

# Clean Air Status and Trends Network (CASTNET)

## 2007 Annual Report



# **Clean Air Status and Trends Network**

**(CASTNET)**

## **2007 Annual Report**

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**MACTEC Engineering and Consulting, Inc.**

**Prepared for:**

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**Clean Air Markets Division**

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## Executive Summary

This report summarizes Clean Air Status and Trends Network (CASTNET) data collected during 2007 and examines trends in air quality since 1990. The report also discusses monitoring activities, network changes and upgrades, and data quality. CASTNET began collecting measurements in 1991 with the incorporation of 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 in order to evaluate the effectiveness of national and regional air pollution control programs.

### *Key Results through 2007*

- ★ Mean annual sulfur dioxide and particulate sulfate concentrations declined significantly over the 18-year period 1990 through 2007. Sulfur dioxide levels declined 40 percent while sulfate concentrations declined 26 percent.
- ★ Total sulfur deposition declined by 34 percent from 1990 through 2007.
- ★ Mean annual concentrations of total nitrate (nitric acid plus particulate nitrate) declined by approximately 20 percent over the 18-year period.
- ★ Total nitrogen deposition declined by 17 percent from 1990 through 2007.
- ★ Precipitation-weighted mean sulfur and nitrogen concentrations in precipitation show an overall decline over the last 18 years with small increases in 2005, 2006, and 2007.
- ★ Ozone levels measured in 2007 continue to corroborate a downward trend that began after a peak in 2002. The median fourth highest daily maximum 8-hour average ozone concentration for 2007 (75 ppb) was the third lowest in the history of the network. For the most recent 3-year period (2005 through 2007), only three eastern and four California sites recorded exceedances of the current 8-hour standard (0.08 ppm). The recent decline in rural ozone and nitrate levels has been attributed to the documented decline in nitrogen oxides emissions that began in 2000.
- ★ Routine quality assurance activities include laboratory intercomparison studies under the direction of the U.S. Geological Survey and National Water Research Institute of Canada. Measurements taken during 2007 and historical data collected over the period 1990 through 2007 were analyzed relative to data quality objectives (DQO) and their numerical measures. These analyses demonstrate that CASTNET data can be used with a high degree of confidence.

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## Chapter 1: CASTNET Overview

CASTNET is a national air quality monitoring network that provides data for determining trends in atmospheric nitrogen, sulfur, and ozone and for determining relationships between emissions, air quality, deposition, and ecological effects. CASTNET began operation in 1991 to provide long-term monitoring of air quality in rural areas. As of December 2007, the network operated 87 monitoring stations throughout the continental United States and Alaska.

### Background

The United States 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 (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>). Congress recognized the need to track real-world environmental results as the ARP was implemented and emission reductions became effective. Congress mandated that CASTNET provide data for determining relationships between changes in emissions and any subsequent changes in air quality, atmospheric deposition, and ecological effects.

The ARP has produced significant reductions in SO<sub>2</sub> and NO<sub>x</sub> emissions from electric generating units (EGUs) since 1995. More recent NO<sub>x</sub> emission control programs also produced substantive declines in NO<sub>x</sub> emissions in the eastern United States. These programs include the Ozone Transport Commission (OTC) NO<sub>x</sub> Budget (1999-2002) and the NO<sub>x</sub> State Implementation Plan (SIP) Call/NO<sub>x</sub> Budget Trading Program (NBP), which was implemented in 2003. The NBP places a cap on total NO<sub>x</sub> emissions from EGUs in 20 states and the District of Columbia during the ozone season (May 1 through September 30) when the potential for photochemical ozone formation is high. The EPA relies on CASTNET and other long-term monitoring networks to measure pollutant concentrations used to assess the effectiveness of emission control programs.

CASTNET started as the National Dry Deposition Network (NDDN), which began operation in 1987. The 50 NDDN sites were transferred to CASTNET in 1991. CASTNET provides a long-term data record with more than 35 sites in operation since the late 1980s. The network, as of December 2007, included 87 monitoring stations at 84 site locations throughout the continental United States and Alaska. 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. The dry deposition estimates are calculated from CASTNET concentration measurements and modeled deposition velocities. The model requires several meteorological parameters and information on vegetation and land use specific to each site.



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This report summarizes CASTNET/NDDN monitoring activities and the resulting concentration and deposition data collected over the 18-year period from 1990 through 2007. 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. Fully validated data are available approximately 10 months following collection.

## Network Partners

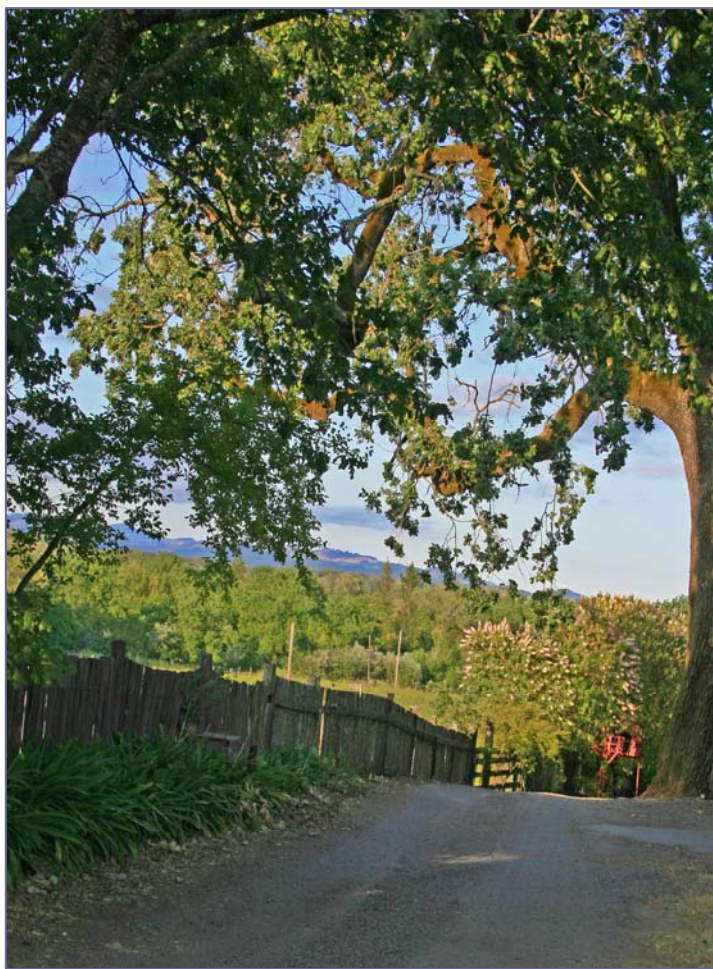
CASTNET is sponsored by EPA and the National Park Service (NPS). NPS began its participation in CASTNET in 1994 under an agreement with EPA. NPS is responsible for the protection and enhancement of air quality and related values in national parks and wilderness areas. The CASTNET sites sponsored by NPS as of December 2007 numbered 27. Additional information on the NPS Air Monitoring program can be found on the Web site: <http://www.nature.nps.gov/air/monitoring/>. In addition to EPA and NPS, CASTNET monitors air quality and deposition in cooperation with other national networks.

- ★ **National Atmospheric Deposition Program/National Trends Network (NADP/NTN)** operates 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. Additional information on NADP/NTN can be found on its Web site: <http://nadp.sws.uiuc.edu/>.
- ★ **Canadian Air and Precipitation Monitoring Network (CAPMoN)** operates 28 measurement sites throughout Canada and one in the United States. CASTNET and CAPMoN both operate samplers at monitoring stations in Ontario, Canada and at Pennsylvania State University. Additional information on CAPMoN can be found on its Web site: [http://www.msc.ec.gc.ca/capmon/index\\_e.cfm](http://www.msc.ec.gc.ca/capmon/index_e.cfm).
- ★ **Interagency Monitoring of Protected Visual Environments (IMPROVE)** measures aerosol pollutants near more than 30 CASTNET sites. IMPROVE measures particulate air pollutants that affect visibility. Additional information on IMPROVE can be found on its Web site: <http://vista.cira.colostate.edu/improve>.
- ★ **Mercury Deposition Network (MDN)** samplers are operated at multiple CASTNET sites. MDN samplers measure mercury in precipitation. Additional information on MDN can be found on its Web site: <http://nadp.sws.uiuc.edu/mdn/>.

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

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different mandates including the Program Assessment Rating Tool (PART), 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 an in-kind service that supports the overall operation of a site including site operation, land use, or both. All of the sites added during the recent expansion of the network (since 2001) have an associated co-sponsor. The CASTNET 2006 Annual Report (MACTEC, 2007a) presented a list of the co-sponsors and the sites they sponsor. Three CASTNET monitoring sites are located on tribal lands: Cherokee Nation in eastern Oklahoma (CHE185), the Alabama-Coushatta in eastern Texas (ALC188), and the Santee Sioux in northern Nebraska (SAN189). Tribal air quality monitoring programs are discussed in Chapter 2. State agencies also operate special purpose air pollutant measurement devices at some CASTNET sites.



As of December 2007, CASTNET sites in rural California ranged from Lassen Volcanic National Park (LAV410) in the north to Joshua Tree National Monument (JOT403) in the south and included Converse Station (CON186), Death Valley National Monument (DEV412), Pinnacles National Monument (PIN414), Sequoia National Park (SEK430), and Yosemite National Park (YOS404).

***AIRNow: EPA-Sponsored CASTNET Sites  
Began Participation in May 2007***

<b>Site ID</b>	<b>Site Name</b>	<b>AQS Code</b>
ARE128	Arendtsville, PA	42-001-9991
CND125	Candor, NC	37-123-9991
CON186	Converse Station, CA	06-071-9991
HWF187	Huntington Wildlife Forest, NY	36-031-9991
LRL117	Laurel Hill State Park, PA	42-111-9991
PED108	Prince Edward, VA	51-147-9991
PND165	Pinedale, WY	56-035-9991
SPD111	Speedwell, TN	47-025-9991
UVL124	Unionville, MI	26-157-9991
WSP144	Washington's Crossing, NJ	34-021-9991

**Notes:** AQS is EPA's Air Quality System for archiving air quality data. NPS-sponsored CASTNET sites have been providing ozone data to AIRNow since May 2002.

AIRNow is a Web site (<http://www.airnow.gov>) designed to allow the public to view air quality data for a specific area in "real-time." The EPA, the National Oceanic and Atmospheric Administration (NOAA), NPS, tribal, state, and local agencies developed the AIRNow Web site to provide the public with easy access to national air quality information. The Web site offers daily forecasts of Air Quality Index (AQI) as well as maps of real-time AQI conditions for over 300 cities across the United States. It also provides links to more detailed state and local air quality Web sites.

The air quality data used in these maps and to generate forecasts are collected using either federal reference or equivalent monitoring techniques approved by EPA. Since the information needed to make the maps must be as "real-time" as possible, the data are displayed as soon as practical after the end of each hour. Although some preliminary data quality assessments are performed, the data are not fully verified and validated through the quality assurance procedures monitoring organizations use to officially submit and certify data on the EPA Air Quality System (AQS). Therefore, data are used on the AIRNow Web site only for the purpose of reporting the AQI.

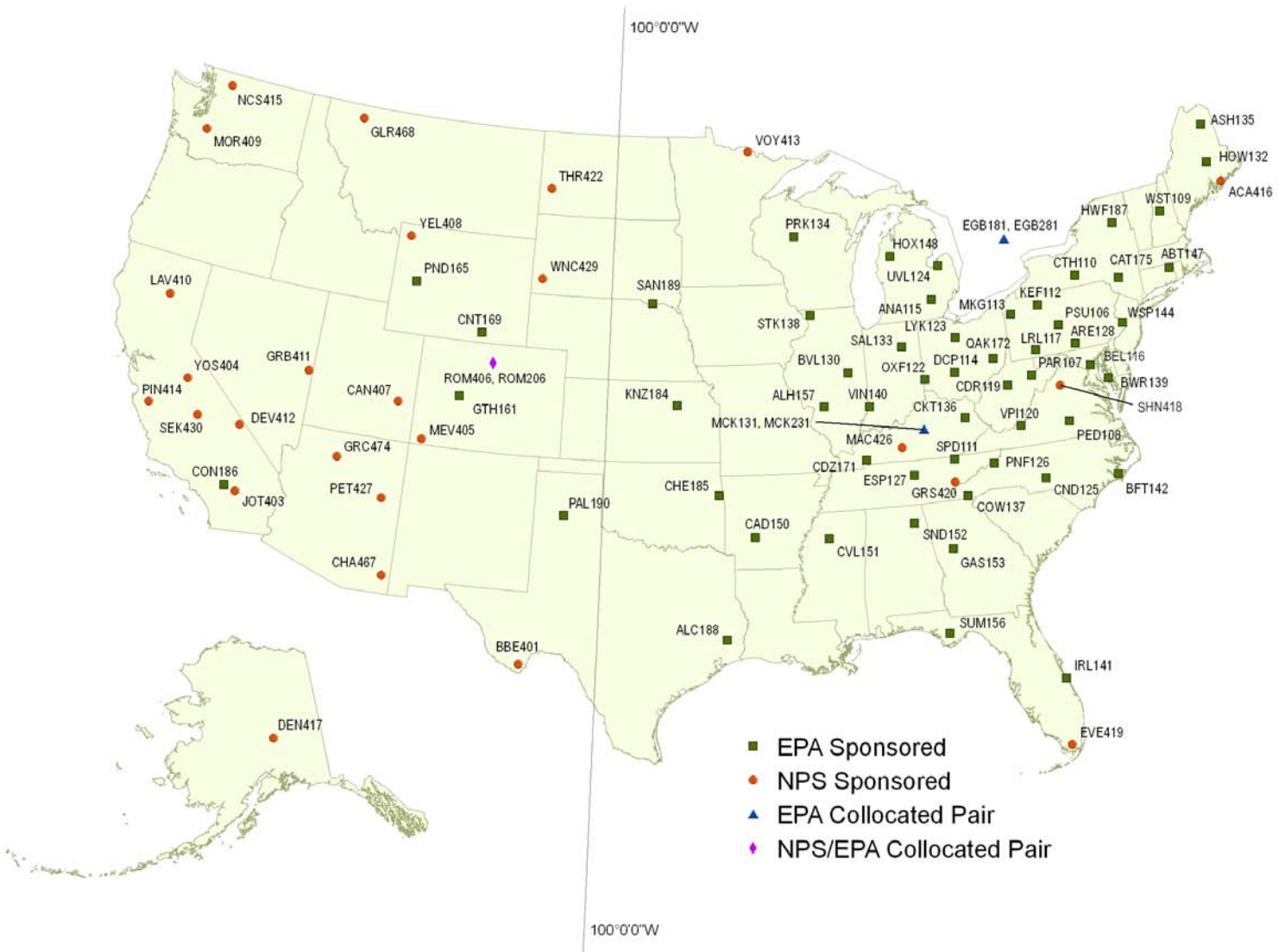
At the end of 2007, 10 EPA-sponsored CASTNET sites were sending 1-hour ozone data to the AIRNow database. The 10 sites were also transmitting 1-hour measurements of temperature, wind speed, wind direction, relative humidity, and solar radiation. Approximately 30 EPA-sponsored CASTNET sites will be participating in AIRNow by the end of 2008, and all sites will be participating by the end of 2009. All NPS-sponsored CASTNET sites are participating.



## Locations of Monitoring Sites

Figure 1-1 shows the locations of CASTNET monitoring sites as of December 2007. Eighty-seven sites were operational at 84 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 and operational characteristics of each site by state including 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.

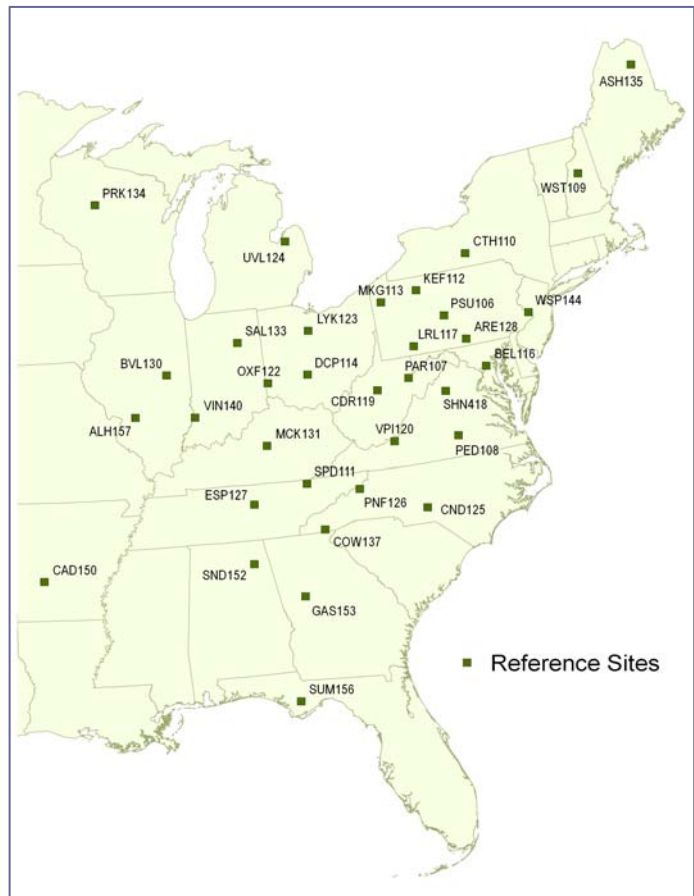
**Figure 1-1** CASTNET Sites as of December 2007



## CASTNET Reference Sites

Chapters 2 through 4 present maps illustrating the magnitude of pollutant concentrations and deposition fluxes across the United States. In addition, measurements from 34 CASTNET reference sites (Figure 1-2) were analyzed for each pollutant in order to determine trends in concentrations and trends in rates of dry, wet, and total deposition. These 34 sites have been reporting CASTNET measurements since at least 1990. The reference sites were selected using criteria similar to those used by EPA in its National Air Quality and Emissions Trends Report (2000). The criteria include site longevity and data completeness. EPA's procedures to interpolate and extrapolate quarterly mean data were also used to replace missing or invalid data. In chapters 2 through 4, the data from the 34 reference sites were aggregated and then presented using box plots for the period 1990-2007.

Figure 1-2 CASTNET Reference Sites



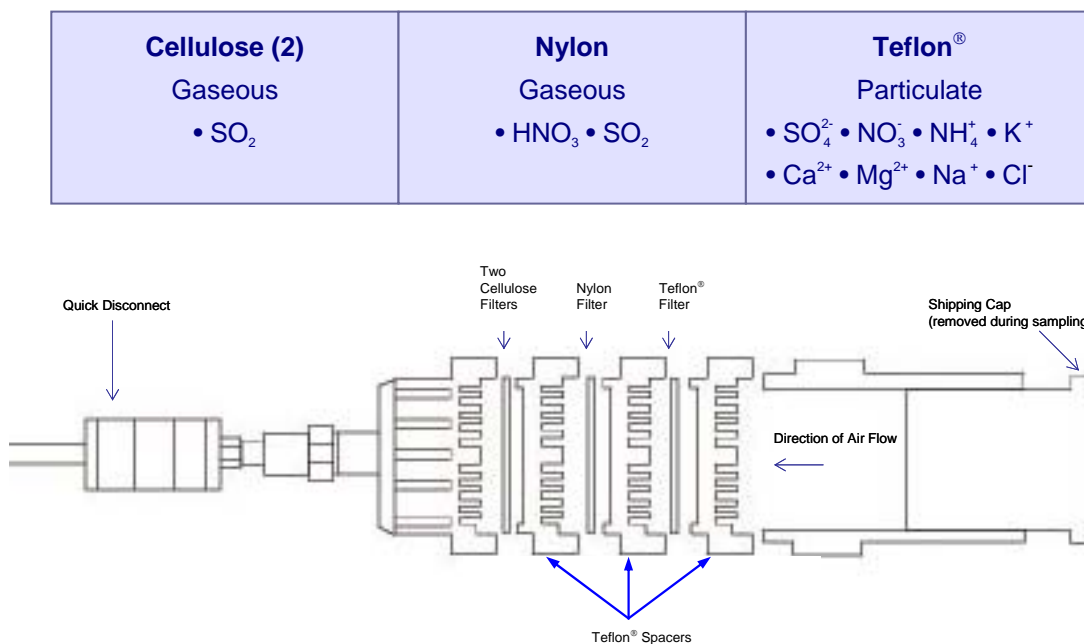
## Measurements Collected at CASTNET Sites

CASTNET was designed primarily to measure trends in seasonal and annual average concentrations and depositions over many years. Consequently, measurement of 7-day average (Tuesday to Tuesday) 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, three-stage filter pack (Figure 1-3) 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<sup>®</sup> filter, the second a nylon filter, and the third holds two potassium carbonate (K<sub>2</sub>CO<sub>3</sub>)-impregnated cellulose filters. The filter pack is changed out each Tuesday and shipped to the analytical chemistry laboratory for analysis.

### CASTNET Ambient Measurements

- Sulfur dioxide (SO<sub>2</sub>)
- Particulate sulfate (SO<sub>4</sub><sup>2-</sup>)
- Particulate nitrate (NO<sub>3</sub>)
- Nitric acid (HNO<sub>3</sub>)
- Particulate ammonium (NH<sub>4</sub><sup>+</sup>)
- Particulate calcium (Ca<sup>2+</sup>)
- Particulate sodium (Na<sup>+</sup>)
- Particulate magnesium (Mg<sup>2+</sup>)
- Particulate potassium (K<sup>+</sup>)
- Particulate chloride (Cl<sup>-</sup>)
- Ozone (O<sub>3</sub>)
- Meteorological variables and information on land use and vegetation

**Figure 1-3** Three-Stage Filter Pack



The filter packs are prepared, loaded, shipped, received, extracted, and analyzed at the MACTEC Gainesville, FL laboratory. Following receipt from the field, exposed Teflon® filters and blanks are extracted and then analyzed for sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and concentrations of chloride (Cl<sup>-</sup>) by micromembrane-suppressed ion chromatography (IC). Teflon® filter extracts are also analyzed for ammonium (NH<sub>4</sub><sup>+</sup>) by the automated indophenol method with the Bran+Luebbe AutoAnalyzer™ 3. The filter extracts are additionally analyzed for calcium (Ca<sup>2+</sup>), sodium (Na<sup>+</sup>), magnesium (Mg<sup>2+</sup>), and potassium (K<sup>+</sup>) by inductively coupled plasma-atomic emission spectrometry (ICP-AES) using a Perkin Elmer Optima 3000 Dual View spectrometer. The cellulose filter extracts are analyzed for SO<sub>2</sub> as sulfate using IC. The nylon filter extracts are analyzed via IC for HNO<sub>3</sub> and for SO<sub>2</sub>. The SO<sub>2</sub> concentrations from the cellulose and nylon filters are summed to obtain total SO<sub>2</sub>.

CASTNET also measures ozone concentrations, one of the major components of smog, and meteorological parameters, which are used to estimate deposition velocities. The meteorological parameters measured at each site include temperature, solar radiation, relative humidity, precipitation, wind speed and direction, standard deviation of wind direction (sigma theta), and surface wetness. These meteorological measurements are used to gauge the transport of air pollutants and as input to the Multi-Layer Model (MLM), a numerical model used for estimating dry deposition values. The ozone and meteorological measurements are recorded continuously and archived as hourly averages.

## Calculating Dry Deposition

Dry deposition processes are modeled as resistances to deposition. The original network design was based on the assumption that dry deposition, or flux, could be estimated as the linear product of measured pollutant concentration (C) and modeled deposition velocity ( $V_d$ ). Measured atmospheric concentrations are calculated based on the mass of each analyte in each filter extract and the volume of air sampled. The rate of deposition of a pollutant, also known as deposition velocity, is influenced by meteorological conditions, vegetation, and atmospheric and plant chemistry. The deposition velocity values for each site are calculated for each hour of each year using the MLM. The MLM is described by Meyers *et al.* (1998) and Finkelstein *et al.* (2000). The data used in the MLM to estimate dry deposition are derived from meteorological measurements and pollutant concentrations taken at the site together with an estimation of the vegetation leaf-out and leaf area index (LAI).

The schematic of the MLM in Figure 1-4 shows the relationships among the various resistances and illustrates the data that are required as model input. An improved version of the MLM (Schwede, 2006) was delivered to MACTEC by EPA's Office of Research and Development (ORD) during 2006. This version includes changes to the soil moisture factor, which affects the stomatal and soil resistances, and to the radiation algorithm, which also affects the stomatal resistance. Deposition velocities and fluxes for the entire network through 2007 were calculated using the updated model.

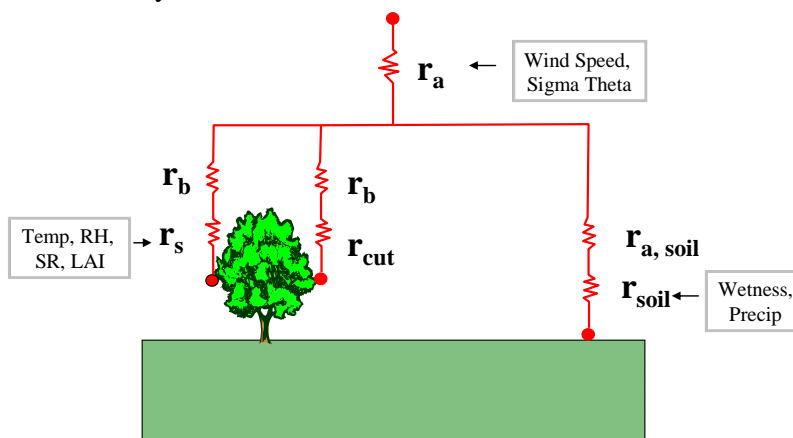
**Figure 1-4** Multi-Layer Model

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

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

### Resistances

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

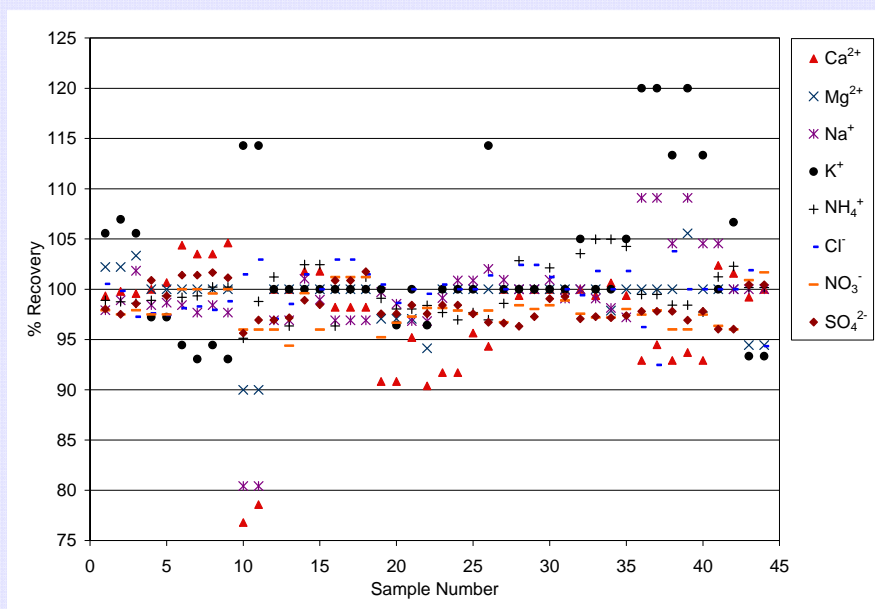




## Results for 2007 Laboratory Intercomparison Studies

The MACTEC laboratory is one of eight laboratories participating in the U.S. Geological Survey (USGS) interlaboratory comparison program ([http://bqs.usgs.gov/precip/interlab\\_data.htm](http://bqs.usgs.gov/precip/interlab_data.htm)). The laboratory receives four samples for chemical analysis from USGS every two weeks. The samples are a mix of 44 synthetically prepared samples, 8 deionized water samples, and 52 natural wet deposition samples. The laboratory reported the eight CASTNET parameters for a total of 104 samples during 2007. Results for the 44 synthetically prepared samples are depicted in the figure below, which presents results as percent recoveries of the median value for all laboratories. As shown in Figure 1-A, only 12 of 352 reported values were more than 10 percent from their corresponding median. All values shown are within two standard deviations of their corresponding median.

**Figure 1-A** Results from Intercomparison of Synthetic and Natural Samples



Additionally, precision results for the 52 natural wet deposition samples were reported as absolute percent differences for replicate analyses. By parameter, less than 5 percent of approximately 260 paired natural wet deposition sample data points exceeded 10 percent difference. These results are presented in the USGS report (USGS, 2008).

The laboratory generally participates in two to three studies each year through the National Water Research Institute (NWRI) of Canada's Proficiency Testing QA Program (<http://www.ec.gc.ca/inre-nwri/Default.asp?lang=En&n=7A20877C-1>) in addition to interlaboratory comparison studies for the USGS. MACTEC's laboratory was rated free of systemic bias for all eight parameters for the two NWRI 2007 studies and was ranked number six out of 36 participating laboratories at the end of 2007 (Environment Canada, 2007).

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## SO<sub>2</sub> and NO<sub>x</sub> Emissions

The ARP, established under Title IV of the 1990 CAAA, was designed to reduce the adverse effects of acid deposition by requiring major reductions of sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>) emissions from the electric power industry. EPA recently assessed the effects of the ARP and related programs in its 2007 progress report (EPA, 2009). The 2007 report summarized 2007 compliance with the ARP and reported progress toward achieving environmental goals.

Under the ARP, SO<sub>2</sub> reductions are achieved using a cap and trade program that sets a permanent cap on the total amount of SO<sub>2</sub> that may be emitted by all regulated EGUs in the contiguous United States. The program began in 1995 and is underway with the final 2010 SO<sub>2</sub> 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 SO<sub>2</sub> 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 SO<sub>2</sub>. 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 SO<sub>2</sub> emission cap. The program encourages early reductions by allowing sources to bank unused allowances in one year and use them in a later year.

Unlike the cap and trade program used to reduce SO<sub>2</sub> emissions, the ARP uses a more traditional approach to achieve NO<sub>x</sub> emission reductions. Rate-based limits apply to most of the coal-fired electric utility boilers subject to the ARP, and no nationwide cap has been placed on NO<sub>x</sub> emissions under the ARP. Other NO<sub>x</sub> emission control programs have resulted in significant reductions in NO<sub>x</sub> emissions during the ozone season (see Chapter 4). Two prominent control programs are the OTC NO<sub>x</sub> Budget (1999-2002) and the NO<sub>x</sub> SIP Call/NBP, which began in 2003.

The ARP is comprised of two phases for the reduction of SO<sub>2</sub> and NO<sub>x</sub>. Phase I applied primarily to the largest coal-fired electric generation sources from 1995 through 1999 for SO<sub>2</sub> and from 1996 through 1999 for NO<sub>x</sub>. Phase II for both pollutants began in 2000. In 2007, the SO<sub>2</sub> Phase II requirements affected 3,536 operating units. The Phase II NO<sub>x</sub> requirements applied to 978 of those operating units that exceeded 25 megawatts and burned coal between 1990 and 1995 (EPA, 2009). Under the ARP's emission reduction requirements, total SO<sub>2</sub> emissions from ARP sources were 8.9 million tons in 2007; total NO<sub>x</sub> emissions from ARP sources were 3.3 million tons.

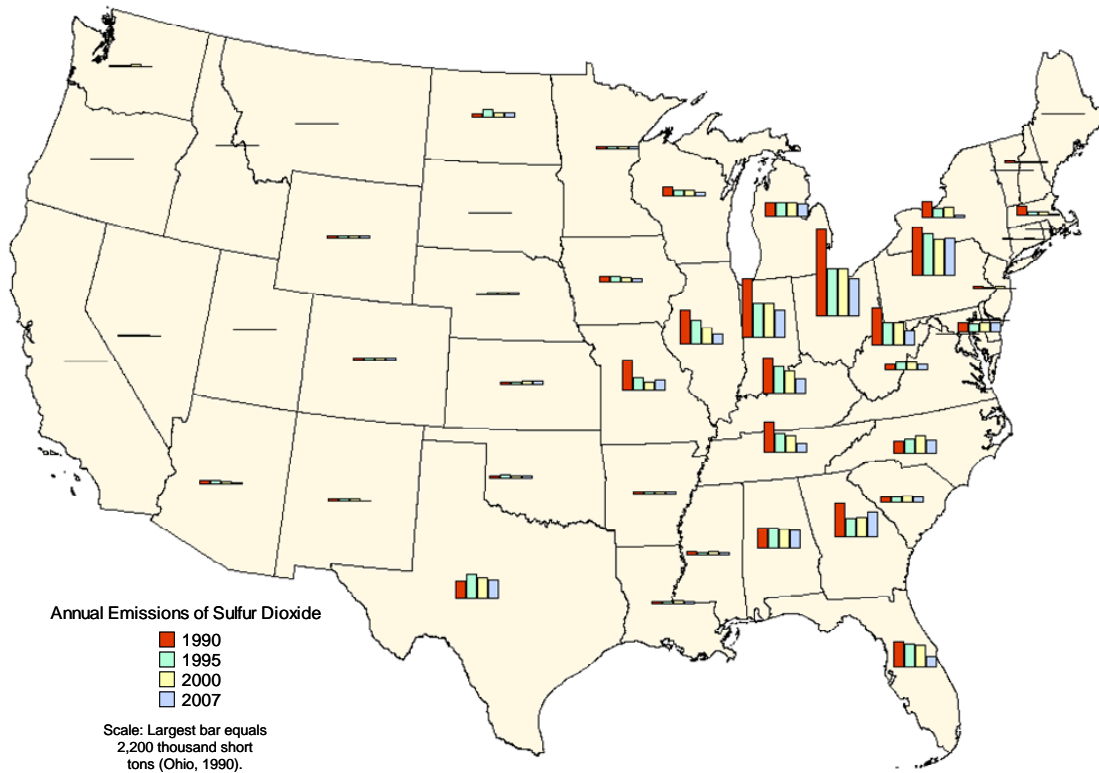
Figure 1-5 presents state-by-state total annual SO<sub>2</sub> emissions for Phase I and Phase II EGUs for four years (1990, 1995, 2000, and 2007). The most significant reduction in SO<sub>2</sub> emissions occurred in 1995 in the Ohio River Valley where a large concentration of coal-fired power plants are located. Many of the states with the highest SO<sub>2</sub> emissions realized the largest reductions. The six states with the greatest annual reductions include Ohio, which has decreased emissions

by 1.3 million tons since 1990, Illinois, Indiana, Missouri, Tennessee, and West Virginia. Annual  $\text{NO}_x$  emissions by state are depicted in Figure 1-6 for the same four years. States with the highest  $\text{NO}_x$  emissions also produced the most significant declines. Every state east of the Mississippi River experienced a significant decline in annual  $\text{NO}_x$  emissions from 2000 through 2007.

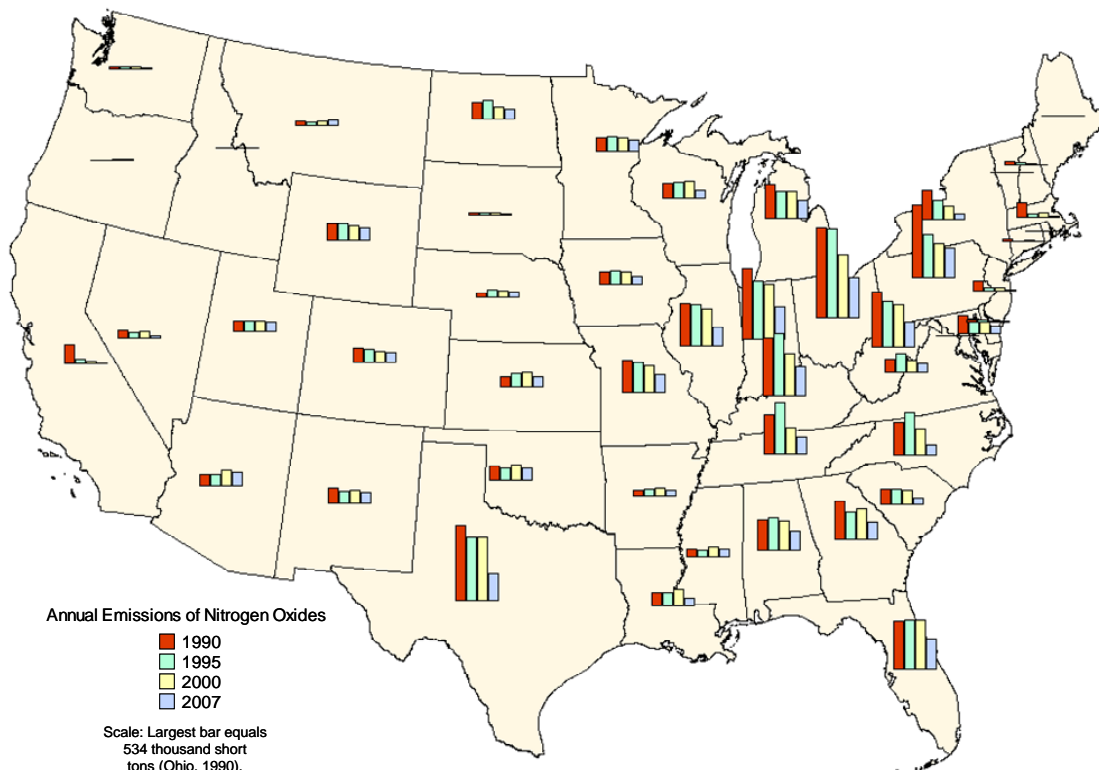


Coweeta, NC (COW137)

**Figure 1-5** Annual Utility SO<sub>2</sub> Emissions (Phase I and Phase II Plants only)



**Figure 1-6** Annual Utility NO<sub>x</sub> Emissions (Phase I and Phase II Plants only)





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## *Significant Events during 2007*

<b>January</b>	EPA-sponsored sites began every-other-day zero/span/precision auto-calibration checks of the ozone analyzer. Daily data submittals started including NPS meteorological data.
<b>February</b>	An independent audit of CASTNET data operations was performed. Overall, CASTNET data operations are managed satisfactorily. Only minor changes were recommended.
<b>April</b>	A new EPA-sponsored site, Palo Duro Canyon State Park, TX (PAL190), began collecting samples on April 24. The site is operated in cooperation with the Texas Agricultural Experiment Station and Palo Duro Canyon State Park.  Ozone sampling was discontinued at the two solar-powered CASTNET sites, Claryville, NY (CAT175) and Lye Brook, VT (LYE145).  The MLM, the deposition model used for estimating dry deposition at CASTNET sites, was run to simulate dry deposition for the entire CASTNET database. The results were archived in the official EPA CASTNET database.
<b>May</b>	Atmospheric sampling at the LYE145, VT site was discontinued, and the site was closed.  Ten EPA-sponsored CASTNET sites began uploading ozone data to AIRNow.
<b>July</b>	The Campbell Scientific, Inc. Model CR3000 data logger was selected as the most suitable model for upgrading the data collection instrumentation at all EPA-sponsored sites.  EPA approved the CASTNET Quality Assurance Project Plan (QAPP) Revision 4.0. (MACTEC, 2007d) <a href="http://www.epa.gov/castnet/docs.html">http://www.epa.gov/castnet/docs.html</a> .
<b>August</b>	All EPA-sponsored CASTNET sites had laptop computers by August.
<b>September</b>	The annual “floor-to-ceiling” CASTNET property audit was conducted by EPA and the Defense Contract Management Agency at MACTEC’s Gainesville, FL facility. The property management system was approved by the audit.  EPA conducted a routine audit of CASTNET financial records. Management of CASTNET finances was deemed satisfactory.
<b>October</b>	Proper removal and disposal of the mercury thermometers at the EPA-sponsored sites began. A third party certified mercury disposal company was contracted for proper disposal.
<b>December</b>	Dry deposition and ozone sampling were discontinued at North Cascades National Park, WA (NCS415). Dry deposition sampling was terminated at Death Valley National Monument, CA (DEV412). Sampling ended as of December 31.  Day-night sampling was discontinued at Egbert, Ontario (EGB281).

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## Chapter 2: Atmospheric Concentrations

Three-stage filter packs are used to measure 7-day average concentrations of sulfur dioxide (SO<sub>2</sub>), sulfate (SO<sub>4</sub><sup>2-</sup>), nitric acid (HNO<sub>3</sub>), nitrate (NO<sub>3</sub><sup>-</sup>), ammonium (NH<sub>4</sub><sup>+</sup>), chloride (Cl<sup>-</sup>), and several earth metals at 87 monitoring stations. Since 1990, measured concentrations of sulfur species have decreased significantly. Concentrations of nitrogen species remained relatively steady from 1990 until 2000 when they began to show a slight decline. Trends in mean annual SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup>, total nitrate, and NH<sub>4</sub><sup>+</sup> concentrations are shown over the period 1990 through 2007 using box plots for each year. All four parameters have declined over the 18-year period.

Separate maps depicting the annual mean concentrations of sulfur dioxide (SO<sub>2</sub>), particulate sulfate (SO<sub>4</sub><sup>2-</sup>), total nitrate (HNO<sub>3</sub> + NO<sub>3</sub><sup>-</sup>), and ammonium (NH<sub>4</sub><sup>+</sup>) are presented in this chapter. Additional maps are provided in CASTNET Quarterly Reports (MACTEC 2007b, 2007e, 2008a, 2008c). The concentration shading in the maps in this chapter was prepared using an algorithm based on inverse distance cubed weighting with a radius of influence of 500 kilometers (km). In addition, trends in concentrations over the 18-year period (1990-2007) were derived from the 34 CASTNET reference sites (Figure 1-2) and are presented using box plots.

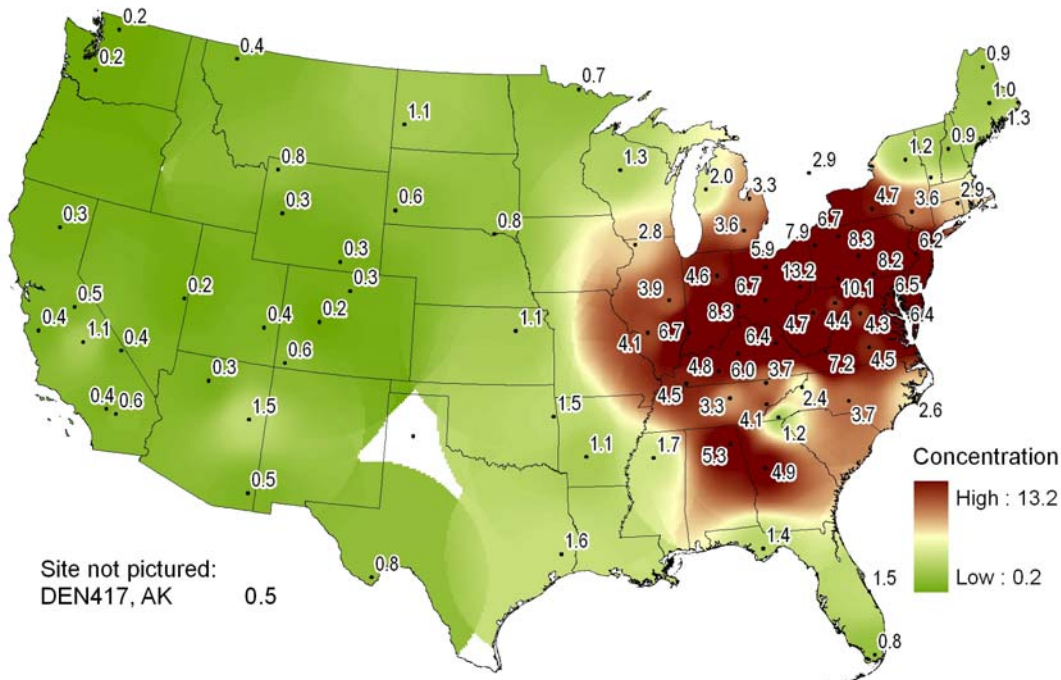
### Sulfur Dioxide

Sulfur dioxide (SO<sub>2</sub>) is a gaseous pollutant emitted through the combustion of coal, oil, and other fossil fuels that contain sulfur. EGUs are the largest source of SO<sub>2</sub> in the United States. SO<sub>2</sub> emissions are controlled by the ARP, and ambient concentrations are regulated by the National Ambient Air Quality Standards (NAAQS), which are designed to protect human health and welfare. The gas reacts in the atmosphere to form various acidic compounds including sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), a significant contributor to acid rain, and ammonium sulfate [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>], a major component of fine particle matter (PM<sub>2.5</sub>). Fine particles are regulated as a criteria pollutant. They are harmful to human health and also contribute to degradation of atmospheric visibility and visual quality.

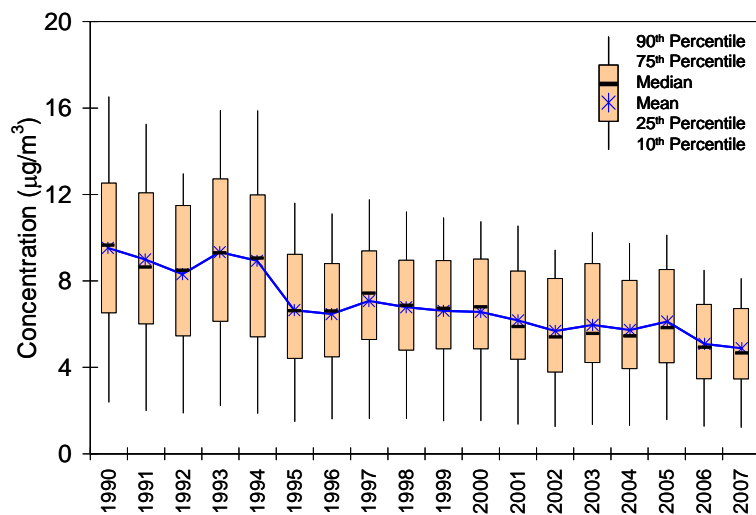
Annual mean SO<sub>2</sub> concentrations measured in 2007 are presented in Figure 2-1. Locations on the map with no value had insufficient data to estimate annual mean concentrations. For 2007, Palo Duro State Park, TX (PAL190) and Lye Brook, VT (LYE145) had insufficient data. PAL190, TX began filter pack sampling at the end of April, and sampling at LYE145, VT was terminated during May. A 7-state region centered on the Ohio River Valley in the eastern United States recorded mean concentrations greater than or equal to 5.0 micrograms per cubic meter (µg/m<sup>3</sup>) of air. The region covers western Virginia, Kentucky, Indiana, Ohio, Pennsylvania, New Jersey, and Maryland. A concentration of 5.3 µg/m<sup>3</sup> was observed at Sand Mountain, AL (SND152). Two sites recorded concentrations in excess of 10.0 µg/m<sup>3</sup> – Quaker City, OH (QAK172) with a value of 13.2 µg/m<sup>3</sup>, the highest in the network, and Laurel Hill State Park, PA (LRL117) with a level of 10.1 µg/m<sup>3</sup>. Only three western sites (i.e., sites west of 100 degrees west longitude) measured an annual mean SO<sub>2</sub> concentration greater than 1.0 µg/m<sup>3</sup>.

Figure 2-2 shows a downward trend in annual mean SO<sub>2</sub> concentrations aggregated over the 34 reference sites (Figure 1-2) from 1990-2007. The most significant reduction in ambient SO<sub>2</sub> was recorded in 1995 at the beginning of the ARP. Three-year mean concentrations for 1990-1992 and 2005-2007 were 8.9 µg/m<sup>3</sup> and 5.3 µg/m<sup>3</sup>, respectively. This change constitutes a 40 percent reduction in 3-year mean SO<sub>2</sub> concentrations between the two time periods. The 2007 mean level was the lowest value measured by the reference sites in the history of the network.

**Figure 2-1** Annual Mean SO<sub>2</sub> Concentrations (µg/m<sup>3</sup>) for 2007



**Figure 2-2** Trend in Annual Mean SO<sub>2</sub> Concentrations (µg/m<sup>3</sup>)



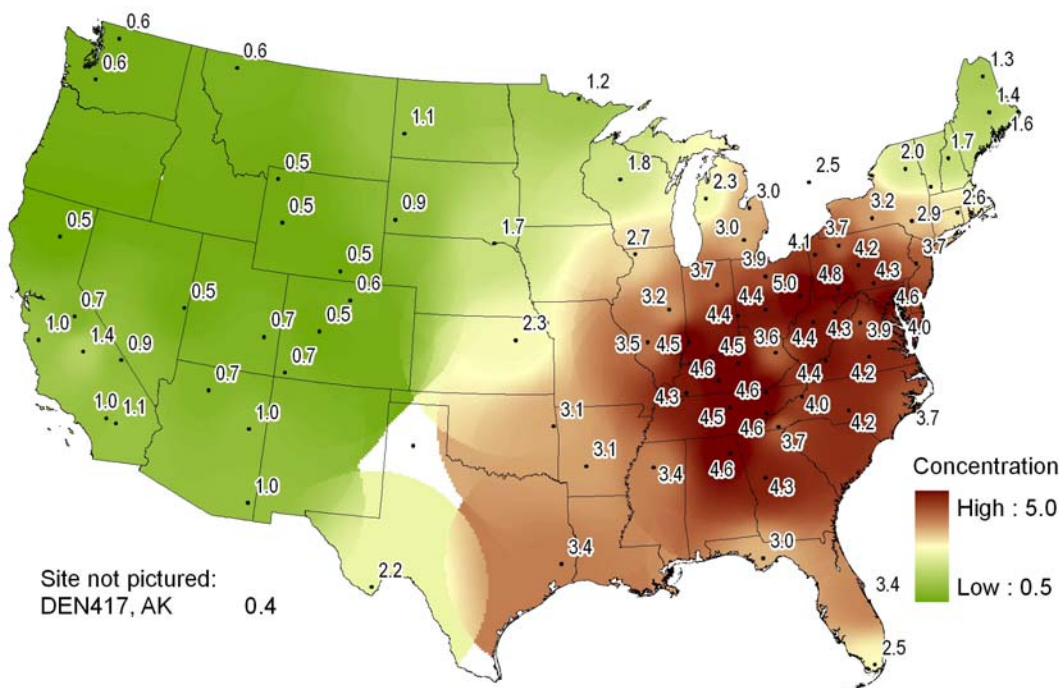
**Note:** All trend diagrams are based on data collected at 34 eastern reference sites.

## Particulate Sulfate

Particulate sulfate ( $\text{SO}_4^{2-}$ ) is formed in the atmosphere through the transformation of  $\text{SO}_2$  via both gas and aqueous (cloud) phase reactions. It typically exists in the atmosphere as ammonium sulfate  $[(\text{NH}_4)_2\text{SO}_4]$ , a major component of  $\text{PM}_{2.5}$ , which is regulated by the NAAQS. A map of annual mean particulate  $\text{SO}_4^{2-}$  concentrations measured during 2007 is provided in Figure 2-3. The map shows a large region of concentrations greater than or equal to  $4.0 \mu\text{g}/\text{m}^3$ . The region extends from Kentucky northeastward to Pennsylvania and also southeastward to Georgia. Concentration levels were fairly uniform throughout the region. Crockett, KY (CKT136) and Shenandoah National Park, VA (SHN418) were the two sites within this region that measured concentrations below  $4.0 \mu\text{g}/\text{m}^3$ . The highest concentration of  $5.0 \mu\text{g}/\text{m}^3$  was measured at QAK172, OH. Sulfate concentrations greater than or equal to  $1.0 \mu\text{g}/\text{m}^3$  were measured in the southwestern United States at four sites in California, two in Arizona, and at Big Bend National Park, TX (BBE401). The western monitoring station at Theodore Roosevelt National Park, ND (THR422) measured a  $\text{SO}_4^{2-}$  concentration of  $1.1 \mu\text{g}/\text{m}^3$ .

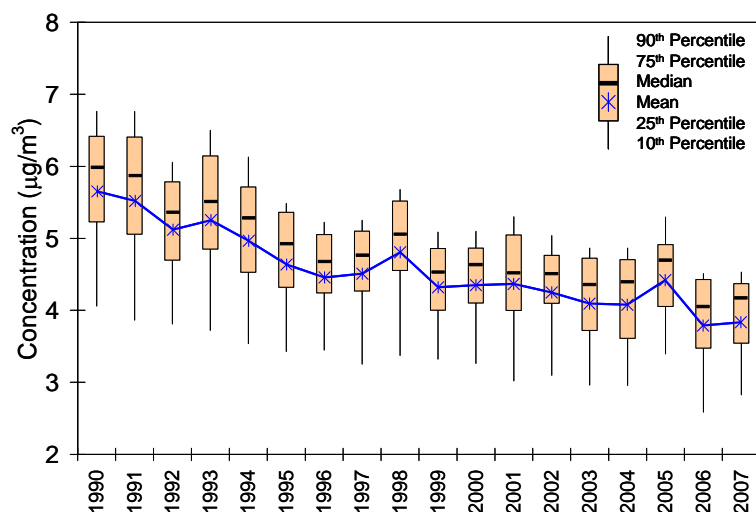
Box plots of annual mean  $\text{SO}_4^{2-}$  concentrations from the eastern reference sites from 1990-2007 are given in Figure 2-4 to illustrate the trend in atmospheric  $\text{SO}_4^{2-}$  over the 18-year period. The figure shows a significant decline in  $\text{SO}_4^{2-}$  with some interannual variability. The difference between 3-year means from 1990-1992 to 2005-2007 is 26 percent, a change from  $5.4 \mu\text{g}/\text{m}^3$  to  $4.0 \mu\text{g}/\text{m}^3$ , respectively. The 2007 mean level was slightly higher than 2006 value. The annual mean concentrations for 2006 and 2007 were the two lowest in the history of the network.

**Figure 2-3** Annual Mean  $\text{SO}_4^{2-}$  Concentrations ( $\mu\text{g}/\text{m}^3$ ) for 2007





**Figure 2-4** Trend in Annual Mean  $\text{SO}_4^{2-}$  Concentrations ( $\mu\text{g}/\text{m}^3$ )



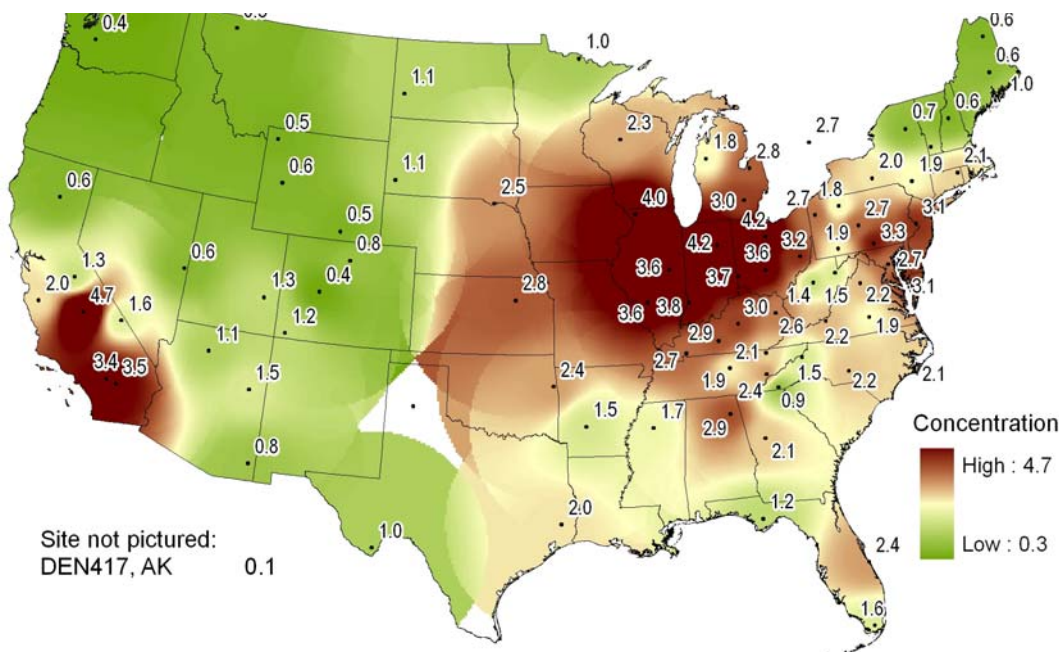
## Total Nitrate

Nitric acid ( $\text{HNO}_3$ ) and particulate nitrate ( $\text{NO}_3^-$ ) are formed from emissions of oxides of nitrogen ( $\text{NO}_x$ ) produced through the combustion of fossil fuels. Mobile sources are the largest contributors of  $\text{NO}_x$  emissions in the United States. Nitric acid exists as a gas and is a contributor to acid rain. Nitrate is a component of fine particles in the form of ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) and as larger particles, e.g., sodium nitrate ( $\text{NaNO}_3$ ) or calcium nitrate [ $\text{Ca}(\text{NO}_3)_2$ ]. Figure 2-5 shows a map of annual mean total nitrate ( $\text{NO}_3^- + \text{HNO}_3$ ) concentrations for 2007. Data on total nitrate levels are discussed in this chapter because total nitrate is more representative of a response to changes in  $\text{NO}_x$  emissions than either of its constituents alone. Also, measurements of the individual constituents are thought to include measurement uncertainties (Lavery *et al.*, 2009).

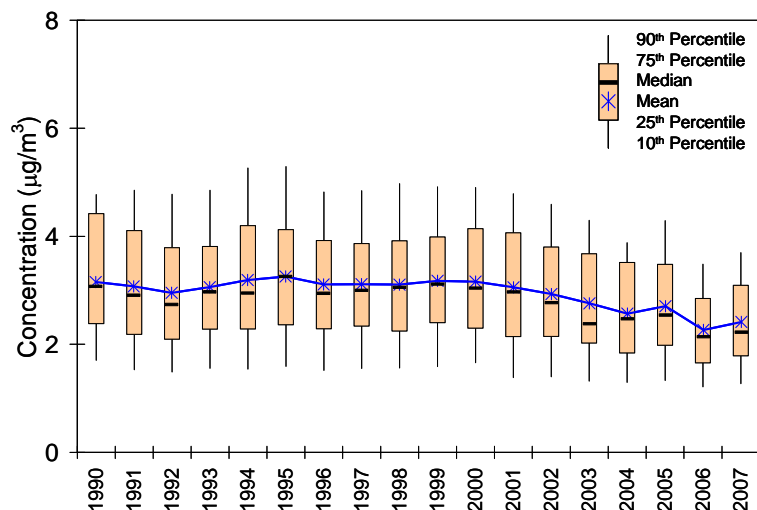
The map of 2007 total nitrate concentrations shows three regions with mean concentrations greater than or equal to  $3.0 \mu\text{g}/\text{m}^3$ . The largest region covers portions of five states in the Midwest, including Illinois, Indiana, Kentucky, Ohio, and Michigan. The second, smaller region covers southeastern Pennsylvania, New Jersey, and one site in Maryland. Three sites in California constitute the third region. Sequoia National Park, CA (SEK430) measured the highest concentration ( $4.7 \mu\text{g}/\text{m}^3$ ) in the network.

Box plots of total nitrate levels for the period 1990-2007 are given in Figure 2-6. These data were aggregated from the 34 eastern reference sites. The data show no trend in mean concentrations until 2000 when nitrate levels began a slow decline in response to  $\text{NO}_x$  emission control programs that were implemented in the eastern United States. See also the discussion in Chapter 4 about ozone season  $\text{NO}_x$  emissions. The difference between 3-year means from 1990-1992 to 2005-2007 is 20 percent, a change from  $3.1 \mu\text{g}/\text{m}^3$  to  $2.5 \mu\text{g}/\text{m}^3$ , respectively. The 2007 mean level was slightly higher than the 2006 value.

**Figure 2-5** Annual Mean Total Nitrate ( $\text{NO}_3^- + \text{HNO}_3$ ) Concentrations ( $\mu\text{g}/\text{m}^3$ ) for 2007



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

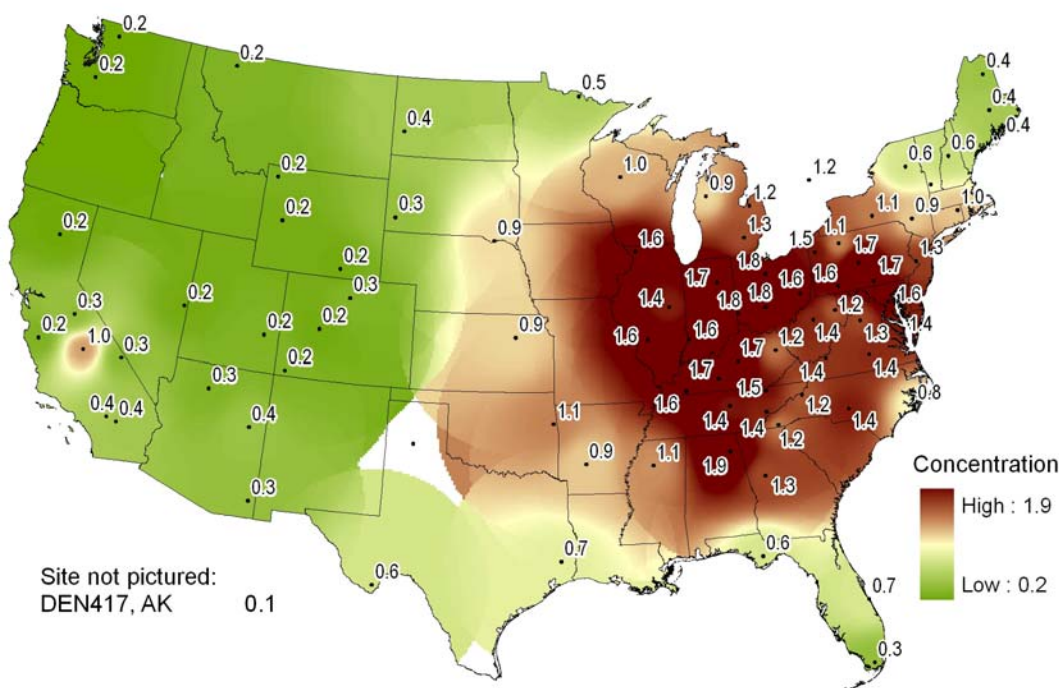


## Particulate Ammonium

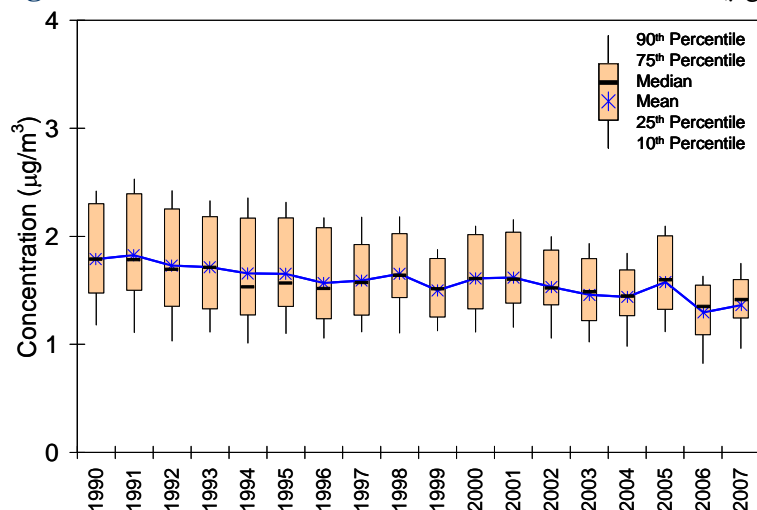
Particulate ammonium ( $\text{NH}_4^+$ ) is formed when ammonia ( $\text{NH}_3$ ) reacts with  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  particles in the atmosphere. Ammonia is formed through combustion processes and volatilization of farm animal wastes and fertilizers. Annual mean  $\text{NH}_4^+$  concentrations for 2007 are depicted in Figure 2-7. No monitors measured annual mean concentrations above  $2.0 \mu\text{g}/\text{m}^3$ . The data show a flat geographic distribution across the eastern United States with most sites reporting values greater than or equal to  $1.0 \mu\text{g}/\text{m}^3$ . The  $\text{NH}_4^+$  concentrations measured at western sites were low

with only one site (SEK430, CA) measuring a concentration greater than or equal to  $1.0 \mu\text{g}/\text{m}^3$ . Box plots of  $\text{NH}_4^+$  concentrations are provided in Figure 2-8. The trend diagram shows a 21 percent reduction in mean  $\text{NH}_4^+$  levels from 1990-1992 to 2005-2007. The 1990-1992 mean concentration was  $1.8 \mu\text{g}/\text{m}^3$ , and the 2005-2007 value was  $1.4 \mu\text{g}/\text{m}^3$ . The 2007 mean value was slightly higher than the 2006 level. This is consistent with the 2007  $\text{SO}_4^{2-}$  and total nitrate concentrations, whose values were higher in 2007 than in 2006, and is also consistent with the relationship between  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  as constituents of ammonium sulfate and the relationship between particulate  $\text{NO}_3^-$  and  $\text{NH}_4^+$  as components of ammonium nitrate.

**Figure 2-7** Annual Mean  $\text{NH}_4^+$  Concentrations ( $\mu\text{g}/\text{m}^3$ ) for 2007



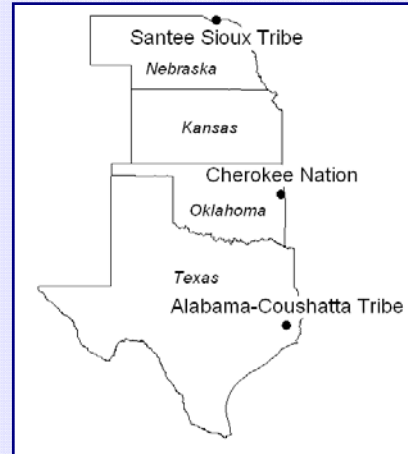
**Figure 2-8** Trend in Annual Mean  $\text{NH}_4^+$  Concentrations ( $\mu\text{g}/\text{m}^3$ )



## *Cherokee Nation Clean Air Program*

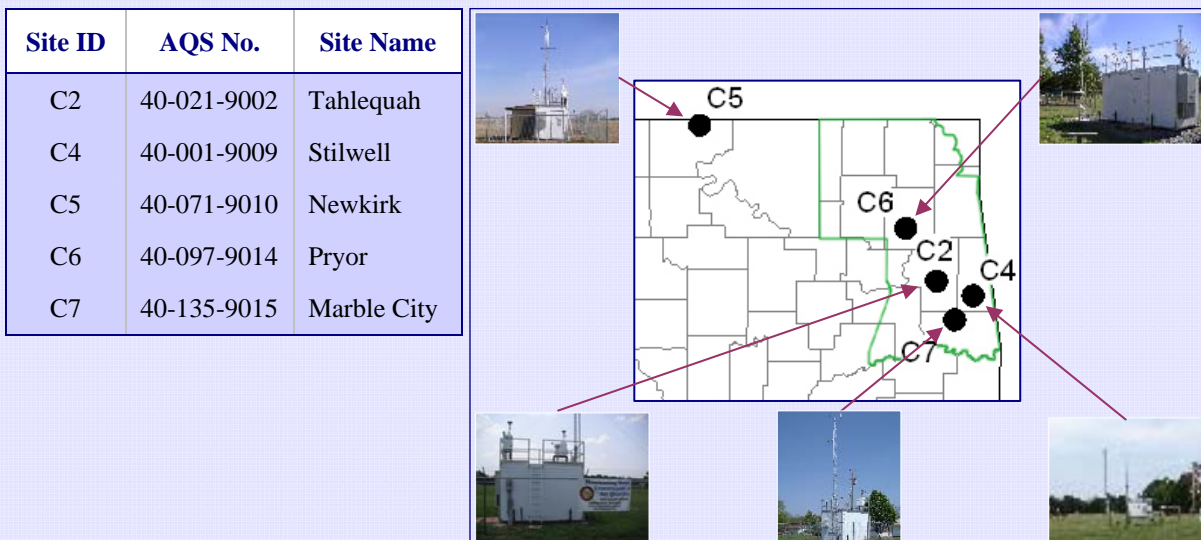
Three CASTNET monitoring sites are located on tribal lands as a result of a unique cooperative effort among EPA Headquarters, EPA Regions, tribal governments, and the Inter-Tribal Environmental Council (ITEC). This collaborative effort has resulted in monitoring sites operating on the tribal lands of the Cherokee Nation in eastern Oklahoma (CHE185), the Alabama-Coushatta in eastern Texas (ALC188), and the Santee Sioux in northern Nebraska (SAN189). Figure 2-A depicts the three tribal CASTNET monitoring sites.

**Figure 2-A** Native American Participation in CASTNET Operations



The Cherokee Nation operates the most extensive air quality monitoring network among tribal organizations. The Cherokee Nation, acting through its Cherokee Nation Environmental Programs (CNEP) office, has been active in ambient air quality monitoring since 1996. The Cherokee Nation currently operates a network of five PM<sub>2.5</sub> monitors and various criteria pollutant monitors at five stations in northeastern Oklahoma (Figure 2-B) with four of the sites on lands within its jurisdictional boundary (green border). The Cherokee Nation also operates meteorological instruments at each of its five stations in Oklahoma.

**Figure 2-B** Cherokee Nation Monitoring Sites in Northeastern Oklahoma



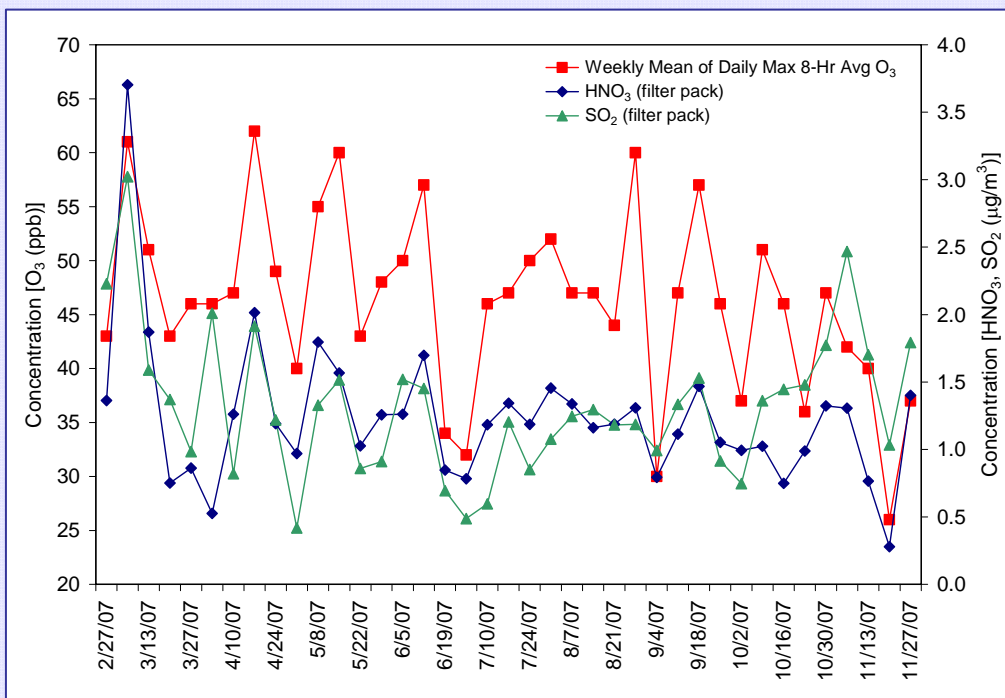


During the past five years, the CNEP has been increasing its participation in EPA, regional, and national monitoring programs and projects. The CNEP operates a CASTNET monitor at its Stilwell station (CHE185), an IMPROVE monitor at its Newkirk station, MDN samplers at its Stilwell and Newkirk stations, and passive ammonia and passive ozone samplers at its Stilwell station. The CNEP participated in the Midwest Ammonia Monitoring Project, operating an ammonia sampler at its Stilwell station from 2003 to 2006, and it has operated a continuous ammonia analyzer at that station since 2003 (Cherokee Nation, 2008).

In the future, the CNEP will continue criteria pollutant and ammonia monitoring at its stations in Oklahoma and continue monitoring as a part of IMPROVE, CASTNET, and MDN. CNEP plans to add monitoring as part of the NADP Atmospheric Mercury Initiative and the NTN. The Cherokee Nation also plans to add trace gas analyzers to the Stilwell station and has requested an NCore designation (<http://epa.gov/ttnamti1/ncore/index.html>) for Stilwell (Cherokee Nation, 2008).

The Cherokee Nation will use the data from these projects to assess the threat of air pollutants to the health of tribal members. The data will also be used to assess trends in air quality and to investigate the interrelationships among the measured air quality parameters such as those depicted in Figure 2-C, a time series of O<sub>3</sub>, SO<sub>2</sub>, and HNO<sub>3</sub> concentrations over the period February 27, 2007 through November 27, 2007.

**Figure 2-C** Time Series of Weekly HNO<sub>3</sub>, SO<sub>2</sub>, and O<sub>3</sub> at CHE185, OK during March through November 2007



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## Chapter 3: Atmospheric Deposition

CASTNET and its predecessor network (NDDN) were designed to provide estimates of the dry deposition of sulfur and nitrogen pollutants across the United States. CASTNET uses a hybrid approach to estimate dry deposition by combining measured pollutant concentrations and modeled deposition velocities. The MLM is the computer model used to calculate hourly deposition velocities for each monitoring site based on meteorological measurements and information on the surrounding vegetation. Total deposition is the sum of estimated dry deposition and measured wet deposition. Since 1990, total sulfur deposition has declined significantly. The data show a 34 percent reduction in 3-year mean sulfur fluxes over the period from 1990-1992 to 2005-2007. Total nitrogen deposition declined by 17 percent over the same period. Dry deposition is responsible for approximately 20 to 80 percent of total deposition depending on location and climate. The percentage is higher in major source regions.

Gaseous and particulate sulfur and nitrogen pollutants are deposited into ecosystems 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 ecosystems based on measured meteorological and other environmental conditions. Flux values are estimated as the product of measured concentration data and MLM-modeled dry deposition velocities. Wet deposition measurements were obtained from NADP/NTN and combined with CASTNET dry deposition data to estimate total deposition. Dry sulfur, total sulfur, and total nitrogen deposition decreased during 2007 while dry nitrogen deposition increased slightly over 2006. Precipitation-weighted mean concentrations of total atmospheric sulfur have declined over the past 18 years. Nitrogen concentrations in precipitation have declined slowly since 1998.

Dry deposition processes were simulated using the MLM (Figure 1-4) as described by Meyers *et al.* (1998) and Finkelstein *et al.* (2000). The most recent version of the MLM (Schwede, 2006) was provided to MACTEC in 2006. For this report, the MLM was run using CASTNET filter pack concentrations and meteorological measurements with information on land use, vegetation, and surface conditions to calculate deposition velocities for sulfur dioxide (SO<sub>2</sub>), nitric acid (HNO<sub>3</sub>), ozone (O<sub>3</sub>), and the particles sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and ammonium (NH<sub>4</sub><sup>+</sup>). The deposition velocities were assumed to be identical for all particle species. Deposition velocity values were calculated for each pollutant species for each hour with valid meteorological data for each CASTNET site for the entire period 1990 through 2007. For a deposition velocity to be estimated, measurements of temperature, solar radiation, relative humidity, wind speed, and standard deviation of the wind direction (sigma theta) must all be valid for the hour. Aggregation rules for CASTNET require three valid quarters for the calculation of an annual value. If an annual value was not available for a specific site, the results were not included on 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

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or extrapolation using valid annual estimates. For example, if an intermediate year was missing, the value for that year was interpolated from adjacent years. If a 2007 value was missing, the 2007 value was assumed equal to the value for 2006.

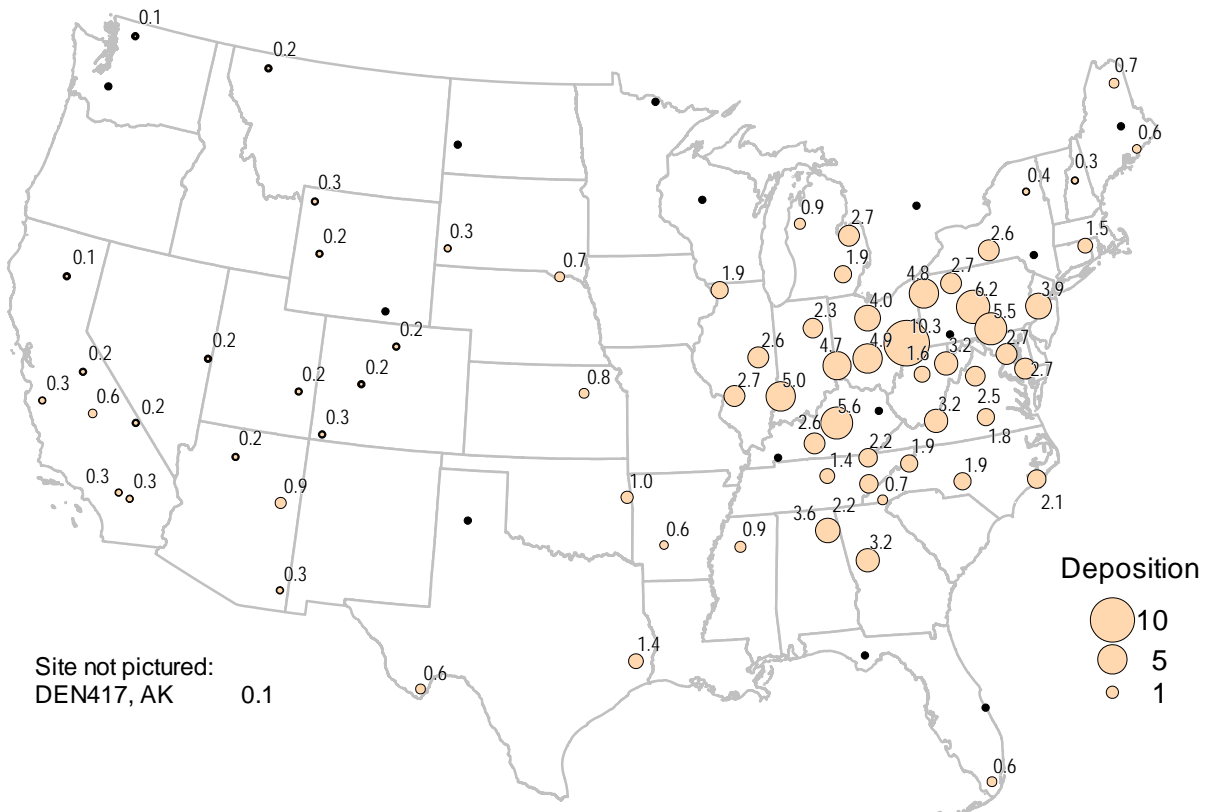
## Sulfur Deposition

MLM simulations were run separately for sulfur dioxide (SO<sub>2</sub>) and sulfate (SO<sub>4</sub><sup>2-</sup>). The model calculations were summed to obtain estimates of dry sulfur deposition [as sulfur (S)]. A map of dry sulfur deposition fluxes for 2007 is provided in Figure 3-1. The magnitude of a deposition rate is illustrated by the size of the circle. The map depicts five CASTNET sites with fluxes greater than or equal to 5.0 kilograms per hectare per year (kg/ha/yr). These sites are located in Kentucky, Indiana, Ohio, and Pennsylvania. Locations on the map with no value had insufficient data to calculate fluxes. The highest deposition rate was estimated for Quaker City, OH (QAK172) with a flux of 10.3 kg/ha/yr. The highest dry sulfur deposition rates coincided with the major SO<sub>2</sub> source region (Figure 1-5) and declined sharply with distance. The dry deposition rates for the western sites were all less than 1.0 kg/ha/yr with the majority of sites less than 0.5 kg/ha/yr.

Wet deposition values used to estimate total deposition represent 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 (when responsibility for wet deposition monitoring activities at CASTNET sites was transferred to NADP/NTN), those values were used in the data set. For sites where no wet concentrations were measured and for all sites after January 1999, values were obtained from a grid of concentration estimates derived from available NADP/NTN sites by using an inverse distance weighting function. Estimated concentrations were multiplied by the precipitation measured at the CASTNET sites to obtain estimates of wet deposition.

Figure 3-2 provides a map of estimates of total sulfur deposition. The map was constructed by adding dry and wet deposition. The diameters of the circles in the figure illustrate the magnitude of total sulfur deposition and also the relative contributions from wet and dry deposition. The dark shading (blue) signifies the percent wet deposition and the light shading (tan) shows the percent dry deposition. Two regions were observed with total (dry + wet) sulfur deposition greater than 10.0 kg/ha/yr. The larger region extended from eastern Ohio and northern West Virginia across Pennsylvania to New Jersey. The second region encompassed Vincennes, IN (VIN140) and Mackville, KY (MCK131). The highest total sulfur deposition rate was estimated for QAK172, OH with a value of 21.0 kg/ha/yr. Sulfur deposition at western sites was less than 2.0 kg/ha/yr. The contribution of dry deposition was much more significant in and near major source regions. For example, the contribution of dry sulfur deposition ranged from about half of total sulfur deposition at MCK131, KY to less than 20 percent at sites in New England.

**Figure 3-1** Dry Sulfur ( $\text{SO}_2 + \text{SO}_4^{2-}$ ) Deposition (kg/ha/yr) for 2007



**Figure 3-2** Total (Dry + Wet) Sulfur Deposition (kg/ha/yr) for 2007

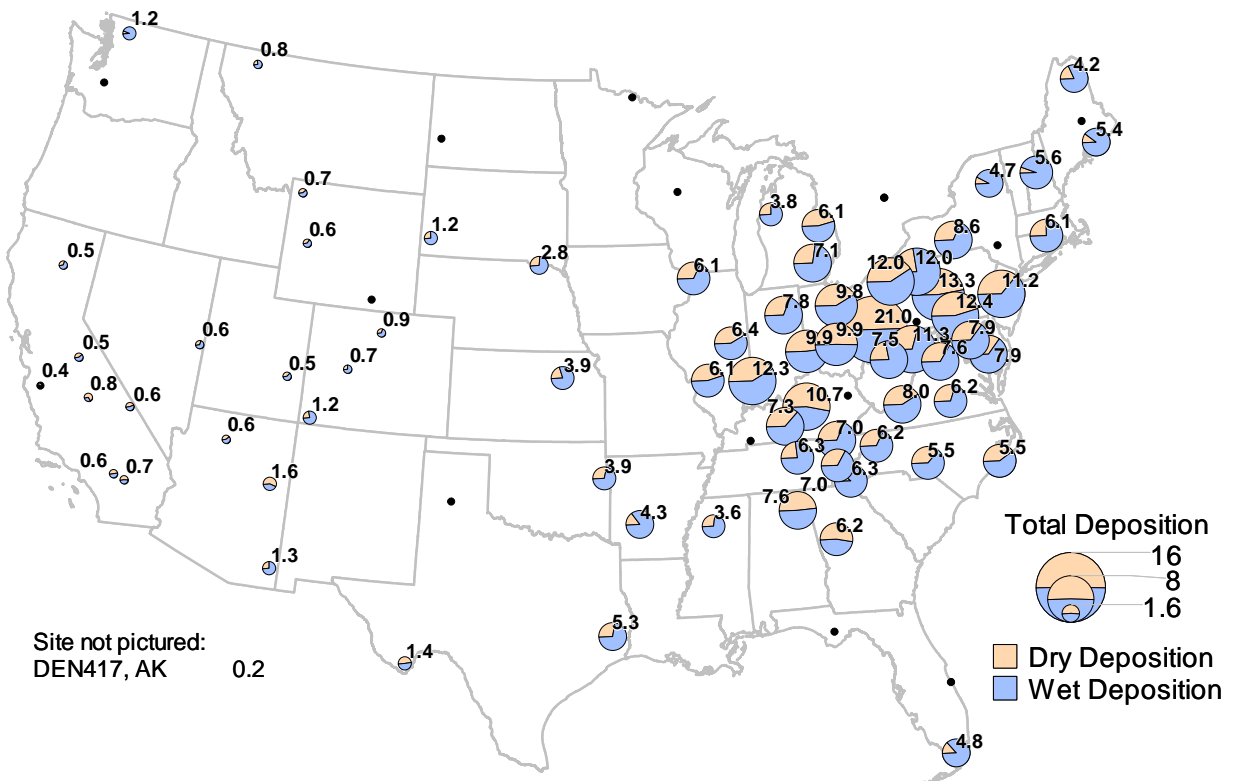
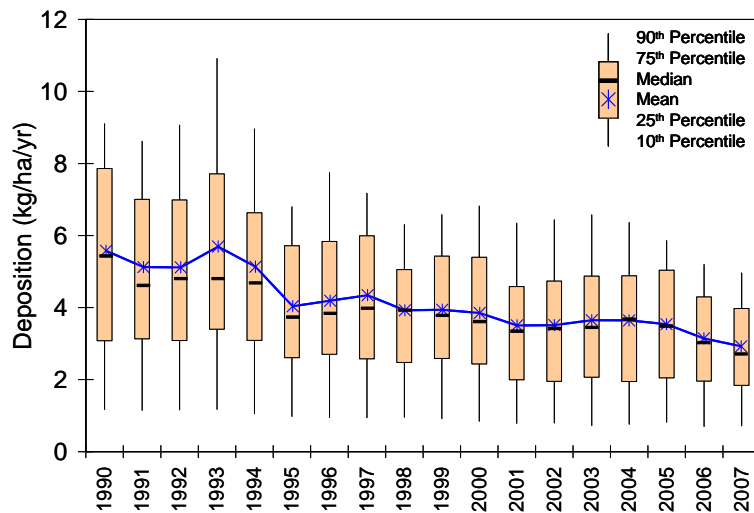




Figure 3-3 presents box plots that show the trend in dry sulfur deposition (as S), and Figure 3-4 shows the trend in annual total (dry + wet) sulfur deposition (as S) over the 18 years, 1990-2007. The box plots were based on data obtained from the 34 eastern CASTNET reference sites (Figure 1-2). Sulfur deposition at these sites declined significantly over the 18 years. Figure 3-5 presents estimates of trends in dry, wet, and total deposition of sulfur (as S) on the same diagram. The trend line for precipitation-weighted mean sulfur concentrations in precipitation shows a decline over the last 18 years with small increases in 2005 through 2007.

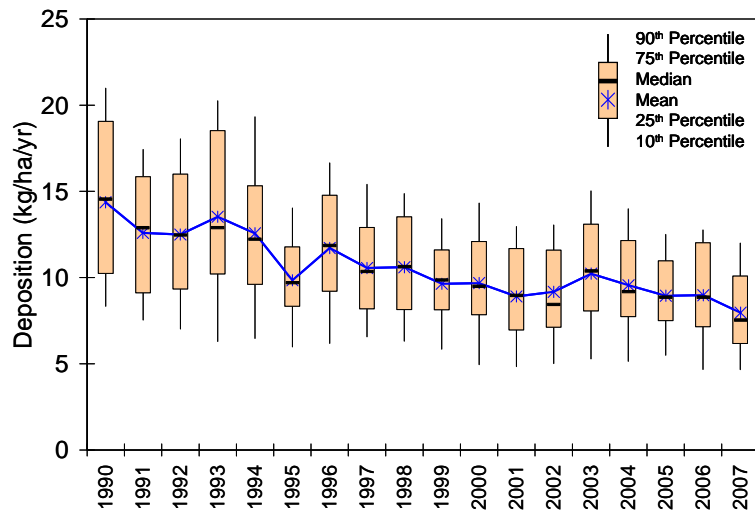
The influence of precipitation on total sulfur deposition is illustrated by comparing the solid (top) line to the dotted blue line in Figure 3-5. The solid line shows total deposition, which depends on sulfur concentrations in precipitation and precipitation amounts. The dotted line shows concentrations in precipitation, which reflect changes in SO<sub>2</sub> emissions. The year 2003 experienced relatively high total sulfur deposition even though the sulfur concentration in precipitation was relatively low. Above average precipitation produced the relatively high total flux of sulfur. Overall, total sulfur deposition declined from a 1990-1992 mean of 13.2 kg/ha/yr to a 2005-2007 mean of 8.7 kg/ha/yr, a 34 percent reduction.

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

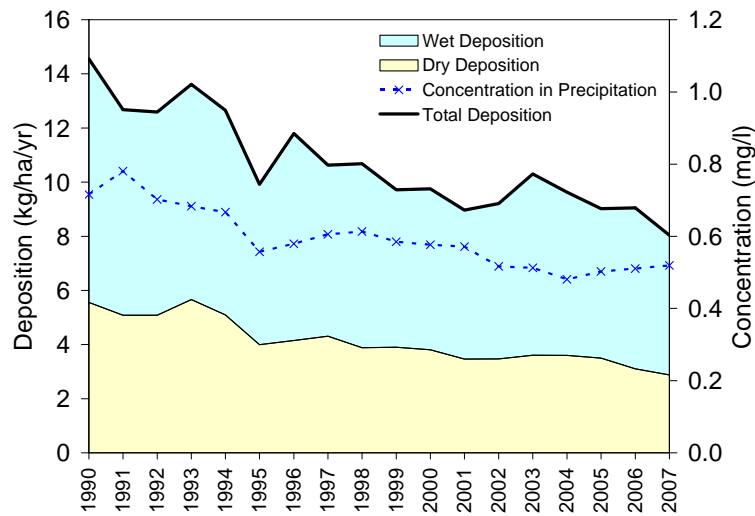


**Note:** All trend diagrams are based on data collected at 34 eastern reference sites.

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



**Figure 3-5** Trend in Sulfur Deposition (kg/ha/yr) with Concentrations in Precipitation (mg/l)



Georgia Station, GA (GAS153)

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## Nitrogen Deposition

Nitrogen deposition, as estimated from CASTNET filter packs, is comprised of fluxes of nitric acid ( $\text{HNO}_3$ ) + nitrate ( $\text{NO}_3^-$ ) + ammonium ( $\text{NH}_4^+$ ). Dry fluxes of nitrogen [as nitrogen (N)] for 2007 are shown in Figure 3-6. This map was constructed by summing the individual MLM simulations for the three species. Almost all of the CASTNET sites in the eastern United States had estimated dry nitrogen deposition rates greater than 1.0 kg/ha/yr. The higher deposition rates were estimated for the Midwestern and Mid-Atlantic States. Deposition rates greater than or equal to 2.0 kg/ha/yr were also calculated for sites in North Carolina, Georgia, Alabama, and at Great Smoky Mountains National Park, TN (GRS420). In the East, the nitrogen fluxes were fairly uniform geographically, reflecting a wide distribution of nitrogen oxides ( $\text{NO}_x$ ) sources (such as motor vehicles). The values at the western sites ranged from 0.2 kg/ha/yr at Mount Rainier National Park, WA (MOR409) and Glacier National Park, MT (GLR468) to 3.0 kg/ha/yr at SEK430, CA. Locations on the map with no value had insufficient data to calculate fluxes.

Figure 3-7 presents a map of total nitrogen deposition (as N) for 2007. The map was constructed by summing the estimates of dry (light shading) and wet (dark shading) deposition. The figure shows that all but a few of the eastern sites recorded deposition rates greater than 5.0 kg/ha/yr. No values above 10.0 kg/ha/yr were observed in 2007. The highest total nitrogen flux (9.8 kg/ha/yr) was estimated for QAK172, OH. Fluxes with values less than 5.0 kg/ha/yr in the eastern United States were estimated for New England, upstate New York, and West Virginia. In the southeastern United States, sites in North Carolina, Tennessee, Florida, Arkansas, and Mississippi had estimated values below 5.0 kg/ha/yr. The values at the western sites ranged from 1.1 kg/ha/yr at Lassen Volcanic National Park, CA (LAV410) to 4.1 kg/ha/yr in southern California at Converse Station (CON186) and Joshua Tree National Monument (JOT403). The contributions of dry nitrogen deposition to total nitrogen were lower than the corresponding contributions of dry sulfur deposition. Dry nitrogen deposition typically contributed less than 50 percent of total deposition in the East. Interestingly, dry nitrogen deposition contributed more than half of total nitrogen deposition at sites in California, a region with elevated concentrations of nitrogen species and limited rainfall. The CASTNET site at QAK172 in eastern Ohio measured both the highest total sulfur and total nitrogen deposition rates.

Figure 3-8 presents box plots that were constructed from data from the 34 reference sites. They show the 18-year trend in dry nitrogen deposition (as N). Figure 3-9 shows the trend in annual total (dry + wet) nitrogen deposition over the period 1990 through 2007. The box plots in Figure 3-8 show a consistent decline in dry nitrogen deposition beginning in 1999. Three-year mean fluxes declined from 2.2 kg/ha/yr in 1990-1992 to 1.7 kg/ha/yr in 2005-2007, a 17 percent decline. Total nitrogen deposition (Figure 3-9) is more variable because the annual wet and total fluxes depend on the amount of precipitation. The figure suggests that total flux has decreased since 1996 with a significant decline over the last four years. Again, the total flux in 2003 was relatively high because of unusually high precipitation.

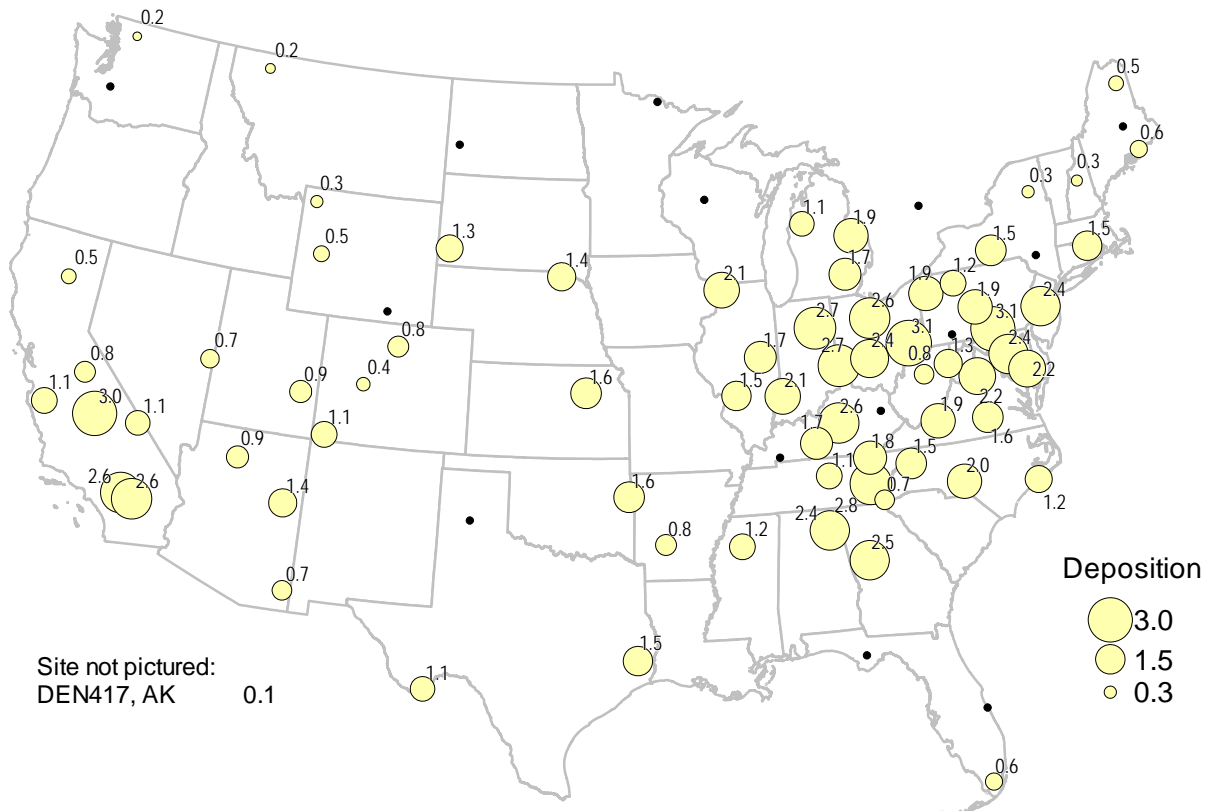
Estimates of trends in wet, dry, and total deposition of atmospheric nitrogen (as N) are presented in Figure 3-10. The trend in precipitation-weighted mean nitrogen concentrations in precipitation is also provided on the same figure. The trend line for precipitation-weighted mean nitrogen concentrations in precipitation shows a downward trend since 1998 with a slight increase over the last three years. This trend line reflects the effect of changes in  $\text{NO}_x$  emissions. Total nitrogen deposition declined 17 percent over the 18 years.



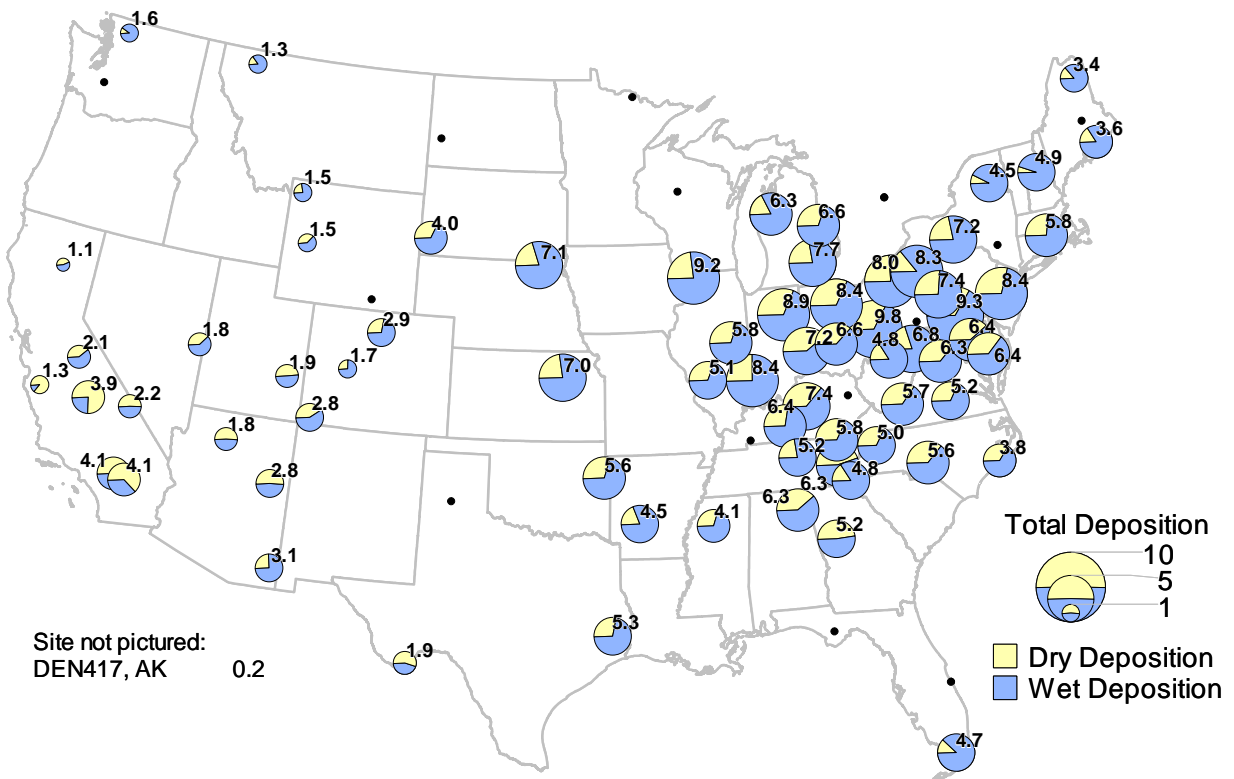
Centennial, WY (CNT169)



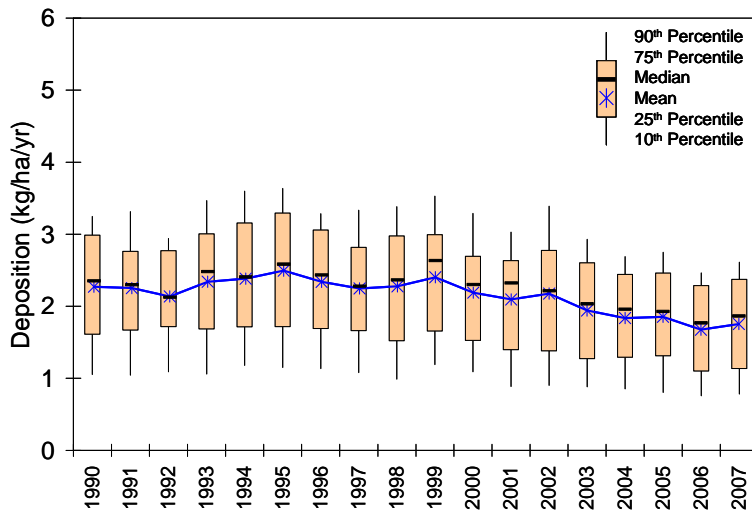
**Figure 3-6** Dry Nitrogen ( $\text{HNO}_3 + \text{NO}_3^- + \text{NH}_4^+$ ) Deposition (kg/ha/yr) for 2007



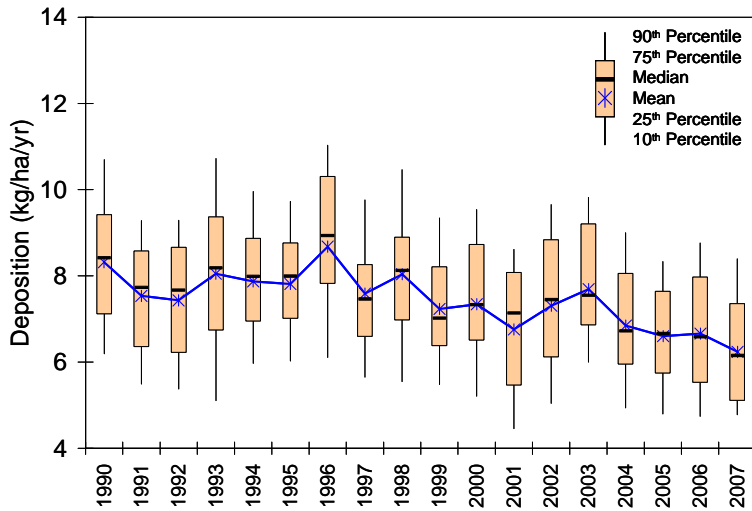
**Figure 3-7** Total (Dry + Wet) Nitrogen Deposition (kg/ha/yr) for 2007



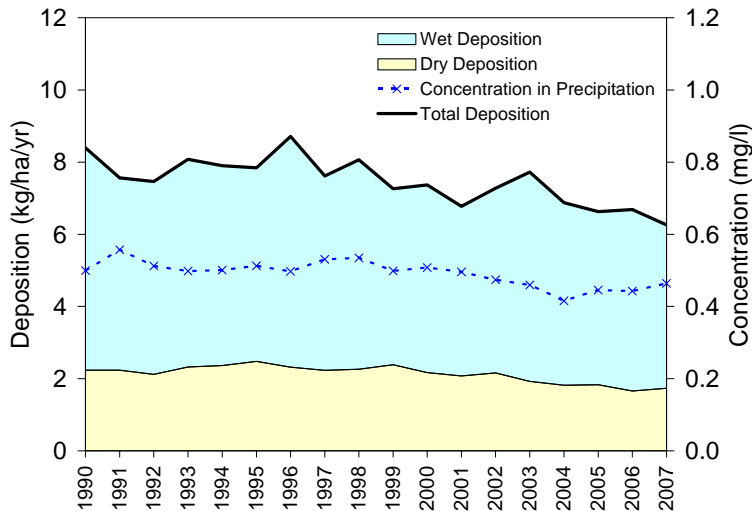
**Figure 3-8** Trend in Dry Nitrogen Deposition (kg/ha/yr)



**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)



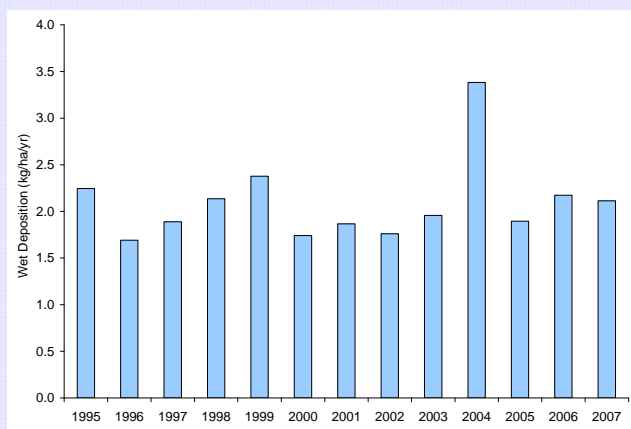
## ***Rocky Mountain Atmospheric Nitrogen and Sulfur (RoMANS) Study***

Rocky Mountain National Park (RMNP) is home to several sensitive, high-elevation ecosystems that are subject to wet and dry deposition of nitrogen and sulfur pollutants. Evidence suggests the park is experiencing changes in ecosystem function and surface water chemistry in addition to visibility degradation as a result of atmospheric pollutants. Consequently, the park has specified a goal for a critical load of 1.5 kg/ha/yr of wet nitrogen deposition (Baron, 2006). This deposition rate is considered protective of park ecosystems. To obtain a better understanding of the effects of air pollution on RMNP, NPS is sponsoring the Rocky Mountain Atmospheric Nitrogen and Sulfur (RoMANS) Study. The objectives of the study are to improve understanding of visibility degradation, improve understanding of atmospheric wet and dry deposition to park ecosystems, provide assessment of sources and contributors to air pollution, and increase knowledge about specific pollutants contributing to air quality problems. The cornerstone of the study was a large field campaign and modeling analysis that was conducted during the spring and summer of 2006 (RoMANS, 2006).

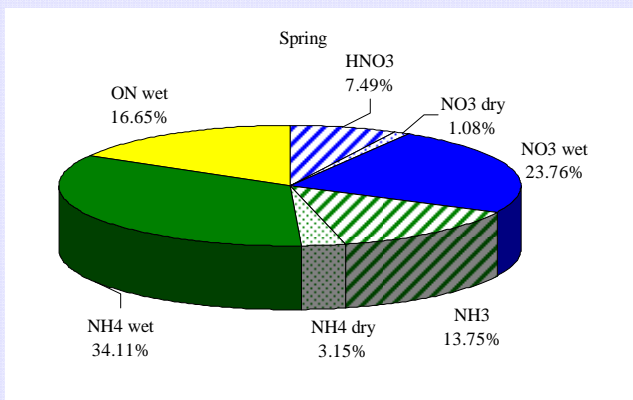
NPS has sponsored standard CASTNET measurements of sulfur and nitrogen pollutants and O<sub>3</sub> near RMNP since 1994, and NADP/NTN measurements of wet nitrogen and sulfur deposition have been collected since 1980. Figure 3-A shows a time series of wet nitrogen deposition fluxes from 1995-2007. These data were interpolated from NADP/NTN measurements for the location of the NPS-sponsored CASTNET site (ROM406) based on methodology discussed in the sulfur deposition section on page 23. It should be noted that NPS policy on nitrogen deposition is based on measurements from the NADP Loch Vale site (CO98). Figure 3-A shows that annual wet nitrogen deposition rates have been consistently higher than the critical load of 1.5 kg/ha/yr. The values ranged from about 1.6 to 3.5 kg/ha/yr. The peak value in 2004 was produced by unusually wet weather.

The spring (3/26-4/28) and summer (7/6-8/11) 2006 RoMANS field studies included precipitation chemistry and gaseous and particulate pollutant and meteorological measurements at 12 to 13 sites in and near RMNP. The core RoMANS site

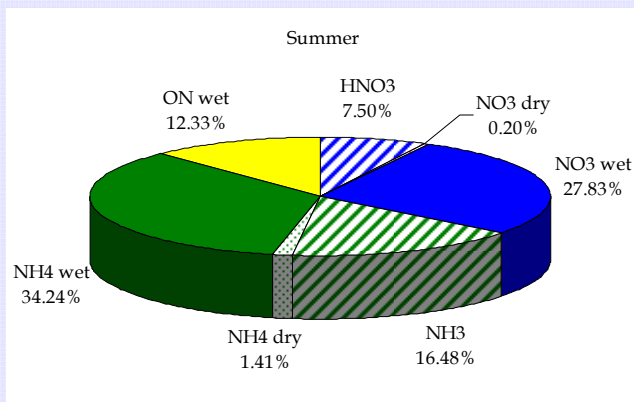
**Figure 3-A** Time Series of Wet Nitrogen Deposition Values at ROM406, CO



**Figure 3-B** Relative contributions of individual N deposition pathways to total measured N deposition at the RoMANS Core Site during the spring campaign



**Figure 3-C** Relative contributions of individual N deposition pathways to total measured N deposition at the RoMANS Core Site during the summer campaign



The two figures illustrate the contributions of wet ON and dry NH<sub>3</sub> deposition to the nitrogen budget. These two parameters are not measured by NADP/NTN and CASTNET. Dry deposition of all species was less than wet deposition during both the spring and summer field campaigns. Wet deposition of nitrogen more than doubled during the summer while sulfur deposition increased by approximately 50 percent. ON in the particle and gas phase was not measured in RoMANS and was not included in the two figures.

Ongoing work includes continuing analyses of the spring and summer field measurements and the identification of meteorological conditions and source types and regions that contribute to deposition in RMNP.

was collocated with CASTNET and IMPROVE measurements at the EPA/NPS collocated site (ROM406/206). Recognizing that CASTNET and NADP/NTN measurements do not represent all nitrogen inputs to deposition, RoMANS sponsored measurement and modeling studies of previously unquantified nitrogen species that contribute to wet and dry deposition. The RoMANS investigators concluded that wet organic nitrate (ON) and ambient levels of ammonia (NH<sub>3</sub>), total reactive oxides of nitrogen (NO<sub>y</sub>), and other nitrogen species contributed to the nitrogen deposition budget. Barna and his colleagues (2008) performed Comprehensive Air Quality Model with Extensions (CAMx) model calculations, which estimated that the deposition of NH<sub>3</sub>, NO<sub>y</sub>, and ON is almost double the quantity of dry nitrogen deposition as currently estimated by CASTNET. Figures 3-B and 3-C show pie charts of nitrogen deposition based on measurements collected during the



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## Chapter 4: Ozone Concentrations

CASTNET is the primary monitoring network for measuring rural, ground-level ozone ( $O_3$ ) concentrations in the United States. The network provides information critical for the evaluation of the NAAQS, which are written in terms of fourth highest daily maximum 8-hour average  $O_3$  concentrations (EPA, 1997). These annual fourth highest daily maximum concentrations are averaged over 3-year periods to determine potential areas of nonattainment.

During early 2008, EPA revised the ozone NAAQS to 0.075 parts per million (ppm; EPA, 2008b). Consequently, the 2007 and 2005-2007  $O_3$  concentrations were analyzed in terms of both the current and the revised standards. Nearly half (24) of CASTNET monitoring stations in the eastern United States and five sites in California would have recorded exceedances of the new standard for the period 2005-2007.

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

On March 12, 2008, EPA revised the NAAQS for ground-level  $O_3$ . EPA revised the 8-hour primary standard to 0.075 ppm (EPA, 2008b). The previous standard, set in 1997 (EPA, 1997), was 0.08 ppm. Because  $O_3$  is measured out to three decimal places, the standard effectively became 0.084 ppm (84 ppb) as a result of rounding. The 1997 ozone NAAQS was exceeded if the fourth highest daily maximum ozone concentration was 0.08 ppm (85 ppb) or higher. EPA also revised the secondary standard to the 0.075 ppm (75 ppb) level making it identical to the primary standard. A primary standard is designed to protect human health while a secondary standard is designed to protect public welfare including protection against visibility impairment and damage to sensitive ecosystems and forests. An area will meet the new standard if the design value, or the 3-year average of the annual fourth highest daily maximum 8-hour average concentrations, is less than or equal to 0.075 ppm. EPA will issue final designations of attainment, nonattainment, and unclassifiable areas no later than March 2010 unless there is insufficient information to make these designation decisions. In that case, EPA will issue designations no later than March 2011. These designations will be made using  $O_3$  measurements from 2006-2008 or later. This chapter presents  $O_3$  concentrations in terms of both standards, 0.08 ppm and 0.075 ppm. More information on trends in ozone concentrations and ozone monitoring requirements can be found in the EPA (2008a) report on National Air Quality Status and Trends through 2007.

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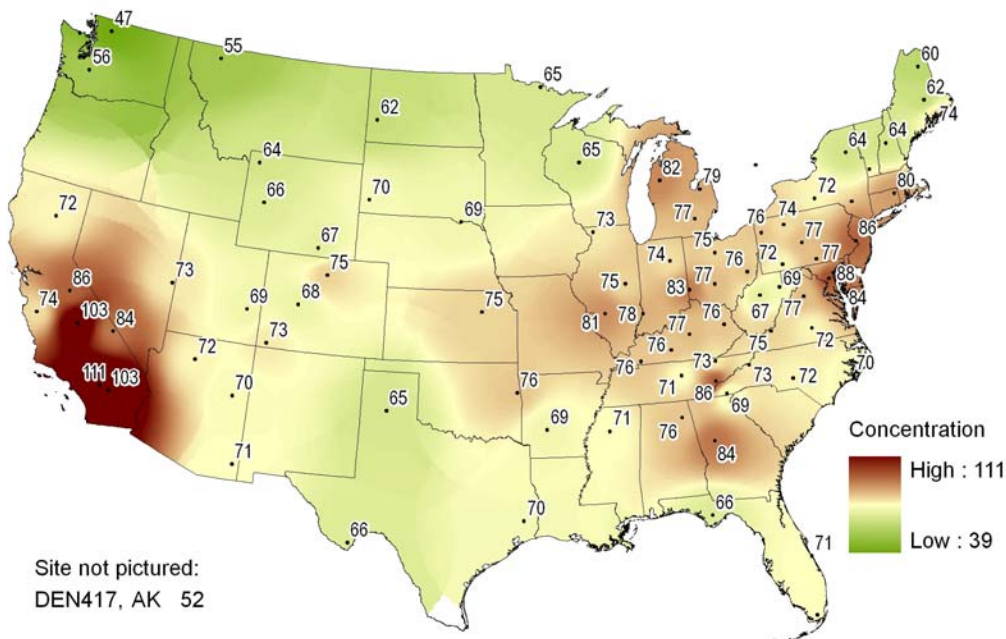
At ground level, O<sub>3</sub> is an air pollutant that can cause harmful effects to the human respiratory system as well as damage to vegetation and ecosystems. Ground-level ozone is formed in the troposphere when volatile organic compounds (VOCs) and NO<sub>x</sub> react in the presence of sunlight. Emissions from motor vehicle exhaust, EGUs, industrial facilities, chemical solvents, and gasoline vapors are the major anthropogenic sources of NO<sub>x</sub> and VOCs. Also, emissions of biogenic VOCs from trees and other vegetation contribute to O<sub>3</sub> formation in rural areas (EPA, 2008d). Emission reduction strategies for VOCs have been successful in reducing high O<sub>3</sub> concentrations in and downwind of urban areas. NO<sub>x</sub> emission reductions were mandated by the ARP, NO<sub>x</sub> SIP Call, and other NO<sub>x</sub> emission control programs to reduce O<sub>3</sub> concentrations. These reductions will continue to be required in order to achieve additional declines in 8-hour average O<sub>3</sub> levels.

The concentration shading for the figures used in this chapter was prepared based on rural ozone concentration data using an algorithm with inverse distance cubed weighting with a radius of influence of 500 km. Consequently, concentration estimates for areas with limited monitoring site coverage, such as much of Texas, and concentrations depicted for urban areas, such as Atlanta, are approximations. Additional maps of ozone concentrations can be viewed on the Web site for the NPS Air Atlas (<http://science.nature.nps.gov/AirAtlas/AirAtlas0105/viewer.htm>).

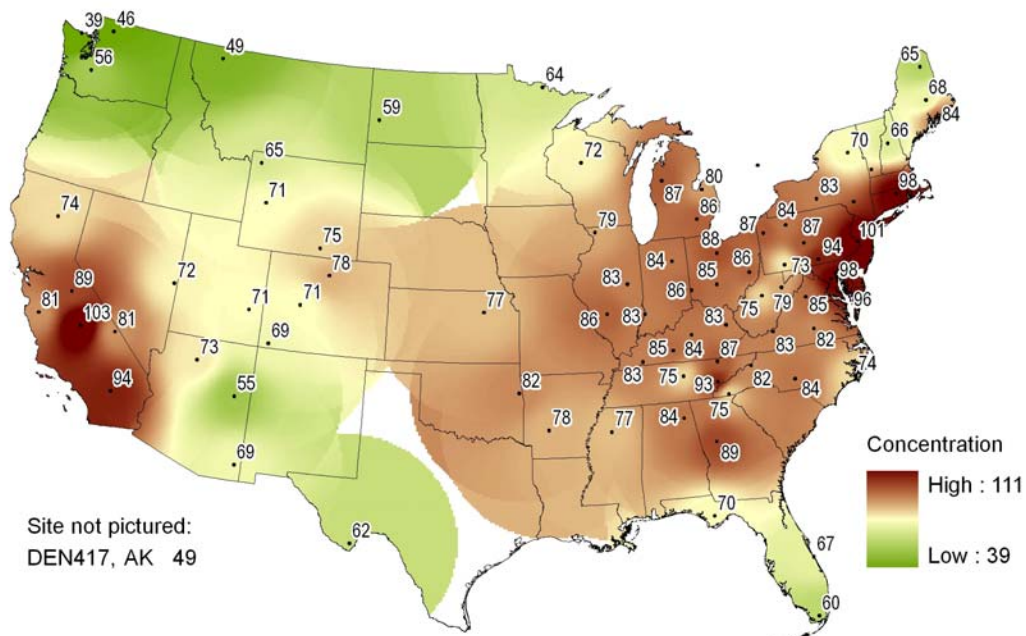
## CASTNET Ozone Concentrations

Figure 4-1 presents 3-year averages of the fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations for 2005-2007. During this period, 3-year averages of the fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations were greater than or equal to 85 ppb at four sites in California, one in New Jersey, one in Maryland, and at Great Smoky Mountains National Park, TN (GRS420). Figure 4-1 also shows the geographic distribution of fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations greater than 75 ppb, which exceed the revised NAAQS (0.075 ppm). The geographic extent of 8-hour levels greater than the new standard is obviously much larger than for values greater than or equal to 85 ppb. Twenty-four eastern and five sites in California exceeded the new standard. Although the CASTNET monitors are not Part 58 compliant, these 3-year average (2005-2007) concentrations represent current design values for the 8-hour NAAQS for O<sub>3</sub> as measured at each site. For example, the value of 111 ppb at Converse Station, CA (CON186) would have to be reduced to 84 ppb to achieve the old standard and to 75 ppb to achieve the new standard. Design values change as a new 3-year database becomes available.

**Figure 4-1** Three-Year Average of Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) for 2005-2007



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



## *National Ambient Air Quality Standards for Ozone*

### **New 8-Hour Ozone Standards**

EPA's lowering of the primary and secondary standards to 0.075 ppm (EPA, 2008b) will improve protection of public health and sensitive vegetation and ecosystems. An area will meet the new standard if the 3-year average of the annual fourth highest daily maximum 8-hour average concentration is less than or equal to 0.075 ppm. The 3-year average reduces the influence that meteorological conditions have on the extent and magnitude of ozone formation. For more information on the revised 8-hour ozone standard, visit the EPA Web site [http://www.epa.gov/air/ozonepollution/pdfs/2008\\_03\\_factsheet.pdf](http://www.epa.gov/air/ozonepollution/pdfs/2008_03_factsheet.pdf). More general information on ozone also can be found at <http://www.epa.gov/groundlevelozone/>.

### **Assessing Compliance with the New Standard**

For regulatory purposes, EPA collects ozone measurements on an hourly basis. To comply with the standard, daily maximum 8-hour average ozone concentrations at a monitor cannot exceed 0.075 ppm during the ozone season. To determine if a monitor is in or out of attainment:

- ★ Hourly ozone measurements are used to compute 8-hour average concentrations.
- ★ The daily maximum 8-hour average is calculated for each day.
- ★ For each year, the fourth highest daily maximum concentration is calculated.
- ★ These annual fourth highest daily maximum concentrations are averaged over 3-year periods.
- ★ If the 3-year average of the fourth highest daily maximum 8-hour average O<sub>3</sub> concentration is greater than 0.075 ppm (or 75 ppb), the area will be designated as a “nonattainment area” and held accountable for reaching the attainment status.

To comply with the new standard, the 3-year average of the daily maximum 8-hour average ozone concentrations at a monitor cannot exceed 0.075 ppm during the ozone season. EPA estimates, based on 2004-2006 data, 345 counties would have exceeded the new NAAQS ([http://www.epa.gov/air/ozonepollution/pdfs/2008\\_03\\_monitors\\_violating\\_2008.pdf](http://www.epa.gov/air/ozonepollution/pdfs/2008_03_monitors_violating_2008.pdf)). EPA will not designate areas as nonattainment based on these three years but will use 2006-2008 or later data. EPA is scheduled to make final determinations for designation of areas as attainment, nonattainment, or unclassifiable no later than March 2011. For further information, see the EPA Fact Sheet for the final revisions to the ozone NAAQS at [http://www.epa.gov/air/ozonepollution/pdfs/2008\\_03\\_factsheet.pdf](http://www.epa.gov/air/ozonepollution/pdfs/2008_03_factsheet.pdf).



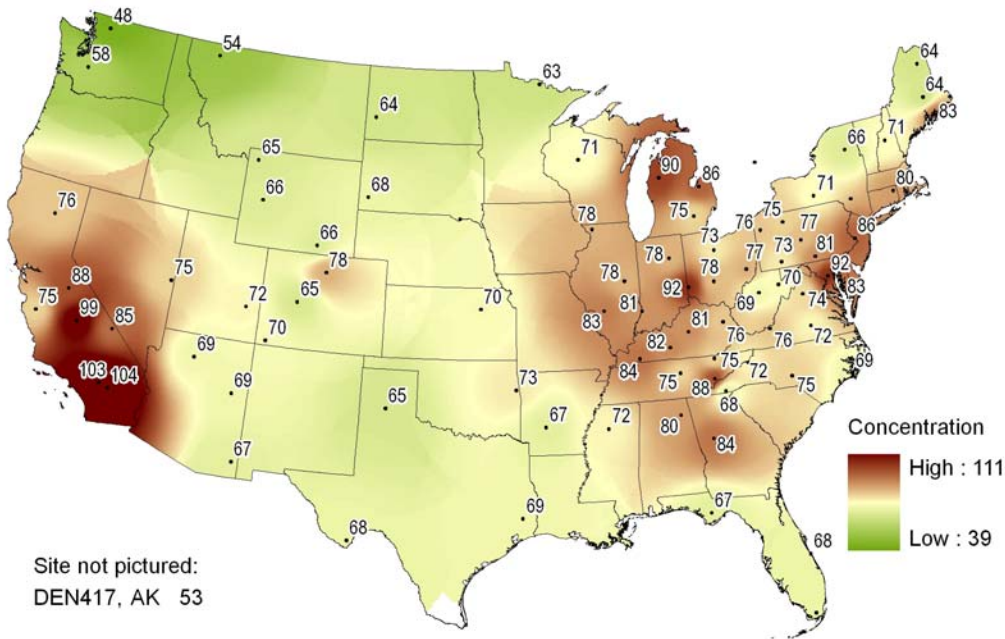
For comparison to current design values, 3-year averages of the fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations for 2000-2002 are presented in Figure 4-2. This period was selected for comparison because the NO<sub>x</sub> SIP Call was implemented in 2003. A comparison of O<sub>3</sub> data for these two 3-year periods illustrates the effectiveness of the emission reduction programs. Using the 84 ppb standard, nineteen eastern and three 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, and Illinois. Exceedances were also recorded in Kentucky, Tennessee, and greater Atlanta. Using the 75 ppb standard, the map shows that most of the Eastern sites measured concentrations greater than 75 ppb and five monitors in California measured concentrations greater than 75 ppb. The period 2005-2007 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 O<sub>3</sub> NAAQS. However, sites in California, along the East coast, and at GRS420, TN continued to measure elevated ozone levels.

Measurements of fourth highest daily maximum 8-hour average concentrations during 2007 (Figure 4-3) were somewhat higher than 2006 but were considerably lower than concentrations measured over the period 2000-2002. Six CASTNET monitors in the East measured fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations greater than or equal to 85 ppb. These sites are widely scattered geographically and are located in five states. Peak values of 92 ppb were measured in the East at Beltsville, MD (BEL116) and Oxford, OH (OXF122). GRS420, TN continued to measure 8-hour average O<sub>3</sub> levels above 85 ppb. Five of seven sites in California measured fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations greater than or equal to 85 ppb. The monitor at Joshua Tree National Monument, CA (JOT403) measured the highest value (104 ppb) in the network during 2007. Figure 4-3 also depicts the geographic distribution of fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations greater than 75 ppb.

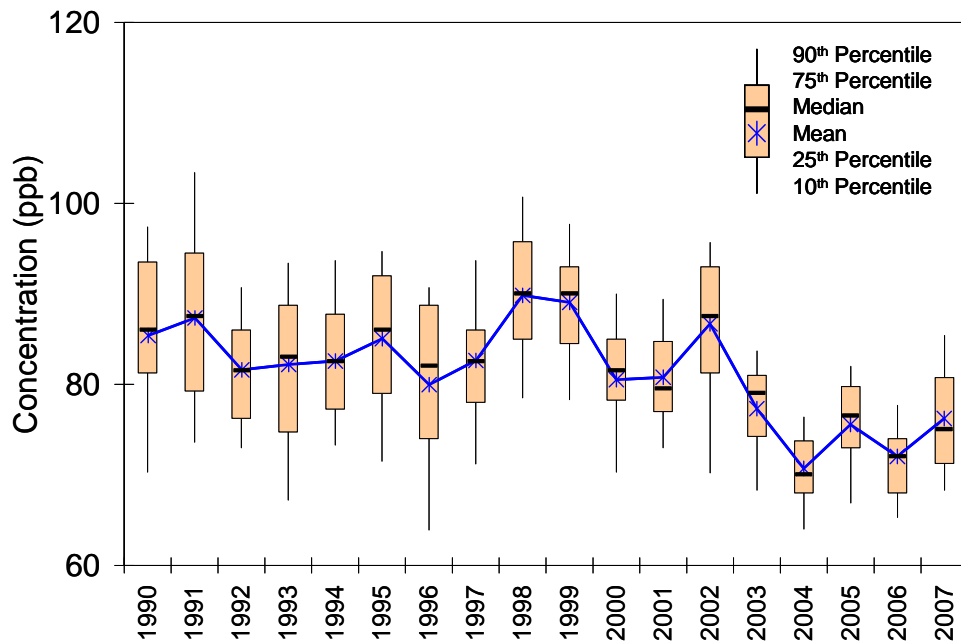
The box plots depicting the trend in fourth highest daily maximum 8-hour average O<sub>3</sub> concentrations from the CASTNET eastern reference sites are provided in Figure 4-4. The median value (75 ppb) from the reference site data for 2007 was the third lowest in the 18-year period. It represents a significant improvement over the 2002 median value of 87.5 ppb and the peak median value of 90 ppb that was measured in 1998 and 1999.



**Figure 4-3** Fourth Highest Daily Maximum 8-Hour Average Ozone Concentrations (ppb) 2007



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



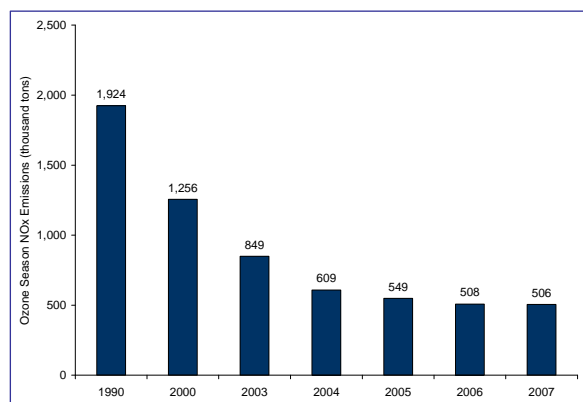
**Note:** All trend diagrams are based on data collected at 34 eastern reference sites

## Nitrogen Oxides and Ozone

EPA recently analyzed changes in NO<sub>x</sub> emissions and O<sub>3</sub> concentrations from 1990-2007. The findings were published in a report (EPA, 2008c) that summarized 2007 compliance and environmental results from the NO<sub>x</sub> SIP Call/NBP. The report discussed the NO<sub>x</sub> and VOCs emission control programs, their effectiveness, subsequent changes in NO<sub>x</sub> and VOCs emissions in the eastern United States, and the resulting changes in ozone levels. Figure 4-5, which was taken from the *NO<sub>x</sub> Budget Trading Program Compliance and Environmental Results* (EPA, 2008c), depicts ozone season NO<sub>x</sub> emissions from all NBP sources in the eastern United States from 1990-2007 (see Figure 4-7 for states covered by the NBP).

Specifically, EPA reported that NBP sources emitted 506,000 tons of NO<sub>x</sub> in 2007, constituting a 74 percent reduction in ozone season NO<sub>x</sub> emissions from NBP sources from 1990 levels. Most of the emission reductions since 1990 were the result of the ARP NO<sub>x</sub> reduction program and other federal, state, and local programs. The significant declines after 2000 resulted from the OTC trading program, which was in effect between 1999 and 2002, and the NBP, which began in 2003.

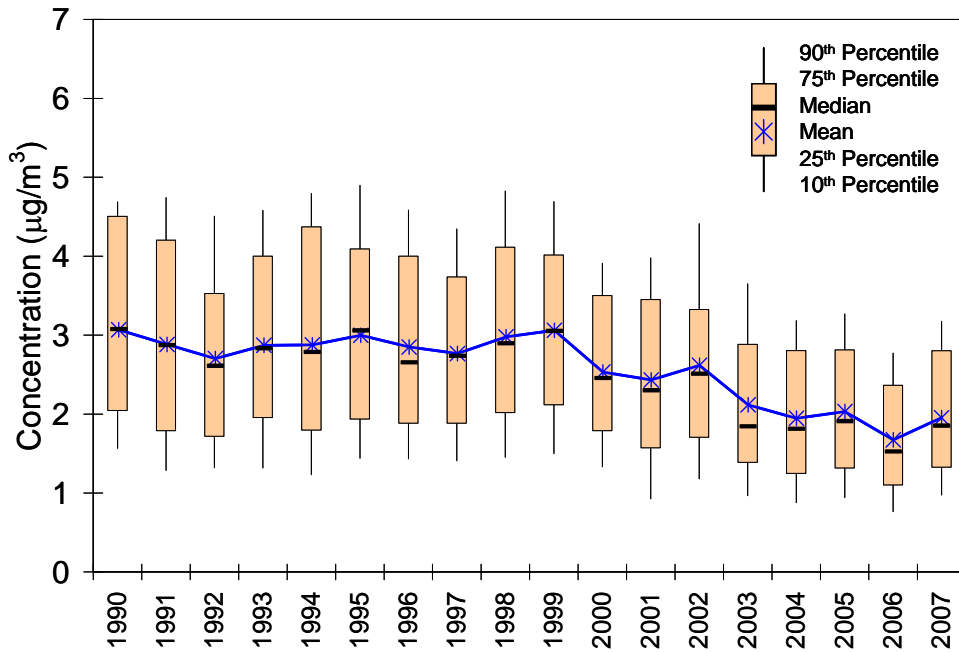
**Figure 4-5** Ozone Season NO<sub>x</sub> Emissions from All NBP Sources



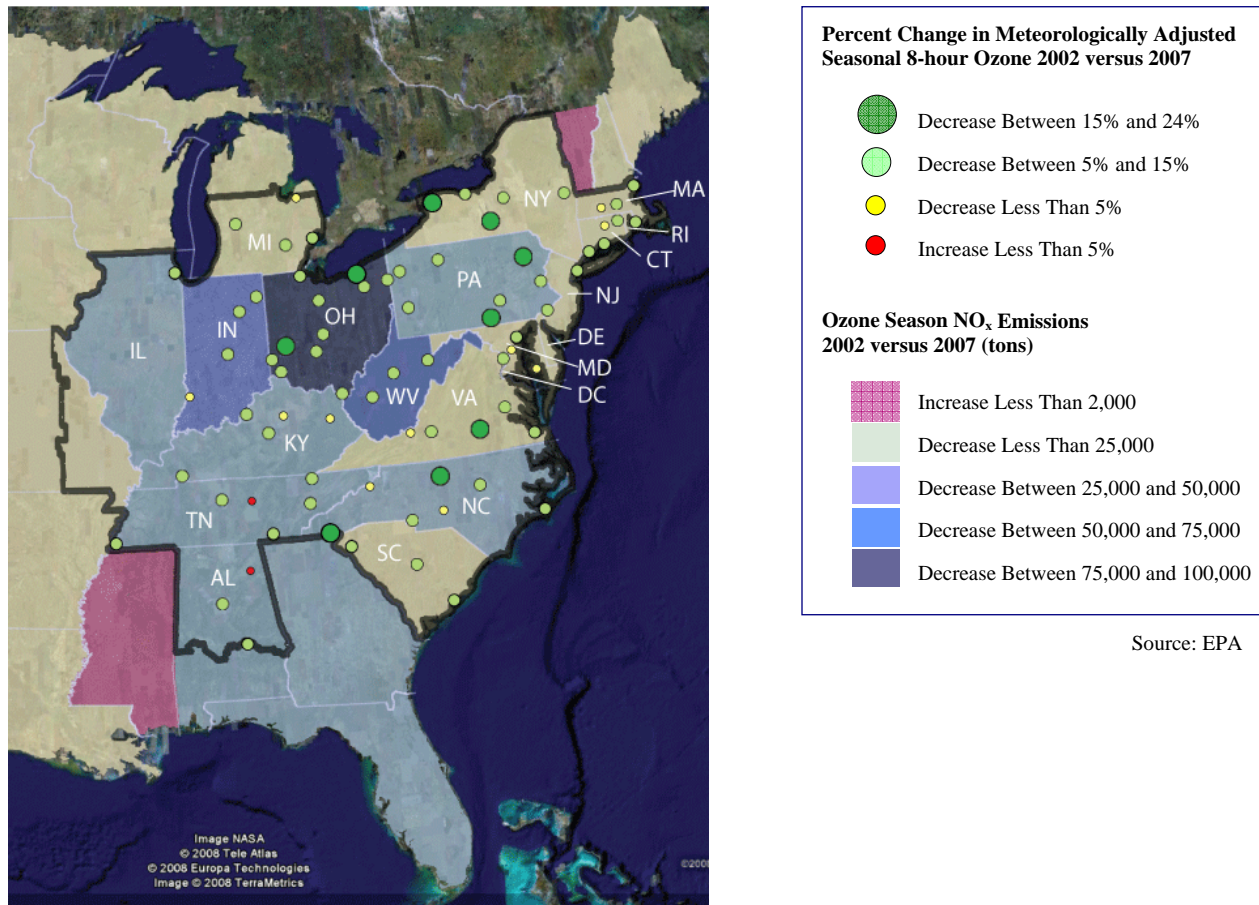
Source: EPA

Trends in CASTNET measurements of total nitrate concentrations (Figure 4-6) measured during the ozone season (May through September) at the 34 eastern reference sites show almost no decline from 1990 through 1999 and a more significant decline thereafter as the emission reductions from the OTC and NBP became effective. Three-year median concentrations before (2000-2002) and after (2003-2005) the implementation of the NBP were 2.43  $\mu\text{g}/\text{m}^3$  and 1.87  $\mu\text{g}/\text{m}^3$ , respectively. The most recent three-year (2005-2007) median value was 1.77  $\mu\text{g}/\text{m}^3$ . Figure 4-7, which was taken from the 2008 EPA Report on NBP compliance (EPA, 2008c), shows the relationship between NO<sub>x</sub> emission reductions from power industry sources and changes in O<sub>3</sub> levels at CASTNET and EPA AQS sites in the eastern United States over the period 2002-2007. The figure shows declines in concentrations in all NBP states. The figure also shows a strong association between regions with greatest emission reductions and downwind monitoring sites showing the greatest improvement in ozone levels.

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



**Figure 4-7** Reductions in Ozone Season Power Industry  $\text{NO}_x$  Emissions and Changes in 8-Hour Ozone, 2002 versus 2007

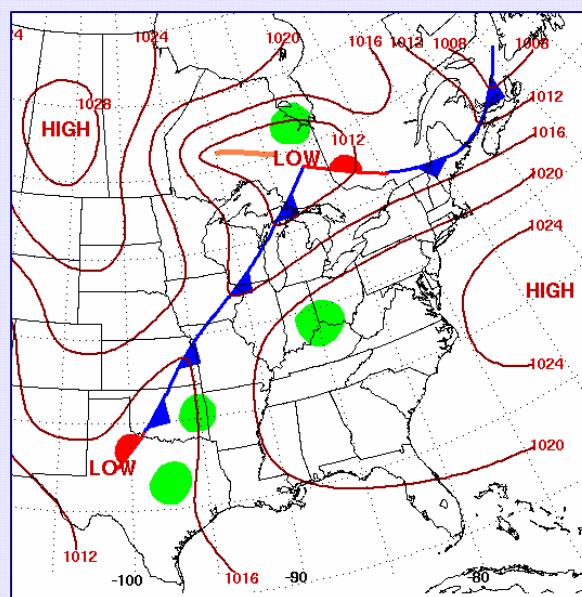




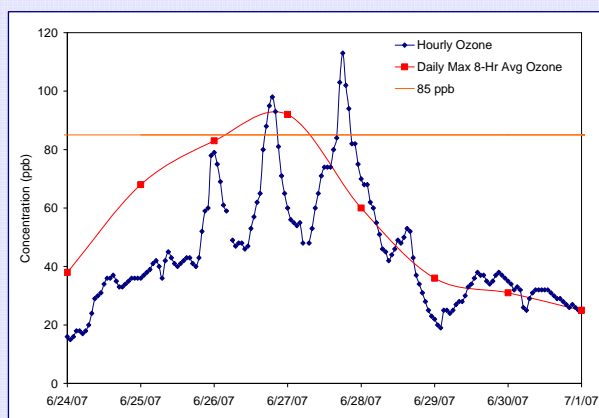
## Ozone Episode in ACA416, ME during 2007

Historical CASTNET O<sub>3</sub> and meteorological data show summer high pressure systems have occasionally produced transport of ozone plumes along the East coast from Washington, DC all the way to Acadia National Park, ME (ACA416). A 3-day ozone episode was observed at the park during June 25-28, 2007. Surface weather conditions are illustrated in Figure 4-A, which shows a Bermuda high off the East coast that produced southwesterly winds over New England. A time series of hourly O<sub>3</sub> concentrations observed at ACA416, ME during June 24-July 1, 2007 is provided in Figure 4-B. Daily maximum 8-hour average O<sub>3</sub> concentrations are also shown. The 8-hour levels are plotted at 00:00 for each day. Figure 4-B shows the gradual buildup of O<sub>3</sub> beginning on June 24 with a subsequent peak hourly value of 113 ppb and a peak daily 8-hour average value of 92 ppb on June 27. The episode was terminated by the passage of a cold front on June 28.

**Figure 4-A** Surface Weather Map for 6/27/07

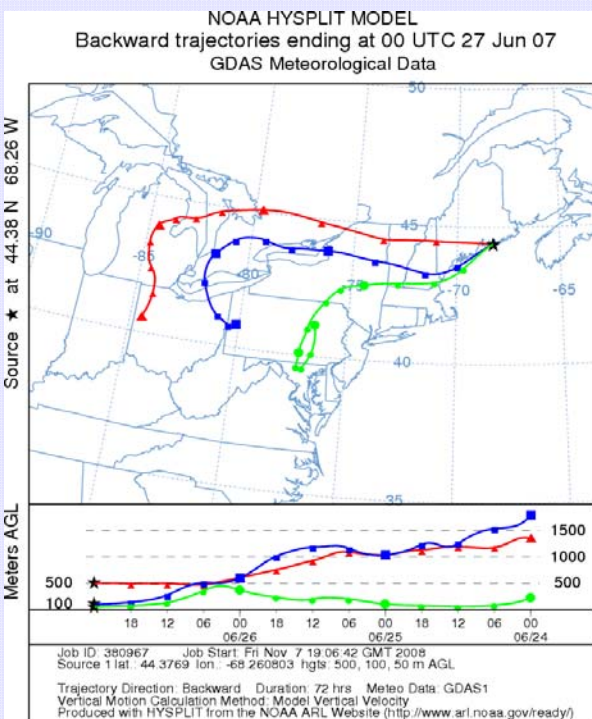


**Figure 4-B** Time Series of Ozone Concentrations at ACA416, ME

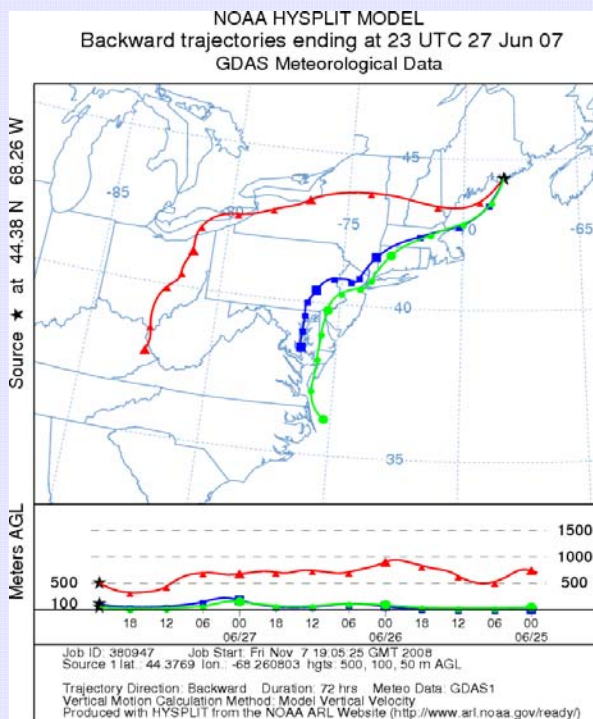


Backward trajectories from the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) Model are provided in Figures 4-C and 4-D for June 27, 2007, 00:00 Coordinated Universal Time (UTC) [June 26, 2007, 19:00 Eastern Standard Time (EST)] and for June 27, 2007, 23:00 UTC (June 27, 2007, 18:00 EST). Trajectories ending at ACA416, ME are given for 50 meters (m), 100 m, and 500 m above ground level. The three trajectories ending at ACA416, ME at 19:00 EST on June 26 show disparate origins. The 50 m trajectory originated in eastern Pennsylvania. The other two trajectories originated further west. All three trajectories show significant subsidence. Hourly ozone concentrations reached 79 ppb at 00:00 EST and 98 ppb at 19:00 EST on June 26. The recording of high concentrations late in the day suggests the transport of ozone and its precursors along the trajectory pathways.

**Figure 4-C** Backward Trajectories ending at June 27, 2007 00:00 UTC



**Figure 4-D** Backward Trajectories ending at June 27, 2007 23:00 UTC



Concentrations increased to an hourly value of 113 ppb at 18:00 EST and a daily maximum 8-hour value of 92 ppb on June 27. These concentrations were the highest measured at ACA416, ME during 2007. The 50 m and 100 m trajectories that ended at the park at 18:00 EST on June 27 both traveled along the East coast from North Carolina and Virginia over the Gulf of Maine to ACA416, ME. The transport of ozone and its precursors along this heavily populated region resulted in elevated ozone concentrations. Ray and his colleagues (Ray *et al.*, 1996) demonstrated that ozone precursors over the Gulf of Maine add to the pool of ozone available for transport to coastal locations by daily sea breezes. In other words, offshore urban plumes are brought inland by sea breezes and contribute to ozone measured at the national park. The combination of long-range transport and local wind patterns resulted in ozone concentrations in excess of the national standard at the park, almost 200 miles from Boston, the nearest major population center.



Acadia National Park, ME (ACA416)  
Photo Courtesy NPS



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## Chapter 5: Data Quality

CASTNET measurements and supporting activities are assessed routinely. This assessment assures that CASTNET data meet data quality objectives (DQO). DQO are evaluated using data quality indicators (DQI) such as precision, accuracy, and completeness. Measurements taken during 2007 and historical data collected over the period 1990-2006 were analyzed relative to DQI 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.

The CASTNET Quality Assurance (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 2007 QA program elements are documented in the QAPP, Revision 4.0 (MACTEC, 2007d). The QAPP is comprehensive and includes standards and policies for all components of project operation from site selection through final data reporting. It includes major sections on field measurements, chemical analysis of field samples, data management, and assessments and response actions. Standard operating procedures are included as appendices.

Data quality indicators (DQI) are quantitative statistics and qualitative descriptors used in interpreting the degree of acceptability and utility of the data collected. The DQI for CASTNET are precision, accuracy, completeness, bias, representativeness, and comparability. Precision, accuracy, and completeness for CASTNET 2007 data were analyzed and compared with historical data collected during the period 1990-2006. The information in this report is supplemented by analyses that are discussed in quarterly CASTNET Quality Assurance Reports (MACTEC, 2007c; 2007f; 2007g; 2008b). These QA reports are produced four times per year with the fourth quarter report including an annual summary.



Huntington Wildlife Forest, NY (HWF187)

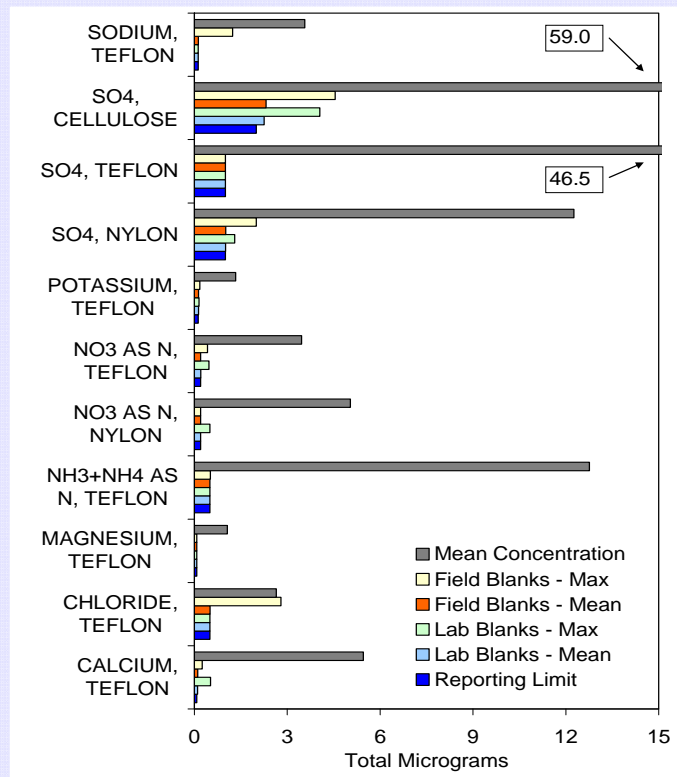
## *Analysis of CASTNET Laboratory and Field Blanks*

CASTNET utilizes an open-face, three-stage filter pack to measure sulfur and nitrogen species. Filter pack handling procedures are designed to minimize handling and transfers and, consequently, any opportunities for contamination and misdirection. Sample integrity is maintained by ensuring that materials in contact with filters do not affect measured analytes. These materials must be traceable to enable documentation of their contact with a sample. These goals are supported by documented chain of custody and evaluation of filter acceptance tests, laboratory blanks, and field (trip) blanks.

Two laboratory blanks are prepared each week while the filter packs are being prepared for the field. Each blank contains a filter from the same lot as the filters used in preparation of the field filter packs. The filters selected for the laboratory blanks are placed directly into the extraction bottles, bypassing the handling involved in loading and unloading the filter packs. These filters then are extracted and analyzed to provide an indication of the effects of filter handling within the laboratory.

Field blanks are prepared quarterly for each CASTNET site. The filter packs used for the field blanks contain a nonstandard connection that cannot be installed in the sampling system in the field. The filter pack with the field blank is prepared and loaded according to standard CASTNET procedures. The field blanks are clearly identified and labeled to prohibit their removal from the resealable plastic bag used for shipping. When the field blank filter is returned from a site, it is unpacked, extracted, and analyzed following procedures for exposed filters. The field blank samples indicate the potential for contamination from the filter pack loading, shipping, and unloading processes.

**Figure 5-A** Summary of Filter Blanks for 2007





The results of the field and laboratory blanks for 2007 are summarized in Figure 5-A and Table 5-A. All results were within the two times reporting limit criterion, with the exception of two cellulose field blank results and one Teflon<sup>®</sup> field blank result. For example, 340 Teflon<sup>®</sup> field blanks were analyzed for NO<sub>3</sub> and only 13 values were above the 0.20 microgram (µg) reporting limit, and the highest value was 0.423 µg. The cellulose filters are impregnated with potassium carbonate (K<sub>2</sub>CO<sub>3</sub>) and undergo additional handling. Consequently, about a third of the cellulose field and laboratory blanks tested positive for small amounts of sulfate. However, no SO<sub>4</sub><sup>2-</sup> values exceeded three times the reporting limit and only two exceeded two times the limit. In short, very few field and laboratory blanks showed significant contamination, and the magnitude of concentrations above detection was small.

**Table 5-A** Evaluation of 2007 Field and Laboratory Filter Blanks

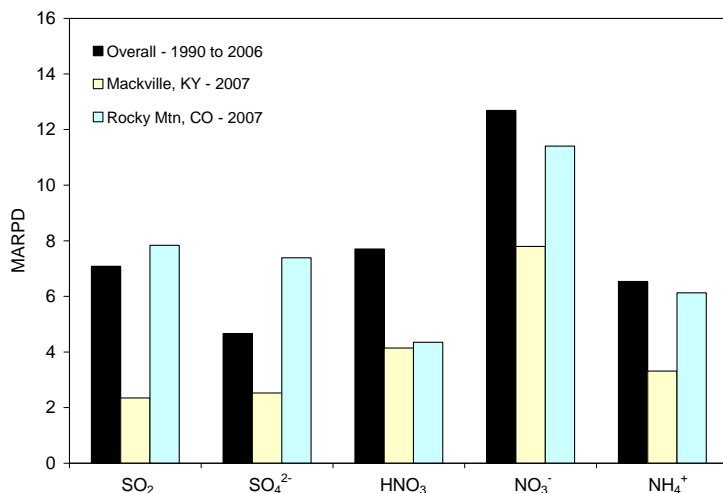
Parameter Name	Reporting Limit Total µg	Total Number	Number > Reporting Limit	Average Total µg	Average Absolute Deviation	Maximum Total µg
<b>FIELD BLANKS</b>						
Teflon <sup>®</sup> - NH <sub>4</sub> <sup>+</sup> -N	0.500	341	1	0.500	0.000	0.515
Teflon <sup>®</sup> - NO <sub>3</sub> -N	0.200	340	13	0.203	0.006	0.423
Teflon <sup>®</sup> - SO <sub>4</sub> <sup>2-</sup>	1.000	340	0	1.000	0.000	1.000
Cl <sup>-</sup>	0.500	340	1	0.507	0.013	2.790
Ca <sup>2+</sup>	0.075	340	26	0.106	0.037	0.251
Mg <sup>2+</sup>	0.075	340	0	0.075	0.000	0.075
Na <sup>+</sup>	0.125	340	4	0.130	0.009	1.230
K <sup>+</sup>	0.125	340	4	0.135	0.012	0.177
Nylon - NO <sub>3</sub> -N	0.200	342	0	0.200	0.000	0.200
Nylon - SO <sub>4</sub> <sup>2-</sup>	1.000	342	33	1.012	0.021	2.000
Cellulose - SO <sub>4</sub> <sup>2-</sup>	2.000	358	138	2.313	0.405	4.550*
<b>LABORATORY BLANKS</b>						
Teflon <sup>®</sup> - NH <sub>4</sub> <sup>+</sup> -N	0.500	96	0	0.500	0.000	0.500
Teflon <sup>®</sup> - NO <sub>3</sub> -N	0.200	92	3	0.205	0.010	0.468
Teflon <sup>®</sup> - SO <sub>4</sub> <sup>2-</sup>	1.000	92	0	1.000	0.000	1.000
Cl <sup>-</sup>	0.500	92	0	0.500	0.000	0.500
Ca <sup>2+</sup>	0.075	92	3	0.101	0.037	0.520
Mg <sup>2+</sup>	0.075	92	0	0.075	0.000	0.075
Na <sup>+</sup>	0.125	92	0	0.125	0.000	0.125
K <sup>+</sup>	0.125	92	0	0.132	0.010	0.150
Nylon - NO <sub>3</sub> -N	0.200	96	0	0.200	0.000	0.500
Nylon - SO <sub>4</sub> <sup>2-</sup>	1.000	96	3	1.006	0.012	1.300
Cellulose - SO <sub>4</sub> <sup>2-</sup>	2.000	96	32	2.250	0.344	4.050

**Note:** Average is defined as average of total number of blanks

Maximum is highest value from total number of blanks

\* A second blank had a value of 4.250 µg compared to the reporting limit of 2.00 µg. No other blanks exceeded the limit.

**Figure 5-1** Historical and 2007 Precision Data for Atmospheric Concentrations



## Precision

### Exposed Filter Concentrations

Figure 5-1 provides a bar chart in which the bars represent precision estimates for five CASTNET analytes. Precision is defined as the mean absolute relative percent difference (MARPD).

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

Where:

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

Historical (1990-2006) data for all 11 collocated site pairs operated over the history of the network and the 2007 data for the current collocated sites at Mackville, KY (MCK131/231) and Rocky Mountain National Park, CO (ROM406/206) are provided. The two sites at Rocky Mountain National Park are operated independently. ROM206 is operated on behalf of EPA and ROM406 on behalf of NPS. Trace cations and chloride are excluded from this figure but are shown later in Figure 5-2. The historical results vary from just over 4 percent for particulate sulfate (SO<sub>4</sub><sup>2-</sup>) to more than 12 percent for particulate nitrate (NO<sub>3</sub><sup>-</sup>). The historical MARPD for SO<sub>4</sub><sup>2-</sup> met the criterion for the CASTNET filter pack measurements shown in Table 5-1. The historical results for sulfur dioxide (SO<sub>2</sub>) and nitric acid (HNO<sub>3</sub>) were above the 5 percent criterion but are considered reasonable. The results for ammonium (NH<sub>4</sub><sup>+</sup>) met the goal of 10 percent. The results for NO<sub>3</sub><sup>-</sup> were significantly above the 5 percent goal. Historically, the precision of NO<sub>3</sub><sup>-</sup> measurements has been consistently worse than for the other analytes, possibly because NO<sub>3</sub><sup>-</sup> concentrations are the lowest of all the pollutants, and nitrate species include sampling artifacts (Lavery *et al.*, 2009).

The 2007 precision results shown in Figure 5-1 indicate that the MARPD data for MCK131/231 were lower than (i.e., more precise than) the historical results for all five parameters. Four parameters (SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup>, HNO<sub>3</sub>, and NH<sub>4</sub><sup>+</sup>) met precision criteria. The 2007 results for ROM406/206 showed greater precision than historical results for HNO<sub>3</sub>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>; the results for HNO<sub>3</sub> and NH<sub>4</sub><sup>+</sup> met the DQI criteria. The precision criterion for NO<sub>3</sub><sup>-</sup> was not met at either site during 2007. The precision results for SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup>, HNO<sub>3</sub>, and NH<sub>4</sub><sup>+</sup> for MCK131/231 are considered acceptable. The precision results were worse for ROM406/206 because of lower concentrations measured at this site. Overall, the filter pack precision results for 2007 were about the same as the 2006 results.

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

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

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

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

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

MARPD = See Equation 5-1

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

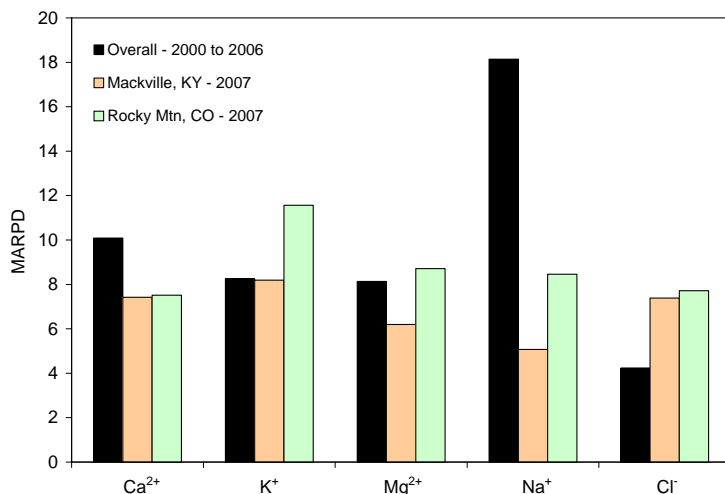
$$\text{RPD} = \left( \frac{S1 - S2}{S1 + S2} \right) \times 200 \quad \text{Equation 5-3}$$

For more information on the analytical methods and associated precision and accuracy criteria used for 2007, see the CASTNET QAPP - Revision 4.0 (MACTEC, 2007d).

Historical and 2007 precision statistics for four cations and chloride (Cl<sup>-</sup>) are summarized in Figure 5-2. The historical MARPD statistics for both MCK131/231 and ROM406/206 did not meet the DQI criterion of 5 percent. As discussed in earlier CASTNET reports (e.g., MACTEC, 2004 and MACTEC, 2003), the very high historical MARPD for sodium (Na<sup>+</sup>) was the result of sample bottle contamination. These bottles are no longer purchased. Also, acceptance testing of the Teflon<sup>®</sup> filters was instituted for the trace cations and Cl<sup>-</sup> in 2003. The 2007 precision results show that Na<sup>+</sup> met the precision criterion at MCK131/231. The 2007 results are somewhat worse than the precision data for 2006.

Table 5-2 summarizes 2007 precision results by quarter for the two sets of collocated sites. See the 2007 Quarterly Data Reports (MACTEC, 2007b; 2007e; 2008c; 2008a) and QA Quarterly Reports (MACTEC, 2007c; 2007f; 2007g; 2008b) for discussions of quarterly precision data.

**Figure 5-2** Historical and 2007 Precision Data for Cation and Cl<sup>-</sup> Concentrations



**Table 5-2** Collocated Precision Results (MARPD) for 2007 Filter Pack Data by Quarter

Site Pairs	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	HNO <sub>3</sub>	SO <sub>2</sub>	Total NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>
<b>MCK 131/231, KY</b>											
Quarter 1	3.76	7.24	6.19	12.72	9.04	8.77	6.57	5.43	3.31	5.78	20.70
Quarter 2	1.67	8.71	1.94	6.15	6.85	3.34	8.37	3.59	1.39	4.17	2.13
Quarter 3	3.40	11.25	3.02	5.95	5.31	5.15	6.94	2.39	1.42	2.91	1.06
Quarter 4	1.27	3.99	2.10	4.86	3.57	3.01	10.87	5.16	3.27	3.62	5.64
2007	2.53	7.80	3.31	7.42	6.19	5.07	8.19	4.14	2.34	4.12	7.38
<b>ROM 406/206, CO</b>											
Quarter 1	7.96	9.15	4.64	6.38	5.82	4.42	12.94	3.44	10.70	4.22	16.64
Quarter 2	3.96	6.60	3.82	4.07	7.17	6.01	7.94	2.29	3.65	2.16	2.95
Quarter 3	9.41	11.85	8.48	10.04	13.08	10.25	14.84	7.22	10.53	7.40	5.55
Quarter 4	8.24	18.02	7.55	9.55	8.76	13.15	10.54	4.44	6.48	6.84	5.71
2007	7.39	11.41	6.12	7.51	8.71	8.46	11.56	4.35	7.84	5.16	7.71

The 2007 analytical precision results for five analytes and the three filter types are presented in Figure 5-3. The results were based on analysis of 5 percent of the samples that were randomly selected for replication in each batch. The results of in-run replicate analyses were compared to the results of the original concentrations. The laboratory precision data met the 5 percent measurement criterion listed in Table 5-1.

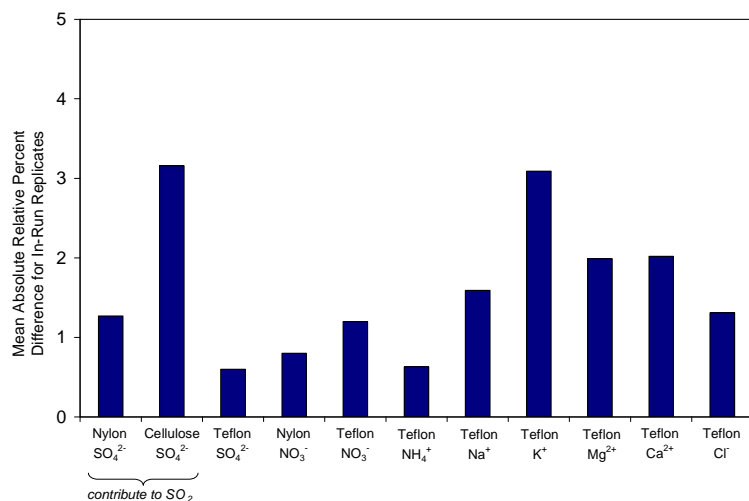


## Ozone Concentrations

CASTNET QA procedures for the EPA-sponsored ozone ( $O_3$ ) analyzers are different from the EPA QA requirements for State and Local Monitoring Stations (SLAMS) monitoring (EPA, 1998). The QA procedures for the  $O_3$  analyzers at the NPS-sponsored CASTNET sites also do not meet the SLAMS requirements. In any event, the operation of the collocated EPA and NPS  $O_3$  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  $O_3$ . The precision criterion for the collocated  $O_3$  data is 10 percent.

MARPD statistics were calculated from hourly  $O_3$  measurements obtained from the collocated sites MCK131/231, KY and ROM406/206, CO during 2007. 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  $O_3$  concentrations by quarter for 2007. The data show the historical results met the 10 percent criterion. The historical data are based on the operation of 11 collocated site pairs over the history of the network. The 2007 precision data also met the 10 percent criterion for the first and second quarters for both MCK131/231 and ROM406/206. The results for MCK131/231 did not meet the criterion during the fourth quarter. The poor results for this quarter were the result of sampling problems and periods of very low concentrations. The two analyzers at ROM406/206 did not meet the precision criterion during the third and fourth quarters. The hourly  $O_3$  data from ROM406 were consistently higher than the data from ROM206 during the third quarter. The reason for this systematic difference has not been determined despite acceptable automatic calibration results and an acceptable external audit. The imprecision of the  $O_3$  data during the fourth quarter was caused by sampling train problems.

**Figure 5-3** Precision Results for Laboratory Replicate Samples (2007)

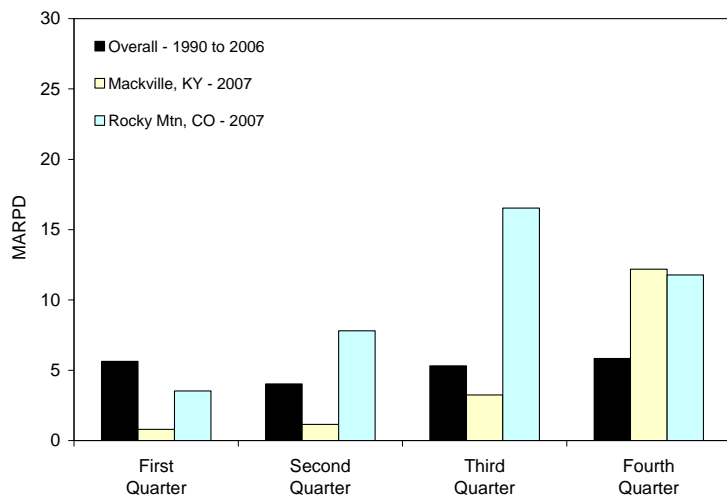


**Table 5-3** Data Quality Indicator Criteria for CASTNET Field Measurements

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

**Note:** °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 2007 Precision Data by Quarter for Ozone Concentrations



**Table 5-4** Collocated Precision Results (MARPD) for 2007 Ozone Concentrations by Quarter

Sampling Period	MCK131/231, KY	ROM406/206, CO
Quarter 1	0.80	3.52
Quarter 2	1.16	7.81
Quarter 3	3.25	16.84
Quarter 4	12.18	11.78
2007	4.35	9.91

## *Comparison of CASTNET and AQS Methods For Determining Precision*

The precision results shown in Table 5-2 for the collocated sampling systems at MCK131/231, KY and ROM406/206, CO were recalculated for the four quarters of 2007 using the methods suggested in the “Air Quality System Precision and Accuracy Summarization Formulas” (EPA, 2004). The results of the recalculations for ozone and six filter pack measurements are shown in Table 5-B.

**Table 5-B** Precision Results for Ozone and Filter Pack Analytes Using AQS Methods

	O <sub>3</sub>		SO <sub>4</sub> <sup>2-</sup>		NO <sub>3</sub>		NH <sub>4</sub> <sup>+</sup>		HNO <sub>3</sub>		SO <sub>2</sub>		Total NO <sub>3</sub>	
	MRPD	Std. Dev.	MRPD	Std. Dev.	MRPD	Std. Dev.	MRPD	Std. Dev.	MRPD	Std. Dev.	MRPD	Std. Dev.	MRPD	Std. Dev.
<b>MCK131/231, KY</b>														
Quarter 1	-0.56	3.22	-1.67	7.15	-4.59	8.79	-0.86	9.00	0.37	8.78	-0.60	4.35	-2.25	7.39
Quarter 2	0.08	1.76	-1.54	1.31	-8.64	5.27	-1.56	1.82	-2.60	4.12	-0.77	1.38	-3.96	3.28
Quarter 3	-0.80	3.97	-3.31	8.32	-9.15	12.37	-3.00	8.15	-1.12	2.70	0.96	1.63	-2.02	3.37
Quarter 4	12.11	24.34	-0.05	1.89	-2.15	4.59	0.34	2.82	0.83	5.60	0.20	4.22	-0.34	4.23
<b>Average</b>	<b>2.71</b>		<b>-1.64</b>		<b>-6.13</b>		<b>-1.27</b>		<b>-0.63</b>		<b>-0.05</b>		<b>-2.14</b>	
<b>ROM406/206, CO</b>														
Quarter 1	2.92	4.23	-4.22	18.89	1.01	13.42	1.74	6.68	-0.44	5.34	-1.29	14.36	0.19	5.81
Quarter 2	7.42	7.84	3.67	2.60	-0.38	9.17	3.46	3.21	2.08	1.84	2.94	4.83	1.56	2.28
Quarter 3	16.23	9.44	1.42	16.07	3.50	17.79	0.24	16.32	-2.80	15.28	-2.23	17.83	-1.77	15.50
Quarter 4	11.78	9.66	4.80	9.34	14.52	24.28	5.15	7.43	4.00	5.90	1.81	9.26	6.66	6.49
<b>Average</b>	<b>9.59</b>		<b>1.42</b>		<b>4.66</b>		<b>2.64</b>		<b>0.71</b>		<b>0.31</b>		<b>1.66</b>	

The principal difference in method is that AQS precision is based on the mean of relative percent difference values, i.e., MRPD, while CASTNET utilizes MARPD. AQS also requires the calculation of the standard deviation of the relative percent difference (RPD) values. MRPD is the mean of the RPD values shown in Equation 5-3.

The precision criterion for O<sub>3</sub> concentrations is 10 percent. The AQS O<sub>3</sub> precision data are consistent with the CASTNET MARPD data. Results for fourth quarter for MCK131/231 and for third and fourth quarter for ROM406/206 exceeded the criterion. As expected, the calculated AQS precision values for the six sets of filter pack concentrations are lower than the corresponding MARPD values. The MARPD criterion is more stringent than MRPD. The filter pack precision criterion is 10 percent for NH<sub>4</sub><sup>+</sup> and 5 percent for the other pollutants. The AQS precision data indicate the precision criteria would have been met for all parameters except particulate nitrate for two quarters at MCK131/231 and one quarter at ROM406/206 and total nitrate at ROM406/206 for the fourth quarter.

## Accuracy

### Laboratory Filter Concentrations

Accuracy of laboratory measurements is assessed through the analysis of reference samples and continuing calibration verification (CCV) spikes. Reference samples and CCV are procured from independent suppliers and are National Institute of Standards and Technology (NIST) traceable. Reference samples are analyzed at the beginning and end of each analytical batch to verify the accuracy and stability of the calibration curve. The target value of the CCV solution is set to the midrange of the calibration curve. In 2007, the CCV were analyzed every tenth sample to verify that instrument calibration had not drifted beyond established limits. Table 5-5 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 2007. Table 5-5 corroborates the precision results that were shown in Figure 5-3.

**Table 5-5** Filter Pack Quality Control Summary for 2007

Filter Type	Parameter	Reference Sample <sup>1</sup> Recovery (%R)			Continuing Calibration Verification Samples (%R)			In-Run Replicate <sup>2</sup> (RPD)		
		Mean	Std. Dev.	Count <sup>3</sup>	Mean	Std. Dev.	Count <sup>3</sup>	Mean	Std. Dev.	Count <sup>3</sup>
Teflon <sup>®</sup>	SO <sub>4</sub> <sup>2-</sup>	99.79	1.64	154	100.11	1.07	809	0.60	1.65	332
	NO <sub>3</sub> <sup>-</sup>	101.71	1.40	154	99.68	1.14	809	1.20	1.98	332
	NH <sub>4</sub> <sup>+</sup>	100.08	2.41	118	99.82	2.09	649	0.63	1.04	269
	Ca <sup>2+</sup>	100.42	2.91	140	100.75	1.28	805	2.02	3.33	334
	Mg <sup>2+</sup>	100.70	1.16	140	99.99	0.89	805	1.99	2.11	334
	Na <sup>+</sup>	97.01	2.11	140	100.12	1.15	805	1.59	2.02	334
	K <sup>+</sup>	98.04	2.91	140	100.21	1.09	805	3.09	3.79	334
	Cl <sup>-</sup>	100.82	1.50	154	99.83	2.19	809	1.31	1.91	332
Nylon	SO <sub>4</sub> <sup>2-</sup>	100.61	1.32	151	98.90	2.11	798	1.27	1.65	331
	NO <sub>3</sub> <sup>-</sup>	99.47	1.73	151	98.76	2.50	798	0.80	1.45	331
Cellulose	SO <sub>4</sub> <sup>2-</sup>	101.64	1.51	187	99.28	1.71	809	3.16	5.33	274

Note: % R = percent recovery (see Equation 5-2)

RPD = relative percent difference (see Equation 5-3)

<sup>1</sup> Results of reference sample analyses provide accuracy estimates

<sup>2</sup> Results of replicate analyses provide precision estimates

<sup>3</sup> Number of QC Samples



## Continuous Measurements

Table 5-6 presents field accuracy results for 2007 based on instrument challenges performed using independent reference standards during site calibration visits. CASTNET sites were calibrated every six months with NIST-traceable standards. The calibration results were evaluated using the accuracy criteria listed in Table 5-3. Each parameter was within its criterion with at least 90 percent frequency with the exception of high (> 85 percent) relative humidity at 88.2 percent, solar radiation at 89.1 percent, and wind direction north at 88.0 percent. However, these results did not adversely affect data collection because data are not considered invalid unless criteria are exceeded by more than two times the criterion. Using the two times standard, the three parameters passed with 95.0, 96.4, and 97.4 percent frequency, respectively.

**Table 5-6** Accuracy Results for 2007 Field Measurements

Parameter	Percent Within Criterion
Temperature (0°C)	98.3 percent
Temperature (ambient)	97.4 percent
Delta Temperature (0°C)	99.1 percent
Delta Temperature (ambient)	98.0 percent
*Relative Humidity > 85%	88.2 percent
Relative Humidity ≤ 50%	98.3 percent
*Solar Radiation	89.1 percent
*Wind Direction North	88.0 percent
Wind Direction South	95.7 percent
Wind Speed < 5 m/s	100.0 percent
Wind Speed ≥ 5 m/s	94.8 percent
Precipitation	100.0 percent
Wetness (w/in 0.5 volts)	100.0 percent
Ozone Slope	98.1 percent
Ozone Intercept	99.1 percent
Flow Rate	95.8 percent

**Note:** °C = degrees Celsius

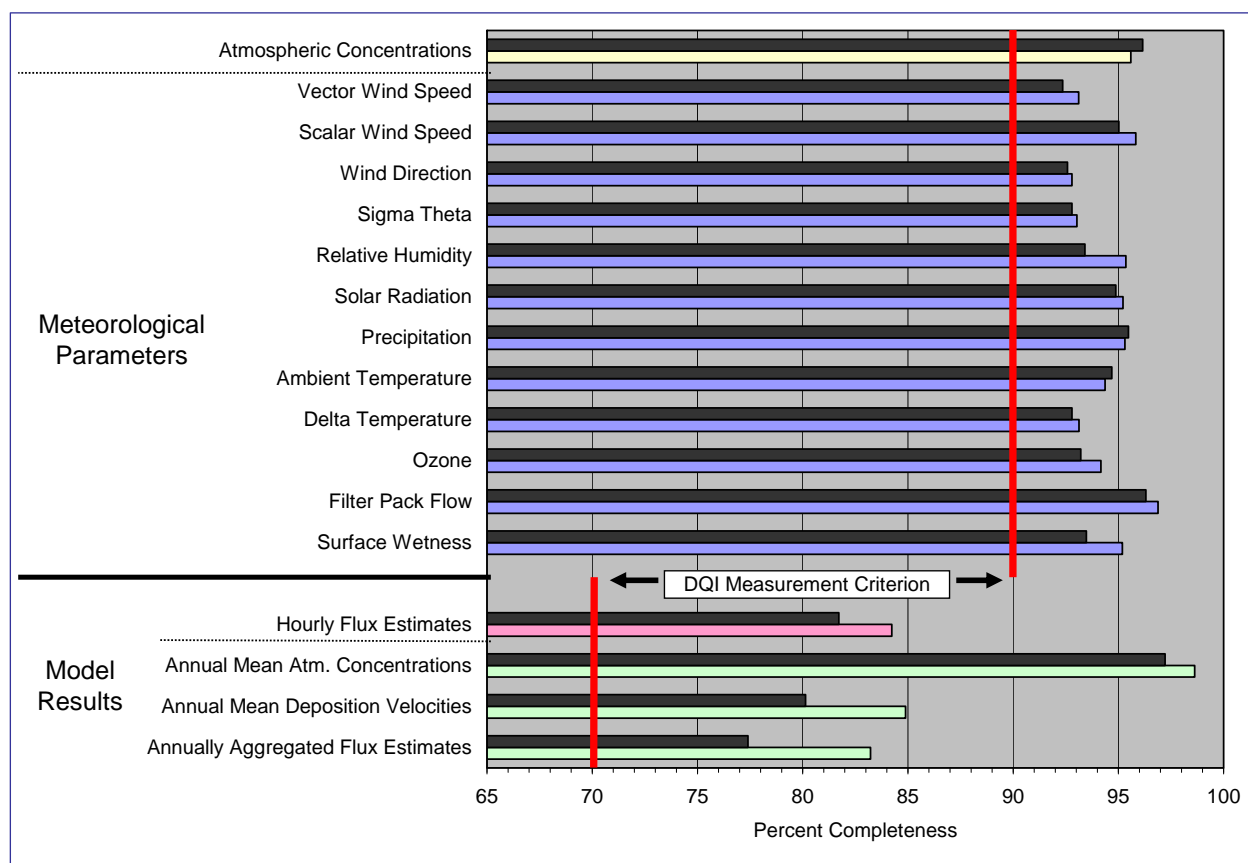
m/s = meters per second

\* Per CASTNET project protocols, data are flagged as “suspect” (S) but still considered valid if the calibration criterion is not exceeded by more than its magnitude (i.e., if within 2x the criterion). The percent within 2x criterion for these parameters ranged from 95.0 percent to 97.4 percent.

## Completeness

Completeness is defined as the percentage of valid data points obtained from a measurement system relative to total possible data points. The CASTNET measurement criterion for completeness requires a minimum completeness of 90 percent for every measurement for each quarter. In addition, data aggregation procedures require approximately 70 percent completeness for hourly fluxes and weekly concentrations/fluxes. Figure 5-5 presents historical (black) and 2007 completeness data for all sites for measured filter concentrations and continuous measurements and calculated parameters. The figure shows that the 2007 direct measurements met the 90 percent completeness goal and exceeded 95 percent for six continuous measurements, including filter pack flow, plus annual mean concentrations. The four parameters derived from model results exceeded 83 percent completeness for 2007. Completeness results for 2007 are better than historical results for all parameters except atmospheric concentrations, temperature, and precipitation.

**Figure 5-5** Historical and 2007 Percent Completeness of Measurements and Modeled Estimates (black bars are 1990-2006)



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## Conclusion

DQI results demonstrate that field and laboratory processes were adequately monitored through QA/QC procedures and were generally free of systemic bias during 2007. Accuracy data met the established criteria for field and laboratory parameters with the exception of relative humidity > 85 percent, solar radiation, and wind direction north. However, since the criterion was exceeded by a value less than its own magnitude, the associated continuous data collected are considered valid.

Precision data for  $\text{SO}_2$ ,  $\text{SO}_4^{2-}$ ,  $\text{HNO}_3$ , and ammonium are considered acceptable. Precision data for nitrate analyses of collocated field samples have not met the established criterion due, most likely, to the low concentrations generally measured and the uncertain nature of the gas-particle equilibrium of the nitrate species.

The 2007 precision data for ozone concentrations also met the 10 percent criterion for the first and second quarters for both MCK131/231 and ROM406/206. The results for MCK131/231 did not meet the criterion during the fourth quarter, and ROM406/206 data did not meet the precision criterion during the third and fourth quarters. The imprecise results for these three quarters were the result of various sampling problems and periods of very low concentrations. Completeness criteria were met for all parameters in 2007. Completeness results for 2007 were generally better than historical results.



Horton Station, VA (VPI120)

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## References

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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 <sup>1</sup>	Ozone and Meteorology <sup>2</sup>	Wet Deposition	Primary Land Use	Terrain	Representative to the MLM <sup>3</sup>	Sponsor
<b>Alabama</b>												
SND152	Sand Mountain	12/27/88	34.2894	85.9704	352	•	•		Agri.	Rolling	Y	EPA
<b>Alaska</b>												
DEN417	Denali National Park	10/06/98	63.7258	148.9633	661	•	•		Forested	Complex	N	NPS
<b>Arizona</b>												
CHA467	Chiricahua National Monument	04/25/89	32.0092	109.3892	1570	•	•	◇	Range	Complex	N	NPS
GRC474	Grand Canyon National Park	05/16/89	36.0597	112.1822	2073	•	•		Forested	Complex	M	NPS
PET427	Petrified Forest National Park	09/24/02	34.8225	109.8919	1723	•	•		Desert	Flat	Y	NPS
<b>Arkansas</b>												
CAD150	Caddo Valley	10/04/88	34.1792	93.0989	71	•	•		Forested	Rolling	N	EPA
<b>California</b>												
CON186	Converse Station	06/17/03	34.1941	116.9130	1837	•	•		Agri./Forested	Complex	N	EPA
DEV412	Death Valley National Monument	02/21/95	36.5092	116.8481	125	•	•		Desert	Complex	Y	NPS
JOT403	Joshua Tree National Monument	02/16/95	34.0714	116.3906	1244	•	•		Desert	Complex	M	NPS
LAV410	Lassen Volcanic National Park	07/25/95	40.5403	121.5764	1756	•	•		Forested	Complex	M	NPS
PIN414	Pinnacles National Monument	05/16/95	36.4850	121.1556	335	•	•		Forested	Complex	M	NPS
SEK430	Sequoia National Park	04/07/05	36.4894	118.8269	457	•	•		Forested	Mountaintop	N	NPS
YOS404	Yosemite National Park	09/25/95	37.7133	119.7061	1605	•	•		Forested	Complex	N	NPS

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

**Table A-1 Locational and Operational Characteristics of CASTNET Sites**

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<b>Indiana</b>												
SAL133	Salamonie Reservoir	06/28/88	40.8164	85.6608	250	•	•		Agri.	Flat	Y	EPA
VIN140	Vincennes	08/04/87	38.7406	87.4844	134	•	•		Agri.	Rolling	M	EPA
<b>Kansas</b>												
KNZ184	Konza Prairie	03/26/02	39.1021	96.6096	348	•	•		Prairie	Flat	Y	EPA
<b>Kentucky</b>												
CDZ171	Cadiz	10/01/93	36.7841	87.8500	189	•	•		Agri.	Rolling	M	EPA
CKT136	Crockett	08/24/93	37.9211	83.0658	455	•	•		Agri.	Rolling	Y	EPA
MAC426	Mammoth Cave National Park	07/24/02	37.1319	86.1478	243	•	•		Agri./Forested	Rolling	M	NPS
MCK131	Mackville	07/31/90	37.7044	85.0483	353	•	•		Agri.	Rolling	M	EPA
<b>Maine</b>												
ACA416	Acadia National Park	12/01/98	44.3769	68.2608	158	•	•		Forested	Complex	M	NPS
ASH135	Ashland	12/20/88	46.6039	68.4142	235	•	•		Agri.	Flat	Y	EPA
HOW132	Howland	11/24/92	45.2158	68.7085	69	•	•		Forested	Rolling	Y	EPA
<b>Maryland</b>												
BEL116	Beltsville	11/01/88	39.0283	76.8175	46	•	•		Urban-Agri.	Flat	N	EPA
BWR139	Blackwater National Wildlife Refuge	07/04/95	38.4448	76.1115	4	•	•		Forest-Marsh	Coastal	M	EPA



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

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

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

**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 <sup>1</sup>	Ozone and Meteorology <sup>2</sup>	Wet Deposition	Primary Land Use	Terrain	Representative to the MLM <sup>3</sup>	Sponsor
<b>Virginia</b>												
PED108	Prince Edward	11/03/87	37.1653	78.3070	150	•	•	◇	Forested	Rolling	M	EPA
SHN418	Shenandoah National Park	06/28/88	38.5231	78.4347	1073	•	•		Forested	Mountaintop	M	NPS
VPI120	Horton Station	06/02/87	37.3300	80.5573	920	•	•		Forested	Mountaintop	N	EPA
<b>Washington</b>												
MOR409	Mount Rainier National Park	08/29/95	46.7583	122.1244	415	•	•		Forested	Complex	N	NPS
NCS415	North Cascades National Park	02/14/96	48.5397	121.4472	109	•	•		Forested	Complex	M	NPS
<b>West Virginia</b>												
CDR119	Cedar Creek State Park	11/10/87	38.8794	80.8478	234	•	•	◇	Forested	Complex	N	EPA
PAR107	Parsons	01/19/88	39.0906	79.6614	510	•	•		Forested	Complex	N	EPA
<b>Wisconsin</b>												
PRK134	Perkinstown	09/27/88	45.2066	90.5972	472	•	•	◇	Agri.	Rolling	M	EPA
<b>Wyoming</b>												
CNT169	Centennial	08/19/91	41.3722	106.2422	3178	•	•		Range	Complex	M	EPA
PND165	Pinedale	12/27/88	42.9214	109.7900	2388	•	•		Range	Rolling	M	EPA
YEL408	Yellowstone National Park	06/26/96	44.5597	110.4006	2400	•	•		Forested	Rolling	N	NPS



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

Teflon<sup>®</sup> = SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, Cl<sup>-</sup>, K<sup>+</sup>, Na<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>

Nylon = SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> (reported as HNO<sub>3</sub>)

Cellulose = SO<sub>4</sub><sup>2-</sup> (reported as SO<sub>2</sub>)

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. O<sub>3</sub> not measured.

5. Solar-powered site.

- Indicates current monitoring.

- ◇ Indicates discontinued monitoring.

Measurements were discontinued at the various sites because of several reasons including:

- (1) rotate limited number of instruments;
- (2) redundant measurements (e.g., with IMPROVE and NADP/NTN); and
- (3) funding limitations.

100 and 200 series = EPA – Operated Sites

400 series = NPS – Operated Sites

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## **Appendix B**

### **Acronyms and Abbreviations**

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## List of Acronyms and Abbreviations

AQI	Air Quality Index
AQS	EPA's Air Quality System
ARP	Acid Rain Program
Ca <sup>2+</sup>	particulate calcium ion
CAAA	Clean Air Act Amendments
CAMx	Comprehensive Air Quality Model with Extensions
Ca(NO <sub>3</sub> ) <sub>2</sub>	calcium nitrate
CAPMoN	Canadian Air and Precipitation Monitoring Network
CASTNET	Clean Air Status and Trends Network
CCV	continuing calibration verification spikes
Cl <sup>-</sup>	particulate chloride ion
CNEP	Cherokee Nation Environmental Programs
DQI	data quality indicator
DQO	data quality objectives
EGU	electric generating unit
EPA	U.S. Environmental Protection Agency
EST	Eastern Standard Time
H <sub>2</sub> SO <sub>4</sub>	sulfuric acid
HNO <sub>3</sub>	nitric acid
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
IC	ion chromatography
ICP-AES	inductively coupled plasma-atomic emission spectrometry
IMPROVE	Interagency Monitoring of Protected Visual Environments
ITEC	Inter-Tribal Environmental Council
K <sup>+</sup>	particulate potassium ion
K <sub>2</sub> CO <sub>3</sub>	potassium carbonate
kg/ha/yr	kilograms per hectare per year
km	kilometer
LAI	leaf area index
lpm	liters per minute
m	meters
MACTEC	MACTEC Engineering and Consulting, Inc.
MARPD	mean absolute relative percent difference
MDN	Mercury Deposition Network
Mg <sup>2+</sup>	particulate magnesium ion
mg/l	milligrams per liter
MLM	Multi-Layer Model
MRPD	mean of relative percent difference

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## List of Acronyms and Abbreviations (continued)

N	nitrogen
Na <sup>+</sup>	particulate sodium ion
NAAQS	National Ambient Air Quality Standards
NADP	National Atmospheric Deposition Program
NaNO <sub>3</sub>	sodium nitrate
NAPAP	National Acid Precipitation Assessment Program
NBP	NO <sub>x</sub> National Budget Trading Program
NDDN	National Dry Deposition Network
NH <sub>3</sub>	ammonia
NH <sub>4</sub> <sup>+</sup>	particulate ammonium
NH <sub>4</sub> NO <sub>3</sub>	ammonium nitrate
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	ammonium sulfate
NIST	National Institute of Standards and Technology
NO <sub>3</sub> <sup>-</sup>	particulate nitrate
NO <sub>y</sub>	total reactive oxides of nitrogen
NO <sub>x</sub>	nitrogen oxides [nitric oxide (NO) + nitrogen dioxide (NO <sub>2</sub> )]
NOAA	National Oceanic and Atmospheric Administration
NPS	National Park Service
NTN	National Trends Network
NWRI	Environment Canada's National Water Research Institute
O <sub>3</sub>	ozone
ON	organic nitrate
ORD	EPA's Office of Research and Development
OTC	Ozone Transport Commission
PART	Program Assessment Rating Tool
PM <sub>2.5</sub>	fine particle matter
ppb	parts per billion
ppm	parts per million
QA	quality assurance
QAPP	Quality Assurance Project Plan
QC	quality control
RMNP	Rocky Mountain National Park
RoMANS	Rocky Mountain Atmospheric Nitrogen and Sulfur Study
RPD	relative percent difference
S	sulfur
SIP	State Implementation Plan
SLAMS	State and Local Monitoring Stations
SO <sub>2</sub>	sulfur dioxide
SO <sub>4</sub> <sup>2-</sup>	particulate sulfate



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## List of Acronyms and Abbreviations (continued)

total NO <sub>3</sub> <sup>-</sup>	gaseous nitric acid (HNO <sub>3</sub> ) + particulate nitrate (NO <sub>3</sub> <sup>-</sup> )
μg	microgram
μg/m <sup>3</sup>	micrograms per cubic meter
USGS	U.S. Geological Survey
UTC	Coordinated Universal Time
V <sub>d</sub>	deposition velocity
VOCs	volatile organic compounds

For More Information

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

**On the Web:**

CASTNET Home Page:

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