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Determination of Fine Particle and Coarse Particle Concentrations and Chemical Composition in the Northeastern United States, 1995

Prepared by

Glen R. Cass

Principal Investigator

California Institute of Technology

Lynn G. Salmon

California Institute of Technology

Daniel U. Pedersen and John l. Durant

Massachusetts Institute of Technology

Ray Bigg, Alex Lunts, and Mark Utell

University of Rochester Medical Center

Project Manager

Praveen K. Amar

Director, Science and Policy

Northeast States for Coordinated Air Use Management

Editor

Ingrid M. Ulbrich

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Lynn G. Salmon and Glen R. Cass

Environmental Engineering Science Department

California Institute of Technology

Pasadena, CA   91125

Daniel U. Pedersen and John L. Durant

Department of Civil and Environmental Engineering

Massachusetts Institute of Technology

Cambridge, MA   02139

Ray Gibb, Alex Lunts, and Mark Utell

University of Rochester Medical Center

Rochester, NY   14642

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1 INTRODUCTION

An air monitoring network was operated at five sites in the Northeastern United States throughout the year 1995. The concentrations and chemical composition of airborne particles were measured using filter samples to characterize the major chemical substances in the aerosol mixture, including sulfates, nitrates, ammonium ion, total organic carbon, elemental carbon, and certain trace metals. The sampling methods used separate the particles measured into two size ranges: fine (less than 2.2 μm in diameter) and coarse (greater than 2.2 μm in diameter).

2 EXPERIMENTAL PROCEDURES

2.1 Air Monitoring Network

During the calendar year 1995, a monitoring network designed to measure the concentration of atmospheric particulate matter was operated at five sampling sites located in the Northeastern United States: two sites in and near Rochester, New York, one rural site in central Massachusetts, and two sites in and near Boston, Massachusetts. A map of the Northeastern US showing the sampling site locations is given in Figure 2.1. The sites were selected so that particle concentrations at two city centers could be compared to those in adjacent rural areas along a west to east transect running from the Great Lakes to the Atlantic Ocean. The three sites chosen in Massachusetts included an urban site located at Kenmore Square in Boston, a suburban location at Reading in the Boston suburbs, and a rural location at Quabbin Reservoir. The Kenmore Square monitoring station was located in a commercial district near the campus of Boston University, approximately one block from the Massachusetts Turnpike. The Reading station was on the roof of the Municipal
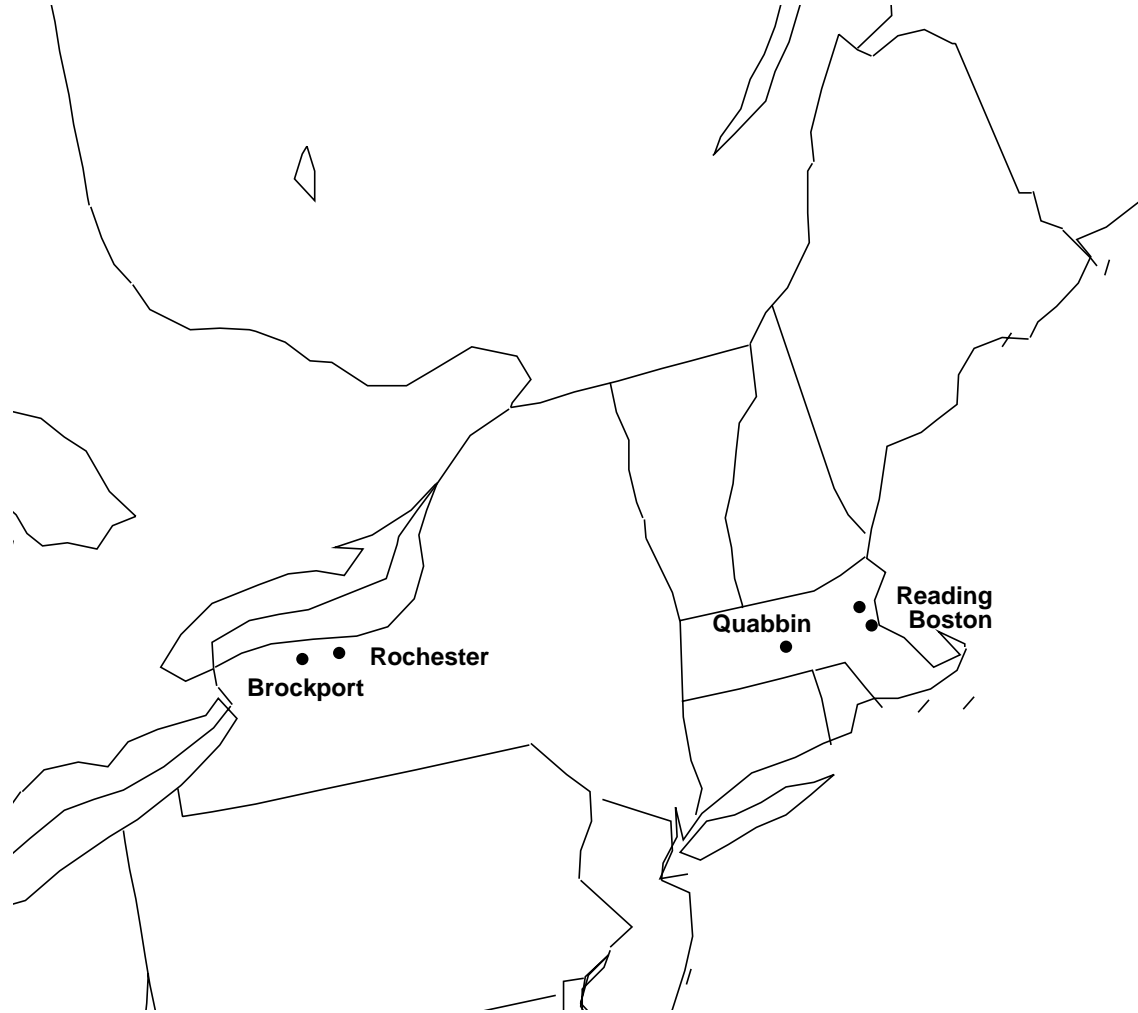
Figure 2.1 - Map of the Northeastern US showing 1995 sampling sites

Light Department office building in a largely residential area but within sight of a railroad right-of-way and a fast food restaurant. The Quabbin Reservoir site was located within a nearly unpopulated protected watershed in central Massachusetts and served as a regional background site that defines the contaminant levels present in a rural location. In New York state, an urban monitoring site was chosen in downtown Rochester (fire station site), and a regional background site outside the city was chosen on the State University of New York (SUNY) Brockport campus. The SUNY Brockport site was located on the roof of a campus building with residences and rural countryside in sight.

Samples were collected every sixth day for 24-hour sampling periods (12 am to 12 am) during the calendar year 1995. The first sample was collected on January 3 to coordinate the timing of this sampling network with the sampling schedule of the national air surveillance network.

2.2   Sampler Design and Sampling Protocol

The sampling system used during this experiment has been described previously (1-3) and is only briefly summarized here. The ambient samplers measured airborne particle concentrations and chemical composition in two size ranges: fine particles (diameter, $d_p \leq 2.2\ \mu$m) and total particles (no size discrimination). Coarse particle concentrations ($d_p > 2.2\ \mu$m) were calculated by subtracting the fine particle concentrations from the total particle concentrations. In each particle size range, samples were taken simultaneously and in parallel on three 47mm diameter filter substrates – one pre-baked quartz fiber filter (Pallflex 2500 QAO) and two polytetrafluoroethylene (PTFE) membrane filters (Gelman Teflo). The filter substrates used to collect particulate matter were chosen to be compatible with subsequent particular chemical analyses. The combination of measurements made

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on the quartz fiber and PTFE filters allows for a nearly complete material balance on the chemical composition of the particles (1), as described in subsequent sections of this report.

A schematic diagram of the sampler used is shown in Figure 2.2. In the fine particle portion of the sampler system, air was pulled at a nominal flow rate of 25 liters per minute (lpm) through an Air and Industrial Hygiene Laboratory (AIHL)-design cyclone separator which, when operated at a flow rate of 25 lpm, removed coarse particles with diameters larger than 2.2 μm (4). Total particles in all size ranges were collected by sampling directly from ambient air onto three open-face filter holder assemblies that were protected from particle deposition by a fallout shield overhead. The flow rate through each filter holder was controlled by a critical orifice. Flow rates were measured each time samples were loaded, and again when samples were unloaded to obtain the volume of air sampled for each sampling event.

Four sampling lines (D, E, G and H; Figure 2.2) collected fine particles or total particles on Teflon filters for subsequent chemical analysis as discussed below. One Teflon filter of each pair was used for mass plus ionic species determination by ion chromatography and the second Teflon filter of each pair was used for mass and trace elements determination by X-ray fluorescence (XRF) analysis. The remaining filter holders (C and F, Figure 2.2) were used to collect particles on quartz fiber filters from which carbonaceous species were determined by thermal evolution and combustion analysis.

2.3   Sample Analysis

Particle mass. PTFE filters used for total particle collection were Gelman Teflo, 2.0 μm pore size. Fine particle samples were collected on Gelman Teflo, 1.0 μm pore size PTFE filters. Atmospheric particle mass concentrations were measured gravimetrically by

TOTAL PARTICLE SAMPLER

OPEN FACE
FILTER
HOLDERS

QUARTZ FILTER	TEFLON FILTER	TEFLON FILTER

F 10 LPM   G   5 LPM   H   3 LPM

→ VACUUM PUMP

FINE PARTICLE SAMPLER

INLET
25 LPM

CYCLONE
SEPARATOR
removes particles
> 2.2µm in
diameter

QUARTZ FILTER	TEFLON FILTER	TEFLON FILTER

C 10 LPM   D 10 LPM   E   5 LPM

→ VACUUM PUMP
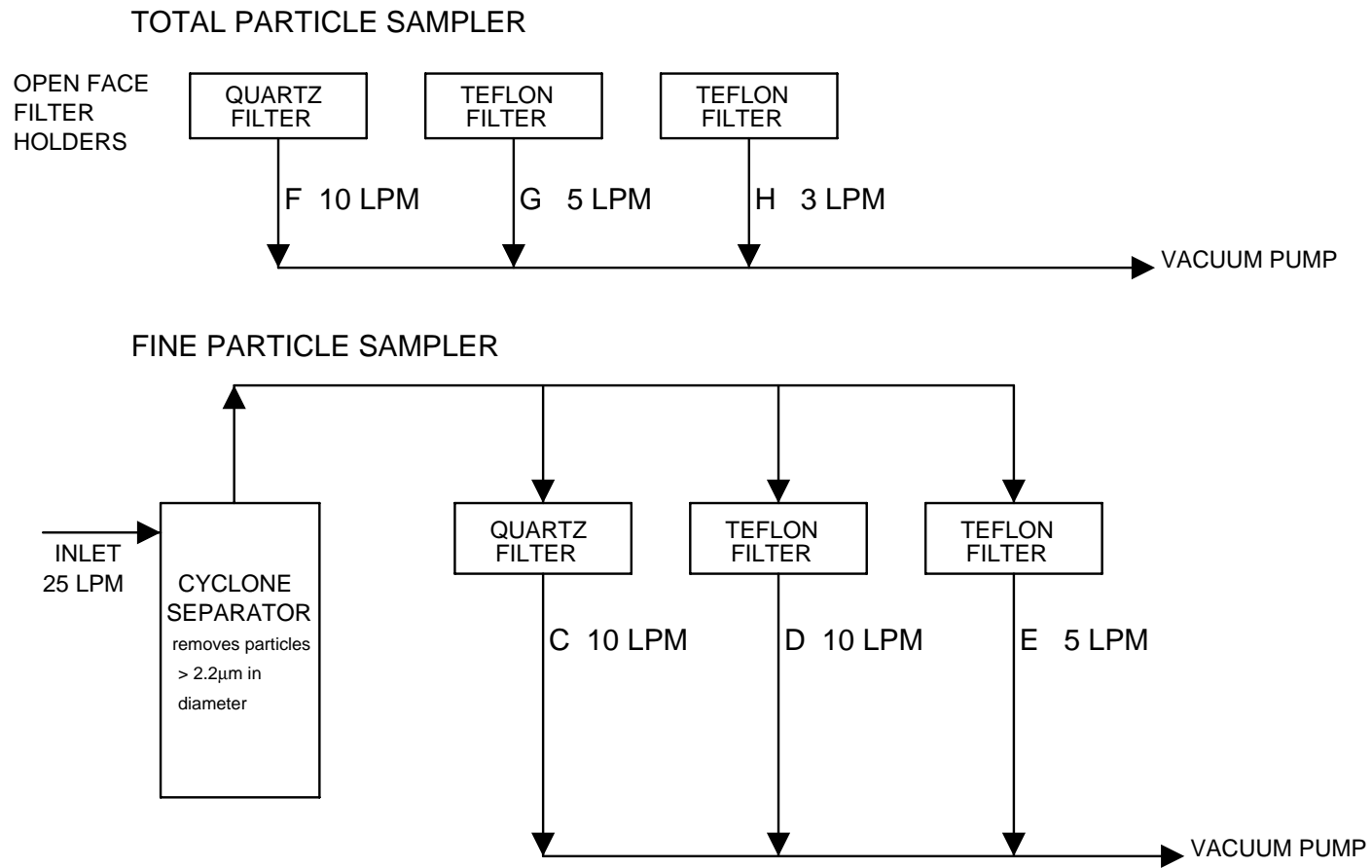
Figure 2.2 - Schematic Diagram of Fine and Total Particle Samplers

weighing each PTFE filter at least twice before and twice after sample collection using a mechanical microgram balance (Model M-5S-A, Mettler Instruments). Unexposed and collected PTFE filters were equilibrated at 21 ± 1° C and 40 ± 3 percent relative humidity for at least 24 hours prior to weighing each filter. To track the calibration of the balance between initial and final weighings, a set of control filters was weighed during each daily weighing period. High precision metal calibration weights also were weighed periodically to check the performance of the balance.

Filter extraction. PTFE filters first were placed in individual extraction cups and then were wetted with 0.2-0.25 ml of ethanol (100 percent) to reduce the hydrophobic nature of this material (5). A Teflon rod was placed on top of each filter to keep it submerged, the extraction cup was sealed with a tight-fitting lid, and then each PTFE filter was extracted by shaking it in a known volume (10-20 ml) of distilled, deionized water for 3 hours or more.

Ionic aerosol species. After extraction, the concentrations of the major water-soluble particulate species (SO_4^{-2}, NO_3^-, and Cl^-) were determined using a Dionex model 2020i ion chromatograph (6,7). The same PTFE filter extracts also were analyzed for particulate ammonium ion (NH_4^+) by an indophenol colorimetric procedure employing a rapid flow analyzer (RFA-300 TM, Alpkem Corp.) (8,9).

It is important to note that the use of PTFE filters for the collection of particulate matter results in a lower limit determination of atmospheric aerosol nitrate concentrations. This negative artifact for aerosol nitrate has been well documented and is most likely due to the vaporization during sampling of a portion of the fine particle NH_4NO_3 from the inert PTFE filter substrate (10–14).

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Organic and elemental carbon. Organic carbon (OC) and elemental carbon (EC) concentrations in fine aerosols were determined from the quartz fiber filters by the thermal-optical method of Birch and Cary (15). Prior to sample collection these filters were heat treated at 550° C in air for at least 8 hours to lower their carbon blank levels. The separate determination of organic and elemental carbon is important because of the effect that elemental carbon can have on atmospheric light absorption and visibility degradation.

Trace elements. The bulk concentrations of 38 major and minor trace elements were measured by X-ray fluorescence (16,17). The species sought were Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Au, Hg, Tl, Pb, and U. Since many of these elements are rare, their concentrations are often below detection limits in the samples.

2.4   Quality Assurance/Quality Control

Field sampling. Samples remained in the field for as short a time period as possible (i.e., they were installed the day before and removed the day after sample collection). All filters were stored in pre-labeled, petri dishes sealed with Teflon tape prior to sample collection. Quartz fiber filters were individually packaged in petri dishes lined with annealed aluminum foil prior to use. After sample collection, filters were placed back into their original pre-labeled petri dish, sealed with Teflon tape, refrigerated until returned to the laboratory, and then frozen at -21° C until sample analysis. Cold storage is employed to prevent the loss of semi-volatile particle-phase species such as ammonium nitrate and certain organic compounds.

Air flow rates through all filter trains were measured before and after sample collection to ensure that the filter holders were not leaking and that the filters did not become

overloaded with particles. Each system had a 24-hour, seven-day on/off timer along with a separate elapsed time indicator. All field data were immediately entered into a field log book at the site when the measurement was obtained. The inlets to the samplers were protected from the sun and from wet or dry fallout.

Chemical analysis. The concentrations of all chemical species analyzed by ion chromatography were determined relative to primary or secondary laboratory standards of known concentrations. Aqueous daily standards were diluted from more concentrated solutions prepared from American Chemical Society (ACS) grade analytical reagents. Whenever possible, the matrix of the daily standards matched that of the leaching solution. Standard log sheets were filled out each time standards or reagents were prepared.

A summary of the instrument detection limits (IDL) and filter blank values for the major species is presented in Table 2.1. The detection limits for the X-ray fluorescence data were supplied by the analytical laboratory performing those analyses (Desert Research Institute, Reno, Nevada). For gravimetric mass determination, the reproducibility of the balance was determined by making a large number (n > 500) of replicate weighings over the course of the experiment. The precision for each weighing was found to be $\pm\,11.2\,\mu$g per filter. The initial and final weighing errors were combined to obtain the precision for sample mass determination. Final error bound estimates were obtained by the statistical propagation of the sample, filter blank, and sampling volume precisions. These error bounds are included in the full data set available from NESCAUM.

Table 2.1. Summary of detection limits and filter blank values for chemical composition determination of particle species.

Species Determined[a]	Instrument Detection Limit (μg/filter)	Filter Type	Filter Blank (μg/filter)	
			Fines	Totals
organic carbon	2.0	quartz	0.25 ± 0.21^c	0.56 ± 0.37^c
elemental carbon	2.0	quartz	0.11 ± 0.16^c	0.26 ± 0.18^c
sulfate	0.4	PTFE	$-^b$	0.25 ± 0.23
nitrate	0.4	PTFE	0.12 ± 0.21	$-^b$
chloride	0.6	PTFE	0.11 ± 0.16	$-^b$
ammonium	0.2	PTFE	$-^b$	$-^b$
sodium	0.4	PTFE	$-^b$	$-^b$
magnesium	0.4	PTFE	$-^b$	$-^e$

Species determined by X-ray fluorescence[d]:

Al	0.091	PTFE	$-^b$	$-^b$
Si	0.057	PTFE	$-^b$	$-^b$
Fe	0.014	PTFE	$-^b$	$-^b$
Ca	0.041	PTFE	$-^b$	$-^b$
S	0.045	PTFE	$-^b$	$-^b$
K	0.055	PTFE	$-^b$	$-^b$
Ti	0.026	PTFE	$-^b$	$-^b$
Cr	0.017	PTFE	$-^b$	$-^b$
Mn	0.015	PTFE	$-^b$	$-^b$

a. Water soluble fraction only for sulfate, nitrate, chloride, ammonium, sodium, and magnesium.

b. Much less than instrument detection limit.

c. Blank value units are μg/cm^2

d. Other trace species determined by XRF: P, Cl, V, Cu, Zn, Ga, As, Co, Ni, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Au, Hg, Tl, Pb and U.

e. Not analyzed.

3 SUMMARY OF AIRBORNE PARTICLE CONCENTRATIONS

Airborne particle concentration data are summarized in graphical form, and the complete data set is available by request from NESCAUM. Annual average coarse and fine particle concentrations subdivided by chemical composition at each site are shown in Figures 3.1–3.5. These data are also presented in Table 3.1. The organic aerosol concentrations shown in the pie charts of Figures 3.1 through 3.5 are estimated as equal to 1.4 times the mass of organic carbon (OC) measured in order to account for the hydrogen, oxygen and nitrogen present in organic compounds. The concentrations of crustal materials derived from soil and rock dust are estimated by converting the elements Si, Al, Fe, Ti, Mn, Ca, and K to their common oxides (i.e., SiO_2, Al_2O_3, Fe_2O_3, TiO_2, Mn_2O_7, CaO, and K_2O) and then summing the concentrations.

The "other" category of material shown in Figures 3.1-3.5 represents the difference between gravimetrically determined mass concentrations and the sum of the masses of chemical species measured. The "other" material can consist of water retained in the samples despite desiccation, as well as contributors to crustal material other than the major crustal oxides, and possibly some organic matter if the presence of highly oxygenated organic compounds leads to a ratio of organic compounds to organic carbon mass that is greater than the estimated value of 1.4. There is no routine method for aerosol water measurement; GC/MS analysis of the organics would be needed to identify a more accurate organic compounds to organic carbon ratio. Analysis of local soils could improve the trace elements to crustal mass conversion. No "other" material appears in the fine particle graph of Figure 3.1 at Kenmore Square. The aerosol mass at that site is slightly overbalanced by the sum of the measured chemical species, possibly due to organic vapor pick-up by the quartz fiber filters in this area with higher motor vehicle traffic.

Coarse
41.3 μg m^{-3}

Organics

Other

EC

Sulfate

Nitrate

Chloride

Trace

Ammonium

Crustal

Fine
16.2 μg m^{-3}

Trace

Crustal

Ammonium

Chloride

Nitrate

Organics

Sulfate

EC
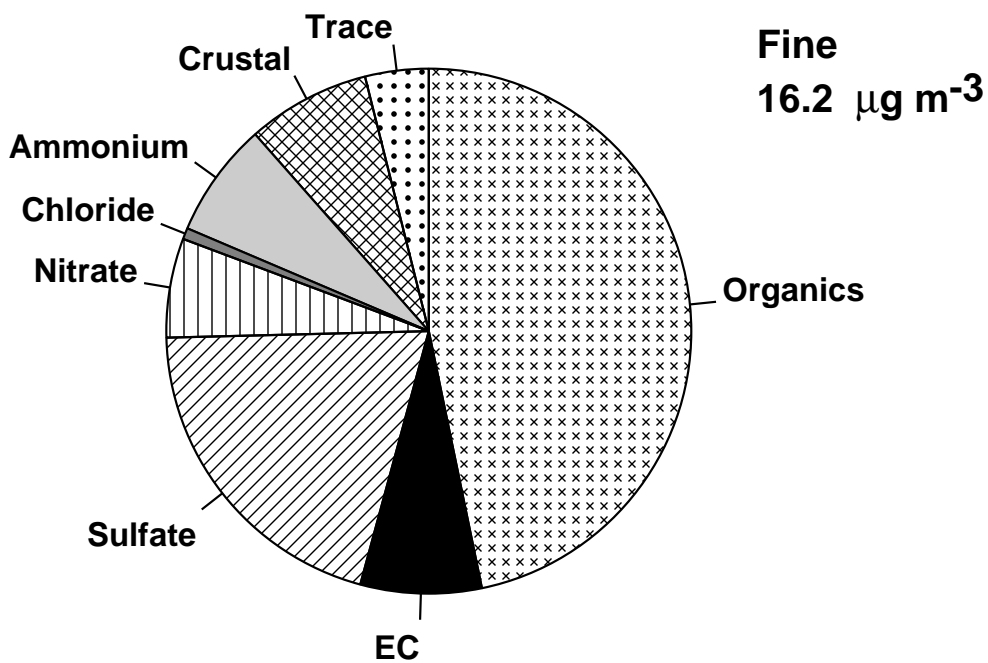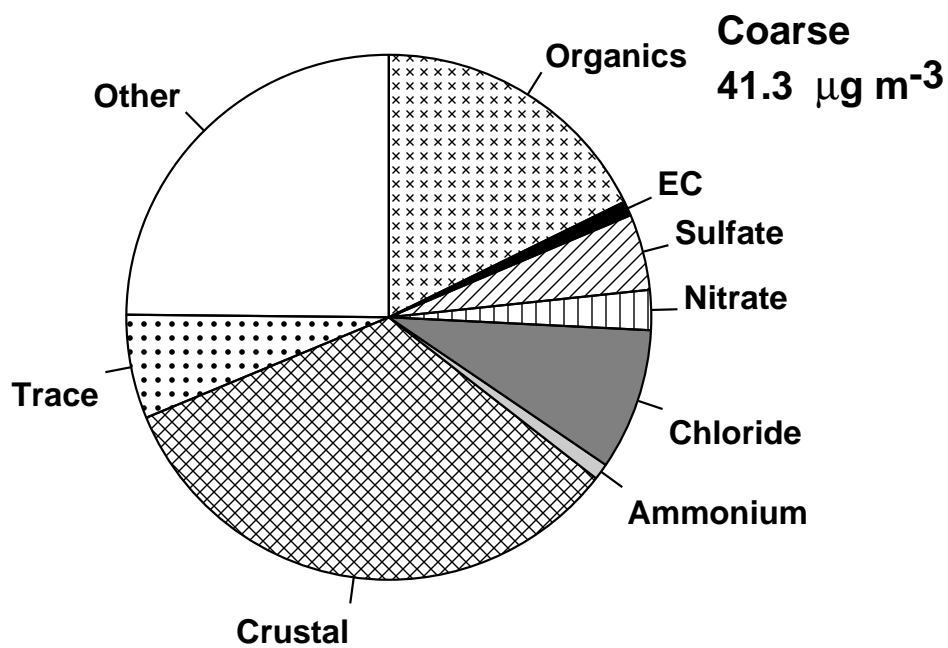
Figure 3.1 - Coarse and Fine Particle Chemical Composition, 1995 Annual Average - Kenmore Square, Boston, MA
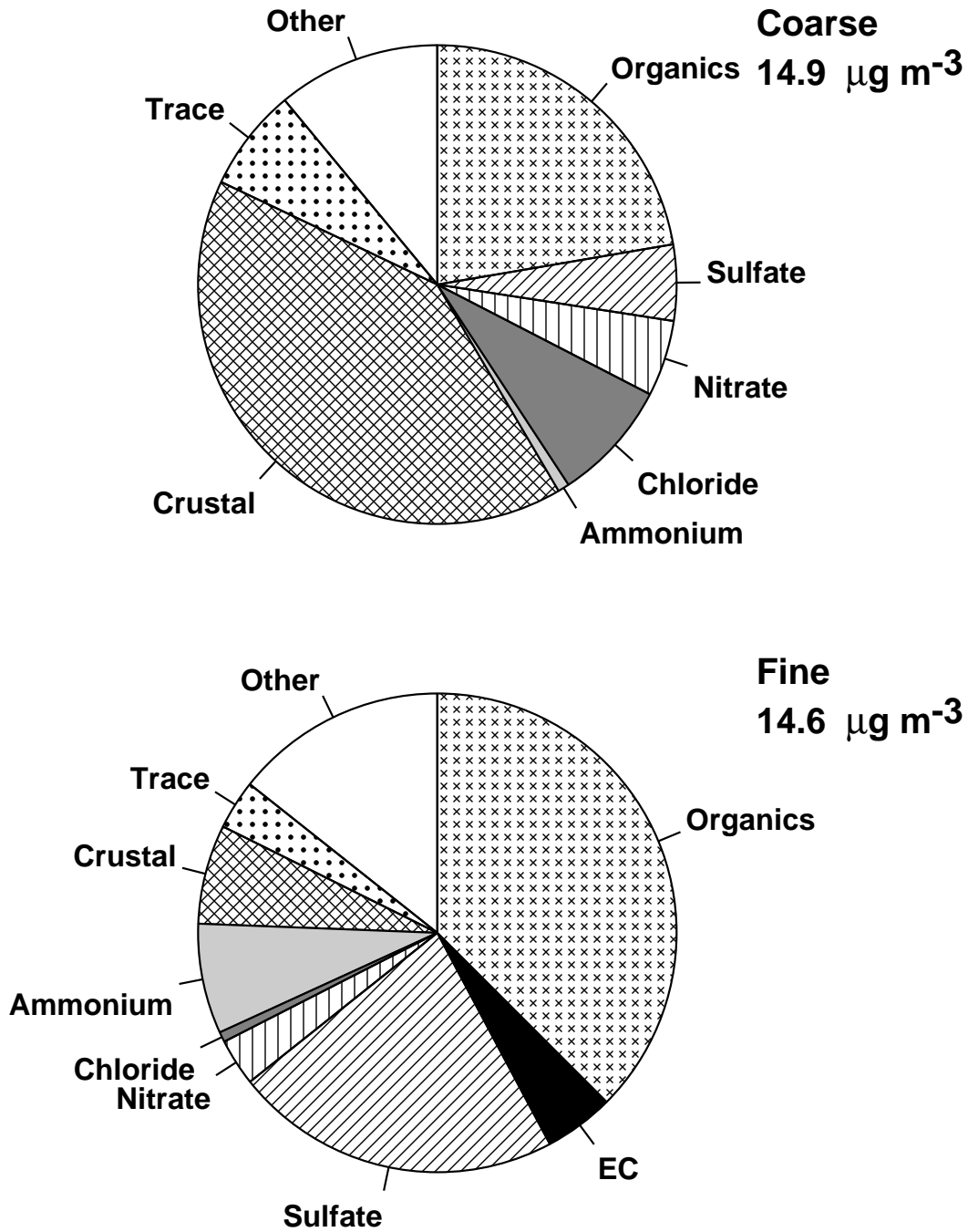
Figure 3.2 - Coarse and Fine Particle Chemical Composition, 1995 Annual Average -- Reading, MA

Coarse
12.1 μg m^{-3}

Organics

Other

Sulfate

Trace

Chloride

Nitrate

Ammonium

Crustal

Fine
12.4 μg m^{-3}

Other

Organics

Trace

Crustal

EC

Ammonium

Chloride

Nitrate

Sulfate
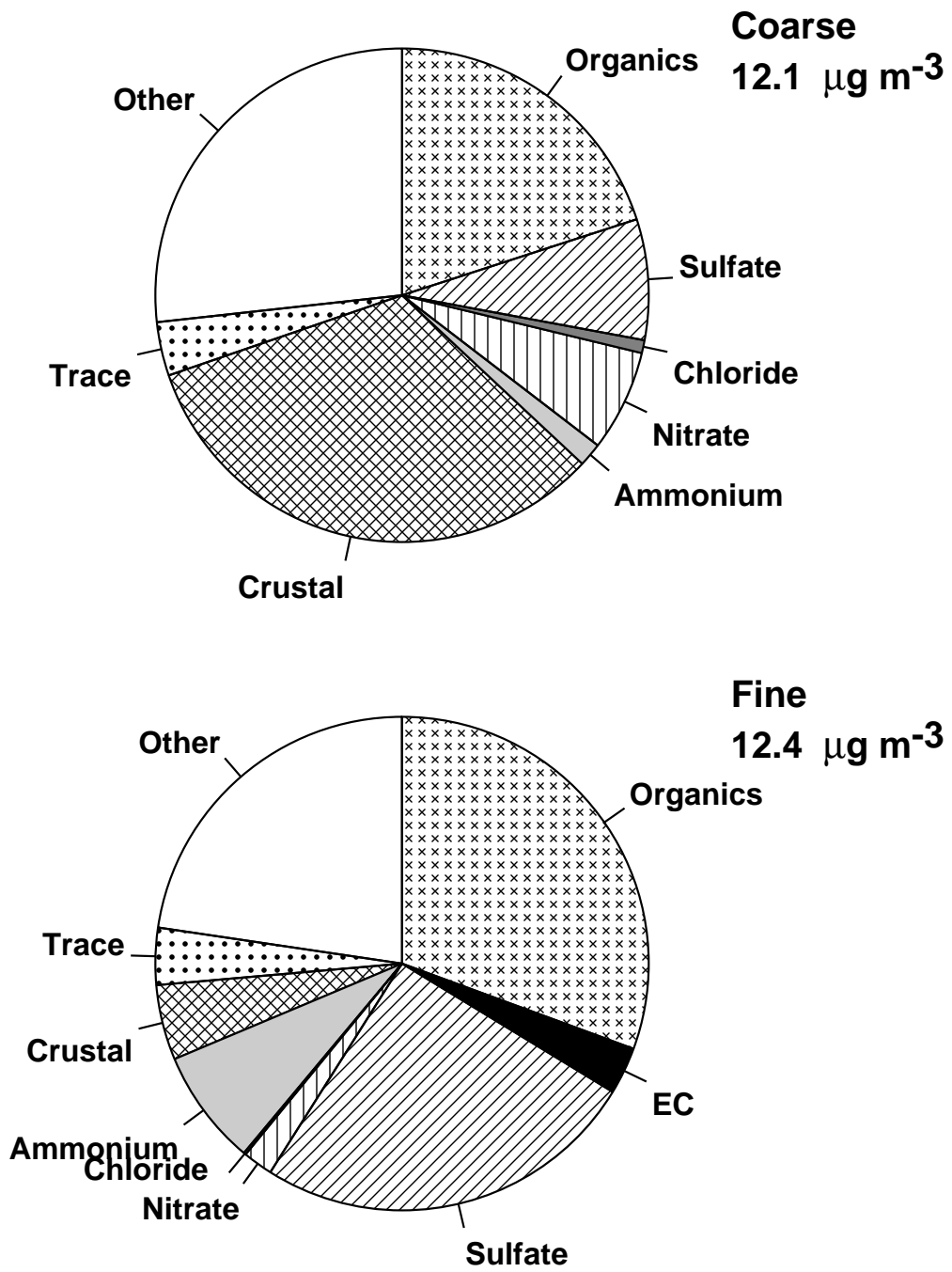
Figure 3.3 - Coarse and Fine Particle Chemical Composition, 1995 Annual Average - Quabbin Reservoir, MA
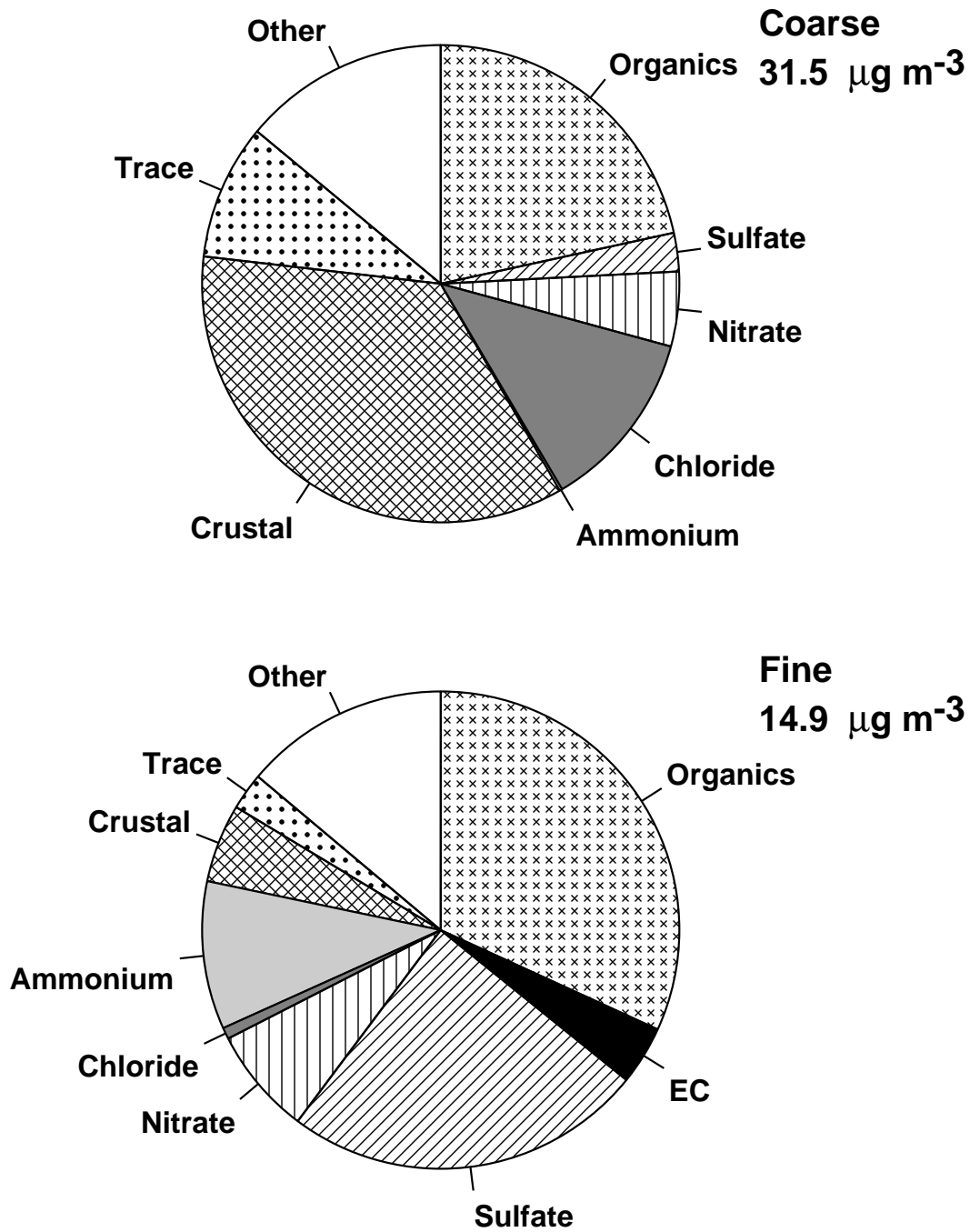
Figure 3.4 - Coarse and Fine Particle Chemical Composition, 1995 Annual Average -- Rochester, NY

Coarse
16.6 μg m^{-3}

Other

Organics

Trace

Sulfate

Nitrate

Chloride

Crustal

Fine
12.8 μg m^{-3}

Other

Trace

Crustal

Organics

Ammonium

EC

Chloride

Nitrate

Sulfate
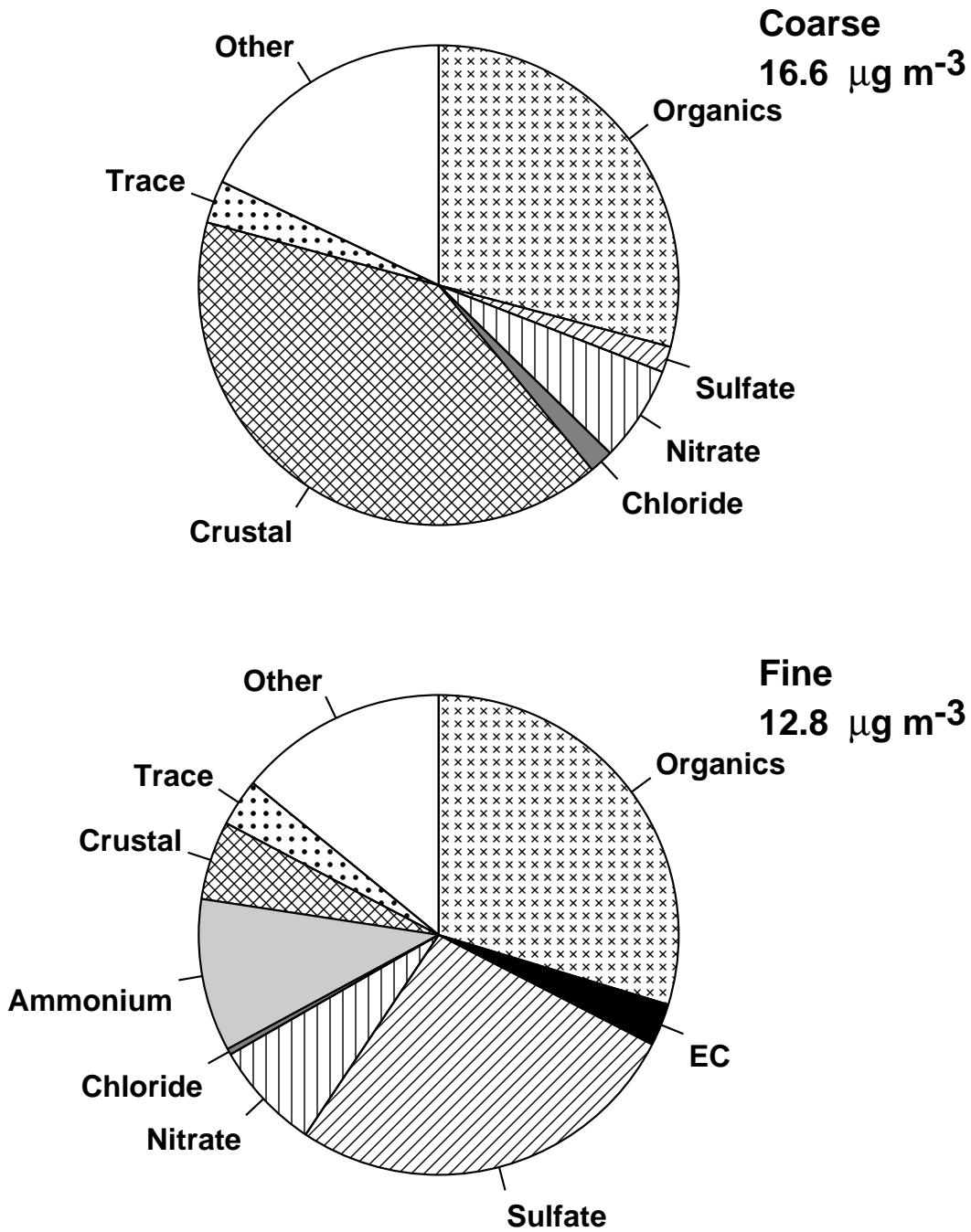
**Figure 3.5 - Coarse and Fine Particle Chemical Composition,
1995 Annual Average -- Brockport, NY**

Table 3.1. Summary of 1995 annual average chemical composition of fine and coarse particle species (given as percent of mass concentration).

	Organics	EC	Sulfate	Nitrate	Chloride	NH$_4$	Crustal	Trace	Other
Kenmore Square:									
fine	47	7	20	6	1	7	8	4	0
coarse	18	1	5	3	9	1	33	6	25
Reading:									
fine	38	5	22	3	1	7	7	3	14
coarse	22	0	5	5	8	1	41	7	11
Quabbin Reservoir:									
fine	31	3	25	2	0	8	5	4	23
coarse	20	0	8	7	1	2	33	4	27
Rochester:									
fine	32	4	24	7	1	10	5	2	14
coarse	22	0	3	5	12	0	35	9	14
Brockport:									
fine	30	3	27	7	0	10	5	3	14
coarse	29	0	2	7	2	0	40	3	18

Time series graphs of the individual 24-h average fine and total particle concentrations are given in Figures 3.6–3.10 along with the coarse particle concentrations determined by subtracting the fine particle concentration from the total particle concentration.

Time series graphs for major individual chemical species are also shown. Figures 3.11a–3.15a and 3.11b–3.15b show the daily time series of fine and total organic species as well as the time series of elemental carbon particle concentrations. Organic carbon data in these figures have been multiplied by 1.4 to provide an estimate of organic compounds mass. The difference between total organics and fine organics is shown as coarse organic

species. Similar data are also shown for coarse elemental carbon species. The coarse particle concentration was set to zero in a few cases where the total concentration was less than the fine concentration in these and subsequent graphs.

Time series plots of fine and total particulate sulfate concentrations measured by ion chromatography (and coarse sulfate by subtraction) are shown in Figures 3.16–3.20. Shown in Figures 3.21 and 3.22 are a comparison of sulfate concentration (molecular weight 96) measured by ion chromatography with three times the sulfur concentration (molecular weight 32) measured by X-ray fluorescence, which serves to demonstrate the equivalence of the work done in the two analytical laboratories used in this study. Figure 3.21 depicts the comparison for fine particle filter samples, while Figure 3.22 shows the same comparison for the total particle data. Fine particle concentrations are more easily measured by XRF than is the case for total particles because total particle measurements require correction for reabsorption of X-rays by the larger particles.

Other species measured by ion chromatography are chloride (Figures 3.23–3.27) and nitrate (Figures 3.28–3.32). Each graph shows time series of fine and total particle concentrations measured at each site with coarse particle concentrations calculated by difference.

Figures 3.33–3.37 show measured soil dust (crustal oxides) concentrations for fine and total particles, and for coarse particles by subtraction. The soil dust concentration is estimated by converting the elements Si, Al, Fe, Ti, Mn, Ca, and K to their common oxides, as described earlier, and then summing the concentrations.

Finally, chemcically speciated monthly average fine and total particle concentrations are shown in Figures 3.38–3.47. In addition, the data is given in Tables 3.2 and 3.3.

4 DISCUSSION

Annual average fine particle concentrations in the Northeastern United States across all of the locations studied are close to the new annual average national ambient air quality standard of 15 μg m^{-3}. At the most rural sites examined, Brockport, NY, and Quabbin Reservoir, MA, the annual average fine particle concentrations were 12.8 and 12.4 μg m^{-3}, respectively, in 1995. These compare to annual average fine particle concentrations of 16.2 μg m^{-3} in downtown Boston and 14.9 μg m^{-3} in downtown Rochester. Some caution must be exercised when comparing these results to the new national ambient air quality standard for fine particles because the measurements in some cases are very close to the standard. Small differences exist between the 2.2 μm particle size cut employed in the present experiments (performed before the national standard was set) versus the 2.5 μm size cut adopted for the Federal reference method samplers. The new Federal reference method samplers operate at a higher filter face velocity than the samplers used in the present work, a feature which may generate small differences in the collection of semi-volatile species such as nitrates and organic aerosols.

The 12.4 μg m^{-3} annual average fine particle concentration seen at Quabbin Reservoir, MA, represents regional "background" concentrations in this part of the northeastern United States. It is hard to identify an area in this part of the county with less local pollutant-generating activity than at the protected watershed at Quabbin Reservoir. Regional background air quality as defined here represents the long distance transport of a widespread diluted air mass that contains the accumulation of the emissions from many distant upwind sources. Regional background values should not be confused with the natural background particle concentrations that would exist in the absence of human activities on the North American continent. For example, upwind of the continent at San

Nicolas Island, CA, we measured annual average fine particle concentrations of 7.7 μg m^{-3} in 1993 (18). In any case, the regional background concentration measurements made in the northeastern United States provide important information because they identify the baseline onto which the effect of local emissions sources is added and thus identify the floor against which an entirely local emission control program will act.

On an annual basis downtown Boston at Kenmore Square shows fine particle concentrations that average 3.8 μg m^{-3} (or 30%) higher than at the remote Quabbin Reservoir site in rural Massachusetts. However, the influence of the cities on coarse particle concentrations is much more significant. The annual average coarse particle concentration at Kenmore Square in downtown Boston was 41.3 μg m^{-3}, and in Rochester, NY the annual average coarse particle concentration was 31.5 μg m^{-3}. By comparison, the rural sites had annual average coarse particle concentrations of 16.6 and 12.1 μg m^{-3} at Brockport, NY and Quabbin Reservoir, MA, respectively.

Material balances on the annual average chemical composition of the coarse and fine airborne particles are shown in Figures 3.1 to 3.5 and in Table 3.1. At the regional background sites in Brockport, NY, and Quabbin Reservoir, MA, ammonium sulfate and carbonaceous particles are of about equal importance, each accounting for roughly 30-35% of the fine particle mass concentration. The dominance of organic carbon over elemental carbon is a general feature observed in most ambient aerosol samples taken in the Northeast as well as elsewhere (1-3, 18). Crustal material makes up the largest fraction of the coarse material at the background sites with carbonaceous particles second.

At the more urban sites, the sulfate contribution to fine particle concentrations remains very similar to that at the background sites, while carbon particle concentrations increase within the more urban atmospheres. This effect is especially pronounced at Kenmore Square in Boston, where particulate organic compounds plus elemental carbon particles

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account for the majority of the fine particle mass concentrations observed. Crustal material is again the most abundant species within the coarse particulate matter with organics second. There is also a significant concentration of chloride particles (8-12%) found in the coarse material at the urban sites in the winter which is absent at the Quabbin Reservoir background site and which is also less pronounced at the rural Brockport site. This coarse particle chloride is logically related to particles generated as vehicles travel over roads on which salt has been used for ice control in the winter.

The highest 24-hour average fine particle concentration measured during the study year was 51.1 μg m^{-3} at Kenmore Square, Boston, on July 14, 1995. That day saw high concentrations throughout the entire air monitoring network; fine particle concentrations at the Quabbin Reservoir site were 47.8 μg m^{-3} on that day, only slightly lower than in downtown Boston. July 14, 1995, also saw much higher than average aerosol sulfate concentrations across the northeast, as did July 26 (see Figures 3.16–3.20). February 20, 1995, experienced the highest 24-hour average fine particle concentrations in the Rochester area. The downtown Rochester site recorded fine particle concentrations of 49.3 μg m^{-3} on a day with much higher than average fine carbon particle concentrations (see Figures 3.9 and 3.14). The new 24-hour average federal ambient air standard for fine particles which is set at 65 μg m^{-3} was not exceeded at any time during the days sampled in 1995. Since the annual average fine particle standard of 15 μg m^{-3} is approached or exceeded at several sites while the 24 hour average standard is not, this situation calls for a sampling strategy that emphasizes accurate determination of annual average values.

The two highest 24-hour coarse particle concentrations measured during the study were both found in the winter at Kenmore Square, Boston. They were 132.8 μg m^{-3} on February 8, 1995, and 119.4 μg m^{-3} on March 16, 1995. The February 8 high concentration event was localized at Kenmore Square due to increased levels of sulfate and chloride

among the coarse particles. The March 16 day was also among the highest days for coarse particle concentrations at Rochester, NY which experienced 69.8 μg m^{-3} on that day.

May 21, 1995, was a high day for coarse particle concentrations at all sites. Quabbin Reservoir experienced its peak coarse particle concentration of 92.4 μg m^{-3} on this day, as did Reading, MA, with 67.3 μg m^{-3}. The other three locations also experienced high coarse particle concentrations on May 21, 1995, with concentrations of 87.8 μg m^{-3} in downtown Boston, 61.8 μg m^{-3} in Brockport, NY, and 34.0 μg m^{-3} in Rochester. The peak days for coarse particle concentrations at the New York locations were August 31, 1995, in Brockport with 70.5 μg m^{-3} and February 14, 1995, in Rochester with 84.9 μg m^{-3}.

Time series graphs of 24-hour average sulfate concentrations for sites monitoring fine particles are shown in Figures 3.16 to 3.20. The general agreement of same-day fine particle and total particle sulfate concentrations is remarkable, confirming that sulfate is primarily a fine particle substance. The degree of agreement of same-day fine particle sulfate concentrations across the Massachusetts sites, and separately the New York sites, also is remarkable. With the exception of late August and early September, 1995, the New York and Massachusetts fine particle sulfate concentrations generally track each other as well. Fine particle sulfates thus comprise a major portion of the regional background air quality discussed earlier that extends across the entire monitoring network.

Aerosol nitrate concentrations are modest contributors to the observed fine particle mass. Fine particle nitrate concentrations are highest in the colder months, as expected since cold temperatures favor NH_4NO_3 formation from gaseous NH_3 and HNO_3. Certain of the days with high coarse particle nitrates (e.g. at Rochester) correspond to days with high coarse particle chloride concentrations and may result from the reaction of nitric acid vapor with NaCl used to salt the roads. A small quantity of material derived from soil or road dust also is present in the fine particles, accounting for about 5-7% of fine particle

mass at most sites. As expected, this material comprises a much greater fraction of the coarse airborne particles.

The samplers used for fine particle collection in this study are similar to the new Federal Reference Method (FRM) samplers in the sense that mass concentrations are determined gravimetrically from weighing 47mm diameter Teflon filters. As in the FRM, no denuder technology was employed and as a result, semi-volatile species such as ammonium nitrate will be lost in part during sampling. However, in the northeastern United States, ammonium nitrate concentrations are generally thought to be small, so that the potential for loss of nitrates by evaporation during sampling probably is low as well. Both positive and negative artifacts for aerosol carbon are possible, and one cannot say with certainty without further experiments exactly what effect the use of denuders ahead of the filters and backup sorbent traps downstream of the filters would have on reported organic particulate matter concentrations. Use of denuder-based sampling technology for organic aerosols is sufficiently complex that it has never been incorporated into previous routine air monitoring networks.

5 CONCLUSION

The picture emerging from these data can be summarized briefly. The areas studied in the northeastern states stretching from the Great Lakes near Rochester to the Atlantic Ocean near Boston experience a high regional background level of fine particulate matter at concentrations just below the new annual average standard for fine particles. The sulfate component of that background is largely the same on the same day across the area studied and is already present at the most upwind site studied. Carbonaceous aerosols and ammonium sulfate are of about equal importance as contributors to fine particle

mass concentrations at the most rural sites, and carbonaceous aerosols become the largest contributor at the most urban site in downtown Boston. The day-to-day variability of carbon particle concentrations is less systematic across the network than is the case for sulfates. These features suggest that local sources as well as regional background are important factors in determining carbon particle concentrations. Coarse particle concentrations are higher in cities than in the more rural areas and reflect the local emissions of coarse particles from sources such as road dust and road salt.

The chemical composition data reported here are suitable for use with trace elements-based receptor-oriented air quality models that seek to apportion increments to primary particle concentrations between the contributing sources (19). Motor vehicle exhaust, paved road dust and biomass burning source contributions can be estimated on that basis using the data provided here on organic carbon, elemental carbon and crustal elements (Si, Al, Fe, Ti, etc.) along with non-soil potassium concentrations (often used as a marker for biomass-combustion aerosol) which can be calculated from the present data. Local paved road dust source profiles and possibly local biomass combustion source profiles would be necessary to support this analysis. The elemental composition data provided in the present report also could be combined with data on the organic compounds present in the particle phase to obtain a more complete account of the motor vehicle, wood smoke, food cooking smoke, paved road dust, tire dust, plant fragments and natural gas combustion contributions to airborne fine particle contributions using the methods of Schauer et al (20).

The organic tracer-based source apportionment method of Schauer et al (20) does require the use of the crustal elements data plus elemental carbon data provided in the present report. In addition, the organic aerosol samples collected as part of this work would need to be extracted and the concentrations of the approximately 50 organic compounds specified by Schauer et al (20) would need to be determined by GC/MS analysis. Then the

organic chemical composition of local wood smoke and paved road dust would need to be determined and combined with existing source profiles for vehicle exhaust, food cooking, and other source categories to complete the source apportionment study.
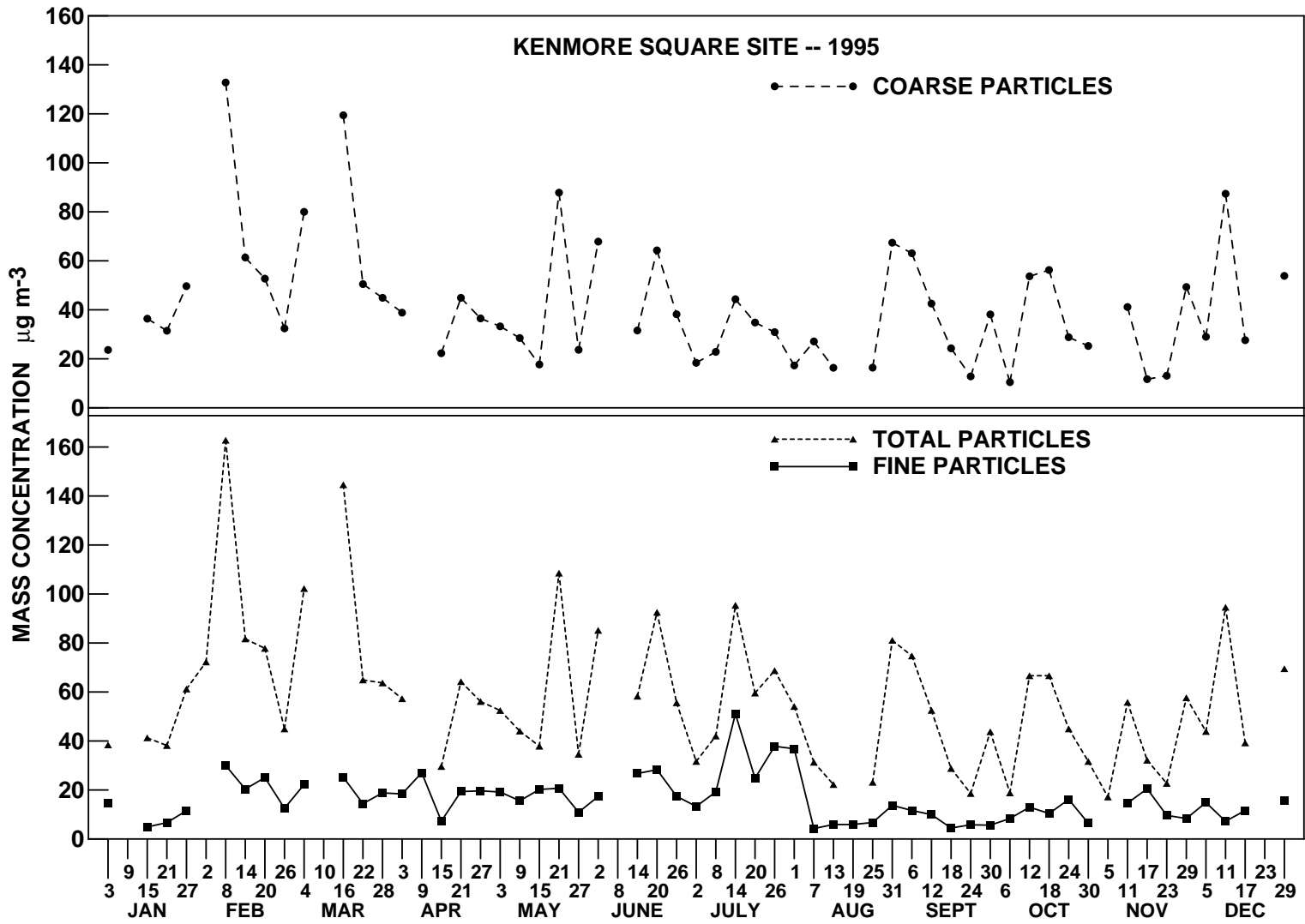
Figure 3.6 - Particle Mass Concentrations at Kenmore Square, Boston, MA
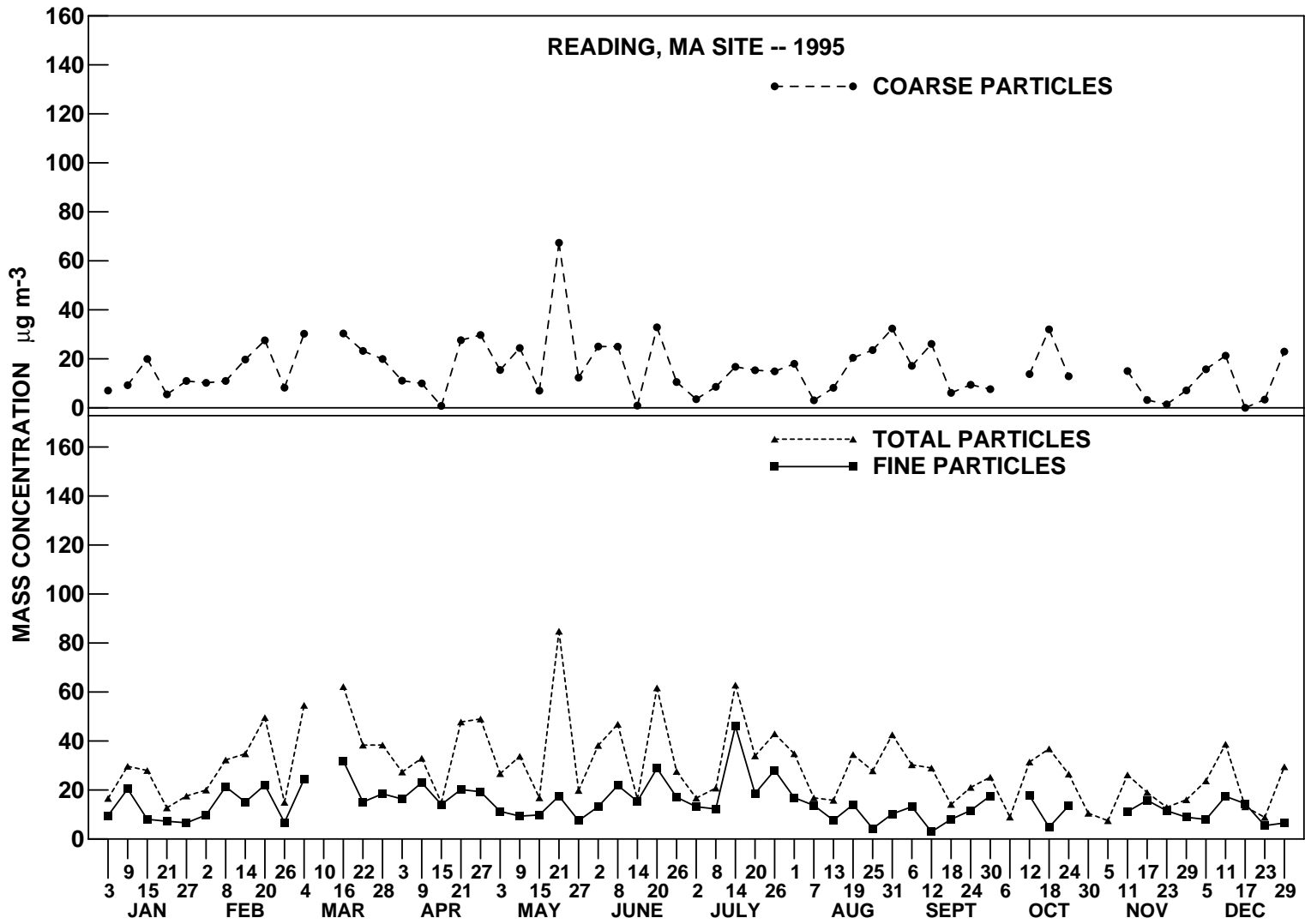
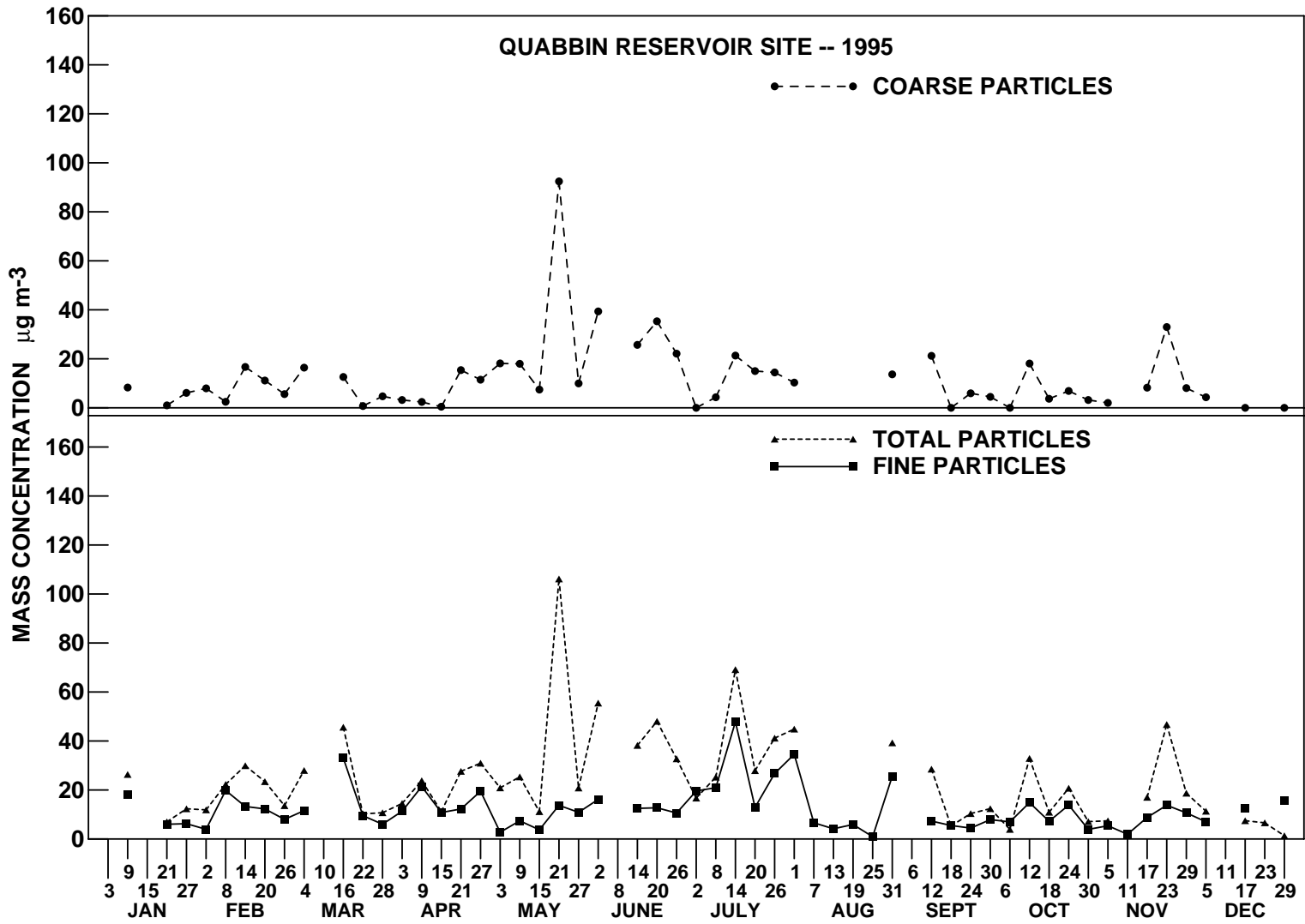**Figure 3.7 - Particle Mass Concentrations at Reading, MA**

Figure 3.8 - Particle Mass Concentrations at Quabbin Reservoir, MA
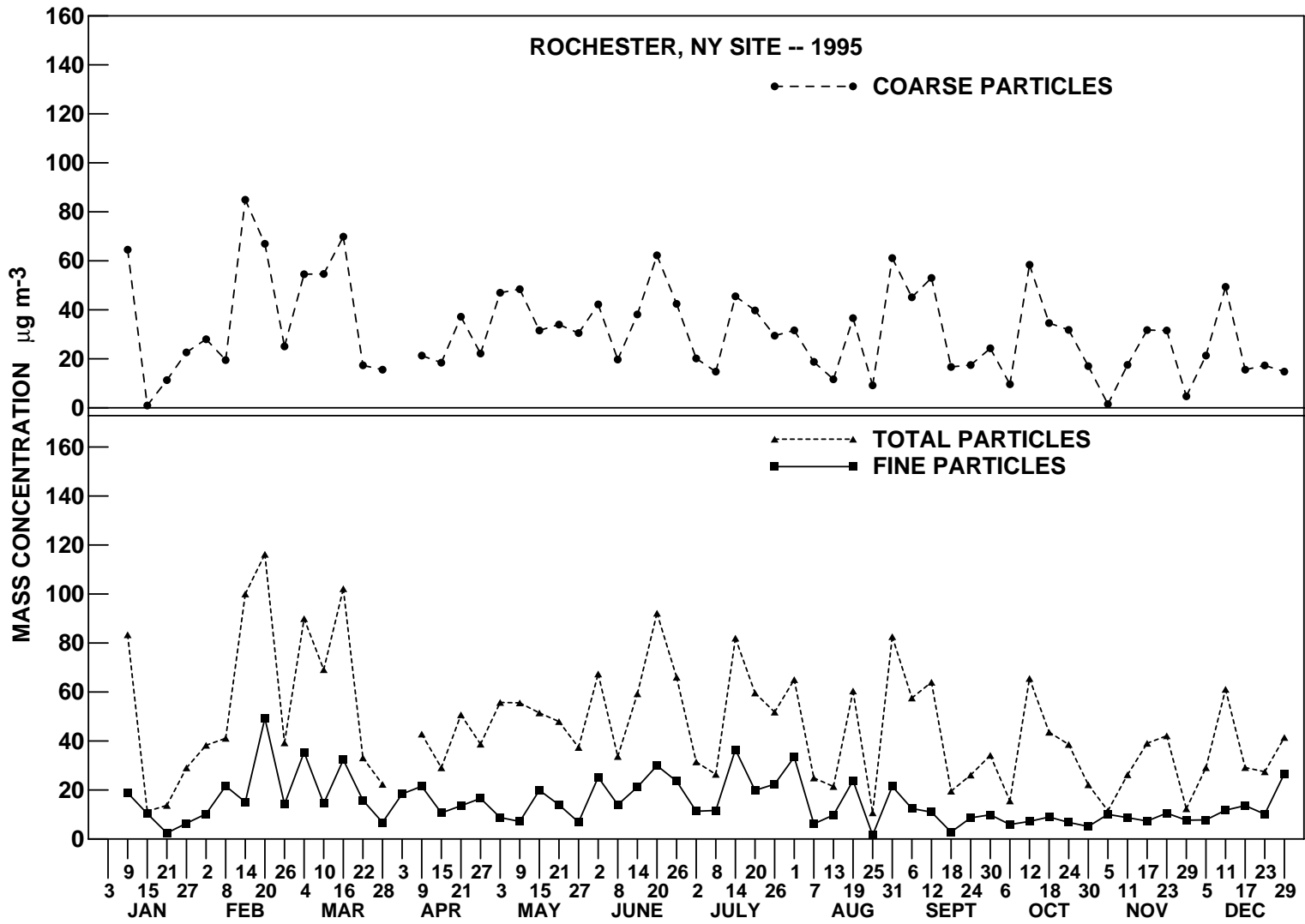
27

Figure 3.9 - Particle Mass Concentrations at Rochester, NY

28
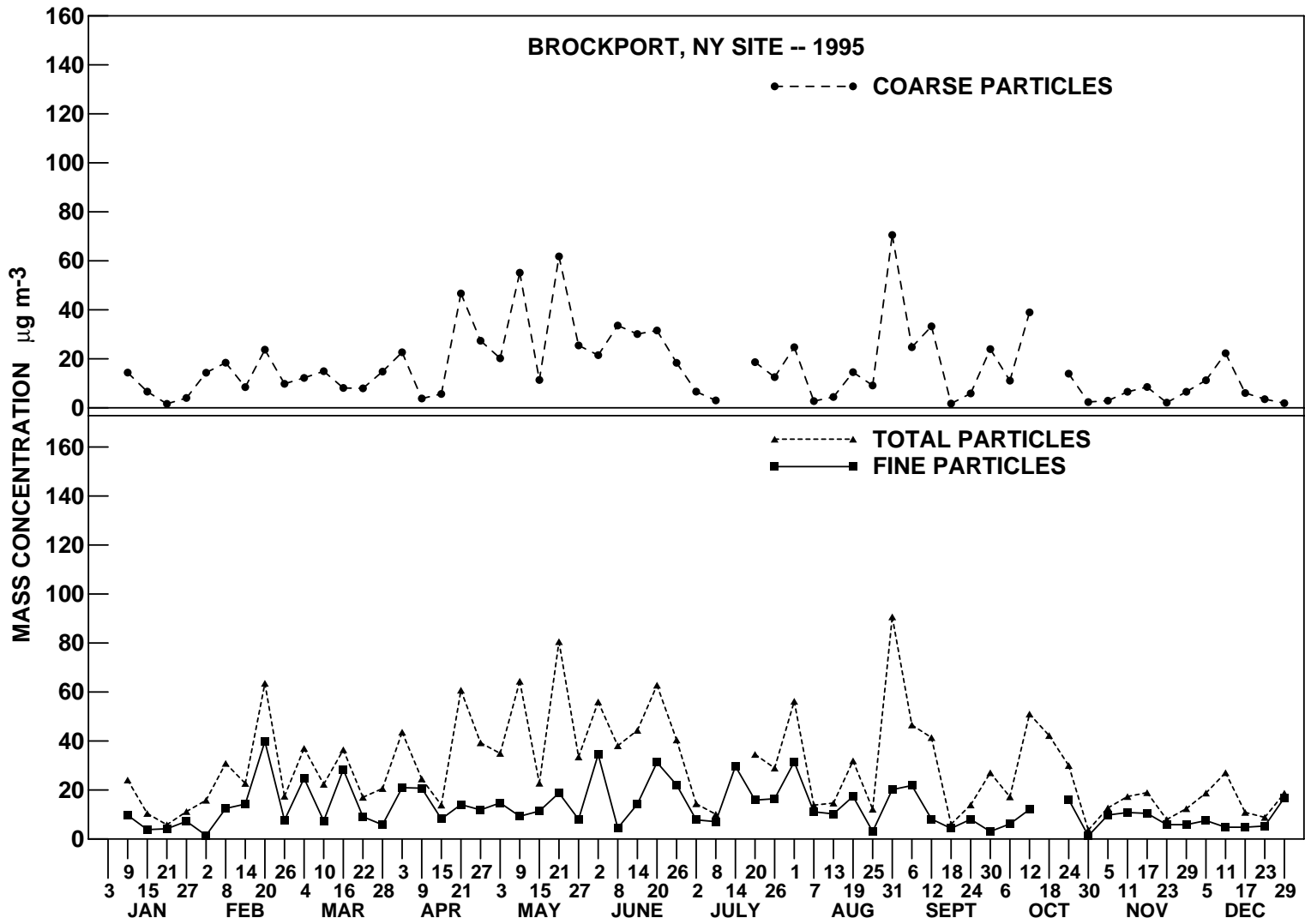
Figure 3.10 - Particle Mass Concentrations at Brockport, NY

29
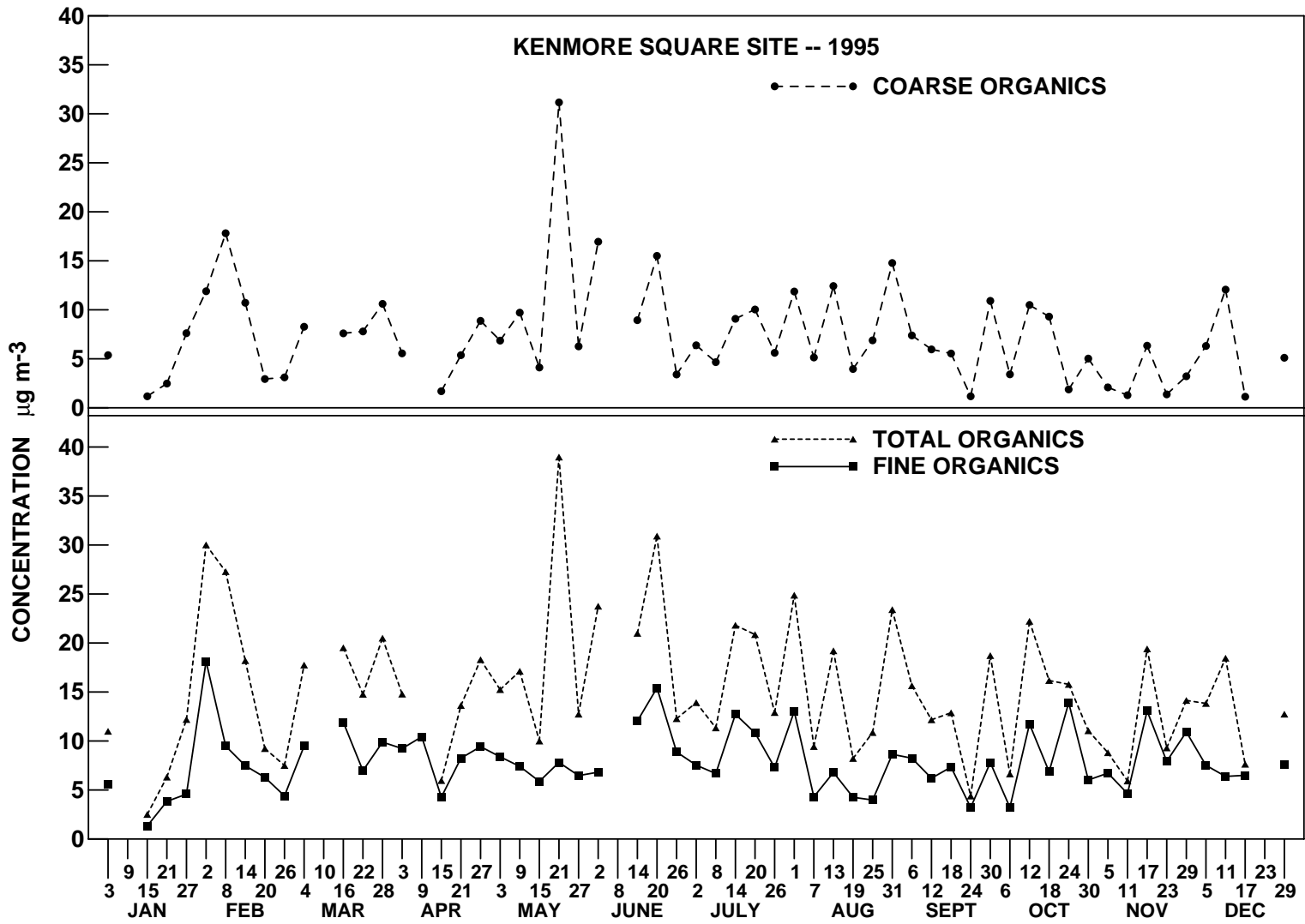
Figure 3.11a - Organic Species at Kenmore Square, Boston, MA
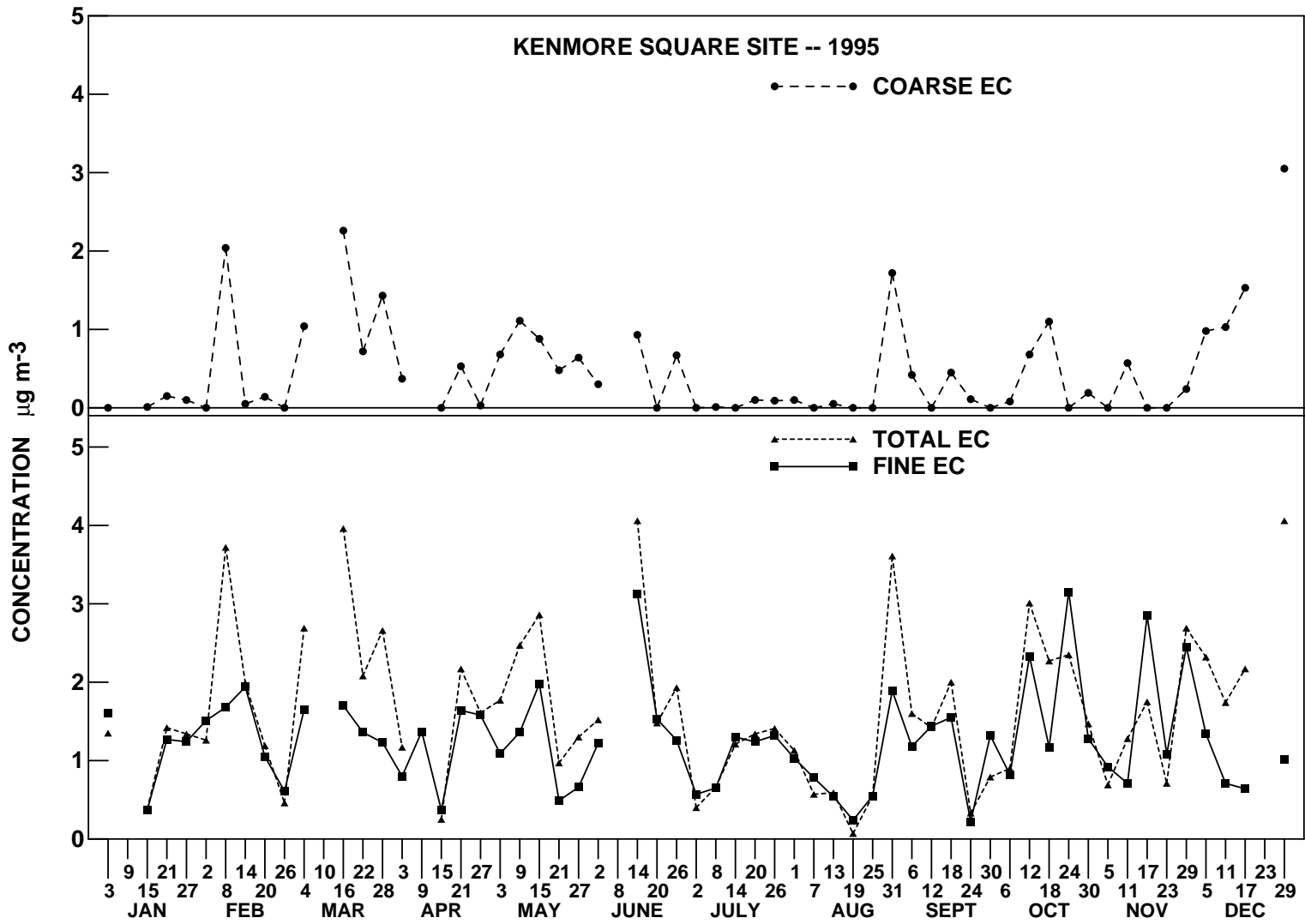
Figure 3.11b - Elemental Carbon at Kenmore Square, Boston, MA
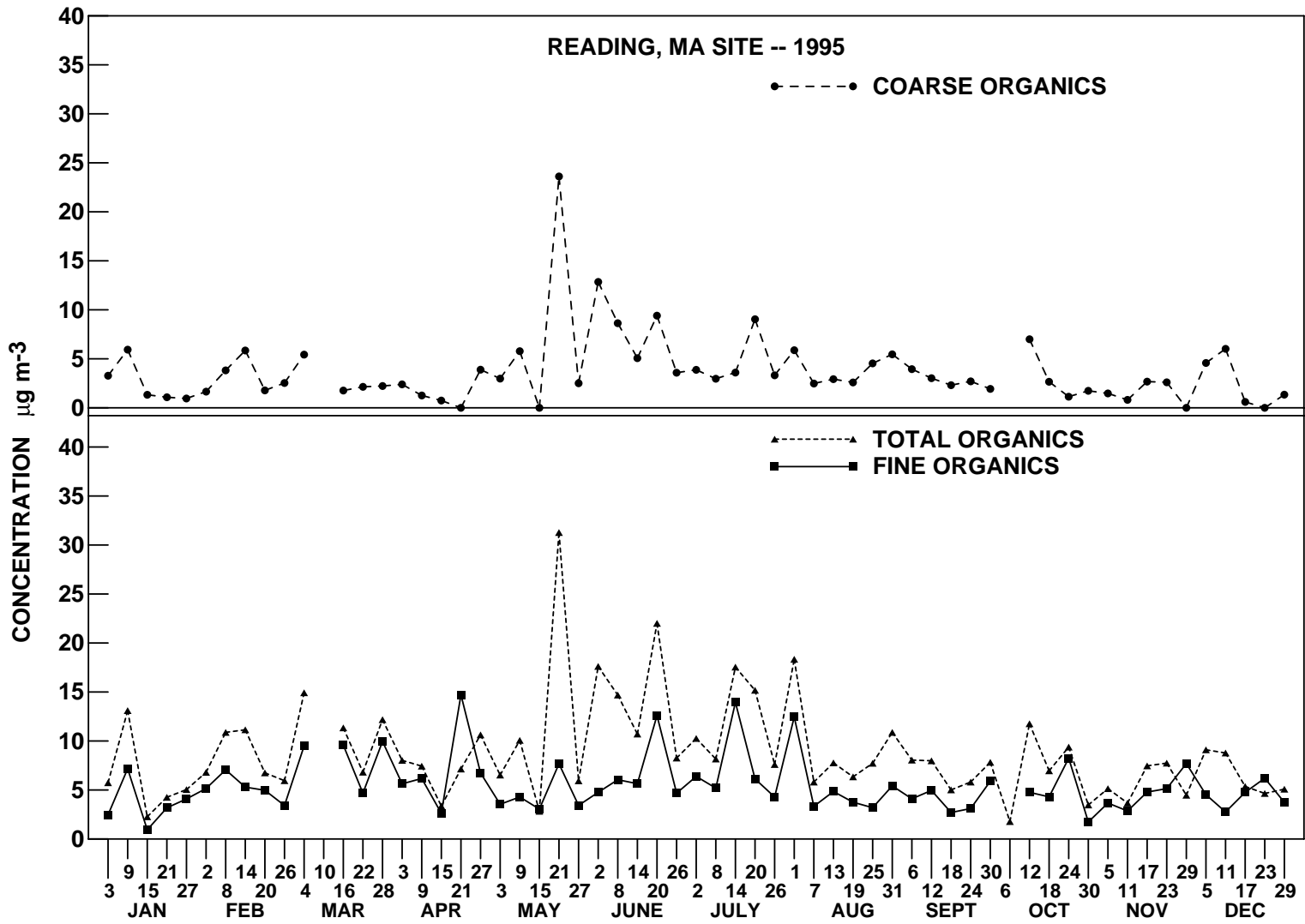
Figure 3.12a - Organic Species at Reading, MA
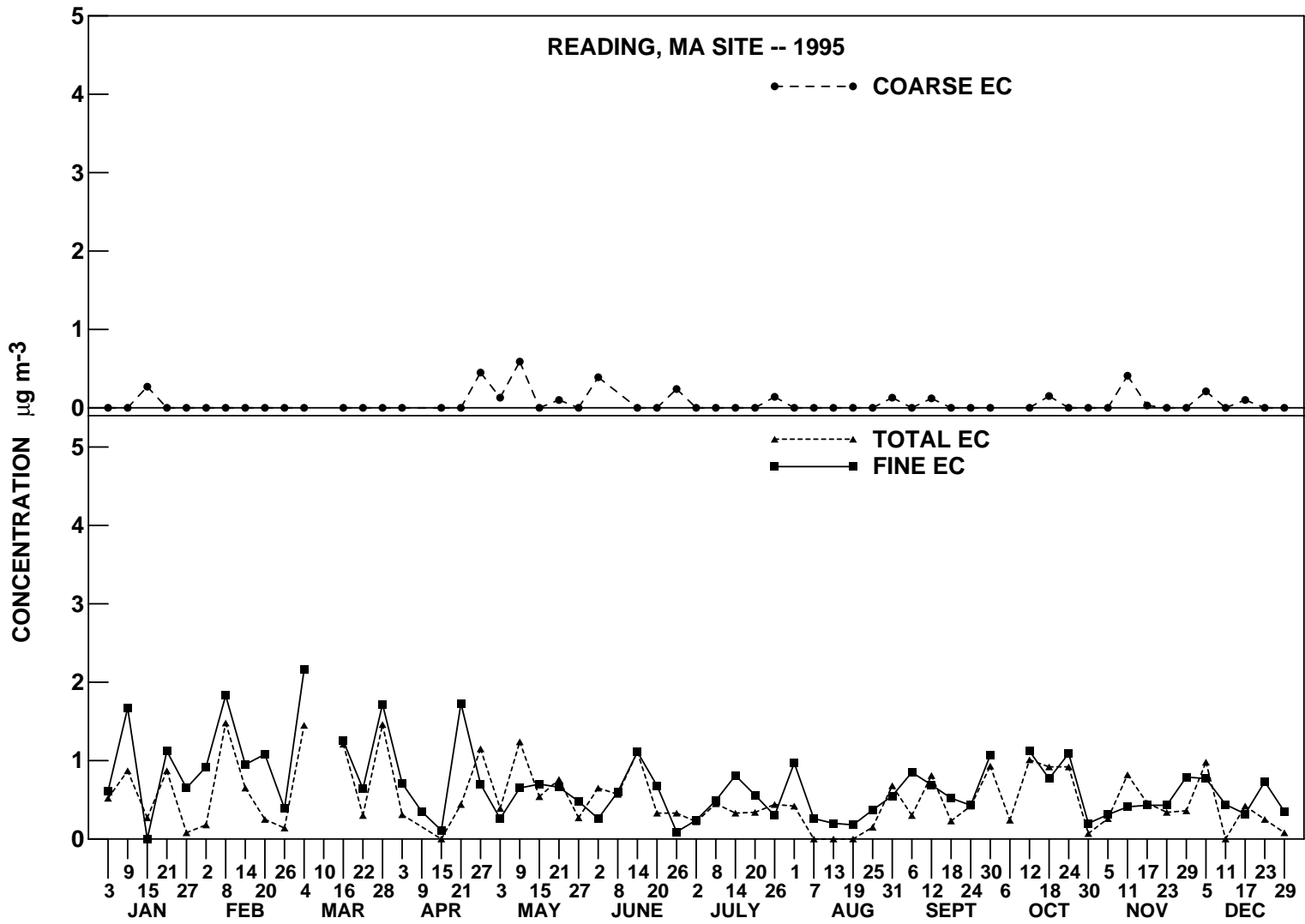
Figure 3.12b - Elemental Carbon at Reading, MA
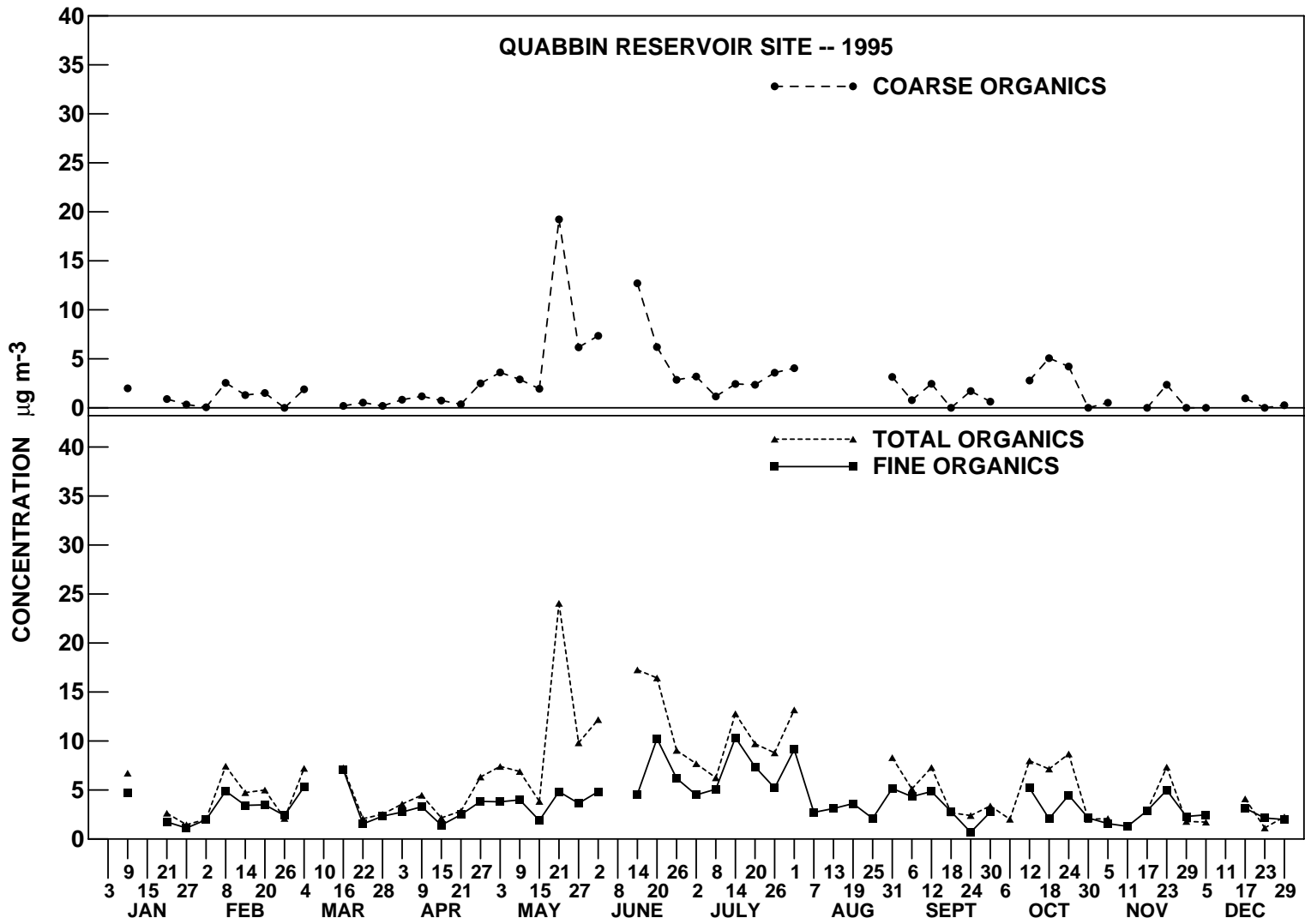
Figure 3.13a - Organic Species at Quabbin Reservoir, MA
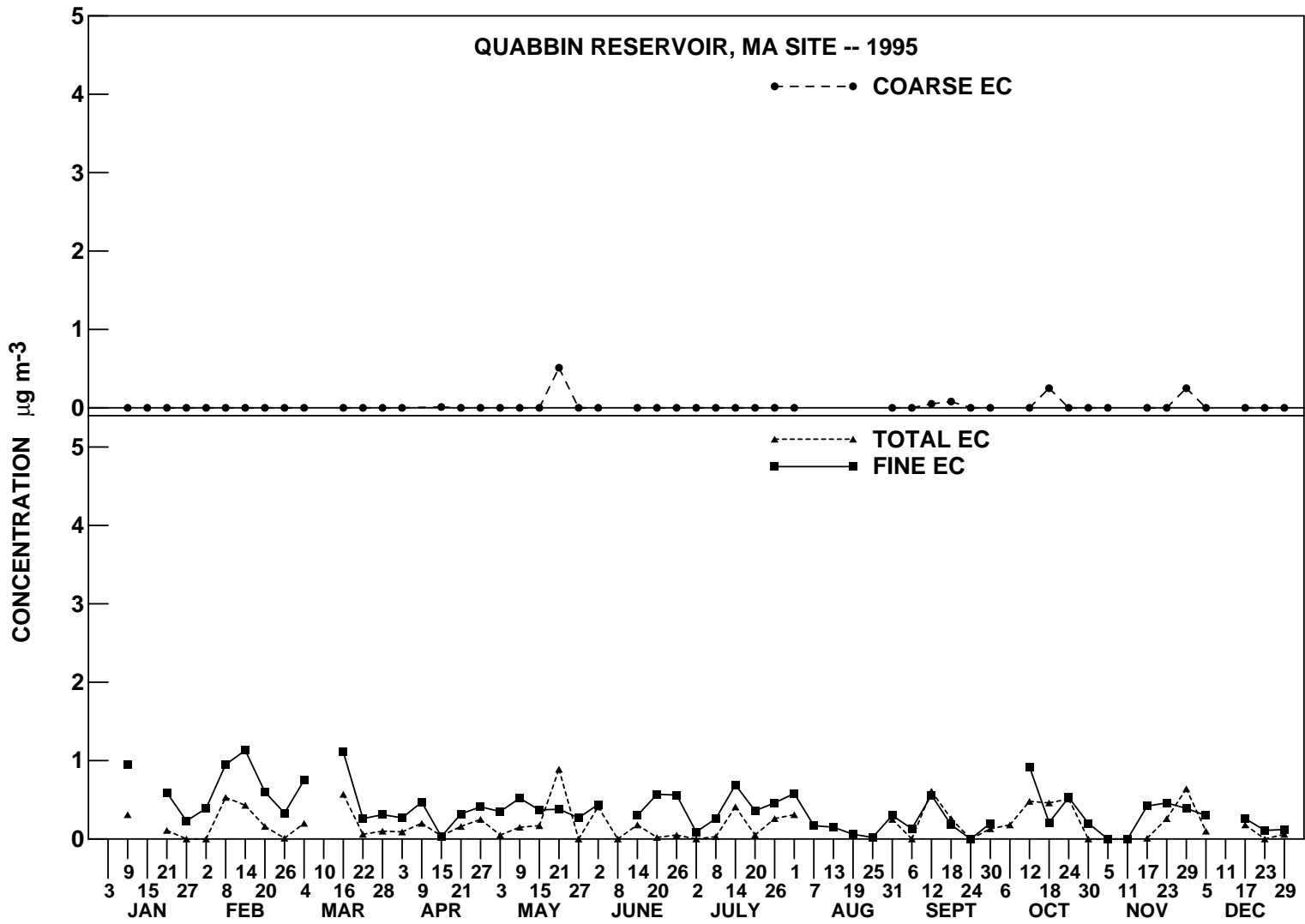
Figure 3.13b - Elemental Carbon at Quabbin Reservoir, MA
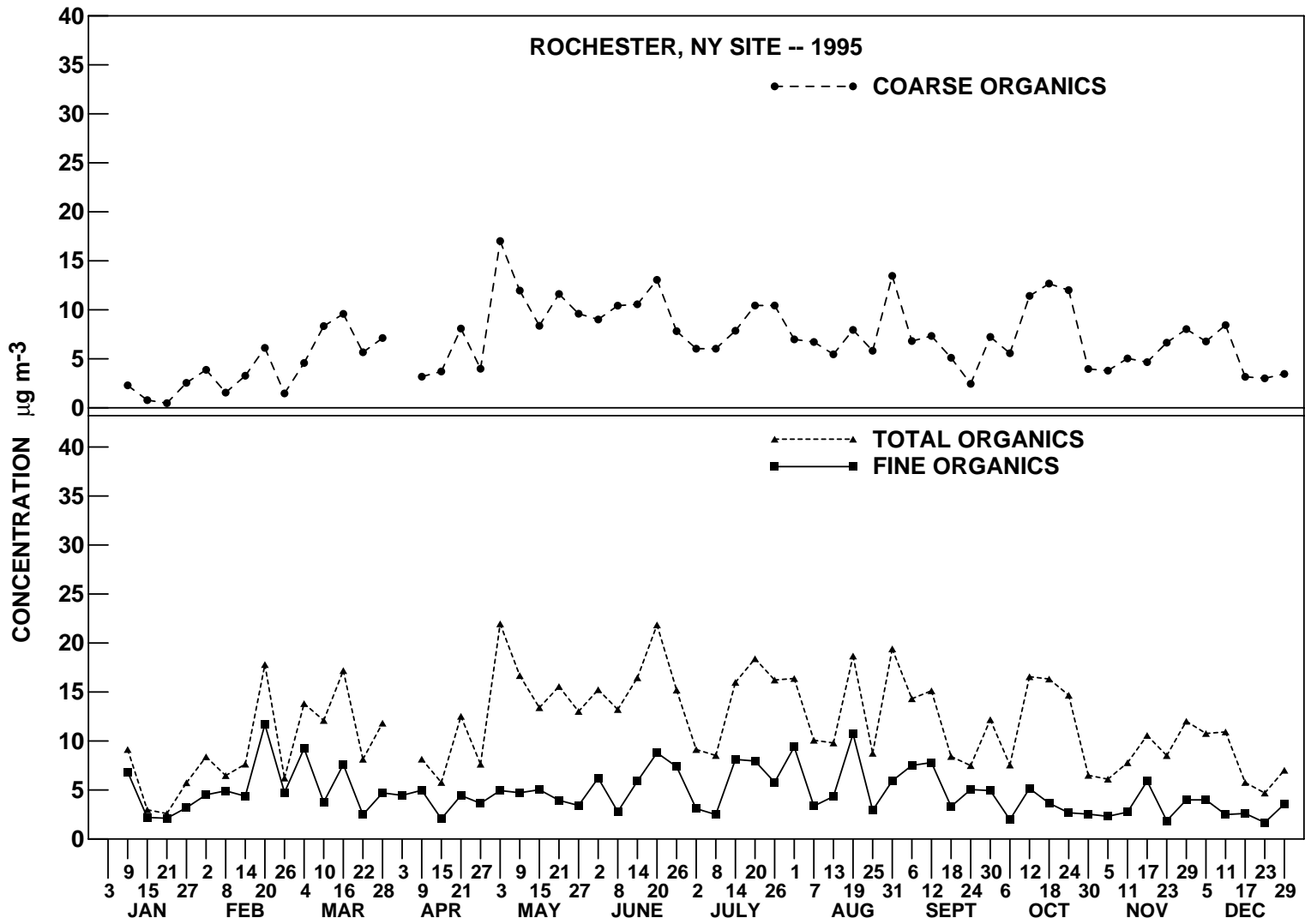
Figure 3.14a - Organic Species at Rochester, NY

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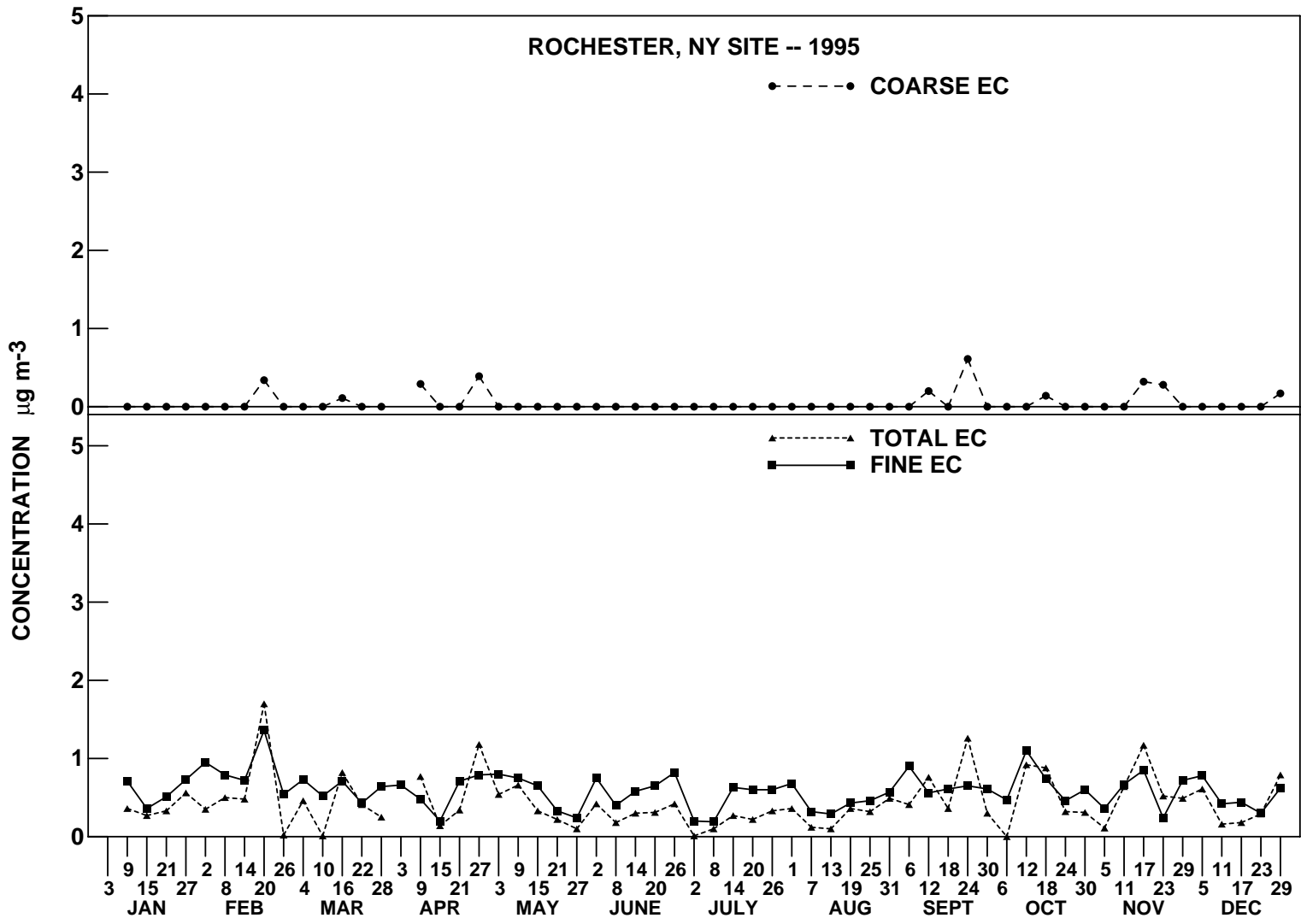
Figure 3.14b - Elemental Carbon at Rochester, NY
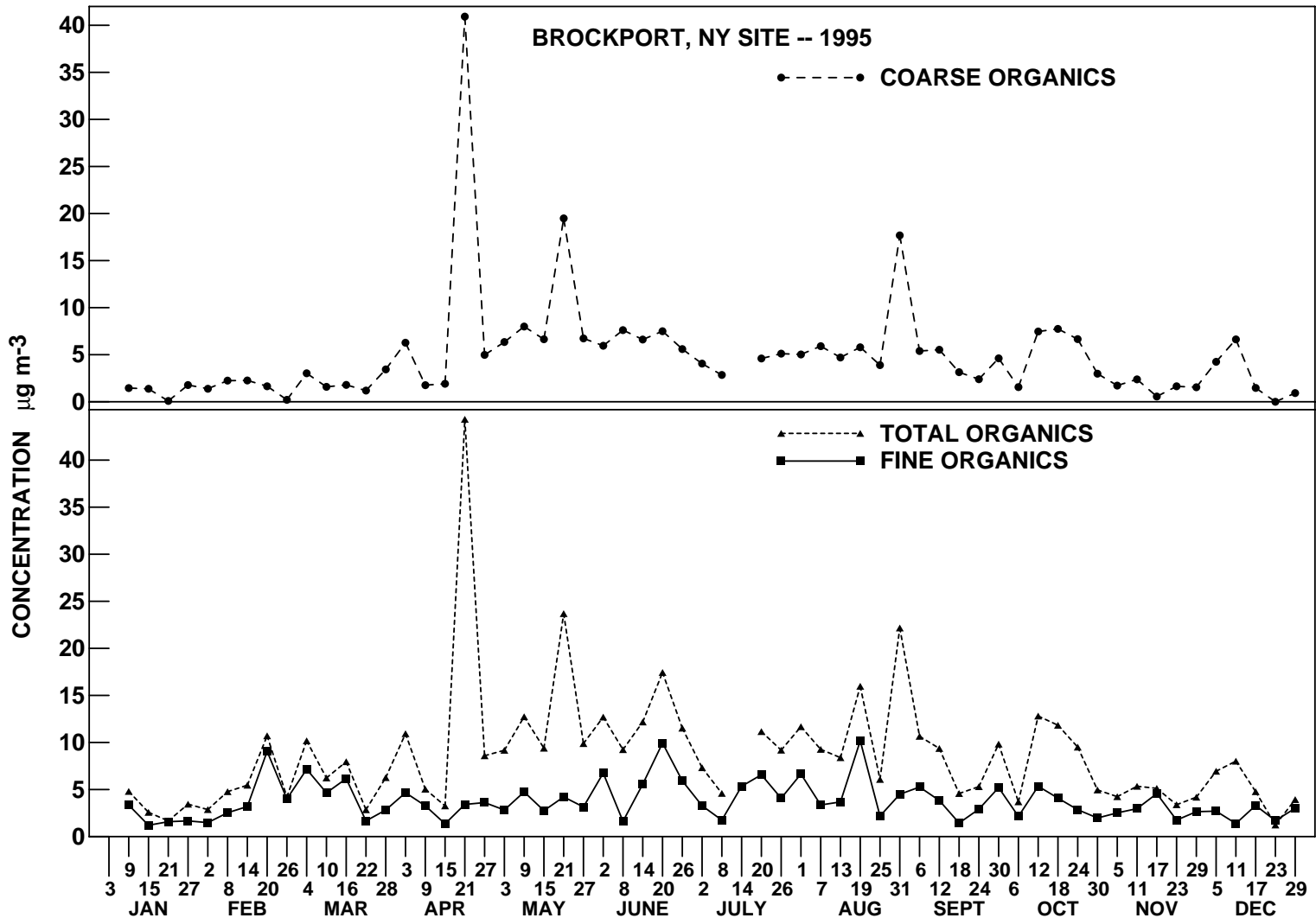
Figure 3.15a - Organic Species at Brockport, NY
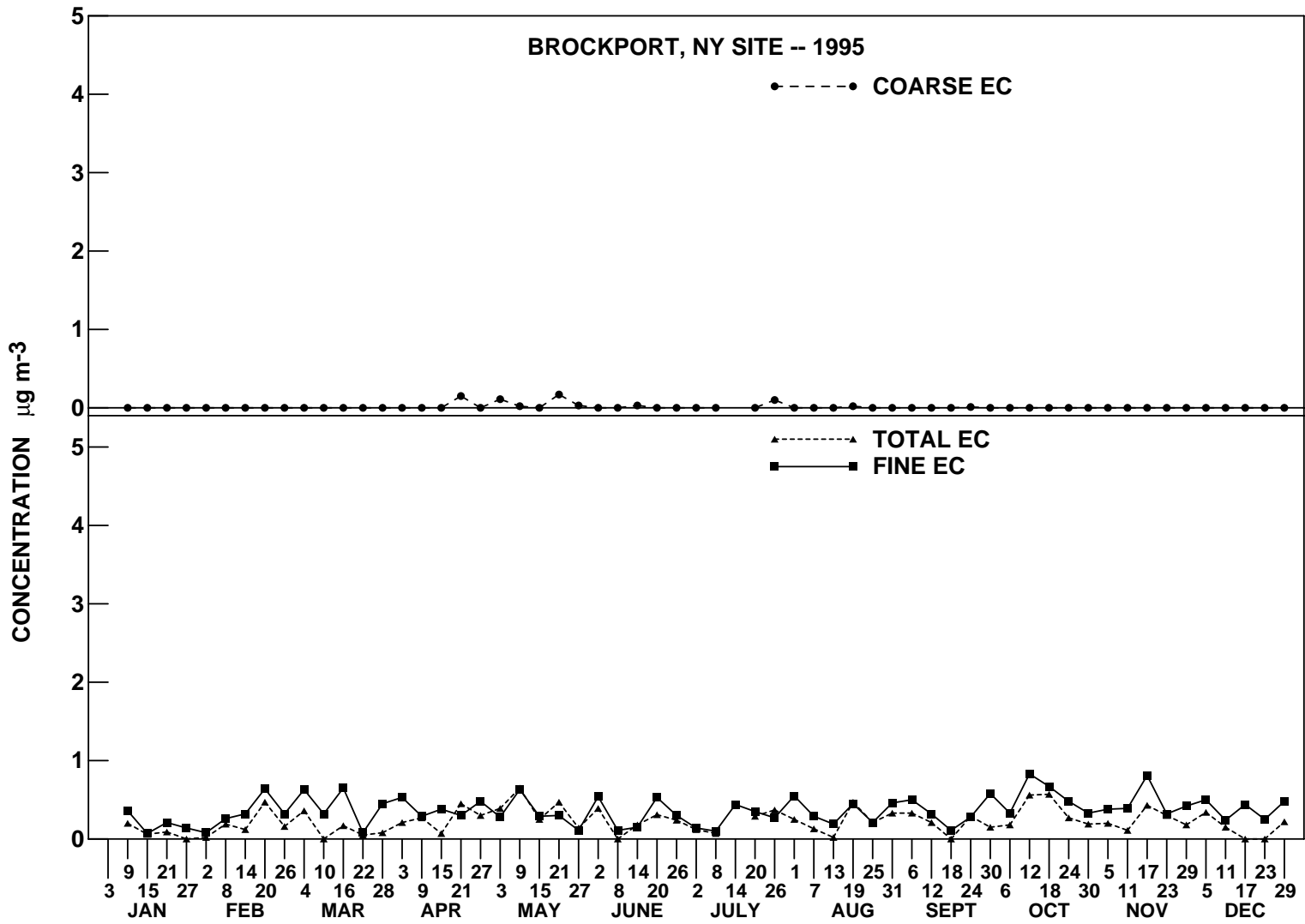
Figure 3.15b - Elemental Carbon at Brockport, NY
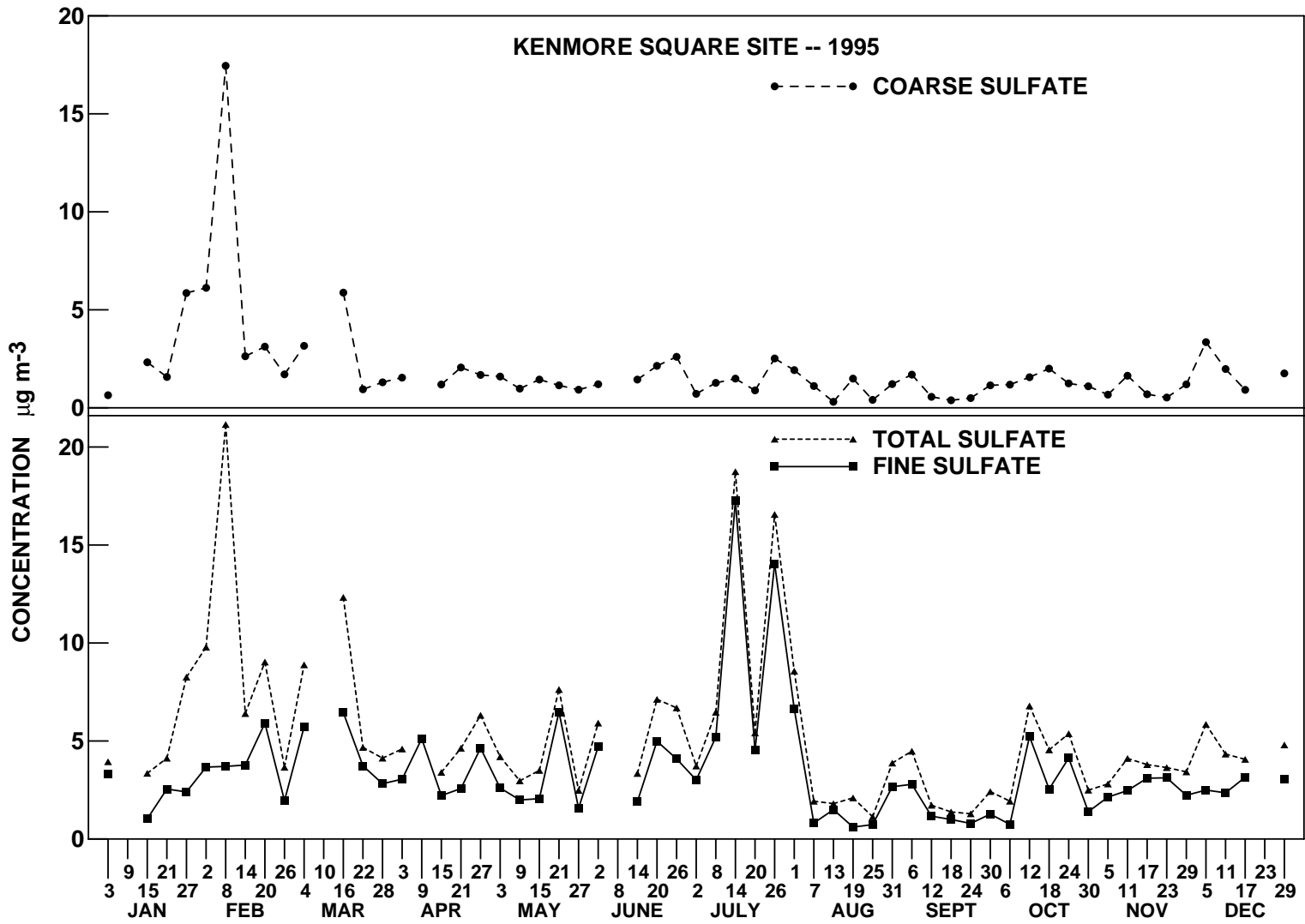
Figure 3.16 - Sulfate Particle Concentrations at Kenmore Square, Boston, MA

40
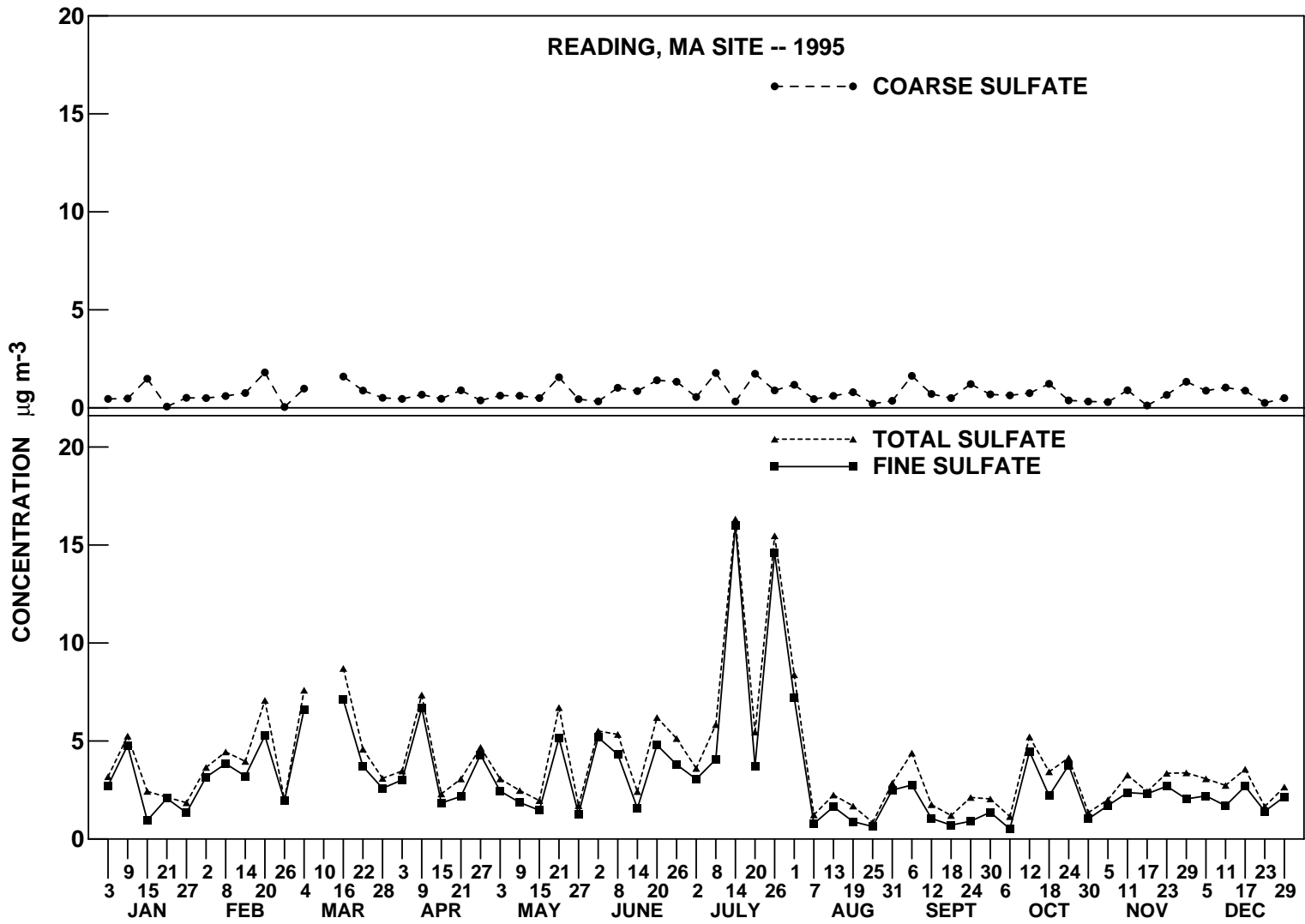
Figure 3.17 - Sulfate Particle Concentrations at Reading, MA

41
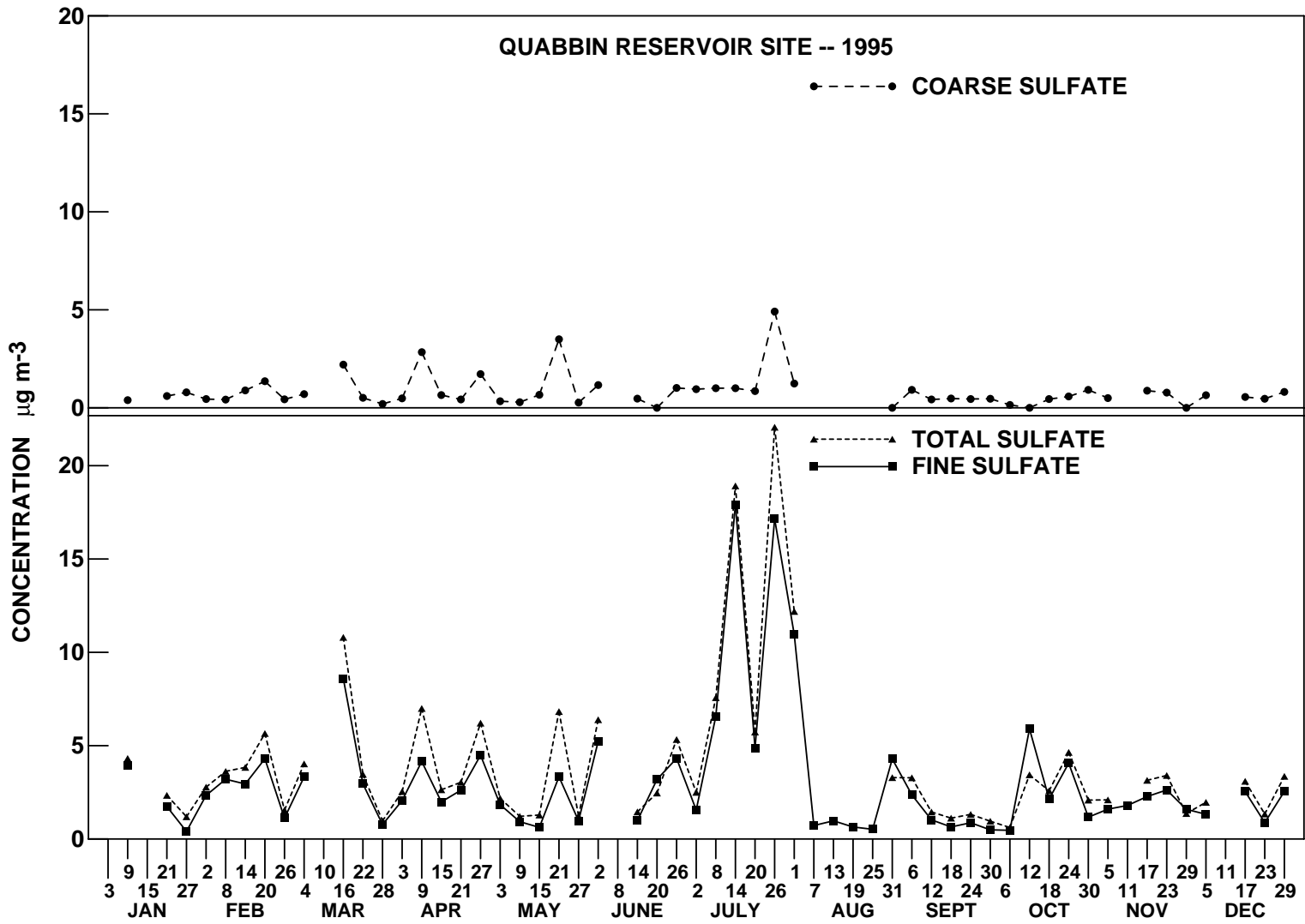
Figure 3.18 - Sulfate Particle Concentrations at Quabbin Reservoir, MA

42
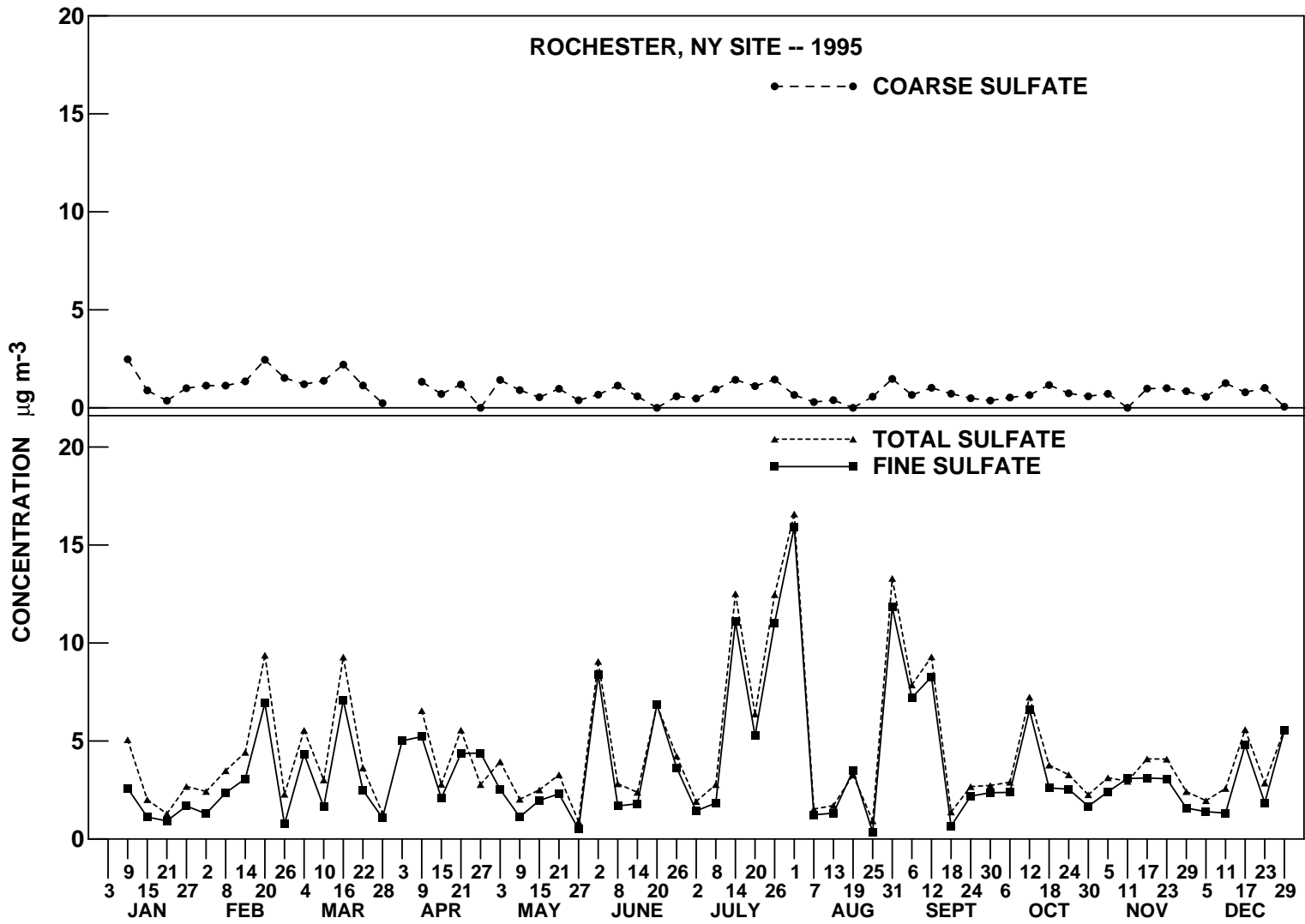
Figure 3.19 - Sulfate Particle Concentrations at Rochester, NY
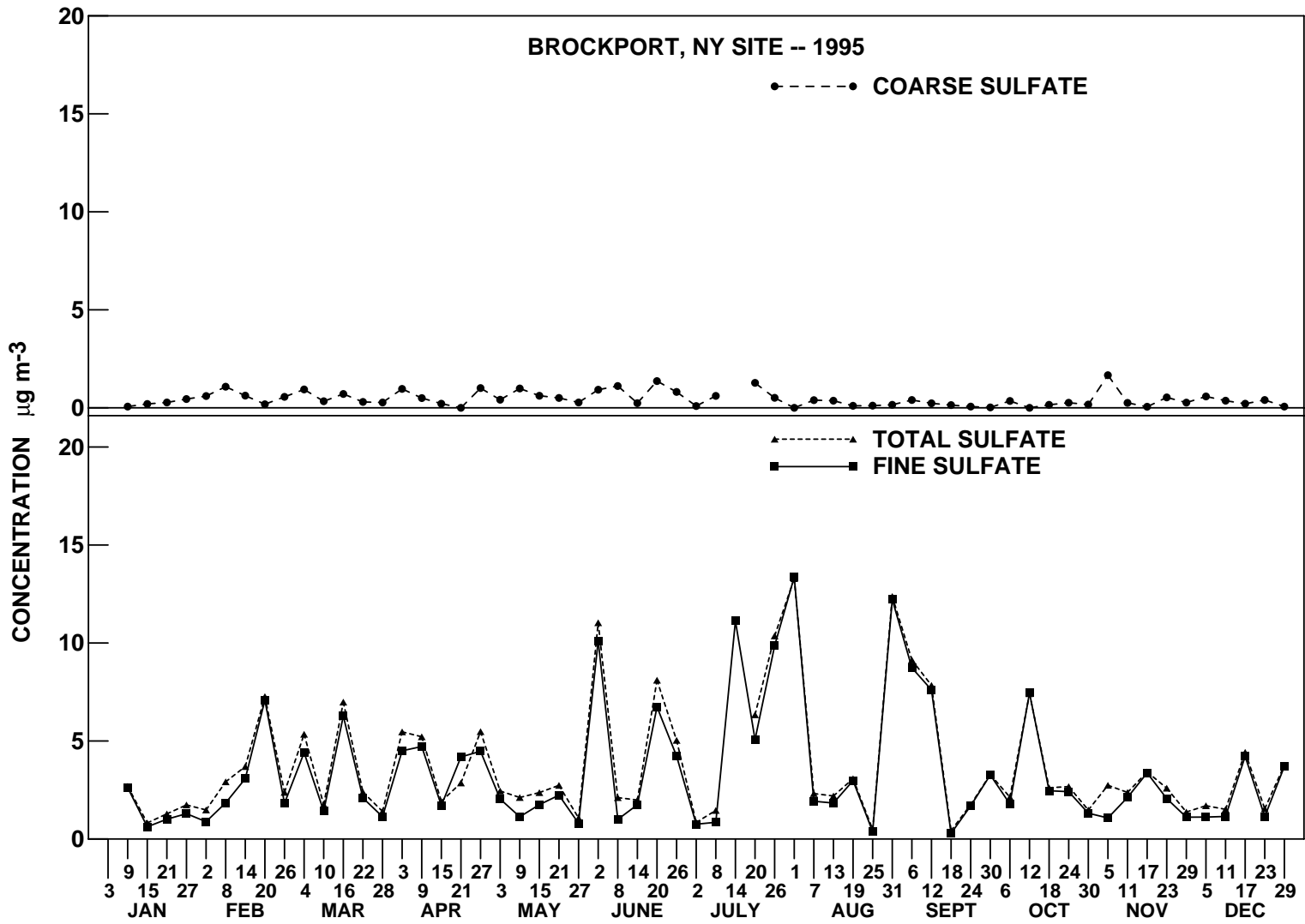
Figure 3.20 - Sulfate Particle Concentrations at Brockport, NY

44
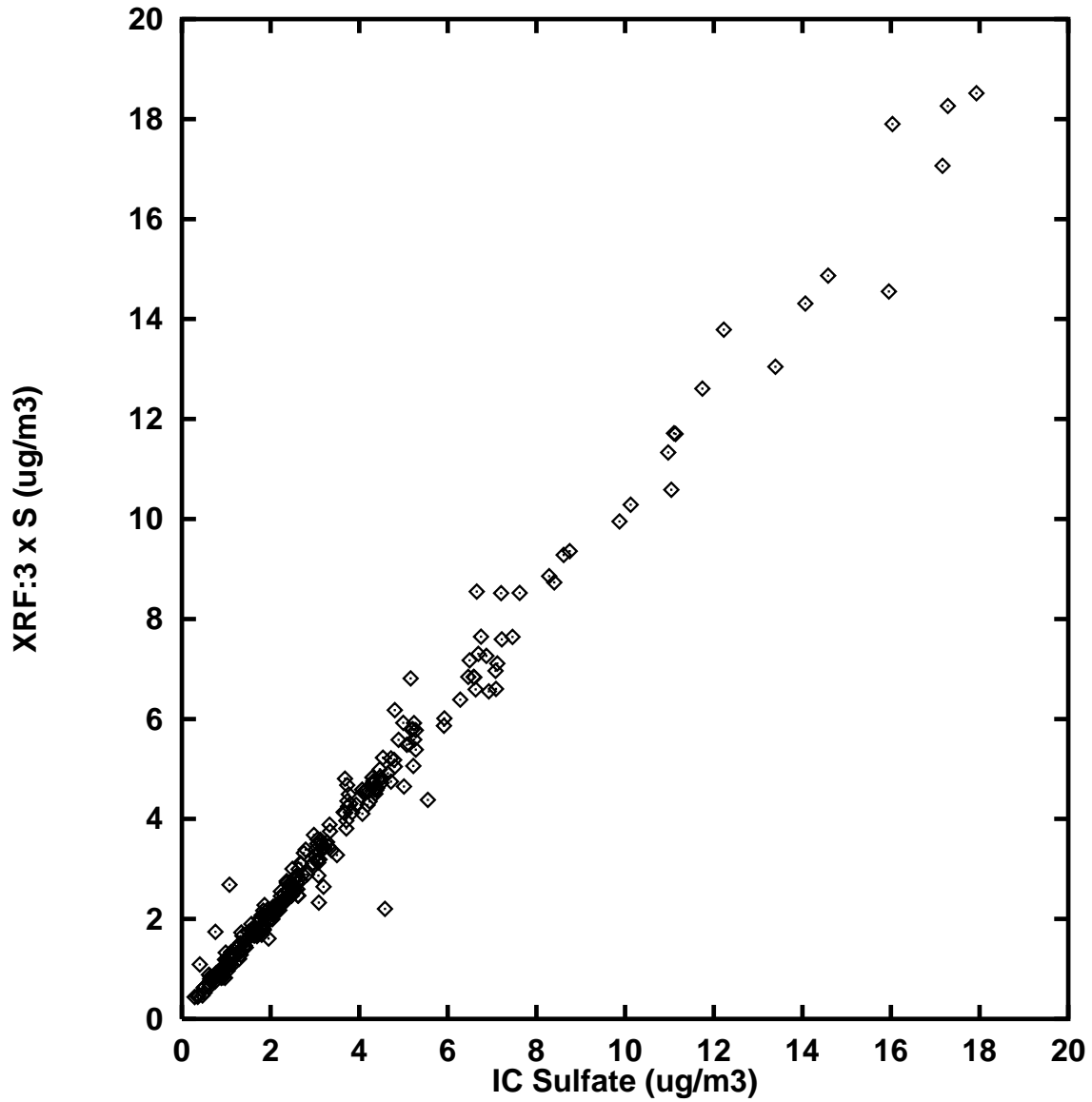
**Figure 3.21 - Comparison of Fine Particle Sulfate Measured by IC
with Fine Particle Sulfur Measured by XRF**
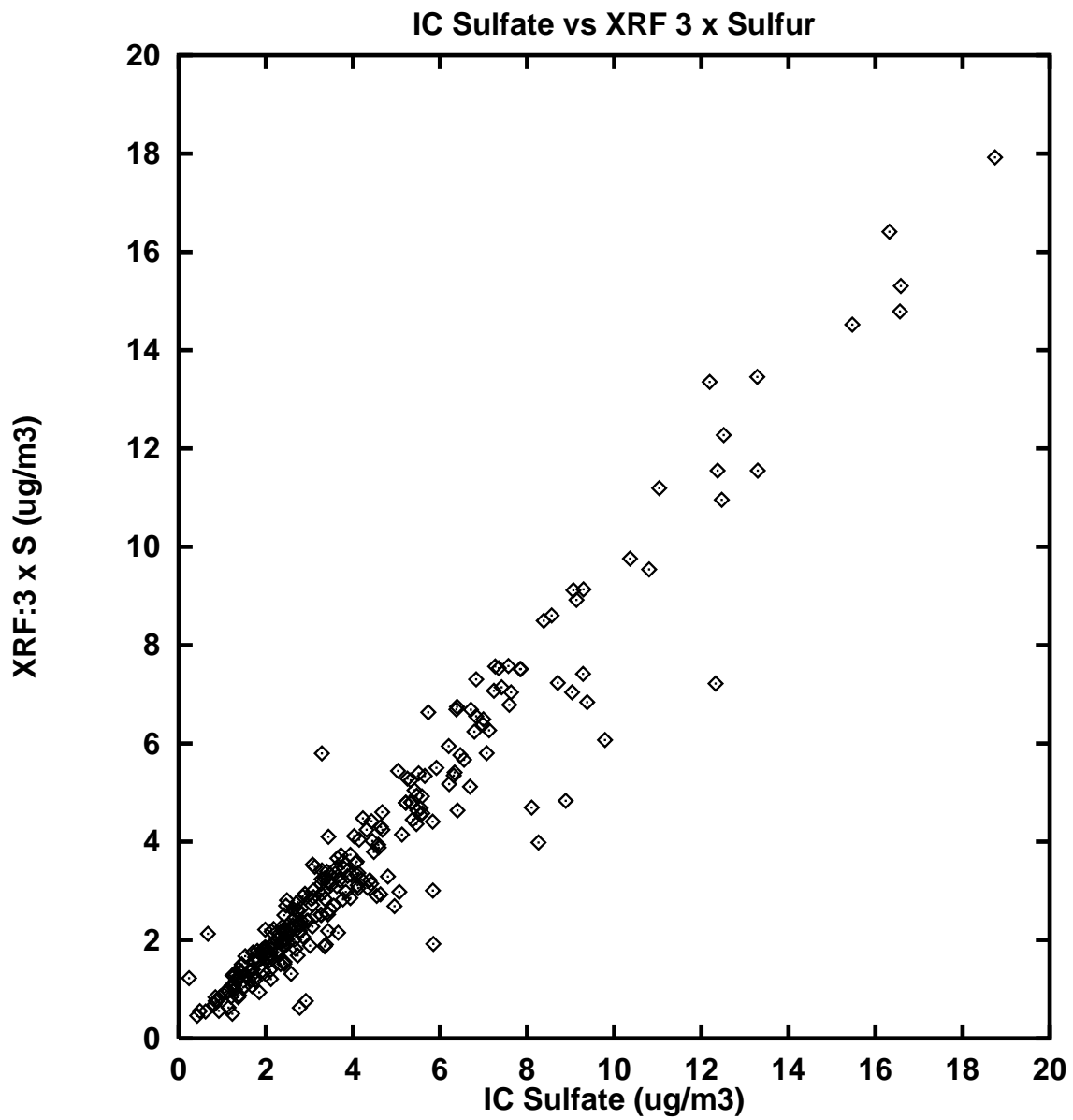
Figure 3.22 - Comparison of Total Particle Sulfate Measured by IC with Total Particle Sulfur Measured by XRF
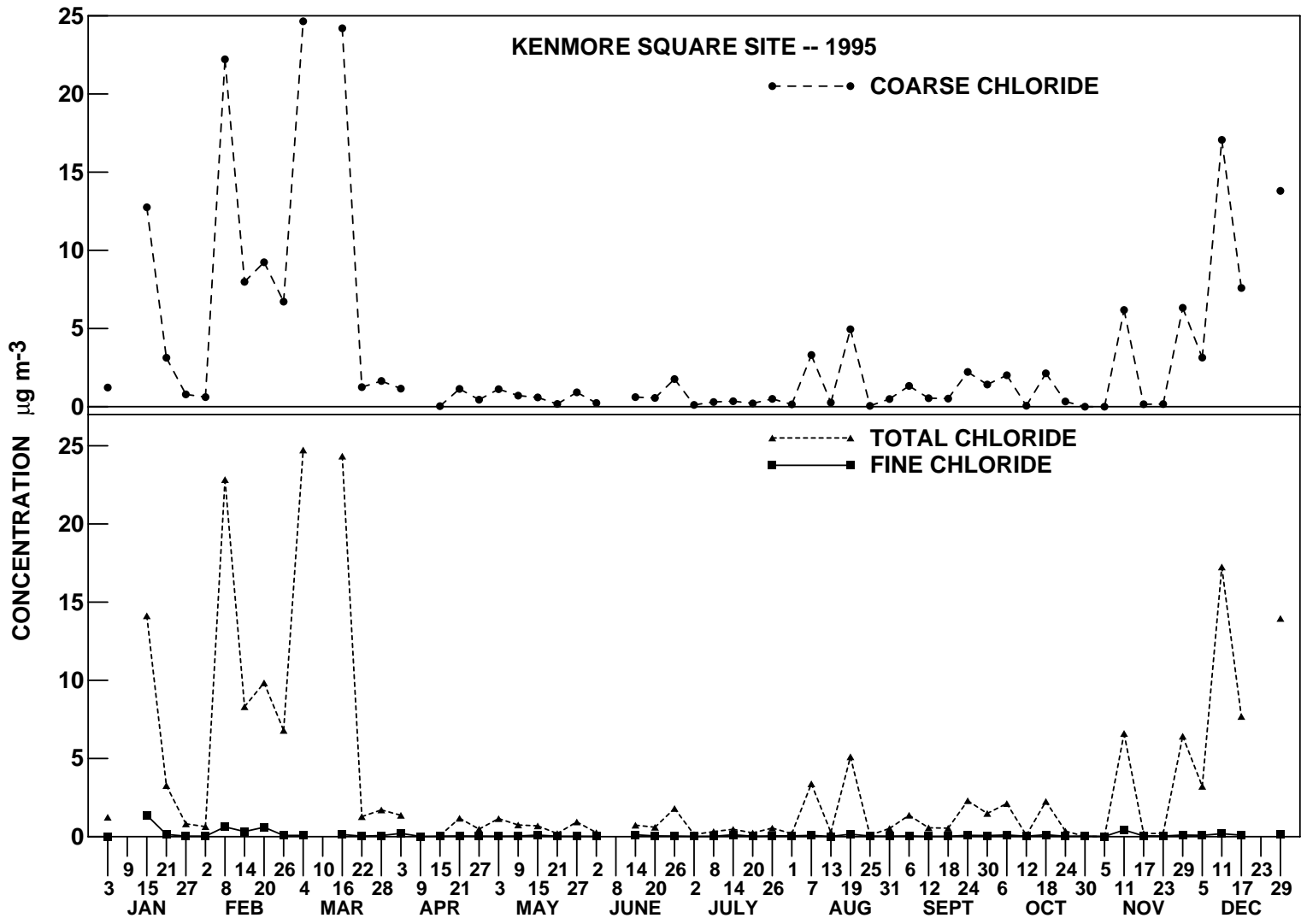
Figure 3.23 - Chloride Particle Concentrations at Kenmore Square, Boston, MA

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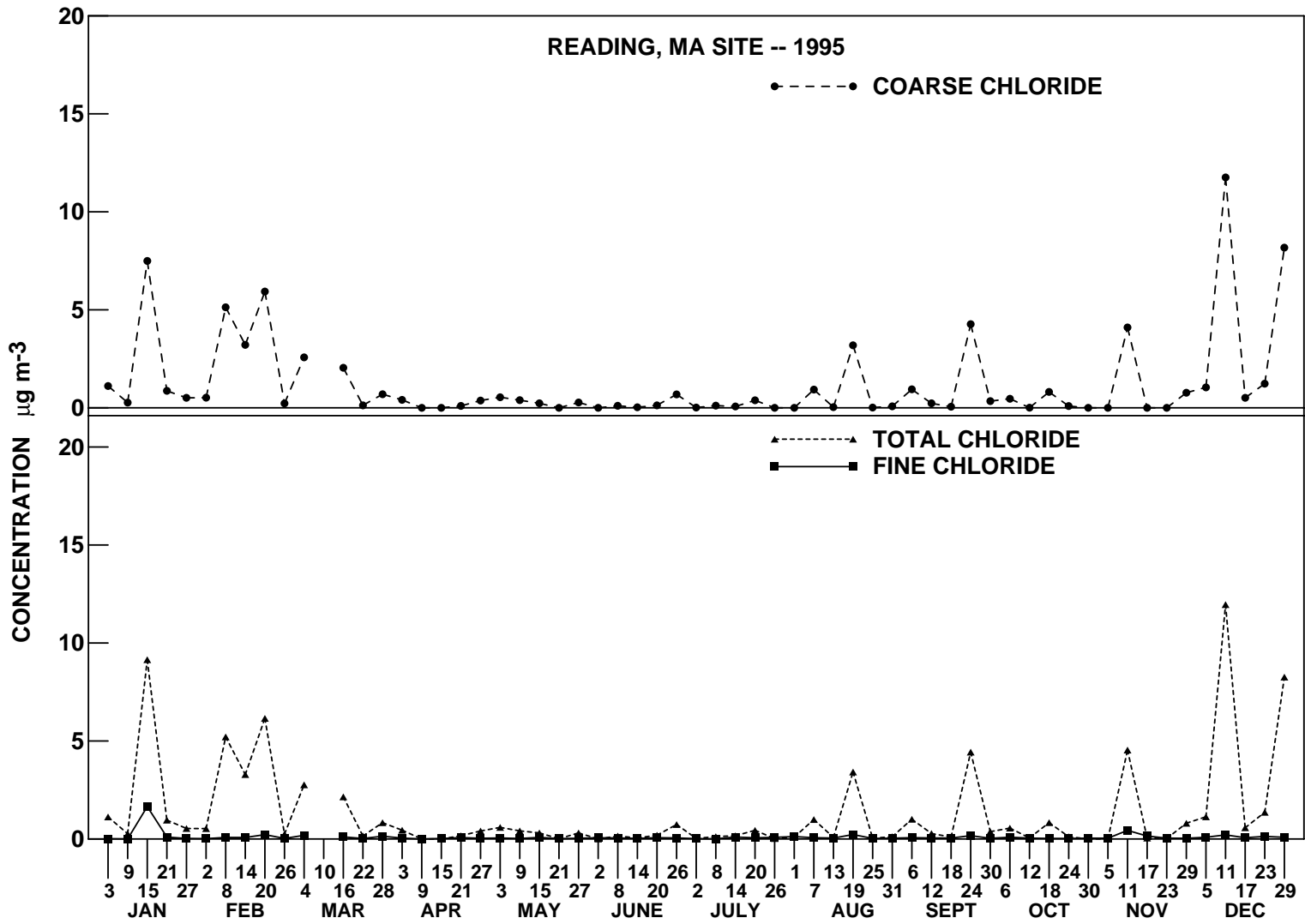
Figure 3.24 - Chloride Particle Concentrations at Reading, MA
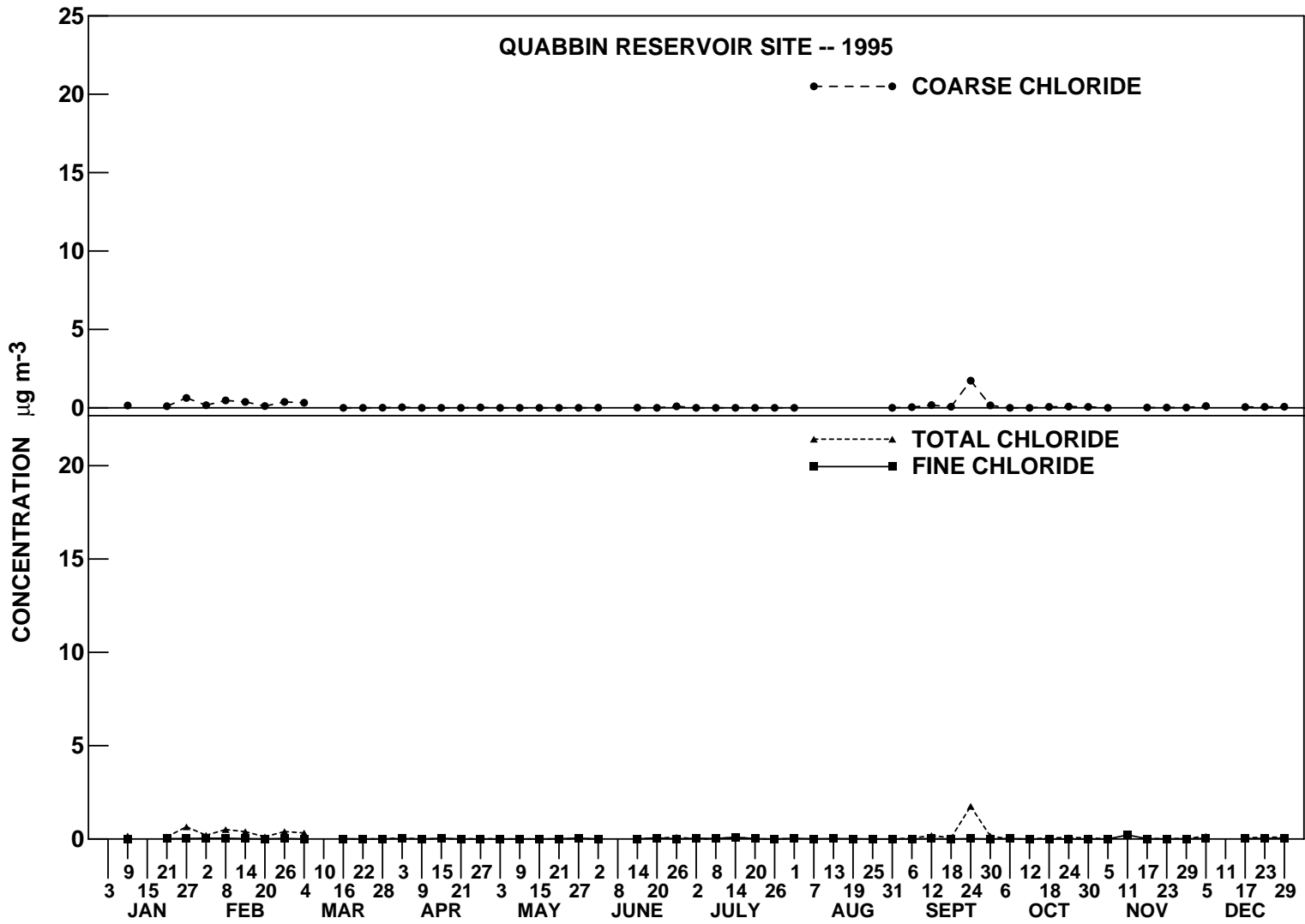
Figure 3.25 - Chloride Particle Concentrations at Quabbin Reservoir, MA

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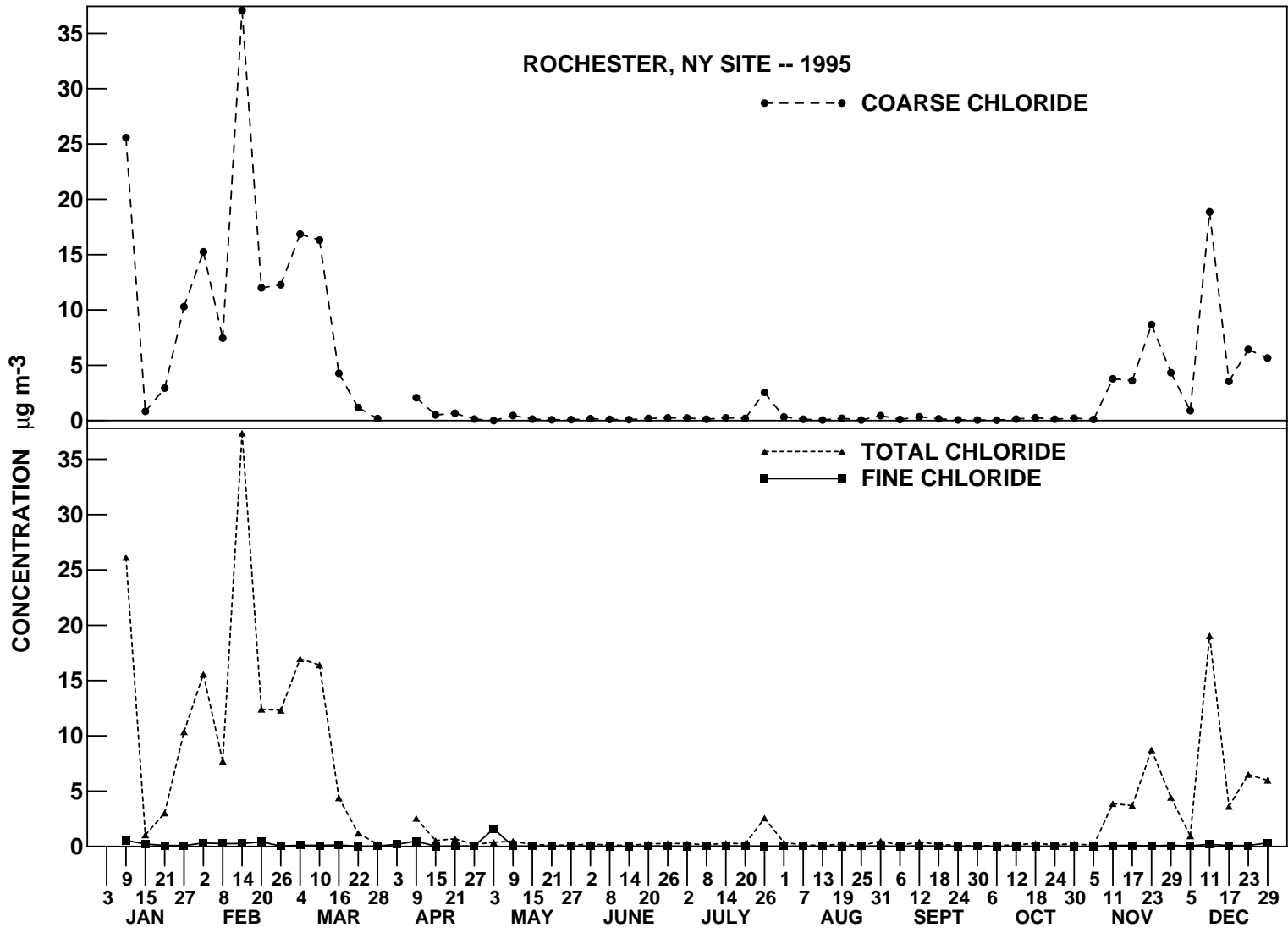
Figure 3.26 - Chloride Particle Concentrations at Rochester, NY
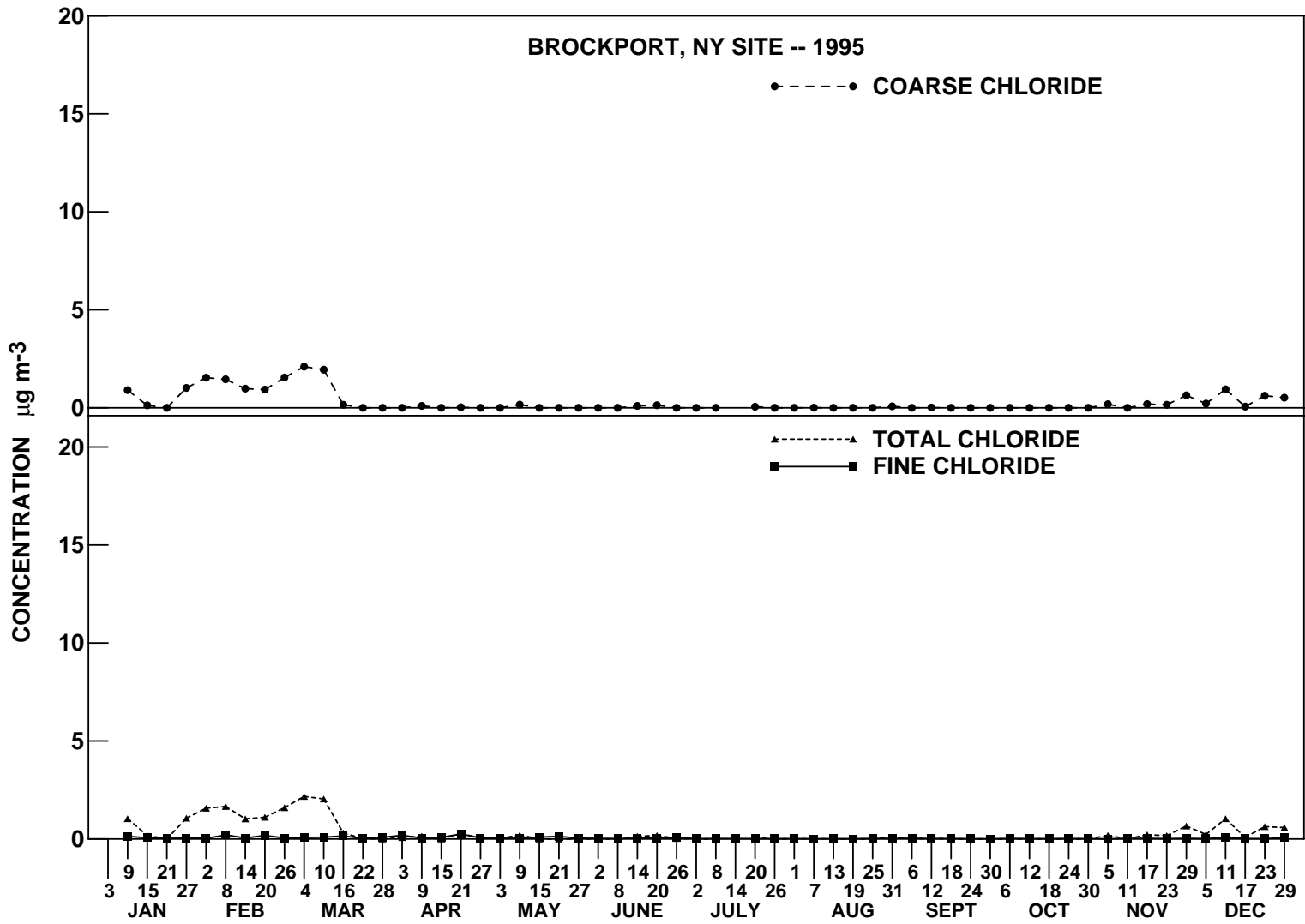
Figure 3.27 - Chloride Particle Concentrations at Brockport, NY

51
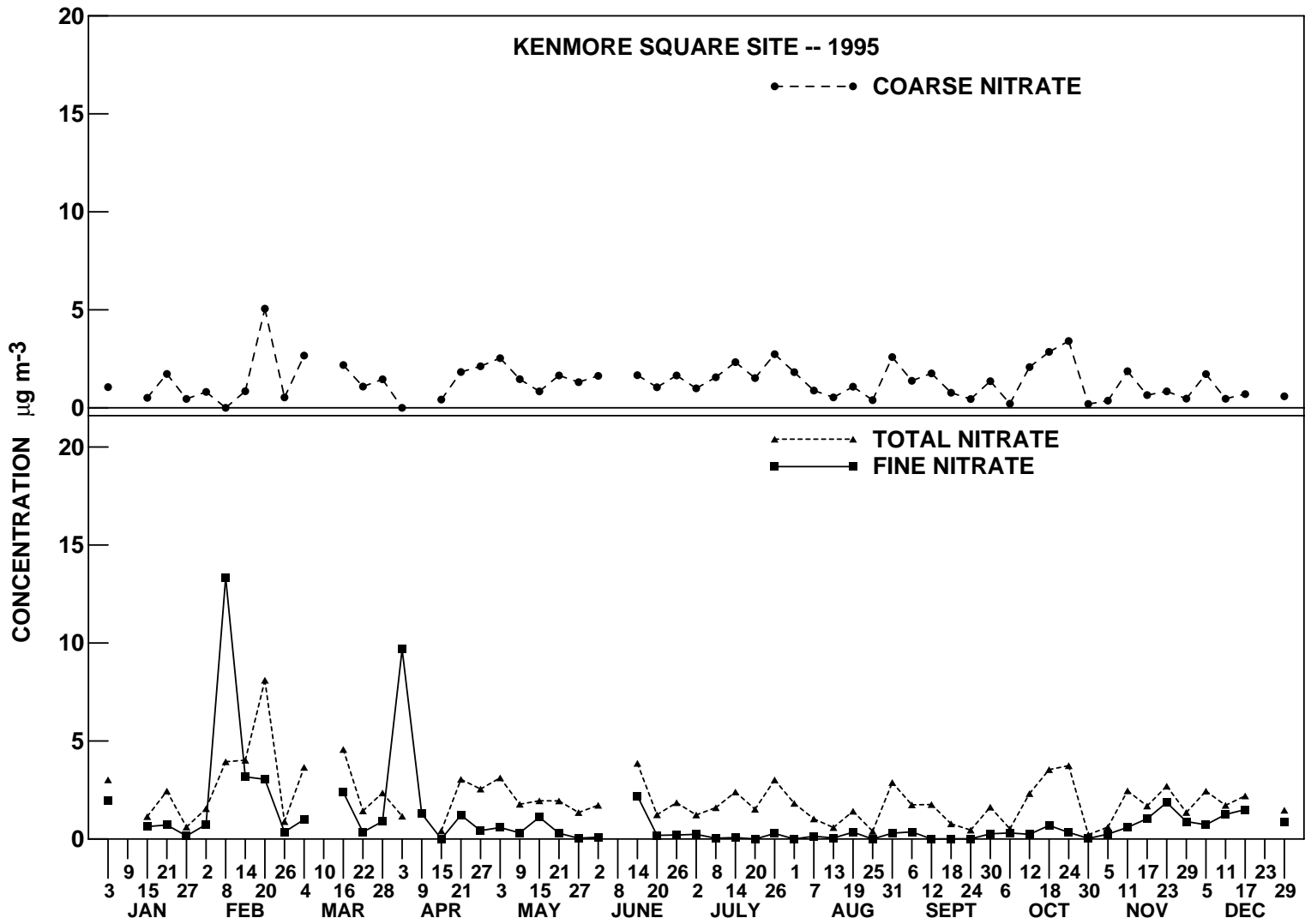
Figure 3.28 - Nitrate Particle Concentrations at Kenmore Square, Boston, MA
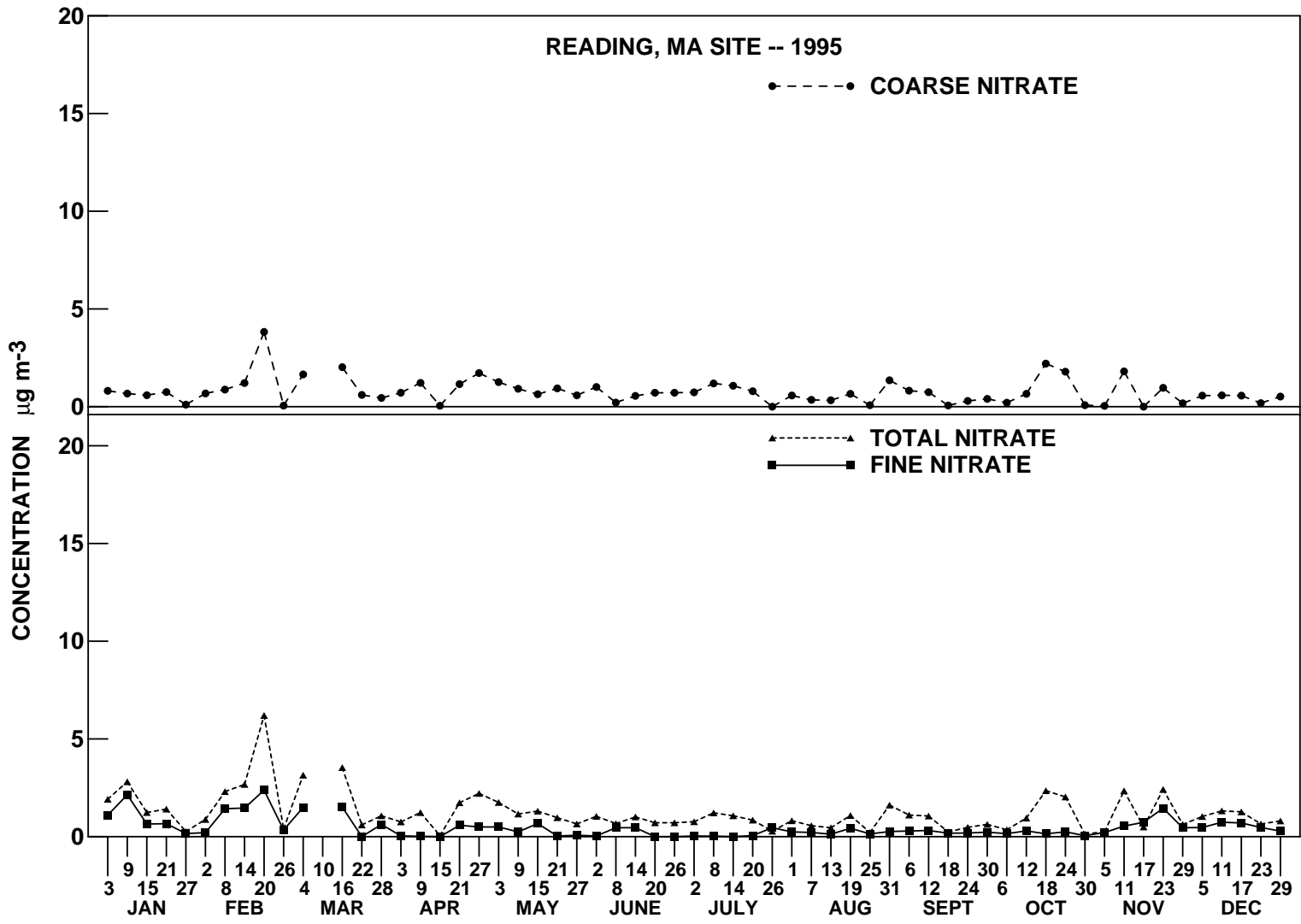
Figure 3.29 - Nitrate Particle Concentrations at Reading, MA

53
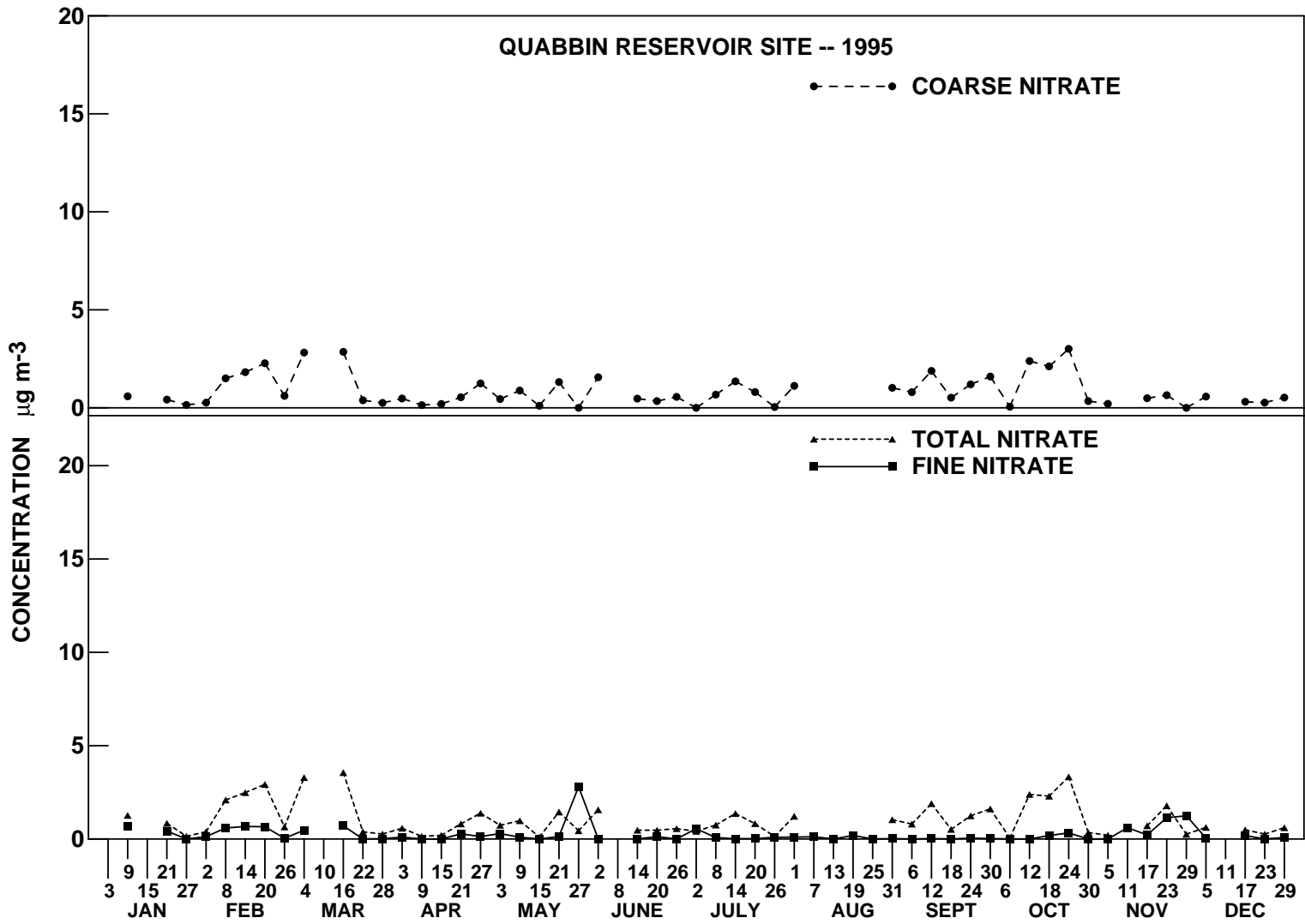
Figure 3.30 - Nitrate Particle Concentrations at Quabbin Reservoir, MA
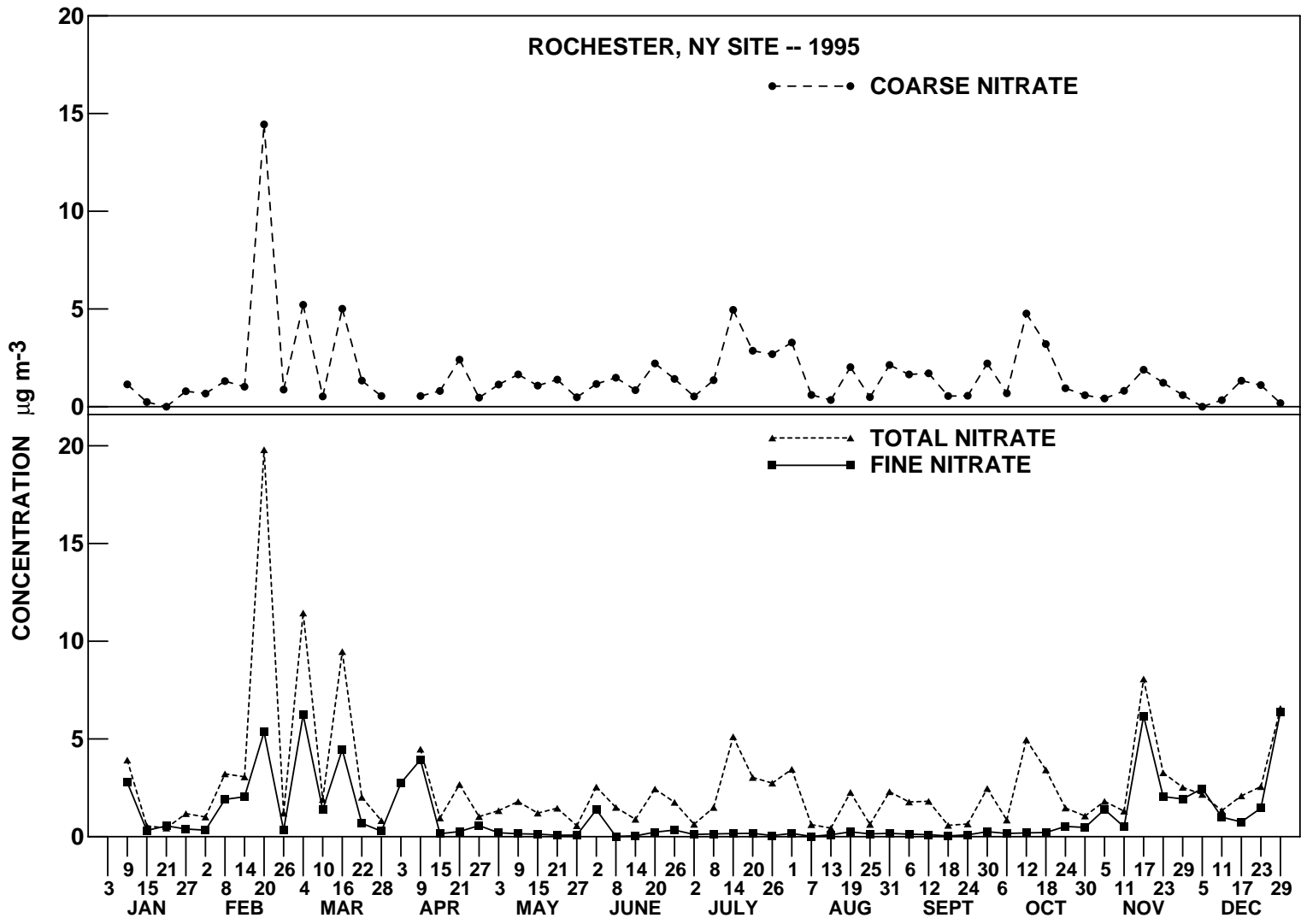
Figure 3.31 - Nitrate Particle Concentrations at Rochester, NY

Figure 3.32 - Nitrate Particle Concentrations at Brockport, NY
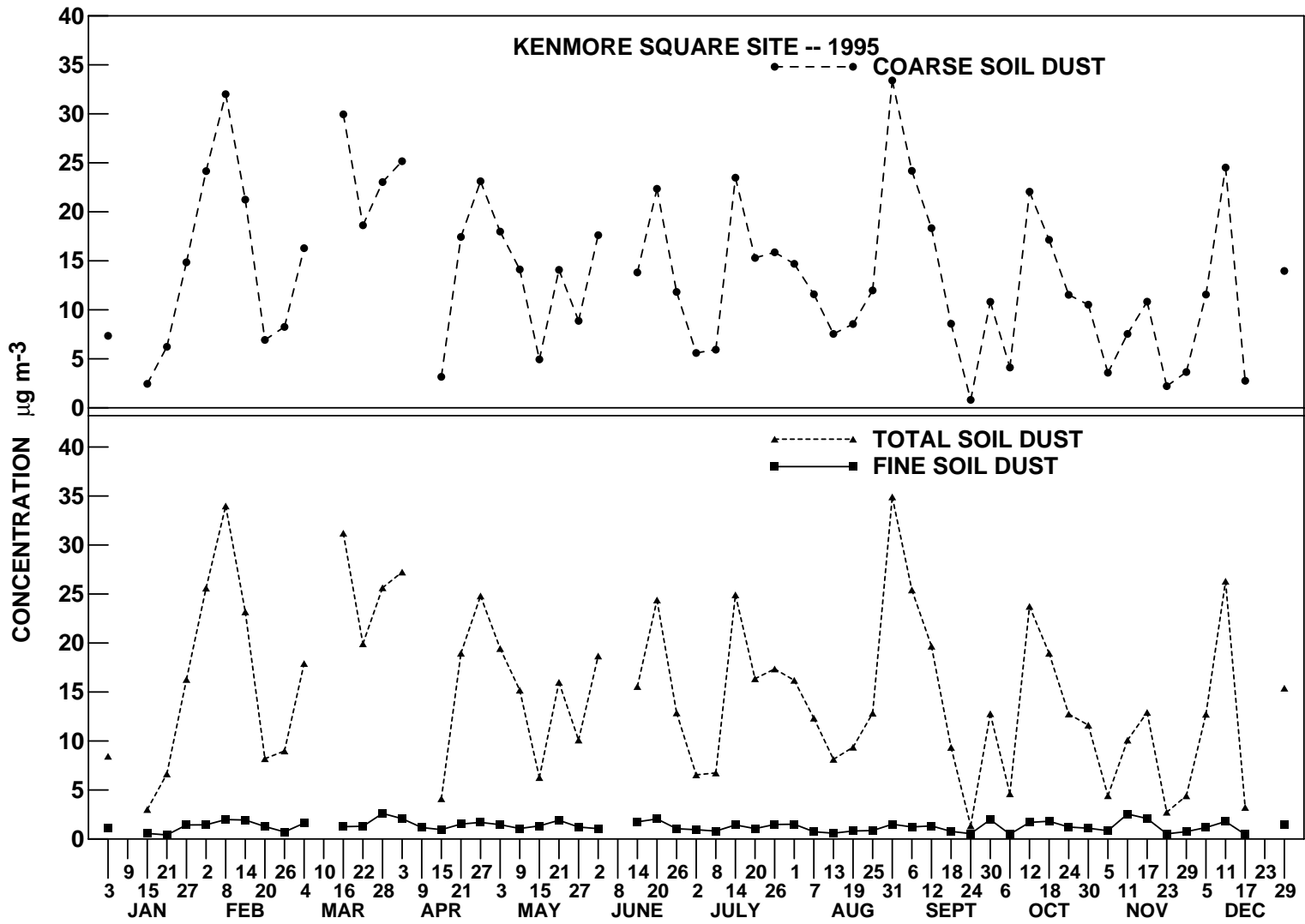
Figure 3.33 - Soil Dust Concentrations at Kenmore Square, Boston, MA

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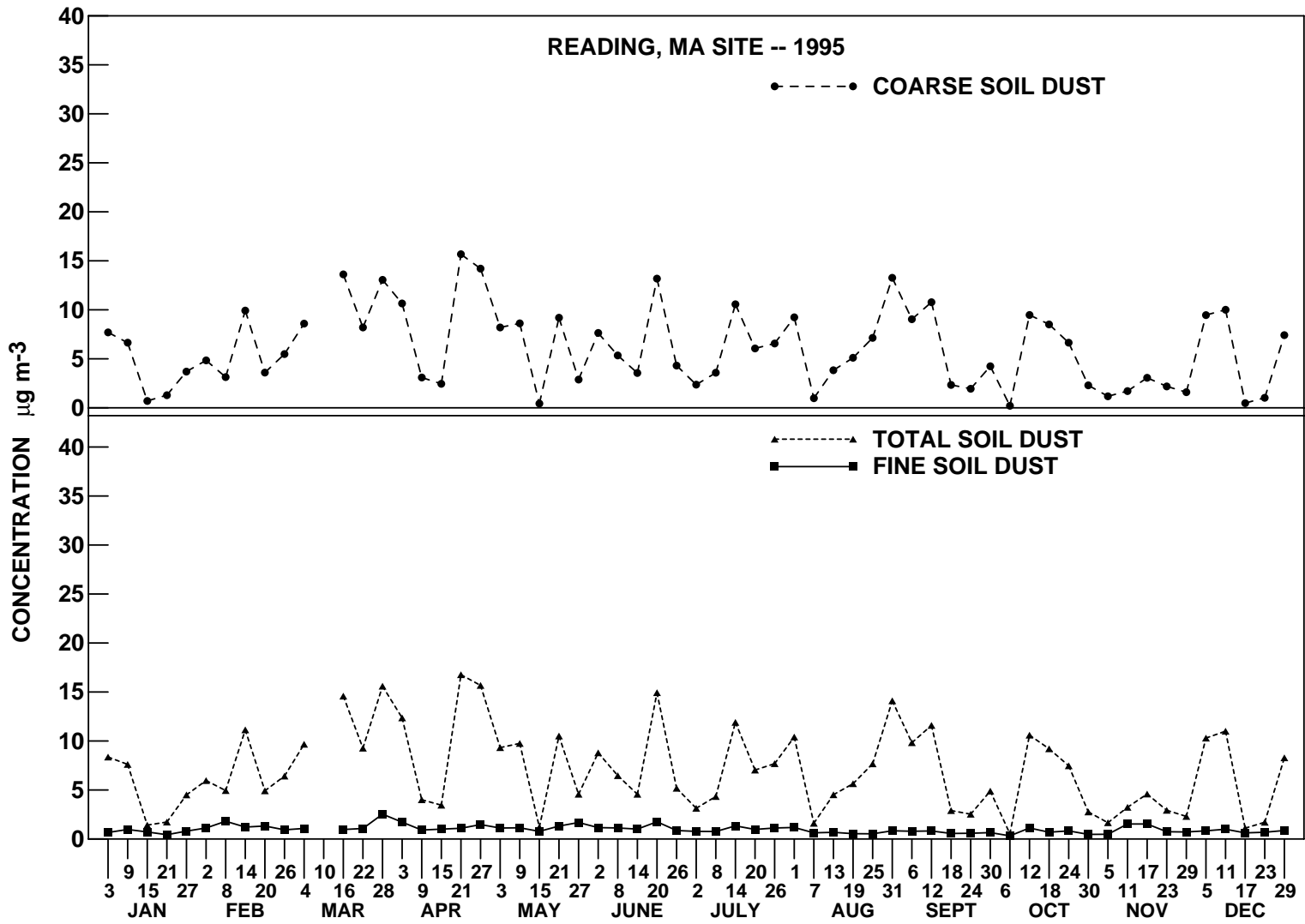
Figure 3.34 - Soil Dust Concentrations at Reading, MA
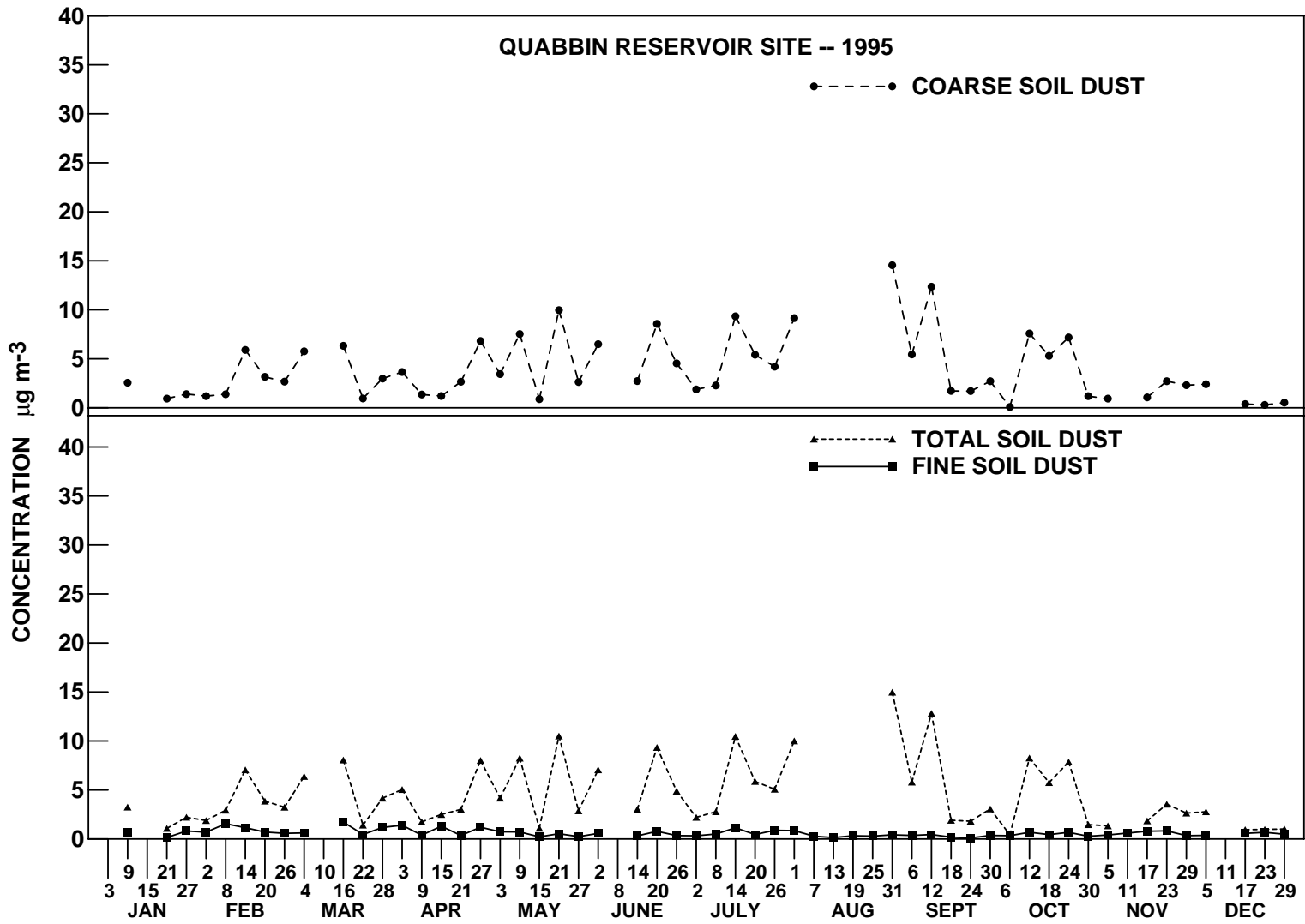
Figure 3.35 - Soil Dust Concentrations at Quabbin Reservoir, MA
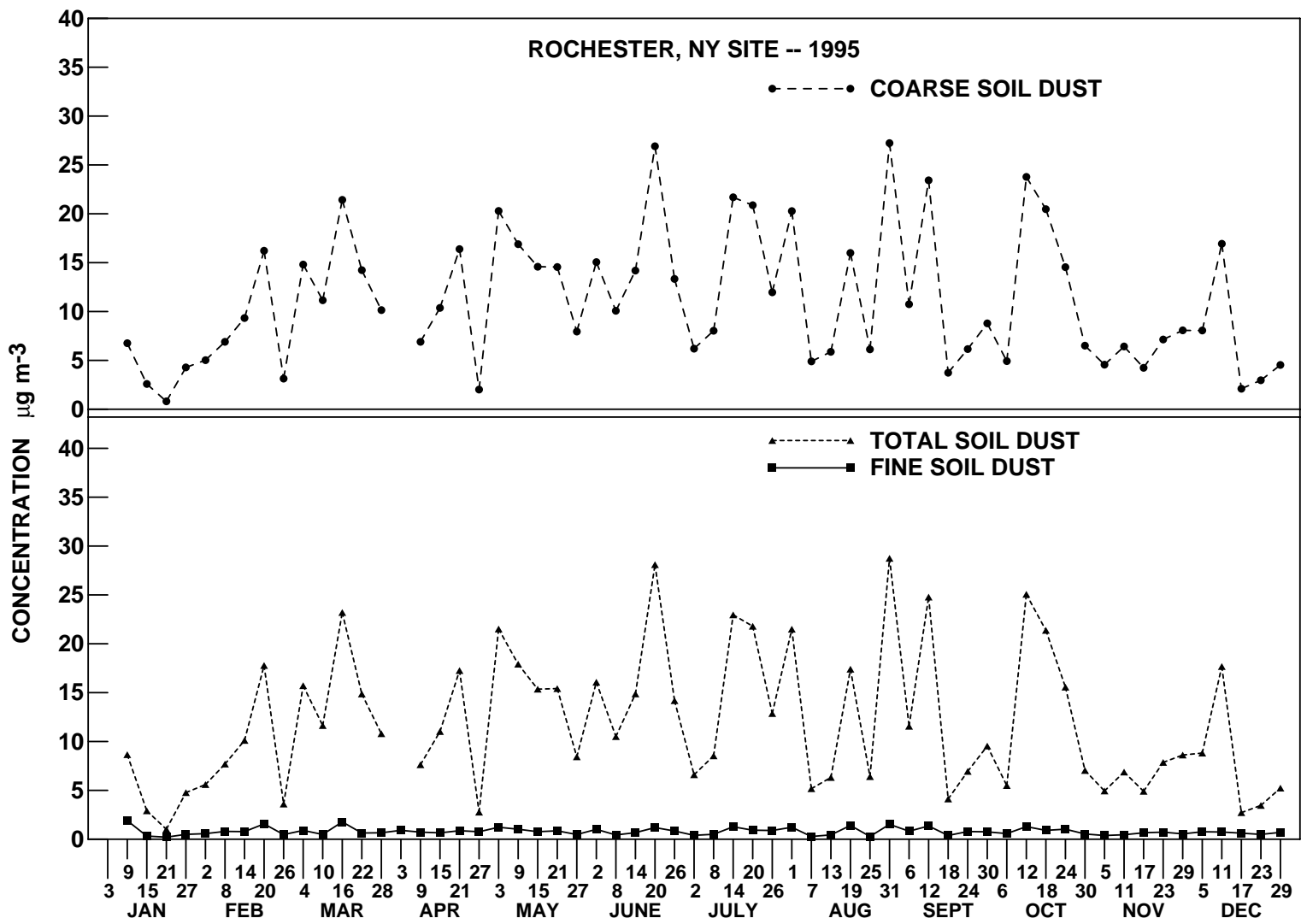
Figure 3.36 - Soil Dust Concentrations at Rochester, NY
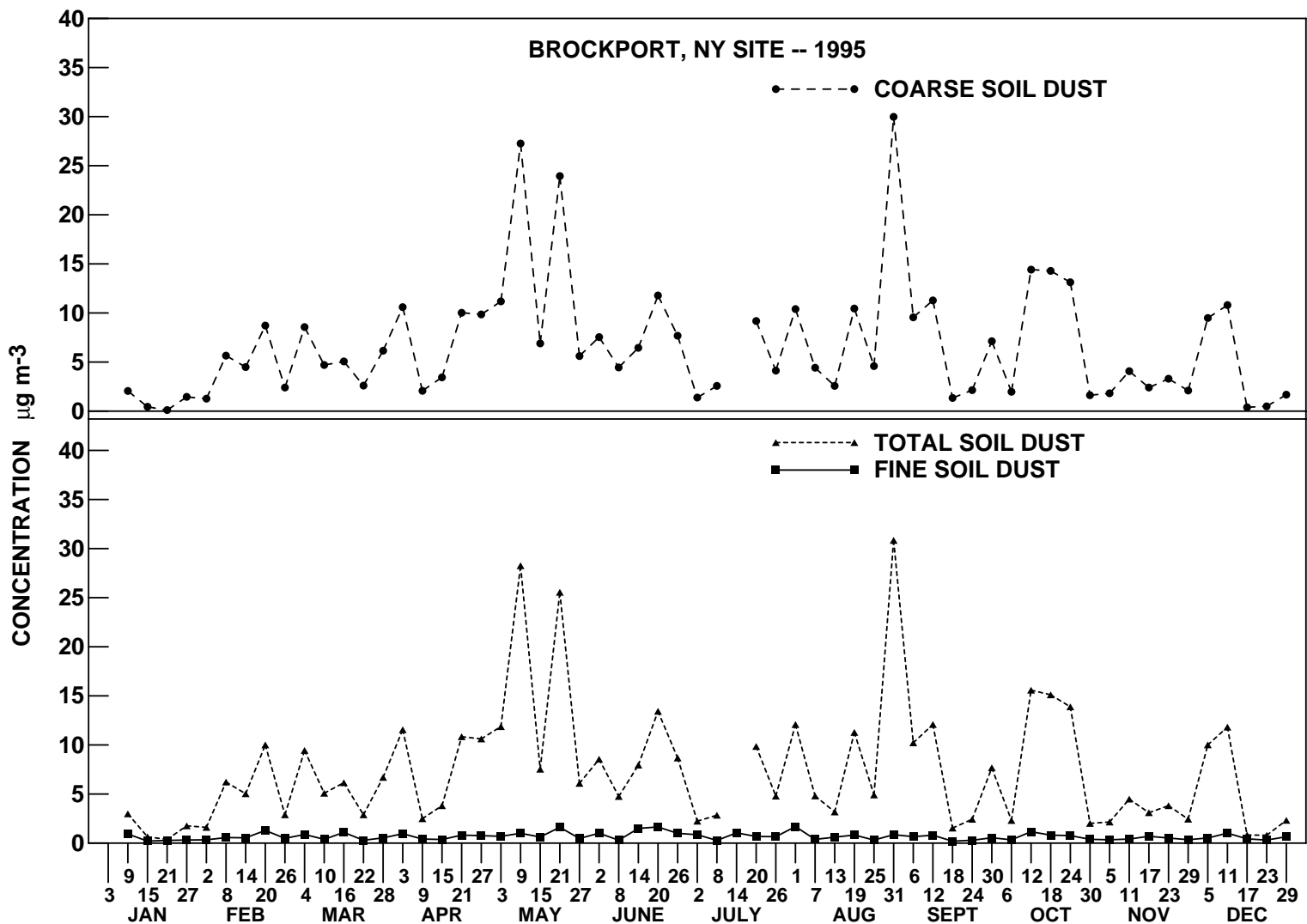
Figure 3.37 - Soil Dust Concentrations at Brockport, NY
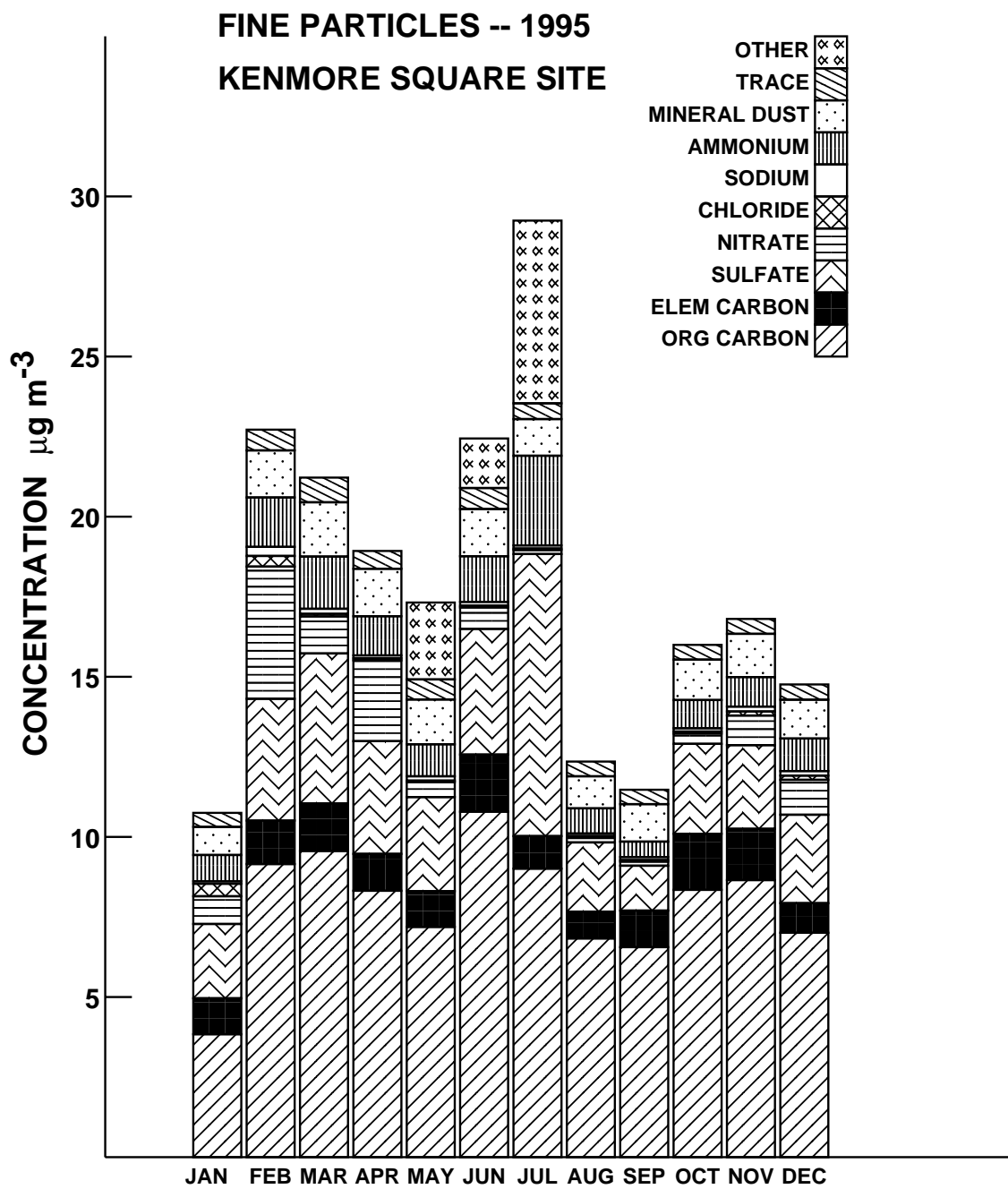
Figure 3.38 - Monthly Average Fine Particle Chemical Composition at Kenmore Square, Boston, MA

FINE PARTICLES -- 1995
READING SITE
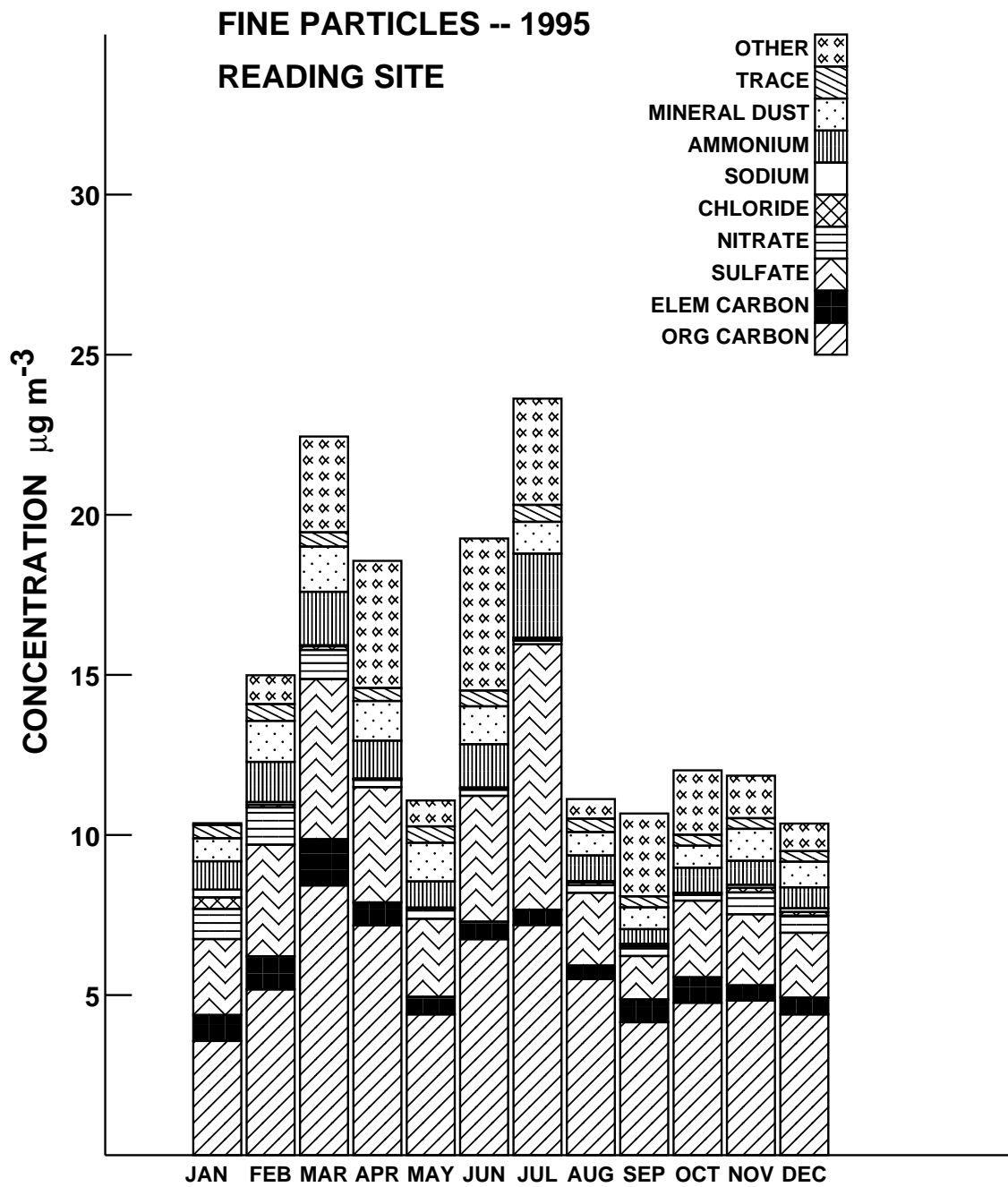
**Figure 3.39 - Monthly Average Fine Particle Chemical Composition
at Reading, MA**

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FINE PARTICLES -- 1995

QUABBIN RESERVOIR SITE

Legend:
OTHER
TRACE
MINERAL DUST
AMMONIUM
SODIUM
CHLORIDE
NITRATE
SULFATE
ELEM CARBON
ORG CARBON

CONCENTRATION μg m^{-3}

JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC
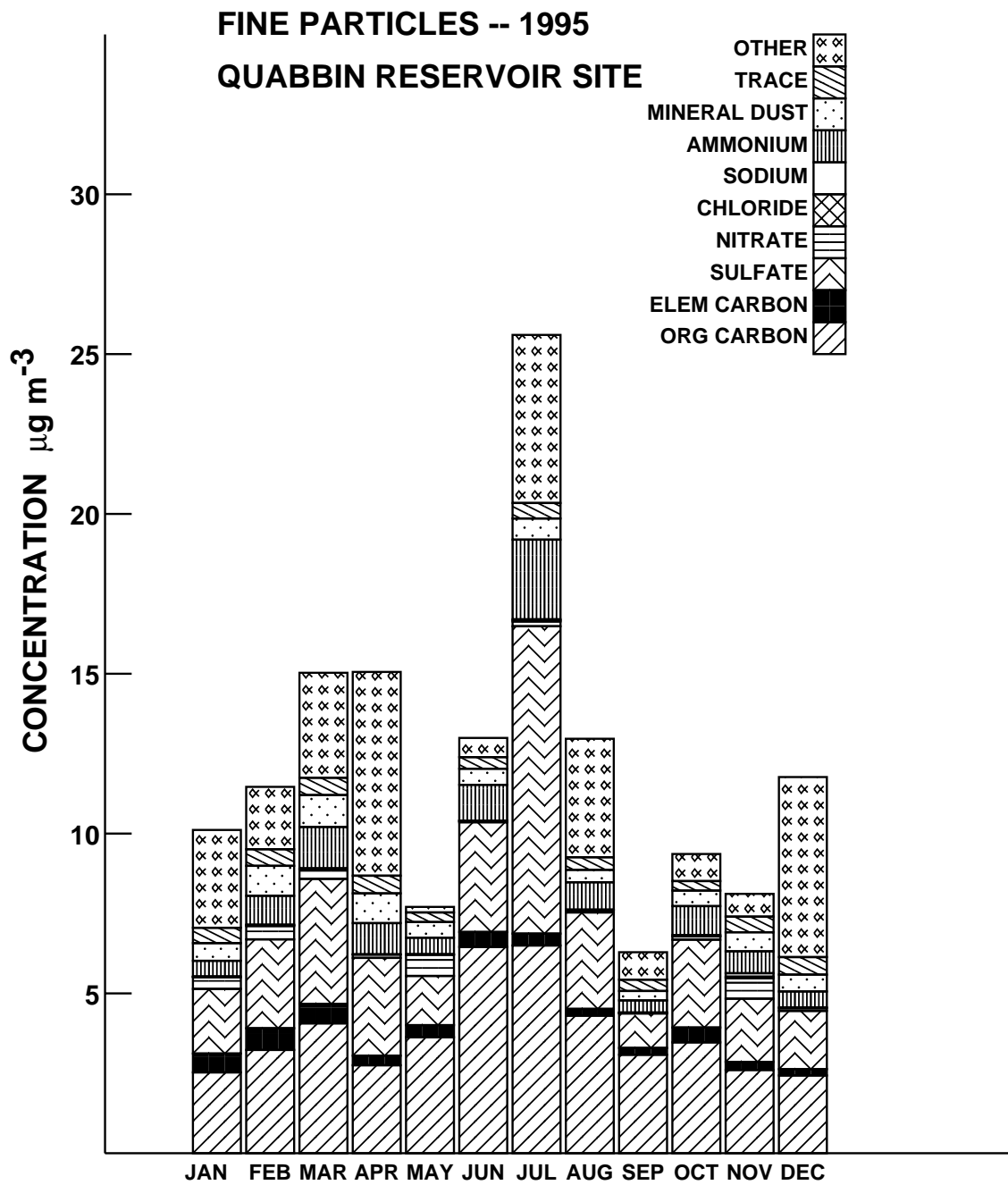
Figure 3.40 - Monthly Average Fine Particle Chemical Composition at Quabbin Reservoir, MA

FINE PARTICLES -- 1995
ROCHESTER SITE
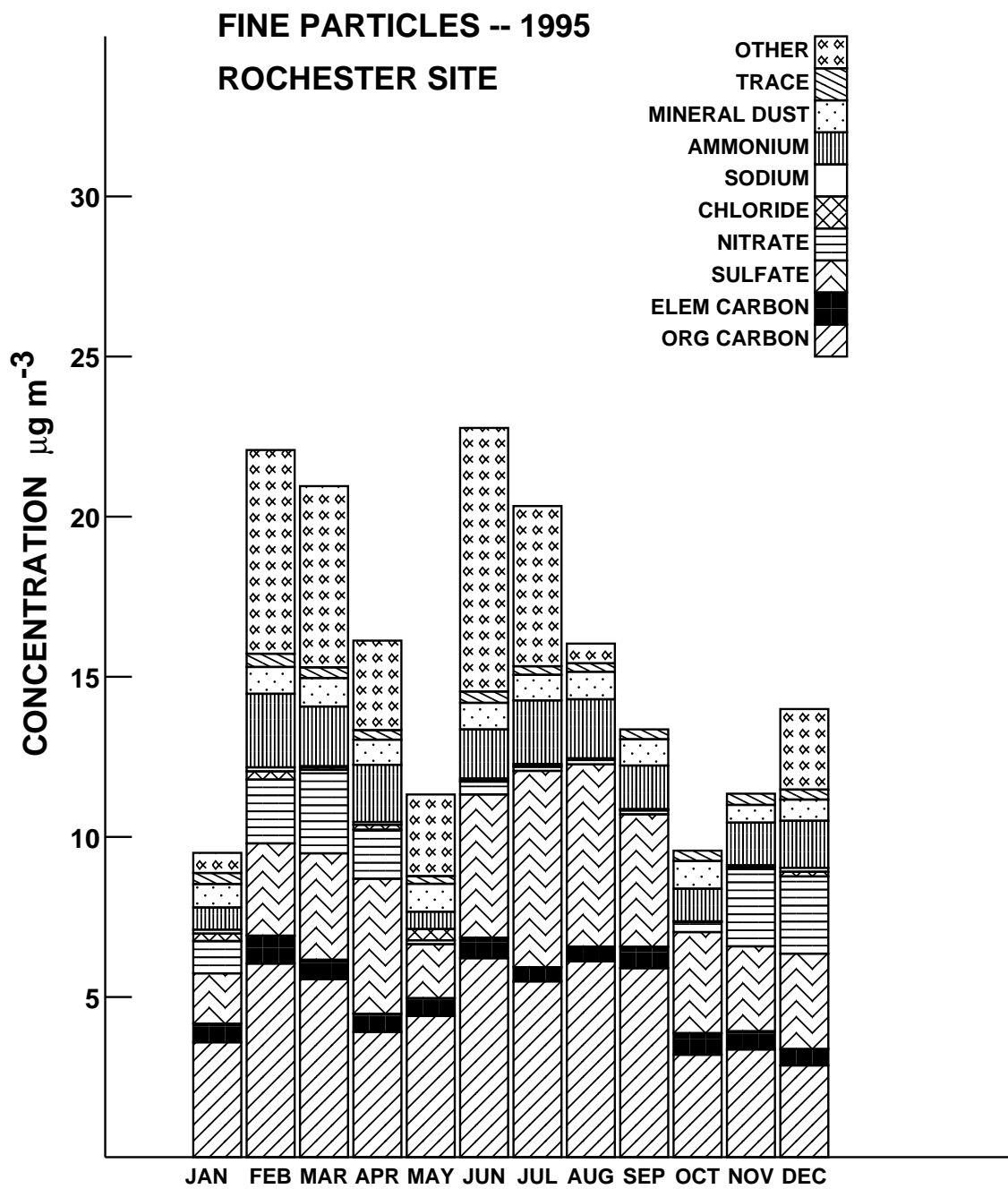
Figure 3.41 - Monthly Average Fine Particle Chemical Composition
at Rochester, NY

FINE PARTICLES -- 1995
BROCKPORT SITE

CONCENTRATION μg m⁻³

Legend:
- OTHER
- TRACE
- MINERAL DUST
- AMMONIUM
- SODIUM
- CHLORIDE
- NITRATE
- SULFATE
- ELEM CARBON
- ORG CARBON

JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC
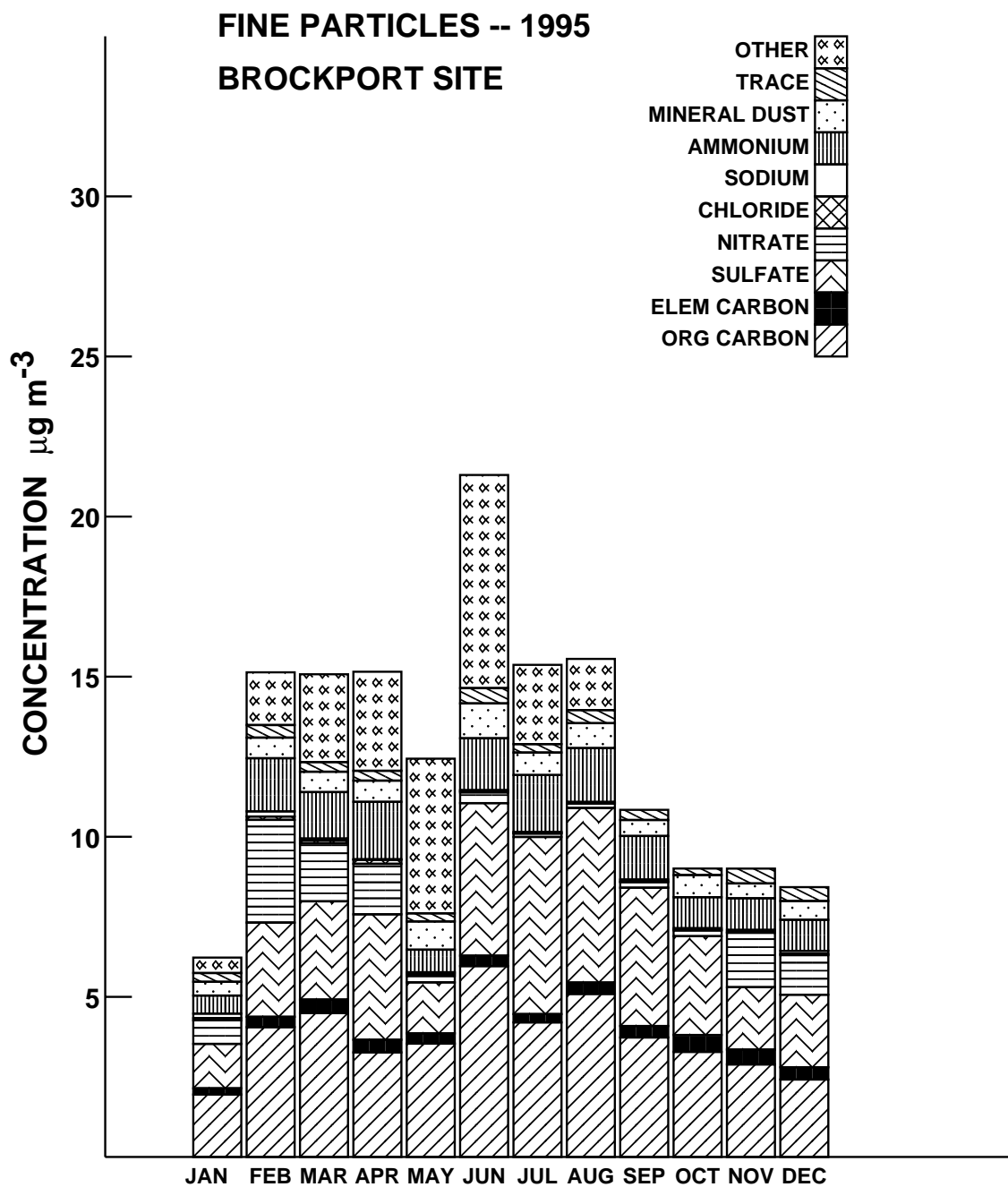
**Figure 3.42 - Monthly Average Fine Particle Chemical Composition
at Brockport, NY**
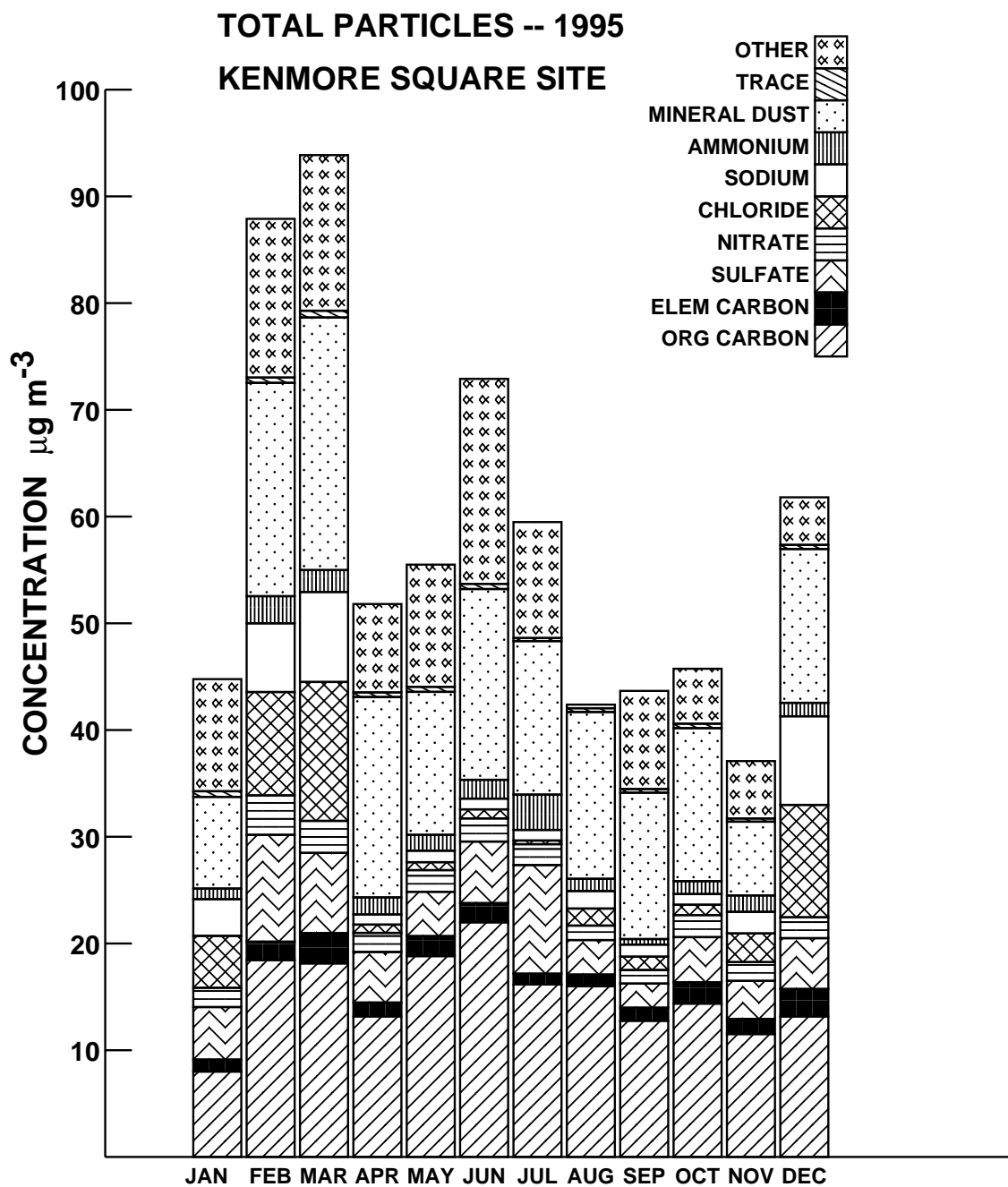
Figure 3.43 - Monthly Average Total Particle Chemical Composition
at Kenmore Square, Boston, MA

TOTAL PARTICLES -- 1995
READING SITE
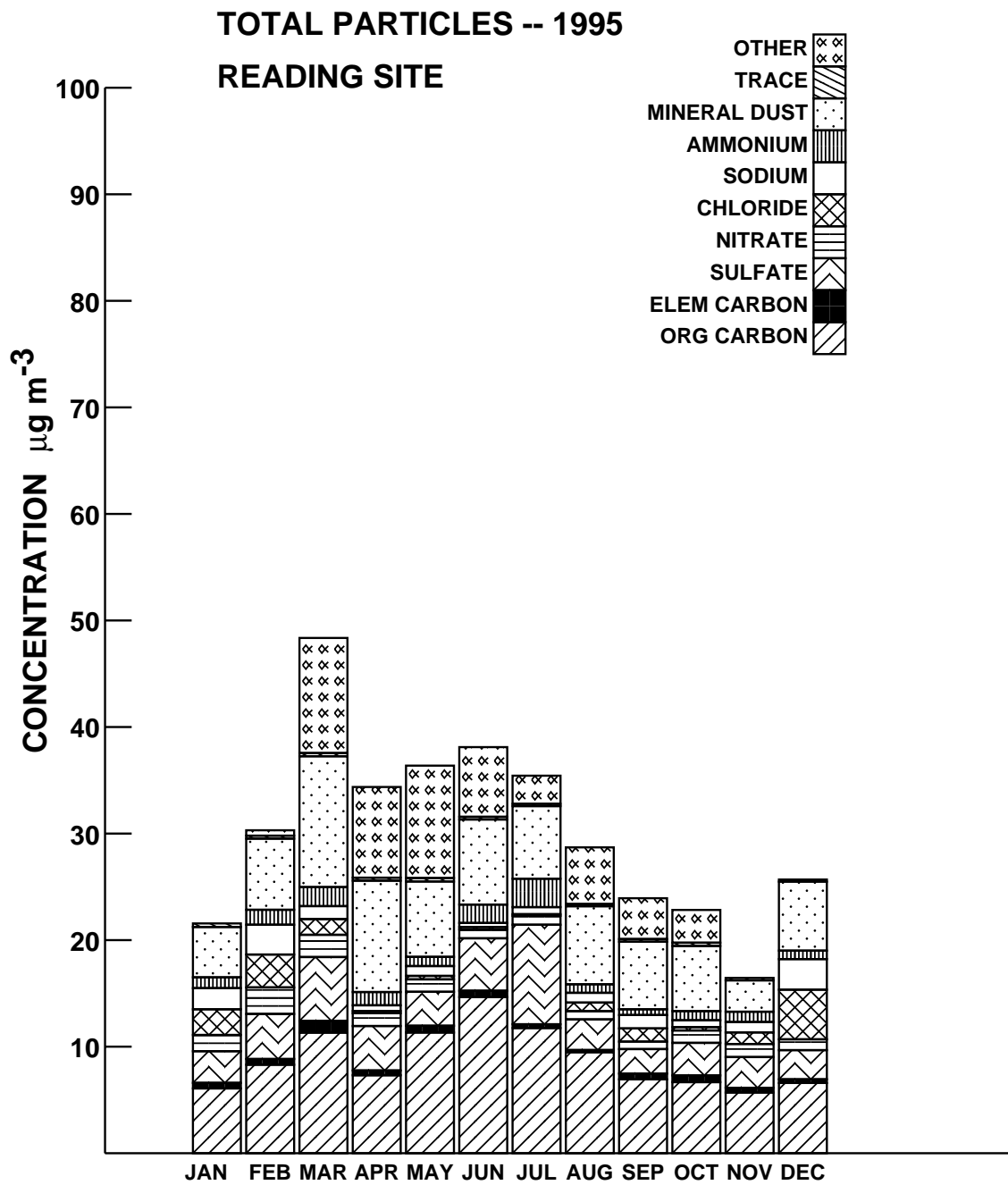
**Figure 3.44 - Monthly Average Total Particle Chemical Composition
at Reading, MA**

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TOTAL PARTICLES -- 1995

QUABBIN RESERVOIR SITE

Legend:
- OTHER
- TRACE
- MINERAL DUST
- AMMONIUM
- SODIUM
- CHLORIDE
- NITRATE
- SULFATE
- ELEM CARBON
- ORG CARBON

Y-axis: CONCENTRATION μg m^{-3}

X-axis: JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC
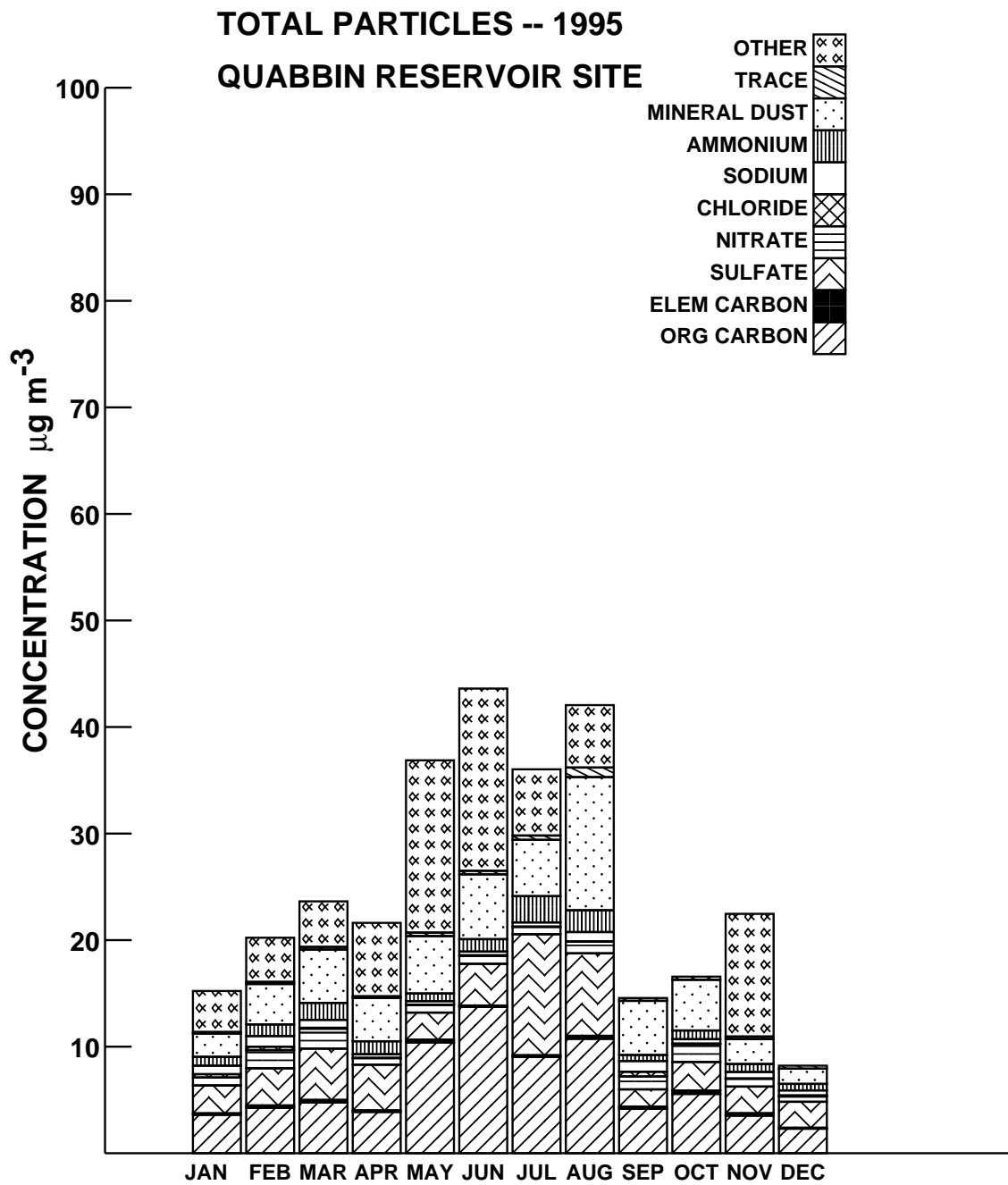
Figure 3.45 - Monthly Average Total Particle Chemical Composition at Quabbin Reservoir, MA

TOTAL PARTICLES -- 1995
ROCHESTER SITE

CONCENTRATION μg m⁻³

Legend:
OTHER
TRACE
MINERAL DUST
AMMONIUM
SODIUM
CHLORIDE
NITRATE
SULFATE
ELEM CARBON
ORG CARBON

JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC
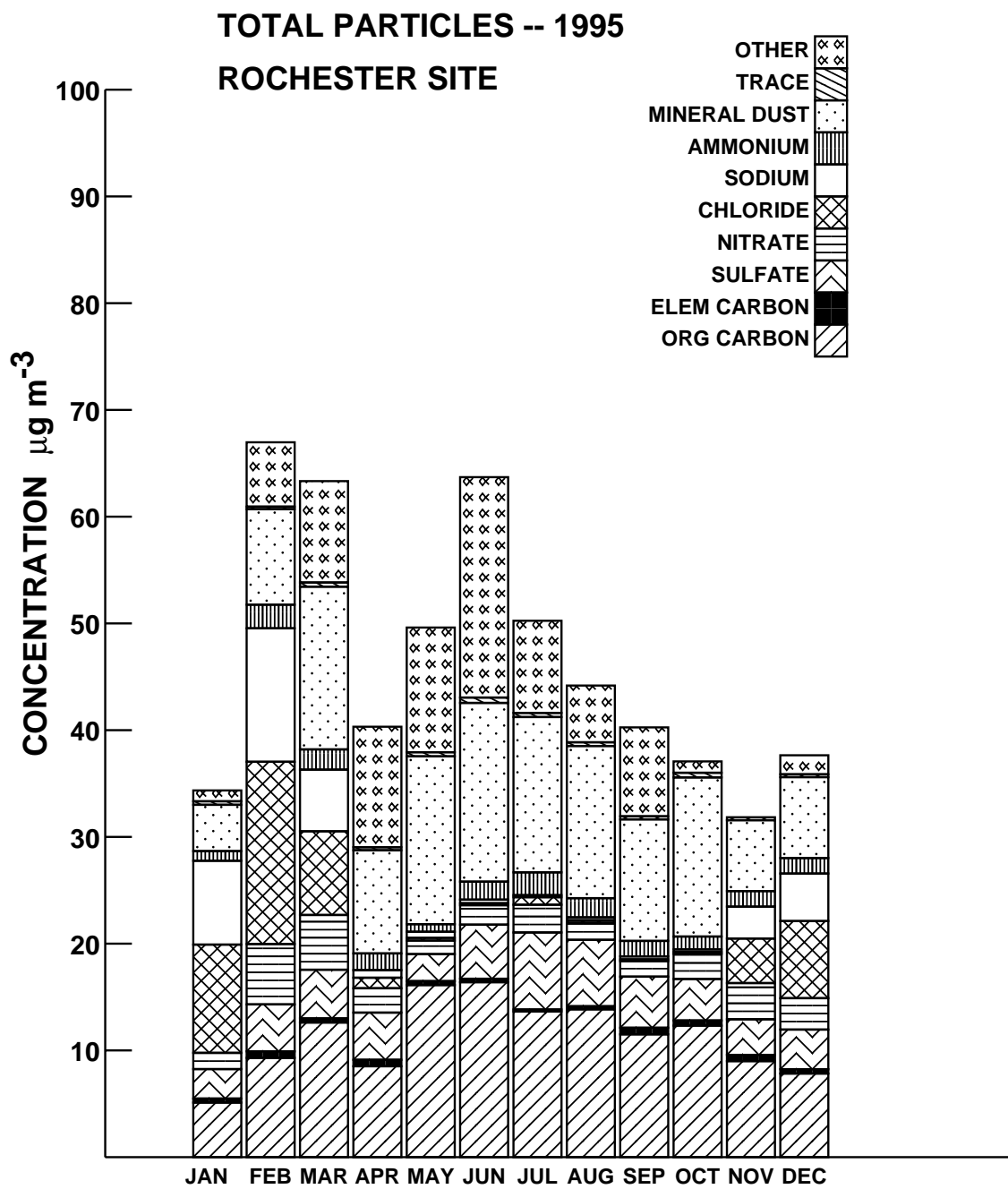
Figure 3.46 - Monthly Average Total Particle Chemical Composition at Rochester, NY

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TOTAL PARTICLES -- 1995
BROCKPORT SITE
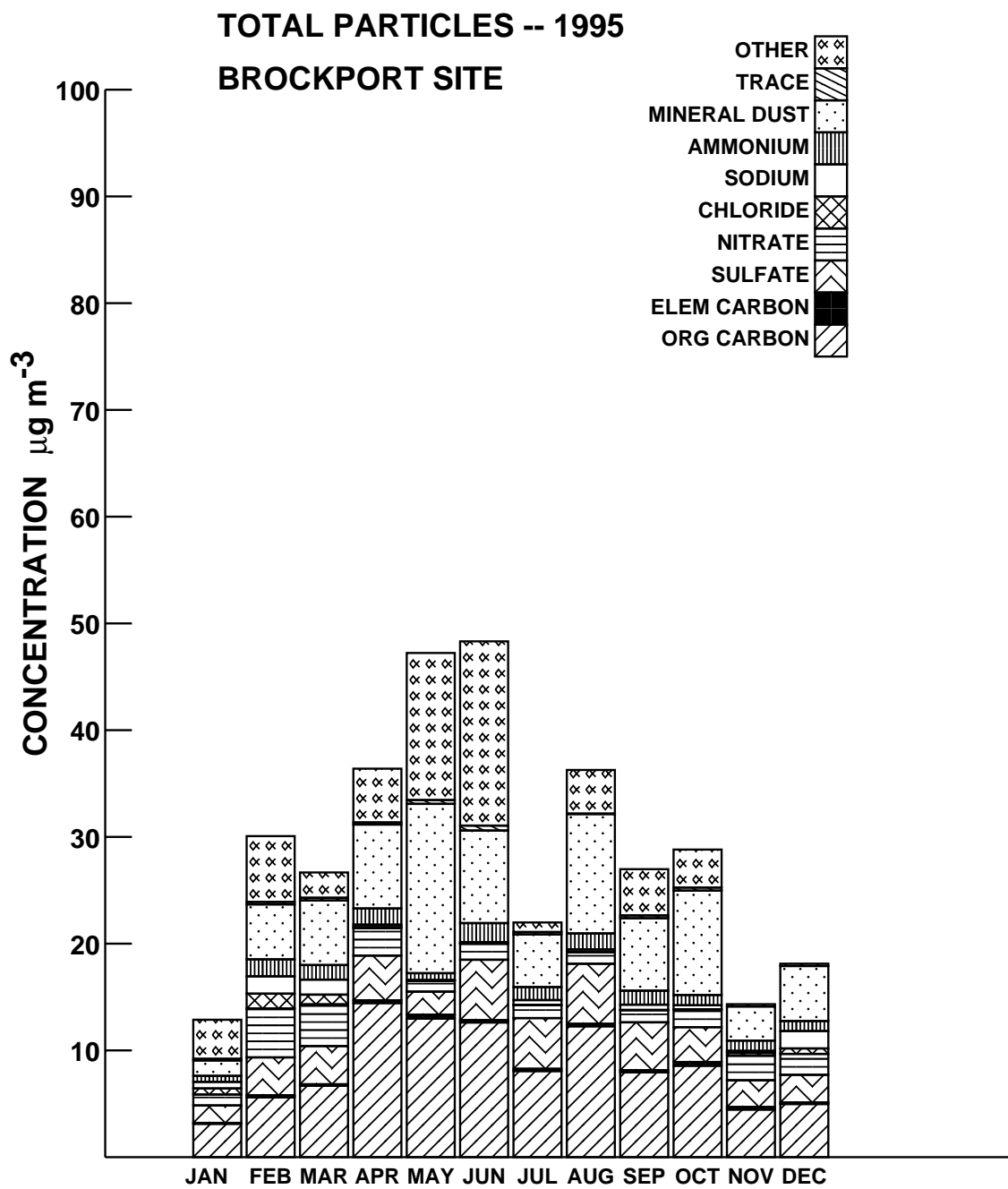
Figure 3.47 - Monthly Average Total Particle Chemical Composition
at Brockport, NY

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Table 3.2. Monthly average chemical composition of fine particle species (μg m^{-3}).

Month	Mass	Organics	EC	SO$_4$	NO$_3$	Cl	Na	NH$_4$	Crustal	Trace	Other
Kenmore Square:											
Jan	9.5	3.8	1.1	2.3	0.9	0.4	0.1	0.8	0.9	0.4	0.0
Feb	22.0	9.2	1.4	3.8	4.1	0.3	0.3	1.5	1.5	0.6	0.0
Mar	20.2	9.6	1.5	4.7	1.2	0.1	0.2	1.6	1.7	0.8	0.0
Apr	18.3	8.3	1.2	3.5	2.5	0.1	0.1	1.2	1.5	0.6	0.0
May	17.3	7.2	1.1	2.9	0.5	0.0	0.1	1.0	1.4	0.6	2.4
Jun	22.4	10.8	1.8	3.9	0.7	0.1	0.1	1.4	1.5	0.7	1.5
Jul	29.2	9.0	1.0	8.8	0.1	0.0	0.1	2.8	1.1	0.5	5.7
Aug	12.2	6.8	0.8	2.2	0.1	0.1	0.1	0.8	1.0	0.5	0.0
Sep	7.5	6.6	1.1	1.4	0.1	0.0	0.1	0.5	1.2	0.4	0.0
Oct	10.9	8.3	1.8	2.8	0.3	0.1	0.1	0.9	1.3	0.5	0.0
Nov	13.3	8.7	1.6	2.6	0.9	0.1	0.2	0.9	1.4	0.5	0.0
Dec	12.3	7.0	0.9	2.8	1.1	0.1	0.1	1.0	1.2	0.5	0.0
Reading:											
Jan	10.4	3.6	0.8	2.4	0.9	0.4	0.2	0.9	0.7	0.4	0.1
Feb	15.0	5.2	1.0	3.5	1.2	0.1	0.1	1.3	1.3	0.5	0.9
Mar	22.4	8.4	1.4	5.0	0.9	0.1	0.0	1.7	1.4	0.4	3.0
Apr	18.6	7.2	0.7	3.6	0.2	0.0	0.0	1.2	1.2	0.4	4.0
May	11.1	4.4	0.6	2.4	0.3	0.0	0.0	0.8	1.2	0.5	0.8
Jun	19.3	6.7	0.5	3.9	0.2	0.1	0.0	1.4	1.2	0.5	4.7
Jul	23.6	7.2	0.5	8.3	0.1	0.1	0.0	2.6	1.0	0.5	3.3
Aug	11.1	5.5	0.4	2.3	0.2	0.1	0.0	0.8	0.7	0.4	0.6
Sep	10.7	4.2	0.7	1.4	0.2	0.1	0.1	0.5	0.7	0.3	2.6
Oct	12.0	4.8	0.8	2.4	0.2	0.0	0.0	0.8	0.7	0.3	2.0
Nov	11.9	4.8	0.5	2.2	0.7	0.1	0.1	0.7	1.0	0.3	1.3
Dec	10.4	4.4	0.5	2.0	0.5	0.1	0.1	0.7	0.8	0.3	0.9
Quabbin Reservoir:											
Jan	10.1	2.5	0.6	2.0	0.4	0.0	0.0	0.5	0.6	0.5	3.1
Feb	11.5	3.2	0.7	2.8	0.4	0.0	0.0	0.9	0.9	0.5	2.0
Mar	15.0	4.1	0.6	3.9	0.3	0.0	0.0	1.3	1.0	0.5	3.3
Apr	15.1	2.8	0.3	3.1	0.1	0.0	0.0	1.0	0.9	0.6	6.4
May	7.7	3.6	0.4	1.5	0.7	0.0	0.0	0.5	0.5	0.3	0.2
Jun	13.0	6.5	0.5	3.4	0.0	0.0	0.0	1.1	0.5	0.4	0.6
Jul	25.6	6.5	0.4	9.6	0.1	0.0	0.0	2.5	0.7	0.5	5.3
Aug	13.0	4.3	0.2	3.0	0.1	0.0	0.0	0.9	0.4	0.4	3.7
Sep	6.3	3.1	0.2	1.1	0.0	0.0	0.0	0.4	0.3	0.4	0.9
Oct	9.4	3.5	0.5	2.7	0.1	0.0	0.0	0.9	0.5	0.3	0.8
Nov	8.1	2.6	0.2	2.0	0.6	0.0	0.1	0.7	0.6	0.5	0.7
Dec	11.8	2.4	0.2	1.8	0.1	0.0	0.0	0.5	0.5	0.5	5.6

Table 3.2 (con'd). Monthly average chemical composition of fine particle species (μg m^{-3}).

Month	Mass	Organics	EC	SO$_4$	NO$_3$	Cl	Na	NH$_4$	Crustal	Trace	Other
Rochester:											
Jan	9.5	3.6	0.6	1.6	1.0	0.2	0.1	0.7	0.7	0.3	0.6
Feb	22.1	6.0	0.9	2.9	2.0	0.3	0.1	2.3	0.8	0.4	6.4
Mar	21.0	5.6	0.6	3.3	2.6	0.1	0.0	1.9	0.9	0.3	5.7
Apr	16.1	3.9	0.6	4.2	1.5	0.2	0.1	1.8	0.8	0.3	2.8
May	11.3	4.4	0.6	1.7	0.1	0.4	0.0	0.5	0.9	0.2	2.5
Jun	22.8	6.2	0.6	4.5	0.4	0.0	0.1	1.5	0.8	0.4	8.2
Jul	20.3	5.5	0.4	6.1	0.1	0.0	0.1	2.0	0.8	0.3	5.0
Aug	16.0	6.1	0.5	5.7	0.1	0.0	0.0	1.8	0.9	0.3	0.6
Sep	8.9	5.7	0.7	4.1	0.1	0.0	0.0	1.4	0.8	0.3	0.0
Oct	6.8	3.2	0.7	3.2	0.3	0.0	0.0	1.0	0.9	0.3	0.0
Nov	8.9	3.4	0.6	2.7	2.4	0.1	0.1	1.3	0.6	0.4	0.0
Dec	14.0	2.9	0.5	3.0	2.4	0.1	0.1	1.5	0.7	0.3	2.5
Brockport:											
Jan	6.2	2.0	0.2	1.4	0.7	0.1	0.1	0.6	0.4	0.3	0.5
Feb	15.1	4.1	0.3	2.9	3.2	0.1	0.2	1.7	0.6	0.4	1.6
Mar	15.1	4.5	0.4	3.1	1.8	0.1	0.1	1.5	0.6	0.3	2.7
Apr	15.2	3.3	0.4	3.9	1.6	0.1	0.0	1.8	0.7	0.3	3.1
May	12.4	3.5	0.3	1.6	0.2	0.1	0.0	0.7	0.9	0.3	4.8
Jun	21.3	6.0	0.3	4.8	0.3	0.0	0.0	1.6	1.1	0.5	6.7
Jul	15.4	4.2	0.3	5.5	0.1	0.0	0.0	1.8	0.7	0.3	2.5
Aug	15.6	5.1	0.4	5.5	0.1	0.0	0.0	1.7	0.8	0.4	1.6
Sep	9.1	3.7	0.4	4.3	0.2	0.0	0.1	1.4	0.5	0.3	0.0
Oct	8.9	3.3	0.5	3.1	0.2	0.0	0.1	1.0	0.7	0.2	0.0
Nov	8.5	2.9	0.5	1.9	1.7	0.0	0.1	1.0	0.5	0.5	0.0
Dec	7.8	2.4	0.4	2.3	1.2	0.0	0.1	1.0	0.6	0.4	0.0

Table 3.3. Monthly average chemical composition of total particle species (μg m^{-3}).

Month	Mass	Organics	EC	SO$_4$	NO$_3$	Cl	Na	NH$_4$	Crustal	Trace	Other
Kenmore Square:											
Jan	44.8	8.0	1.1	4.9	1.8	4.9	3.4	1.0	8.6	0.5	10.5
Feb	87.9	18.4	1.7	10.0	3.7	9.7	6.4	2.5	20.0	0.5	14.9
Mar	93.9	18.1	2.8	7.5	3.0	13.0	8.4	2.1	23.7	0.6	14.6
Apr	51.8	13.2	1.3	4.7	1.8	0.8	1.0	1.6	18.8	0.4	8.3
May	55.5	18.8	1.9	4.2	2.0	0.7	1.1	1.5	13.4	0.5	11.4
Jun	72.9	22.0	1.8	5.8	2.2	0.8	1.0	1.8	17.9	0.5	19.2
Jul	59.5	16.2	1.0	10.2	2.0	0.3	1.0	3.3	14.4	0.3	10.9
Aug	42.4	16.0	1.1	3.2	1.4	1.6	1.6	1.1	15.6	0.4	0.3
Sep	43.7	12.8	1.2	2.3	1.3	1.3	1.1	0.5	13.7	0.3	9.2
Oct	45.7	14.4	2.0	4.2	2.1	1.0	1.0	1.2	14.3	0.4	5.1
Nov	37.1	11.5	1.4	3.6	1.8	2.7	2.0	1.5	6.9	0.3	5.4
Dec	61.8	13.2	2.6	4.8	2.0	10.5	8.3	1.3	14.4	0.4	4.5
Reading:											
Jan	20.9	6.1	0.5	3.0	1.5	2.4	2.0	1.0	4.7	0.3	0.0
Feb	30.3	8.3	0.5	4.2	2.5	3.1	2.8	1.4	6.7	0.3	0.5
Mar	48.4	11.3	1.1	6.0	2.1	1.5	1.2	1.8	12.3	0.3	10.8
Apr	34.4	7.3	0.5	4.2	1.2	0.2	0.5	1.2	10.5	0.3	8.5
May	36.4	11.3	0.6	3.2	1.2	0.3	0.9	0.9	7.1	0.3	10.5
Jun	38.1	14.7	0.6	4.9	0.8	0.2	0.4	1.7	8.0	0.3	6.5
Jul	35.4	11.7	0.4	9.3	0.8	0.2	0.7	2.7	6.8	0.2	2.6
Aug	28.7	9.5	0.2	2.9	0.8	0.8	0.9	0.8	7.3	0.2	5.3
Sep	23.9	6.9	0.5	2.3	0.7	1.2	1.3	0.5	6.3	0.2	3.8
Oct	22.8	6.7	0.6	3.1	1.2	0.3	0.7	0.9	6.1	0.3	3.1
Nov	16.3	5.7	0.4	2.9	1.2	1.1	1.0	1.0	2.9	0.2	0.0
Dec	22.8	6.6	0.3	2.7	1.0	4.7	2.9	0.8	6.5	0.2	0.0
Quabbin Reservoir:											
Jan	15.2	3.6	0.1	2.6	0.8	0.3	0.8	0.8	2.2	0.1	3.9
Feb	20.2	4.3	0.2	3.5	1.7	0.3	1.0	1.1	3.8	0.2	4.1
Mar	23.6	4.8	0.2	4.8	1.9	0.1	0.7	1.6	5.0	0.2	4.3
Apr	21.6	3.9	0.1	4.3	0.6	0.0	0.4	1.2	4.1	0.2	6.9
May	36.9	10.4	0.3	2.5	0.7	0.0	0.3	0.7	5.4	0.3	16.2
Jun	43.6	13.7	0.1	3.9	0.8	0.0	0.3	1.2	6.1	0.3	17.1
Jul	36.0	9.0	0.2	11.4	0.7	0.0	0.4	2.5	5.3	0.4	6.2
Aug	42.1	10.7	0.3	7.7	1.1	0.0	0.9	2.0	12.5	0.9	5.9
Sep	14.1	4.2	0.2	1.6	1.2	0.4	1.0	0.6	5.1	0.3	0.0
Oct	15.1	5.6	0.3	2.7	1.7	0.0	0.4	0.8	4.8	0.3	0.0
Nov	22.5	3.5	0.2	2.5	0.7	0.0	0.6	0.7	2.3	0.2	11.5
Dec	6.7	2.3	0.1	2.4	0.5	0.1	0.5	0.6	1.4	0.3	0.0

Table 3.3 (con'd). Monthly average chemical composition of total particle species (μg m^{-3}).

Month	Mass	Organics	EC	SO$_4$	NO$_3$	Cl	Na	NH$_4$	Crustal	Trace	Other
Rochester:											
Jan	34.4	5.1	0.4	2.8	1.5	10.1	7.9	0.9	4.3	0.3	1.0
Feb	67.0	9.3	0.6	4.4	5.7	17.1	12.5	2.2	9.0	0.2	6.0
Mar	63.3	12.6	0.4	4.6	5.1	7.8	5.8	1.9	15.2	0.4	9.5
Apr	40.3	8.5	0.6	4.4	2.3	1.0	0.7	1.6	9.7	0.3	11.3
May	49.6	16.1	0.4	2.5	1.3	0.3	0.6	0.7	15.7	0.4	11.7
Jun	63.7	16.4	0.3	5.1	1.8	0.2	0.3	1.7	16.7	0.5	20.6
Jul	50.2	13.7	0.2	7.2	2.6	0.7	0.2	2.1	14.6	0.4	8.6
Aug	44.2	13.8	0.3	6.2	1.6	0.2	0.3	1.8	14.3	0.3	5.3
Sep	40.3	11.5	0.6	4.8	1.5	0.2	0.3	1.5	11.4	0.3	8.3
Oct	37.1	12.3	0.5	3.9	2.3	0.2	0.2	1.2	14.9	0.4	1.1
Nov	26.3	9.0	0.6	3.3	3.4	4.2	3.0	1.5	6.6	0.3	0.0
Dec	37.6	7.8	0.4	3.7	2.9	7.2	4.5	1.5	7.6	0.3	1.8
Brockport:											
Jan	12.9	3.1	0.1	1.6	1.0	0.6	0.6	0.6	1.4	0.2	3.7
Feb	30.1	5.6	0.2	3.5	4.6	1.4	1.6	1.6	5.1	0.2	6.2
Mar	26.7	6.7	0.1	3.6	3.9	0.9	1.4	1.4	6.1	0.2	2.4
Apr	36.4	14.4	0.3	4.2	2.6	0.1	0.2	1.5	7.9	0.2	5.0
May	47.2	13.0	0.4	2.1	1.0	0.0	0.0	0.6	15.9	0.4	13.8
Jun	48.3	12.6	0.2	5.7	1.5	0.1	0.1	1.8	8.7	0.5	17.3
Jul	22.0	8.1	0.2	4.8	1.2	0.0	0.5	1.2	4.9	0.2	0.9
Aug	36.6	12.3	0.2	5.6	1.1	0.0	0.2	1.5	11.2	0.3	4.1
Sep	27.0	7.9	0.2	4.5	1.2	0.0	0.5	1.3	6.8	0.3	4.3
Oct	28.8	8.6	0.4	3.3	1.7	0.0	0.4	1.0	9.8	0.3	3.6
Nov	13.9	4.5	0.2	2.5	2.3	0.2	0.2	1.0	3.2	0.2	0.0
Dec	16.8	5.0	0.1	2.6	2.0	0.5	1.6	0.9	5.2	0.2	0.0

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