sector in 1980. Use of a market-based cap and trade mechanism to reduce SO₂ emissions allows flexibility for individual combustion units to select their own methods of compliance. Currently, one allowance provides a regulated unit limited authorization to emit one ton of SO₂. The CAAA allocated allowances to regulated units based on historic fuel consumption and specific emission rates prior to the start of the program. The total allowances allocated for each year equal the SO₂ emission cap. The program encourages early reductions by allowing sources to bank unused allowances in one year and use them in a later year.

The ARP uses a more traditional approach to achieve NO_x emission reductions. Rate-based limits apply to most of the coal-fired electric utility boilers subject to the SO₂ emission reductions, and no nationwide cap has been placed on NO_x emissions. Other NO_x emission control programs have resulted in significant reductions in NO_x emissions during the ozone season (see Chapter 4). Two prominent control programs are the OTC NO_x Budget (1999–2002) and the NO_x SIP Call/NBP, which began in 2003 and continued through 2008. The NBP placed a cap on total NO_x emissions from EGUs during the ozone season (May 1 through September 30) when the potential for ozone formation is high. The CAIR NO_x ozone season trading program replaced the NBP in 2009. Although CAIR was remanded back to EPA in 2008, the CAIR programs remain in effect while EPA works to develop a replacement rule.

The ARP is comprised of two phases for the reduction of SO₂ and NO_x. Phase I applied primarily to the largest coal-fired EGUs from 1995 through 1999 for SO₂ and from 1996 through 1999 for NO_x. Phase II for both pollutants began in 2000. In 2008, the SO₂ Phase II requirements affected 3,572 operating units. The Phase II NO_x requirements applied to 969 of those operating units that exceeded 25 megawatts and burned coal between 1990 and 1995 (EPA, 2009a). Under the ARP's emission reduction requirements, total SO₂ emissions from ARP sources were 7.6 million tons in 2008; total NO_x emissions from ARP sources were 3.0 million tons.

Figure I-6 presents state-by-state total annual SO₂ emissions for Phase I and Phase II electric utility plants for four years (1990, 1995, 2000, and 2008). The most significant reduction in SO₂ emissions occurred in and near the Ohio River Valley where many coal-fired power plants are located. Many of the states with the highest SO₂ emissions realized the largest reductions. The seven states with the greatest annual reductions include Ohio, which decreased emissions by

1.5 million tons since 1990, and Illinois, Indiana, Kentucky, Missouri, Tennessee, and West Virginia. Annual NO_x emissions by state are depicted in Figure I-7 for the same four years. States with the highest NO_x emissions also produced the most significant declines. Every state east of the Mississippi River experienced a significant decline in annual NO_x emissions from 2000 to 2008. For example, NO_x emissions from Illinois power plants declined by 47 percent from 2000 to 2008.

Figure I-6 Annual Utility SO₂ Emissions (Phase I and Phase II Plants only)

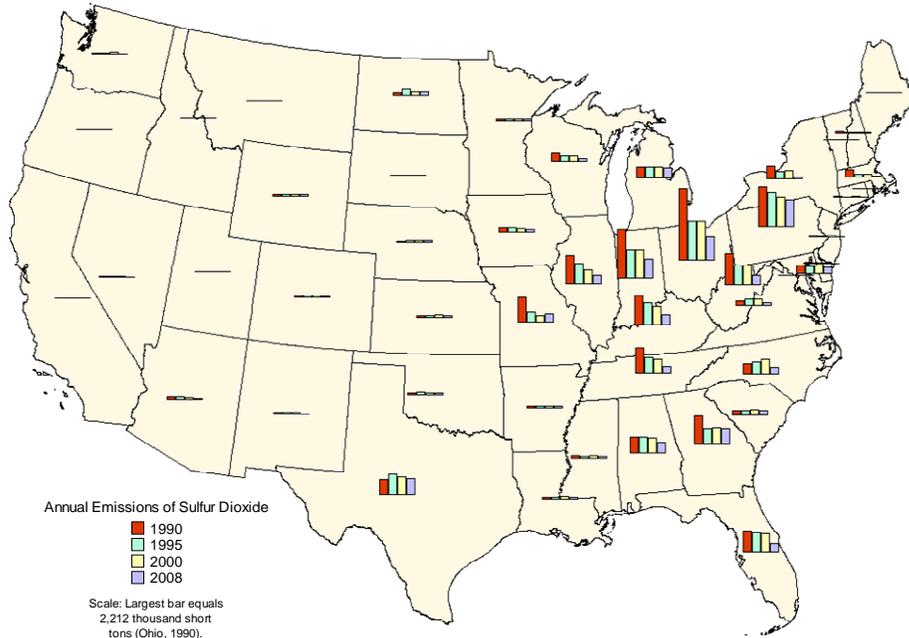
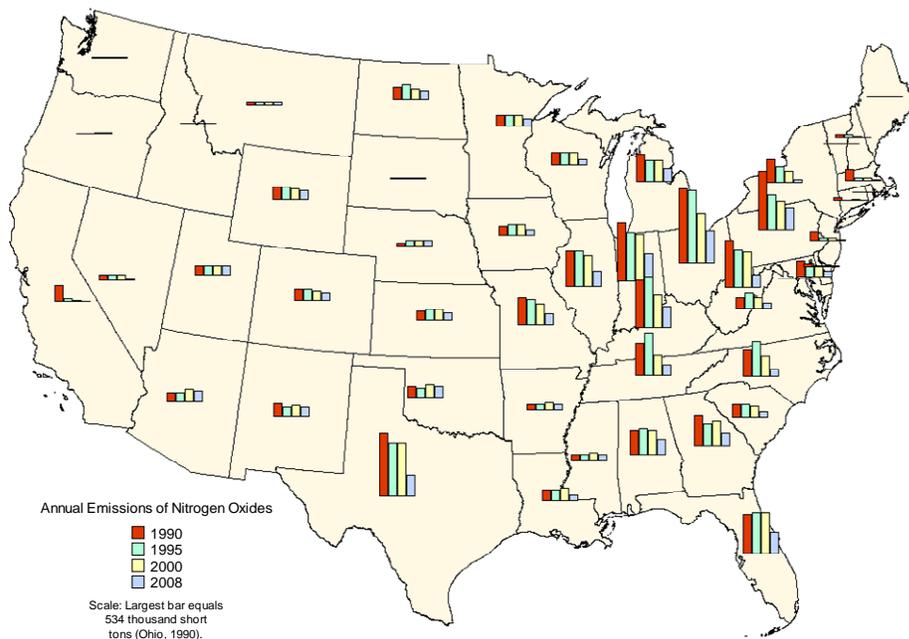


Figure I-7 Annual Utility NO_x Emissions (Phase I and Phase II Plants only)



2008 Significant Events

January
The first Campbell Scientific CR3000 data logger was installed at Palo Duro Canyon State Park, TX.
February
Analog cellular service ended. First installations of the AirLink Raven X cellular Ethernet modems were completed to provide access and communication to sites without landlines.
May
Due to budget constraints, it was decided that cloud water sampling would not be conducted at CLD303, TN for the 2008 sampling season under the Mountain Acid Deposition Program (MADPro). This is the first season without sampling since MADPro began sampling in 1994.
July
MACTEC determined the date and time when model runs initialized with drought and saturated model runs coalesced for each site for the entire record of MLM simulations. This point of coalescence is now the starting point for each site's dry deposition record.
September
MACTEC's CASTNET analytical laboratory tied for first place out of 39 participating laboratories in Environment Canada's laboratory intercomparison Proficiency Test 0092.
October
Yagi directional antennae were installed at select remote sites to boost the cellular signal at each site and allow use of Raven X Cellular Ethernet modems for Internet Protocol (IP) connections. Supplemental CASTNET training videos became accessible through a dedicated MACTEC Web site. Operators can access videos from the sites.
November
MACTEC's CASTNET analytical laboratory began using the Dionex AS22 analytical column/AG22 guard column for the impregnated cellulose filter analyses, replacing the Dionex AS4A analytical column /AG4A guard column. The new column provides improved peak separation and is less susceptible to matrix interferences.

NPS produces a biannual CASTNET newsletter called *The Monitor* that contains information about some of the changes and important events affecting CASTNET operations. For an electronic library of current and previous newsletters, go to:
<http://www.nature.nps.gov/air/Pubs/theMonitor.cfm>.

Chapter 2: Atmospheric Concentrations

Three-stage filter packs are used to measure 7-day average concentrations of SO_2 , SO_4^{2-} , HNO_3 , NO_3^- , NH_4^+ , Cl^- , and four earth metals at 84 CASTNET monitoring stations. Measured annual mean concentrations of sulfur species have decreased steadily since 1990. Concentrations of nitrogen species remained relatively steady from 1990 until 2000 when they began to decline as NO_x emission reduction programs were implemented. Trends in mean annual SO_2 , SO_4^{2-} , total NO_3^- , and NH_4^+ concentrations aggregated over the 34 eastern reference sites are shown over the period 1990 through 2008 and for the 17 western sites for the period 1996 through 2008 using box plots for each year. All four parameters declined over the 19-year period. SO_2 and SO_4^{2-} concentrations declined significantly since 2005. SO_2 concentrations measured at the 17 western reference sites also declined. Concentrations of the other three pollutants measured at the western sites did not change significantly.

Separate maps depicting the annual mean concentrations of SO_2 , SO_4^{2-} , total nitrate ($\text{HNO}_3 + \text{NO}_3^-$), and NH_4^+ are presented in this chapter. Additional maps are provided in CASTNET quarterly reports (MACTEC 2008a, 2008d, 2009c, 2009a). The concentration shading used in the maps in this chapter was prepared using an algorithm based on inverse distance weighting and the scales specified on each map. In addition, trends in concentrations over the 19-year period (1990 through 2008) were derived from the 34 eastern CASTNET reference sites (Figure I-2) and are presented using box plots. Trends in annual mean concentrations were also estimated from measurements aggregated over the 17 western CASTNET reference sites (Figure I-3) for the period 1996 through 2008.

Sulfur Dioxide

Sulfur dioxide is a gaseous pollutant emitted during the combustion of coal, oil, and other fossil fuels that contain sulfur. EGUs constitute the largest source of SO_2 in the United States. SO_2 is a criteria pollutant regulated by the NAAQS, which are designed to protect human health and welfare. To be compliant under the SO_2 NAAQS, the annual average concentration must be below 80 micrograms per cubic meter ($\mu\text{g}/\text{m}^3$) of air. SO_2 gas reacts in the atmosphere to form various acidic compounds including sulfuric acid (H_2SO_4), a significant contributor to acid rain, and ammonium sulfate [$(\text{NH}_4)_2\text{SO}_4$], a major component of fine particle matter ($\text{PM}_{2.5}$). Fine particles are regulated by the NAAQS since $\text{PM}_{2.5}$ contributes to negative health effects and also to degradation of atmospheric visibility, including range and detail. The NAAQS for an annual average $\text{PM}_{2.5}$ concentration is $15 \mu\text{g}/\text{m}^3$.

Annual mean SO_2 concentrations measured in 2008 are presented in Figure 2-1. Six states centered on and downwind of the Ohio River Valley in the eastern United States recorded

mean concentrations greater than or equal to $5.0 \mu\text{g}/\text{m}^3$ of air. The region covers Indiana, Ohio, Pennsylvania, New Jersey, Maryland, and western Virginia. Quaker City, OH (QAK172) measured the highest concentration in the network with a value of $11.4 \mu\text{g}/\text{m}^3$. Only two western sites (Figure 1-1) measured an annual mean SO_2 concentration greater than or equal to $1.0 \mu\text{g}/\text{m}^3$. The western sites are Palo Duro Canyon State Park, TX (PAL190) and Theodore Roosevelt National Park, ND (THR422).

Figure 2-1 Annual Mean SO_2 Concentrations ($\mu\text{g}/\text{m}^3$) for 2008

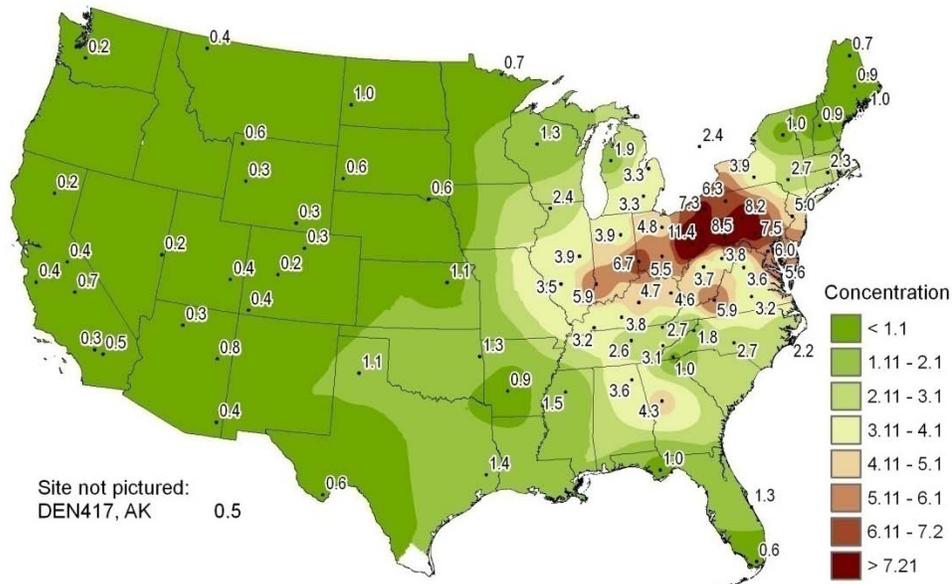
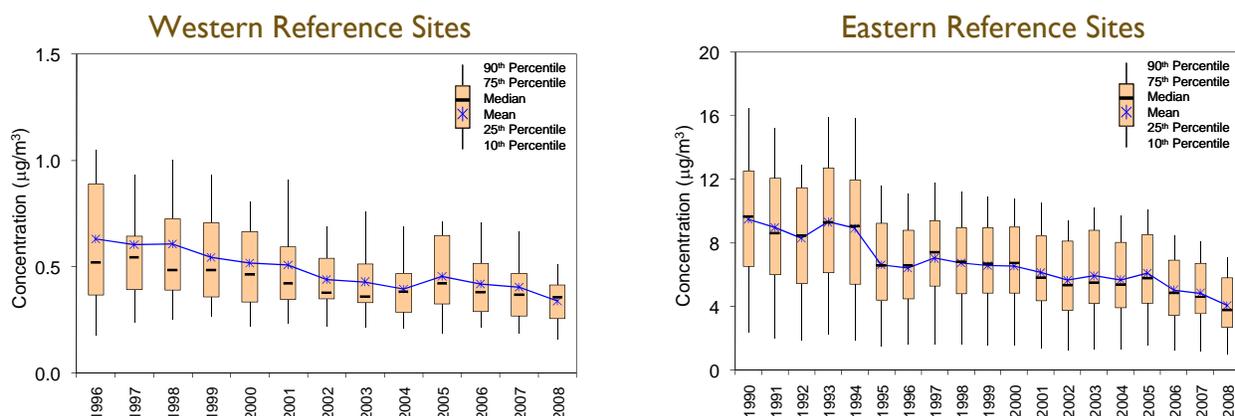


Figure 2-2 shows box plots of annual mean SO_2 concentrations aggregated over the 34 eastern reference sites from 1990 through 2008 (right side) and the 17 western reference sites from 1996 through 2008 (left side). The y-axes on the western and eastern plots have different scales. The box plots for the eastern sites show a downward trend. The most substantial reduction in ambient SO_2 was recorded in 1995 at the beginning of the ARP. A noteworthy decline was also recorded from 2005 through 2008. Three-year mean concentrations for 1990–1992 and 2006–2008 were $8.9 \mu\text{g}/\text{m}^3$ and $4.6 \mu\text{g}/\text{m}^3$, respectively. This change constitutes a 48 percent reduction in 3-year mean SO_2 concentrations between the two time periods. The 2008 mean level of $4.1 \mu\text{g}/\text{m}^3$ was the lowest mean value measured by the eastern reference sites in the history of the network.

The box plots for the western reference sites indicate a decline in annual mean SO_2 concentrations aggregated over the 17 sites. Three-year mean SO_2 concentrations for 1990–1992 and 2006–2008 were $0.6 \mu\text{g}/\text{m}^3$ and $0.4 \mu\text{g}/\text{m}^3$, respectively. This change constitutes a 37 percent reduction in 3-year mean SO_2 concentrations over the 13 years. The aggregated mean SO_2 concentrations from the western reference sites were more than 10 times lower than mean concentrations measured at the eastern reference sites.

Figure 2-2 Trend in Annual Mean SO₂ Concentrations (µg/m³)

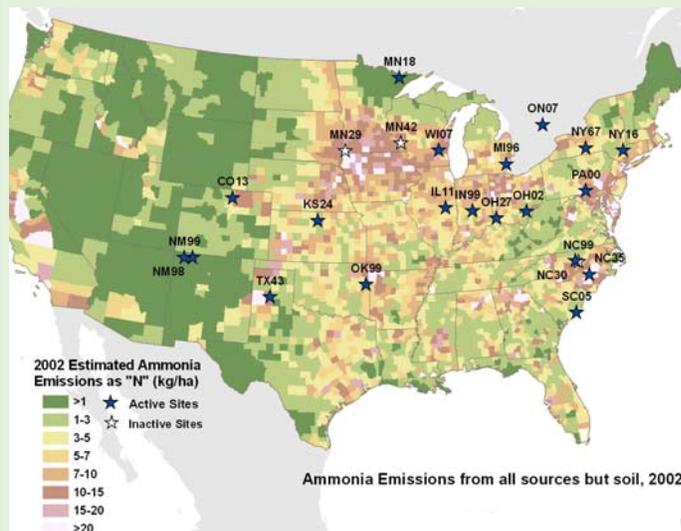


Ammonia Monitoring Network (AMoN)

Gaseous NH₃ affects chemical deposition and biogeochemical processes in ecosystems. NH₃ reacts with sulfur (S), nitrogen (N), and other acidic species to form (NH₄)₂SO₄ and ammonium nitrate (NH₄NO₃) particles, which exist in the atmosphere as PM_{2.5}. These aerosols, along with carbon aerosols, constitute the major fraction of PM_{2.5} pollution in the atmosphere. PM_{2.5} affects human health, visibility, and visual quality. PM_{2.5} is subject to long-range transport and contributes to international transboundary air pollutant issues.

Domesticated animals and fertilizer are the largest global source of anthropogenic atmospheric NH₃ emissions. Current estimates of NH₃ emissions are uncertain because of the diverse nature of agricultural and feedlot operations. Figure 2a illustrates modeled NH₃ emissions for 2002.

Figure 2a Ammonia Emissions: All Sources but Soil



Source: Carnegie Mellon University emissions model described in <http://www.cmu.edu/ammonia>.

A 2007 United States-Canada workshop on NH₃ (NADP, 2009) called for the creation of an extensive long-term network of passive NH₃ samplers with NADP serving as the coordinating organization. The participants also recommended the use of Radiello diffusive passive samplers throughout the network because of low detection limits and high reproducibility.

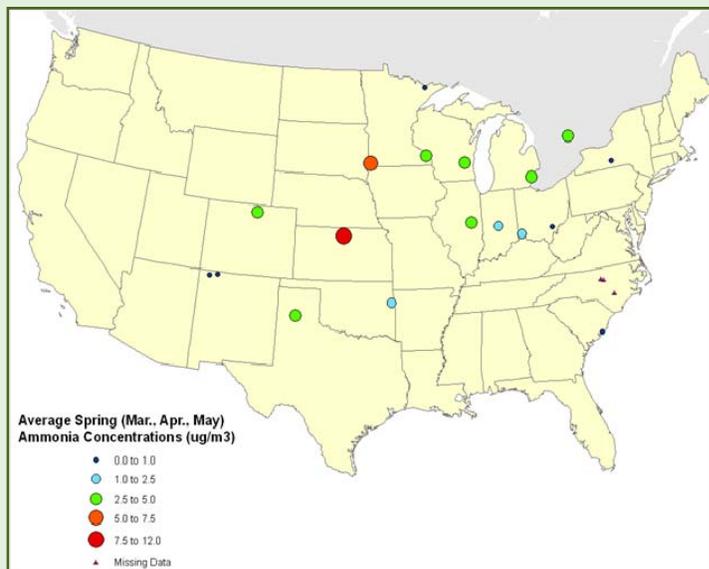
Table 2a AMoN Sampling Locations

Site	Name	Latitude (°N)	Longitude (°W)	Site	Name	Latitude (°N)	Longitude (°W)
CO13	Fort Collins, CO	40.5592	105.0781	NM99	Farmington, NM	36.7358	108.2380
IL11	Bondville (QAQC), IL	40.0520	88.3725	NY16	Millbrook, NY	41.7858	73.7414
IN99	Indianapolis, IN	39.7333	86.2833	NY67	Ithaca, NY	42.4014	76.6589
KS24	Glen Elder State Park, KS	37.0378	95.6264	OH02	Athens Super Site, OH	39.3167	82.1000
MI96	Detroit, MI	42.2500	83.2000	OH27	Cincinnati, OH	39.1500	84.5167
MN18	Fernberg, MN	48.0500	91.8166	OK99	Stilwell, OK	35.7514	94.6717
MN29	Blue Mounds, MN	43.6667	96.2333	ON07	Egbert, ON	44.2333	79.7833
MN42	Great River Bluffs, MN	44.0500	91.6333	PA00	Arendtsville, PA	39.9231	77.3078
NC30	Duke Forest, NC	35.7789	78.8003	SC05	Cape Romain Refuge, SC	32.9419	79.6591
NC35	Clinton Research Station, NC	35.0258	78.2783	TX43	Canonceta, TX	34.8803	101.6649
NC99	Durham, NC	35.8106	76.8978	WI07	Mayville, WI	43.4833	88.5300
NM98	Navajo Lake, NM	36.8097	107.6520				

Note: Collocated CASTNET sites are highlighted in yellow.

In response, NADP initiated AMoN near the end of 2007 as a network of passive NH₃ samplers across the United States with a site also operating in Egbert, ON. Table 2a lists the AMoN locations. In addition to providing data that describe atmospheric NH₃ concentrations over space and time, NADP is evaluating the Radiello passive NH₃ sampler against other passive sampler types and denuder-based measurement systems to record the accuracy, precision, and repeatability of the passive NH₃ samplers in order to determine the best sampler for the lowest cost.

Figure 2b Average Spring 2008 NH₃ Concentrations



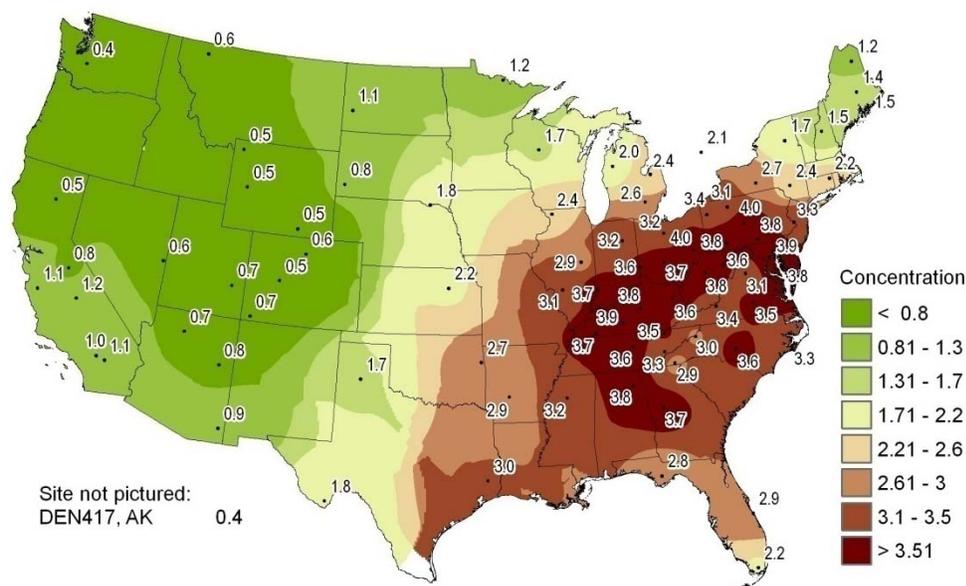
Information about AMoN can be found on NADP NH₃ Web site (<http://nadp.sws.uiuc.edu/nh3Net/>). The Web site discusses the sampling system and related information. Each site operates triplicate Radiello samplers over 2-week periods. The samplers are removed at the end of the two weeks and sent to the NADP Central Analytical Laboratory at the Illinois State Water Survey for chemical analysis. The site also maintains a database of NH₃ measurements.

Figure 2b was constructed from the triplicate 2-week data and shows spring 2008 mean seasonal NH₃ concentrations. The highest concentrations for 2008 were observed during the spring, perhaps associated with fertilizer application. Nine sites measured NH₃ values greater than 2.5 µg/m³.

Particulate Sulfate

Particulate sulfate is formed in the atmosphere through the transformation of SO_2 via both gas and aqueous (cloud) phase reactions. It typically exists in the atmosphere as $(\text{NH}_4)_2\text{SO}_4$, a major component of $\text{PM}_{2.5}$. Figure 2-3 provides a map of annual mean particulate SO_4^{2-} concentrations measured during 2008. Pennsylvania State University, PA (PSU106) and QAK172, OH each measured a concentration of $4.0 \mu\text{g}/\text{m}^3$. The map shows significant geographic gradients in SO_4^{2-} levels from the monitors in the vicinity of the Ohio River Valley northeastward to sites in New England and northwestward to sites in Wisconsin and northern Minnesota. Sulfate concentrations greater than or equal to $1.0 \mu\text{g}/\text{m}^3$ in the western region were measured at four sites in California, two in Texas, and the monitoring station at THR422, ND.

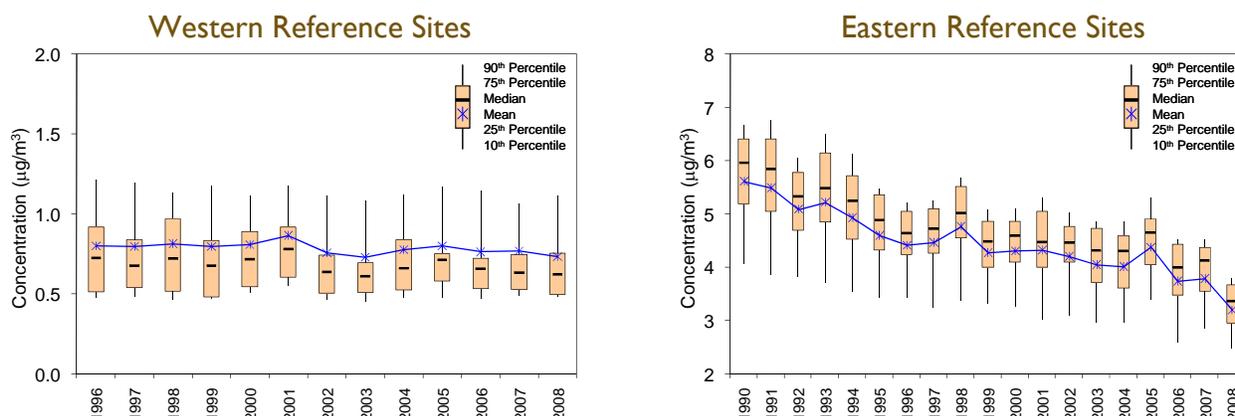
Figure 2-3 Annual Mean SO_4^{2-} Concentrations ($\mu\text{g}/\text{m}^3$) for 2008



Box plots of annual mean SO_4^{2-} concentrations from the 34 eastern reference sites from 1990 through 2008 are presented in the right half of Figure 2-4. The plots illustrate the trend in atmospheric SO_4^{2-} over the 19-year period. The figure shows a substantial decline in SO_4^{2-} with some interannual variability. The difference between 3-year means from 1990–1992 to 2006–2008 represents a 26 percent reduction in SO_4^{2-} from $5.4 \mu\text{g}/\text{m}^3$ to $3.6 \mu\text{g}/\text{m}^3$, respectively. The 2008 mean SO_4^{2-} level of $3.2 \mu\text{g}/\text{m}^3$ was the lowest in the history of the network.

The box plots for the western reference sites are provided on the left side of Figure 2-4. The boxes show no real change in annual mean SO_4^{2-} concentrations aggregated over the 17 sites. Mean sulfate concentrations measured at the western sites were lower than mean values measured at eastern sites.

Figure 2-4 Trend in Annual Mean SO_4^{2-} Concentrations ($\mu\text{g}/\text{m}^3$)

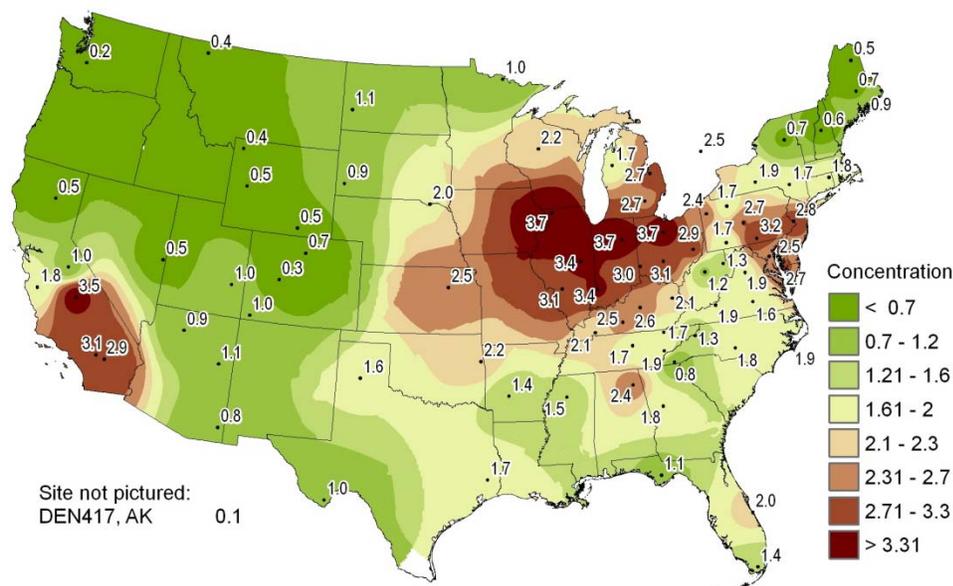


Total Nitrate

Total nitrate is the sum of HNO_3 and NO_3^- , which are formed from emissions of NO_x produced through the combustion of fossil fuels. Transportation sources are the largest contributors of NO_x emissions in the United States. Nitric acid exists as a gas and contributes to acid deposition. Nitrate is a component of $\text{PM}_{2.5}$ in the form of NH_4NO_3 and as larger particles, e.g., sodium nitrate (NaNO_3) or calcium nitrate [$\text{Ca}(\text{NO}_3)_2$]. A map of annual mean total nitrate concentrations for 2008 is shown in Figure 2-5. This chapter discusses data on total nitrate levels because of the uncertainty in individual species measurements and because total nitrate is more representative of a response to changes in NO_x emissions than either of its constituents alone. The map of 2008 total nitrate concentrations shows three regions with mean concentrations greater than or equal to $2.5 \mu\text{g}/\text{m}^3$. The largest region extends from eastern Kansas to Ontario. CASTNET monitors in six states in the Midwest, including Kansas, Illinois, Indiana, Kentucky, Ohio, and Michigan, measured concentrations greater than or equal to $2.5 \mu\text{g}/\text{m}^3$. The monitor at Egbert, ON (EGB181) also measured an annual mean concentration of $2.5 \mu\text{g}/\text{m}^3$. The second, smaller region covers sites in eastern Pennsylvania, New Jersey, and Maryland. Three sites in California constitute the third region. Three sites, including Stockton, IL (STK138); Salamonie Reservoir, IN (SAL133); and Lykens, OH (LYK123), measured the highest concentration ($3.7 \mu\text{g}/\text{m}^3$) in the network.

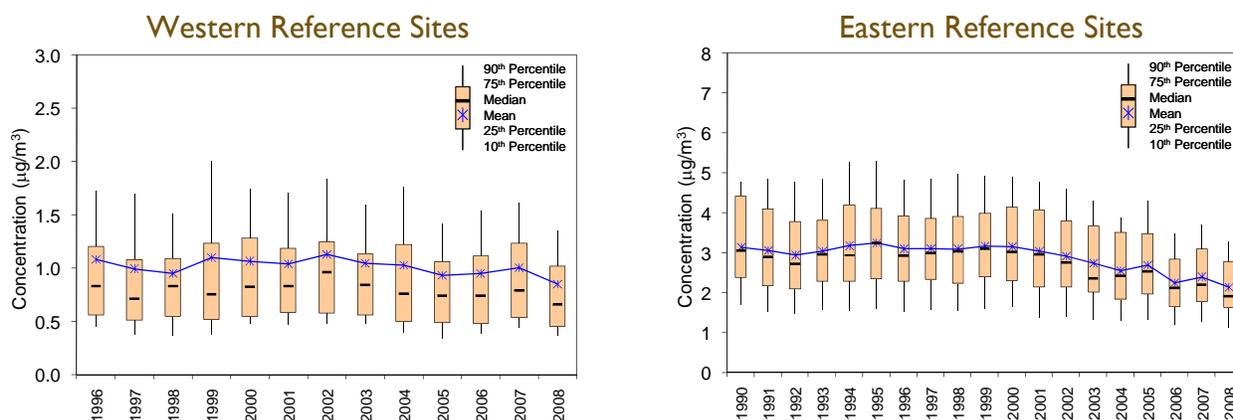
Box plots of total nitrate levels for 1990 through 2008 are given in Figure 2-6. The data shown on the right side of the figure were aggregated from the 34 eastern reference sites. The data show no trend in mean concentrations until 2000 when nitrate levels began to decline in response to NO_x emission control programs. Three-year mean levels declined from $3.0 \mu\text{g}/\text{m}^3$ to $2.3 \mu\text{g}/\text{m}^3$ from 1990–1992 to 2006–2008, producing a 25 percent reduction in total nitrate. Total nitrate levels measured at the eastern reference sites declined from a 2000 mean value of $3.2 \mu\text{g}/\text{m}^3$ to a 2008 level of $2.1 \mu\text{g}/\text{m}^3$, the lowest in the history of CASTNET.

Figure 2-5 Annual Mean Total Nitrate ($\text{NO}_3^- + \text{HNO}_3$) Concentrations ($\mu\text{g}/\text{m}^3$) for 2008



Data aggregated from the 17 western sites are shown on the left side of Figure 2-6. The 3-year mean total nitrate concentration for 2006–2008 was lower than the corresponding 1990–1992 level. However, this change is not considered substantive.

Figure 2-6 Trend in Annual Total Nitrate ($\text{NO}_3^- + \text{HNO}_3$) Concentrations ($\mu\text{g}/\text{m}^3$)

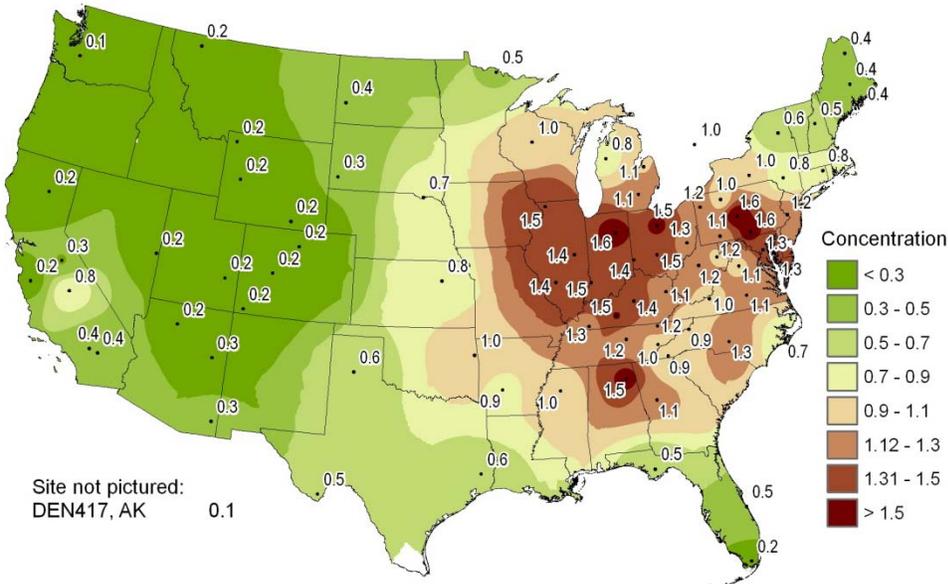


Particulate Ammonium

Particulate ammonium is formed when NH_3 , HNO_3 , SO_4^{2-} , and NO_3^- particles interact in the atmosphere. NH_3 is formed through volatilization of farm animal wastes and fertilizers and some combustion processes. Annual mean NH_4^+ concentrations for 2008 are depicted in Figure 2-7. No monitors measured concentrations above $2.0 \mu\text{g}/\text{m}^3$. The data show a flat geographic distribution across the eastern United States with most sites reporting values greater than or equal to $1.0 \mu\text{g}/\text{m}^3$. Higher concentrations were measured in the agricultural

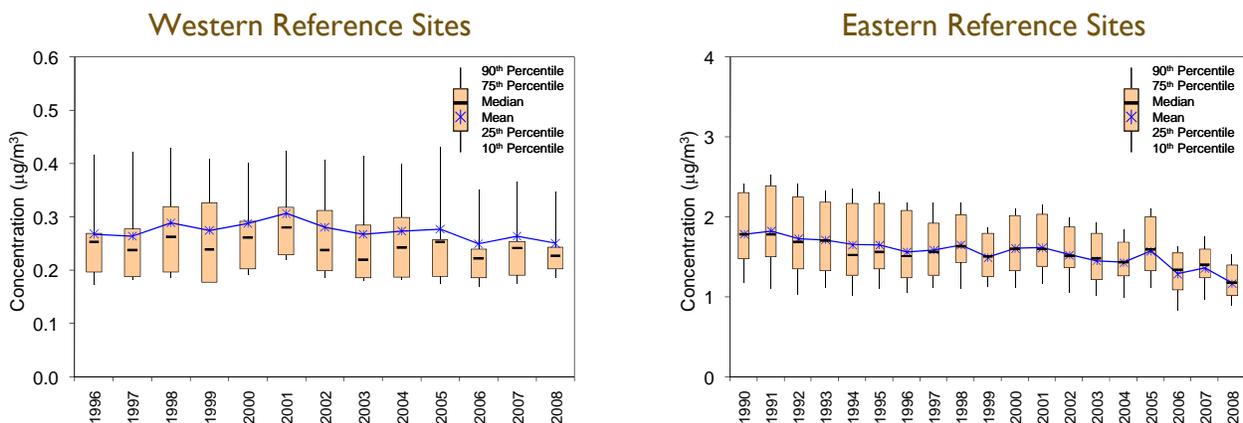
Midwest, Pennsylvania, and northern Alabama. The NH_4^+ concentrations measured at western sites were low with all sites measuring concentrations less than $1.0 \mu\text{g}/\text{m}^3$.

Figure 2-7 Annual Mean NH_4^+ Concentrations ($\mu\text{g}/\text{m}^3$) for 2008



Box plots of NH_4^+ concentrations are provided in Figure 2-8. The trend diagram for the eastern sites shows a reduction in mean NH_4^+ levels from 1990–1992 to 2006–2008. The 1990–1992 mean concentration was $1.8 \mu\text{g}/\text{m}^3$ and the 2006–2008 value was $1.3 \mu\text{g}/\text{m}^3$. The box plots characterizing the western sites show no change.

Figure 2-8 Trend in Annual Mean NH_4^+ Concentrations ($\mu\text{g}/\text{m}^3$)



Trends in Air Quality at CASTNET Western Reference Sites

Trends in concentrations over the 13-year period (1996–2008) were derived from the 17 CASTNET western reference sites (Figure 1-3). Box plots constructed from data aggregated from the 17 sites are given in Figures 2-2, 2-4, 2-6, and 2-8. Additional trend plots are shown in this discussion.

Figure 2c presents mean SO₂ trend lines for eight western sites selected to depict a range of results. Declines in annual mean SO₂ levels were observed at six of the sites. Noticeable reductions were measured at Chiricahua National Monument, AZ (CHA467) and Mesa Verde National Park, CO (MEV405). The box plots (Figure 2-2) derived from aggregated data show a 37 percent decline.

Trend lines for annual mean total nitrate concentrations measured at four California CASTNET sites are shown in Figure 2d. A significant decline was measured at Joshua Tree National Park, CA (JOT403) with about a 17 percent reduction between 3-year averages at the beginning and end of the 13-year period. Concentrations measured at JOT403, which is downwind of the Los Angeles Basin, were about three times higher than concentrations measured at Yosemite National Park, CA (YOS404).

Figure 2e depicts trends in all four annual mean pollutants measured at Mount Rainier National Park, WA (MOR409). The figure shows that all four pollutants declined with especially noticeable declines in SO₂ and SO₄²⁻ levels during 2000 to 2002. The decline in sulfur pollutants is attributed to the installation of SO₂ scrubbers at the Centralia Power Plant and a corresponding reduction in emissions.

Figure 2c Trends in Mean SO₂ Concentrations (µg/m³) at Eight Western Sites

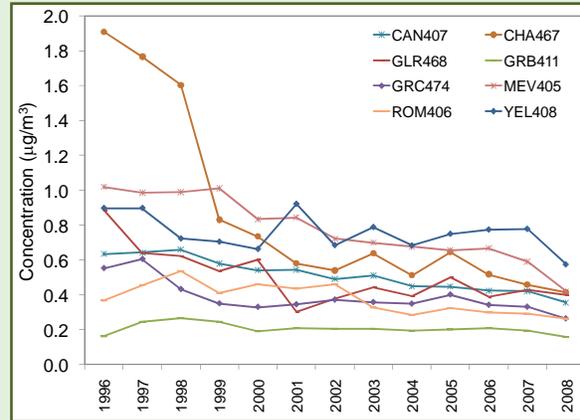


Figure 2d Trend in Annual Mean Total Nitrate Concentrations (µg/m³) at Four California Sites

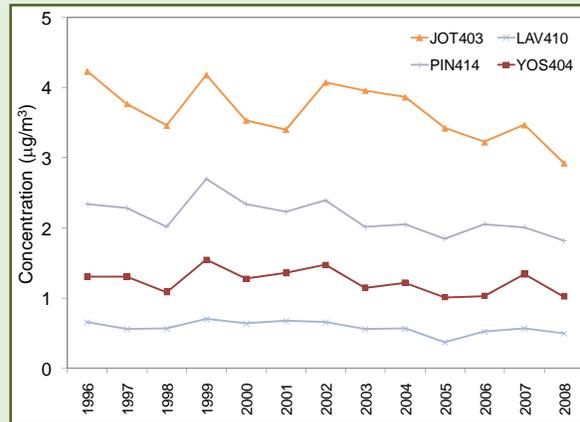
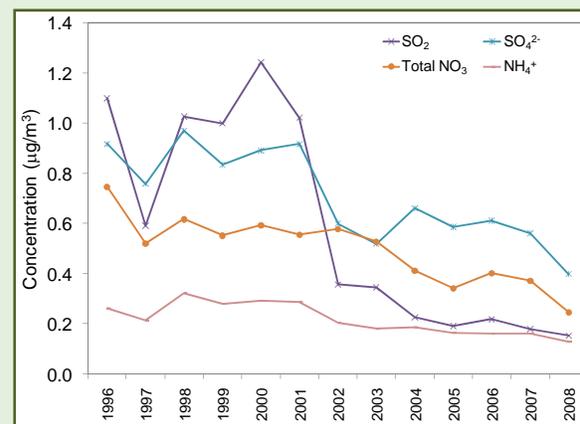


Figure 2e Trends in Annual Mean Pollutant Concentrations (µg/m³) at Mount Rainier National Park, WA



Chapter 3: Atmospheric Deposition

CASTNET was designed to provide estimates of the dry deposition of sulfur and nitrogen pollutants across the United States. CASTNET uses a hybrid approach to estimate dry deposition by combining measured pollutant concentrations and modeled deposition velocities. The Multi-Layer Model is used to calculate hourly deposition velocities for each monitoring site based on meteorological measurements and information on the vegetation within 1 km of each site. Total deposition is the sum of estimated dry deposition and measured wet deposition. Total sulfur deposition estimated for eastern reference sites has declined since 1990. The data show a 38 percent reduction in 3-year mean sulfur fluxes over the period from 1990–1992 to 2006–2008. Dry and total sulfur deposition estimated for 17 western sites declined over the period 1996 through 2008. Total nitrogen deposition estimated for the eastern reference sites declined by 19 percent. Total nitrogen deposition did not change at the western reference sites over the 13 years. Dry deposition is responsible for a significant percentage of total deposition, especially in major emission source regions.

Gaseous and particulate sulfur and nitrogen pollutants are deposited through dry and wet atmospheric processes. A critical objective of CASTNET is to estimate the rate, or flux, of dry deposition from the atmosphere to sensitive ecosystems based on measured meteorological and other environmental conditions. Flux values are estimated as the product of measured concentration data and MLM-modeled dry deposition velocities. Wet deposition measurements were obtained from NADP/NTN and combined with CASTNET dry deposition data to estimate total deposition. Dry sulfur, total sulfur, dry nitrogen, and total nitrogen deposition decreased during 2008. Precipitation-weighted mean concentrations of atmospheric sulfur declined over the past 19 years. Nitrogen concentrations in precipitation have declined slowly since 1998.

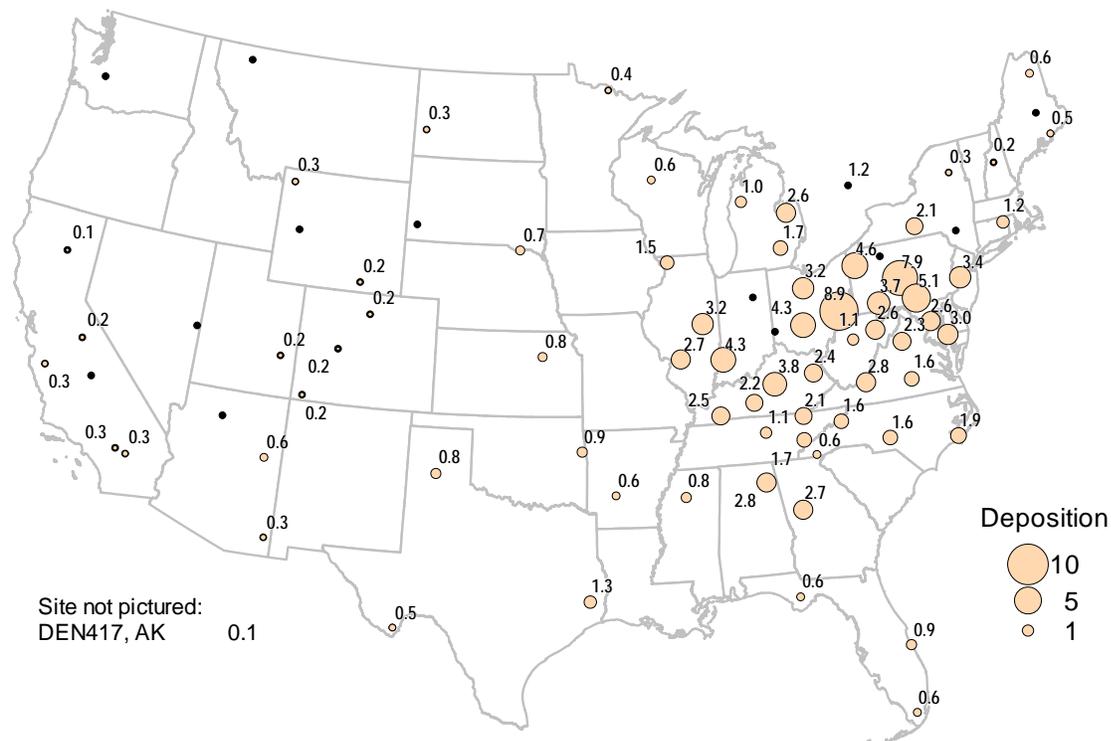
Dry deposition processes were simulated using the MLM (Figure I-5) as described by Meyers *et al.* (1998) and Finkelstein *et al.* (2000). The most recent version of the MLM (Schwede, 2006) was used for this report. The MLM was run using CASTNET filter pack concentrations and meteorological measurements with information on land use, vegetation, and surface conditions, to calculate deposition velocities for SO₂, HNO₃, O₃, and the particles, SO₄²⁻, NO₃⁻, and NH₄⁺. The deposition velocities were assumed to be identical for all particle species. Deposition velocity values were calculated for each of the pollutant species for each hour with valid meteorological data for each CASTNET site for the entire period 1990 through 2008. For a deposition velocity to be estimated, measurements of temperature, solar radiation, relative humidity, wind speed, and standard deviation of the wind direction (sigma theta) must all be

valid for the hour. Aggregation rules for CASTNET require three valid quarters for the calculation of an annual value. If an annual value was not available for a specific site, the results were not included on the maps presented in this chapter. Sites with no deposition estimates are shown as dots with no value. For trends analyses, missing values were replaced by interpolation or extrapolation using valid annual estimates. For example, if an intermediate year was missing, the value for that year was interpolated from adjacent years. If a 2008 value was missing, the 2008 value was assumed equal to the value for 2007.

Sulfur Deposition

MLM runs were made separately for SO_2 and SO_4^{2-} . The model calculations were summed to obtain estimates of dry sulfur deposition (as S). A map of dry sulfur deposition fluxes for 2008 is provided in Figure 3-1. The magnitude of a deposition rate is illustrated by the size of the circle. The map depicts three CASTNET sites with estimated fluxes greater than 5.0 kilograms per hectare per year (kg/ha/yr). These sites are located in Ohio at Quaker City (QAK172) and in Pennsylvania at Pennsylvania State University (PSUI06) and Arendtsville (ARE128). The highest deposition rate was estimated for QAK172, OH with a flux of 8.9 kg/ha/yr. The highest dry sulfur deposition rates coincided with the major SO_2 source region (Figure 1-6) and declined sharply with distance. The dry deposition rates for the western sites were all less than 1.0 kg/ha/yr with the majority of sites less than 0.5 kg/ha/yr.

Figure 3-1 Dry Sulfur ($\text{SO}_2 + \text{SO}_4^{2-}$) Deposition (kg/ha/yr) for 2008



Wet deposition values used to estimate total deposition were based on a combination of historical CASTNET wet deposition data and NADP/NTN wet deposition data. For CASTNET sites where wet concentrations were measured prior to January 1999, those values were used in the data set. For sites where no wet concentrations were measured and for all sites after January 1999, values were obtained from a grid of concentration estimates derived from available NADP/NTN sites by using an inverse distance weighting function. Estimated concentrations were multiplied by the precipitation measured at the CASTNET sites to obtain estimates of wet deposition.

Figure 3-2 provides a map of estimates of total sulfur deposition. The map was constructed by adding dry and wet deposition. The diameters of the circles in the figure illustrate the magnitude of total sulfur deposition and also the relative contributions from wet and dry deposition. The dark shading (blue) signifies the percent wet deposition, and the light shading (tan) shows the percent dry deposition. Seven CASTNET sites in Indiana, Ohio, and Pennsylvania had total (dry + wet) sulfur deposition fluxes greater than 10.0 kg/ha/yr. The highest total sulfur deposition rate was estimated for QAK172, OH with a value of 17.2 kg/ha/yr. Total sulfur deposition at western sites was less than or equal to 2.0 kg/ha/yr. Centennial, WY (CNT169) had the highest estimated flux for the western sites with a value of 2.0 kg/ha/yr. The contribution of dry deposition was much more significant in and near major source regions. For example, the contribution of dry sulfur deposition ranged from about half of total sulfur deposition at Horton Station, VA (VPI120) to less than 20 percent at sites in New England.

Figure 3-3 presents box plots that show the trend in dry sulfur deposition (as S) for the eastern (right side) and western (left side) reference sites. Figure 3-4 shows the trend in annual total (dry + wet) sulfur deposition (as S). The box plots were based on data obtained from the 34 CASTNET eastern reference sites (Figure 1-2) for the period 1990 through 2008 and from the 17 western reference sites (Figure 1-3) for 1996 through 2008. The y-axes on the two figures have different scales. Aggregated sulfur deposition declined considerably at the eastern and western reference sites over their respective periods. Figure 3-5 presents estimates of trends in dry, wet, and total deposition of sulfur (as S) on the same diagram for the eastern reference sites only. The trend line for precipitation-weighted mean sulfur concentrations in precipitation shows a decline in 2008 after small increases during the previous three years.

Figure 3-2 Total (Dry + Wet) Sulfur Deposition (kg/ha/yr) for 2008

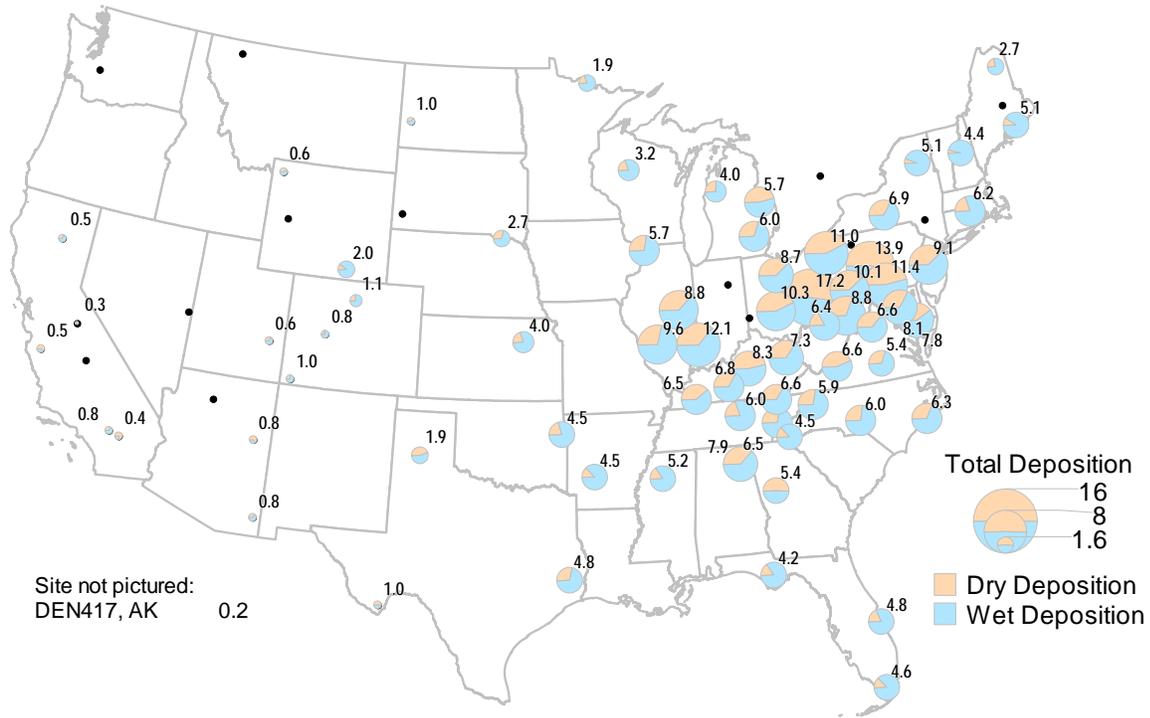


Figure 3-3 Trend in Dry Sulfur Deposition (kg/ha/yr)

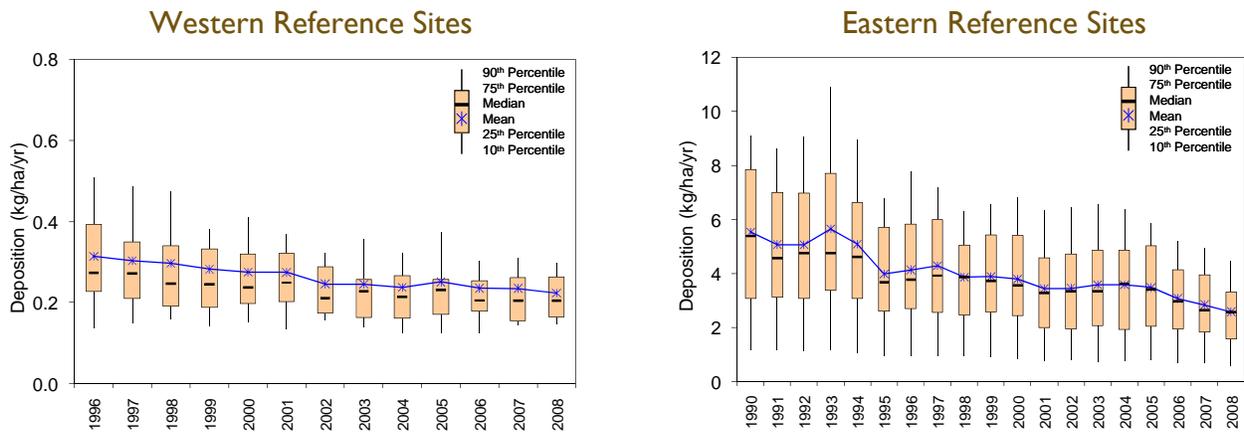
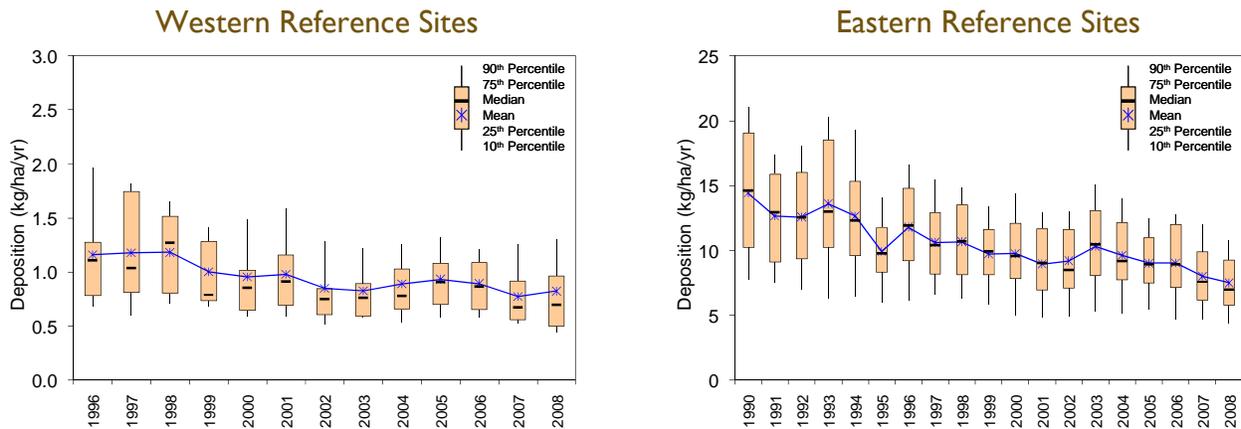
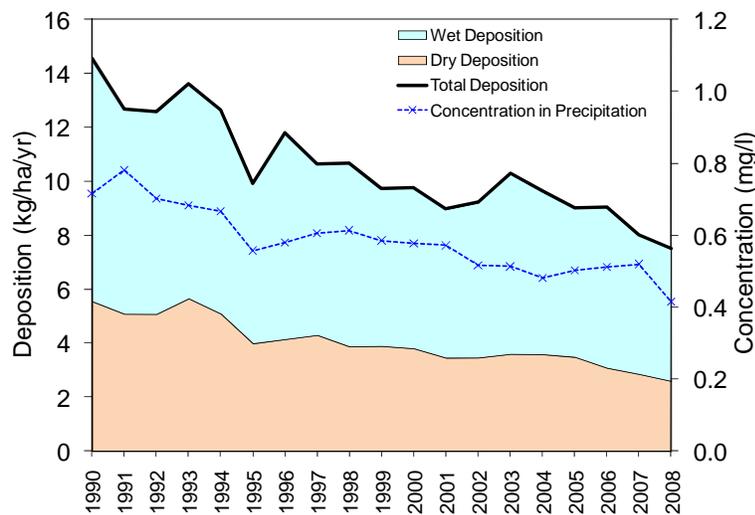


Figure 3-4 Trend in Total Sulfur Deposition (kg/ha/yr)



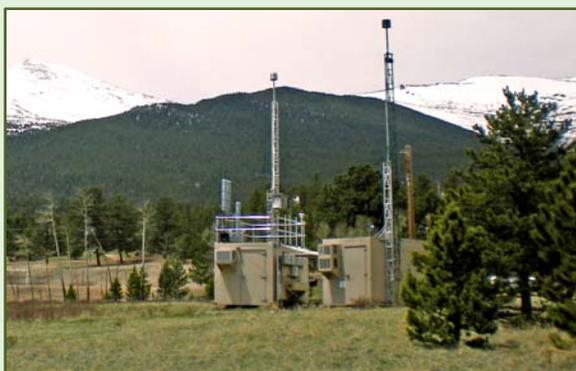
The influence of precipitation on total sulfur deposition is illustrated by comparing the solid (top) line to the dotted blue line in Figure 3-5. The solid line shows total deposition, which depends on sulfur concentrations in precipitation and precipitation amounts. The dotted line shows concentrations in precipitation, which are more representative of changes in SO₂ emissions. The relatively wet year 2003 experienced high total sulfur deposition even though the sulfur concentration in precipitation was relatively low. Overall, total sulfur deposition declined at the eastern reference sites from a 1990–1992 mean of 13.2 kg/ha/yr to a 2006–2008 mean of 8.2 kg/ha/yr, a 38 percent reduction. Total sulfur deposition declined at the western reference sites from a 1996–1998 mean of 1.17 kg/ha/yr to a 2006–2008 mean of 0.83 kg/ha/yr, a 29 percent reduction.

Figure 3-5 Trend in Sulfur Deposition (kg/ha/yr) with Concentrations in Precipitation (mg/l): Eastern Reference Sites



Using CASTNET Data to Assess Quantity of Atmospheric Deposition Compared with Critical Loads

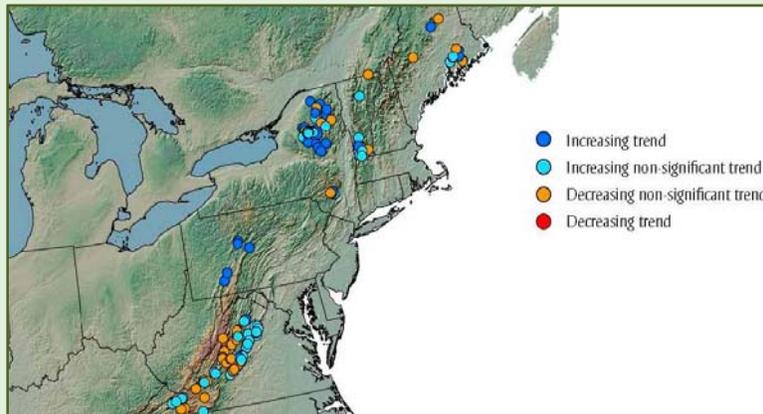
A critical load is a measure of the exposure to one or more pollutants found in an ecosystem, below which significant harmful effects on sensitive members of the ecosystem do not occur (Nilsson and Grennfelt, 1988). Measures of critical load have been most often applied to atmospheric deposition of sulfur and nitrogen pollutants. The critical load of pollutants provides a measure to gauge the extent to which the ecosystem has recovered from past deposition or is potentially at risk due to current or future deposition loads. For this reason, the critical load approach has become an important assessment tool for examining the environmental benefits that result from emission reduction programs. For example, NPS has specified a goal for a critical load of 1.5 kg/ha/yr of wet nitrogen deposition (Baron, 2006) for Rocky Mountain National Park. In other words, annual wet deposition fluxes less than the specified goal will not have harmful effects on the high-elevation ecosystems in the park.



Rocky Mountain National Park, CO (ROM406/206)

Two EPA-administered monitoring networks, the Temporally Integrated Monitoring of Ecosystems (TIME) and the Long-Term Monitoring (LTM) programs, provide surface water chemistry measurements in four regions known to be sensitive to acid precipitation (EPA, 2009b): New England, the Adirondack Mountains, the Appalachian Plateau, and the Blue Ridge Mountains (Figure 3a). Sulfate and NO_3^- ion concentrations, base cation (Ca^{2+} , Mg^{2+} , K^+ , Na^+) concentrations, acid neutralizing capacity (ANC), and other parameters are monitored in the TIME/LTM networks in order to track changes in surface water chemistry and the level of acidification produced in response to changes in acidic deposition. Surface water chemistry data from TIME/LTM and other programs and deposition loadings from NADP and CASTNET can be used to determine whether a critical load is exceeded, i.e., whether the amount of deposition is greater than the critical level needed to reach and/or maintain a healthy ecosystem.

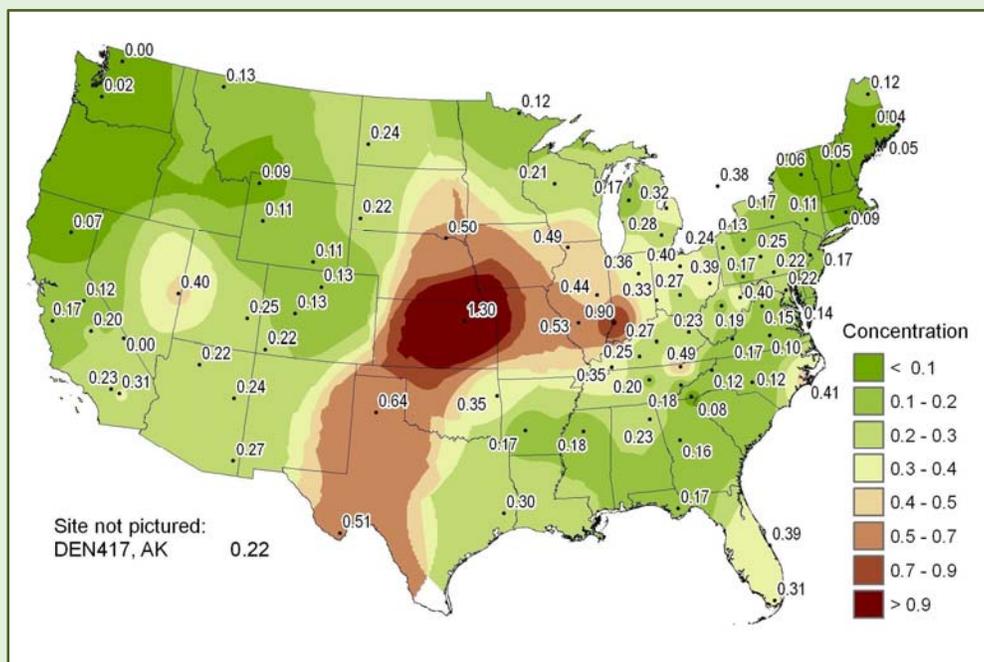
Figure 3a Trends in Lake and Stream ANC Concentrations at LTM Sites, 1990-2006



Source: EPA

CASTNET provides weekly concentration data for acidic pollutants and base cations, which could be utilized to estimate dry deposition fluxes and gauge exceedances of critical loads. Figure 3b shows 2008 annual mean Ca^{2+} concentrations. These data and other CASTNET ion concentration measurements can be used to model dry deposition loading of base cations to an ecosystem. This information is typically missing from critical load and biogeochemical hydrological models. The benefits of using CASTNET data for input to these models are that CASTNET (1) provides long-term trends of pollutants measured near sensitive ecosystems and (2) provides a more complete estimate of the ions that contribute to depositional loading to an ecosystem.

Figure 3b CASTNET 2008 Annual Mean Ca^{2+} Concentrations ($\mu\text{g}/\text{m}^3$)



Nitrogen Deposition

Dry fluxes of nitrogen (as N) for 2008 are shown in Figure 3-6. These fluxes are the sum of fluxes of $\text{HNO}_3 + \text{NO}_3^- + \text{NH}_4^+$, based on the individual MLM simulations for the three species. A majority of CASTNET sites in the eastern United States had estimated dry nitrogen deposition rates greater than 1.0 kg/ha/yr. Deposition rates greater than 2.0 kg/ha/yr were calculated for sites in Ohio, Pennsylvania, New Jersey, Kentucky, and at Great Smoky Mountains National Park (GRS420) in Tennessee. Two monitoring sites in California had fluxes greater than or equal to 2.0 kg/ha/yr. Locations on the map with no value had insufficient data to calculate fluxes.

Figure 3-6 Dry Nitrogen ($\text{HNO}_3 + \text{NO}_3^- + \text{NH}_4^+$) Deposition (kg/ha/yr) for 2008

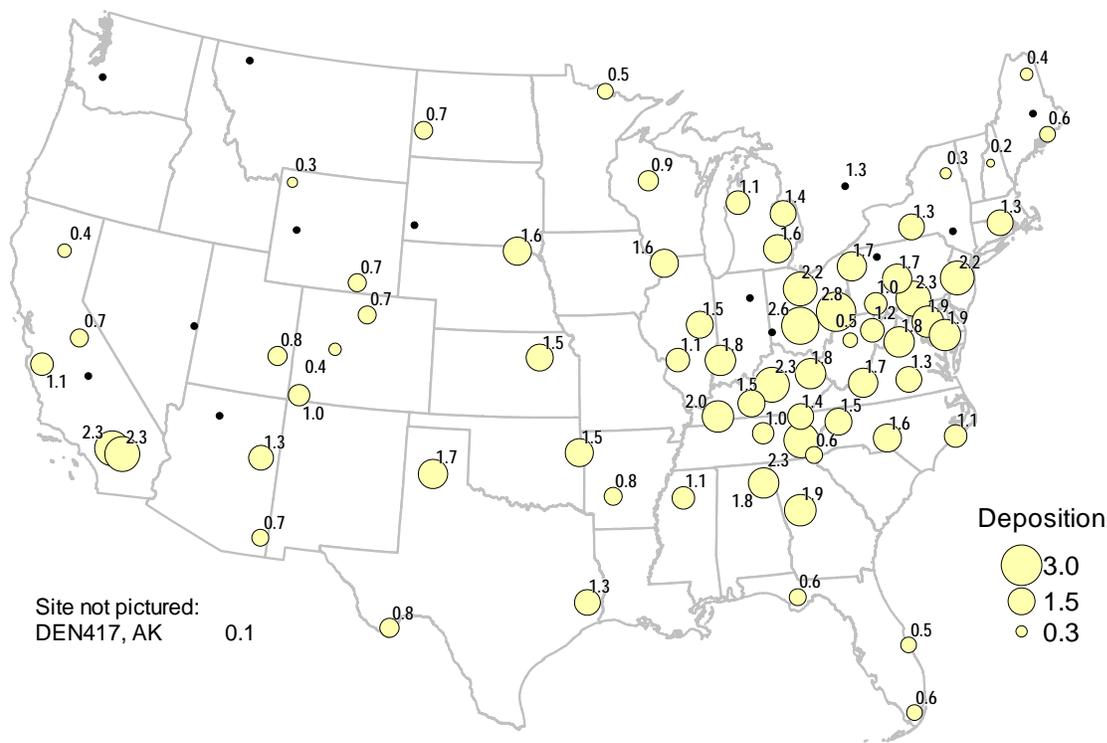


Figure 3-7 presents a map of total nitrogen deposition (as N) for 2008. The map was constructed by summing the estimates of dry (light shading) and wet (dark shading) deposition. The figure shows that a majority of the eastern sites recorded deposition rates greater than 5.0 kg/ha/yr. No values above 10.0 kg/ha/yr were estimated for 2008. The highest total nitrogen flux (9.6 kg/ha/yr) was estimated for Vincennes, IN (VIN140). The values at the western sites ranged from 1.0 kg/ha/yr at Yosemite National Park, CA (YOS404) to 3.8 kg/ha/yr in southern California at Converse Station (CON186). The contributions of dry nitrogen deposition to total nitrogen were lower than the corresponding contributions of dry sulfur deposition. Dry nitrogen deposition contributed less than 50 percent of total deposition in the East. On the

other hand, dry nitrogen deposition contributed more than half of total nitrogen deposition at four of five monitoring sites in California, a region with elevated concentrations of nitrogen species and limited rainfall.

Figure 3-7 Total (Dry + Wet) Nitrogen Deposition (kg/ha/yr) for 2008

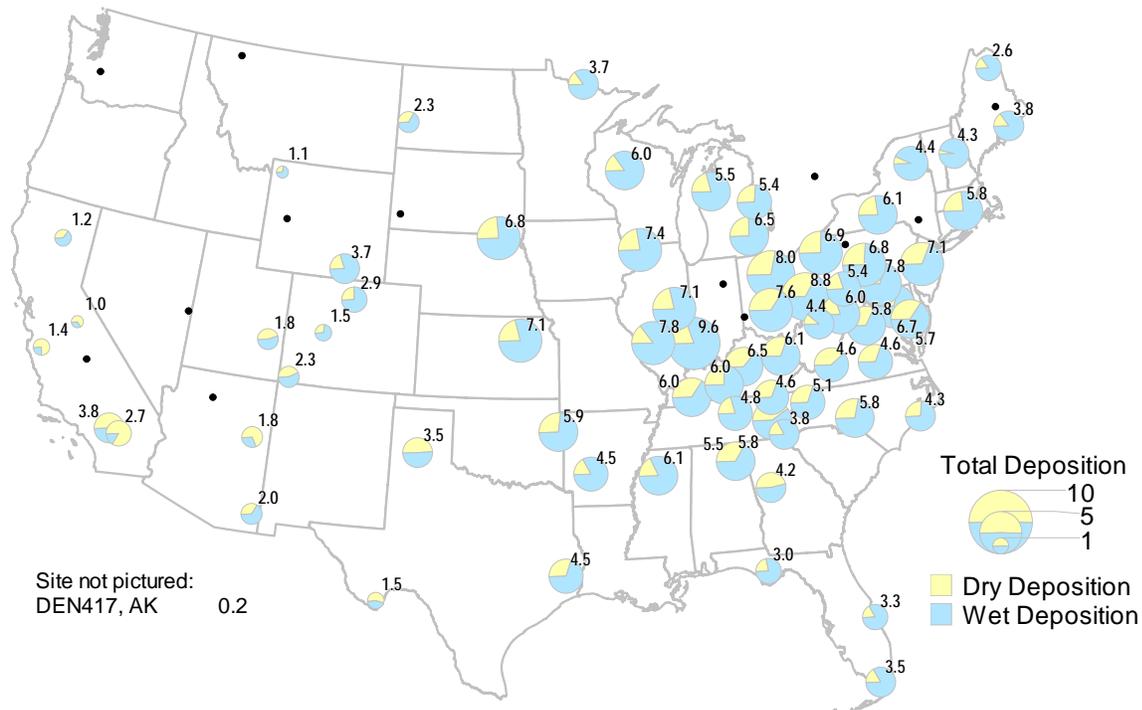


Figure 3-8 presents box plots that were constructed using data from the 34 eastern reference sites over the period 1990 through 2008 (right side) and 17 western reference sites for 1996 through 2008 (left side). The box plots show the trends in dry nitrogen deposition (as N). The box plots for the eastern sites in Figure 3-8 show a reduction in dry nitrogen deposition beginning in 1999. Three-year mean fluxes declined from 2.2 kg/ha/yr in 1990–1992 to 1.6 kg/ha/yr in 2006–2008, a 27 percent reduction over the 19 years. The box plots for the western sites show a smaller 15 percent decline, a change from 0.90 kg/ha/yr in 1996–1998 to a mean of 0.76 kg/ha/yr in 2006–2008.

Figure 3-8 Trend in Dry Nitrogen Deposition (kg/ha/yr)

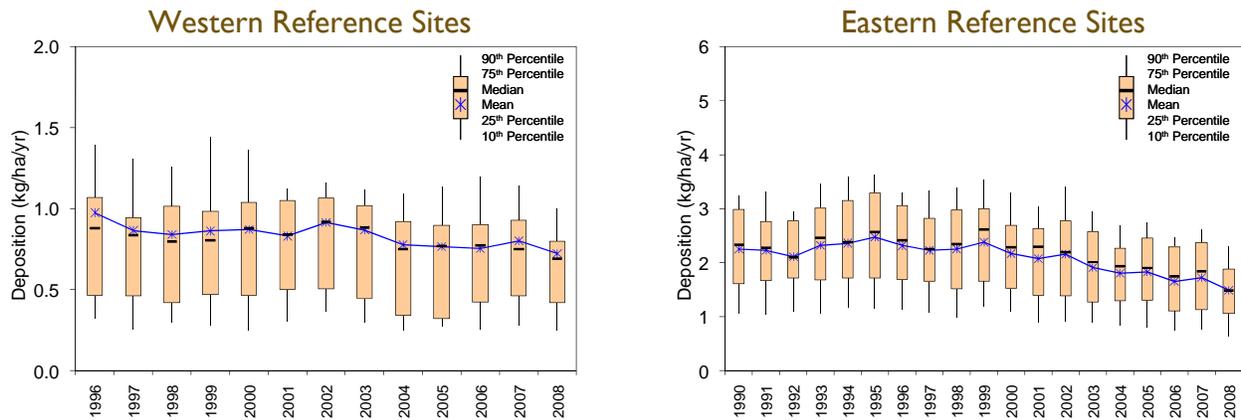


Figure 3-9 shows the trends in annual total (dry + wet) nitrogen deposition for the eastern (right side) and western (left side) reference sites. Total nitrogen deposition aggregated over the eastern sites is more variable because the annual wet and total fluxes depend on the amount of precipitation. The figure suggests that total flux has decreased since 1996 with a substantial decline over the last five years. Total nitrogen flux for the western sites shows a relatively flat distribution. Estimates of trends in wet, dry, and total deposition of atmospheric nitrogen (as N) are presented in Figure 3-10 for the eastern reference sites only. The trend line (dotted) for precipitation-weighted mean nitrogen concentrations in precipitation shows a slow downward trend since 1998 with a drop in 2008. This trend line is reasonably representative of the effect of changes in NO_x emissions because it does not depend on precipitation amounts. Total nitrogen deposition declined 19 percent in the East over the 19 years.

Figure 3-9 Trend in Total Nitrogen Deposition (kg/ha/yr)

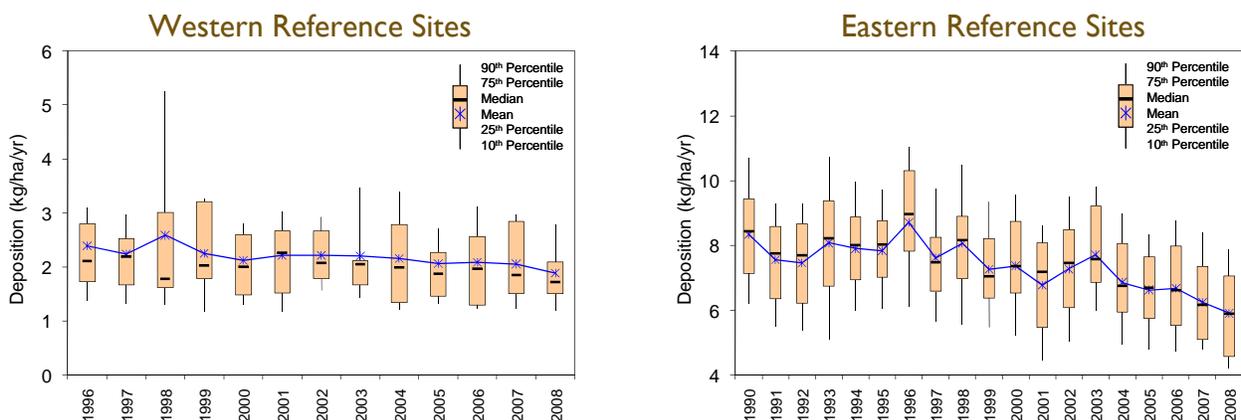
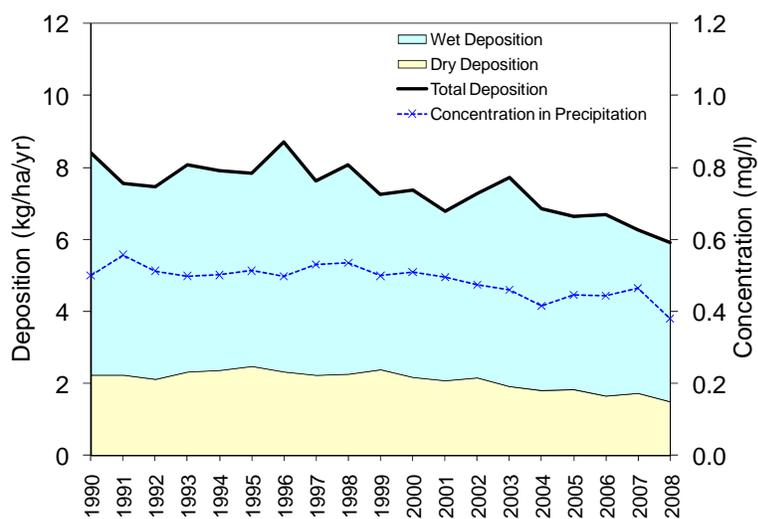


Figure 3-10 Trend in Nitrogen Deposition (kg/ha/yr) with Concentrations in Precipitation (mg/l): Eastern Reference Sites



Uncertainties in NO_3^- and HNO_3 Concentrations and Their Effect on Dry Nitrogen Deposition

The uncertainties of NO_3^- and HNO_3 concentrations were estimated using results of the Maryland Aerosol Research and Characterization (MARCH)/CASTNET data intercomparison (Lavery *et al.*, 2009). MARCH data were collected at Fort Meade, MD, and the CASTNET measurements were taken at Beltsville, MD (BELI 16). The CASTNET filter pack technology causes uncertainty in the measured nitrogen species. The effects of these uncertainties on weekly and annual dry deposition fluxes of NO_3^- , HNO_3 , and total nitrogen deposition ($\text{NO}_3^- + \text{HNO}_3 + \text{NH}_4^+$, as N) were analyzed by adjusting the HNO_3 and NO_3^- concentration values for BELI 16, MD based on the MARCH/CASTNET intercomparison results and then recalculating the total nitrogen flux.

NO_3^- concentrations were decreased by a factor of 3.5 to characterize the fact that filter-sampled NO_3^- was comprised primarily of larger particles. HNO_3 concentrations were increased by a factor of 1.15 based on the MARCH/CASTNET intercomparison results. Total nitrogen deposition values were then remodeled using the MLM based on measurements collected at BELI 16, MD for the years 1998 through 2007.

As Table 3a shows, differences from the recalculated concentrations are relatively small (approximately 10 percent or less) reflecting the fact that the HNO_3 component dominates the total nitrogen dry deposition. Dry deposition of particulate NO_3^- has a

relatively small contribution to the total nitrogen flux, and thus, the large decrease in NO_3^- concentrations does not have a significant effect on total deposition.

Table 3a Changes to Dry Deposition of Total Measured Nitrogen after Adjustments to NO_3^- and HNO_3 Fluxes

Site ID	Year	Total Measured N	NO_3^- Adjustment Only	HNO_3 Adjustment Only	Both Adjustments
BEL116	1998	2.62	2.58	2.93	2.89
BEL116	1999	3.00	2.94	3.35	3.29
BEL116	2000	1.85	1.77	2.05	1.97
BEL116	2001	2.41	2.36	2.68	2.64
BEL116	2002	2.57	2.53	2.87	2.84
BEL116	2003	2.23	2.18	2.49	2.44
BEL116	2004	2.56	2.50	2.85	2.79
BEL116	2005	2.25	2.18	2.51	2.44
BEL116	2006	2.63	2.59	2.94	2.90
BEL116	2007	2.40	2.38	2.69	2.66



Beltsville, MD (BEL116)

Chapter 4: Ozone Concentrations

CASTNET provides the primary platform for monitoring rural, ground-level ozone (O_3) concentrations in the United States and for providing information on geographic patterns in rural O_3 levels. CASTNET was not designed to operate as a network for demonstration of compliance with O_3 NAAQS. However, the network provides data on the extent to which rural areas potentially exceed concentration values mandated by NAAQS. Ozone measurements collected during 2006–2008 were used to evaluate both the 1997 (EPA, 1997) and 2008 (EPA, 2008a) O_3 standards. To attain the 1997 standard, the 3-year average of the fourth-highest daily maximum 8-hour average O_3 concentrations measured at each monitor within a specified area over each year must not exceed 0.08 parts per million (ppm) or be greater than or equal to 85 ppb in practice. The 2008 standard is achieved when the 3-year average of the fourth-highest daily maximum 8-hour average O_3 concentrations measured at each monitor within a specified area over each year do not exceed 0.075 ppm (75 ppb in practice). The year 2008 is the first year when no eastern CASTNET sites measured 8-hour average O_3 concentrations greater than or equal to 85 ppb.

Almost all CASTNET sites operate an O_3 analyzer that measures hourly concentrations. While CASTNET is not a compliance network, the data collected provide useful information on geographic patterns in regional O_3 and the extent to which rural areas potentially exceed concentration values mandated by the NAAQS. The 8-hour O_3 standard is a useful measure for assessing the status and trends in rural O_3 levels in order to gauge the success of EPA emission reduction programs such as the NO_x SIP Call/NBP and CAIR. The analyses presented in this section provide maps and examine trends in the fourth highest daily maximum 8-hour average O_3 concentrations measured annually.

Ground-level O_3 is an air pollutant that can cause harmful effects to the human respiratory system and damage to vegetation and ecosystems. Ground-level O_3 is formed in the lower atmosphere when volatile organic compounds (VOCs) and NO_x react in the presence of sunlight. Emissions from motor vehicle exhaust, industrial facilities, chemical solvents, gasoline vapors, and power plants are the major anthropogenic sources of NO_x and VOCs. Also, biogenic VOC emissions from trees and other vegetation contribute to O_3 formation in rural areas. VOC emission reduction strategies have been successful in reducing higher, short-term O_3 concentrations in and downwind of urban areas. However, as discussed in this chapter, implementation of the NO_x emission reductions that were mandated by the ARP and other NO_x emission control programs contributed to reductions in O_3 concentrations, especially in

rural areas with elevated 8-hour average levels, because regional transport of pollutants contributes to ozone formation.

Meteorological conditions play a significant role in O₃ formation. Dry, hot, and sunny days are most conducive to O₃ production. Ozone concentrations typically increase during daylight and peak in the late afternoon after peak temperature and sunlight intensity. Concentrations then drop in the evening. The warm months in spring, summer, and fall typically define the O₃ season. EPA has defined a specific ozone season for each state and has proposed recently that the ozone season be designated as year round in most locations.

On May 1, 2009, the CAIR NO_x ozone season trading program began, replacing the NBP in states covered under the CAIR NO_x ozone season program, requiring further NO_x emission reductions from the power sector.

National Ambient Air Quality Standards for Ozone				
	Primary Standards		Secondary Standards	
	Level	Averaging Time	Level	Averaging Time
Ozone	0.075 ppm	8-hour ¹	Same as Primary	
	0.08 ppm	8-hour ²		

Notes:

- 1 (a) On September 5, 2009, EPA announced that it is reconsidering the current levels of the ozone primary and secondary standards.
- (b) To attain this standard, the 3-year average of the fourth-highest daily maximum 8-hour average O₃ concentrations measured at each monitor within a specified area over each year must not exceed 0.075 ppm (effective May 27, 2008).
- 2 (a) To attain this standard, the 3-year average of the fourth-highest daily maximum 8-hour average O₃ concentrations measured at each monitor within an area over each year must not exceed 0.08 ppm (effective 1997).
- (b) The 1997 standard—and the implementation rules for that standard—will remain in place for implementation purposes as EPA undertakes rulemaking to address the transition from the 1997 O₃ standard to the 2008 O₃ standard.

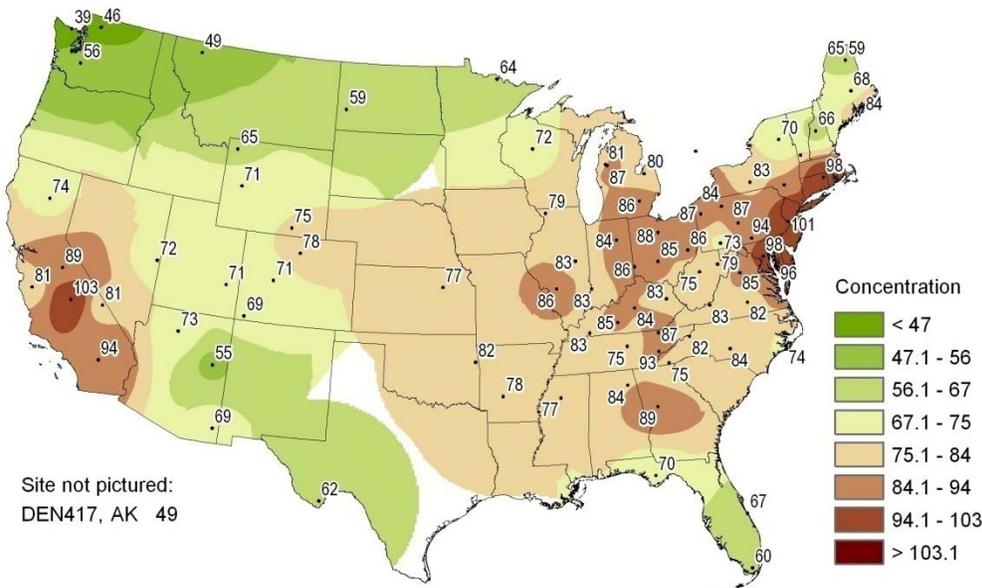
The concentration shading for the figures used in this chapter was prepared using the scales shown on each map. Additional maps of ozone concentrations can be viewed on the Web site for the NPS Air Atlas (<http://science.nature.nps.gov/AirAtlas/AirAtlas0105/viewer.htm>). The Air Atlas incorporates all ozone monitoring reporting to AQS plus the CASTNET ozone measurements.

Eight-Hour Ozone Concentrations

Figure 4-1 presents 3-year averages of the fourth highest daily maximum 8-hour average O₃ concentrations for 2006–2008. During this period, 3-year averages of the fourth highest daily

from northern Virginia to Connecticut, extending almost to Maine, and in the central states of Pennsylvania, Ohio, Michigan, Indiana, and Illinois. Exceedances were also recorded in Kentucky, Tennessee, and greater Atlanta. Using the 75 ppb standard, the map in Figure 4-2 shows that most of the eastern sites measured O₃ concentrations greater than 75 ppb, as well as five monitors in California and the site at Rocky Mountain National Park (ROM406). The period 2006–2008 represents a significant improvement in air quality and is one of the best periods of air quality since the inception of CASTNET in terms of having the fewest sites with exceedances of the 8-hour O₃ NAAQS. However, sites in California, Maryland, and at GRS420, TN continued to measure elevated ozone levels.

Figure 4-2 Three-Year Average of Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) for 2000–2002



Measurements of 8-hour average O₃ concentrations during 2008 (Figure 4-3) were lower than 2007 and considerably lower than concentrations measured over the period 2000–2002. Five sites in California measured O₃ concentrations greater than or equal to 85 ppb. The five California sites include Yosemite National Park (YOS404), Pinnacles National Monument (PIN414), Sequoia National Park (SEK430), Converse Station (CON186), and Joshua Tree National Park (JOT403). The monitor at SEK430, CA measured the highest value (112 ppb) in the network during 2008. The year 2008 is the first year when no eastern CASTNET sites measured 8-hour average O₃ concentrations greater than or equal to 85 ppb.

Figure 4-3 also depicts the geographic distribution of fourth highest daily maximum 8-hour average O₃ concentrations greater than 75 ppb. Eleven eastern sites in eight states and six California monitors measured ozone concentrations above 75 ppb. The monitor at

ROM406, CO measured a concentration of 76 ppb. The eastern states include Pennsylvania, New Jersey, Maryland, Virginia, North Carolina, Tennessee, Georgia, and Texas. In addition to the five California sites already mentioned, Lassen Volcanic National Park (LAV410) measured a fourth highest daily maximum 8-hour average O₃ concentration of 83 ppb.

Figure 4-3 Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) for 2008

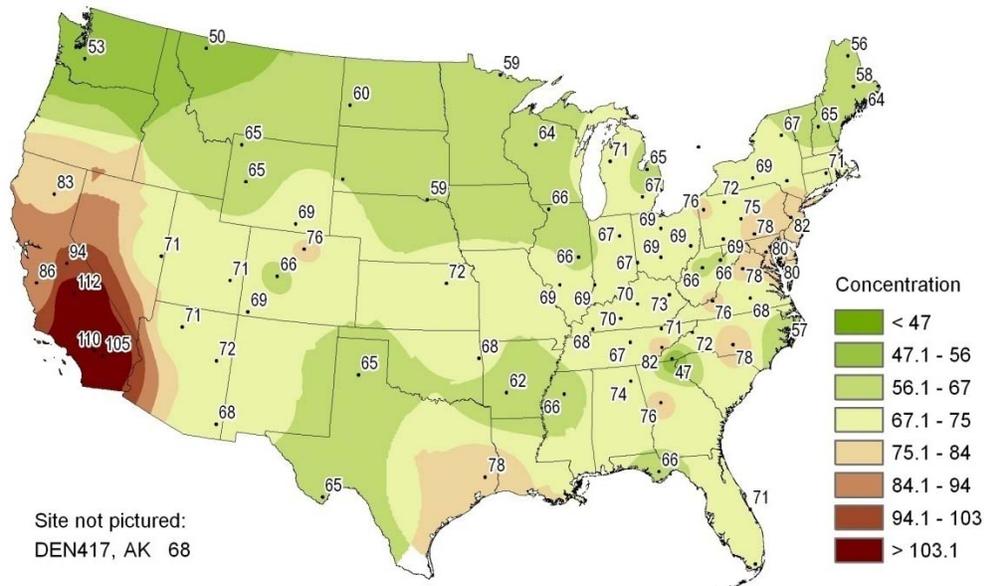
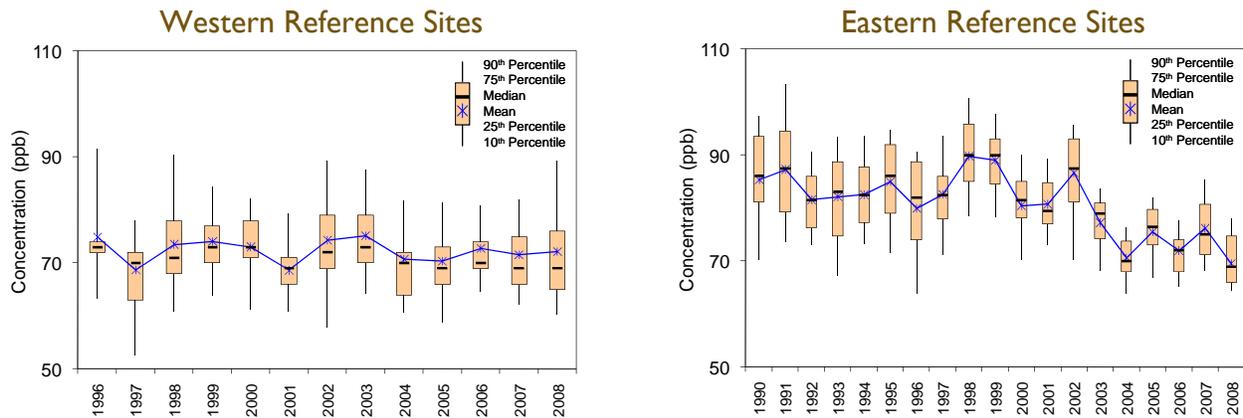


Figure 4-4 provides box plots depicting trends in fourth highest daily maximum 8-hour average O₃ concentrations from the 34 CASTNET eastern reference sites (right side) and 17 western reference sites (left side). The eastern ozone data show a decline since 2002. The mean aggregated value (69 ppb) for 2008 was the lowest in the history of the network. It represents an improvement over the mean value of 90 ppb measured in 1998. The box plots constructed from the aggregated western O₃ concentrations show no trend. The 2006–2008 average of the fourth highest daily maximum 8-hour average O₃ concentrations for the western reference sites was 72 ppb.

Figure 4-4 Trend in Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb)

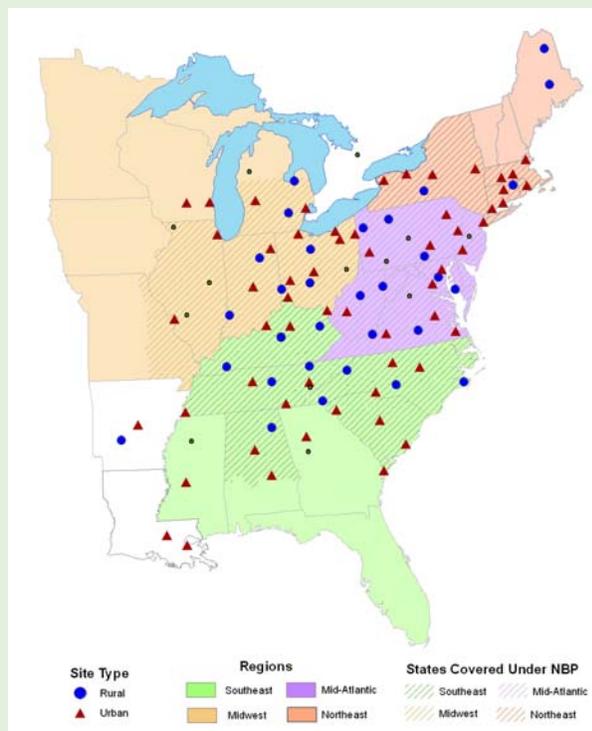


Impacts of NO_x Budget Program and Clean Air Interstate Rule on Rural and Urban Ozone Levels in the Eastern United States

Researchers at Cornell University and the Cary Institute of Ecosystem Studies used random coefficient models to assess the impact of reduced NO_x emissions during the O₃ season on daily maximum 8-hour average O₃ concentrations for various regions of the eastern United States for the period 1997 through 2007. This assessment extended previous analyses by Butler *et al.* (2003, 2005) that investigated the impact of reduced NO_x emissions on NO₃ concentrations in precipitation and ambient concentrations of HNO₃.

Random coefficient models were used because O₃ concentration values are not independent but clustered for each site. In addition, the models can examine monitoring sites on a regional basis (e.g., the Northeast) or in other combinations (e.g., urban versus rural sites). In this

Figure 4a Locations of Ozone Monitoring Sites



Note: Four sites in eastern Texas were included in the analysis for Eastern United States but are not shown on map.

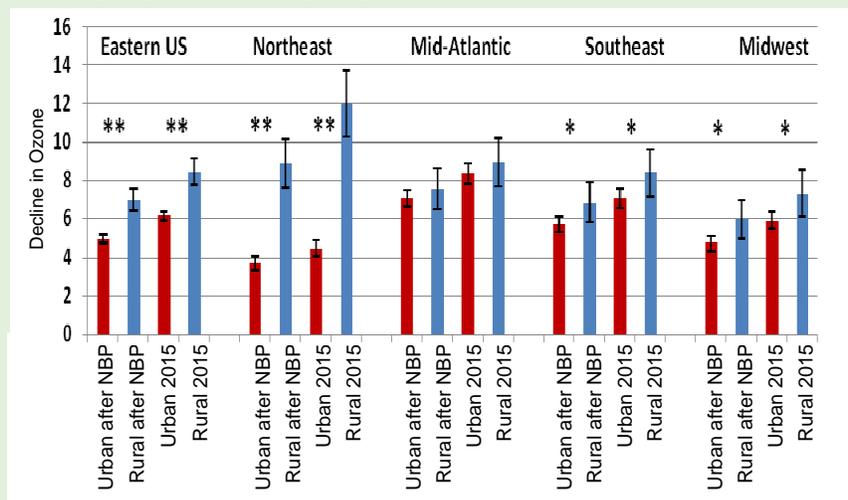
Source: T. J. Butler

analysis, O₃ data from 95 CASTNET and AQS sites were evaluated for the 11-year time period for (a) the Eastern United States as a whole, (b) by region (Northeast, Mid-Atlantic, Southeast, and Midwest), and (c) by rural CASTNET sites (32 sites) versus urban AQS sites (63 sites). Figure 4a shows the locations of the sites and the defined regions used in this analysis, as well as the NBP regulated states/areas.

Source regions for NO_x emissions were derived from multiyear 24-hour air mass back trajectories at 500 meters above ground level (AGL) and 1,000 meters AGL. The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT)-4 model was applied to generate the trajectories. State-level emissions for stationary sources, which were derived from the EPA ARP emissions database, provided the most complete record of O₃ season emissions for the period 1997 through 2007 and are considered the best estimate of NO_x emission changes from stationary sources before and after NBP implementation. CAIR, as promulgated, was designed to further reduce power plant pollutant emissions, including a 2 million ton (61 percent) reduction in NO_x emissions, from 2003 levels. The models were used to estimate the impact of CAIR in terms of reducing O₃ concentrations in the eastern United States in 2015.

Ozone season O₃ concentrations were meteorologically-adjusted to account for year-to-year variations in humidity and temperature, which influence the production of O₃. Statistical model results were generated by dividing each region into rural and urban sites. The models were used to evaluate how reduced NO_x emissions would impact the sites. Figure 4b illustrates the declines in daily maximum 8-hour average O₃ concentrations for the different regions and for rural and urban sites for pre-NBP (1997–2000) and post-NBP (2004–2007) implementation, as well as for projected 2015 NO_x emissions.

Figure 4b Declines (ppb) in Daily Maximum 8-Hour Average O₃ Concentrations for Urban (red) and Rural (blue) Sites



Source: T. J. Butler

The double asterisk (**) indicates the urban and rural site differences for a region are statistically significant at $p < 0.05$. Similarly, a single asterisk (*) indicates statistically significant differences between rural and urban sites in a region at $p < 0.10$. Error bars are based on the standard error of the slope of the model regression lines. For the eastern United States as a whole, O_3 data from rural and urban sites are statistically different ($p < 0.05$). Greater declines in ozone concentrations occurred at the rural sites, which show a 7 ppb (12 percent) decline, while urban sites show a 5 ppb (8 percent) decline.

The Northeast region also shows statistically different ($p < 0.05$) daily maximum 8-hour average O_3 concentrations between rural and urban sites and the greatest difference between declines in O_3 levels at these sites. Urban sites show a post-NBP decline of 4 ppb (9 percent) while the rural sites show a decline of 9 ppb (18 percent). Modeled differences between rural and urban sites for the Southeast and the Midwest regions were less statistically significant ($p < 0.10$), with urban declines of 9 percent and 8 percent for the two regions and rural declines of 12 percent and 11 percent. The difference between urban and rural sites in the Mid-Atlantic region was not statistically significant.

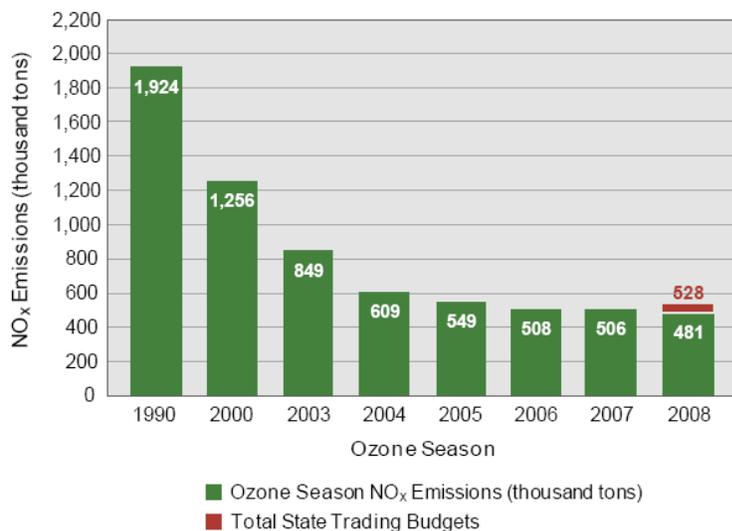
Rural monitoring sites are typically affected little by local emissions. They are likely more affected by transported emissions such as those from large stationary sources (EGUs). The greater declines at rural sites, except for the Mid-Atlantic region, may be explained by the fact that stationary source emissions have been controlled to a larger extent than comparable urban and transportation NO_x sources. Urban sites are more affected by a variety of local sources such as vehicular NO_x emissions, which are more difficult to regulate and control. The Northeast had the greatest decline in rural O_3 concentrations. The Northeast rural monitoring sites are “downwind” of a large number of stationary sources that are now being regulated. Further ozone reductions at urban locations may require a greater emphasis on controlling local NO_x and VOC emissions.

Nitrogen Oxides and Ozone

In May 2009, EPA published *The NO_x Budget Trading Program: 2008 Emission, Compliance, and Market Analyses*. The report analyzed changes in NO_x emissions from 1990 through 2008. Figure 4-5, which was taken from the EPA report, depicts O_3 season NO_x emissions from all NBP sources in the eastern United States from 1990 through 2008. The NBP was in effect from 2003 through 2008. The CAIR NO_x ozone season program began on May 1, 2009 and replaced the NBP in affected states. The OTC NO_x Budget was in effect from 1999 through 2002. Other emission control programs such as mobile source controls, VOC Reasonably Available Control Technology (RACT) and Maximum Available Control Technology (MACT), new source review, and the ARP contributed to NO_x and VOC emission reductions.

NBP sources emitted 481,420 tons of NO_x in the 2008 O₃ season (EPA, 2009c). Emissions in 2008 decreased 24,880 tons from 2007 and were 62 percent below 2000 levels and 75 percent below 1990 emissions. NBP regulated 2,568 units in 2008. EGUs constituted 88 percent of the regulated units; industrial sources constituted most of the remaining 12 percent.

Figure 4-5 Ozone Season NO_x Emissions from All NBP Sources



Notes: Data reflect full ozone season emissions in all years for all states. The year 2000 baseline value has been adjusted to correct a misprint in Figure 5 of the 2007 NBP report. The 2008 total state trading budgets include opt-in allowances, where applicable (New York, Ohio, and West Virginia).

Source: EPA (2009c)

Trends in CASTNET measurements of total nitrate concentrations (Figure 4-6) measured during the ozone season (May through September) at the 34 eastern reference sites show almost no change from 1990 through 1999 followed by a decline as the emission reductions from the OTC and NBP became effective. Three-year median concentrations before (2000–2002) and after (2003–2005) the implementation of the NBP were 2.43 µg/m³ and 1.87 µg/m³, respectively. The most recent three-year (2006–2008) median value was 1.67 µg/m³. Figure 4-7, which was taken from the EPA report on NBP compliance (EPA, 2009c), shows the relationship between NO_x emission reductions from power industry sources and changes in O₃ levels at CASTNET and EPA AQS-compliant sites in the eastern United States over the period 2002–2008. The figures show declines in concentrations in all NBP states. The figure also shows a strong association between regions with the greatest emission reductions and downwind monitoring sites showing the greatest improvement in O₃ levels.

Figure 4-6 Trend in Ozone Season (May through September) Mean Total Nitrate ($\text{NO}_3^- + \text{HNO}_3$) Concentrations ($\mu\text{g}/\text{m}^3$) – Eastern Reference Sites

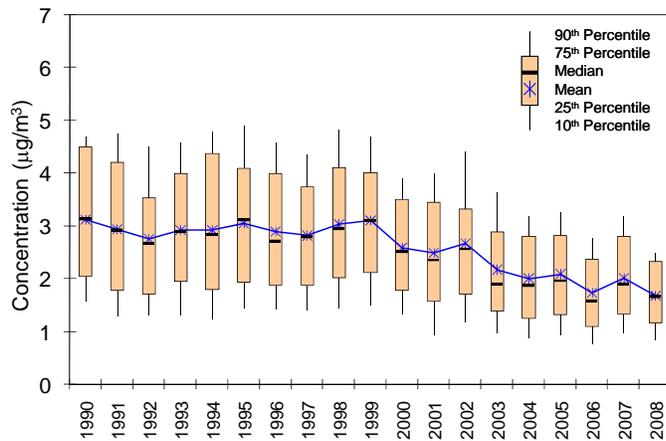
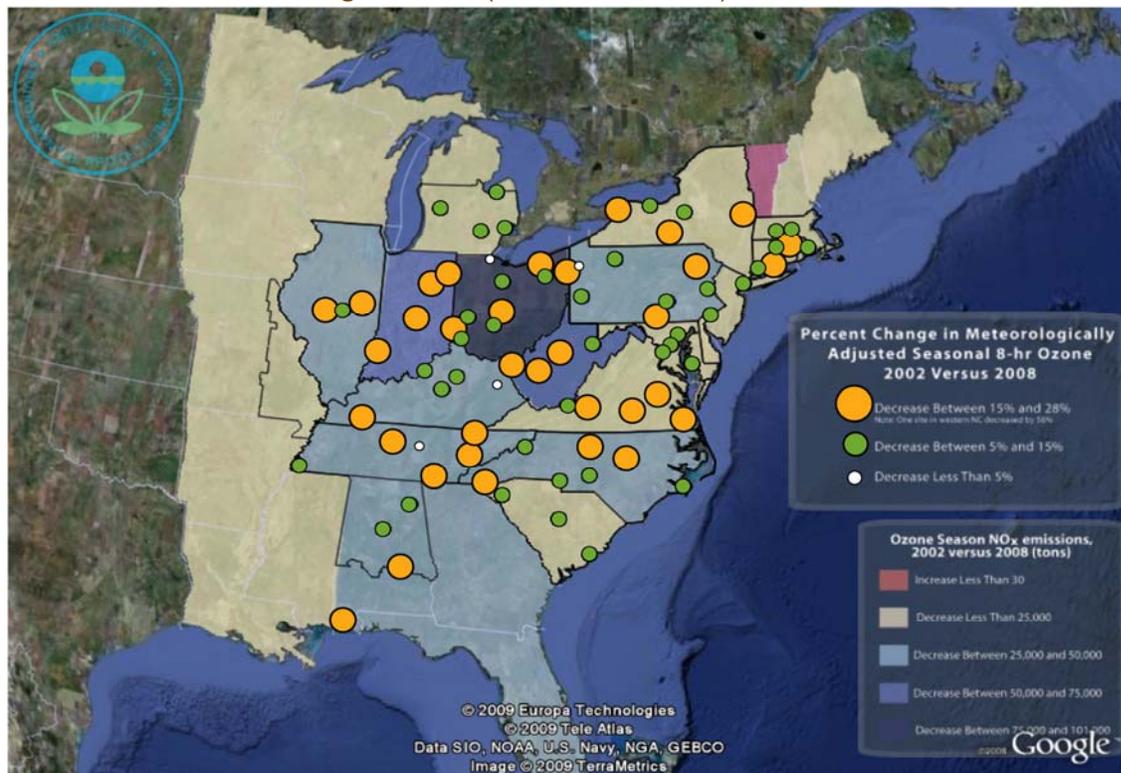


Figure 4-7 Reductions in Ozone Season Power Industry NO_x Emissions and Changes in 8-Hour Average Ozone, (2002 versus 2008)



Notes:

- From 1999 to 2002, states in the Northeast reduced emissions from EGUs and industrial boilers under the OTC NO_x Budget Program. OTC states include Connecticut, Delaware, the District of Columbia, Main, Maryland, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, Vermont, and Virginia.
- Meteorologically-adjusted ozone data are from AQS and CASTNET sites that met completeness criteria.
- Google Earth was used to display the information shown in this figure. To access the data layers shown here as well as other data, including unite-level emissions and controls, visit the Clean Air Market Division's interactive mapping site at www.epa.gov/airmarkt/progress/interactivemapping.html.

Source: EPA (2009c)

Ozone Episode in the Great Smoky Mountains, TN (GRS420) in July 2008

Periods with extended high ozone concentrations have occurred occasionally over the last 10 years at the Look Rock site in Great Smoky Mountains National Park, TN (GRS420). Measured fourth highest daily maximum 8-hour average O_3 concentrations have exceeded 85 ppb causing potential contravention of both the 1997 0.08 ppm and 2008 0.075 ppm NAAQS. A four-day O_3 episode was observed at the park during July 17–20, 2008. Surface weather conditions are illustrated in Figure 4c, which shows a Bermuda High off the East Coast and Tropical Storm Cristobal off the coast of North Carolina. The circulation around the tropical storm was sufficiently developed to produce northeasterly winds along the East Coast from New Jersey to the park. However, the cloud pattern produced by the storm was limited and did not inhibit O_3 formation.

A time series of hourly O_3 concentrations observed at GRS420, TN during July 13–24, 2008 is provided in Figure 4d. Daily maximum 8-hour O_3 average concentrations are also shown. The 8-hour levels are plotted at 0000 local time for each day. Figure 4d shows the gradual buildup of O_3 beginning on July 15th with a subsequent peak hourly value of 90 ppb on the 19th and a peak daily 8-hour value of 87 ppb, shown at 0000 on July 19th. The episode was terminated by a shift to westerly winds on July 20th.



Great Smoky Mountains National Park, TN
Source: NPS

Figure 4c Surface Weather Map for July 20, 2008

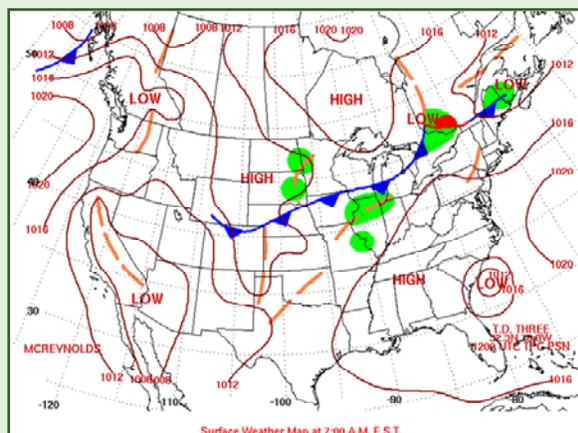


Figure 4d Time Series of Ozone Concentrations

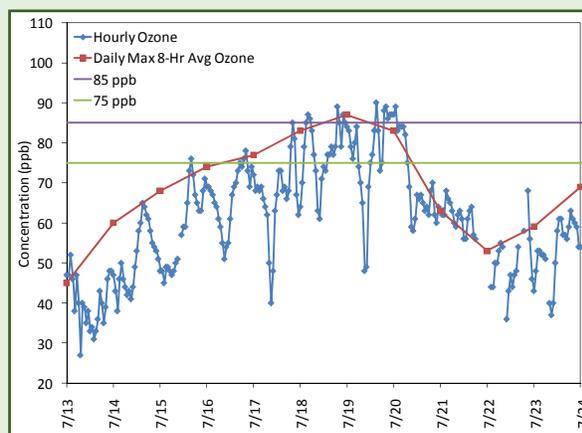


Figure 4d shows a fairly steady increase in peak hourly O₃ values during the ozone buildup. The peak 1-hour concentration during the episode was not much higher than the peak 8-hour value. The figure also shows a diurnal cycle daily with lower O₃ concentrations observed around 0900. Figure 4e shows two plots of composite O₃ concentrations. The top curve was constructed by averaging all hours for each hour of the day for the four-day episode. The bottom curve was created from a composite of the remaining hours in July. The two daytime O₃ peaks in the top curve might have been produced by pollutants from two different source regions or by recirculated pollutants. The minimum concentrations around 0900 to 1000 can be explained by the end of nighttime dry deposition or by fresh NO_x emissions that scavenge O₃ around that time.

Figure 4e Composite Diurnal Ozone Concentrations

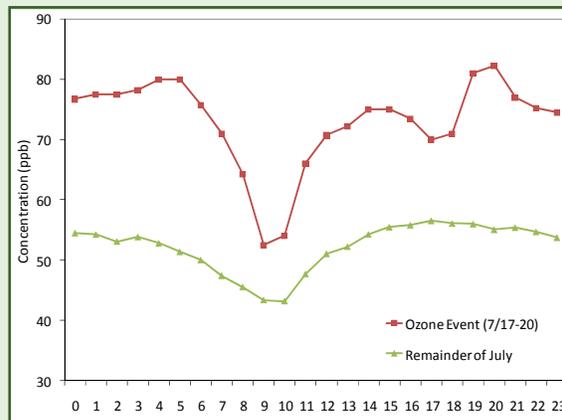


Figure 4f Backward Trajectories Ending at 1000 EDT July 19, 2008

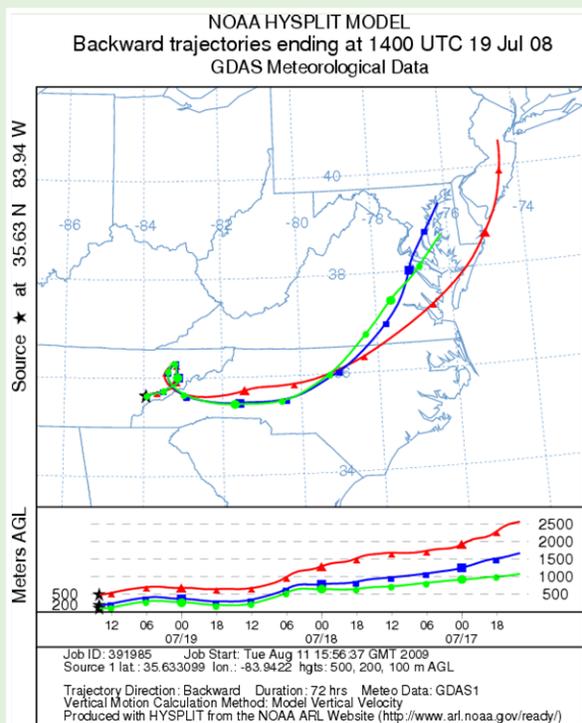
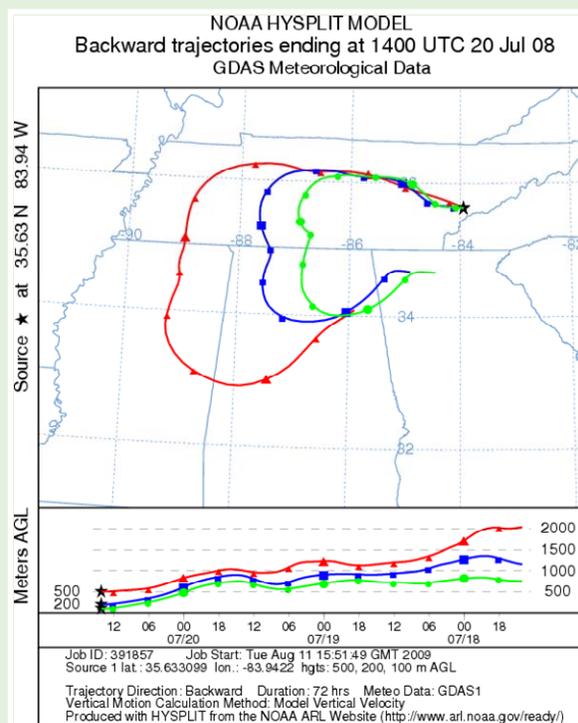


Figure 4g Backward Trajectories Ending at 1000 EDT July 20, 2008



Backward trajectories from the National Oceanic and Atmospheric Administration (NOAA) HYSPLIT Model for July 19, 2008 at 1400 UTC (1000 EDT) and for July 20, 2008 at 1400 UTC are provided in Figures 4f and 4g, respectively. Trajectories ending at GRS420, TN are given for 100 m, 200 m, and 500 m AGL. The three trajectories ending at GRS420, TN at 1000 EDT on July 19th show similar origins. The 100 m and 200 m trajectories originated in Maryland. The 500 m trajectory originated in New Jersey. All three trajectories show significant subsidence and recirculation of air near their endpoint at the park. The 100 m trajectory originated at an altitude of approximately 500 m three days earlier. Hourly ozone concentrations reached 90 ppb during the afternoon of July 19th, and the highest 8-hour average daily maximum level was recorded at 0000 on the 19th. The recording of high concentrations in the afternoon suggests the transport of ozone and its precursors along the trajectory pathways. The episode ended on July 20, 2008 when the winds switched to westerly as shown by the trajectories in Figure 4g.

The occurrence of northeasterly winds with significant atmospheric subsidence is somewhat unusual during the summer in eastern Tennessee. Evidently, the northeasterly winds transported precursor pollutants to the park from the Mid-Atlantic states, and the pollutants were trapped under a subsidence inversion. These pollutants produced relatively high ozone concentrations over the 4-day period, which ended with the wind shift.

*Views from the Look Rock (GRS420, TN) air quality monitoring station.
Photo on left shows a day with 83 mile visual range. Hazeiness (16 mile visual range) in
the photo on right is caused primarily by tiny sulfate particles.*



July 14, 2008 – 83 mile visual range

Source: NPS



July 19, 2008 – 16 mile visual range

Chapter 5: Data Quality

CASTNET measurements and supporting activities are assessed routinely in order to provide high-quality information to meet project objectives. The CASTNET Quality Assurance program is based on specific data quality objectives that are evaluated using data quality indicators (DQI) such as precision, accuracy, and completeness. Measurements taken during 2008 and historical data collected over the period 1990–2007 were analyzed relative to DQI and their associated metrics. These analyses demonstrate that CASTNET data can be used with confidence and that CASTNET continues to produce information of the highest quality.

The CASTNET QA program was designed to ensure that all reported data are of known and documented quality in order to meet CASTNET objectives and to be reproducible and comparable with data from other monitoring networks and laboratories. The 2008 QA program elements are documented in the CASTNET Quality Assurance Project Plan (QAPP), Revision 4.1 (MACTEC, 2008c). The QAPP is comprehensive and includes standards and policies for all components of project operation from site selection through final data reporting. It includes major sections on field measurements, chemical analysis of field samples, data management, and assessments and response actions. Standard operating procedures are included as appendices.

Data quality indicators (DQI) are quantitative statistics and qualitative descriptors used in interpreting the degree of acceptability and utility of the data collected. The DQI for CASTNET are precision, accuracy, completeness, bias, representativeness, and comparability. Precision, accuracy, and completeness for CASTNET 2008 data were analyzed and compared with historical data collected during the period 1990 through 2007. The information in this report is supplemented by analyses that are discussed in quarterly CASTNET Quality Assurance Reports (MACTEC, 2008b; 2008e; 2008f; 2009b). These QA reports are produced four times per year with the fourth quarter report including an annual summary.

Precision

Exposed Filter Concentrations

Figure 5-1 provides a bar chart in which the bars represent precision estimates for five CASTNET analytes. Table 5-1 lists the precision and accuracy criteria for laboratory filter pack measurements. Precision is defined as the mean absolute relative percent difference (MARPD).

Figure 5-1 Historical and 2008 Precision Data for Atmospheric Concentrations

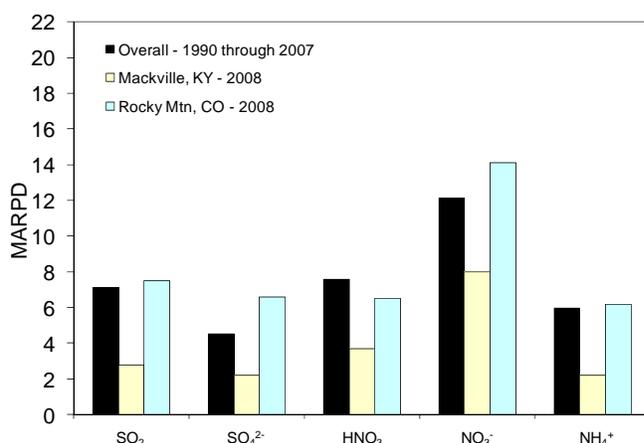


Table 5-1 Data Quality Indicator Criteria for CASTNET Laboratory Measurements

Analyte	Method	Precision ¹ (MARPD)	Accuracy ² (%)	Nominal Reporting Limits
Ammonium (NH ₄ ⁺)	Automated colorimetry	10	90 - 110	0.020 mg-N/L
Sodium (Na ⁺)	ICP-AES	5	95 - 105	0.005 mg/L
Potassium (K ⁺)	ICP-AES	5	95 - 105	0.006* mg/L
Magnesium (Mg ²⁺)	ICP-AES	5	95 - 105	0.003 mg/L
Calcium (Ca ²⁺)	ICP-AES	5	95 - 105	0.006* mg/L
Chloride (Cl ⁻)	Ion chromatography	5	95 - 105	0.020 mg/L
Nitrate (NO ₃ ⁻)	Ion chromatography	5	95 - 105	0.008 mg-N/L
Sulfate (SO ₄ ²⁻)	Ion chromatography	5	95 - 105	0.040 mg/L

Note: ¹ This column lists precision goals for both network precision calculated from collocated filter samples and laboratory precision based on replicate samples. The goal for the ICP-AES precision relative percent difference (RPD) criterion changed from 10 percent to 5 percent at the onset of the CASTNET III contract beginning on July 30, 2003. The precision criterion is applied as described below: QC conditions: (v1 = initial response; v2 = replicate response; RL = nominal reporting limit)
 Condition 1: if (v1 or v2 < RL and the absolute value of (v1 - v2) < RL) = OK
 Condition 2: if (v1 - v2) < RL and v1 < 5 x RL = OK
 Condition 3: if (v1 > 5*RL and RPD < 5%) = OK
 Status: one of the conditions is OK = Precision QC Passes

² This column lists laboratory accuracy goals based on reference standards and continuing calibration verification spikes. The goal for the ICP-AES accuracy criterion changed from 90–110 percent recovery to 95–105 percent for continuing calibration verification spikes beginning on July 30, 2003. The criterion remains 90–110 percent for ICP-AES reference standards.

* Effective September 26, 2007, changed to 0.006 mg/L from 0.003 mg/L for calcium and 0.005 mg/L for potassium

$$\text{MARPD} = \frac{1}{k} \sum_{j=1}^k \left(\frac{|S1 - S2|}{S1 + S2} \right)_j \times 200 \quad \text{Equation 5-1}$$

Where:

- S1 = The value for the primary sampler
- S2 = The value for the collocated sampler
- k = The number of pairs of valid data

$$\text{Percent recovery} = \frac{S1}{S2} \times 100 \quad \text{Equation 5-2}$$

$$\text{RPD} = \left(\frac{S1 - S2}{S1 + S2} \right) \times 200 \quad \text{Equation 5-3}$$

For more information on the analytical methods and associated precision and accuracy criteria used for 2007, see the CASTNET QAPP Revision 4.1 (MACTEC, 2008c).

Historical (1990 through 2007) data for all 11 collocated site pairs operated over the history of the network and the 2008 data for the current collocated sites at Mackville, KY (MCK131/231) and Rocky Mountain National Park, CO (ROM406/206) are provided. The two sites at Rocky Mountain National Park are operated independently. ROM206 is operated on behalf of EPA and ROM406 on behalf of NPS. Trace cations and chloride are excluded from this figure but are shown later in Figure 5-2. The historical results vary from just over 4 percent for particulate SO_4^{2-} to more than 12 percent for particulate NO_3^- . The historical MARPD for SO_4^{2-} met the criterion for the CASTNET filter pack measurements shown in Table 5-1. The historical results for SO_2 and HNO_3 were above the 5 percent criterion but are considered reasonable. The results for NH_4^+ met the goal of 10 percent. The results for NO_3^- were significantly above the 5 percent goal. Historically, the precision of NO_3^- measurements has been consistently worse than for the other analytes, possibly because NO_3^- concentrations are the lowest of all the pollutants, and nitrate species include sampling artifacts (Lavery *et al.*, 2009).

The 2008 precision results shown in Figure 5-1 indicate that the MARPD data for MCK131/231 were lower than (i.e., more precise than) the historical results for all five parameters. Four parameters (SO_2 , SO_4^{2-} , HNO_3 , and NH_4^+) met precision criteria. The 2008 results for ROM406/206 showed better precision than historical results for HNO_3 , and the result for NH_4^+ met the DQI criteria. The precision criterion for NO_3^- was not met at either site during 2008. The precision results were worse for ROM406/206 because of lower concentrations measured at this site. Overall, the filter pack precision results for 2008 were about the same as the 2007 results.

Historical and 2008 precision statistics for four cations and chloride (Cl^-) are summarized in Figure 5-2. The historical MARPD statistics for both MCK131/231 and ROM406/206 did not meet the DQI criterion of 5 percent. As discussed in earlier CASTNET annual reports (e.g., MACTEC, 2004 and MACTEC, 2003), the very high historical MARPD for Na^+ was the result of sample bottle contamination. These bottles are no longer purchased. Also, acceptance testing of the Teflon filters was instituted for the trace cations and Cl^- in 2003. The 2008 precision results show that Cl^- met the precision criterion at MCK131/231, and the results for the four cations were reasonable. The 2008 precision results for MCK131/231 showed some improvement over the 2007 results. The 2008 precision data for ROM406/206 are somewhat worse than 2007.

Table 5-2 summarizes 2008 precision results by quarter for the two sets of collocated sites. See the 2008 Quarterly Data Reports (MACTEC, 2008a; 2008d; 2009a; 2009c) and QA Quarterly Reports (MACTEC, 2008b; 2008e; 2008f; 2009b) for discussions of quarterly precision data.

Figure 5-2 Historical and 2008 Precision Data for Cation and Cl⁻ Concentrations

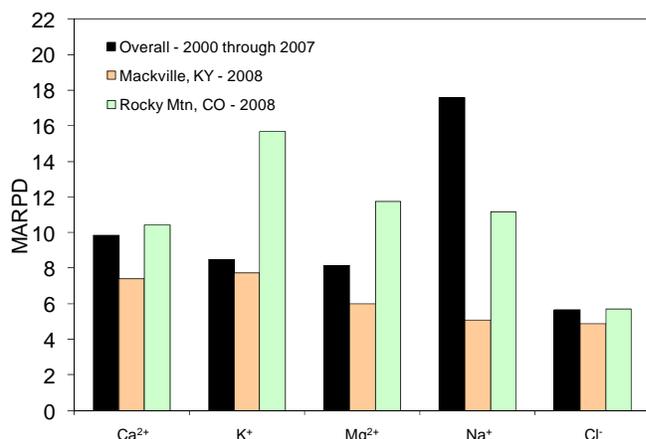
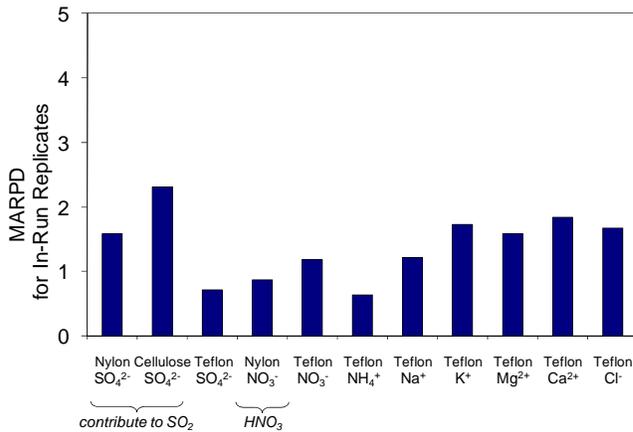


Table 5-2 Collocated Precision Results (MARPD) for 2008 Filter Pack Data by Quarter

Site Pairs	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HNO ₃	SO ₂	Total NO ₃ ⁻	Cl ⁻
MCK131/231, KY											
Quarter 1	1.78	4.68	2.00	7.67	4.66	2.39	2.45	2.66	1.89	2.17	5.04
Quarter 2	2.01	13.16	1.65	12.21	11.67	8.88	6.94	3.18	2.86	4.26	5.04
Quarter 3	2.36	9.40	2.38	3.78	3.73	4.10	4.39	3.91	1.94	3.25	4.63
Quarter 4	2.64	4.80	2.69	5.97	3.82	4.86	17.11	4.99	4.40	3.31	4.79
2008	2.20	8.01	2.18	7.41	5.97	5.06	7.72	3.68	2.77	3.25	4.87
ROM406/206, CO											
Quarter 1	7.03	10.22	5.82	9.08	11.34	9.05	14.60	6.45	5.14	6.73	4.38
Quarter 2	6.74	12.77	6.67	8.22	11.46	8.13	11.20	4.18	8.04	6.13	5.28
Quarter 3	7.76	17.79	7.74	11.55	10.26	10.82	7.29	8.63	9.25	8.06	6.70
Quarter 4	4.84	15.66	4.40	12.88	14.00	16.72	29.56	6.77	7.59	4.65	6.46
2008	6.59	14.11	6.16	10.43	11.76	11.18	15.66	6.51	7.50	6.39	5.71

The 2008 analytical precision results for five analytes and the three filter types are presented in Figure 5-3. The results were based on analysis of 5 percent of the samples that were randomly selected for replication in each batch. The results of in-run replicate analyses were compared to the results of the original concentrations. The laboratory precision data met the 5 percent measurement criterion listed in Table 5-1.

Figure 5-3 Precision Results for Laboratory Replicate Samples (2008)



Ozone Concentrations

CASTNET QA procedures for the EPA-sponsored O₃ analyzers are different from the EPA QA requirements for State and Local Monitoring Stations (SLAMS) monitoring (EPA, 1998). However, as discussed in Chapter I, MACTEC is preparing CASTNET monitoring shelters to meet the 40 CFR Part 58 monitoring requirements. The QA procedures for the O₃ analyzers at the NPS-sponsored CASTNET sites also do not meet the SLAMS requirements, but the NPS sites utilize the appropriate procedures and equipment, including an on-site O₃ transfer standard. In any event, the operation of the collocated EPA and NPS O₃ analyzers at ROM406/206, CO provides an opportunity to evaluate the precision of the independent systems. Table 5-3 provides the DQI criteria for the CASTNET continuous measurements including O₃. The precision criterion for the collocated O₃ data is 10 percent.

MARPD statistics were calculated from hourly O₃ measurements obtained from the collocated sites MCK131/231, KY and ROM406/206, CO during 2008. In addition, quarterly historical precision statistics were compiled for all collocated sites. Quarterly precision results are summarized in Figure 5-4. Table 5-4 provides precision results for O₃ concentrations by quarter for 2008. The data show the historical and 2008 results met the 10 percent criterion. The historical data are based on the operation of 11 collocated site pairs over the history of the network. The 2008 O₃ precision data for ROM406/206 were excellent and demonstrate the comparability of the two independent O₃ sampling systems.

Table 5-3 Data Quality Indicator Criteria for CASTNET Field Measurements

Measurement		Criteria*	
Parameter	Method	Precision	Accuracy
Wind Speed	Anemometer	± 0.5 m/s	The greater of ± 0.5 m/s for winds < 5 m/s or ± 5% for winds ≥ 5 m/s
Wind Direction	Wind Vane	± 5°	± 5°
Sigma Theta	Wind Vane	Undefined	Undefined
Relative Humidity	Thin Film Capacitor	± 10% (of full scale)	± 5%, relative humidity > 85% ± 20%, relative humidity ≤ 85%
Solar Radiation	Pyranometer	± 10% (of reading taken at local noon)	± 10%
Precipitation	Tipping Bucket Rain Gauge	± 10% (of reading)	± 0.05 inch†
Ambient Temperature	Platinum RTD	± 1.0°C	± 0.5°C
Delta Temperature	Platinum RTD	± 0.5°C	± 0.5°C
Surface Wetness	Conductivity Bridge	Undefined	Undefined
Ozone	UV Absorbance	± 10% (of reading)	± 10%
Filter Pack Flow	Mass Flow Controller	± 10%	± 5%

Note: Mean Absolute Difference (MAD) is the precision measure for difference criteria such as for wind speed.

MARPD is the precision measure for percentage criteria such as for relative humidity.

°C = degrees Celsius

m/s = meters per second

RTD = resistance-temperature device

UV = ultraviolet

* Precision criteria apply to collocated instruments, and accuracy criteria apply to calibration of instruments.

† For target value of 0.50 inch.

Figure 5-4 Historical and 2008 Precision Data by Quarter for Ozone Concentrations

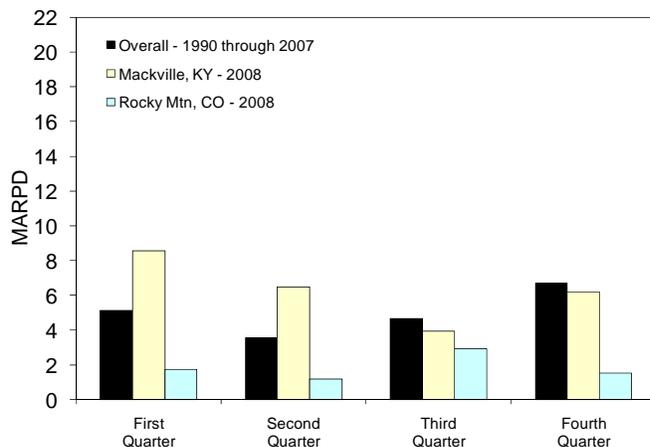


Table 5-4 Collocated Precision Results (MARPD) for 2008 Ozone Concentrations by Quarter

Sampling Period	MCK131/231, KY	ROM406/206, CO
Quarter 1	8.57	1.71
Quarter 2	6.49	1.16
Quarter 3	3.91	2.90
Quarter 4	6.19	1.51
2008	6.29	1.82

Continuous Measurements

Precision criteria for the continuous measurements are listed in Table 5-3. Precision was calculated in terms of the MARPD or mean absolute difference (MAD) of hourly measurements collected at all collocated pairs operated over the history of the network and also for MCK131/231, KY and ROM406/206, CO for 2008. Figure 5-5 provides precision results for historical data through 2007 and precision statistics for the two collocated sites for 2008. All historical precision results met the DQI criteria. Table 5-5 gives precision data by quarter for 2008 for the two collocated sites. The results show excellent instrument precision. All 2008 annual and quarterly precision data met the DQI criteria. The first quarter MAD value for wind direction at ROM 406/206, CO was not available because of invalid data from ROM206, CO during that quarter. Precipitation measurements were invalid from MCK231, KY during the second and third quarters and delta temperature data were invalid for the second quarter from ROM406, CO.

Figure 5-5 Historical and 2008 Precision Data for Continuous Measurements

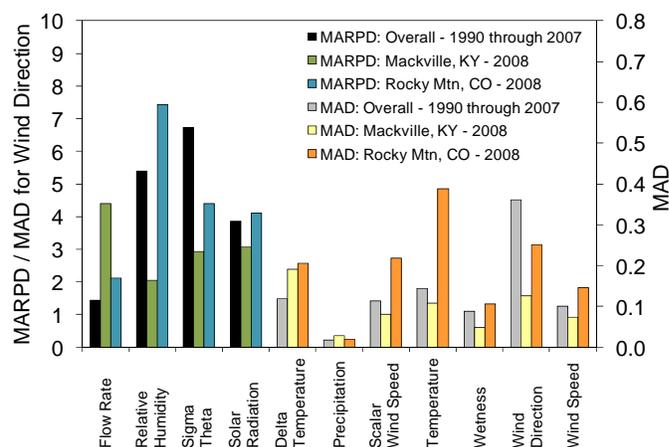


Table 5-5 Collocated Precision Results for 2008 Continuous Measurements by Quarter

Site Pairs	MARPD				MAD						
	Flow Rate	Relative Humidity	Sigma Theta	Solar Radiation	Delta Temperature	Precipitation	Scalar Wind Speed	Temperature	Wetness	Wind Direction	Wind Speed
MCK131/231, KY											
Quarter 1	1.31	2.87	2.21	1.96	0.34	0.04	0.10	0.12	0.05	1.32	0.09
Quarter 2	9.66	2.47	2.97	3.03	0.08	na	0.09	0.09	0.07	1.01	0.08
Quarter 3	2.22	0.83	4.21	4.33	0.08	na	0.08	0.04	0.05	1.36	0.07
Quarter 4	4.46	2.03	2.29	2.96	0.26	0.02	0.05	0.18	0.03	2.60	0.05
2008	4.41	2.05	2.92	3.07	0.19	0.03	0.08	0.11	0.05	1.57	0.07
ROM406/206, CO											
Quarter 1	0.90	8.19	4.53	4.59	0.11	0.02	0.28	0.49	0.23	na	0.19
Quarter 2	0.91	7.51	4.49	5.56	na	0.04	0.23	0.35	0.05	2.96	0.15
Quarter 3	1.28	4.86	4.74	3.58	0.23	0.01	0.17	0.36	0.09	4.82	0.11
Quarter 4	5.33	9.18	3.88	2.65	0.28	0.01	0.20	0.36	0.06	1.66	0.14
2008	2.11	7.44	4.41	4.10	0.21	0.02	0.22	0.39	0.11	3.14	0.15

Accuracy

Laboratory Filter Concentrations

Accuracy of laboratory measurements is assessed through the analysis of reference and continuing calibration verification (CCV) samples. Reference samples and CCV are procured from independent suppliers and are traceable to the National Institute of Standards and Technology (NIST). Reference samples are analyzed at the beginning and end of each analytical batch to verify the accuracy and stability of the calibration curve. The target value of the CCV solution is set to the midrange of the calibration curve. In 2008, the CCV were analyzed every tenth sample to verify that instrument calibration had not drifted beyond established limits. Table 5-6 presents the percent recoveries and standard deviations for reference samples and CCV relative to target concentrations. The table shows that the DQI goals (see Table 5-1) were met in 2008. Table 5-6 corroborates the precision results that were shown in Figure 5-3.

Table 5-6 Filter Pack Quality Control Summary for 2008

Filter Type	Parameter	Reference Sample ¹ Recovery (%R)			Continuing Calibration Verification Samples (%R)			In-Run Replicate ² (RPD)		
		Mean	Standard Deviation	Count ³	Mean	Standard Deviation	Count ³	Mean	Standard Deviation	Count ³
Teflon	SO ₄ ²⁻	98.62	1.69	153	99.57	1.39	717	0.71	1.29	331
	NO ₃ ⁻	100.02	1.33	153	99.57	1.28	717	1.18	1.87	331
	NH ₄ ⁺	101.72	1.88	144	99.61	1.22	696	0.63	0.71	326
	Ca ²⁺	104.42	2.87	146	100.83	1.21	710	1.84	2.90	332
	Mg ²⁺	102.66	1.50	146	100.05	0.80	710	1.58	1.97	332
	Na ⁺	98.21	2.32	146	100.17	1.12	710	1.21	2.31	332
	K ⁺	97.83	2.46	146	100.05	0.96	710	1.73	1.90	332
	Cl ⁻	99.80	1.53	153	100.01	1.24	717	1.67	3.11	327
Nylon	SO ₄ ²⁻	99.83	1.77	144	99.88	1.54	692	1.58	1.99	317
	NO ₃ ⁻	99.60	1.38	144	100.27	1.43	692	0.87	1.07	317
Cellulose	SO ₄ ²⁻	100.23	1.17	173	99.48	0.96	686	2.31	3.04	318

Note: % R = percent recovery (see Equation 5-2)
 RPD = relative percent difference (see Equation 5-3)

¹ Results of reference sample analyses provide accuracy estimates

² Results of replicate analyses provide precision estimates

³ Number of QC Samples

Continuous Measurements

Table 5-7 presents field accuracy results for 2008 based on instrument challenges performed using independent reference standards during site calibration visits. CASTNET sites were calibrated every six months with NIST-traceable standards. The calibration results were evaluated using the accuracy criteria listed in Table 5-3. Each parameter was within its criterion with at least 90 percent frequency with the exception of high (> 85 percent) relative humidity at 87.3 percent and solar radiation at 85.0 percent. However, these results did not adversely affect data collection because data are not considered invalid unless criteria are exceeded by more than two times the criterion. Using the two times standard, the two parameters passed with 98.2 and 98.1 percent frequency, respectively.

Table 5-7 Accuracy Results for 2008 Field Measurements

Parameter	Percent Within Criterion
Temperature (0°C)	98.3 percent
Temperature (ambient)	99.1 percent
Delta Temperature (0°C)	98.3 percent
Delta Temperature (ambient)	96.6 percent
*Relative Humidity > 85%	87.3 percent
Relative Humidity ≤ 50%	99.1 percent
*Solar Radiation	85.0 percent
Wind Direction North	92.2 percent
Wind Direction South	92.2 percent
Wind Speed < 5 m/s	98.2 percent
Wind Speed ≥ 5 m/s	98.2 percent
Precipitation	100.0 percent
Wetness (w/in 0.5 volts)	99.1 percent
Ozone Slope	97.2 percent
Ozone Intercept	98.2 percent
Flow Rate	100.0 percent

Note: °C = degrees Celsius

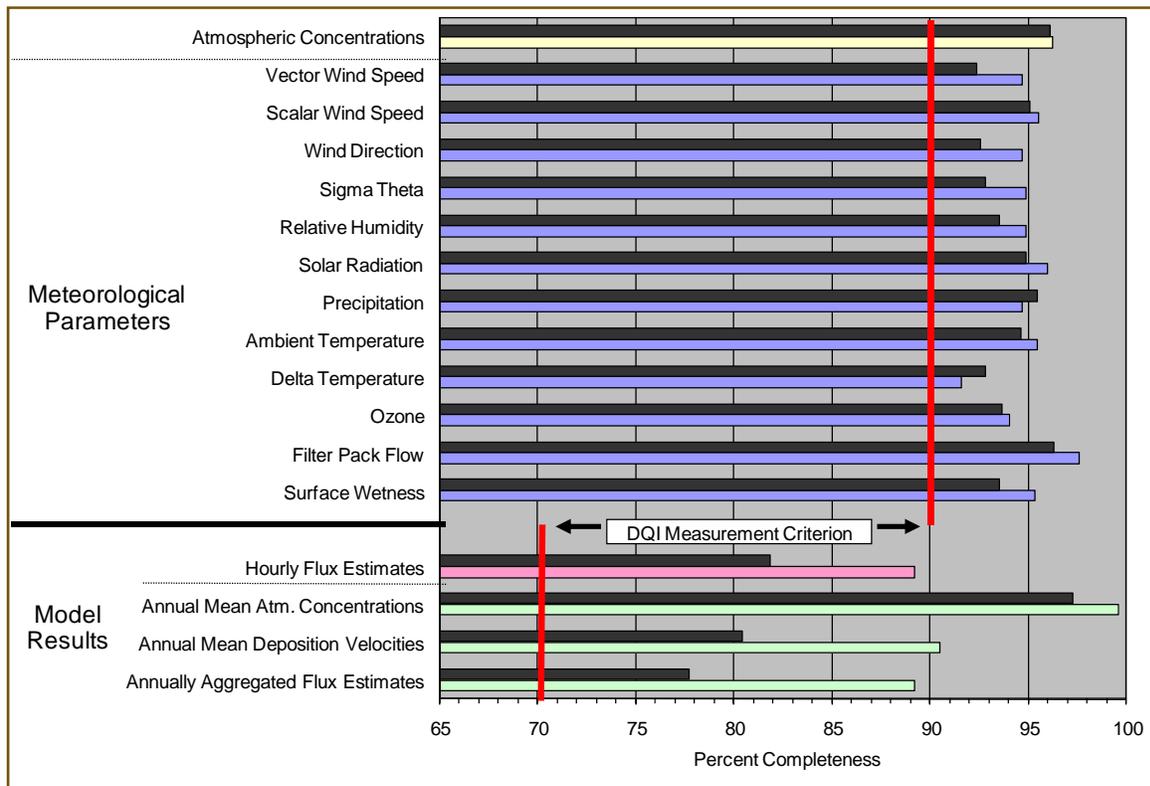
m/s = meters per second

* Per CASTNET project protocols, data are flagged as “suspect” (S) but still considered valid if the calibration criterion is not exceeded by more than its magnitude (i.e., if within 2x the criterion). The percent within 2x criterion for these parameters was about 98 percent.

Completeness

Completeness is defined as the percentage of valid data points obtained from a measurement system relative to total possible data points. The CASTNET measurement criterion for completeness requires a minimum completeness of 90 percent for every measurement for each quarter. In addition, data aggregation procedures require approximately 70 percent completeness for hourly fluxes and weekly concentrations/fluxes. Figure 5-6 presents historical (black) and 2008 completeness data for all sites for measured filter concentrations and continuous measurements and calculated parameters. The figure shows that the 2008 direct measurements met the 90 percent completeness goal, and five continuous measurements exceeded 95 percent, including filter pack flow. Atmospheric concentrations also exceeded 95 percent. The four parameters derived from model results exceeded 89 percent completeness for 2008. Completeness results for 2008 are better than historical results for all parameters except delta temperature and precipitation.

Figure 5-6 Historical and 2008 Percent Completeness of Measurements and Modeled Estimates (black bars are 1990–2007)

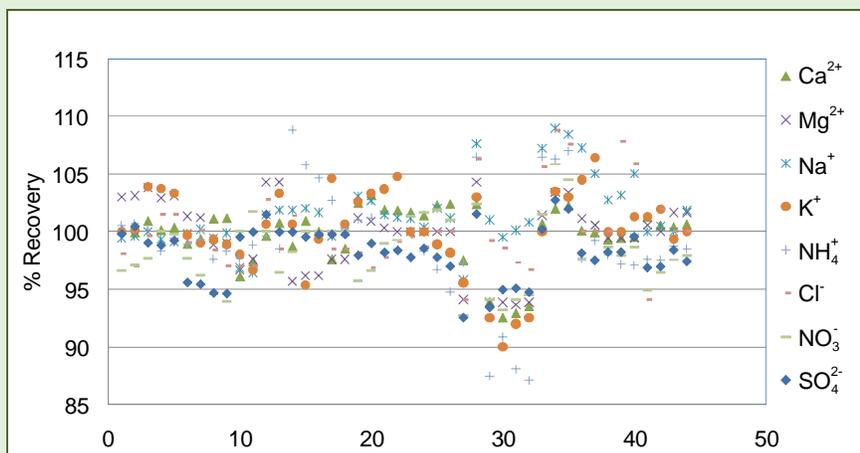


Interlaboratory Comparison Results

The MACTEC laboratory is one of eight laboratories that participate in the US Geological Survey (USGS) interlaboratory comparison program. The laboratory receives four samples for chemical analysis from USGS every two weeks. The samples are a mix of 44 synthetically prepared samples, 8 deionized water samples, and 52 natural wet deposition samples. The laboratory reported the eight CASTNET parameters for a total of 104 samples during 2008. Results for the 44 synthetically prepared samples are depicted in the figure below, which presents results as percent recoveries of the median value for all laboratories.

As shown below, only 3 of 352 reported values were more than 10 percent from their corresponding median. All values shown are within two standard deviations of their corresponding median.

Figure 5a Results from Intercomparison of Synthetic and Natural Samples



Additionally, precision results for the 52 natural wet deposition samples were reported as absolute percent differences for replicate analyses. None of the paired natural wet deposition sample data points exceeded 10 percent difference. These results are presented in the USGS report (USGS, 2008).

The laboratory generally participates in two to three studies each year through Environment Canada's National Water Research Institute (NWRI) Proficiency Testing QA Program in addition to interlaboratory comparison studies for the USGS. MACTEC's laboratory was rated free of systemic bias for all eight parameters for the two NWRI 2008 studies and was tied for first place out of 39 participating laboratories at the end of 2008 (Environment Canada, 2008).

Summary of Data Quality Results

DQI results demonstrate that field and laboratory processes were adequately monitored through QA/QC procedures and were generally free of systemic bias during 2008. Accuracy data met the established criteria for field and laboratory parameters with the exception of relative humidity > 85 percent and solar radiation. However, since the criterion was exceeded by a value less than its own magnitude, the associated continuous data collected are considered valid.

Precision data for SO_2 , SO_4^{2-} , HNO_3 , and NH_4^+ are considered acceptable. Precision data for nitrate analyses of collocated field samples have not met the established criterion due, most likely, to the low concentrations generally measured and the uncertain nature of the gas-particle equilibrium of the nitrate species.

The 2008 precision data for O_3 concentrations met the 10 percent DQI criterion. The 2008 precision data for ROM406/206 were excellent and demonstrate the comparability of the two independent O_3 sampling systems. Completeness criteria were met for all parameters in 2008. Completeness results for 2008 were better than historical results for all but two parameters.

The precision of continuous measurements for 2008 was excellent. Quarterly and annual precision results for 2008 met the DQI criteria.

The laboratory intercomparison results show the CASTNET analytical chemistry laboratory was rated free of systematic bias and was tied for first place out of 39 participating laboratories at the end of 2008 in Environment Canada's NWRI Proficiency Testing QA Program.

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Appendix A

Locational and Operational Characteristics of CASTNET Sites

Table A-I Locational and Operational Characteristics of CASTNET Sites

Site ID	Site Name	Start date	Latitude (°N)	Longitude (°W)	Elevation (m)	Deposition Filter Pack ¹	Ozone and Meteorology ²	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Alabama											
SND152	Sand Mountain	12/27/88	34.2894	85.9704	352	•	•	Agri.	Rolling	Y	EPA
Alaska											
DEN417	Denali National Park and Preserve	10/06/98	63.7258	148.9633	661	•	•	Forested	Complex	N	NPS
Arizona											
CHA467	Chiricahua National Monument	04/25/89	32.0092	109.3892	1570	•	•	Range	Complex	N	NPS
GRC474	Grand Canyon National Park	05/16/89	36.0597	112.1822	2073	•	•	Forested	Complex	M	NPS
PET427	Petrified Forest National Park	09/24/02	34.8225	109.8919	1723	•	•	Desert	Flat	Y	NPS
Arkansas											
CAD150	Caddo Valley	10/04/88	34.1792	93.0989	71	•	•	Forested	Rolling	N	EPA
California											
CON186	Converse Station	06/17/03	34.1941	116.9130	1837	•	•	Agri./Forested	Complex	N	EPA
JOT403	Joshua Tree National Park	02/16/95	34.0714	116.3906	1244	•	•	Desert	Complex	M	NPS
LAV410	Lassen Volcanic National Park	07/25/95	40.5403	121.5764	1756	•	•	Forested	Complex	M	NPS
PIN414	Pinnacles National Monument	05/16/95	36.4850	121.1556	335	•	•	Forested	Complex	M	NPS
SEK430	Sequoia National Park	04/07/05	36.4894	118.8269	457	•	•	Forested	Mountaintop	N	NPS
YOS404	Yosemite National Park	09/25/95	37.7133	119.7061	1605	•	•	Forested	Complex	N	NPS

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Colorado											
GTH161	Gothic	05/16/89	38.9573	106.9854	2926	•	•	Range	Complex	N	EPA
MEV405	Mesa Verde National Park	01/10/95	37.1983	108.4903	2165	•	•	Forested	Complex	M	NPS
ROM206	Rocky Mountain National Park	07/03/01	40.2778	105.5453	2743	•	•	Forested	Complex	M	EPA
ROM406	Rocky Mountain National Park	12/20/94	40.2778	105.5453	2743	•	•	Forested	Complex	M	NPS
Connecticut											
ABT147	Abington	12/28/93	41.8402	72.0111	209	•	•	Urban-Agri.	Rolling	M	EPA
Florida											
EVE419	Everglades National Park	10/06/98	25.3911	80.6806	2	•	• ⁴	Swamp	Flat	Y	NPS
IRL141	Indian River Lagoon	07/09/01	30.1065	80.4554	2	•	•	Beach	Flat	Y	EPA
SUM156	Sumatra	12/28/88	30.1065	84.9938	14	•	•	Forested	Flat	Y	EPA
Georgia											
GAS153	Georgia Station	06/28/88	33.1812	84.4100	270	•	•	Agri.	Rolling	M	EPA
Illinois											
ALH157	Alhambra	06/28/88	38.8690	89.6229	164	•	•	Agri.	Flat	Y	EPA
BVL130	Bondville	02/09/88	40.0520	88.3725	212	•	•	Agri.	Flat	Y	EPA
STK138	Stockton	12/28/93	42.2872	89.9998	274	•	•	Agri.	Rolling	M	EPA

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Site ID	Site Name	Start date	Latitude (°N)	Longitude (°W)	Elevation (m)	Deposition Filter Pack ¹	Ozone and Meteorology ²	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Indiana											
SAL133	Salamonie Reservoir	06/28/88	40.8164	85.6608	250	•	•	Agri.	Flat	Y	EPA
VIN140	Vincennes	08/04/87	38.7406	87.4844	134	•	•	Agri.	Rolling	M	EPA
Kansas											
KNZ184	Konza Prairie	03/26/02	39.1021	96.6096	348	•	•	Prairie	Flat	Y	EPA
Kentucky											
CDZ171	Cadiz	10/01/93	36.7841	87.8500	189	•	•	Agri.	Rolling	M	EPA
CKT136	Crockett	08/24/93	37.9211	83.0658	455	•	•	Agri.	Rolling	Y	EPA
MAC426	Mammoth Cave National Park	07/24/02	37.1319	86.1478	243	•	•	Agri./Forested	Rolling	M	NPS
MCK131	Mackville	07/31/90	37.7044	85.0483	353	•	•	Agri.	Rolling	M	EPA
Maine											
ACA416	Acadia National Park	12/01/98	44.3769	68.2608	158	•	•	Forested	Complex	M	NPS
ASH135	Ashland	12/20/88	46.6039	68.4142	235	•	•	Agri.	Flat	Y	EPA
HOW132	Howland	11/24/92	45.2158	68.7085	69	•	•	Forested	Rolling	Y	EPA
Maryland											
BEL116	Beltsville	11/01/88	39.0283	76.8175	46	•	•	Urban-Agri.	Flat	N	EPA
BWR139	Blackwater National Wildlife Refuge	07/04/95	38.4448	76.1115	4	•	•	Forest-Marsh	Coastal	M	EPA

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Site ID	Site Name	Start date	Latitude (°N)	Longitude (°W)	Elevation (m)	Deposition Filter Pack ¹	Ozone and Meteorology ²	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Michigan											
ANA115	Ann Arbor	06/28/88	42.4164	83.9019	267	•	•	Forested	Flat	M	EPA
HOX148	Hoxeyville	10/31/00	44.1809	85.7390	298	•	•	Forested	Flat	Y	EPA
UVL124	Unionville	06/28/88	43.6139	83.3597	201	•	•	Agri.	Flat	Y	EPA
Minnesota											
VOY413	Voyageurs National Park	06/13/96	48.4128	92.8292	429	•	•	Forested	Rolling	M	NPS
Mississippi											
CVL151	Coffeeville	12/27/88	34.0028	89.7989	134	•	•	Forested	Rolling	M	EPA
Montana											
GLR468	Glacier National Park	12/27/88	48.5103	113.9956	976	•	•	Forested	Complex	N	NPS
Nebraska											
SAN189	Santee Sioux	07/05/06	42.8292	97.8541	429	•	•	Agri.	Rolling	N	EPA
Nevada											
GRB411	Great Basin National Park	05/16/95	39.0053	114.2158	2060	•	•	Forested	Complex	M	NPS
New Hampshire											
WST109	Woodstock	12/27/88	43.9446	71.7008	258	•	•	Forested	Complex	N	EPA
New Jersey											
WSP144	Washington's Crossing	12/27/88	40.3133	74.8726	61	•	•	Urban-Agri.	Rolling	M	EPA

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Site ID	Site Name	Start date	Latitude (°N)	Longitude (°W)	Elevation (m)	Deposition Filter Pack ¹	Ozone and Meteorology ²	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
New York											
CAT175	Claryville	05/10/94	41.9423	74.5519	765	•	• ^{4, 5}	Forested	Complex	N	EPA
CTH110	Connecticut Hill	09/28/87	42.4010	76.6535	515	•	•	Forested	Rolling	N	EPA
HWF187	Huntington Wildlife Forest	05/28/02	43.9732	74.2232	502	•	•	Forested	Complex	N	EPA
North Carolina											
BFT142	Beaufort	12/28/93	34.8843	76.6213	2	•	•	Agri.	Flat	Y	EPA
CND125	Candor	09/25/90	35.2643	79.8382	198	•	•	Forested	Rolling	M	EPA
COW137	Coweeta	11/04/87	35.0605	83.4302	686	•	•	Forested	Complex	N	EPA
PNF126	Cranberry	12/27/88	36.1040	82.0448	1250	•	•	Forested	Mountaintop	M	EPA
North Dakota											
THR422	Theodore Roosevelt National Park	10/06/98	46.8947	103.3778	850	•	•	Range	Rolling	Y	NPS
Ohio											
DCP114	Deer Creek State Park	09/28/88	39.6358	83.2600	267	•	•	Agri.	Rolling	Y	EPA
LYK123	Lykens	01/10/89	40.9169	82.9981	303	•	•	Agri.	Flat	M	EPA
OXF122	Oxford	08/18/87	39.5314	84.7231	284	•	•	Agri.	Rolling	N	EPA
QAK172	Quaker City	09/28/93	39.9431	81.3378	372	•	•	Agri.	Rolling	M	EPA
Oklahoma											
CHE185	Cherokee Nation	04/02/02	35.7507	94.6700	299	•	•	Agri.	Rolling	Y	EPA
Ontario											
EGB181	Egbert, Ontario	12/27/94	44.2317	79.7840	251	•	• ⁴	Agri.	Rolling	Y	EPA

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Site ID	Site Name	Start date	Latitude (°N)	Longitude (°W)	Elevation (m)	Deposition Filter Pack ¹	Ozone and Meteorology ²	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Pennsylvania											
ARE128	Arendtsville	06/28/88	39.9231	77.3078	269	•	•	Agri.	Rolling	M	EPA
KEF112	Kane Experimental Forest	01/03/89	41.5981	78.7683	622	•	•	Forested	Rolling	Y	EPA
LRL117	Laurel Hill State Park	12/15/87	39.9883	79.2522	615	•	•	Forested	Complex	N	EPA
MKG113	M.K. Goddard State Park	01/12/88	41.4250	80.1447	384	•	•	Forested	Rolling	N	EPA
PSU106	Penn. State University	01/06/87	40.7209	77.9316	376	•	•	Agri.	Rolling	M	EPA
South Dakota											
WNC429	Wind Cave National Park	11/18/03	43.5578	103.4839	1292	•	•	Prairie	Rolling	M	NPS
Tennessee											
ESP127	Edgar Evins State Park	03/22/88	36.0389	85.7330	302	•	•	Forested	Rolling	N	EPA
GRS420	Great Smoky Mountains National Park	10/06/98	35.6331	83.9422	793	•	•	Forested	Complex	N	NPS
SPD111	Speedwell	06/12/89	36.4698	83.8265	361	•	•	Agri.	Rolling	Y	EPA
Texas											
ALC188	Alabama-Coushatta	04/02/04	30.4210	94.4045	101	•	•	Forested	Rolling	Y	EPA
BBE401	Big Bend National Park	07/18/95	29.3022	103.1772	1052	•	•	Forested	Complex	M	NPS
PAL190	Palo Duro Canyon State Park	04/24/07	34.8803	101.6649	1050	•	•	Prairie	Complex	M	EPA
Utah											
CAN407	Canyonlands National Park	01/24/95	38.4586	109.8211	1809	•	•	Desert	Complex	M	NPS

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Site ID	Site Name	Start date	Latitude (°N)	Longitude (°W)	Elevation (m)	Deposition Filter Pack ¹	Ozone and Meteorology ²	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Virginia											
PED108	Prince Edward	11/03/87	37.1653	78.3070	150	•	•	Forested	Rolling	M	EPA
SHN418	Shenandoah National Park	06/28/88	38.5231	78.4347	1073	•	•	Forested	Mountaintop	M	NPS
VPI120	Horton Station	06/02/87	37.3300	80.5573	920	•	•	Forested	Mountaintop	N	EPA
Washington											
MOR409	Mount Rainier National Park	08/29/95	46.7583	122.1244	415	•	•	Forested	Complex	N	NPS
West Virginia											
CDR119	Cedar Creek State Park	11/10/87	38.8794	80.8478	234	•	•	Forested	Complex	N	EPA
PAR107	Parsons	01/19/88	39.0906	79.6614	510	•	•	Forested	Complex	N	EPA
Wisconsin											
PRK134	Perkinstown	09/27/88	45.2066	90.5972	472	•	•	Agri.	Rolling	M	EPA
Wyoming											
CNT169	Centennial	08/19/91	41.3722	106.2422	3178	•	•	Range	Complex	M	EPA
PND165	Pinedale	12/27/88	42.9214	109.7900	2388	•	•	Range	Rolling	M	EPA
YEL408	Yellowstone National Park	06/26/96	44.5597	110.4006	2400	•	•	Forested	Rolling	N	NPS

1. The dry deposition filters are analyzed for the following constituents:

- Teflon = SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, K⁺, Na⁺, Mg²⁺, Ca²⁺
- Nylon = SO₄²⁻, NO₃⁻ (reported as HNO₃)
- Cellulose = SO₄²⁻ (reported as SO₂)

2. Meteorological sensors: temperature, delta temperature, relative humidity, solar radiation, vector wind speed, scalar wind speed, wind direction, sigma theta, surface wetness, and precipitation via tipping bucket rain gauge.

3. N = No; Y = Yes; M = Marginal.

4. O₃ not measured.

5. Solar-powered site.

• Indicates current monitoring.

100 and 200 series = EPA – Operated Sites

400 series = NPS – Operated Sites

Appendix B

Acronyms and Abbreviations

List of Acronyms and Abbreviations

AGL	above ground level
AMoN	Ammonia Monitoring Network
ANC	acid neutralizing capacity
AQS	EPA's Air Quality System
ARP	Acid Rain Program
Ca ²⁺	particulate calcium ion
CAAA	Clean Air Act Amendments
Ca(NO ₃) ₂	calcium nitrate
CAIR	Clean Air Interstate Rule
CMAQ	Community Multiscale Air Quality
CAPMoN	Canadian Air and Precipitation Monitoring Network
CASTNET	Clean Air Status and Trends Network
CCV	continuing calibration verification samples
CDX	AQS Central Data Exchange
Cl ⁻	particulate chloride ion
CO	carbon monoxide
DMC	Data Management Center
DQI	data quality indicator
EAC	Early Action Compact areas
EGU	electric generating unit
EPA	U.S. Environmental Protection Agency
EDT	Eastern Daylight Time
H ₂ SO ₄	sulfuric acid
HNO ₃	nitric acid
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
IC	ion chromatography
ICP-AES	inductively coupled plasma-atomic emission spectrometry
IMPROVE	Interagency Monitoring of Protected Visual Environments
IP	Internet protocol
K ⁺	particulate potassium ion
K ₂ CO ₃	potassium carbonate
kg/ha/yr	kilograms per hectare per year
km	kilometer
LAI	leaf area index
lpm	liters per minute
LTM	Long-Term Monitoring program
m	meters
MACTEC	MACTEC Engineering and Consulting, Inc.
MACT	Maximum Available Control Technology
MAD	mean absolute difference

List of Acronyms and Abbreviations (continued)

MADPro	Mountain Acid Deposition Program
MARCH	Maryland Aerosol Research and Characterization
MARPD	mean absolute relative percent difference
MDN	Mercury Deposition Network
Mg ²⁺	particulate magnesium ion
mg/l	milligrams per liter
MLM	Multi-Layer Model
N	nitrogen
Na ⁺	particulate sodium ion
NAAQS	National Ambient Air Quality Standards
NADP	National Atmospheric Deposition Program
NaNO ₃	sodium nitrate
NAPAP	National Acid Precipitation Assessment Program
NBP	NO _x Budget Trading Program
NDDN	National Dry Deposition Network
NH ₃	ammonia
NH ₄ ⁺	particulate ammonium
NH ₄ NO ₃	ammonium nitrate
(NH ₄) ₂ SO ₄	ammonium sulfate
NIST	National Institute of Standards and Technology
NO ₃ ⁻	particulate nitrate
NO _x	nitrogen oxides
NOAA	National Oceanic and Atmospheric Administration
NPS	National Park Service
NTN	National Trends Network
NWRI	Environment Canada's National Water Research Institute
O ₃	ozone
OTC	Ozone Transport Commission
PM _{2.5}	fine particle matter
ppb	parts per billion
ppm	parts per million
QA	quality assurance
QAPP	Quality Assurance Project Plan
QC	quality control
RACT	Reasonably Available Control Technology
RPD	relative percent difference
S	sulfur
SIP	State Implementation Plan

List of Acronyms and Abbreviations (continued)

SLAMS	State and Local Monitoring Stations
SO ₂	sulfur dioxide
SO ₄ ²⁻	particulate sulfate
TIME	Temporally Integrated Monitoring of Ecosystems
total NO ₃ ⁻	gaseous nitric acid (HNO ₃) + particulate nitrate (NO ₃ ⁻)
µg/m ³	micrograms per cubic meter
USGS	U.S. Geological Survey
V _d	deposition velocity
VOCs	volatile organic compounds
z/s/p	zero/span/precision

For More Information:

**U.S. Environmental Protection Agency
Office of Air and Radiation
Clean Air Markets Division
Washington, DC**

On the Web:

CASTNET Home Page:

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Division Home Page:

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