CHAPTER 2

AEROSOL MASS BUDGETS AND SPATIAL DISTRIBUTIONS

A detailed discussion and associated assumptions of how aerosol species mass is derived from IMPROVE aerosol measurements have been presented in Malm et al. [1994], Sisler et al. [1993], Sisler [1996] and only a summary will be presented here.

2.1 DETERMINATION OF AEROSOL SPECIES MASS

Most fine sulfates are the result of oxidation of sulfur dioxide (SO_2) gas to sulfate particles. In humid atmospheres, the oxidation typically occurs in clouds where sulfuric acid is formed within water droplets. If there is inadequate ammonia in the atmosphere to fully neutralize the sulfuric acid, as is sometimes the case, then the resulting aerosols are acidic. Under these circumstances solutions of continuously varying acidity are formed. The extremes of this continuum are ammonium sulfate (neutral) and sulfuric acid. The mass associated with the dry ammoniated sulfate ion can be estimated from independent measurements of sulfate (SO_4) and ammonium (NH_4) ions using:

$$[SULFATE] = (0.944)[NH_4^+] + (1.02)[SO_4^{2-}] \tag{2.1}$$

where [SULFATE] is the mass of the ammoniated sulfate ion. If only the sulfate ion is measured, as is the case in nearly every IMPROVE site, then one must assume a form of sulfate and multiply by an appropriate multiplication factor: 1.37*SO_4 or 4.125*S, if ammonium sulfate is assumed.

An average ambient particulate organic compound is assumed to have a constant fraction of carbon by weight. Organic carbon mass concentration from module C (OMC) is simply:

$$[OMC] = (1.4)[OC] \tag{2.2}$$

The factor of 1.4 was selected to adjust the organic carbon mass [OC] for other elements assumed to be associated with the organic carbon molecule [Watson et al., 1988].

Organic mass can also be estimated from the concentrations of H and S measured on the module A Teflon filter if certain assumptions are made [Malm et al., 1994]. It is assumed that during exposure to the vacuum of module A particle induced x-ray emission (PIXE) and proton elastic scattering (PESA) analyses all nitrates and water volatilize and do not contribute to the mass of H. It is further assumed that the remaining hydrogen can be apportioned between sulfates and organic

carbon. Assuming full neutralization of the sulfate ion, organic carbon by hydrogen (OCH) is calculated using:

$$OCH = 11(H - 0.25S) \tag{2.3}$$

The sulfur factor, H/S ratio, for ammonium sulfate is 8/32 or 0.25. The C/H ratio is 11 and operationally defined by forcing OCH to equal OC. Comparison of OCH to OC is used in data validation procedures and OCH is used to estimate organic mass when carbon is not explicitly measured.

Assuming that the collected nitrate ion is associated with fully neutralized nitrate aerosol, [NH_4NO_3], the ammonium nitrate mass, [NITRATE], is estimated from the nitrate ion mass concentration by using a multiplication factor of 1.29.

Soil mass concentration, [SOIL], is estimated by summing the elements predominantly associated with soil, plus oxygen for the common compounds (Al_2O_3, SiO_2, CaO, K_2O, FeO, Fe_2O_3, TiO_2), plus a correction for other compounds such as MgO, Na_2O, water, and carbonate [Malm et al., 1994].

The sum of the above four composites plus elemental carbon, referred to in this report as light-absorbing carbon (LAC), provides a reasonable estimate of the ambient fine mass concentration. The equation for this reconstruction of fine mass (RCFM) is then:

$$[RCFM] = [SULFATE] + [NITRATE] + [LAC] + [OMC] + [SOIL] . \tag{2.4}$$

Coarse mass (CM) is estimated gravimetrically by subtracting gravimetric fine mass ($PM_{2.5}$) concentration from total gravimetric mass (PM_{10}) concentration:

$$[CM] = [PM_{10}] - [PM_{2.5}] . \tag{2.5}$$

In the IMPROVE program, additional chemical analysis is not carried out on the coarse fraction. It is assumed that in rural or remote areas of the country the primary constituent of coarse mass is naturally occurring wind-blown dust along with some vegetative material [Noll et al., 1985; Noll, 1991].

The self-consistency and overall quality of the aerosol measurements are assured by redundancy and intercomparisons between independently measured species. A detailed description of validation and quality assurance procedures is available in Malm et al. [1994], Sisler et al. [1993], and Eldred et al. [1988]. In the most general sense, validation is a matter of comparing chemically related species that have been measured in different modules. Fortunately, the design of the IMPROVE sampler allows for redundancy between certain module A measurements and modules B and C measurements of the ions and carbons enabling quality control checks. For example, elemental sulfur mass times three should agree with the sulfate ion measured in module B. However, when comparing measured fine mass to RCFM, two complicating factors must be dealt with. First, under some conditions, a large portion of the nitrates (≥50%) can volatilize from the module A Teflon

filter; and second, the amount of residual water on the filter is a function of the relative humidity that the filter is weighed at because of water retention by soluble aerosol species.

2.2 ENSEMBLE AVERAGES

When reconstructed mass and/or extinction are presented as averages for some time period, the data must be aggregated into subsets of the larger database, then averages and related statistics are calculated for each of the subsets. There are a number of ways to aggregate the data and the strategy applied to any given aggregation is determined, at least in part, by the question being entertained. In this report, annual and seasonal aggregations of three years of data are presented. For instance, for summer averages data is aggregated over the months of June, July and August for the three years under consideration. This results in between 75 to 80 data points assuming up to nine samples per month. Other aggregations presented in this report require sorting the data into subgroups representing the days with the 20% lowest fine mass loadings, median 20%, and 20% highest fine mass loadings using measured fine mass as the sort variable. Sorting the three-year data set into three quintile yield data sets that contain fewer data points. Doing this on an annual basis yields even fewer data points, typically 18–20 observations.

Whatever criteria is used for sorting the data into subsets, at least two approaches reflecting different ways of handling missing values can be used for calculating summary statistics. The two approaches can yield significantly different results, particularly when the subsets of data are small. To understand this, consider the following extreme case; the sampling periods that correspond to the 20% highest fine mass concentrations for one season in one year of data yielding an ensemble of five data points (Table 2.1). In this example, there are two missing observations for nitrate, and one missing observation for the carbon data yielding only two observational periods without any missing values. The issue is, when reconstructing a variable such as reconstructed fine mass (RCFM): are only sampling periods with all variables present used in the reconstruction or are averages of each species calculated and then reconstructions carried out on the basis of these averages? By going with the former, (removing three observations) the RCFM is 10.75 $\mu g/m^3$ with a 14% contribution from OC. By using the second method, a RCFM value of 11.6 $\mu g/m^3$ is calculated with a 21% contribution from OC.

When removing the three observations with missing data either method yields identical results. By removing the observation with the high OC value, because nitrate is missing, skews the contribution of OC to the average RCFM for this ensemble as evidenced by the fact the measured fine mass is also highest for this observation. Other examples could be created and the question of the "best" way to do the ensemble averaging is currently under investigation and will be the subject of a forthcoming report and peer-reviewed journal articles.

In this report, the latter method is used to summarize average reconstructed mass and extinction as well as species concentrations/extinction. Specifically, by using measured fine mass as opposed to reconstructed fine mass as the sort variable larger, ensembles of data points are obtained. Then the average concentration for each species is calculated and the averages are then summed to estimate RCFM/extinction and the associated "budgets."

Table 2.1 *Five hypothetical observations that correspond to the 20% highest fine mass concentrations for one season as sorted by gravimetric fine mass (FM). Concentrations are given in $\mu g/m^3$.*

Observations	FM	Sulfate	Nitrate	Organics	Light-Absorbing Carbon (LAC)	Soil	RCFM
1	10	6	1	1	1	2	11
2	13	4	---	6	0.8	1	---
3	10	7	1.1	---	---	1	---
4	10	5	0.8	2	0.7	2	10.5
5	12	8	---	1	0.5	1	---
AVERAGE	11	6	0.96	2.5	0.75	1.4	10.75(11.61)

2.3 SPATIAL TRENDS IN AEROSOL CONCENTRATIONS

This section discusses the observed spatial variations in aerosol concentration and chemical composition throughout the United States on the basis of the IMPROVE measurements for the three-year period March 1996 through February 1999.

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. In addition, concentrations are highest when atmospheric dilution is minimal such as what occurs in stagnation periods or periods of limited mixing. Because 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 stationary sources. Organic carbon concentrations tend to be highest in regions such as the Rocky Mountains and Pacific Northwest due in part to forest management practices and forest-product industries. 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.

Average concentrations and chemical composition are calculated on the basis of measurements for each region. The heading of fine mass in the following discussion and tables is fine mass estimated from the summation of the individual aerosol species as described by Equation (2.4). The reconstructions are based on the summation of average concentrations for the time period reported as opposed to reconstructing the fine mass and then averaging. Data recovery is typically high

[Eldred et al., 1997] so a criteria for the number of data points required for a given average was not established. However, the validity of this assumption is currently being investigated and will be the topic for a future report and peer-reviewed publications.

Tables 2.2 and 2.3 show the reconstructed fine mass concentrations (RCFM) and coarse aerosol (CM) and the chemical composition (mass budgets) of the fine aerosol for each of the 21 regions in the United States, respectively. 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.

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.

2.4 CHARACTERISTICS OF THE REGIONS

Alaska. The Alaska region has only one monitoring site, Denali National Park and Preserve, and it began operation in March 1988. The average concentrations of fine and coarse aerosols over the three-year period were 1.4 and 3.0 $\mu g/m^3$, respectively. The fine aerosol concentration was the lowest measured anywhere in the United States during this period. Both fine and coarse aerosol concentrations were largest in summer and smallest in winter. Organics were the largest contributor of fine particle mass (46.5%), followed by sulfate (31.4%), soil (11.7%), light-absorbing carbon (6.7%), and nitrate (3.6%). The concentrations of organics were largest in summer, perhaps due to the prescribed burning and forest fires that usually occurred during that season. The concentrations of light-absorbing carbon were largest in autumn.

Appalachian Mountains. This region has four sites reported here: Great Smoky Mountains and Shenandoah National Parks, both initiated in March 1988, Dolly Sods Wilderness Area in West Virginia, initiated in September 1991, and Shining Rock in North Carolina, which began monitoring in September 1994.

The average concentrations of fine and coarse aerosols for this region were 9.8 $\mu g/m^3$ and 4.5 $\mu g/m^3$, respectively. Both fine and coarse aerosol concentrations were maximum in summer and minimum in winter. Sulfate was by far the largest component of the fine particle mass. At 64%, sulfate was more than twice that of the next largest contributor, organics (23.8%). Other contributors included nitrate and light-absorbing carbon (both at 3.8%), and soil (4.6%). Except for nitrate and light-absorbing carbon, which had their maximum concentrations in winter and autumn, respectively, all other species had maximum concentrations in summer. The seasonal variation in sulfate concentrations is particularly strong with summer concentrations more than three times the winter concentrations.

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 module A only.

Table 2.2 *Measured fine and coarse aerosol concentrations (in $\mu g/m^3$) for the 21 regions in the IMPROVE Network. Fine mass is reconstructed from the sum of individual species.*

Season	Reconstructed Fine Mass	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil	Coarse Mass
Alaska							
ANNUAL	1.4	0.45	0.05	0.67	0.10	0.17	3.0
Spring	1.6	0.70	0.06	0.46	0.09	0.28	3.2
Summer	2.1	0.44	0.03	1.37	0.10	0.15	3.2
Autumn	1.1	0.30	0.04	0.48	0.11	0.15	3.2
Winter	1.0	0.38	0.08	0.31	0.09	0.10	2.4
Appalachian							
ANNUAL	9.8	6.28	0.37	2.34	0.37	0.45	4.5
Spring	8.8	5.18	0.56	2.12	0.36	0.54	5.1
Summer	15.5	11.1	0.22	3.14	0.38	0.61	5.1
Autumn	9.2	5.66	0.35	2.33	0.41	0.42	4.0
Winter	5.6	2.98	0.34	1.71	0.33	0.20	3.5
Boundary Waters							
ANNUAL	4.5	1.89	0.57	1.47	0.19	0.34	3.6
Spring	4.0	1.90	0.50	1.05	0.14	0.40	3.3
Summer	5.8	2.17	0.12	2.83	0.23	0.41	4.1
Autumn	4.0	1.48	0.45	1.52	0.22	0.33	4.4
Winter	4.6	2.06	1.18	1.00	0.18	0.22	2.7
Cascade Mountains							
ANNUAL	3.4	1.01	0.21	1.66	0.27	0.28	3.2
Spring	3.0	1.04	0.20	1.16	0.20	0.36	2.6
Summer	5.3	1.83	0.29	2.42	0.36	0.38	4.5
Autumn	3.7	0.79	0.18	2.13	0.33	0.28	3.2
Winter	1.6	0.33	0.16	0.85	0.16	0.08	2.5
Central Rocky Mountains							
ANNUAL	2.6	0.81	0.16	0.96	0.13	0.54	3.4
Spring	3.0	0.87	0.22	0.86	0.12	0.90	3.7
Summer	3.4	1.01	0.14	1.44	0.15	0.62	4.0
Autumn	2.5	0.83	0.12	0.97	0.14	0.42	3.3
Winter	1.4	0.50	0.13	0.49	0.09	0.19	2.5
Colorado Plateau							
ANNUAL	3.0	1.03	0.20	0.94	0.17	0.64	4.3
Spring	3.3	0.89	0.24	0.98	0.16	1.06	5.3
Summer	3.8	1.44	0.19	1.23	0.18	0.79	4.8
Autumn	2.8	1.14	0.14	0.91	0.18	0.46	3.7
Winter	1.8	0.64	0.21	0.62	0.14	0.21	3.2
Great Basin							
ANNUAL	2.6	0.59	0.13	1.05	0.14	0.69	4.5
Spring	2.5	0.64	0.16	0.84	0.12	0.75	3.5
Summer	3.8	0.78	0.15	1.59	0.16	1.16	6.7
Autumn	2.4	0.63	0.12	0.94	0.13	0.58	4.0
Winter	1.2	0.26	0.10	0.61	0.12	0.12	2.7
Mid Atlantic							
ANNUAL	9.9	5.75	0.80	2.39	0.51	0.44	13.1
Spring	9.0	5.47	0.90	1.77	0.42	0.45	19.3
Summer	12.9	8.40	0.47	3.03	0.47	0.57	13.3
Autumn	9.0	4.92	0.69	2.38	0.59	0.44	8.2
Winter	8.8	4.36	1.08	2.47	0.58	0.30	10.6

***Table 2.2** Continued.*

Season	Reconstructed Fine Mass	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil	Coarse Mass
Mid South							
ANNUAL	11.4	6.60	0.76	2.86	0.48	0.72	5.2
Spring	10.2	5.55	0.91	2.68	0.44	0.63	5.7
Summer	15.4	9.80	0.34	3.52	0.46	1.33	6.6
Autumn	11.5	6.64	0.60	3.10	0.57	0.57	4.9
Winter	8.1	4.09	1.21	2.07	0.43	0.32	3.4
Northeast							
ANNUAL	5.3	2.96	0.32	1.56	0.25	0.25	3.7
Spring	4.5	2.55	0.33	1.14	0.21	0.30	4.0
Summer	7.8	4.70	0.21	2.37	0.28	0.28	3.6
Autumn	4.5	2.29	0.28	1.41	0.25	0.24	3.8
Winter	4.4	2.20	0.47	1.31	0.26	0.17	3.4
Northern Great Plains							
ANNUAL	4.0	1.71	0.52	1.13	0.14	0.45	5.5
Spring	4.3	1.96	0.73	0.91	0.13	0.53	5.7
Summer	4.4	1.91	0.14	1.71	0.15	0.52	6.1
Autumn	4.0	1.59	0.44	1.23	0.16	0.53	5.6
Winter	3.2	1.37	0.77	0.67	0.12	0.23	4.4
Northern Rocky Mountains							
ANNUAL	4.6	0.93	0.23	2.52	0.36	0.57	6.2
Spring	4.6	1.19	0.23	2.25	0.31	0.64	4.4
Summer	4.9	0.91	0.14	2.66	0.30	0.93	10.8
Autumn	5.5	0.84	0.20	3.48	0.51	0.52	6.1
Winter	3.3	0.78	0.35	1.70	0.31	0.18	3.3
Pacific Coastal Mountains							
ANNUAL	3.7	1.24	0.64	1.33	0.19	0.27	6.6
Spring	3.5	1.22	0.60	1.17	0.17	0.36	6.7
Summer	4.0	1.99	0.59	1.07	0.12	0.27	7.1
Autumn	4.0	1.11	0.55	1.77	0.26	0.32	7.4
Winter	3.0	0.64	0.82	1.24	0.20	0.11	5.4
Sierra-Humboldt							
ANNUAL	2.4	0.50	0.13	1.24	0.18	0.37	3.1
Spring	2.3	0.54	0.16	0.89	0.14	0.55	3.0
Summer	3.7	0.80	0.16	1.94	0.25	0.51	3.9
Autumn	2.6	0.46	0.12	1.52	0.21	0.31	2.6
Winter	0.9	0.17	0.07	0.47	0.11	0.08	2.6
Sierra Nevada							
ANNUAL	6.6	1.29	0.94	2.78	0.38	1.20	7.0
Spring	5.9	1.22	0.86	2.11	0.33	1.40	6.4
Summer	9.7	2.30	0.63	4.09	0.49	2.15	9.5
Autumn	7.2	1.18	0.93	3.56	0.47	1.09	8.6
Winter	3.7	0.50	1.27	1.48	0.23	0.22	3.6
Sonoran Desert							
ANNUAL	3.9	1.53	0.25	1.10	0.18	0.85	6.5
Spring	4.2	1.25	0.31	1.14	0.15	1.36	8.1
Summer	4.7	2.18	0.23	1.11	0.16	0.97	7.5
Autumn	4.1	1.77	0.19	1.25	0.22	0.62	5.9
Winter	2.6	0.90	0.27	0.88	0.18	0.41	4.1

Table 2.2 *Continued.*

Season	Reconstructed Fine Mass	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil	Coarse Mass
Southeast							
ANNUAL	9.8	5.22	0.40	2.86	0.51	0.81	9.5
Spring	9.9	5.61	0.49	2.73	0.50	0.60	10.4
Summer	11.3	6.12	0.32	2.71	0.35	1.86	10.8
Autumn	9.6	5.35	0.32	2.81	0.53	0.56	8.7
Winter	8.4	3.83	0.47	3.17	0.63	0.26	8.2
Southern California							
ANNUAL	7.2	1.47	2.49	2.04	0.37	0.83	6.5
Spring	9.0	1.53	3.72	2.25	0.40	1.06	6.8
Summer	9.6	2.45	2.60	3.03	0.50	1.06	8.8
Autumn	5.6	1.23	1.69	1.58	0.31	0.80	6.4
Winter	3.5	0.44	1.54	0.99	0.22	0.30	3.1
Wasatch							
ANNUAL	4.5	1.05	0.59	1.56	0.33	0.92	5.1
Spring	4.2	1.04	0.50	1.30	0.28	1.05	4.5
Summer	5.8	1.24	0.30	2.53	0.42	1.33	6.8
Autumn	3.9	1.06	0.36	1.35	0.33	0.76	4.7
Winter	3.9	0.86	1.26	1.01	0.28	0.47	4.4
Washington, D.C.							
ANNUAL	14.5	7.22	1.57	3.84	1.15	0.74	4.9
Spring	13.2	6.83	1.69	2.94	1.01	0.71	5.1
Summer	17.8	11.1	0.76	4.09	1.07	0.80	4.4
Autumn	13.9	6.12	1.64	3.99	1.29	0.85	5.9
Winter	13.1	4.68	2.25	4.36	1.26	0.59	4.4
West Texas							
ANNUAL	5.5	2.40	0.25	1.23	0.17	1.43	7.2
Spring	6.8	2.23	0.31	1.70	0.19	2.34	10.1
Summer	6.8	3.02	0.29	1.25	0.13	2.10	7.1
Autumn	5.0	2.91	0.15	1.12	0.18	0.60	5.3
Winter	3.3	1.53	0.23	0.79	0.16	0.61	5.9

Table 2.3 *Measured fine aerosol mass budgets (in %) for the 21 regions in the IMPROVE Network.*

Season	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil
Alaska					
ANNUAL	31.4	3.6	46.5	6.7	11.7
Spring	44.2	3.6	28.9	5.8	17.5
Summer	20.9	1.6	65.4	4.8	7.3
Autumn	27.9	3.3	44.7	9.9	14.2
Winter	39.6	8.7	32.3	9.2	10.2
Appalachian					
ANNUAL	64.0	3.8	23.8	3.8	4.6
Spring	59.1	6.3	24.2	4.1	6.2
Summer	71.9	1.4	20.2	2.5	4.0
Autumn	61.7	3.8	25.4	4.5	4.6
Winter	53.6	6.1	30.8	5.9	3.6

Table 2.3 *Continued.*

Season	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil
Boundary Waters					
ANNUAL	42.4	12.8	33.0	4.2	7.7
Spring	47.5	12.5	26.3	3.6	10.0
Summer	37.7	2.2	49.1	3.9	7.1
Autumn	37.0	11.2	38.1	5.4	8.2
Winter	44.4	25.4	21.5	3.8	4.8
Cascade Mountains					
ANNUAL	29.5	6.1	48.6	7.7	8.0
Spring	35.1	6.8	39.2	6.8	12.1
Summer	34.8	5.5	45.8	6.7	7.2
Autumn	21.3	4.8	57.4	8.9	7.6
Winter	21.1	10.1	53.8	10.2	4.8
Central Rocky Mountains					
ANNUAL	31.1	6.0	37.1	4.9	20.9
Spring	29.2	7.5	28.9	4.0	30.3
Summer	30.0	4.2	43.0	4.4	18.4
Autumn	33.5	5.0	39.0	5.7	16.9
Winter	35.7	9.3	35.2	6.4	13.4
Colorado Plateau					
ANNUAL	34.6	6.6	31.7	5.6	21.6
Spring	26.7	7.1	29.5	4.8	31.9
Summer	37.6	4.9	32.2	4.8	20.6
Autumn	40.3	5.0	32.2	6.2	16.4
Winter	35.1	11.7	34.1	7.8	11.3
Great Basin					
ANNUAL	22.6	5.1	40.4	5.3	26.6
Spring	25.6	6.4	33.6	4.7	29.8
Summer	20.3	3.9	41.5	4.1	30.1
Autumn	26.4	5.0	39.1	5.5	24.0
Winter	21.4	8.3	50.2	10.1	10.1
Mid Atlantic					
ANNUAL	58.1	8.1	24.2	5.2	4.4
Spring	60.6	10.0	19.6	4.7	5.0
Summer	65.0	3.6	23.4	3.6	4.4
Autumn	54.6	7.7	26.3	6.5	4.9
Winter	49.6	12.3	28.1	6.6	3.4
Mid South					
ANNUAL	57.8	6.6	25.0	4.2	6.3
Spring	54.4	8.9	26.3	4.4	6.2
Summer	63.5	2.2	22.8	3.0	8.6
Autumn	57.8	5.3	27.0	4.9	5.0
Winter	50.3	15.0	25.5	5.3	3.9
Northeast					
ANNUAL	55.4	6.0	29.2	4.7	4.7
Spring	56.3	7.3	25.1	4.6	6.7
Summer	60.0	2.7	30.2	3.6	3.6
Autumn	51.3	6.3	31.4	5.7	5.3
Winter	50.0	10.6	29.7	5.9	3.9

***Table 2.3** Continued.*

Season	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil
Northern Great Plains					
ANNUAL	43.2	13.2	28.6	3.6	11.4
Spring	46.0	17.3	21.3	3.0	12.4
Summer	43.1	3.2	38.5	3.5	11.7
Autumn	40.2	11.2	31.1	4.1	13.3
Winter	43.3	24.4	21.2	3.8	7.4
Northern Rocky Mountains					
ANNUAL	20.1	5.0	54.7	7.8	12.3
Spring	25.8	5.1	48.5	6.8	13.9
Summer	18.4	2.8	53.8	6.1	18.8
Autumn	15.1	3.5	62.8	9.3	9.3
Winter	23.4	10.7	51.2	9.4	5.3
Pacific Coastal Mountains					
ANNUAL	34.0	17.5	36.2	5.1	7.3
Spring	34.6	17.1	33.3	4.7	10.3
Summer	49.4	14.6	26.5	2.9	6.6
Autumn	27.8	13.6	44.2	6.4	7.9
Winter	21.4	27.2	41.0	6.6	3.8
Sierra-Humboldt					
ANNUAL	20.7	5.3	51.2	7.5	15.3
Spring	23.6	6.9	39.3	6.2	24.1
Summer	21.7	4.5	53.0	6.8	14.0
Autumn	17.3	4.6	58.1	8.1	11.8
Winter	18.7	7.5	52.5	12.6	8.7
Sierra Nevada					
ANNUAL	19.6	14.3	42.1	5.7	18.2
Spring	20.7	14.5	35.6	5.5	23.7
Summer	23.8	6.5	42.3	5.0	22.3
Autumn	16.3	12.8	49.3	6.5	15.1
Winter	13.6	34.4	39.8	6.3	6.0
Sonoran Desert					
ANNUAL	39.2	6.4	28.1	4.5	21.8
Spring	29.6	7.3	27.1	3.7	32.3
Summer	46.9	4.9	24.0	3.4	20.9
Autumn	43.7	4.7	30.9	5.3	15.4
Winter	34.1	10.1	33.4	7.0	15.4
Southeast					
ANNUAL	53.3	4.1	29.2	5.2	8.3
Spring	56.5	4.9	27.5	5.1	6.1
Summer	53.9	2.8	23.8	3.1	16.4
Autumn	55.9	3.3	29.3	5.5	5.9
Winter	45.8	5.6	38.0	7.6	3.1
Southern California					
ANNUAL	20.4	34.7	28.3	5.1	11.5
Spring	17.1	41.6	25.1	4.5	11.8
Summer	25.4	26.9	31.4	5.2	11.0
Autumn	21.9	30.2	28.1	5.5	14.2
Winter	12.6	44.2	28.3	6.3	8.6

Table 2.3 *Continued.*

Season	Sulfate	Nitrate	Organics	Light-Absorbing Carbon	Soil
Wasatch					
ANNUAL	23.6	13.3	35.1	7.3	20.6
Spring	24.9	12.1	31.3	6.7	25.1
Summer	21.3	5.2	43.5	7.2	22.9
Autumn	27.4	9.2	35.0	8.6	19.8
Winter	22.1	32.6	26.0	7.1	12.1
Washington, D.C.					
ANNUAL	49.7	10.8	26.4	7.9	5.1
Spring	51.8	12.9	22.3	7.6	5.4
Summer	62.3	4.2	23.0	6.0	4.5
Autumn	44.1	11.8	28.7	9.3	6.1
Winter	35.6	17.1	33.2	9.6	4.5
West Texas					
ANNUAL	43.9	4.6	22.4	3.0	26.1
Spring	32.9	4.6	25.1	2.8	34.5
Summer	44.4	4.3	18.4	1.9	30.9
Autumn	58.7	3.0	22.6	3.7	12.0
Winter	46.0	6.9	23.7	4.9	18.5

The average fine and coarse aerosol concentrations were 4.5 and 3.6 $\mu g/m^3$, respectively. The highest concentrations occurred during summer for fine aerosols and in the autumn for coarse aerosols, 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 (42.4%), followed by organics (33%), and more distantly by nitrate (12.8%), soil (7.7%), and light-absorbing carbon (4.2%).

Cascade Mountains. This region in the states of Washington and Oregon has three monitoring sites reported here. Mount Rainier National Park southeast of Seattle was initiated in March 1988, Three Sisters Wilderness Area on the Willamette National Forest, and Snoqualmie Pass on the Snoqualmie National Forest were implemented in July 1993 and became fully operational September 1994.

Here the average fine and coarse aerosol concentrations were 3.4 and 3.2 $\mu g/m^3$, respectively. Fine and coarse aerosol concentrations reached their maxima in summer and minima in winter. Sulfate and nitrate concentrations had strong seasonal variations, 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, organics were the single most significant contributor (48.6%) to fine particle mass. Sulfate (29.5%) contributed much less than organics. Nitrate contributed 6.1%, followed by soil (8%) then light-absorbing carbon (7.7%).

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 2.6 and 3.4 $\mu g/m^3$, respectively. 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 (37.1%), followed by sulfate (31.1%), soil (20.9%), nitrate (6.0%), and light-absorbing carbon (4.9%).

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. A seventh site, Arches National Park, was discontinued in May 1992. In this region, fine and coarse aerosol concentrations averaged 3.0 and 4.3 $\mu g/m^3$, respectively. Fine and coarse aerosol concentrations were greatest in summer and spring, respectively, and least in winter. Concentrations of sulfate, organics, and light-absorbing carbon were also greatest in summer and smallest in winter. However, nitrate was highest in the spring and lowest in the autumn. Sulfate (34.6%) and organics (31.7%) contributed the most followed by soil (21.6%), nitrate (6.6%), and light-absorbing carbon (5.6%).

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. The fine and coarse aerosol concentrations averaged 2.6 and 4.5 $\mu g/m^3$, respectively. The fine mass concentration was the lowest of any of the regions in the contiguous 48 states. Perhaps this was due to the fact that this site was relatively remote from high emission density areas and was generally well ventilated. Both fine and coarse aerosol concentrations, as well as all of the fine aerosol components, experienced largest concentrations in the summer (except nitrate) and lowest concentrations in the winter (both winter and spring for light-absorbing carbon). The largest single contributors to fine particle mass at this region were organics (40.4%) and soil (26.6%). Sulfate was a smaller contributor (22.6%), followed by light-absorbing carbon (5.3%) and nitrate (5.1%).

Mid Atlantic. This region is represented by the Edwin B. Forsythe National Wildlife Refuge west of Atlantic City, New Jersey and began monitoring in September 1991. Fine and coarse aerosol concentrations averaged 9.9 and 13.1 $\mu g/m^3$, respectively. A moderate seasonality was evident with the highest fine and coarse aerosol concentrations occurring in the summer and spring, and the least in the winter and autumn, respectively. Sulfate, organics, and fine soil follow the same seasonal trend as for fine aerosol mass. Nitrates peaked in the winter at about two times its summer concentration, and light-absorbing carbon peaked in the winter as well but only showed a small seasonality. Sulfate comprises the bulk of the fine aerosol mass (58.1%) followed by organics (24.2%), nitrate (8.1%), light-absorbing carbon (5.2%), and soil (4.4%).

Mid South. Three sites are monitored for this 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 aerosols was 11.4 and 5.2 $\mu g/m^3$, respectively. Outside of Washington, D.C., which is an urban site, this region had the highest average concentration of fine aerosol. A modest seasonality was evident for fine and coarse aerosols with the minima occurring in the winter and the maxima in the summer. All fine aerosol constituents except nitrate and light-absorbing carbon follow the seasonality of fine aerosol. Nitrate had its maximum concentrations in the winter, while light-absorbing carbon was highest in the autumn. Sulfate (57.8%) composed the

bulk of fine aerosol followed by organics (25.0%), nitrate (6.6%), soil (6.3%), and light-absorbing carbon (4.2%).

Northeast. The northeastern United States is represented by measurements at three sites: Acadia National Park on the coast of Maine, which began monitoring in March 1988, Lye Brook Wilderness Area in southern Vermont, which began in September 1991, and Moosehorn National Wildlife Refuge on the border of Maine and New Brunswick, which began in December 1994. Fine and coarse aerosol concentrations averaged 5.3 and 3.7 $\mu g/m^3$, respectively. Although fine concentrations were largest in summer and least in winter, there was not a strong seasonal variation for coarse aerosol concentrations. Sulfate, organics, and light-absorbing carbon concentrations were also largest in summer. Nitrate concentrations reached their maximum in winter. The contributors to fine particle mass included sulfate (55.4%), organics (29.2%), nitrate (6.0%), light-absorbing carbon (4.7%), and soil (4.7%).

Northern Great Plains. Only one set of measurements was made in this region, at Badlands National Park in South Dakota. Fine and coarse aerosol concentrations averaged 4.0 and 5.5 $\mu g/m^3$, respectively. The maximum concentrations for fine mass and coarse mass occurred in the summer and were least in the winter. Sulfate (43.2%) contributed the most to fine mass and second were organics (28.6%), followed by nitrate (13.2%), soil (11.4%), and light-absorbing carbon (3.6%).

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 4.6 and 6.2 $\mu g/m^3$, respectively. The strongest seasonality was shown by nitrate, with a significant winter peak, and coarse mass, which peaked in the summer. Sulfates peaked in the spring and organics and light-absorbing carbon peaked in the autumn. Organics were by far the largest contributor to fine particle mass (54.7%) followed by sulfate (20.1%), soil (12.3%), light-absorbing carbon (7.8%), and nitrate (5.0%).

Pacific 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 Redwood National Park. In this region, the fine and coarse aerosol concentrations averaged 3.7 and 6.6 $\mu g/m^3$, respectively. There was no strong seasonal variation for fine and coarse concentrations, however, sulfate concentration was greatest in summer and least in winter, while nitrate showed the opposite trend, peaking in the winter and low in summer. Organics in this region were the largest single component of fine aerosol (36.2%), followed by sulfate (34%), nitrate (17.5%), soil (7.3%), and light-absorbing carbon (5.1%).

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 Volcanic 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 2.4 and 3.1 $\mu g/m^3$, respectively. Summer concentration of fine mass was generally about four times than during the winter. Organics contributed most of the fine particle mass (51.2%), followed by sulfate (20.7%), soil, (15.3%), light-absorbing carbon (7.5%), and nitrate (5.3%).

Sierra Nevada. The Sierra Nevada Mountains in California were monitored at two sites: Yosemite and Sequoia National Parks. Yosemite National Park has been monitored since March 1988. Sequoia National Park had modules A and D since March 1992 but was not fully instrumented until July 1993. Average fine and coarse aerosol concentrations were 6.6 and 7.0 $\mu g/m^3$, respectively. There was a strong seasonal variation of fine and coarse mass, with maximum concentrations in summer and minimum concentrations in winter. Sulfate concentrations, followed by organics, showed the largest seasonal variation between summer and winter, while nitrate concentration was greatest in the winter and least in summer. On a relative basis organics contributed more than twice what sulfate contributed (42.1% and 19.6%, respectively). Soil was the next largest contributor (18.2%), followed by nitrate (14.3%), and light-absorbing carbon (5.7%).

Sonoran Desert. This region in southeastern Arizona was monitored at two sites: Chiricahua and Tonto National Monuments that were initiated in March 1988. The three-year averages of fine and coarse mass concentrations in this region were 3.9 and 6.5 $\mu g/m^3$, respectively. These concentrations were highest in spring, summer, and fall and lowest in winter. The sulfate, organics, and soil components of fine particle mass had maxima in the summer, autumn, and spring, respectively, and minima in the winter. The contributions to fine particle mass were distributed between sulfate (39.2%) then organics (28.1%), followed by soil (21.8%), nitrate (6.4%), and light-absorbing carbon (4.5%).

Southeast. Previously, this region was designated as Florida, and is now represented by three sites at Chassahowitzka National Wildlife Refuge on the Gulf Coast north of Tampa, Florida, Okefenokee National Wildlife Refuge an inland site on the Georgia-Florida border, and Cape Romain National Wildlife Refuge on the South Carolina coast. Monitoring at these three sites began in April 1993, September 1991, and September 1994, respectively. The fine and coarse aerosol concentrations averaged 9.8 and 9.5 $\mu g/m^3$, respectively, with their concentrations highest in summer and lowest in winter. Sulfate and soil concentrations were greatest in the summer and least in the winter. Organics and light-absorbing carbon had their greatest concentrations in the winter, while nitrate concentrations were greater in the spring. Sulfate was found to be the largest contributor to fine particle mass (53.3%), followed by organics (29.2%), soil (8.3%), light-absorbing carbon (5.2%), and nitrate (4.1%).

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 7.2 and 6.5 $\mu g/m^3$, respectively. Like many sites in the IMPROVE Network, concentrations were highest in summer and lowest 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 (34.7%), organics (28.3%), sulfate (20.4%), soil (11.5%), and light-absorbing carbon (5.1%).

Wasatch. This area is monitored at Lone Peak Wilderness Area above Provo, Utah to the north and east in the Wasatch Mountain Range. Monitoring began in December 1993 but suffered equipment failure in September 1994. It has since operated reliably. Fine and coarse aerosol concentrations averaged 4.5 and 5.1 $\mu g/m^3$, respectively, and were highest in the summer and least during winter. Concentrations of sulfate, organics, light-absorbing carbon, and soil were also highest in the summer and least in the winter. Nitrate, however, was exactly the opposite with

winter concentrations being more than four times as in summer. On average, organics comprised the bulk of fine mass (35.1%), followed by sulfate (23.6%), soil (20.6%), nitrate (13.3%), and light-absorbing carbon (7.3%).

Washington, D.C. This is a single monitoring site in the nation's capital, the only urban site of the network. Fine aerosol concentrations were higher here than anywhere in the IMPROVE Network. Fine and coarse mass concentrations averaged 14.5 and 4.9 $\mu g/m^3$, respectively. There was a moderate seasonal variation in fine aerosol concentrations; they ranged from 13.1 to 17.8 $\mu g/m^3$ 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. Fine particle mass consisted of sulfate (49.7%), organics (26.4%), nitrate (10.8%), light-absorbing carbon (7.9%), and soil (5.1%).

West Texas. Two measurement sites in west Texas were included: Big Bend and Guadalupe Mountains National Parks on or near the Mexico border in southwestern Texas, respectively, and have operated since March 1988. The fine and coarse aerosol concentrations averaged 5.5 and 7.2 $\mu g/m^3$, respectively. Minimum concentrations generally occurred during winter, while maximum concentrations occurred in summer for fine mass and spring for coarse mass. All components of fine mass showed seasonal variability except light-absorbing carbon, which remained relatively constant. Nitrate, and soil concentrations peaked during the spring, while sulfate and organics were lowest in the winter and nitrate was lowest in the autumn. Concentrations of organics were highest in the spring, while those of sulfates were highest in the summer. The contributions to fine particle mass was sulfate (43.9%), soil (26.1%), organics (22.4%), nitrate (4.6%), and light-absorbing carbon (3%).

In general, the following observations can be made. With few exceptions, aerosol concentrations were highest in summer and lowest in winter. In the eastern United States, sulfates contributed most to fine mass, while in southern California nitrates were the single largest contributor. In the desert Southwest, carbon, sulfates, and soil all contributed about equally to fine mass, while in the Northwest carbon and sulfate were the largest contributors.

2.5 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 1996 through February 1999 could be drawn. Figures 2.1 through 2.8 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. Because Washington, D.C. is an urban site it is not included in the isopleth presentations.

2.5.1 PM_{10} Aerosol

Figure 2.1 shows isopleths of the PM_{10} gravimetric mass concentration measured during this three-year period. The highest concentrations occurred in the eastern United States. All the areas

east of the Mississippi River had concentrations in excess of 8 μg/m^3. The highest concentrations were in Edwin B. Forsythe National Wildlife Refuge in New Jersey at 23 μg/m^3, followed by the Southeast region, Sequoia National Park, and the Mid South, which experienced concentrations in excess of 15 μg/m^3. Outside of southern California and the Northern Rockies the least amount of PM_{10} concentrations occurred in the western United States, where there was a large swath extending from Oregon, northern California, Nevada, Utah, Wyoming, into northern Arizona, northern New Mexico and western Colorado, where the concentration of PM_{10} was less than 8.0 μg/m^3. The lowest concentration in the contiguous 48 states occurred at Lassen Volcanic National Park at only 5.1 μg/m^3 on average; the least was recorded at Denali National Park in Alaska at 4.5 μg/m^3. The strongest gradients were between regions of California and Nevada, where concentrations varied from 5.9 μg/m^3 at Great Basin National Park to an excess of 19 μg/m^3 at Sequoia National Park and Edwin B. Forsythe National Wildlife Refuge and Lye Brook Wilderness Area, where concentrations decreased from 23 to 8.2 μg/m^3.

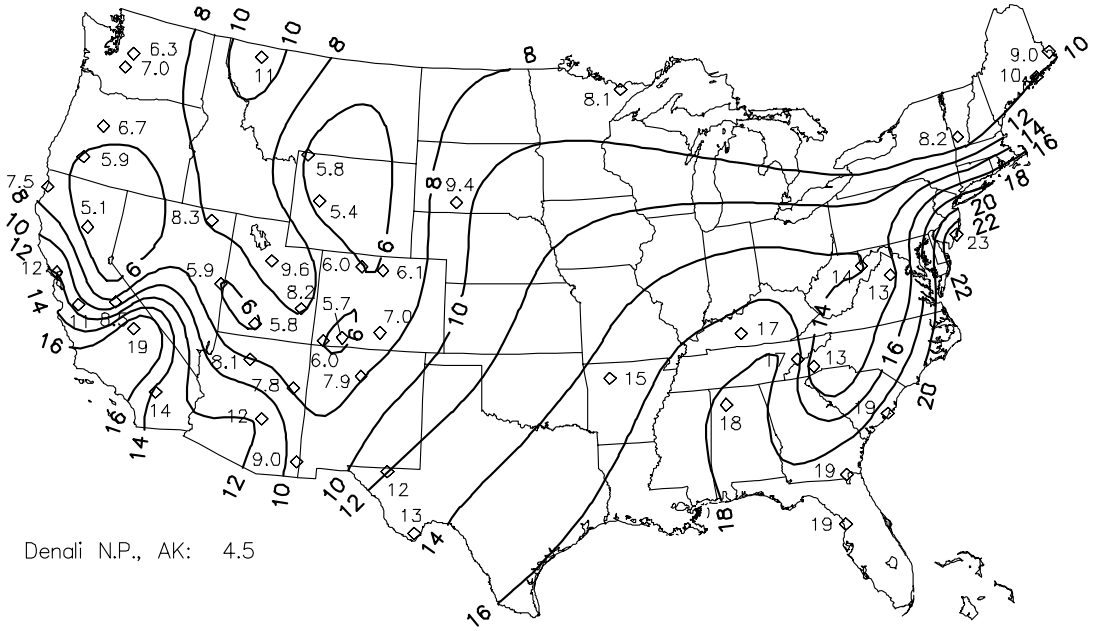

Figure 2.1 *Average PM_{10} mass concentrations (in μg/m^3) for each site in the IMPROVE Network, excluding Washington, D.C.*

2.5.2 Fine Aerosol

Figure 2.2 shows isopleths of the average reconstructed fine aerosol concentrations measured during the three-year period. Note the strong gradient in fine particle concentrations from southern California, a local maximum of 8.9 μg/m^3 to minima of 2.2 to 2.7 μg/m^3 observed in southern Oregon, Nevada, southern Utah, western Colorado, and Wyoming. This is a factor of four variations in average fine aerosol concentration. Also, note that fine aerosol concentrations increased again as one moves to the eastern United States with levels in excess of 9 μg/m^3 in Mammoth Cave National Park and Upper Buffalo and Sipsy Wilderness Areas. Thus, from the

minima in the western United States to the maxima in the eastern United States, there was about a factor of six difference in average concentration.

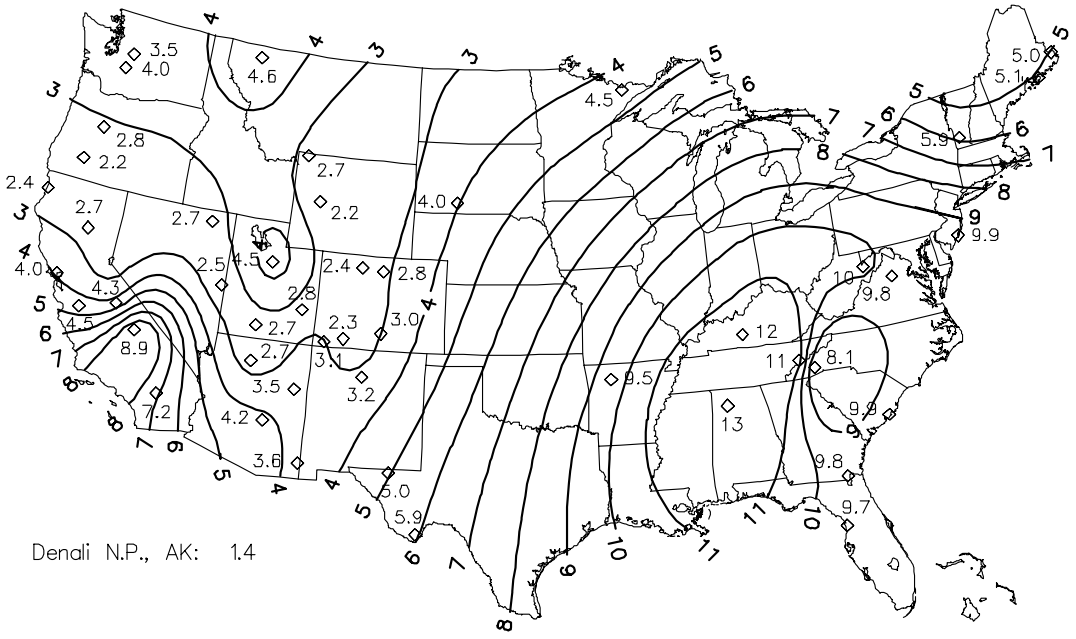

Figure 2.2 *Average fine mass aerosol concentrations (in μg/m^3) for each site in the IMPROVE Network, excluding Washington, D.C.*

2.5.3 Coarse Aerosol

Figure 2.3 shows isopleths of the three-year average coarse mass (CM) concentrations. There were a few local maxima from 7.5 to 13 μg/m^3 that were noticeable near southern Arizona, southern California, San Francisco, and the New Jersey coast. The lowest coarse aerosol concentrations occurred in the swath from the Pacific Northwest through Nevada to southern Utah. Concentrations in this region averaged around 3.5 μg/m^3. Throughout the United States coarse aerosol concentrations were generally in the factor-of-four range from 3 to 11 μg/m^3. The patterns in the eastern United States showed a steady north-south trend of increasing coarse aerosol concentrations with steepest gradients near the coast. Coarse aerosol concentrations in Alaska were not significantly lower than in the contiguous 48 states.

2.5.4 Fine Sulfate Aerosol

The average sulfate component of the fine aerosol measured over the three-year period is shown in Figure 2.4. Since sulfate is one of the two major components of fine particle mass, it was not surprising to observe gradients across the United States similar to what was observed for total fine particle mass. There was a strong gradient from high concentrations in southern California to low concentrations in southern Oregon and Nevada. There was also a strong gradient from the relatively low concentrations in the West to those in the East. There was about a factor of 18 between the lowest concentrations measured in Nevada and Oregon to the highest concentrations measured in

the Mid-South and Southern Appalachian Mountains. A relative maximum in sulfate concentration was observed in southern Arizona. The lower map in Figure 2.4 shows that sulfate constituted as little as 19% of fine particle mass in the Sierra-Nevada area to as much as 66% of total fine mass in Shining Rock Wilderness Area in North Carolina. On the Colorado Plateau sulfate was 31 to 37% of the fine particle mass.

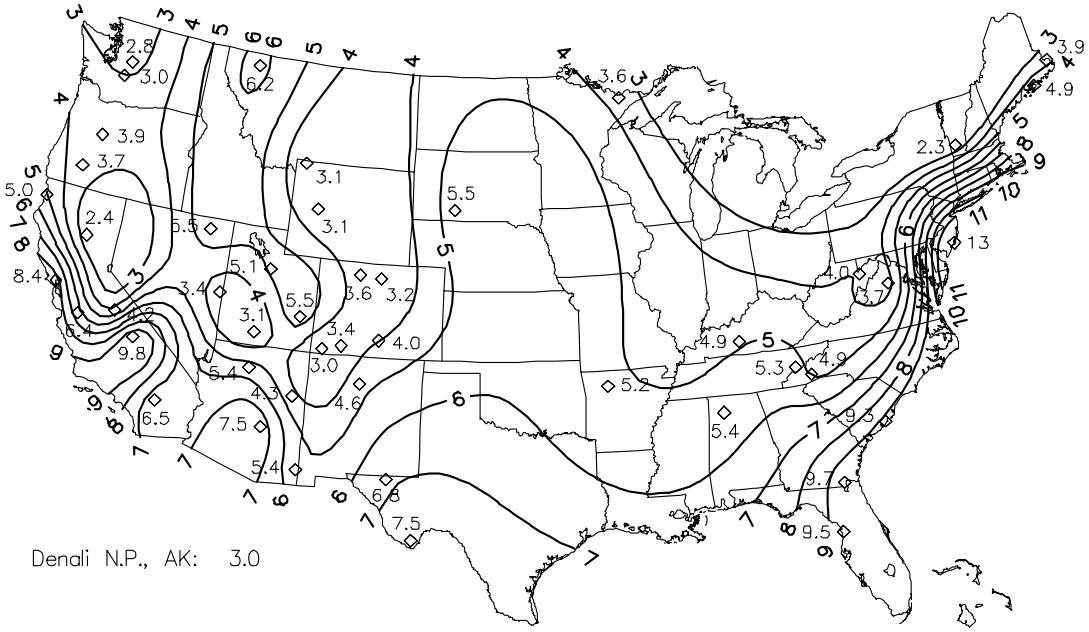

Figure 2.3 *Average coarse particle mass concentrations (in μg/m^3) for each site in the IMPROVE Network, excluding Washington, D.C.*

In the eastern United States, sulfate was the largest single component of fine particle mass. In the Boundary Waters, Sonoran Desert, and West Texas regions, sulfate is still the largest contributor to fine mass, however, it is followed closely by organic carbon. Sulfate was 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.

2.5.5 Fine Nitrate Aerosol

Figure 2.5 shows isopleth maps of the nitrate concentration and nitrate mass fraction of fine aerosol, averaged over the three-year period. Note, the highest average concentration of 2.5 μg/m^3 was measured in San Gorgonio Wilderness, just east of the Los Angeles metropolitan area. There was a strong gradient from the high concentrations in the California coastal areas to the minima of 0.1 μg/m^3 measured in Oregon. There was 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 μg/m^3). Nitrate mass fractions were typically 4 to 12% except in California where they were 30% and higher. In the north central part of the United States, nitrates constituted over 12% of the fine aerosol mass. Nitrates were the largest single component of fine aerosol mass in southern California at San Gorgonio Wilderness Area.

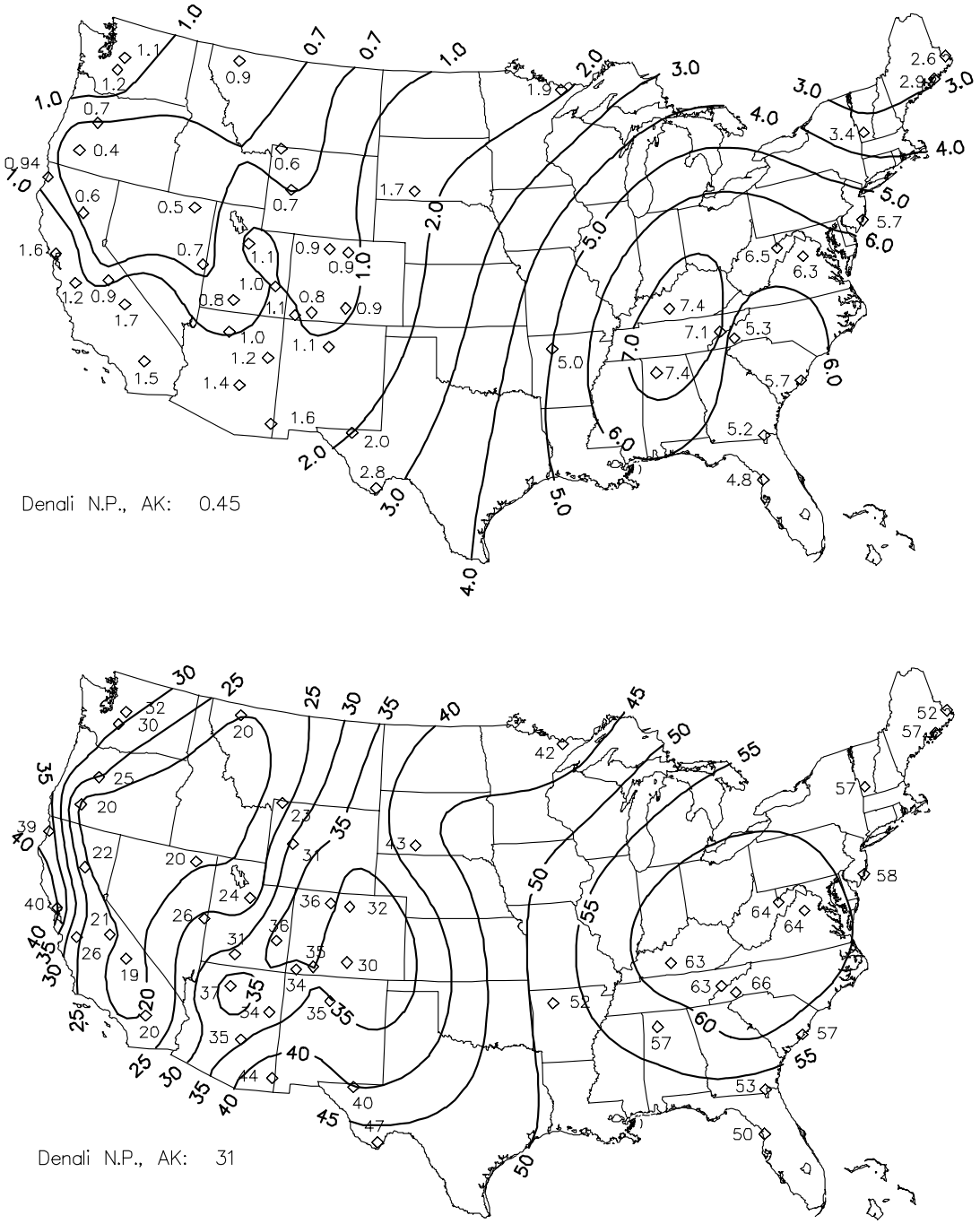

Figure 2.4 *Average fine sulfate aerosol concentrations (in μg/m^3) (top map) and sulfate fine mass (in %) (bottom map) for each site in the IMPROVE Network, excluding Washington, D.C.*

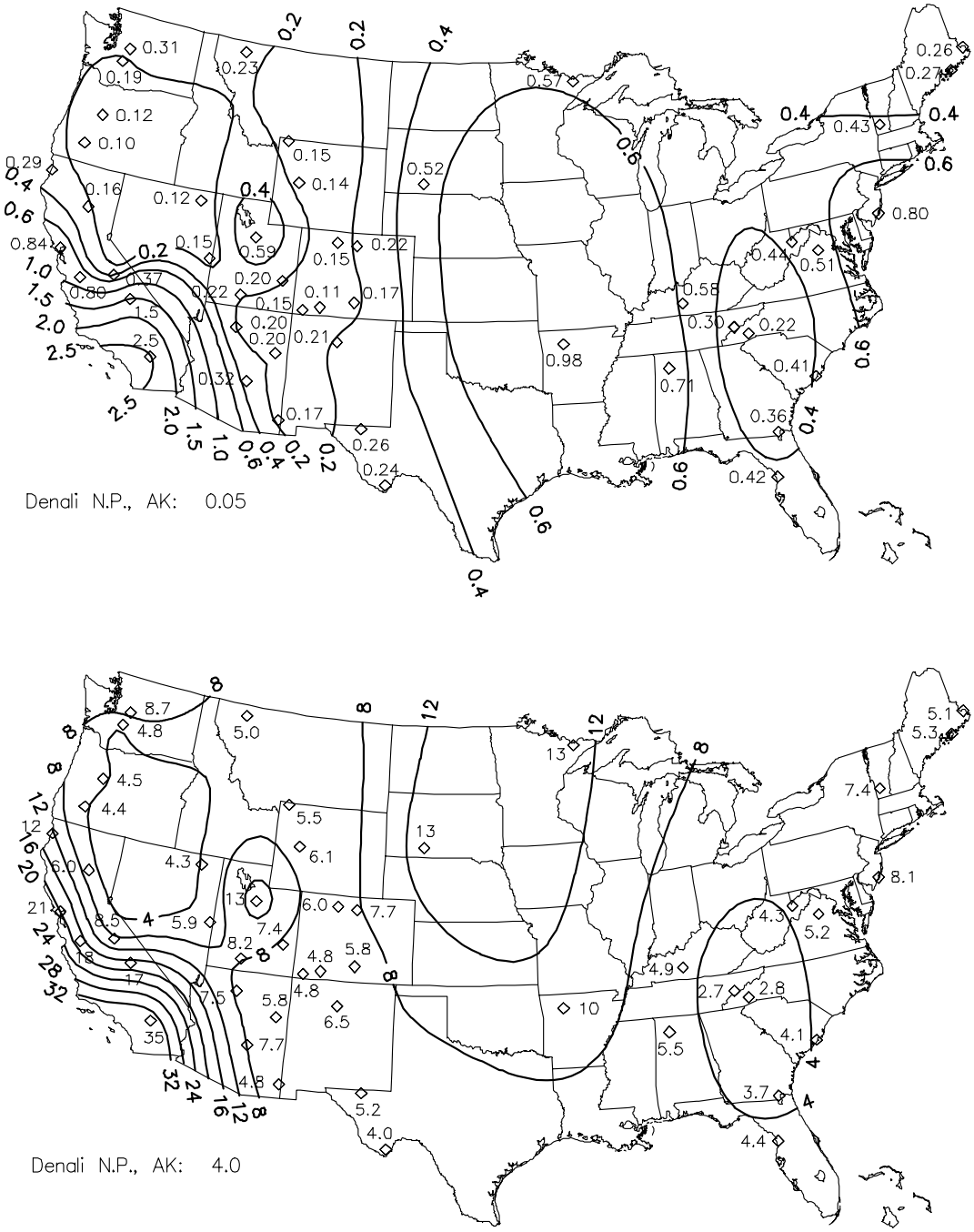

Figure 2.5 *Average fine nitrate aerosol concentrations (in μg/m^3) (top map) and nitrate fine mass (in %) (bottom map) for each site in the IMPROVE Network, excluding Washington, D.C.*

2.5.6 Fine Organic Aerosol

Figure 2.6 shows isopleth maps of the organic carbon mass concentration and organic mass fraction of the fine aerosol concentration, averaged over the three-year period. There was a significant spatial gradient from the lower Sierra-Nevada region, with average concentrations of 3.3 $\mu g/m^3$ to the inner-mountain region of Wyoming, Colorado, Oregon, Utah, and Arizona of 1.0 $\mu g/m^3$ or less. In the eastern United States, organics ranged generally between 1.5 to 3.4 $\mu g/m^3$. Except in the Sierra and Cascade Mountain regions, where organics were over half of the fine particle mass, organics generally constitute between 20 to 40% of the fine particle mass.

2.5.7 Fine Light-Absorbing Carbon Aerosol

Figure 2.7 shows isopleth maps of the light-absorbing carbon concentration and mass fraction of the fine aerosol, averaged over the three-year period. Note, light-absorbing carbon concentrations were lowest in the inner-mountain west and on the Colorado Plateau where light-absorbing carbon was generally less than 0.2 $\mu g/m^3$. Mass fractions were typically 4-5% of fine mass except in the Pacific Northwest where light-absorbing carbon contributed as much as 8.8% of the fine particle mass.

2.5.8 Fine Soil Aerosol

Figure 2.8 shows isopleth maps for fine soil. The contribution of soil to the fine aerosol in the United States was generally small, except for the elevated concentrations (>1 $\mu g/m^3$) in the southern tier of the United States. There was a quite noticeable north-south trend of increasing soil concentrations with the Northeast being the lowest. Soil contributed approximately 5 to 10% of the fine aerosol mass in the East. Except for Florida, all of the areas east of the Mississippi, the Pacific Northwest, and parts of California, soil contributed less than 10% to fine aerosol mass with much of the inner-mountain west in excess of 20%.

2.6 SUMMARY

The following were the major patterns observed in the three-year period of IMPROVE from March 1996 through February 1999:

(1) Spatial Patterns. Concentrations of fine particles (those most important in determining visibility) were 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 was sulfate at 60-65% of the mass, while in the Pacific Northwest it was organics, and in southern California it was nitrates. In general, the largest mass fractions of the fine aerosol were sulfates, organics and in soil/dust. Of the 21 regions in the IMPROVE Network, carbon (organic plus light-absorbing carbon) was the largest single component in

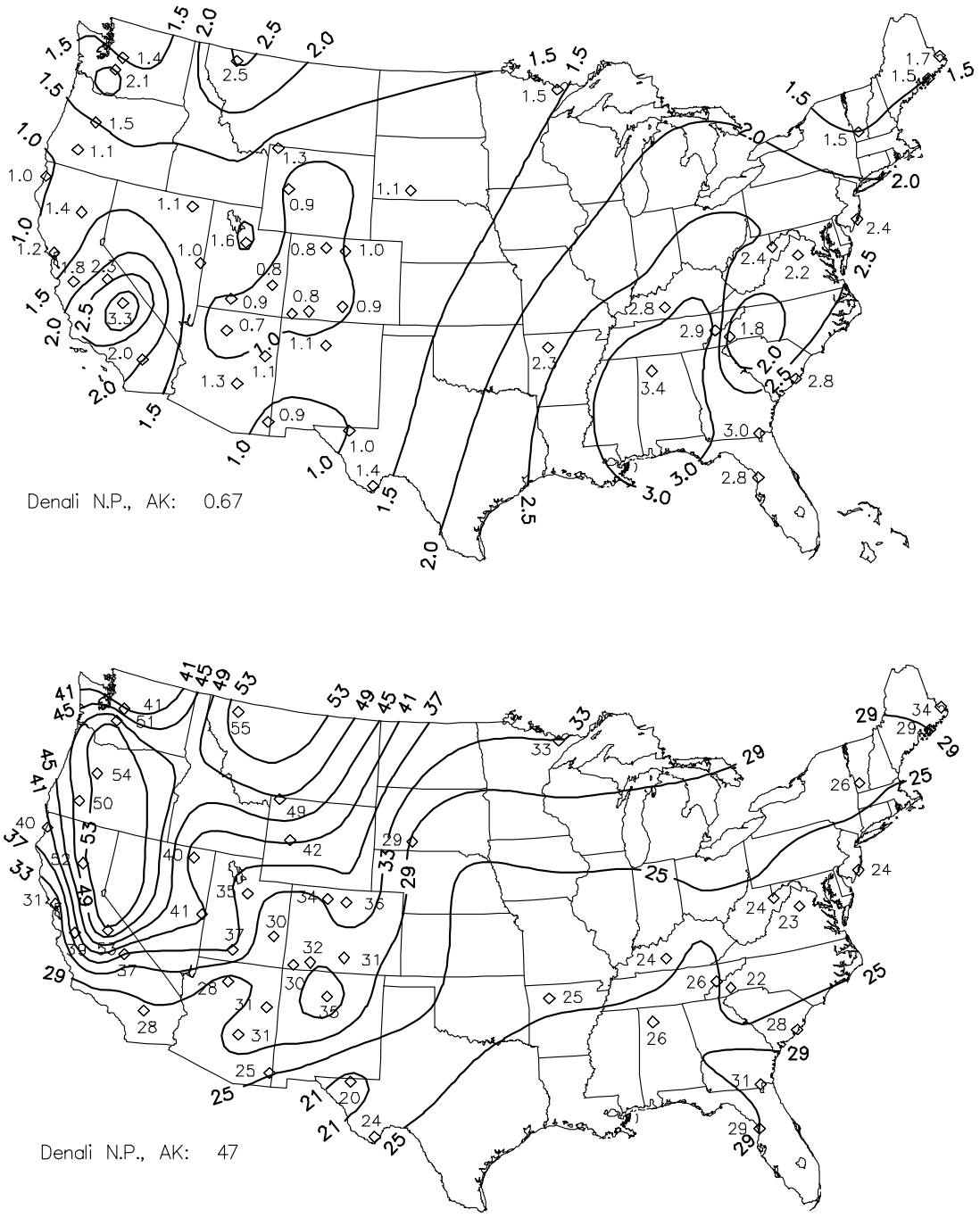

Figure 2.6 *Average fine organic aerosol concentrations (in μg/m^3) (top map) and organic fine mass (in %) (bottom map) for each site in the IMPROVE Network, excluding Washington, D.C.*

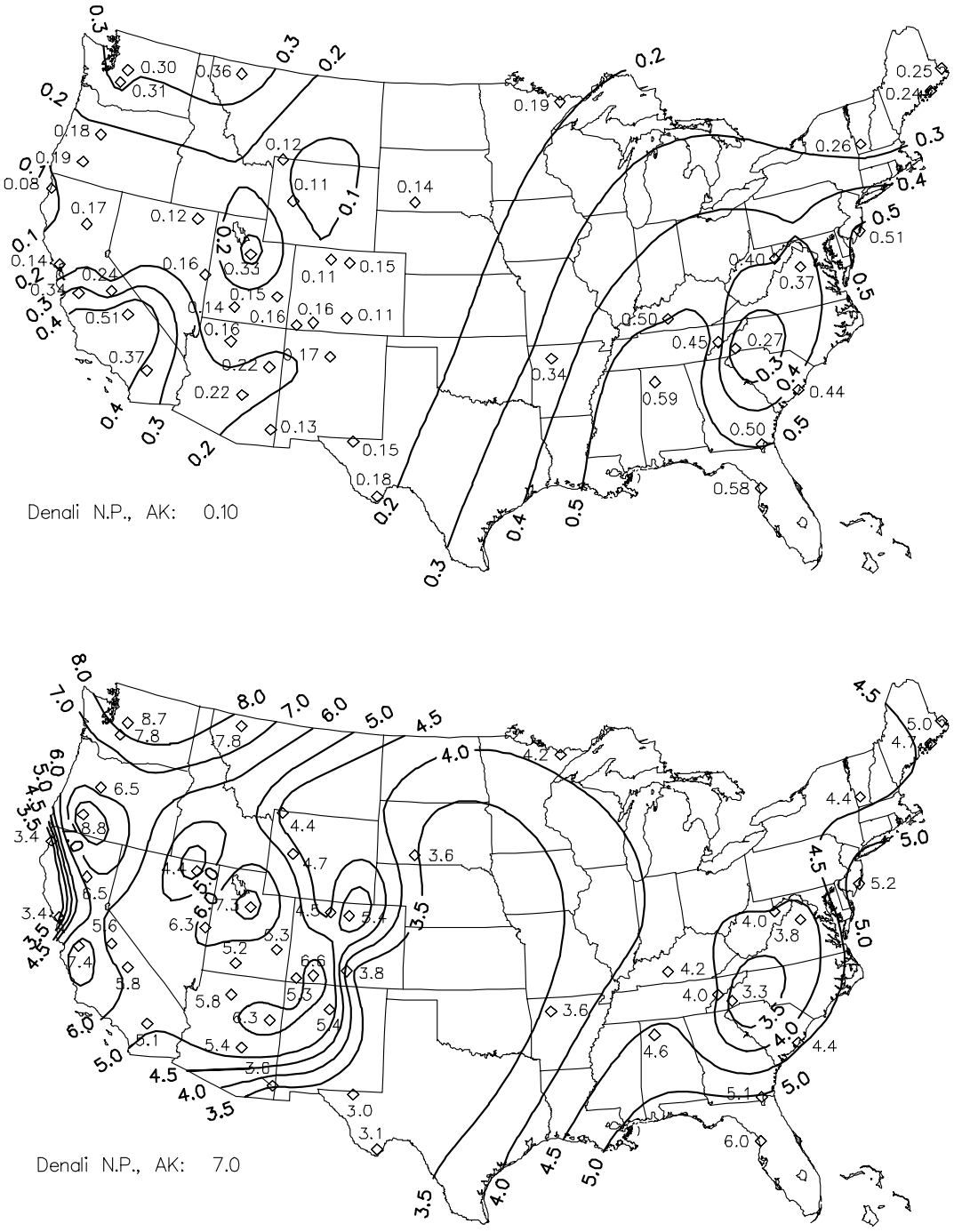

Figure 2.7 *Average light-absorbing carbon concentrations (in μg/m^3) (top map) and light-absorbing carbon fine mass (in %) (bottom map) for each site in the IMPROVE Network, excluding Washington, D.C.*

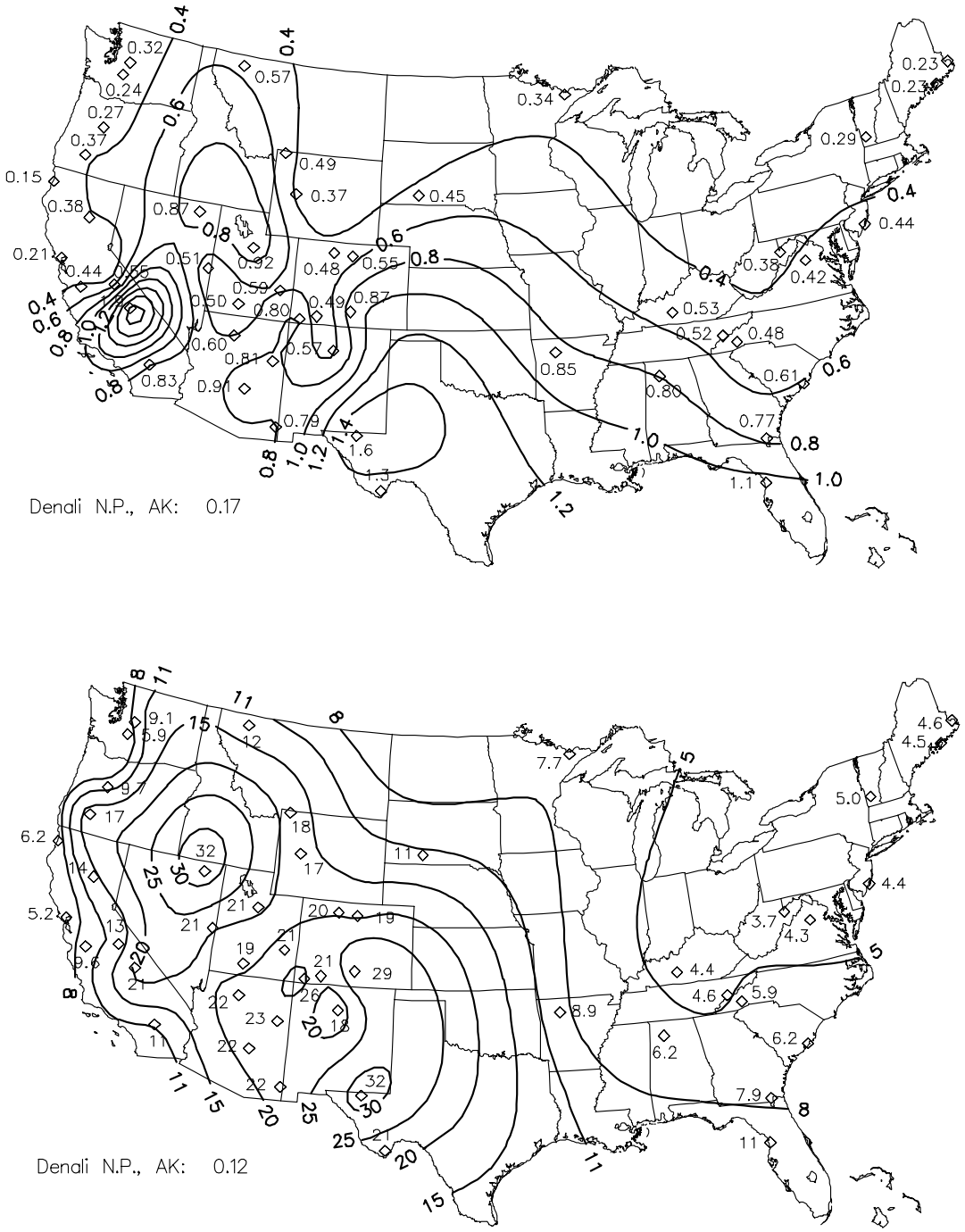

Figure 2.8 *Average fine soil aerosol concentrations (in μg/m^3) (top map) and soil fine mass (in %) (bottom map) for each site in the IMPROVE Network, excluding Washington, D.C.*

10 regions (Alaska, Cascade Mountains, Central Rocky Mountains, Colorado Plateau, Great Basin, Northern Rocky Mountains, Pacific Coastal Mountains, Sierra-Nevada, Sierra-Humboldt, and Wasatch). Sulfate was the largest single component of fine aerosol in 10 regions, primarily in the East (Appalachian Mountains, Boundary Waters, Mid Atlantic, Mid South, Northeast, Northern Great Plains Sonoran Desert, Southeast, Washington D.C. and West Texas), while nitrates were slightly greater than carbon in Southern California. Sulfate and carbon were approximately the same on the Colorado Plateau and Sonoran Desert.

(3) Smaller Contributors. After the contributions of organics and sulfate, soil was the next largest, followed by nitrate (except for nitrate in Southern California) and light-absorbing carbon.

(4) Seasonality. With a few exceptions, average fine mass concentrations, organics and sulfate components of fine mass were highest in summer. Soil concentrations were highest in spring or summer. On the other hand, nitrate concentrations were generally highest in winter or spring. Light-absorbing carbon exhibited relatively little seasonal variation.

(5) PM_{10}. The highest concentrations of PM_{10} 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 PM_{10}.

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