CHAPTER 5

SPATIAL AND SEASONAL DISTRIBUTION OF AEROSOL CONCENTRATION

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 (see Chapter 3) for the three-year period, March 1988 through February 1991.

Aerosol concentrations and chemical composition vary because of a number of factors, including the spatial distribution of natural and anthropogenic emission sources and the meteorological conditions of the area. 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 SO_2 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 (SO_2) emissions such as the eastern United States where SO_2 is emitted from coal-fired power plants, and in the Southwest due to copper smelter and Mexican SO_2 emissions. Organic carbon concentrations tend to be highest in regions such as the Pacific Northwest due to forests and forest-products industries and in areas such as Southern California from motor vehicle emissions. 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 descriptive statistics of the three years of measurements (Appendix F), time lines of the individual measurements as shown in (Appendix G), and mass budgets. Mass budgets are the contribution of individual aerosol species to the reconstructed fine particle mass (see Chapter 3). 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 first three years of the IMPROVE network are presented. The 36 IMPROVE sites are grouped into regions according to their relative location, climatology, sulfate acidity, and similarities in concentrations and seasonal trends (see Chapter 1 for a list of the sites in each region). Average concentrations and chemical composition are calculated on the basis of the measurements for each region. Tables 5.1 and 5.2 show the mass concentrations of fine and coarse aerosol and the chemical composition (mass budgets) of the fine aerosol for each of the 19 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. Figures 5.1 and 5.2 present these data in graphical form. The seasonal and annual averages of concentrations and mass budgets are presented as bar charts for each region and overlaid on maps of the United States.

Figures 5.3 and 5.4 show isopleth maps of measured three-year averages of fine and coarse particle concentrations, respectively. Figures 5.5 through 5.9 show isopleth maps of the three-year average concentrations and mass budgets for all the sites in the United States for the sulfate, nitrate, organic carbon, light-absorbing carbon, and soil fractions of the fine aerosol, respectively. The top map in each figure shows the concentration, and the bottom map shows the percentage contribution (mass budget) of the given species to total reconstructed fine-particle mass.

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

5.1 Characteristics of the Regions

Alaska. The Alaska region has only one monitoring site at Denali National Park. The average concentrations of fine and coarse aerosol over the three-year period were 1.9 and 4.2 $\mu g/m^3$, 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. Organic carbon is the largest contributor of fine particle mass (at 44%), followed by sulfate (33%), soil (17%), and nitrate and light-absorbing carbon (each at 3%). The concentrations of organic 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 monitors at two sites: Great Smoky Mountains and Shenandoah National Parks. The average concentrations of fine and coarse aerosol over the three-year period were 10.9 and 6.2 $\mu g/m^3$, respectively. The fine aerosol concentration was the second largest in the entire United States, exceeded only by the concentration of 16.2 $\mu g/m^3$ measured in Washington, D.C. 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 58 percent, it is more than twice that of the next largest contributor, organic carbon (27%). Other contributors include nitrate (6%), soil (5%), and light-absorbing carbon (4%). Except for nitrate that has its largest concentration in the winter, the other components of fine aerosol all have maximum concentrations in summer. The seasonal variation in sulfate concentrations is particularly strong with summer concentrations of 10.5 $\mu g/m^3$ more than three times winter concentrations.

Table 5.1 Measured fine and coarse aerosol concentrations (in $\mu g/m^3$) for the 19 regions in the IMPROVE network, averaged over the three-year period, March 1988 through February 1991.

Season	Fine mass	Sulfate	Nitrate	Organics	Elemental carbon	Soil	Coarse mass
ALASKA							
Winter	1.6	0.7	0.1	0.6	0.1	0.2	4.0
Spring	2.4	0.9	0.1	0.7	0.1	0.6	3.9
Summer	2.7	0.5	0.0	1.5	0.1	0.4	5.4
Autumn	1.2	0.4	0.1	0.6	0.1	0.1	3.2
ANNUAL	1.9	0.6	0.1	0.9	0.1	0.3	4.2
APPALACHIAN							
Winter	6.5	3.0	0.8	2.0	0.4	0.3	3.1
Spring	10.6	6.0	0.8	2.7	0.5	0.6	4.5
Summer	16.6	10.5	0.3	4.4	0.5	0.8	11.2
Autumn	9.7	5.6	0.5	2.7	0.5	0.4	5.5
ANNUAL	10.9	6.3	0.6	3.0	0.5	0.5	6.2
BOUNDARY WATERS							
Winter	5.2	2.0	1.4	1.4	0.2	0.2	3.2
Spring	5.4	2.6	0.4	1.8	0.2	0.4	5.1
Summer	6.2	2.2	0.1	3.1	0.3	0.5	8.2
Autumn	4.3	1.6	0.4	1.8	0.2	0.3	5.8
ANNUAL	5.3	2.0	0.6	2.1	0.2	0.3	5.7
CASCADES							
Winter	3.8	0.6	0.1	2.6	0.5	0.1	2.9
Spring	5.2	1.4	0.2	2.7	0.5	0.3	3.1
Summer	6.7	2.4	0.4	3.0	0.5	0.3	4.6
Autumn	5.3	1.3	0.2	3.1	0.5	0.2	3.9
ANNUAL	5.1	1.3	0.2	2.8	0.5	0.2	3.5
COLORADO PLATEAU							
Winter	2.9	0.9	0.5	1.1	0.2	0.3	3.2
Spring	3.4	0.9	0.2	1.0	0.1	1.1	5.3
Summer	4.1	1.3	0.2	1.6	0.2	0.9	6.4
Autumn	3.2	1.2	0.1	1.2	0.2	0.5	3.7
ANNUAL	3.4	1.1	0.2	1.2	0.2	0.7	4.7
CENTRAL ROCKIES							
Winter	2.0	0.5	0.2	0.9	0.1	0.3	3.0
Spring	3.4	0.9	0.3	1.1	0.1	1.1	4.3
Summer	4.8	1.0	0.1	2.4	0.2	0.9	7.5
Autumn	2.9	0.8	0.1	1.3	0.1	0.5	4.0
ANNUAL	3.3	0.8	0.2	1.5	0.1	0.7	4.8
CENTRAL COAST							
Winter	5.6	0.9	1.9	2.3	0.4	0.2	7.7
Spring	4.2	1.4	0.8	1.5	0.2	0.3	9.3
Summer	4.5	1.9	0.8	1.4	0.1	0.2	10.7

Autumn	5.7	1.4	1.0	2.7	0.4	0.3	7.8
ANNUAL	5.0	1.4	1.1	1.9	0.3	0.2	8.9

Table 5.1 Continued

Season	Fine mass	Sulfate	Nitrate	Organics	Elemental carbon	Soil	Coarse mass
FLORIDA							
Winter	5.5	2.4	0.7	1.9	0.4	0.2	8.5
Spring	7.7	3.8	0.9	2.1	0.3	0.7	8.0
Summer	9.1	2.5	0.5	3.0	0.3	2.7	13.6
Autumn	6.9	3.1	0.5	2.3	0.4	0.5	8.6
ANNUAL	7.1	2.9	0.7	2.3	0.4	0.9	9.6
GREAT BASIN							
Winter	1.1	0.3	0.1	0.5	0.0	0.1	1.0
Spring	2.4	0.5	0.1	0.9	0.0	0.9	3.7
Summer	4.5	0.7	0.1	1.7	0.1	1.9	8.2
Autumn	3.1	0.6	0.1	1.4	0.1	1.0	5.1
ANNUAL	2.8	0.5	0.1	1.1	0.1	1.0	5.0
HAWAII							
Winter	4.0	2.8	0.1	0.9	0.1	0.1	3.0
Spring	3.6	2.5	0.1	0.8	0.1	0.2	7.4
Summer	1.6	0.9	0.1	0.5	0.0	0.1	10.3
Autumn	3.4	2.5	0.1	0.8	0.1	0.1	9.3
ANNUAL	3.2	2.2	0.1	0.7	0.1	0.1	8.2
NORTHEAST							
Winter	6.6	3.3	0.8	1.8	0.5	0.2	3.1
Spring	6.1	3.6	0.4	1.5	0.3	0.3	4.1
Summer	8.6	4.5	0.3	3.0	0.4	0.3	6.7
Autumn	5.6	3.0	0.4	1.6	0.4	0.2	4.1
ANNUAL	6.7	3.6	0.5	2.0	0.4	0.2	4.5
NORTHERN GREAT PLAINS							
Winter	3.4	1.2	0.6	1.1	0.1	0.5	3.9
Spring	5.0	1.9	0.6	1.3	0.1	1.0	6.0
Summer	5.6	1.8	0.2	2.2	0.2	1.2	9.7
Autumn	4.0	1.2	0.2	1.5	0.1	1.0	5.8
ANNUAL	4.5	1.5	0.4	1.5	0.1	0.9	6.3
NORTHERN ROCKIES							
Winter	5.3	1.0	0.6	3.0	0.5	0.3	2.5
Spring	4.6	1.1	0.2	2.4	0.3	0.6	4.2
Summer	5.4	0.9	0.2	3.0	0.3	1.0	9.2
Autumn	6.7	0.9	0.3	4.3	0.6	0.6	5.7
ANNUAL	5.5	1.0	0.3	3.1	0.4	0.6	5.5
SOUTHERN CALIFORNIA							
Winter	4.6	0.5	2.2	1.2	0.2	0.4	4.2
Spring	13.6	1.7	6.9	3.2	0.6	1.2	9.8
Summer	13.8	2.4	4.6	4.2	0.8	1.8	15.2
Autumn	8.1	1.1	3.1	2.0	0.4	1.5	13.2

ANNUAL	9.8	1.4	4.2	2.5	0.5	1.2	10.4

Table 5.1 Continued

Season	Fine mass	Sulfate	Nitrate	Organics	Elemental carbon	Soil	Coarse mass
SONORA							
Winter	3.2	1.2	0.3	1.1	0.2	0.4	3.3
Spring	4.4	1.2	0.3	1.3	0.1	1.5	7.5
Summer	5.6	2.1	0.2	1.8	0.2	1.2	7.6
Autumn	4.5	1.7	0.2	1.7	0.2	0.8	5.8
ANNUAL	4.4	1.5	0.3	1.5	0.2	0.9	6.0
SIERRA							
Winter	2.5	0.4	0.7	1.1	0.1	0.2	2.1
Spring	4.3	1.0	0.6	1.7	0.2	0.8	4.8
Summer	7.2	1.7	0.6	3.6	0.5	0.9	7.0
Autumn	4.4	0.9	0.6	2.1	0.3	0.5	5.3
ANNUAL	4.5	1.0	0.6	2.1	0.3	0.6	4.7
SIERRA/HUMBOLDT							
Winter	1.7	0.2	0.1	1.0	0.1	0.3	2.9
Spring	3.0	0.6	0.2	1.4	0.1	0.6	2.9
Summer	4.0	0.7	0.2	2.2	0.3	0.6	5.6
Autumn	2.8	0.4	0.1	1.7	0.2	0.4	2.7
ANNUAL	2.9	0.5	0.2	1.6	0.2	0.5	3.7
WASHINGTON DC							
Winter	16.3	5.4	3.4	4.9	2.0	0.6	30.1
Spring	16.8	7.3	2.6	4.2	1.7	1.0	10.2
Summer	16.7	8.6	1.2	4.4	1.6	0.9	13.5
Autumn	15.3	6.6	1.6	4.4	2.0	0.8	8.4
ANNUAL	16.2	6.9	2.2	4.5	1.8	0.8	16.4
WEST TEXAS							
Winter	3.6	1.5	0.2	1.1	0.1	0.6	5.1
Spring	6.4	2.2	0.3	1.7	0.2	2.1	10.4
Summer	6.6	2.5	0.3	1.7	0.1	1.9	7.4
Autumn	4.8	2.3	0.2	1.4	0.2	0.8	7.0
ANNUAL	5.4	2.1	0.3	1.5	0.1	1.4	7.5

Table 5.2 Measured aerosol mass budgets (in percent) for the 19 regions in the IMPROVE network, averaged over the three-year period, March 1988 through February 1991.

Season	Sulfate	Nitrate	Organics	Elemental carbon	Soil
ALASKA					
Winter	42.1	6.2	36.5	3.4	11.8
Spring	39.5	3.1	30.5	2.3	24.6
Summer	20.7	1.2	57.9	3.2	16.9
Autumn	32.1	4.3	49.2	4.9	9.5
ANNUAL	32.6	3.3	43.9	3.3	17.0
APPALACHIAN					
Winter	45.8	12.8	31.3	6.2	3.8
Spring	56.8	7.9	25.1	4.4	5.8
Summer	63.5	2.0	26.5	2.9	5.1
Autumn	58.0	4.9	28.1	5.0	4.0
ANNUAL	58.0	5.7	27.2	4.2	4.8
BOUNDARY WATERS					
Winter	38.0	27.4	27.0	3.8	3.9
Spring	48.7	6.8	32.6	3.6	8.3
Summer	35.8	2.1	50.6	4.2	7.3
Autumn	37.9	10.1	40.9	4.6	6.6
ANNUAL	38.9	11.0	39.5	4.1	6.5
CASCADES					
Winter	14.6	3.5	67.2	12.0	2.7
Spring	26.7	4.7	53.2	8.8	6.7
Summer	35.7	6.1	45.1	8.1	5.0
Autumn	24.6	3.7	58.7	9.7	3.3
ANNUAL	25.7	4.5	55.7	9.5	4.5
COLORADO PLATEAU					
Winter	33.0	13.1	37.3	6.1	10.5
Spring	27.9	7.0	29.9	2.6	32.6
Summer	31.9	4.3	39.0	4.2	20.6
Autumn	36.3	4.6	38.4	5.0	15.7
ANNUAL	31.9	7.2	36.3	4.3	20.3
CENTRAL ROCKIES					
Winter	27.8	11.2	45.1	3.8	12.2
Spring	27.6	7.8	32.0	2.1	30.5
Summer	24.0	3.2	48.7	4.6	19.4
Autumn	27.9	4.5	45.4	4.3	18.0
ANNUAL	25.8	5.9	43.7	3.9	20.7
CENTRAL COAST					
Winter	16.8	29.3	44.7	6.3	2.9
Spring	33.6	18.7	36.5	4.1	7.1
Summer	43.4	17.1	31.5	2.9	5.0
Autumn	24.2	16.3	47.9	6.9	4.7
ANNUAL	28.5	21.1	40.3	5.2	4.8

Table 5.2 Continued

Season	Sulfate	Nitrate	Organics	Elemental carbon	Soil
FLORIDA					
Winter	43.3	12.5	34.0	6.9	3.2
Spring	48.5	11.2	27.4	3.7	9.2
Summer	27.1	5.9	33.3	3.4	30.2
Autumn	45.8	7.8	33.3	6.2	6.9
ANNUAL	40.9	9.2	31.9	5.0	13.0
GREAT BASIN					
Winter	25.9	12.3	48.0	1.4	12.3
Spring	22.1	5.9	35.6	1.1	35.3
Summer	14.9	2.5	38.8	2.2	41.6
Autumn	17.7	4.6	44.5	2.6	30.6
ANNUAL	18.3	4.7	40.1	2.0	34.9
HAWAII					
Winter	70.8	1.6	22.9	2.4	2.4
Spring	67.8	2.2	22.1	1.8	6.1
Summer	56.7	5.3	30.6	2.6	4.8
Autumn	72.0	1.6	22.1	2.0	2.3
ANNUAL	68.5	2.2	23.4	2.1	3.7
NORTHEAST					
Winter	50.6	11.4	27.8	7.2	3.0
Spring	58.5	7.1	24.4	5.3	4.6
Summer	52.4	4.0	35.1	4.9	3.6
Autumn	53.5	7.1	29.4	6.6	3.5
ANNUAL	53.5	7.2	29.8	5.9	3.7
NORTHERN GREAT PLAINS					
Winter	34.5	16.6	31.7	3.6	13.6
Spring	38.6	11.8	26.7	2.4	20.5
Summer	32.1	2.9	39.5	3.2	22.3
Autumn	30.0	5.2	37.1	3.6	24.1
ANNUAL	34.0	8.5	33.9	3.1	20.6
NORTHERN ROCKIES					
Winter	18.6	10.6	56.7	9.4	4.8
Spring	23.3	5.2	52.2	6.7	12.5
Summer	17.1	3.1	54.5	6.1	19.2
Autumn	12.8	4.3	64.7	9.4	8.8
ANNUAL	17.7	5.7	57.3	7.9	11.4
SOUTHERN CALIFORNIA					
Winter	11.3	47.8	26.2	5.3	9.4
Spring	12.2	51.1	23.5	4.2	8.9
Summer	17.2	33.4	30.6	5.7	13.1
Autumn	13.4	38.6	24.3	5.1	18.6
ANNUAL	13.9	43.0	25.9	4.9	12.3

Table 5.2 Continued

Season	Sulfate	Nitrate	Organics	Elemental carbon	Soil
SONORA					
Winter	38.6	8.6	34.6	5.2	13.0
Spring	26.5	6.9	29.8	2.9	33.9
Summer	37.7	3.8	33.0	3.2	22.3
Autumn	37.5	3.7	37.1	5.1	16.5
ANNUAL	35.4	5.5	33.4	4.1	21.6
SIERRA					
Winter	14.9	27.1	46.7	4.2	7.2
Spring	24.2	14.3	39.4	4.0	18.1
Summer	23.4	8.0	49.6	6.7	12.2
Autumn	20.6	13.2	48.3	6.5	11.4
ANNUAL	21.7	13.6	46.4	5.6	12.7
SIERRA/HUMBOLDT					
Winter	14.2	7.2	56.6	6.6	15.4
Spring	18.6	8.2	48.5	4.8	19.9
Summer	18.2	4.7	55.1	6.5	15.5
Autumn	15.5	3.5	59.9	7.4	13.7
ANNUAL	17.1	5.7	54.7	6.3	16.2
WASHINGTON DC					
Winter	33.2	20.9	29.9	12.4	3.6
Spring	43.6	15.5	24.9	10.1	5.9
Summer	51.4	7.4	26.1	9.8	5.3
Autumn	43.3	10.5	28.5	12.8	4.9
ANNUAL	42.4	13.8	27.5	11.4	4.9
WEST TEXAS					
Winter	40.6	6.2	31.4	3.8	18.0
Spring	33.6	4.7	26.1	2.5	33.0
Summer	38.7	4.7	25.9	2.0	28.7
Autumn	46.8	3.4	29.1	3.5	17.2
ANNUAL	39.3	4.7	27.6	2.8	25.6

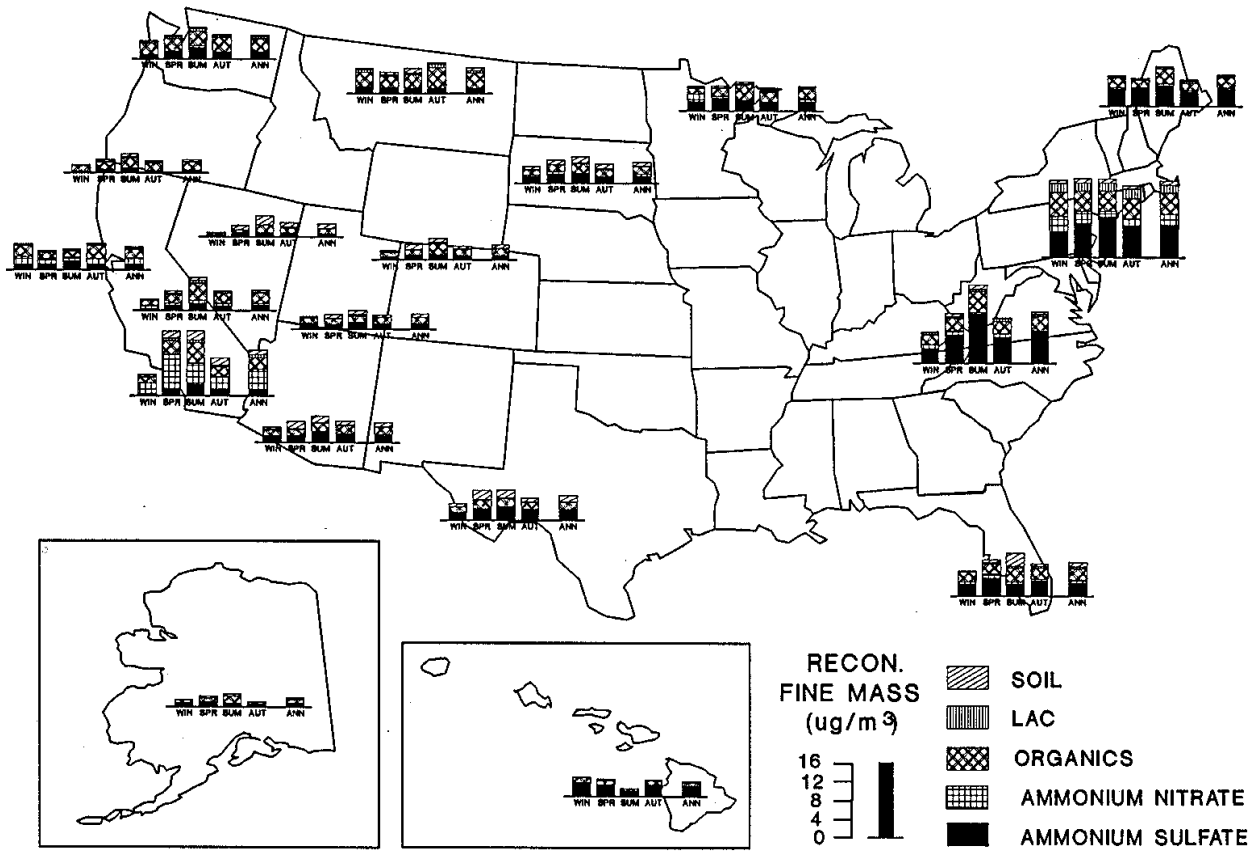

Figure 5.1 Seasonal and annual average concentrations of the fine particle mass and its components (in $\mu g/m^3$) for the 19 regions in the IMPROVE network in the United States for the three-year period, March 1988 through February 1991. For each region the bars from left to right are for winter, spring, summer, autumn, and annual averages.

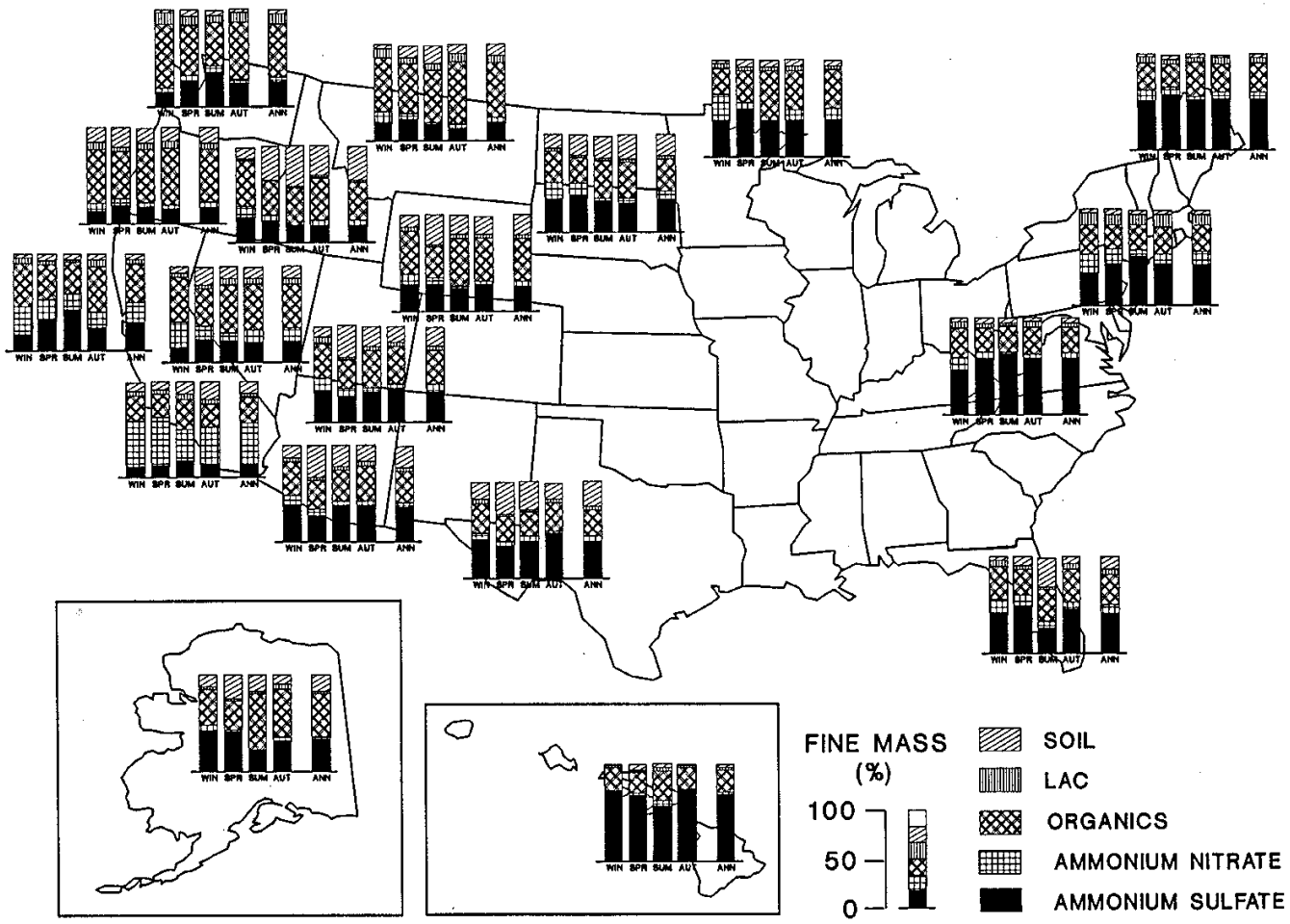

Figure 5.2 Seasonal and annual average fine particle mass budgets (in percent) for the 19 regions in the IMPROVE network in the United States for the three-year period, March 1988 through February 1991. For each region the bars from left to right are for winter, spring, summer, autumn, and annual averages.

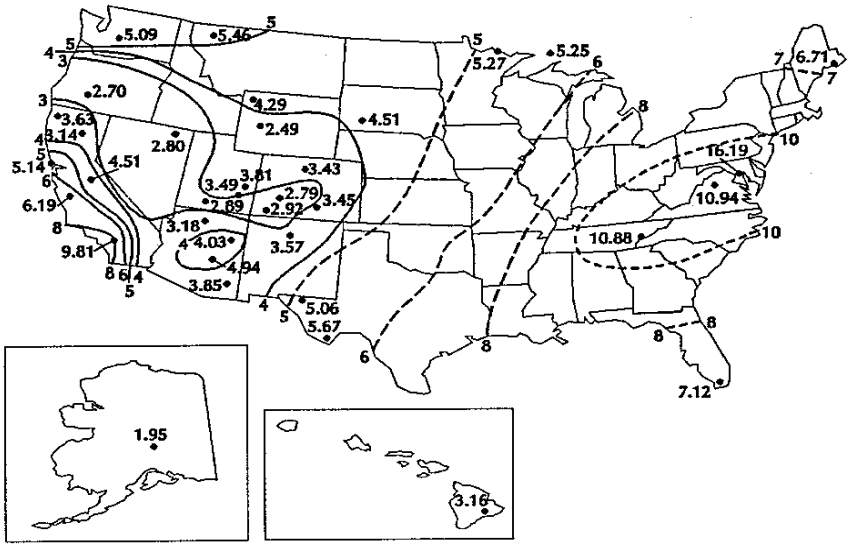

Figure 5.3 Three-year averages of fine particle mass concentrations (in $\mu m/m^3$) for each of the sites in the IMPROVE network in the U.S. for the three-year period, March 1988 - February 1991.

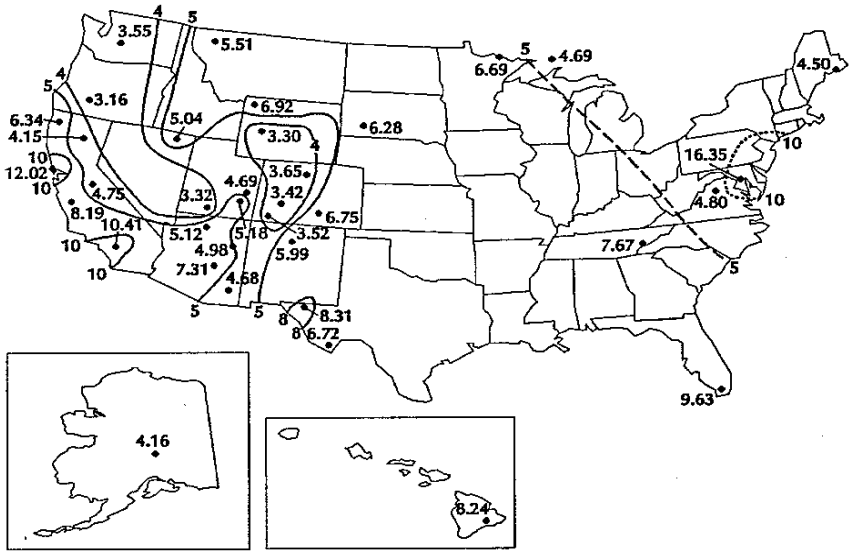

Figure 5.4 Three-year averages of coarse particle mass concentrations (in $\mu g/m_3$) for each of the sites in the IMPROVE network in the U.S. for the three-year period, March 1988 - February 1991.

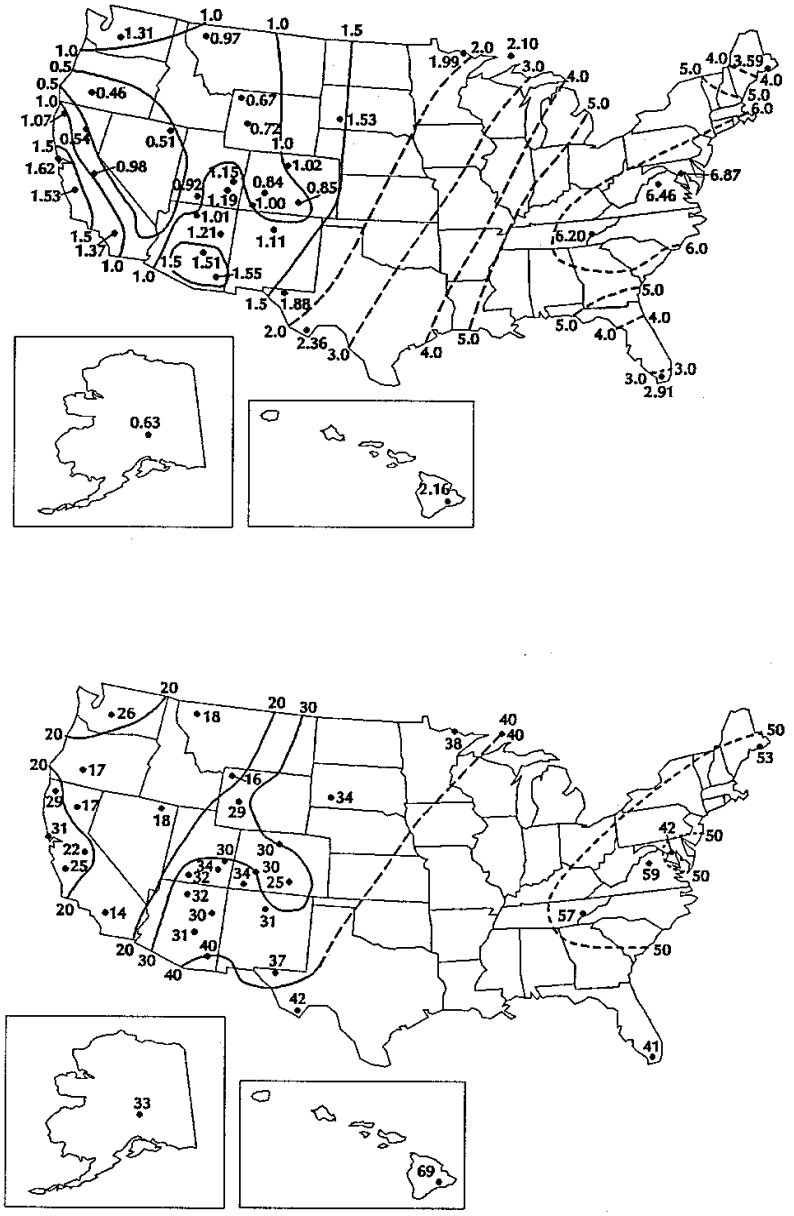

Figure 5.5 Three-year averages of fine sulfate aerosol concentrations (in $\mu g/m^3$) and sulfate fine mass fractions (in percent) for each of the sites in the IMPROVE network in the U.S. for the three-year period, March 1988 through February 1991.

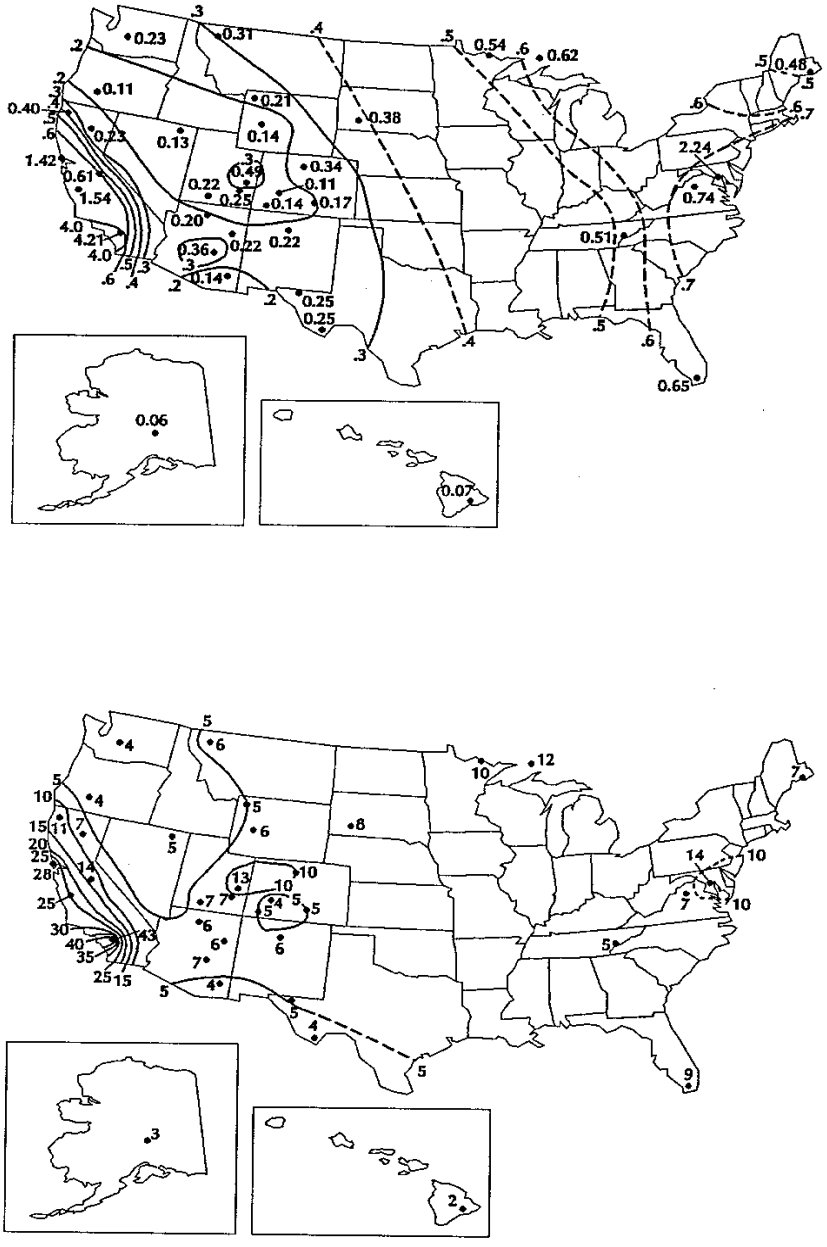

Figure 5.6 Three-year averages of fine nitrate aerosol concentrations (in $\mu g/m^3$) and nitrate fine mass fractions (in percent) for each of the sites in the IMPROVE network in the United States for the three-year period, March 1988 through February 1991.

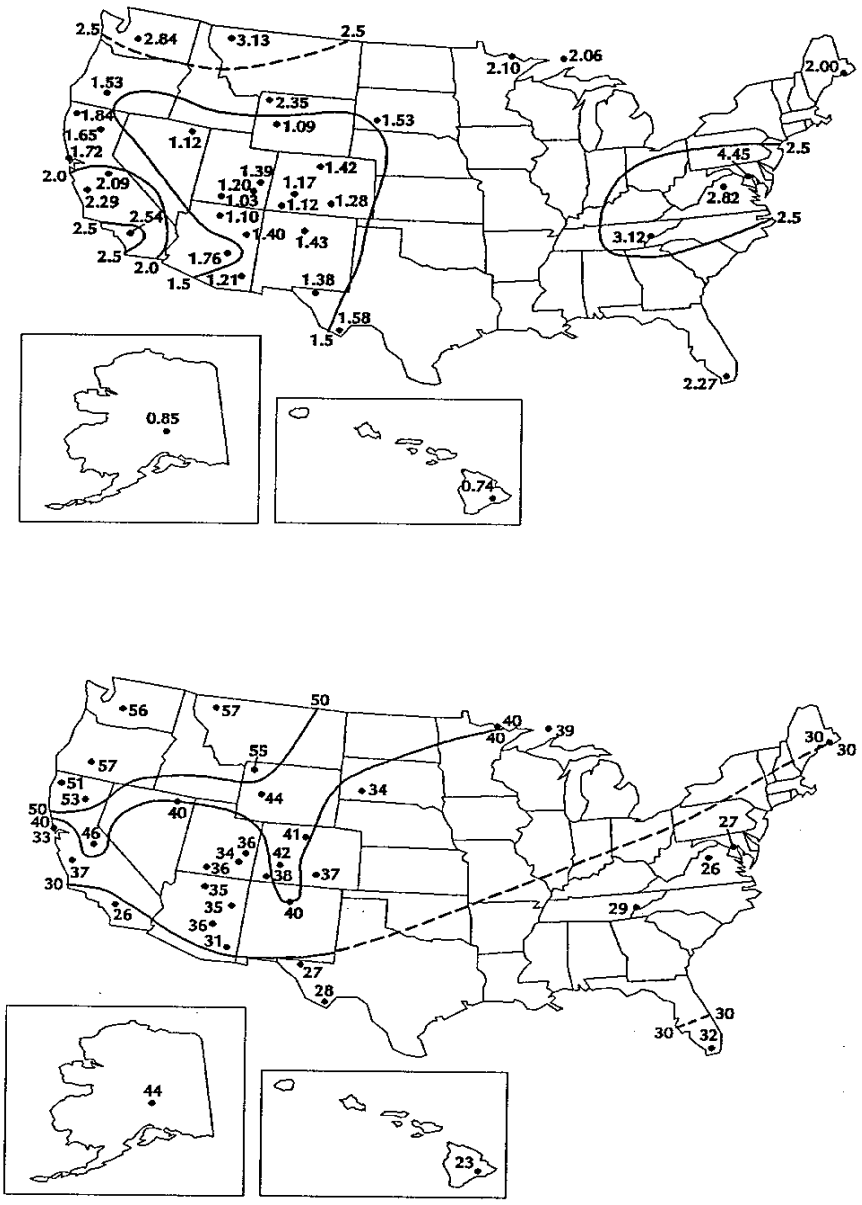

Figure 5.7 Three-year averages of fine organic carbon aerosol concentrations (in $\mu g/m^3$) and organic carbon fine mass fractions (in percent) for each of the sites in the IMPROVE network in the United States for the three-year period, March 1988 through February 1991.

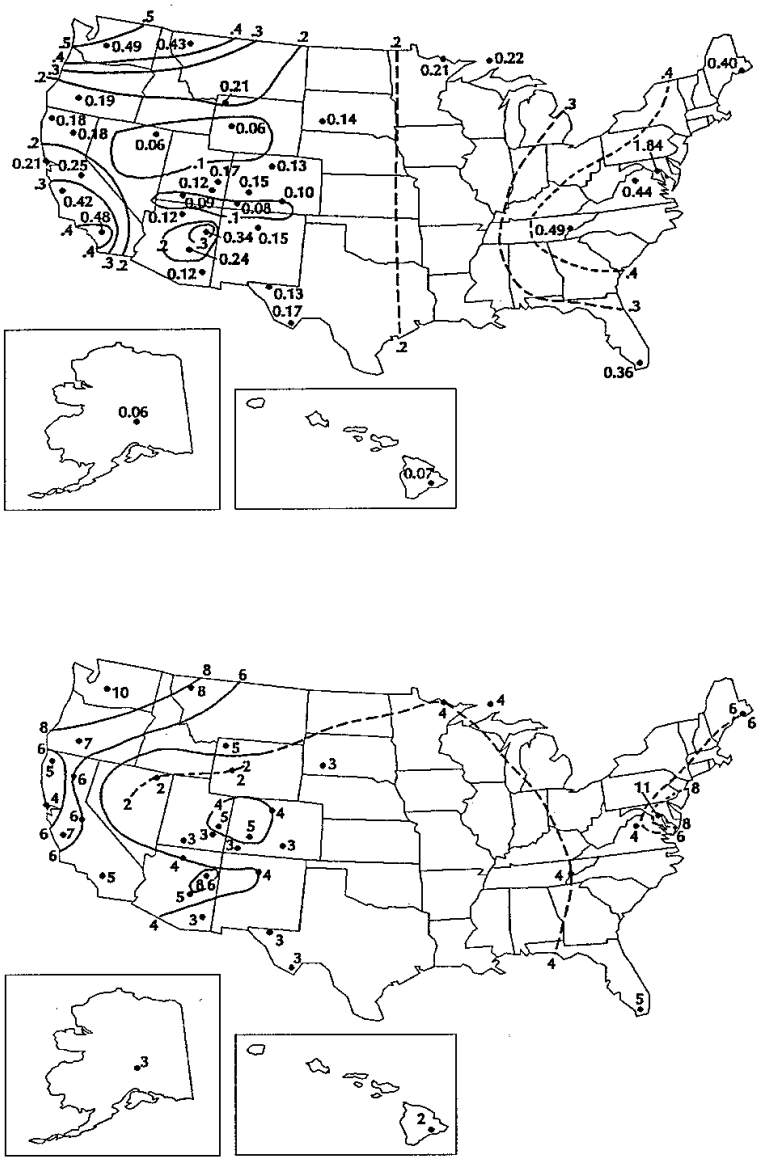

Figure 5.8 Three-year averages of fine light-absorbing carbon aerosol concentrations (in $\mu g/m^3$) and light-absorbing carbon fine mass fractions (in percent) for each of the sites in the IMPROVE network in the United States for the three-year period, March 1988 through February 1991.

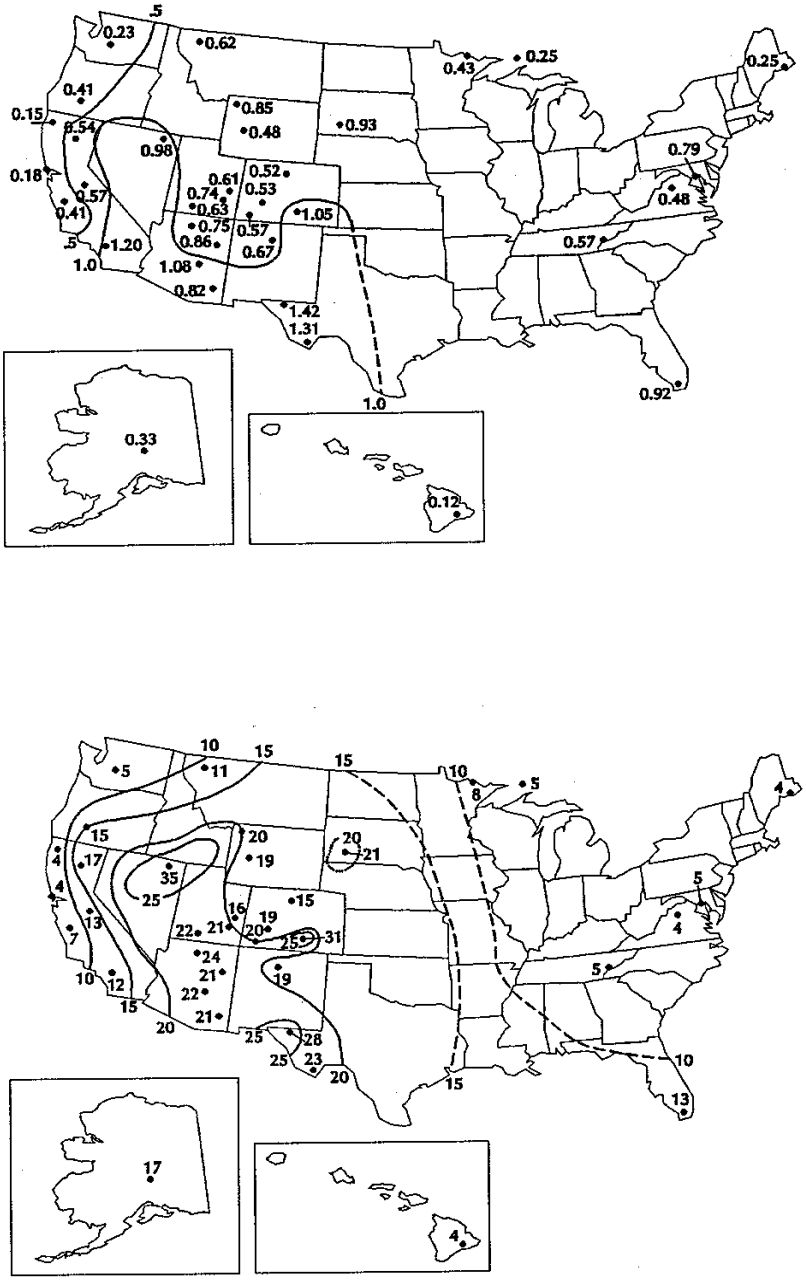

Figure 5.9 Three-year averages of fine soil aerosol concentrations (in $\mu g/m^3$) and soil fine mass fractions (in percent) for each of the sites in the IMPROVE network in the United States for the three-year period, March 1988 through February 1991.

Boundary Waters. This region, in Minnesota and Michigan, has two sets of measurements: in Isle Royale and Voyageurs National Parks. Over the three-year period, the average fine and coarse aerosol concentrations were 5.3 and 5.7 $\mu g/m^3$, respectively. Thus, the fine aerosol concentration is between the minimum measured in Alaska and the near maximumin the Appalachian Mountains. The highest fine and coarse aerosol concentrations occur during summer, but there is not as strong a seasonal variation as in Alaska and the Appalachian Mountains. In this region organic carbon is the largest fraction of fine particle mass at 40%, followed closely by sulfate (39%), and more distantly by nitrate (11%), soil (6%), and light-absorbing carbon (4%).

Cascade Mountains. This region in Washington State has only one set of measurements at Mount Rainier National Park, southeast of Seattle. Here the three-year average fine and coarse aerosol concentrations are 5.1 and 3.5 $\mu g/m^3$, respectively. Fine and coarse aerosol concentrations reach their maxima in summer and minima in winter. Sulfate and nitrate concentrations have strong seasonal variations, also with maxima in summer and minima in winter. This seasonal variation could be, in part, the result of seasonal variations in mixing and in photochemistry. In this region organic carbon is the single most significant contributor (at 56%) to fine particle mass. Sulfate (at 26%) is less than half the contribution of organics. Light-absorbing carbon contributes 10%, followed by soil (5%) and nitrate (4%).

Central Rocky Mountains. The measurements in this region are 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 3.3 and 4.8 $\mu g/m^3$ over the three-year period. Like many of the other regions, concentrations, especially of sulfate, organic carbon, light-absorbing carbon, and coarse aerosol, are highest in summer and lowest in winter. The largest contributor to fine particle mass in this region was organic carbon (44%), followed by sulfate and soil at 25% and 21%, nitrate (6%), and light-absorbing carbon (4%).

Coastal Mountains. 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 averaged 5.0 and 8.9 $\mu g/m^3$ over the three-year period. There was no strong seasonal variation in concentration, except for sulfate that had maxima and minima in summer and winter, respectively, and nitrate that showed the opposite trend, with maxima and minima in winter and summer, respectively. One would expect sulfate to reach its maximum concentrations 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. Organic carbon in this region was the largest single component of fine aerosol (at 39%), followed by sulfate and nitrate (28%, 22%), and soil and light-absorbing carbon (5% each).

Colorado Plateau. This region in the Four Corners states of the Southwest is the most intensively monitored in the IMPROVE network. There are seven sites, most of them within the so-called Golden Circle of National Parks: Arches, Bandelier, Bryce Canyon, Canyonlands, Grand Canyon, Mesa Verde, and Petrified Forest National Parks. This region is of particular concern to the newly established Grand Canyon Visibility Transport Commission as required by Congress in the 1990 amendments to the Clean Air Act. In this region fine and coarse aerosol concentrations averaged 3.4 and 4.7 $\mu g/m^3$, respectively. Fine and coarse aerosol concentrations here were greatest in summer and minimum in winter. Sulfate and organic carbon concentrations were also greatest in summer and smallest in winter. However, nitrate and light-absorbing carbon were both largest in winter. Here organic carbon and sulfate contributions are nearly equal (36% and 32%, respectively), followed by soil

(20%), nitrate (7%), and light-absorbing carbon (5%).

Florida. This region has only one monitoring site, Everglades National Park. At Everglades the fine and coarse aerosol concentrations averaged 7.1 and 9.6 $\mu g/m^3$ over the three-year period of IMPROVE. Fine and coarse aerosol concentrations were highest in summer. Fine aerosol concentrations were smallest in winter, while coarse aerosol concentrations were smallest in spring. Sulfate was found to be the largest contributor to fine particle mass (41%), followed by organic carbon (32%), soil (13%), nitrate (9%), and light-absorbing carbon (5%).

Great Basin. The Great Basin of Nevada was represented by only one set of measurements at Jarbidge Wilderness Area in northeastern Nevada. Here the fine and coarse aerosol concentrations averaged 2.8 and 5.0 $\mu g/m^3$. 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, experienced largest concentrations in the summer and lowest concentrations in the winter. The largest single contributors to fine particle mass at this site were organic carbon (40%) and soil (35%). Sulfate was a smaller contributor (18%), followed by nitrate (5%) and light-absorbing carbon (2%).

Hawaii. The Hawaiian Islands were represented by a single measurement site at Hawaii Volcanoes National Park. The fine and coarse aerosol concentrations were 3.2 and 8.2 $\mu g/m^3$, respectively. This site experienced quite a different seasonal pattern, with minimum fine aerosol concentrations in summer and maximum concentrations in winter. The sulfate, organic carbon, and light-absorbing carbon fractions of fine aerosol mass also exhibited this pattern. Coarse particle concentrations, however, had maximum concentrations in summer and minimum concentrations in winter. Sulfate was by far the largest contributor to fine particle mass, at 69%. Organic carbon contributed the next largest amount (23%). Soil, nitrate, and light-absorbing carbon were all minor contributors (4%, 2%, and 2%, respectively). Perhaps much of the sulfate measured in Hawaii is due to the natural emissions from the volcanic activity on this island.

Northeast. The northeastern United States is represented by the set of measurements at Acadia National Park on the coast of Maine. Here fine and coarse aerosol concentrations averaged 6.7 and 4.5 $\mu g/m^3$. Although fine and coarse aerosol concentrations were both largest in summer, there was not a strong seasonal variation. Sulfate, organic carbon, and soil concentrations were also largest in summer. Nitrate concentrations reached their maximum in winter. The contributors to fine particle mass included sulfate (53%), organic carbon (30%), nitrate (7%), light-absorbing carbon (6%), and soil (4%).

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 6.3 $\mu g/m^3$ over the three-year IMPROVE monitoring period. Like many sites, the maximum and minimum fine and coarse aerosol concentrations occurred in summer and winter, respectively. All of the fine aerosol constituents except nitrate also exhibited this seasonal trend. Sulfate and organic carbon each contributed 34% of the fine particle mass, followed by soil (21%), nitrate (8%), and light-absorbing carbon (3%).

Northern Rocky Mountains. This region has measurements made at Glacier National Park in Montana, close to the Canadian border. Fine and coarse aerosol concentrations averaged 5.5 $\mu g/m^3$ each here. There were no strong seasonal variations except nitrate showed a strong winter peak.

Organic carbon was by far the largest contributor to fine particle mass: 57%. Sulfate contributed 18%, soil 11%, light-absorbing carbon 8%, and nitrate 6%.

Sierra Nevada. The Sierra Nevada mountains in California were monitored at Yosemite National Park. Average fine and coarse aerosol concentrations were 4.5 and 4.7 $\mu g/m^3$. There was a strong seasonal variation, with maximum concentrations in summer and minimum concentrations in winter. The only exception was nitrate, which was relatively constant throughout the year. Organic carbon contributed more than twice what any other fine particle constituent contributed (46%). Its contribution was followed by sulfate (22%), nitrate (14%), soil (13%), and light-absorbing carbon (6%).

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 2.9 and 3.7 $\mu g/m^3$, respectively. Summer concentrations were generally about twice those during the winter. Organic carbon contributed most (55%) of the fine particle mass, with nearly equal contributions from sulfate and soil (17% and 16%) and from nitrate and light-absorbing carbon (6% each).

Sonoran Desert. This region in southeastern Arizona was monitored at two sites: Chiracahua and Tonto National Monuments. The three-year average fine and coarse mass concentrations in this region were 4.4 and 6.0 $\mu g/m^3$, respectively. These concentrations were highest in summer and lowest in winter. The sulfate, organic carbon, 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 and organic carbon (35% and 34%), followed by soil (22%), nitrate (6%), and light-absorbing carbon (4%).

Southern California. Measurements in this region were made in San Gorgonio National Monument, east of the Los Angeles metropolitan area. Fine and coarse aerosol concentrations were highest of any western U.S. site here (9.8 and 10.4 $\mu g/m^3$); concentrations were only higher in the eastern United States. Like many sites in the IMPROVE network, concentrations are 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 (43%), organic carbon (26%), sulfate (14%), soil (12%), and light-absorbing carbon (5%).

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 16.2 and 16.4 $\mu g/m^3$ over the three-year period from March 1988 through February 1991. There was not strong seasonal variation in fine aerosol concentrations; they only ranged from 15.3 $\mu g/m^3$ in autumn to 16.8 $\mu g/m^3$ in spring. 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 (42%), organic carbon (27%), nitrate (14%), light-absorbing carbon (11%), and soil (5%).

West Texas. Two measurement sites in west Texas were included in the IMPROVE network:

Big Bend and Guadalupe Mountains National Parks. Both sites are near the Mexico border in southwestern Texas. The fine and coarse aerosol concentrations averaged 5.4 and 7.5 $\mu g/m^3$ over the three-year period. Minimum concentrations generally occurred during winter, while maxima occurred in summer. The only exception was soil and coarse aerosols which peaked in spring, presumably during windy periods. The contributions to fine particle mass averaged 39% for sulfate, 28% for organic carbon, 25% for soil, 5% for nitrate, and 3% 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 organic carbon 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 and in Hawaii than in the western United States and in Alaska. Since most of the sulfate is anthropogenic in origin, regional SO_2 control would be a generally effective way to reduce fine aerosol concentrations in the United States.

5.2 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 1988 through February 1991 could be drawn. Since there are relatively fewer sites in the eastern United States, isopleths there are much less accurate; this is indicated by dashed lines. Figures 5.3 through 5.9 show isopleth maps of the three-year average aerosol concentrations (fine mass, coarse mass, sulfate, nitrate, organic carbon, light-absorbing carbon, and soil). These figures provide us with information on how aerosol concentrations and mass budgets vary over the United States.

5.2.1 Fine Aerosol

Figure 5.3 shows isopleths of the three-year average fine aerosol concentrations measured during the first three-year period of the IMPROVE network. Note the strong gradient in fine particle concentrations from Southern California, a local maximum of 9.8 $\mu g/m^3$ to the 2.7-2.9 $\mu g/m^3$ minima observed in southern Oregon, Nevada, southern Utah, and southwestern Colorado. This is a factor of 3.5 variation 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 11 $\mu g/m^3$ in Shenandoah and Great Smoky Mountain National Parks and over 16 $\mu g/m^3$ 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 variation in average concentration. Average fine aerosol concentrations in Denali National Park of 2.0 $\mu g/m^3$ in Alaska are lower than any measured in the lower 48. There is a factor of 8 variation between the average measured in Alaska and that measured in Washington, D.C.

5.2.2 Coarse Aerosol

Figure 5.4 shows isopleths of the three-year average coarse aerosol concentrations throughout the IMPROVE network. There are a few local maxima from 12 to 16 $\mu g/m^3$ 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 less than 4 ìg/m^3. However, coarse aerosol concentrations are generally in the factor-of-two range from 4 to 8 ìg/m^3. The patterns in the eastern United States are more difficult to discern. Coarse aerosol concentrations in Alaska and Hawaii are not significantly lower than in the lower 48 states. There is approximately a factor-of-five range from the lowest average concentrations measured in Oregon and Utah and the highest measured in Washington, D.C.

5.2.3 Fine Sulfate Aerosol

The average sulfate component of the fine aerosol measured over the first three-year period of the IMPROVE network is shown in Figure 5.5 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 the high concentrations in California urban areas to the 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 13.5 variation from the lowest concentration measured in Nevada to the highest concentration measured in Washington, D.C. This gradient is mostly 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 SO_2 emission densities. A relative maximum in sulfate concentration is observed in southern Arizona, which is near copper smelters that emit large quantities of SO_2. The lower map in Figure 5.5 shows that sulfate constitutes as little as 14% of fine particle mass in Southern California to as much as 59% of total fine mass in Shenandoah National Park. In the Golden Circle of parks in the Four Corners states, sulfate is 30-34% of the fine particle mass.

In the eastern United States and in Hawaii, 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).

5.2.4 Fine Nitrate Aerosol

Figure 5.6 shows isopleth maps of the nitrate concentration and nitrate mass fraction of fine aerosol, averaged over the first three years of the IMPROVE measurement program. Note that the highest concentration of 4.2 ìg/m^3 was measured in San Gorgonio Wilderness, just east of the Los Angeles metropolitan area. Other high concentrations occur in Washington, D.C. (2.2 ìg/m^3), and near the San Francisco area (1.4-1.5 ìg/m^3). There is a strong gradient from the high concentrations in the California urban areas to the minima of 0.1 ìg/m^3 measured in Oregon, Nevada, Wyoming, and Colorado. There is a local maximum of 0.4 ìg/m^3 near the Phoenix metropolitan area. The long swath of low nitrate concentrations (<0.15 ìg/m^3) extending from Oregon, Nevada, and Idaho into Utah, Wyoming, and Colorado is interrupted by higher concentrations in southeastern Utah, including a local maximum of 0.5 ìg/m^3 at Arches National Park. Nitrate mass fractions are typically 4-7 percent, except in California where they are 25 percent and higher in eastern Utah, western Colorado, Minnesota, Michigan, and in the Washington, D.C. area where they range from 10-14 percent. Nitrate concentrations generally reach their maxima in the winter when colder temperature 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 Gorgonio Wilderness.

5.2.5 Fine Organic Carbon Aerosol

Figure 5.7 shows isopleth maps of the organic carbon fraction of the fine aerosol concentration, averaged over the first three years of the IMPROVE measurement program. There is a significant spatial gradient from California and the Pacific Northwest with average concentrations of 2.0 to 3.1 $\mu g/m^3$ to the desert areas of the western United States with concentrations of 1.0 to 1.5 $\mu g/m^3$. In the eastern United States organics range generally from 2.0 to 4.5 $\mu g/m^3$. In Alaska and Hawaii organic aerosol concentrations are the lowest, from 0.7 to 0.9 $\mu g/m^3$.

Except in the northwestern United States where organic carbon is over half of the fine particle mass, organic carbon generally constitutes between 30-40 percent of the fine particle mass. Moreover, organic carbon is the largest single component of fine particle mass in most of the regions in the United States. Exceptions include the eastern United States and Hawaii where sulfate is the dominant component and Southern California where nitrate is the dominant component.

5.2.6 Fine Light-Absorbing Carbon Aerosol

Figure 5.8 shows isopleth maps of the light absorbing carbon concentration and mass fraction of the fine aerosol, averaged over the first three years of IMPROVE. Note that light absorbing carbon concentrations are highest in the Northeast, Pacific Northwest, 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 2-6 percent 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 10-11 percent of the fine particle mass.

5.2.7 Fine Soil Aerosol

Figure 5.9 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 $\mu g/m^3$) in the desert areas of the Southwest. Soil contributes approximately 5-10 percent of the fine aerosol mass, except in the desert Southwest where contributions are generally greater than 20 percent.

5.3 Summary

The following are the major patterns observed in the first three years of IMPROVE:

1. Spatial Patterns. Concentrations of fine particles (those most important in determining visibility) are highest in the eastern U.S. and in Southern California and lowest in the relatively unpopulated areas of the West.

2. Major Contributions to Fine Aerosol. The most significant components of the fine particles are organic carbon and sulfate. Of the 19 regions studied, organic carbon was the largest component in nine regions (Alaska, Cascades, Colorado Plateau, Central Rockies, Coast Mountains, Great Basin, Northern Rockies, Sierra Nevada, and Sierra-Humboldt). Sulfate was the dominant component of fine particle mass in six regions, mainly in the East (Appalachian Mountains, Florida, Hawaii, Northeast, Northern Great Plains, and Washington, D.C.). The contributions of organic carbon and sulfate were approximately equal in three regions (Boundary Waters, Sonoran Desert, and West Texas). Nitrate was the largest component of the fine aerosol in Southern California.

3. Smaller Contributors. After the contributions of organic carbon and sulfate, soil is the next largest, followed by nitrate and light absorbing carbon.

4. Seasonality. With a few exceptions, average fine mass concentrations, and the sulfate and organic carbon 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.