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**COMPOSITION OF AEROSOLS OVER
THE CONTINENTAL U.S.**

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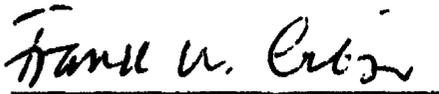
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<p>This study examines and integrates two databases containing aerosol species for the fine mass from the NPS-NFPN and NESCAUM monitoring networks. The coarse mass for the NPS-NFPN network is also examined. These networks operated in different parts of the United States, and over different time periods. The final database is then partitioned into the major aerosol types for the four quarters of the year.</p>			
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Equations are developed to partition the fine and coarse aerosol mass into sulfate, ammonia, water, organic, soil, soot, and salt aerosol types. The equations estimate these aerosol types by scaling up tracer species characteristic for specific source types. Any scaling factors which could not be determined from the assumed chemical composition of the aerosol types were found through a fitting process. Results of the partitioning agree with previous studies and alternative means of estimation. However, more reasonable results are found if the two databases are treated separately than combined.

The largest observed fine particle mass occurs in the eastern part of the U.S. where it is dominated by sulfur aerosol types (sulfate, ammonia, water) and organics constituting approximately 80% of the mass. These aerosol types reach a maximum during the third quarter of the year. In the Southwest fine soil is also a major component accounting for between 25-30% of the fine mass. The soil aerosols have maximum concentrations during the second quarter, about 1300 ng/m³ while the sulfur aerosol types are maximum during quarter three. The Northwest differs significantly from the rest of the country. Organics are the major aerosol type accounting for over 40% of mass for quarters 1, 2, and 4. The sulfate and soil each account for about 15% of the mass. The largest amount of "unknown" mass, approximately 20%, occurs in this region. The coarse mass is found to be dominated by soil, and the maximum concentrations are in the Southwest.

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DISCLAIMER

The NESCAUM data is routinely subjected to several levels of quality assurance screening prior to formal release. The results presented here have not yet been subject to review by the state site operating agencies, and are considered "preliminary."

1. INTRODUCTION

Reduction of visibility, or transmission of visible radiation, is due to scattering and absorption by atmospheric particles and gases. Usually the visible light extinction due to gases is small compared to that by particles. The light extinction due to scattering and absorbing particles is dependent on the particle size, shape and its refractive index. If a particle size and refractive index is known, and it is assumed to be spherical then the particle light extinction can be calculated through the Mie Theory (van de Hulst, 1957⁽¹⁾).

The optical properties of the particles are dependent upon their composition. Consequently, the properties of the aerosol greatly influence their ability to reduce human or electronic vision. Therefore to understand and model the visibility and the propagation of visible radiation, the behavior and composition of the aerosols must be known.

1.1 Related Reports

This report is one of the summary reports presenting the results from the past four years of research as part of this contract. Other summary reports include the *Organization, Access, and Exploration Facilities for Large Geophysical Databases* (September 22, 1989) and *Visibility Data Filters for Europe* (April 14, 1990). Additional summary reports will include presentation of the haze climate of North America and Europe, and a corresponding database to be used as input to the radiation transmission models. In addition to the extinction coefficients for North America and Europe this last report will also contain the apportionment of the extinction coefficient by aerosol types. These aerosol types are derived from the results of this report.

1.2 Purpose

The purpose of this study is two fold. First, aerosol data from two networks which sampled aerosols in rural locations in the U.S. are analyzed and integrated. Then the fine aerosol mass for both networks is apportioned into aerosol types which should

be suitable for the creation of extinction budgets, i.e. apportionment of total light extinction into contributions from different aerosol source types.

1.3 Background

A significant amount of work has been done on the properties of aerosols and their compositions. From this work a number of regular features have been found. Whitby, Husar and Liu (1972⁽²⁾) found that most aerosols have a bi-modal size distribution. This distribution has a fine mode centered at about 0.3 μm and a coarse mode centered between 5 and 30 μm . These two modes are usually unrelated, having different compositions, sources, and lifetimes (EPA 1979⁽³⁾).

It has been determined that the fine particles are generally made up of secondary and primary particles (Whitby et al., 1972⁽²⁾; EPA, 1979⁽³⁾). Secondary particles are formed through a gas to particle transformation that takes place in the atmosphere. This is a common source for much of the sulfates, particulate organics, and ammonium compounds. Primary particles are directly emitted from combustions, industrial, and natural sources. Fine particles have a slow removal mechanism and can travel large distances (Patterson et al., 1981⁽⁴⁾; Clarke et al., 1983⁽⁵⁾).

Coarse particles usually are derived from primary emission. They generally have a short residence time in the atmosphere settling out within hours or less (Husar, 1991⁽⁶⁾). In urban centers coarse particles are a result of both anthropogenic and natural sources. Due to the short residence time of coarse particles, anthropogenic sources contribute little to the coarse aerosol mass in rural areas. The primary source in these areas is soil dust (NAPAP, 1991⁽⁷⁾).

Many investigations have shown that the fine particles are the chief contributors to light extinction. In a study in Los Angeles White and Roberts (1976⁽⁸⁾) found excellent correlation between light extinction and fine particle mass. Using Mie calculations, Charlson et al., (1978⁽⁹⁾) showed that particles with a diameter between 0.1 and 1 μm are the most efficient light scatterers. For this reason, the determination of the sources of fine aerosol mass and the apportionment of this mass into aerosol types has received much attention.

By apportioning the aerosol into its source types the light extinction due to each aerosol type can be determined. The total light extinction would then be the sum of the apportioned light extinction, this is known as an extinction budget. (NAPAP, 1991⁽⁷⁾; Ouimette and Flagan 1982⁽¹⁰⁾; Friedlander, 1977⁽¹¹⁾). However, the legitimacy of the extinction budget is dependent on how the aerosol is apportioned. As noted by White (1986⁽¹³⁾), the light extinction contributed by distinct particles, externally mixed species, are additive, but the light extinction contributed by distinct chemical fractions within a single particle, internally mixed species, are not additive.

Most particles contain a number of chemical species internally mixed. Consequently, partitioning the extinction into the contribution by each aerosol species will be in error (NAPAP, 1991⁽⁷⁾). These errors can be reduced by lumping together chemical species which are known to be internally mixed. For example, ammonium is usually grouped with another chemical species such as sulfate or nitrate (White, 1986⁽¹²⁾).

The aerosol can be apportioned into aerosol types by various means. One method is to determine the particulate loading at a location from the major air pollution sources. Once the loading from each source is known, the aerosol types can be established through emission inventories. This has the added benefit of being able to estimate changes in the aerosol types by changes in emissions.

There are several methods to determine the source of aerosols from a particular location, known as receptor models. Friedlander developed one of the first models known as chemical mass balance, CMB, (Friedlander 1977⁽¹¹⁾, White, 1991⁽¹³⁾). This is a simple and straight forward method which can be applied to individual ambient samples. It uses chemical tracers, measured at a particular location, and emission inventories from the major local sources to partition the fine mass into source contributions.

Factor analysis is a more flexible apportioning scheme which can be used when multiple data samples are available (Hopke, 1985⁽¹⁴⁾; Hwang et al., 1984⁽¹⁵⁾). Instead of having to determine the tracer species a priori as in CMB, factor analysis determines them through the data. Thus it can be used to check and refine tracers used in a CMB.

Factor analysis determines the tracers based on the concept that two aerosol species from the same source should correlate well together (White, 1991⁽¹³⁾).

A problem with CMB and factor analysis is that they do not account for secondary species or aerosol growth due to water. This can lead to a large fraction of the fine mass going unaccounted for (White, 1986⁽¹²⁾). However, secondary aerosols can be taken into account by estimating their total mass from measured data. (Friedlander, 1977⁽¹¹⁾; Dzubay, 1980⁽¹⁶⁾) The sources of the gases from which the species came are not identified.

Linear regression is a receptor model which can take into account the secondary particles and growth of particles due to water condensation during transport (Belsley et al., 1980⁽¹⁷⁾; Kleinman, 1980⁽¹⁸⁾). This method works by estimating coefficients of independent and dependent variables, tracers, in a linear relationship using multiple data samples.

All three of these methods rely on tracers to identify sources and be scaled up to the sources mass contribution to the aerosol. To have reliable results these chemical tracers must be stable and measurable species. In many wilderness areas and national parks the concentrations of these tracers are often below the detectable limits of the measuring instruments (White, 1991⁽¹³⁾). This can greatly hinder the attempts at correctly determining the sources and species of the aerosol.

If only the aerosol types need to be determined, then there is an alternative to receptor modeling. The gross features of a mass apportionment can be estimated from chemical data by assuming that there are certain aerosol types that dominate the aerosol mass. A common chemical element in the aerosol type that is measured from the sample can be scaled up to estimate the aerosol types mass. An example would be to assume that one dominate aerosol type is ammonium sulfate, and all sulfur in the aerosol is associated with ammonium sulfate. To find the mass of the ammonium sulfate the measured sulfur would be scaled up by the mass ratio of ammonium sulfate to sulfur. This technique has been used in many studies both in rural and urban areas (Eldred et al., 1990⁽¹⁹⁾; Poirot et al., 1990⁽²⁰⁾; White and Macias, 1990⁽²¹⁾).

A technique which reconstructs the fine mass through linear regression can also be applied to obtain the major aerosol types. This method has the advantage that no

prior knowledge or assumptions about the chemical form of the aerosol types is needed. Using data from Glen Canyon, AZ, Sutherland and Bhardwaja (1990(22)) partitioned the fine mass into aerosol types with a multivariate regression method.

1.4 Scope

The report covers two main sections. The first section is concerned with the raw data. Here, the time trends of several species will be examined. The second part of the report apportions the seasonally averaged aerosol data into aerosol types. This will be accomplished through a process which assumes that the aerosol is constituted primarily by several aerosol types. The aerosol mass for each type is found by scaling up certain chemical species which are assumed to be associated only with that particular aerosol type. The results from this apportionment are then compared to previous studies. Also the spatial and temporal variations of each aerosol type for the U.S. is examined.

2. DATABASE DESCRIPTION

This study was conducted using two data bases created from the National Park Service - National Fine Particle Network (NPS - NFPN) (Eldred et al., 1986⁽²³⁾) and the Northeast States for Coordinated Air Use Management (NESCAUM) (Poirot et al., 1990⁽²⁰⁾) monitoring networks. The locations of the sites in each network were chosen based on the same criteria. However, the aerosol samplers used and the time periods of collection were different.

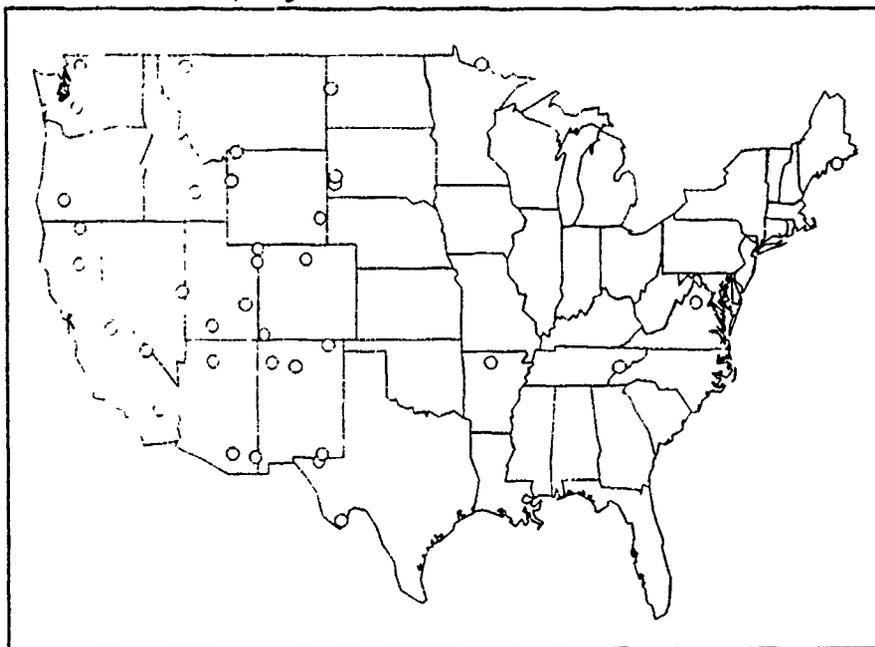
These rich databases allow the spatial and temporal trends of atmospheric aerosols across the U.S. to be analyzed. The large number of aerosol species measured will allow many meaningful studies to be conducted. They should also prove to be useful in the near future to help in monitoring reduction in air pollution due to the Clean Air Act of 1990. This section describes and analyzes the data from these two networks, and discusses the suitability for integration into one national data set.

2.1 NPS - NFPN monitoring Network

The NPS - NFPN network consisted of 37 stations located across the continental U.S., Figure 1. As shown, these stations were located primarily in the western U.S. with five sites in the East. All samplers were located inside national parks and wilderness areas far from any urban centers, industrial sources, and highways. Inside the parks the samplers were kept away from roads, parking lots and chimneys.

This network, which began operating in 6/82, originated from the Western Particulate Monitoring Network (WPMN) which operated from 8/79 to 9/81. The database used in this study contains some data from the WPMN network. Also several stations were added since the initial establishment of the NPS-NFPN network. Consequently, the sampling period for the database is station dependent. The beginning and ending dates for each station is presented in Table 1. Note that three stations, Fort Laramie WY, Yellowstone WY, and Mount Rushmore SD, have data only between 8/79 and 9/81, and the three stations, Acadia ME, Voyageurs, MN, and Saquaro, AZ, operated for less than a year.

Sampling Sites for NPS-NFPN Network



Sampling Sites for the NESCAUM Network

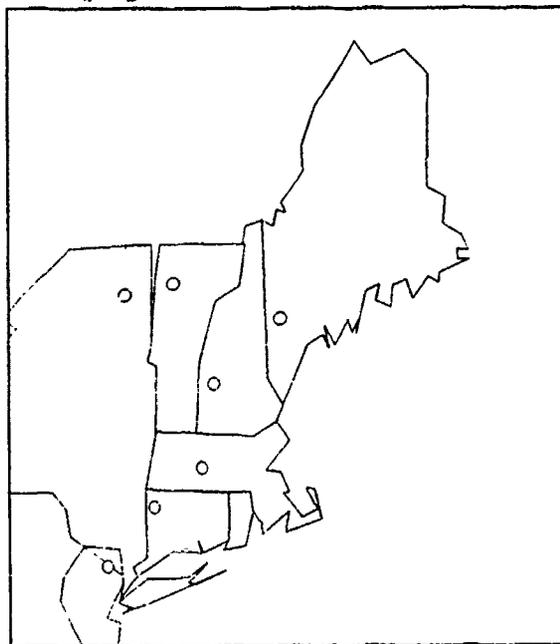


Figure 1. Sampling sites for the NPS-NFPN and NESCAUM monitoring networks.

Table 1. Data collection time span for each station in the NPS - NFPN network

Station Name	Elevation (ft.)	Begin	End	Latitude (rad.)	Longitude (rad.)
CRATER LAKE NP, OR	6500	10/82	6/86	0.7484	-2.1415
MOUNT RAINIER NP, WA	400	6/83	6/86	0.8170	-2.1241
LASSEN VOLCANIC NP, CA.	900	6/82	6/84	0.7010	-2.1218
LAVA BEDS NM, CA.	800	1/83	6/86	0.7280	-2.1206
NORTH CASCADES NP, WA	400	6/84	6/86	0.8472	-2.120
YOSEMITE NP, CA	5300	9/82	6/86	0.6536	-2.0831
DEATH VALLEY NM, CA	400	6/82	6/86	0.6370	-2.0394
JOSHUA TREE NM, CA	4600	6/82	6/86	0.5934	-2.0242
LEHMAN CAVES NM, NV	6800	6/82	6/86	0.6807	-1.9935
GLACIER NP, MT	4500	6/82	6/86	0.8465	-1.990
CRATERS OF MOON NM, ID	5900	6/82	6/86	0.7540	-1.9783
BRYCE CANYON NP, UT	8000	9/79	6/86	0.6557	-1.9580
SAGUARO NM, AZ	3100	6/85	6/86	0.5615	-1.9326
GRAND CANYON NP, AZ,	800	9/79	6/86	0.6295	-1.9577
GRAND TETON NP, WY	3100	7/85	6/86	0.7624	-1.9326
YELLOWSTONE NP, WY	6300	7/79	9/81	0.7840	-1.9272
CANYONLANDS NP, UT	5900	7/79	6/86	0.6710	-1.9167
CHIRICAHUA NM, AZ	5400	6/83	6/86	0.5585	-1.9060
DINOSAUR NM, CO	6000	9/84	6/86	0.7025	-1.9021
BROWNS PARK NWR, CO	5500	7/79	7/84	0.7121	-1.9019
MESA VERDE NP, CO	7200	6/82	6/86	0.6493	-1.8933
CHACO CULTURE NHP, NM.	400	9/79	6/86	0.6289	-1.8834
BANDELIER NM, NM	6500	10/82	6/86	0.6254	-1.8558
ROCKY MOUNTAIN NP, CO	7900	9/79	6/86	0.7046	-1.8425
GUADALUPE MTNS NP, TX	5400	1/83	6/86	0.5561	-1.8267
FORT LARAMIE NHS, WY	4300	9/79	9/81	0.7355	-1.8239
CARLSBAD CAVERN NP, NM	4400	1/82	1/83	0.5615	-1.8227
CAPULIN MOUNTAIN NM, NM	7300	6/82	6/86	0.6418	-1.8146
ROOSEVELT NMP/NORTH ND	2600	7/79	6/86	0.8308	-1.8107
WIND CAVE NP, SD	4800	6/82	6/86	0.7601	-1.8061
MOUNT RUSHMORE NMP, SD	4100	9/79	9/81	0.7659	-1.8055
BIG BEND NP, TX	3500	6/82	6/86	0.5114	-1.8008
VOYAGEURS NP, MN	1100	8/85	6/86	0.8478	-1.6261
BUFFALO NATL RIVER, AR	1000	1/84	6/86	0.6283	-1.6158
GREAT SMOKY MTNS NP, TN	2500	1/84	6/86	0.6240	-1.4574
SHENANDOAH NP, VA	3600	6/82	6/86	0.6716	-1.3634
ACADIA, ME:	400	9/85	6/86	0.7144	-1.1915

2.1.1 Sample Collection

Samples were collected over a 72 hour duration using a two stage stacked filter. Two samples were taken each week. The filter consisted of an 8 μm pore diameter Nuclepore filter followed by a 100% efficient wetted Teflon filter mounted in a plastic filter cassette. The inlet of the sampler had a 50% capture efficiency for particles 15 μm in diameter. This was for an airflow of about 10 liters per minute into the filter. This airflow was maintained by an Aero-Vironment SFS-500 unit. In 1983 Rockwell International performed field flow audits on the samplers, and found that 16% of the samplers had differences in the flow calibration exceeding 20%. The flow system was modified in 1984 which eliminated this problem (Eldred et al., 1986⁽²³⁾).

The aerosol mass is separated into a fine and coarse mode via the capture efficiency of the first filter. This filter had a capture efficiency of 50% for particles 2.5 μm in diameter at normal air flow rates. The first filter was coated with Apiezon L grease to reduce both particle bounce and reentrainment. All filters used in the first stage came from the same batch of precoated filters to maintain consistent collection properties. The second Teflon filter captured all of the fine particles that passed the first filter. This filter had a collection area 1/4 that of the first stage. This was done to increase the sensitivity of the analysis technique.

The sampling technique proved to be very reliable producing about 92% of the possible valid samples. Also, intercomparison with other samplers showed very good accuracy (Eldred et. al., 1986⁽²³⁾).

2.1.2 Sample Analysis

All analysis was conducted at the University of California, Davis using four nondestructive techniques. These techniques measured the fine and coarse mass, the fine optical absorption, elements from sodium to lead, and hydrogen. On average, there was a one month time lag between sampling and analysis. During the time lag the filters were stored in the plastic cassettes used during sampling.

The fine and coarse mass concentrations were measured by gravimetrically weighing the filters on a Cahn 25 electrobalance. The filters were weighed before and after sampling the aerosol to account for differences in mass of the unused filters. The

uncertainty in the actual weighing of the filters was about 2%. Unfortunately there was a large uncertainty added to this measurement due to a mass gain acquired by a filter stored in a cassette. Both the Nuclepore and Teflon filters experience this mass gain. The increased mass was accounted for by determining the mass gain on unused filters stored in plastic cassettes for one month. It was found that the Teflon filter's mass increased by 16 ug and the nuclepore filter increased by 10 ug. The uncertainty in these mass gains increased the uncertainty in the mass measurement to about 4% (Eldred et al., 1986(23)). Note, that this uncertainty does not account for the variation in flow rate to the sampler. Consequently the actual uncertainty in the fine mass concentration will be larger than 4%. Also, the composition of the mass gain is not known.

The optical absorption measurements were made only on the Teflon filters, collecting the fine particle mode aerosol, using a laser integrating plate method, IPM. This analysis was not conducted on the data until 1982. Excellent agreement was found between a comparison of this method and a photospectrometer. The minimum detectable limit for this measurement was determined to be about $0.2 \cdot 10^{-6} \text{ m}^{-1}$, and the uncertainty was around 7% (Eldred et al., 1986(23)).

Elemental concentration for sodium to lead were measured using a particle induced x-ray emission, PIXE. Precisions for these measurements were assessed by reanalyzing about 15 filters. Precisions were 4% for sulfur, 6 - 10% for silicon, potassium, calcium, iron, and zinc, and 10 - 15% for copper, lead, and bromide. The reported minimum detectable limits for the elements varied between 2 ng/m^3 for sulfur and 0.5 ng/m^3 for selenium.

Matrix corrections were made for each element due to absorption of some of the X-rays by the samples. These corrections were largest for light elements on the coarse filter. It was estimated that for coarse silicon 30% of the X-rays were absorbed, but for coarse iron only 5% were absorbed. These corrections were greatly reduced for the fine particles, about 1% for both fine sulfur and iron (Eldred et al., 1986(23)).

A measurement of the hydrogen content of the aerosol using a proton elastic scattering analysis, PESA, was added to the analysis of the samples in June 1984. This measurement was conducted only on the fine particle mass. The minimum detectable

limit was around 10 ng/m^3 , and precision of the measurement was found to be about 15% (Eldred et al., 1986⁽²³⁾).

This network is no longer in operation. It has been superseded by the IMPROVE (Interagency Monitoring of Protected Visual Environments) monitoring network beginning in 1988.

2.2 The NESCAUM Monitoring Network

The NESCAUM network consisted of seven stations located in the northeastern U.S., Figure 1. These sites were located in rural locations which were found to be consistent with EPA, NPS, and IMPROVE siting criteria (Poirot et al., 1990⁽²⁰⁾). The data used in this study were collected over the two year time period 2/89 to 1/91. This network is still in operation.

2.2.1 Sample Collection

Each site contained two samplers, one to sample for PM-10 particles, and the other to sample fine particles. Additional samplers were deployed at the Whiteface and Quabbin sites to sample for organic and elemental carbon.

The fine particles were sampled for a 24 hour duration on every Wednesday, Saturday, and every 6th day. The fine particles were separated from the bulk aerosol using a U. C. Davis cyclone sampler, and collected on Teflon filters mounted in Nuclepore cassettes. This cyclone had a 50% capture efficiency for particles $2.5 \mu\text{m}$ in diameter at a flow rate of 23 l/min. The flow rate was maintained using a critical orifice between the filters and pump. The flow rate was measured before and after the filter by two independent methods. Third party audits showed that the uncertainty in the flow rate was approximately 3% (Flocchini et al., 1990⁽²⁴⁾). The samplers used to collect the fine organic and elemental carbon were the same as the fine particle samplers except two pre-fired quartz filters in tandem were used instead of the Teflon filter.

2.2.2 Sample Analysis

The samples containing the fine mass were analyzed by the U. C. Davis laboratory. There the mass, absorption, hydrogen concentration and elemental concentrations, sodium and heavier elements, were analyzed using the same techniques as those used for the NPS - NFPN network.

The quartz filters were analyzed for carbon concentrations by the Desert Research Institute (DRI) using the thermal optical reflectance method (TOR). This method determines the contribution to carbon mass by both organics and elemental carbon. Two problems arose for this analysis technique. First, large and variable artifacts were present from the quartz filters, collection process, and storage in the plastic cassettes. Quartz filters had a positive organic artifact due to adsorption of organic gases during collection. The second quartz filter in the tandem pair was used to estimate the total artifact. Normally the adsorption reaches saturation very quickly. However, some of the samples have much larger artifact concentrations than the average which increases the overall uncertainty. Corrections for this artifact causes large precisions and minimum quantifiable limits (MQL). The MQL is defined as twice the uncertainty for zero loading on the filter. The MQL was about 530 ng/m^3 for the organic carbon and 145 ng/m^3 for elemental carbon (Eldred et al., 1990(19))

The second problem is that the TOR method does not agree with other methods on where to separate the organic and elemental carbon. In a comparison of the TOR and thermal manganese dioxide oxidation method (TMO), using data from the WHITEX study, it was found that the TOR elemental carbon concentrations were five times those of the TMO. However, the TOR elemental carbon was approximately equal to the soot concentrations estimated from optical absorption (Eldred et al., 1990(19)).

2.3 The Raw Data

The meaningful interpretation of aerosol data depends strongly on the understanding of the basic characteristics of the data. This is best accomplished by examining the raw data. To present the main features of the raw data, three stations from the NPS-NFPN network and two stations from the NESCAUM network for four

aerosol species, fine mass, sulfur, silicon, and lead are plotted in Figures 2-7. Superimposed on the raw data is a heavy line representing the monthly average.

Each of the NPS-NFPN sites comes from a specific region of the U.S., and represents the basic features of that region. The sites associated with each region are Shenandoah, Va in the eastern U.S., Grand Canyon, AZ in the Southwest, and Glacier, MT in the Northwest. The two NESCAUM sites, Mohawk, CT and Underhill, VT, present the general characteristics of the Northeast.

The raw data for the fine mass is shown in Figures 2 & 3. At Grand Canyon, AZ, the time spans the period from 9/72 to 6/86. Note that the data before 1983 is rather variable, and it is difficult to recognize any trends in the data. However, after 1983 there is a distinct seasonal pattern with a high during the summer months. This variability was found for most stations which had data prior to 1983.

The seasonal pattern is also seen at all of the other NPS-NFPN sites located in the Southwest and the East. The NESCAUM sites also exhibit a seasonal pattern, although it is not as distinct as it is for the NPS-NFPN sites, Figure 3. The only deviation from this pattern is in the Northwest, where there are a number of sites, such as Glacier, MT in Figure 2, with no discernable seasonal pattern. This presents a definite difference between the Northwest sites and those in the rest of the country. However, at all of the sites in both networks the fine mass showed no trend towards increasing or decreasing concentrations.

Since all of the stations in the Southwest and East exhibited a seasonal pattern for the fine mass, the variability in the data prior to 1983 at the Grand Canyon most likely is not solely due to a varying aerosol mass. One possible explanation for the suspect data is a varying sampler airflow rate during collection. As discussed in section 2.1.1 there was up to a 20% deviation in the airflow rate to the samplers which was not corrected until 1984. Due to the uncertainty in the early data, all averaging and examination on the NPS-NFPN data took place from 1/83 to 12/85.

The fine silicon and sulfur are presented for the five stations in Figures 4-6. Again at the Grand Canyon and Shenandoah, there is the same seasonal pattern. However, the maximum concentration for the silicon occurs more towards the spring

months, whereas this maximum occurred during the summer month for the fine mass and sulfur. At the Northwest station, Glacier, MT, the silicon pattern is similar to the rest of the country, but the sulfur, like the fine mass, does not exhibit any seasonal pattern. Again, for both the silicon and sulfur at all sites there is no trend towards increasing or decreasing concentrations.

The silicon and sulfur trends at the NESCAUM sites are similar to those at the Grand Canyon and Shenandoah. These species exhibit a slight seasonal pattern with the maximum concentrations occurring during the same months as those for the NPS-NFPN sites. Also there is no increasing or decreasing time trend.

The fine lead is shown in Figures 6 & 7. At the NPS-NFPN sites there are fewer data points for the lead than for the other aerosol species examined. This is due to a large number of measurements falling below the minimum detectable limit, MDL. It was not uncommon for over 50% of the data for lead to be below the MDL. However, for the NESCAUM sites, there is a higher proportion of the lead data above the MDL. This difference between the two networks is a common occurrence for the trace species.

The fine lead time trends for both networks show no seasonal patterns. However, the lack of a seasonal pattern may be due to the large amount of data falling below the MDL. Unlike the other aerosol species examined, the lead has a definite decreasing concentration with time at most stations. This was to be expected with the removal of lead from gasoline which generally is the leading source of aerosol lead. However, at several sites, such as the Grand Canyon, smelters contribute a significant fraction of the fine particulate lead, so the decreasing concentrations were not as evident.

Based on the analysis of all of the raw data from both networks, two points were evident. All of the NESCAUM sites and the NPS-NFPN sites located in the East and the Southwest had a seasonal pattern in the time trend for the major aerosol species, and there was no trend to increasing or decreasing concentrations. Due to the similarity of the data from the two networks we felt that the data from these networks could be combine.

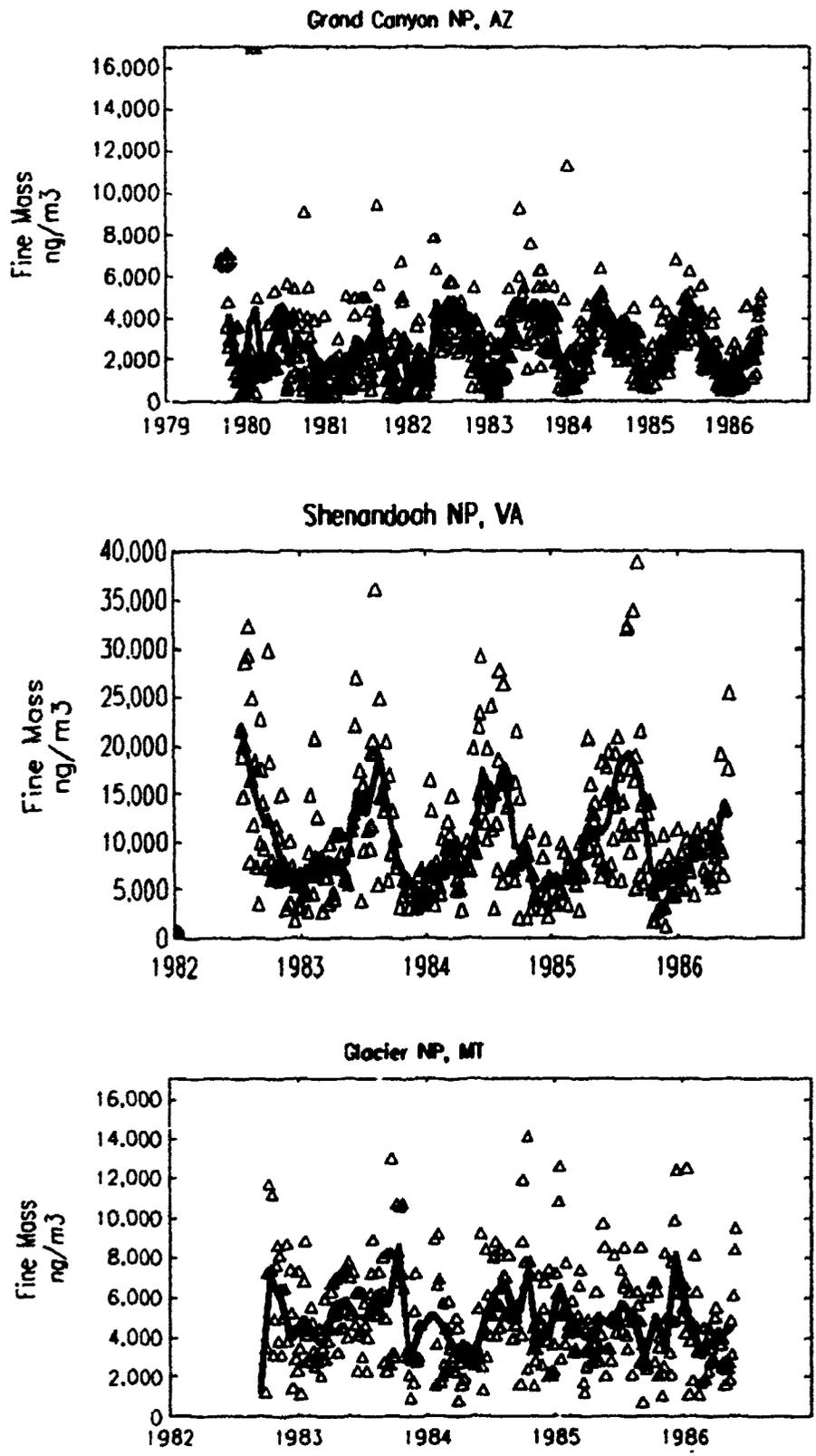


Figure 2. The raw fine mass data for three NPS-NFPN stations.

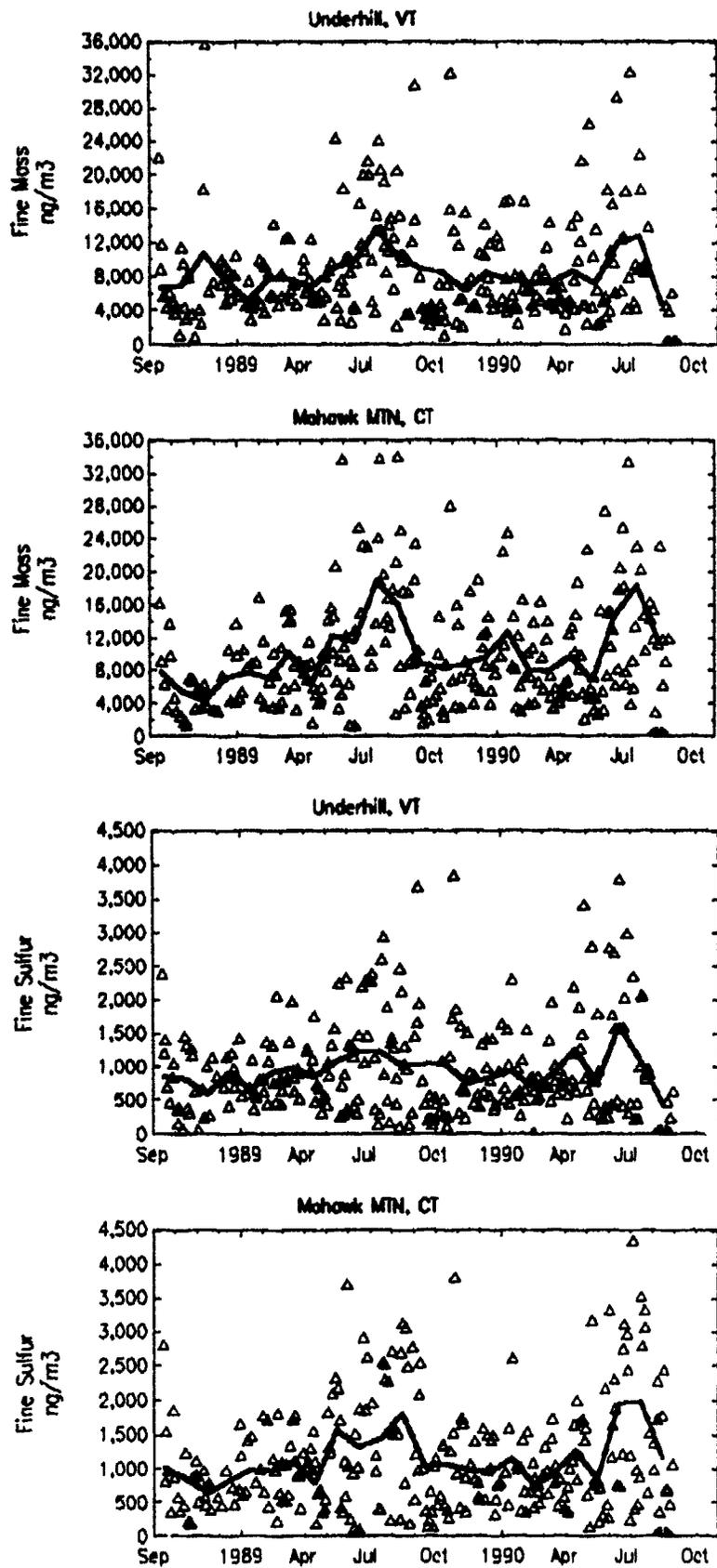


Figure 3. The raw data at two NESCAUM sites for the fine mass and sulfur.

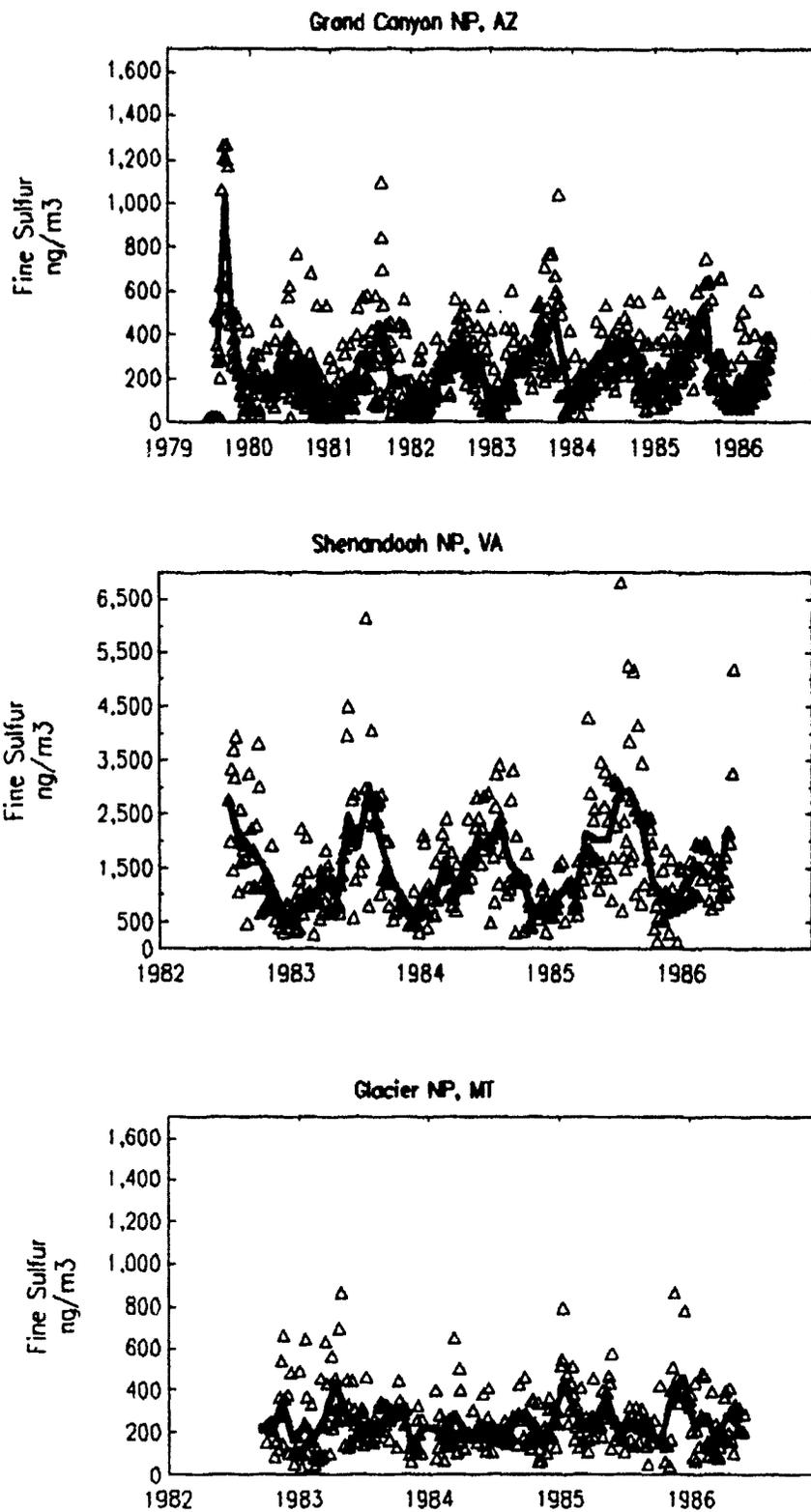


Figure 4. The raw sulfur data for three NPS-NPPN stations.

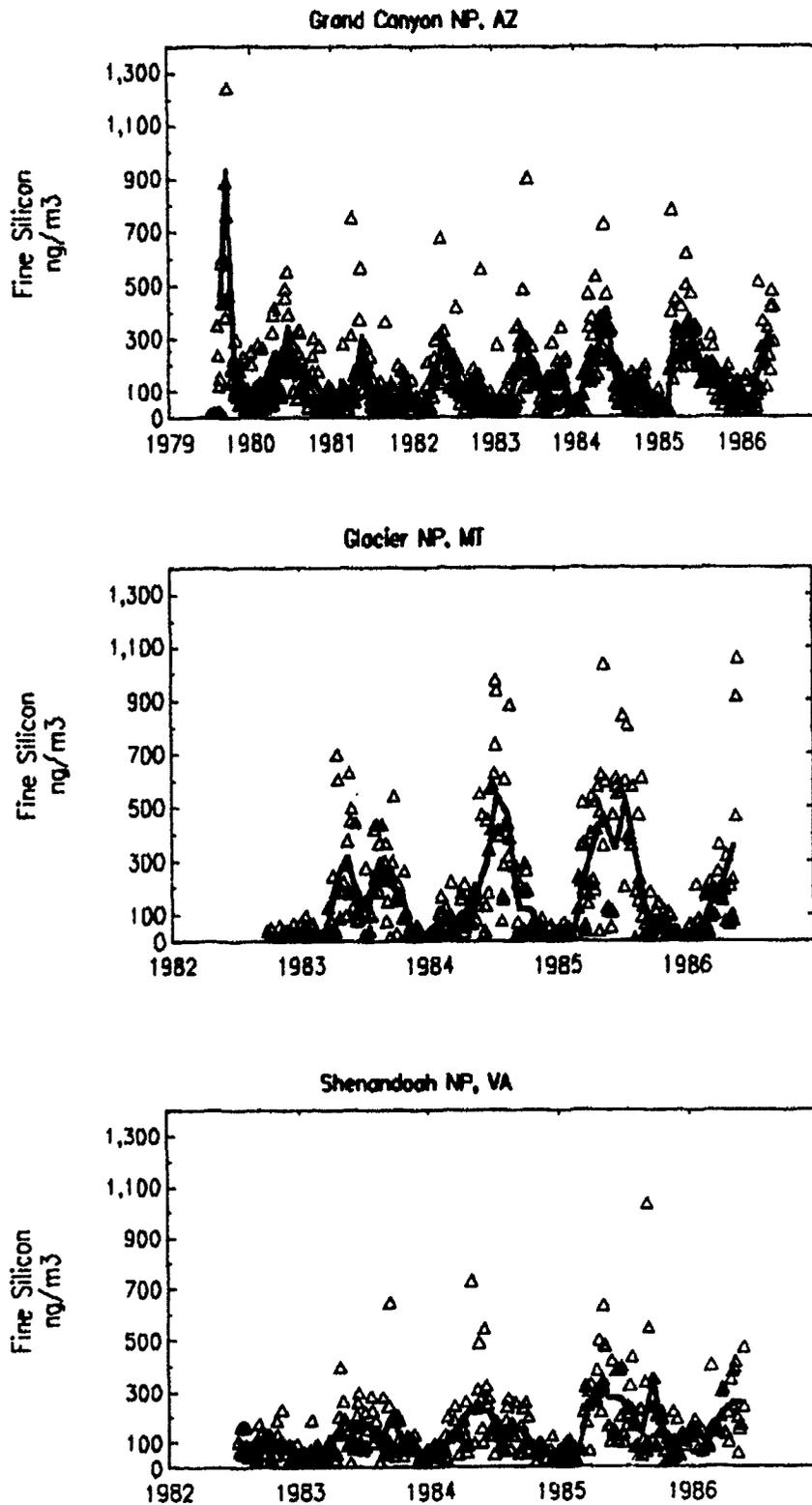


Figure 5. The raw fine silicon data for three NPS-NFPN stations.

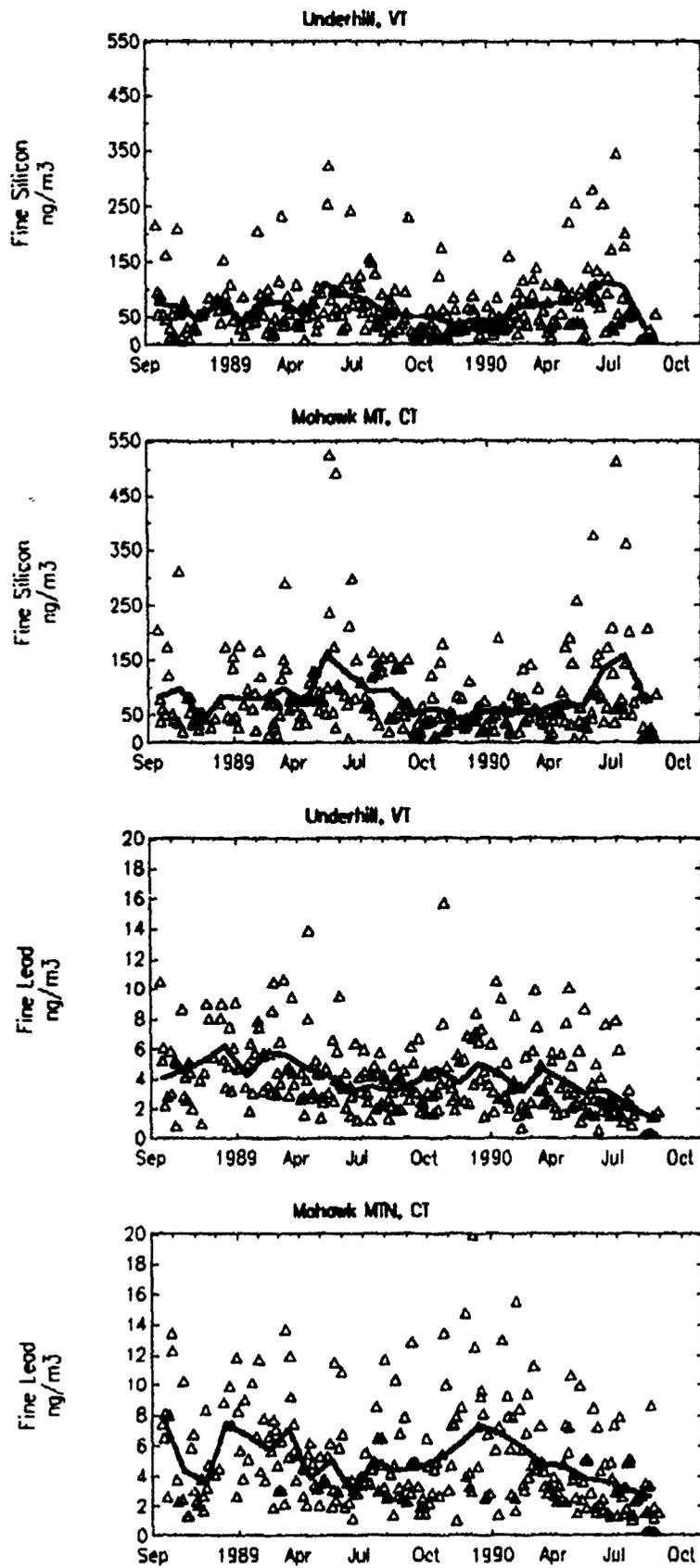


Figure 6. The raw data at two NESCAUM sites for the fine silicon and lead.

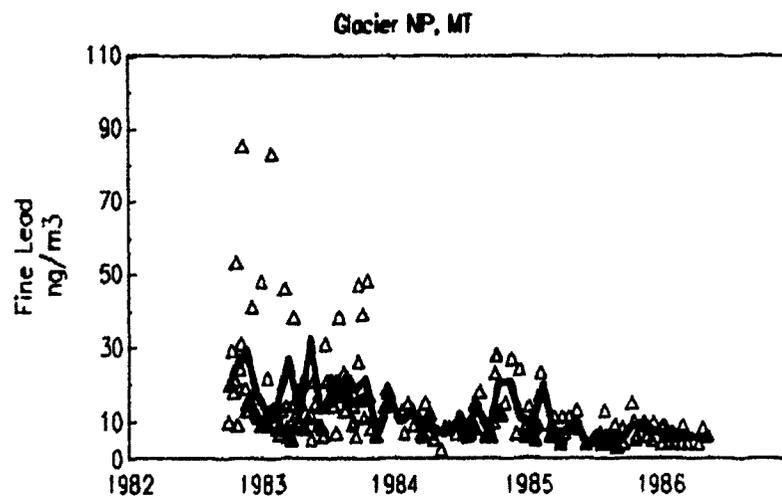
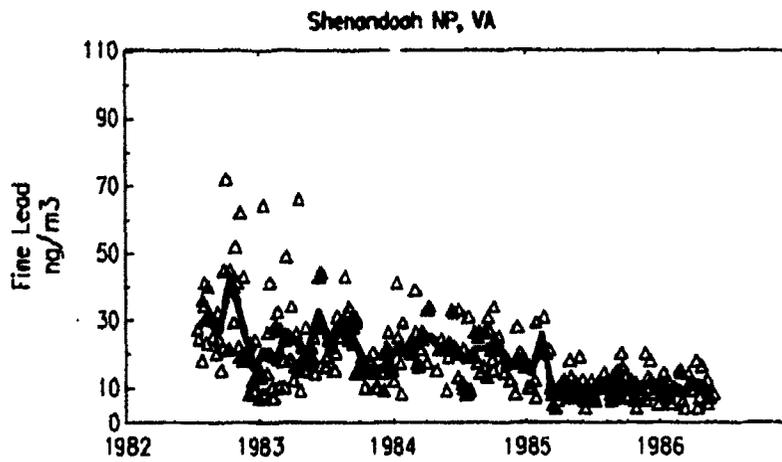
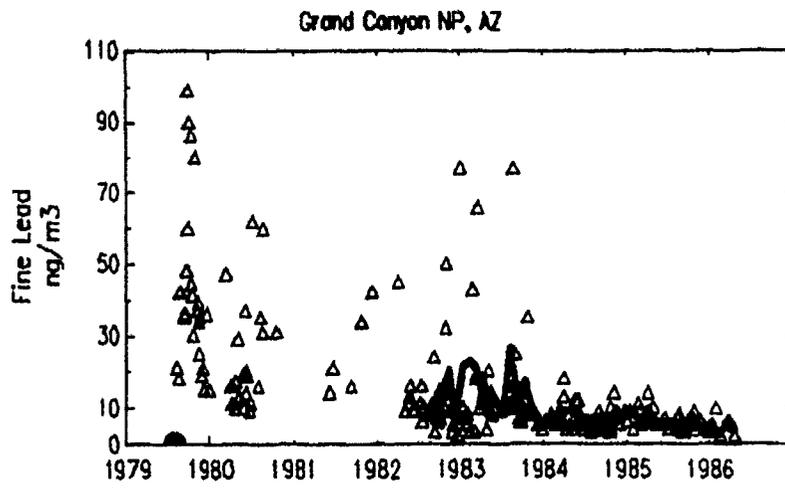


Figure 7. The raw fine lead data for three NPS-NFPN stations.

3. DETERMINATION OF AEROSOL TYPES

The aerosol mass in the atmosphere contains many different aerosol types. This is especially true in urban centers where anthropogenic sources can contribute a large number of different types of aerosol to the atmosphere. In rural areas the particulate mass contains generally fewer aerosol types. Six common aerosol types which normally constitute the bulk of the nonurban fine particulate mass are: sulfate, water, soot, soil, organics, and nitrates (Cahill et al., 1989⁽²⁵⁾; Friedlander, 1977⁽¹¹⁾). A seventh aerosol type, salt, is often important for coastal sampling sites (Junge, 1972⁽²⁶⁾). The coarse mass is primarily made up of soil constituents. Other aerosol types such as sulfates and organics can constitute a small fraction of the coarse mass. (Lewis and Macias et al., 1980⁽²⁷⁾).

This section discusses the process used to partition the fine and coarse aerosol mass into the aerosol types. This was accomplished by using a two step process. The first part follows the work of Cahill et al., (1989⁽²⁵⁾), in partitioning the NPS-NFPN data into aerosol types by scaling up the concentrations of aerosol species used as tracers. The next part used a fitting process to estimate any unknown tracer scaling factors.

The equations developed were applied to the quarterly averaged data. That is, all of the data over the given time span was averaged together for each quarter. Quarter one began in January, and the time span used was from 1/83 to 12/85 for the NPS-NFPN data and 9/88 to 9/90 for the NESCAUM data.

3.1 Equations for Aerosol Types

The aerosol types were found by creating equations which multiplied a tracer species by a scaling factor. An equation for this would be Aerosol Type = $C_i * T_i$, where C_i is the scaling factor, and T_i is the concentration of an aerosol species, such as silicon. An assumption of this method is that the tracer constitutes a constant fraction of the aerosol type. This is in accordance with the source apportionment presented by Friedlander (1977⁽¹¹⁾).

3.1.1 Sulfate and Water

The sulfate particle which often has water associated with it is the most important aerosol type in terms of its fraction of the fine mass and light scattering. Nearly all fine particulate sulfur exists as a sulfate anion, SO_4^{-2} , and an associated cation. The form of the cation depends on the degree of neutralization by ammonium. The three common forms of particulate sulfate are ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$), bisulfate (NH_4HSO_4), and sulfuric acid (H_2SO_4). (Poirot al., 1990(20); Pierson & Brachaczek, 1983(28)). In this analysis, the sulfate aerosol was divided into two parts, the sulfate anion which will be call sulfate and the sulfate cation which will be called cation. Both the sulfate and cation will be referred to as aerosol types.

In order to calculate the concentration of the sulfate aerosol type, we multiplied the sulfur concentrations by the ratio of the molecular weights of sulfur to sulfate. Therefore, the sulfate aerosol equation was sulfate = $3 \cdot \text{S}$.

The cation concentrations in the West were found by assuming that there was sufficient ammonium to neutralize the sulfate completely, producing only ammonium sulfate, $(\text{NH}_4)_2\text{SO}_4$. (White and Macias, 1990(21), Ouimette and Flagan, 1982(10)). In the cation aerosol equation for the West, the sulfur concentrations were again used as the tracer, and a scaling constant of 1.125, the ratio of the molecular weights of $(\text{NH}_4)_2$ to sulfur was used.

Due to the high sulfur concentrations in the eastern U.S., the sulfate generally is not fully neutralized by ammonium. Consequently, the sulfate aerosol is found in its three forms. To reflect this diversity, the cation concentration in the East was represented by $(\text{NH}_4)_x\text{H}_y$. All sulfate aerosol is in the form of ammonium sulfate if $y = 0$ and $x = 2$ and sulfuric acid if $y=2$ and $x = 0$. Variations of x and y between these extreme values present a mixture of the three possible sulfate cations. However, the constraint $x+y = 2$ must be met. The values for x and y can vary both spatially and temporally.

Due to the variation of the eastern sulfate aerosol the constant used to scale the sulfur tracer was not known. Therefore, the equation Cation = $A \cdot 1.125 \cdot \text{S}$ was developed where the constant A can have a maximum value of 1 for ammonium sulfate,

and a minimum value of 0.056 for sulfuric acid. The actual constant varied within this range depending on the sulfate constituents. The constant A was found through the fitting process discussed latter in this section.

It has not been conclusively demonstrated that the coarse sulfur is in the form of an ammonium cation and sulfate anion. However, several studies have made this assumption in partitioning the coarse data (Macias et al., 1981⁽²⁹⁾, Lewis and Macias, 1980⁽²⁷⁾). Therefore, for this study we assumed that the coarse sulfur had the same speciation as the fine sulfur, and used the same constants as those found in the fitting process for the fine aerosols to estimate the cation and sulfate concentrations.

The aerosol type water becomes associated with the aerosol mass through absorption by the soluble salts. Of these salts, sulfate has the highest concentration and a very high affinity for water (Sloane, 1986⁽³⁰⁾; Lewis and Macias, 1980⁽²⁷⁾). For this reason, all water calculated was assumed to be associated with the sulfate aerosol.

Prior to analysis, the samples were stored in the lab for an extended period of time. This allowed the water in the sample to reach an equilibrium with the laboratory atmosphere. Consequently, the water associated with the aerosol was dependent upon the relative humidity of the laboratory, not that of the sampling conditions. The water concentration was estimated by scaling up the sulfate and cation concentrations; $\text{water} = B \cdot (\text{Sulfate} + \text{cation})$. The constant B was determined through the fitting process discussed in section 3.2.

Although, the water associated with the aerosol samples was dependent upon the laboratory R.H., the weather conditions during sampling could affect the aerosol's water concentration. An aerosol at a relative humidity below the deliquescence point will contain more water, approximately two times, if it had previously been at the deliquescence point then if it had not (Winkler and Junge, 1972⁽³¹⁾). Assuming the sample had not been dried out. This is known as the hysteresis effect. Therefore, the water constant can vary with sampling time and location.

It has been shown that the coarse aerosol mass has little water gain even at high humidities (Lewis and Macias, 1980⁽²⁷⁾). Therefore, no water was apportioned for the coarse mass.

3.1.2 Soot

The optical absorption measured for each fine mode air sample was attributed to elemental carbon. Using an absorption efficiency of $10 \text{ m}^2/\text{g}$, the absorption was converted to soot concentrations (Eldred et al., 1986⁽²³⁾). The absorption efficiency used was based on the assumption that the absorption was primarily due to diesel exhaust. On examining the soot concentrations, the average values were found to be over 8% of the measured fine mass. This seemed high for remote areas. In a study conducted at Zilnez Mesa, AZ by J.R. Ouimette and R.C. Flagan (1982⁽¹⁰⁾) a soot concentration of 3.5% was found.

Two major sources of soot are automobiles and the burning of wood (Pierson and Brachaczek, 1983⁽²⁸⁾; Cooper, 1980⁽³²⁾, Valaoras et al., 1987⁽³³⁾). The major source of lead is only automobiles. During the summer months the burning of wood for heat would not be present, so the contribution to organics via wood burning would be reduced. Consequently, we believed that during the summer months vehicle traffic, particularly diesel vehicles, was the primary contributor to lead and soot.

Figure 8 is a scatter plot of soot and lead for the two summer quarters of the NPS-NFPN data. As can be seen, there is a good correlation between these two species, except for two outliers. Outlier A occurred during the third quarter in Yosemite. The abnormally high soot concentration was most likely due to forest fires in the area. Outlier B is associated with Chiricahua, AZ, which has a number of copper smelters located near by. The high lead concentration is most likely due to the smelters and not automobiles. If these two data points are removed the correlation coefficient increases to 0.84 and the Y offset becomes $250 \text{ ng}/\text{m}^3$.

If the lead and soot concentrations originate from the same source, then as the lead concentration approaches zero, the soot concentration should also diminish. According to the regression line in the scatter plot, Figure 8, at small concentrations of lead the soot concentration is around $250 \text{ ng}/\text{m}^3$. During the summer months a soot background will probably exist due to wild fires, however we believe that this background level will be much less than $250 \text{ ng}/\text{m}^3$. For this reason, we felt the reported soot concentrations were systematically over estimated. Therefore, these concentrations were reduced for the NPS data set by $200 \text{ ng}/\text{m}^3$. This brings the average soot concentration to less than 4% of the measured fine mass. This value appears more reasonable, and is on the order found by Ouimette and Flagan (1982⁽¹⁰⁾).

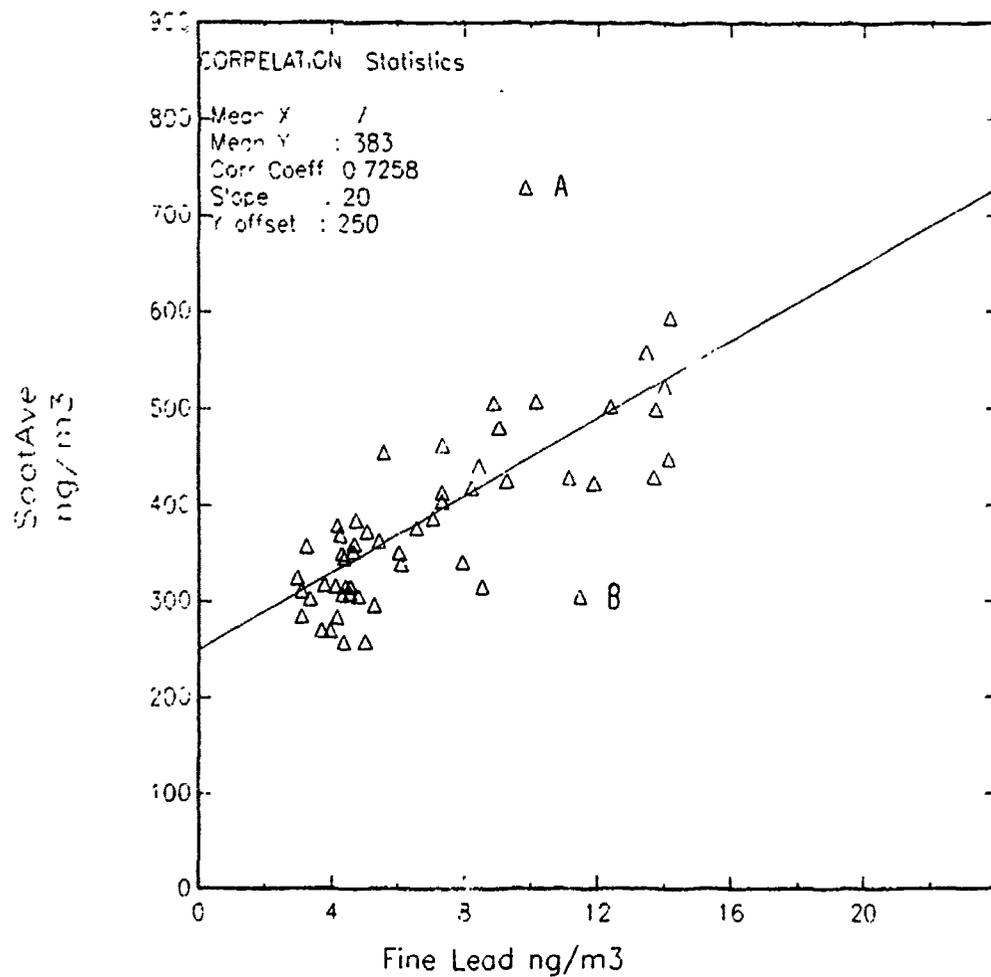


Figure 8. A comparison between the quarterly averaged soot and fine lead for quarters 2 and 3 of the NPS-NFPN data.

The soot concentrations at the seven NESCAUM stations are also very large. In order to reduce these values, we divided the reported concentrations by 2, as suggested by UCD (1991⁽³⁴⁾). This corresponds to using an absorption efficiency of 20 m²/g to calculate the soot concentration from the absorption measurements.

No absorption measurements were performed on the coarse mode aerosol, so we did not have any means of estimating the coarse soot. However, since the coarse soot is derived primarily from anthropogenic sources, such as diesel exhaust, we do not expect the coarse soot to contribute any sizable fraction of the mass. This is because all of the samplers were located in remote areas away from all roads and industrial sources.

3.1.3 Soil

The fine soil aerosol was calculated by adding the concentrations of the normal oxide forms of the major soil elements: Al₂O₃, SiO₂, CaO, K₂O, FeO, and Fe₂O₃ (Bohn et al., 1979⁽³⁵⁾, Mason and Moore, 1982⁽³⁶⁾). It was assumed that the iron was split equally between FeO and Fe₂O₃. (Eldred et al., 1986⁽²³⁾)

The fine potassium has two major sources, soil and smoke (Eldred et al., 1990⁽¹⁹⁾; Lewis et al., 1986⁽³⁷⁾). Consequently, the soil potassium had to be estimated from the data. This was accomplished by multiplying the fine silicon by a soil K to soil Si ratio. This ratio was ascertained using the coarse data in the NPS data set and assuming that all potassium in this mode was soil derived. As can be seen from Figure 9, coarse silicon and potassium correlated very well, with an average K to Si ratio of 0.12. The resulting soil equation used was:

$$\text{Fine Soil} = 1.89 \cdot \text{Al} + 2.14 \cdot \text{Si} + 1.4 \cdot \text{Ca} + 1.35 \cdot \text{Fe} + 1.2 \cdot \text{SoilK}$$

where SoilK = 0.12 · Si.

Titanium has often been included as a major soil element (Eldred et al., 1990⁽¹⁹⁾). It was not included in this calculation, because the Ti concentrations were often below the MDL. This made it very difficult to obtain an accurate average. By neglecting Ti in the soil equation an error on the order of 0.5 to 2% was introduced.

This same equation can be applied to the coarse soil, however, all of the potassium would be assumed to be soil derived. Another method which has often been used to calculate both fine and coarse soil aerosols scales up one of the soil elements

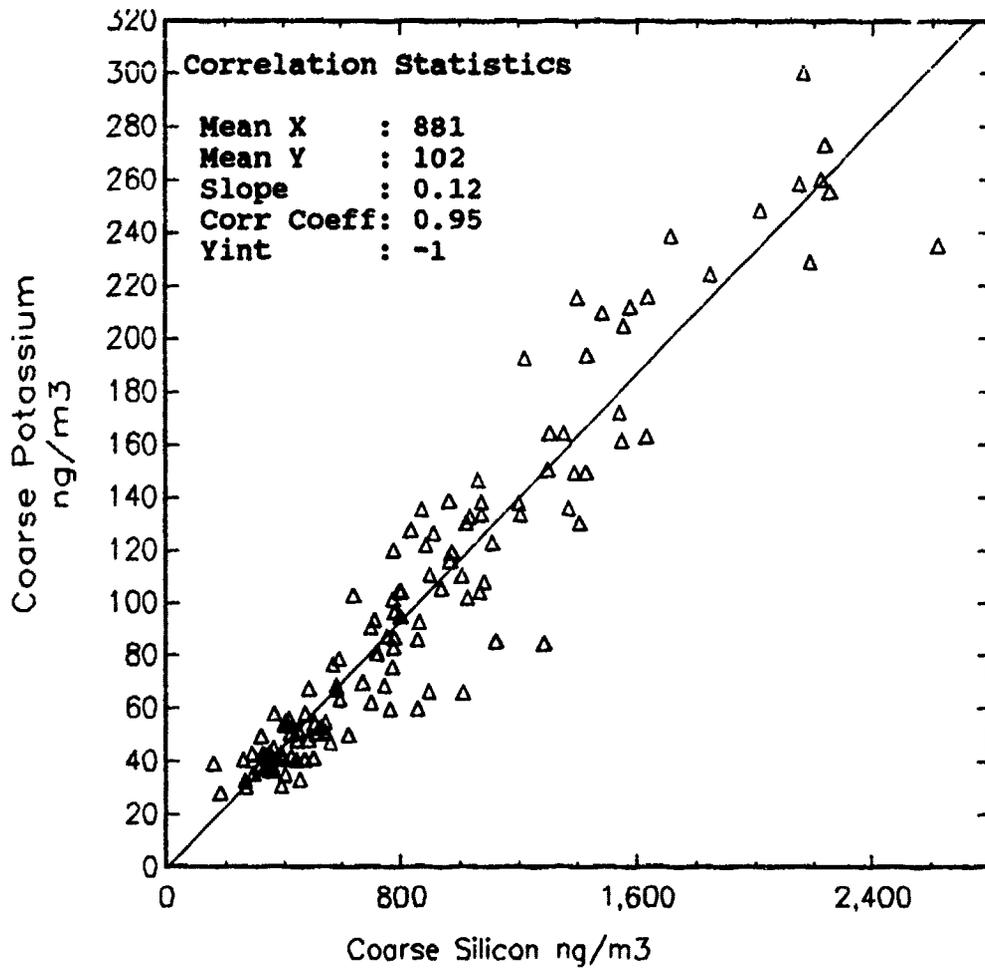


Figure 9. A comparison between the quarterly averaged coarse potassium and coarse silicon for the NPS-NFPN locations.

based on an assumed continental crust concentration (White and Macias, 1990⁽²¹⁾; Cass and McRae, 1983⁽³⁸⁾). Two common scalars used are silicon and iron. Mason and Moore (1982⁽³⁶⁾) estimated that silicon accounted for about 28% of the upper 16 km of the continental crust while iron accounted for 4%. These assumed fractions of soil for silicon and iron translate to scaling factors of 3.6 for Si and 25 for Fe.

3.1.4 Sea Salt

Fine sodium is a tracer often used to estimate the concentration of fine sea salt. Both Kowalczyh et al., (1978⁽³⁹⁾) and Taback et al., (1979⁽⁴⁰⁾) have used a Na to sea salt ratio of 0.31 to calculate the sea salt concentration. We also used this value to scale up the fine sodium to fine sea salt via the equation: sea salt = $3.2 * (\text{Na} - 15)$. The 15 ng/m³ was subtracted from the sodium concentration, because the averages from the raw data were biased high. This bias arose from the large fraction of sodium measurements below the minimum detectable limit, MDL, which were not taken into account when calculating the straight arithmetic average. To overcome this problem a log normal scheme was used to estimate a less biased arithmetic average. This was accomplished by fitting the raw sodium data to a log normal distribution to estimate the data below the MDL (Helsel, 1990; Travis and Land, 1990⁽⁴¹⁾). By calculating the average using a log normal scheme it was found that the straight arithmetic average was about 15 ng/m³ larger than the less biased log normal average, Figure 10. After subtracting 15 ng/m³ from the sodium concentrations, four data points were below zero. These data points were subsequently set to zero.

In Figure 10, there is one data point which has a much larger log normal arithmetic average than the straight arithmetic average. This outlier is due to the percentage of data points below the MDL biasing the log normal average. Due to the incomplete reliability of this averaging scheme we chose not to use its results, but rather use the mean of the results. The NA for the NESCAUM data was not scaled down by 15 ng/m³, because a concentration below the MDL was estimated by using half the MDL.

Another possible tracer for sea salt is chlorine. However, Cl is chemically reactive in the atmosphere and dissociates from the sea salt with time (Junge, 1972⁽²⁶⁾).

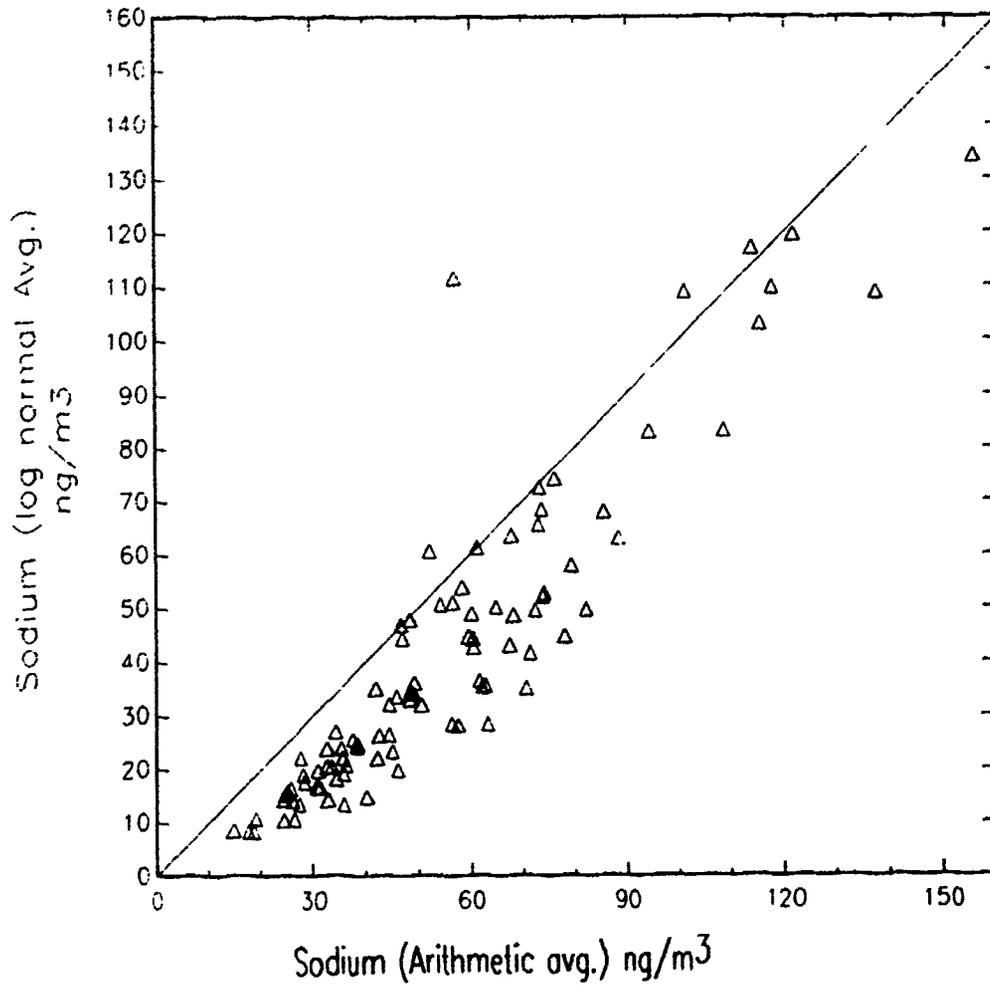


Figure 10. A comparison of the arithmetic and log normal averaging schemes to calculate the quarterly averages of fine sodium for the NPS-NFPN data.

The Cl⁻ also tends to volatilize from Teflon filters during collection (UCD, 1991⁽³⁴⁾). For this reason, Cl was not used in the sea salt estimates.

The sodium for the coarse data was rarely above the minimum detectable limit. Due to this a coarse sea salt could not be obtained.

3.1.5 Organics

Organics are one of the major constituents of the fine aerosol mass. Unfortunately, they are difficult to measure accurately. One method used to estimate organic matter scales up the tracer, organic hydrogen. This method has been used with success on aggregated data by Cahill et al., (1989⁽²⁵⁾) for the NPS-NFPN data set and by Poirot et al., (1990⁽²⁰⁾) for the NESCAUM data set. Organics can also be estimated by the remaining mass method which assumes that organics constitute a constant fraction of the remaining mass (Cahill et al., 1989⁽²⁵⁾). The remaining mass is calculated by the equation: Remaining Mass = Fine Mass - SO₄⁻² - Cation - Soil - Salt - Soot.

The NESCAUM data also contains the fine organic carbon collected on quartz filters at the NY and MA sites. The reported concentrations have already been corrected for artifact. The organic carbon can be used as a tracer for organics, and scaled up to estimate the organic mass (Wolff et al., 1986⁽⁴³⁾; Ferman et al., 1981⁽⁴⁴⁾). The hydrogen method is used to estimate the organic concentration for this study while the other two methods are used to check this organic mass.

The major problem with estimating organics via organic hydrogen is separating the inorganic from the organic hydrogen. There are three main contributors to inorganic hydrogen: ammonium sulfate, ammonium nitrate and, water (Cahill et al., 1989⁽²⁵⁾). In order to measure the hydrogen concentration the sample is placed into a vacuum. This causes the majority of the water to desorb from the sample before the hydrogen concentration is measured. The ammonium nitrate aerosol is extremely volatile on Teflon filters, and it is believed that most of it would also be removed when the sample is placed into the vacuum (Shaw et al., 1982⁽⁴⁵⁾, UCD 1991⁽³⁴⁾). The contributors to the remaining hydrogen should be only organics and ammonium due to the ammonium sulfate. Consequently, if the ammonium hydrogen is removed, only the

organic hydrogen is left. It should be noted that a fraction of volatile organic matter will also be removed in the vacuum which must be taken into account.

We estimated the organic concentration using the equation; $\text{organic} = C \cdot (\text{H-Cation}/D)$. The constant C was found through the fitting process to be discussed later. The constant D is determined by the assumed makeup of the cation term associated with sulfate. For ammonium sulfate $D=4.5$, for bisulfate $D=3.8$, and $D=1$ for sulfuric acid.

In order to estimate the organics via the remaining mass method the ratio of organic material to the remaining mass needs to be known. Cahill et al., (1989⁽²⁵⁾) calculated this ratio from two different studies. They found that from the VISTTA study conducted in Northern AZ 53% of the remaining mass was contributed by organic matter. From the RESOLVE study in southern California 67% of the remaining mass was organics.

The equation we used to estimate the organics from the carbon measurements is that suggested by UCD (1991⁽³⁴⁾): $\text{Organic} = 1.4 \cdot (\text{OCLT} + \text{OCHT}) + 400$. The 1.4 coefficient accounts for the non-carbon contribution to organic mass. OCLT and OCHT refer to the organic carbon measured at low and high temperatures respectively. The 400 is added, because they felt that the organic artifact correction was too large. The large correction was due to the high uncertainty in this estimate, see section 2.2.2 on NESCAUM sampling analysis.

The coarse organic aerosol type has been found to have varying importance to the coarse mass. In a study conducted in southern California (Appel et al., 1976⁽⁴⁶⁾) no coarse organic mass was found. However, coarse organic carbon was found in studies conducted in Charleston, WV (Lewis and Macias, 1980⁽²⁷⁾), St. Louis, MO, and Covina, CA (Spicer, 1976⁽⁴⁷⁾) where it accounted for about 10% of the coarse mass. All of these studies were conducted in cities where part of the coarse organic mass could result from primary emissions such as automobile lubricants (Cukor et al., 1972⁽⁴⁸⁾).

In this study we did not calculate the organic mass for several reasons. First, since the samplers were located in remote areas few coarse organic particles from anthropogenic sources would be found. Consequently we believed that the coarse organic mass would be much less than the 10% of the coarse mass found in the studies

located in sites which makes the organic fraction rather unimportant. Also, the NPS-NFPN data did not contain hydrogen measurement for the coarse data, so the hydrogen method could not be used to estimate the coarse organic mass. A revised remaining mass method could be used, but any estimates of the organic concentration would be rather suspect, and we have no other estimates to compare it to.

3.1.6 Nitrates

Nitrates are a very elusive aerosol type to measure. They tend to volatilize from the Teflon filters making it very difficult to obtain a measurement representative of their true atmospheric concentrations (Shaw et al., 1982⁽⁴⁵⁾; Appel et al., 1981⁽⁴⁹⁾; White and Macias, 1987⁽⁵⁰⁾). The data sets used in this study do not contain any measurements that could be used to estimate nitrates. We feel that nitrates are the only remaining large aerosol type which has not been taken into account. Therefore, the difference between the measured mass and calculated mass, sum of the sulfate, water, cation, salt, soot, soil, and organic, will be assumed to be constituted mainly of nitrate.

Since the nitrates are generally a secondary aerosol type, their contribution to the coarse mode should be very small. Consequently, the difference between the calculated and measured coarse mass cannot be assumed to be primarily nitrates.

3.2 Determining the Unknown Aerosol Constants for the Fine Aerosol Types

The aerosol equations discussed to this point have fully defined the sulfate, soil, soot, and salt for all stations, and the sulfate cation for the western sites, for the fine aerosol mass. The constants needed to determine the remaining fine aerosol types, the cation concentration in the East, water, and organics at all locations were found by fitting the calculated fine mass to the measured fine mass. The calculated mass is the sum of all seven estimated aerosol types.

To allow for regional variation of aerosol scaling constants, the U.S. was divided into three regions, the Northwest, Southwest, and the East as defined in Figure 11. In the fitting process, it was assumed that the organic constant was the same for each region. This implies that the average organic composition, carbon to hydrogen ratio, is not spatially dependent, and a constant fraction of the organic material is removed from the sample when it is placed into the vacuum for hydrogen measurement.

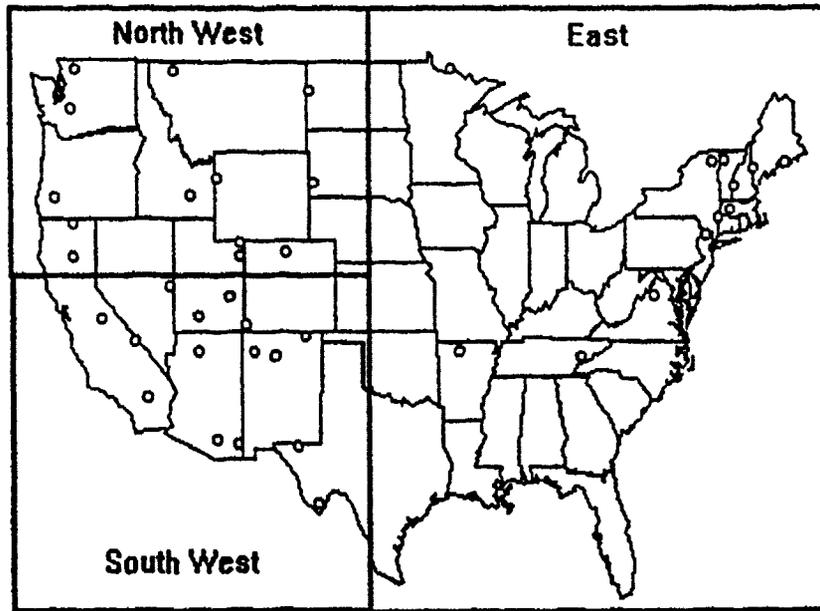


Figure 11. The three regions, and the stations located in each region, that the fitting process was conducted over to find the undetermined aerosol constants.

In performing such a fitting process to calculate the remaining aerosol type concentrations we are becoming dependent upon the adjustments to the fine mass concentrations which were used to compensate for the increase between sampling and analysis, see section 2.1.2. This can introduce uncertainty in the fitting process, because we do not know the chemical composition of the material responsible for this increased mass.

3.2.1 Fitting the Southwest Region

In the Southwest there were two unknown constants which needed to be estimated to approximate the water and organic aerosols. Since the organic constant was assumed not to be spatially dependent, the constant determined in this region was applied to the other two regions. It was assumed that the water constant in the Southwest was invariant with season. The R.H. in this region rarely exceeds 80%, hence, the soluble salts seldom reach the deliquescence point, and the hysteresis effect is negligible.

In order to perform the fitting process we also assumed that the nitrate concentration was negligible. The nitrate concentrations in the rural Southwest have been found to be small by White and Macias (1987⁽⁵⁰⁾) using data from the WRAQS study. Also the nitrate aerosol is very volatile on Teflon filters, and a fraction of the nitrate collected will volatilize from the filters before measurement, reducing the importance of nitrates further. Therefore, we assumed that the measured mass was estimated by the seven aerosol equations already defined.

The fitting process involved first estimating the two unknown constants then comparing the calculated mass to the measured mass by linear regression. The two constants were adjusted in order to get as close to a 1 to 1 correspondence between the two masses as possible. The best fit was found when the correlation coefficient and slope were as close to one as possible without greatly overestimating the measured mass. By not allowing the calculated mass to overestimate the measured mass the stations where the nitrate assumption is not valid will underestimate the measured mass. The difference is assumed to be made up of primarily nitrates.

The two constants found through this fitting process were, $C_{\text{water}} = 0.1$ and $C_{\text{organic}} = 13$. The water concentration of 10% of the sulfate salt is within the range

found by Winkler and Junge (1972⁽³¹⁾) for sulfate in relative humidities between 40 and 60 percent, which is the estimated R.H. of the lab.

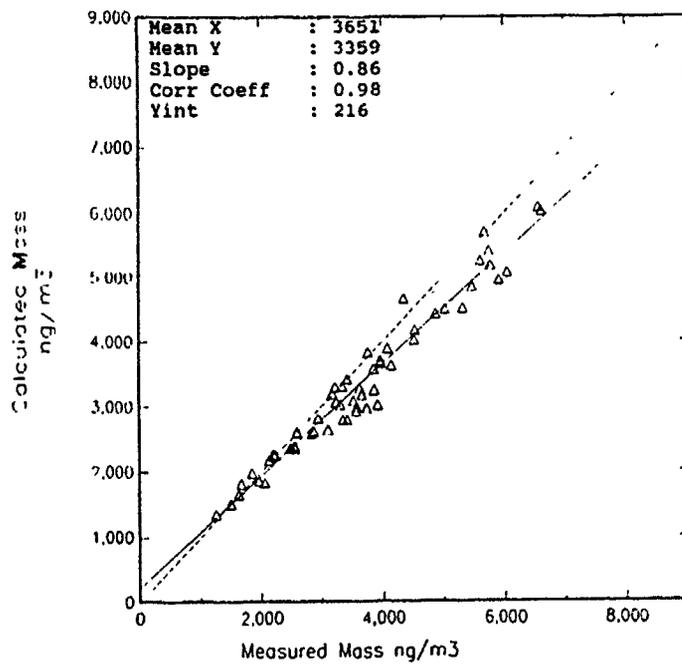
An estimate of the average organic molecule contains 9% hydrogen and 71% carbon (Eldred et al., 1990⁽¹⁹⁾; and Poirot et al., 1990⁽²⁰⁾). Based on this hydrogen content, the organic constant would be approximately 11. The increase in this constant accounts for that part of the organic concentration that volatilized from the filter during analysis. UCD (1989⁽³⁴⁾) using the IMPROVE data set found that approximately 25% of the organic matter volatilizes from the filters at low temperature. Assuming that all of this organic material will be removed from the filter during the hydrogen analysis, the C_{Organic} becomes 13.75 which is close to the constant we found.

The results for the calculated mass using these constants are presented in Figure 12. Figure 12A presents the scatter plot and correlation statistics between the two masses. The dotted line in this plot, represents the one to one correspondence between the two masses. The solid line is a linear regression line fitted to the data. Figure 12B plots the residues of the calculated mass. As can be seen, there is a high correlation between the two masses, $r = 0.98$. Also, the overestimation was held to only a few data points, and generally under seven percent. The slope of the regression line is only 0.86. To increase this any substantial amount without changing any of the assumptions would require over estimating some data points by more than 15% which seemed unreasonable.

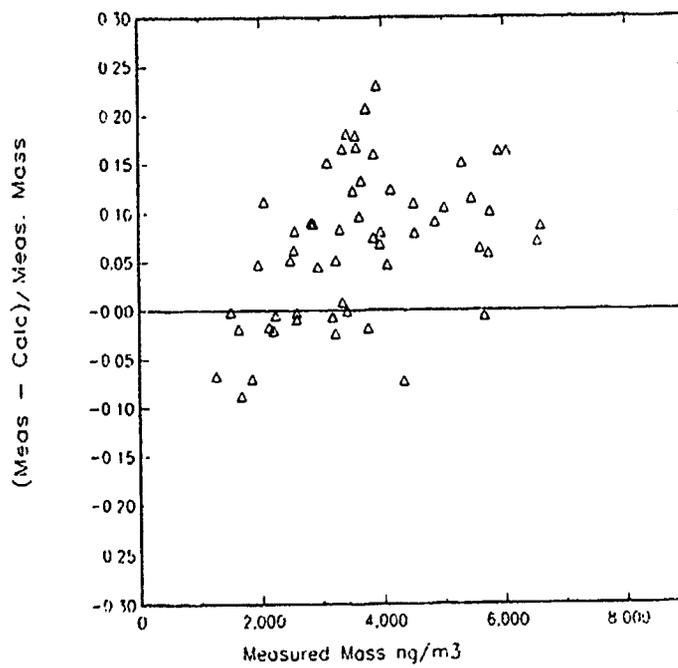
3.2.2 Fitting the Eastern Region

The constants for the eastern region were fitted next. It has been found that there are major differences between the North and Southeast (Husar and Wilson, 1990⁽⁵¹⁾). Unfortunately, there are only three NPS-NFPN stations which can be considered to be in the Southeast. Therefore, the constants were fitted to all eastern stations at once.

In the fitting process, we again assumed that the nitrate concentrations were negligible. The eastern aerosol is more acidic due to the incomplete neutralization of the sulfate anion. Therefore, when the sulfate aerosol comes into contact with the nitrate, such as on a filter, the sulfate causes the nitrate to volatilize and associates with the ammonium cation. Shaw et al., (1982⁽⁴⁵⁾) showed at a site in North Carolina between 37 and 90% of the particulate nitrate was volatilized from the Teflon filters.



A)



B)

Figure 12. A) A comparison between the calculated fine mass and the measured fine mass for the Southwest. B) A plot of the residuals of the calculated fine mass.

Due to the assumption of a spatial independent organic constant, the organic constant found in the Southwest was used for the East. This left the water and cation constants to be determined. Both of these constants were allowed to vary with time, and the cation constant could vary with space for the three southern stations from the N.E. stations. To allow for the temporal variation, each quarter was fitted separately. Figure 13 illustrates the resultant scatter plot of the calculated and measured mass from the fitting process, and the residuals of the calculated fine mass. As can be seen, there is excellent correlation, $r = 0.98$, between the two masses with a slope close to one, and little overestimation. The constants used to create this fit are presented in Table 2.

Table 2. The best fitted aerosol constants for the Northeast, by quarter.

	Q1	Q2	Q3	Q4
Cation				
Northeast	0.75	0.85	0.41	0.75
Shenandoah	0.75	0.85	0.41	0.75
Great Smokey Mnt.	0.75	0.85	0.41	0.75
Buffalo River	0.8	0.8	0.8	0.8
Water				
All Eastern Stations	0.17	0.15	0.25	0.20
Organic				
All Stations	13	13	13	13

Although the cation constant was allowed to vary spatially, the only station which significantly differed from the other stations was Buffalo River where the constant equaled 0.8 for all four quarters. Buffalo River, AR was located close to the east - west boundary as we defined it. In the West, the sulfate was assumed to be fully neutralized for all seasons. Consequently, for Buffalo River, AR to have a nonvarying cation constant close to 1 (fully neutralized sulfate anion) seemed reasonable.

All the other stations in the East had approximately the same coefficients for a given season. As shown in the table, the highest cation constant occurred in the second quarter, the lowest in the third quarter, and the same for the first and fourth quarters. The constant for the first and fourth quarter is roughly equivalent to a 50% mixture of ammonium sulfate and bisulfate. Poirot et al., (1990⁽²⁰⁾) suggest that this is the likely composition of sulfate for the Northeast during the winter seasons.

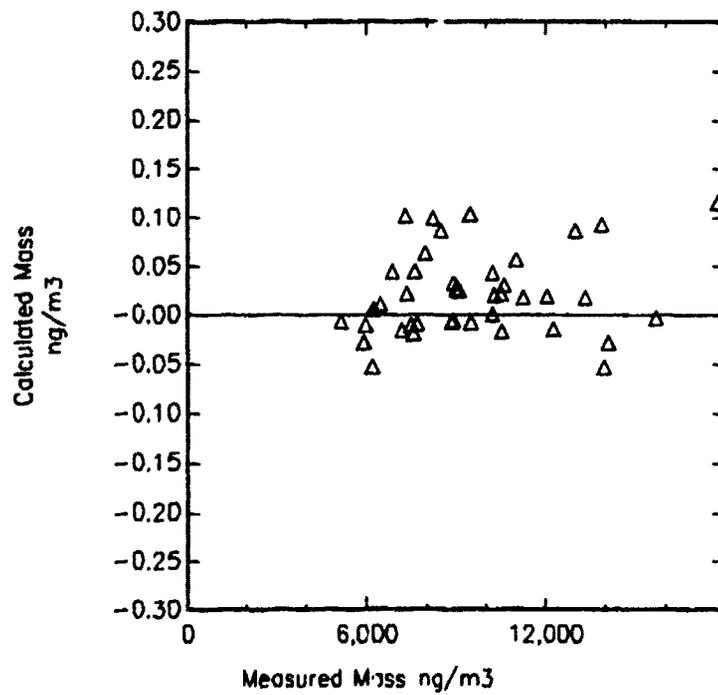
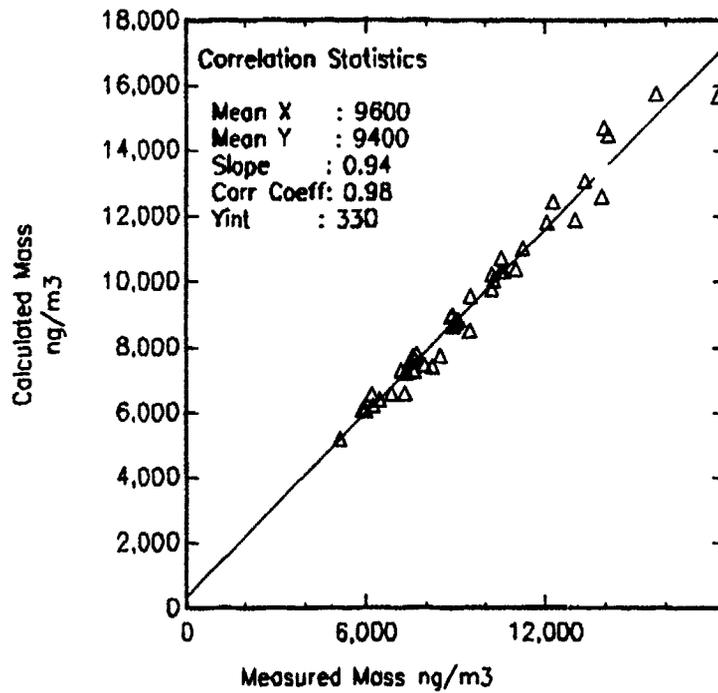


Figure 13. A) A comparison between the calculated fine mass and the measured fine mass for the East. B) A plot of the residuals of the calculated fine mass.

From the National Atmospheric Deposition Program (NADP) precipitation chemistry data (NADP, 1982⁽⁵²⁾) it was found that the maximum cation concentrations in the northeastern U.S. occurred during the second quarter months. This coincides with the fitted cation constants where the second quarter had the largest values. Two other rain chemistry studies located in the Midwest had similar results (Ramundo and Seastedt, 1990⁽⁵³⁾).

The third quarter has the lowest cation constant, yet the rainfall studies showed this quarter to have some of the highest cation concentrations. This dilemma will be rectified in section 4 where it will be shown that this quarter had the highest eastern sulfate concentrations thus the least neutralization. The value of the third quarter constant represents a slightly more acidic sulfate aerosol than ammonium bisulfate.

The water constants found were relatively constant for all four quarters. Interesting, the water constants seem to be associated with the acidity of the aerosol. The largest water constant occurred during quarter 3 which was also the most acidic quarter while the smallest water constant occurred during quarter 2, the least acidic quarter. These water constants are approximately twice that found for the Southwest. This increase was most likely due to the hysteresis effect.

3.2.3 Sensitivity Analysis for the Eastern Stations

The cation constant in the system of equations is very important, because the inferred water and organic concentrations are dependent upon its value. To understand the effect of this constant on the calculated mass, a sensitivity analysis was conducted on the eastern data with the water constant set equal to 0.1. The results of this analysis for the third quarter are presented as pie charts for each station in Figures 14 & 16.

The sensitivity analysis was conducted by letting the cation constant vary from its maximum value to its minimum value. The maximum value physically possible is 1 which corresponds to an aerosol type of pure ammonium sulfate. Figure 14 presents the resultant aerosol type concentrations assuming a value of one for the third quarter. A large percentage of the measured mass is undetermined. We have called this part of the mass "unknown." Also, the organic concentration in the Northeast is greater than 25% of the fine mass which is more than half the concentration attributable to the sulfur

aerosol types, sulfate, cation and water. At the two southern stations, Shenandoah and Smokey Mnts., the organic concentration is nearly nonexistent, and the sulfur aerosol types contribute about 65% compared to 45% for the northern stations.

At Buffalo River the aerosol makeup is different from that of the other two southern stations. At this site, the sulfur aerosol types account for less of the fine mass than at the other two southern stations, but both the organics and soil constitute more of the fine mass, about 20% each compared to 5% at Shenandoah and Smokey Mnts. These differences are most likely due to the western location of Buffalo River where it experiences a different regional aerosol composition than the other eastern stations.

The variations between the northern and southern stations in the eastern U.S. may be due to differences in regional aerosol chemistry, or it may be due to the different sampling techniques used in the NPS-NFPN and NESCAUM monitoring networks. Figure 15 is the same plot as Figure 14 using the fourth quarter data. This figure contains data for Acadia ME from the NPS data set which lies in the NESCAUM region. Again, the sulfur aerosol types concentrations are greater in the two southern stations than in the Northeast, except for Acadia which has an aerosol make up similar to that of the southern stations. This illustrates that the differences between the North and Southeast were probably due to differences in the sampling techniques than the regional aerosol.

Figure 16 presents the break down of the aerosol types assuming a minimum value for the cation constant. The value for the minimum cation constant changed for each quarter, and was dependent on two constraints. The first constraint was due to the defined aerosol equations, and a decreasing cation term. As the cation constant was decreased the aerosol concentration attributed to the sulfur aerosol types decreased due to the decreases in cation and water. However, the organic concentrations acted in the reverse manner and increased. The increase in organics was greater than the decrease in the sulfur aerosol types causing the unknown term to decrease. The minimum cation constant was that value which corresponded to an unknown of zero. However, if the cation constant was equal to 0.056 which corresponds to a sulfuric acid aerosol before the unknown became zero, then 0.056 was the minimum cation value.

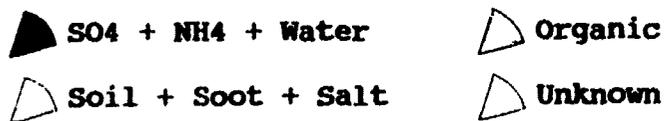
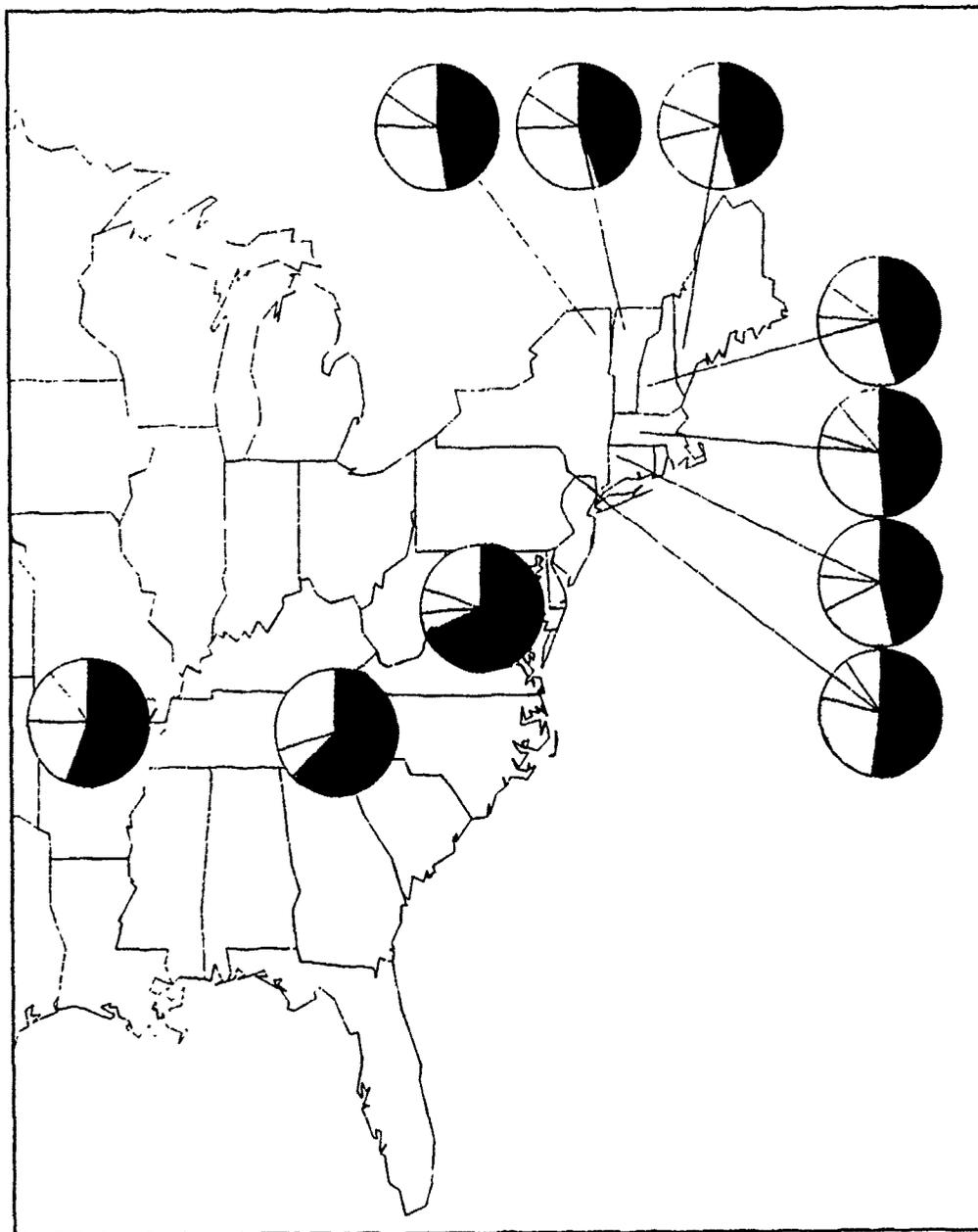


Figure 14. The 3rd quarter aerosol constituents at each eastern location. The sulfate is assumed to be in the form of ammonium sulfate, and the water content was estimated to be 10% of the ammonium sulfate.

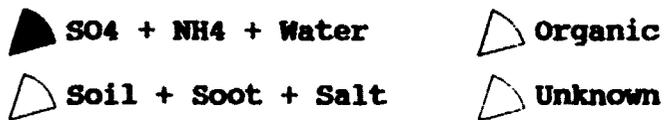
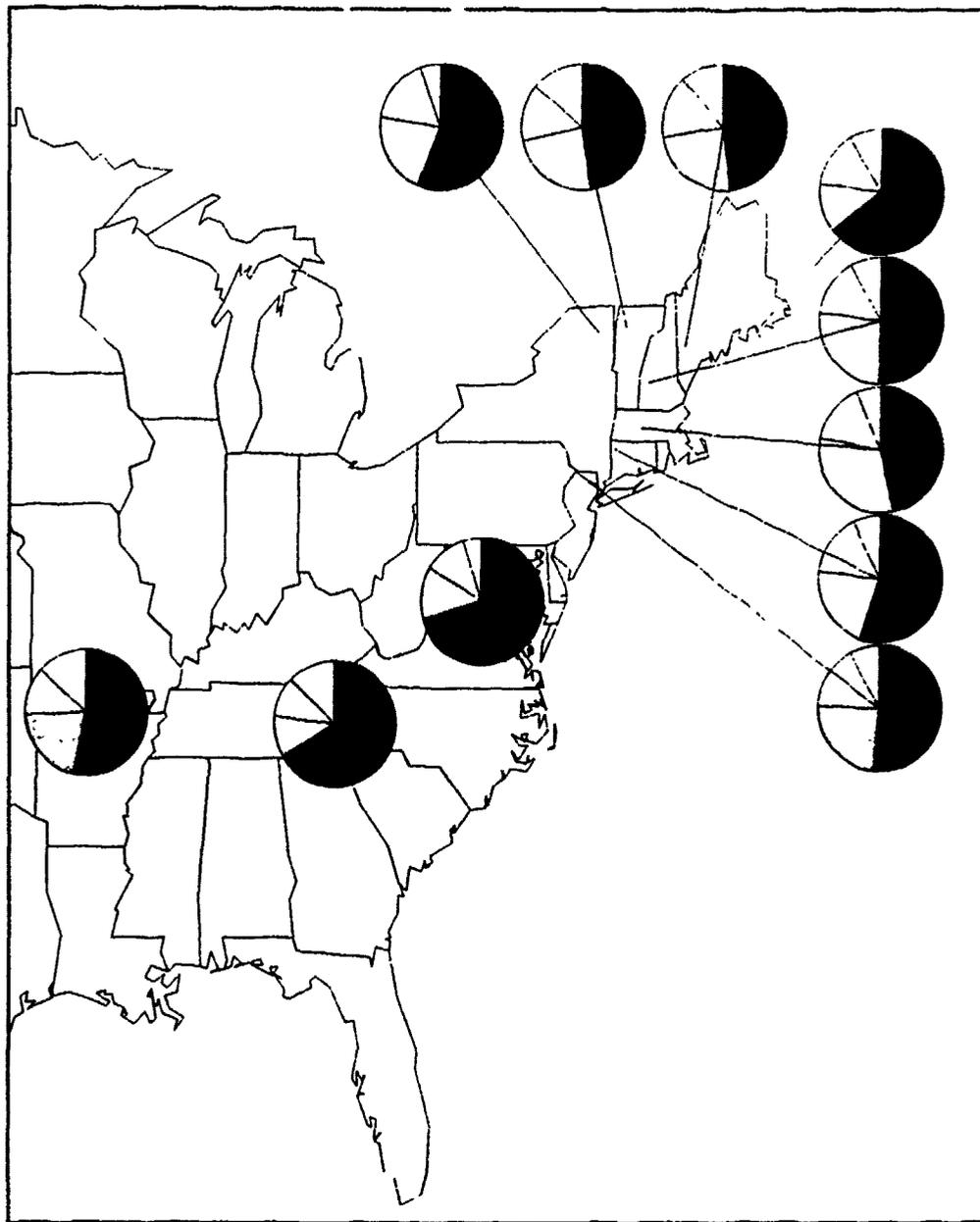


Figure 15. The 4th quarter aerosol constituents at each eastern location. The sulfate is assumed to be in the form of ammonium sulfate, and the water content was estimated to be 10% of the ammonium sulfate.

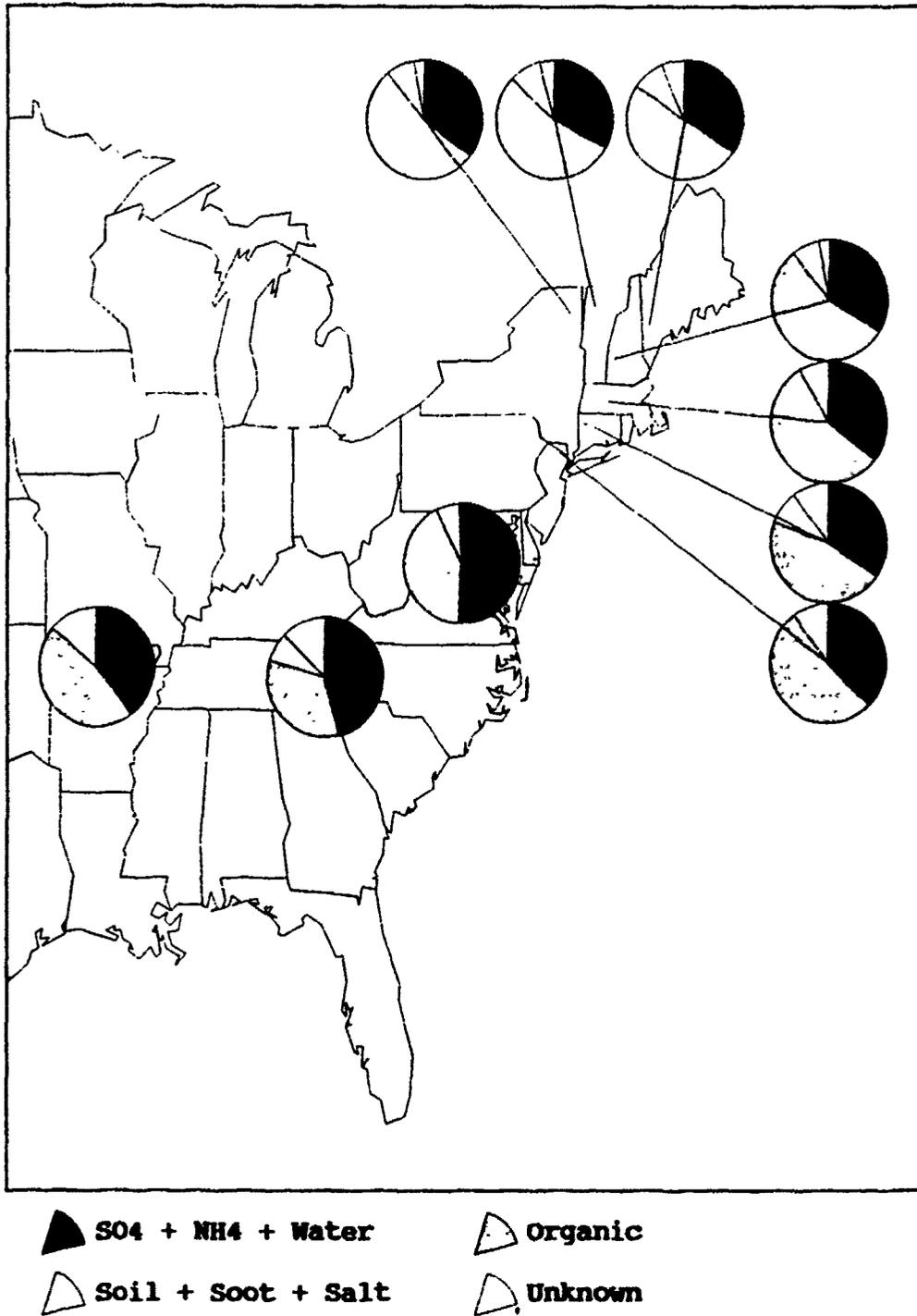


Figure 16. The 3rd quarter aerosol constituents at each eastern location. The sulfate is assumed to be in the form of sulfuric acid, and the water content was estimated to be 10% of the sulfuric acid.

It was found that for the third quarter, the minimum constant was 0.056 which corresponded to an sulfate cation of sulfuric acid. The resultant aerosol types concentrations are presented in Figure 16. The six defined aerosol types account for virtually all of the measured mass except at Smokey Mnts, TN and Mohaw' CT. The organic concentrations increased by more than 30% for all stations over those presented in Figure 14. The sulfates decreased by about 15%. This is a wide range of variation for these aerosol types which placed much importance on finding the correct cation constant.

The minimum cation constant for the other three quarters was limited by the unknown aerosol function reaching zero. This occurred at a cation aerosol type corresponding to ammonium bisulfate. Therefore, the possible variation in the organic, cation and unknown concentrations for these three quarters was smaller than for the third quarter.

3.2.4 Fitting The Northwest Region

In the Northwest, only the organic and water constants were undetermined. Since we assumed that the organic constant did not vary with season or location we used the value found from the Southwest fitting process. Consequently, only the water constant needed to be determined. In order to calculate the water constant, we used relative humidity charts to estimate the average local R.H. values and temperature for each season (Ruffner and Bair, 1979⁽⁵⁵⁾). The constants applied were those found in the Southwest for periods where the R.H. was below 70% on average, and those determined in the East for higher R.H. periods. If the average temperature was below freezing we used the Southwest constant reasoning that the water would be in a crystalline state and the aerosol could not absorb it.

Table 3 presents the water constants used for this region. The temperature and relative humidity varied over a warm and cold season, so the constants were defined for quarters 1 and 4, (cold season), and quarters 2 and 3, (warm season). The cold season generally has higher constants than the warm season for the coastal and more southern stations. This is because the highest R.H. values were found during this season and the temperature was usually above freezing.

A best fitting process was not used in this region because the nitrate aerosol may not be negligible (White and Macias, 1987⁽⁵⁰⁾). We had no means of estimating the nitrate aerosol to determine if this was true or not. If the nitrates are not negligible, then a best fitting process would overestimate the water contribution to the sample mass.

Table 3. The water constants for the Northwest Stations.

<u>Station</u>	<u>Cold Season (O1,O4)</u>	<u>Warm Season (O2,O3)</u>
Crater Lake	0.2	0.15
Mount Rainier	0.2	0.15
Lassen Volcanic	0.15	0.1
North Cascades	0.2	0.15
Lava Beds	0.2	0.15
Craters of Moon	0.15	0.1
Grand Teton	0.15	0.1
Glacier	0.1	0.15
Dinosaur	0.1	0.1
Browns Park	0.1	0.1
Rocky Mnt.	0.1	0.1
Roosevelt	0.1	0.15
Wind Cave	0.1	0.1
Voyager	0.1	0.15

Figure 17 presents the scatter plot of the calculated vs measured mass, and the residuals. As can be seen from the scatter plot, the correlation is very good. The slope is only 0.77, but the intercept is not large. The low slope and small intercept is an indication that we did not account for some of the aerosol mass such as nitrates. As shown in the residual plot, only one data point overestimates the measured mass. Also, of the three regions, the Northwest has the largest residuals. The majority are greater than 10% with two data points even greater than 30%. The possible constituents of this large unknown are discussed in section 5.

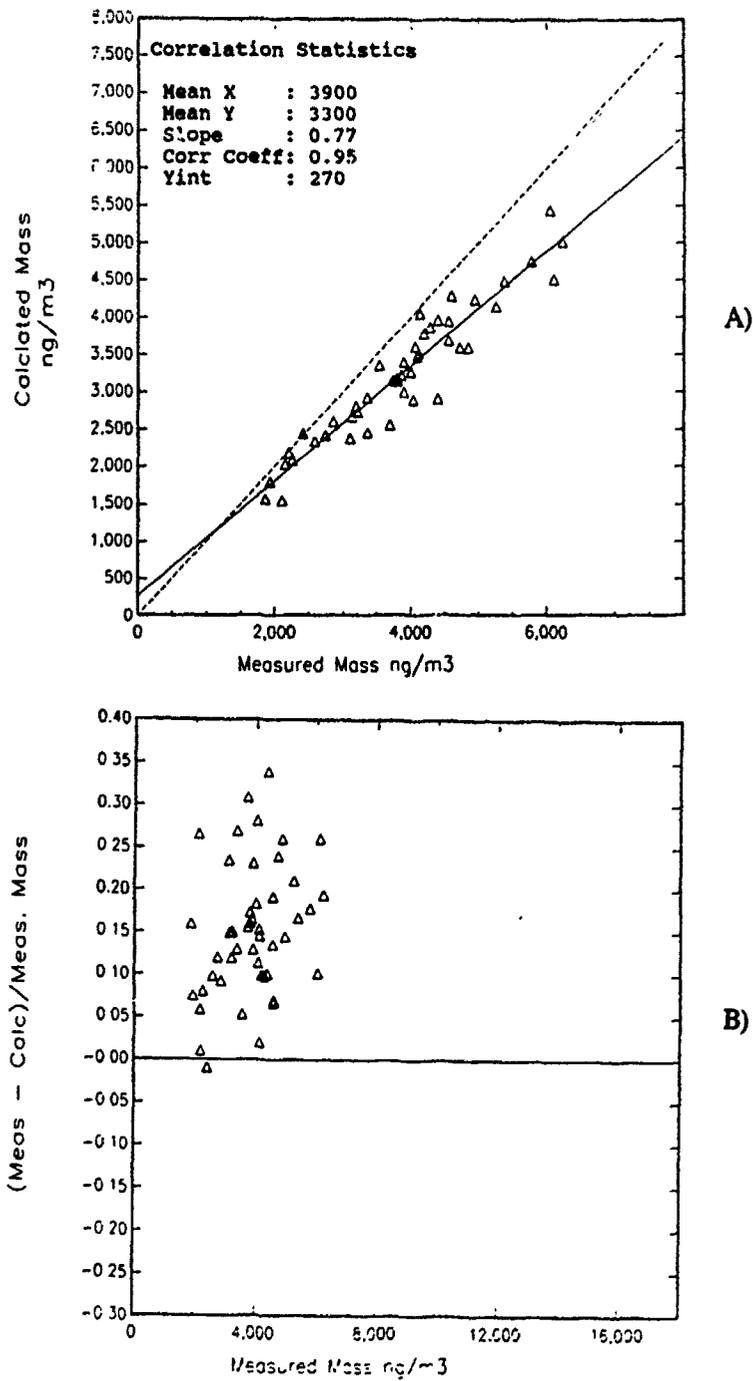


Figure 17. A) A comparison between the calculated fine mass and the measured fine mass for the Northwest. B) A plot of the residuals of the calculated fine mass.

3.2.5 The Entire U.S.

The comparison of the calculated and the measured fine aerosol mass for the U.S. is presented in Figure 18. In performing the fitting process we were trying to satisfy three criteria based on a linear regression comparison of the measured and calculated mass. These criteria were to have a correlation coefficient and slope as close to one as possible, and keep the overestimation of the measured mass by the calculated mass to a minimum. As can be seen from the scatter plot, the three criteria have been met. Both the slope and correlation coefficient are 0.99 and the overestimation of the measured mass has been kept to a minimum with only six data points overestimating the measured mass by more than 5%. The variation of the residuals have also been kept to a reasonable level with the bulk of the residuals falling between -5 and 15 percent. There are, however, a number of points which lie outside this range and must be examined closer.

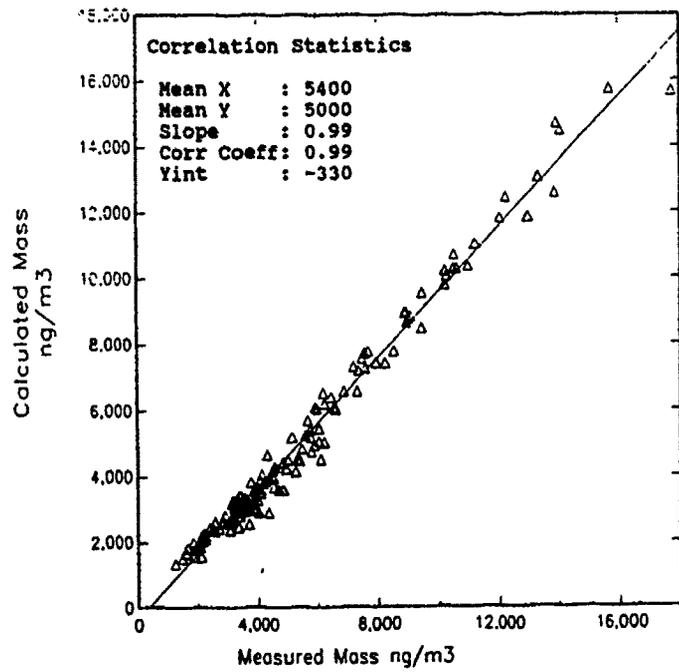
3.3 Examining Alternative Fine Aerosol Equations

Both the fine soil and organic aerosols had more than one method to calculate their concentrations. In order to perform the fitting process the fine soil aerosol was calculated using the oxide method, and the organic aerosol was calculated using the hydrogen method. This section compares the alternative methods available to estimate the soil and organic concentrations with those used in the fitting process.

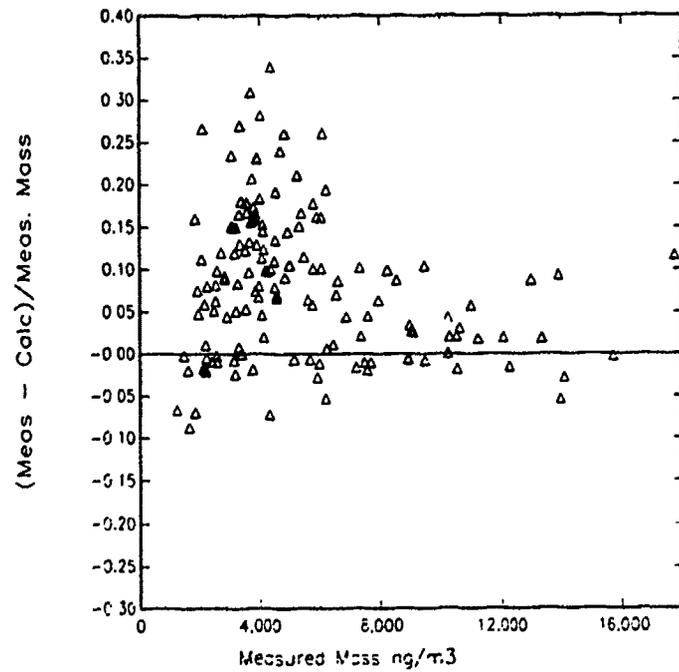
3.3.1 Soil

The soil equation used in this study scaled up the major soil elements according to their assumed soil oxide forms. As discussed in section 3.1.3, an alternative method simply scales up one of the soil elements based on an assumed continental crust concentration. Using this alternative method the fine silicon concentration was multiplied by 3.6 to calculate the fine soil.

A comparison between the two methods is presented as a scatter plot in Figure 19. As shown, there is excellent agreement between the two methods. It should be noted that the soil aerosol is most likely a combination of crustal material and clay. Clay has a Si content about 25% (Mason and Moore, 1982⁽³⁶⁾) less than the continental



A)



B)

Figure 18. A) A comparison between the calculated fine mass and the measured fine mass for all three regions. B) A plot of the residuals of the calculated fine mass.

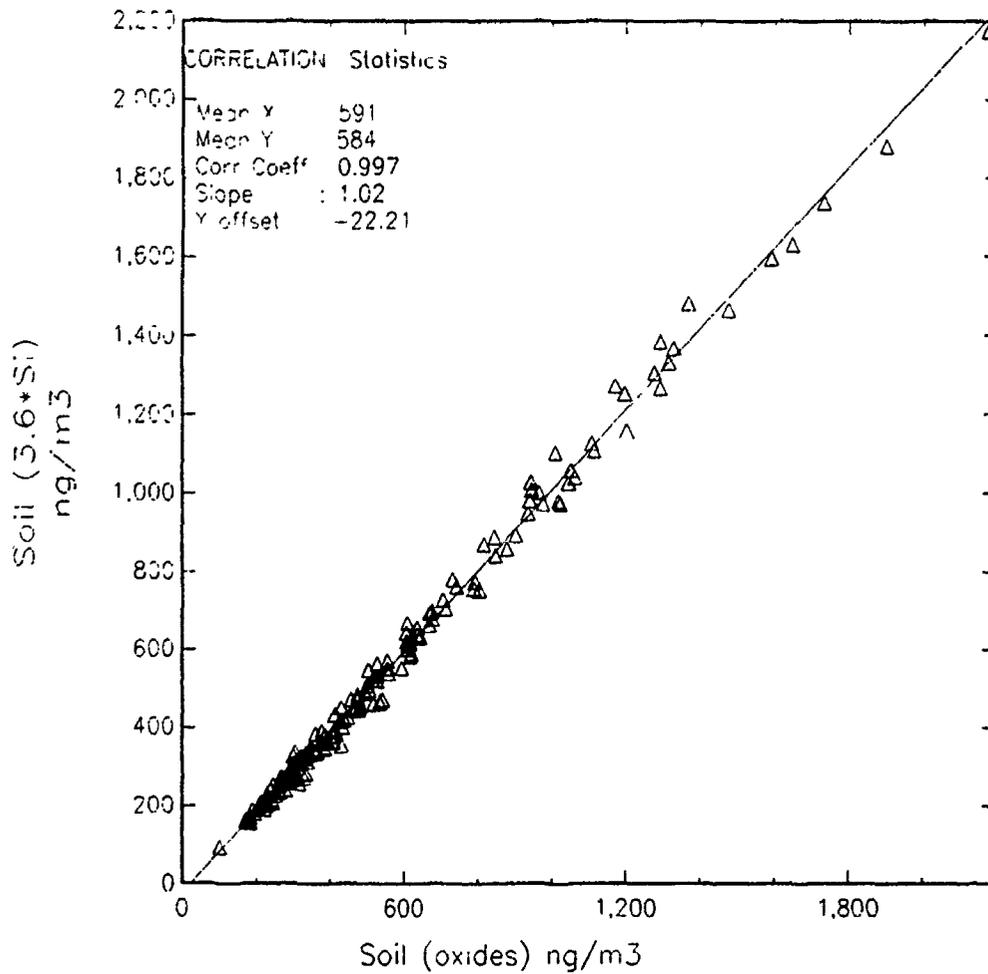


Figure 19. A comparison between the oxide method and scaling silicon to estimate the fine soil concentrations.

crust, so the soil estimated based on the Si may be underestimated. Also, Cahill et al., (1989⁽²⁵⁾) believe that the oxide soil equation accounts for only 84% of the fine soil mass, so the oxide method may also be slightly underestimated. However, using this same scaling factor and coarse Si, White and Macias (1990⁽²¹⁾) found near one to one correlation between coarse soil and mass at Spirit Mnt, NV.

3.3.2 Organics

The hydrogen method was used in the fitting process to calculate the organic concentrations. As discussed in section 3.1.5, two alternative methods which use the remaining mass and organic carbon can also be used to estimate the organic concentration. The remaining mass method can be applied to all stations, but only two stations in the NESCAUM data set contain the organic carbon measurements.

Figure 20 presents a comparison of organics by the hydrogen and remaining mass methods for all locations and quarters. The dotted line is the one to one line while the solid line is the linear regression line. The organic concentrations from the hydrogen method are those obtained from the fitting process. The constant used to estimate the organics by remaining mass was the larger constant of 0.67 found from the RESOLVE study. As can be seen, these two techniques correlate very well, $r = 0.96$. This is consistent with the analysis of the NESCAUM data by Poirot et al., (1990⁽²⁰⁾) The high correlation adds a degree of confidence that the two very different methods are estimating the same quantity. The only problem is that the magnitudes of the two methods differ. At high organic concentrations the organics by the hydrogen method are about 15% larger than those by the remaining mass method. However, at low organic concentrations this is reversed with organics by remaining mass being about 15% larger.

A comparison between the remaining mass and hydrogen methods for the stations west of the Mississippi is presented in Figure 21A. This constitutes the lower half of Figure 20 where the concentrations are much smaller. In this region, the organics by remaining mass are larger than those estimated by hydrogen, especially for the southwestern stations which have the lowest organic concentrations. This difference can be resolved by using a smaller constant for the remaining mass method. For example, using a scaling factor of 0.6 as apposed to 0.67 reduces the remaining mass

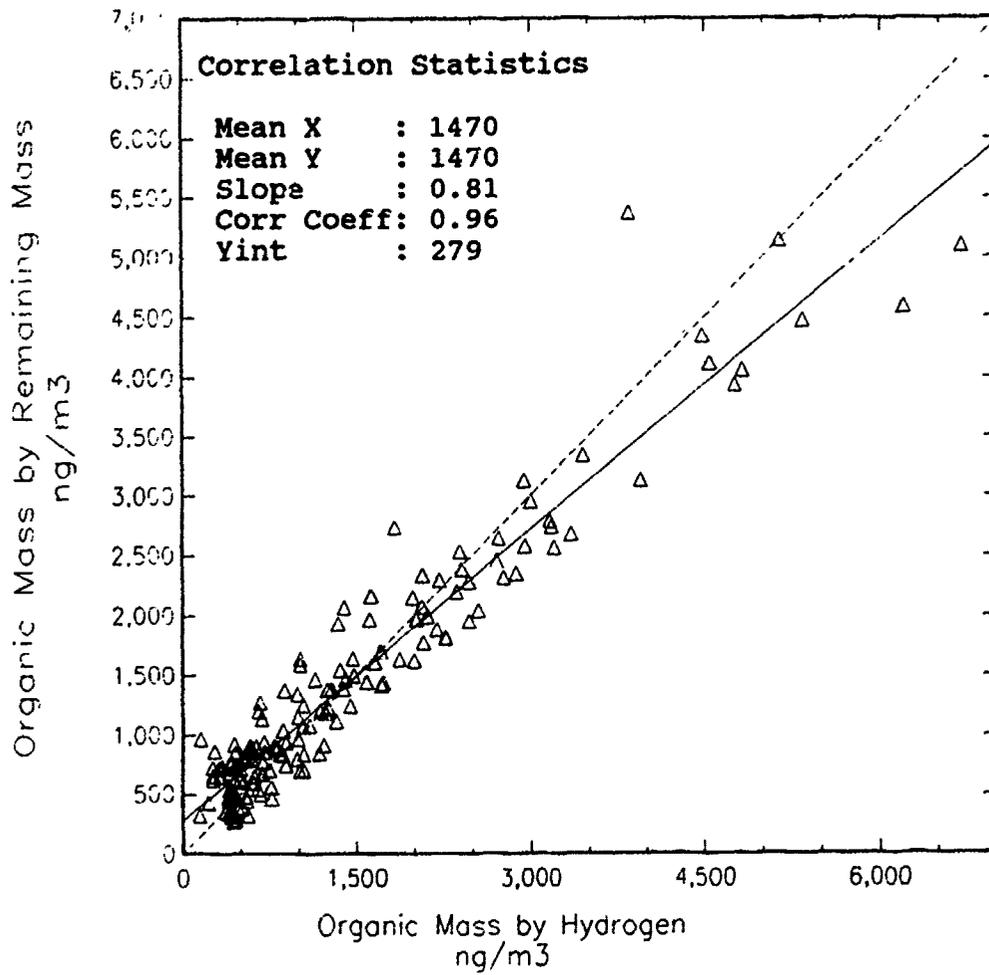
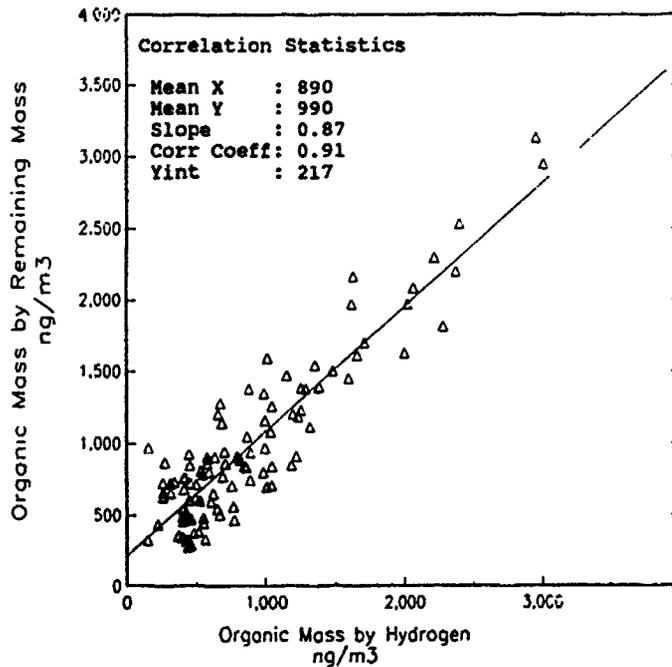
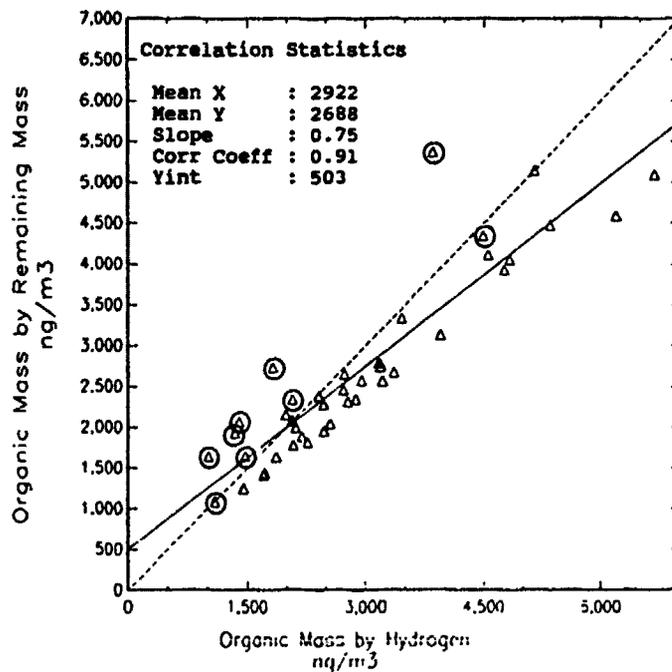


Figure 20. A comparison between the remaining mass and hydrogen methods to estimate the organics for all stations and quarters.



A)



B)

Figure 21. A comparison between the remaining mass and hydrogen methods to estimate the organics for A) all of the western sites and B) all of the eastern sites.

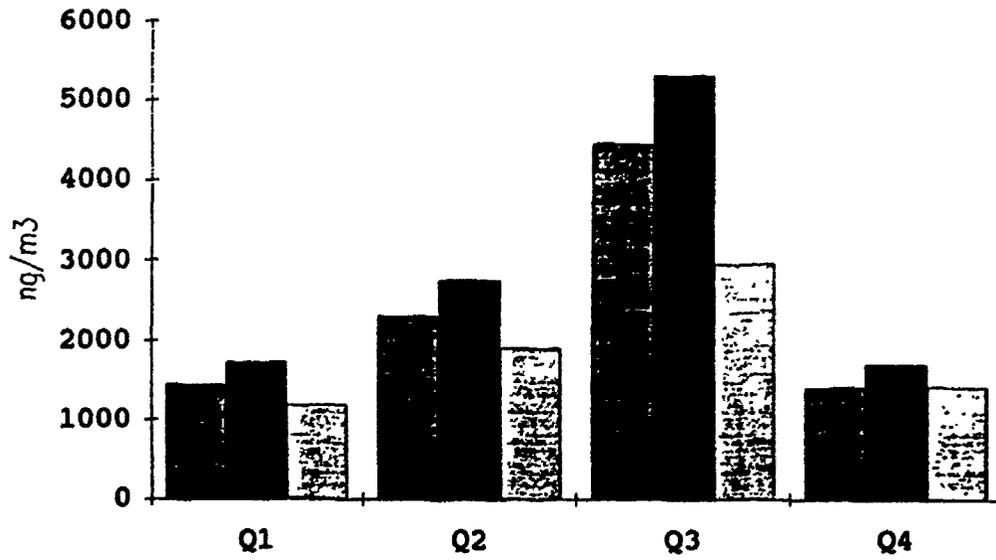
average to about that of the organics by hydrogen with only slightly changing the slope and correlation coefficient of the regression line.

A comparison of the two methods in the East is presented in Figure 21B. In this region the organics by the remaining mass is generally lower than that by the hydrogen method. Since the remaining mass constant used was a higher bound, the difference cannot be resolved by increasing this constant. At Qubbin Summit, MA and Whiteface Mnt. NY the data contains measurements of the organic carbon. In Figure 22, all three estimates of organic mass for these two stations are presented. At both sites and all four quarters the hydrogen method has a larger concentration than the other two methods which are comparable in concentration. This suggests that the hydrogen method overestimated the true organic concentration in this region.

The overestimations seems to be restricted to the NESCAUM data. As was determined previously, the organic mass at the NPS-NFPN sites in the Southeast and at Acadia, ME all accounted for a smaller fraction of the mass than the NESCAUM stations, see section 3.2.3 concerning the sensitivity analysis. In Figure 21B the data points from these stations have been circled. The organic matter at all of these points estimated by the remaining mass is either larger than or equal to that estimated by the organic hydrogen.

The overestimation at the NESCAUM sites may have resulted from making the sulfate too acidic, for as presented in the eastern sensitivity analysis in section 3.2.3, the organic concentration by hydrogen was highly dependent on the cation concentration. The more acidic the sulfate, the smaller the cation concentration, and the larger the estimate for the organic mass. It was found that by performing the fitting process on only the NESCAUM stations an equivalent fit was found by assuming pure ammonium sulfate and increasing the water constant. Poirot et al., (1991⁽⁵⁶⁾) found very good agreement between the three methods by assuming such complete neutralization of the sulfate.

Organic Mass Estimation at Whiteface Mt.



Organic Mass Estimation at Quabbin Summit

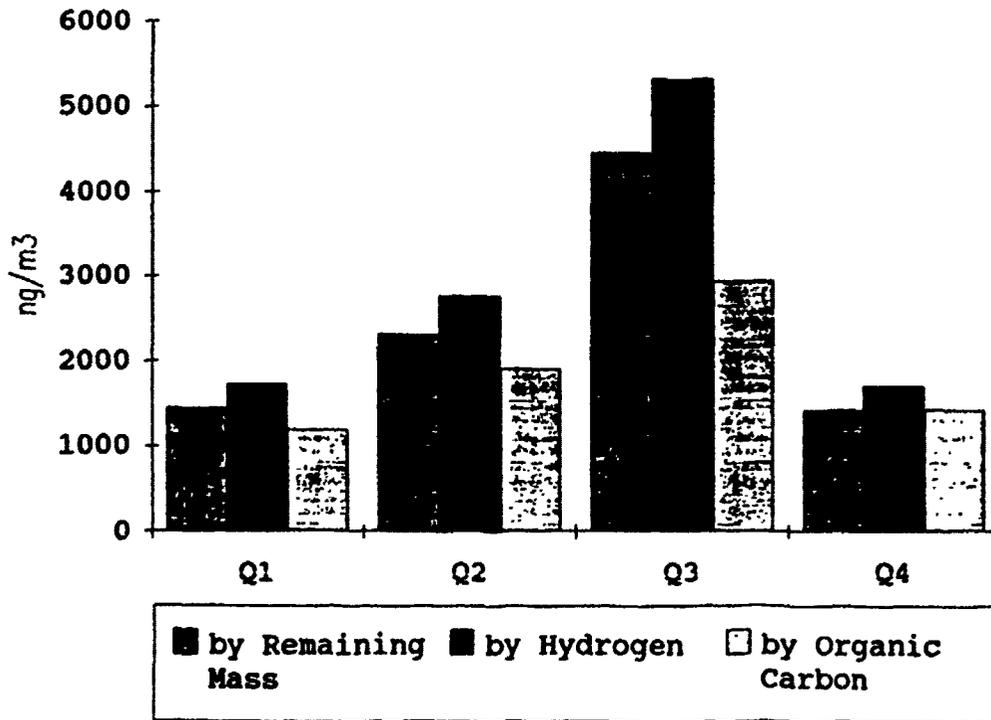


Figure 22. A comparison between the three methods available to estimate the organic concentrations at Whiteface Mt., NY and Quabbin Summit, MA.

By assuming pure ammonium sulfate for the eastern NPS-NFPN station we obtain very different results. As seen in Figure 14, using pure ammonium sulfate in the third quarter reduced the organic mass to less than 5%. This presents a definite incompatibility between the NESCAUM and NPS-NFPN data sets.

3.4 Comparison of Aerosol Equations and Constants to Other Studies

The final equations and constants used to find the aerosol types concentrations are summarized in Tables 2,3, and 4. Table 5 presents the equations and constants used by Cahill et al., (1989(25)) on the NPS-NFPN data, and Poirot et al., (1991(56)) on the NESCAUM data. Both of these studies were conducted on data averaged over the data's time span. These studies estimated all constant a priori, consequently, a fitting process was not used.

There are several differences between the equations and constants used for the NPS-NFPN data from this study and those in Table 5. First of all, Cahill et al., (1989(25)) did not calculate the water and salt concentrations. In their soil equation they did not separate the soil and smoke potassium, and they felt that the soil equation underestimated the true soil concentration by 16%. For the soot, they did not make a correction of 200 ng/m^3 as we did. Last, their organic constant is only 11 compared to our 13. This difference is due to their not accounting for the loss of volatile organics. The cation constants, which are similar to ours, were found by estimating the acidity of the sulfate aerosols.

A number of differences also exist between the equations and constants used in this study and those used by Poirot et al., (1991(56)) on the NESCAUM data. For the cation constant, they assumed a fully neutralized sulfate whereas in this study the sulfate was assumed to be acidic. Also, like Cahill et. al., (1989(25)), they increased the estimated soil concentration by 16%. The organic constant they used was 13.75 which is close the constant of 13 which we used. They determined this constant by comparing the low and high temperature organics by combustion of the organic carbon collected at two of the NESCAUM sites.

Table 4. Equations and assumptions used to estimate the fine aerosol types in this study.

Sulfate:	<p>Sulfate = $3 \cdot S$ -all sulfur is in the form of sulfate, SO_4^{-2}</p>
Cation:	<p>A) West Cation = $1.125 \cdot S$ -all sulfur is ammonium sulfate B) East Cation = $C_{amm} \cdot 1.125 \cdot S$ -the cation represents $(NH_4^+)_x H_y$</p>
Water:	<p>Water = $C_{water} \cdot (Sulfate + Cation)$ -Majority of water is associated with sulfate</p>
Soil:	<p>Soil = $1.89 \cdot Al + 2.14 \cdot Si + 1.4 \cdot Ca + 1.35 \cdot Fe + 1.2 \cdot SoilK$ -all elements are in their oxide forms -these elements account for the majority of the soil -SoilK is the non smoke potassium equal to $0.12 \cdot Si$ -Fe is equally split between FeO and Fe_2O_3</p>
Soot:	<p>A) NPS Stations: Soot = $(Abs - 200)$ -all absorption is caused by soot -the absorption efficiency is $10 \text{ m}^2/\text{g}$ -the Abs coefficient is systematically overestimated by $200 \text{ ng}/\text{m}^3$</p> <p>B) NESCAUM Stations: Soot = $Abs/2$ -the soot absorption efficiency is $20 \text{ m}^2/\text{g}$</p>
Sea Salt:	<p>A) NPS Stations: Sea Salt = $3.2 \cdot (NA - 15)$ -sodium constitutes 31% of sea salt -the averaged sodium was biased by $15 \text{ ng}/\text{m}^3$</p> <p>B) NESCAUM Stations: Sea Salt = $3.2 \cdot (NA)$ -all Na concentrations below the detection limit were set to 1/2 the MDL</p>
Organics:	<p>Organic = $C_{org} \cdot (H - Cation/Const)$ -all hydrogen is associated only with sulfate and organics</p>

Table 5. Equations and constants used on the NPS-NFPN and NESCAUM data sets in previous studies.

NPS-NFPN Aerosol Equations (Cahill et al., 1989⁽²⁵⁾)

Sulfate: Sulfate = $3 \cdot S$

Cation: A) West Cation = $1.125 \cdot S$
 B) East Cation = $C_{\text{cation}} \cdot 1.125 \cdot S$

Soil: Soil = $1.16 \cdot (1.89 \cdot Al + 2.14 \cdot Si + 1.4 \cdot Ca + 1.35 \cdot Fe + 1.2 \cdot K)$

Soot: Soot = Absorption coefficient

Organics: Organic = $C_{\text{org}} \cdot (H - \text{Cation}/\text{Const})$

NPS-NFPN Aerosol Constants

Cation Constant

Acadia	:	3.77
Shenandoah	:	3.77
Smokey Mnt.	:	3.77
Buffalo River	:	3.94

Organic Constant

All Stations	:	11
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NESCAUM Aerosol Equations (Poirot et al., 1991⁽⁵⁶⁾)

Sulfate: Sulfate = $3 \cdot S$

Cation: Cation = $1.125 \cdot S$

Soil: Soil = $1.16 \cdot (1.89 \cdot Al + 2.14 \cdot Si + 1.4 \cdot Ca + 1.35 \cdot Fe + 1.2 \cdot \text{Soil}K)$

Soot: Soot = Absorption coefficient/2

Organics: Organic = $C_{\text{org}} \cdot (H - \text{Cation}/\text{Const})$

NESCAUM Aerosol Constants

Organic Constant

All Stations:		11
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3.5 Application of the Aerosol Equations to the Coarse Mass

Three aerosol equations, soil, sulfate, and cation, were defined to partition the coarse mass. The soil and sulfate aerosol equations were fully defined. This left only the cation constant to be determined. Instead of performing another fitting process we chose to use the same constants found from fitting the fine aerosol mass.

Although no fitting process was necessary, there were two possible aerosol equations for the calculation of the coarse soil. In this section, we will look at these two equations to determine the best approach to calculating the coarse soil concentration. We will then compare the calculated coarse mass, the sum of the soil, sulfate, and cation, to the measured coarse mass.

3.5.1 Coarse Soil

The two methods available to calculate the coarse soil were the oxide method, which scaled up the common soil elements to their assumed oxide forms, and a method which scaled up one aerosol species by its assumed soil content ratio, see section 3.1.3. The oxide method was first examined, and the results can be seen in Figure 23, a comparison between the coarse soil and the coarse mass. Note that the slope of a line estimating the scatter changes at the tail end of the plot where the coarse mass concentration is small. The effect of the changing slope can be seen in Figure 23B where the normalized soil and coarse mass are compared. At the data points with low mass concentrations there is a sudden increase in the normalized coarse mass. This abnormality at low mass concentrations was not seen in the fine soil and is not reasonable.

On examining the raw coarse data, it was found that the aluminum concentrations were the cause of this deviation. Figure 24 presents a scatter plot comparing the quarterly averaged aluminum and the coarse mass. The aluminum has been scaled by its assumed soil content, and normalized by the coarse mass. As shown, the scatter plot is greatly skewed for those data points with low coarse mass. These are the same points which were skewed in Figure 23.

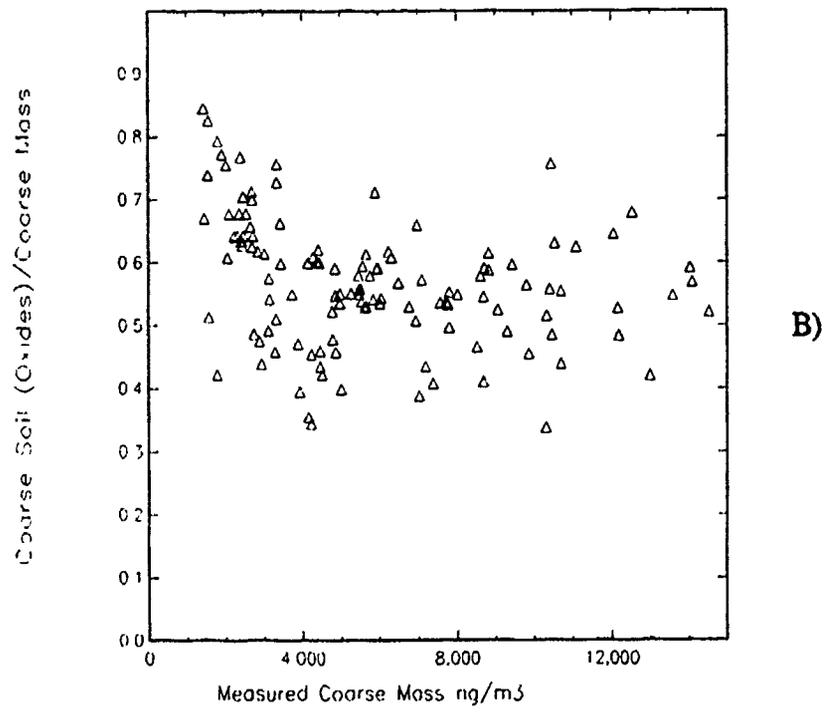
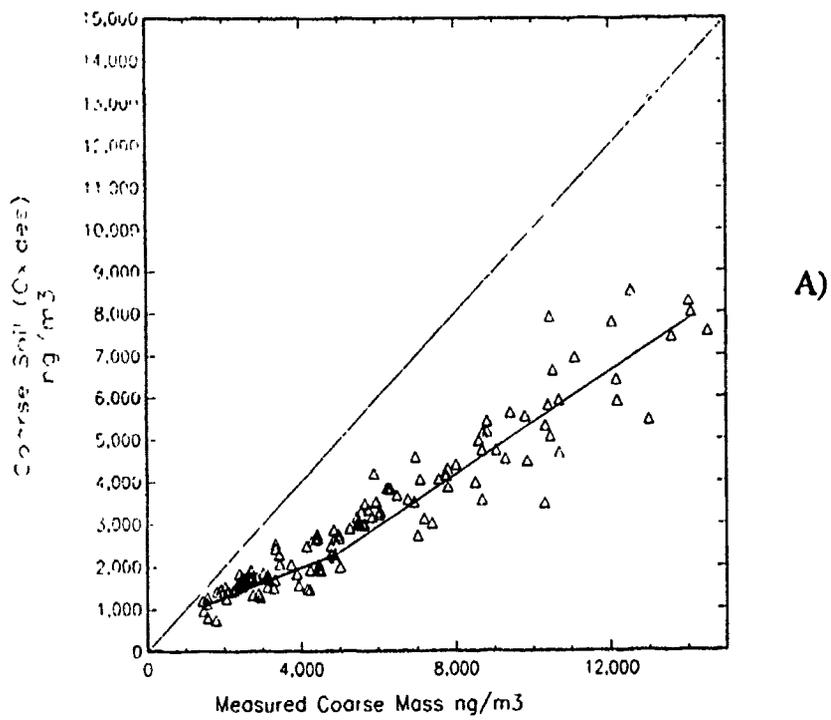


Figure 23. A) A comparison between the coarse soil and the measured coarse mass. The soil was estimated by the oxide method. B) The calculated coarse soil normalized by the coarse mass is compared to the coarse mass.

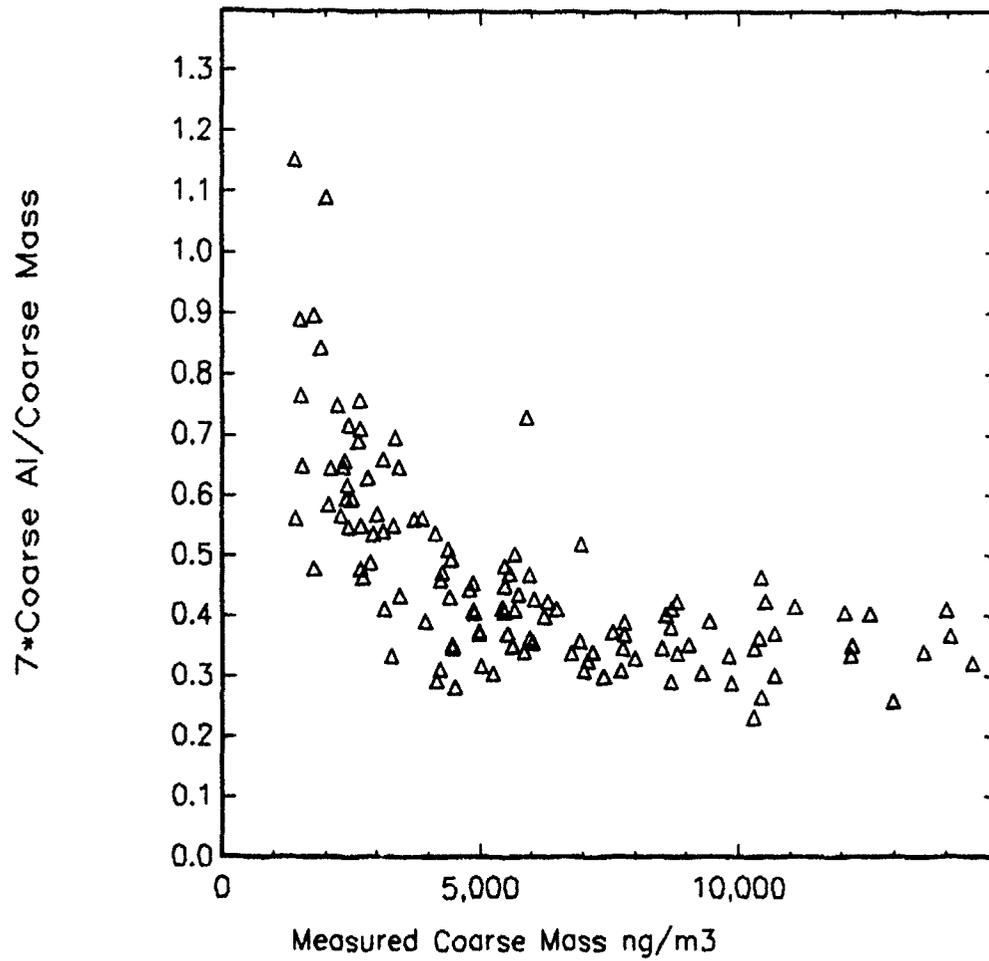


Figure 24. A comparison between the coarse aluminum and the coarse mass. The aluminum has been normalized by the coarse mass, and scaled to represent the coarse soil.

The cause of the skewed data could be due to an inadequate matrix correction during the PIXE analysis of the elemental data. As discussed in section 2.1, the lower the molecular weight, the larger the corrections that were necessary. Examination of the silicon data showed only a slight bias, and the iron data showed no bias at all. Both silicon and iron have greater molecular weights than aluminum with iron being more than twice as heavy as aluminum. Due to the bias of the aluminum data the oxide method could not be used to estimate the coarse soil.

To employ the alternative method we examined silicon and iron as the tracer species. As discussed in section 3.1.3, these species were scaled by assuming that the silicon constituted 28% of the soil and iron constituted 4%. In Figure 25, these two soil estimates are compared. These methods give different results with the iron estimating about 20% more for the coarse soil than the Si. Consequently, the choice in species does make a difference and adds some uncertainty to this method. After examining the coarse silicon it was found that there was a slight bias to the data like that seen for the coarse Al. This is one possible reason for the difference in soil concentrations estimated from using Si and Fe. For this reason, we chose to use the coarse Fe to calculate the coarse soil.

3.5.2 The Calculated Coarse Mass

The application of the three aerosol equations, soil, sulfate, and cation, to the coarse aerosol is presented in Figure 26. As can be seen in the scatter plot, there is excellent correlation between the calculated and measured mass, however, the slope is only 0.74. As seen from the residual plot, the unaccounted mass is approximately 30% on average. As previously discussed the majority of the coarse aerosol mass is soil. In light of this, having 30% of the coarse mass unaccounted for seems high. However, other researchers have found similar result (Macias et al., 1981⁽²⁹⁾; Lewis and Macias 1980⁽²⁷⁾).

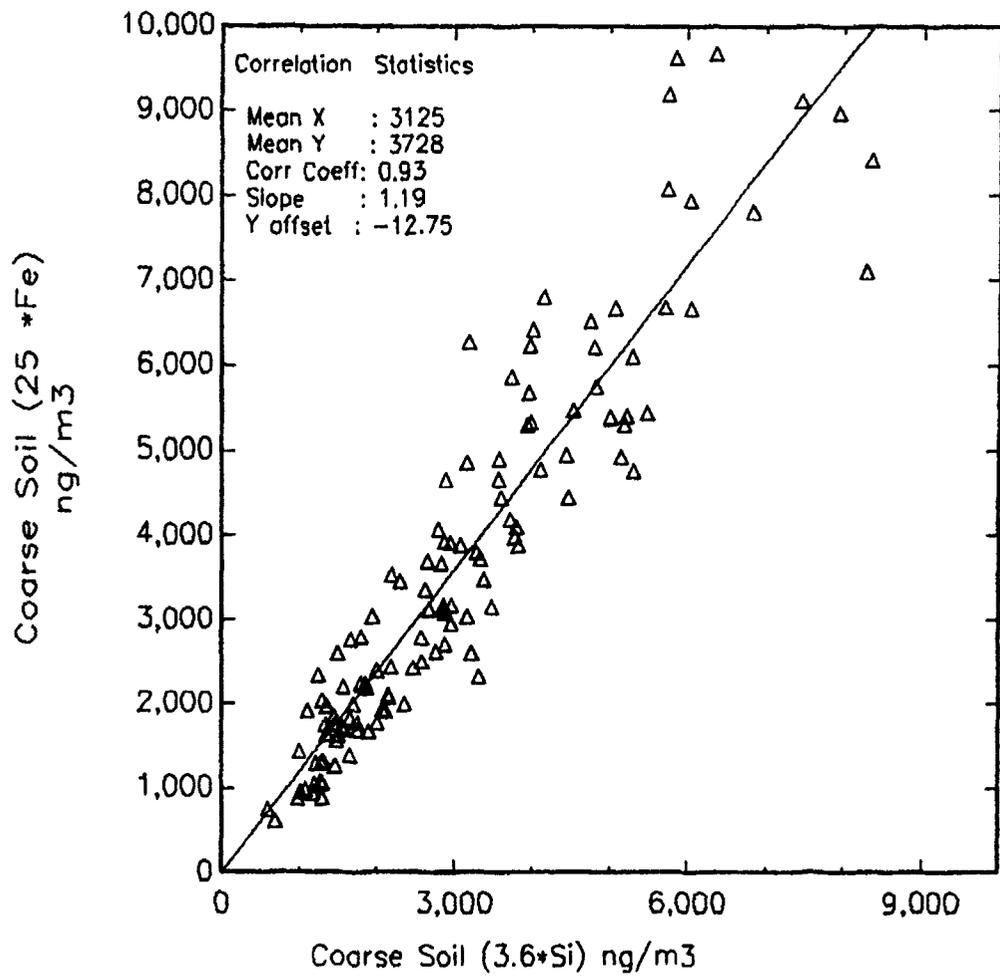


Figure 25. A comparison between estimating the soil by scaling up iron and by scaling up silicon.

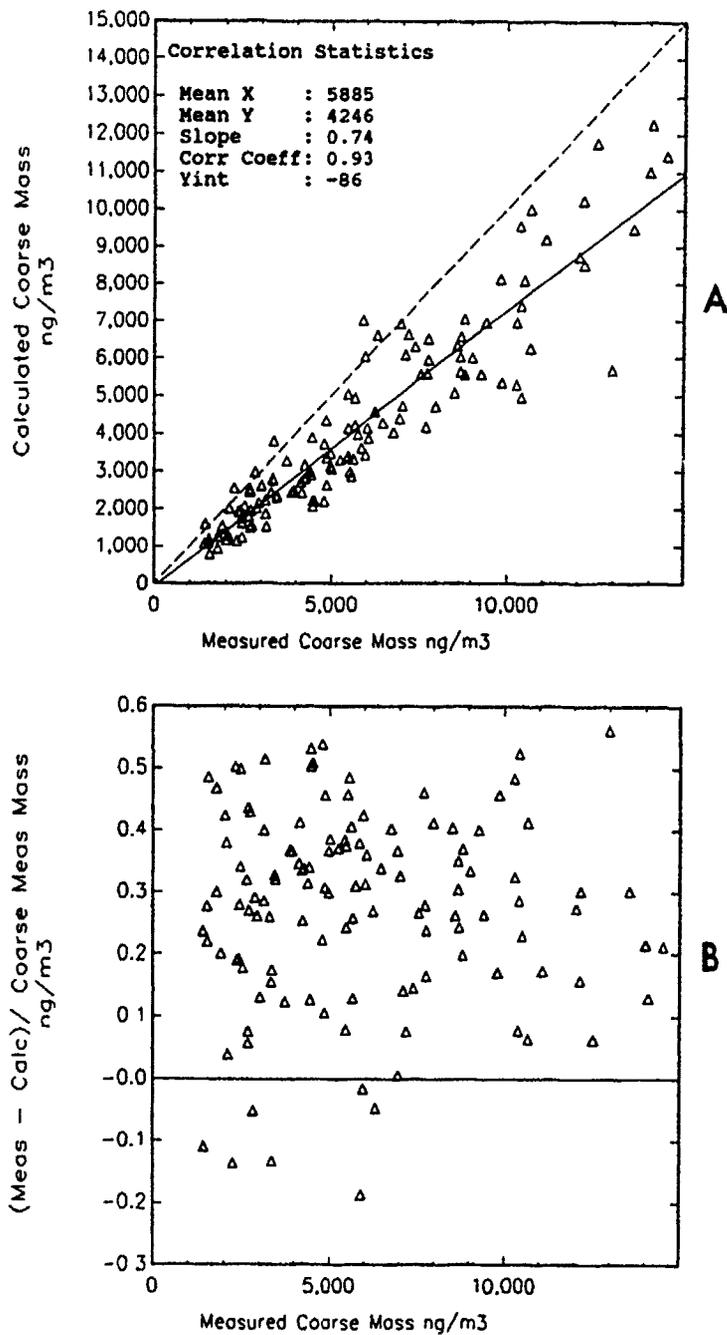


Figure 26. A) A comparison between the calculated coarse mass and the measured coarse mass.
 B) The residuals of the calculated coarse mass.

4. SPATIAL AND SEASONAL DISTRIBUTIONS OF AEROSOL TYPES

This section presents the results of the partitioning of the aerosol data into aerosol types. Both the fine mode of NPS-NFPN and NESCAUM data and the coarse mode of the NPS-NFPN data will be examined. The fine mode data were partitioned for both data sets by applying all seven aerosol type equations and fitted constants in Table 2, 3, & 4. For the coarse mode, only equations to estimate the soil, sulfate, and cation were applied to the NPS-NFPN data set.

4.1 The Aerosol Types for the Fine Mass

The results of the partitioning of the quarterly averaged fine mass are presented in Figures 27-34. These figures have been divided into spatial and regional categories. The spatial plots in Figures 27-30 depict the mass fractions of the different aerosol types for each station, while the regional plots in Figures 31-34 present the average mass fractions and concentrations for specified regions.

To create the regional plots, five areas were defined which represented the characteristics of each region, Figure 35. Each region in this figure has been numbered which will be referred to as 1-Northeast, 2-Southeast, 3-Northwest, 4-Southwest, and 5-Smelter region. Figures 31 & 32 presents the mass fractions of the average aerosol types for each area defined in Figure 35 as pie charts for each quarter. The concentrations of these average aerosol types are presented in Figures 33 & 34 as bar charts. The data from which these figures were created from is presented in Appendix A in Tables 7 & 8.

In this section, the magnitudes and trends of each aerosol type will be discussed using these plots. Also, the mass ratios of the sulfates and organics will be contrasted with those of previous studies, and we will reexamine the validity of the applied aerosol equations developed in section 3.

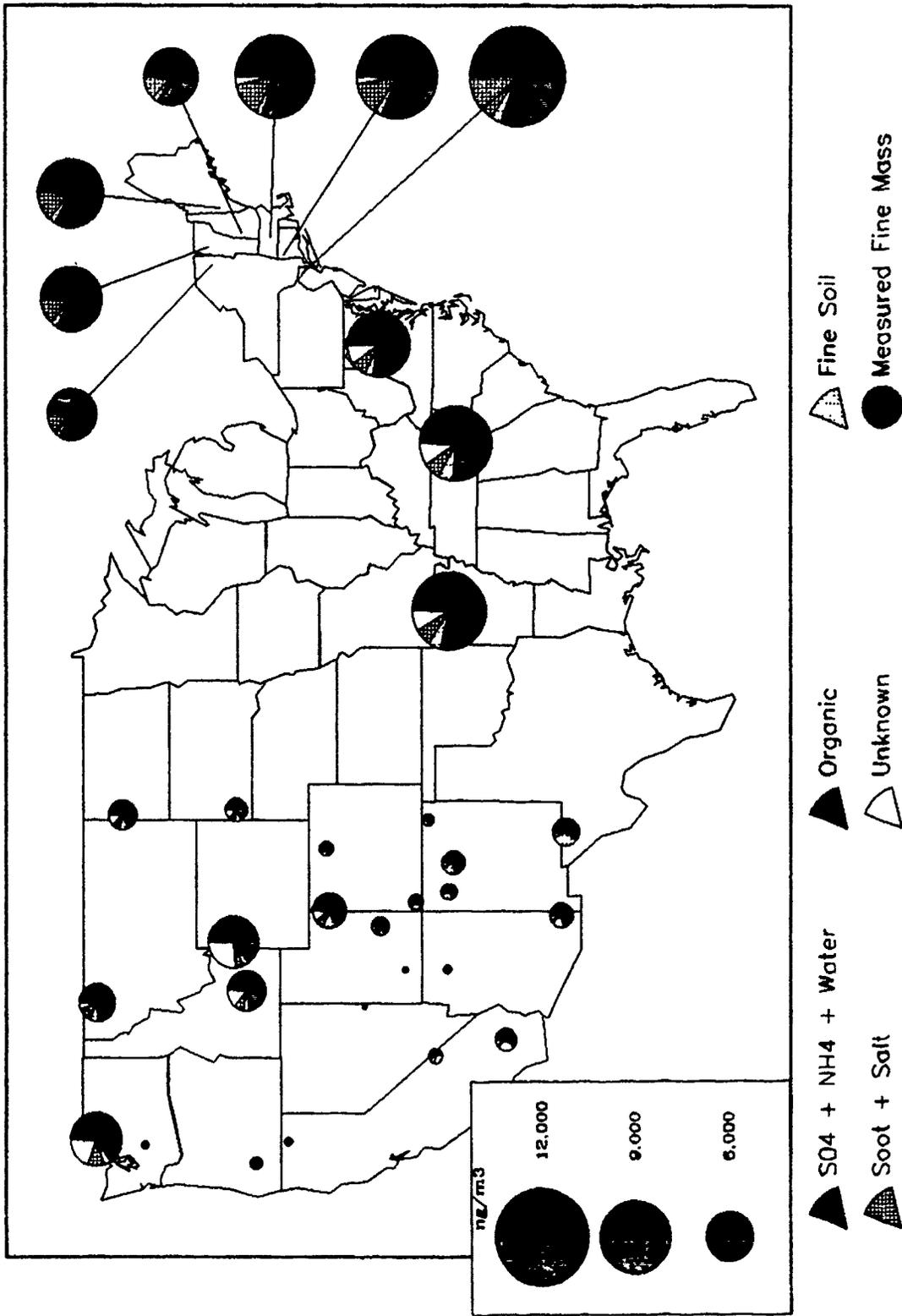


Figure 27. The mass fraction of each fine aerosol type at every location for quarter 1. The size of each pie chart is dependent on the fine mass concentration.

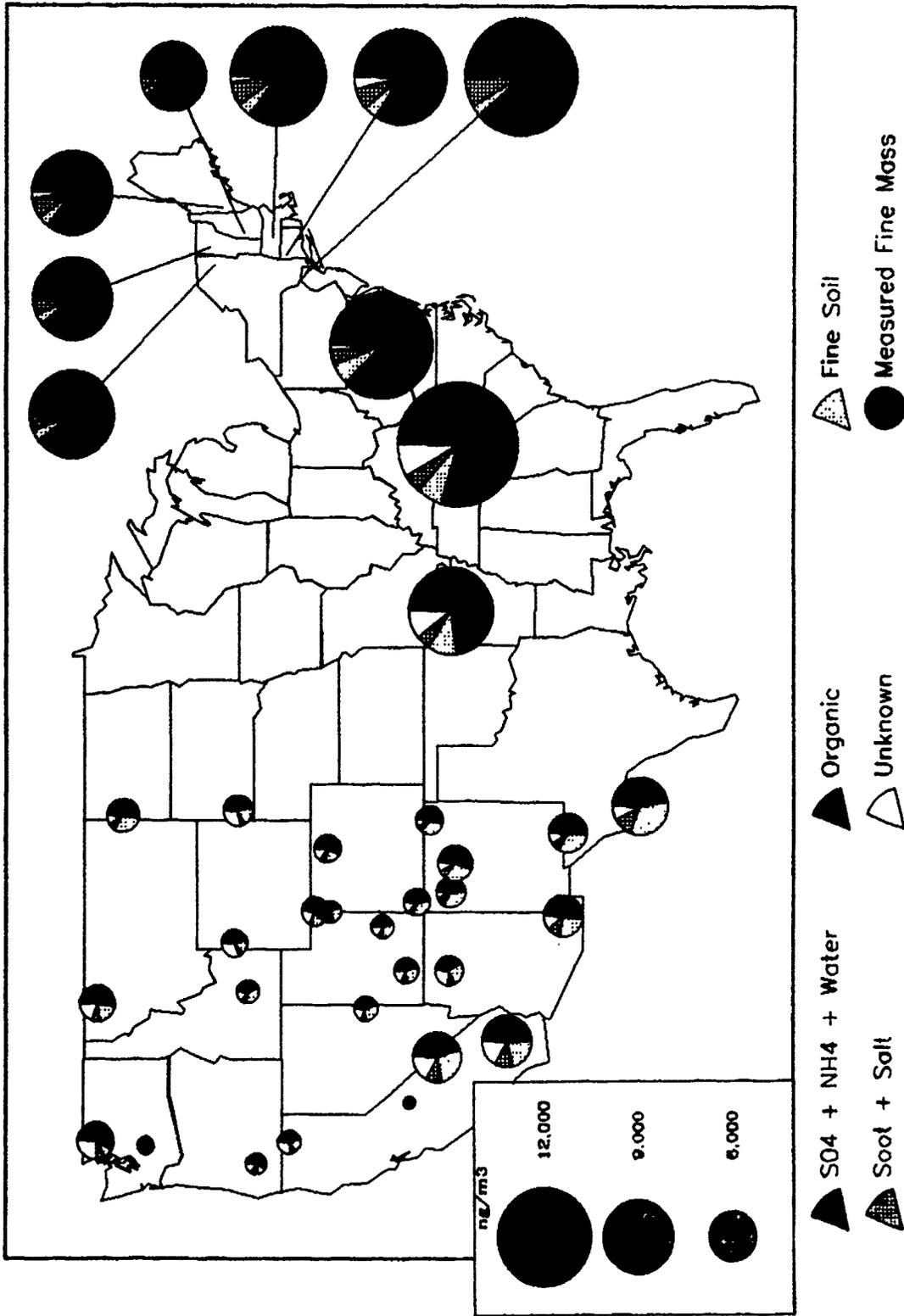


Figure 28. The mass fraction of each fine aerosol type at every location for quarter 2. The size of each pie chart is dependent on the fine mass concentration.

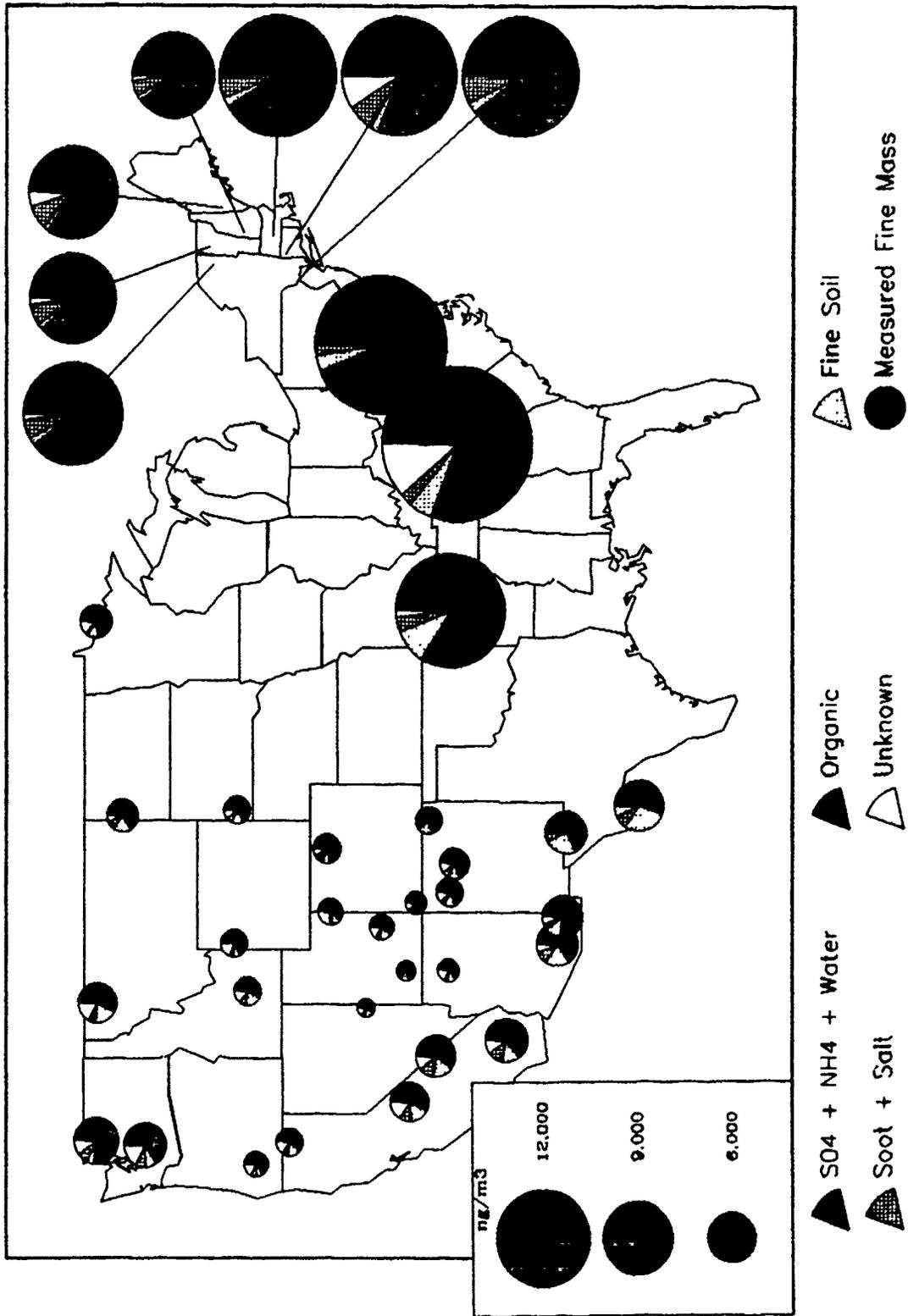


Figure 29. The mass fraction of each fine aerosol type at every location for quarter 3. The size of each pie chart is dependent on the fine mass concentration.

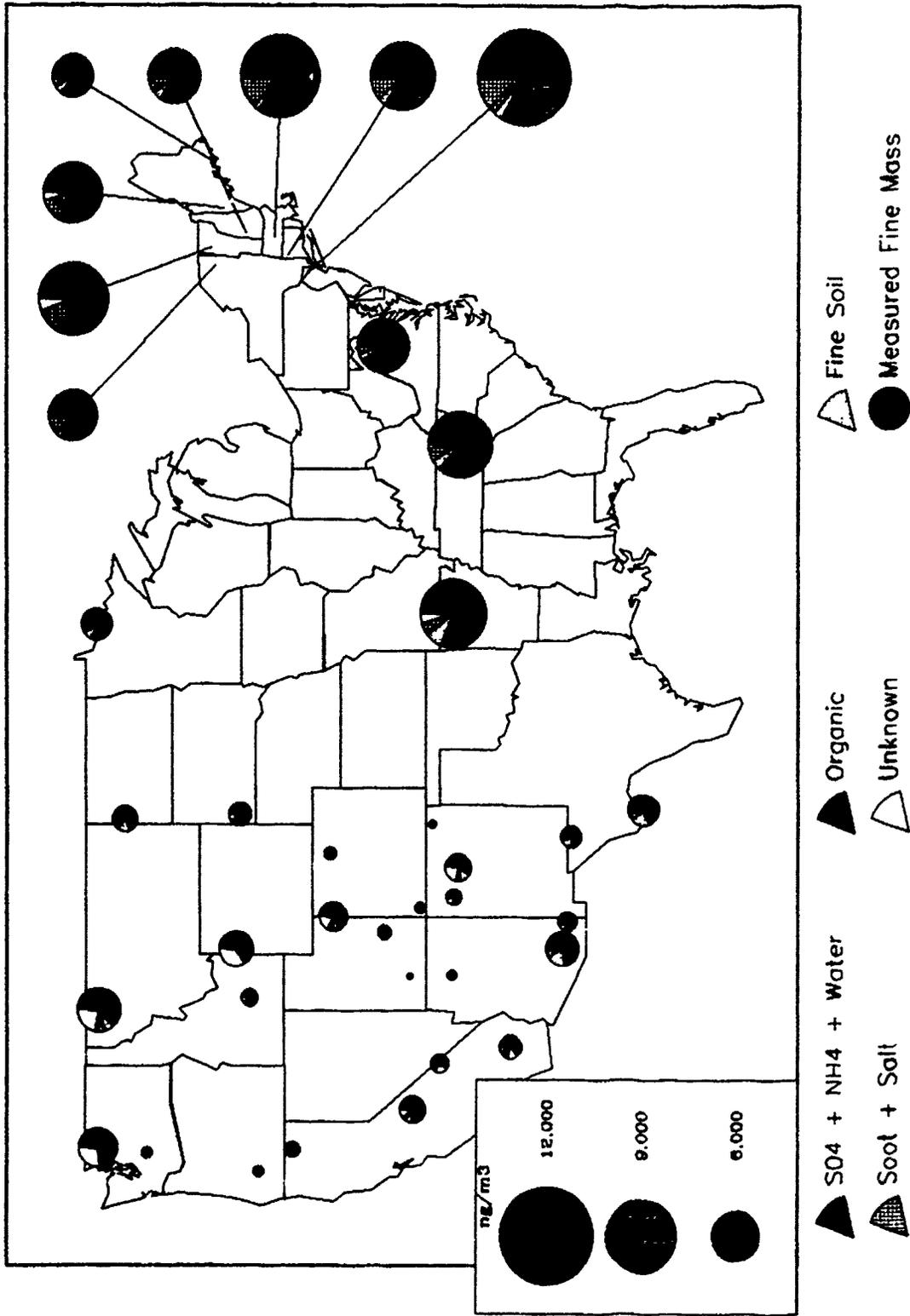


Figure 30. The mass fraction of each fine aerosol type at every location for quarter 4. The size of each pie chart is dependent on the fine mass concentration.

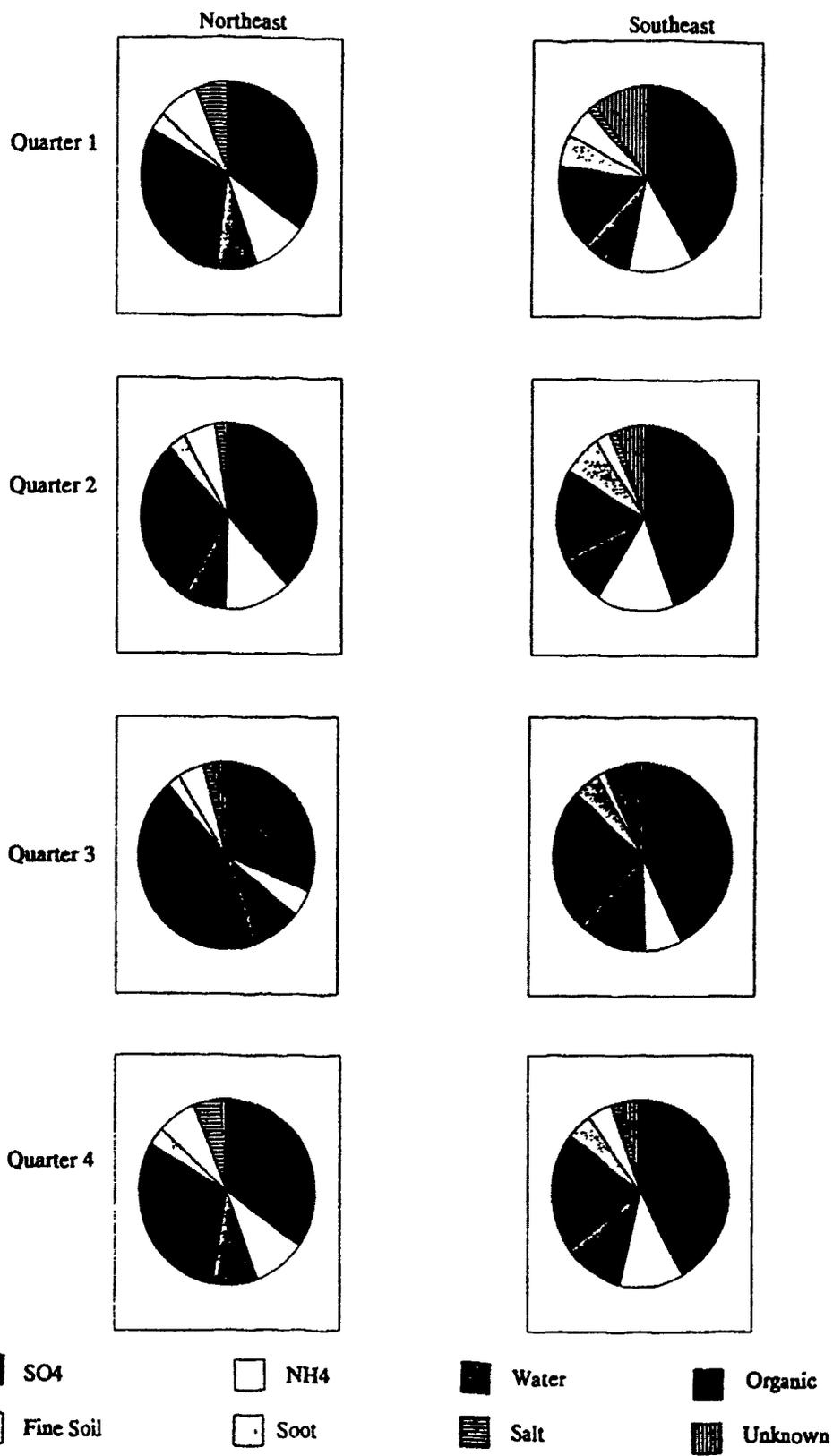


Figure 31. The average mass fraction of the seven aerosol types and the unknown for the Northeast and Southeast as defined in Figure 35.

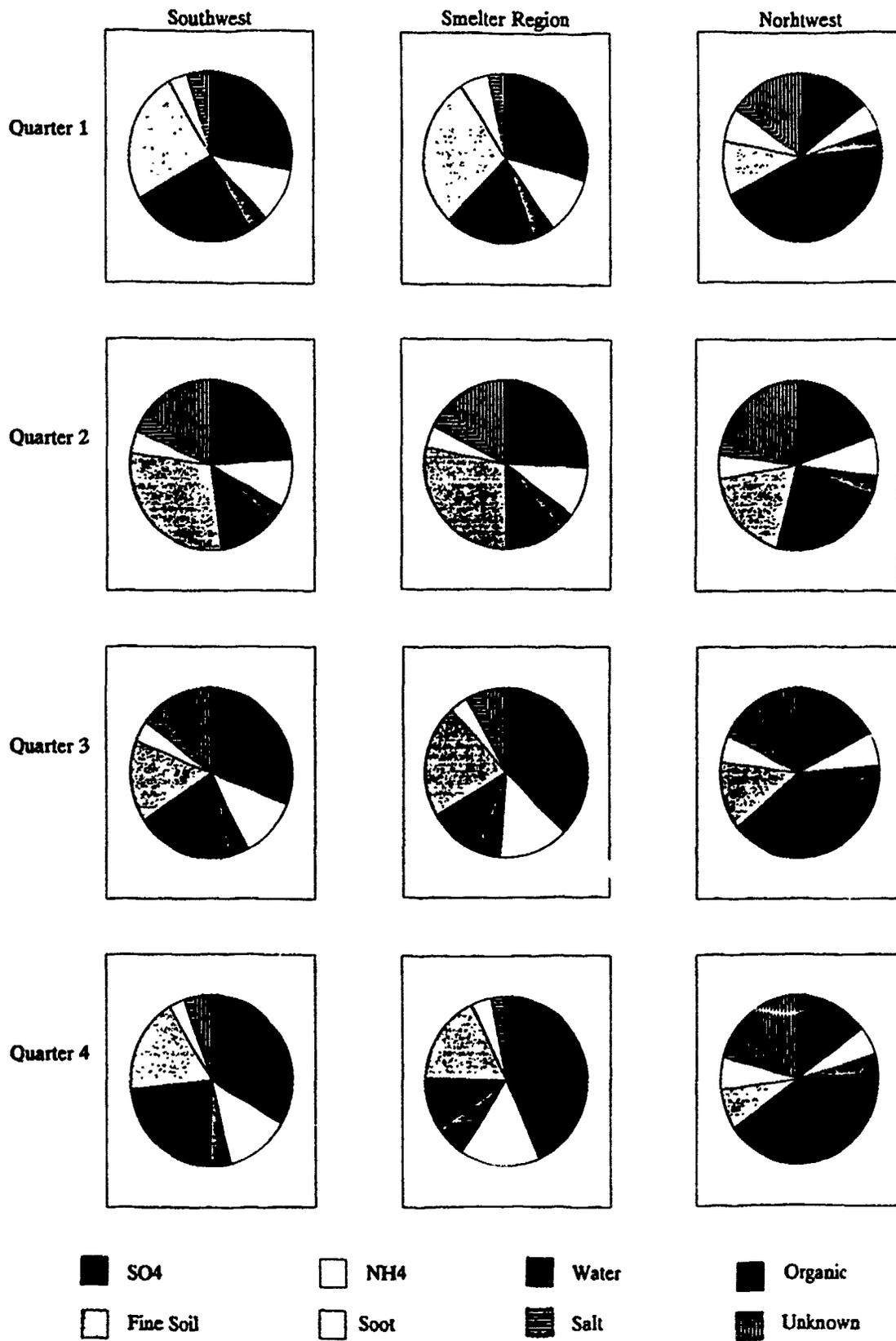


Figure 32. The average mass fraction of the seven aerosol types and the unknown for the Northwest, Southwest, and Smelter region as defined in Figure 35.

Best Fit Aerosol Composition

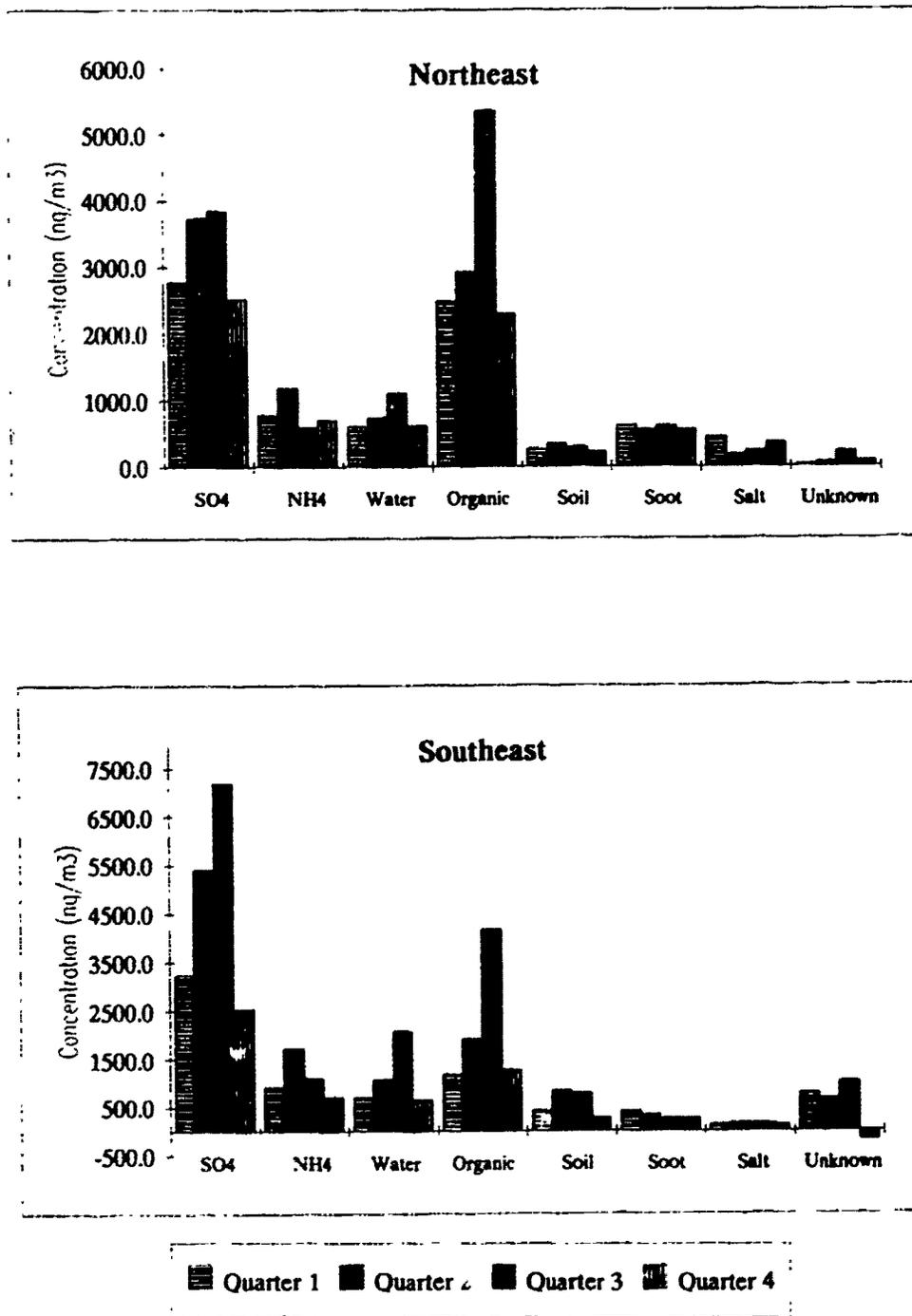


Figure 33. The concentrations of the seven aerosol types and the unknown for each quarter in the Northeast and Southeast as defined in Figure 35.

Best Fit Aerosol Composition

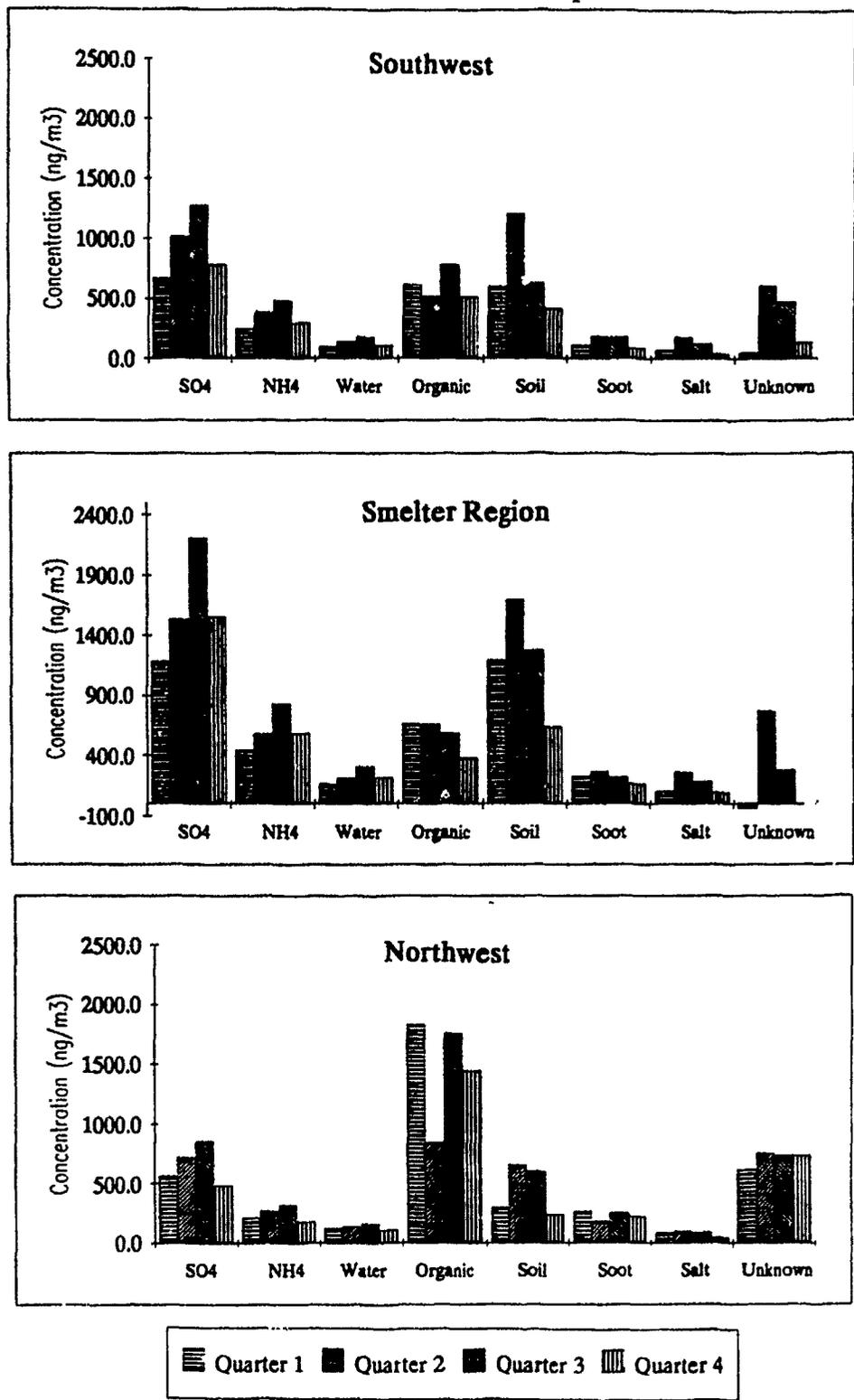


Figure 34. The concentrations of the seven aerosol types and the unknown for each quarter in the Northwest, Southwest, and Smelter region as defined in Figure 35.

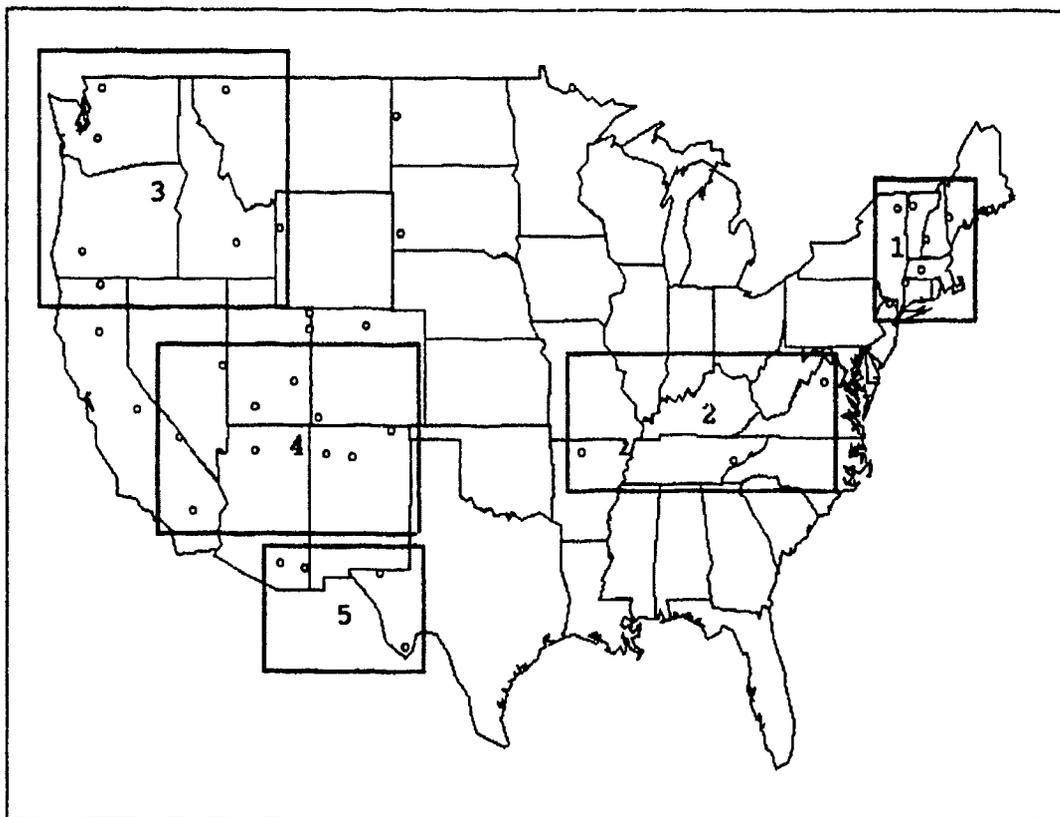


Figure 35. The stations that were averaged over to define the five regions of the U.S.. The five regions are: 1) Northeast, 2) Southeast, 3) Northwest, 4) Southwest, 5) Smelter Region.

4.1.1 Fine Mass

The concentration of the fine mass for each quarter and station is displayed in Figure 36. As shown, there is a substantial increase in the mass concentration from the cold season Q1 and Q4, to the warm season Q2 and Q3 in the East and southwestern U.S.. The mass in the Northwest is rather invariant, but there are a few exceptions. Mount Rainier WA, has a mass increase of 300% between quarters one and three. During the third quarter the Southwest and East experience the largest aerosol concentrations of the year.

The mass concentration in the East is larger than the West for all four quarters. The Southeast generally has larger concentrations during the warm season than the Northeast, but this reverses during the cold season. In the West, the stations located in the central states have smaller mass concentrations than the other stations for all quarters. During quarters 1 and 4 the mass in the West increases from the south to the north, but for the warm season the concentration is much more uniform.

4.1.2 Sulfate, Cation, and Water

Sulfate, cation, and water are three aerosol types which are all associated with particulate sulfur. As discussed in section 3.1.1, these concentrations were all calculated by scaling up the measured sulfur concentration. The fine mass fraction that these three aerosol types constitute are presented in Figures 27-30 where the fine mass is divided into aerosol types using pie charts for every station. The size of each pie is dependent on the fine mass concentration.

The mass fraction of the sulfur aerosol types varies both temporally and spatially. In the Northeast there is little variation between stations. As can be seen from the regional pie diagrams, Figures 31, the sulfur aerosol types, sulfate, cation, and water, account for approximately 50% of the mass for all four quarters. This fraction increases in the Southeast where they make up between 55 and 65% of the fine mass.

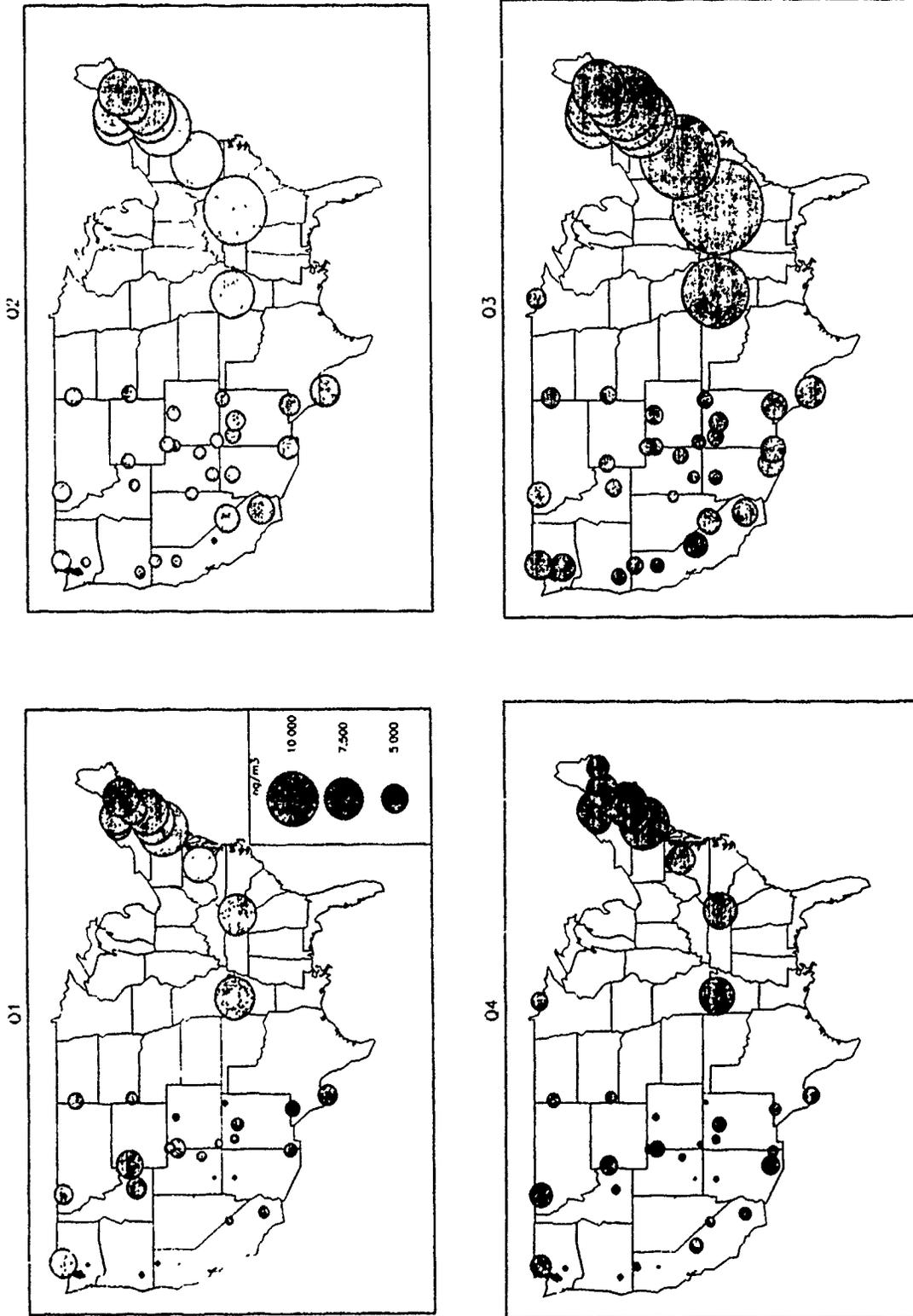


Figure 36. The fine mass concentration at each station and quarter.

In the West, sulfur aerosol types generally constitutes a smaller fraction of the fine mass. This fraction though, is quite variable over different region in the West. At the stations located more to the East and in the Smelter region, defined in Figure 35, these aerosol types are an important component to the mass accounting for about 50% during the warm season and somewhat less during the cold season. This fraction decreases for the stations farther to the north and west. During the cold seasons at the stations in the upper Northwest the sulfur aerosol types account for less than 25% of the mass.

Spatial plots of the sulfur aerosol types, sulfate, cation, and water, concentrations are displayed in Figures 37-39. As can be seen, sulfate is the major constituent accounting for about 60% of the mass of the sulfur aerosol types. The sulfate concentration closely follows the trends of the fine mass. Consequently, higher concentrations are seen in the warmer seasons than the colder seasons. Also, there is more sulfate in the East than the West, and the third quarter experiences the largest concentrations for the year. In the East, there is a decreasing sulfate concentration gradient from the south to the north. In the West, the concentrations decrease from the Southwest to the Northwest with the smallest concentrations occurring in northern California and Oregon during quarters 1 and 4. The largest western concentrations are seen in the Smelter region during the warm season.

Like the sulfate, the cation in the West was assumed to be a constant fraction of the sulfur. In the East where the sulfate acidity is variable, the cation trends vary considerably from the sulfate. The estimated cation concentrations were found from the fitting process discussed in section 3.2.2. As displayed in Figure 38, the cation concentrations are largest during the second quarter with the highest concentrations occurring at the two southern stations Great Smokey Mnts, TN and Shenandoah, VA where they are about 1700 ng/m^3 . Although the third quarter has the largest sulfate concentrations, it also is the most acidic quarter. Consequently, this quarter has the smallest cation concentration with less than 700 ng/m^3 for the Northeast. The third quarter cation concentrations is the only quarter where the Southwest has a larger concentration than the East. The cation concentrations at Buffalo River, AR differed from the rest of the East, because the aerosol acidity remained constant for all four quarters.

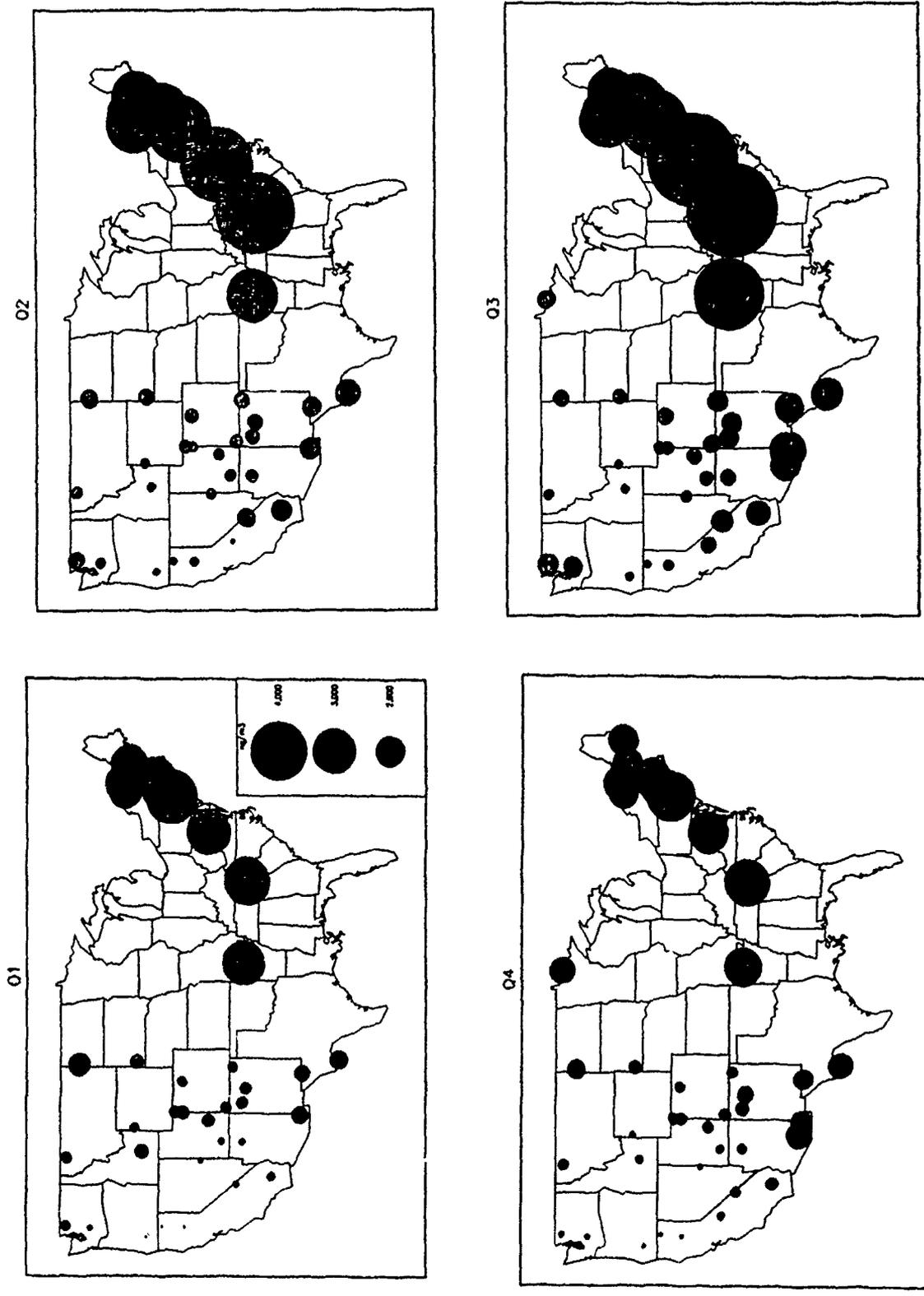


Figure 37. The calculated sulfate concentrations at each station and quarter.

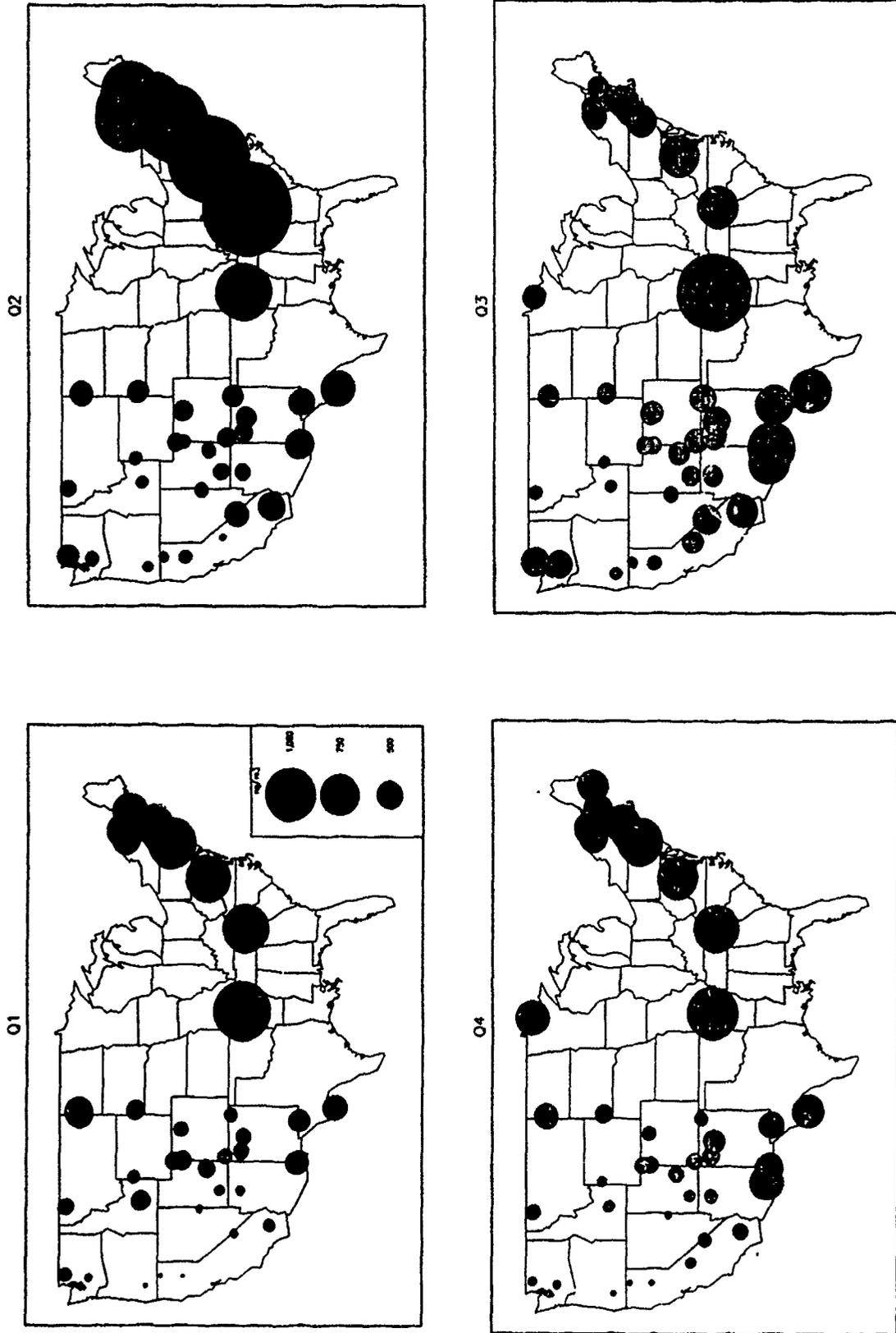


Figure 38. The calculated ammonia concentrations at each station and quarter.

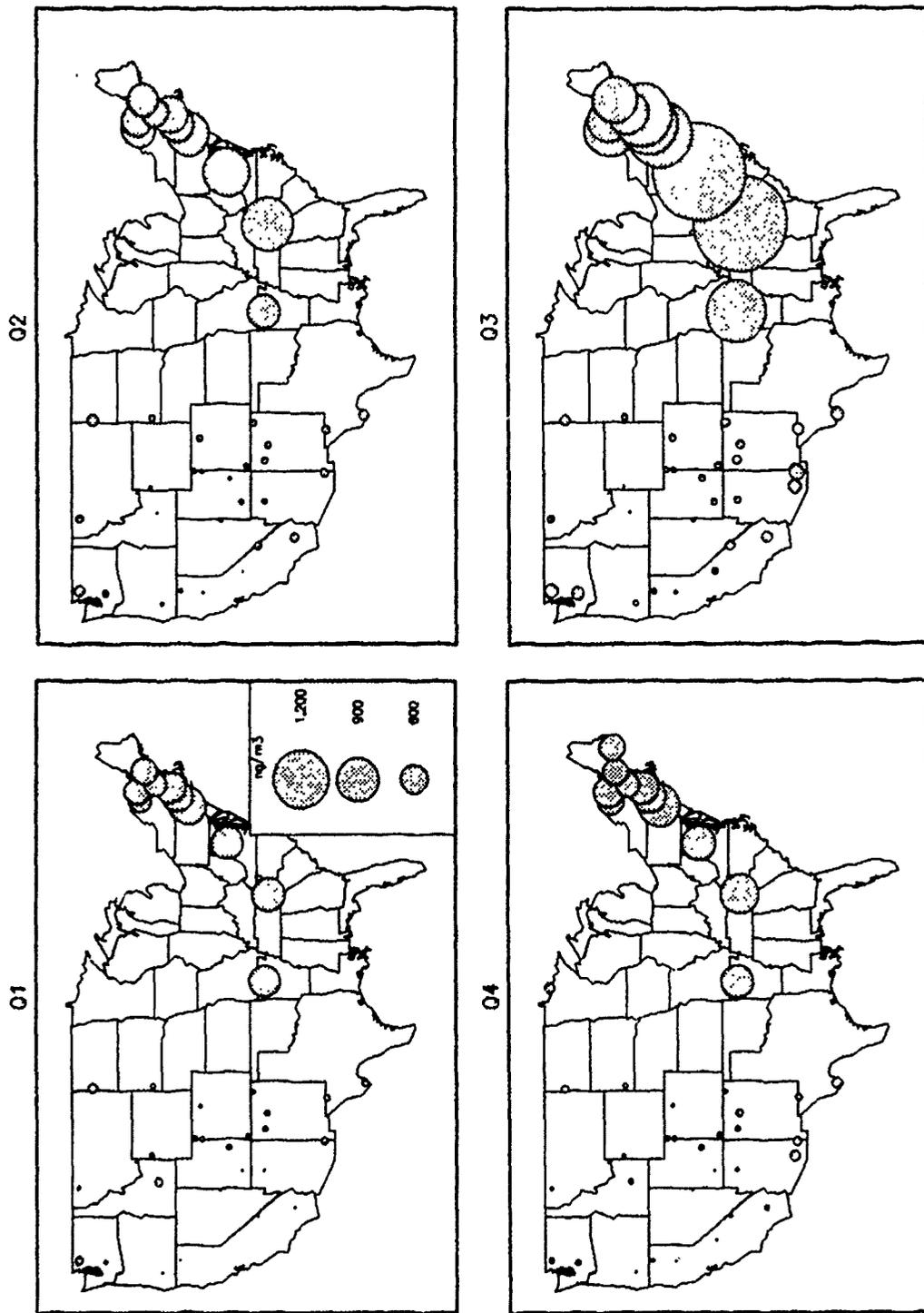


Figure 39. The calculated water concentrations at each station and quarter.

Since the samples acclimatized to the U.C. Davis laboratory climate before analysis, the magnitude of the water concentrations determined in this study offers little information to the water content of the *in situ* aerosol. However, the water content is partly dependent on the relative humidity during collection which enters our estimation through the hysteresis effect.

As shown in Figure 39, the estimated water content of the laboratory-equilibrated aerosol is largest in the East where the high sulfur concentrations exist. On the other hand, the water is relatively constant throughout the entire West with the highest concentrations occurring in the state of Washington and in the Smelter region. The water content is largest during the third quarter in all regions, and rather constant for the other three quarters.

4.1.2.1 Comparison of Sulfate Mass Fractions To Other Studies

It has been well established that the majority of particulate sulfur is in the form of SO_4^{2-} . Since sulfate can be measured accurately, the mass fraction of sulfur should be generally equivalent for different data sets if the sampling periods are similar. Table 6 presents the sulfate and organic mass fraction from various studies. All stations are located in a rural area, and the values presented reside in the fine mass fraction. Also, in the table are values from this study presented for those stations which are the same or in the same area as a station in the literature. The literature values in the table can be compared to Figures 27-32 and Table 7 in Appendix A.

As shown in Table 6, the two southeastern stations, Smokey Mnts and Shenandoah, have data analyzed in previous studies, and their sulfate values are about equivalent. This is not the case in the Northeast. The Lenox, MA is the only northeastern station from the literature we have. Its sulfate mass fraction is about 48% compared to about 35% for the NESCAUM sites. However, the NESCAUM data are comparable to that at Laurel Hill and Allegheny Mnt, PA reported by Japar et al., (1990(58)).

Table 6. Sulfate and organic mass fractions from various studies.

Reference	Location	Period	Sampling Duration	FM ug/m ³	Sulfate%FM.....	Organics
<u>EAST</u>						
57	Lenox, MA	Q3, 1984	24 h	20	48	9
This Study	Ringwood St. Park, NJ	Q3	24 h	14	35	44
58	Allegheny Mnt, PA	Q3, 1983	24 h	64	30	4
58	Laurel Hill, PA	Q3, 1983	24 h	59	33	5
43	Lewis, DE	Q3, 1983	24 h	17	43	34
43	Lewis, DE	Q1, 1984	24 h	15	35	22
44	Luray, VA	Q3, 1980	24 h	26	45	25
This Study	Shenandoah, VA	Q3	72 h	16	45	29
59	Great Smokey Mnts., TN	Q3, 1980	24 h	24	50	13
This Study	Great Smokey Mnts., TN	Q3,	72 h	16	46	24
<u>North West</u>						
60, 61 ^a	Little Butte, ID	1986-87	day	5	20	39
60, 61 ^a	Harlowton, MT	1986-87	day	4	20	41
60, 61 ^a	Pathfinder, WY	1986-87	day	5	16	33
62 ^b	Portland, OR	1977-78	day	17	12	45
This Study	Crater Lake, OR	Year	72 h	3	14	43
<u>South West</u>						
29 ^c	Zilnez Mesa, AZ	6/26-7/13/79	24 h	6	31	29
21 ^d	Spirit Mountain, NV	1985-86	24 h	4	29	20
7 ^c	Spirit Mountain, NV	1986-87	24 h	4	29	21
7 ^c	Meadview, AZ	1986-87	24 h	4	33	22
7 ^c	Prescott, AZ	1986-87	24 h	3	41	23
7 ^c	Grand Canyon, AZ	1986-87	24 h	3	33	20
This Study	Grand Canyon, AZ	Year	72 h	3	29	18
63	Grand Canyon, AZ	Winter '86-87	24 h	3	34	14
This Study	Grand Canyon, AZ	Q1, Q4	72 h	2	30	24
7 ^c	Page, AZ	1986-87	24 h	4	31	24
63	Page, AZ	Winter '86-87	24 h	3	28	40
7 ^c	Bryce Canyon, UT	1986-87	24 h	3	31	19
This Study	Bryce Canyon, UT	Year	72 h	2	31	23
60, 61 ^a	Encino, NM	1981-82	day	4	29	30
60, 61 ^a	Walsenburg, CO	1981-82	day	5	20	28
60, 61 ^a	Ouray, UT	1981-82	day	6	12	31
60, 61 ^a	Delta, UT	1981-86	day	5	21	30
60, 61 ^a	Fish Creek Ranch	1981-86	day	6	11	20

Table 6, Continued;

Reference	Location	Period	Sampling Duration	FM ug/m ³	Sulfate%FM.....	Organics
Smelter Region						
60, 61 ^a	Sierra Vista, AZ	1981-82		day	8	31 19
This Study	Chiricahua, AZ	year	72 h	4	40	11

Notes:

- a. The average values were obtained from Table 4.2-1 in the NAPAP (1991⁽⁷⁾) report.
- b. Organic carbon was collected on glass fiber filters. The averages were obtained from the two rural sites in the study.
- c. The averages at Zilnez Mesa, AZ excluded the data from July 5 which was dominated by smoke from near by wildfires
- d. The organic values were obtained from Table 4.2-1 in the NAPAP (1991⁽⁷⁾) report.
- e. The data at this station was collected by the SCENES network. The average values were obtained from Table 4.2-1 in the NAPAP (1991⁽⁷⁾) report.

In the Northwest there is excellent agreement between this study and the literature. At Portland OR, the sulfate fraction is 12%. At two NPS-NFPN stations located close to this site, Crater Lake, OR and North Cascades, WA the yearly averaged mass fraction is about 15%. Those stations in the vicinity of Little Butte, ID and Harlowton, MT all have yearly average values around 20%.

The Southwest also agrees very well with the literature. The identical stations in the literature, Table 6, and this study have very good agreement. Additionally, the overall average of the sulfate mass ratios of all the southwestern literature ratios is 28%, and the average for all of the southwestern data in this study is also 28%. Although the Sierra Vista is located in the Smelter region, its sulfur mass fraction is similar to the stations in the Southwest. Its sulfate mass fraction is about 10% less than the stations in this region from this study.

4.1.3 Organics

The organic aerosol is a very important component of the fine particle mass. The method used to estimate the organic aerosol was described in section 3.1.4. As displayed in Figures 27-30, the organics in the Northeast account for a little more than 30% of the fine mass for quarters 1,2 and 4. During quarter 3 this fraction increases to over 40% of the fine mass. Note that during the fourth quarter, organics at Acadia, ME account for about 24% of the fine mass compared to 32% for the surrounding stations.

The seasonal variation of the organics to fine mass ratio for the three Southeastern stations is the same as that in the Northeast, except the organic fraction is about 10% less for all four quarters. The differences between Acadia, ME and the southeastern station from the NESCAUM stations will be addressed in the following section.

In the West, the mass fraction of the organics varies widely with space and time. It becomes a larger part of the fine mass at those stations more to the north and west. At the Grand Canyon, during the second quarter the organic mass accounts for less than 5% of the fine mass, but during the fourth quarter at North Cascades, WA it accounts for almost 50% of the fine mass. The temporal pattern for the West follows closely that of the East, except for the upper Northwest corner of the country. Here the organics

account for a large fraction of the mass particularly during the winter season where more than 45% of the fine mass is due to organics. Although Yosemite is located in the southwestern part of the country, its organic patterns are more like those in the upper Northwest.

The spatial distribution of the organic concentrations are displayed in Figure 40. The Northeast has much higher concentrations than the rest of the country for all four quarters. In the East, the largest concentrations occur during the third quarter where the Southeast has organic concentrations on the order of 4000 ng/m³, and Northeast has organic masses between 4500 - 6500 ng/m³.

In the West, two spatial concentration gradients are apparent. The organics increase from east to west, and from the south to the north. The Northwest contains the largest western concentrations during quarter 1 where the concentrations are generally greater than 2000 ng/m³ and even reach 3000 ng/m³ at North Cascades, WA. The stations located in the Southwest, east of CA, show the smallest concentrations in the country, generally less than 700 ng/m³ for all four quarters. The concentrations here are rather invariant with time, but the third quarter does experiences a slightly higher concentration.

4.1.3.1 Comparison of Organic Mass Fractions To Other Studies

Organic mass fractions determined in other studies are presented in Table 6. These values were all calculated from organic carbon collected on quartz filters except at the Portland Oregon site. The WRAQS stations have been corrected for organic artifact however, the stations from the other studies have not. The artifact concentrations on the uncorrected filters should be a small percentage of the total organic mass, because the artifact concentrations should be a function of the filter and sampling time. The longer the sampling period, the smaller the percentage of artifact to total organics (White, 1991⁽⁶⁴⁾). All of the studies taken from the literature in Table 6, except the WRAQS study, sampled over a 24 hour period which should be long enough to make the artifact negligible.

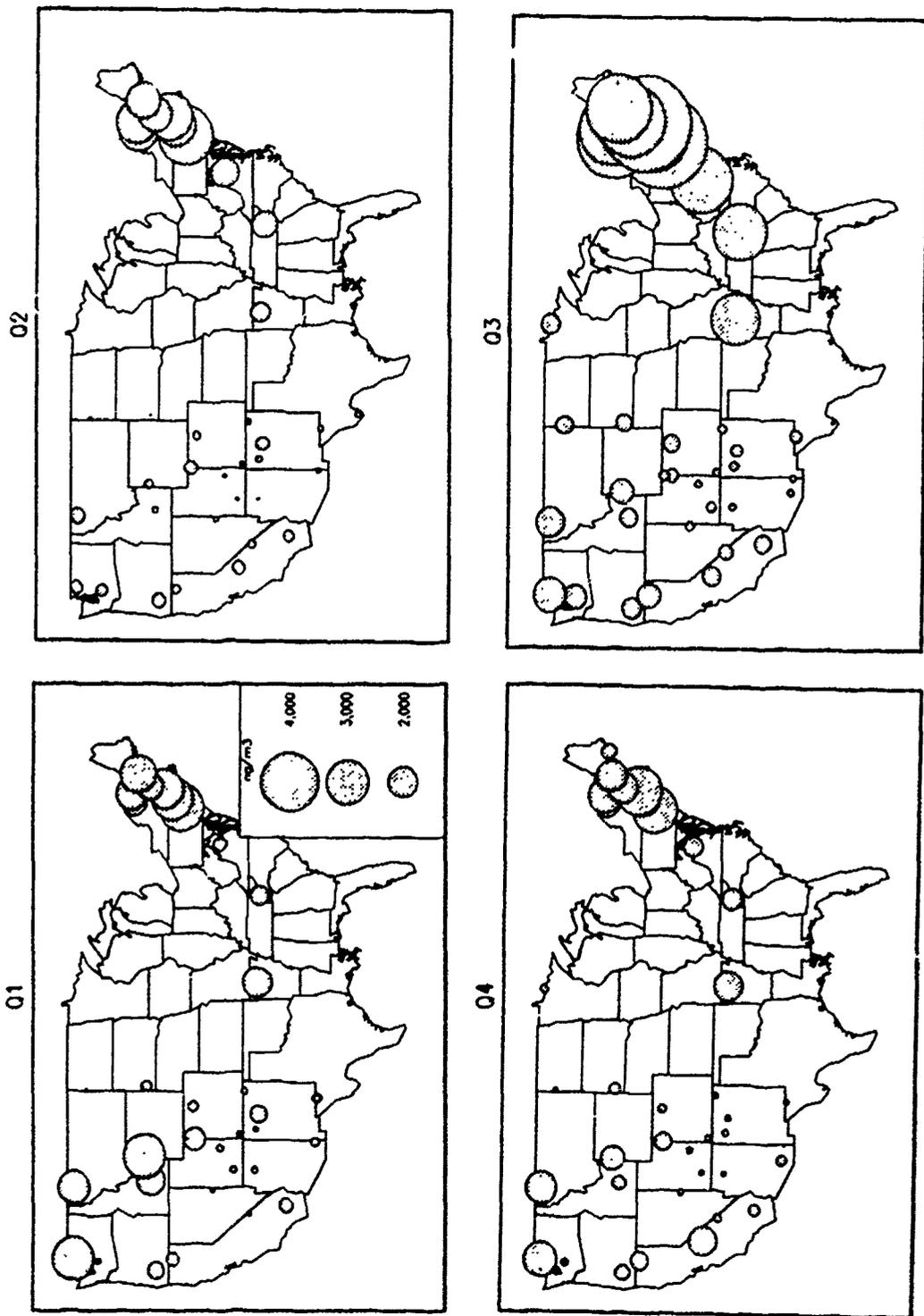


Figure 40. The calculated organic concentrations at each station and quarter.

The organic fraction we found for Shenandoah is very close to that at Luray, VA, located in the Shenandoah valley and Lewis, DE, located just northeast of the Shenandoah valley. In this study, the organic fraction at Smokey Mnts is comparable to the Shenandoah site as would be expected, however, in the literature the organic mass fraction at Smokey Mnts was only half of the value we calculated.

The NESCAUM sites in the Northeast have much larger organic mass fractions than any of the values found in the literature. The two PA sites, Allegheny and Laurel Hill, which have comparable sulfate fractions to the NESCAUM sites have organics an order of magnitude smaller, 4% compared to 45% for the NESCAUM sites. This supports the conclusion determined in section 3.3.2, that the organics for these stations were overestimated by the hydrogen method. However, estimating the organics by the hydrogen method, assuming a fully neutralized sulfate cation, as previous discussed reduces the organic mass fraction to about 25% of the fine mass. This is comparable to the literature for the more southern stations, but the northern stations in the literature are still much less.

Unfortunately, we have found only one study in the East that collected data outside of August. As shown, the organic mass fraction at Lewis, DE during January and February was about 22%. During the first quarter the two southeastern stations had organic fractions around 15% and the corrected organics at the NESCAUM sites averaged about 23%.

As seen by comparing the values in Table 6 and Table 7, the yearly average organic mass fraction for the stations in the West from this study are all about equal to those in the literature. The average organic fraction of all the station in the Northwest from the literature is about 39%. The stations in this study designated in the Northwest region from Figure 35 have a yearly average of 37%. We do not have any values from the literature averaged over less than a year for the Northwest. Consequently, we cannot compare the seasonal variation of our data with that in the literature.

In the Southwest, the yearly average organic mass fraction for this study is about 20% where as from the literature values in Table 6, the same average is approximately 24%. These two averages are rather close, however, it appears that our estimation is low. This low estimation may be due to underestimations in all four quarters, or it may

be due to an underestimation in one or two quarters. Unfortunately, we do not have enough quarterly average organic mass fractions from the literature to examine this question fully. However, two stations in Table 6 located in the Southwest, Grand Canyon, AZ and Page, AZ collected data only during the winter. These two sites have very different organic mass fractions, 14 and 40% respectively, and give a range that the organic fraction probably varies between in this region. As shown in Figures 27-30, most of our data for quarters 1 and 4 in the Southwest fall within this range.

Overall, the NPS-NFPN sites in the East and West are consistent with that found in the literature and the alternative estimation methods discussed in section 3.3.2. This helps to justify the process by which the organics at these stations were determined, and the degree of neutralization of the sulfate anion for the eastern sites. The organic mass fraction at the NESCAUM sites, however, are much larger than the values found in the literature and those found using alternative methods previously discussed.

It was found that much more reasonable results were obtained for the NESCAUM sites by assuming full neutralization of the sulfate cation and increasing the water constants. This makes water a substantial part of the fine mass accounting for almost 25% in the third quarter. Although this fraction is much larger than for the NPS-NFPN stations, it is not unreasonable, for in the study conducted at the PA sites in Table 6, the authors estimated the water to account for 38% at Allegheny Mnt and 44% at Laurel Hill after the sample had acclimated to the laboratory climate.

These results suggest that the two data sets, NESCAUM and NPS-NFPN are not fully compatible. However, the equations and fitting process used to determine the final concentrations appears to produce reasonable results for both data sets if the fitting process is performed on each data set separately.

4.1.4 Fine Soil

The fine soil aerosol is relatively unimportant in the eastern U.S. accounting for a few percent of the fine mass in all four quarters, Figures 27-30. However, in the West, soil is a very important constituent, and makes up over 30% of the fine mass at several stations. For the western stations, there is generally a north to Southwest increasing gradient for the fine soil mass fraction. The largest soil mass fraction occurs in the

Smelter region where soil constitutes more than 30% of the fine mass in quarters 1 and 2. The smallest fraction occurs in the Northwest corner of the country where less than 15% of the fine mass is attributable to soil for quarter 4. At most stations the fine soil mass fraction is largest during the second quarter.

Spatial plots of the fine soil concentration are presented in Figure 41 for all four quarters. One of the more striking features of these plots is the temporal variation. The second quarter contains much larger concentrations than the other three quarters, while the fourth quarter has the smallest concentration. In the Southwest, the second quarter has a concentration greater than 1000 ng/m^3 while it is less than 500 ng/m^3 during the fourth quarter. There is also a distinct temporal pattern between the Northwest and Southwest. In the southwestern region, quarters one and three have comparable average concentrations, on the order of 750 ng/m^3 . In the Northwest, quarter three has a much larger concentration than quarter one, 300 compared to 600 ng/m^3 , but quarter four has comparable concentrations to quarter one.

The smallest fine soil concentrations exist during the winter seasons in the Northeast where they are generally less than 300 ng/m^3 . However, these concentrations increase for the eastern stations more to the south and west. In the West, the soil concentrations increase from north to the south. The largest concentrations are in the Smelter region most notably at Big Bend Texas. The two sites, Glacier National Park and Wind Cave, SD located in the Northwest both have soil concentrations comparable to those in the Southwest.

4.1.5 Soot

As shown in Figures 31 & 32, the mass fraction of soot is variable across the country. The largest mass fraction occurs in the Northwest and Northeast during quarters 1 and 4 accounting for approximately 7% of the fine mass, while the smallest mass fraction occurs during quarter 3 of the Southeast accounting for about 2%. There is a definite warm and cold season variation in the upper Northwest and East with the largest mass fractions occurring during the cold season. The soot mass fraction in the Southwest is very consistent accounting for between 4 and 5% of the fine mass.

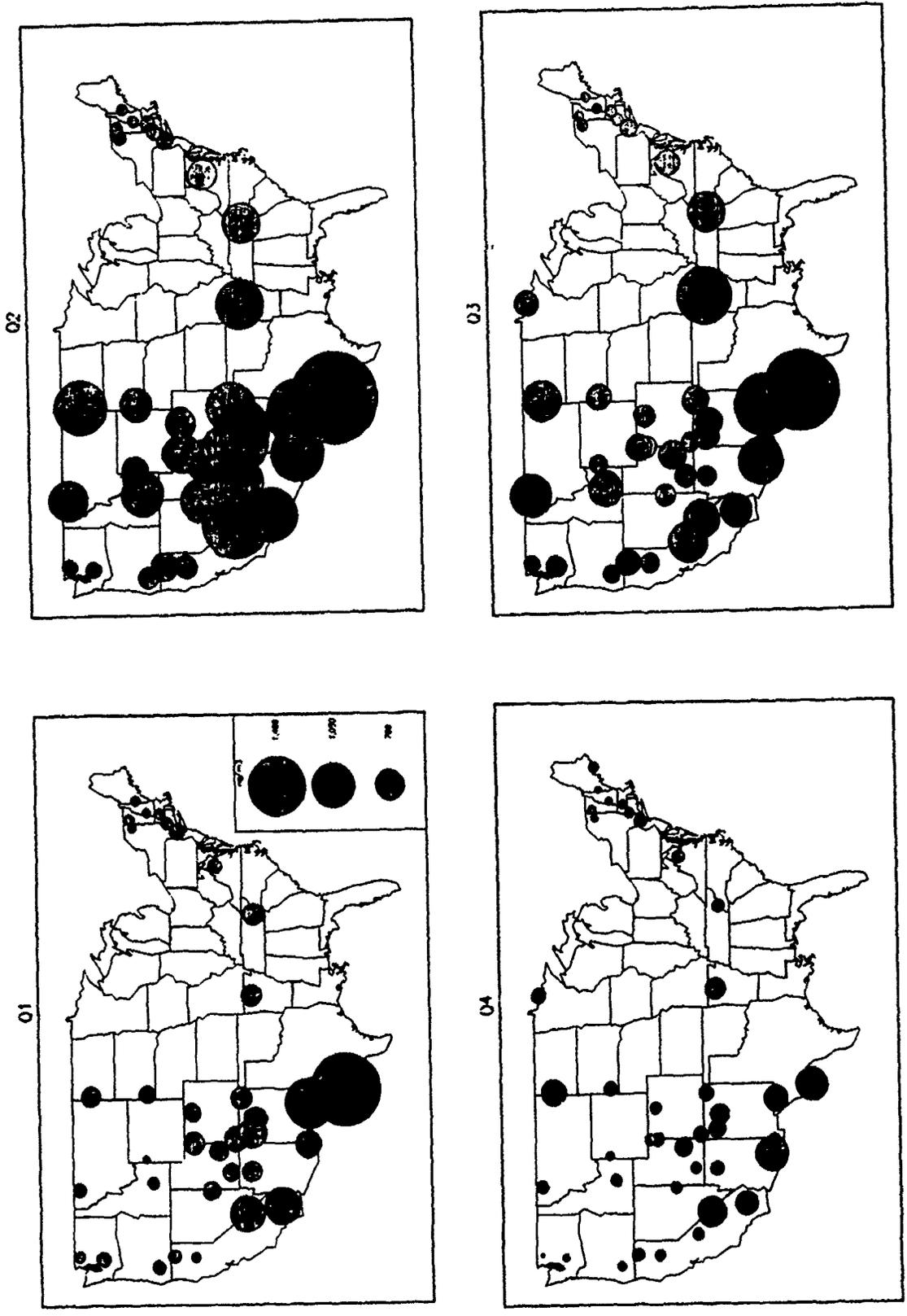


Figure 41. The calculated fine soil concentrations at each station and quarter.

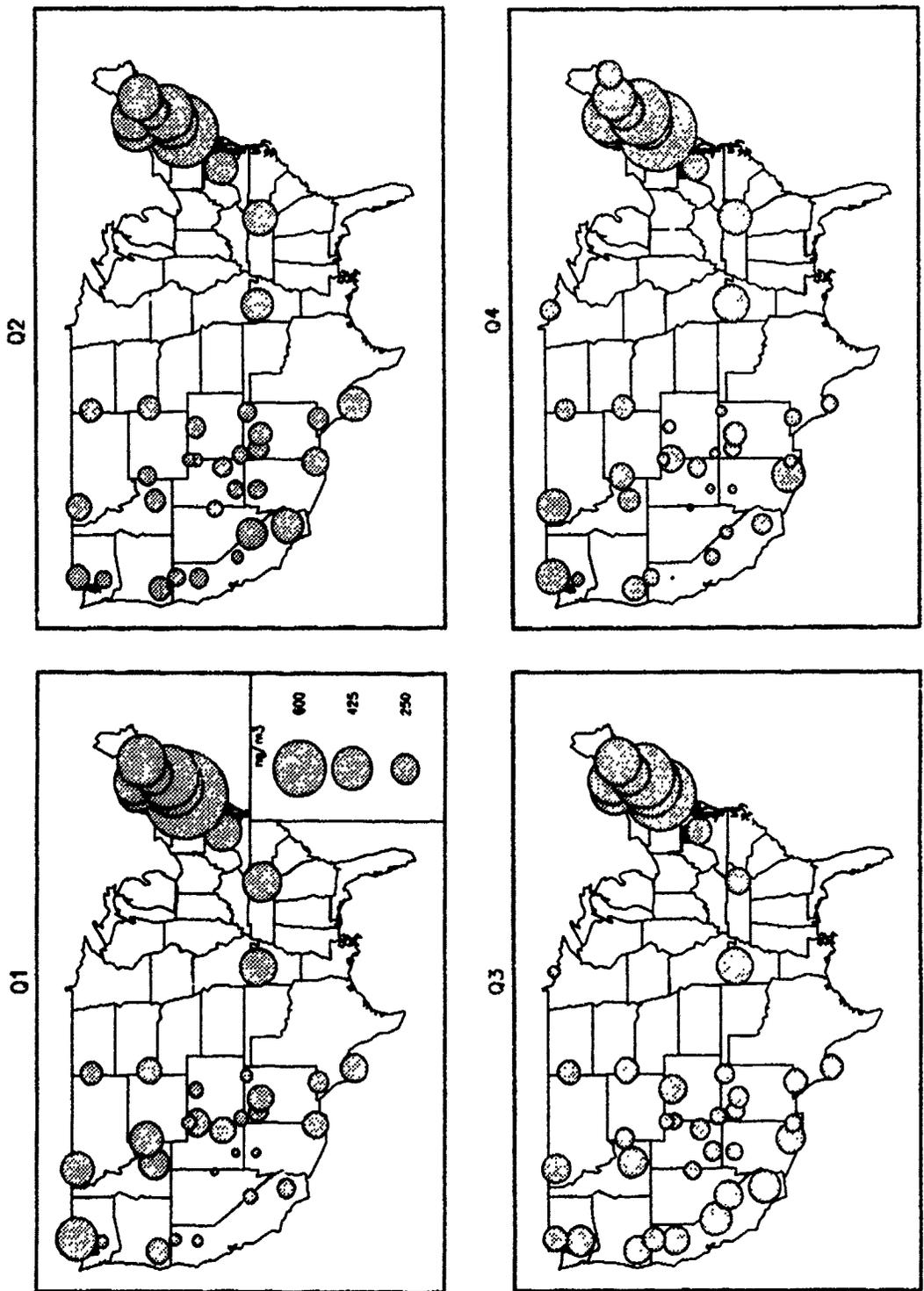


Figure 42. The calculated soot concentrations at each station and quarter.

As shown in Figure 42, the concentrations in the Northeast, the NESCAUM sites, are rather constant containing about 600 ng/m^3 . These concentrations are much larger than the rest of the country where the soot concentrations are less than 300 ng/m^3 . In the West, the concentrations are rather uniform during quarters two and three with the central states having the smallest concentrations around 150 ng/m^3 . During quarters one and four the soot concentrations increase from south to north. The largest western concentrations are in the upper Northwest for these two quarters where they are generally larger than 300 ng/m^3 .

4.1.6 Salt

The salt mass fraction is very variable with the coastal sites generally having higher mass fractions than the inland sites. This variation can range from less than 1% of the fine mass in the central western states to up to 10% in Southern California during quarter 1, Table 7 and Figures 31 & 32.

In the Northeast, there is a strong seasonal dependence with the cold seasons accounting for about 5% of the fine mass while the warm seasons account for less than 2%. The salt mass fraction in the Southeast is invariant with season and accounts for less than 2% of the mass for all seasons. In the West, there is little seasonal dependence, but the second quarter in the Southwest generally has the highest mass fractions where the salt can account for up to 7% of the fine mass.

As shown in Figure 43, the largest salt concentrations occur in the Northeast during the cold season where they are about 400 ng/m^3 . The largest concentrations in the West occur during the warm season at the sites in southern CA and the Smelter region where they can exceed 350 ng/m^3 . The inland sites have the smallest concentrations and are approximately zero during the cold season.

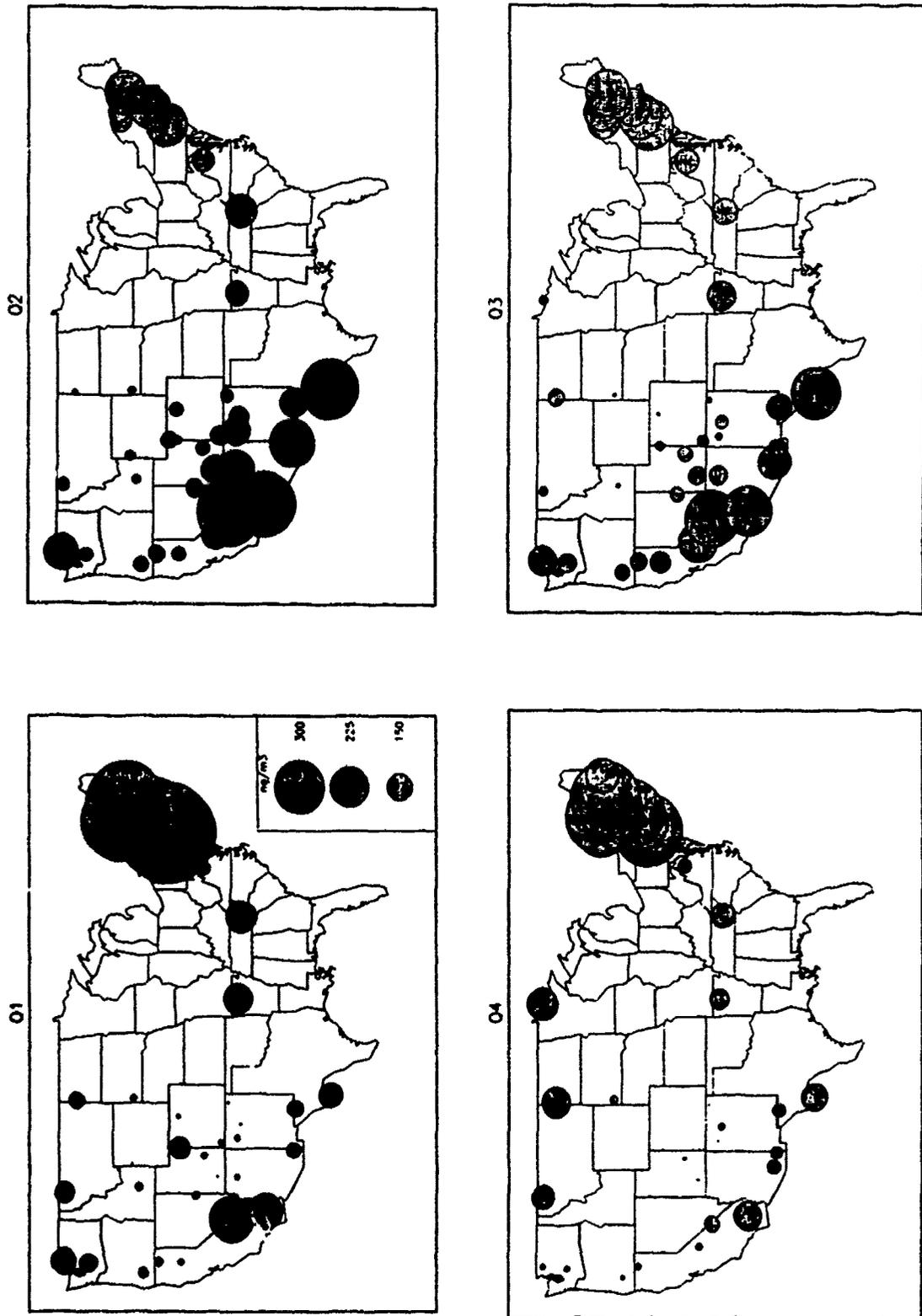


Figure 43. The calculated salt concentrations at each station and quarter.

4.1.7 Unknown

The unknown is the difference between the calculated mass and the measured mass. Assuming we have taken into account all of the major aerosol types except nitrate, define in section 3.1, the unknown should represent the nitrate aerosol. The trends of the unknown may be representative of nitrate, but the magnitudes probably are not. This is because, part of the magnitude of the unknown is a result of any errors in the aerosol equations, assumptions, and the data itself. Also, while we have probably accounted for the major aerosol types, the unknown also contains all of the minor aerosol types such as fly ash.

The best way to examine the trends of the unknown is through its fraction of the fine mass in Figures 27-30. The unknown at the northeastern sites accounts for a very small portion of the mass for all four quarters. In the Southeast the mass fraction increases.

At the western sites the mass fraction of the unknown is dependent upon the season and location. In the Southwest during the cold season the mass of the unknown is very small. During the warm season it increases dramatically to account for almost 10% of the fine mass on average. The largest unknown mass fraction occurs in the Northwest where up to 35% of the mass is unaccounted for. In this region, the unknown exhibits little seasonal variation.

4.2 The Coarse Mass Aerosol Types

This section presents the results from the partitioning of the coarse aerosol mass. These results are presented as temporal and spatial distributions for the coarse aerosol mass, and the three aerosol types, coarse soil, sulfate, and cation, for the NPS-NFPN data network in Figures 44-46. Note that in these plots, the sulfate and cation have been combined and will be referred to as sulfate.

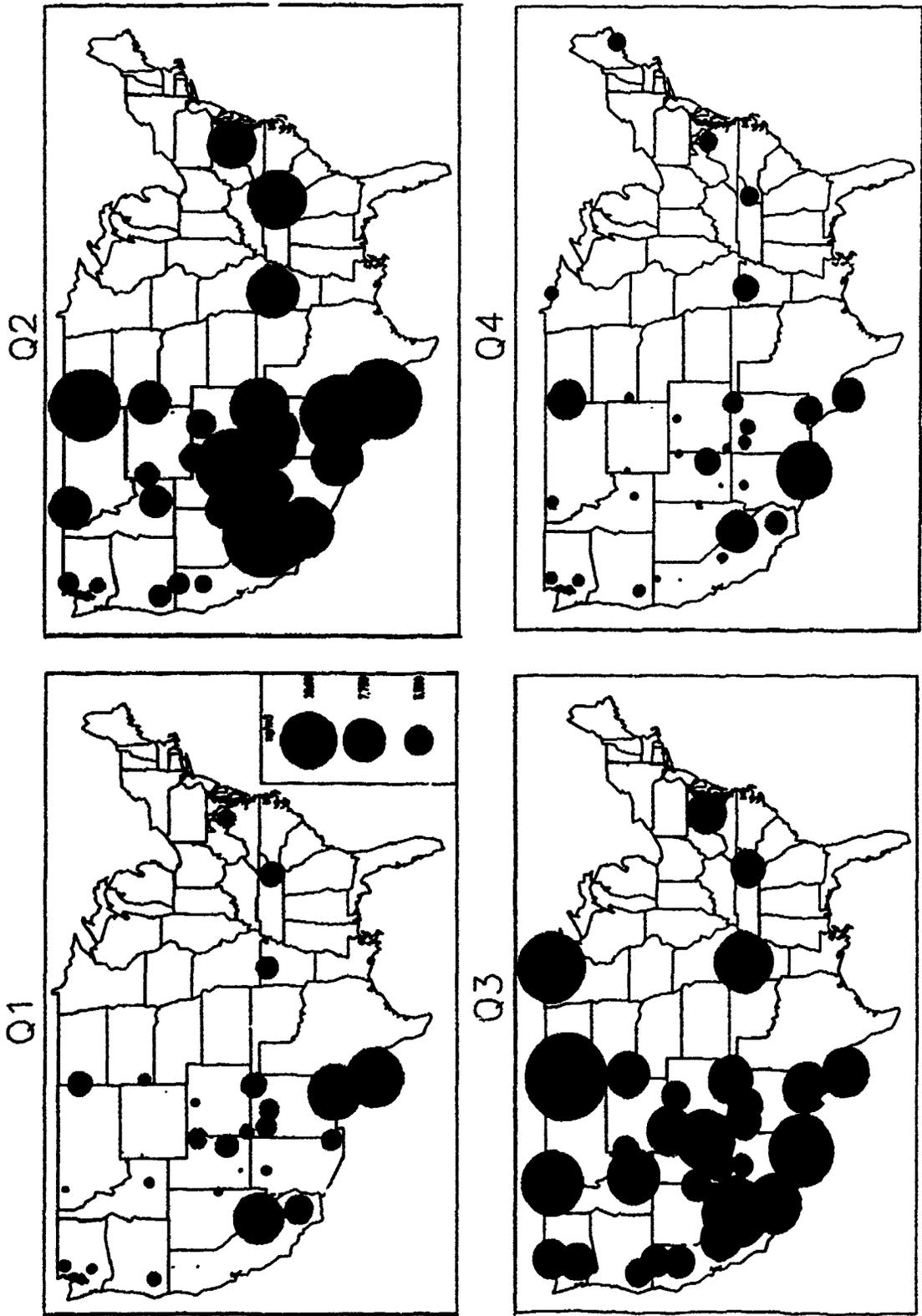


Figure 44. The coarse mass concentrations at each station and quarter.

4.2.1 Coarse Mass

The spatial and seasonal distributions of the coarse mass for the NPS-NFPN data are presented in Figure 44. As shown, there is a strong seasonal variation with the second and third quarter concentrations about twice that of quarters 1 and 4. In the East, the coarse mass concentrations are relatively constant for each quarter. These concentrations are similar to those for the central west for all four quarters.

In the West, the Southwest has the largest concentrations for quarters 1, 2, and 4. However, the coarse mass concentrations are rather uniform over the West during quarter 3, at about 7000 ng/m^3 . The largest coarse particle concentrations in the country occurs during quarter 2 in the Southwest where they can exceed $10,000 \text{ ng/m}^3$, while the smallest are found in the Northwest during quarters 1 and 4 where they are generally less than 3000 ng/m^3 . The stations located in the eastern part of the Northwest, such as Roosevelt, ND have concentrations comparable to those in the Southwest.

4.2.2 Temporal and Seasonal Distribution of the Coarse Aerosol Types

The distribution of the sulfate and cation is shown in Figure 45 for all four quarters. The largest concentrations are found in the East during the second and third quarters where the concentrations can exceed 1000 ng/m^3 . In the West, the concentrations are very uniform for all locations and quarters with an average concentration about 250 ng/m^3 .

The coarse soil concentrations are presented in Figure 46. The coarse soil follows closely the patterns of the coarse mass with the largest concentrations occurring during quarters 2 and 3. The West has larger concentrations than the East for quarters 1 and 4. However, the coarse soil concentrations in the Northwest and East are comparable for quarters 1 and 4, at about 1500 ng/m^3 . The concentrations in the Southwest are larger than those in the Northwest for all quarters except the third where they are comparable at about 6000 ng/m^3 .

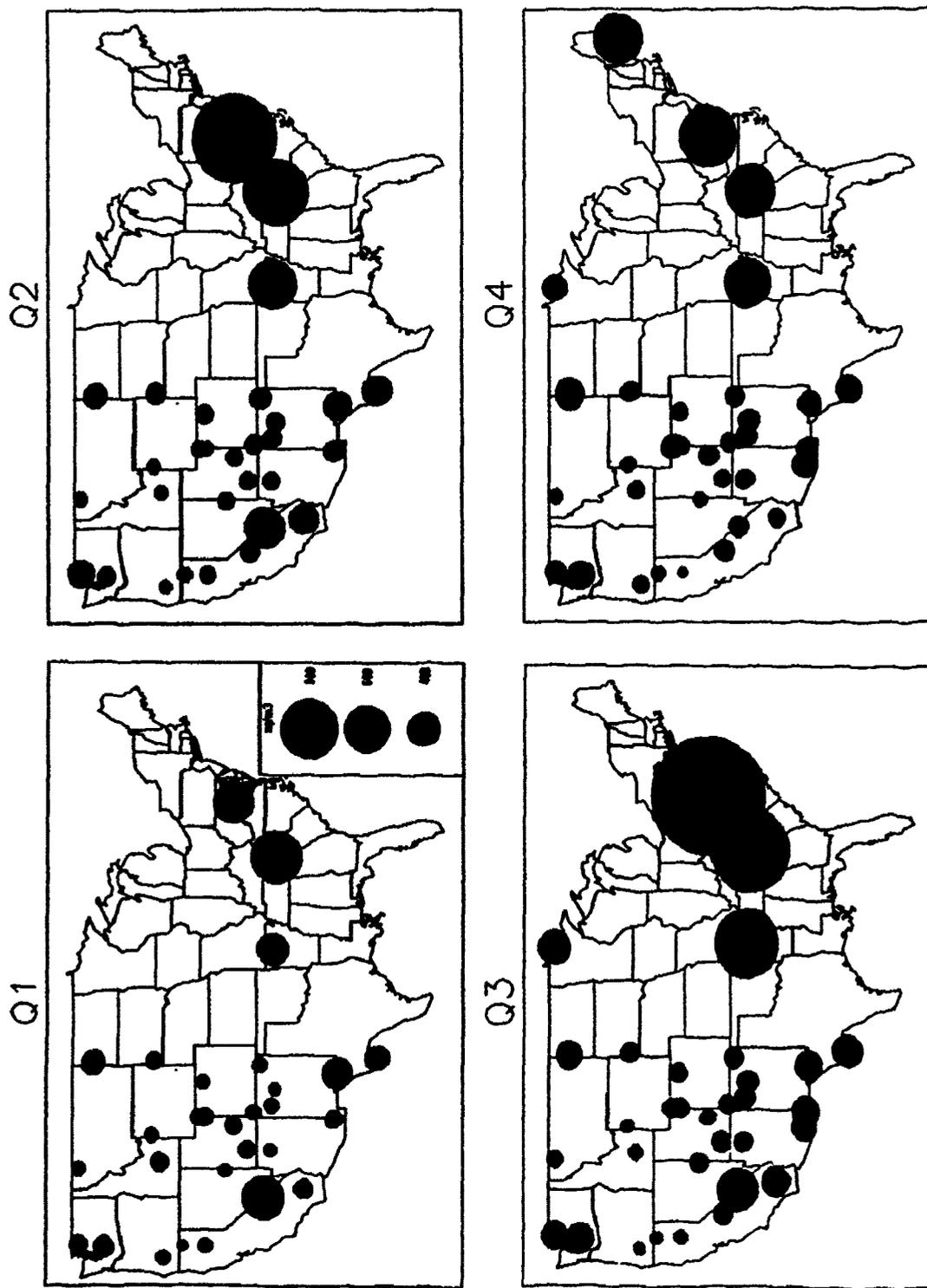


Figure 45. The calculated coarse sulfate + ammonia concentrations at each station and quarter.

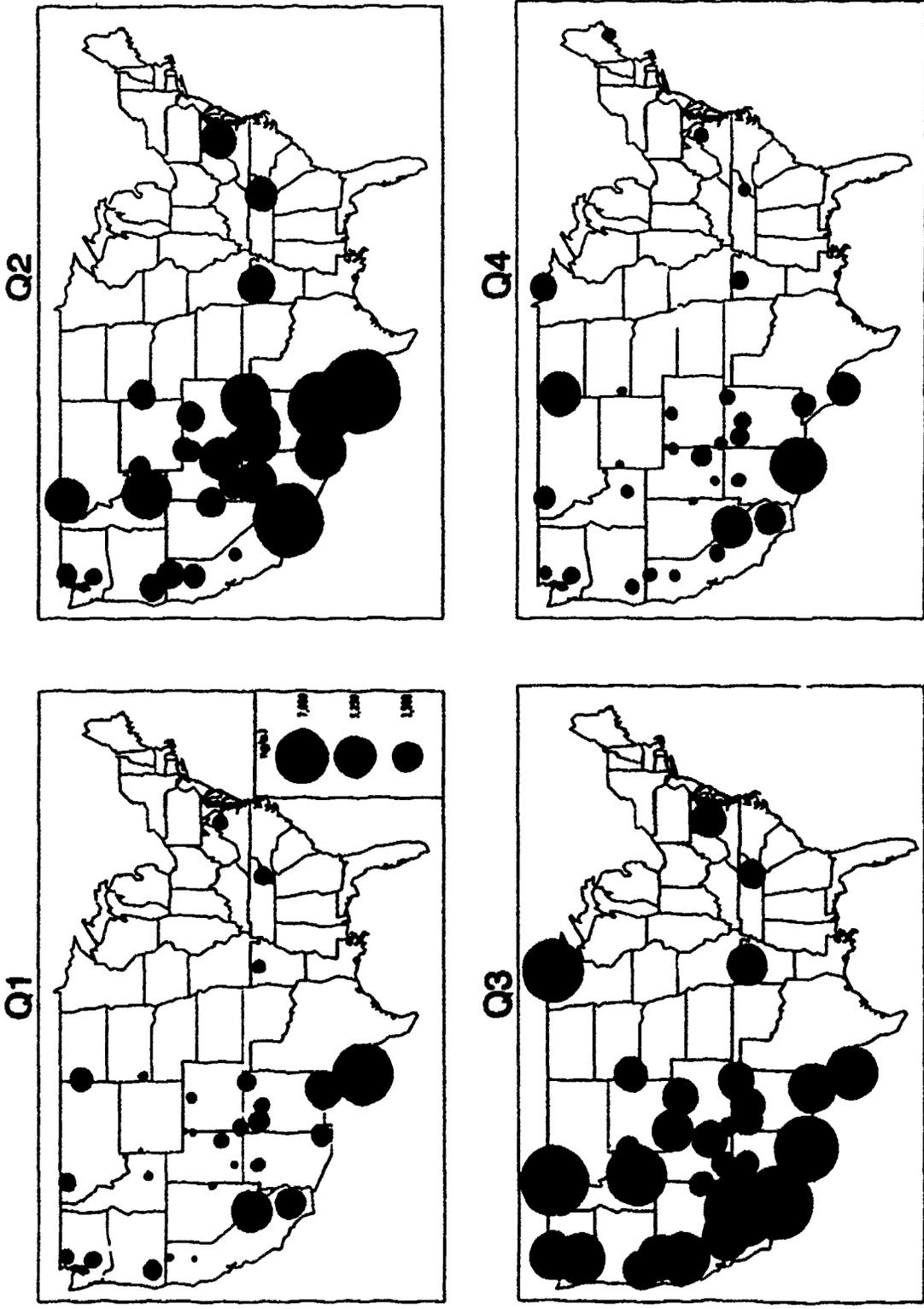


Figure 46. The calculated coarse soil concentrations at each station and quarter.

5. SUMMARY AND DISCUSSION

In this study the concentrations of the major atmospheric aerosol types in rural areas of the U.S. were examined. It was determined that there are at least eight aerosol types, sulfate, cation, water, soot, soil, organics, salt, and nitrates, which constitute the bulk of the fine aerosol mass. The main purpose of this work was to partition the fine aerosol mass into these eight aerosol types.

The aerosol partitioning was conducted on a database which combined the two databases from the NPS-NFPN and NESCAUM monitoring networks. These two databases were combined on the grounds that both networks had their samples analyzed at U.C. Davis in California using the same techniques. Also, examination of the raw data showed that the data from each network behaved in similar manners, and the difference in sampling periods should not introduce any errors. We recognized, however, that the sampling techniques of the two networks were different, and that this could cause some incompatibilities between the data sets.

In order to partition the aerosol mass into aerosol types, aerosol equations were developed. These equations estimated the aerosol types by scaling up tracer species characteristic for specific source types. The tracers were any aerosol species which was solely attributable to one aerosol type. Any scaling factors which could not be determined from the assumed chemical composition of the aerosol types were found through a fitting process.

Seven of these equations were developed for the fine mass defining the sulfate, cation, water, organics, soot, soil, and salt. The nitrate was estimated by the difference between the measured fine mass and the sum of the seven defined aerosol types. The results from the application of these equations were presented as mass fraction in Figures 27-30 and as concentrations in Figures 37-43 for the fine aerosol types. The temporal and spatial trends of the aerosol types were discussed at length in section 4. In this section the aerosol type's concentrations and the equations used to estimate these concentrations will be examined and discussed on a regional basis.

5.1 The Regional Aerosol Types

The variation of aerosol types within and between regions is best illustrated in the regional plots in Figures 31-34. As previously discussed, these figures present the bulk average of the fine mass fraction and aerosol types for each quarter, for the five regions defined in Figure 35.

5.1.1 The Northeast and Southeast

As shown in Figure 31, the organic and sulfur aerosol types (sulfate, water and cation) constitute the vast majority of the aerosol mass in the eastern regions, accounting for nearly 80% of the fine mass. This fraction is smaller during the cold season than the warm season, and is maximum during the third quarter. The Northeast has larger organic mass fractions and smaller sulfur aerosol type fractions than the Southeast. Also, the organic mass fractions were much larger than was found in the literature, section 4.1.3.1. If the organics are reduced in the Northeast as discussed in section 3.3.2, this difference between the two regions no longer exists, and the results of this study agree better with those in the literature. Of the other aerosol types, the soot is largest for the Northeast accounting for about 7% of the fine mass, and soil is largest for the Southeast making up about 6%.

The unknown aerosol fraction accounts for a larger fraction in the Southeast than Northeast. This difference is most likely due to an underestimation of an aerosol type than an increase in nitrate in the sample. As discussed in section 3.2, the nitrate values in the East are very low. The most likely aerosol type which was underestimated was the water. In calculating the water constant these stations were fitted along with the NESCAUM stations. As discussed previously, it appears as though the water was underestimated in the Northeast. If this is the case, then it is reasonable to assume that the water may also have been underestimated in this region.

The variation of the average concentrations for the eastern regions is displayed in Figure 33. Quarters one and four in the Southeast have approximately the same concentrations for each aerosol type which is generally less than quarters 2 and 3. During the warm season quarter 3 has larger concentrations for sulfate and organics than quarter 2. However, quarter 2 concentrations for cation are larger due to the

highly neutralized sulfate aerosol during quarter 2, and the high acidic sulfate aerosol for quarter 3. In the Northeast the larger variation in second and third quarter sulfate values is not seen. Between the two regions the Southeast has larger sulfate concentrations while the Northeast has more organic mass.

5.1.2 The Southwest

The average mass fraction for each aerosol type in the three western regions are presented in Figure 32. As shown, there are three major aerosol types in the Southwest; sulfate (including cation and water), organic, and soil. The soot and salt each account for about 4% of the mass in all four quarters. The sulfate and soil exhibit a seasonal pattern. During quarters 1 and 2 the sulfate and soil each account for about 25% of the mass. But during quarters 3 and 4 the sulfate increases to about 30% while the soil fraction decreases to approximately 20%.

Comparing Figure 32 to Figure 34, it is seen that the concentrations do not follow the mass fraction trend. The sulfate concentrations increase from quarters one through three where a maximum concentration of 1300 ng/m^3 is seen, but falls off sharply during quarter four which had the largest sulfate mass fraction. The soil concentration is constant for quarters one and three while it has the largest concentration for quarter 2, at about 1300 ng/m^3 . In fact, quarter 2 has a soil concentration about twice that of the other quarters.

The mass fraction of the organics in the Southwest is largest for the cold season where it is approximately 20% of the mass. This fraction decreases for the warm season where organics account for only about 10% during quarter two. The concentration of the organics does not exhibit the warm/cold pattern. The largest organic mass occurs during quarters one and three.

The unknown in this region has a definite warm/cold season variation. During the warm season the unknown accounts for more than 10% of the mass, but during the cold season its about 4%. We have assumed that the major constituent of the unknown is nitrate. However, a nitrate concentration on a filter of 10% appears high for the warm seasons. For, from the WRAQS network nitrates measured on the filters were found to constitute less than 3% of the fine mass in the Southwest (NAPAP, 1991(7)). Also the nitrate concentration is larger during the cold seasons than the warm season

(White and Macias, 1987⁽⁵⁰⁾), and an unknown mass fraction of only 4% was found for the cold season. Consequently, it appears as though one or more of the calculated aerosol types was underestimated during the warm season. Therefore, one or more of the aerosol equations or assumptions used in the fitting process are flawed.

There are a few possible explanations for the large unknown aerosol fraction. First, the water may be underestimated as in the East. This is probably not the case due to the dry sampling conditions in the Southwest. Also, the majority of the unknown would remain even if the water concentration was doubled. Another possibility is that the soil is underestimated. As discussed previously the equation we used for the soil may underestimate the true soil mass. This will decrease the unknown, but if we increase the soil by the 16% that Cahill et al., (1989⁽²⁵⁾) suggests we still have an unknown accounting for about 8% of the mass for quarters 2 and 3 which is still high.

A third explanation is that the assumption that the organic constant does not vary with time or space, used to fit the data, was not fully correct, see section 3.1.4. If the percentage of volatile organics in the organic mass changes with site location and season, then the amount lost when the sample is placed into the vacuum for measurement will vary. This variation would effect the organic constant both spatially and temporally.

If the warm season in the Southwest had a larger fraction of its organic mass made up of volatile organics, then the organic constant used to scale the organic hydrogen for these quarters would have been underestimated. This is a result of the criteria of not overestimating the mass in the fitting process. By relaxing the assumption of an invariant organic constant, and increasing this constant for quarter 2 and 3 the organics would increase and the unknown would decrease for these seasons. This, along with increasing the soil, would reduce the unknown to acceptable levels. Also, an increase in the summer organic concentrations would increase the yearly average organic mass fraction. This would decrease the difference in the mass fraction in this study and those compared to the WRAQS study in Table 6.

5.1.3 The Smelter Region

The first two quarters for the Smelter region have about the same mass fractions and trends as the Southwest, except its total mass is larger, Figures 32 and 36. Differences arise between the two regions for the third and fourth quarters. During these seasons the sulfate mass fraction is 10 - 15% larger for the smelter region, and the organic mass fraction is about 10% smaller. However, the soil fraction is about the same in each region. The mass fraction of the unknown for quarters 3 and 4 in the Smelter region is less than the mass fraction of the unknown in the Southwest. Interestingly, the third quarter organics and unknown in the Smelter region is about half of that in the Southwest. This helps to support the idea of a varying organic constant for space and time, for if the organic concentration is underestimated than the smaller its mass fraction the smaller the error in the unknown.

5.1.4 The Northwest

The Northwest region differs significantly from the rest of the West. The sulfate fraction is the smallest in the country accounting for about 15% of the mass. The major aerosol type is organics which account for more than 40% of the mass during quarters 1, 3, and 4. The second quarter organic fraction is much smaller than the other seasons accounting for less than 25% of the mass, but the largest sulfate and soil mass fractions in this region are found during this quarter.

The seasonal variation of the organic concentration in Figure 34, shows that the organic concentrations are roughly equal for quarters 1, 3, and 4, about 1600 ng/m^3 , and that quarter 2 contains about half the organic mass of the other three quarters. The soil and sulfate concentrations during the warm season are all about 700 ng/m^3 . The other aerosol types have small concentrations with salt contributing less than 100 ng/m^3 .

The Northwestern region has a large percentage of its mass unaccounted for by the aerosol equations, about 20% for all four quarters. This is much higher than could be accounted for by nitrates. In the WRAQS study, for the Northwestern stations the nitrates measured on the filters varied between 1 and 6% of the mass (NAPAP, 1991(7)). Part of the unknown mass may be due to underestimating the soil aerosol. Also, the organics may be underestimated, for it is possible that we did not fully account for the loss of volatile organic matter, as was suggested for the Southwest. However,

this seems like a small source of error, for the average yearly organic values did agree very well with the literature.

The most likely cause is that the water had been underestimated. The Northwest region experiences high humidity so the aerosol mass will be affected by the hysteresis effect. Consequently, as was found in the East, the water scaling factors may be too small. Also, to calculate the water we scaled up only the sulfate aerosol. It is believed that a fraction of the organics may also absorb a significant amount of water (Sloane, 1986⁽³⁰⁾; Malm, 1991⁽⁵⁴⁾). Not including the water due to organics could cause a large underestimation in this region, because the sulfate fraction is very low and the organic mass fraction is large.

6. FUTURE WORK

The partitioned data can be used as an initial step to several other studies. One such study would involve the partitioning of the extinction coefficient over the U.S. by using the partitioned data and incorporating any of the suggested changes in section 5, a data set containing the extinction coefficient over the U.S. can be apportioned into contributions from the different aerosol types. This will allow the examination of the greatest contributors to the haze, and may provide insight into means of reducing anthropogenic generated haze.

The partitioned data can also be used as a base of comparison for future aerosol studies. This will allow the monitoring of the aerosol concentrations in the U.S. to test the changes in anthropogenic pollution due to the Clean Air Act of 1990 and any other large scale changes in air pollution generation.

APPENDIX A: AEROSOL TYPE CONCENTRATION TABLES

Table A-1. The aerosol type mass fractions for every station and quarter.

NESCAUM Sites	Q1	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		35.0	9.8	7.6	30.5	3.7	6.7		-1.7	10528.6
Whiteface Mt., NY		37.4	10.5	8.1	28.9	3.4	5.9	1.1	-1.1	8976.6
Mohawk Mt., CT		32.8	9.2	7.1	32.8	3.2	6.5	1.5	2.7	9009.1
Proctor Maple R. F.		35.7	10.1	7.8	31.6	3.6	6.5	6.3	-1.6	7190.0
Quabbin Summit, MA		33.5	9.4	7.3	30.3	3.0	8.0	5.1	3.4	8941.3
Sunapee Mt., NH		36.0	10.1	7.8	29.0	3.4	6.5	6.1	1.1	6452.1
Bridgton, ME		34.3	9.7	7.5	33.2	3.0	7.3	4.1	-1.0	7695.6
AVERAGE		35.0	9.8	7.6	30.9	3.3	7.6	5.5	0.2	7970.5
NESCAUM Sites	Q2	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		37.6	12.0	7.4	32.2	3.4	6.8	2.0	-1.5	12267.8
Whiteface Mt., NY		40.3	12.9	8.0	29.2	3.7	5.3	1.3	-0.8	9482.3
Mohawk Mt., CT		38.2	12.2	7.6	26.7	3.8	5.6	1.7	4.3	10221.2
Proctor Maple R. F.		38.3	12.2	7.6	32.2	3.6	5.3	1.4	-0.6	8919.1
Quabbin Summit, MA		36.9	11.8	7.3	30.1	3.5	5.8	2.4	2.1	10512.8
Sunapee Mt., NH		39.1	12.5	7.7	32.7	3.5	5.0	1.3	-1.9	7569.7
Bridgton, ME		39.0	12.4	7.7	27.2	3.0	5.4	2.7	2.6	9084.1
AVERAGE		38.5	12.3	7.6	30.1	3.5	5.6	1.8	0.6	9722.4
NESCAUM Sites	Q3	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		34.7	5.3	10.0	44.4	2.9	5.9	2.1	-5.4	13958.4
Whiteface Mt., NY		31.5	4.9	9.1	44.3	2.4	4.3	1.7	2.0	12052.9
Mohawk Mt., CT		30.7	4.7	8.9	37.0	2.3	5.1	1.9	9.4	13885.7
Proctor Maple R. F.		29.2	4.5	8.4	45.5	2.3	4.9	2.0	3.1	10612.5
Quabbin Summit, MA		32.2	5.0	9.3	47.4	2.2	5.1	1.6	-2.8	14093.1
Sunapee Mt., NH		30.0	4.6	8.7	46.4	2.2	4.4	1.6	4.1	10266.4
Bridgton, ME		30.2	4.7	8.7	41.3	2.2	4.7	2.4	5.7	10999.9
AVERAGE		31.2	4.8	9.0	43.8	2.4	4.9	1.9	2.0	12266.6

Table A-1. The aerosol type mass fractions for every station and quarter. (Continued)

NPS/CAUK Sites	Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		34.1	9.6	8.7	31.2	2.9	9.3	4.1	0.1	10215.2
Whiteface Mt., NY		37.3	10.5	9.6	29.0	3.2	7.5	5.7	-2.8	5913.7
Mohawk Mt., CT		36.4	10.2	9.3	29.3	3.2	7.8	4.6	-1.0	7484.7
Proctor Maple R. F.		31.6	8.9	8.1	30.4	2.6	7.0	5.1	6.4	7939.0
Quabbin Summit, MA		30.8	8.7	7.9	37.8	2.5	8.5	4.4	-0.7	8876.4
Sunapee Mt., NH		33.5	9.4	8.6	33.4	2.9	6.6	5.1	0.6	6234.4
Bridgton, ME		31.9	9.0	8.2	30.8	2.8	7.1	6.1	4.4	6864.1
AVERAGE		33.6	9.5	8.6	31.7	2.9	7.7	5.0	1.0	7646.8
NPS-NPPW East	Q1	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Shenandoah, VA		43.0	12.1	9.4	13.8	4.7	5.7	1.1	10.2	7321.0
Great Smoky MNT, TN		40.3	11.3	8.8	16.3	6.2	5.0	2.2	9.9	8229.0
Buffalo Natl R., AR		38.7	10.7	7.9	24.4	5.9	4.6	2.1	8.8	8504.0
AVERAGE		39.7	11.4	8.7	18.2	5.6	5.1	1.8	9.6	8018.0
NPS-NPPW East	Q2	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Shenandoah, VA		46.0	14.7	9.1	17.7	6.5	3.0	1.2	1.8	11232.0
Great Smoky MNT, TN		43.4	13.8	8.6	14.1	7.2	2.7	1.5	8.7	13002.0
Buffalo Natl R., AR		38.6	11.6	7.5	14.7	12.4	3.3	1.5	10.4	9466.0
AVERAGE		42.7	13.4	8.4	15.5	8.7	3.0	1.4	7.0	11233.3
NPS-NPPW East	Q3	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Shenandoah, VA		45.2	7.0	13.0	28.6	3.9	1.7	0.9	-0.3	15697.0
Great Smoky MNT, TN		45.7	7.0	13.2	24.2	5.9	1.6	1.0	13.0	15878.9
Buffalo Natl R., AR		37.0	11.1	9.6	25.9	10.3	2.9	1.3	1.9	13320.0
AVERAGE		42.7	8.4	12.0	26.3	6.7	2.0	1.1	4.9	14965.3
NPS-NPPW East	Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Acadia, ME		42.6	12.0	10.9	21.3	4.9	5.2	3.8	-0.7	5138.0
Shenandoah, VA		46.4	13.1	11.9	23.4	4.8	4.3	1.3	-5.3	6192.0
Great Smoky MNT, TN		41.6	12.3	11.2	19.9	4.1	4.9	1.9	2.2	7356.0
Buffalo Natl R., AR		35.3	10.6	9.2	27.2	6.6	5.1	1.6	4.5	7604.0
AVERAGE		42.0	12.0	10.8	23.0	5.1	4.9	2.1	0.2	6572.5

Table A-1. The aerosol type mass fractions for every station and quarter. (Continued)

NPS-NPPN N. West Q1	SO ₄	NH ₄	Water	Organic	Soil	soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	10.0	3.8	2.8	53.8	15.2	10.5	2.9	1.0	2203.0
MOUNT RAINIER, WA	21.0	7.9	5.8	21.8	18.4	3.7	5.4	16.0	1867.0
LASSEN VOLCANIC, CA	13.6	5.1	2.8	--	16.5	4.2	2.5	--	1500.0
LAVA BEDS, CA	12.3	4.6	3.4	50.7	16.0	2.6	2.8	7.5	1942.0
NORTH CASCADES, WA	10.9	4.1	3.0	48.2	4.6	7.3	2.5	19.4	6227.0
GLACIER, MT	17.1	6.4	2.3	49.5	7.2	7.9	2.6	7.0	4599.0
CRATERS OF MOON, ID	19.8	7.4	4.1	40.8	5.9	6.4	1.0	14.5	4951.0
GRAND TETON, WY	10.5	3.9	2.2	48.3	3.0	5.9	0.2	26.1	6100.0
DINOSAUR, CO	21.6	8.1	3.0	36.2	12.2	6.0	2.8	10.0	4396.0
BROWNS PARK, CO	28.7	10.8	4.0	--	5.8	3.3	0.4	--	2976.0
ROCKY MOUNTAIN, CO	29.7	11.1	4.1	31.6	18.6	4.8	1.3	-1.0	2416.0
ROOSEVELT, ND	40.1	15.0	5.5	6.8	12.2	5.0	2.5	12.9	3896.0
WIND CAVE, SD	28.5	10.7	3.9	23.5	12.2	7.0	1.1	12.9	3360.0
Average	20.3	7.6	3.6	37.4	11.4	5.7	2.1	11.5	3571.6

NPS-NPPN N. West Q2	SO ₄	NH ₄	Water	Organic	Soil	soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	16.3	6.1	3.4	33.1	14.8	6.6	2.8	15.0	3136.0
MOUNT RAINIER, WA	26.1	9.8	5.4	29.5	13.2	4.3	2.6	9.2	2858.0
LASSEN VOLCANIC, CA	20.7	7.7	2.8	--	17.8	5.3	2.6	--	3125.0
LAVA BEDS, CA	16.2	6.1	3.3	20.4	20.2	3.9	3.0	27.0	3364.0
NORTH CASCADES, WA	23.7	8.9	1.9	21.4	8.2	4.6	4.4	24.0	4728.0
GLACIER, MT	17.6	6.6	3.6	25.2	20.7	5.4	1.7	19.2	4864.0
CRATERS OF MOON, ID	19.5	7.3	2.7	15.5	32.7	5.8	1.5	15.0	3222.0
GRAND TETON, WY	18.5	6.9	2.5	18.2	17.3	4.2	1.5	30.9	3694.0
DINOSAUR, CO	22.4	8.4	3.1	8.5	30.2	2.4	1.6	23.5	3104.0
BROWNS PARK, CO	24.2	9.1	3.3	26.3	16.8	1.9	2.4	16.1	3792.0
ROCKY MOUNTAIN, CO	26.8	10.1	3.7	17.0	20.0	4.7	2.1	15.6	3730.0
ROOSEVELT, ND	30.0	11.2	6.2	6.3	30.8	4.7	0.9	9.8	4200.0
WIND CAVE, SD	28.0	10.5	3.8	3.8	19.7	5.0	1.0	28.2	4036.0
Average	22.3	8.4	3.8	18.8	20.3	4.5	2.2	19.5	3657.9

Table A-1. The aerosol type mass fractions for every station and quarter. (Continued)

NPS-NFFN N. West Q3	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	15.7	5.9	3.2	38.3	11.1	6.9	2.3	16.5	3858.0
MOUNT RAINIER, WA	24.4	9.1	5.0	28.0	8.6	5.2	1.9	17.8	5779.0
LASSEN VOLCANIC, CA	20.7	7.6	2.8	--	13.3	7.4	3.3	--	3624.0
LAVA BEDS, CA	13.8	5.2	2.8	40.4	18.0	5.0	2.5	15.3	4103.0
NORTH CASCADES, WA	23.8	8.9	4.9	39.2	6.5	3.8	2.8	10.1	6050.0
GLACIER, NT	12.6	4.7	2.6	38.4	18.8	5.2	1.0	16.7	5379.0
CRATERS OF MOON, ID	15.3	5.7	2.1	33.6	20.5	7.5	0.7	14.6	4119.0
GRAND TETON, WY	15.7	5.9	2.2	42.8	10.8	4.3	0.0	18.4	4001.0
DINOSAUR, CO	22.4	8.4	3.1	25.4	13.3	3.0	1.3	23.1	3894.0
BROWNS PARK, CO	22.8	8.4	3.1	20.2	15.6	2.8	-	--	3974.0
ROCKY MOUNTAIN, CO	27.5	10.3	3.8	29.3	12.4	6.3	0.5	9.8	4283.0
ROOSEVELT, ND	24.6	9.2	5.1	26.8	21.0	4.4	2.2	6.7	4597.0
WIND CAVE, SD	25.2	9.5	3.5	29.3	15.1	5.6	0.6	11.3	4072.0
VOYAGEURS, MN	26.4	9.9	3.6	30.8	13.1	1.4	1.2	13.5	4560.0
Average	20.7	7.8	3.4	32.5	13.9	4.9	1.6	14.5	4449.5

NPS-NFFN N. West Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	14.2	5.3	3.9	48.5	10.7	10.1	1.5	5.9	2159.0
MOUNT RAINIER, WA	10.7	7.8	5.7	25.2	8.7	3.7	1.6	26.6	2112.0
LASSEN VOLCANIC, CA	18.8	7.1	3.9	--	16.4	0.0	0.0	--	1729.0
LAVA BEDS, CA	12.8	4.8	3.5	51.1	11.7	4.6	1.6	9.8	2988.0
NORTH CASCADES, WA	8.9	3.3	2.4	49.4	2.1	7.1	0.7	26.0	4857.0
GLACIER, NT	14.3	5.3	2.0	42.1	6.1	6.5	2.6	21.1	5253.0
CRATERS OF MOON, ID	21.5	8.1	4.4	36.3	10.7	7.1	0.0	12.0	2744.0
GRAND TETON, WY	11.9	4.4	2.4	37.1	4.9	5.8	0.0	34.0	4395.0
DINOSAUR, CO	21.9	8.2	3.0	33.9	8.8	6.5	0.3	17.3	3798.0
BROWNS PARK, CO	32.3	12.1	4.4	--	10.2	2.5	0.3	--	2600.0
ROCKY MOUNTAIN, CO	32.2	12.1	4.4	27.0	13.2	3.2	0.0	8.0	2262.0
ROOSEVELT, ND	36.7	13.7	5.0	11.8	17.6	4.9	5.0	5.3	3539.0
WIND CAVE, SD	28.9	10.8	4.0	25.5	10.6	6.7	1.5	12.0	3190.0
VOYAGEURS, MN	42.3	15.9	5.8	16.6	8.8	4.1	4.5	2.0	4137.0
Average	24.4	9.2	4.2	36.8	10.8	5.6	1.5	16.3	3489.5

Table A-1. The aerosol type mass fractions for every station and quarter. (Continued)

NPS-NFFN S. West Q1	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine mass
DEATH VALLEY, CA	18.9	7.1	2.6	15.9	33.2	3.9	10.2	8.2	2586.0
JOSHUA TREE, CA	20.3	7.6	2.8	32.8	27.5	5.2	6.4	-2.4	3211.0
LENNAN CAVES, NV	26.2	9.8	3.6	29.0	28.8	0.0	3.0	-0.5	1501.0
BRUCE CANYON, UT	31.4	11.8	4.3	33.8	26.0	0.7	0.7	-8.8	1672.0
GRAND CANYON, AZ	26.3	9.9	3.6	28.2	24.4	1.2	1.7	4.7	1962.0
CANYONLANDS, UT	31.0	11.6	4.3	18.3	16.3	8.3	1.3	8.9	2872.0
CHIRICAHUA, AZ	34.5	12.9	4.7	19.2	19.0	7.3	2.4	-0.1	3409.0
NEHA VERDE, CO	30.9	11.4	4.2	17.2	24.1	4.6	1.7	6.2	2543.0
CHACO CULTURE, NM	33.1	12.4	4.5	15.5	27.3	6.0	1.4	-0.2	2583.0
BANDELIER, MN	25.5	9.5	3.5	26.6	16.7	6.9	0.5	0.8	3330.0
GUADALUPE MTS, TX	29.5	11.1	4.1	17.9	32.0	4.9	2.4	-1.9	3786.0
CAPULIN MNT, MN	33.5	12.6	4.6	23.4	23.9	3.3	0.8	-2.1	2208.0
BIG BEND, TX	27.1	10.2	3.7	--	37.2	5.5	3.0	--	4681.0
AVERAGE	28.3	10.6	3.9	23.9	25.9	4.4	2.7	1.1	2788.5

NPS-NFFN S. West Q2	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
YOSHUTE, CA	15.4	5.8	2.1	45.3	21.5	3.0	7.5	-0.6	2235.0
DEATH VALLEY, CA	21.8	8.2	3.0	11.1	27.9	5.2	6.5	16.3	5905.0
JOSHUA TREE, CA	23.8	8.9	3.3	14.5	21.4	5.8	6.2	16.2	6045.0
LENNAN CAVES, NV	22.1	8.3	3.0	12.2	30.0	3.2	3.1	18.1	3410.0
BRUCE CANYON, UT	23.5	8.8	3.2	9.6	28.6	4.0	4.3	17.9	3566.0
GRAND CANYON, AZ	22.3	8.4	3.1	7.1	27.2	3.5	5.5	23.0	3912.0
CANYONLANDS, UT	23.4	8.8	3.2	9.6	31.4	4.7	2.4	16.5	3338.0
CHIRICAHUA, AZ	29.3	11.0	4.0	8.1	26.3	5.3	5.4	10.5	5014.0
NEHA VERDE, CO	25.5	9.6	3.5	11.2	30.4	3.7	2.9	13.2	3658.0
CHACO CULTURE, NM	25.6	9.6	3.5	11.3	33.5	4.2	4.2	8.0	3967.0
BANDELIER, MN	24.5	9.2	3.4	19.6	28.2	4.4	2.8	7.9	4529.0
GUADALUPE MTS, TX	27.6	10.3	3.8	9.4	32.8	3.8	3.3	9.0	4859.0
CAPULIN MNT, MN	28.2	10.6	3.9	12.8	31.1	3.9	2.1	7.4	3855.0
BIG BEND, TX	27.2	10.2	3.7	6.9	32.9	5.2	5.3	8.6	6596.0
AVERAGE	24.3	9.1	3.3	13.5	28.8	4.3	4.4	12.3	4349.2

Table A-1. The aerosol type mass fractions for every station and quarter. (Continued)

NPS-NTPN S. West Q3	SO4	NR4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
YOSEMITE, CA	20.2	7.6	2.8	25.5	18.4	6.2	4.3	15.1	5310.0
DEATH VALLEY, CA	27.6	10.4	3.8	19.1	16.6	5.1	5.9	11.5	5459.0
JOSHUA TREE, CA	28.9	10.8	4.0	21.7	13.7	5.8	4.9	10.1	5763.0
LENNAN CAVES, NV	25.3	9.5	3.5	22.9	16.7	4.3	2.8	15.1	3100.0
BRUCE CANYON, UT	30.8	11.6	4.2	23.3	16.8	5.0	3.2	8.1	3226.0
GRAND CANYON, AZ	31.3	11.7	4.3	16.4	12.9	3.6	3.1	16.7	3576.0
SAGUARO, AZ	37.6	14.1	5.2	9.5	19.3	5.3	3.3	5.8	5736.0
CANYONLANDS, UT	29.6	11.1	4.1	15.1	17.4	4.6	2.2	15.9	3857.0
CHRISCAHUA, AZ	45.0	16.9	6.2	10.4	10.9	2.4	1.9	6.4	5595.0
MESA VERDE, CO	34.4	12.9	4.7	16.9	13.5	3.4	1.9	12.2	3521.0
CRACO CULTURE, MN	33.9	12.7	4.7	17.0	14.7	3.6	1.1	12.4	4135.0
BANDOLIER, MN	32.5	12.2	4.5	19.2	14.9	4.1	1.7	11.0	4512.0
GUADALUPE MNTS, TX	34.6	13.0	4.8	15.6	26.1	4.0	2.6	-0.6	5661.0
CAPULIN MNT, MN	35.6	13.4	4.9	17.5	17.1	3.9	0.9	6.8	3955.0
BIG BEND, TX	33.8	12.7	4.6	4.8	29.1	3.4	4.6	7.0	6541.0
AVERAGE	32.1	12.0	4.4	17.0	17.2	4.3	2.0	10.0	4663.1

NPS-NTPN S. West Q4	SO4	NR4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
YOSEMITE, CA	15.1	9.7	2.1	55.1	8.1	3.0	1.2	9.7	3628.0
DEATH VALLEY, CA	25.1	9.4	3.5	22.0	25.2	2.5	3.3	9.0	2837.0
JOSHUA TREE, CA	25.4	9.5	3.5	26.0	16.9	5.2	5.1	8.3	3291.0
LENNAN CAVES, NV	32.1	12.0	4.4	36.5	22.2	0.0	0.5	-5.5	1261.0
BRUCE CANYON, UT	36.3	13.6	5.0	26.2	17.6	2.2	1.0	-1.9	1626.0
GRAND CANYON, AZ	34.5	12.9	4.7	19.4	16.2	0.5	0.5	11.1	2066.0
SAGUARO, AZ	40.4	15.1	5.6	17.7	18.8	7.8	1.5	-7.2	6338.0
CANYONLANDS, UT	33.5	12.6	4.6	19.6	17.7	6.9	1.0	5.2	2481.0
CHRISCAHUA, AZ	49.9	18.7	6.9	5.0	10.6	2.2	2.2	4.4	2942.0
MESA VERDE, CO	39.0	14.6	5.4	22.5	17.8	2.3	0.3	-1.8	2139.0
CRACO CULTURE, MN	38.2	14.3	5.3	21.5	16.0	5.1	0.5	-0.9	2584.0
BANDOLIER, MN	32.0	12.0	4.4	11.9	12.2	5.6	1.1	20.7	3740.0
GUADALUPE MNTS, TX	41.8	15.7	5.7	11.8	19.5	3.8	2.4	-0.7	3163.0
CAPULIN MNT, MN	40.0	15.0	5.2	23.8	19.6	2.3	0.8	-7.0	1853.0
BIG BEND, TX	41.5	15.6	5.7	5.5	19.8	3.5	3.8	4.7	4072.0
AVERAGE	35.0	13.1	4.8	21.6	17.2	3.5	1.7	3.2	2801.6

Table A-2. The aerosol type concentrations for each station and quarter.

NEECAUN Sites	Q1	SO ₄	NH ₄	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		3684.0	1036.1	802.4	3207.2	385.4	1018.4	578.8	-183.7	10528.6
Whiteface Mt., NY		2233.8	628.3	486.6	1728.8	197.2	430.6	338.6	-67.3	5976.6
Nohawk Mt., CT		2954.0	830.8	643.4	2955.9	291.8	677.3	408.9	247.0	9009.1
Proctor Maple R. P.		2549.8	722.7	589.7	2270.0	258.6	476.5	452.1	-116.4	7190.0
Quabbin Summit, MA		2998.3	843.3	653.1	2712.4	264.8	713.5	454.1	301.9	8941.3
Sunapee Mt., NH		2322.0	653.1	508.8	1670.3	216.9	421.7	392.8	69.5	6452.1
Bridgton, ME		2642.4	743.2	578.5	2851.1	228.6	564.4	467.3	-76.9	7695.6
AVERAGE		2772.0	779.6	603.8	2470.8	262.9	614.6	441.8	24.9	7970.5

NEECAUN Sites	Q2	SO ₄	NH ₄	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		4618.8	1472.2	913.7	3948.0	413.1	839.1	247.1	-184.1	12267.8
Whiteface Mt., NY		3822.7	1218.5	756.2	2771.2	353.0	505.9	127.6	-72.8	9482.3
Nohawk Mt., CT		3903.0	1244.1	772.1	2731.5	388.2	571.6	171.6	439.1	10221.2
Proctor Maple R. P.		3413.3	1088.0	678.2	2878.6	318.0	675.4	124.2	-80.6	8919.1
Quabbin Summit, MA		3884.1	1238.1	768.3	3169.5	366.5	610.3	280.2	225.9	10612.8
Sunapee Mt., NH		2960.0	943.5	588.5	2476.3	267.8	381.4	99.4	-144.3	7569.7
Bridgton, ME		3542.0	1129.0	700.7	2468.6	274.3	489.0	248.5	235.1	9084.1
AVERAGE		3734.8	1190.5	738.8	2920.1	340.1	653.2	180.8	64.0	9722.4

NEECAUN Sites	Q3	SO ₄	NH ₄	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		4839.2	748.8	1396.3	6192.0	410.8	827.4	290.9	-746.7	13958.4
Whiteface Mt., NY		3793.0	584.6	1094.4	5341.3	292.0	512.6	199.1	235.9	12082.9
Nohawk Mt., CT		4260.8	656.7	1229.4	5142.9	322.7	706.2	268.4	1298.7	13888.7
Proctor Maple R. P.		3103.4	478.3	898.4	4830.3	247.2	515.6	215.3	327.0	10612.5
Quabbin Summit, MA		4539.5	700.0	1310.0	6467.1	305.9	720.2	220.5	-390.0	14093.1
Sunapee Mt., NH		3079.7	474.7	888.6	4764.8	230.6	488.2	180.2	212.6	10266.4
Bridgton, ME		3324.9	512.4	989.3	4848.3	240.8	520.6	264.0	630.7	10999.9
AVERAGE		3848.6	593.2	1110.5	5357.7	292.8	608.2	231.5	224.0	12366.6

Table A-2. The aerosol type concentrations for each station and quarter. (Continued)

NBSCADW Sites	Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Ringwood, NJ		3483.8	979.7	892.6	3188.9	299.7	946.4	416.7	10.6	10215.2
Whiteface Mt., NY		2204.0	619.9	864.8	1713.6	191.4	444.4	338.7	-143.2	8913.7
Rehawk Mt., CT		2725.9	766.7	698.8	2192.9	244.3	886.9	346.0	-76.8	7484.7
Proctor Maple R. F.		2805.6	704.7	642.1	2410.9	206.3	587.7	407.3	504.4	7939.0
Quabbin Summit, MA		2737.1	769.8	701.4	3383.8	224.0	758.7	391.6	-60.1	8876.4
Sunapee Mt., NH		2086.0	886.7	834.8	2080.4	183.7	411.0	316.6	35.4	6234.4
Bridgton, ME		2187.9	618.4	860.7	2112.6	178.0	489.7	421.3	301.4	6864.1
AVERAGE		2861.4	720.4	686.4	2435.7	217.8	599.3	376.9	78.9	7646.8
NPS-NYPW East	Q1	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Shenandoah, VA		3146.6	885.0	685.4	1013.4	346.7	414.9	79.6	749.6	7321.0
Great Smoky Mtn, TN		3315.0	932.3	722.1	1342.9	806.9	410.2	183.8	815.7	8229.0
Buffalo Natl R., AR		3038.9	910.8	670.9	2071.0	802.0	389.6	178.2	745.4	8504.0
AVERAGE		3168.8	909.4	692.8	1478.8	481.9	404.9	147.2	770.2	8018.0
NPS-NYPW East	Q2	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Shenandoah, VA		8168.8	1647.8	1022.4	1988.7	733.4	341.3	129.8	200.8	11232.0
Great Smoky Mtn, TN		8639.6	1797.6	1118.6	1629.8	942.0	381.4	189.5	1136.5	13002.0
Buffalo Natl R., AR		3682.4	1098.7	712.2	1391.1	1174.4	312.7	142.7	984.7	9466.0
AVERAGE		4820.2	1813.6	980.1	1736.8	949.9	338.1	184.0	773.9	11233.3
NPS-NYPW East	Q3	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Shenandoah, VA		7093.2	1093.2	2046.0	4483.4	610.9	267.4	144.7	-41.8	18497.0
Great Smoky Mtn, TN		7263.2	1119.4	2098.7	3880.0	944.4	248.3	151.0	2065.8	17748.9
Buffalo Natl R., AR		4931.8	1479.8	1282.2	3486.5	1369.9	380.9	170.6	249.0	13320.0
AVERAGE		6429.3	1230.7	1808.0	3930.0	978.0	298.9	188.4	757.6	14965.3
NPS-NYPW East	Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
Acadia, ME		2188.8	618.8	860.8	1096.0	282.3	266.0	196.2	-37.1	8138.0
Shenandoah, VA		2878.1	808.6	736.8	1480.4	300.0	269.2	81.6	-329.7	6192.0
Great Smoky Mtn, TN		3205.1	901.4	821.3	1466.9	301.1	342.9	139.0	158.2	7356.0
Buffalo Natl R., AR		2682.7	804.8	697.8	2064.8	804.8	390.0	118.0	342.0	7604.0
AVERAGE		2737.9	782.6	704.1	1819.8	339.8	322.0	133.7	33.3	6872.8

Table A-2. The aerosol type concentrations for each station and quarter. (Continued)

NPS-NPPN N. West Q1	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	221.4	83.0	60.9	1184.2	335.9	232.3	63.1	22.2	2203.0
MOUNT RAINIER, WA	392.1	147.0	107.8	406.4	344.4	49.4	101.3	298.5	1867.0
LASSEN VOLCANIC, CA	204.1	76.5	42.1	--	247.6	63.4	37.1	--	1800.0
LAVA BEDS, CA	237.9	89.2	65.4	985.6	311.6	51.1	55.1	146.1	1942.0
NORTH CASCADES, WA	677.4	254.0	186.3	3002.1	286.9	456.6	153.3	1210.4	6227.0
GLACIER, MT	784.9	294.3	107.9	2277.8	331.1	362.7	119.5	320.9	4599.0
CRATERS OF MOON, ID	982.7	368.5	202.7	2022.3	294.0	317.3	47.3	716.1	4951.0
GRAND TETON, WY	639.5	239.8	131.8	2946.3	180.6	360.4	11.8	1889.8	6100.0
DINOSAUR, CO	951.4	356.8	130.8	1593.4	536.0	262.1	124.4	441.1	4396.0
BROWNS PARK, CO	855.2	320.7	117.6	--	173.9	98.9	12.6	--	2976.0
ROCKY MOUNTAIN, CO	716.6	268.7	98.5	763.9	449.2	114.9	25.5	-23.3	2414.0
ROOSEVELT, ND	1560.7	545.3	214.6	265.5	474.0	196.0	96.3	503.7	3896.0
WIND CAVE, SD	959.3	359.7	131.9	790.0	411.2	234.9	38.0	435.0	3360.0
Average	706.4	264.9	123.0	1476.1	336.6	216.9	68.1	514.6	3571.6
NPS-NPPN N. West Q2	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	510.8	191.5	105.3	1037.6	527.2	206.3	86.4	468.9	3136.0
MOUNT RAINIER, WA	744.8	279.3	183.6	841.7	377.3	123.8	75.0	262.5	2858.0
LASSEN VOLCANIC, CA	645.8	242.2	88.8	--	557.0	166.4	80.4	--	3125.0
LAVA BEDS, CA	544.4	204.2	112.3	686.0	678.5	129.6	100.5	908.4	3364.0
NORTH CASCADES, WA	1119.8	419.9	231.0	1014.0	386.6	216.8	206.0	1133.8	4728.0
GLACIER, MT	803.0	301.1	165.6	1149.7	946.8	246.3	76.4	874.9	4564.0
CRATERS OF MOON, ID	627.5	235.3	86.3	498.2	1053.5	188.1	48.8	484.4	3222.0
GRAND TETON, WY	683.6	256.4	94.0	672.6	637.9	153.5	54.8	1141.2	3694.0
DINOSAUR, CO	696.5	261.2	95.8	262.6	935.9	74.8	48.4	725.9	3104.0
BROWNS PARK, CO	916.3	343.6	126.0	996.3	638.8	70.5	91.6	608.8	3792.0
ROCKY MOUNTAIN, CO	1000.4	375.2	137.6	634.5	744.3	176.5	78.2	583.3	3730.0
ROOSEVELT, ND	1258.9	472.1	259.7	266.0	1293.7	196.9	39.7	413.1	4200.0
WIND CAVE, SD	1129.3	423.5	155.3	153.3	794.4	200.6	41.4	1138.2	3036.0
Average	821.6	308.1	139.3	684.4	736.3	165.4	79.2	728.9	3657.9

Table A-2. The aerosol type concentrations for each station and quarter. (Continued)

NPS-NPPN N. West Q3	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	607.1	227.7	125.2	1477.4	428.1	265.4	89.5	637.6	3854.0
MOUNT RAINIER, WA	1408.5	528.2	290.5	1619.0	497.4	298.3	107.8	1029.5	5779.0
LASSEN VOLCANIC, CA	729.7	273.6	100.3	--	482.0	266.9	118.0	--	3624.0
LAVA BEDS, CA	865.9	212.2	116.7	1658.3	614.7	203.8	102.6	628.8	4103.0
NORTH CASCADES, WA	1440.5	840.2	297.1	2369.4	392.8	226.9	169.5	613.7	6050.0
GLACIER, MT	678.1	254.3	140.0	2066.9	1010.5	280.7	82.0	896.5	5379.0
CRATERS OF MOON, ID	629.4	236.0	86.5	1385.4	846.4	307.5	27.2	600.5	4119.0
GRAND TETON, WY	627.9	235.5	86.3	1711.1	430.1	173.4	0.9	735.7	4001.0
DINOSAUR, CO	871.6	326.8	119.8	990.1	516.6	118.1	50.3	900.6	3894.0
BROWNS PARK, CO	893.4	338.0	122.8	804.5	618.6	111.1	--	--	3974.0
ROCKY MOUNTAIN, CO	1179.0	442.1	162.1	1254.1	529.7	271.5	23.6	420.9	4283.0
ROOSEVELT, ND	1131.2	424.2	233.3	1234.0	965.5	203.8	98.9	306.1	4597.0
WIND CAVE, SD	1026.9	385.1	141.2	1192.6	613.1	225.3	23.3	461.7	4072.0
VOYAGEURS, MN	1205.4	452.0	165.7	1406.0	595.6	61.8	56.2	617.3	4560.0
Average	928.2	348.1	156.3	1474.5	610.1	215.5	70.7	654.1	4449.5

NPS-NPPN N. West Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
CRATER LAKE, OR	307.0	115.1	84.4	1046.1	230.6	217.4	32.0	126.4	2159.0
MOUNT RAINIER, WA	438.0	164.3	120.5	533.0	184.2	78.0	33.2	940.9	2112.0
LASSEN VOLCANIC, CA	325.3	122.0	67.1	--	284.4	0.0	0.0	--	1729.0
LAVA BEDS, CA	332.3	124.6	91.4	1322.1	303.8	119.3	41.2	253.3	2588.0
NORTH CASCADES, WA	431.3	161.7	118.6	2397.7	103.1	347.2	33.7	1263.6	4857.0
GLACIER, MT	749.0	280.9	103.0	2213.7	318.3	343.6	137.4	1107.1	5283.0
CRATERS OF MOON, ID	589.6	221.1	121.6	996.4	294.9	195.4	0.0	329.4	2744.0
GRAND TETON, WY	521.1	195.4	107.5	1629.1	214.6	293.2	0.0	1492.6	4395.0
DINOSAUR, CO	832.7	312.3	114.5	1288.1	334.7	246.1	11.7	657.8	3798.0
BROWNS PARK, CO	840.0	318.0	115.5	--	244.3	65.1	8.8	--	2600.0
ROCKY MOUNTAIN, CO	728.8	273.3	100.2	609.9	299.4	72.5	0.0	180.7	2242.0
ROOSEVELT, ND	1297.4	486.5	178.4	418.7	621.9	172.8	175.5	187.9	3539.0
WIND CAVE, SD	920.4	345.1	126.6	814.1	339.7	214.9	47.4	381.9	3190.0
VOYAGEURS, MN	1750.0	656.2	240.6	687.7	362.8	171.0	186.1	82.6	4137.0
Average	774.1	290.3	130.0	1268.8	319.7	192.0	54.4	602.2	3489.5

Table A-2. The aerosol type concentrations for each station and quarter. (Continued)

NPS-NPPN S. West Q1	SO ₄	NH ₄	Water	Organic	Soil	Soot	Salt	Unknown	Fine mass
DEATH VALLEY, CA	483.1	181.2	66.4	408.4	849.1	99.6	261.3	210.0	2586.0
JOSHUA TREE, CA	653.2	244.9	89.8	1044.9	881.6	168.2	208.7	-77.4	3211.0
LEHMAN CAVES, NV	393.3	147.8	54.1	435.9	432.2	0.0	48.0	-6.8	1501.0
BRUCE CANYON, UT	525.8	197.1	72.3	565.1	435.3	11.9	11.2	-146.3	1672.0
GRAND CANYON, AZ	516.4	193.6	71.0	552.5	478.7	22.7	34.3	92.8	1962.0
CANYONLANDS, UT	890.1	333.8	122.4	524.3	468.4	238.4	38.5	286.2	2872.0
CHIRICAHUA, AZ	1175.6	440.9	161.7	654.6	647.0	249.7	82.2	-2.5	3409.0
MESA VERDE, CO	776.3	291.1	106.7	436.8	612.3	117.3	44.1	158.4	2543.0
CRACO CULTURE, NM	854.0	320.3	117.4	400.8	705.4	154.4	36.7	-6.1	2543.0
MANDELIER, NM	847.9	317.9	116.6	1217.8	556.1	230.2	16.9	26.7	3330.0
GUADALUPE MTS, TX	1109.3	416.0	182.8	671.3	1202.8	182.8	91.9	-70.4	3756.0
CAPULIN MTN, NM	738.8	277.1	101.6	515.4	527.7	72.8	17.6	-48.6	2205.0
BIG BEND, TX	1260.8	472.7	173.3	--	1731.4	254.7	138.4	--	4651.0
AVERAGE	786.5	294.9	108.1	618.7	732.9	138.6	78.7	32.4	2788.5
NPS-NPPN S. West Q2	SO ₄	NH ₄	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
YOSEMITE, CA	348.1	129.4	47.8	1012.1	480.1	66.3	167.2	-12.6	2235.0
DEATH VALLEY, CA	1285.3	482.0	176.7	657.7	1649.8	306.0	386.0	961.8	5908.0
JOSHUA TREE, CA	1436.0	538.5	197.4	877.3	1293.6	349.8	372.3	980.1	6048.0
LEHMAN CAVES, NV	753.1	282.4	103.6	416.9	1023.9	107.9	105.9	616.3	3410.0
BRUCE CANYON, UT	839.7	314.9	115.8	342.6	1019.6	141.1	154.9	637.7	3866.0
GRAND CANYON, AZ	872.2	327.1	119.9	276.4	1063.0	136.8	216.6	900.0	3912.0
CANYONLANDS, UT	781.7	293.1	107.8	319.1	1047.0	186.3	81.4	551.8	3338.0
CHIRICAHUA, AZ	1471.4	551.8	202.3	406.8	1318.9	268.5	270.8	526.5	5014.0
MESA VERDE, CO	932.0	349.5	128.1	409.0	1112.7	135.4	107.8	483.4	3658.0
CRACO CULTURE, NM	1014.9	380.6	139.8	446.8	1330.0	168.2	168.1	318.9	3967.0
MANDELIER, NM	1108.6	415.7	152.4	888.7	1278.3	199.6	126.8	359.1	4529.0
GUADALUPE MTS, TX	1339.0	502.1	184.1	455.3	1892.1	186.7	160.8	439.3	4859.0
CAPULIN MTN, NM	1088.8	408.3	149.7	493.9	1197.3	149.8	80.4	286.9	3885.0
BIG BEND, TX	1791.8	671.9	246.4	482.0	2172.2	345.2	347.1	569.6	6896.0
AVERAGE	1078.7	403.4	147.9	532.8	1258.6	193.9	196.1	544.2	4349.2

Table A-2. The aerosol type concentrations for each station and quarter. (Continued)

NPS-NTPN S. West Q3	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
YOSHUTE, CA	1070.3	401.3	147.2	1355.4	976.0	327.7	230.2	601.9	5310.0
DEATH VALLEY, CA	1507.5	545.3	207.3	1045.3	906.3	277.9	323.7	625.8	5459.0
JOSHUA TREE, CA	1664.7	624.3	228.9	1252.7	791.0	333.3	284.3	583.8	5763.0
LENNAN CAVES, NV	782.9	293.6	107.7	709.6	518.4	132.9	86.7	448.3	3100.0
BRUCE CANYON, UT	994.3	372.8	136.7	750.8	542.4	160.8	102.9	165.2	3226.0
GRAND CANYON, AZ	1117.9	419.2	153.7	585.4	461.0	129.6	111.6	597.6	3576.0
SAGUARO, AZ	2184.8	808.1	296.3	545.1	1106.8	302.2	188.2	334.5	5736.0
CANYONLANDS, UT	1143.2	428.7	157.2	581.3	669.9	176.1	86.1	614.5	3857.0
CHIRICAHUA, AZ	2516.0	943.5	345.9	582.4	608.9	137.1	104.1	357.2	5595.0
NEHA VERDE, CO	1211.4	454.3	166.6	596.2	475.2	120.9	65.4	431.1	3521.0
CHACO CULTURE, NM	1402.1	525.8	192.8	702.5	608.0	147.5	44.4	512.0	4135.0
BANDELIER, NM	1465.8	549.7	201.8	864.1	672.7	186.8	77.4	496.0	4512.0
GUADALUPE MTS, TX	1960.0	735.0	269.5	885.8	1477.3	224.7	145.6	-36.8	5661.0
CAPULIN MTS, NM	1409.0	528.4	193.7	692.2	676.1	152.6	35.3	267.7	3955.0
BIG BEND, TX	2209.1	828.4	303.8	316.6	1901.8	221.6	299.6	460.1	6541.0
AVERAGE	1507.3	545.2	207.2	764.4	826.1	202.0	145.7	445.3	4663.1

NPS-NTPN S. West Q4	SO4	NH4	Water	Organic	Soil	Soot	Salt	Unknown	Fine Mass
YOSHUTE, CA	548.5	205.7	75.4	1999.7	294.5	109.3	44.1	351.0	3628.0
DEATH VALLEY, CA	712.1	267.0	97.9	622.8	718.8	71.5	94.1	255.7	2837.0
JOSHUA TREE, CA	835.0	313.1	114.8	857.0	557.5	170.6	168.4	274.5	3291.0
LENNAN CAVES, NV	405.2	151.9	55.7	459.9	280.5	0.0	6.7	-49.6	1261.0
BRUCE CANYON, UT	590.7	221.5	81.2	425.8	286.5	35.0	16.9	-31.6	1626.0
GRAND CANYON, AZ	712.3	267.1	97.9	401.3	325.2	10.8	10.9	230.3	2066.0
SAGUARO, AZ	1752.0	657.0	240.9	769.7	817.3	337.6	77.2	-313.7	4338.0
CANYONLANDS, UT	831.4	311.8	114.3	461.8	439.4	170.1	24.0	128.1	2481.0
CHIRICAHUA, AZ	1468.5	550.7	201.9	148.6	310.7	65.1	65.8	130.8	2942.0
NEHA VERDE, CO	833.8	312.7	114.6	490.7	380.1	48.4	6.4	-37.7	2139.0
CHACO CULTURE, NM	987.9	370.5	135.8	554.7	414.2	133.0	12.1	-24.3	2584.0
BANDELIER, NM	1197.8	449.2	164.7	445.1	457.2	211.3	41.4	773.5	3740.0
GUADALUPE MTS, TX	1320.7	495.3	181.6	373.8	618.3	121.4	75.4	-23.5	3163.0
CAPULIN MTS, NM	741.3	278.0	101.9	441.4	362.6	43.2	14.4	-129.8	1853.0
BIG BEND, TX	1690.3	633.9	232.4	223.5	806.1	141.9	154.6	190.4	4072.0
AVERAGE	975.2	365.7	134.1	577.7	471.7	111.3	54.2	113.6	2801.4

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