

# CHAPTER 4: TEMPORAL TRENDS IN FINE AEROSOL SPECIES CONCENTRATIONS AND AEROSOL EXTINCTION

## INTRODUCTION

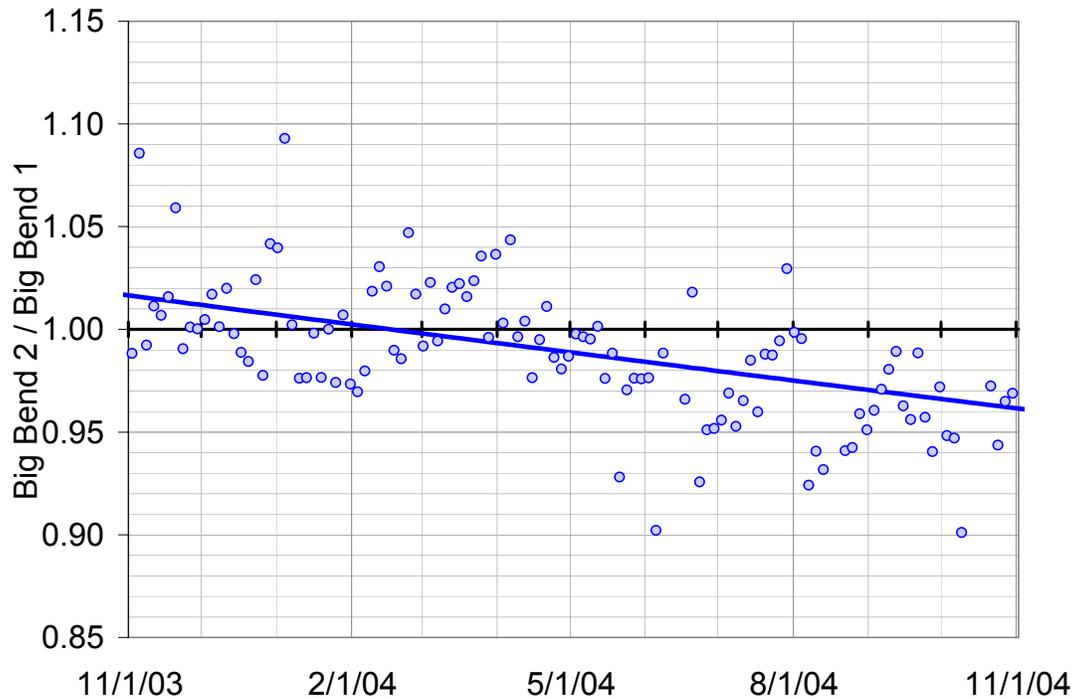
The results of several studies investigating monotonic trends in fine aerosol species concentrations will be summarized here, with the originals available in Appendix D. Topics explored since the last IMPROVE report in 2000 include the 10-year spatial and temporal trends in sulfate concentrations and SO<sub>2</sub> emissions [Malm et al., 2002], the uncertainty in sulfate concentration trends [White et al., 2005], 10-year trends in visibility [NPS, 2006], >7-year trends in organic and elemental carbon [Schichtel et al., 2004], and the Visibility Information Exchange Web System (VIEWS) annual summary trends tools. The IMPROVE program often uses Theil regression statistics for calculating trends; the original reference is not widely available so there is a discussion of Theil regression and associated statistics available from the IMPROVE website. A detailed discussion and algorithm for performing Theil regression in SAS, by Hess et al. [2002], is available at [http://vista.cira.colostate.edu/improve/Publications/GrayLit/026\\_TheilReg/TheilRegressionl.pdf](http://vista.cira.colostate.edu/improve/Publications/GrayLit/026_TheilReg/TheilRegressionl.pdf).

### 4.1 Estimating Measurement Uncertainty in an Ambient Sulfate Trend

The Regional Haze Rule seeks restoration of natural visibility conditions through steady improvements over the next six decades. Documenting change on this timescale entails particle measurements that will support accurate comparisons between different eras, even as monitoring methods evolve [White, 1997; Weatherhead et al., 1998]. Such absolute measurement stability is a requirement not encountered in the shorter, more intensive field campaigns typically mounted to support source apportionment and model validation studies.

It is sometimes asserted that measurement errors should have little impact on trend estimates. Random errors are as likely to cancel as to reinforce each other in successive observations, and the estimated imprecision of an average accordingly declines with the square root of the number of observations going into it. Fixed biases are even less of a concern; zero offsets cancel when observations from different periods are subtracted, and scaling factors cancel when ratios are taken.

The problem with such theoretical analyses is that actual measurement series generally include nonideal errors. Figure 4.1 shows an example of such nonideality, a year-long trend in observed differences between collocated measurements of SO<sub>4</sub><sup>-</sup> at Big Bend National Park. The errors that generate these differences are clearly neither independent in successive observations nor constant across all observations. The collocated precision of the measurements is an excellent 2.5%, but there is a difference of about 5%/year between the time trends of the two measurement series.



**Figure 4.1. Ratio of 24-hour sulfate concentrations measured by collocated and routine IMPROVE B modules at Big Bend NP.**

A case study of the trend uncertainty produced by actual patterns of measurement error was undertaken by White et al. [2005] with data from Shenandoah National Park. IMPROVE and the Clean Air Status and Trends Network (CASTNet) have conducted collocated particle monitoring at this site since 1988. The analysis was based on the three independent series recorded for 24-hour sulfur (IMPROVE proton induced X-ray emission/X-ray fluorescence—PIXE/XRF), 24-hour sulfate (IMPROVE ion chromatography—IC), and 168-hour sulfate (CASTNet IC). These redundant determinations allowed the uncertainties of measurement to be isolated from those of meteorological variability and incomplete sampling.

Table 4.1 presents a selection of the results. Five-year sulfate trends can be estimated from both the sulfur and sulfate series, and these trends are expected to agree in the absence of measurement error. Equivalently, any trends observed in the series of ratios can be attributed to the effects of errors. It can be seen that spurious trends of as much as 3% per year were recorded over 5-year periods. The overall uncertainty of 5-year sulfate trends was estimated to be at least 1% per year from measurement error alone, significantly more than would be expected under naive statistical assumptions from the demonstrated precision of the measurements. The excess uncertainty arises from subtle trends in the errors themselves.

**Table 4.1. Five-year trends in measurement differences at Shenandoah NP.**

Period	SO <sub>4</sub> /3S	
	Decrease/yr	+/-
6/88 – 5/93	0.3%	0.6%
6/89 – 5/94	-0.5%	0.7%
6/90 – 5/95	0.1%	0.7%

Period	SO <sub>4</sub> /3S	
	Decrease/yr	+/-
6/91 – 5/96	2.3%	0.8%
6/92 – 5/97	2.8%	0.7%
6/93 – 5/98	2.6%	0.7%
6/94 – 5/99	2.0%	0.6%
6/95 – 5/00	0.8%	0.5%
6/96 – 5/01	-0.6%	0.6%
6/97 – 5/02	-1.5%	0.7%
6/98 – 5/03	-3.5%	0.8%
$rms/\sqrt{2}$	1.3%	0.5%

## 4.2 A 10-YEAR SPATIAL AND TEMPORAL TREND OF SULFATE ACROSS THE UNITED STATES

### 4.2.1 Introduction

Emissions of sulfur dioxide (SO<sub>2</sub>) into the atmosphere result in the formation of fine (<2.5 mm) particulate sulfate via a number of physiochemical mechanisms. Fine sulfates significantly impair visibility, deposit to susceptible aquatic and terrestrial ecosystems, and have the potential to be harmful to human health. A number of legislative and regulatory mandates have reduced SO<sub>2</sub> emissions both in the eastern and the western United States in an attempt to ameliorate these potentially deleterious effects. In response to the regulations and other socioeconomic pressures, SO<sub>2</sub> emissions have been declining over much of the United States [U.S. EPA, 2001].

This chapter builds upon previous work by examining the spatial and temporal trends of SO<sub>4</sub><sup>=</sup> over the United States. The CASTNet SO<sub>4</sub><sup>=</sup> data and the IMPROVE monitoring program data were combined to investigate changes in the spatial trends of the 90<sup>th</sup> percentile SO<sub>4</sub><sup>=</sup> concentrations across the United States between two time periods (1990–1994 and 1995–1999) and the yearly temporal trends of the 20<sup>th</sup> and 80<sup>th</sup> percentile. Also, the trends in SO<sub>2</sub> emissions and SO<sub>4</sub><sup>=</sup> concentrations were compared for regions in both the western and eastern United States. The spatial trends in the 90<sup>th</sup> percentile will not be discussed here but can be found in the full article in Appendix D.

The IMPROVE and CASTNet monitoring sites used in this analysis are presented in Figure 1 in Malm et al. [2002]. The CASTNet, or CASTNet Deposition Network (CDN), was established as a result of the 1990 Amendments to the Clean Air Act, with the goal to determine the effect of emissions reductions mandated by the act on air quality and deposition. Important differences between the samplers deployed by the CASTNet and IMPROVE networks are summarized in Table 4.2 and should be kept in mind when comparing their respective data. The trends in SO<sub>2</sub> emissions were examined using annual SO<sub>2</sub> emission rates from the EPA's National Emission Trends (NET) database [U.S. EPA, 2000]. The data were obtained by downloading 15 years (1985–1999) of county-level emission data from the EPA's AIRData website.

**Table 4.2. Important differences between samplers deployed in the CASTNet and IMPROVE networks.**

Protocol	CASTNet	IMPROVE
Sampler type	Filter pack	Modular
Sampling frequency	Weekly	Twice weekly (Wednesday and Saturday), 24-hour
Collection substrate	Teflon	Teflon (for elemental S), nylon (Nylasorb) for NO <sub>3</sub> <sup>-</sup> ions
Inlet size cut	Non-size-selective	PM <sub>2.5</sub> cyclone
Inlet denuder	None	Na <sub>2</sub> CO <sub>3</sub> (for nylon collection substrate, intended to remove HNO <sub>3</sub> )
Inlet height	10 m agl	~ 3 m above ground level (agl)

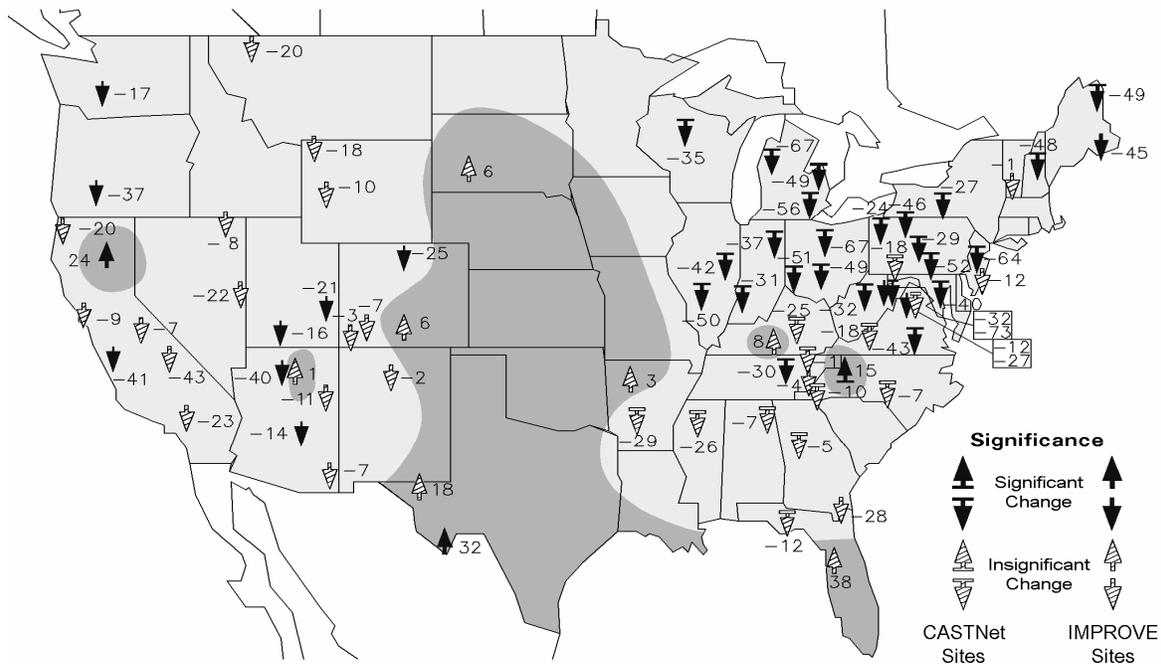
#### 4.2.2 Yearly Temporal Trends of the 20<sup>th</sup> and 80<sup>th</sup> Percentile SO<sub>4</sub><sup>=</sup> Concentrations

In this section, the temporal trends in the yearly 20th and 80th percentile sulfate mass concentrations are examined. Characterization of trends can be a highly subjective exercise in that slopes and their significance can vary depending on the technique employed. Using an ordinary least squares (OLS) regression approach is questionable with such small data sets, as the results can be highly influenced by outliers. In another approach developed by Theil [1950], outlier data points do not as significantly influence the results. Slopes of trend lines are calculated for each site by first finding the slope between all possible pairs of data points. The median value in the case of an odd number of pairs is selected as the estimated slope, or in the case of an even number of pairs, the average of the two slopes that straddle the median is used as the estimate. The significance of the Theil trend is found by calculating the probability that a random re-ordering of the same observations would yield a trend as consistent as the one observed. The slope estimates ( $\mu\text{g}/\text{m}^3/\text{yr}$ ) are the trend lines for the periods under consideration. For the IMPROVE data set, slope estimates are only shown for those sites that have at least 75 data points (two 24-hour samples per week  $\approx 72\%$  data recovery) for 7 years out of approximately 12 years of data. For the CASTNet data set, trends were calculated for sites with 40 data points (weekly samples  $\approx 77\%$  data recovery) for at least 7 out of 11 years of data.

Trends in yearly 80<sup>th</sup> percentile sulfate concentration at all, except two, monitoring sites west of the Rocky Mountains show some decrease in sulfate concentration (Figure 4.2). The exceptions are Lassen Volcanic National Park, California, where the sulfate concentration shows a statistically significant increase of about 24%, and Hopi Point in the Grand Canyon National Park, which had an insignificant increase of 1%. There are two sites, Indian Gardens in the Grand Canyon National Park, Arizona (4000 feet below rim), and Pinnacles National Monument, California, that exhibit about a 40% reduction that is statistically significant. Six additional sites also show statistically significant decreases.

In the East, the largest decreases occurred north of the Ohio River valley, while all monitoring sites south of Kentucky and Virginia showed increasing and decreasing trends that are not statistically significant. The maximum percent decrease occurred at Dolly Sods Wilderness Area, West Virginia, at a rate of 73% for the 11-year period, while many sites showed statistically significant decreases in excess of 50%.

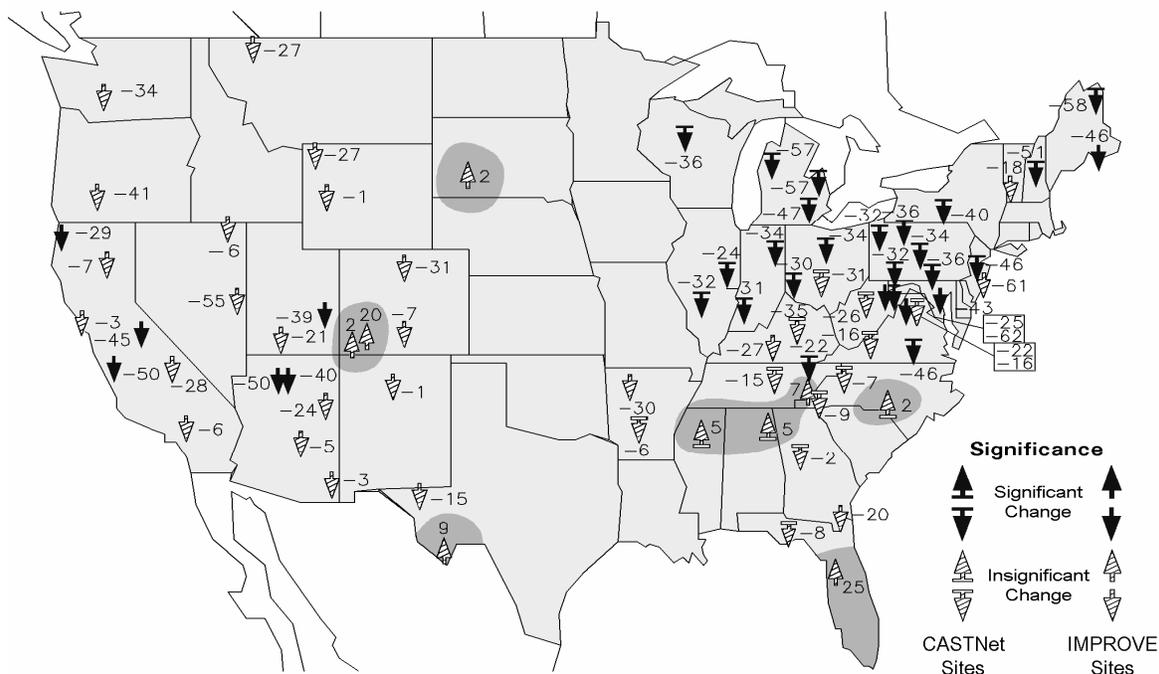
East of the Rocky Mountains, Big Bend, Texas, and Cranberry, North Carolina, are the only areas that show statistically significant increases in  $\text{SO}_4^{2-}$  mass concentrations of 32% and 15%, respectively, while Guadalupe Mountains, Great Sand Dunes, Badlands and Mammoth Cave National Parks, Upper Buffalo Wilderness Area, and Chassahowitzka National Wildlife Refuge showed increasing trends that were not statistically significant.



**Figure 4.2. Summary of the results of Theil regressions for the 80<sup>th</sup> percentile  $\text{SO}_4^{2-}$  ( $3^*S$  for IMPROVE program) concentrations from 1989 to 1999. Solid up or down arrows show which sites have trends with a significance level of at least 10%. Arrows with enclosed hatch lines show whether the trend was up or down but not statistically significant. Arrows with a bar across the tail represent CASTNet sites, while arrows without the bar show IMPROVE monitoring sites. The numbers are the percent changes from the overall median of the 80th percentile.**

The spatial patterns of the 20th percentile  $\text{SO}_4^{2-}$  concentrations are similar to the 80th percentile map shown in that most of the western United States shows a downward trend in 20th percentile sulfate concentrations (Figure 4.3). In the East, all but eight sites north of the northern border of Tennessee and North Carolina have statistically significant downward trends, while those sites south of this border show statistically insignificant increasing and decreasing trends.

It is of interest to point out that for the 80th percentile trends, the largest decreases were observed in the central eastern United States with somewhat smaller but still significant trends in the Northeast, while for the 20th percentile sulfate concentrations the spatial relationship of trends between the central and northeastern United States is reversed.



**Figure 4.3. Summary of the results of Theil regressions for the 20<sup>th</sup> percentile  $SO_4^{2-}$  ( $3^*S$  for IMPROVE program) concentrations from 1989 to 1999. Solid up or down arrows show which sites have a trend with a significance level of at least 10%. Arrows with enclosed hatch lines show whether the trend was up or down but not statistically significant. Arrows with a bar across the tail represent CASTNet sites, while arrows without the bar show IMPROVE monitoring sites. The numbers are the percent changes from the overall median of the 20th percentile.**

### 4.2.3 Yearly Temporal Trends of NET $SO_2$ Emissions

The trends in the NET  $SO_2$  emission fields were examined for each state by first aggregating the county-level emissions over each state for each year from 1990 to 1999. The slope of the 10-year trend for each state was estimated using the Theil regression. In the northern half of the eastern United States from Missouri to Maine, most states had statistically significant decreasing trends from 10% to 60% and ~30% over the entire region (Figure 4.4). States along the Ohio River valley, which have the highest  $SO_2$  emissions in the country, had statistically significant decreases as high as 35% in Ohio and 44% in Indiana. Over most of the southeastern states from Arkansas to Florida, the  $SO_2$  emissions have increased with significant increases in Arkansas (87%), Louisiana (48%), and North Carolina (37%). Tennessee and Georgia, with some of the highest  $SO_2$  emission rates in the region, had significant decreases of 35% and 42%, respectively.

In the central part of the United States extending from Texas to North Dakota, the  $SO_2$  emissions have also been increasing. Statistically significant increases were found in Texas (14%), New Mexico (10%), Colorado (11%), Wyoming (31%), North Dakota (35%), and Minnesota (9%). Decreases were found for Oklahoma (26%) and South Dakota (4%); however, these were not significant. Throughout most of the western United States, the  $SO_2$  emissions have decreased from 8% in Idaho to 37% in California. Statistically significant decreases of 30% were also found in Oregon and Montana.

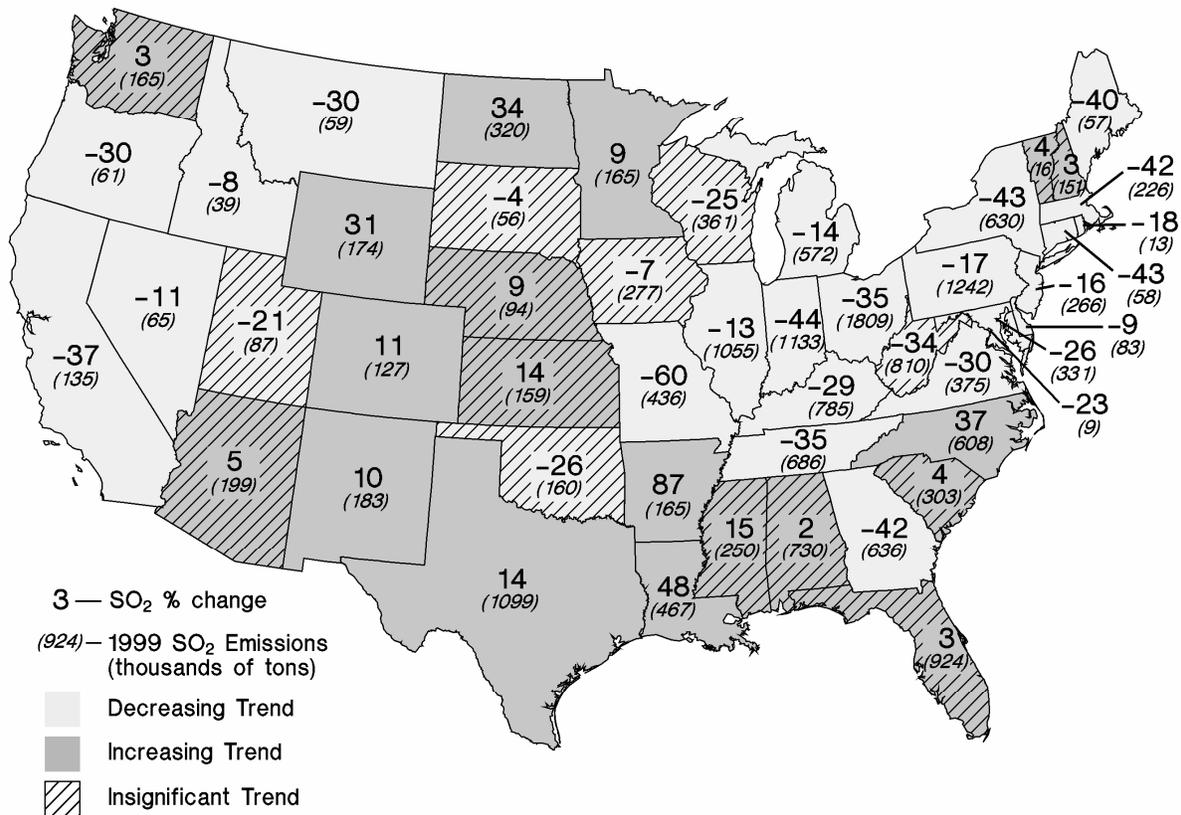


Figure 4.4. The percent change in the NET SO<sub>2</sub> emissions for each state in the conterminous United States from 1990 through 1999. The light gray states have decreasing trends, while the dark gray states have increasing trends. States without hatch marks have trends that are significant with two-sided P values below 0.1. The percent changes were calculated by dividing the change in emissions over the 10-year period by the 1990 emissions estimated from the trend line. The 1999 SO<sub>2</sub> emission rates for each state are in parentheses.

#### 4.2.4 Regional Comparisons of SO<sub>2</sub> Emissions and SO<sub>4</sub><sup>2-</sup> Concentrations

The 1990 to 1999 annual SO<sub>2</sub> emission and 80<sup>th</sup> percentile SO<sub>4</sub><sup>2-</sup> concentration time series were qualitatively compared over broad northeastern, southeastern, south-middle, and western United States regions. Each region had a unique time series pattern, with the SO<sub>4</sub><sup>2-</sup> concentrations and SO<sub>2</sub> emissions closely tracking each other over the 10-year time period (Figure 4.5). In the western United States from Arizona to Washington, both SO<sub>2</sub> emissions and concentrations steadily declined about 15% throughout the 1990s. In the south-middle United States, they steadily increased about 15%. In the northeastern United States, the SO<sub>2</sub> emissions and SO<sub>4</sub><sup>2-</sup> concentrations declined ~7% from 1990 to 1994, then decreased nearly 20% between 1994 and 1995, and leveled off afterward. In the southeastern United States, the SO<sub>4</sub><sup>2-</sup> concentrations and SO<sub>2</sub> emissions did not change appreciatively over the time period, but each had a decreasing trend in the early 1990s followed by an increasing trend since 1995–1996.

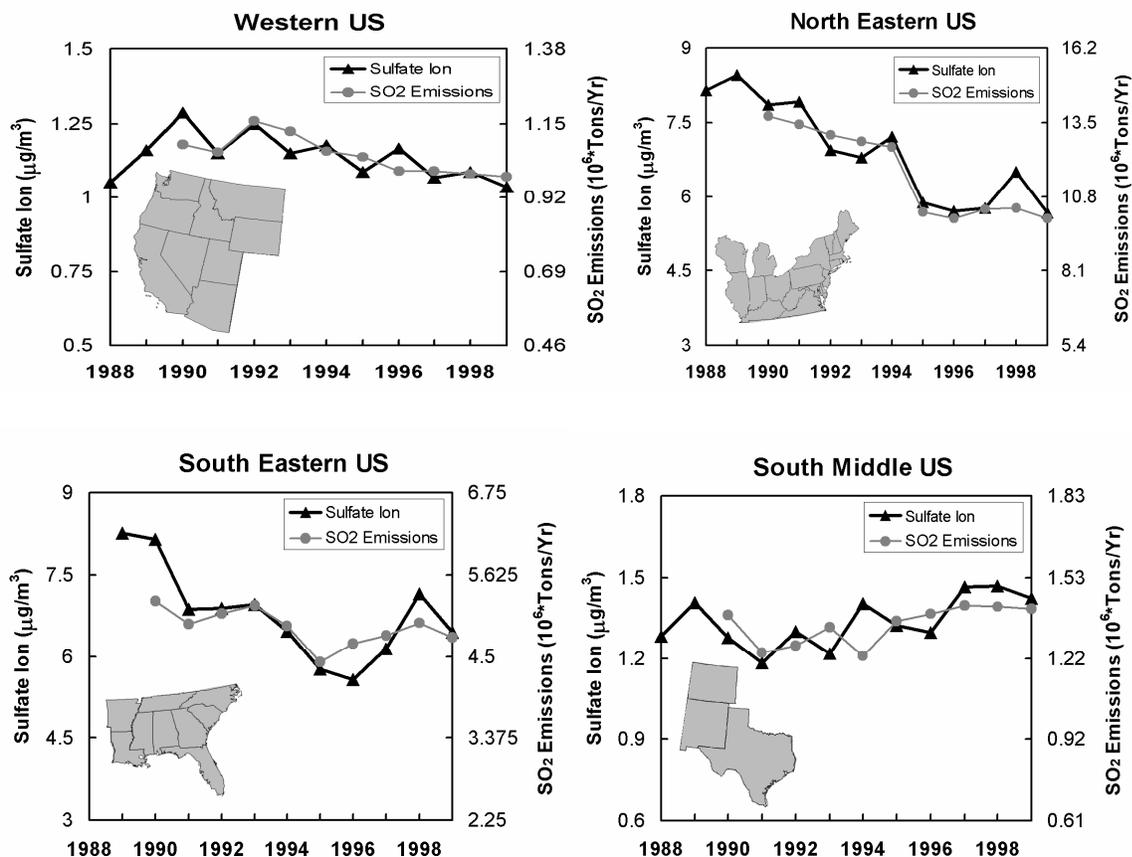


Figure 4.5. Comparison of the 80<sup>th</sup> percentile  $\text{SO}_4^{2-}$  concentrations (3\*S for IMPROVE program) and NET  $\text{SO}_2$  emissions aggregated over northeastern, southeastern, south-middle, and western United States regions. In each plot the  $\text{SO}_4^{2-}$  and  $\text{SO}_2$  emission scales have a factor of 3 change between the low and high values.

### 4.3 Trends in the Haze Index

Trends in the haze index were examined for the 10-year period 1995–2004 by the National Park Service [NPS, 2006]. The haze index is measured in deciviews, a visibility metric based on the light extinction coefficient that expresses incremental changes in perceived visibility (Chapter 2). The haze index values were calculated as prescribed under the Regional Haze Rule (RHR) guidance for tracking progress [U.S. EPA, 2003]. The RHR guidance outlines missing variable treatment and completeness criteria for constructing annual average deciview values for the 20% best and 20% worst visibility days. Trends in the annual average 20% best and worst days were examined using the Theil regression method (see section 4.2 for a description of the regression method) for the IMPROVE sites with at least 6 complete years out of the 10-year period. Visibility was stable (insignificant trends) or improving at all IMPROVE sites at the 0.05 significance level. Acadia, Moosehorn, Lye Brook, Dolly Sods, and Shenandoah showed statistically significant improving visibility trends for the clearest days at eastern national park monitoring sites (Figure 4.6). Great Smoky Mountains, Okefenokee, Mammoth Cave, and Washington, D.C., also had improving trends on the haziest visibility days (Figure 4.7). Statistically significant improving trends for the clearest visibility days were observed at 17 sites in the western United States including Alaska. Mount Rainer also had an improving trend

on the haziest visibility days. No site included in the analysis had a significant worsening trend on either the clearest or haziest visibility days.

### Trends in Haze Index (Deciview) on Clearest Days, 1995-2004



Figure 4.6. Their trends in the haze index of the annual average 20% best visibility days.

## Trends in Haze Index (Deciview) on Haziest Days, 1995-2004

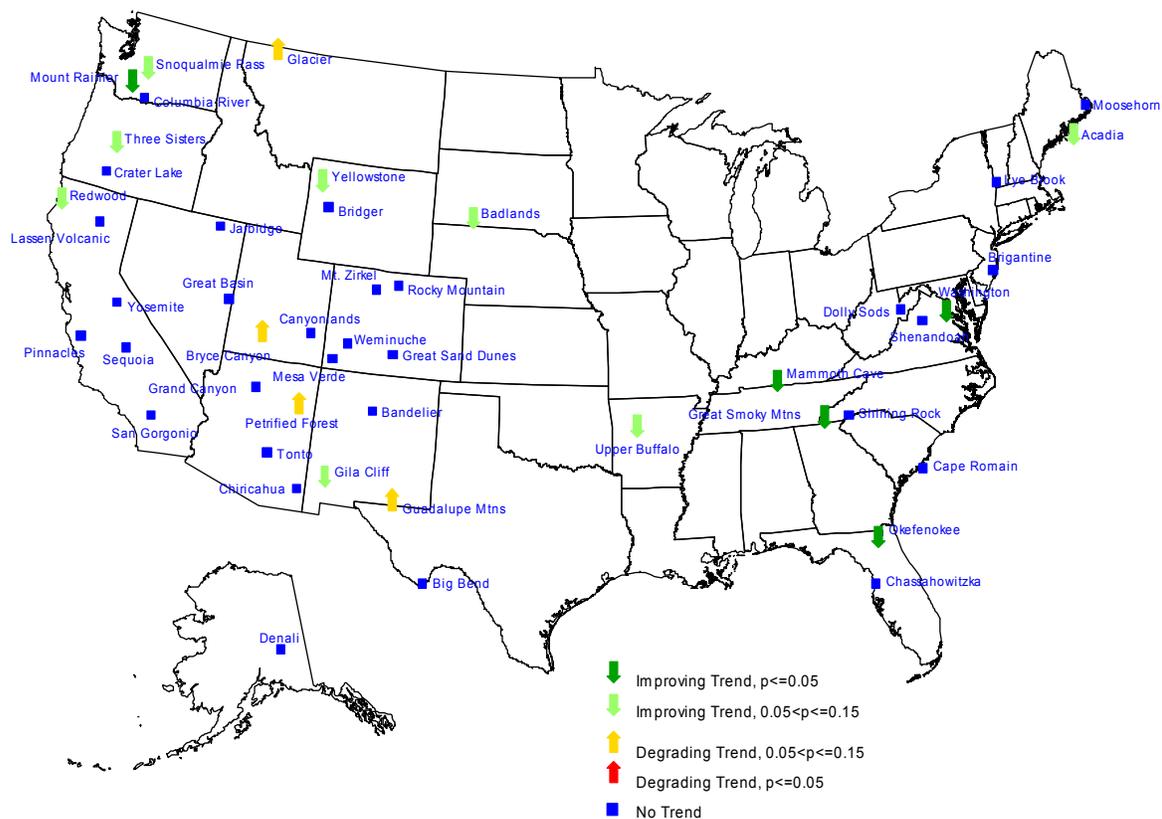


Figure 4.7. Their trends in the haze index of the annual average 20% worst visibility days.

### 4.4 Organic and Elemental Carbon Long-Term Trends and Spatial Patterns in the Rural United States

#### 4.4.1 Introduction

Carbonaceous aerosols are a complex mixture of organic carbon compounds and soot composed predominately of elemental carbon. These compounds are a major component of the fine aerosol mass [Malm et al., 2004], and their mass fraction is increasing as sulfate aerosols decrease [Malm et al., 2002]. Scattering and absorption by organic and elemental carbon are key factors in atmospheric transparency and the balance of solar radiation [White and Macias, 1989; Malm et al., 1994; Hegg et al., 1997; Hansen et al., 2000], which is important to the regional haze regulations and climate change. Carbonaceous emissions from wood burning and fossil fuel combustion may also have possible health effects [Lewis et al., 1988; U.S. EPA, 2000].

Carbonaceous aerosols arise from a wide array of sources, including diesel combustion and cooking in urban areas and smoke from fires and biogenic emissions from plants in rural areas. Planned changes in wildfire management are likely to increase the carbon contributions of smoke [Fox et al., 1999]. Combined with new diesel engine standards [Lloyd and Cackette, 2001] and other vehicle technology trends, the concentrations and chemical character of the

carbonaceous materials will change over time. In addition, other changes in the chemical environment can alter the rates of formation of secondary organic aerosols, further changing carbonaceous materials' concentrations and chemical character.

The long-term trends in carbonaceous material are a response to past changes in emissions and reactive environment. Understanding these responses and their causes will aid in future actions to reduce carbonaceous aerosol concentrations. Seasons are examined because source emissions and relevant atmospheric processes often vary with season, which could lead to seasonal variations in the trends of carbonaceous material.

#### 4.4.2 EC and OC Long-Term Trends

Theil regression was used to examine trends in winter and summer elemental and organic carbon at the 54 sites with 7 or more years of data. Winter elemental carbon (EC) concentrations decreased significantly at most monitoring sites in the Pacific coastal states and throughout the eastern United States, with median EC concentrations decreasing from 50% to 75% over a 10-year time period (Figure 4.8). Winter organic carbon (OC) concentrations (Figure 4.9) from Washington State to northern California showed similar significant decreases, but Acadia, Maine, was the only monitoring site in the eastern United States with a significant downward trend. Unlike EC, wintertime OC increased at a number of monitoring sites in the southeastern United States, though not significantly. Most sites in the intermountain West did not show significant winter trends in either EC or OC.

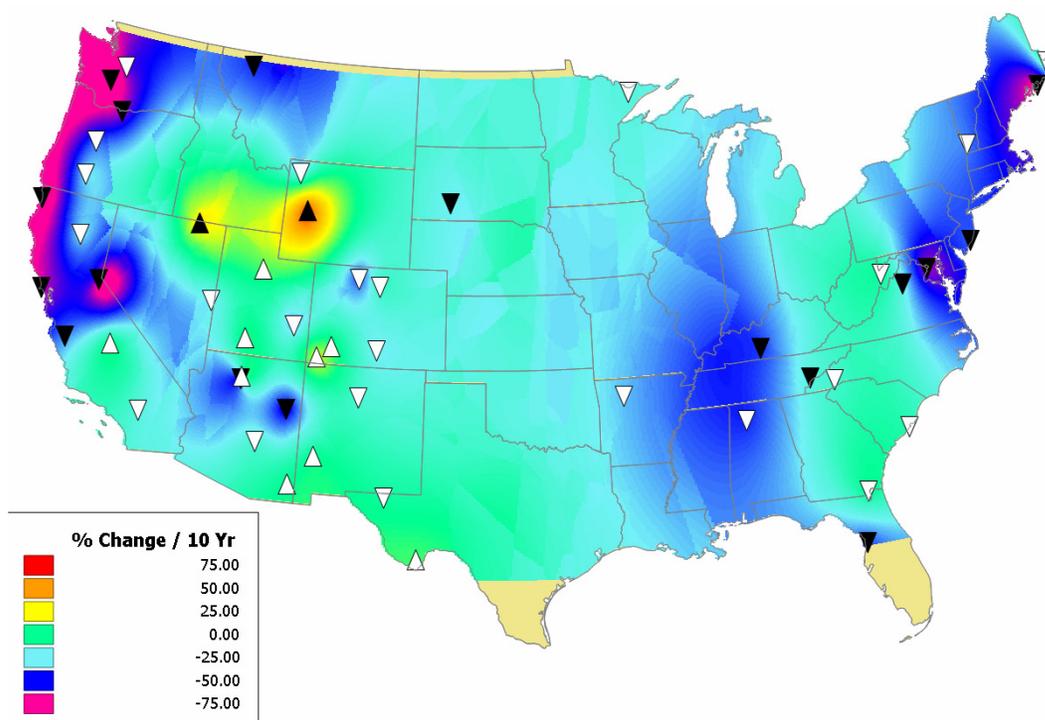
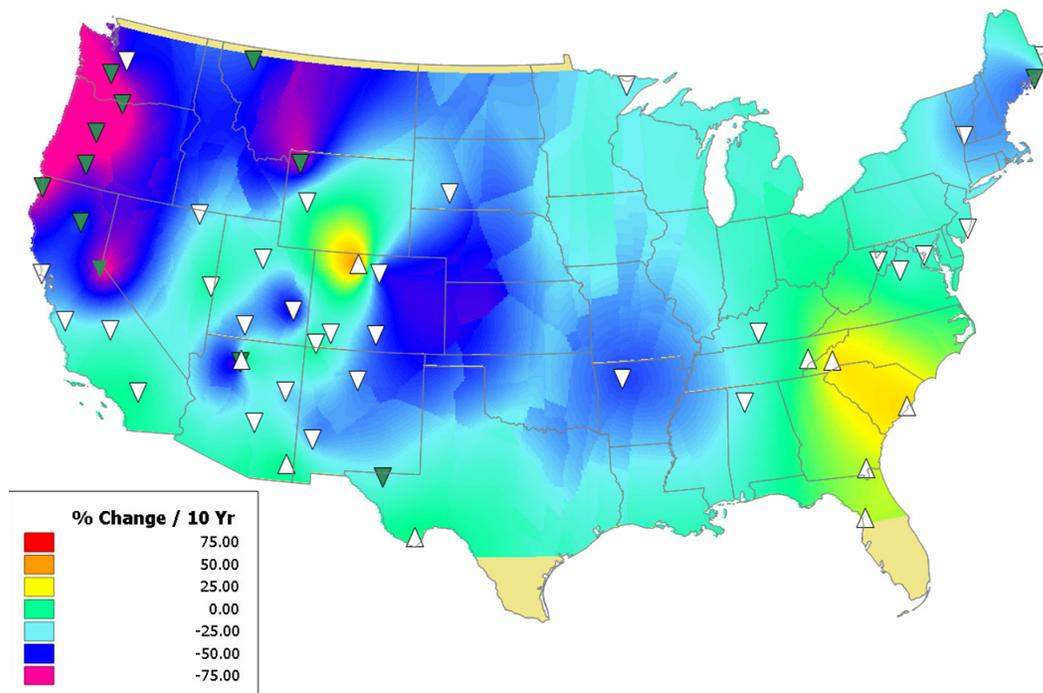


Figure 4.8. The wintertime elemental carbon trend using IMPROVE data from monitoring sites with a minimum of 7 years of data. The triangles indicate a increasing (up) or decreasing (down) trend, and black arrows have a significant trend at the 0.05 level. The isopleths are the slope of the trend line as the % change from the median EC concentration per 10 years.



**Figure 4.9. The wintertime organic carbon trend using IMPROVE data from monitoring sites with a minimum of 7 years of data.**

EC and OC displayed significant summer concentration trends at only a few sites (Figures 4.10 and 4.11). However, there were some coherent spatial patterns in the trends that were observed. Summertime EC and OC both decreased at most sites from Washington State to northern California and in Maine. A number of monitoring sites from Montana to New Mexico had increases of over 30% in the median summer OC and EC concentrations over a 10-year period, though the trends were not statistically significant. In the middle and late 1990s this region experienced increased wild fire, which likely drove the large increases in the observed carbon.

## Elemental Carbon, Summer

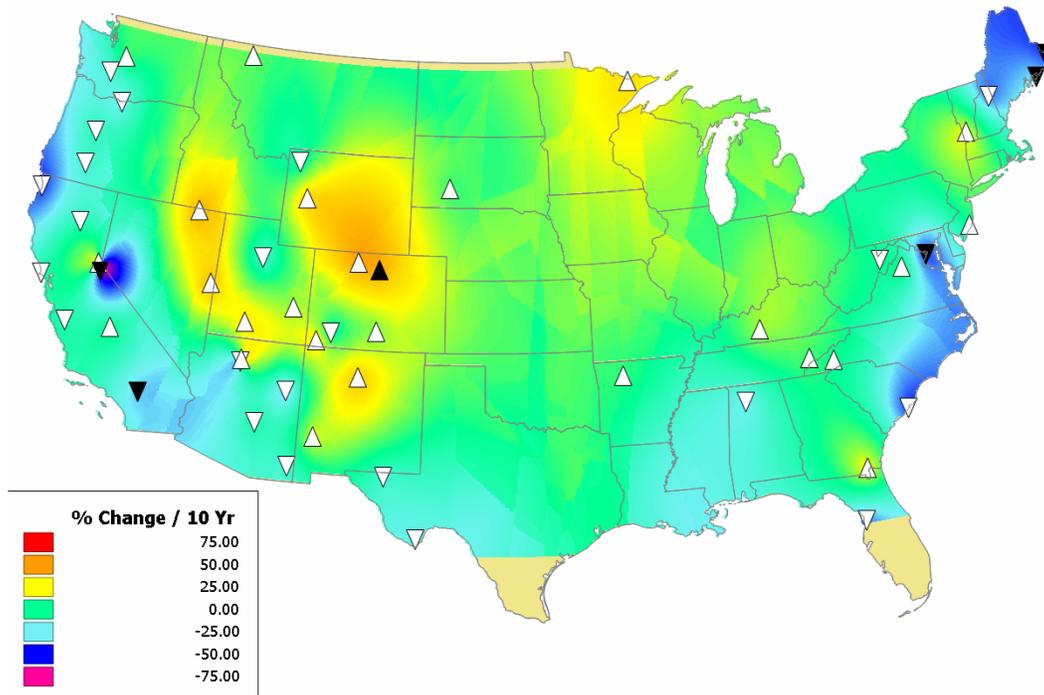
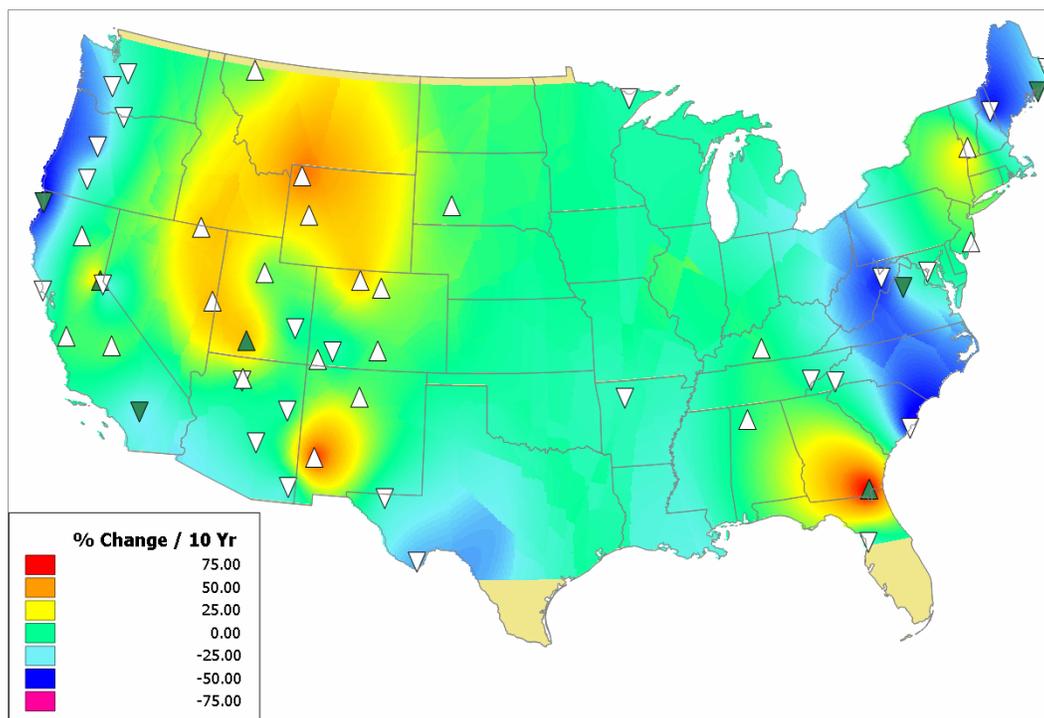


Figure 4.10. The summertime elemental carbon trend using IMPROVE data from monitoring sites with a minimum of 7 years of data.



**Figure 4.11. The summertime organic carbon trend using IMPROVE data from monitoring sites with a minimum of 7 years of data.**

OC and EC can be emitted from separate sources, e.g., secondary biogenic organic carbon, or at different ratios from the same source, e.g., fire and automobile traffic. The differing trends in OC and EC in the Northeast imply a change in the mix of sources contributing to carbonaceous aerosols there. Alternatively, these trends could be the result of changes or drifts in the sampling and analysis of the carbonaceous material. The samples have been analyzed using the same set of thermal optical reflectance (TOR) instruments over the past 15 years, and subtle changes may have occurred as the instruments have aged. In addition, the positive OC and EC artifact corrections have changed over time. For example, the EC artifact correction has been decreasing since the early 1990s.

#### **4.5 VIEWS Annual Summary Trends Tools**

VIEWS is an online exchange of air quality data, research, and ideas designed to understand the effects of air pollution on visibility and to support the Regional Haze Rule enacted by the U.S. EPA to reduce regional haze and improve visibility in national parks and wilderness areas. The Annual Summary section of the VIEWS web site provides access to data products described in the VIEWS scope of work. It also provides browsing capabilities of the IMPROVE aerosol database using graphical interfaces.

On the trends page of the annual summary section, the user can click on a map icon to view trend data for a selected parameter at that site (currently, <http://vista.cira.colostate.edu/views/web/AnnualSummaryDev/Trends.aspx> ). Annual data aggregations can be selected by calculated aerosol extinction percentile group and averaging period (either annual averages or 5-year rolling averages). The timeline plot has two modes, “Single” and “Multi”. In “Single” mode, the timeline will display the most recent selection. In “Multi” mode, each additional selection adds another timeline to the display. Up to four timelines can be superimposed in “Multi” mode.

The annual data aggregations for the 20% best and 20% worst visibility days represent the finalized calculation procedures as specified in guidance documents for the RHR. Daily IMPROVE data, including daily data substitutions where applicable, are available in the Annual Summary and by selecting the IMPROVE regional haze (IRHR) program in other VIEWS tools.

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