

**Winter Fine Particle Composition in the Northeast:
Preliminary Results From the NESCAUM Monitoring Network**

Richard L. Poirot
Air Pollution Control Division
Vermont Agency of Natural Resources, Waterbury, VT 05676

Robert G. Flocchini
Land, Air and Water Resources and Crocker Nuclear Laboratory
University of California, Davis, CA 95616

Rudolf B. Husar
Center for Air Pollution Impact and Trend Analysis
Washington University, St. Louis, MO 63130

Presented at the 83rd Annual Meeting of the Air and Waste
Management Association, Pittsburgh, PA, June, 1990.

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INTRODUCTION

A regional particle monitoring network was initiated in the fall of 1988 by the Northeast States for Coordinated Air Use Management (NESCAUM), to promote a better understanding of regional-scale particulate matter concentrations and composition. The NESCAUM states felt such information was needed for the effective implementation of a number of current state regulatory programs, including attainment, maintenance, and PSD programs for PM-10, visibility protection in Class I Federal areas, and various state air toxics programs. The resultant data might also help support development of potential future regulatory programs, such as Phase II (regional haze) regulations for Class I areas, secondary fine particle standard(s) to protect visibility in all areas, or primary standards for acid aerosols, and could be particularly useful in tracking the long-term effects of proposed Clean Air Act revisions.

The Network is operated by the participating state air quality programs in NY, NJ, CT, MA, ME, NH, and VT, in cooperation with the Air Quality Group at the University of California at Davis (UCD). Technical advice and assistance has been provided by the Air Quality Division of the National Park Service (NPS) and the Environmental Monitoring Systems Laboratory of the Environmental Protection Agency (EPA). Funding to support the first two years of operation has been provided by Region I EPA and participating states.

The Voyager Data Exploration Software¹ is being utilized by the NESCAUM states to help screen, edit, exchange, interpret, and report the results of the regional monitoring network. This paper presents some of the currently available NESCAUM data, and displays some simple interpretations using the Voyager software. An electronic version of this paper is available as a Voyager file, which includes a textual narrative, several prepared views and interpretations of the data, and the raw data itself--which may be viewed, manipulated, compared to related data sets, etc., with the Voyager software. The authors hope that this will encourage others to utilize the detailed NESCAUM data set and promote a more rapid and comprehensive exchange of information than has previously been possible.

METHODS

Sites

Rural, regionally representative monitoring sites have been established at Bridgton, ME; Mt. Sunapee, NH; Mohawk Mtn., CT; Underhill, VT; Quabbin Summit, MA; Ringwood State Park, NJ; and Whiteface Mtn., NY. Site locations (see Table I and Figure 1), have been audited by UCD and found consistent with EPA, NPS, and IMPROVE siting criteria. Several NESCAUM sites are (or were) co-located with other monitoring programs (including National Atmospheric Deposition Program, National Dry Deposition Network, Operational Evaluation Network and Eastern Fine Particle Visibility Network) providing opportunities for supplemental data and methods intercomparisons.

Sampling and Analysis

Sampling is conducted at each site for particles ≤ 10 microns and ≤ 2.5 microns aerodynamic diameter. PM-10 samples are collected using Weddington critical flow high volume PM-10 samplers (EPA reference method #RFPS-1087-062), equipped with standard EPA calendar year quartz PM-10 filters, run 24 hours (midnight to midnight, EST) on the standard EPA 1 in 6 day sampling schedule. These filters are currently analyzed for mass (gravimetric) and sulfate (ion chromatography) in participating state laboratories.

Fine particles are collected using UCD cyclone (IMPROVE-type) samplers, equipped with stretched teflon filters run 24 hours (midnight to midnight, EST) every Wednesday, Saturday, and every 6th day. The samplers are sufficiently automated that this sampling schedule can generally be maintained with weekly site visits. Fine particle filters are analyzed by UCD for mass (gravimetric), optical absorption (Integrating Plate Method), hydrogen (Proton Elastic Scattering Analysis), and multiple elements--Na and heavier (Proton Induced X-ray Emission). Additional UCD cyclone samplers are deployed at the Whiteface and Quabbin sites, equipped with pre-fired quartz filters, run on the Wednesday, Saturday, 6th day schedule, and analyzed for elemental carbon and organic carbon (thermal optical reflectance method) by the Desert Research Institute (DRI).

The cyclone samplers, Wednesday-Saturday sampling schedule, and filter analysis are essentially identical to a portion of the particle sampling conducted by the IMPROVE (Interagency Monitoring of PROtected Visual Environments) and NPS sampling networks. Consequently, the NESCAUM network enhances the spatial coverage of these more comprehensive (but predominantly western) networks. In return, NESCAUM reaps considerable informational, financial, and personnel benefits from the continuing methods development, QA/QC programs, automated filter processing, and data interpretation techniques which result from the IMPROVE and NPS programs. Additional details on the UCD cyclone sampler^{2,3}, IMPROVE network⁵⁻⁷, and sampling procedures²⁻⁴, analytical methods and uncertainties⁵⁻⁷, and NESCAUM sampling procedures⁸ have been previously reported.

Data Management and Availability

Raw data resulting from the UCD and DRI filter analyses are routinely screened by UCD⁴ and conveyed to each NESCAUM state. These data are also converted to Voyager format by Lantern Corp. and submitted to the states in this form. (The individual states use a variety of different and not always very compatible, computerized data management systems, and Voyager is one system they all have in common. It is particularly useful for screening, interpreting, intercomparing and reporting large multivariate data sets of this nature). Each state conducts additional screening, flags any suspect data values, and reports these along with the PM-10 and sulfate data back to Lantern Corp. and UCD. As network operations become routine, it is anticipated that this database will be updated within four months of the end of each calendar quarter, and will be available as ASCII file from UCD or Voyager file from Lantern Corp.

With complete data recovery, an annual NESCAUM data set would be composed of approximately 150 days of fine mass, optical absorption, hydrogen and PIXE measurements and 60 days of PM-10 mass and sulfate for each of seven sites, plus 150 days of elemental carbon and organic carbon measurements from two of these sites. There would be 104 days/site year with NESCAUM fine particle results concurrent with the (Wednesday-Saturday) NPS and IMPROVE measurements, and 60 days/site year with NESCAUM PM-10 and fine particle results concurrent with standard EPA (one in six day) PM-10 measurements.

Although we anticipate release of the first full year of NESCAUM data (September 88-August 89) by early summer 1990, at the present time (spring 1990), only a portion of the fine particle filters (Wednesday-Saturday teflon) from the first two quarters have been analyzed. The fall (SON) 1988 sampling period is incomplete, as no sites began operation prior to the third week of September and two sites (Quabbin and Mt. Sunapee) did not begin operation until the winter quarter (DJF). There were a large number of missing or invalid samples during these first two quarters, due in part to unexpected complexities in mastering the unusual sampling schedule, and in part to experimentation with flow restrictors in an attempt to improve detection limits for Se. These problems have since been resolved, and a much higher data recovery efficiency has occurred in more recent sampling periods. The results presented here are based on only about 1/4 of the filters which will ordinarily/eventually be analyzed from a six month sampling period.

RESULTS

Table II lists the average winter (DJF) 1988-89 values for fine mass, optical absorption, and elements above detection limits in every sample. Given the relatively small sample sizes, caution should be exercised in comparing these seasonal mean values for some of the sites. Taken as a whole, however, the data suggest a gradual spatial gradient of increasing concentrations as one moves from northwest to southeast in the region, with the highest average concentrations of fine mass and most elements generally less than a factor of two greater than those at the cleanest sites. Optical absorption is about twice as high at Ringwood, NJ (about 30 miles from New York City) as at Whiteface Mountain (250 miles to the north). Bridgton, ME exhibits relatively high concentrations of K, but otherwise the concentrations of most elements in Table II are relatively uniform and in general proportion to mass concentrations at most sites.

Other elements detectable on most or some of the winter filters include Cu (detectable on 95% of filters), Mn (94%), V (84%), Ti (80%), Ni (65%), Se (57%), Al (38%), As (25%), and Cr (15%). Like potassium, many of these other trace elements exhibit much more dynamic spatial (and temporal) variation than most of the variables listed in Table II, and may provide unique opportunities to identify local and distant source influences by receptor methods.

Data Views

Figures 1 through 5 are example Voyager views which display some of the spatial, temporal, and relational aspects of the data. They are intended to demonstrate the kinds of information inherent to this particular suite of measurements, and the efficiency of the Voyager system for viewing and interpreting large multivariate data sets of this nature.

Figure 1 displays site locations and demonstrates the potential utility of the trace element data for identification of complex source-type influences. Figure 1a displays relative fine mass concentrations and sulfate fractions on 2/18/89, both of which are highest at the Quabbin, MA site in the center of the region. (Sulfate includes estimated associated cations, and is assumed in this case to be composed of a 50:50 mixture of ammonium sulfate and ammonium bisulfate). It might be considered a "typical day" in the sense that mass values and sulfate fractions are similar to seasonal means, and show a general pattern of higher concentrations at the more southerly sites. Figures 1b, c, and d display the relative concentrations of V, As, and K on this date and suggest a much more complex pattern of potential source influences than the gradual mass and sulfur gradients would indicate.

Figures 2 and 3 display some of the relationships between fine mass, sulfur, and hydrogen, and also demonstrate the utility of Voyager as a screening tool. Figures 2a and b show the temporal patterns in fine sulfur and fine mass at all the sites, and suggest relatively uniform regional scale variations for these two variables. An obvious exception to these uniform regional patterns is the apparent isolated fine mass episode at Underhill, Vermont on 11/19/88. Figure 3a compares the temporal patterns in fine mass, sulfur, and hydrogen at the Vermont site. (Sulfur and hydrogen are multiplied by factors of 8 and 20 respectively, to allow a more direct comparison on the fine mass scale). Again, the mass value on 11/19/88 stands out as dramatic exception to the otherwise strong co-variation of these variables at this site.

Figure 3b shows the S vs. mass and H vs. mass relationships for all samples from all sites for the fall and winter quarters. The 11/19/88 VT mass value is distinctly out of line with all other data points and has subsequently been flagged as suspect. Also noteworthy in Figures 2b and 3b are network-wide S/mass and H/mass ratios of about 1/8 and 1/20 respectively, and the fact that hydrogen (though composing only 5% of mass) correlates even more strongly with mass than sulfur does. For the winter season, the network-wide correlation coefficients (r) for S vs. mass and H vs. mass were 0.90 and 0.96 respectively.

Undoubtedly, a large portion of the particulate hydrogen is chemically associated with sulfur as ammonium sulfate, bisulfate, and/or sulfuric acid, but even assuming a maximum feasible H to S molar ratio of 8 to 1 (ammonium sulfate), substantial remaining hydrogen must be associated with non-sulfur compounds which may help account for additional variance of the mass concentrations.

UCD researchers have suggested that organic matter is the predominant contributor to non-sulfate hydrogen on the teflon filters, since other potentially significant hydrogen containing ambient species, including water and ammonium nitrate, are likely to be lost during sampling, prior to analysis, and/or^{5,9} under the vacuum conditions employed in the PIXE and PESA analyses. If assumptions are made about average sulfate speciation and the average hydrogen fraction of organic matter, then estimates of organic matter concentrations can be readily derived⁵ from the S and H measurements on the teflon filters. Eldred et al.⁵ have recently applied such estimates for organics to a large number of sites in the IMPROVE and NPS networks and found an excellent agreement with organic carbon measured by DRI on quartz filters (corrected for artifact) from these same sites and sample dates.

Estimated NESCAUM Winter Fine Mass Composition

Using the extensive long-term NPS and IMPROVE data sets, the UCD group has developed and refined methods for estimating the major components of fine particulate matter which are most relevant to visibility effects.^{5,9} Sulfates (sulfate and associated ammonium) are estimated from fine sulfur concentrations and assumptions regarding sulfur speciation (i.e., ammonium sulfate in the West, bisulfate in the East). Soil is estimated from concentrations of Si, Al, Ca, Fe, Ti, and non-smoke K assumed to be in their common oxide forms. Soot is estimated from optical absorption measurements and assumptions regarding the absorption efficiency of soot carbon. Organic matter is estimated by the non-sulfate hydrogen method referred to earlier, and also by the remaining mass method, which takes a fixed fraction of the mass remaining after sulfates, soil and soot have been subtracted. Additional details on these UCD⁹ estimation methods and assumptions may be found in Cahill et al.⁹ and Eldred et al.⁵

The estimation factors and assumptions which we will apply to the winter season NESCAUM data are listed in Table III. Several differ slightly from assumptions employed in the previously cited UCD studies, in an attempt to more directly reflect the winter, northeastern aspects of this data set. Specifically, we assume that fine sulfur is divided equally between ammonium sulfate and ammonium bisulfate. Previous UCD estimates assumed ammonium bisulfate for annual average composition at eastern sites, but a somewhat less acidic composition seems appropriate for the winter season. For soil, we simply assume that silicon constitutes 25% of soil. There are several reasons why a more detailed soil estimate was not conducted here, including the fact that Al was frequently below detection limits on the winter NESCAUM filters and also a relative lack of confidence that some of the traditional soil elements like Fe and Ti are in fact soil derived in the Northeast. Soil would be a minor component of this NESCAUM data set under any range of assumptions, and our seasonal estimates using ⁴ Si are generally within 10 nanograms/m³ of the UCD estimates⁸ of NESCAUM Winter soil concentrations reported by Flocchini et al.⁸ For soot we assume an absorption efficiency of 10 m²g⁻¹. We assume that organic matter^{5,10} comprises 70% of remaining mass, and is composed of 9% hydrogen.

When the assumptions listed in Table III are applied to the pooled NESCAUM data for all sites for the winter season, the resulting regional winter apportionment consists of 48% sulfates, 23% organic matter (by both remaining mass and non-sulfate hydrogen methods), 15% soot, 4% soil, and 10% unexplained. If organic matter is composed of 71% carbon (OC), mean winter OC concentrations would be 1.2 ug/m^3 ; winter EC (soot, from optical absorption) would be 1.1 ug/m^3 . These values compare well with previously reported annual 1975 hi-vol measurements of 1.6 ug/m^3 OC and 1.0 ug/m^3 EC from Acadia, ME¹⁰; summer, 1984 fine particle measurements of 1.3 ug/m^3 OC and 0.8 ug/m^3 EC from Lenox, MA¹¹; and winter, 1983 fine particle measurements of 2.4 ug/m^3 OC and 1.1 ug/m^3 EC from Lewes, DE¹².

Estimated winter season compositions for the individual NESCAUM sites (summarized in Table IV) are each within a few percentage points of the regional means. They show excellent agreement between the organic matter estimates by the remaining mass and non-sulfate hydrogen methods - adding confidence to the results of these two fundamentally different approaches. Figure 4 displays estimated daily mass compositions for the Whiteface Mtn., NY and Bridgton, ME sites. Proportionate contributions from estimated sulfate compounds range from about 30% to 70% of fine mass. Daily organic matter mass fractions range from about 10% to 40%, occasionally exceeding sulfate contributions, as on 11/2/88 at the Bridgton, ME site.

These daily estimates, particularly for organic matter, should be viewed with caution. Although it might be anticipated that the remaining mass and non-sulfate hydrogen estimates could produce reasonably consistent (and accurate) estimates for long-term averages, there are a number of reasons to expect these estimates to differ from each other and from actual organic concentrations on a day to day basis. Proportionate contributions of unmeasured species to remaining mass could exhibit considerable short-term variations in response to changing ambient concentrations, and/or to changes in volatile or reactive species on filters during sampling and prior to analysis. Reported values for the organic fraction of remaining mass in western measurement programs have varied from about 50 to 80 percent. The sulfur species composition could also be quite variable on a daily basis (particularly during the warm season in the East). Nevertheless, the daily estimates for the NESCAUM winter season exhibited a surprisingly high daily correspondence between the organic estimates [$r=.77$ for Org (Rem) vs. Org (NSH)].

Figures 5a and b compare the daily estimates of organic matter by remaining mass and non-sulfate hydrogen from the Whiteface Mtn., NY and Bridgton, ME sites. Despite the fundamentally different conceptual approaches to the estimation methods, and the sensitivity of the estimates to uncertainties regarding potential variations in sulfate speciation and in unmeasured components of remaining mass, the daily winter season estimates agree remarkably well, both in terms of absolute magnitude and temporal variation. Note, however, that the fall season estimates at Whiteface Mtn., though similar on average, do not agree as well on a daily basis.

DISCUSSION

It is possible that these estimates of organic matter will prove less useful in the East during warm season conditions when a highly variable sulfate speciation is anticipated. On the other hand, it should be recognized that the org (NSH) estimate is dependent on the sulfate composition on the filter, rather than in the ambient air. No extraordinary precautions are taken during or after sampling to prevent neutralization of acidic species on the filter by gaseous ammonia. So the sulfur-hydrogen relationship on the analyzed filters may be more consistently stable (neutralized) than might otherwise be anticipated. For the future, we look forward to comparing these estimates to the organic (and elemental) carbon measurements from the quartz filters from the Whiteface and Quabbin sites, especially for the summer season.

A potentially interesting variation on the analysis procedure might be deliberate exposure of the filters to ammonia prior to a second hydrogen analysis--in which case, we could be relatively certain of the "non-sulfate" hydrogen concentrations. If methods could also be developed to prevent neutralization of acidic species during sampling and prior to an initial hydrogen analysis, these sampling and analysis procedures may eventually prove useful in determining aerosol acidity concentrations, while at the same time reducing the uncertainties in the organic matter estimates at eastern sites.

It was not intended to draw definitive conclusions from the limited, currently available NESCAUM data and suggested interpretations reported here. The mass composition estimates in particular, are preliminary and based on assumptions which are subject to testing and revision. The objectives of this paper were to formally introduce the data set to the research (and regulatory) communities, to demonstrate the potential utility of this suite of measurement variables in identifying particulate matter composition and source contributions, and to illustrate the utility of Voyager as a management tool for complex multivariate data sets of this nature.

Within a few months, a much larger set of NESCAUM measurements will be available, and will include additional variables (PM-10, EC, OC), enhanced sample frequency (Wed., Sat., 6th day), and additional sample periods (Spring and Summer, 1989). NESCAUM is eager to share this data with any interested parties, and looks forward to sharing the increased understanding of northeastern particulate matter which may result from subsequent interpretations.

ACKNOWLEDGMENTS

The NESCAUM Regional Particle Monitoring Network has only been possible with funding support from EPA Region I, and the in-kind technical support of the Ambient Monitoring Sections from the states of CT, ME, MA, NH, NJ, NY and VT.

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Table I. NESCAUM Regional Particle Monitoring Site Locations.

	Lat.	Long.	Elev.	Start
1. Whiteface Mtn., NY	44°23'	73°51'	2100'	9/17/88
2. Underhill, VT	44°32'	72°52'	1300'	9/14/88
3. Mt. Sunapee, NH	43°19'	72°04'	2700'	12/7/89
4. Bridgton, ME	44°06'	70°44'	728'	9/14/88
5. Quabbin, MA	42°18'	72°20'	1020'	12/3/89
6. Mohawk Mtn., CT	41°50'	73°18'	1500'	9/14/88
7. Ringwood, NJ	41°07'	74°14'	605'	9/14/88

**Table II. Average NESCAUM Winter Fine Particle Concentrations.
(December, 1988 - February, 1989)**

	NY	VT	NH	ME	MA	CT	NJ
Number	23	20	23	16	16	11	14
Mass	5782	7156	6260	7310	7812	8698	8208
LR*	887	944	799	1100	1327	1402	1589
S	768	918	768	915	880	1098	1017
H	264	353	287	349	340	421	388
Na	154	225	171	230	207	233	213
Si	72	74	74	58	72	99	92
K	33	48	39	82	48	51	50
Ca	24	29	24	30	26	35	34
Fe	21	24	24	19	26	34	31
Zn	8.2	11	9.1	9.3	11	13	11
Pb	4.7	5.5	5.3	5.0	5.7	7.4	7.2
Br	2.0	2.7	2.3	2.1	2.4	3.4	3.4

*LR=Optical Absorption in 10^{-8} m^{-1} ; all other values in nanograms m^{-3}

Table III. Factors and Assumptions for Estimating Mass Composition.

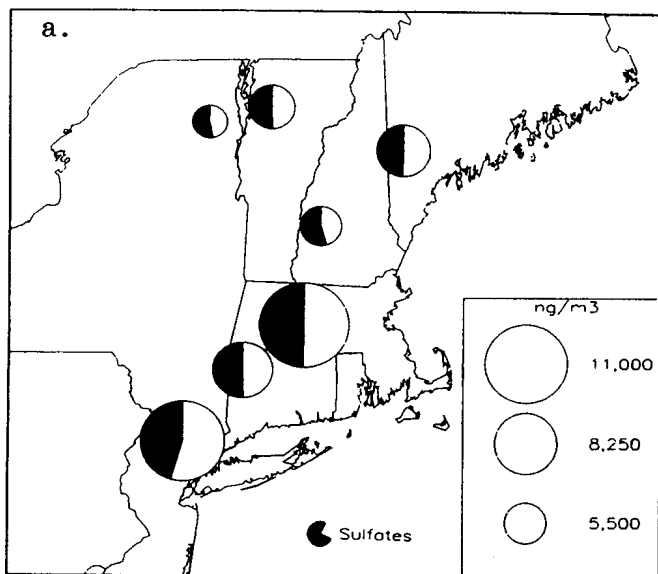
-
-
1. Sulfates = 3.85 S:
 - a. All sulfur is sulfate;
 - b. 50% $(\text{NH}_4)_2\text{SO}_4$ and 50% NH_4HSO_4 .
 2. Soil = 4 Si:
 - a. All silicon is soil derived;
 - b. 25% of soil is silicon.
 3. Soot (ng m^{-3}) = Optical Absorption (10^{-8} m^{-1}):
 - a. Optical absorption caused primarily by soot;
 - b. Soot absorption efficiency is $10 \text{ m}^2 \text{ g}^{-1}$.
 4. Org (Rem) = 0.7 (Fine mass-sulfates-soil-soot):
 - a. Sulfates, soil and soot are reasonable estimates;
 - b. 70% of remaining mass is organic matter.
 5. Org (NSH) = 11(H-0.2S):
 - a. Sulfate speciation as in 1.b. above;
 - b. All non-S Hydrogen is organic;
 - c. Organic composition is 9% hydrogen, 71% carbon.
-
-

**Table IV. Estimated NESCAUM Fine Mass Composition (Winter '88-'89).
(Mean Daily Percentage of Fine Mass \pm Standard Deviation)**

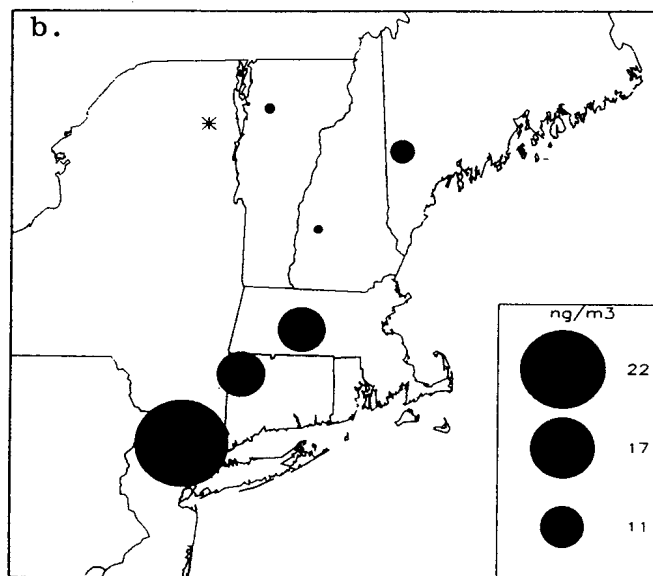
	NY	VT	NH	ME	MA	CT	NJ
Sulfates	51 \pm 7	49 \pm 8	47 \pm 7	48 \pm 7	44 \pm 7	49 \pm 13	48 \pm 9
Soot	15 \pm 2	14 \pm 3	13 \pm 2	15 \pm 3	16 \pm 3	16 \pm 3	19 \pm 5
Soil	5 \pm 2	4 \pm 2	4 \pm 1	3 \pm 1	4 \pm 1	4 \pm 2	5 \pm 2
Org (Rem)	21 \pm 4	24 \pm 6	22 \pm 4	24 \pm 4	22 \pm 5	25 \pm 5	24 \pm 4
Org (NSH)	21 \pm 4	24 \pm 6	23 \pm 4	24 \pm 4	22 \pm 6	25 \pm 5	24 \pm 4
Other?	7 \pm 7	9 \pm 11	13 \pm 6	10 \pm 7	15 \pm 12	7 \pm 12	4 \pm 12

Figure 1. Fine Mass, Sulfates and Trace Elements on February 18, 1989

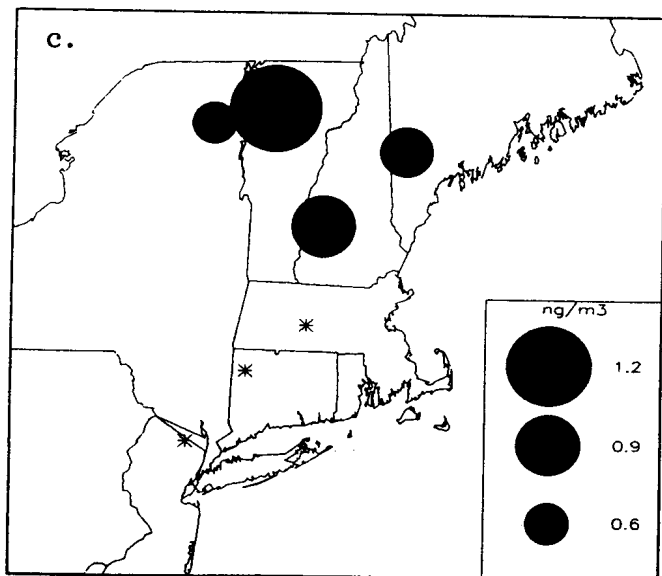
Fine Mass and Sulfates, 2/18/89



Fine Particle Vanadium, 2/18/89



Fine Particle Arsenic, 2/18/89



Fine Particle Potassium, 2/18/89

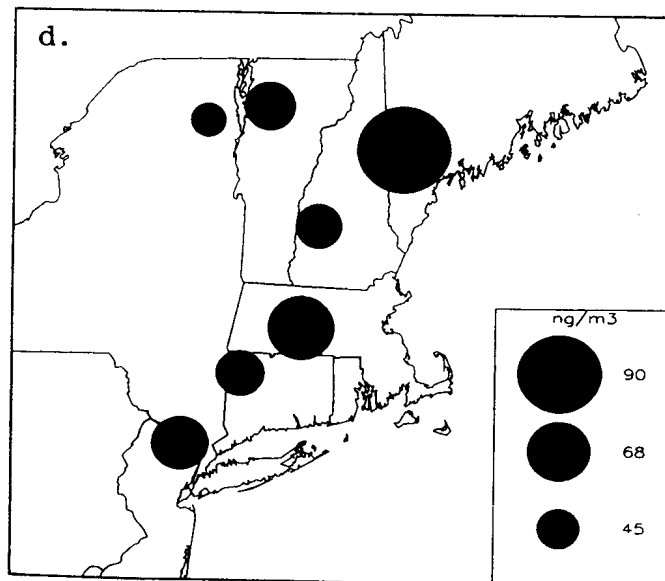
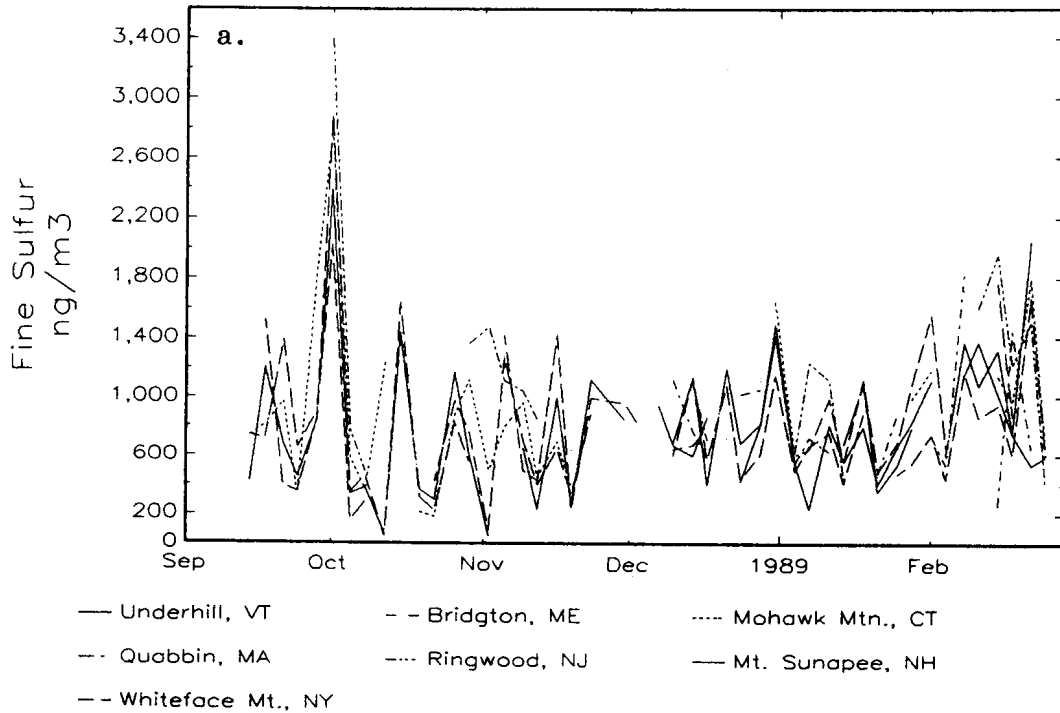


Figure 2. NESCAUM Regional Sulfur and Mass Concentrations (9/88-2/89)

Fine Sulfur Concentrations at All NESCAUM Sites
September, 1988 – February, 1989



Fine Mass Concentrations at All NESCAUM Sites
September, 1988 – February, 1989

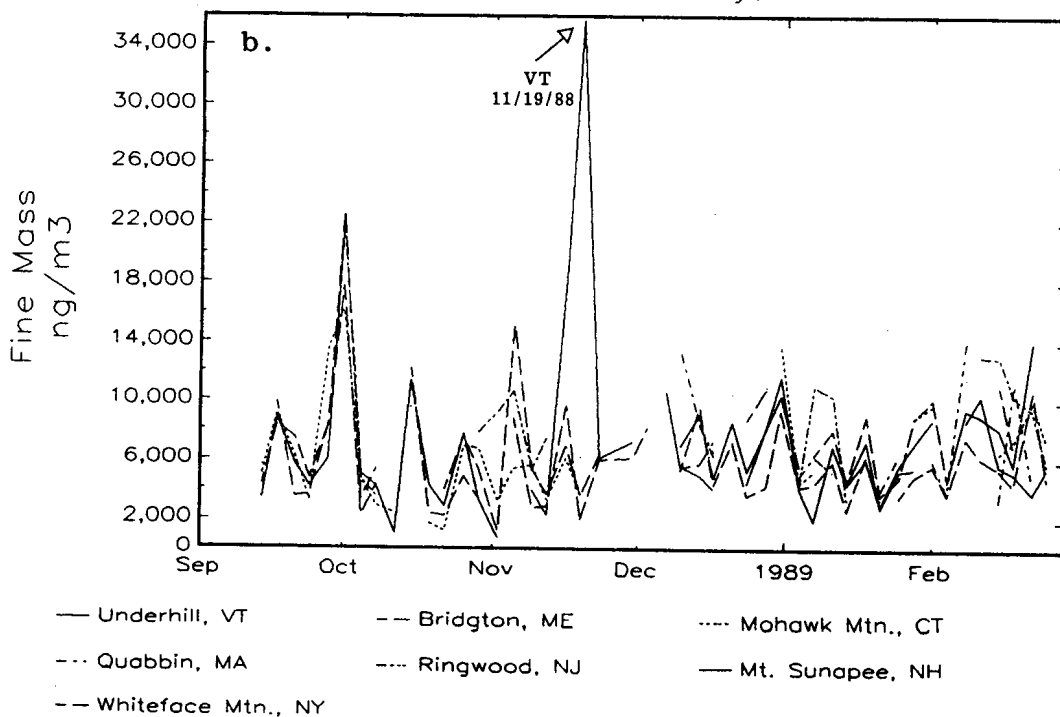


Figure 3. NESCAUM Mass, Sulfur, Hydrogen Relationships (9/88-2/89)

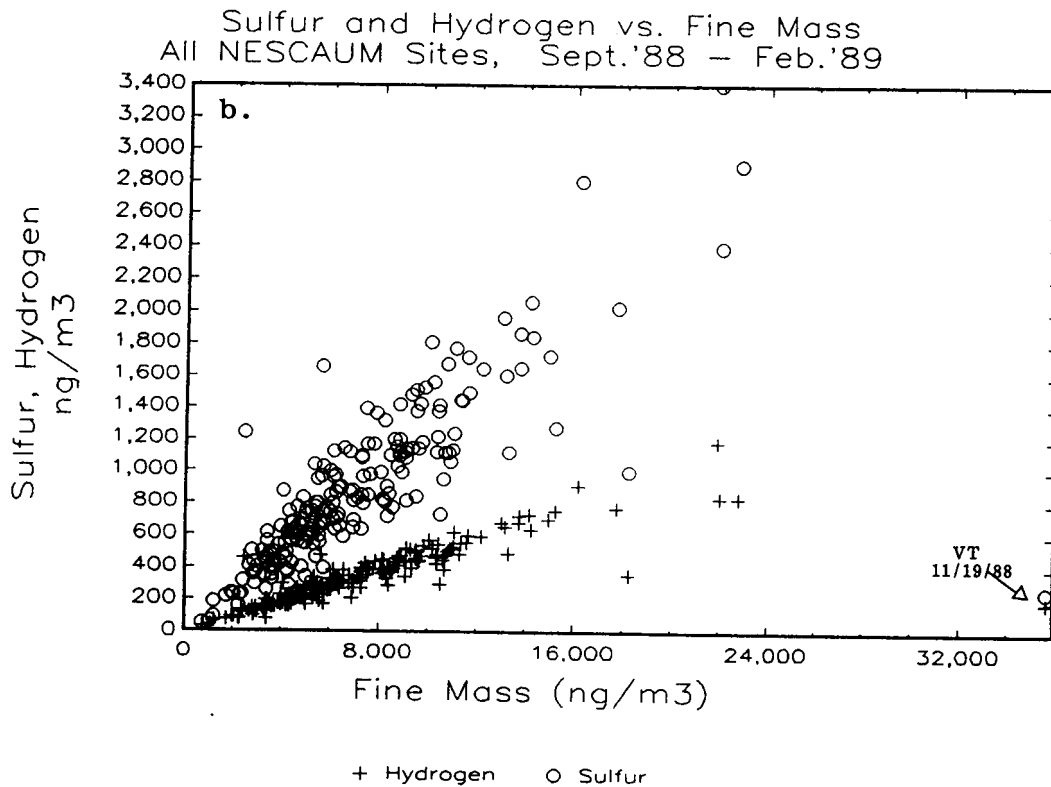
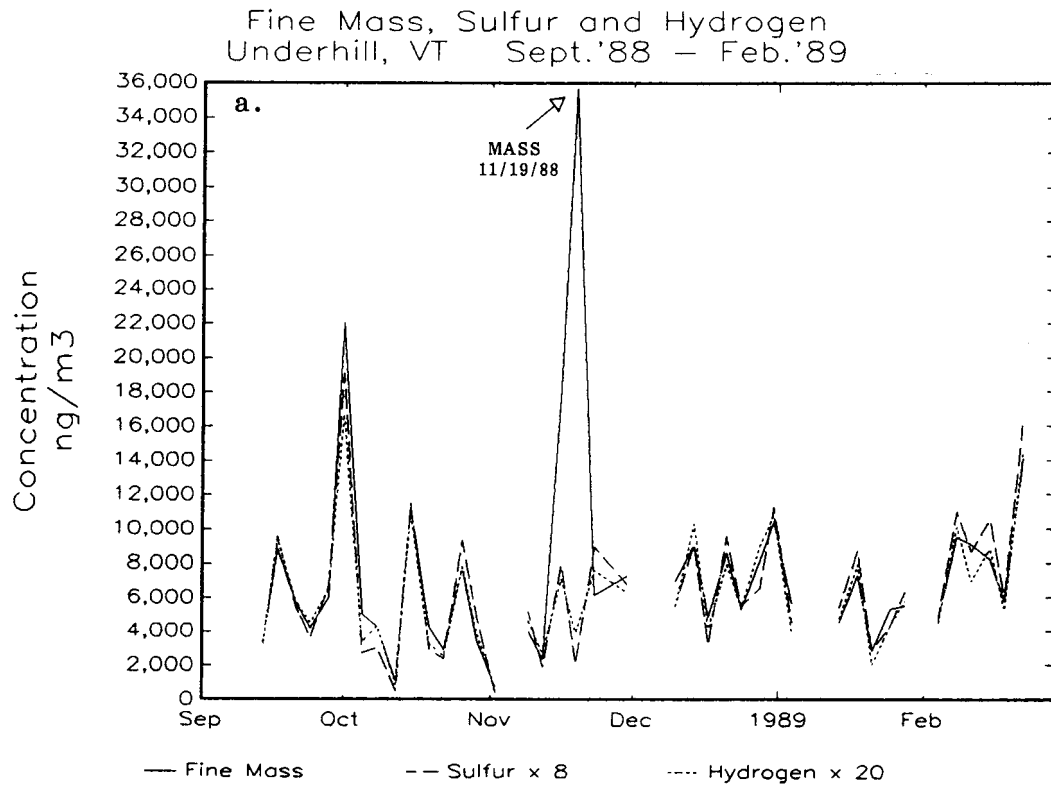
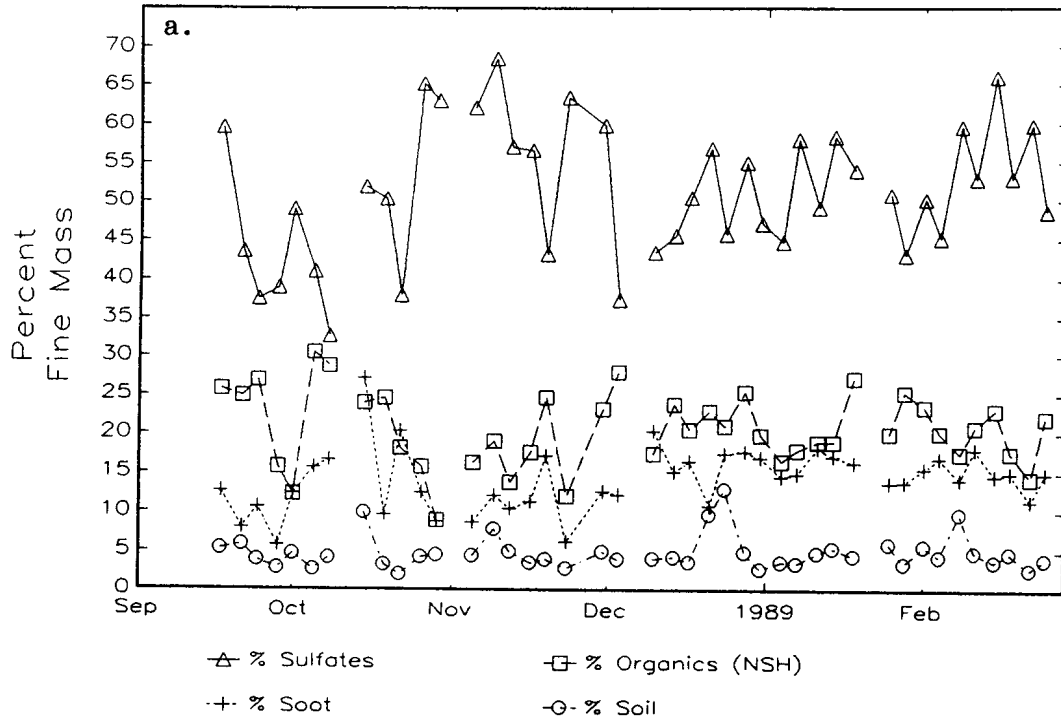


Figure 4. Estimated Daily Mass Composition: Whiteface Mtn. and Bridgton, Me.

Estimated Fine Mass Composition, Whiteface Mtn., NY
September, 1988 – February, 1989



Estimated Fine Mass Composition, Bridgton, ME
September, 1988 – February, 1989

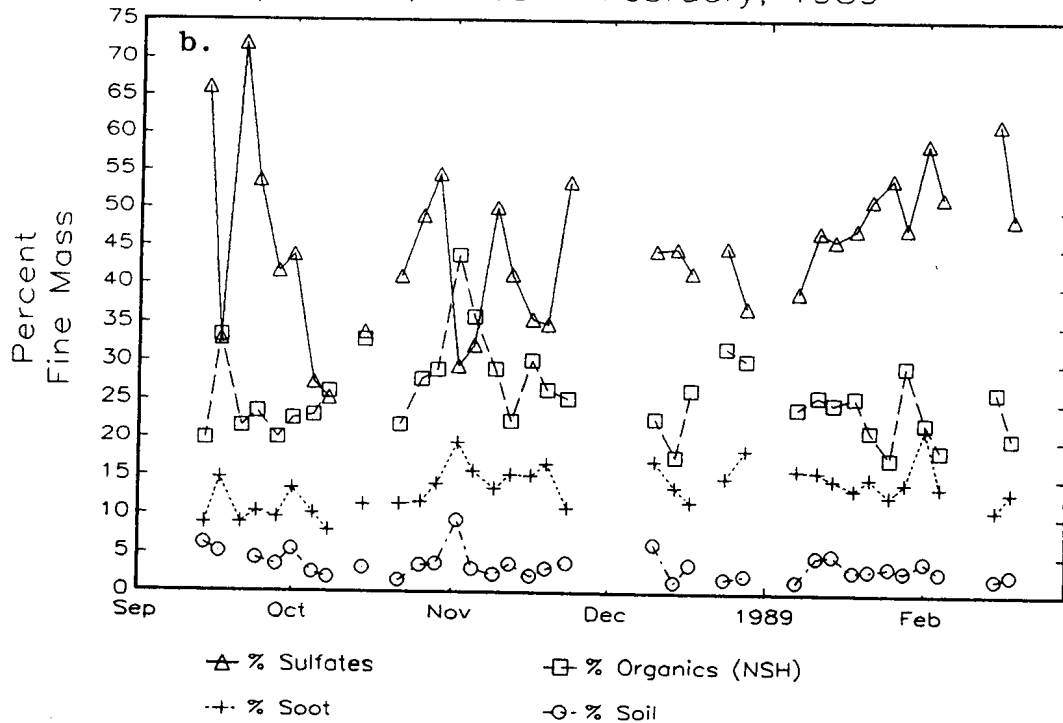


Figure 5. Organic Matter Estimates By Remaining Mass and Non-S Hydrogen

