Thomas Cahill, University of California-Davis

"Trends s Analysis of Sulfates and Nitrates"

Trends Analysis of Sulfates and Nitrates

A presentation concerning critical needs in

"$PM_{2.5}$ Sampling and Analysis Workshop".

Sept. 30, 1997, DOE Federal Energy Technology Center, ~~Morgantown, WV~~ PITTSBURGH, PA

by Thomas A. Cahill, Prof. (Emeritus), Atm. Sciences /Physics and
Principal Investigator and Prime Contractor, IMPROVE Aerosol Program

Problem:

The nation needs prompt and accurate information on whether important legislation regarding energy and the environment, such as the 1991 CAA Amendments, are working as intended.

Are beneficial aspects being achieved?

Are there unexpected negative consequences?

Was the estimated cost/benefit ratio accurate?

This analysis needs measurements, as most environmental legislative actions are based on model predictions.

Trends Analysis of Sulfates and Nitrates

Example:

Trends in sulfates in national parks, 1982 - 1996

Problem:

There have been increases in sulfates at eastern national parks, 1982 - 1996.

However, there have been decreases in national SO_2 emissions in the same period.

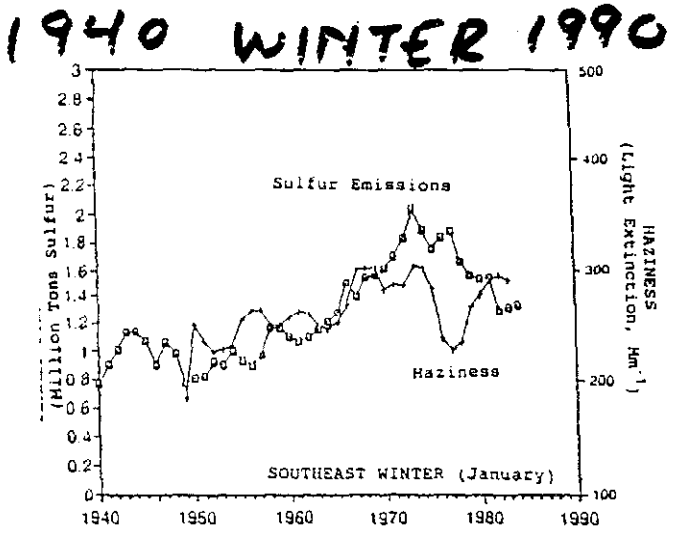
If this result is accurate, does it challenge the linear roll back model used in the 1991 CAA Amendments?

My answer:

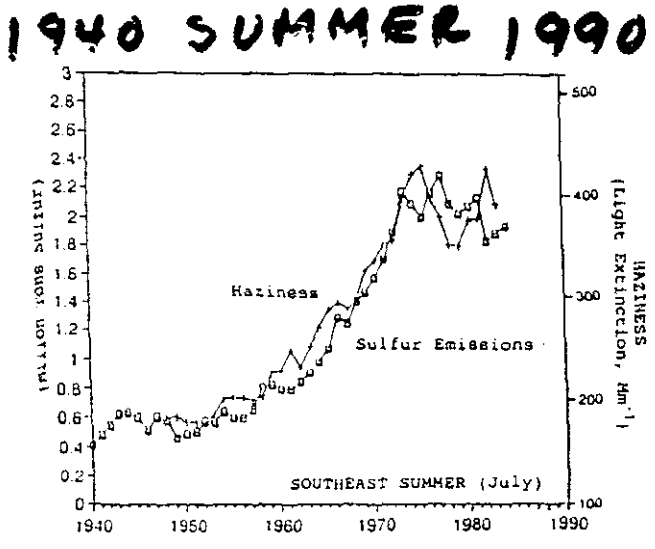
The upward trend in sulfates is real, but reflect local SO_2 increases rather than regional or national SO_2 patterns.

n the 1950's and 1960's with a significant increase in the ate 1960's. Since about 1970, there is no significant lecline except for 1983. Overall, the emission and haze rends show certain correspondences, particularly in the 950's and 1960's, but they deviate somewhat since the 970's.

The winter haze and emission trends for the southeast are hown in Figures 24-36a and 24-36b. Winter emissions ise moderately until the early 1970's, when a slight ecline begins. The winter haziness also shows a moder-



igure 24-36a Comparison of sulfur emission trends (□) and extinction coefficient (+) for the southeastern region during the winter months.



ure 24-36b Comparison of sulfur emission trends (□) and extinction coefficient (+) for the southeastern region during the summer months.

ate increase up to the early 1970's. The dip in winter haziness around 1977 has no correspondence in the emission trends. The comparison of summer sulfur emissions and haziness for the southeast is found in Figure 24-36b. There is a remarkable correspondence, increasing values from the late 1940's through the early 1970's followed by a leveling off since then.

The statistical relationship between historical sulfur emissions and extinction coefficient is further illustrated as a scatterplot in Figure 24-37. Here, yearly emissions and extinction coefficient are aggregrated over the entire East (states east of the Mississippi). In Figure 24-37, the correspondence between yearly sulfur emissions and yearly extinction coefficient is fairly close. As noted previously, the relationship between extinction coefficient and sulfur emissions depends on region and season. However, one aspect that is the same for both regions and both seasons is that a 50% change in sulfur emissions from current levels is statistically associated with about a 30% change in light extinction.

In conclusion, these data show that trends in the seasonal sulfur emissions provide a plausible explanation for the observed seasonal trends of atmospheric extinction coefficient over the eastern United States. However, such qualitative comparisons do not provide conclusive evidence of a cause-effect relationship. Also, the pattern of haze and sulfur emissions for the northeast and southeast tend to deviate at times. The causes of such deviations may include variabilities due to meteorology as well as poten-

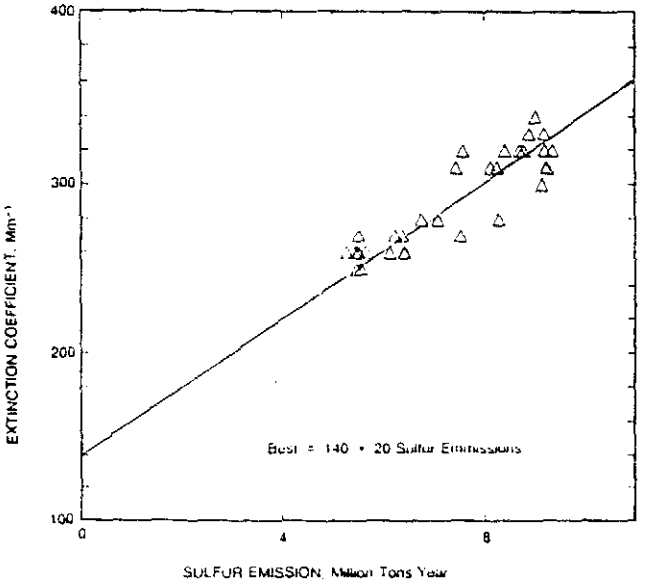
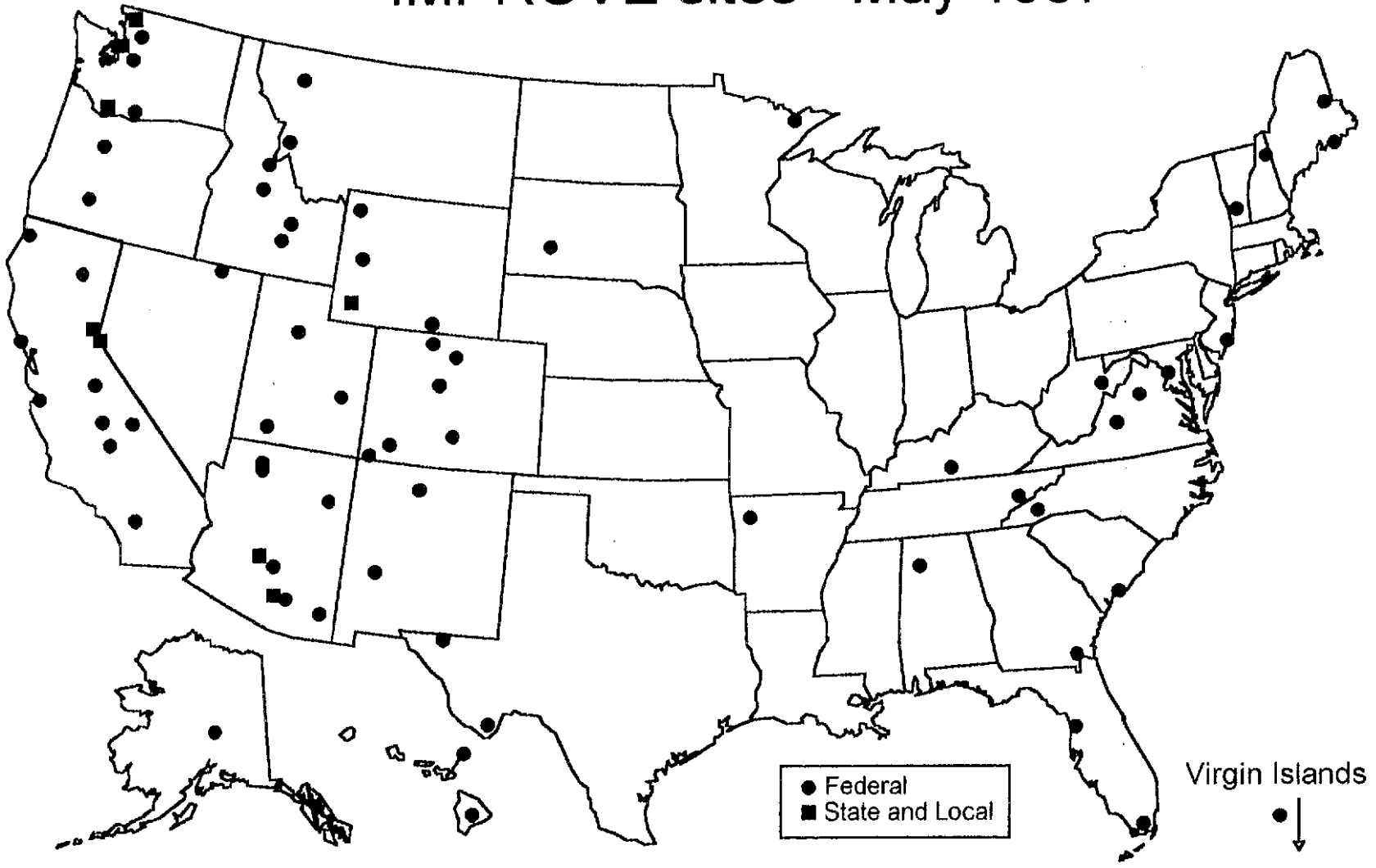
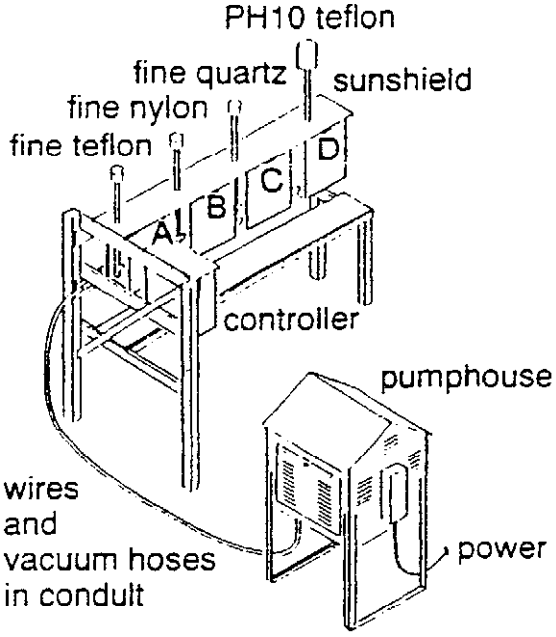


Figure 24-37 Relationship between yearly extinction coefficients and yearly sulfur emissions for the entire East.

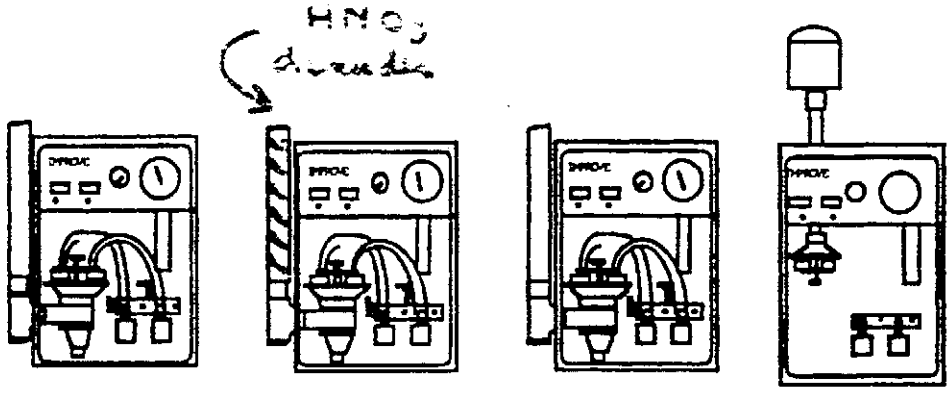
IMPROVE sites - May 1997





Layout of the IMPROVE modular aerosol monitor.

Each of the 4 filters are analyzed in the following way:



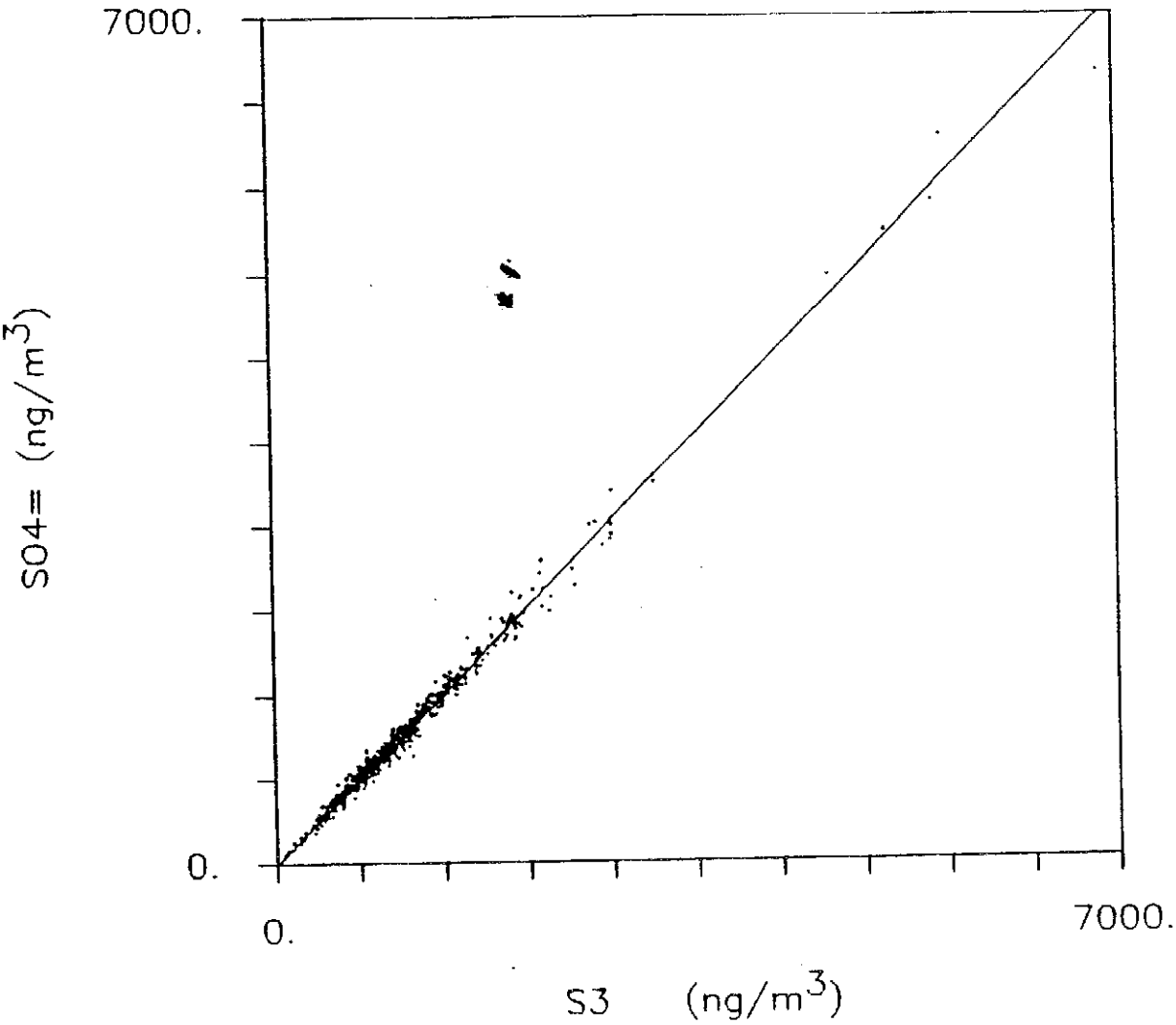
	A	B	C	D
sizes	0-2.5 µm	0-2.5 µm	0-2.5 µm	0-2.5 µm
filter	Teflon	nylon	quartz	Teflon
area	2.2 cm^2	14 cm^2	3.8 cm^2	3.8 cm^2
analysis	mass absorption PIXE PESA	IC	thermal carbon	mass

Schematic of IMPROVE sampling system. System is highly redundant, with four units sharing only clock to ensure on and off times are accurate.

SELECT WESTERN
B92

N = 463
R = 0.995 R^2 = 0.990
Y = 1.0180*X − 11.4688
σ_a= 0.0095 σ_b= 10.1444

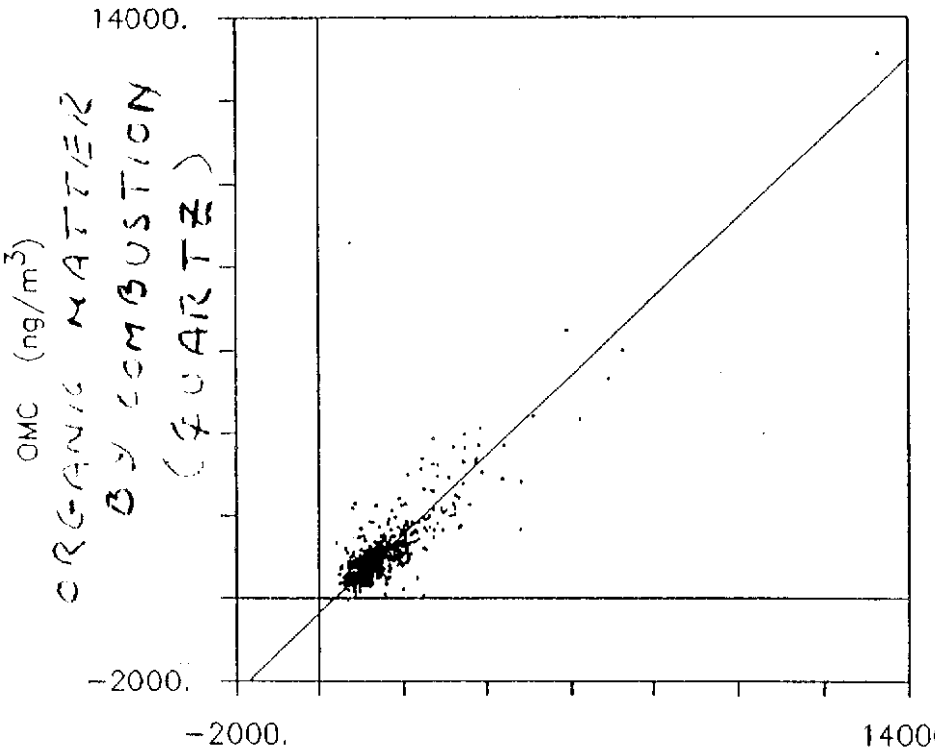
$\bar{x}$ = 1065.39 $\sigma_{\bar{x}}$= 33.77 σ_x= 726.68
$\bar{y}$ = 1073.09 $\sigma_{\bar{y}}$= 34.38 σ_y= 739.69



SELECT WESTERN
A88

N = 440
R = 0.888 R^2 = 0.788
OMC = 0.9549*OMH − 354.6689
σ_a = 0.0477 σ_b = 77.5473

$\bar{x}$ = 1624.64 $\sigma_{\bar{x}}$ = 50.73 σ_x = 1064.02
$\bar{y}$ = 1196.69 $\sigma_{\bar{y}}$ = 48.69 σ_y = 1021.30

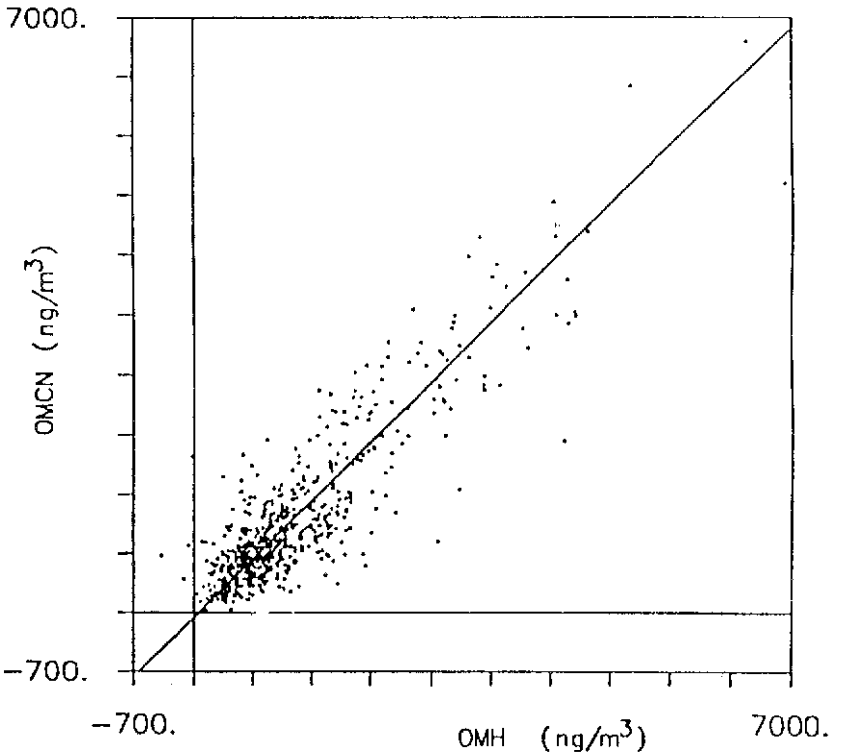


ORGANIC MATTER BY COMBUSTION (QUARTZ)

SELECT WESTERN
A94

N = 617
R = 0.875 R^2 = 0.766
Y = 0.9910*X − 67.0284
σ_a = 0.0454 σ_b = 53.7497

$\bar{x}$ = 1182.92 $\sigma_{\bar{x}}$ = 36.45 σ_x = 905.33
$\bar{y}$ = 1105.19 $\sigma_{\bar{y}}$ = 36.16 σ_y = 898.16

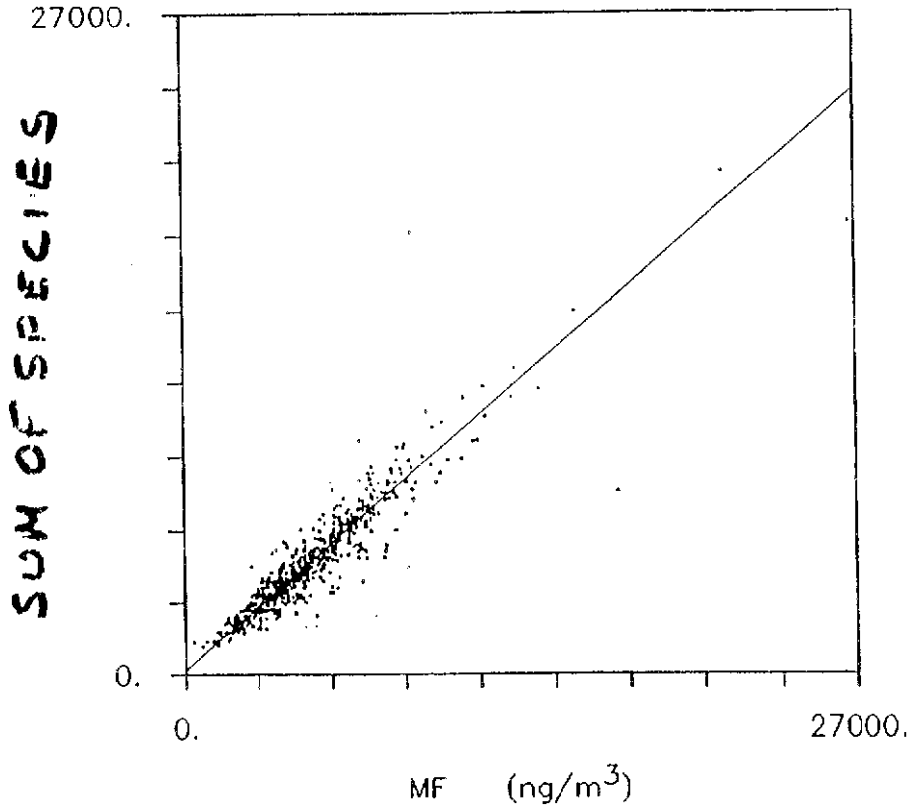


ORGANIC MATTER BY PESA (TEFLON)

SELECT WESTERN
B88

N = 523
R = 0.917 R^2 = 0.840
RCMA= 0.8769•MF + 196.4854
σ_a= 0.0326 σ_b= 167.5649

$\bar{x}$ = 5145.60 $\sigma_{\bar{x}}$= 114.75 σ_x=2624.19
$\bar{y}$ = 4708.53 $\sigma_{\bar{y}}$= 101.72 σ_y=2326.29



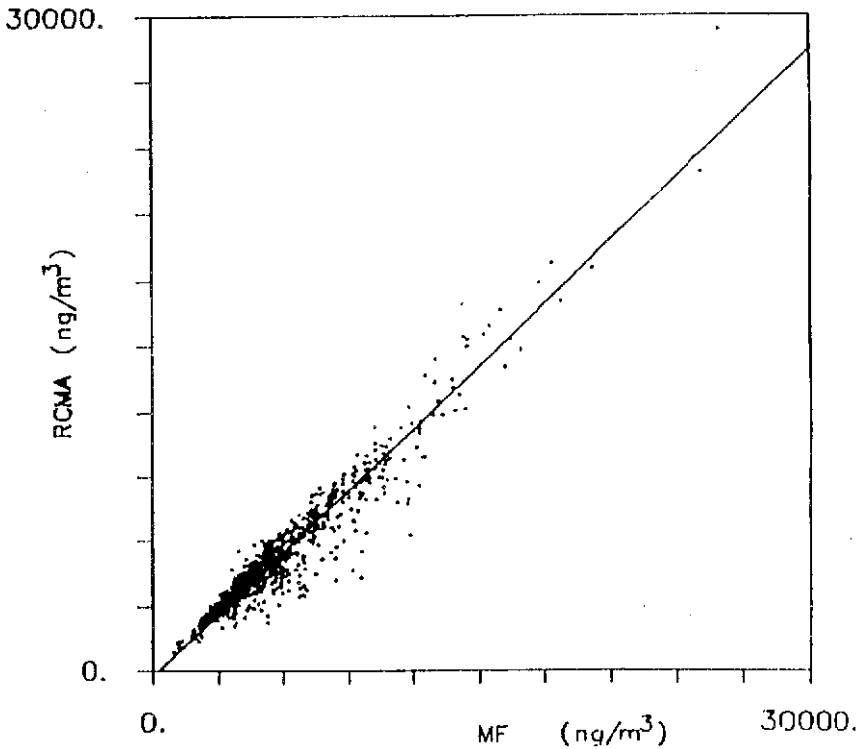
SUM OF SPECIES

FINE MASS

SELECT WESTERN
B94

N = 749
R = 0.956 R^2 = 0.914
Y = 0.9553•X − 264.9922
σ_a= 0.0212 σ_b= 124.9941

$\bar{x}$ = 5904.31 $\sigma_{\bar{x}}$= 112.02 σ_x=3065.72
$\bar{y}$ = 5375.24 $\sigma_{\bar{y}}$= 107.22 σ_y=2934.46



FINE MASS

JOURNAL OF GEOPHYSICAL RESEARCH VOL 99, NO D1 PAGES 1347-1370, JANUARY 20, 1994

Spatial and seasonal trends in particle concentration and optical extinction in the United States

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CIRA, Colorado State University, Fort Collins

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Abstract. In the spring of 1988 an interagency consortium of Federal Land Managers and the Environmental Protection Agency initiated a national visibility and aerosol monitoring network to track spatial and temporal trends of visibility and visibility-reducing particles. The monitoring network consists of 36 stations located mostly in the western United States. The major visibility-reducing aerosol species, sulfates, nitrates, organics, light-absorbing carbon, and wind-blown dust are monitored as well as light scattering and extinction. Sulfates and organics are responsible for most of the extinction at most locations throughout the United States, while at sites in southern California nitrates are dominant. In the eastern United States, sulfates contribute to about two thirds of the extinction. In almost all cases, extinction and the major aerosol types are highest in the summer and lowest during the winter months.

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TRENDS IN ELEMENTAL CONCENTRATIONS OF FINE PARTICLES AT REMOTE SITES IN THE UNITED STATES OF AMERICA

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Abstract—The University of California at Davis has been monitoring particulate concentrations in remote sites throughout the United States of America since 1979 in networks operated for the National Park Service and the Environmental Protection Agency. Twelve sites in remote class I visibility areas have almost complete records from June 1982 to August 1992, seven in the southwest, three in the northwest, and two in the east. During this period, two samples of fine particles (0–2.5 μm) on Teflon filters were collected every week and analysed for elemental concentration by Particle Induced X-ray Emission (PIXE). This paper will examine the historical trends for sulfur, zinc, lead, and the soil elements. Measurements during the past four years have verified that the sulfur is present as sulfate. In the southwest, 80% of the sulfur trends in spring, summer and fall decreased, while most of the winter trends increased. The annual trends decreased at six of the seven sites. The trends in the northwest increased slightly. The two eastern sites had the most important trends, with significant increases of almost 4% per year in summer, 1–3% increases in spring and fall, and 2% decreases in winter. The annual increases were between 2 and 3%. Generally there were no significant trends for zinc and the soil elements. Lead at all sites decreased sharply through 1986, corresponding to the shift to unleaded gasoline, but has since leveled off at around 18% of the 1982 means. The most important conclusion of this study is that through the use of stable sampling and analytical protocols, we have been able to determine statistically significant historical trends of as small as 1–2% per year for sites with 10-year records.

Key word index: Particulate monitoring networks, sulfur trends, aerosol trends, National Parks, class I visibility areas.

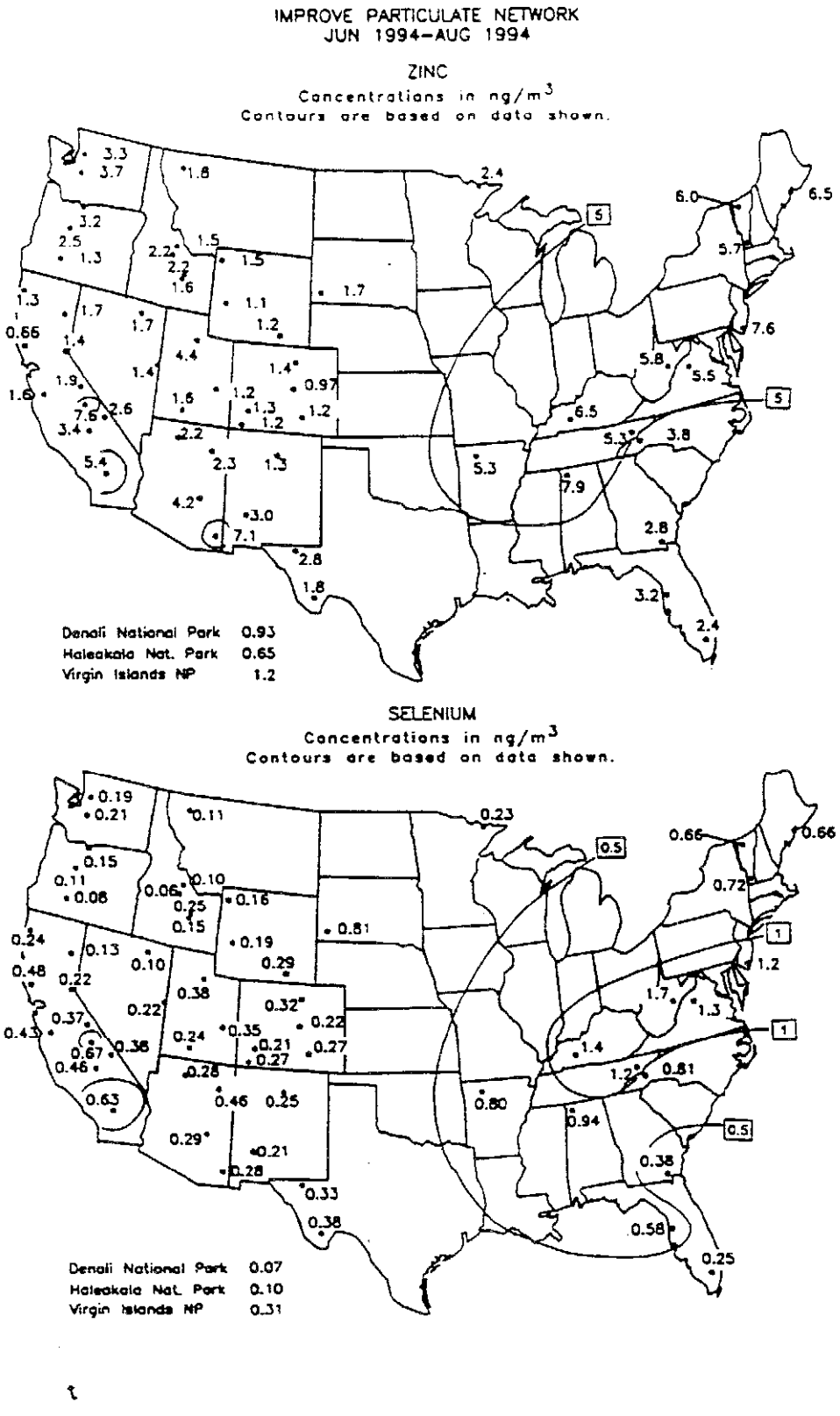


Figure 27. Con our plots of fine zinc and fine selenium for summer 1994 (June-August).

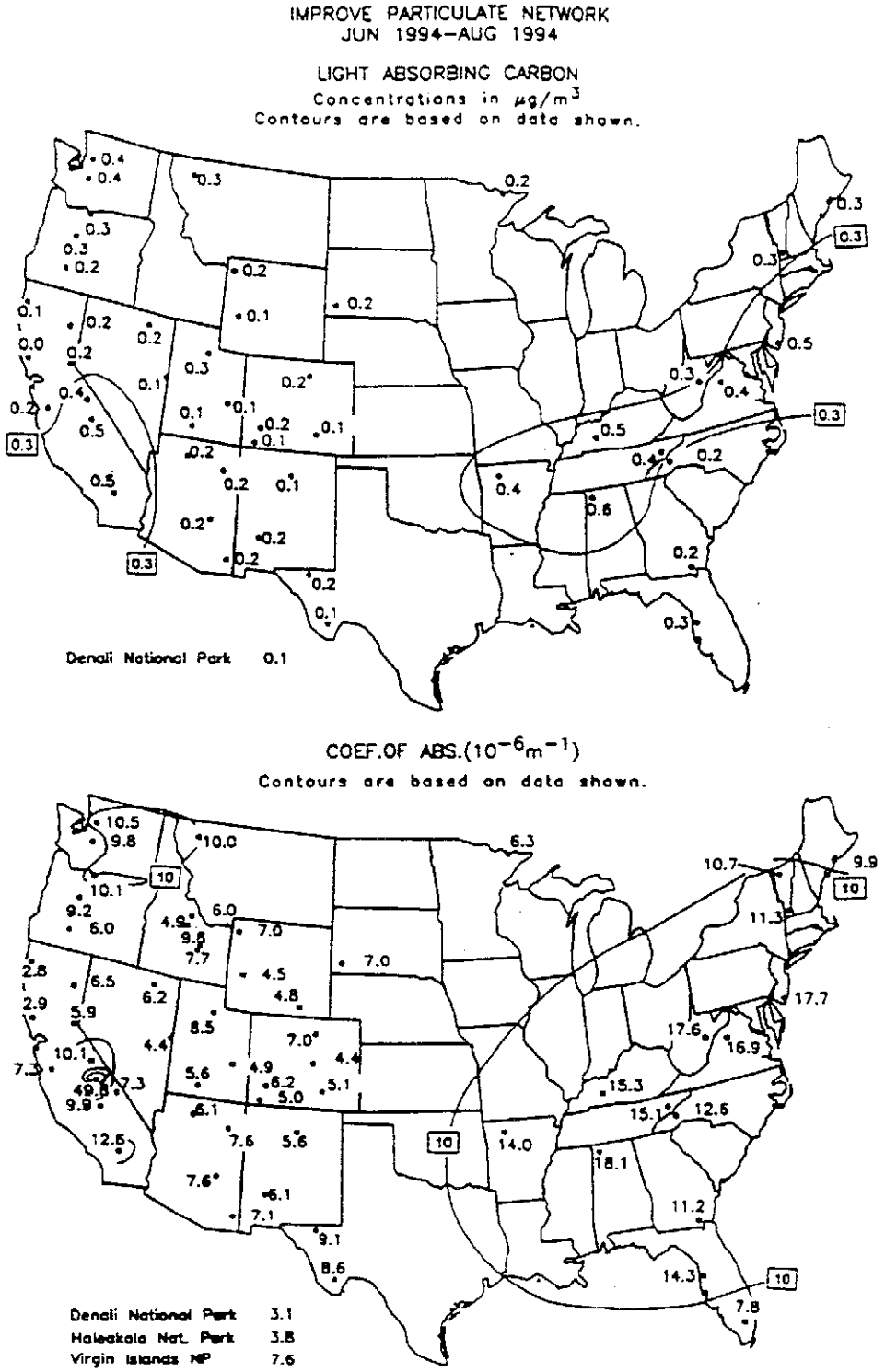


Figure 23. Contour plots of fine light-absorbing carbon and the coefficient of absorption for summer 1994 (June-August).

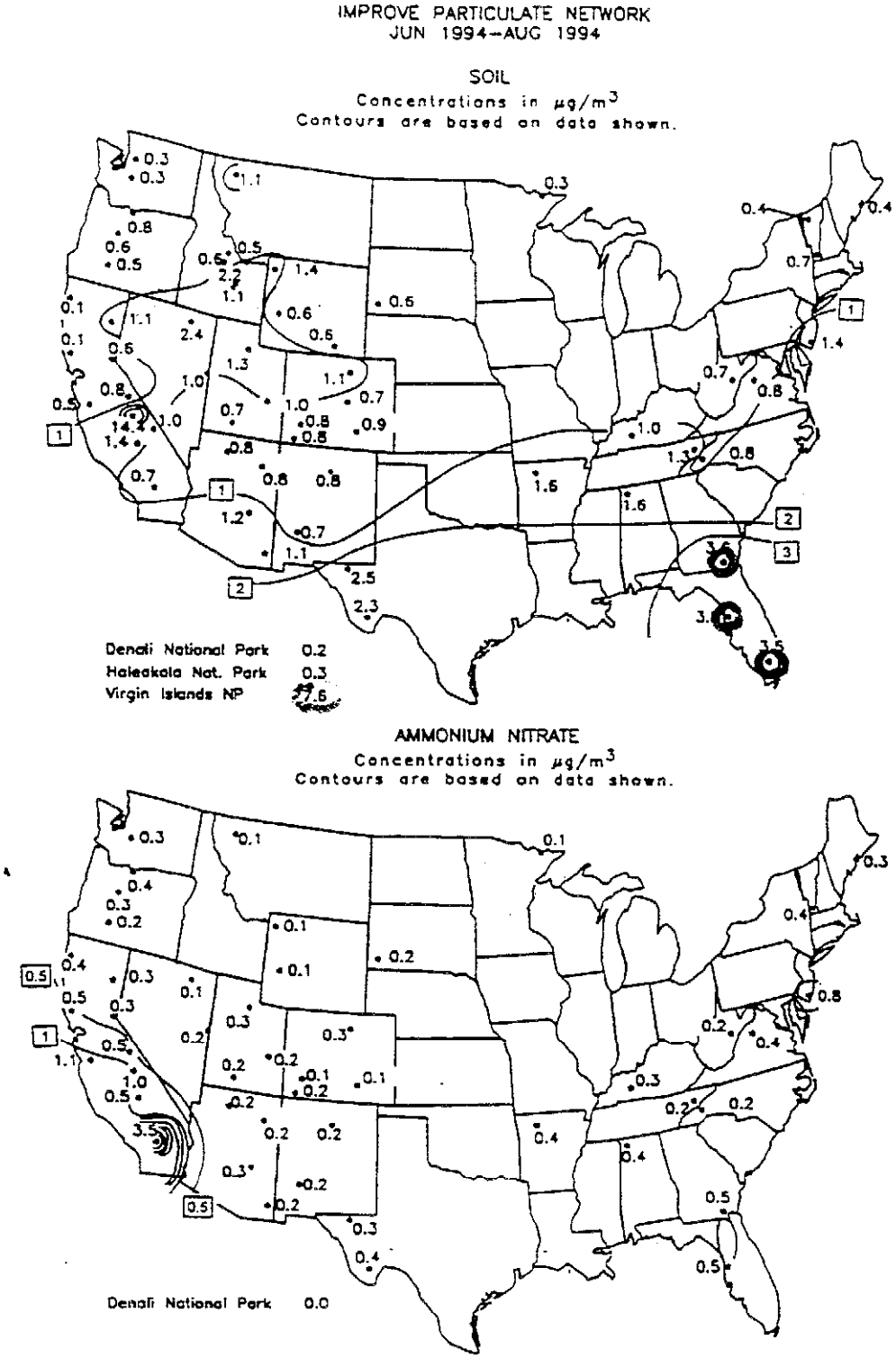


Figure 19. Contour plots of fine soil and fine ammonium nitrate for summer 1994 (June-August).

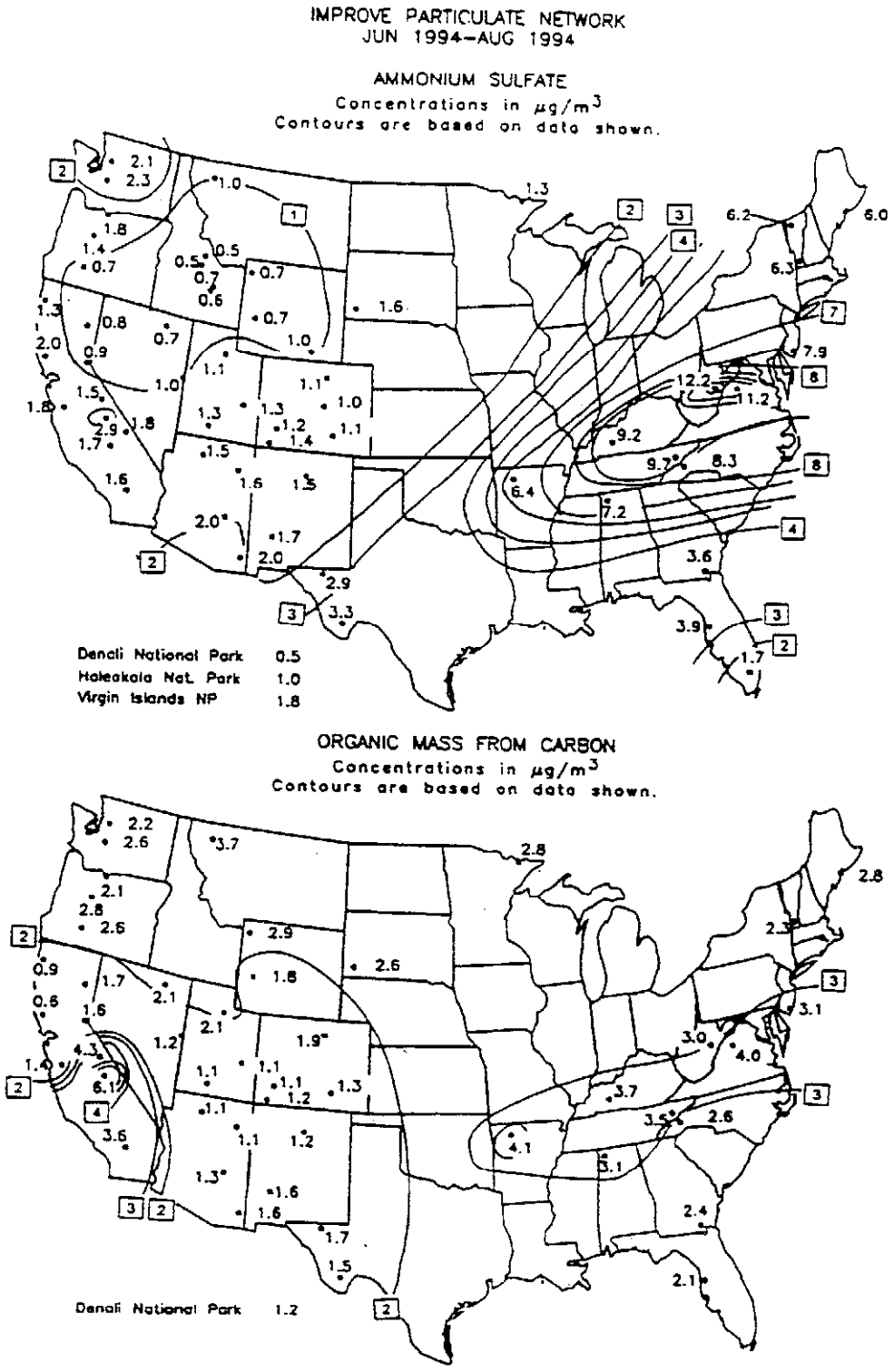


Figure 15. Contour plots of fine ammonium sulfate and fine organic mass by carbon for summer 1994 (June-August).

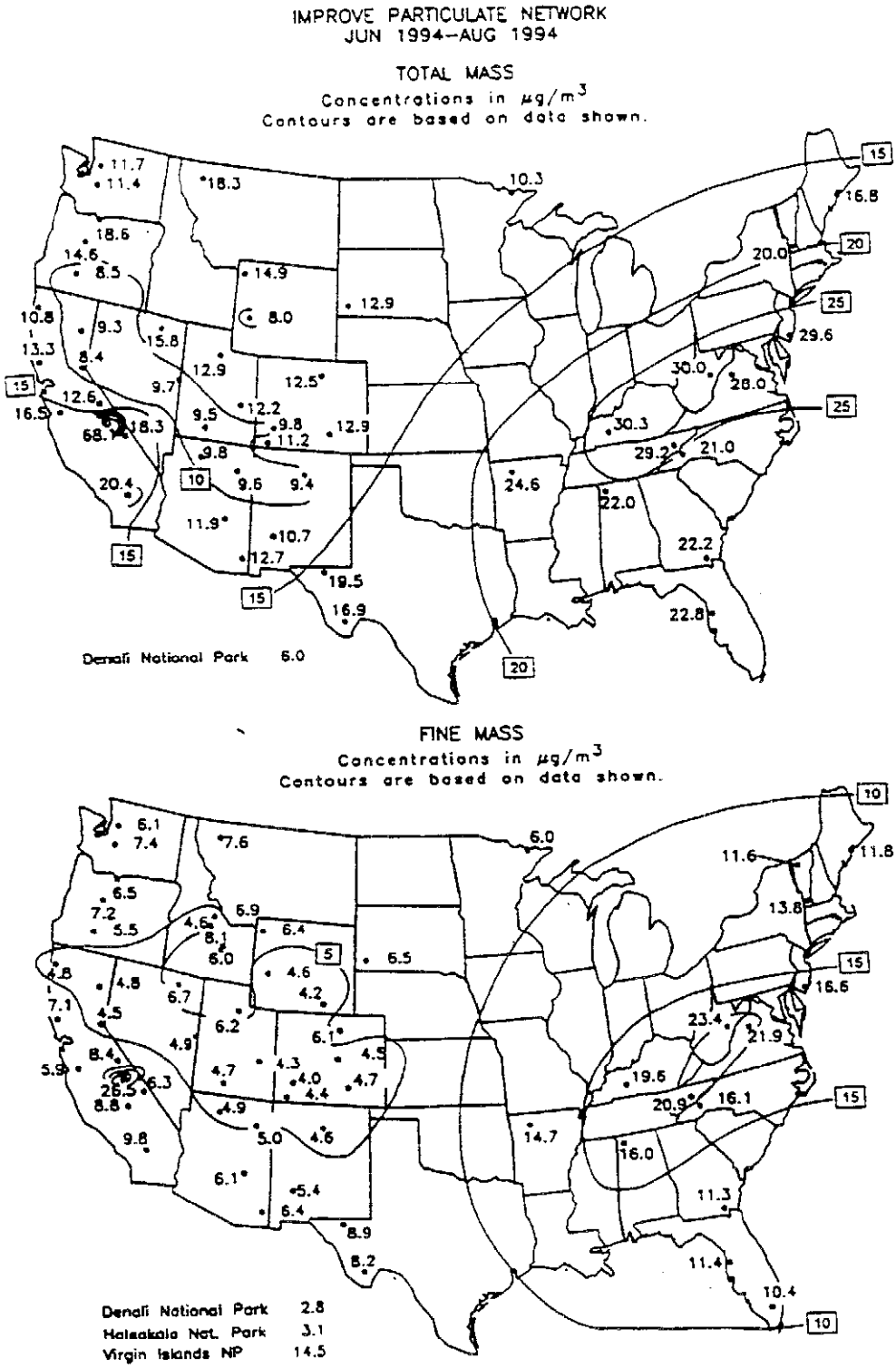


Figure 11. Contour plots of PM_{10} and $PM_{2.5}$ mass for summer 1994 (June-August).

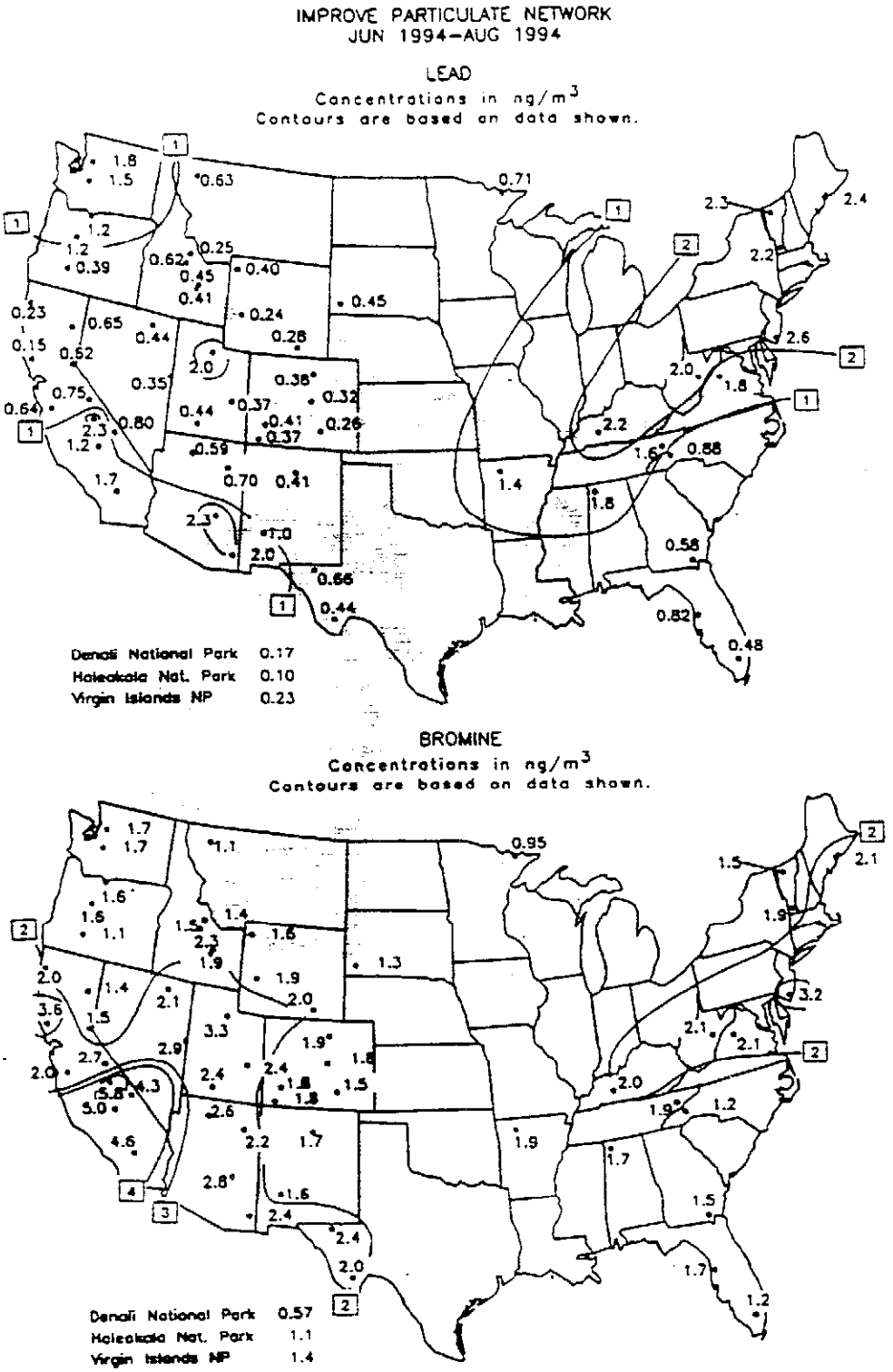
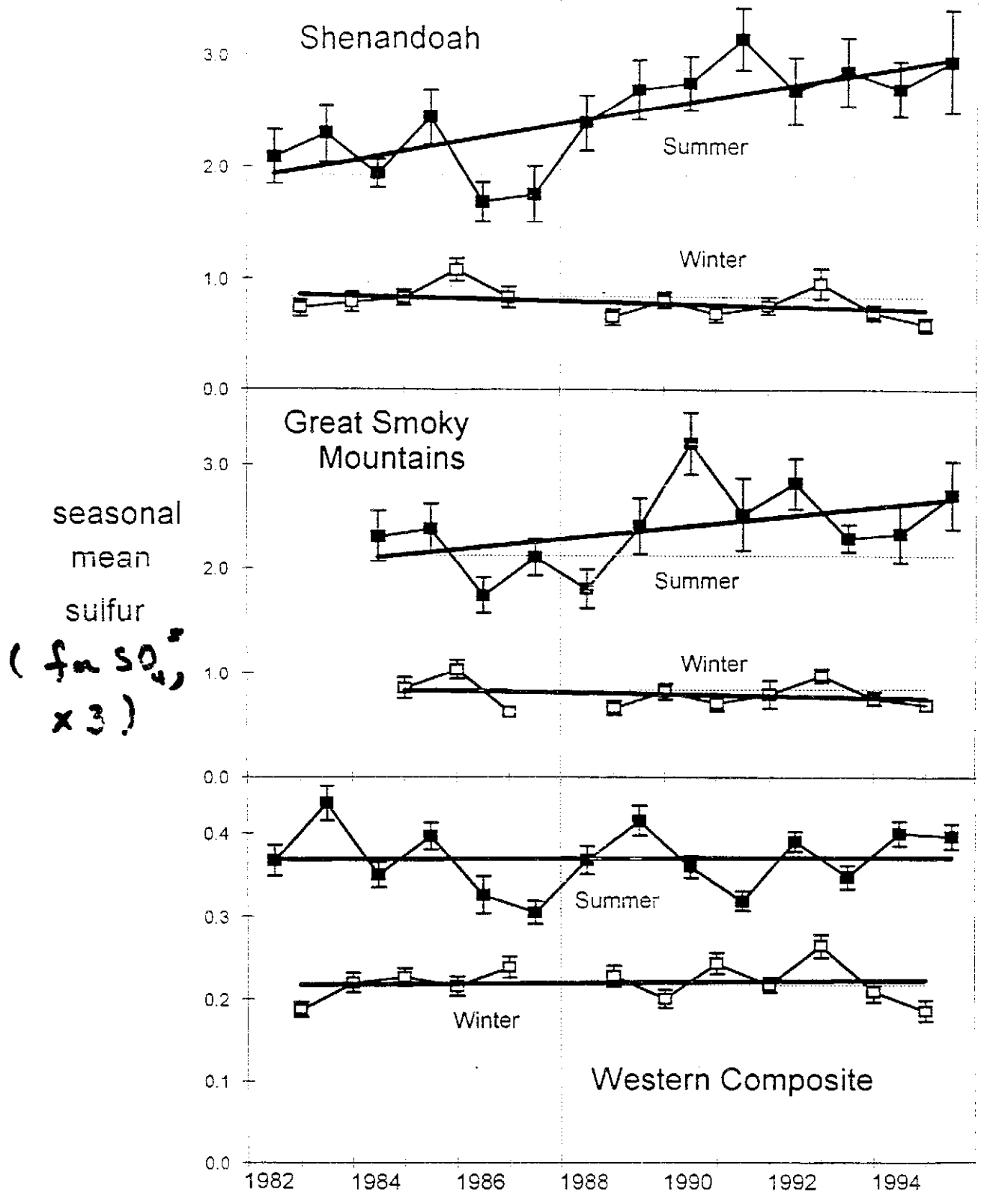


Figure 31. Contour plots of fine lead and fine bromine for summer 1994 (June-August).

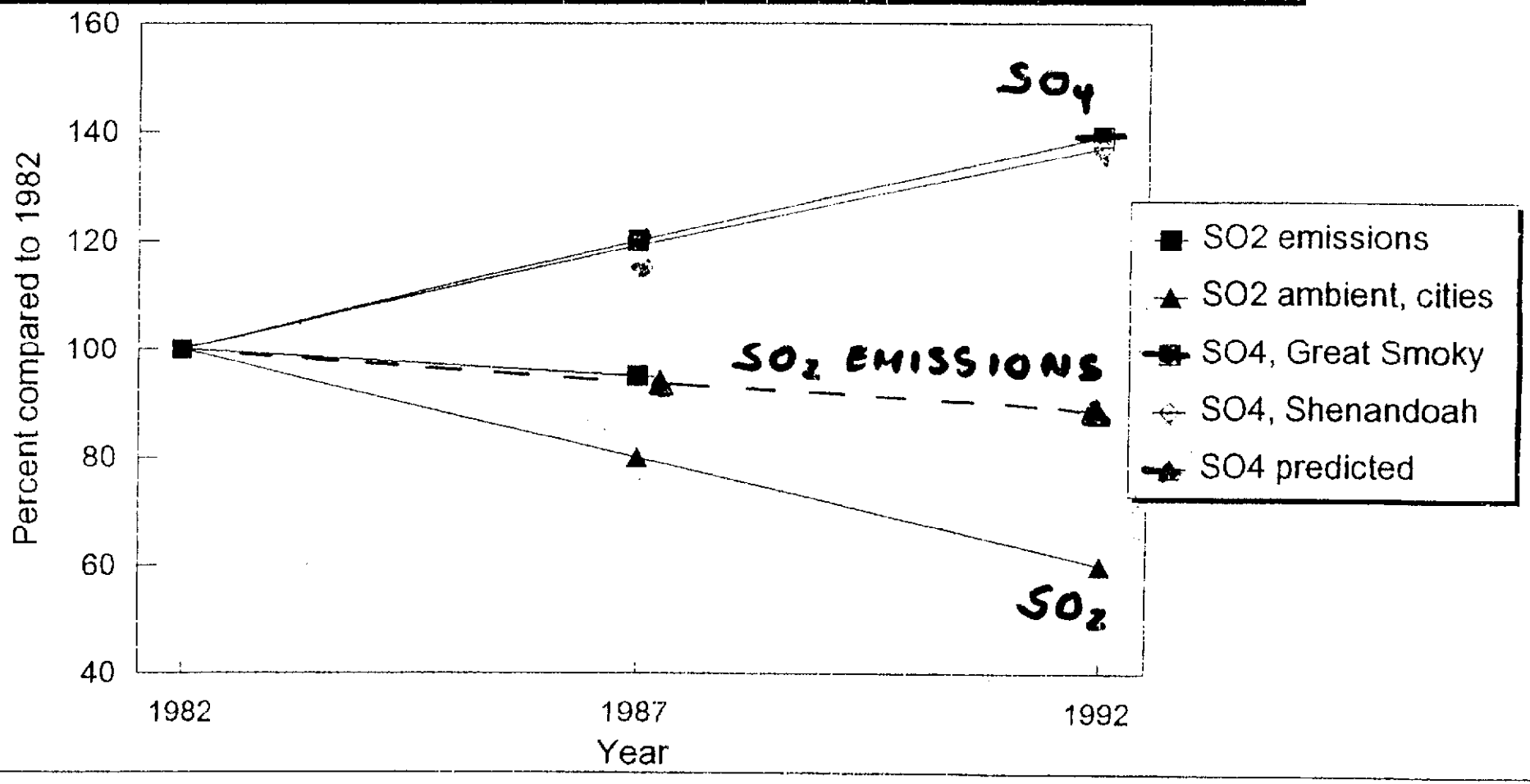


seasonal mean sulfur ($f_m SO_4^=$ x 3)

The error bars indicate the standard error of the concentrations.
The solid lines indicates the best fit for all point.
The dotted lines indicates the horizontal.
The vertical dotted line indicates the change from stacked filter unit to IMPROVE sampler.

Sulfur Emissions and Observations in the East

NPS/IMPROVE Program



JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 102, NO. D6, PAGES 6813–6821, MARCH 27, 1997

Deteriorating air or improving measurements? On interpreting concatenate time series

Warren H. White

Chemistry Department, Washington University, Saint Louis, Missouri

Abstract. Measurements reported by *Eldred and Cahill* [1994] provide an extended record of fine-particle sulfur concentrations at Shenandoah National Park. Begun in 1982, the measurements were upgraded in 1986 to improve time resolution and again in 1988 to improve sensitivity and particle size discrimination. Standard hypothesis rejection tests indicate a statistically significant upward trend in measured concentrations from 1982 to 1992. However, similar tests also indicate statistically significant differences between measurements made by the different methods, even when the apparent trend is accounted for. Does the trend in measured concentrations reflect increasing atmospheric concentrations, or is it an artifact of evolving measurement methods? This paper frames the question in rudimentary Bayesian terms and shows that one's interpretation of the trend is sensitive to one's prior confidence in the measurements.

Introduction

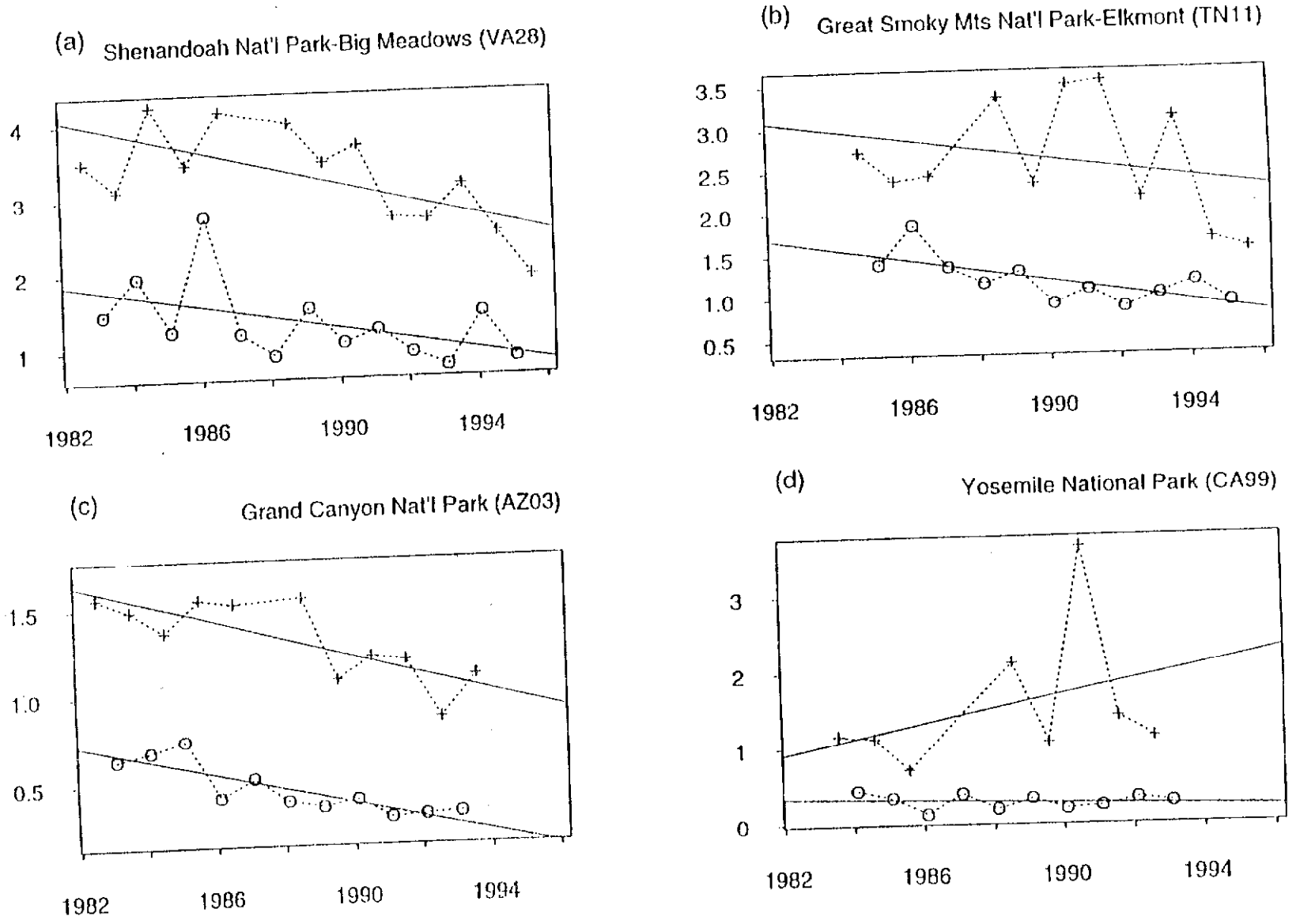
Shenandoah National Park is a scenic preserve in the Blue Ridge Mountains of western Virginia. This area lies within the core of a large region now covered by regular summertime haze [*Husar and Wilson*, 1993]. Visitors to the park are greeted by signs advising them of current air quality [*Frampton*, 1995], and the sulfate aerosol that limits visibility has been the subject of several studies [*Ferman et al.*, 1981; *Stevens et al.*, 1984; *Malm and Sisler*, 1987; *Gebhart et al.*, 1994]. Although aggregate sulfur dioxide emissions over the eastern United States are expected to decline as a result of the 1990 Clean Air Act Amendments [*Malm et al.*, 1994], several major new coal-fired boilers are currently under development in the vicinity of the park [*Carr*, 1995].

Ambient particles have been routinely monitored at Shenandoah since 1982, and the resulting data now form the longest continuing record we have for the regional sulfate aerosol [*Allan and Mueller*, 1995]. Figure 1 shows the 1982-1992 summer average concentrations of fine-particle sulfur [*Eldred and Cahill*, 1994]. The Shenandoah data exhibit an upward trend that is supported by measurements since 1984 at Great Smoky Mountains, another national park in the southern Appalachians. A nonparametric fit [*Sen*, 1968] to the Shenandoah data yields an average annual increase of 3.3%, which compounds to 38% over the 10 year record. Such a large increase comes as something of a suprise [*McDonald*, 1994], because estimated eastern U.S. [*National Acid Precipitation Assessment Program (NAPAP)*, 1991] and mid-Atlantic (Figure 2) sulfur dioxide emissions were flat during this period. It is not supported by a recent comparison of data from the 1977-1978 Sulfur Regional Experiment and 1988-1989 Eulerian Model Evaluation Field Study [*Shreffler and Barnes*, 1996].

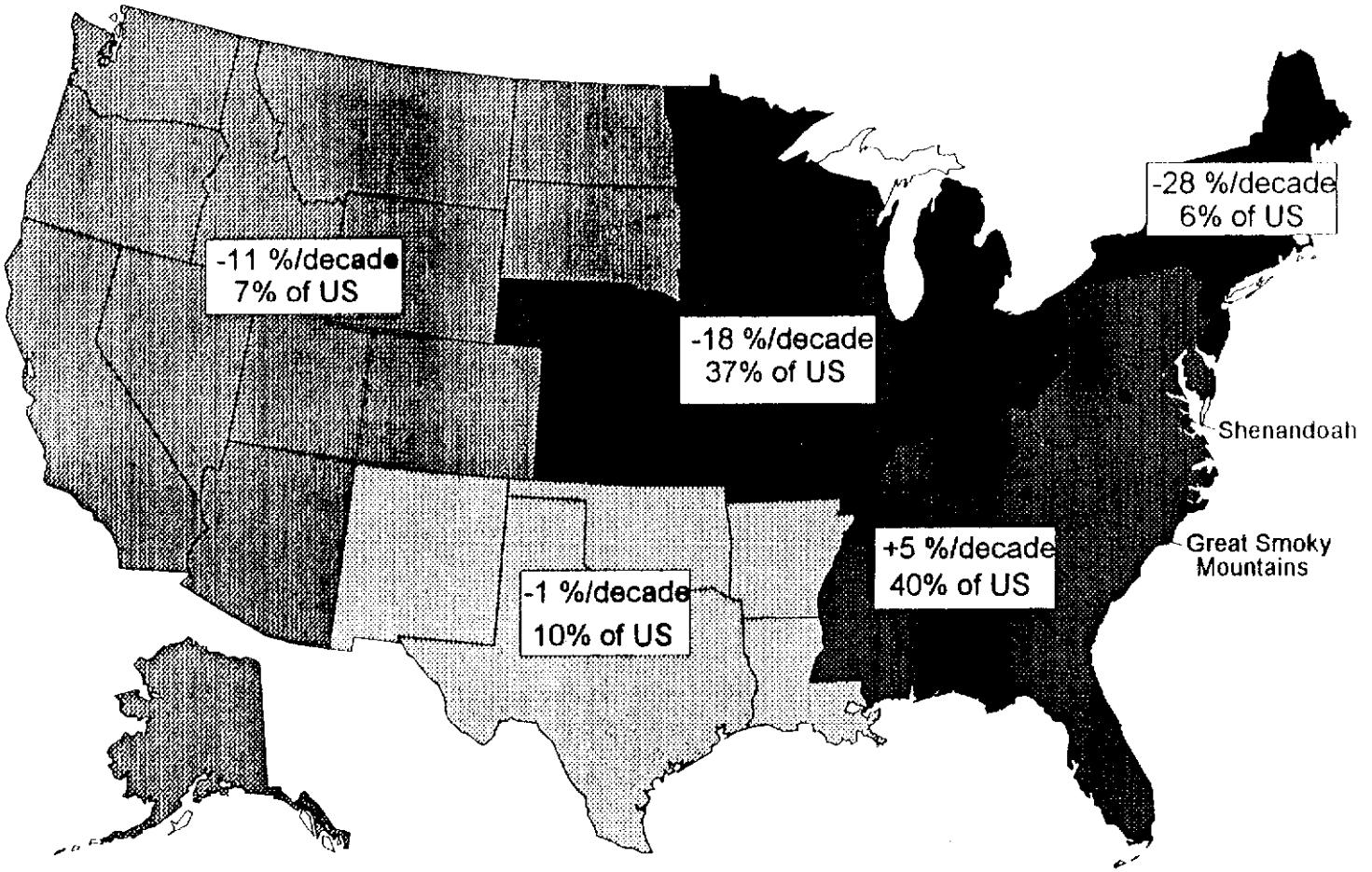
Paper number 97JD00030.
0148-0227/97/97JD-00030$09.00

Figure 2 shows the two largest year-to-year shifts in concentration to have coincided with significant changes in the method used to sample ambient particles (Table 1). The first change occurred between 1985 and 1986, when the sampling interval was reduced from 72 hours to 24 hours with the filters masked to retain adequate areal loadings. The coincident concentration drop was not replicated in other southern Appalachian measurement series that might have been expected to track regional sulfate concentrations (Figure 3): summer average particle sulfur concentrations at Kingston, Tennessee [*Koutrakis et al.*, 1988; G. Allen and P. Koutrakis, personal communication, 1995], summer median visual ranges at Shenandoah [*O'Leary*, 1988], summer precipitation-weighted average sulfate concentrations in Shenandoah rain [*National Atmospheric Deposition Program (NADP)*, 1995], and annual average particulate matter (PM10) concentrations at Roanoke, Virginia [*U.S. Environmental Protection Agency (USEPA)*, 1995, R.B. Husar, personal communication, 1995]. The second change occurred between 1987 and 1988, when the stacked filter unit was replaced by the IMPROVE sampler [*Cahill et al.*, 1990]. The sharp rise in sulfur concentrations is not replicated in the Roanoke PM10 data, the only overlapping series in Figure 3.

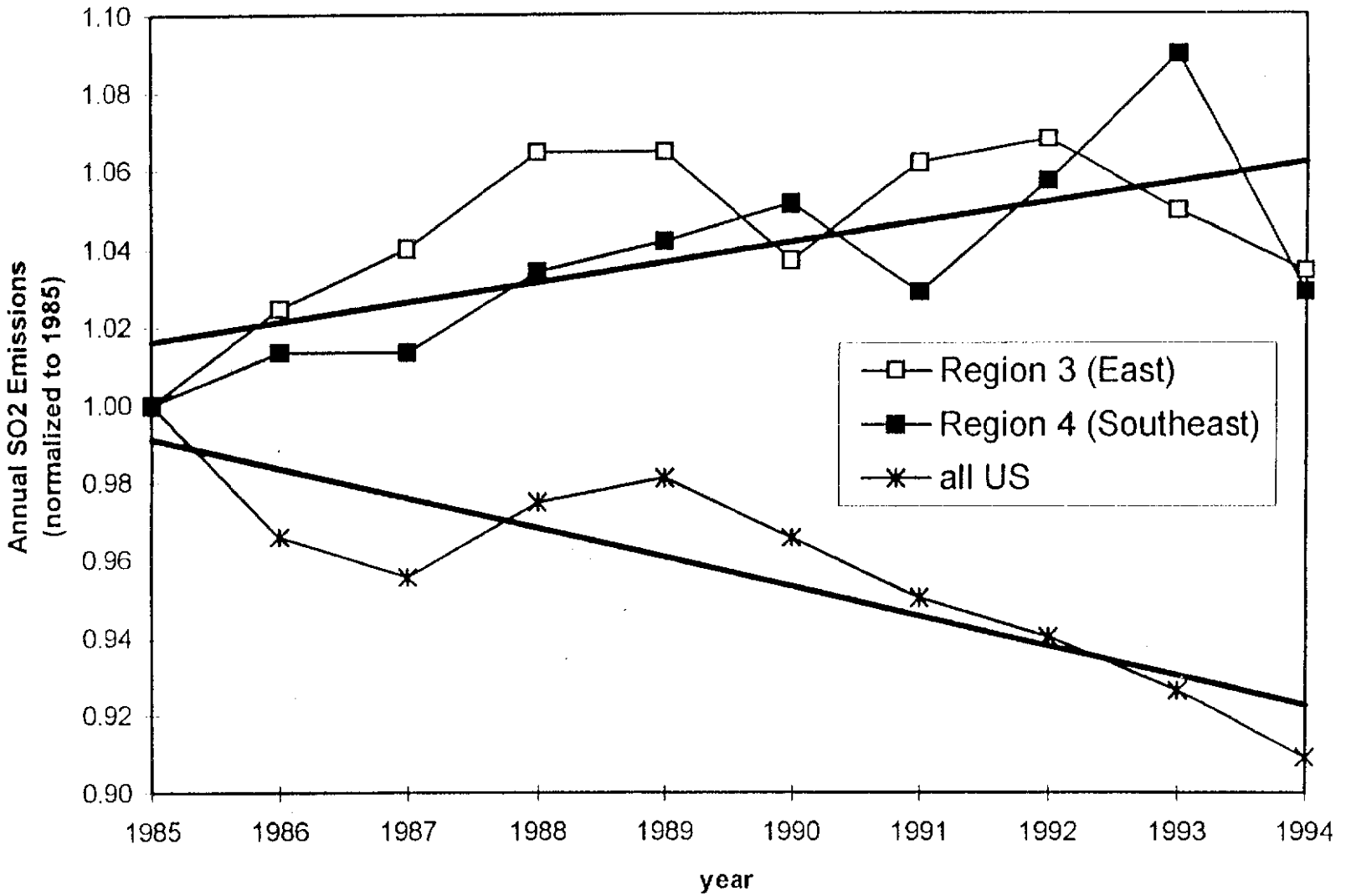
The Shenandoah sulfur trend is statistically significant at the 5% two-sided error level [*White*, 1996]. Moreover, it is based on data from a continuous measurement program, a program administered and executed by a stable team that evaluates equivalence when introducing new methods and maintains consistency as an explicit goal [*Cahill et al.*, 1990, 1996]. As substantial and well-documented as the measured increase is, however, it is also "startling" and "a paradox" [*McDonald*, 1994], because it is as yet unexplained by known emissions and meteorological trends. It is clearly remarkable that each of the year-to-year differences coinciding with the two major sampling changes is larger than the cumulative 1982-1992 increase. In addition to the obvious interpretation in terms of unrecognized trends in emissions and/or climate, an analyst might therefore wish to consider an alternative inter-

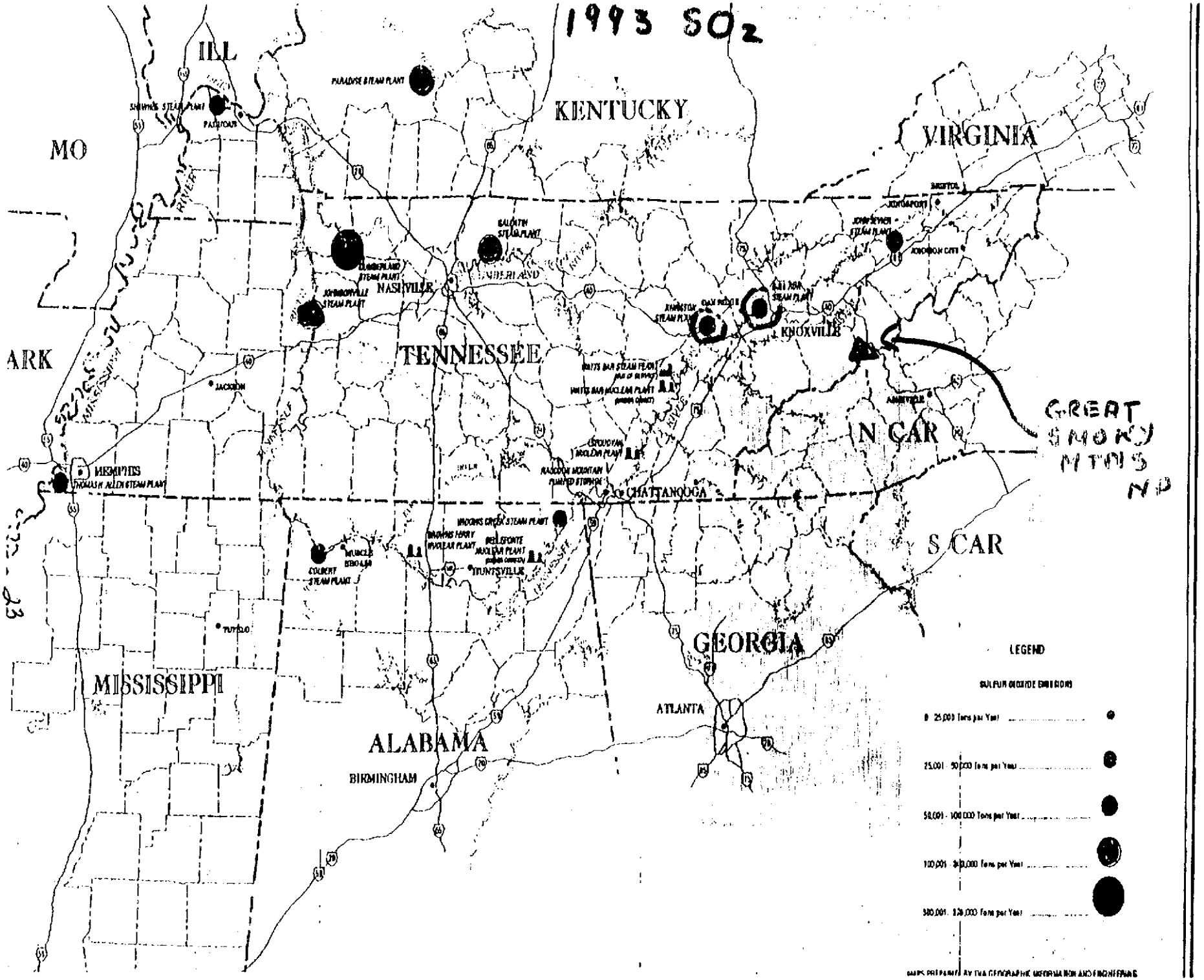

(a) Shenandoah Nat'l Park-Big Meadows (VA28)
4
3
2
1
1982
1986
1990
1994
(b) Great Smoky Mts Nat'l Park-Elkmont (TN11)
3.5
3.0
2.5
2.0
1.5
1.0
0.5
1982
1986
1990
1994
(c) Grand Canyon Nat'l Park (AZ03)
1.5
1.0
0.5
1982
1986
1990
1994
(d) Yosemile National Park (CA99)
3
2
1
0
1982
1986
1990
1994

ACID RAIN



Increase in SO_2 Emissions (percent per decade)
Fraction of US total in 1990

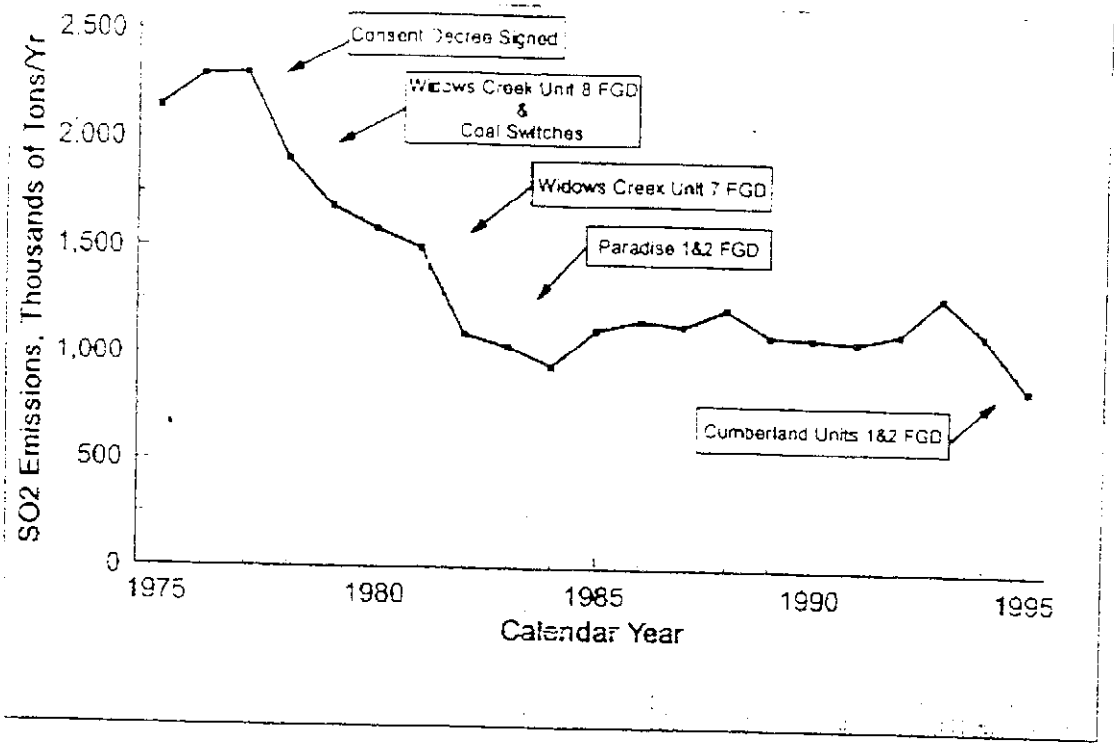

Annual SO2 Emissions
(normalized to 1985)
1.10
1.08
1.06
1.04
1.02
1.00
0.98
0.96
0.94
0.92
0.90
1985
1986
1987
1988
1989
1990
1991
1992
1993
1994
year
Region 3 (East)
Region 4 (Southeast)
all US


1993 SO2
ILL
MO
KENTUCKY
VIRGINIA
ARK
TENNESSEE
NASHVILLE
KNOXVILLE
MEMPHIS
CHATTANOOGA
N CAR
S CAR
GEORGIA
MISSISSIPPI
ALABAMA
BIRMINGHAM
ATLANTA
GREAT SMOKY MTNS NP
LEGEND
SULFUR DIOXIDE EMISSIONS
0 - 25,000 Tons per Year
25,001 - 50,000 Tons per Year
50,001 - 100,000 Tons per Year
100,001 - 300,000 Tons per Year
300,001 - 570,000 Tons per Year

TOTAL P.05

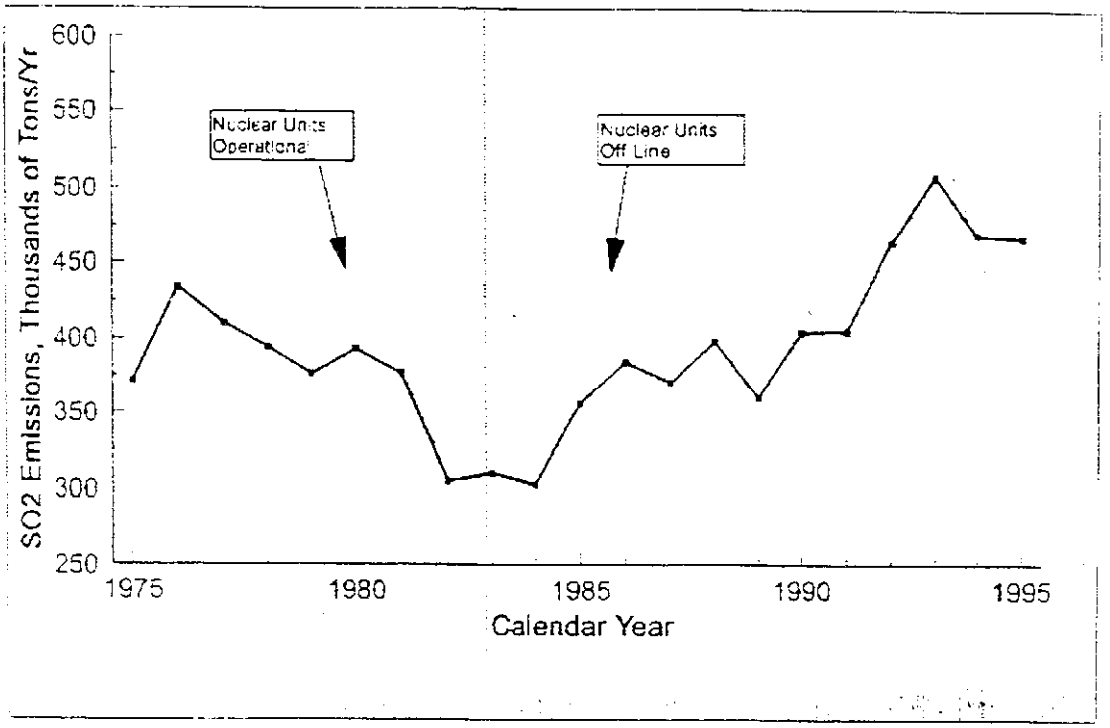
TITLE IV PROGRAMS

TVA's Historical SO2 Emissions

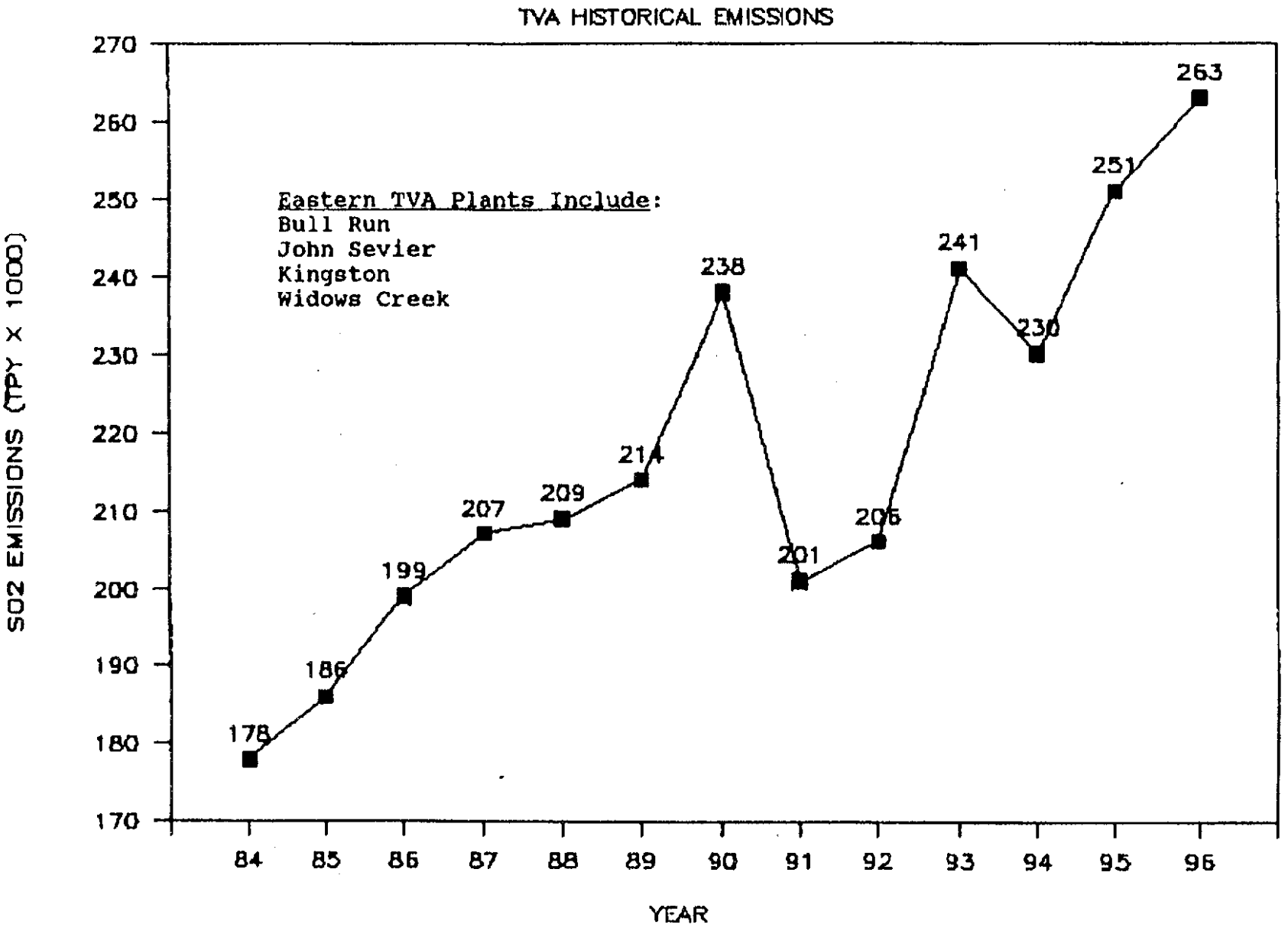


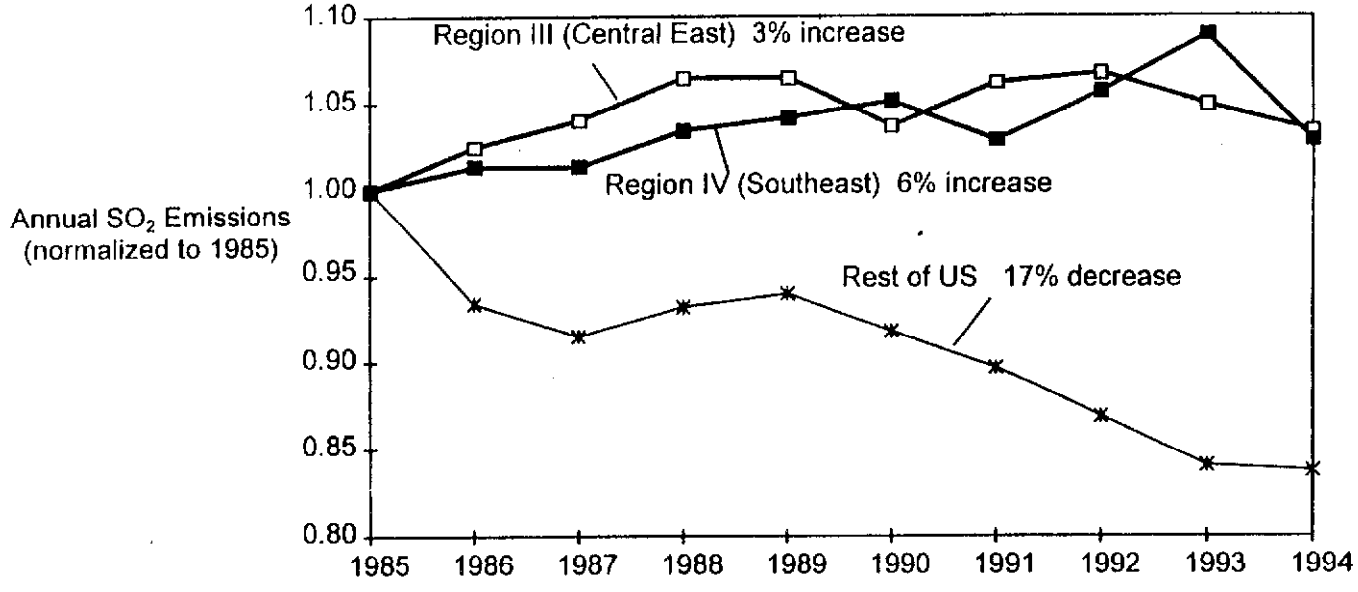
TITLE IV PROGRAMS

TVA's Historical NOx Emissions



TVA SO2 EMISSIONS (EASTERN PLANTS)




Region III (Central East) 3% increase
Region IV (Southeast) 6% increase
Rest of US 17% decrease
Annual SO_2 Emissions
(normalized to 1985)
1.10
1.05
1.00
0.95
0.90
0.85
0.80
1985
1986
1987
1988
1989
1990
1991
1992
1993
1994

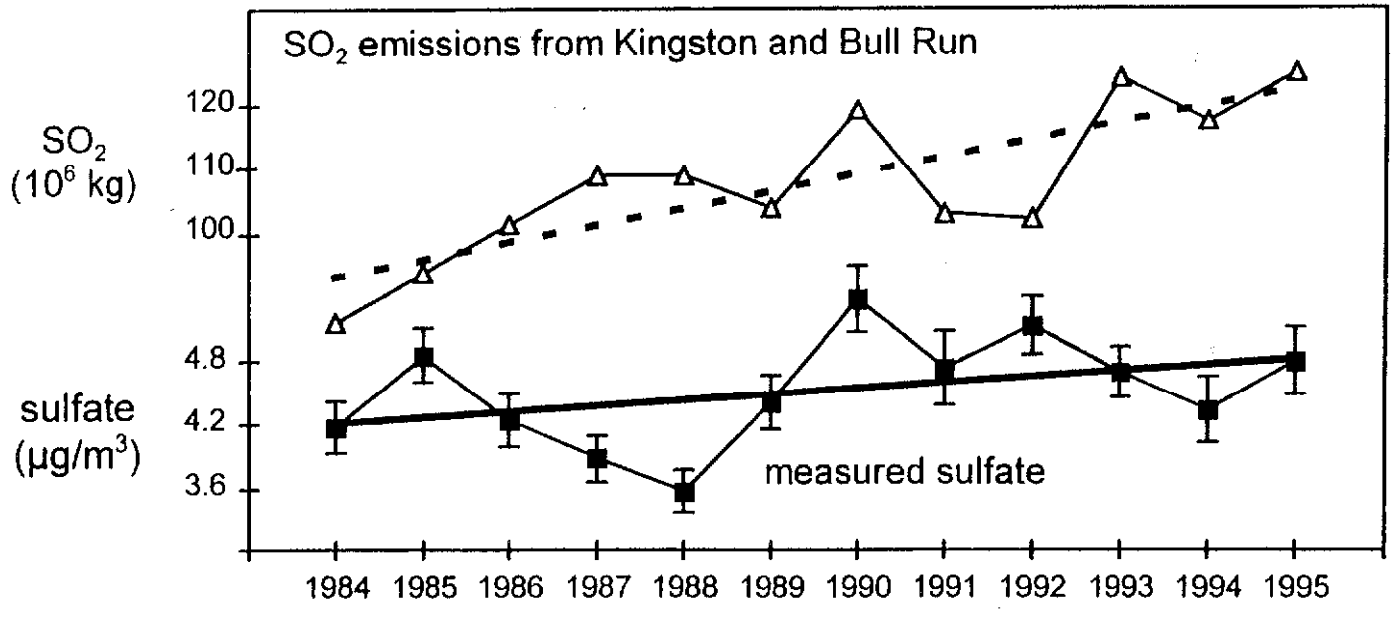

SO_2 emissions from Kingston and Bull Run
SO_2
(10^6 kg)
120
110
100
sulfate
($\mu g/m^3$)
4.8
4.2
3.6
measured sulfate
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
1994
1995

Table 1. Trends in SO_2 emissions from Kingston Steam Plant and measured sulfate at Great Smoky Mountains National Park from 1985 to 1994. The values indicate the percentage per decade.

	summer	fall	winter	spring
SO_2 emissions	+26%	+18%	+6%	-1%
measured sulfate	+26%	+20%	-7%	-6%

Southeastern Aerosol and Visibility Study

EPRI

To provide basic scientific information that will serve air quality management in the eastern United States

Background/ Motivation

Microscopic atmospheric particles with diameter less than 2.5 microns (PM-2.5) contribute to haze formation. They may also play a role in acute and chronic respiratory disease; exposure to trace substances ("air toxics"); transfer of pollutants to aquifers, vegetation, and soils; and climate modulation. The U.S. Environmental Protection Agency is currently reassessing the existing National Ambient Air Quality Standard (NAAQS) for particulate matter (PM) and its policies for regional haze. Thus a need exists for reliable information about the relationship between source emissions and particle characteristics and their potential environmental consequences.

Southeastern Aerosol and Visibility Study (SEAVS) is a partnership composed of electric utilities, National Park Service (NPS), Electric Power Research Institute (EPRI), universities, and consulting firms. Its purpose is to fill crucial gaps in knowledge of atmospheric fine particle characteristics and visibility under humid conditions typical of the southeastern United States. The study will produce instruments, data, and simulation methods that will improve our ability to predict the changes in airborne particle characteristics and in visibility that result from changes in emissions

Photo courtesy of Susanne Hering

The Great Smoky Mountains National Park is the measurement site chosen for the Southeastern Aerosol and Visibility Study.

The Study

SEAVS's goals are to determine the dynamics and composition of suspended particles (aerosols) in the southeastern United States and produce tested and reliable models to simulate the formation of these aerosols and their optical properties. Over a 2 ½-year period it will use the following steps to accomplish these goals:

Step 1

Develop reliable instrumentation to measure aerosol properties. (completed)

- Technologies previously used in the southwestern United States have been modified for application in humid eastern conditions

Step 2

Conduct 6 weeks of synchronized measurements in the Great Smoky Mountains National Park. (set to begin July 1995)

- Field test the instruments.
- Measure specific properties of PM-2.5 — with emphasis on the contribution of water and organics (carbon-containing compounds) to aerosols, trace substance concentrations, and human perceptions of visibility.

Step 3

Characterize air quality and test simulation methods.

- Synthesize the observations to present an integrated picture of the composition of the atmospheric aerosol (particle and gas) and the

Summary of SEAVS Aerosol Results

Tom. Cahill, UC Davis

Results from the IMPROVE network

- **Summer, 1995, was a typical summer**

  Average values for all major and most minor aerosol components were within a few percent of average values, 1988-1994

  Comparison, summer, 1995 (SEAVS) to previous summers:

	Summer, 1995	All summers, 1988-1994
Fine Mass	20.8 ± 10.6	21.4 ± 10.2
Sulfate (from SO4)	8.5 ± 6.0	8.5 ± 4.0
Soil	1.25 ± 2.0	1.23 ± 2.5
Organic matter (from C)	4.0 ±1.3	4.0 ± 0.8

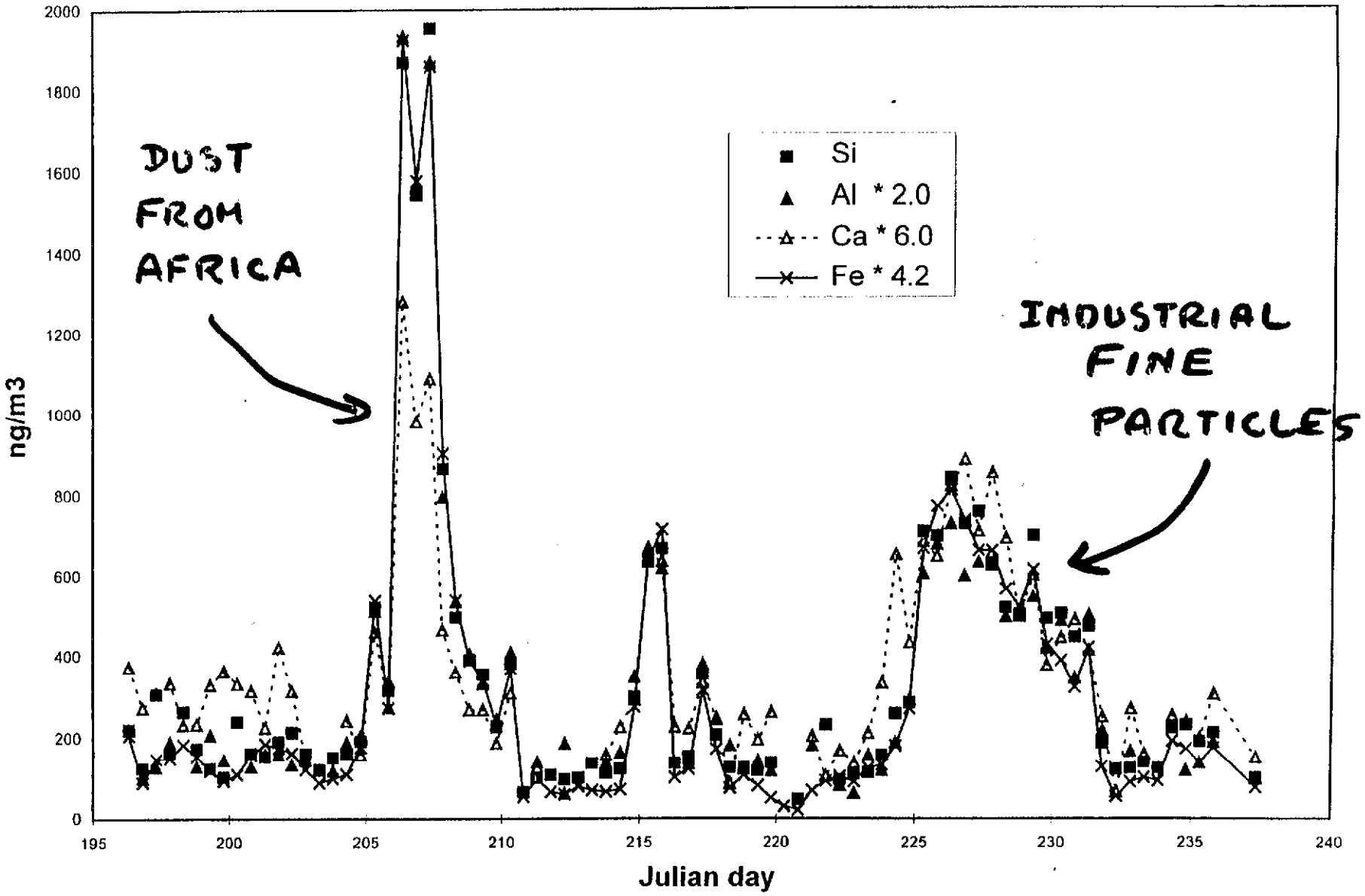
- **Variability in Time**

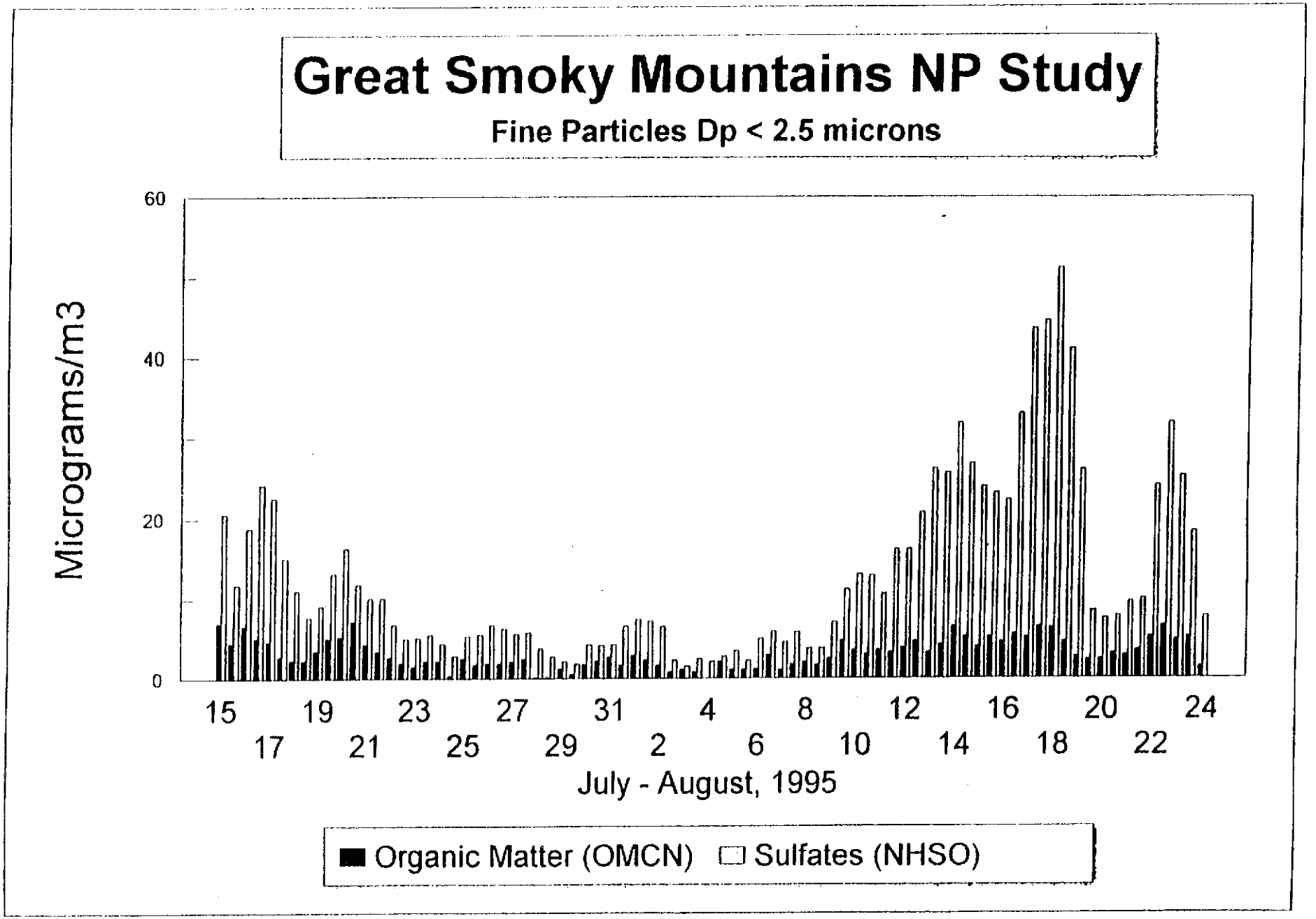
  Sulfates and soils had high variability during the summer, organics and nitrates much less so

- **Variability in Space**

  Periods of high sulfate concentration were concentrated into a region no more than a few hundred miles across.

  Peaks of high organic concentrations were much more regional over the eastern US

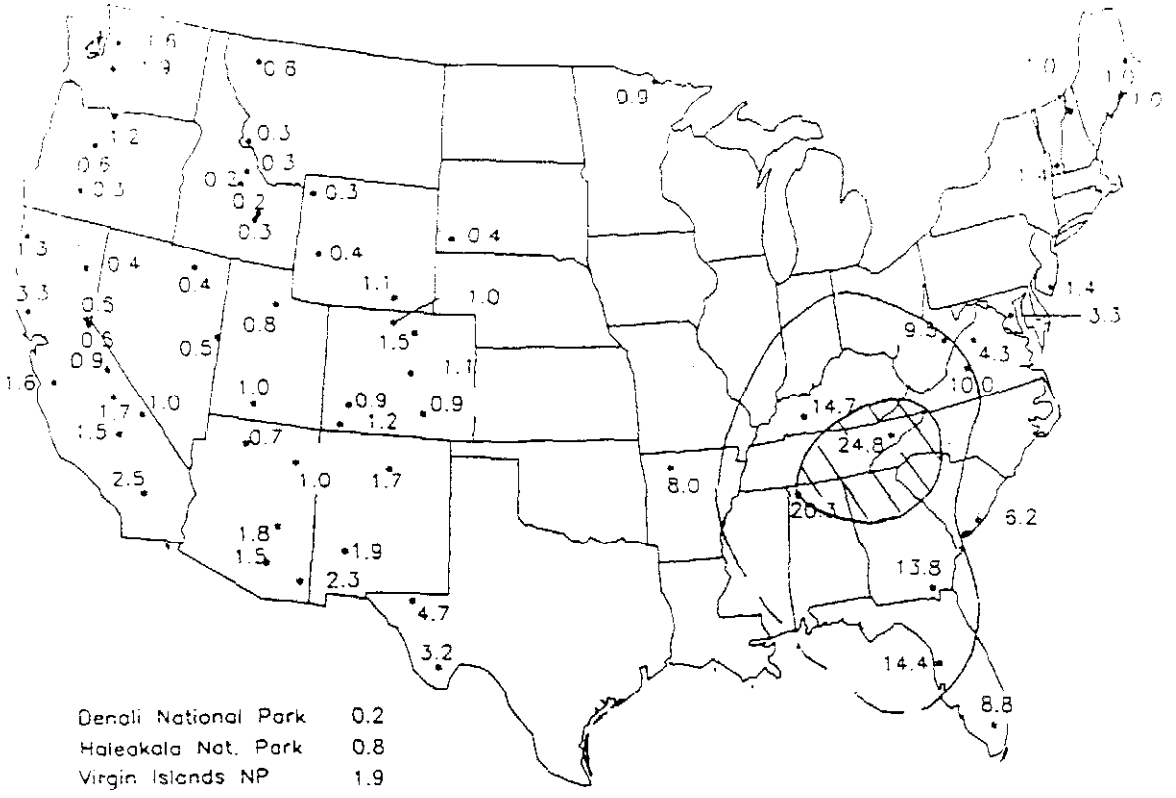

DUST FROM AFRICA
INDUSTRIAL FINE PARTICLES
Si
Al * 2.0
Ca * 6.0
Fe * 4.2
ng/m3
Julian day
0
200
400
600
800
1000
1200
1400
1600
1800
2000
195
200
205
210
215
220
225
230
235
240


Great Smoky Mountains NP Study
Fine Particles Dp < 2.5 microns
Micrograms/m3
60
40
20
0
15
17
19
21
23
25
27
29
31
2
4
6
8
10
12
14
16
18
20
22
24
July - August, 1995
Organic Matter (OMCN)
Sulfates (NHSO)

AMMONIUM SULFATE

Concentrations in $\mu g/m^3$

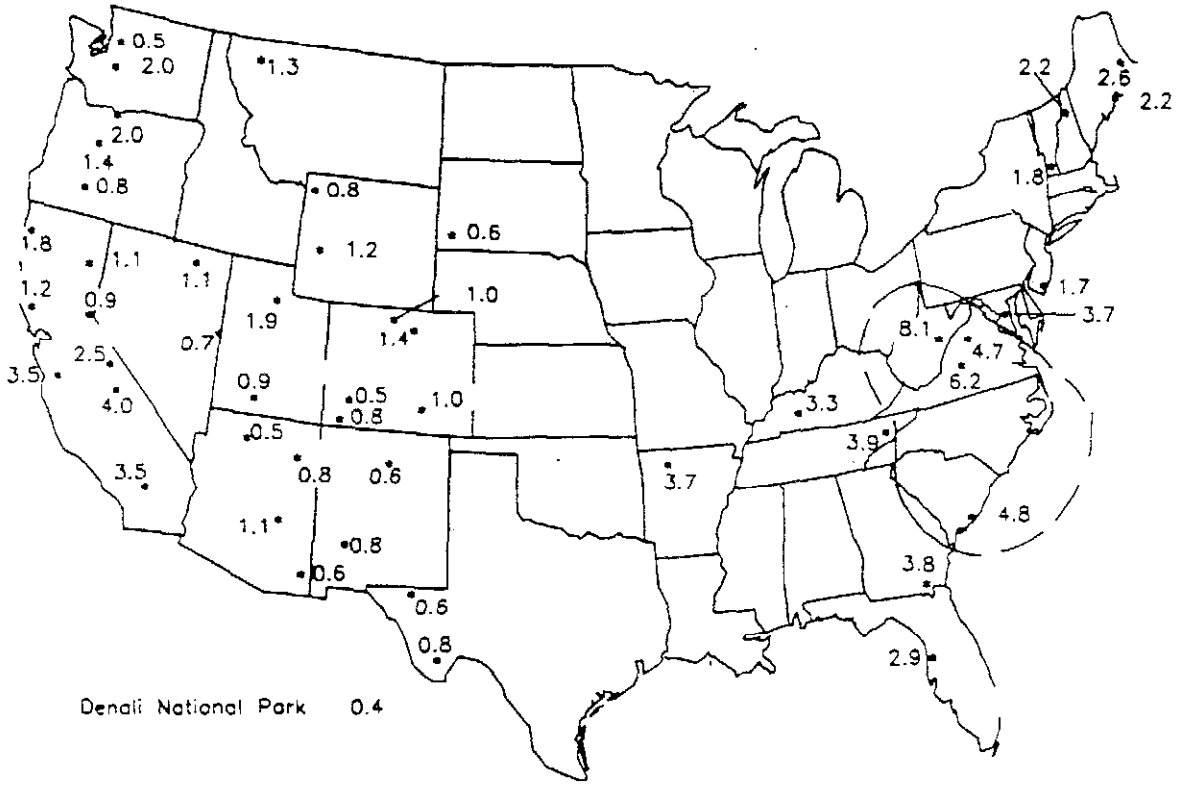
Contours are based on data shown

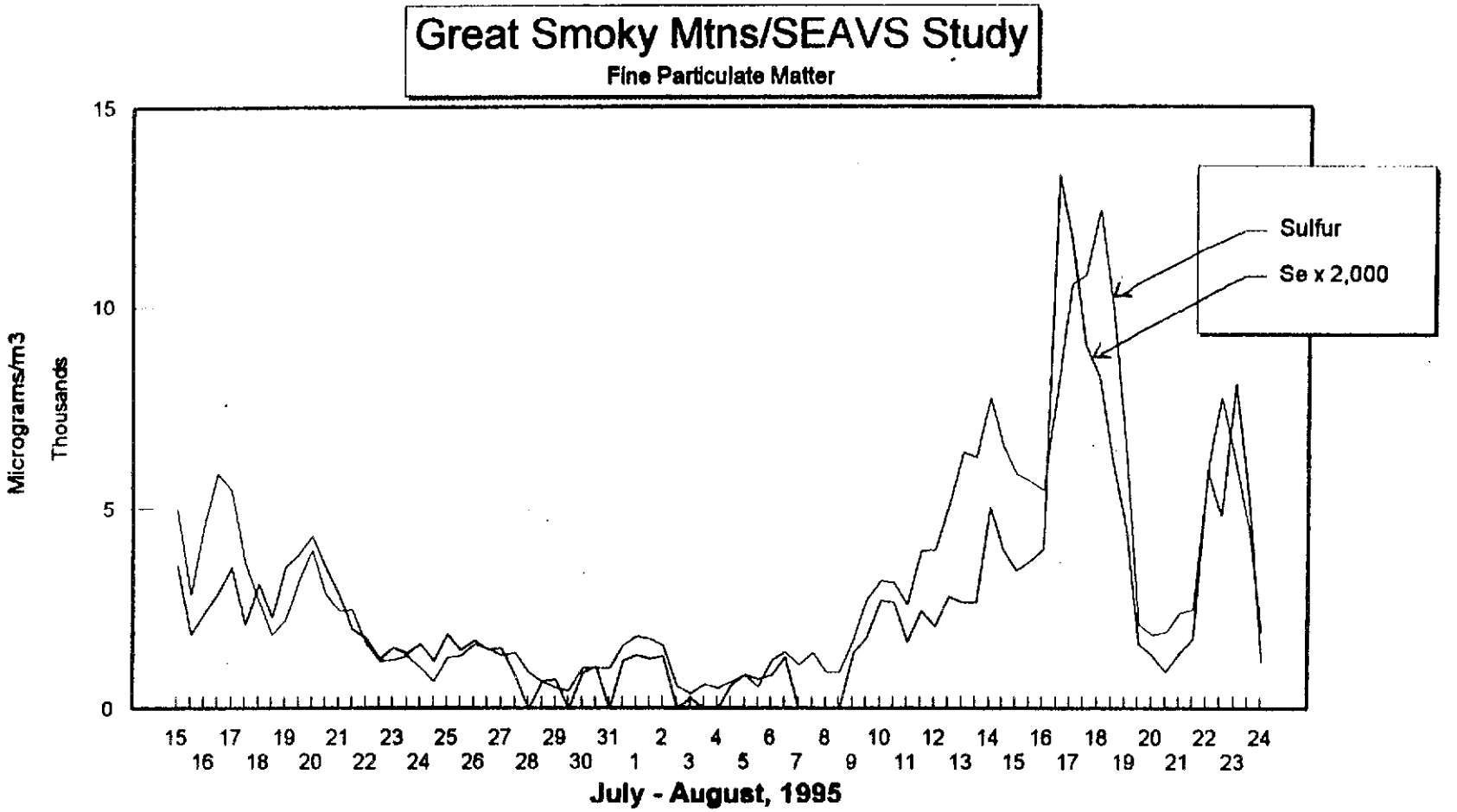


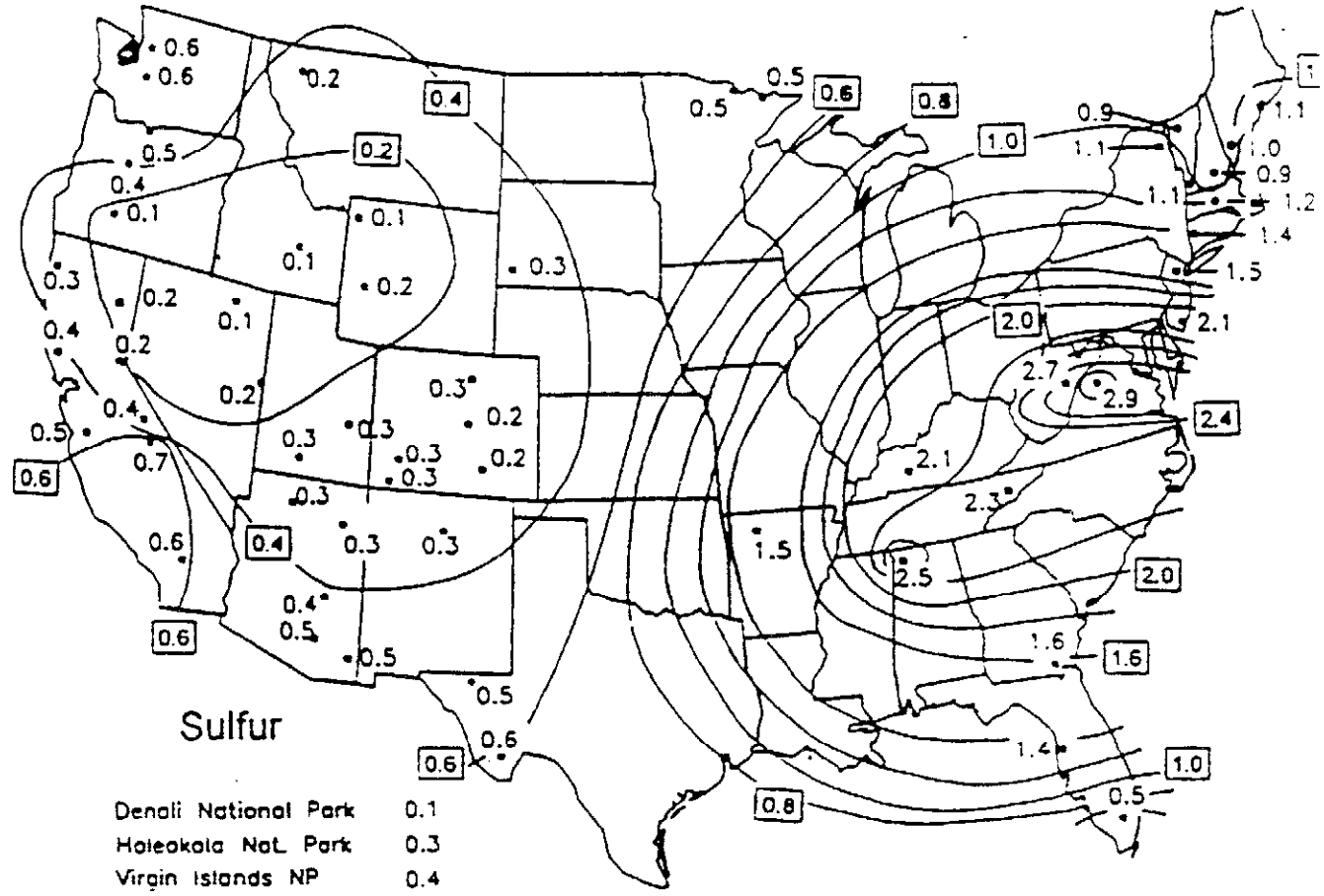
ORGANIC MASS FROM CARBON

Concentrations in $\mu g/m^3$

Contours are based on data shown.




Great Smoky Mtns/SEAVS Study
Fine Particulate Matter
Micrograms/m3
Thousands
15
10
5
0
Sulfur
Se x 2,000
15 17 19 21 23 25 27 29 31 2 4 6 8 10 12 14 16 18 20 22 24
16 18 20 22 24 26 28 30 1 3 5 7 9 11 13 15 17 19 21 23
July - August, 1995



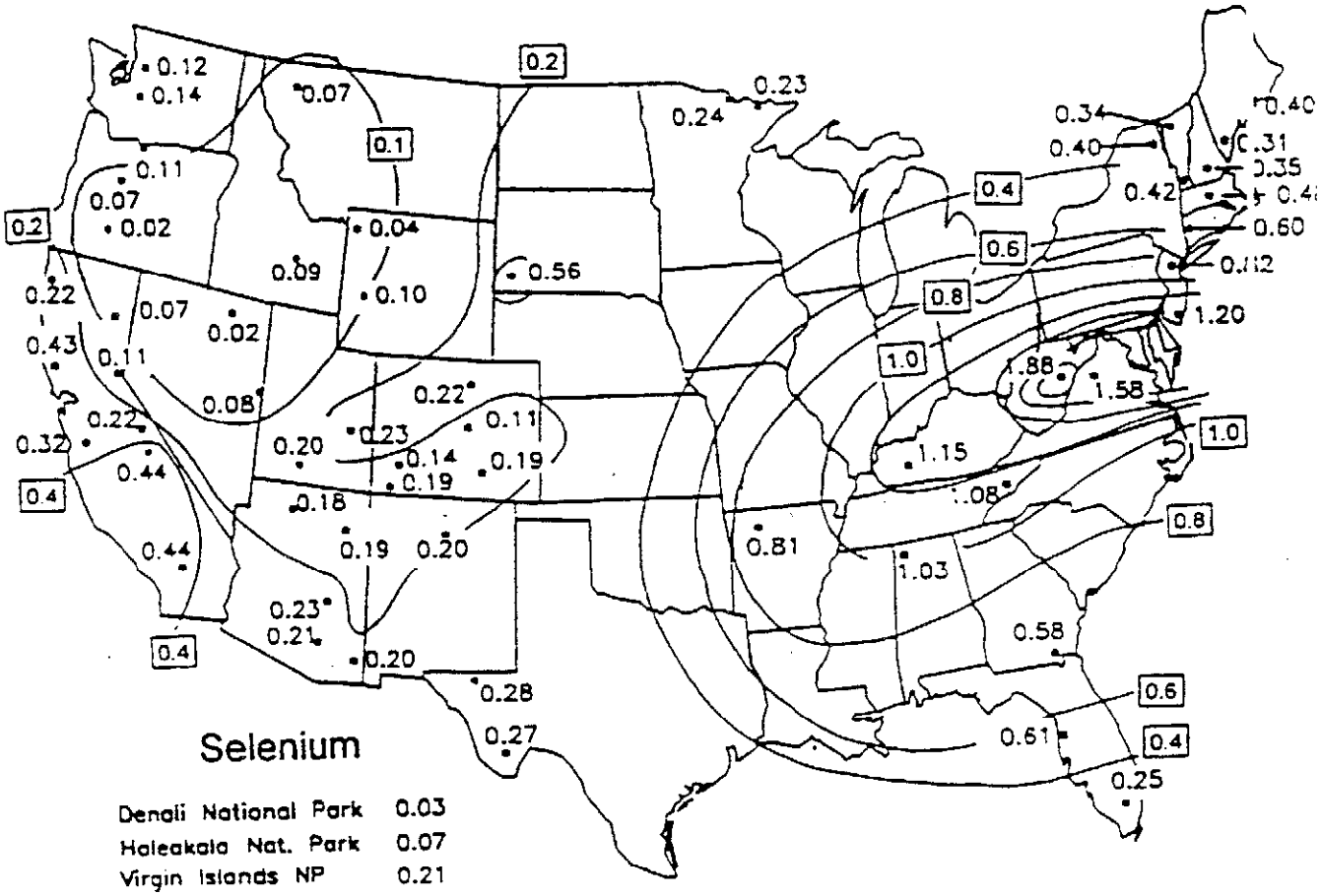
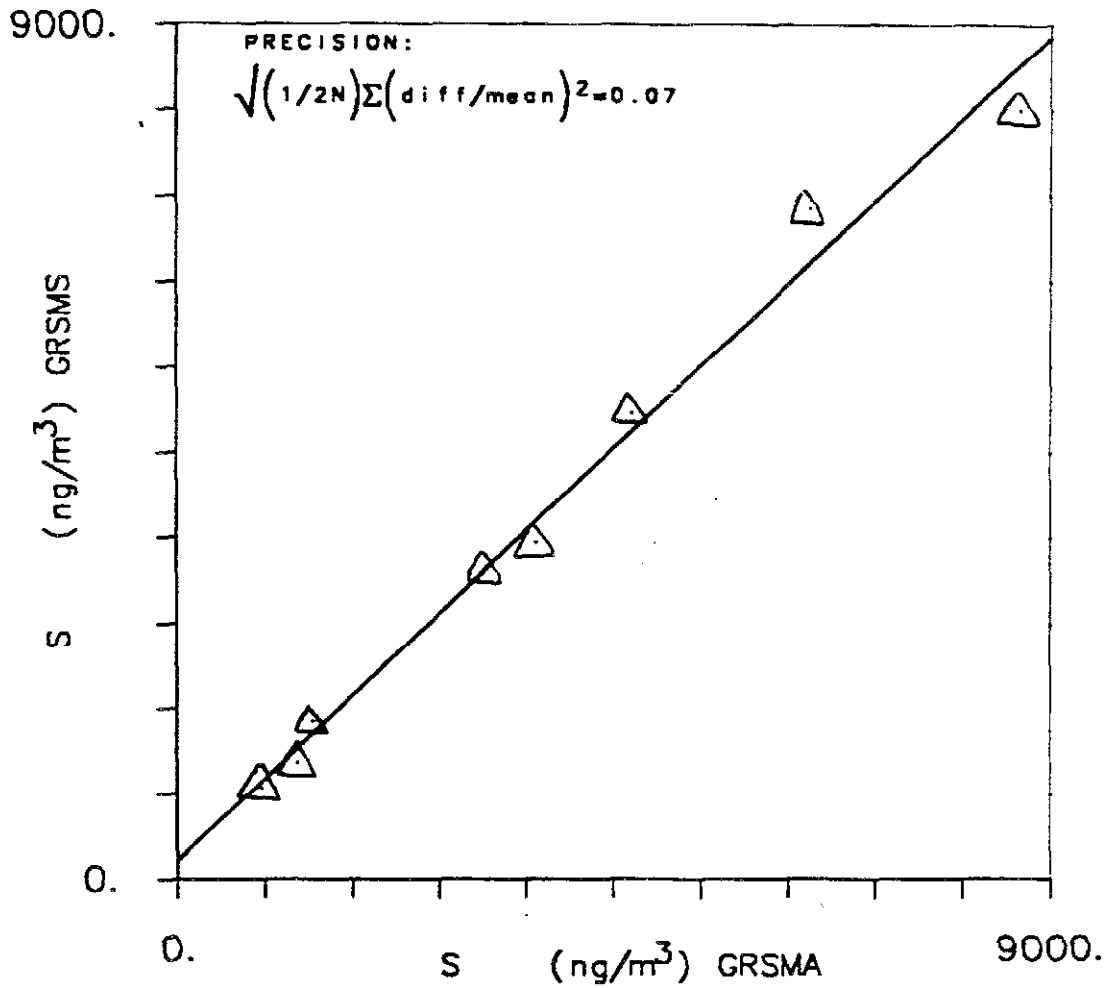
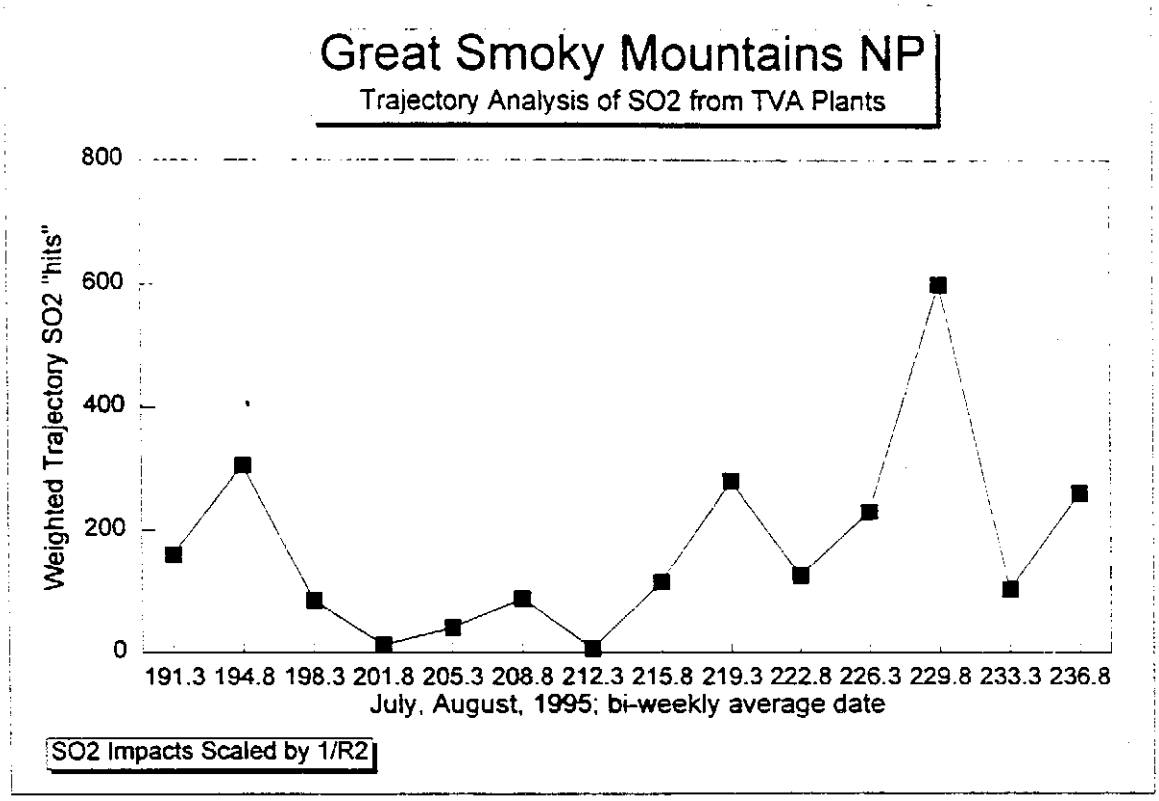


Figure 2. Contour maps of mean sulfur in µg/m³ and mean selenium in ng/m³ for summer 1993 (Jun
August).

Comparison of Stacked Filter Units (1982-1987) with IMPROVE cyclones (1988-present)

Great Smoky Mountains NP Study, Summer, 1995





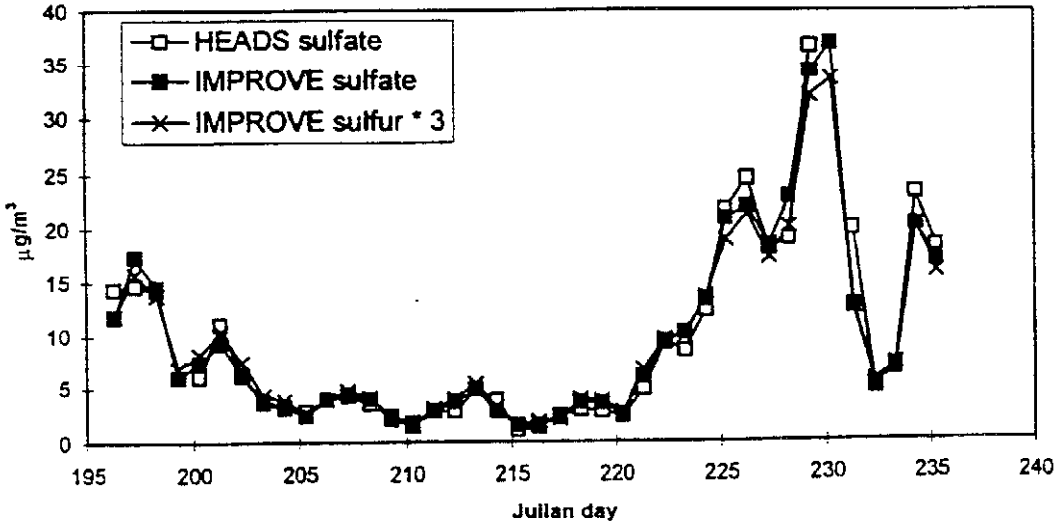


FIGURE: COMPARISON OF SO_2 TRAJECTORIES AND AMBIENT SULFATES

Comparison of SO_2 trajectories for TVA sources to great Smoky Mountains National Park, Summer, 1995. The bottom figure is preliminary sulfate data for the National Park Service/SEAVS for the same period.

Trends Analysis of Sulfates and Nitrates

Analysis:

A $1/r^2$ weighted residence time analysis accurately ties emissions to observed sulfate concentrations at Great Smoky Mountains NP.

Reasoning:

Mean summer sulfate levels are dominated by the number of high sulfate events that occur, not the median concentration.

High sulfate events occur in stagnation periods, when source emissions are injected into limit volumes of air.

Summer stagnation periods are characterized by low wind speeds and high temperatures, maximizing SO_2 to sulfate conversion in local areas.

Conclusion:

Therefore, weak, local sources are much more important than strong, remote sources during high sulfate events.

Corollary:

If a utility such as TVA wishes to improve air quality at Great Smoky Mountains NP, it should upgrade the old (and increasingly ineffective ?) SO_2 scrubbers on the two nearest coal fired power plants, Kingston and Bull Run.

Trends Analysis of Sulfates and Nitrates

Example:

Road kill on the $PM_{2.5}$ highway?

Pittsburgh averages 21.5 $\mu g/m^3$ $PM_{2.5}$ annual average fine mass.

13 $\mu g/m^3$. - upwind Sites

The new $PM_{2.5}$ FPM standard is 15 $\mu g/m^3$.

Analysis:

Therefore,

a) **Pittsburgh would have to reduce local emissions by 76% to achieve the FPM standard,**
b) **Pittsburgh must do chemical speciation and CMB analysis to distinguish its (controllable) sources from regional background,**
c) **Pittsburgh can not do such planning without knowledge of future levels of upwind FPM values.**