

CHAPTER 3

AEROSOL MASS BUDGETS AND SPATIAL DISTRIBUTIONS

This chapter discusses the observed spatial and temporal variations in aerosol concentration and chemical composition throughout the United States on the basis of the IMPROVE measurements [Sisler *et al.*, 1993] for the three-year period, March 1992 through February 1995.

Aerosol concentrations and chemical composition vary because of a number of factors, including the spatial distribution of natural and anthropogenic emission sources and meteorological conditions. The highest aerosol concentrations tend to occur in significant urban or industrialized areas where emission densities are high. Also, concentrations are highest when atmospheric dilution is minimal such as what occurs in stagnation periods or periods of limited mixing. In addition, since sulfate and nitrate aerosols are formed from SO₂ and NO_x emissions and chemical reactions in the atmosphere, these aerosols are highest when photochemistry is strongest.

For example, concentrations of sulfates tend to be highest in areas of significant sulfur dioxide (SO₂) emissions such as the eastern United States where SO₂ is emitted from coal-fired stationary sources, and in the Southwest due to copper smelters, power plants, and SO₂ emissions from Mexico. Organic carbon concentrations tend to be highest in regions such as the Pacific Northwest and Southeast due in part to forests and forest-product industries, which cause organics to dominate fine aerosol mass in the Pacific Northwest. Nitrates tend to be most prevalent in California where both NO_x emissions from motor vehicles and industry are high.

Spatial and temporal variations in aerosol composition and concentrations can be qualitatively examined through the use of annual and seasonal mass budgets. Mass budgets are the contribution of individual aerosol species to the reconstructed fine particle mass [Sisler *et al.*, 1993]. Mass budgets are calculated by dividing the average concentration of each species by the average reconstructed fine particle mass for each region and time period of interest.

In this chapter, the observed spatial and seasonal trends in aerosol concentrations and chemical composition from the three-year period, March 1992 through February 1995, of the IMPROVE network are presented. There are 58 sites in the IMPROVE network that are fully instrumented for aerosol monitoring (channels A-D). Only 43 sites with data for this three-year period are summarized in this report. Since the last IMPROVE report [Sisler *et al.*, 1993] five sites have been downgraded to channel A only or were discontinued and are not summarized here. The downgraded sites are at Everglades and Voyageurs National Parks. The discontinued sites are Arches, Isle Royale, and Hawaii Volcanoes National Parks.

The 43 IMPROVE sites are grouped into 21 regions according to their relative location, climatology, similarities in concentrations, and seasonal trends. Since the last IMPROVE report, three new regions have been introduced, the Mid-South, Mid-Atlantic, and Lake Tahoe region, while one region was dropped, Hawaii. Average concentrations and chemical composition are calculated on the basis of measurements for each region. Tables 3.1 and 3.2 show the mass concentrations of fine and coarse aerosol and the chemical composition (mass budgets) of the fine aerosol for each of the 21 regions in the United States. These concentrations and mass budgets are averaged over the entire three-year period to provide the annual average and over the three years for each of the four seasonal averages.

First, the characteristics of each of the regions (in alphabetic order) are discussed, followed by the spatial and temporal trends of the fine and coarse mass concentrations and the constituents of the fine-particle mass.

3.1 Characteristics of the Regions

Alaska. The Alaska region has only one monitoring site, Denali National Park. The average concentrations of fine and coarse aerosols over the three-year period were 1.8 and 3.3 $\mu\text{g}/\text{m}^3$, respectively. The fine aerosol concentration was the lowest measured anywhere in the United States during this period. Both fine and coarse aerosol concentrations are largest in summer and smallest in autumn. Organics are the largest contributor of fine particle mass (52%), followed by sulfate (28.6%), soil (10.2%), light-absorbing carbon (4.6%), and nitrate (3.3%). The concentrations of organics and light-absorbing carbon are largest in summer, perhaps due to the prescribed burning and forest fires that usually occur during that season.

Appalachian Mountains. This region has five sites of which three are reported here: Great Smoky Mountains and Shenandoah National Parks, both initiated in March 1988, and Dolly Sods Wilderness Area in West Virginia, initiated in September 1991. The other two sites, Shinning Rock in North Carolina and James River Face in Virginia have less than one year of data.

The average concentrations of fine and coarse aerosol for this region were 11.3 $\mu\text{g}/\text{m}^3$ and 4.8 $\mu\text{g}/\text{m}^3$, respectively. Both fine and coarse aerosol concentrations are maximum in summer and minimum in winter. Sulfate is by far the largest component of the fine particle mass. At 59.9%, it is more than twice that of the next largest contributor, organics (26%). Other contributors include nitrate (5.5%), soil (4.7%), and light-absorbing carbon (3.7%). Except for nitrate and light-absorbing carbon, which have their maximum concentrations in the winter and autumn, respectively, all other species have maximum concentrations in summer. The seasonal variation in sulfate concentrations is particularly strong with summer concentrations more than three times the winter concentrations at 11.2 $\mu\text{g}/\text{m}^3$.

Table 3.1. Measured fine and coarse aerosol concentration (in micro-g/m³) for the 21 regions in the IMPROVE network.

Season	Fine Mass	Sulfate	Nitrate	Organics	Elemental Carbon	Soil	Coarse Mass
Alaska							
Spring	2.0	0.8	0.1	0.8	0.1	0.3	2.8
Summer	2.4	0.5	0.0	1.6	0.1	0.2	2.9
Autumn	1.4	0.4	0.0	0.7	0.1	0.1	1.9
Winter	1.5	0.5	0.1	0.7	0.1	0.1	1.7
ANNUAL	1.8	0.5	0.1	1.0	0.1	0.2	2.3
Appalachian Mountains							
Spring	10.3	6.0	0.8	2.7	0.4	0.5	5.5
Summer	16.8	11.2	0.3	3.7	0.4	1.2	6.3
Autumn	10.6	6.3	0.5	3.0	0.5	0.3	4.3
Winter	7.1	3.2	0.9	2.3	0.4	0.2	3.3
ANNUAL	11.3	6.7	0.6	2.9	0.4	0.6	4.8
Boundary Waters							
Spring	5.4	2.8	0.5	1.5	0.2	0.4	3.4
Summer	5.2	1.9	0.1	2.8	0.2	0.2	3.8
Autumn	4.3	1.7	0.5	1.7	0.2	0.2	3.0
Winter	5.5	2.1	1.4	1.5	0.2	0.2	2.6
ANNUAL	5.1	2.1	0.6	1.9	0.2	0.3	3.2
Cascade Mountains							
Spring	5.6	1.6	0.3	3.0	0.4	0.3	4.0
Summer	6.3	2.4	0.4	2.9	0.4	0.3	3.7
Autumn	5.3	1.3	0.2	3.2	0.5	0.2	3.9
Winter	3.5	0.5	0.1	2.3	0.4	0.1	2.5
ANNUAL	5.2	1.5	0.2	2.8	0.4	0.2	3.5
Central Rocky Mountains							
Spring	3.3	0.9	0.2	1.1	0.1	0.9	5.0
Summer	4.0	1.0	0.2	1.8	0.2	0.8	5.7
Autumn	3.1	0.8	0.2	1.4	0.2	0.5	3.9
Winter	2.1	0.6	0.2	1.0	0.1	0.2	2.8
ANNUAL	3.1	0.8	0.2	1.3	0.2	0.6	4.4

Table 3.1 Continued

Season	Fine Mass	Sulfate	Nitrate	Organics	Elemental Carbon	Soil	Coarse Mass
Colorado Plateau							
Spring	3.5	1.1	0.2	1.2	0.1	0.9	4.2
Summer	3.9	1.3	0.2	1.5	0.2	0.7	5.3
Autumn	3.4	1.1	0.2	1.5	0.2	0.4	4.0
Winter	2.6	0.8	0.2	1.1	0.2	0.2	3.0
ANNUAL	3.3	1.1	0.2	1.3	0.2	0.6	4.1
Florida							
Spring	11.0	6.0	0.6	3.3	0.6	0.6	6.8
Summer	11.8	4.9	0.5	3.0	0.4	3.1	10.1
Autumn	8.8	4.5	0.4	2.9	0.6	0.4	6.4
Winter	8.9	3.9	0.6	3.3	0.8	0.2	6.3
ANNUAL	10.1	4.8	0.5	3.1	0.6	1.1	7.4
Great Basin							
Spring	3.2	0.7	0.2	1.2	0.1	1.0	5.4
Summer	4.2	0.8	0.2	1.7	0.2	1.4	7.1
Autumn	3.0	0.6	0.1	1.5	0.2	0.6	4.6
Winter	2.1	0.4	0.2	1.1	0.2	0.2	2.6
ANNUAL	3.1	0.7	0.2	1.4	0.2	0.8	4.9
Lake Tahoe							
Spring	5.7	0.9	0.4	2.6	0.6	1.1	6.8
Summer	5.4	1.1	0.3	2.6	0.6	0.8	5.4
Autumn	6.8	0.8	0.4	3.9	1.1	0.6	5.2
Winter	8.0	0.4	0.5	5.0	1.5	0.6	9.2
ANNUAL	6.4	0.8	0.4	3.5	0.9	0.8	6.6
Mid Atlantic							
Spring	10.1	5.6	1.2	2.4	0.5	0.4	10.1
Summer	13.9	8.3	0.7	3.5	0.6	0.9	11.4
Autumn	10.5	5.3	1.1	3.1	0.7	0.4	8.6
Winter	11.1	4.5	2.2	3.3	0.8	0.3	7.2
ANNUAL	11.4	5.9	1.3	3.1	0.6	0.5	9.1

Table 3.1 Continued

Season	Fine Mass	Sulfate	Nitrate	Organics	Elemental Carbon	Soil	Coarse Mass
Mid South							
Spring	11.8	6.3	1.2	3.3	0.5	0.6	5.3
Summer	15.3	8.3	0.4	3.9	0.5	2.2	8.9
Autumn	11.0	5.8	0.7	3.6	0.6	0.4	5.6
Winter	10.3	4.5	1.8	3.1	0.6	0.3	4.5
ANNUAL	12.1	6.2	1.0	3.5	0.5	0.9	6.0
Northeast							
Spring	5.7	3.0	0.4	1.6	0.3	0.3	4.4
Summer	8.4	4.8	0.3	2.6	0.3	0.3	4.5
Autumn	5.7	3.0	0.4	1.8	0.3	0.2	4.0
Winter	5.6	2.5	0.7	1.9	0.4	0.2	4.0
ANNUAL	6.4	3.4	0.5	2.0	0.3	0.2	4.2
Northern Great Plains							
Spring	4.7	1.9	0.6	1.3	0.2	0.7	5.6
Summer	4.5	1.7	0.2	2.0	0.2	0.5	5.6
Autumn	4.2	1.3	0.5	1.7	0.2	0.5	5.5
Winter	4.7	1.7	1.2	1.3	0.2	0.2	3.1
ANNUAL	4.5	1.7	0.6	1.6	0.2	0.5	5.0
Northern Rocky Mountains							
Spring	4.7	1.0	0.2	2.6	0.3	0.6	5.0
Summer	5.2	1.0	0.1	3.0	0.3	0.8	8.2
Autumn	7.4	1.0	0.3	4.7	0.6	0.7	6.9
Winter	5.3	1.1	0.6	2.9	0.5	0.3	2.8
ANNUAL	5.7	1.0	0.3	3.3	0.4	0.6	5.8
Pacific Coast							
Spring	4.2	1.4	0.7	1.5	0.2	0.3	9.3
Summer	4.2	1.8	0.7	1.4	0.2	0.3	8.9
Autumn	5.4	1.4	0.8	2.5	0.4	0.4	8.2
Winter	4.7	0.8	1.6	1.9	0.3	0.1	6.0
ANNUAL	4.6	1.3	0.9	1.8	0.2	0.3	8.2

Table 3.1 Continued

Season	Fine Mass	Sulfate	Nitrate	Organics	Elemental Carbon	Soil	Coarse Mass
Sierra-Humboldt							
Spring	3.1	0.7	0.3	1.4	0.2	0.7	3.7
Summer	3.8	0.8	0.2	2.1	0.3	0.6	4.1
Autumn	3.3	0.6	0.2	1.8	0.2	0.4	3.0
Winter	1.9	0.3	0.2	1.0	0.3	0.1	1.8
ANNUAL	3.1	0.6	0.2	1.6	0.2	0.5	3.2
Sierra Nevada							
Spring	4.6	1.2	0.6	1.9	0.2	0.7	5.3
Summer	6.8	1.6	0.5	3.7	0.4	0.7	6.2
Autumn	4.9	1.0	0.5	2.5	0.3	0.6	5.1
Winter	1.9	0.3	0.2	1.1	0.1	0.1	3.3
ANNUAL	4.5	1.0	0.4	2.3	0.2	0.5	5.0
Sonoran Desert							
Spring	4.6	1.5	0.3	1.5	0.2	1.1	6.4
Summer	5.0	1.9	0.2	1.7	0.2	1.0	6.6
Autumn	4.3	1.6	0.2	1.7	0.3	0.5	4.9
Winter	3.1	1.1	0.2	1.3	0.2	0.3	4.0
ANNUAL	4.3	1.5	0.2	1.6	0.2	0.7	5.5
Southern California							
Spring	12.5	1.8	6.1	3.2	0.5	0.9	8.9
Summer	12.0	2.4	4.2	4.0	0.6	0.7	11.1
Autumn	7.5	1.3	2.5	2.2	0.4	1.1	11.8
Winter	3.4	0.6	1.3	1.1	0.2	0.2	2.9
ANNUAL	9.0	1.5	3.7	2.6	0.4	0.7	8.4
Washington D.C.							
Spring	17.1	8.0	2.6	4.2	1.4	0.9	7.6
Summer	23.0	13.9	1.3	5.1	1.4	1.3	6.9
Autumn	18.6	7.9	2.4	5.4	2.0	0.9	7.4
Winter	18.4	5.9	3.9	5.9	1.9	0.8	7.8
ANNUAL	19.2	9.0	2.5	5.2	1.7	1.0	7.4
West Texas							
Spring	5.3	2.1	0.2	1.7	0.2	1.1	7.6
Summer	7.0	2.7	0.3	1.8	0.2	2.0	7.8
Autumn	4.6	2.0	0.2	1.4	0.2	0.7	6.8
Winter	3.8	1.6	0.3	1.3	0.2	0.4	5.4
ANNUAL	5.2	2.1	0.2	1.5	0.2	1.1	6.9

Table 3.2 Measured fine aerosol mass budgets (in percent) for the 21 regions in the IMPROVE network.

Season	Sulfate	Nitrate	Organics	Elemental Carbon	Soil
Alaska					
Spring	38.1	3.7	38.0	4.7	15.5
Summer	19.4	1.8	65.8	4.9	8.1
Autumn	26.5	3.1	51.9	7.9	10.7
Winter	34.0	5.6	46.7	7.2	6.5
ANNUAL	28.6	3.3	52.0	5.9	10.2
Appalachian Mountains					
Spring	58.4	7.4	25.9	3.8	4.6
Summer	66.8	1.9	22.0	2.3	7.0
Autumn	58.9	5.0	28.5	4.3	3.2
Winter	46.0	12.6	32.6	5.9	2.9
ANNUAL	59.9	5.5	26.0	3.7	4.9
Boundary Waters					
Spring	51.8	9.1	28.1	3.6	7.5
Summer	36.2	2.2	53.2	3.8	4.6
Autumn	40.2	11.3	38.4	4.7	5.4
Winter	38.9	26.0	27.1	3.9	4.0
ANNUAL	41.8	11.9	37.0	4.0	5.4
Cascade Mountains					
Spring	27.8	5.4	53.1	7.5	6.2
Summer	38.4	5.6	45.6	6.4	4.0
Autumn	24.1	3.6	59.4	9.4	3.6
Winter	15.4	3.8	66.6	10.9	3.3
ANNUAL	28.1	4.7	54.6	8.2	4.4
Central Rocky Mountains					
Spring	27.8	7.3	33.4	3.8	27.7
Summer	24.2	4.1	46.3	4.9	20.5
Autumn	27.1	5.4	45.5	5.8	16.2
Winter	27.6	8.2	47.8	6.3	10.0
ANNUAL	26.5	5.9	42.9	5.1	19.6

Table 3.2 Continued

Season	Sulfate	Nitrate	Organics	Elemental Carbon	Soil
Colorado Plateau					
Spring	30.0	6.2	33.6	4.2	26.0
Summer	34.5	4.8	37.7	4.8	18.1
Autumn	33.1	5.2	43.1	6.0	12.6
Winter	33.1	9.2	42.9	7.2	7.6
ANNUAL	32.6	6.1	39.1	5.4	16.7
Florida					
Spring	54.0	5.5	30.1	5.3	5.1
Summer	41.9	3.9	25.0	3.2	25.9
Autumn	51.0	4.9	33.3	6.5	4.4
Winter	43.8	7.3	37.5	8.8	2.8
ANNUAL	47.4	5.3	30.9	5.7	10.7
Great Basin					
Spring	22.2	5.6	37.3	4.2	30.7
Summer	19.8	3.7	41.1	3.6	31.9
Autumn	20.9	4.0	48.8	5.9	20.4
Winter	20.9	8.1	53.7	8.1	9.2
ANNUAL	21.1	5.0	44.3	5.1	24.5
Lake Tahoe					
Spring	16.3	7.6	46.4	10.1	19.6
Summer	20.6	5.8	48.3	10.2	15.0
Autumn	11.9	6.1	57.3	15.5	9.2
Winter	5.3	6.5	62.6	18.3	7.2
ANNUAL	13.0	6.5	54.3	13.9	12.3
Mid Atlantic					
Spring	55.6	11.8	23.7	4.8	4.1
Summer	59.5	5.3	25.0	4.0	6.2
Autumn	50.0	10.0	29.1	6.8	4.0
Winter	40.5	19.8	29.8	7.1	2.9
ANNUAL	51.8	11.3	26.8	5.6	4.4

Table 3.2 Continued

Season	Sulfate	Nitrate	Organics	Elemental Carbon	Soil
Mid South					
Spring	53.3	9.8	27.6	4.5	4.8
Summer	54.2	2.7	25.2	3.3	14.5
Autumn	52.4	6.1	32.6	5.2	3.7
Winter	43.1	17.9	30.3	5.7	3.0
ANNUAL	51.3	8.4	28.6	4.5	7.2
Northeast					
Spring	53.5	7.8	28.7	4.8	5.3
Summer	57.7	3.6	30.8	4.1	3.8
Autumn	52.1	7.4	31.2	6.1	3.2
Winter	45.0	12.4	33.1	6.4	3.0
ANNUAL	52.9	7.2	30.9	5.2	3.8
Northern Great Plains					
Spring	41.1	12.9	28.4	3.3	14.3
Summer	37.6	3.4	45.0	3.6	10.4
Autumn	31.8	11.3	39.7	4.3	12.8
Winter	37.5	25.2	27.9	4.0	5.4
ANNUAL	37.2	13.3	35.1	3.8	10.7
Northern Rocky Mountains					
Spring	20.9	4.4	55.0	6.7	13.1
Summer	18.7	2.5	58.5	5.8	14.6
Autumn	14.0	4.3	63.7	8.4	9.6
Winter	20.0	11.6	53.9	9.5	5.0
ANNUAL	17.9	5.6	58.4	7.7	10.4
Pacific Coast					
Spring	33.2	17.8	36.3	4.5	8.2
Summer	41.5	15.3	33.5	3.6	6.0
Autumn	25.2	15.3	46.3	6.5	6.7
Winter	17.2	33.8	40.3	6.3	2.5
ANNUAL	29.0	20.2	39.6	5.3	5.9

Table 3.2 Continued

Season	Sulfate	Nitrate	Organics	Elemental	Soil
Sierra-Humboldt					
Spring	20.7	8.0	43.8	5.5	21.9
Summer	19.7	4.1	54.8	6.6	14.8
Autumn	18.4	6.8	54.2	6.9	13.7
Winter	16.5	9.1	53.6	13.1	7.7
ANNUAL	19.1	6.5	51.7	7.4	15.2
Sierra Nevada					
Spring	25.3	13.2	41.1	4.3	16.0
Summer	23.6	7.0	54.0	5.8	9.7
Autumn	21.4	10.1	51.4	5.7	11.4
Winter	17.8	10.5	57.8	7.0	7.0
ANNUAL	22.8	9.8	50.3	5.5	11.6
Sonoran Desert					
Spring	31.5	6.8	32.5	4.3	24.9
Summer	38.0	4.3	33.7	4.4	19.6
Autumn	37.9	4.4	39.6	6.1	12.0
Winter	34.0	7.8	42.8	7.3	8.2
ANNUAL	35.4	5.7	36.6	5.3	17.1
Southern California					
Spring	14.3	49.2	25.3	3.8	7.4
Summer	19.9	35.3	33.5	5.0	6.2
Autumn	17.2	33.5	28.8	5.3	15.3
Winter	16.5	38.9	33.1	6.5	5.0
ANNUAL	17.0	40.8	29.3	4.7	8.2
Washington D.C.					
Spring	46.6	15.1	24.9	8.0	5.5
Summer	60.4	5.6	22.1	6.1	5.7
Autumn	42.7	12.8	29.0	10.8	4.8
Winter	32.0	21.5	32.1	10.2	4.3
ANNUAL	46.6	12.9	26.8	8.6	5.1
West Texas					
Spring	40.2	4.3	31.5	3.6	20.4
Summer	38.6	4.4	25.6	2.4	29.1
Autumn	44.1	4.2	31.6	3.9	16.2
Winter	42.2	7.1	34.1	5.0	11.5
ANNUAL	40.8	4.8	30.0	3.5	20.9

Boundary Waters. This region in northern Minnesota is monitored at Boundary Waters Canoe Area in the Superior National Forest, which began monitoring in August 1991. Previously, this region was represented by two sites, Isle Royale National Park, which was discontinued in July 1991, and Voyageurs National Park, which has been downgraded to channel-A only.

The average fine and coarse aerosol concentrations were 5.1 and 3.7 $\mu\text{g}/\text{m}^3$, respectively. The highest fine and coarse aerosol concentrations occurred during summer, but there was not as strong a seasonal variation as in Alaska and the Appalachian Mountains. In this region, sulfate was the largest fraction of fine particle mass (41.8%), followed closely by organics (37%), and more distantly by nitrate (11.9%), soil (5.4%), and light-absorbing carbon (4%).

Cascade Mountains. This region in the states of Washington and Oregon has two monitoring sites out of the four reported here. Mount Rainier National Park, initiated in March 1988, is southeast of Seattle, and the Columbia River Gorge on the Hood River National Forest, east of Portland, began monitoring in June 1993. The other two sites, Three Sisters Wilderness Area on the Willamette National Forest and Snoqualamie Pass on the Snoqualamie National Forest, were implemented in July 1993 but were not fully operational until September 1994.

Here the average fine and coarse aerosol concentrations are 5.2 and 3.5 $\mu\text{g}/\text{m}^3$, respectively. Fine and coarse aerosol concentrations reach their maxima in summer and minima in winter. Sulfate and nitrate concentrations have strong seasonal variations, with maxima for sulfate in summer and nitrate in winter. This seasonal variation could be, in part, the result of seasonal variations in mixing and in photochemistry. In this region, organics are the single most significant contributor (54.6%) to fine particle mass. Sulfate (28.1%) is about half the contribution of organics. Nitrate contributes 4.7%, followed by light-absorbing carbon (8.2%) then soil (5.4%).

Central Rocky Mountains. The measurements in this region were made at five locations in the mountainous Class I areas of Colorado and Wyoming, including the Bridger and Weminuche Wilderness Areas, Rocky Mountain and Yellowstone National Parks, and Great Sand Dunes National Monument. Fine and coarse aerosol concentrations in this region averaged 3.1 and 4.4 $\mu\text{g}/\text{m}^3$ over the three-year period. Like many of the other regions, concentrations, especially of sulfate, organics, light-absorbing carbon, and coarse aerosol, were highest in summer and lowest in winter. The largest contributor to fine particle mass in this region was organics (42.9%), followed by sulfate (26.5%), soil (19.6%), nitrate (5.9%), and light-absorbing carbon (5.1%).

Colorado Plateau. This region in the Four Corners' states of the Southwest is the most intensively monitored in the IMPROVE network. There are six sites, most of them within the so-called Golden Circle of National Parks: Bandelier, Bryce Canyon, Canyonlands, Grand Canyon, Mesa Verde, and Petrified Forest National Parks. A seventh site, Arches National Park, was discontinued in May 1992. This region is of particular concern to the Grand Canyon Visibility Transport Commission as required by Congress in the 1990 amendments to the Clean Air Act [Stensvaag, 1991].

In this region, fine and coarse aerosol concentrations averaged 3.3 and 4.1 $\mu\text{g}/\text{m}^3$, respectively. Fine and coarse aerosol concentrations here were greatest in summer and minimum in winter. Concentrations of sulfate and organics were also greatest in summer and smallest in winter. However, nitrate and light-absorbing carbon were both largest in winter. Here organics (39.1%) and sulfate (32.6%), contribute the most followed by soil (16.7%), nitrate (6.1%), and light-absorbing carbon

(5.4%).

Florida. Previously, this region had its monitoring site at Everglades National Park, which has now been downgraded to a channel-A only site. This region is now represented by two sites at Chassahowitzka Wildlife Refuge on the Gulf Coast north of Tampa, and Okefenokee Wilderness Area on the Georgia-Florida border. Monitoring at these two sites began in April 1993 and September 1991, respectively. Only Chassahowitzka is reported here for the three-year averages.

The fine and coarse aerosol concentrations averaged 10.1 and 7.4 $\mu\text{g}/\text{m}^3$, their concentrations were highest in summer. Fine and coarse aerosol concentrations were smallest in winter. Sulfate was found to be the largest contributor to fine particle mass (47.4%), followed by organics (30.9%), soil (10.7%), light-absorbing carbon (5.7%), and nitrate (5.3%).

Great Basin. The Great Basin of Nevada has two sets of measurements at Jarbidge Wilderness Area in northeastern Nevada and Great Basin National Park, which began monitoring in March 1988 and May 1992, respectively. Here the fine and coarse aerosol concentrations averaged 3.1 and 4.9 $\mu\text{g}/\text{m}^3$. The fine mass concentration was the lowest of any of the regions in the lower 48 states. Perhaps this is due to the fact that this site is relatively remote from high emission density areas and is generally well ventilated. Both fine and coarse aerosol concentrations, as well as all of the fine aerosol components, except nitrate and light-absorbing carbon, experienced largest concentrations in the summer and lowest concentrations in the winter. The largest single contributors to fine particle mass at this region were organics (44.3%) and soil (24.6%). Sulfate was a smaller contributor (21.1%), followed by light-absorbing carbon (5.1%) and nitrate (5%).

Lake Tahoe. Two sites are monitored for this region: one site is in Bliss State Park in southern California and a bit east of the lake and began sampling in March 1989. The other is close to the urban area of Lake Tahoe and sampling started in November 1990. Fine and coarse aerosol concentrations averaged 6.4 and 6.6 $\mu\text{g}/\text{m}^3$, respectively; there is a modest seasonality with highest concentrations occurring in the winter, and the least for fine aerosols in the summer and for coarse aerosol in the autumn. Sulfate, nitrate, organics, and light-absorbing carbon have strong seasonal trends with sulfate concentrations being more than twice as high in the summer than in the winter; however, nitrates, organics, and light-absorbing carbon have winter maxima at least twice their summer concentrations. The largest contributor to fine aerosol is organics (54.3%), followed by light-absorbing carbon (13.9%), sulfate (13%), soil (12.3%), and nitrate (6.5%).

Mid Atlantic. This new region is represented by the Edmond D. Forsyth Wildlife Refuge west of Atlantic City, New Jersey and began monitoring in September 1991. Fine and coarse aerosol concentrations averaged 11.4 and 9.1 $\mu\text{g}/\text{m}^3$, respectively. A moderate seasonality is evident with the highest fine and coarse aerosol concentrations occurring in the summer, and the least in the spring and winter, respectively. Sulfate, organics, and fine soil are the fine aerosol constituents that follow the seasonal trend for fine aerosol mass. Nitrate peaks in the winter at three times its summer concentration, and light-absorbing carbon peaks in the winter as well but only shows a small seasonality. Sulfate comprises the bulk of the fine aerosol mass (51.8%) followed by organics (26.8%), nitrate (11.3%), light-absorbing carbon (5.8%), and soil (4.4%).

Mid South. Three sites are monitored for this new region: Upper Buffalo Wilderness Area in north central Arkansas initiated in December 1991, Mammoth Cave National Park in Kentucky initiated in September 1991, and Sipsey Wilderness Area in northern Alabama initiated in March 1992. The

average concentration of fine and coarse aerosol was 12.1 and 6.0 $\mu\text{g}/\text{m}^3$, respectively. Outside of Washington D.C., which is an urban site, this region has the highest average concentration of fine aerosol. A modest seasonality is evident for fine and coarse aerosols with the minima occurring in the winter and the maxima the summer. All fine aerosol constituents except nitrate and light-absorbing carbon follow the seasonality of fine aerosol. Nitrate has its maximum concentrations in the winter, while light-absorbing carbon is fairly constant between seasons. Sulfate (51.3%) composes the bulk of fine aerosol followed by organics (28.6%), nitrate (8.4%), soil (7.2%), and light-absorbing carbon (4.5%).

Northeast. The northeastern United States is represented by measurements at two sites: Acadia National Park on the coast of Maine, which began monitoring in March 1988, and Lye Brook Wilderness Area in southern Vermont, which began in September 1991. Here fine and coarse aerosol concentrations averaged 6.4 and 4.2 $\mu\text{g}/\text{m}^3$. Although fine and coarse aerosol concentrations were both largest in summer, there was not a strong seasonal variation. Sulfate, organics, and soil concentrations were also largest in summer. Nitrate concentrations reached their maximum in winter. The contributors to fine particle mass included sulfate (52.9%), organics (30.9%), nitrate (7.2%), light-absorbing carbon (5.2%), and soil (3.8%).

Northern Great Plains. Only one set of measurements was made in this region, at Badlands National Monument in South Dakota. Here fine and coarse aerosol concentrations averaged 4.5 and 5.0 $\mu\text{g}/\text{m}^3$, respectively. The maximum concentrations for fine mass occurred in the winter and spring and was least in the autumn. The maximum for coarse mass occurred in the spring and summer and was least during the winter. Sulfate (37.2%) and organics (35.1%) each contributed to fine mass about equally, followed by nitrate (13.3%), soil (10.7%), and light-absorbing carbon (3.8%).

Northern Rocky Mountains. This region has measurements made at Glacier National Park in Montana, close to the Canada border. Fine aerosol and coarse aerosol concentrations averaged 5.5 $\mu\text{g}/\text{m}^3$ each. There were no strong seasonal variations except for nitrate, which showed a strong winter peak, and coarse mass, which peaked in the winter. Organics are by far the largest contributor to fine particle mass (58.4%) followed by sulfate (17.9%), soil (10.4%), light-absorbing carbon (7.7%), and nitrate (5.6%).

Pacific Coast. This region includes three Class I areas along and near the coast of northern California: Pinnacles National Monument, Point Reyes National Seashore, and Redwoods National Park. In this region, the fine and coarse aerosol concentrations over the three-year period averaged 4.6 and 8.2 $\mu\text{g}/\text{m}^3$. There was no strong seasonal variation in concentration, except for sulfate that had maxima and minima in summer and winter, and nitrate that showed the opposite trend, with maxima and minima in winter and summer, respectively. One would expect sulfate to reach its maximum concentration in summer because of photochemistry. Nitrate would be expected to reach its peak during the colder months of winter because of the extreme thermal volatility of ammonium nitrate. Organics in this region are the largest single component of fine aerosol (39.6%), followed by sulfate (29%), nitrate (20.2%), soil (5.9%), and light-absorbing carbon (5.3%).

Sierra-Humboldt. The region further north in the Sierra Nevada and Humboldt Mountain Ranges was measured with sites at Crater Lake National Park in Oregon and Lassen Volcanoes National Park in northern California. This region is relatively remote from high emission density areas. Its fine and coarse aerosol concentrations were relatively low, at 3.1 and 3.2 $\mu\text{g}/\text{m}^3$, respectively. Summer concentrations were generally about twice those during the winter. Organics contributed most of the

fine particle mass (51.7%), followed by sulfate (19.1%), soil, (15.2%), light-absorbing carbon (7.4%), and nitrate (6.5%).

Sierra Nevada. The Sierra Nevada Mountains in California was monitored at two sites: Yosemite and Sequoia National Parks. Yosemite National Park has been monitored since March 1988. Sequoia National Park had channel A and D since March 1992 but was not fully instrumented until July 1993.

Average fine and coarse aerosol concentrations were 4.5 and 5.0 $\mu\text{g}/\text{m}^3$. There was a strong moderate variation, with maximum concentrations in summer and minimum concentrations in winter. The only exception was nitrate, which was relatively constant throughout the year. Organics contributed more than twice what sulfate contributed (50.3% and 22.8%, respectively). Soil was the next largest contributor (11.6%), followed by nitrate (9.8%), and light-absorbing carbon (5.5%).

Sonoran Desert. This region in southeastern Arizona was monitored at two sites: Chiricahua and Tonto National Monuments and were initiated in March 1988. The three-year average of fine and coarse mass concentrations in this region were 4.3 and 5.5 $\mu\text{g}/\text{m}^3$, respectively. These concentrations were highest in summer and lowest in winter. The sulfate, organics, and soil components of fine particle mass also had maxima and minima in these seasons. The contributions to fine particle mass were distributed nearly equally between sulfate (35.4%) and organics (36.6%), followed by soil (17.1%), nitrate (5.7%), and light-absorbing carbon (5.3%).

Southern California. Measurements in this region were made in San Geronio National Monument, east of the Los Angeles metropolitan area. Fine and coarse aerosol concentrations were highest of any western United States site (9.0 and 8.4 $\mu\text{g}/\text{m}^3$); concentrations were only higher in the eastern United States. Like many sites in the IMPROVE network, concentrations were highest in summer and lowest in winter. This trend was also observed for nitrate: actually nitrate was highest in spring and lowest in winter, but concentrations in summer were twice those in winter. This site was the only site in the IMPROVE network in which nitrate was a larger contributor to fine particle mass than either sulfate or organic carbon. The contributions were nitrate (40.8%), organics (29.3%), sulfate (17%), soil (8.2%), and light-absorbing carbon (4.7%).

Washington, D.C. This is a single monitoring site in the nation's capital. Fine and coarse aerosol concentrations were higher here than anywhere in the IMPROVE network. They averaged 19.2 and 7.4 $\mu\text{g}/\text{m}^3$ over the three-year period. There was a moderate seasonal variation in fine aerosol concentrations; in spring they ranged from 17.1 to 23 $\mu\text{g}/\text{m}^3$ in summer. However, the sulfate and nitrate components varied significantly by season: sulfate concentrations were largest in summer and smallest in winter, while nitrate concentrations were largest in winter and smallest in summer. The sulfate behavior could be caused by the seasonal variation in photochemistry. The nitrate behavior may be due to the extreme volatility of nitrate in warm weather. Over the entire three-year period, fine particle mass was constituted of sulfate (46.6%), organics (26.8%), nitrate (12.9%), light-absorbing carbon (8.6%), and soil (45.1%).

West Texas. Two measurement sites in west Texas were included: Big Bend and Guadalupe Mountains National Parks. Both sites are near the Mexico border in southwestern Texas and have operated since March 1988. The fine and coarse aerosol concentrations averaged 5.2 and 6.9 $\mu\text{g}/\text{m}^3$ over the last three years. Minimum concentrations generally occurred during winter, while maximum concentrations occurred in summer. The only exception was light-absorbing carbon, which remained constant. The contributions to fine particle mass averaged 40.8% for sulfate, 30% for organics, 20.9% for soil, 4.8% for nitrate, and 3.5% for light-absorbing carbon.

In general, the following observations can be made. With few exceptions, aerosol concentrations are highest in summer and lowest in winter. This is consistent with the fact that sulfate formation rates, natural organic carbon emissions, and mixing into mountainous regions are all maximum in summer and minimum in winter. With the notable exception of southern California where nitrate is dominant, sulfate and organics are the two principal components of the fine particle mass throughout the United States. Sulfate's contribution is much higher in the eastern United States than in the western United States and in Alaska.

3.2 Spatial Trends in Aerosol Concentrations in the United States

Because of the relatively large number of IMPROVE aerosol monitoring sites in the western United States, isopleth maps of the average aerosol concentrations measured over the three-year period from March 1992 through February 1995 could be drawn. Figures 3.1 through 3.8 show isopleth maps of the three-year average aerosol concentrations (PM_{10} , fine mass, coarse mass, sulfate, nitrate, organics, light-absorbing carbon, and soil). These figures provide us with information on how aerosol concentrations and mass budgets vary over the United States.

3.2.1 PM_{10} Aerosol

Figure 3.1 shows isopleths of the PM_{10} aerosol mass concentration measured during this three-year period. The highest concentrations occur in the eastern United States. With the exceptions of the Northern Great Plains states, almost all the area east of Colorado and New Mexico has concentrations in excess of $10 \text{ } \mu\text{g}/\text{m}^3$. The highest concentrations are in Washington D.C. at $22 \text{ } \mu\text{g}/\text{m}^3$, followed by Florida and the Mid South, which experienced concentrations in excess of $18 \text{ } \mu\text{g}/\text{m}^3$. Outside of California and the Northern Rockies the least amount of PM_{10} concentrations occur in the western United States, where there is a large swath extending from Oregon, northern California, Nevada, Utah, Wyoming, into northern Arizona and northern New Mexico and western Colorado, where the concentration of PM_{10} is less than $8.5 \text{ } \mu\text{g}/\text{m}^3$. The lowest concentration in the lower 48 states occurs at Bridger Wilderness Area in Wyoming with only $5.7 \text{ } \mu\text{g}/\text{m}^3$ on average, the least was recorded at Denali National Park in Alaska at $4.2 \text{ } \mu\text{g}/\text{m}^3$. The strongest gradient is between northern California and Utah and the coastal regions of California, where concentrations vary from $6.4 \text{ } \mu\text{g}/\text{m}^3$ to an excess of $15 \text{ } \mu\text{g}/\text{m}^3$.

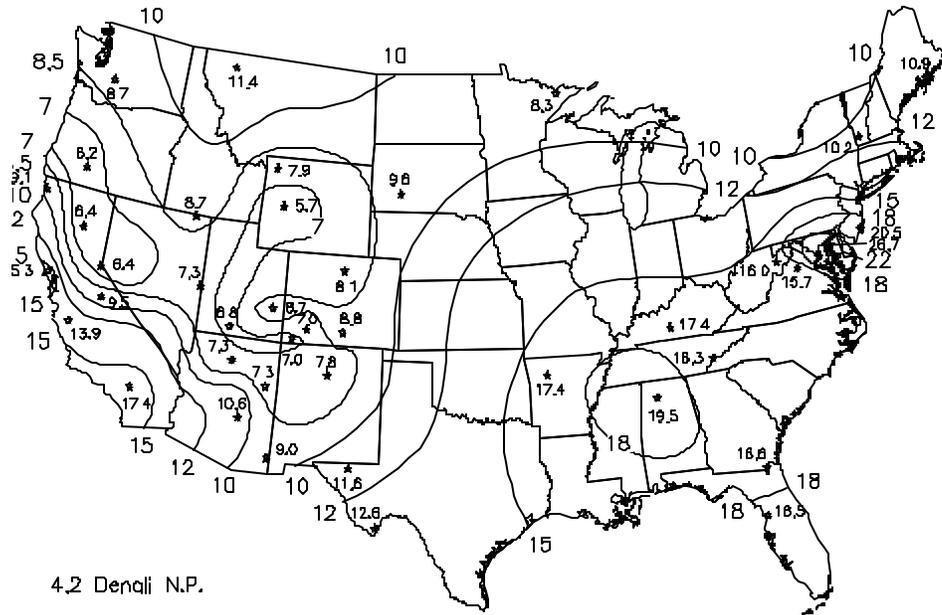


Figure 3.1 Average PM_{10} mass concentration (in $\mu\text{g}/\text{m}^3$) for each site in the IMPROVE network.

3.2.2 Fine Aerosol

Figure 3.2 shows isopleths of the average fine aerosol concentrations measured during the three-year period. Note the strong gradient in fine particle concentrations from southern California, a local maximum of $9 \mu\text{g}/\text{m}^3$ to minima of 2.7 to $3.1 \mu\text{g}/\text{m}^3$ observed in southern Oregon, Nevada, southern Utah, western Colorado, and Wyoming. This is a factor of three variations in average fine aerosol concentration. Also, note that fine aerosol concentrations increase again as one moves to the eastern United States with maxima of about $13.5 \mu\text{g}/\text{m}^3$ in Shenandoah and Great Smoky Mountains National Parks and over $19 \mu\text{g}/\text{m}^3$ in Washington D.C. Thus, from the minima in the western United States to the maxima in the East, there is a factor of six variations in average concentration. Average fine aerosol concentrations in Denali National Park of $1.8 \mu\text{g}/\text{m}^3$ are lower than any measured in the lower 48 states. There is a factor of 10 variations between the average measured in Alaska and that measured in Washington D.C.

The lower map in Figure 3.2 shows isopleths that depict the fraction of PM_{10} that is fine aerosol ($PM_{2.5}$). Almost all of the country outside of the intermountain west has a fine mass fraction of PM_{10} that exceeds 50%. East of the Mississippi and south of the Great Lakes there is a broad region that exceeds 65%. The highest values encompass a region that covers the Ohio Valley, parts of the Mid South, West Virginia, Shenandoah, and Washington D.C., where fine mass fraction is greater than 70%. The smallest fine mass fraction occurs in the Great Basin Region, central Utah, and portions of Colorado, where less than 40% of PM_{10} is fine mass.

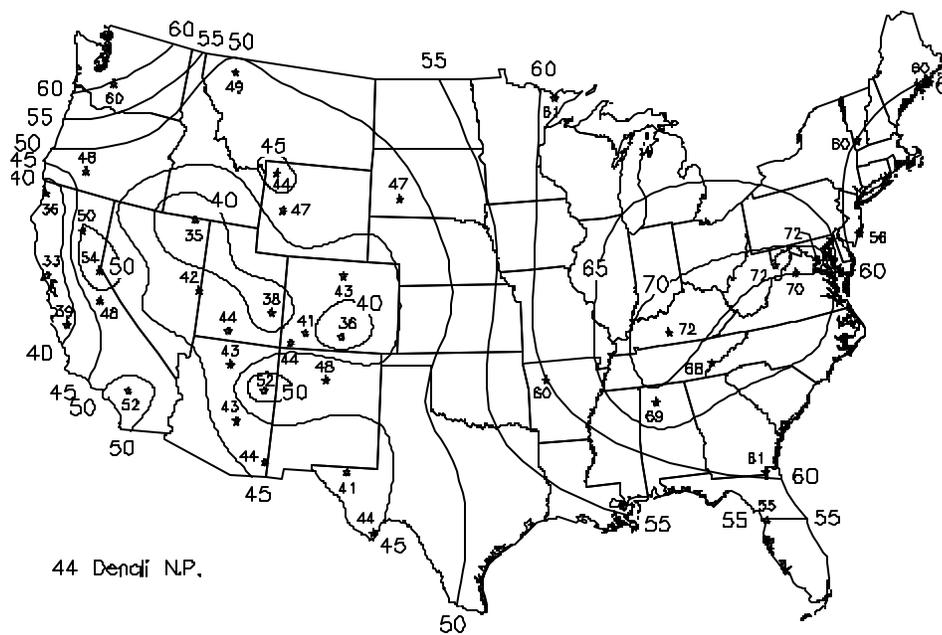
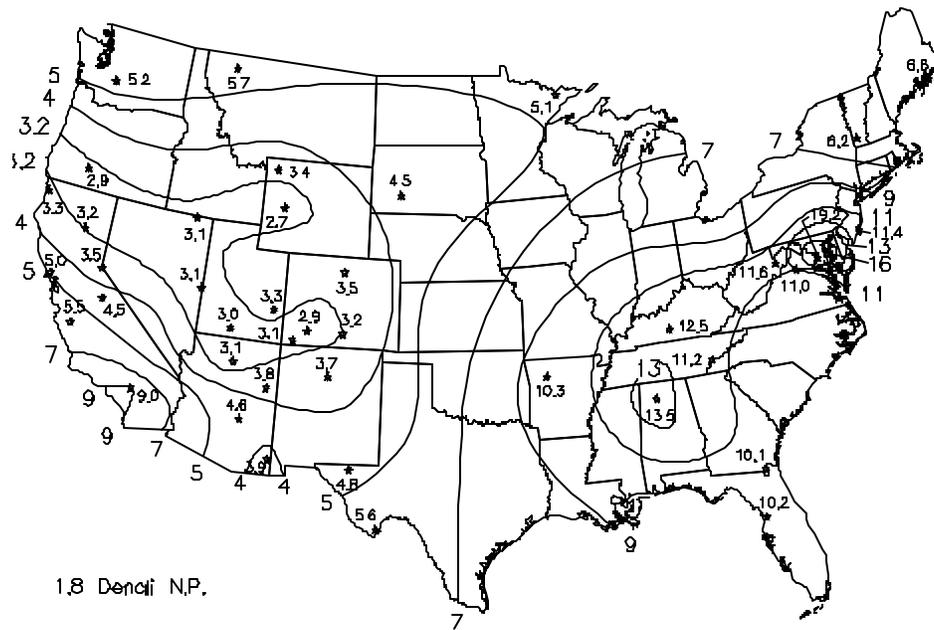


Figure 3.2 Average fine mass aerosol concentrations (in $\mu\text{g}/\text{m}^3$) (top) and fine mass fraction of PM_{10} (bottom) for each site in the IMPROVE network.

3.2.3 Coarse Aerosol

Figure 3.3 shows isopleths of the three-year average coarse aerosol concentrations. There are a few local maxima from 7.4 to 10.3 $\mu\text{g}/\text{m}^3$ that are noticeable near Los Angeles, San Francisco, and Washington D.C. The lowest coarse aerosol concentrations occur in the swath from the Pacific Northwest through Nevada to southern Utah. Concentrations in this region average around 4 $\mu\text{g}/\text{m}^3$. Throughout the United States coarse aerosol concentrations are generally in the factor-of-two range from 4 to 8 $\mu\text{g}/\text{m}^3$. The patterns in the eastern United States, with the exception of Washington D.C., shows a steady north-south trend of increasing coarse aerosol concentrations. Coarse aerosol concentrations in Alaska are not significantly lower than in the lower 48 states. There is approximately a factor-of-three range from the lowest average concentrations measured in Oregon and Utah and the highest measured in Washington D.C.

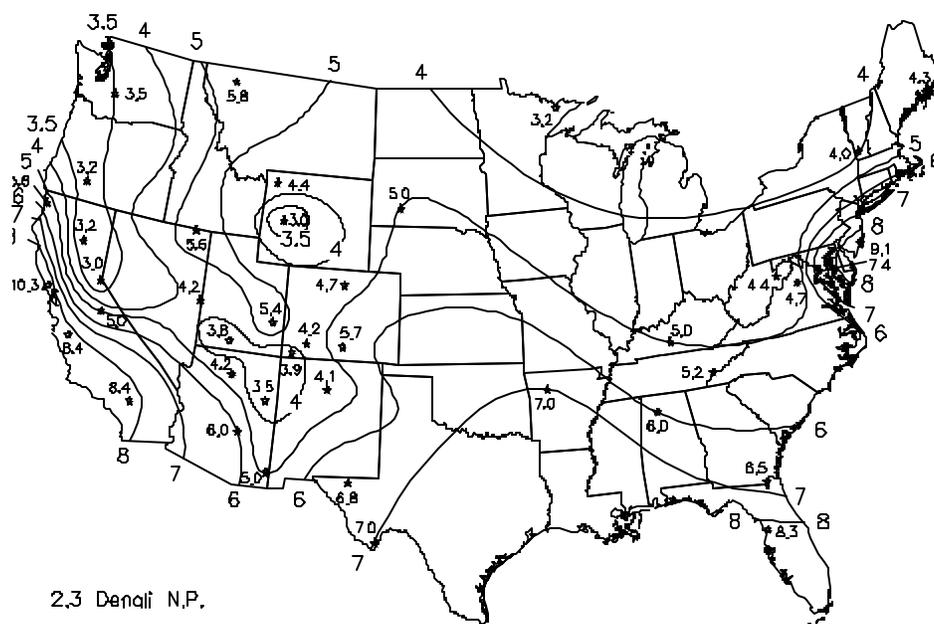


Figure 3.3 Average coarse particle mass concentration (in $\mu\text{g}/\text{m}^3$) for each site in the IMPROVE network.

3.2.4 Fine Sulfate Aerosol

The average sulfate component of the fine aerosol measured over the three-year period is shown in Figure 3.4. Since sulfate is one of the two major components of fine particle mass, it is not surprising to observe similar gradients across the United States to what was observed for total fine particle mass. There is a strong gradient from high concentrations in California urban areas to low concentrations in southern Oregon and Nevada. There is also a strong gradient from the relatively low concentrations in the West to those in the East. There is a factor of 15 variations from the lowest concentration measured in Nevada to the highest concentration measured in Washington, D.C. This gradient is most likely indicative of the strong regional gradient in SO_2 emission density. The eastern United States has a concentration of power plants that burn high sulfur coal, while the western United States has relatively

low SO₂ emission densities. A relative maximum in sulfate concentration is observed in southern Arizona, which is near copper smelters that emit large quantities of SO₂. The lower map in Figure 3.4 shows that sulfate constitutes as little as 17% of fine particle mass in southern California to as much as 61% of total fine mass in Shenandoah National Park. In the Golden Circle of parks in the Four Corners' states, sulfate is 31 to 35% of the fine particle mass.

In the eastern United States sulfate is the largest single component of fine particle mass. In the Boundary Waters, Sonoran Desert, and West Texas regions, sulfate is tied with organic carbon as the largest component of fine particle mass. Sulfate is the second largest component of fine mass in all other regions studied except southern California and the Great Basin (where sulfate is the third largest component).

3.2.5 Fine Nitrate Aerosol

Figure 3.5 shows isopleth maps of the nitrate concentration and nitrate mass fraction of fine aerosol, averaged over the three-year period. Note that the highest concentration of 3.7 $\mu\text{g}/\text{m}^3$ was measured in San Geronio Wilderness, just east of the Los Angeles metropolitan area. Other high concentrations occur in Washington, D.C. (2.5 $\mu\text{g}/\text{m}^3$), and near the San Francisco area (1.3 $\mu\text{g}/\text{m}^3$). There is a strong gradient from the high concentrations in the California urban areas to the minima of 0.1 $\mu\text{g}/\text{m}^3$ measured in Oregon, Nevada, Wyoming, and Colorado. There is a long swath of low nitrate concentrations extending from Oregon, Nevada, and Idaho into Utah, Wyoming, Colorado and into southern Arizona and southern New Mexico (<0.2 $\mu\text{g}/\text{m}^3$). Nitrate mass fractions are typically 4 to 12% except in California where they are 30% and higher. In the north central part of the United States and the mid-Atlantic region nitrates constitute over 12% of the fine aerosol mass. Nitrates generally reach their maxima in the winter when colder temperatures favor the formation of ammonium nitrate aerosol from nitric acid vapor. Nitrate is the largest single component of fine aerosol mass in southern California at San Geronio Wilderness Area.

3.2.6 Fine Organic Aerosol

Figure 3.6 shows isopleth maps of the organic carbon mass fraction of the fine aerosol concentration, averaged over the three-year period. There is a significant spatial gradient from the Pacific Northwest, with average concentrations of 2.0 to 3.0 $\mu\text{g}/\text{m}^3$ to the intermountain region of Wyoming, Colorado, Utah, and Arizona of 1.2 $\mu\text{g}/\text{m}^3$ or less. In the eastern United States, organics range generally from 2.0 to 4 $\mu\text{g}/\text{m}^3$. In Alaska, organic aerosol concentrations are the lowest at 1 $\mu\text{g}/\text{m}^3$.

Except in the northwestern United States, where organics are over half of the fine particle mass, organics generally constitute between 25 to 40% of the fine particle mass. Moreover, organics are the largest single component of fine particle mass in most of the regions in the United States. Exceptions include the Mid South and eastern United States where sulfate is the dominant component and southern California, where nitrate is the dominant component.

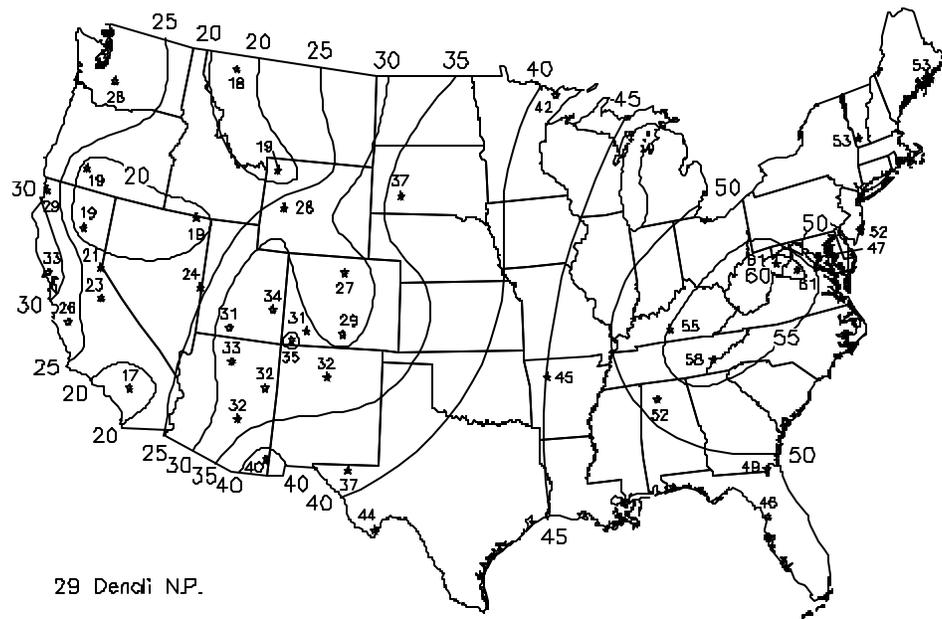
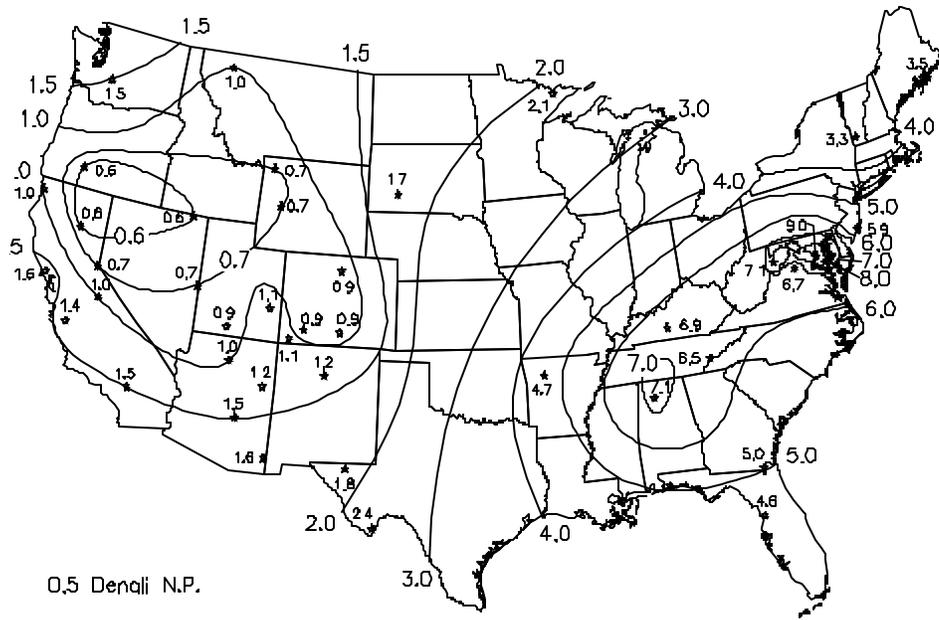


Figure 3.4 Average fine sulfate aerosol concentrations (in $\mu\text{g}/\text{m}^3$) (top) and sulfate fine mass fractions (bottom) for each site in the IMPROVE network.

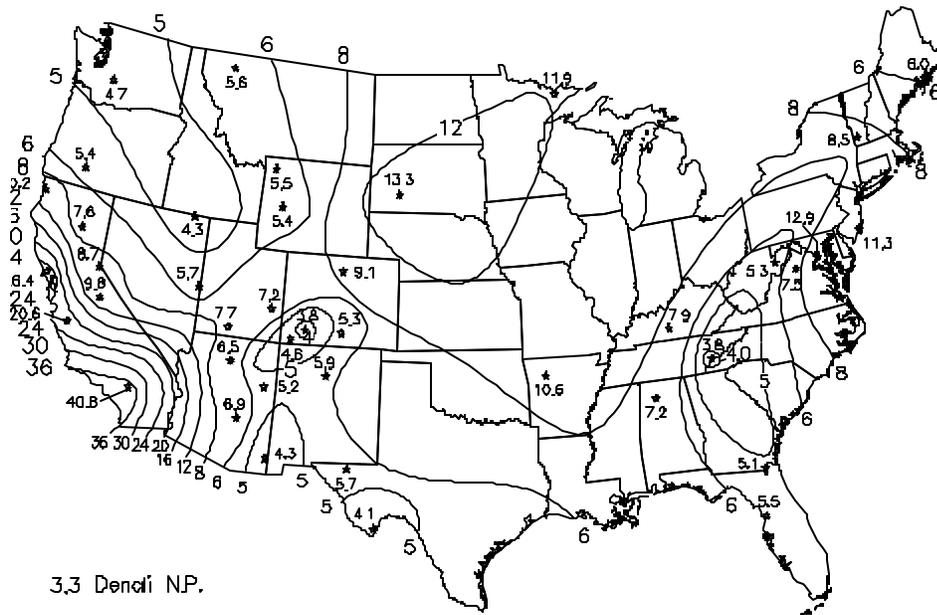
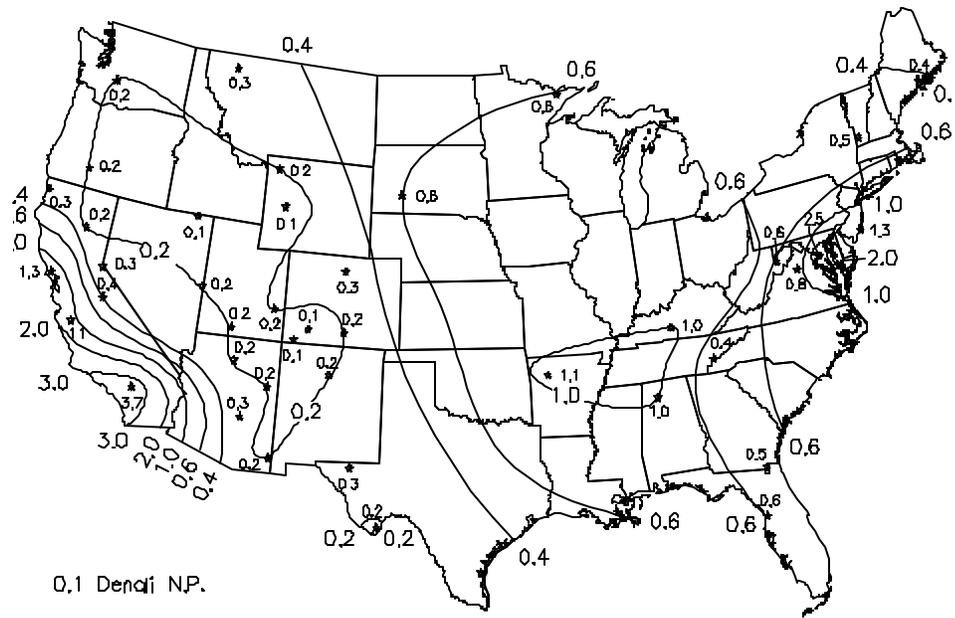


Figure 3.5 Average fine nitrate aerosol concentrations (in $\mu\text{g}/\text{m}^3$) (top) and nitrate fine mass fractions (bottom) for each site in the IMPROVE network.

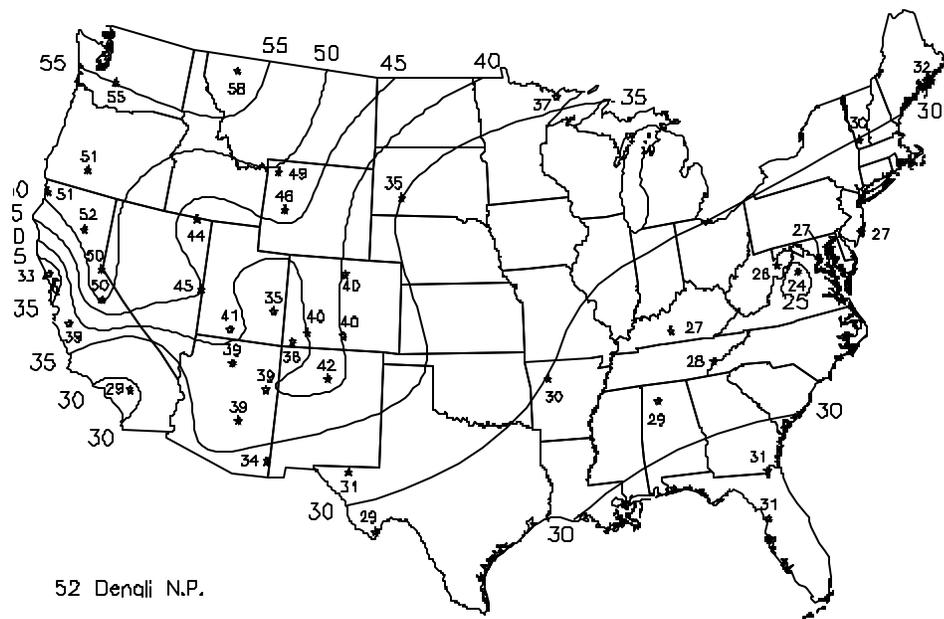
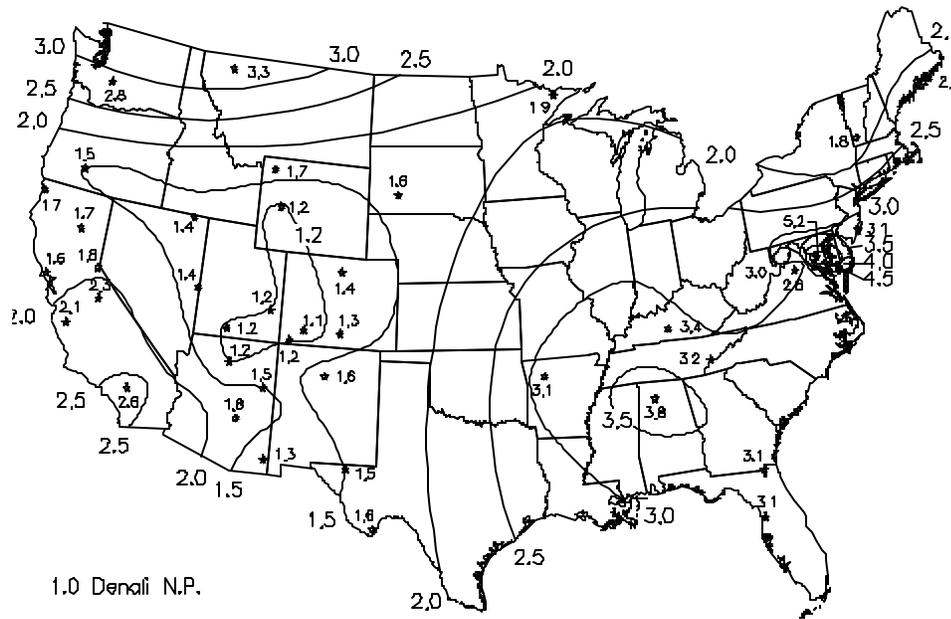


Figure 3.6 Average fine organic aerosol concentrations (in $\mu\text{g}/\text{m}^3$) (top) and organic fine mass fractions (bottom) for each site in the IMPROVE network.

3.2.7 Fine Light-Absorbing Carbon Aerosol

Figure 3.7 shows isopleth maps of the light-absorbing carbon concentration and mass fraction of the fine aerosol, averaged over the three-year period. Note that light-absorbing carbon concentrations are highest in the Pacific Northwest, the area east of the Mississippi and south of the Great Lakes, and southern California, while concentrations are much lower in much of the West (Wyoming, Utah, and Nevada). Light-absorbing carbon is the smallest contributor to fine particle mass, constituting generally 3 to 5% of the fine particle mass. Exceptions to this are the Pacific Northwest and Washington, D.C. areas where light-absorbing carbon contributes as much as 8% of the fine particle mass.

3.2.8 Fine Soil Aerosol

Figure 3.8 shows isopleth maps for fine soil. The contribution of soil to the fine aerosol in the United States is generally small, except for the elevated concentrations ($<1 \mu\text{g}/\text{m}^3$) in the southern tier of the United States. There is a quite noticeable north-south trend of increasing soil concentrations with the Northeast being the lowest. Soil contributes approximately 5 to 10% of the fine aerosol mass in the East. Except for Florida, all of the area east of the Mississippi, the Pacific Northwest, and parts of California, soil contributes less than 10% to fine aerosol mass with much of the intermountain west in excess of 20%.

3.3 Summary

The following are the major patterns observed in the three-year period of IMPROVE from March 1992 through February 1995:

1. Spatial Patterns. Concentrations of fine particles (those most important in determining visibility) are highest in the eastern United States and in southern California and lowest in the relatively unpopulated areas of the West.
2. Major Contributions to Fine Aerosol. The largest single component of the fine aerosol in the East is sulfate, while in the Pacific Northwest it is organics, and in southern California it is nitrate. In general, the largest mass fractions of the fine aerosol are sulfates and organics. Of the 21 regions in the IMPROVE network, organic carbon is the largest single component in ten regions (Alaska, Cascades, Colorado Plateau, Central Rockies, Pacific Coastal Mountains, Great Basin, Northern Rockies, Sierra Nevada, Sierra-Humboldt, and Lake Tahoe). Sulfate is the largest single component of fine aerosol in seven regions, primarily in the East (Appalachian Mountains, Florida, Northeast, Mid South, Mid Atlantic, Washington D.C., and West Texas). The contributions of organic carbon and sulfate are approximately equal in three regions (Boundary Waters, Sonoran Desert, and Northern Great Plains). Soil is the next largest contributor, followed by nitrate and light-absorbing carbon. Nitrate is the largest component of fine aerosol in southern California only.
3. Smaller Contributors. After the contributions of organics and sulfate, soil is the next largest, followed by nitrate and light-absorbing carbon.

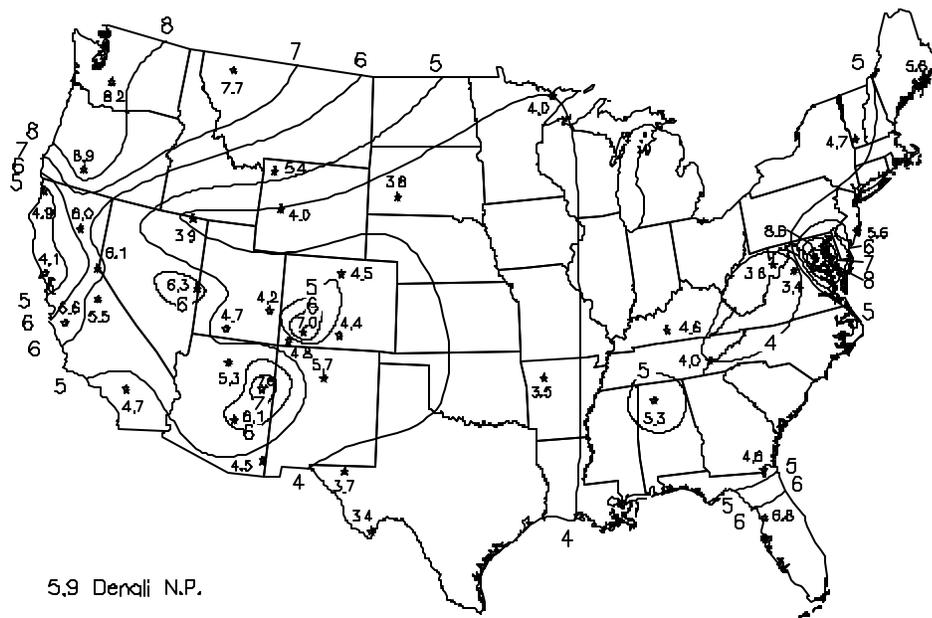
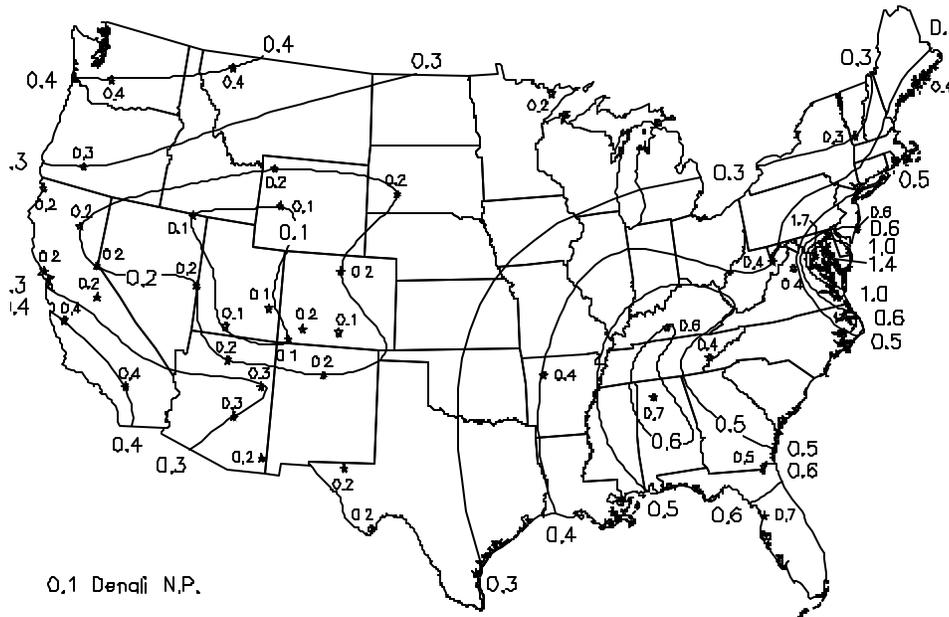


Figure 3.7 Average fine elemental carbon aerosol concentrations (in $\mu\text{g}/\text{m}^3$) (top) and elemental carbon fine mass fractions (bottom) for each site in the IMPROVE network.

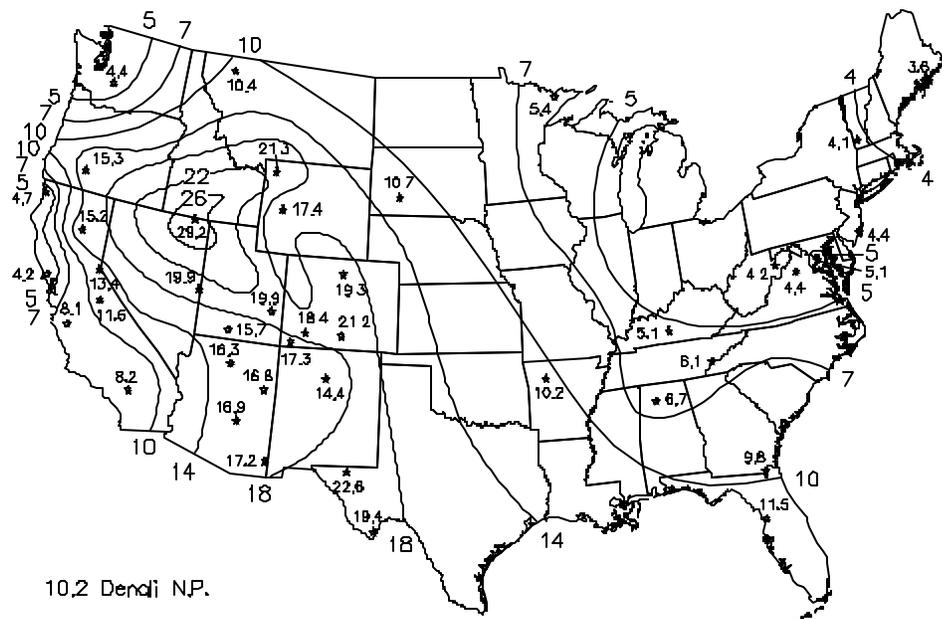
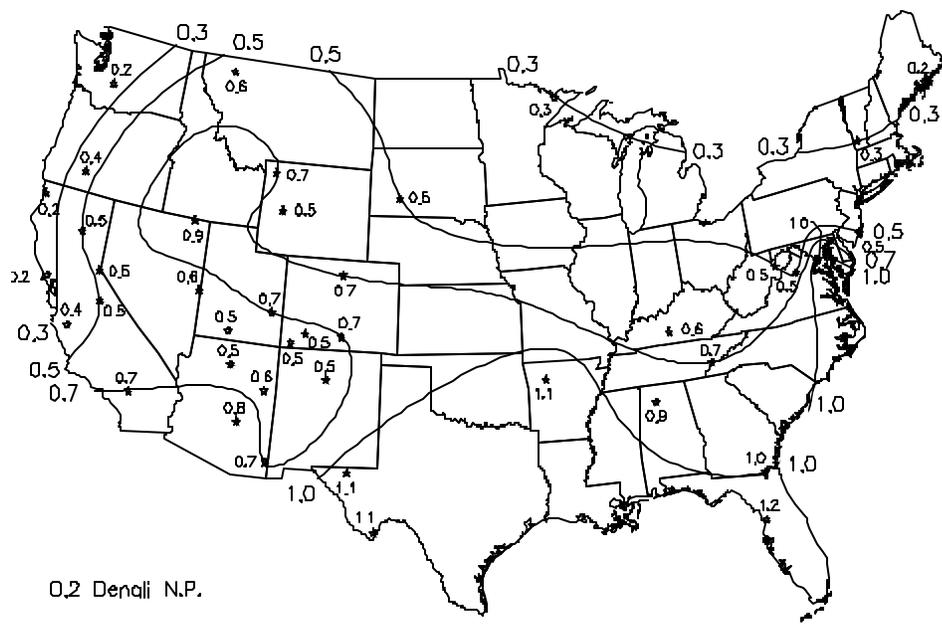


Figure 3.8 Average fine soil aerosol concentrations (in $\mu\text{g}/\text{m}^3$) (top) and soil fine mass fractions (bottom) for each site in the IMPROVE network.

4. **Seasonality.** With a few exceptions, average fine mass concentrations, organics and sulfate components of fine mass are highest in summer. Soil concentrations are highest in spring or summer. On the other hand, nitrate concentrations are generally highest in winter or spring. Light-absorbing carbon exhibits relatively little seasonal variation.
5. **PM₁₀.** The highest concentrations of PM₁₀ occur in a region east of the Mississippi and south of the Great Lakes, followed by coastal and southern California. In the East, the high concentrations are driven by high fine mass, which contributes as much as 70% of PM₁₀.

3.4 References

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