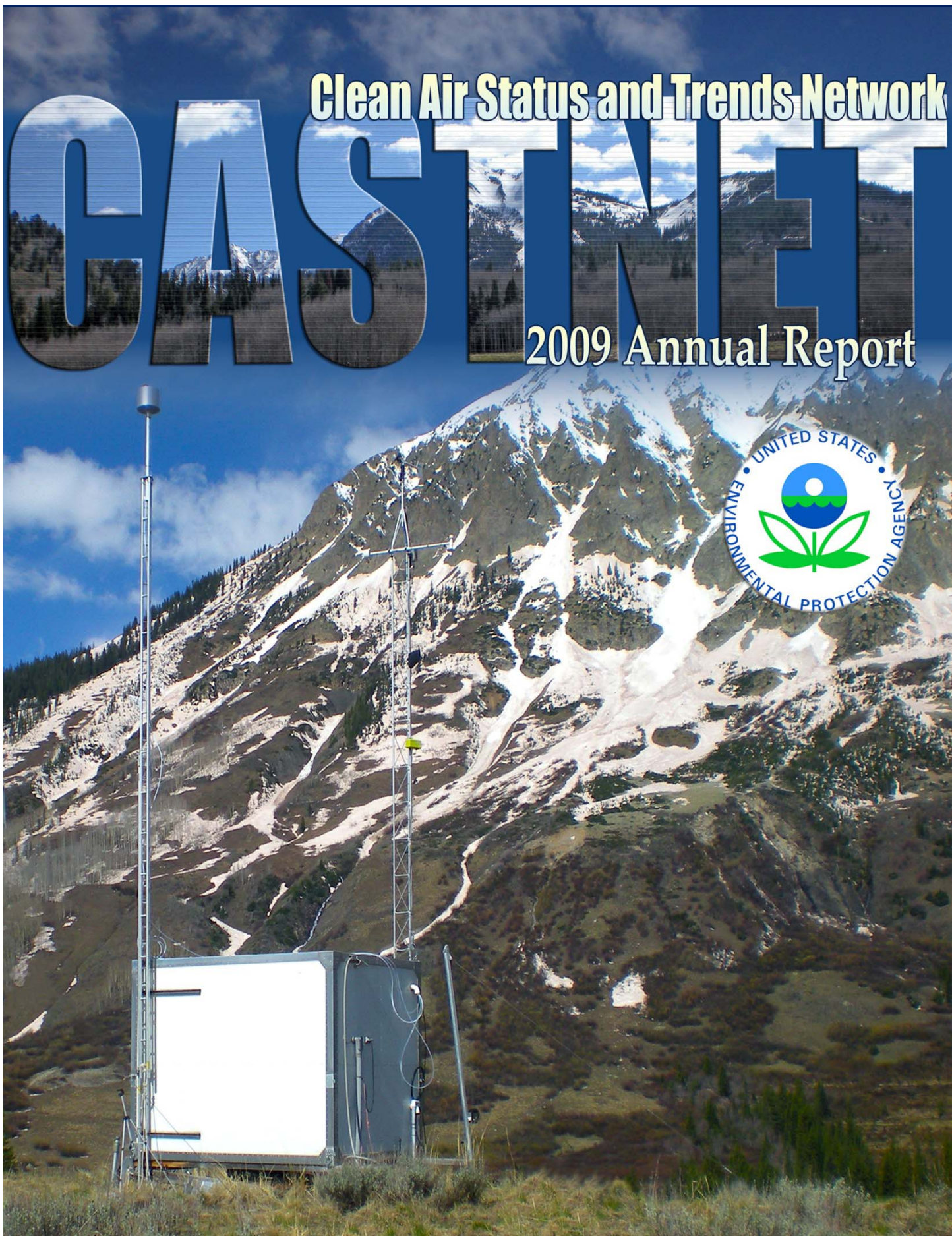


Clean Air Status and Trends Network

CASSTNET

2009 Annual Report



**Clean Air Status and Trends Network
(CASTNET)**

2009 Annual Report

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MACTEC Engineering and Consulting, Inc.

Prepared for:

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

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Executive Summary

The EPA Clean Air Status and Trends Network (CASTNET) provides real-world air pollution concentration data in order to evaluate the effectiveness of national and regional air pollution control programs. This report examines trends in air quality since 1990 and presents maps of the geographic distribution of ambient pollutant concentrations and deposition fluxes measured in 2009. CASTNET began collecting measurements in 1991 with the incorporation of 50 sites from the National Dry Deposition Network (NDDN), which had been in operation since 1987. CASTNET measures rural, regionally representative concentrations of sulfur and nitrogen species and ozone at 84 sites in the United States and Canada.

Key Results through 2009

- ❖ CASTNET pollutant measurements collected in 2009 showed the best air quality in the history of the network.
- ❖ Mean annual sulfur dioxide and particulate sulfate concentrations declined significantly over the 20-year period 1990 through 2009 and showed a sharp decline since 2005. Sulfur dioxide levels declined 56 percent while sulfate concentrations declined 41 percent at eastern reference sites.
- ❖ Total (dry + wet) sulfur deposition measured in the East declined by 46 percent from 1990 through 2009.
- ❖ Mean annual concentrations of total nitrate (nitric acid plus particulate nitrate) declined 30 percent over the 20-year period.
- ❖ Total nitrogen deposition declined by 26 percent from 1990 through 2009 at eastern reference sites.
- ❖ Ozone levels measured in 2009 continue to show a downward trend. The aggregated mean fourth highest daily maximum 8-hour average ozone concentration for 2009 (64 ppb) was the lowest in the history of the network. Only four eastern and five California sites recorded exceedances of the 8-hour standard of 0.075 ppm during the most recent 3-year period (2007–2009). During 2009, no eastern site recorded a fourth highest daily maximum 8-hour average ozone concentration above 75 ppb.
- ❖ Measurements taken during 2009 and historical data collected over the period 1990–2008 were analyzed relative to data quality objectives and their numerical measures. These analyses demonstrate that CASTNET data can be used with confidence and that CASTNET continues to produce information of the highest quality.

Chapter 1: CASTNET Overview

CASTNET is a national long-term environmental monitoring program operated by the Environmental Protection Agency and the National Park Service. The network was designed to provide data for assessing trends in air quality, atmospheric deposition, and ecological effects that result from air pollutant emission reduction regulations. CASTNET was established under the 1990 Clean Air Act Amendments and was developed from the National Dry Deposition Network, which began in 1987. As of December 2009, the network operated 84 monitoring stations throughout the contiguous United States, Alaska, and Canada. Measurements collected in 2009 show the continuing improvement in air quality with significant declines in sulfur and nitrogen pollutants and ozone concentrations measured throughout the network.

Background

The Environmental Protection Agency (EPA) established the Clean Air Status and Trends Network (CASTNET) under a provision in the 1990 Clean Air Act Amendments (CAAA) 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). The U.S. 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 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. The SO₂ program set a permanent cap on the total amount of SO₂ emissions allowed annually from EGUs and includes provisions for trading and banking emission allowances. The program has been phased in with the final 2010 SO₂ emissions cap set at 8.95 million tons, a level about one-half of the total SO₂ emissions from the power sector in 1980. Actual SO₂ emissions in 2009 from ARP regulated sources were about 5.7 million tons.

NO_x reductions under the ARP are achieved through a program that applies to a subset of EGUs by limiting the NO_x emission rate. 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. Actual NO_x emissions from ARP-regulated EGUs in 2009 were about 2.0 million tons.

The Clean Air Interstate Rule (CAIR), as promulgated in March 2005, was intended to permanently lower SO₂ and NO_x emissions in the eastern United States. The rule was designed to continue the progress achieved under the ARP and NBP. CAIR 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 CAIR annual and ozone season NO_x requirements began in 2009. The requirement to hold allowances in the SO₂ program began on January 1, 2010. On July 6, 2010, EPA proposed the Transport Rule, which, if finalized as proposed, will replace the CAIR. The Transport Rule, as proposed, would require 31 states and the District of Columbia to achieve additional reductions in power plant SO₂ and NO_x emissions. EPA further expects that by 2014, the Transport Rule and other state and EPA actions would reduce power plant SO₂ emissions by 71 percent and NO_x emissions by 52 percent from 2005 levels.

CASTNET was developed from 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 2009, included 84 monitoring stations at 82 site locations throughout the contiguous United States, Alaska, and Canada. 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, which are estimated 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 may also provide input to ecological models, such as the Model of Acidification of Groundwater in Catchments (MAGIC).

This report summarizes CASTNET monitoring activities and the resulting concentration and deposition data collected over the 20-year period from 1990 through 2009. 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 National Park Service (NPS)-sponsored CASTNET sites. Fully validated data are available approximately 10 months following collection. NPS data collected during 2009 from the NPS-sponsored CASTNET sites and the NPS Gaseous Pollutant Monitoring Program are available in the NPS 2009 Annual Data Summary Report (Ray, 2010) and the NPS 2009 Data Quality Assurance Report (NPS, 2010).

Cooperating Networks

CASTNET monitors air quality and deposition in cooperation with other national networks.

- ❖ **National Atmospheric Deposition Program (NADP)** operates:
 - 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.
 - Mercury Deposition Network (MDN)**, which operates samplers to measure mercury in precipitation. MDN samplers are operated at several CASTNET sites.
 - Ammonia Monitoring Network (AMoN)**, a pilot network, which operates triplicate passive ammonia (NH₃) samplers at about 20 locations. AMoN, in operation for two years, provides information on 2-week average NH₃ concentrations.
 - Atmospheric Mercury Network (AMNet)**, which measures atmospheric concentrations of gaseous oxidized, particulate-bound, and elemental mercury at about 20 locations in the United States and Canada in order to estimate dry and total mercury deposition.Visit NADP's Web site for more information: <http://nadp.isws.illinois.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. Visit CAPMoN's Web site <http://www.ec.gc.ca/rs-mn/default.asp?lang=En&n=752ce271-1> for more information.
- ❖ **Interagency Monitoring of Protected Visual Environments (IMPROVE)** measures aerosol pollutants at or near more than 30 CASTNET sites. IMPROVE measures particulate air pollutants that affect visibility. For more information on IMPROVE, see <http://vista.cira.colostate.edu/IMPROVE/>.

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.

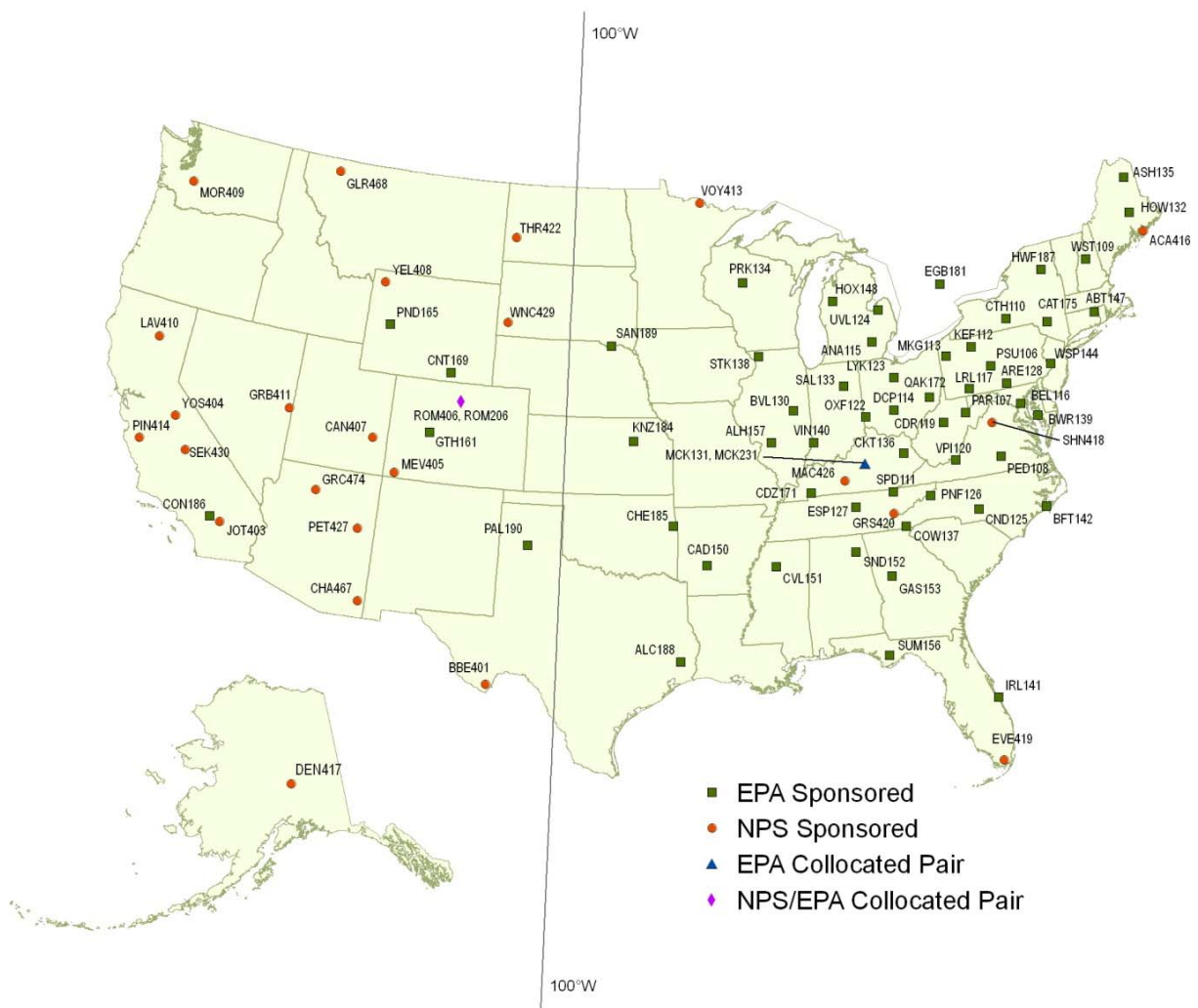
Although EPA and NPS are the primary sponsors of CASTNET, other organizations, Native American tribes, universities, and government agencies play a role in sponsoring individual CASTNET sites. These co-sponsors provide in-kind services that support the overall operation of a site, including site operation, 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 1-1 shows the locations of CASTNET monitoring sites as of December 2009. 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 1-1 CASTNET Sites as of December 2009



CASTNET Reference Sites

Maps of pollutant concentrations and deposition fluxes across the United States are presented in Chapters 2 through 4. In addition, measurements from 34 CASTNET eastern reference sites (Figure 1-2) were analyzed for each pollutant in order to determine trends in concentrations and 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. Trends in pollutant concentrations measured at CASTNET western reference sites (Figure 1-3) are also presented. The 17 western reference sites have been operating since at least 1996.

The data from the 34 eastern reference sites were aggregated and then presented in Chapters 2 through 4 using box plots for the 20-year period 1990 through 2009. Data from the 17 western reference sites were aggregated and then presented using box plots for the period 1996 through 2009.



Speedwell, TN (SPD111)

Figure 1-2 CASTNET Eastern Reference Sites



Figure 1-3 CASTNET Western Reference Sites



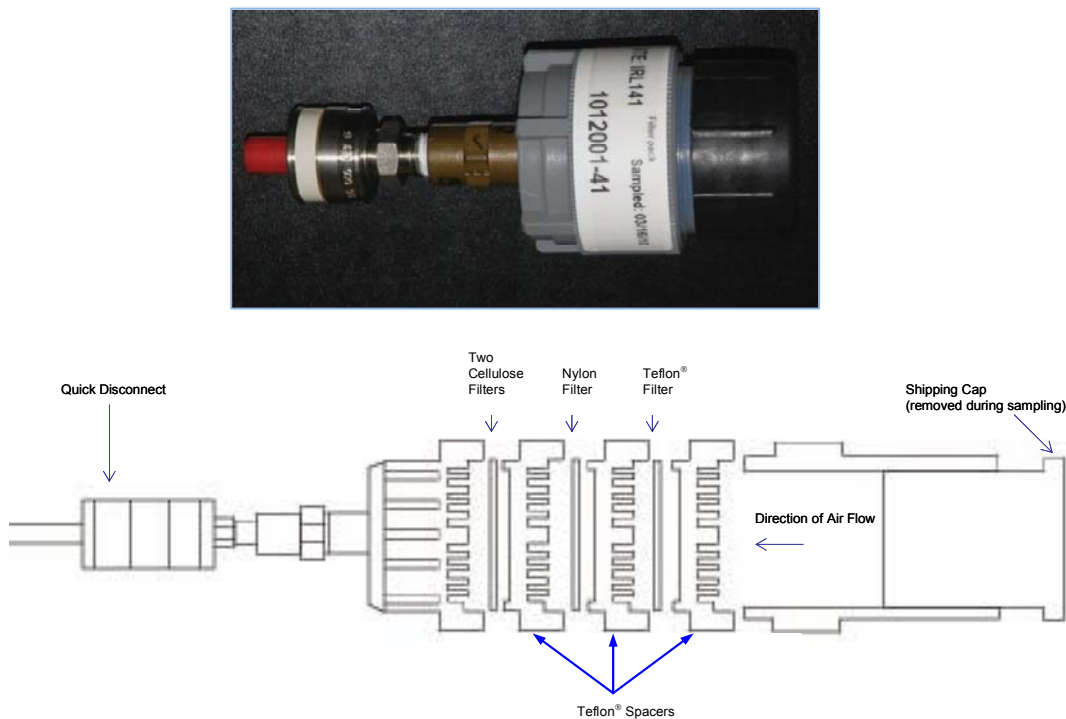
Measurements Collected at CASTNET Sites

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 concentrations was selected as the basic sampling strategy. Over the course of the seven days, air is drawn at a controlled flow rate through an open-face 3-stage filter pack (Figure 1-4) 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-impregnated cellulose filters. The filter pack is changed out each Tuesday and shipped to the analytical chemistry laboratory for analysis.

CASTNET Ambient Measurements

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

Figure 1-4 Three-Stage Filter Pack



Cellulose = Gaseous: SO_2
 Nylon = Gaseous: HNO_3 , SO_2
 Teflon = Particulate: SO_4^{2-} , NO_3^- , NH_4^+ , K^+ , Ca^{2+} , Mg^{2+} , Na^+ , Cl^-

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) and also for ammonium (NH_4^+) by the automated indophenol method with the Bran+Luebbe AutoAnalyzer 3. Additionally, Teflon filter extracts are 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 7300 Dual View spectrometer. The cellulose filter extracts are analyzed for SO_2 as SO_4^{2-} using IC. The nylon filter extracts are analyzed via IC for nitric acid (HNO_3) as nitrogen and for SO_2 as SO_4^{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 pollutant concentrations, CASTNET sites collect hourly meteorological measurements, which are used as input to the MLM, a numerical model used for estimating dry deposition to ecosystems from the atmospheric boundary layer. The meteorological data are also used by other investigators for hydrogeochemical and ecosystem modeling. The O_3 and meteorological measurements are recorded continuously and archived as hourly averages.

CASTNET Meteorological Measurements

- Temperature:
 - Temperature (at 2 and 9 meters)
 - Delta temperature (difference between 2 and 9 meters)
- Relative Humidity
- Solar Radiation
- Winds:
 - Speed
 - Direction
 - Sigma theta (standard deviation of direction)
- Precipitation
- Surface Wetness



Edgar Evins State Park, TN (ESP127)

One-Hour Sulfur Dioxide Concentrations Measured at Beltsville, MD and Cherokee Nation, OK

Hourly average trace-level concentrations of carbon monoxide (CO), total reactive oxides of nitrogen (NO_x), and SO₂ have been collected at the Beltsville, MD (BEL116) CASTNET site since spring 2005. The continuous monitors were initiated to establish baseline concentrations of these trace-level pollutants and to help analyze chemical and meteorological measurements. The Cherokee Nation established an NCore monitoring station in Stilwell, OK (CHE185) in July 2008, which also measures trace-level pollutants.

On June 2, 2010, EPA strengthened the primary National Ambient Air Quality Standards (NAAQS) for SO₂. EPA replaced the existing annual and 24-hour primary SO₂ standards with a new 1-hour SO₂ standard set at 75 parts per billion (ppb) to better protect public health by reducing human exposure to high short-term (5 minutes to 24 hours) concentrations of SO₂. The new standard is based on the 3-year average of the 99th percentile of the yearly distribution of daily maximum 1-hour SO₂ concentrations. For more information on the revised standard visit the EPA Web site <http://www.epa.gov/air/sulfurdioxide/pdfs/20100602fs.pdf>.

Assessing Compliance with the New Standard

SO₂ measurements are collected on an hourly basis. For compliance purposes:

- ❖ The daily maximum 1-hour SO₂ concentration is recorded for each day.
- ❖ For each year, the 99th percentile of the yearly distribution of daily maximum 1-hour values is calculated.
- ❖ These annual 99th percentile values are averaged over rolling 3-year periods.
- ❖ If the 3-year average of these 99th percentile values exceeds 75 ppb, the area is designated as a “nonattainment area” and is held accountable for reaching the attainment status.

EPA estimates that, based on 2007–2009 data, 59 of 249 monitored counties would have exceeded the new SO₂ NAAQS. EPA plans to use 2009 to 2011 or later data to designate areas as nonattainment. EPA is scheduled to make final determination for area designations no later than June 2012.

2009 SO₂ Concentration Data from BEL116, MD and CHE185, OK

SO₂ concentration data from CASTNET sites BEL116, MD and CHE185, OK can be used to evaluate the compliance assessment procedure. Figure 1a presents the cumulative frequency distribution of daily maximum 1-hour SO₂ concentrations for 2009 for BEL116, and Figure 1b presents SO₂ data from CHE185. The 99th percentile values are below the 75 ppb NAAQS. The figures present data from 2009, only. Compliance with the NAAQS must be evaluated with 3-year averages of 99th percentile values.

Figure 1a BEL116 Cumulative Frequency for 2009

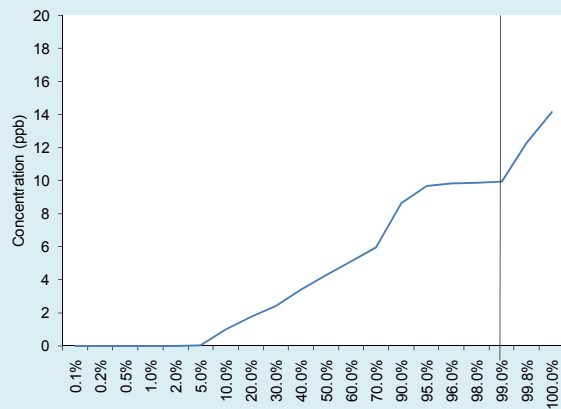
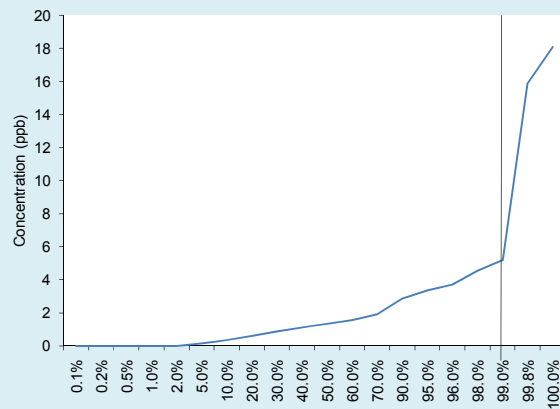


Figure 1b CHE185 Cumulative Frequency for 2009



Modeling 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 1-5. 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 summarized 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 1-5 shows the relationships among the various resistances and illustrates the meteorological and other data that are required as model input.

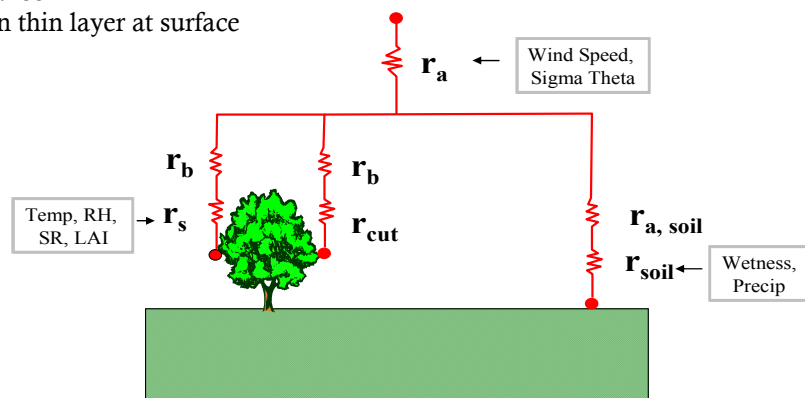
Figure 1-5 Multi-Layer Model

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

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

Resistances

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



Atmospheric Mercury Network

Mercury (Hg) is commonly emitted to the atmosphere through power generation from coal-fired power plants, the single largest source of anthropogenic Hg in the United States. Other sources include incineration of waste containing products with Hg, combustion of coal and petroleum, metal reprocessing facilities, cement kilns, and natural sources such as volcanoes. Atmospheric deposition is the dominant source of Hg to surface waters and wetlands over most of the globe. Methylmercury (MeHg) is produced from ionic Hg by microorganisms in aquatic systems. It is not readily eliminated from organisms, and it is biomagnified in aquatic food chains. Because fish and animals can accumulate MeHg faster than they eliminate it, fish and animals consume higher concentrations of Hg at each successive level of the food chain. Small environmental concentrations of MeHg can thus readily accumulate to potentially harmful concentrations in fish, fish-eating wildlife, and humans.

Despite the widespread Hg pollution and its serious health risks, a comprehensive and integrated mercury monitoring network does not exist. However, considerable progress has been made in the design of a national mercury monitoring program. In fall 2009, the NADP established the Atmospheric Mercury Network (AMNet) to measure Hg species concentrations in ambient air. Data from the AMNet are used to estimate dry deposition, assess regulatory emission reduction programs, evaluate atmospheric models, and estimate long-term trends. The NADP's MDN has been monitoring wet deposition of Hg at 113 sites, including approximately 25 sites that measure MeHg in precipitation, since 1996. The measurement of wet deposition at MDN sites and the modeled dry deposition estimates at AMNet sites will enable improved estimates of total Hg deposition. All AMNet sites operate a Tekran analyzer, which provides a continuous analysis of gaseous oxidized Hg, elemental Hg, and particulate-bound Hg. Chemical analysis is based on cold vapor atomic fluorescence spectroscopy.

AMNet Goals

The NADP established three major goals for AMNet, including determining the status and trends in concentrations of atmospheric Hg fractions (gaseous oxidized, particulate-bound, and elemental) in select locations; offering high-quality meteorological measurements to estimate dry and total deposition of atmospheric Hg to aquatic ecosystems and other areas of interest on the local, regional, and global scale; and providing data for atmospheric Hg model development, validation, and improvement (NADP/AMNet, 2010).

Figure 1c shows the 20 AMNet sites that are currently operating in the United States and Canada. Most sites are collocated with MDN sites and include meteorological measurements. Dual sampling systems are operated at Beltsville, MD (AMNet sites MD98 and MD99: collocated with NADP/MDN site MD99 and CASTNET site BEL116), Grand Bay National Estuarine Research Reserve (NERR), MS (AMNet sites MS12 and MS99: collocated with NADP/MDN site MS12) and Rochester, NY (AMNet sites NY43 and NY95: collocated with NADP/MDN sites NY43 and NY95, respectively).

Table 1a lists the AMNet sites, their locations, and sponsoring organizations. Information about AMNet can be found on the NADP Web site (<http://nadp.isws.illinois.edu/amn>). The Web site discusses the sampling system and related information. It includes a network description, a list of network participants, site information, and network contacts. Mercury concentration data will be posted on the Web site when they are available.

Figure 1c Atmospheric Mercury Network Sites



Table 1a Atmospheric Mercury Network Site Locations and Sponsors

Site	Site ID	Name	Location	Latitude	Longitude	Organization
1	MD99	Beltsville	Maryland	39.0284	-76.8172	NOAA/EPA
	MD98	Beltsville Collocated				
2	MD08	Piney Reservoir	Maryland	39.7053	-79.0122	State MD
3	MS12	Grand Bay NERR	Mississippi	30.4294	-88.4277	NOAA
	MS99	Grand Bay NERR Collocated				
4	NJ05	Brigantine	New Jersey	39.4020	-74.3790	State NJ
5	NJ32	Chester	New Jersey	40.7876	-74.6763	State NJ
6	NJ54	Elizabeth Lab	New Jersey	40.6414	-74.2084	State NJ
7	NJ30	New Brunswick	New Jersey	40.4728	-74.4225	State NJ
8	NY20	Huntington Wildlife Forest	New York	43.9731	-74.2232	Clarkson U.
9	NY06	NYC	New York	40.7146	-74.0058	State NY
10	NY43	Rochester	New York	43.1544	-77.6160	Clarkson U.
11	NY95	Rochester Collocated	New York	43.1544	-77.6160	State NY
12	OH02	Athens	Ohio	39.3000	-82.1167	Ohio U.
13	OK99	Stilwell	Oklahoma	35.7514	-94.6717	Cherokee Nation
14	VT99	Underhill	Vermont	44.5283	-72.8689	Ecosystems Res. Gr.
15	WV99	Canaan Valley Institute	West Virginia	39.0636	-79.4222	NOAA
16	UT96	Antelope Island	Utah	41.0467	-112.0248	U Utah
17	UT97	Salt Lake City	Utah	40.7118	-111.9609	State Utah
18	NH06	Thompson Farm	New Hampshire	43.1100	-70.9500	U New Hampshire
19	NU15	Alert	Nunavut	54.0000	-90.0000	Env. Canada
20	NS02	Kejimikujik	Nova Scotia	44.4300	-65.2100	Env. Canada

Note: Collocated CASTNET sites are highlighted in yellow.

Source: NADP/AMNet, 2010

Sulfur Dioxide and Nitrogen Oxides Emissions

Title IV of the 1990 CAAA established the ARP, which was designed to reduce the 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 2009 progress report (EPA, 2010a). The report summarizes 2009 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 annually by all regulated EGUs in the contiguous United States. The program, which began in 1995, has required incremental reductions in SO₂ emissions yearly with the final 2010 SO₂ emissions 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. 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 O₃ formation is high. The CAIR NO_x ozone season trading program replaced the NBP in 2009. The CAIR annual NO_x controls were also instituted in 2009. Although CAIR was remanded back to EPA in 2008, the CAIR programs continue in effect temporarily. If the Transport Rule is finalized as proposed, the CAIR programs would continue in effect through the completion of all 2011 control period activities before being replaced.

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 emissions and from 1996 through 1999 for NO_x emissions. Phase II for both pollutants began in 2000. In 2009, the SO₂ Phase II requirements affected 3,572 operating units. The Phase II NO_x requirements applied to 960 of those operating units that exceed 25 megawatts and burned coal between 1990 and 1995 (EPA, 2010a). Under the ARP's emission reduction requirements, total SO₂ emissions from ARP sources were 5.7 million tons in 2009; total NO_x emissions from ARP sources were 2.0 million tons.

Figure 1-6 presents state-by-state total annual SO₂ emissions for Phase I and Phase II electric utility plants for five years (1990, 1995, 2000, 2005, and 2009). 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 in 1990 realized the largest reductions. Annual NO_x emissions by state are depicted in Figure 1-7 for the same five years. States with the highest NO_x emissions also had the most significant declines. Every state east of the Mississippi River experienced a significant decline in annual NO_x emissions from 2000 to 2009. For example, NO_x emissions from power plants in more than 20 eastern states declined by at least 65 percent from 2000 to 2009. Also, EGU NO_x emissions in Texas and Missouri declined by 65 percent or more from 2000 to 2009.

Figure 1-6 Annual Utility SO₂ Emissions (Phase I and Phase II Plants only)

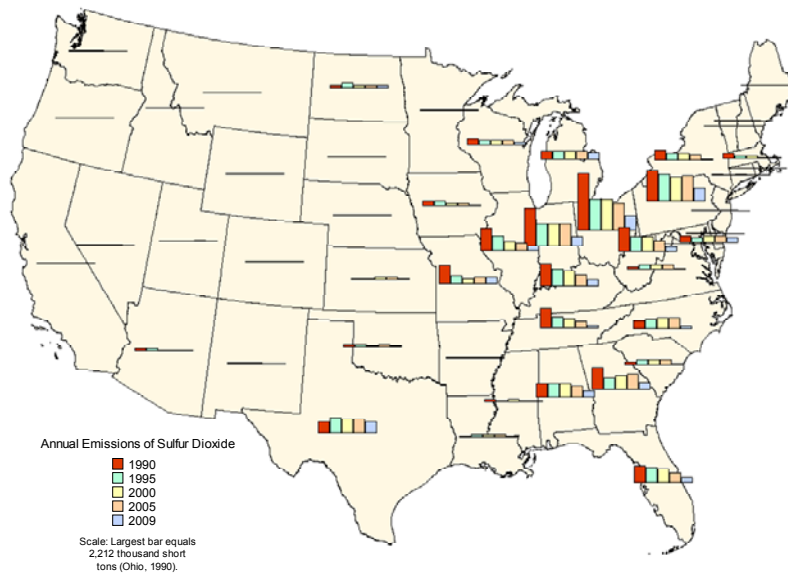
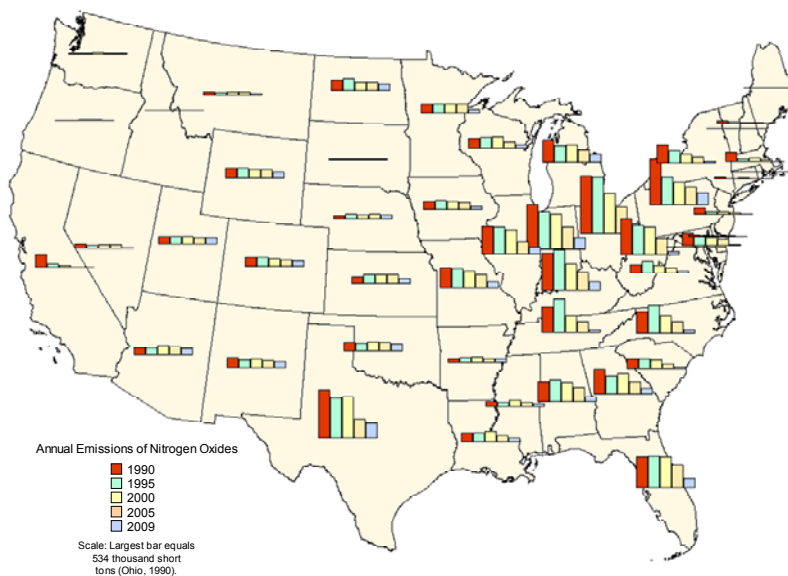


Figure 1-7 Annual Utility NO_x Emissions (Phase I and Phase II Plants only)



2009 Significant Events

January
<p>The span for O₃ multipoint calibrations changed from 400 ppb to 450 ppb. The checkpoints are now 0 ppb, 450 ppb, 300 ppb, 200 ppb, 100 ppb, and 60 ppb.</p> <p>As of January 2009, the field calibration accuracy criterion for relative humidity changed to ± 10 percent for all relative humidity measurements.</p>
March
<p>MACTEC tied for first place with two of 36 other laboratories in the intercomparison proficiency test (PT) Study 0093 with Environmental Canada. For the U.S. Geological Survey 2008 Interlaboratory Comparison samples, MACTEC achieved the target value or within less than 5 percent for all of the 144 samples analyzed.</p>
May
<p>Shelter temperature data from EPA-sponsored CASTNET sites were included in the publically-available CASTNET database hosted by EPA.</p>
July
<p>The Campbell Scientific CR3000 data logger rollout was completed. With the exception of the Cherokee Nation, OK (CHE185) site, all EPA-sponsored CASTNET sites operate CR3000 data loggers. The CHE185, OK site will continue to operate an Environmental Systems Corporation data logger, which is also used at all NPS-sponsored CASTNET sites.</p>
August
<p>The CASTNET IV contract (EP-W-09-028) officially began on August 11, 2009.</p> <p>By early August 2009, all EPA-sponsored CASTNET sites that measure O₃ (see Appendix A) were uploading hourly O₃ and meteorological data to AIRNow. NPS-sponsored CASTNET sites have been included in AIRNow since 2004.</p> <p>Beginning on August 11, 2009, precision measurement criteria for filter pack concentrations changed from a mean absolute relative percent difference (MARPD) of 5.0 percent for all collocated CASTNET filter pack measurements (except for particulate NH₄⁺, which had a criterion of 10.0 percent MARPD) to a single MARPD value of 20.0 percent.</p>

NPS produced a biannual CASTNET newsletter called *The Monitor* that contained information about some of the changes and important events affecting CASTNET operations. Recently, NPS replaced *The Monitor* by publishing an e-mail newsbrief called “Eye on Air Quality,” which will be published each April, July, and October. This newsbrief can be found at <http://ard-request.air-resource.com/project/documents.aspx>. For an electronic library of previous newsletters, go to <http://www.nature.nps.gov/air/Pubs/theMonitor.cfm>.

Chapter 2: Atmospheric Concentrations

Weekly average concentrations of SO₂, SO₄²⁻, HNO₃, NO₃⁻, NH₄⁺, Cl⁻, and four earth metals were measured at 84 CASTNET monitoring stations using 3-stage filter packs. Measured annual mean concentrations of SO₂ and SO₄²⁻ have decreased steadily since 1990 with a sharp decline since 2005. Concentrations of total nitrate (HNO₃ + NO₃⁻) showed little change from 1990 until 2000 when they began to decline in response to NO_x emission reduction programs. Trends in mean annual SO₂, SO₄²⁻, total NO₃⁻, and NH₄⁺ concentrations aggregated over the 34 eastern reference sites are shown over the period 1990 through 2009 and for 17 western reference sites for the period 1996 through 2009 using box plots for each year. All four parameters measured at the eastern sites declined over the 20-year period. SO₂ concentrations measured at the 17 western reference sites also declined over the 14 years. Concentrations of the other three pollutants measured at the western sites did not change significantly but did decline in 2009.

Maps of annual mean concentrations of SO₂, SO₄²⁻, total nitrate (HNO₃ + NO₃⁻), and NH₄⁺ are presented in this chapter. Additional maps are provided in CASTNET quarterly reports (MACTEC 2009a; 2009c; 2010a; 2010c). The concentration shading used in the maps was prepared using an algorithm based on inverse distance weighting and the scales specified on each map. In addition, trends in concentrations over the 20-year period (1990 through 2009) were derived from the 34 eastern CASTNET reference sites (Figure 1-2) 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 1-3) for the period 1996 through 2009.

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 SO₂ in the United States. As discussed in Chapter 1, SO₂ is a criteria pollutant regulated by the NAAQS. To be compliant under the new SO₂ NAAQS, the 3-year average of the 99th percentile of the yearly distribution of daily maximum 1-hour SO₂ concentrations must be at 75 ppb [215 micrograms per cubic meter (µg/m³) of air]. SO₂ gas reacts in the atmosphere to form various acidic compounds including sulfuric acid (H₂SO₄), a significant contributor to acid rain, and ammonium sulfate [(NH₄)₂SO₄], a major component of fine particle matter (PM_{2.5}). Fine particles are also regulated by the NAAQS since PM_{2.5} adversely affects health and contributes to degradation of atmospheric visibility.

Annual mean SO₂ concentrations recorded in 2009 are presented in Figure 2-1. Only two sites in the eastern United States measured mean concentrations greater than 5.0 µg/m³ of air. The sites are Quaker City, OH (QAK172), which measured the highest concentration in the network with a value of 8.3 µg/m³, and Laurel Hill State Park, PA (LRL117), which measured a concentration of 5.5 µg/m³. SO₂ concentrations recorded in 2009 were significantly lower than SO₂ levels measured in previous years. For example, in 2008, concentrations greater than 5.0 µg/m³ were measured in a six-state region centered on the Ohio River Valley. Only two western sites (Figure 1-1) measured a 2009 annual mean SO₂ concentration equal to 1.0 µg/m³. The western sites are Palo Duro Canyon State Park, TX (PAL190) and Theodore Roosevelt National Park, ND (THR422).

Figure 2-1 Annual Mean SO₂ Concentrations (µg/m³) for 2009

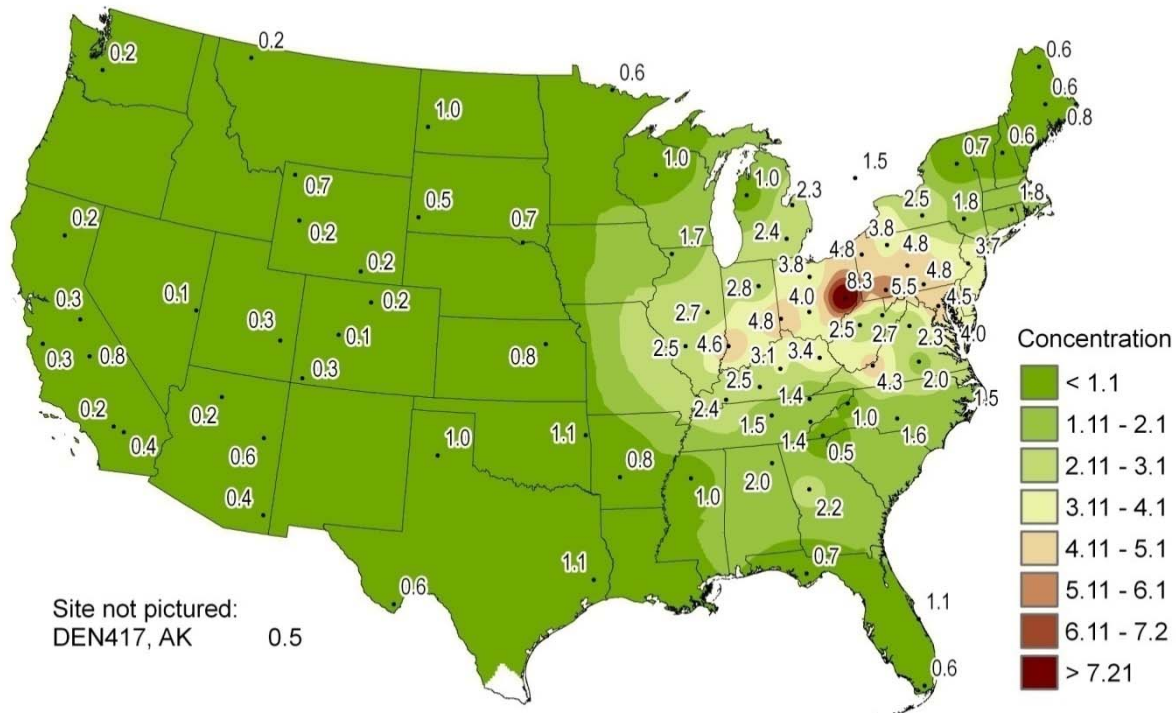
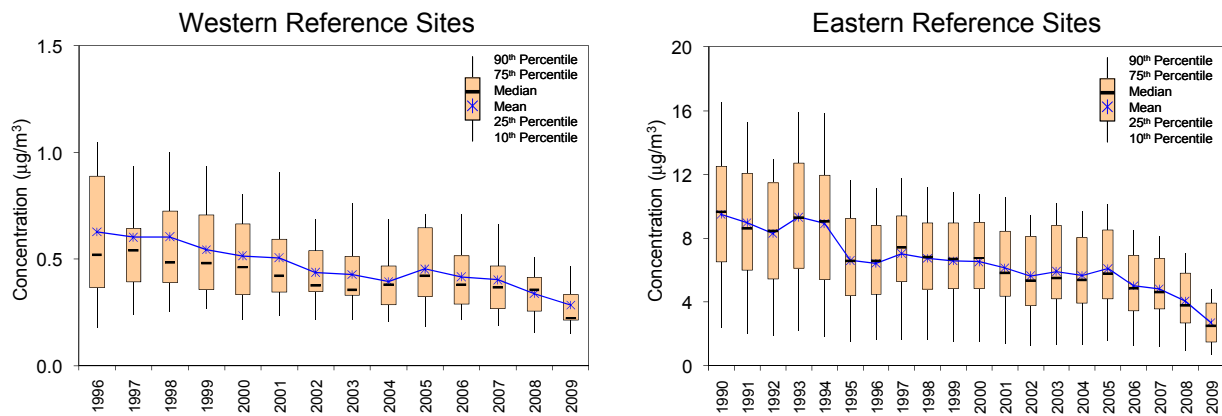


Figure 2-2 shows box plots of annual mean SO₂ concentrations aggregated over the 34 eastern reference sites from 1990 through 2009 (right side) and the 17 western reference sites from 1996 through 2009 (left side). The y-axes on the western and eastern plots have different scales since concentrations measured at the western CASTNET sites are generally much lower than concentrations measured at the eastern sites. The box plots for the eastern sites show a downward trend. A significant reduction in ambient SO₂ was recorded in 1995 at the beginning of the ARP. A sharp decline was also recorded from 2005 through 2009. Three-year mean concentrations for 1990–1992 and 2007–2009 were 8.9 µg/m³ and 3.9 µg/m³, respectively. This change constitutes a 56 percent reduction in 3-year mean SO₂ concentrations between the two time periods. The 2009 mean level of 2.7 µg/m³ was the

lowest mean value measured by the eastern reference sites in the history of the network and represents a dramatic decline over the past four years from the 2005 concentration of $6.1 \mu\text{g}/\text{m}^3$.

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 1996–1998 and 2007–2009 were $0.61 \mu\text{g}/\text{m}^3$ and $0.34 \mu\text{g}/\text{m}^3$, respectively. This change constitutes a 44 percent reduction in 3-year mean SO_2 concentrations over the 14 years. The aggregated mean SO_2 concentrations from the western reference sites were typically more than 10 times lower than mean concentrations measured at the eastern reference sites.

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



Georgia Station, GA (GAS153)

Spatial Variability of Western Nitrogen Concentrations

CASTNET measures weekly concentrations of the nitrogen containing compounds HNO_3 , NH_4^+ , and NO_3^- . The dry deposition of nitrogen to natural areas is known to acidify soils, induce biological species changes, and act as a fertilizer that helps promote the growth of exotic plant species. In the western United States, CASTNET stations are typically 100-200 miles apart, often with mountainous terrain between sites. Although the concentrations and deposition fluxes of nitrogen compounds in the West are generally much lower than in the East, western desert and alpine ecosystems are sensitive to changes in nitrogen deposition.

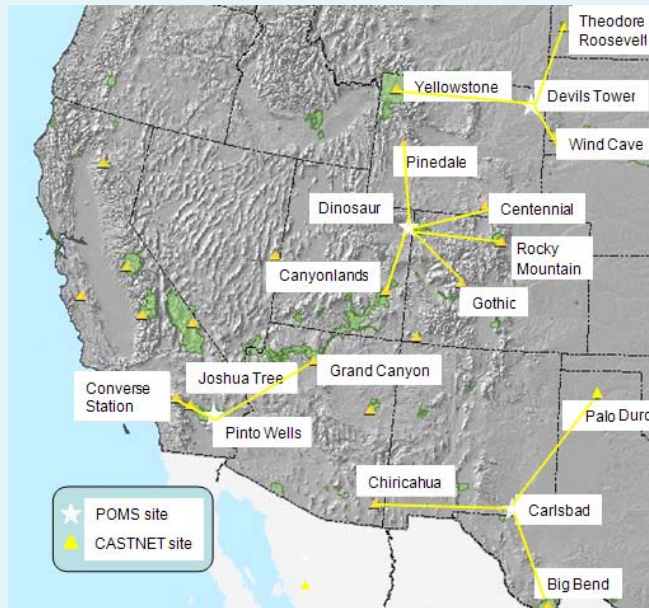
Taking into account the complexity and variability of western landscapes and terrain, how dense a network of sites is needed in the West to adequately capture the rural spatial variability of the dry chemistry species? An independent NPS program consisting of a network of 18-20 portable O_3 monitoring systems (POMS) that operate during the summer months mostly at western locations provides some insight into this question. Four of the POMS have masts and enough solar power to run CASTNET filter pack systems. Weekly samples are taken on the CASTNET schedule. The filter packs are prepared and analyzed by the CASTNET laboratory to obtain results comparable to those from nearby CASTNET stations. The POMS season runs from May through September to correspond with higher O_3 levels. The POMS sites with CASTNET filter pack systems provide “fill-in” measurements to help gauge areas that may need a permanent monitor.

Table 2a Comparison of Total Nitrogen (as N) Concentrations ($\mu\text{g}/\text{m}^3$) at POMS and Nearby CASTNET Sites

SITE ID	Site	Years	Miles	Mean	Mean	Max	Max
			Distance	Total_N	% Diff	Total_N	% Diff
CAV100	Carlsbad Caverns National Park, NM (POMS)	4	0	1.2		4.1	
BBE401	Big Bend National Park, TX (CASTNET)	5	214	1.0	-16.5	5.9	44
DET100	Devils Tower National Monument, WY (POMS)	2	0	0.6		0.9	
WNC429	Wind Cave National Park, SD (CASTNET)	5	93	0.8	19	1.4	56
DIN100	Dinosaur National Monument, CO & UT (POMS)	5	0	0.7		1.4	
CAN407	Canyonlands National Park, UT (CASTNET)	5	140	0.7	5	1.0	-29
JOT100	Joshua Tree National Park (Pinto Wells), CA (POMS)	4	0	1.6		3.5	
JOT403	Joshua Tree National Park, CA (CASTNET)	5	57	2.0	55	3.4	-7
CON186	Converse Station, CA (CASTNET)	5	88	2.2	94	4.2	50
ROM406	Rocky Mountain National Park, CO (NPS) (CASTNET primary site)	5	0	0.6		1.3	
ROM206	Rocky Mountain National Park, CO (EPA) (CASTNET collocated site)	5	0	0.6		1.1	

Source: J.D. Ray

Figure 2a Western CASTNET Sites and POMS Locations



Source: J.D. Ray

Table 2a compares the mean and maximum total nitrogen (Total_N) content of the HNO_3 , NH_4^+ , and NO_3^- concentrations measured at four POMS and nearby CASTNET sites, plus the CASTNET collocated site pair at Rocky Mountain National Park, CO (ROM406/ROM206) during the 5-month sampling season. Both samplers at ROM406/ROM206, CO, where the separation between sampling sites is about 50 feet, measured a mean concentration of $0.6 \mu\text{g}/\text{m}^3$. The recent (Malm and Collett, 2009) Rocky Mountain Atmospheric Nitrogen and Sulfur (RoMANS) Study, a research study of high-elevation ecosystems performed over two 6-week time periods, showed that large episodic events accounted for much of the wet and dry nitrogen deposition in and near Rocky Mountain National Park. Table 2a also shows that the distances between the POMS and their nearby CASTNET sites do not explain

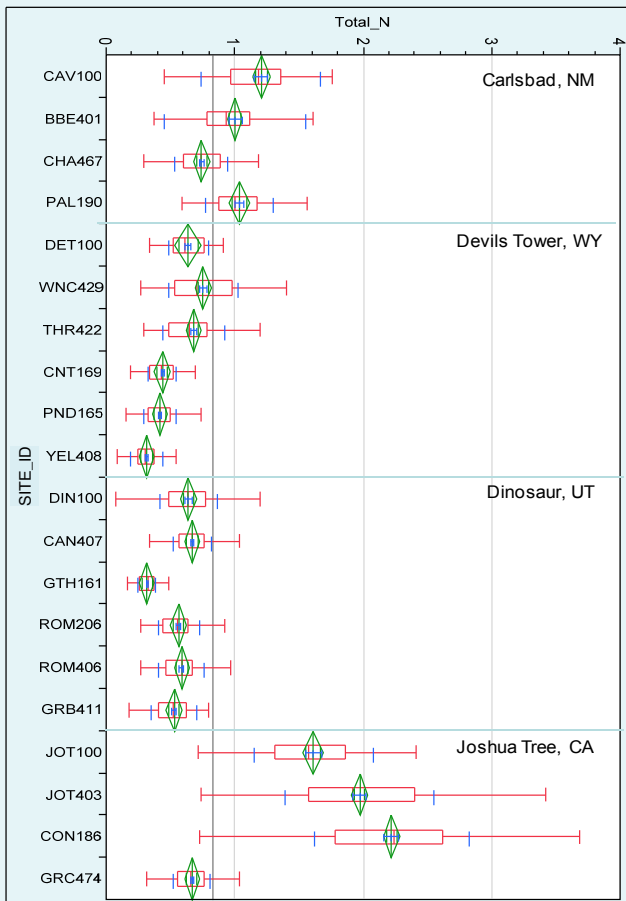
the differences in 5-month Total_N concentrations. For example, concentrations measured at Dinosaur National Monument and Canyonlands National Park show little difference (5 percent) in mean Total_N. On the other hand, the two sites in Joshua Tree National Park and Converse Station in southern California measured the highest concentrations and showed a significant increasing east to west gradient in concentrations toward Los Angeles over about the same distance.

A map of western CASTNET and POMS sites is presented in Figure 2a. The POMS and nearby CASTNET site groups are shown with the related groups linked with yellow lines.



Converse Station, CA (CON186)

Figure 2b Comparison of POMS and Nearby CASTNET Site Total Nitrogen Concentrations ($\mu\text{g}/\text{m}^3$)



Source: J.D. Ray

The sites within a broad area of Utah, Colorado, Wyoming, and the Dakotas recorded lower total nitrogen concentrations with similar means. Most of the monitors in this region had concentrations below $0.8 \mu\text{g}/\text{m}^3$ and are probably fairly well represented by the widely spaced network in those areas of the West. No attempt was made in this analysis to account for elevation differences.

More information can be gained by analyzing the other dry chemistry compounds measured on the filters along with the high weekly concentrations that indicate episodes. The Devils Tower and Dinosaur National Monuments filter pack sampler data do not add much new information to the network, despite oil and gas development near Dinosaur National Monument. There was also a question whether pollution from the Salt Lake City area was transported to the park. Since the nitrogen species and O_3 concentrations are not elevated, any impact from those source areas to Dinosaur National Monument is not detectable with the POMS O_3 and filter pack systems. The Pinto Wells site in Joshua Tree National Park, CA and the Carlsbad Caverns, NM site, however, do indicate transport from pollution source areas.

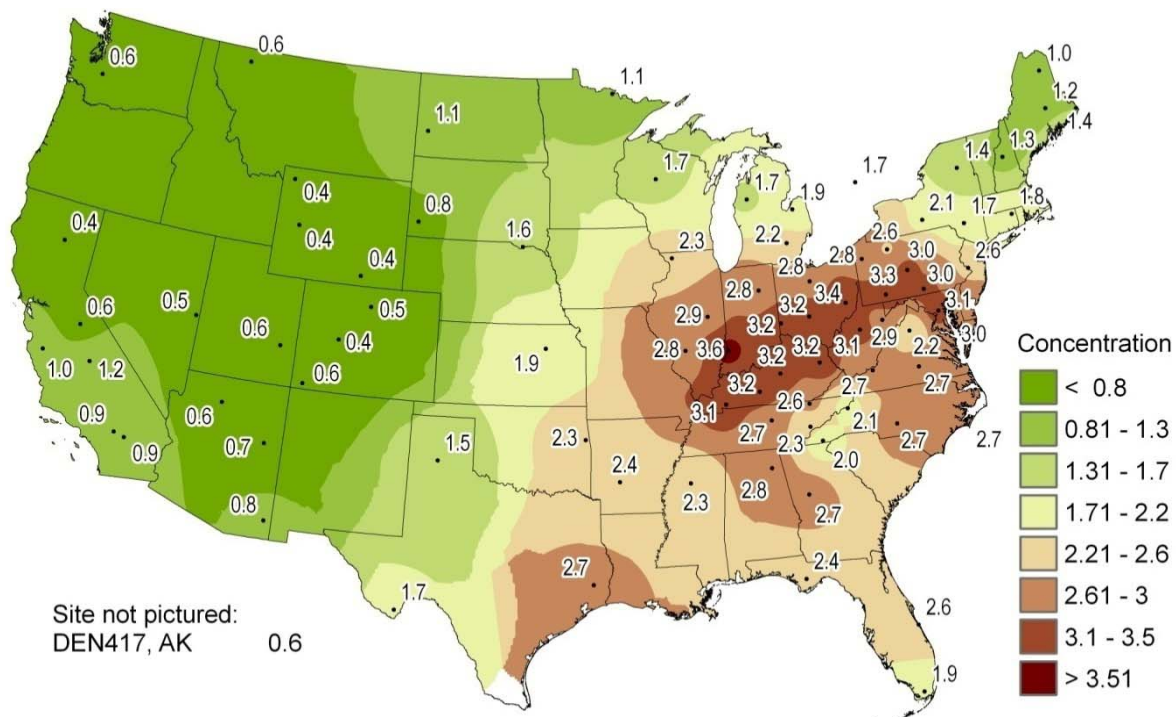
Figure 2b presents a broad statistical picture of the western Total_N data by comparing the POMS data with data from additional nearby CASTNET sites. In Figure 2b, statistical distributions of all the weekly filter data for total nitrogen are represented using box and whisker plots. The 10th, 25th, 50th, 75th and 90th percentiles are shown for each site. The sites are organized in site groups for comparison. The diamonds represent the confidence interval about the mean and are useful for comparing to nearby sites to see if the means are different. If the diamonds overlap, the mean values are not statistically significant.

Gradients in total nitrogen were observed at the western sites. The concentration gradient increases from east to west and also from north to south. The southern California sites measured the highest total nitrogen concentrations among the western sites. The Carlsbad Caverns, NM and two Texas sites observed higher nitrogen levels than the remaining sites. The data suggest that there are strong gradients associated with major source areas from Los Angeles along the southern tier to Texas that may not be fully represented spatially by the current

Particulate Sulfate

Particulate SO_4^{2-} is formed in the atmosphere through the transformation of SO_2 via both gas and aqueous (cloud) phase reactions. SO_4^{2-} typically exists in the atmosphere as $(\text{NH}_4)_2\text{SO}_4$, a major component of $\text{PM}_{2.5}$. A map of annual mean particulate SO_4^{2-} concentrations measured during 2009 is given in Figure 2-3. The map shows a fairly uniform distribution of SO_4^{2-} concentrations greater than or equal to $3.0 \mu\text{g}/\text{m}^3$ centered on the major SO_2 source region in the vicinity of the Ohio River Valley. SO_4^{2-} concentrations greater than or equal to $1.0 \mu\text{g}/\text{m}^3$ in the western region were measured at two sites in California, two in Texas, and the monitoring station at THR422, ND.

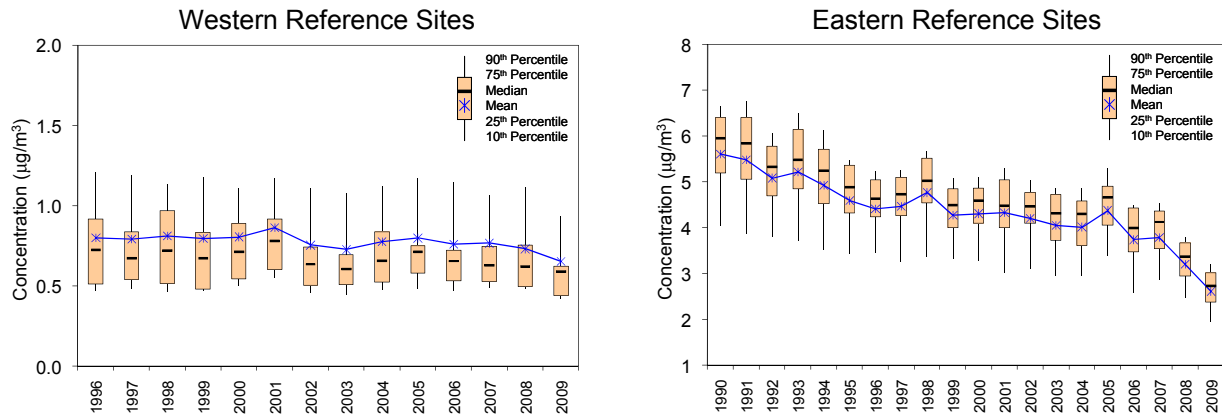
Figure 2-3 Annual Mean SO_4^{2-} Concentrations ($\mu\text{g}/\text{m}^3$) for 2009



Box plots of annual mean SO_4^{2-} concentrations from the 34 eastern reference sites from 1990 through 2009 are presented in the right half of Figure 2-4. The figure shows a substantial decline in SO_4^{2-} with some interannual variability. Like SO_2 , sulfate concentrations declined significantly from 2005 through 2009. The difference between 3-year means from 1990–1992 to 2007–2009 represents a 41 percent reduction in SO_4^{2-} from $5.4 \mu\text{g}/\text{m}^3$ to $3.2 \mu\text{g}/\text{m}^3$, respectively. The 2009 mean SO_4^{2-} level of $2.6 \mu\text{g}/\text{m}^3$ 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, although the 2009 aggregated mean concentration was the lowest recorded. Mean SO_4^{2-} 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 ($\mu\text{g}/\text{m}^3$)



Palo Duro Canyon State Park, TX (PAL190)

Ten-Year Changes in SO₂ and Total Nitrate Concentrations

Emission reductions of SO₂ and NO_x resulting from the implementation of the ARP in 1995, the NBP in 1999, and the CAIR in 2009, resulted in a dramatic improvement in air quality in the eastern United States. SO₂ and NO_x emissions are shown in Figures 1-6 and 1-7. Three-year mean SO₂ emissions from ARP units declined from 13.2 million tons in 1997–1999 to about 7.4 million tons in 2007–2009, a 44 percent reduction.

Aggregated 3-year mean SO₂ concentrations declined approximately 42 percent over that period. More recently, SO₂ emissions declined from 10.2 million tons in 2005 to 5.7 million tons in 2009. This significant decrease in emissions was produced mostly from the installation of SO₂ emission control technology because of early compliance planning to meet CAIR requirements (EPA, 2010a).

NO_x emissions from ARP units declined from a 1997–1999 mean of 5.2 tons per year to a 2007–2009 mean of 2.8 tons per year, a 46 percent reduction. Three-year mean total NO₃ concentrations declined from 3.1 to 2.1 µg/m³, a 33 percent reduction over that period. The drop in NO_x emissions from 2008 to 2009 was the largest single year decline in the history of the ARP. This large reduction in emissions can be primarily attributed to compliance with the annual and O₃ season CAIR programs for NO_x, which began in 2009 (EPA, 2010a).



Beaufort, NC (BFT142)



The maps shown in Figures 2c and 2d show 3-year mean SO₂ concentrations for 1997–1999 and 2007–2009, respectively. The maps show a significant reduction in SO₂ over the 10 years. Both the magnitude and geographic extent of high concentrations declined. For example, the 3-year mean SO₂ concentration measured at Quaker City, OH (QAK172) declined from 12.5 µg/m³ to 8.1 µg/m³.

Figure 2c Three-year Mean SO₂ Concentrations (µg/m³) for 1997–1999

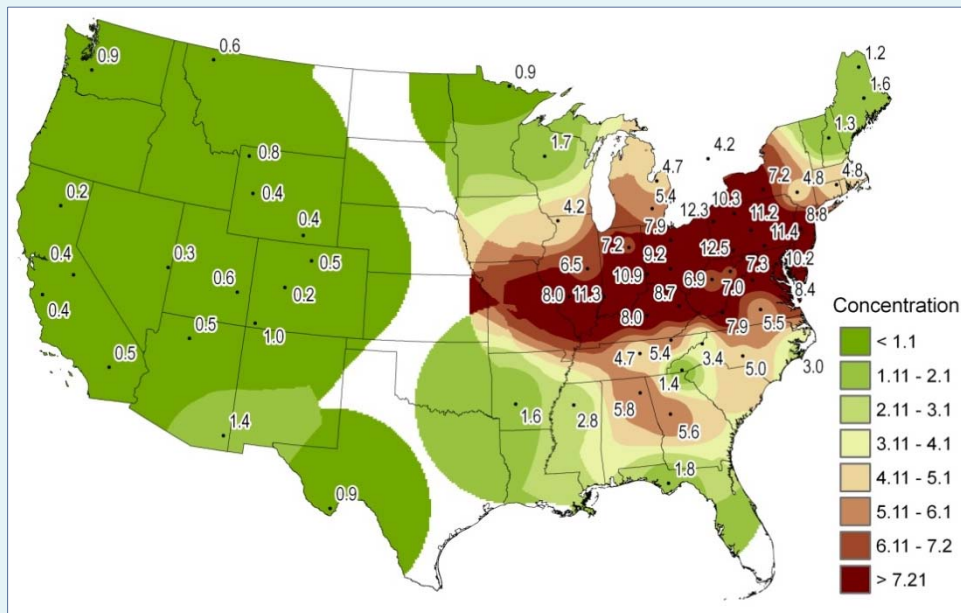
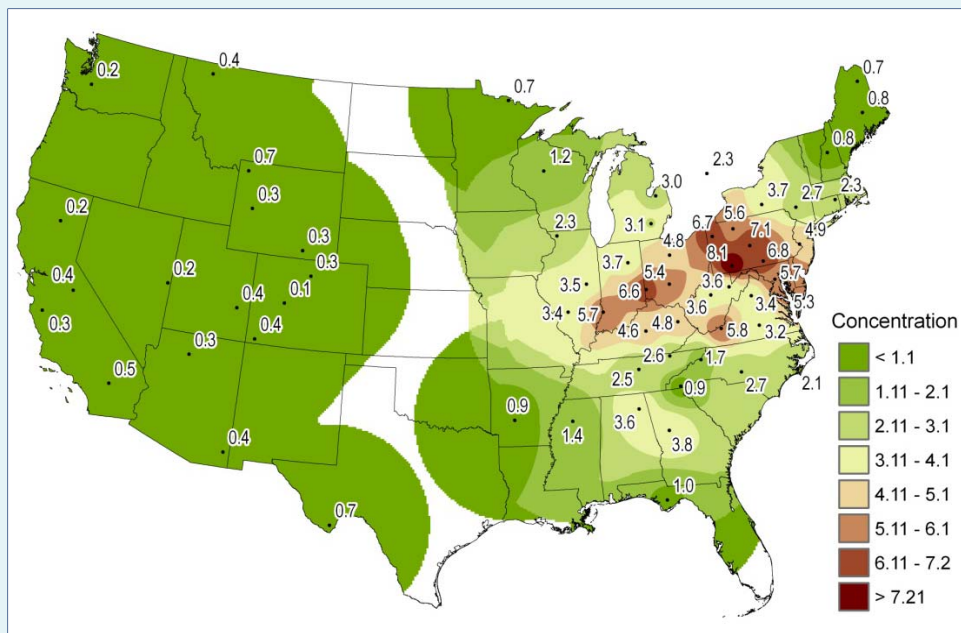


Figure 2d Three-year Mean SO₂ Concentrations (µg/m³) for 2007–2009



Maps of 3-year mean total nitrate concentrations are depicted in Figures 2e and 2f for 1997–1999 and 2007–2009. Again, both the magnitude and geographic extent of the high concentrations have been reduced.

Figure 2e Three-year Mean Total Nitrate Concentrations ($\mu\text{g}/\text{m}^3$) for 1997–1999

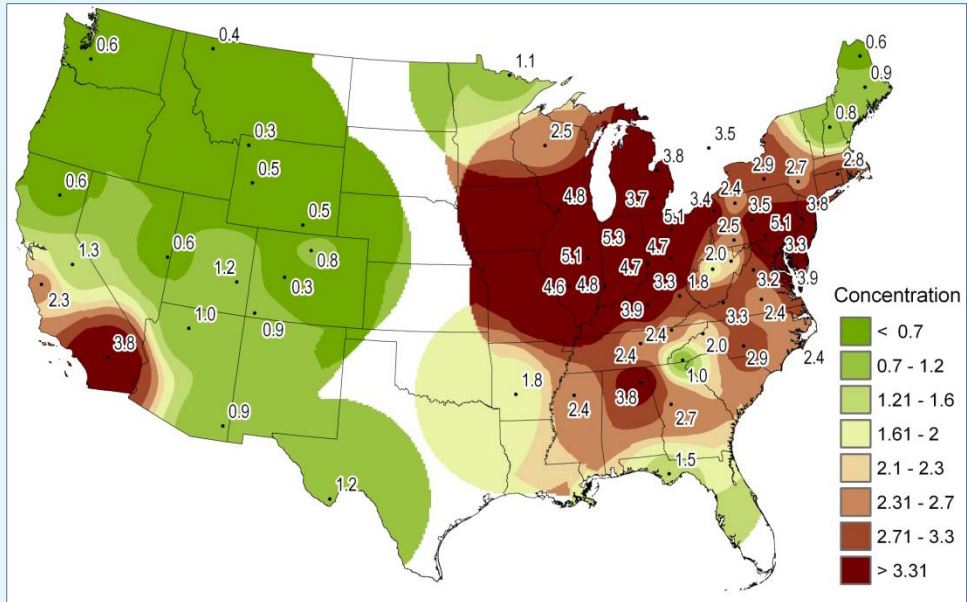
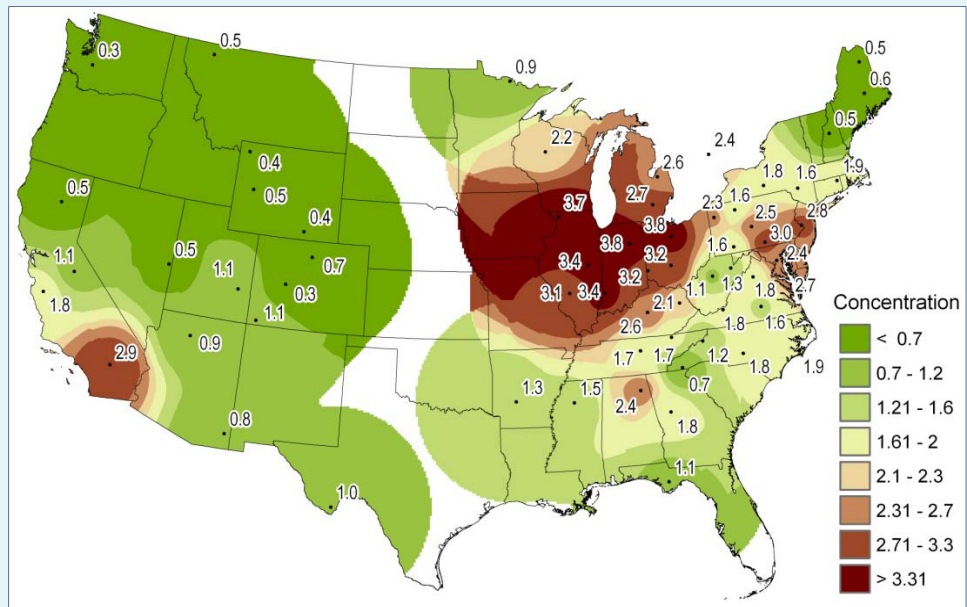


Figure 2f Three-year Mean Total Nitrate Concentrations ($\mu\text{g}/\text{m}^3$) for 2007–2009



Total Nitrate

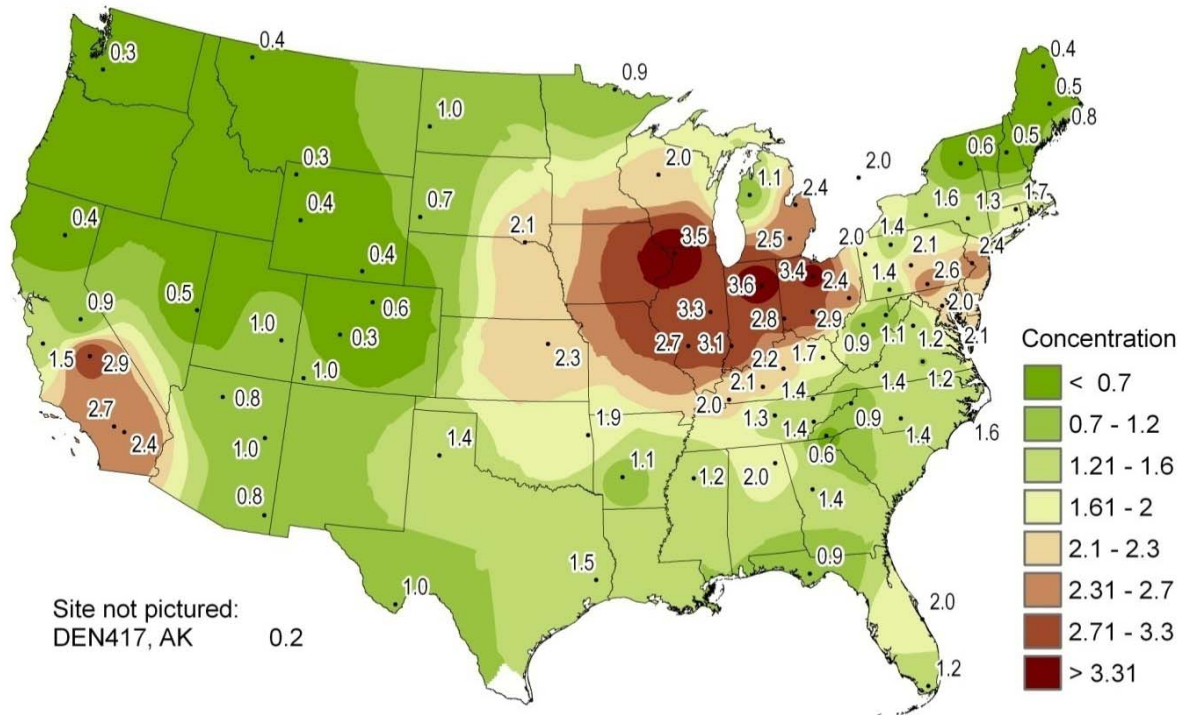
Total nitrate is the sum of HNO_3 and NO_3^- , which are formed in the atmosphere from NO_x emitted during combustion of fossil fuels. Transportation sources (e.g., automobiles and trucks) are the largest contributors of NO_x emissions in the United States. Nitric acid exists as a gas and contributes to acid deposition. Nitrate is a component of $\text{PM}_{2.5}$ in the form of ammonium nitrate and as larger particles, e.g., sodium nitrate or calcium nitrate. Figure 2-5 provides a map of annual mean total nitrate concentrations for 2009. 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 2009 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 includes the four states of Illinois, Indiana, Ohio, and Michigan. The area around Arendtsville, PA (ARE128) is the second region. The ARE128 monitor measured a concentration of $2.6 \mu\text{g}/\text{m}^3$. Two sites, Sequoia National Park (SEK430) and Converse Station (CON186), in California constitute the third region. Salamonie Reservoir, IN (SAL133) measured the highest concentration ($3.6 \mu\text{g}/\text{m}^3$) in the network.

Box plots of total nitrate levels for 1990 through 2009 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 $2.1 \mu\text{g}/\text{m}^3$ from 1990–1992 to 2007–2009, producing a 30 percent reduction in total nitrate. Total nitrate levels measured at the eastern reference sites declined from a mean value of $3.1 \mu\text{g}/\text{m}^3$ in 2000 to a mean value of $1.8 \mu\text{g}/\text{m}^3$ in 2009, which is the lowest in the history of CASTNET.



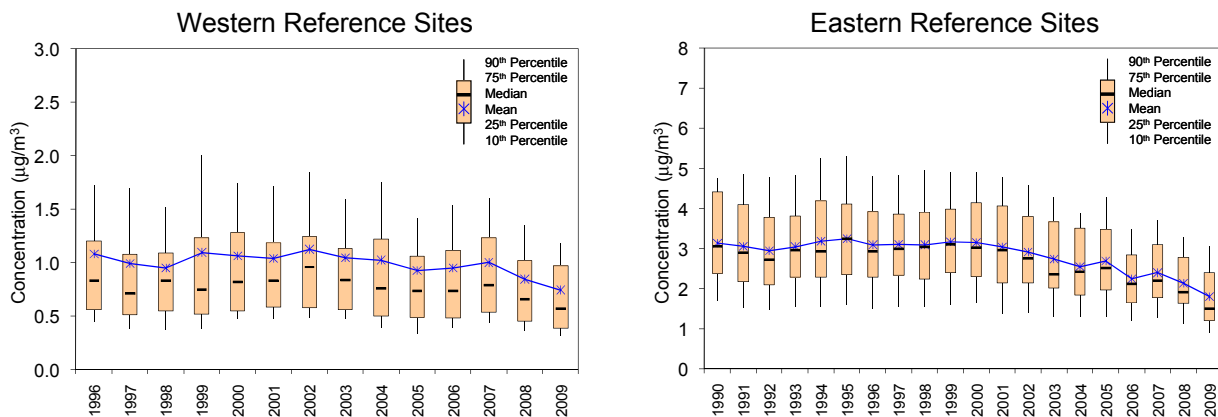
Cranberry, NC (PNF126)

Figure 2-5 Annual Mean Total Nitrate ($\text{NO}_3 + \text{HNO}_3$) Concentrations ($\mu\text{g}/\text{m}^3$) for 2009



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 2007–2009 was lower than the corresponding 1996–1998 level, and the two mean concentrations for 2008 and 2009 were the lowest over the 14-year period.

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



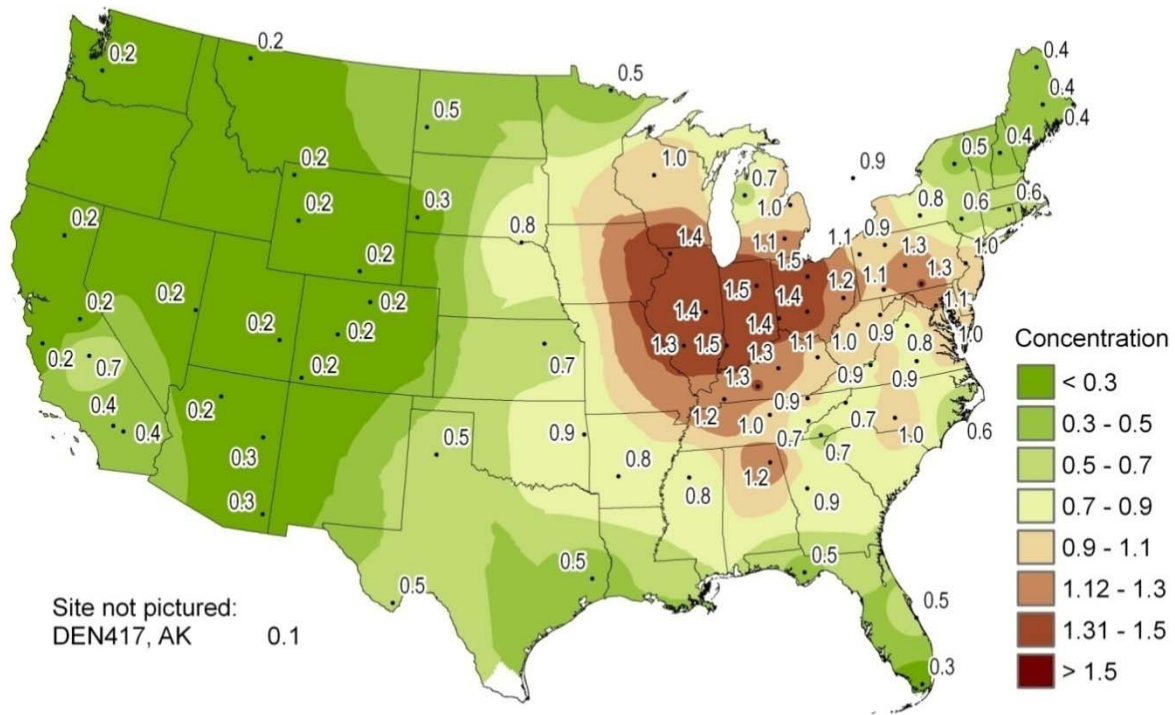
Particulate Ammonium

Particulate ammonium is formed when NH_3 reacts with other gases and particles in the atmosphere including HNO_3 , particulate SO_4^{2-} , and particulate NO_3^- . A map of annual mean NH_4^+ concentrations for 2009 is 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 central and eastern United States with about half the 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, Kentucky, and northern Alabama. The NH_4^+ concentrations measured at western sites were low with all sites measuring concentrations less than $1.0 \mu\text{g}/\text{m}^3$.



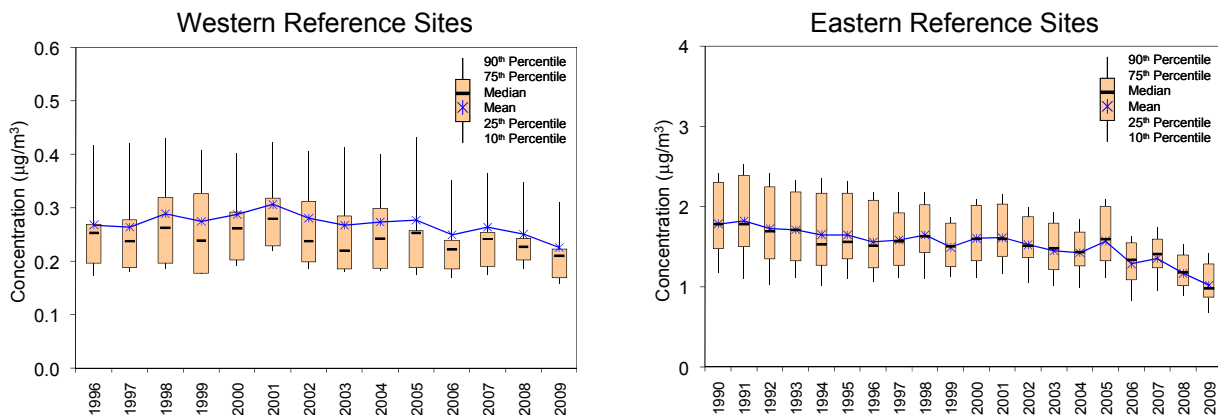
Sumatra, FL (SUM156)

Figure 2-7 Annual Mean NH_4^+ Concentrations ($\mu\text{g}/\text{m}^3$) for 2009



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 2007–2009. The 1990–1992 mean concentration was $1.8 \mu\text{g}/\text{m}^3$, and the 2007–2009 value was $1.2 \mu\text{g}/\text{m}^3$, a 34 percent decline. The box plots characterizing the western sites show no significant change.

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



Trends in Sulfur Pollutants and Mercury Deposition

EGUs and other sources, such as incinerators and gold mines, emit significant quantities of Hg. These emissions produce concentrations of Hg in the atmosphere and contribute to subsequent Hg deposition, which leads to contamination of sensitive ecosystems. In some ecosystems, excess sulfur contributes to increased Hg methylation—the transformation of Hg into MeHg, a highly toxic form of Hg associated with a range of adverse effects in humans and animals. EPA is currently developing air toxics emissions standards for power plants under Section 112 of the CAAA, which gives EPA the authority to regulate power plant Hg emissions by establishing performance standards or maximum achievable control technology (MACT).

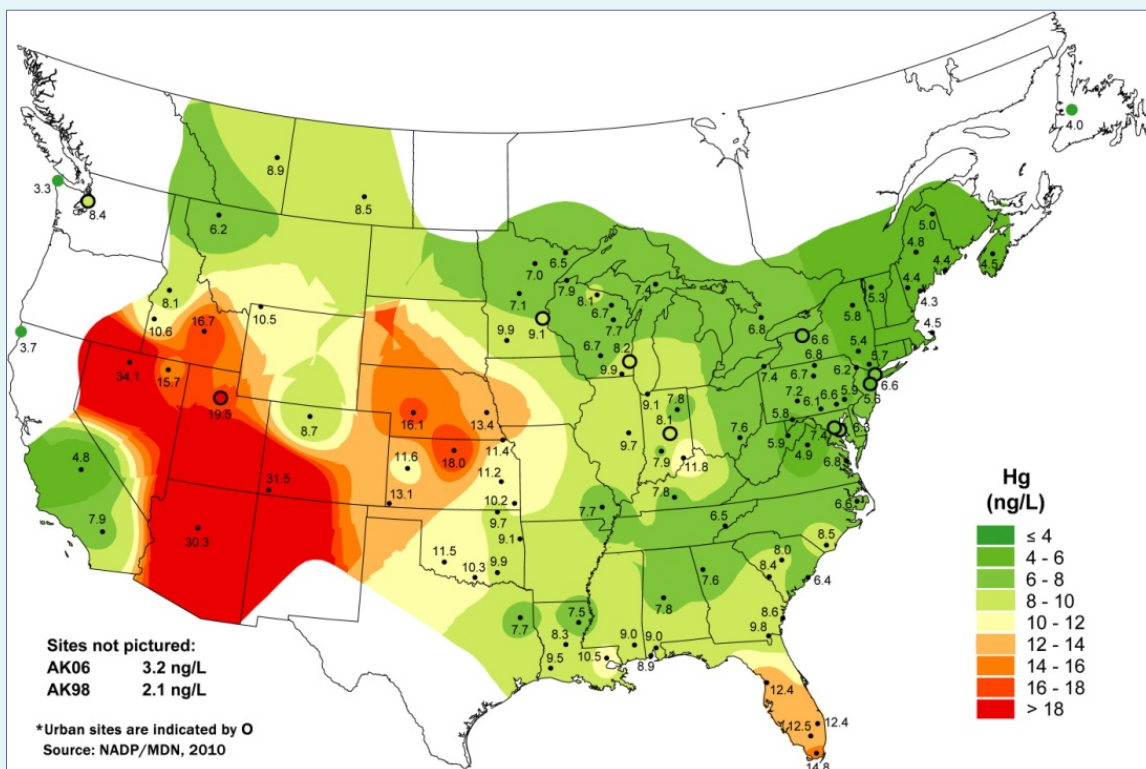
A recent study investigated if a secondary benefit of reduced Hg in precipitation accompanied the decline in SO₂ and NO_x emissions and the related decline in ambient sulfur and nitrogen concentrations and depositions. The study examined the relationships between CASTNET measurements of SO₂ and particulate SO₄²⁻ concentrations and NADP's MDN measurements of concentrations of Hg in precipitation. The study also investigated long-term trends in the three parameters. Eighteen collocated MDN and CASTNET sites were identified (Figure 2g), and data sets were developed by matching weekly samples. Selected sites were divided into three groups consisting of eastern, western, and “other” sites (Table 2b), which were outside of or located on the periphery of the eastern and western groups. Analyses were performed on all sites individually and for each regional group.

Figure 2g CASTNET/MDN Collocated Site Pairs



Figure 2h depicts 2009 annual mean concentrations of Hg in precipitation from all MDN sites. Annual mean SO₂ and SO₄²⁻ concentrations for 2009 are shown in Figures 2-1 and 2-3. Figure 2h shows relatively high Hg concentrations in precipitation were measured in the western states and southern Florida while Figures 2-1 and 2-3 show elevated ambient SO₂ and SO₄²⁻ concentrations were observed in and near the major SO₂ source region in the Ohio River Valley.

Figure 2h 2009 Mercury Concentrations in Precipitation* [nanograms per liter (ng/L)]



Three primary approaches were used. The initial approach included correlation analyses and stepwise regression to determine relationships between Hg (as a dependent variable) and SO_4^{2-} and SO_2 (as independent variables). Then, long-term trends in Hg, SO_2 , and SO_4^{2-} were investigated by nonparametric trend analyses using the Mann-Kendall test and also by parametric trend analyses using regression analysis. Since the data sets were not normally distributed even after log-transformation of the data, nonparametric analysis was applied. Regression approaches require normally distributed data for technical validity. Nonparametric tests do not rely on/are insensitive to assumptions about the data distribution making them more rigorous than parametric tests. However, nonparametric procedures are less likely to observe a trend even if a trend exists.

Correlation of Hg with SO_2 and SO_4^{2-}

The first approach investigated whether Hg concentrations were correlated with either SO_2 or SO_4^{2-} concentrations. These analyses were performed for the entire data set and for individual stations. The results are summarized in Table 2b.

Taken as a group, the eastern sites exhibited significant positive correlation between SO_4^{2-} and Hg but significant negative correlation between SO_2 and Hg. Most of the individual sites exhibited similar relationships. The correlations were interpreted as significant at the 0.10 level of significance. All 10 of the eastern sites showed a positive correlation between Hg and SO_4^{2-} ; 7 of the 10 exhibited a negative correlation between Hg and SO_2 . The western sites positively correlated both SO_4^{2-} and SO_2 with Hg, though results varied for individual sites. The “other sites” were similar to the eastern group.

Table 2b Correlations between Log-Transformed Hg and SO₄²⁻ and SO₂

Site	SO ₄ ²⁻		SO ₂		N
	Sign	α	Sign	α	
All	+	<0.001	-	<0.001	3140
Eastern Sites					
ACA416/ME98	+	<0.001	-	<0.001	375
ARE128/PA00	+	<0.001	-	0.09	278
BEL116/MD99	+	0.001			96
BVL130/IL11	+	<0.001	-	0.002	311
CAT175/NY68	+	<0.001	-	0.02	133
CND125/NC26	+	0.01			61
HWF187/NY20	+	<0.001	-	<0.001	259
MAC426/KY10	+	<0.001	-	0.002	234
SHN418/VA28	+	<0.001	-	0.03	200
WST109/NH02	+	0.07			39
Eastern	+	<0.001	-	<0.001	1986
Western Sites					
CON186/CA94	+	0.03	+	0.006	26
GLR468/MT05	+	0.08			109
MEV405/CO99	+	<0.001			147
SEK430/CA75			+	<0.001	61
YEL408/WY08			+	0.006	21
Western	+	<0.001	+	<0.001	364
Other Sites					
CHE185/OK99					173
EGB181/ON07	+	<0.001	-	<0.001	300
EVE419/FL11					310

Table 2c Individual Stations, Mann-Kendall, SO₄²⁻ Trends

Station	N	P	Significant trend?
Eastern Sites			
ACA416/ME98	514	0.000	Decreasing
ARE128/PA00	411	0.002	Decreasing
BEL116/MD99	168	0.009	Decreasing
BVL130/IL11	464	0.001	Decreasing
CAT175/NY68	195	0.003	Decreasing
CND125/NC26	84	0.010	Increasing
HWF187/NY20	339	0.000	Decreasing
MAC426/KY10	323	0.337	
SHN418/VA28	292	0.751	
WST109/NH02	50	0.001	Decreasing
Western Sites			
CON186/CA94	139	0.095	Decreasing
GLR468/MT05	260	0.024	Decreasing
MEV405/CO99	310	0.806	
SEK430/CA75	179	0.044	Decreasing
YEL408/WY08	277	0.626	
Other Sites			
CHE185/OK99	84	0.013	Decreasing
EVE419/FL11	446	0.075	Decreasing

The positive correlation between Hg and SO₄²⁻ is of particular concern because SO₄²⁻-reducing bacteria play a primary role in methylation of Hg to MeHg in the environment. Although atmospheric-ecosystem chemical reactions are complex, scientists (e.g., Jeremiason *et al.*, 2006; Gilmour *et al.*, 1992; Hines and Brezonik, 2004) have shown through laboratory and field studies that the addition of SO₄²⁻ to experimental wetlands has increased MeHg concentrations.

Trends in Hg, SO₂, and SO₄²⁻

The Mann-Kendall test is a nonparametric statistical test that was used to analyze trends in the three parameters for individual monitoring sites as well as in the four groups of sites (i.e., eastern, western, other, and all sites). The individual results showed downward trends for SO₂ and SO₄²⁻ at most of the sites and downward trends for Hg at two sites (HWF187/NY20, NY and WST109/NH02, NH). The results for the four groups showed only a downward trend for SO₄²⁻ measured at western sites. Table 2c summarizes the Mann-Kendall trend analyses for SO₄²⁻ for the individual monitoring sites. Seven eastern, three western, and two other sites exhibited statistically significant decreasing trends. The Mann-Kendall test also showed significant declines in SO₂ concentrations at 15 of 17 sites.

The parametric trend analyses used regression techniques to gauge trends in the eastern and western subsets after the statistical elimination of annual cycles in the raw data. The results are summarized in Figure 2i and Table 2d. The results show that in the west, SO₄²⁻ increased from 2002 to 2008 at approximately 2.8 percent/year, while in the east, no statistically significant trend was observed. However, the apparent, but not significant, trend in the east was a decline of 0.7 percent/year. The trend in SO₄²⁻ concentrations measured at the CASTNET eastern reference sites shown in Figure 2-4 gives a 41 percent reduction in SO₄²⁻ over 20 years. Figure 2i and Table 2d also show that SO₂ decreased in both the eastern and western regions at a rate of 2.3 percent/year in the east and 7.6 percent/year in the west, and that Hg declined at a rate of 1.1 percent/year in the east and 5.2 percent/year in the west.

Figure 2i Trends in SO₄²⁻, SO₂, and Hg

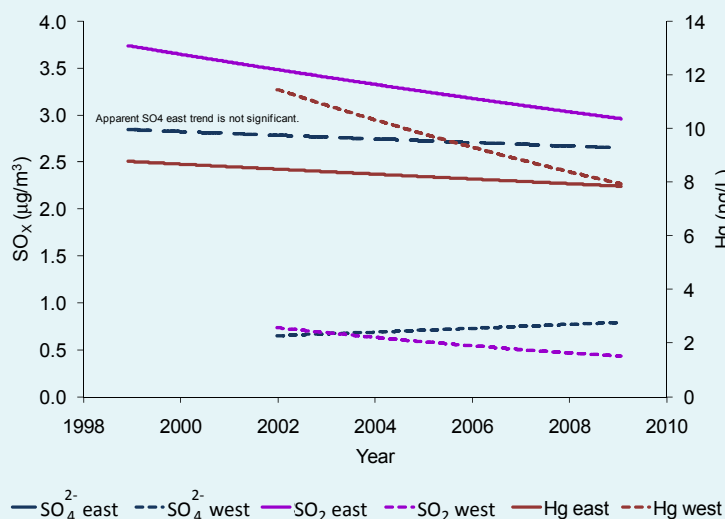


Table 2d Parametric Trend Results

	Region	Direction	Rate (%/year)
SO ₄ ²⁻	Eastern	Not significant	-0.7±0.5
	Western	Increasing	2.8±0.8
SO ₂	Eastern	Decreasing	-2.3±0.7
	Western	Decreasing	-7.6±1.1
Hg	Eastern	Decreasing	-1.1±0.6
	Western	Decreasing	-5.2±2.3

Summary

Hg was positively correlated with SO₄²⁻ at 14 of 18 sites evaluated. Exceptions were SEK430/CA75, CA; YEL408/WY08, WY; EVE419/FL11, FL; and CHE185/OK99, OK. Hg was also positively correlated with SO₄²⁻ in the eastern and western groups. Overall, this study demonstrated a positive correlation between SO₄²⁻ concentrations and Hg in precipitation.

Hg was negatively correlated with SO₂ at 7 of the 10 eastern sites but was positively correlated with SO₂ in the west. This was related to the seasonal cycle of SO₂ emissions near the western sites where SO₂ is higher in the summer months because of increased demand for electrical power.

A strong declining trend for SO₂ was demonstrated consistently by both the nonparametric and parametric analyses. No significant declining trend for SO₄²⁻ in either the eastern or western subsets was demonstrated by the parametric analyses while, in contrast, the Mann-Kendall nonparametric procedure identified significant decreasing trends for SO₄²⁻ at a number of sites. Because Hg and SO₄²⁻ are strongly correlated, similar trends would be expected for Hg and SO₄²⁻; however, the trends at individual stations and within regional subsets were not consistent. The nonparametric procedure indicates SO₄²⁻ concentrations were generally declining while decreasing trends in Hg were observed at relatively few stations. Parametric trend analysis reveals significant decreases for Hg in both the western and eastern subsets, but these clear trends were not observed for SO₄²⁻ in the regional subsets. These results may indicate that geographic differences have not been correctly accounted for in the regional analyses.

The trends analyses demonstrated that both Hg and SO₂ have declined at individual monitoring stations and the eastern and western subsets. However, the results do not demonstrate that a secondary benefit of a reduction in Hg concentrations accompanied the declines in SO₂ concentrations.

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 measurement-modeling approach to estimate dry deposition. The hybrid approach combines measured pollutant concentrations and modeled deposition velocities. The MLM is used to calculate hourly deposition velocities for each monitoring site based on meteorological measurements and information on the vegetation within 1 kilometer (km) of each site. Dry deposition is then calculated as the product of the modeled deposition velocity and measured pollutant concentration. Total deposition is the sum of estimated dry deposition and measured wet deposition. Three-year mean total sulfur fluxes aggregated over the eastern reference sites declined by 46 percent over the period from 1990–1992 to 2007–2009. Three-year mean total sulfur deposition estimated for 17 western sites declined by about a third over the period 1996–1998 to 2007–2009. Total nitrogen deposition estimates declined 26 percent for the eastern reference sites and 21 percent at the western reference sites. 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 to the environment through dry and wet atmospheric processes. A principal objective of CASTNET is to estimate the rate, or flux, of dry deposition from the atmosphere to sensitive ecosystems. 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 2009 at the eastern reference sites. These same parameters, with the exception of total sulfur deposition, declined during 2009 at the western reference sites. Precipitation-weighted mean concentrations of atmospheric sulfur declined over the past 20 years at the eastern sites, and mean concentrations of atmospheric nitrogen in precipitation have declined slowly since 1998 at the same sites.

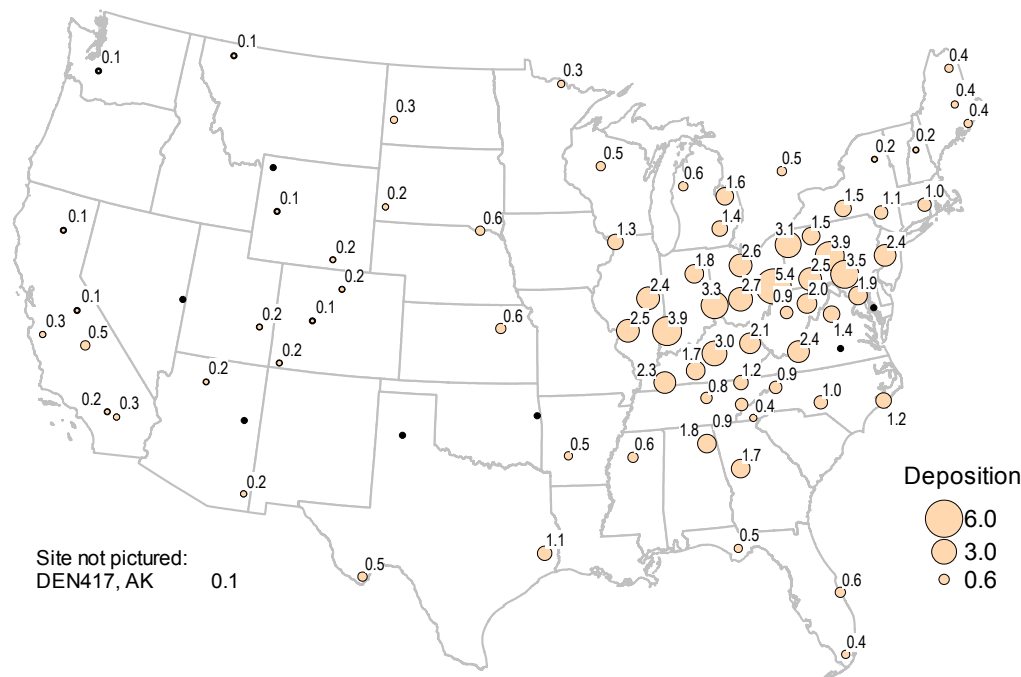
The MLM (Figure 1-5) simulates dry deposition processes. The MLM was summarized by Meyers *et al.* (1998) and Finkelstein *et al.* (2000). The MLM was run using CASTNET filter pack concentrations and meteorological measurements, with information on percent leaf-out of nearby vegetation, LAI, and surface 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 2009. Valid measurements of temperature, solar radiation,

relative humidity, wind speed, and standard deviation of the wind direction (sigma theta) must all be available for the hour in order to estimate deposition velocity. 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.

Sulfur Deposition

The MLM was run separately for SO₂ and SO₄²⁻. The model calculations were summed to obtain the 2009 estimates of dry sulfur deposition (as S) shown in the map in Figure 3-1. The magnitude of a deposition rate is illustrated by the size of the circle. The map shows only seven CASTNET sites in four states with estimated fluxes greater than or equal to 3.0 kilograms per hectare per year (kg/ha/yr). These sites are located in Indiana, Kentucky, Ohio, and Pennsylvania. The geographic extent of elevated deposition rates was significantly smaller than in 2008. The highest 2009 deposition rate was estimated for QAK172, OH with a flux of 5.4 kg/ha/yr compared to a flux of 8.9 kg/ha/yr in 2008. The highest dry sulfur deposition rates coincided with the major SO₂ source region (Figure 1-6) and declined 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 (SO₂ + SO₄²⁻) Deposition (kg/ha/yr) for 2009



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.

A map of estimates of total sulfur deposition is given in Figure 3-2. 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. Twelve CASTNET sites in the states of Illinois, Indiana, Kentucky, Ohio, Pennsylvania, and Maryland had total (dry + wet) sulfur deposition fluxes greater than 7.0 kg/ha/yr. The highest total sulfur deposition rate was estimated for QAK172, OH with a value of 9.7 kg/ha/yr. The total sulfur deposition value was 17.2 kg/ha/yr for this site in 2008. Total sulfur deposition at western sites was less than or equal to 1.7 kg/ha/yr, which was the estimate for Centennial, WY (CNT169). 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 QAK172, OH to less than 20 percent at Indian River Lagoon, FL (IRL141).

Figure 3-2 Total (Dry + Wet) Sulfur Deposition (kg/ha/yr) for 2009

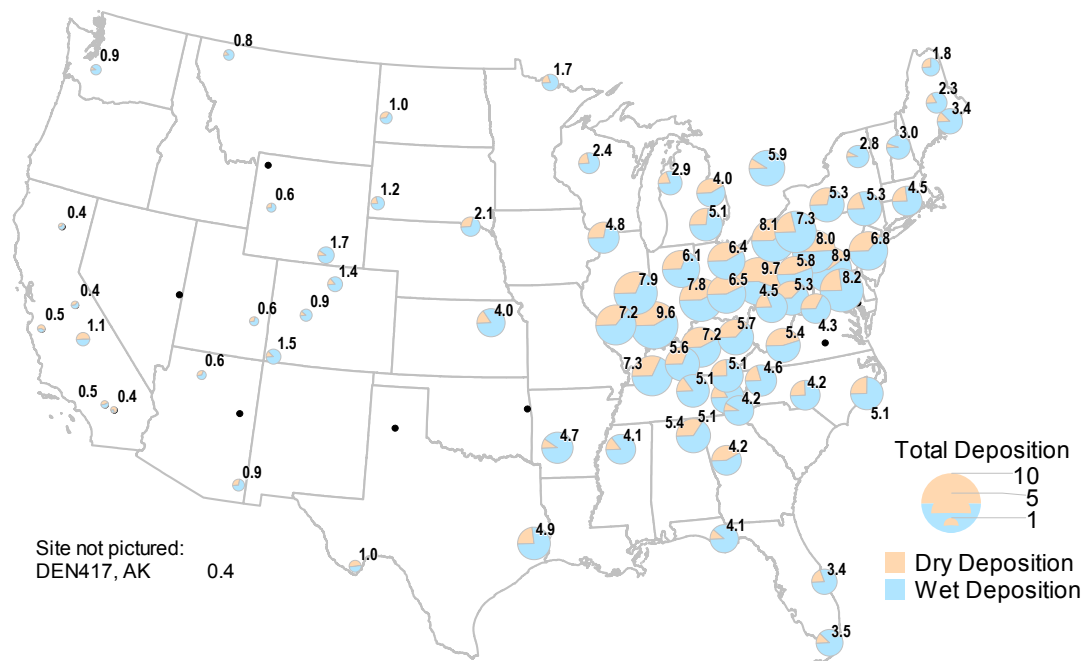


Figure 3-3 presents box plots that show the trend in dry sulfur deposition (as S) for the eastern (right side) and western (left side) reference sites. Figure 3-4 shows the trend in annual total (dry + wet) sulfur deposition (as S). The box plots were based on data obtained from the 34 CASTNET eastern reference sites (Figure 1-2) for the period 1990 through 2009 and from the 17 western reference sites (Figure 1-3) for 1996 through 2009. 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. Overall, total sulfur deposition declined at the eastern reference sites from a 1990–1992 mean of 13.2 kg/ha/yr to a 2007–2009 mean of 7.1 kg/ha/yr, a 46 percent reduction. Total sulfur deposition declined at the western reference sites from a 1996–1998 mean of 1.2 kg/ha/yr to a 2007–2009 mean of 0.8 kg/ha/yr, a 31 percent reduction.

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

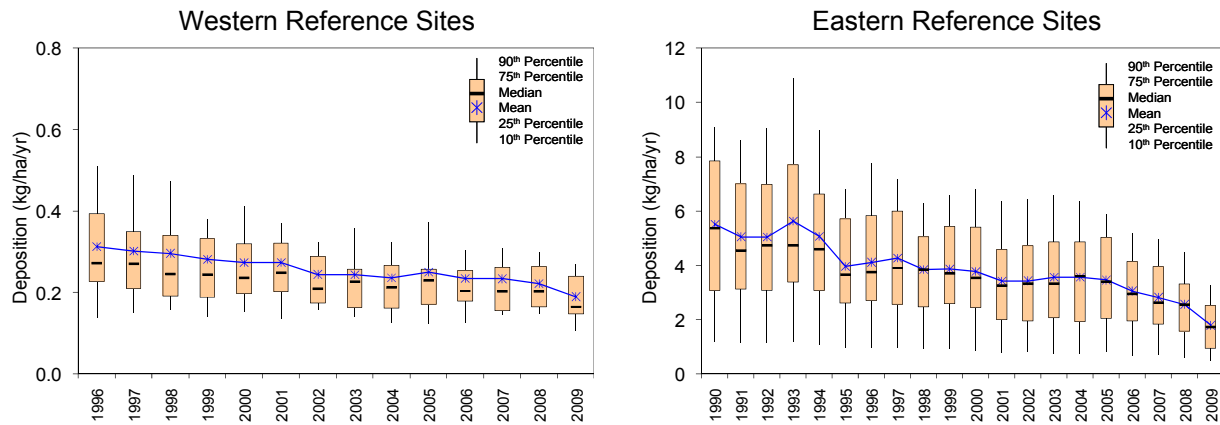


Figure 3-4 Trend in Total Sulfur Deposition (kg/ha/yr)

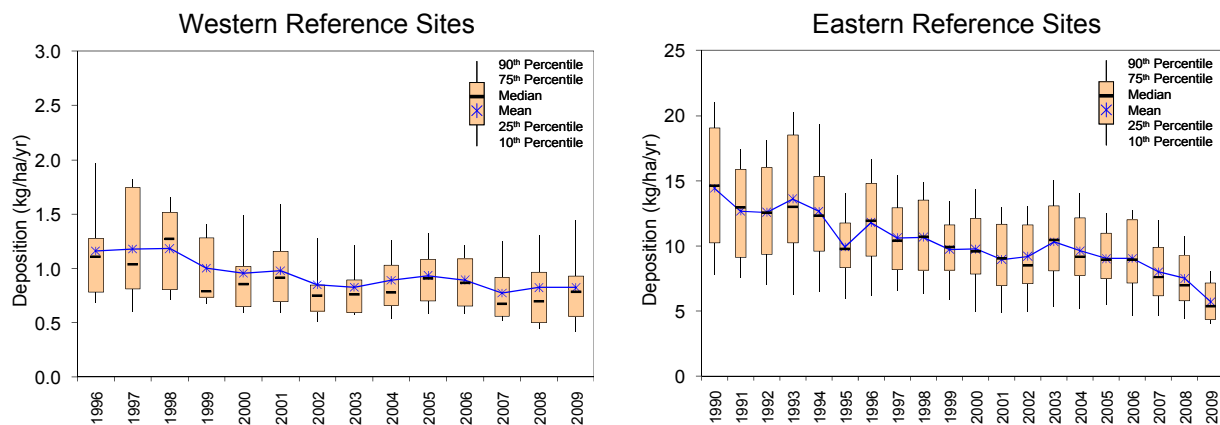
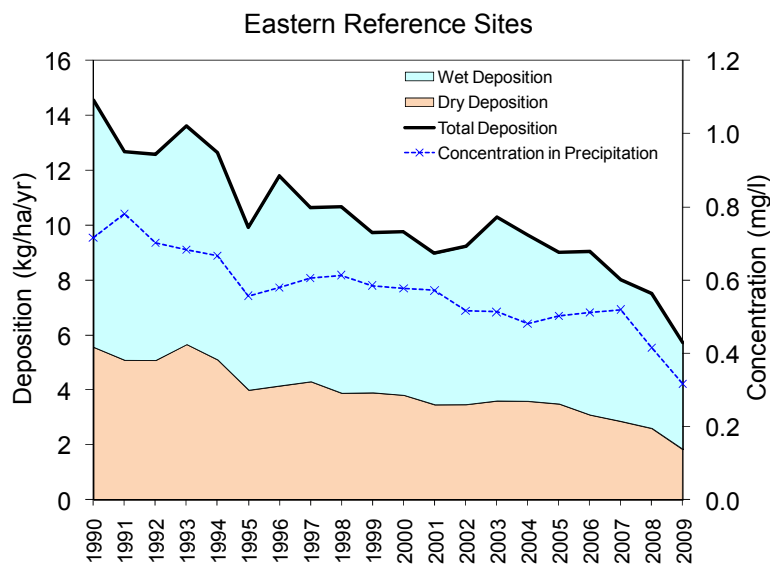


Figure 3-5 presents estimates of trends in dry, wet, and total deposition of sulfur (as S) for the eastern reference sites. The influence of precipitation on total sulfur deposition is illustrated by comparing the solid (top) line to the dotted blue line in Figure 3-5. The solid line shows total deposition, 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 trend line for precipitation-weighted mean sulfur concentrations in precipitation shows a decline in 2008 and 2009 after small increases during the previous three years.

Figure 3-5 Trend in Sulfur Deposition (kg/ha/yr) with Concentrations in Precipitation [milligrams per liter (mg/l)]

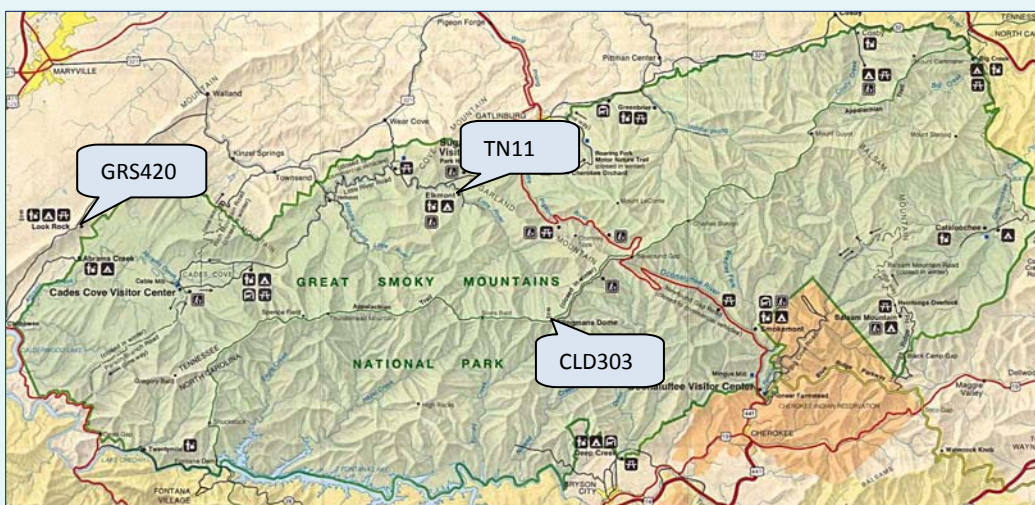


Laurel Hill State Park, PA (LRL117)

Trends in Pollutants in Cloud Water and a Comparison to Trends in Sulfate and Nitrate in Precipitation

The CASTNET Mountain Acid Deposition Program (MADPro) has been monitoring cloud water and its chemical constituents at Clingmans Dome, TN (CLD303) in the Great Smoky Mountains National Park (Figure 3a) since 1994. High-elevation (typically higher than 800 meters) ecosystems such as Clingmans Dome are subject to substantial levels of acid deposition from clouds that originate in polluted areas and contain high concentrations of dissolved acidic ions. High levels of acid deposition result from frequent cloud immersion, orographically enhanced precipitation, high wind speeds, and the large leaf areas typical of mountain tree species.

Figure 3a Great Smoky Mountains National Park Monitoring Sites



Sulfur and nitrogen species concentrations and total (dry + wet) deposition fluxes have declined steadily over the last 10 to 15 years at the lower elevation CASTNET sites in the eastern United States. Changes in cloud water concentrations and deposition rates based on CLD303 estimates (MACTEC, 2010d) have been more sporadic yet also show a decline.



Close Up View of Cloud Collector

Recently, significant reductions in sulfur and nitrogen species have been measured at CLD303, perhaps associated with emission reductions at Tennessee Valley Authority (TVA) power plants during the period from 2007 through 2009 (TVA, 2010). Mean seasonal (June through September) reductions in SO_4^{2-} and NO_3^- cloud water concentrations were 60.4 percent and 49.3 percent, respectively, between 2007 and 2009 (MACTEC, 2010d). SO_2 emissions from all TVA plants declined by 47 percent from 2007 to 2009. The NO_x emission reduction over the same period was 70 percent (TVA, 2010; Figure 3b).

Because of the substantial decline in both mean SO_4^{2-} and mean NO_3^- cloud water concentrations,

Figure 3b Sulfur Dioxide and Nitrogen Oxides Emissions at TVA Power Plants

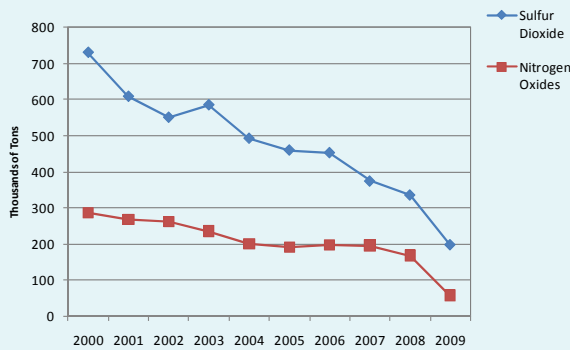


Figure 3c Mean Seasonal Cloud Water and Mean Seasonal Precipitation Sulfate Concentrations, 2000–2009

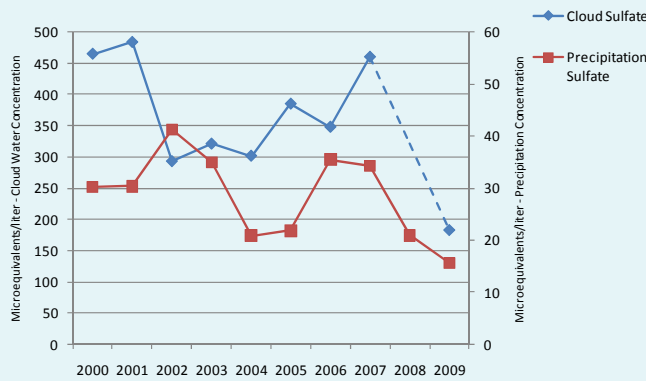
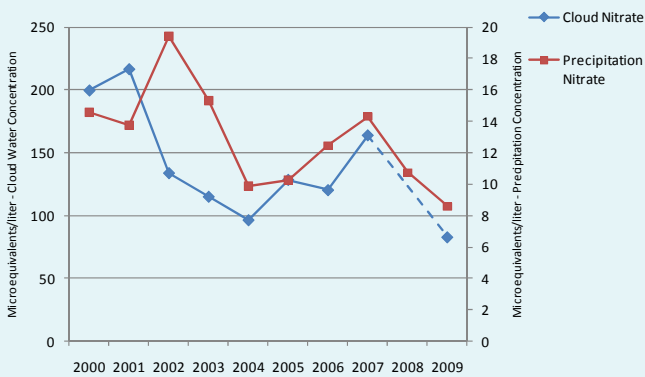


Figure 3d Mean Seasonal Cloud Water and Mean Seasonal Precipitation Nitrate Concentrations, 2000–2009



precipitation concentration data were obtained from the nearby NADP/NTN site at Elkmont, TN (TN11) to assess whether the 2009 mean seasonal (June through September) SO_4^{2-} and NO_3^- precipitation concentrations exhibited a similar reduction. The data were compared over the same period from 2000 through 2007 and 2009.

Figures 3c and 3d show mean seasonal cloud water and precipitation concentrations for SO_4^{2-} and NO_3^- , respectively. Cloud water concentrations are plotted on the left y-axis, and precipitation concentrations are plotted on the right y-axis. No data are available for 2008 because cloud sampling was not performed during that year. Both figures show that the large drop in the 2009 cloud water SO_4^{2-} and NO_3^- concentrations was mirrored by a decline in precipitation SO_4^{2-} and NO_3^- concentrations. The 60 percent decrease in 2009 cloud water SO_4^{2-} concentrations from 2007 concentrations is paralleled by a 55 percent decrease in precipitation SO_4^{2-} concentrations. The 49 percent decrease in 2009 cloud water NO_3^- concentrations is tracked by a 40 percent decrease in 2009 precipitation NO_3^- concentrations.

SO_4^{2-} and NO_3^- mean seasonal cloud water and wet deposition fluxes as kilograms per hectare (kg/ha) for 2000 through 2007 and 2009 are shown in Figures 3e and 3f. Cloud water deposition rates are plotted on the left y-axis and precipitation fluxes are plotted on the right y-axis. Deposition fluxes for both species basically follow the same pattern starting in 2004. The main exception is that the precipitation SO_4^{2-} deposition value for 2009 decreased with respect to the 2007 value but was higher than the 2008 deposition rate. The NO_3^- depositions for both cloud and precipitation deposition show increases in 2009 with respect to the 2007 value. The increases in the cloud water SO_4^{2-} and NO_3^- and the wet NO_3^- deposition are opposite to the decrease in seasonal concentrations of these parameters.

Figure 3e Mean Seasonal Cloud Water and Wet Sulfate Deposition Estimates, 2000-2009

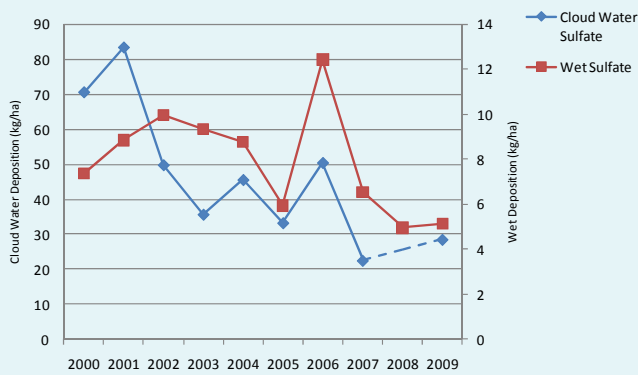
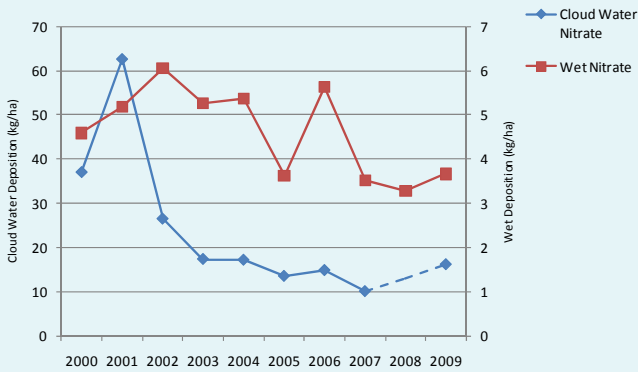


Figure 3f Mean Seasonal Cloud Water and Wet Nitrate Deposition Estimates, 2000-2009



The increases in cloud water deposition for 2009 have been attributed to higher water deposition (Lovett, 2010). Water deposition for 2009 was 9.1 centimeters per month (cm/month) versus 3.5 cm/month for 2007 (MACTEC, 2010d). The year 2007 was characterized by drought.

Although substantial decreases were observed in both cloud water and precipitation SO_4^{2-} and NO_3^- concentrations since 2007, these decreases were not accompanied by decreases in cloud water and wet deposition. Consequently, sensitive high-elevation ecosystems of the Smoky Mountains may continue to experience damage from acid deposition and acidification because of poorly buffered soils and other ecological factors. Therefore, it is important to continue cloud water collection and calculation of SO_4^{2-} and NO_3^- deposition fluxes at the Clingmans Dome site.



View from the Clingmans Dome Tower

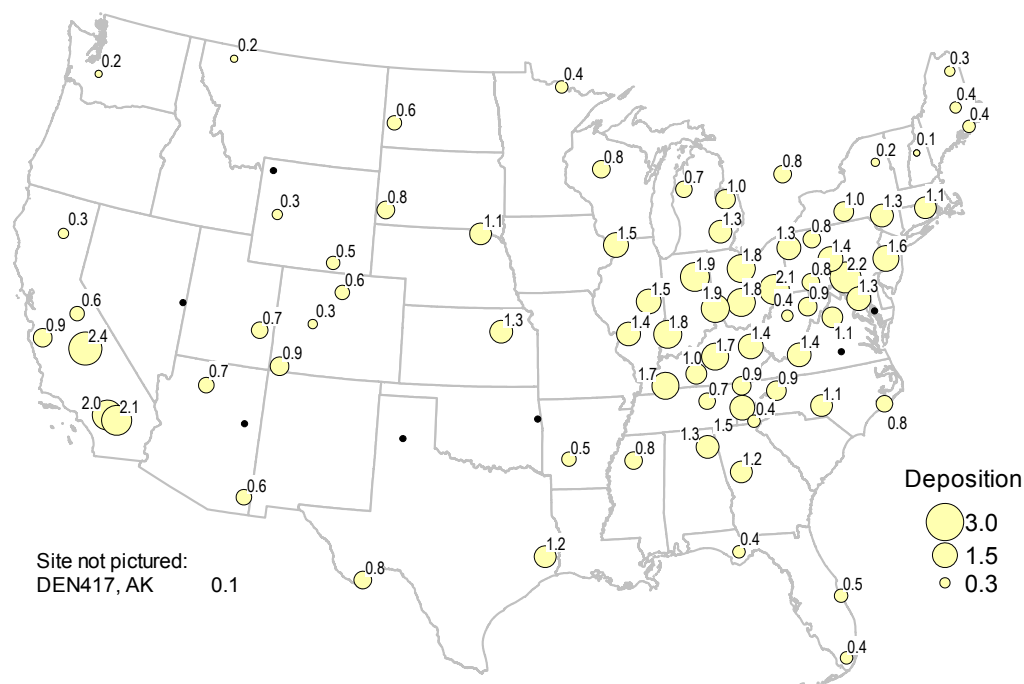


Clingmans Dome Tower

Nitrogen Deposition

Figure 3-6 illustrates dry fluxes of nitrogen (as N) for 2009. 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. QAK172, OH and ARE128, PA were the only two eastern sites with deposition rates greater than 2.0 kg/ha/yr. Three monitoring sites in California had fluxes greater than or equal to 2.0 kg/ha/yr. The highest value was calculated for the CASTNET site in Sequoia National Park (SEK430) with a dry nitrogen flux of 2.4 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 2009



A map of total nitrogen deposition (as N) for 2009 is given in Figure 3-7. 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 estimated deposition rates greater than 5.0 kg/ha/yr. No values above 10.0 kg/ha/yr were estimated for 2009. Vincennes, IN (VIN140) recorded the highest total nitrogen flux (8.8 kg/ha/yr). The values at the western sites ranged from 0.8 kg/ha/yr at Mount Rainier National Park, WA (MOR409) to 4.2 kg/ha/yr in California at SEK430. 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. Dry nitrogen deposition contributed 85 percent of the total nitrogen deposition at Joshua Tree National Park, CA (JOT403).

Figure 3-7 Total (Dry + Wet) Nitrogen Deposition (kg/ha/yr) for 2009

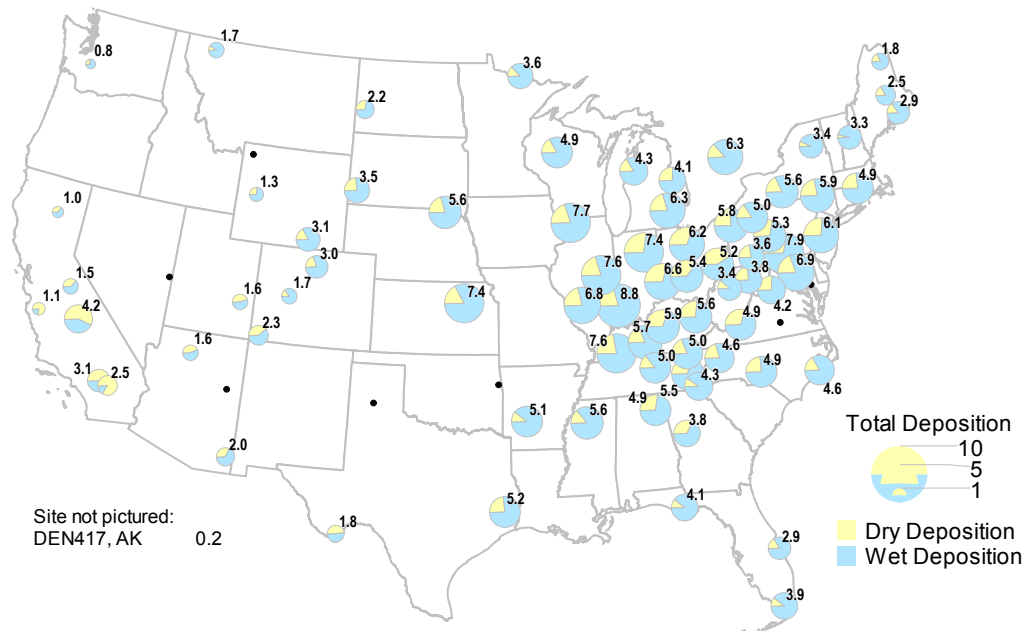


Figure 3-8 presents box plots that were constructed using data from the 34 eastern reference sites over the period 1990 through 2009 (right side) and 17 western reference sites for 1996 through 2009 (left side). The box plots show the trends in dry nitrogen deposition (as N). The box plots for the eastern 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 for 1990–1992 to 1.5 kg/ha/yr for 2007–2009, a 32 percent reduction over the 20 years. The box plots for the western sites show a smaller 20 percent decline, a change from a mean of 0.9 kg/ha/yr for 1996–1998 to 0.7 kg/ha/yr for 2007–2009.



Rocky Mountain National Park, CO (ROM206)

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

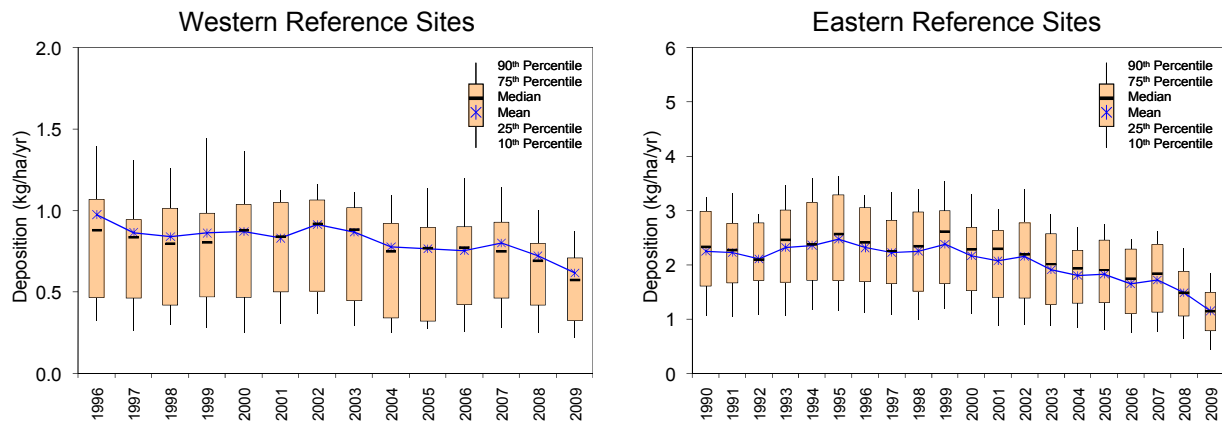


Figure 3-9 shows the trends in annual total (dry + wet) nitrogen deposition for the eastern (right side) and western (left side) reference sites. Total nitrogen deposition aggregated over the eastern sites is more variable because the annual wet and total fluxes depend on the amount of precipitation. The figure suggests that total flux in the east 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 2009. 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 26 percent in the East over the 20 years and 21 percent for the aggregated western sites over the 14-year period.

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

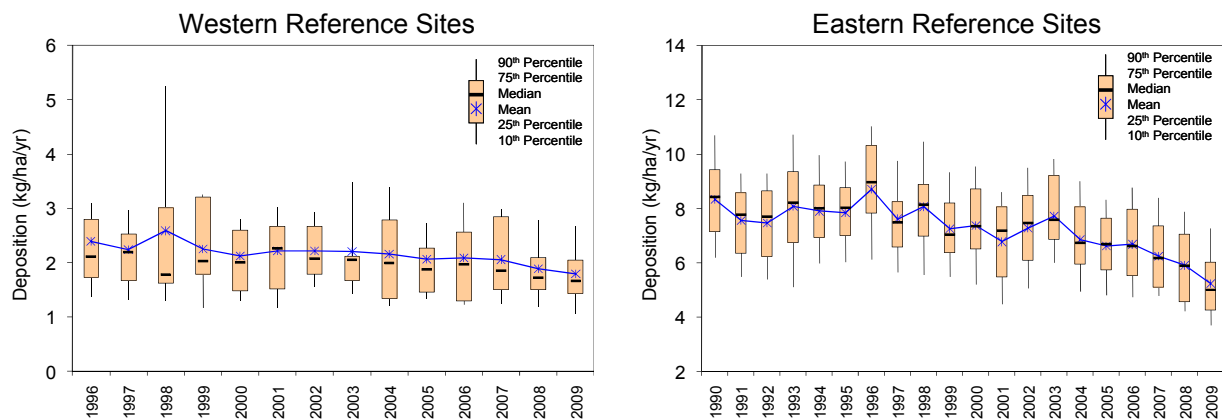
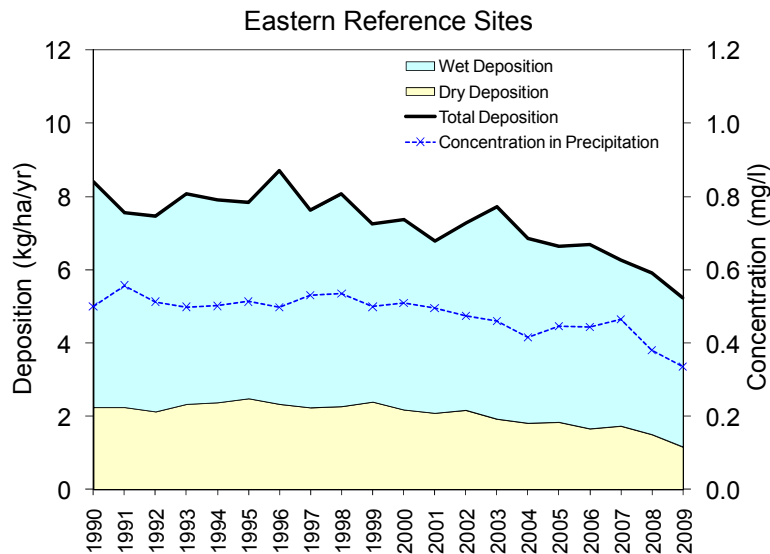


Figure 3-10 Trend in Nitrogen Deposition (kg/ha/yr) with Concentrations in Precipitation (mg/l)



Estimates of Critical Loads for the Shenandoah Region

The mid-Appalachian Mountains in the eastern United States have been strongly impacted by acidic deposition. Many of the small forested watersheds, particularly on ridge and mountaintops, have soils and surface waters that are unable to buffer the acidity from pollutant deposition, causing the ecosystems to acidify. As a result, the health of some forest trees and aquatic biota has declined, and some species, such as brook trout, have been lost in the small streams in these watersheds.

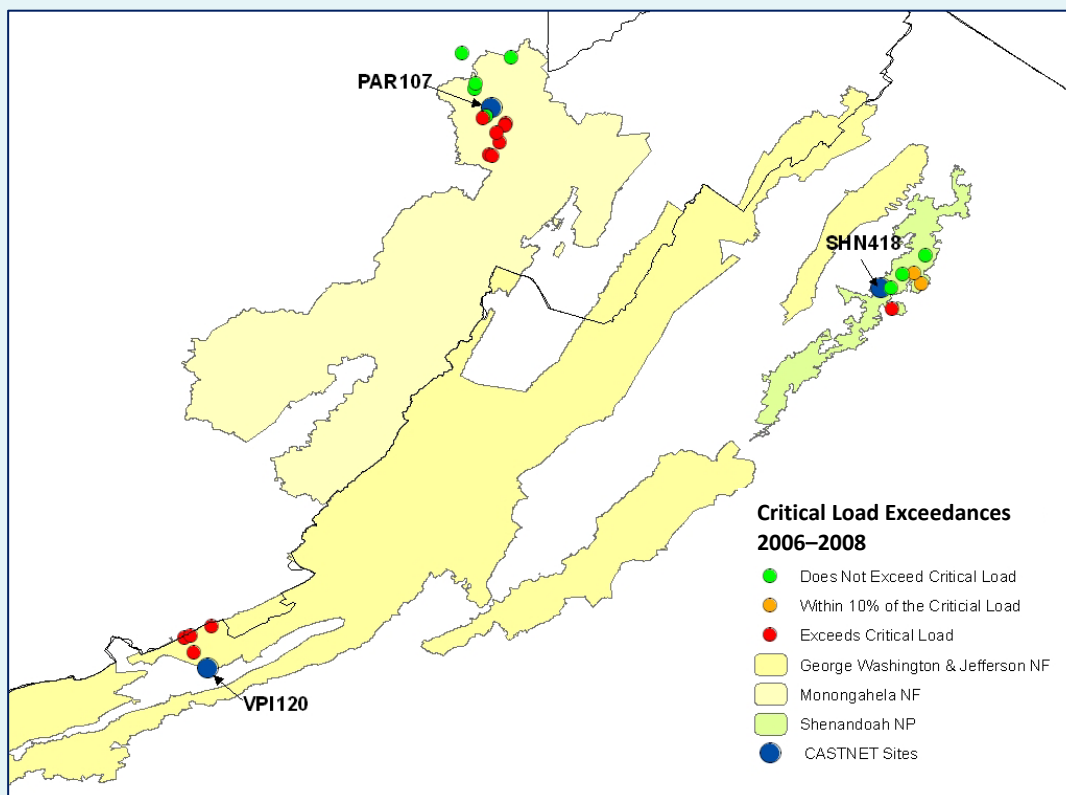
The critical load approach is an emerging assessment tool that can be used to determine the degree to which acidic deposition may be affecting ecological health. A critical load is a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specific sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt, 1988). If pollutant exposure is less than the critical load, adverse ecological effects (e.g., reduced reproductive success, stunted growth, or loss of biological diversity) are not anticipated. A critical load exceedance is the measure of pollutant exposure above the critical load and indicates the ecosystem is exposed to damaging levels of pollutants.

Building on previous surface water modeling of this region (EPA, 2008a), this critical load study examines 22 streams in the Shenandoah National Park and George, Jefferson, and Monongahela National Forests. Three CASTNET monitoring sites, Parsons, WV (PAR107); Shenandoah National Park, VA – Big Meadows (SHN418); and Horton Station, VA (VPI120), are located near the streams. For this particular analysis, the critical load represents the deposition load of sulfur to which a stream could be subjected and still have an acid neutralizing capacity (ANC) of 50 microequivalents per liter or higher, which would support a healthy aquatic ecosystem. Critical loads of total sulfur deposition are expressed in terms of ionic charge balance as milliequivalents per square meter per year.

Critical loads were calculated for the 22 streams using the Steady-State Water Chemistry Model (Dupont *et al.*, 2005). Catchment weathering rates were derived from the Model of Acidification of Groundwater in Catchments (MAGIC; Cosby *et al.*, 1985). CASTNET (dry) and NADP/NTN (wet) deposition data were used to determine total base cation, Cl⁻, and sulfur deposition each watershed received. Dry deposition was estimated from data from the CASTNET sites closest to the streams. Wet deposition was estimated from NADP/NTN data collected at sites within 0.2 km of the three CASTNET sites. The 22 streams for which critical loads were calculated represent small trout streams that are highly sensitive to acidic deposition and should not be considered characteristic of all streams across this region (Sullivan *et al.*, 2008).

For the period from 2006 through 2008, 60 percent of the modeled streams received levels of sulfur deposition that exceeded the critical load (Figure 3g). While reductions in acidic deposition over the past decade have helped to improve water quality and support healthy aquatic ecosystems, many of the highly sensitive trout streams in the mid-Appalachian Mountains are still at risk from acidification at current deposition levels.

Figure 3g Stream Exceedances of Modeled Critical Loads for Total Sulfur Deposition for the Period 2006–2008



Source: EPA Clean Air Markets Division

Chapter 4: Ozone Concentrations

CASTNET is the principal network for monitoring rural, ground-level O₃ concentrations in the United States and for providing information on geographic patterns in rural O₃ levels. CASTNET was not originally designed to operate as a regulatory network. Even so, the network provides data that can be evaluated in the context of the ozone NAAQS levels. In this report, O₃ measurements collected during 2007–2009 are evaluated with respect to both the 1997 O₃ standard of 0.08 parts per million (ppm) and the 2008 standard of 0.075 ppm.

On January 6, 2010, EPA initiated a revision of the NAAQS for O₃. EPA proposed lowering the primary standard to a level within the range of 0.060 to 0.070 ppm and establishing a secondary standard based on the measure W126 within a range of 7 to 15 ppm-hours. EPA plans to promulgate final standards by July 2011.

All but three CASTNET sites operate an O₃ analyzer that measures hourly concentrations. CASTNET's geographic coverage of the United States provides data that are useful in terms of presenting information on geographic patterns in regional O₃ and for evaluating O₃ concentrations in context of the O₃ NAAQS. The analyses presented in this chapter provide maps of 8-hour average O₃ concentrations and W126 values and examine trends in the annual fourth highest daily maximum 8-hour average O₃ concentrations using data derived from O₃ concentration measurements at CASTNET sites. The concentration shading for the figures used in this chapter was prepared using the scales shown on each map.

Additional maps of O₃ 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 O₃ monitoring reported to AQS plus the CASTNET O₃ measurements.

Ground-level O₃ is formed in the lower atmosphere when volatile organic compounds (VOCs) and NO_x react in the presence of sunlight and is the main component of smog. It is an air pollutant that can cause a number of human respiratory effects and damage to vegetation and ecosystems. 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 O₃ formation in rural areas. Meteorological conditions play a significant role in O₃ formation. O₃ is mainly a summertime pollutant because sunlight and hot weather accelerate its formation. O₃ concentrations typically increase during daylight hours and peak in the late afternoon after temperature and sunlight intensity have peaked. Concentrations typically fall in the evening although many rural sites measure peak O₃ concentrations after sunset. O₃ concentrations are generally higher in the warmer months but can vary by region of country.

The 8-hour average O₃ standard can be used to assess the status and trends in rural O₃ levels in order to gauge the success of EPA emission reduction programs such as the NO_x SIP Call/NBP and now CAIR. On May 1, 2009, the CAIR NO_x O₃ season trading program began, replacing the NBP in states covered by the new program and requiring further NO_x emission reductions from the power sector. Implementation of the NO_x emission reductions that were mandated by the ARP and other NO_x emission control programs have contributed to reductions in O₃ concentrations, especially in rural areas with elevated 8-hour average levels, because regional transport of pollutants contributes to O₃ formation. VOC emission reduction strategies also have been successful in reducing higher, short-term O₃ concentrations in and downwind of urban areas.

The current O₃ NAAQS are summarized in the box on the following page. On January 6, 2010, EPA proposed revising the NAAQS for O₃ by lowering the primary standard to a level between 0.060 and 0.070 ppm based on the 3-year average of the fourth highest daily maximum 8-hour average concentration. The primary standard is designed to protect public health, including the health of at-risk populations. EPA also proposed establishing a secondary standard based on the measure W126 within a range of 7 to 15 ppm-hours. The secondary standard is designed to protect public welfare and the environment, including sensitive vegetation and ecosystems. W126 is a cumulative metric that sums weighted hourly O₃ concentrations during daylight during the O₃ season. For this report, W126 was calculated for the months of May through September. The highest of the rolling 3-month sums during the five months is the W126 value. EPA plans to promulgate final standards by the end of July 2011.



Unionville, MI (UVL124)

National Ambient Air Quality Standards for Ozone

	Primary Standards		Secondary Standards	
	Level	Averaging Time	Level	Averaging Time
Ozone ¹ (Current)	0.075 ppm 0.08 ppm	8-hour ² 8-hour ³	0.075 ppm 0.08 ppm	8-hour ² 8-hour ³

- Notes:**
- ¹ On January 6, 2010, EPA announced that it is revising the O₃ primary and secondary standards (EPA, 2010c).
 - ² 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 must not exceed 0.075 ppm or 75 ppb in practice (effective May 27, 2008).
 - ³ (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 must not exceed 0.08 ppm (EPA, 1997) or 84 ppb in practice (effective 1997).
(b) The 1997 standard—and the implementation rules for the standard—will remain in place for implementation purposes as EPA undertakes rulemaking to address the transition to the revised O₃ standard.

Eight-Hour Ozone Concentrations

Figure 4-1 presents 3-year averages of the fourth highest daily maximum 8-hour average O₃ concentrations for 2007–2009. During this period, 3-year averages of the fourth highest daily maximum 8-hour average O₃ concentrations were greater than or equal to 85 ppb at four sites in California, including Yosemite National Park (YOS404), Sequoia National Park (SEK430), Converse Station (CON186), and Joshua Tree National Park (JOT403). Four eastern and five California CASTNET sites measured concentrations above the 2008 NAAQS of 0.075 ppm. The four eastern sites include Washington’s Crossing, NJ (WSP144); Beltsville, MD (BEL116); Blackwater National Wildlife Refuge, MD (BWR139); and Great Smoky Mountains National Park, TN (GRS420). The California sites include Pinnacles National Monument (PIN414) in addition to the four sites listed previously. Three-year average concentrations were generally lower in 2007–2009 than in 2006–2008.

The 2007–2009 3-year average of fourth highest daily maximum 8-hour average O₃ concentrations constitute the current design values for achieving the 8-hour O₃ NAAQS. For example, the value of 107 ppb at Converse Station, CA (CON186) would have to be reduced to 84 ppb to achieve the level of the 1997 standard and to 75 ppb to achieve the level of the 2008 standard. A design value is a statistic that describes the air quality status of a given area relative to the level of the NAAQS. Design values are typically used to classify nonattainment areas, assess progress towards meeting the NAAQS, and develop control strategies to achieve the NAAQS. Design values change as each new 3-year database of monitored O₃ concentrations become available.

Figure 4-1 Three-year Average of Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) for 2007–2009

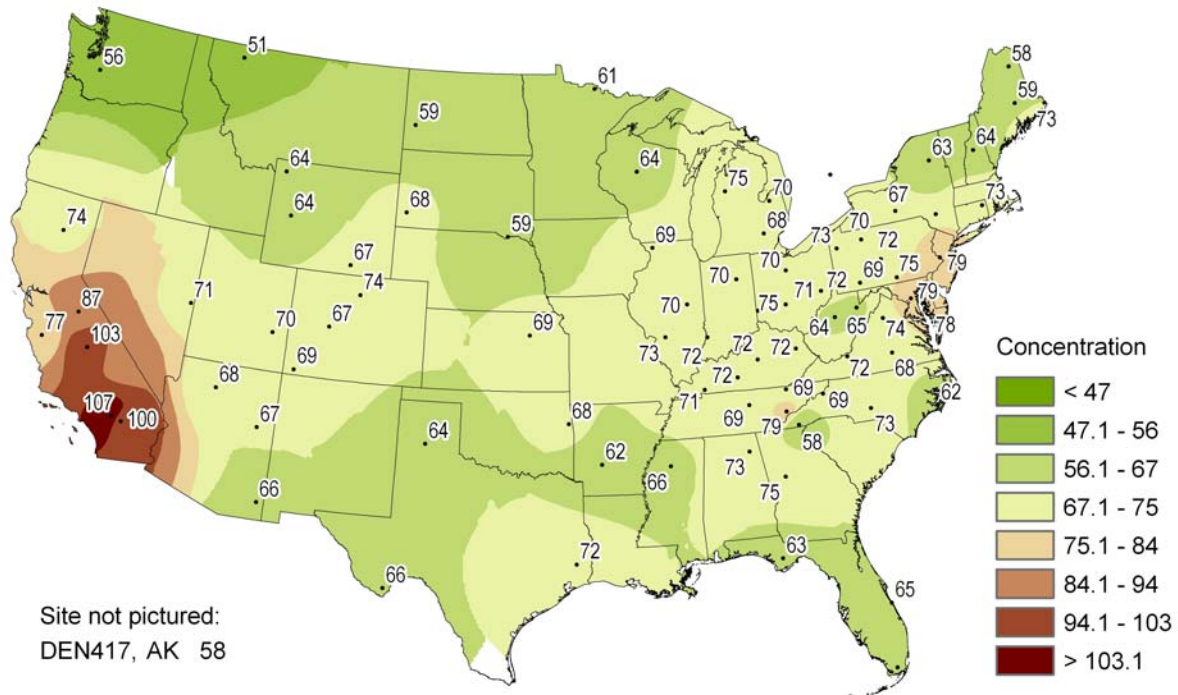
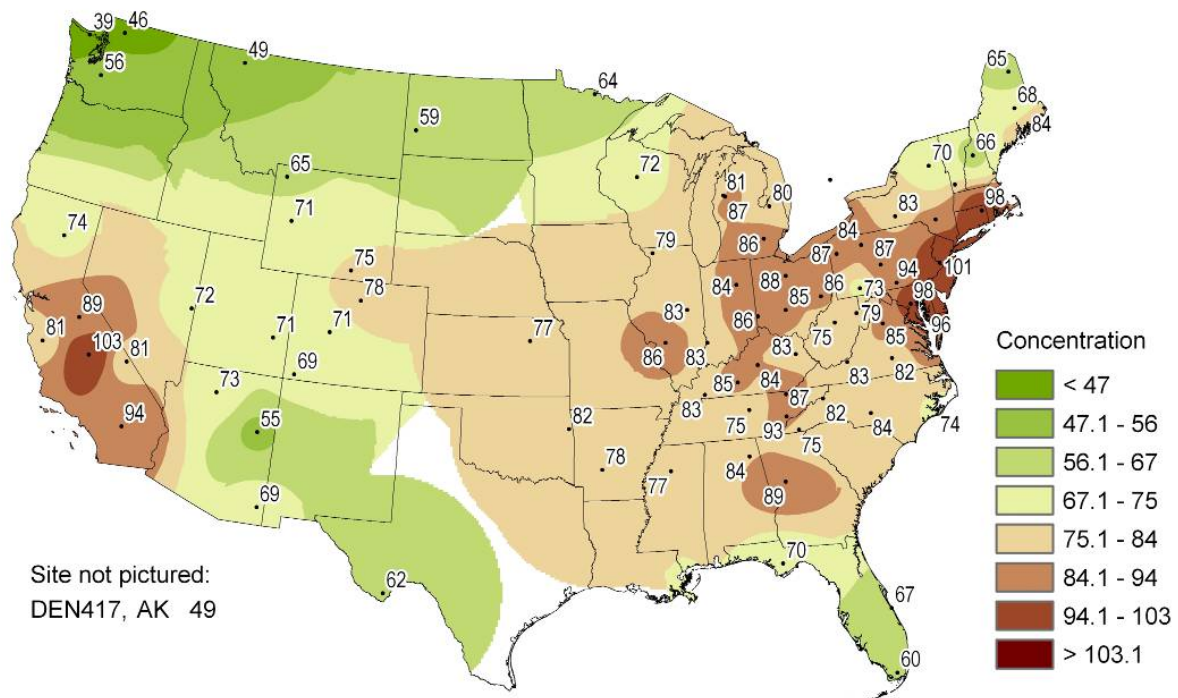


Figure 4-2 Three-year Average of Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) for 2000–2002



Three-year averages of the fourth highest daily maximum 8-hour average O₃ concentrations for 2000–2002 are presented in Figure 4-2 for comparison to 2007–2009 design values. The period 2000–2002 was selected because the NO_x SIP Call/NBP began in 2003 for the eastern United States. The O₃ data for these two 3-year periods illustrates the effectiveness of the emission reduction programs. Based on the 1997 NAAQS level (84 ppb), 19 eastern and 3 California sites recorded 3-year averages greater than or equal to 85 ppb during 2000–2002. The regions with elevated concentrations were located along the East Coast 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 the greater Atlanta area. Using the 2008 standard level (75 ppb), the map in Figure 4-2 shows that most of the eastern sites measured O₃ concentrations greater than 75 ppb. Five monitors in California and the site at Rocky Mountain National Park (ROM406) also recorded concentrations greater than 75 ppb. The period 2007–2009 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.

Measurements of 8-hour average O₃ concentrations during 2009 (Figure 4-3) were lower than 2008 and considerably lower than concentrations measured during the period 2000–2002. No eastern CASTNET sites measured concentrations above 75 ppb. Four sites in California measured O₃ concentrations greater than 75 ppb. The four California sites include YOS404, SEK430, CON186, and JOT403. Three of the four sites measured concentrations above 84 ppb. The monitor at CON186, CA measured the highest value (108 ppb) in the network during 2009. The year 2009 is the first year when no eastern CASTNET sites measured 8-hour average O₃ concentrations greater than 75 ppb.



Laurel Hill State Park, PA (LRL117)

Figure 4-3 Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) for 2009

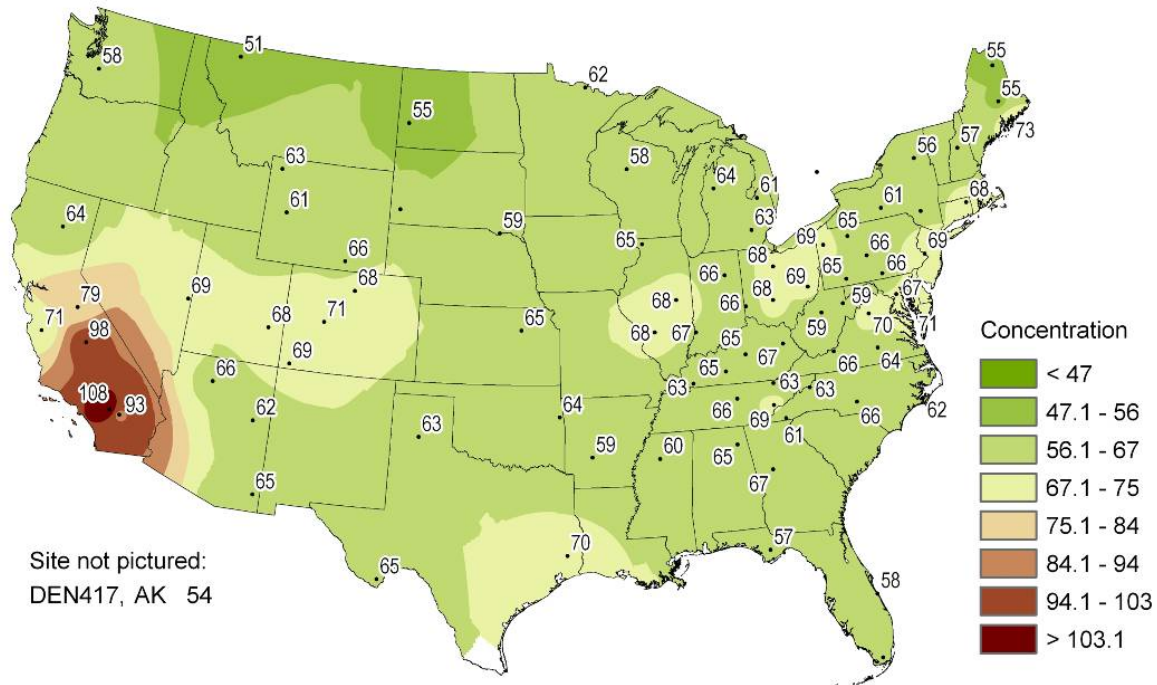
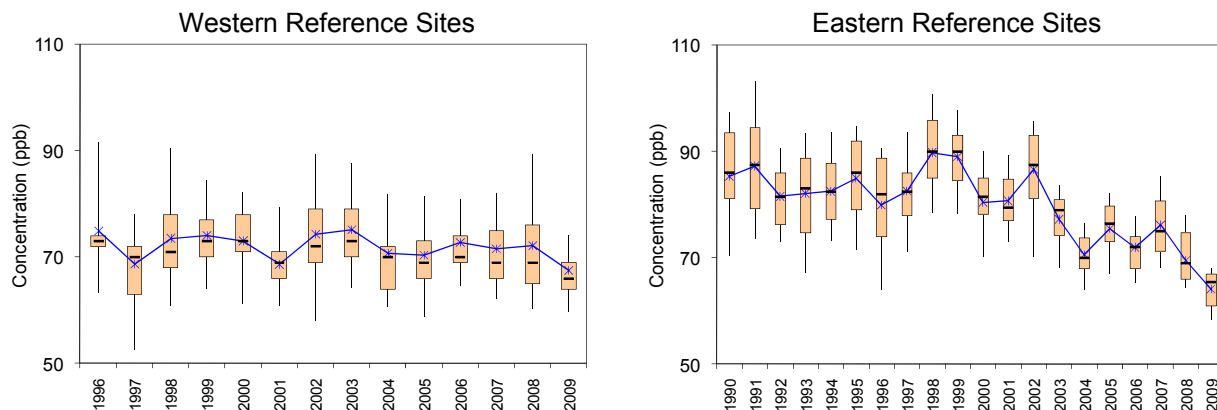


Figure 4-4 provides box plots depicting trends in fourth highest daily maximum 8-hour average O₃ concentrations from the 34 CASTNET eastern reference sites (right side) and 17 western reference sites (left side). The eastern O₃ data show a reduction since 2002. The mean aggregated value (64 ppb) for the eastern reference sites for 2009 was the lowest in the history of the network. It represents a substantive improvement over the mean value of 90 ppb measured in 1998. The box plots constructed from the aggregated western O₃ sites show no trend. The 2007–2009 average of the fourth highest daily maximum 8-hour average O₃ concentrations for the western reference sites was 70 ppb.

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



W126 Values for 2009

The W126 O₃ exposure index is a biologically-based cumulative exposure index (Lefohn and Runeckles, 1987) that was described and used in the EPA's Ozone Criteria Documents (EPA, 1996; 2006) for both characterizing O₃ trends and relating vegetation yield reduction losses with O₃ exposure. W126 is a seasonal metric derived from the sum of weighted hourly values measured during the 12-hour daylight period during the O₃ season, with the higher hourly concentrations being weighted more heavily than lower concentrations (EPA, 2010c). Many researchers (e.g., Musselman *et al.*, 2006) concluded that both peak concentrations and cumulative effects are important in relating vegetation damage to atmospheric O₃. W126 characterizes high concentrations over a seasonal 3-month exposure period.

The W126 index is represented as the sum of all hourly O₃ concentrations, where each measured concentration is weighted by a function, w_i , to assign greater emphasis to the higher measured concentrations. This weighting function provides a weighting value that is unique for each hourly O₃ concentration. The weighting function, as described by Lefohn *et al.* (1988) is:

$$\text{Hourly Weighting} = w_i = \frac{1}{1 + 4403 \exp(-0.126c_i)} \quad \text{Equation 4-1}$$

Where:

w_i = weighting value for hourly concentration i , and
 c_i = hourly concentration i in ppb.

Each hourly weighting value is multiplied by its corresponding hourly concentration. This product is summed over all valid hours to calculate the W126 exposure. Thus, the W126 exposure is:

$$W126 = \sum_{i=1}^n w_i c_i \quad \text{Equation 4-2}$$

Where:

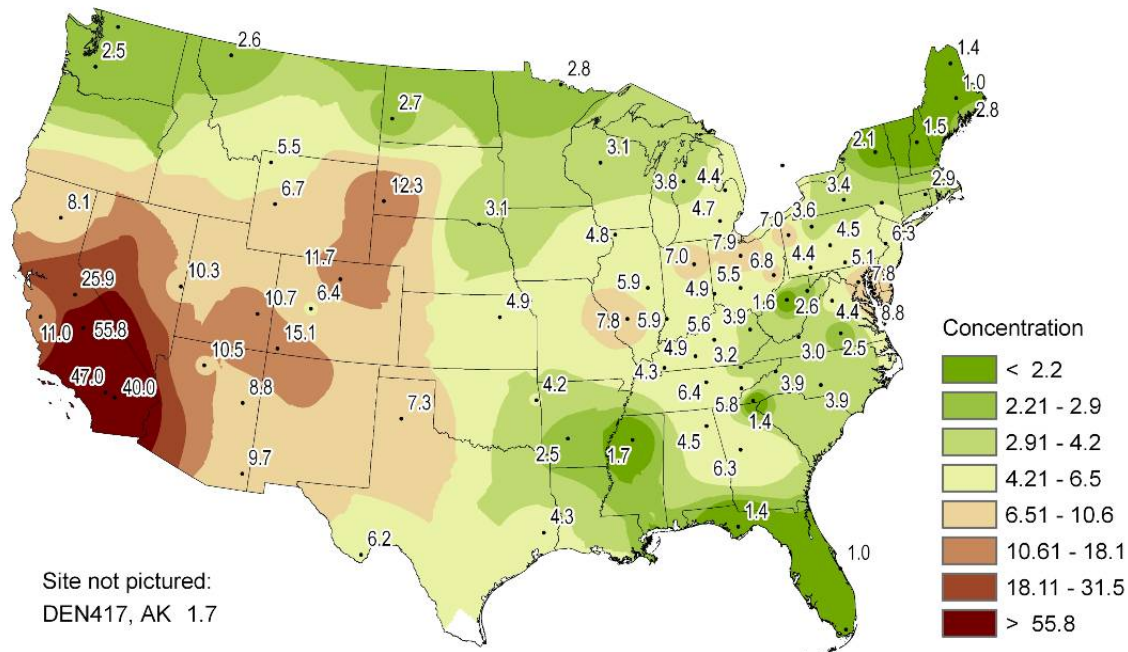
n = number of hours with valid O₃ concentrations.

The exposure unit is ppm-hours. For this report, W126 exposures were calculated using O₃ values measured during the 12 hours from 8:00 a.m. to 8:00 p.m. in the months of May through September of each year.

CASTNET hourly O₃ concentrations were used to calculate W126 values for 2009. First, the daily index value for each site was calculated. The daily index values were then used to calculate the monthly index value for the months of May through September. The highest of the rolling 3-month sums for a consecutive 3-month period was the W126 value for each site.

W126 levels for 2009 are presented in Figure 4-5. The higher W126 values were measured in California and at western sites in high terrain. O₃ concentrations measured at high elevation sites (MACTEC, 2003) typically show a flat diurnal pattern with little variability from hour to hour because dry deposition and scavenging of O₃ by nitric oxide are low at night. Nighttime dry deposition is low because shallow boundary layers typically do not form at elevated sites, and scavenging is low because little fresh NO is available to react with the existing O₃. The persistence of moderate O₃ concentrations at night produces steady exposure and high W126 levels.

Figure 4-5 W126 Values (ppm-hour) for 2009



Changes in Ozone Design Values

A design value is a statistic that describes the air quality status of a given area relative to the level of the NAAQS. Design values are typically used to classify nonattainment areas, assess progress towards meeting the NAAQS, and develop control strategies to achieve the NAAQS. Because the NAAQS for O₃ are written in terms of the 3-year average of the fourth highest daily maximum 8-hour average concentrations, design values change as each new 3-year database of monitored O₃ concentrations becomes available.

CASTNET was not designed to operate as a network for demonstration of compliance with O₃ NAAQS. However, the network provides data that can be evaluated in the context of the O₃ NAAQS, and the data can be used to provide reasonable estimates of design values.

Figures 4-1 and 4-2 in this chapter provide maps of design values for 2007–2009 and 2000–2002, respectively. For example, the design value for Beltsville, MD (BEL116) for 2007–2009 was 79 ppb. This value would have to be reduced to 75 ppb to achieve the level of the 2008 standard.

No sites in the East measured design values greater than or equal to 85 ppb in 2009 or 2007–2009 though four sites had values above 75 ppb. Figure 4a provides a time series of annual design values from 2000–2002 through 2007–2009 for three eastern CASTNET sites, which are Abington, CT (ABT147); Beaufort, NC (BFT142);

and Bondville, IL (BVL130), and Figure 4b shows design values for three western CASTNET sites, Grand Canyon National Park, AZ (GRC474); Sequoia National Park, CA (SEK430); and Converse Station, CA (CON186). These two figures demonstrate the progress made toward achieving the 2008 NAAQS level of 0.08 ppm (84 ppb in practice). ABT147, CT began the 10-year period with a 3-year design value of 99 ppb. The 2007–2009 value was 76 ppb. The other two eastern sites in Figure 4a are below the 2008 NAAQS level. Figure 4a shows design values above 100 ppb for the two California sites. The design value measured at CON186, CA decreased from 128 ppb in 2001–2003 to 107 ppb in 2007–2009. The site at the Grand Canyon shows a level below the 2008 standard.

Figure 4a Trends in Ozone Design Values

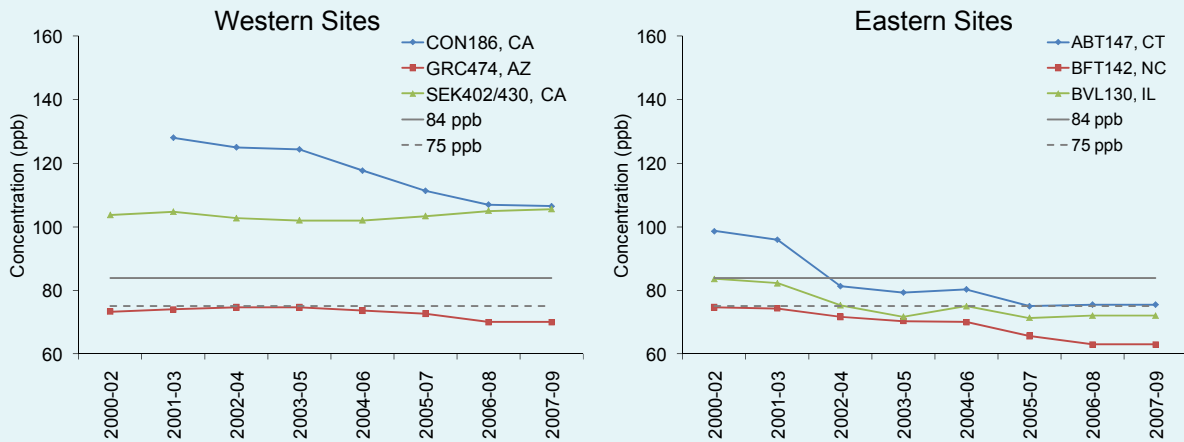
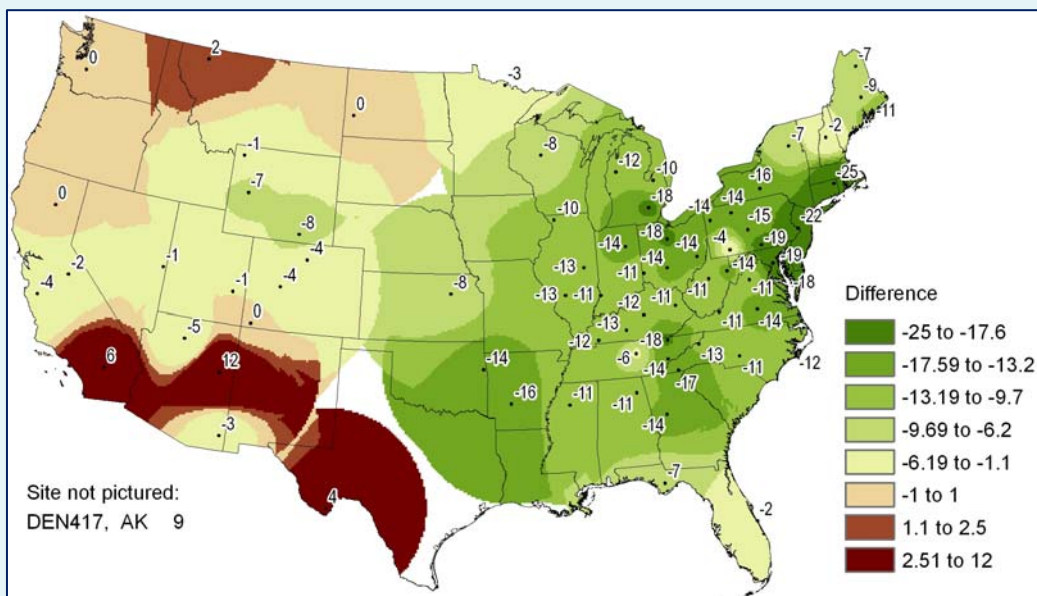


Figure 4b was constructed by taking the difference (in ppb) between the design values at each CASTNET site from 2000–2002 to 2007–2009. Negative values indicate an improvement in O₃ air quality. The map shows widespread reduction in O₃ concentrations in the eastern United States. In fact, every eastern site shows a reduction ranging from -2 to -25 ppb. The figure shows a mixture of some increasing and many decreasing O₃ values at the western sites.

Figure 4b Change (ppb) in Ozone Design Values 2000–2002 to 2007–2009



Chapter 5: Data Quality

Data quality indicators (DQI) such as precision, accuracy, and completeness are used to assess CASTNET measurements and supporting activities. Routine assessment helps guarantee the production of high-quality information to meet project objectives. Measurements taken during 2009 and historical data collected over the period 1990–2008 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 2009 QA program elements are documented in the CASTNET Quality Assurance Project Plan (QAPP), Revision 4.1 (MACTEC, 2008). The QAPP is comprehensive and includes standards and policies for all components of project operation from site selection through final data reporting. Sections on field measurements, chemical analysis of field samples, data management, and assessments and response actions are included. Standard operating procedures for all aspects of CASTNET operations are provided in the appendices.

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 2009 data were analyzed and compared with historical data collected during the period 1990 through 2008. The information in this report is supplemented by analyses that are discussed in quarterly CASTNET Quality Assurance Reports (MACTEC, 2009b; 2009d; 2009e; 2010b). 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 filter pack pollutants. Precision is expressed as mean absolute relative percent difference (MARPD). Table 5-1 lists the precision and accuracy criteria for laboratory filter pack measurements. Precision DQI criteria for filter pack concentrations changed during third quarter 2009. Previously, DQI criteria required a MARPD of 5.0 percent for all collocated CASTNET filter pack measurements, except for NH_4^+ , which had a criterion of 10.0 percent MARPD. Beginning on August 11, 2009, the DQI criteria were changed to a

single MARPD value of 20.0 percent. Since the new criterion became effective during the middle of the third quarter, this report uses the old criteria to evaluate filter pack precision.

Figure 5-1 Historical and 2009 Precision Data for Atmospheric Concentrations

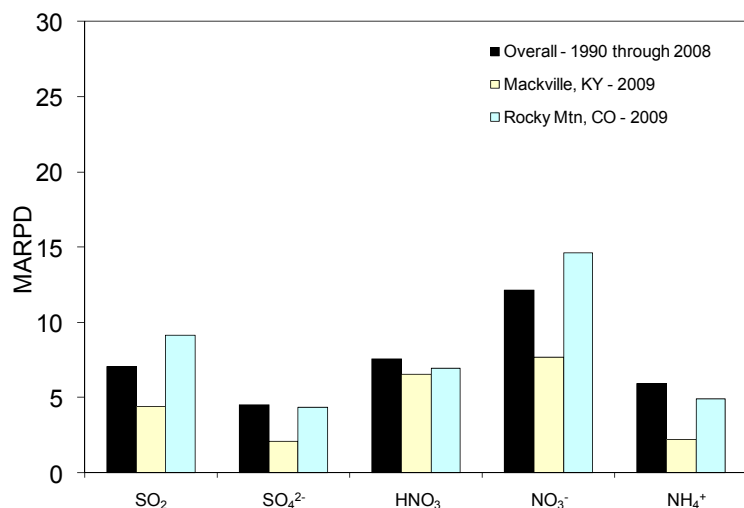


Table 5-1 Data Quality Indicator Criteria for CASTNET Laboratory Measurements

Analyte	Medium	Method	Through 08-10-09 Precision ¹ (MARPD)	Beginning 08-11-09 Precision ¹ (MARPD)	Accuracy ² (%)	Nominal Reporting Limits	
						mg/l	µg/Filter
Ammonium (NH ₄ ⁺)	F	AC	10	20	90 – 110	0.020*	0.5
Sodium (Na ⁺)	F	ICP-AES	5	20	95 – 105	0.005	0.125
Potassium (K ⁺)	F	ICP-AES	5	20	95 – 105	0.006	0.15
Magnesium (Mg ²⁺)	F	ICP-AES	5	20	95 – 105	0.003	0.075
Calcium (Ca ²⁺)	F	ICP-AES	5	20	95 – 105	0.006	0.15
Chloride (Cl ⁻)	F	IC	5	20	95 – 105	0.020	0.5
Nitrate (NO ₃ ⁻)	F	IC	5	20	95 – 105	0.008*	0.2
Sulfate (SO ₄ ²⁻)	F	IC	5	20	95 – 105	0.040	1.0

Note: ¹ This column lists precision goals for both network precision calculated from collocated filter samples and laboratory precision based on replicate samples.

² This column lists laboratory accuracy goals based on reference standards and continuing calibration verification spikes. The criterion is 90-110 percent for ICP-AES reference standards.

- F = filter pack samples
- AC = automated colorimetry
- ICP-AES = inductively coupled plasma-atomic emission spectrometry
- IC = ion chromatography
- MARPD = mean absolute relative percent difference
- L = liter
- µg/Filter = micrograms per filter
- * = as nitrogen

Historical (1990 through 2008) data for all 11 collocated site pairs operated over the history of the network and the 2009 data for the current collocated sites at Mackville, KY (MCK131/231) and Rocky Mountain National Park, CO (ROM406/206) are provided in Figure 5-1. 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 Cl⁻ are excluded from Figure 5-1 but are shown later in Figure 5-2. The historical results vary from just over 4 percent for particulate SO₄²⁻ to more than 12 percent for particulate NO₃. The historical MARPD for SO₄²⁻ met the 5 percent precision criterion for the CASTNET filter pack measurements shown in Table 5-1. The historical results for SO₂ and HNO₃ were above the 5 percent criterion but are considered reasonable. The results for NH₄⁺ met the goal of 10 percent. The results for NO₃ were significantly above the 5 percent goal. Historically, the precision of NO₃ measurements has been consistently worse than for the other analytes, possibly because NO₃ and HNO₃ concentrations include sampling artifacts. The historical precision data met the new DQI criterion of 20 percent MARPD.

MARPD values were not calculated for SO₂, HNO₃, and total NO₃ for the fourth quarter for MCK131/231. These data are labeled “NC” in Table 5-2. Beginning on October 20, 2009, with approval from EPA, MACTEC changed the analysis protocol for the nylon filter fraction analyses for the remainder of 2009 to perform a short study. Samples for weeks 42 through 52 were extracted using the Teflon filter extraction fluid (deionized water) and included automated colorimetry (AC) analyses for NH₃ + NH₄⁺ (as N) in addition to the regular IC analyses for SO₂ as SO₄²⁻ and HNO₃ as N. Because of this change in extraction fluid, the three parameters were invalidated and precision was not calculated.

The 2009 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 parameters. Three parameters (SO₂, SO₄²⁻, and NH₄⁺) met the MARPD precision criteria. The results for HNO₃ were above the 5 percent criterion but are considered reasonable. The 2009 data for MCK131/231 met the new criterion of 20 percent. The 2009 results for ROM406/206 showed better precision than historical results for SO₄²⁻, HNO₃, and NH₄⁺. The results for SO₄²⁻ and NH₄⁺ met their respective DQI criteria. The 5 percent precision criterion for NO₃ was not met at either site during 2009. All the parameters met the new 20 percent criterion. The MARPD results were higher for ROM406/206 because of lower concentrations measured at this site. Overall, the filter pack precision results for 2009 were about the same as the 2008 results.

Historical and 2009 precision statistics for four metal cations and 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. The 2009 precision results show that Na⁺ and Cl⁻ met the 5 percent precision criterion at MCK131/231, and Cl⁻ met the 5 percent MARPD criterion at ROM406/206. The overall results for four of five ions were reasonable, and they met the new DQI criterion of 20 percent MARPD.

The MARPD values for K^+ exceeded the former and new DQI criteria at both sites. Several pairs of K^+ concentrations were different by as much as a factor of 2.0 during the year. The poor results are currently being investigated. The 2009 precision results were about the same as for 2008.

Table 5-2 summarizes 2009 precision results by quarter for the two sets of collocated sites. See the 2009 Quarterly Data Reports (MACTEC, 2009a; 2009c; 2010a; 2010c) and QA Quarterly Reports (MACTEC, 2009b; 2009d; 2009e; 2010b) for discussions of quarterly precision data.

Figure 5-2 Historical and 2009 Precision Data for Cation and Cl⁻ Concentrations

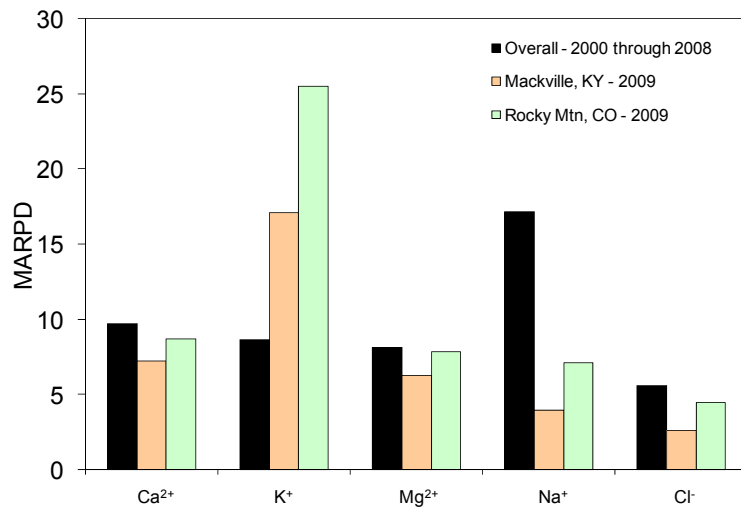
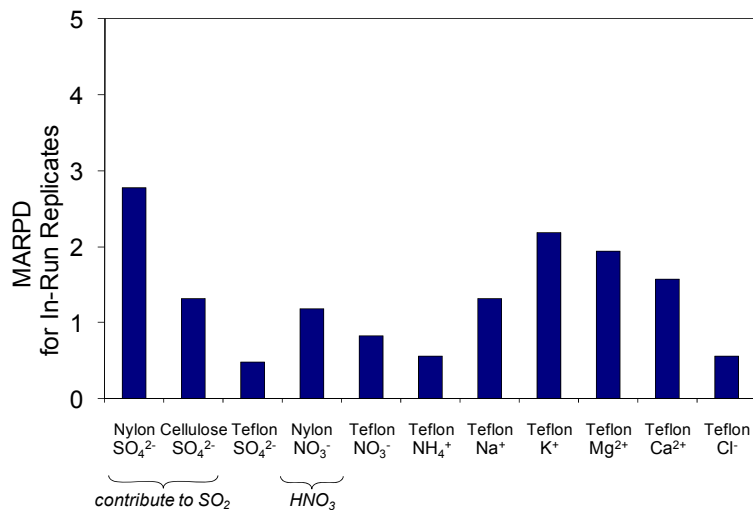


Table 5-2 Collocated Precision Results (MARPD) for 2009 Filter Pack Data by Quarter

Site Pairs	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	Cl ⁻	HNO ₃	SO ₂	Total NO ₃ ⁻
MCK131/231, KY											
Quarter 1	1.80	3.77	1.65	8.84	4.97	2.89	14.76	5.79	2.95	1.78	2.78
Quarter 2	2.52	9.89	2.49	6.17	6.62	3.53	6.09	1.59	4.50	3.23	5.54
Quarter 3	1.44	10.47	1.75	5.81	6.54	3.35	22.95	0.25	6.41	6.99	5.52
Quarter 4	2.69	6.61	2.88	8.07	6.96	5.89	24.59	2.72	NC	NC	NC
2009	2.11	7.68	2.19	7.22	6.27	3.92	17.10	2.59	4.62	4.00	4.61
ROM406/ROM206, CO											
Quarter 1	4.15	15.16	3.57	9.42	6.55	9.57	16.43	2.12	5.96	8.61	5.61
Quarter 2	5.29	9.69	5.35	5.08	6.82	5.47	17.36	5.51	7.16	16.06	4.94
Quarter 3	1.92	11.89	3.90	8.21	8.07	5.09	21.47	5.85	4.95	3.54	3.34
Quarter 4	6.01	21.83	6.80	12.03	9.86	8.26	46.70	4.20	9.62	8.31	10.61
2009	4.34	14.64	4.90	8.68	7.82	7.09	25.49	4.42	6.92	9.13	6.12

The 2009 analytical precision results for eight 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 original concentration results. The laboratory precision data met the 5 percent measurement criterion listed in Table 5-1.

Figure 5-3 Precision Results for Laboratory Replicate Samples (2009)



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, 2010b; 2008b). 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 2009. 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 2009. The data show the historical and 2009 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.

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

Measurement Parameter	Method	MAD/MARPD	Criteria *	
			Precision	Accuracy
Wind Speed	Anemometer	MAD	± 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	MAD	± 5°	± 5°
Sigma Theta	Wind Vane	MAD	Undefined	Undefined
Relative Humidity	Thin Film Capacitor	MARPD	± 10% (of full scale)	± 10 percent
Solar Radiation	Pyranometer	MARPD	± 10% (of reading taken at local noon)	± 10%
Precipitation	Tipping Bucket Rain Gauge	MARPD	± 10% (of reading)	± 0.05 inch [†]
Ambient Temperature	Platinum RTD	MAD	± 1.0°C	± 0.5°C
Delta Temperature	Platinum RTD	MAD	± 0.5°C	± 0.5°C
Surface Wetness	Conductivity Bridge	MAD	Undefined	Undefined
Ozone	UV Absorbance	MARPD	± 10% (of reading)	± 10%
Filter Pack Flow	Mass Flow Controller	MARPD	± 10%	± 5%

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

MARPD is the precision measure for percentage criteria such as 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 2009 Precision Data by Quarter for Ozone Concentrations

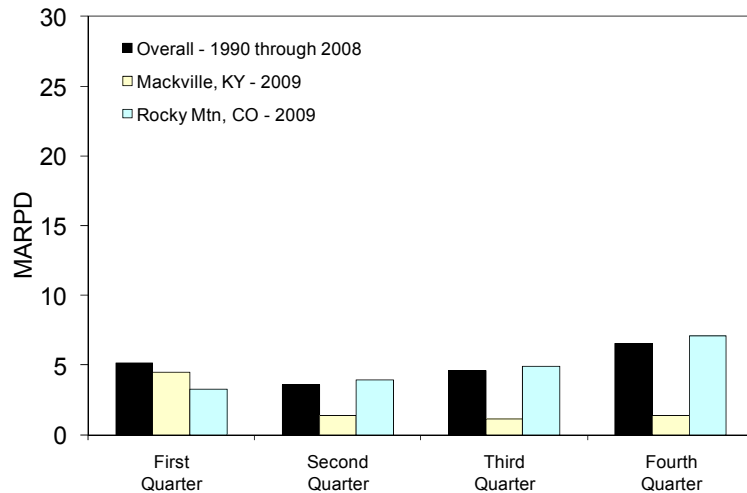


Table 5-4 Collocated Precision Results (MARPD) for 2009 Ozone Concentrations by Quarter

Sampling Period	MCK131/231, KY	ROM406/206, CO
Quarter 1	4.47	3.28
Quarter 2	1.40	3.92
Quarter 3	1.13	4.90
Quarter 4	1.38	7.09
2009	2.09	4.80

Historical Precision of Daily Maximum 8-Hour Average O₃ Concentrations

Precision is a DQI for CASTNET continuous measurements including O₃ values. Precision is calculated as the MARPD of hourly concentration data from collocated sites. The O₃ precision goal is a MARPD of less than or equal to 10 percent. CASTNET sites at Mackville, KY (MCK131/231) and Rocky Mountain National Park, CO (ROM406/206) currently host collocated O₃ analyzers. Nine other CASTNET sites have run collocated O₃ analyzers over the history of the network. MCK131/231 has operated collocated analyzers since 1992 and ROM406/206 since 2001. MCK131 is considered the primary site of the MCK131/231 pair. ROM406 is considered the primary site of the Rocky Mountain pair and is operated on behalf of NPS. The two sites at Rocky Mountain National Park are operated independently. ROM206 is operated on behalf of EPA.

The two Rocky Mountain sites are served by different site operators and calibration technicians, have operated different instruments, and currently have different sampling configurations. Until recently, ROM206 did not meet the EPA SLAMS monitoring requirements, as described in 40 CFR Part 58, Appendix A (EPA, 2010b). ROM406 met the site configuration requirements but not all of the contemporaneous operational requirements including frequency of calibrations. In any event, the operation of the collocated NPS and EPA O₃ analyzers at ROM406/206 provides an opportunity to evaluate the precision of independent systems. A discussion of the precision of 2009 hourly O₃ concentrations is presented in this chapter. Both sites met the 10 percent precision goal for each quarter of 2009 (Table 5-4). Another useful measure of precision is the MARPD of daily maximum 8-hour average (DM8A) O₃ concentrations since these data are used to gauge compliance with the NAAQS. The NAAQS for O₃ are written in terms of the 3-year average of the fourth-highest daily maximum 8-hour average O₃ concentration. Table 5a presents MARPD values based on DM8A O₃ levels for the two collocated sites over the period 2002 through 2009. The MARPD values were within 10 percent except for the 2007 ROM406/206 results. The cause of the problems in 2007 was not determined. During 2007, zero/span/ precision (z/s/p) checks and calibration results were acceptable. Audit results from an independent audit were also acceptable, although the problems began shortly after completion of the site audit.

Table 5a Ozone MARPD Values Based on DM8A Levels

Year	\bar{x}	\bar{y}	MARPD
MCK131/231, KY			
2002	43.7	44.5	3.3
2003	41.9	41.5	1.5
2004	40.2	40.3	1.3
2005	43.7	43.5	2.0
2006	47.9	45.8	5.8
2007	47.4	47.1	3.4
2008	39.8	39.2	6.4
2009	40.0	39.7	1.9
ROM406/206, CO			
2002	56.4	53.5	5.1
2003	54.5	51.0	6.4
2004	52.0	48.0	8.1
2005	49.9	50.0	5.9
2006	53.7	51.0	6.4
2007	53.0	47.8	10.8
2008	51.5	51.1	1.8
2009	51.2	48.3	5.8

Note: \bar{x} is the mean DM8A value for the primary site and \bar{y} for the collocated site.

Table 5b provides overall MARPD values based on hourly O₃ concentrations for every collocated monitoring site over the history of the network. The MARPD values for the entire period for MCK131/231 and ROM406/206 are 1.3 percent and 3.5 percent.

Table 5b Overall Ozone MARPD Values for all Collocated Sites

Site	Annual MARPD	Years Collocated	Site	Annual MARPD	Years Collocated
ALH157/257, IL	0.7	1	MCK131/231, KY*	1.3	17
ARE128/228, PA	1.2	6	PAR107/207, WV	0.4	1
ASH135/235, ME	1.5	7	RCK163/263, ID	0.9	3
CHA167/267, AZ	0.9	3	ROM406/206, CO*	3.5	9
DCP114/214, OH	2.4	3	SUM156/256, FL	2.7	4
GAS153/253, GA	0.6	1			

Note: *Currently active collocated sites

Figure 5a Fourth Highest Daily Maximum 8-Hour Average Concentration (ppb) MCK131/231, KY

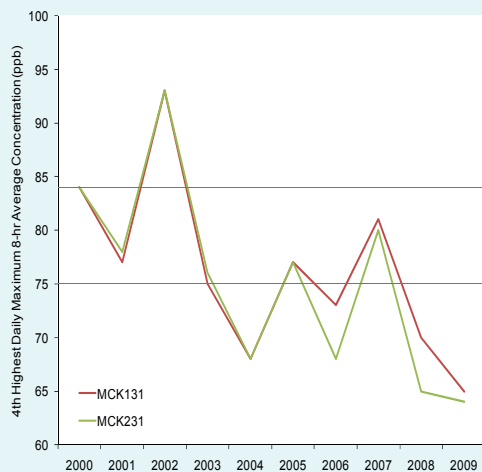
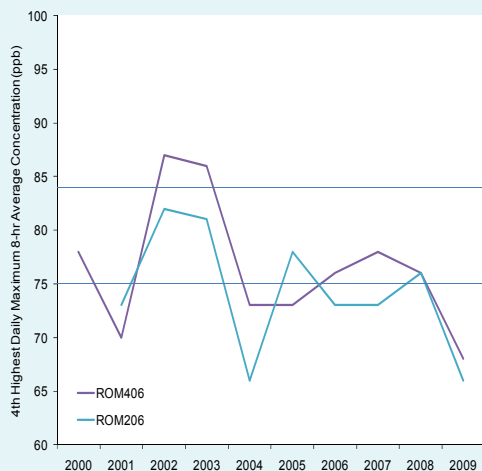


Figure 5b Fourth Highest Daily Maximum 8-Hour Average Concentration (ppb) ROM406/206, CO



Another way of evaluating the collocated data is to compare fourth-highest DM8A O₃ concentrations for both collocated sites for each year. Figure 5a presents these results for MCK131/231 for 2000 through 2009. Figure 5b presents the results for ROM406/206 for 2001 through 2009. The figures also depict the 2008 and 1997 NAAQS of 75 ppb and 85 ppb. The results from the two analyzers at MCK131/231 track well. In particular, both analyzers showed NAAQS exceedances for the same years. The worst years for precision were 2006 and 2008 when data completeness was lower than normal.

Although the fourth highest values for the ROM406/206 site pair were generally close, the results from ROM206 were generally lower. At times ROM406 showed exceedances, and ROM206 did not. For example, ROM406 measured a fourth-highest DM8A value of 76 ppb in 2006, and ROM206 recorded 73 ppb. These results led to the analysis of the instruments, sampling systems, and data processing procedures at the two Rocky Mountain monitoring sites. Historically, ROM206 was not operated for regulatory monitoring purposes and did not meet SLAMS requirements. The ROM206 system did not include a second O₃ analyzer as a transfer standard. In 2010, the O₃ sampling system at ROM206 was replaced with a system that uses a separate site analyzer and site transfer standard and is otherwise configured to comply with 40 CFR Part 58 requirements.

The review of the ROM406/206 sampling systems also showed that differences in O₃ sampling trains may explain the consistent differences in concentrations. The ROM406 sampling line has a diameter of 1/4 inch and does not utilize an inline filter and knockout bottle. The ROM206 sampling train includes an inline filter and knockout bottle. Until recently, the ROM206 sampling line had a diameter of 3/8 inch. The wider sampling line combined with the inline filter and knockout bottle resulted in a longer residence time in the ROM206 O₃ system, which could result in collection of some O₃ in the sampling system, perhaps resulting in lower concentrations measured by the analyzer. The 3/8-inch tubing was subsequently replaced with 1/4 inch tubing. The concentration differences might also be explained by differences in the altitude of the location where the O₃ analyzers are certified, even though all analyzers utilize an altitude compensation algorithm. The ROM406 analyzer and transfer standards are certified at an altitude near the actual location of the monitoring system while the ROM206 analyzer standards are certified at a location near sea level, a difference of approximately 2,700 meters.

The assessment of the ROM406/206 O₃ measurements and sampling systems showed the importance of collocated measurements and replacing older equipment when failure rates increase. Records indicate that use of older equipment results in extended periods of lost data and requires more significant efforts to troubleshoot, maintain, and replace instruments than any other cause. O₃ measurements are sensitive to sampling train differences (materials, integrity, and length). To maximize precision of O₃ measurements, sites must be standardized for sampling train materials, configuration, and minimum residence time, and sampling train integrity must be properly maintained and routinely monitored for leaks, moisture, and cleanliness.

Continuous Measurements

Precision criteria for the continuous measurements are listed in Table 5-3. Figure 5-5 provides precision results for historical data (1990 through 2008) at all collocated site pairs and 2009 precision statistics for the two current collocated site pairs. Precision was calculated in terms of the MARPD or mean absolute difference (MAD) of hourly measurements. All historical precision results met the DQI criteria. Table 5-5 gives precision data by quarter for 2009 for the two collocated sites. The results show excellent instrument precision. All but five 2009 annual and quarterly precision results met the DQI criteria. The third and fourth quarter and annual MARPD values for solar radiation at MCK131/231 did not meet the 10 percent goal. The first quarter MARPD value for relative humidity and the fourth quarter MARPD value for solar radiation at ROM406/206 did not meet the 10 percent criterion.

Figure 5-5 Historical and 2009 Precision Data for Continuous Measurements

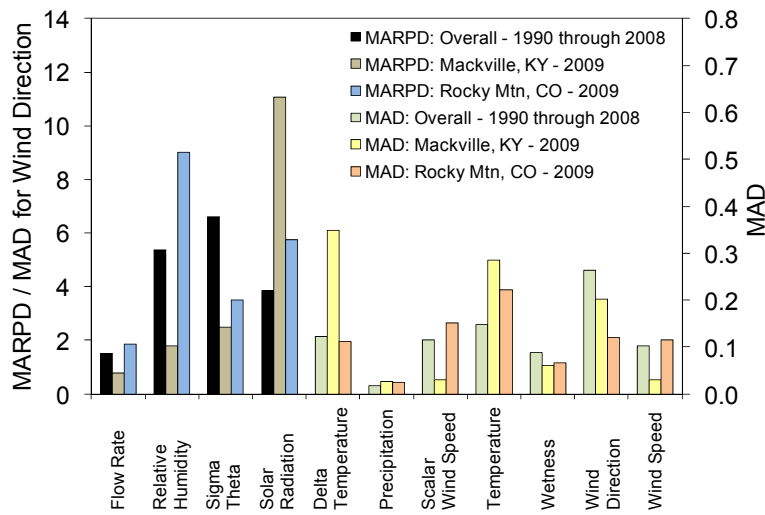


Table 5-5 Collocated Precision Results for 2009 Continuous Measurements by Quarter

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	2.39	3.16	2.58	5.85	0.56	0.03	0.04	0.49	0.04	3.18	0.04
Quarter 2	0.66	1.17	2.47	7.28	0.32	0.04	0.03	0.27	0.06	3.84	0.03
Quarter 3	0.07	0.79	3.11	15.30	0.25	0.02	0.02	0.22	0.08	3.52	0.02
Quarter 4	0.02	2.09	1.77	15.87	0.26	0.02	0.03	0.16	0.06	3.64	0.03
2009	0.79	1.80	2.48	11.07	0.35	0.03	0.03	0.29	0.06	3.55	0.03
ROM406/206, CO											
Quarter 1	1.00	13.74	2.83	6.40	0.07	0.02	0.14	0.09	0.06	2.06	0.13
Quarter 2	1.36	7.04	3.89	4.35	0.11	0.04	0.16	0.19	0.08	2.57	0.09
Quarter 3	2.35	5.42	NC	2.03	0.17	0.02	0.16	0.35	0.06	NC	NC
Quarter 4	2.76	9.90	3.82	10.29	0.09	0.02	0.14	0.26	0.07	1.72	0.13
2009	1.87	9.03	3.51	5.77	0.11	0.02	0.15	0.22	0.07	2.12	0.11

Note: NC = not calculated

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. 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 2009.

Table 5-6 Filter Pack Quality Control Summary for 2009

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⁻	97.71	1.05	143	98.81	0.96	689	0.48	0.98	309
	NO ₃ ⁻	99.37	1.03	143	98.83	0.95	689	0.82	1.08	309
	NH ₄ ⁺	100.83	2.99	135	99.84	1.36	677	0.56	0.65	317
	Ca ²⁺	104.15	3.16	132	100.44	1.01	676	1.57	2.46	310
	Mg ²⁺	102.33	1.53	132	100.08	0.77	676	1.94	2.62	310
	Na ⁺	100.16	2.03	132	100.20	1.02	676	1.31	1.35	310
	K ⁺	98.10	2.87	132	100.06	0.84	676	2.18	2.80	310
	Cl ⁻	101.97	1.66	143	99.32	1.06	689	0.56	0.76	309
Nylon	SO ₄ ²⁻	98.82	1.08	139	99.13	1.22	668	2.77	3.17	307
	NO ₃ ⁻	100.15	1.28	139	99.39	1.27	668	1.18	1.69	307
Cellulose	SO ₄ ²⁻	98.65	1.24	179	99.43	1.14	691	1.31	1.57	336

Note: % R = percent recovery
RPD = relative percent difference

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 2009 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.

Table 5-7 Accuracy Results for 2009 Field Measurements

Parameter	Percent Within Criterion
Temperature (0°C)	99.1 percent
Temperature (ambient)	99.1 percent
Delta Temperature (0°C)	98.8 percent
Delta Temperature (ambient)	98.1 percent
Relative Humidity	100.0 percent
Solar Radiation	91.1 percent
Wind Direction North	95.7 percent
Wind Direction South	95.7 percent
Wind Speed < 5 m/s	100.0 percent
Wind Speed ≥ 5 m/s	90.4 percent
Precipitation	100.0 percent
Wetness (within 0.5 volts)	97.4 percent
Ozone Slope	98.2 percent
Ozone Intercept	98.2 percent
Flow Rate	96.6 percent

Note: °C = degrees Celsius
m/s = meters per second

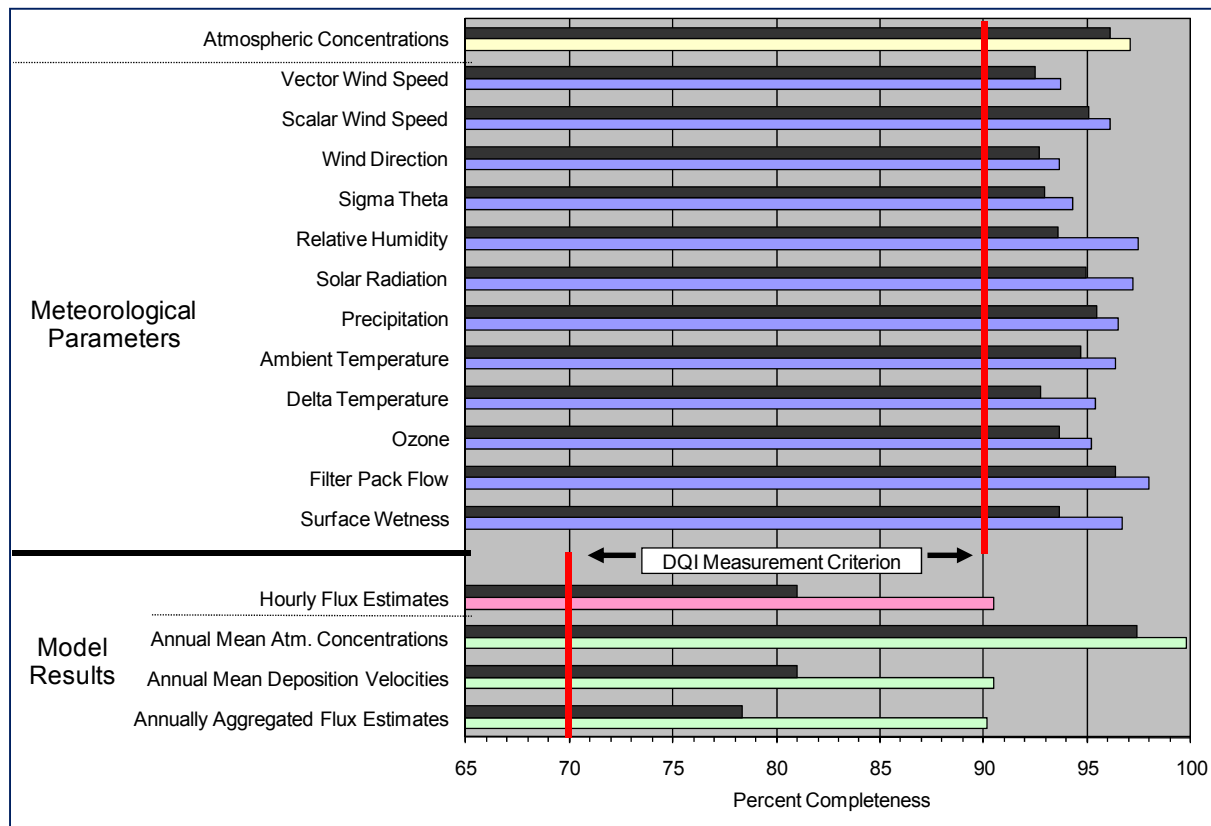


Vincennes, IN (VIN140)

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 2009 completeness data for all sites for measured filter concentrations and continuous measurements and calculated parameters. The figure shows that the 2009 direct measurements met the 90 percent completeness goal, and nine continuous measurements exceeded 95 percent, including filter pack flow. Atmospheric concentrations also exceeded 95 percent. The four parameters derived from model results exceeded 90 percent completeness for 2009. Completeness results for 2009 are better than historical results for all parameters.

Figure 5-6 Historical and 2009 Percent Completeness of Measurements and Modeled Estimates (black bars are 1990–2008)



Results for 2009 Environment Canada Proficiency Testing

The MACTEC laboratory is one of 33 laboratories that participated in the Environment Canada (ECAN) Proficiency Testing Program for Inorganic Environmental Substances during 2009 (<http://www.ec.gc.ca/inre-nwri/default.asp?lang=En&xml=7A20877C-359F-44C5-9497-EB904806131C>). Specifically, the MACTEC laboratory participates in the Rain and Soft Waters round robin studies, which consist of natural waters supplied by the National Laboratory for Environmental Testing. The laboratory receives ten samples of mixed rain and Canadian Shield waters for chemical analysis from ECAN every six months. The MACTEC laboratory reported the eight CASTNET parameters for samples in two studies (study codes PT 0094 and PT 0095) during 2009.

The participating laboratories' reported results are evaluated for systematic bias and precision. Systematic bias is evaluated using Youden (1969) non-parametric analysis, while precision is evaluated using algorithm A from International Organization for Standardization (ISO) standard 13528 [ISO 13528:2005(E); ISO, 2005].

Individual sample results are flagged based on the robust standard deviation obtained from the ISO 13528 computation. Sample results within 2 standard deviations of the assigned (median laboratory) value are not flagged, sample results between 2 and 3 standard deviations are given a warning flag, and sample results greater than 3 standard deviations from the assigned value are flagged as above the action limit (remedial action is required). Laboratory results are considered to be systematically biased when individual parameters are ranked by the Youden analysis as consistently and significantly higher or lower than the assigned value regardless of deviations from the assigned (median laboratory) value.

The MACTEC laboratory's 2009 ECAN results (ECAN, 2010) for the eight CASTNET parameters are presented in Table 5c below. All eight parameters received the highest possible rating of "ideal," indicating an unbiased score.

Table 5c MACTEC Results for ECAN Studies PT 0094 and PT 0095

Test Parameter	Analytical Method	Reference Method	Laboratory Performance Rating	
			PT Study 0094	PT Study 0095
Ammonia	AC	EPA Method 350.1	Ideal	Ideal
Calcium	ICP-AES	EPA Method 6010	Ideal	Ideal
Chloride	IC	EPA Method 300.0	Ideal	Ideal
Magnesium	ICP-AES	EPA Method 6010	Ideal	Ideal
Nitrate + Nitrite	IC	EPA Method 300.0	Ideal	Ideal
Potassium	ICP-AES	EPA Method 6010	Ideal	Ideal
Sodium	ICP-AES	EPA Method 6010	Ideal	Ideal
Sulfate	IC	EPA Method 300.0	Ideal	Ideal

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 2009. Precision data for SO_2 , SO_4^{2-} , HNO_3 , and NH_4^+ are considered acceptable. Precision data for NO_3^- 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 nitrogen species. The precision of K^+ filter pack measurements exceeded the established criteria and are not considered acceptable. The poor K^+ precision is under investigation.

The 2009 precision data for O_3 concentrations met the 10 percent DQI criterion. The 2009 O_3 precision data for ROM406/206 demonstrate the comparability of the two independent O_3 sampling systems. The precision of continuous measurements for 2009 was excellent. All but four quarterly and one annual precision result for 2009 met the DQI criteria.

Accuracy data met the established criteria for field and laboratory parameters. Completeness criteria were met for all parameters in 2009. Completeness results for 2009 are considered excellent and were better than historical results for all parameters. ECAN proficiency test studies (0094 and 0095) showed that the MACTEC laboratory performed well on the interlaboratory chemical analyses with all parameters rated ideal.



Alhambra, IL (ALH157)

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Appendix A

Locational and Operational Characteristics of CASTNET Sites

Table A-1 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	Nearby NADP Site	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Alabama												
SND152	Sand Mountain	12/27/88	34.2894	85.9704	352	•	•	AL99	Agri.	Rolling	Y	EPA
Alaska												
DEN417	Denali National Park	10/06/98	63.7258	148.9633	661	•	•	AK03	Forested	Complex	N	NPS
Arizona												
CHA467	Chiricahua National Monument	04/25/89	32.0092	109.3892	1570	•	•	AZ98	Range	Complex	N	NPS
GRC474	Grand Canyon National Park	05/16/89	36.0597	112.1822	2073	•	•	AZ03	Forested	Complex	M	NPS
PET427	Petrified Forest National Park	09/24/02	34.8225	109.8919	1723	•	•	AZ97	Desert	Flat	Y	NPS
Arkansas												
CAD150	Caddo Valley	10/04/88	34.1792	93.0989	71	•	•	AR03	Forested	Rolling	N	EPA
California												
CON186	Converse Station	06/17/03	34.1941	116.9130	1837	•	•	CA94	Agri./Forested	Complex	N	EPA
JOT403	Joshua Tree National Monument	02/16/95	34.0714	116.3906	1244	•	•	CA67	Desert	Complex	M	NPS
LAV410	Lassen Volcanic National Park	07/25/95	40.5403	121.5764	1756	•	•	CA96	Forested	Complex	M	NPS
PIN414	Pinnacles National Monument	05/16/95	36.4850	121.1556	335	•	•	CA66	Forested	Complex	M	NPS
SEK430	Sequoia National Park	04/07/05	36.4894	118.8269	457	•	•	CA75	Forested	Mountaintop	N	NPS
YOS404	Yosemite National Park	09/25/95	37.7133	119.7061	1605	•	•	CA99	Forested	Complex	N	NPS
Colorado												
GTH161	Gothic	05/16/89	38.9573	106.9854	2926	•	•	CO10	Range	Complex	N	EPA
MEV405	Mesa Verde National Park	01/10/95	37.1983	108.4903	2165	•	•	CO99	Forested	Complex	M	NPS
ROM206	Rocky Mountain National Park	07/03/01	40.2778	105.5453	2743	•	•	CO98	Forested	Complex	M	EPA
ROM406	Rocky Mountain National Park	12/20/94	40.2778	105.5453	2743	•	•	CO98	Forested	Complex	M	NPS

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Connecticut												
ABT147	Abington	12/28/93	41.8402	72.0111	209	•	•	CT15	Urban-Agri.	Rolling	M	EPA
Florida												
EVE419	Everglades National Park	10/06/98	25.3911	80.6806	2	•	• ⁴	FL11	Swamp	Flat	Y	NPS
IRL141	Indian River Lagoon	07/09/01	30.1065	80.4554	2	•	•	FL99	Beach	Flat	Y	EPA
SUM156	Sumatra	12/28/88	30.1065	84.9938	14	•	•	FL23	Forested	Flat	Y	EPA
Georgia												
GAS153	Georgia Station	06/28/88	33.1812	84.4100	270	•	•	GA41	Agri.	Rolling	M	EPA
Illinois												
ALH157	Alhambra	06/28/88	38.8690	89.6229	164	•	•	IL46	Agri.	Flat	Y	EPA
BVL130	Bondville	02/09/88	40.0520	88.3725	212	•	•	IL11	Agri.	Flat	Y	EPA
STK138	Stockton	12/28/93	42.2872	89.9998	274	•	•	IL18	Agri.	Rolling	M	EPA
Indiana												
SAL133	Salamonie Reservoir	06/28/88	40.8164	85.6608	250	•	•	IN20	Agri.	Flat	Y	EPA
VIN140	Vincennes	08/04/87	38.7406	87.4844	134	•	•	IN22	Agri.	Rolling	M	EPA
Kansas												
KNZ184	Konza Prairie	03/26/02	39.1021	96.6096	348	•	•	KS31	Prairie	Flat	Y	EPA
Kentucky												
CDZ171	Cadiz	10/01/93	36.7841	87.8500	189	•	•	KY99	Agri.	Rolling	M	EPA
CKT136	Crockett	08/24/93	37.9211	83.0658	455	•	•	KY35	Agri.	Rolling	Y	EPA
MAC426	Mammoth Cave National Park	07/24/02	37.1319	86.1478	243	•	•	KY10	Agri./Forested	Rolling	M	NPS
MCK131	Mackville	07/31/90	37.7044	85.0483	353	•	•	KY03	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	Nearby NADP Site	Primary Land Use	Terrain	Representative to the MLM ³	Sponsor
Maine												
ACA416	Acadia National Park	12/01/98	44.3769	68.2608	158	•	•	ME98	Forested	Complex	M	NPS
ASH135	Ashland	12/20/88	46.6039	68.4142	235	•	•	ME00	Agri.	Flat	Y	EPA
HOW132	Howland	11/24/92	45.2158	68.7085	69	•	•	ME09	Forested	Rolling	Y	EPA
Maryland												
BEL116	Beltsville	11/01/88	39.0283	76.8175	46	•	•	MD99	Urban-Agri.	Flat	N	EPA
BWR139	Blackwater National Wildlife Refuge	07/04/95	38.4448	76.1115	4	•	•	MD15	Forest-Marsh	Coastal	M	EPA
Michigan												
ANA115	Ann Arbor	06/28/88	42.4164	83.9019	267	•	•	MI52	Forested	Flat	M	EPA
HOX148	Hoxeyville	10/31/00	44.1809	85.7390	298	•	•	MI53	Forested	Flat	Y	EPA
UVL124	Unionville	06/28/88	43.6139	83.3597	201	•	•	MI51	Agri.	Flat	Y	EPA
Minnesota												
VOY413	Voyageurs National Park	06/13/96	48.4128	92.8292	429	•	•	MN32	Forested	Rolling	M	NPS
Mississippi												
CVL151	Coffeeville	12/27/88	34.0028	89.7989	134	•	•	MS30	Forested	Rolling	M	EPA
Montana												
GLR468	Glacier National Park	12/27/88	48.5103	113.9956	976	•	•	MT05	Forested	Complex	N	NPS
Nebraska												
SAN189	Santee Sioux	07/05/06	42.8292	97.8541	429	•	•	SD99	Agri.	Rolling	N	EPA
Nevada												
GRB411	Great Basin National Park	05/16/95	39.0053	114.2158	2060	•	•	NV05	Forested	Complex	M	NPS
New Hampshire												
WST109	Woodstock	12/27/88	43.9446	71.7008	258	•	•	NH02	Forested	Complex	N	EPA

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New Jersey												
WSP144	Washington's Crossing	12/27/88	40.3133	74.8726	61	•	•	NJ99	Urban-Agri.	Rolling	M	EPA
New York												
CAT175	Claryville	05/10/94	41.9423	74.5519	765	•	• ^{4,5}	NY68	Forested	Complex	N	EPA
CTH110	Connecticut Hill	09/28/87	42.4010	76.6535	515	•	•	NY67	Forested	Rolling	N	EPA
HWF187	Huntington Wildlife Forest	05/28/02	43.9732	74.2232	502	•	•	NY20	Forested	Complex	N	EPA
North Carolina												
BFT142	Beaufort	12/28/93	34.8843	76.6213	2	•	•	NC06	Agri.	Flat	Y	EPA
CND125	Candor	09/25/90	35.2643	79.8382	198	•	•	NC36	Forested	Rolling	M	EPA
COW137	Coweeta	11/04/87	35.0605	83.4302	686	•	•	NC25	Forested	Complex	N	EPA
PNF126	Cranberry	12/27/88	36.1040	82.0448	1250	•	•	NC45	Forested	Mountaintop	M	EPA
North Dakota												
THR422	Theodore Roosevelt National Park	10/06/98	46.8947	103.3778	850	•	•	ND00	Range	Rolling	Y	NPS
Ohio												
DCP114	Deer Creek State Park	09/28/88	39.6358	83.2600	267	•	•	OH54	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	•	•	OH09	Agri.	Rolling	N	EPA
QAK172	Quaker City	09/28/93	39.9431	81.3378	372	•	•	OH49	Agri.	Rolling	M	EPA
Oklahoma												
CHE185	Cherokee Nation	04/02/02	35.7507	94.6700	299	•	•	AR27	Agri.	Rolling	Y	EPA
Ontario												
EGB181	Egbert, Ontario	12/27/94	44.2317	79.7840	251	•	• ⁴	NY10	Agri.	Rolling	Y	EPA

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Pennsylvania												
ARE128	Arendtsville	06/28/88	39.9231	77.3078	269	•	•	PA00	Agri.	Rolling	M	EPA
KEF112	Kane Experimental Forest	01/03/89	41.5981	78.7683	622	•	•	PA29	Forested	Rolling	Y	EPA
LRL117	Laurel Hill State Park	12/15/87	39.9883	79.2522	615	•	•	MD08	Forested	Complex	N	EPA
MKG113	M.K. Goddard State Park	01/12/88	41.4250	80.1447	384	•	•	NY10	Forested	Rolling	N	EPA
PSU106	Penn. State University	01/06/87	40.7209	77.9316	376	•	•	PA42	Agri.	Rolling	M	EPA
South Dakota												
WNC429	Wind Cave National Park	11/18/03	43.5578	103.4839	1292	•	•	SD04	Prairie	Rolling	M	NPS
Tennessee												
ESP127	Edgar Evins State Park	03/22/88	36.0389	85.7330	302	•	•	KY10	Forested	Rolling	N	EPA
GRS420	Great Smoky Mountains National Park	10/06/98	35.6331	83.9422	793	•	•	TN11	Forested	Complex	N	NPS
SPD111	Speedwell	06/12/89	36.4698	83.8265	361	•	•	TN04	Agri.	Rolling	Y	EPA
Texas												
ALC188	Alabama-Coushatta	04/02/04	30.7017	94.6742	101	•	•	TX10	Forested	Rolling	Y	EPA
BBE401	Big Bend National Park	07/18/95	29.3022	103.1772	1052	•	•	TX04	Forested	Complex	M	NPS
PAL190	Palo Duro Canyon State Park	04/24/07	34.8803	101.6649	1050	•	•	TX43	Prairie	Complex	M	EPA
Utah												
CAN407	Canyonlands National Park	01/24/95	38.4586	109.8211	1809	•	•	UT09	Desert	Complex	M	NPS
Virginia												
PED108	Prince Edward	11/03/87	37.1653	78.3070	150	•	•	VA24	Forested	Rolling	M	EPA
SHN418	Shenandoah National Park	06/28/88	38.5231	78.4347	1073	•	•	VA28	Forested	Mountaintop	M	NPS
VPI120	Horton Station	06/02/87	37.3300	80.5573	920	•	•	VA13	Forested	Mountaintop	N	EPA

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Washington												
MOR409	Mount Rainier National Park	08/29/95	46.7583	122.1244	415	•	•	WA99	Forested	Complex	N	NPS
West Virginia												
CDR119	Cedar Creek State Park	11/10/87	38.8794	80.8478	234	•	•	WV05	Forested	Complex	N	EPA
PAR107	Parsons	01/19/88	39.0906	79.6614	510	•	•	WV18	Forested	Complex	N	EPA
Wisconsin												
PRK134	Perkinstown	09/27/88	45.2066	90.5972	472	•	•	WI35	Agri.	Rolling	M	EPA
Wyoming												
CNT169	Centennial	08/19/91	41.3722	106.2422	3178	•	•	WY95	Range	Complex	M	EPA
PND165	Pinedale	12/27/88	42.9214	109.7900	2388	•	•	WY06	Range	Rolling	M	EPA
YEL408	Yellowstone National Park	06/26/96	44.5597	110.4006	2400	•	•	WY08	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

AC	automated colorimetry
ANC	acid neutralizing capacity
AMNet	Atmospheric Mercury Network
AMoN	Ammonia Monitoring Network
ARP	Acid Rain Program
Ca ²⁺	particulate calcium ion
CAAA	Clean Air Act Amendments
CAIR	Clean Air Interstate Rule
CAPMoN	Canadian Air and Precipitation Monitoring Network
CASTNET	Clean Air Status and Trends Network
CCV	continuing calibration verification samples
Cl ⁻	particulate chloride ion
CLD303	MADPro cloud water monitoring site at Clingmans Dome, TN
CMAQ	Community Multiscale Air Quality modeling system
cm/month	centimeters per month
DM8A	daily maximum 8-hour average
DQI	data quality indicator
ECAN	Environment Canada
EGU	electric generating unit
EPA	U.S. Environmental Protection Agency
H ₂ SO ₄	sulfuric acid
Hg	mercury
HNO ₃	nitric acid
IC	ion chromatography
ICP-AES	inductively coupled plasma-atomic emission spectrometry
IMPROVE	Interagency Monitoring of Protected Visual Environments
ISO	International Organization for Standardization
K ⁺	particulate potassium ion
kg/ha	kilograms per hectare
kg/ha/yr	kilograms per hectare per year
km	kilometer
LAI	leaf area index
lpm	liters per minute
MACT	maximum achievable control technology
MACTEC	MACTEC Engineering and Consulting, Inc.
MAD	mean absolute difference
MADPro	Mountain Acid Deposition Program
MAGIC	Model of Acidification of Groundwater in Catchments
MARPD	mean absolute relative percent difference
MDN	Mercury Deposition Network
MeHg	methylmercury

List of Acronyms and Abbreviations (continued)

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
NAPAP	National Acid Precipitation Assessment Program
NBP	NO _x Budget Trading Program
NDDN	National Dry Deposition Network
NERR	National Estuarine Research Reserve
ng/L	nanograms per liter
NH ₃	ammonia
NH ₄ ⁺	particulate ammonium
(NH ₄) ₂ SO ₄	ammonium sulfate
NIST	National Institute of Standards and Technology
NO ₃ ⁻	particulate nitrate
NO _x	nitrogen oxides [nitric oxide (NO) + nitrogen dioxide (NO ₂)]
NO _y	total reactive oxides of nitrogen
NPS	National Park Service
NTN	National Trends Network
O ₃	ozone
OTC	Ozone Transport Commission
PM _{2.5}	fine particle matter
POMS	portable ozone monitoring system
ppb	parts per billion
ppm	parts per million
PT	proficiency test
QAPP	Quality Assurance Project Plan
RoMANS	Rocky Mountain Atmospheric Nitrogen and Sulfur study
S	sulfur
SIP	State Implementation Plan
SLAMS	State and Local Monitoring Stations
SO ₂	sulfur dioxide
SO ₄ ²⁻	particulate sulfate
TN11	NADP wet deposition sampling site at Elkmont, TN
total NO ₃ ⁻	gaseous nitric acid (HNO ₃) + particulate nitrate (NO ₃ ⁻)
Total_N	total nitrogen content of HNO ₃ , NH ₄ ⁺ , and NO ₃ ⁻
TVA	Tennessee Valley Authority
µg/m ³	micrograms per cubic meter
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:

Clean Air Markets
Division Home Page:

EPA Home Page:

www.epa.gov/castnet

www.epa.gov/airmarkets

www.epa.gov