DATA REPORT

CASCO BAY AIR DEPOSITION STUDY

prepared by

Byard W. Mosher, Ph.D.
Lakeshore Consulting
Barrington, NH 03825

for

The Casco Bay Estuary Project

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Executive Summary

Overview

The Casco Bay Plan developed by Casco Bay Estuary Project (CBEP) has identified a number of important environmental issues which are already influencing or could potentially adversely effect environmental quality in Casco Bay such as:

- Nuisance algal blooms from excess nitrogen loading in a small, rural bay of Casco Bay resulted in mortality of shellfish and other marine animals.
- Elevated levels of mercury, cadmium and polycyclic aromatic hydrocarbons in the estuary.
- Fish advisories have been issued because of elevated levels of methylmercury in fish from fresh water lakes and ponds in the Casco Bay Estuary watershed.

In September 1997, the Casco Bay Air Deposition Study was initiated to address some of the unanswered questions raised by these initial findings presented in the Casco Bay conservation and management plan. The Air Deposition Study, which is funded by the Great Waters Program, is managed by a interagency group comprised of staff from the Casco Bay Estuary Project, the Maine Department of Environmental Protection, and EPA Region 1. A monitoring site was established on the shore of Casco Bay at Wolfe's Neck in Freeport, ME. The primary objective of the study was to monitor atmospheric deposition of pollutants for a three year period in order to assess the magnitude and impact of atmospheric deposition to the Casco Bay ecosystem. This monitoring program was designed to address the following environmental questions:

- Is air deposition a current source of pollution to the bay?
- What is the relative contribution of atmospheric deposition to the total pollution measured in the water column and sediments?
- How does Maine fit into the larger regional pattern of air pollution?

The ultimate goal of the Study is to use the information gathered in this study to develop an air deposition loading estimation protocol which may be used by other community-based programs. The following sampling equipment were installed at the site:

- An IMPROVE (Interagency Monitoring of Protected Visual Environments) aerosol collection system.
- A NADP (National Atmospheric Deposition Program) precipitation collector.
- A MDN (Mercury Deposition Network) mercury precipitation sampler.
- A PAH (polyaromatic hydrocarbon) wet and dry deposition sampler designed by

personnel from the University of Massachusetts - Lowell.

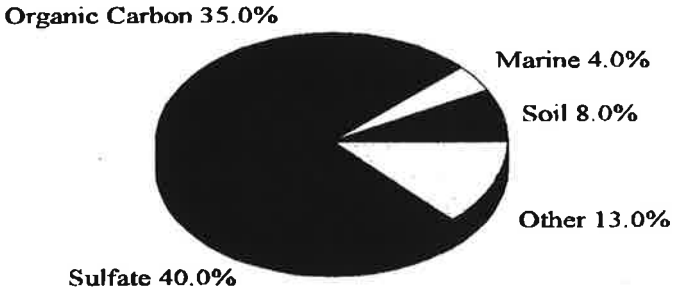
- A Belford Rain Gage.
- A meteorological station.

Significant Findings

Initial results of the air deposition sampling study at Casco Bay provide valuable insights on the magnitude, spacial patterns, and local/regional sources of atmospheric pollutants measured along the south coast of Maine. The coastal environment appears to experience higher concentrations of pollutants in air and in precipitation than inland sites. The Casco Bay Study demonstrates that atmospheric deposition of pollutants such as mercury and nitrogen can be significant and comparable in magnitude to inputs from point sources of pollution such as sewage treatment and industrial facilities. The local/regional mix of emission sources of these airborne pollutants appear to be varied and complex. Air pollution emissions from the metropolitan areas located along the eastern seaboard of the United States and Canada, and more distant emission sources in the mid-west appear to be important contributors of air pollution and contaminated precipitation that are measured along the Maine coast.

Atmospheric Particulates at Freeport

The dry deposition of particulate borne pollutants such as toxic trace elements, sulfate, nitrate, and organic compounds is an important mechanism which can rival wet deposition as a source of pollutants to terrestrial and marine ecosystems. Analysis of the IMPROVE aerosol data for the period of January 1988 - September 1999, indicates that the chemical composition of the aerosol found at Freeport is dominated by pollutant species. The average aerosol composition during this period is shown below.



Aerosol Composition at Freeport, ME

Concentrations pollutants in aerosols including trace metals such as arsenic, lead, and chromium were typically lower than those observed in more urban locations such as Boston, MA. It was not possible to develop definitive information on pollutant source regions because of limited data. Trajectory analysis suggests that local, regional, and inter-regional emission sources

(in the U.S. and Canada) influence the level of pollutants and air-quality in the coastal Maine region.

Major Ion Chemistry in Maine

The availability of weekly precipitation data from four NADP sites in Maine (Freeport, Bridgton, Acadia, and Greenville) provides valuable information on regional deposition patterns in the both the coastal and in-land Maine regions.

Precipitation chemistry at Freeport was dominated sulfates and nitrates. These two ionic species constitute approximately 51% of the dissolved ions measured in wet deposition measured at Freeport.

The marine elements, sodium and chlorine were major constituents of the precipitation at Freeport, contributing on average about 42% of the measured ions. As expected, marine contributions to precipitation chemistry in Maine were much more pronounced at the coastal sites of Freeport and Acadia. Similar concentrations of sulfates and nitrates were found at the inland and coastal monitoring sites.

Precipitation acidity was greatest at Acadia, and least at the inland site of Greenville. The acidity pattern in Maine suggests that the influence of emissions from industrial and metropolitan centers generally decreases south-to-north and east-to-west.

Wet and Dry Deposition of Nitrogen to Casco Bay

The annual direct wet deposition of 1.8×10^5 kg N/yr to Casco Bay during 1998 was calculated based on the Wolfe's Neck (Freeport) wet deposition data assuming a surface area for Casco Bay of 518 km^2. Approximately 70% of this nitrogen was in the form of nitrate. The remaining nitrogen in precipitation was in the form of ammonium. Previous studies of wet and dry nitrogen deposition have shown significant annual variations in the contribution of wet and dry components to total atmospheric deposition. These studies have also shown that wet and dry deposition are typically similar in magnitude. In the Casco Bay Study, dry nitrogen deposition was assumed to be of similar magnitude as wet deposition. Together wet and dry deposition of atmospheric nitrogen was estimated to be 3.6×10^5 kg N/yr to the surface of the Bay. To put this in context, NOAA estimated in 1991, point-source discharges from sewage treatment effluent introduced roughly 5.4×10^5 kg N/yr into Casco Bay. These data indicate that the atmosphere may be a significant source of nitrogen to the Bay, contributing about two-thirds as much nitrogen as do point sources in the Bay. In addition, atmospheric deposition of nitrogen occurs throughout the watershed of the Bay. A small portion (1-5%)of the nitrogen entering Casco Bay via riverine input may be atmospherically derived.

Mercury in Precipitation in Maine

Mercury in precipitation was measured at four MDN sites in Maine (Freeport, Acadia, Bridgton, and Greenville), two New Hampshire sites; New Castle, on the coast and Laconia in the lakes region. In addition, data from two Canadian maritime sites; St Andrews New Brunswick, and an inland site at Kejimkujik, Nova Scotia was examined.

The MDN data indicated that a few large deposition events contribute a significant portion of the annual mercury deposition at the Freeport site. The highest weekly deposition occurred during the week of June 9-16, 1998. During this one week, roughly 18% of the annual mercury

deposition occurred at Freeport.

Most of the MDN sites received more mercury deposition during the summer months. Higher summer temperatures (higher volatility of mercury) and oxidant levels (more rapid conversion to soluble mercury) play an important role in the removal of mercury from the atmosphere in precipitation during the summer.

The greatest wet deposition of mercury occurred at Freeport. The lowest mercury deposition occurred at the Nova Scotia site. Mercury deposition at the in-land sites was considerably lower than mercury deposition at the coastal New Hampshire or Maine sites.

Wet Deposition of Mercury to Casco Bay

The predominant form of mercury found in the atmosphere is non-reactive gaseous phase mercury, which does not readily deposit on surfaces. Therefore, dry deposition of non-reactive gaseous mercury is probably not an important mercury input factor to the surface of Casco Bay. The MDN wet deposition measurements were used to calculate the input of mercury to Casco Bay assuming an area for Casco Bay of 518 km^2. The calculated annual input of 4.7 kg/Hg to the surface of the Bay must be considered a conservative estimate of atmospherically derived mercury input since some of the mercury deposited on the surrounding watershed enters into the Bay in the form of runoff and riverine flow. Mercury input from the surrounding watershed may be significant during the spring snow melt when runoff from spring rains flushes rapidly into the Bay.

Estimates of point source inputs from NOAA for the year 1991 place the atmospheric contribution into perspective. Direct input from the atmosphere was estimated to be 25-30% that of the known point sources. In the future, as these point sources come under control, the relative significance of mercury from the atmosphere to Casco Bay will increase.

PAH Wet and Dry Chemistry at Freeport

Twelve (12) weekly wet deposition samples and sixteen (16) weekly dry deposition samples were collected over the sampling period (March 1997 through February 1998). The average hourly dry deposition rate for total PAHs was 8.7 $ng/m^2/hr$, with values ranged from 2.7 to 20.1 $ng/m^2/hr$. Dry deposition rates were lowest in the summer and highest during the winter months. The major PAH compounds found in the dry deposition samples were fluoranthene (25%), pyrene (21%), and fluorene (8%). The increased use of fuels (oil and wood) during the winter may explain the higher concentrations of PAHs found in dry deposition in the winter. In addition, atmospheric photochemistry is lowest during the cold months and photochemically induced removal of ambient PAHs is significantly reduced.

The average weekly wet deposition rate was 2,234 ng/m^2 and samples ranged from 587 to 5664 ng/m^2. Major PAH compounds found in the wet deposition were phenanthrene (21%), fluorene (17%), and fluoranthene (11%). Unlike the dry deposition data, no clear seasonal variations in the wet PAH deposition rates were evident.

Deposition of PAHs to Casco Bay

Using the results of the weekly wet and dry deposition sampling at Freeport, thre total direct annual atmospheric input of PAHs to Casco Bay was calculated to be 64 kg PAHs/yr. It is difficult to put this deposition number into context due to a lack of either estimates of inputs from other sources or Casco Bay sediment PAH burdens.

Casco Bay Air Deposition Study Data Report

The Casco Bay Air Deposition Data report is divided into four main sections. Section A deals with the IMPROVE aerosol data set. The major composition of the aerosols collected at Freeport is examined and regional sources of the pollutants found in the aerosol are discussed using air mass trajectory analysis.

Section B of the report details the results of NADP precipitation sampling. Wet deposition chemistry and seasonal trends are examined. Variations in wet deposition patterns within the NADP sites in Maine are explored and calculations of wet and dry deposition of nitrogen to Casco Bay are presented.

In Section C, the magnitude and regional variations in mercury in precipitation at a number of northeastern MDN sites are discussed. An examination of the relationship between Hg in precipitation and precipitation depth is followed by an estimation of mercury inputs to Casco Bay.

Finally, in Section D, PAH concentrations in wet and dry deposition at Freeport are discussed, potential sources for these PAHs are examined, and PAH inputs to Casco Bay are estimated.

A. IMPROVE Data

This section discusses IMPROVE data from the Freeport, Maine site located on Wolfe's Neck, focusing on the period of January 23, 1998 through June 6, 1999 (eighty 24-hour aerosol samples were collected during this time). Additional data for the components of fine mass that were collected through September 3, 1999 have also been included in this analysis. Details of the sampling system and analytical procedures used in the collection and analysis of IMPROVE network samples are presented in the IMPROVE Quality Assurance Project Plan (Casco Bay Estuary Project, 2001).

Several different analytical methods were used to analyze the filters that captured the aerosols. Chlorine was determined by three independent analytical techniques, elemental chlorine by X-ray Fluorescence (XRF) and Proton Induced X-ray Emission (PIXE), and chloride ion by ion-chromatography (IC). The chloride ion values have been used in all calculations. These data are much more comprehensive and appear more reliable than either the XRF or PIXE results.

Sulfur compounds were determined as elemental sulfur by PIXE analysis and as sulfate ion ($SO_4^=$) by ion chromatography. An examination of these data reveals significant differences. The mean ratio of PIXE sulfur to IC sulfate ion sulfur was calculated to be 0.57 ± 0.45 ($n = 56$), which was significantly less than the expected value of 1.0. Sulfate ion concentrations have been deemed to be more reliable and thus this data will be used in this report.

The data presented for the trace metal mercury should also be discussed. While only six values are available for particulate mercury, the reported values are exceedingly high compared with those reported by other studies. A mean of 0.958 ng/m^3 was reported for these IMPROVE samples. Typically, particulate mercury concentrations are observed to be very low and to represent a very small fraction of total atmospheric mercury (usually less than a few percent). For example, Fitzgerald et al. (1991) reported a mean value of 0.062 ± 0.048 ng/m^3 ($n = 22$) particulate mercury at Avery Point, CT, a coastal location on Long Island Sound. More recent

data (1998) from the EPA New England regional Environmental Monitoring and Assessment Program (REMAP) indicate particulate mercury to vary at rural to urban locations. Average mercury concentrations from particulate samples collected at Underhill (VT) and Providence (RI) were 0.010 ng/m^3 and 0.019 ng/m^3, respectively. These values are a factor of 15 to ~100X less than the mean of the data reported for the six IMPROVE samples from Freeport. Mercury concentrations in the IMPROVE samples were determined by XRF while most studies reporting particulate mercury data employ cold-vapor atomic fluorescence spectroscopy, a method which has a much lower detection limit as well as the accuracy and precision necessary to accurately measure the extremely low levels of particulate mercury present in the atmosphere. For this reason, the mercury data for the IMPROVE data set should not be considered representative of actual particulate concentrations.

An examination of the fine mass (FM) values suggests that there may be problems with these data. FM is determined gravimetrically by weighing one of the IMPROVE filters prior to analysis of the individual species present on the filter. For a significant portion of the aerosol samples, the FM has been significantly underestimated. A calculation and examination of the variable Sum of Species/Fine Mass clearly indicates the FM to be less than the Sum of Species. The ΣSP/FM should be equal to or less than 1.0 for several reasons: 1) not all compounds and elements have been detected in each sample, and 2) the sum of elemental species does not include oxygen in the form of metal and mineral oxides nor the hydrogen and oxygen associated with organic material. For a significant portion of the data under consideration, this value is found to be greater than 1.0. The mean value for 77 samples was calculated to be 1.89 ± 3.91 suggesting that FM may be underestimated in a significant portion of the samples in question. Underestimates are most apparent during the summer and fall months when higher temperatures and increased relative humidity lead to sampling and/or analytical artifacts. The calculated variable Sum of Species has been used rather than FM in calculations of aerosol component composition. The use of this calculation may result in underestimations of soil and marine aerosol contributions to the total fine mass because not all elemental species or compounds are detected in many of the samples.

Summary statistics for 35 of the measured species detected in these samples are presented in Table 1. A correlation matrix for 30 variables is found in Table 2. Seasonal summary statistics were compiled for this data set and they are shown in Table 3.

Summary and Seasonal Statistics for Measured Variables

The natural crustal and marine components contribute roughly 12 % of the fine mass of the IMPROVE aerosol data. Anthropogenic pollutants emitted by combustion and transportation sources are by far the largest fraction of the $PM_{2.5}$ aerosol fraction at Freeport, ME.

Crustal Species

The concentrations of crustal species observed at the Freeport site are similar to those found by other recent studies in coastal regions. For example, at Point Reyes, CA Chow et al. (1996) found average aluminum, iron, and silicon concentrations of 16, 18, and 10 ng/m^3, respectively, compared with 24, 15, and 31 ng/m^3 at Freeport. Data (1988-1997) from the IMPROVE site located at the Acadia National Park (ANP, ME) compare well with those from the Freeport site (Al - 15 ng/m^3, Fe - 18.5 ng/m^3, and Si - 47.9 ng/m^3). The Freeport values are much lower than concentrations typically found in urban locations, however. In Watertown, MA, Thurston and

Spengler (1985) found mean concentrations of iron and silicon of 75 and 104 ng/m^3 respectively. Time series plots for the crustal species Al, Ca, Fe, Si, and Ti are shown in Figures 1-5 respectively. Iron, calcium and silicon exhibit slightly lower concentrations during the summer. Interesting, correlations between the crustal species are not exceptionally strong in most cases. For instance, iron exhibits it's best correlations with zinc and copper (r = 0.68 and 0.63 respectively), two species generally considered to have major pollutant sources. In the case of aluminum, the strongest correlations are found with H and sodium (r = 0.68 for both pairs). Correlations are also strong between rubidium, yttrium, and zirconium, and several of the crustal species, suggesting that crustal material is a major source for these species as well.

Marine Species

Times series plots for the two major marine species, sodium and chloride ion, are presented in Figures 6 and 7 respectively. Chloride was determined by three analytical methods, XRF, PIXE, and ion chromatography (IC). Only the chloride ion IC data will be discussed here as this is the most extensive and reliable data set. The chloride ion plot indicates that in general concentrations of this species are less than 100 ng/m^3 with occasional events where concentrations increase by a factor of three to four. Chloride ion and sodium are well correlated (r = 0.92) since this site is close to the ocean. Both these species exhibit a strong correlation with magnesium (r = 0.79 and 0.86 respectively. These data suggest that a significant portion of the aerosol magnesium seen at Freeport is of marine origin. Much weaker correlations are observed for chloride ion and sodium with bromine (r = 0.38 and 0.64, respectively), suggesting that bromine may have a significant non-marine component in the fine aerosol phase as well as a marine component. Concentrations of both sodium and chloride ion peak during the spring months (see Table 2 and Figures 6 and 7). The average Na/Cl ratio in aerosols is 0.84, considerably higher than that of bulk seawater, 0.56, but only slightly higher than the Na/Cl ratio for ANP (0.7). As alkaline seasalt aerosols react with acidic species such as nitric acid in the atmosphere, chloride ion tends to be lost from the aerosol through volatilization of species such as HCl (hydrochloric acid). This local chemical process may explain some of the deficit of chloride in the small, sub-2.5μm, size range collected by the IMPROVE sampler. It should be pointed out that the 2.5μm particle-size cut-off of the IMPROVE sampler excludes a significant fraction of both crustal and seasalt derived aerosols generally found in the super-micron size range.

Trace Metals

The trace metal species arsenic, chromium, copper, lead, mercury, nickel, selenium, vanadium and zinc originate in large measure from anthropogenic emission sources such as fossil fuel combustion, industrial processes, incineration of waste (medical and municipal), and transportation. Time series plots for eight of these trace metal species (excluding mercury, see discussion above) are shown in Figures 8-15. The levels of particulate trace metals observed at the Freeport, ME IMPROVE site appear to be consistent with levels found by other studies at similar coastal locations. For comparison purposes IMPROVE data from a coastal location in Acadia National Park (ME) and from maritime Canada (St. Andrews, New Brunswick) are presented in Table 4. Data from 125 one day IMPROVE samples collected from May 15, 1996 through July 26, 1997 at the Huntsman Marine Science Centre, St Andrews, New Brunswick, Canada, and

980 one day samples (April 1988 through September 1997) collected at the IMPROVE sampler at Acadia National Park (ANP) are tabulated. Arsenic, zinc and selenium concentrations at the sites are very similar, while the remainder of the trace metal species appear to be present at higher levels at the Freeport site than at the other sites.

The elements bromine and lead show a relatively strong correlation ($r = 0.75$) at the Freeport site. These variables are plotted in Figure 16 and 11 respectively, and a time series plot of the Br/Pb ratio is presented in Figure 17. The mean Br/Pb ratio observed at Freeport was 1.09 ± 0.783 ($n = 59$), much higher than typically observed in previous studies conducted when leaded gasoline was widely used. For example, Thurston and Spengler (1985) measured a Br/Pb of 0.263 in Watertown MA and Chan et al (1997) reported an average ratio of 0.36 in Brisbane, Australia. Leaded-gasoline was commonly used at the time these two studies were conducted. These results are consistent with the findings of many other studies which have concluded that the elimination of lead as a fuel additive has resulted in a significant decrease in the lead content of fine particulate matter in the atmosphere. Similar ratios were calculated for the Bridgton and ANP IMPROVE sites (Br/Pb = 0.73 for both sites).

Sulfate, Nitrite and Nitrate Ions

Time series plots of sulfate, nitrite, and nitrate ion are shown in Figures 18, 19 and 20 respectively. The major production mechanism for these three species is the particulate conversion of gaseous precursors such as sulfur and nitrogen oxides (SO_2, NO and NO_2) and nitric acid (HNO_3). The combustion of fossil fuels to produce heat and electricity and vehicle emissions are major sources of these gas phase compounds. Sulfate exhibits higher concentrations during the summer months, while the concentration of nitrate appears to peak in the winter with a secondary peak in the summer. Highest values for nitrite ion are found during the winter and spring, with values typically a factor of 3 to 6 lower in the summer and fall, respectively. This is due to the more rapid conversion of nitrogen compounds such as HNO_3 and nitrite ion, during the warmer months when higher temperatures, humidity, and increased photochemical activity result in faster conversion to nitrate. Nitrate represents the major oxidative particulate nitrogen compound measured at Freeport, with nitrate values typically 30-70 times those of nitrite.

Remaining Trace Element Species

Time series plots for the remaining species, hydrogen, potassium, magnesium, manganese, rubidium, strontium, yttrium, and zirconium are shown in Figures 21- 28. The peak in hydrogen (aerosol acidity) in the summer months (Figure 21) was due to significantly higher concentrations of acidic compounds such as sulfate present during the warmer months. Conversely, potassium is seen to peak in the winter months (Figure 22) and this may be related to increased regional combustion of wood for residential heating. Consistent seasonal trends for magnesium, manganese, rubidium, strontium, yttrium, and zirconium were less evident (Figures 23-28). This is different from the long-term data that exist for Bridgton (1989-1993) and Acadia National Park (1988-1997) which show weak seasonal trends (higher concentrations during the winter and lower concentrations during the summer).

Summary Statistics for Composite Variables

A number of composite variables have also been calculated using the IMPROVE elemental compositional data. Definitions of these variables are found in Table 5 and summary statistics are presented in Table 6. Elemental carbon (light absorbing carbon, LAC) originates predominantly from burning processes and vehicle exhaust (diesel exhaust in particular). Sources of organic carbon (OC) include fuel burning, vehicles, biomass burning and oil refineries, etc. Time series plots for elemental and organic carbon are shown in Figures 29 and 30, respectively. Organic carbon values were typically three times those of elemental carbon (OC/LAC = 3.78 ± 1.31, n = 71) and these two species were well correlated (r = 0.92) suggesting that both carbon species have similar sources. There are no seasonal trends apparent in the time series plot for these two species or their ratio (Figure 31).

It has been suggested that non-soil potassium (KNON) may serve as a qualitative tracer for smoke originating from combustion sources such as wood burning and forest fires. As can be seen in Figure 32, this variable exhibits peaks in the winter months and thus may be indicative of the impact of local wood combustion on the sea coast of Maine. Ammonium sulfate calculated using the IMPROVE sulfate ion-sulfur concentrations exhibits higher concentrations during the summer months (Figure 33) due to the elevated levels of sulfate ion present during this period. Conversely, ammonium nitrate concentrations appear to be highest during the winter with a secondary peak in the summer (Figure 34). Seasonal trends for the variable RCMC (reconstructed mass without nitrate) are less evident (Figure 35).

Major Component Analysis

The typical general sources of many of the individual species need to be considered. The species have been broadly characterized as shown in Table 7. The measured Fine Mass reported for a significant portion of the IMPROVE samples may be too low as discussed above, and thus in the major component analysis calculations the derived value *Sum of Species* has been used. The soil and marine aerosol components were calculated as shown in Table 5 and the results are presented in Table 8 and Figure 36.

Soil Aerosol

As seen in Table 8, on average, soil derived aerosol contributes almost 8% of the fine aerosol loading at Freeport. Minimum values are observed during the summer while maximum input of soil material occurs during the windy fall months. Similar crustal inputs were measured at an IMPROVE coastal site at St. Andrews, New Brunswick, Canada where researchers calculated that crustal inputs contributed 3.9 ± 0.20 % of the fine mass over a 15 month period (J.L. Campbell, personal communication). The relatively low crustal component contribution is not surprising in-light of the fact that the 2.5μm cut-off of the IMPROVE sampler excludes a significant fraction of airborne crustal material which is present predominantly in the super-micron range in continental regions.

Marine aerosol

Aerosol derived from the nearby ocean constitutes a relatively small fraction of the fine aerosol mass at Freeport, roughly 4% on average, with the highest inputs occurring during the spring, winter, and fall.. As in the case of the soil component, the 2.5μm cut-off of the

IMPROVE sampler discriminates against sea salt particles which are generally found in the super-micron size range. Together, these two natural aerosol sources contribute approximately 12% of the annual fine aerosol loading at this site. Maximum natural inputs are observed to occur during the fall months when about 18% of the fine mass loading originates from these two natural sources.

Trace metals

Loading of trace metal species contributed a small fraction of the input at this site (< 1%). However, due to their toxic nature and potential for bio-accumulation in the marine ecosystem, inputs of these species may still have significant ecological effects. Metal inputs appear to be lowest in the spring and at a maximum during the fall period.

Organic Carbon

Anthropogenically derived particulates constitute the major portion of the fine mass at Freeport. As is evident in Table 8, organic carbon makes up a significant fraction, roughly 35%, of the fine mass at this site. There do not appear to be distinct seasonal changes in Organic Carbon input at Freeport.

Sulfate

Sulfate is the other major contributor to fine mass at Freeport, on average constituting 40% of the mass. Sulfate input was observed to be at a minimum during the winter and fall months and maximum during the spring and summer. This is certainly related to the much higher oxidation rates typical in the warmer months due to increased temperature and photochemical processing of the gas phase precursor species, sulfur dioxide (SO_2).

It is evident that the fine mass at Freeport is dominated by anthropogenic aerosol throughout the year. This is consistent with observations at other IMPROVE sites. On average, these four components account for a large fraction (87 %) of the fine mass collected at the Freeport site annually. Seasonal totals also appear reasonable, ranging from 78% in the winter to 93% in the fall.

Regional Sources of $PM_{2.5}$ Aerosol at Freeport

An analysis of the chemical composition of the fine aerosol mass at Freeport indicates that the vast majority ($\geq$ 75%) of the ambient aerosol is derived from pollutant sources. An examination of air-mass trajectories can provide some evidence about the potential sources of aerosols and the relative strengths of the sources. On-line (internet web site) backward trajectories ending at the Freeport ME, IMPROVE sampling site have been generated using software available at the NOAA Air Resources Laboratory web site (www.arl1.noaa.gov). These trajectories are isentropic in nature. They follow lines of equal potential temperature in the atmosphere. Two heights in the atmosphere were examined, 500m and 1000m above ground level (AGL). Each of the trajectories span a 24-hour period, with 6-hour time-steps indicated on the plots. Although archived meteorological data were not available at the NOAA web-site for all the time periods of interest, twelve trajectories were generated using the accessible archived on-line data. The trajectories were chosen to illustrate sampling periods when concentrations of pollutants and natural aerosol at Freeport were found to be among the highest during the sampling

period. Together they demonstrate that aerosols found at the Freeport site originate from diverse regions, both in the vicinity of the sampling site and at distant locations. Information on these trajectories is tabulated in Table 9. Interpretation of these air mass trajectories must be undertaken with some caution. Trajectories are generally indicative of the atmospheric flow immediately upwind of the Freeport sampling site during the sampling period. The travel pathways shown on the trajectories provide qualitative information on the potential origin of aerosols observed at the sampling site. Certainly, diffusion and dispersion of pollutants occurs in the atmosphere as the air mass travels along the calculated trajectory line.

The first trajectory run (Figure 37) was for the sampling day January 29, 1998 when the highest nickel concentration was observed at Freeport, ME. The trajectory indicates that air arriving at the site had passed over northern coastal Maine and Nova Scotia, Canada prior to its arrival in Freeport. It is difficult to conjecture as to what the (significant) source of nickel would be along this flow pattern.

High levels of chromium, lead and titanium were found in the aerosol sample at Freeport on February 22, 1998. In this case the 24-hour back trajectory (Figure 38) of the air mass arriving at coastal Maine was from the metropolitan Quebec and Montreal regions.

On March 18, 1998, high concentrations of a number of trace metal were collected on the IMPROVE filters at the Freeport sampling site. The highest concentrations of lead, rubidium, selenium, zinc, zirconium, and bromine were recorded and elevated concentrations of iron, nickel, vanadium, calcium, copper, and nitrate were also found. In this case (Figure 39) air mass flow was rather slow as indicated by the relatively short 24-hour trajectory flow pattern. The trajectory suggests that the emission sources of these pollutants lies within the northern New England region. It would appear that the elevated pollutant levels observed in this sample may be from several local sources. For instance, the high zinc concentration may result from incinerator emissions while vanadium is considered to be indicative of oil combustion.

For the April 23, 1998 IMPROVE sample, trajectory analysis (Figure 40) indicates flow at Freeport was in off the oceans with indications of air mass re-circulation in the central New England coastal region. High chromium and rubidium concentrations were evident on this occasion.

Flow from the northern mid-west (Detroit) and southern Quebec (Toronto) regions (Figure 41) resulted in elevated levels of sulfate and organic carbon on May 29, 1998. The highest levels of sulfate observed during the IMPROVE sampling were seen on July 16, 1998. Again, these elevated sulfate levels were accompanied by high OC concentrations. Meteorological data was not available in this case.

Similarly, on July 22, 1998, high sulfate levels at the Freeport site result when flow came from the same mid-west region (Figure 22). Six days later, on July 28, 1998, high sulfate levels were again observed. This time flow was from a more southwesterly/southerly direction, appearing to originate in the midwest (Figure 43).

The aerosol event which occurred at Freeport on December 7, 1998 is interesting in that it shows high levels for a number of pollutants such as nitrate, sulfate, organic and elemental (light absorbing) carbon as well as the marine derived species sodium and chlorine. In this case (Figure 44) the flow near the surface (500 meters above ground level) was much slower than flow higher in the atmosphere at 1000m. Low level flow came up from the metropolitan areas along the eastern seaboard while upper-level flow originated in the Ohio River valley and the aerosol

collected at Freeport during this period may reflect this mixture. The high nitrate and marine component appears to be brought in with the low-level flow from the east coast while the high sulfate and total carbon concentrations may have originated in the heavily industrialized regions of the Ohio River valley, as previous trajectories have indicated.

It has been suggested that potassium and the derived variable nonsoil potassium, (KNON = [K] - 0.6[Fe]) may serve as a qualitative tracer for wood combustion. The highest levels of potassium and KNON at Freeport were found on December 31, 1998. Trajectory analysis (Figure 45) indicates that during this time period, flow in the atmosphere, both near the surface and at altitude, originated near the Canadian cities of Ottawa and Montreal and passed over northern New England. The highest arsenic concentrations of the sampling period were also observed in this sample. Arsenic is considered to be a tracer of coal combustion and ore smelting.

Trajectory analysis for the sample collected on January 6, 1999 (Figure 46) suggests local sources of aerosols found in the Freeport atmosphere. In this case, high concentrations of vanadium, LAC, OC, TC, and zinc were observed. Air mass movement at both levels in the atmosphere was slow and appeared to pass over southern New Hampshire and western Massachusetts. Six days later, on January 12, 1999, much more vigorous flow was established as indicated by the much longer 24-hour trajectories (Figure 47). Again, in this case, divergent flow in the atmosphere was apparent, with low level flow passing over the east coast of the U.S., and the air mass associated with upper level flow coming from a more westerly direction, the Ohio River valley.

Finally, on March 25, 1999, high levels of just one pollutant, aerosol nitrate, were collected at Freeport. Backward trajectory analysis for this sample revealed flow at both levels passing over the metropolitan Montreal, Canada region.

The trajectory analysis presented here suggest local, regional and distant sources of air pollution, both in the U.S. and Canada, influence the level of pollutants measured along south coastal Maine. Many factors influence the concentration of polluted and natural aerosols in the coastal Maine region. The geographic distribution of emission sources, meteorology factors such as air mass flow patterns, clouds, precipitation, temperature, relative humidity and photochemical activity, as well as aerosol chemistry, all play a role in influencing the mixture of aerosol measured at the Freeport IMPROVE monitoring site.

B. NADP Data

This section discusses the National Atmospheric Deposition Program (NADP) data from four NADP sites in Maine (including the Freeport site). The period of record is December 30, 1997 through January 12, 1999. Details of the sampling system and analytical procedures used in the collection and analysis of NADP network samples are presented in the NADP section of the Casco Bay Air Deposition Study QAPP (Casco Bay Estuary Project, 2001). The locations of the four NADP sampling sites (Bridgton, Greenville, Freeport, Acadia) are shown in Figure 49.

ME02 - Bridgton
ME09 - Greenville
ME96 - Freeport
ME98 - Acadia
NH02 - Laconia

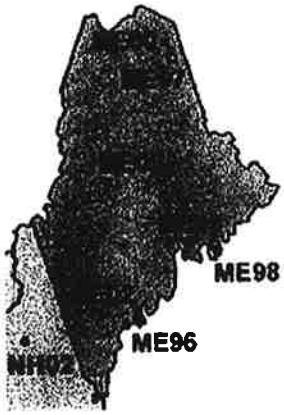


Figure 49. Maine and New Hampshire NADP Sampling Sites

Wet Deposition Chemistry and Seasonal Trends at Freeport

Summary information on annual and seasonal precipitation chemistry at the Freeport site are shown in Table 10 and Figure 50. Precipitation chemistry at Freeport is dominated by sulfates and nitrates. This was also the case for IMPROVE aerosol chemistry. Although the integration times of the IMPROVE aerosol (one-day-in-six) and NADP precipitation samples (weekly integrated samples) differ, comparing the partitioning of nitrate and sulfate in precipitation and in aerosols provides insight into the role of atmospheric chemistry and local sources. The average nitrate/sulfate ratio measured in the aerosol was 0.23. This ratio was found to be much higher in the precipitation phase, 0.77. This difference may be partially explained by the fact that nitric acid is much more volatile than sulfuric acid and thus as cloud droplets form and dissipate, nitrate tends to be concentrated in the gas phase, while sulfate remains in the dissolved or aerosol phase. Certainly, differences in aerosol and precipitation origin also have a profound influence on precipitation and aerosol chemistry. Also, the proximity to the ocean and the potential for sulfate enriched ocean aerosols cannot be dismissed. Seasonal trends in precipitation chemistry also differ from those observed with the aerosol phase suggesting that different factors may be influencing aerosol and precipitation chemistry at Freeport. Aerosol phase nitrate is highest in the winter and lowest in the fall, while nitrate in precipitation peaks in the fall and summer and is lowest in the winter. In contrast to nitrate, aerosol sulfates peak in the summer, but are lowest in the fall, while precipitation sulfate is highest during the summer and fall months and lowest in the winter. Changes in motor vehicle, residential and commercial fuel consumption, and seasonal changes in atmospheric nitrogen chemistry due to seasonal differences in temperature and photochemical activity, combine to influence the partitioning and concentration of nitrogen species in aerosol and precipitation chemistry.

The marine environment is a source of atmospheric aerosols. Physical processes such as breaking waves and bursting bubbles generate sea salt aerosols observed at the Freeport site. Sodium and chlorine ions are significant constituents in precipitation at Freeport, contributing on average 42% of the measured species. The sodium/chloride ratio in seawater is 0.558 and precipitation chemistry exhibits a very similar ratio, 0.60 on average, indicating that the coastal

marine environment has a significant influence on precipitation chemistry.

East-West Variations in Precipitation Chemistry

Annual precipitation data for four Maine NADP sites are presented in Table 11. The Freeport and Acadia NADP sites are located along the coast, while the Bridgton and Greenville sites serve as their inland counterparts. The geographical distribution of these four sites provides an opportunity to identify gradients for inland versus coastal, east-west variations in precipitation chemistry, and north-south variations at both coastal and inland locations.

A strong influence of the marine environment is especially evident when precipitation chemistry at the two coastal Maine sites is compared with that of the two inland sites (Table 11). Annual volume weighted concentrations of sodium and chloride are a factor of five to ten lower in precipitation at the inland sites. The Na/Cl ratio at both Freeport and Acadia (0.60 and 0.57, respectively) is similar to that of bulk sea-water (0.56) while the Na/Cl ratio at Bridgton and Greenville is lower (0.52 and 0.41, respectively). Several other elements show similar trends. Magnesium concentrations in precipitation are seen to be much lower at the inland sites, suggesting a marine component may be important at coastal locations. The Mg/Na ratio at the four sites supports this hypothesis. This ratio is similar to that of bulk sea water (0.12) at the two coastal sites, Freeport and Acadia (0.11 and 0.12, respectively) while it is much higher at the inland sites of Bridgton and Greenville (0.16 and 0.20, respectively).

Potassium concentrations in precipitation are also lower at the inland sites. The K/Na ratio in precipitation appears to increase the further north and inland one moves. Examination of the K/Na ratio at the four sites suggests that the marine environment may not be the major source of potassium in precipitation at the inland sites. The K/Na ratio at two of the sites is significantly greater than the bulk sea-water ratio of 0.036. This ratio is higher at the in-land sites (0.13 and 0.27 at Bridgton and Greenville, respectively) than at the coastal locations (Freeport and Acadia 0.043 and 0.073). The examination of IMPROVE aerosol concentrations indicated that combustion of wood contributed to aerosol potassium at Freeport. The K/Na ratios in precipitation suggest that this influence is small at Freeport where this ratio is slightly elevated over the sea water ratio. At Freeport, aerosol potassium concentrations were highest during the winter and fall months. However, potassium concentrations in precipitation are highest during the summer and fall months. Concentrations of calcium in precipitation exhibit no pronounced seasonal variation among the four sites.

Ammonium concentrations in precipitation are remarkably constant among the four sites. The NADP sampler at Freeport is located on the grounds of a cattle farm where animal waste is routinely spread on the fields as a fertilizer. The data seem to suggest that revolatilization of ammonia from the farm and subsequent entrainment in precipitation is not a factor in ammonia deposition in precipitation at Wolfe's Neck.

Spatial trends between coastal and inland sites for nitrate and sulfate are not strong. Concentrations of both these anions are lower at the inland sites compared with the coastal sites.

North-South Variations in Precipitation Chemistry

Concentrations of the marine derived species (Na, Mg, and Cl) are very similar at the two coastal locations. There is little change in the concentration of these species at the coastal sites as one moves north. At the inland sites however, where the marine influence is much less

pronounced, a significant south-to-north decrease (a factor of 1.5 to 2) is evident. Sodium, magnesium and chloride input from industrial and construction activities may be more important at these inland sites and thus as one moves north to the more rural, less populated regions in the region of Greenville, this influence should be less pronounced.

There are differences in the concentration of nitrate and sulfate along the coast. Precipitation acidity (pH) is highest at the coastal Acadia site. The pattern suggests that the influence of emissions from industrial and metropolitan centers generally decreases as one moves north through Maine. Concentrations of nitrate, sulfate and acidity are highest at the three southern NADP sites.

Atmospheric input of pollutants to Casco Bay

Annual precipitation measurements during 1998 are shown in Table 13 for the four sites. It is evident that the coastal sites receive more precipitation and that Acadia was the wettest site of the four, receiving about 14% more precipitation than Freeport. Annual wet deposition for all ions were higher at the coastal sites than at inland counterparts.

One of the objectives of the Casco Bay Air Deposition Project is to estimate the amount of atmospheric nitrogen deposition to the Bay. As point sources of water pollution come under more stringent controls, sources such as the atmosphere will play a more important role in the introduction of species such as nitrogen and some trace elements if this source is not also controlled. Excess levels of nitrogen in marine ecosystems can lead to increased algal productivity and eutrophication of coastal bays and estuaries. Wet deposition of nitrate collected at the Freeport site can be used to estimate direct atmospheric input to the Bay. The surface of the Bay and its surrounding watershed receive nitrogen from the atmosphere in the form of wet deposition, as measured by the NADP sampler, and as dry deposition (gas phase and particulate nitrogen). While the NADP sampler integrates wet deposition in continuous one-week intervals, the IMPROVE sampling network, because of it's one-day-in-six sampling protocol provides a discontinuous record of the atmospheric concentrations of nitrogen species. Thus, rather than using the IMPROVE data to estimate the dry depositional component of atmospheric input to the Bay, we shall use the NADP data to provide a semi-quantitative estimate of both wet and dry nitrogen deposition to Casco Bay. Calculations of annual wet deposition (kg N/ha) supplied by the NADP Program, based on the weekly precipitation chemistry data and rain fall depths for the four Maine NADP sites are presented in Table 12. Using the Freeport wet deposition data and assuming a surface area for Casco Bay of 518 km^2 (Casco Bay Report, 1995), we calculate an annual direct wet deposition of 1.81×10^5 kg N/yr to Casco Bay. Typically wet and dry deposition are similar in magnitude. Thus we may use our calculation of wet deposition to make a semi-quantitative estimate of dry nitrogen deposition by assuming that they are of similar magnitude. Wet and dry deposition of atmospherically borne nitrogen species introduce approximately 3.62×10^5 kg N/yr to Casco Bay (Table 14). Approximately 70% of this nitrogen input is in the form of nitrate with the remainder entering as ammonium. Similar deposition studies of wet and dry nitrogen have shown significant annual variation in the contribution of wet and dry components to total atmospheric deposition. For instance, the dry deposition of nitrogen to Chesapeake Bay has been estimated to be 20-55% of direct wet deposition (US EPA, 1996). To put this estimate for Casco Bay in context, NOAA estimated that annual point-source inputs to Casco Bay (including sewage treatment effluent) was roughly 5.4×10^5 kg N/yr for the year

1991. The data suggest the atmosphere is a significant source of nitrogen to the coastal marine environment of the Bay, directly contributing perhaps 50+% as much nitrogen as do point sources in the Bay. This direct deposition estimate is important considering that it is just to the surface of the Bay. Atmospheric deposition of nitrogen also occurs in the surrounding watershed and a portion of this nitrogen entering Casco Bay via riverine input. The Casco Bay watershed area is estimated to be 2550 km^2 or roughly five times the surface area of the Bay itself. If identical wet and dry depositional rates are assumed within the watershed, and that possibly 5-20% (based on Chesapeake Bay inputs) of the nitrogen deposited in the watershed from the atmosphere ultimately reaches Casco Bay, then an additional atmospherically derived input of 0.5 - 2.8 x 10^5 kg N/yr occurs from the watershed.

C. Mercury Deposition Network Data

This section discusses 1998 MDN data from four sites in Maine, two New Hampshire sites (New Castle, on the coast and Laconia in the lakes region) and two Canadian sites (St Andrews, New Brunswick, and an in-land site at Kejimkujik, Nova Scotia). The reader is referred to the MDN Quality Assurance Project Plan (Casco Bay Estuary Project, 2001) for details on the sampling system and analytical procedures used in the collection and analysis of the MDN network samples. Locations of these MDN sampling sites are shown in Figure 51 (except the Kejimkujik Park site).

NH00 - Laconia
NH05 - New Castle
ME02 - Bridgton
ME09 - Greenville
ME96 - Freeport
ME98 - Acadia
NB02 - St Andrews

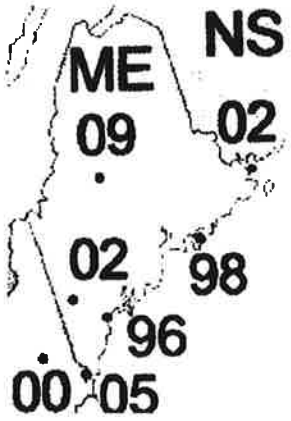


Figure 51. MDN Sites

Mercury Concentrations and Deposition at the Freeport site

Mercury concentrations in precipitation measured in the composite, one-week samples collected at Freeport, are presented in Figure 52. This figure also shows the wet mercury deposition values associated with each of the weekly samples. Clearly, samples containing high mercury concentrations do not always result in large depositional inputs of mercury. A few large deposition events contribute a significant portion of the annual mercury wet deposition at Freeport. The highest weekly deposition occurred during the week of June 9, 1998. During this one week period, over 18% of the annual mercury deposition occurred at Freeport. This week was also the largest single contributor to annual deposition at the Laconia, New Castle, Bridgton

and Greenville MDN sites. In fact, deposition during seven of the fifty one weeks (14%) at Freeport accounted for 53% of the total annual input at the site.

Mercury Concentrations in Precipitation at Eight MDN sites

A summary of weekly wet mercury concentrations recorded at the MDN sites during 1998 is presented in Table 15. The Laconia site began operation on April 28, 1998 and did not have a complete year of data. All other sites operated during the entire year. One pattern is evident in the annual mean volume weighted mean concentrations at these eight sites; the relatively uniform nature of the mercury concentrations. There is only roughly a factor of 1.5 difference seen in the annual mean values, and this is much less than observed for many of the other atmospheric aerosol data we have examined. Uniformity such as this occurred for the nitrogen and sulfur compounds, calcium and pH measured at the NADP sites, but not for any other chemical components of precipitation. Mercury vapor exhibits a relatively long residence time in the atmosphere (~ year) and unlike other pollutants (except nitrogen), mercury may be revolatilized or re-emitted into the atmosphere from surfaces. In general, the annual mean mercury concentrations in precipitation are highest at the two southern coastal sites (Freeport and New Castle), and decease as one moves northward and inland.

Seasonal trends in the concentration of mercury at the MDN sites are also very similar (Table 15). Highest concentrations are observed during the summer months at all sites. The lowest concentrations occur at all sites during the winter months. On average summer mercury concentrations in precipitation are 1.8 to 3.7 times those found in the winter across the seven sites (excluding Laconia, NH). This seasonal change may be due to the relatively volatile nature of mercury. Mercury in the atmosphere is found almost exclusively in the gas phase (as Hg^0). A significant portion of this gas phase mercury may be re-volatilized mercury that has been deposited on the surface. Recycling of previously deposited mercury will be most important during the summer and less significant during the winter when colder temperatures and lower photochemical activity result in lower evasion of mercury.

North-south patterns in the average mercury concentrations are evident across the network during 1988. During three of the four seasons, (winter, summer, and fall) the average concentrations are highest are highest at the two southernmost sites of New Castle and Freeport (Table 15). During the spring, the highest concentrations are observed at the coastal sites with St Andrews recording the highest concentration of mercury. During the winter the highest average concentrations were observed at the in-land Bridgton site, while in the fall the volume weighted mean concentration was greatest at western and southern sites (Laconia, New Castle, and Freeport). The northernmost sites (Greenville, ME and Kejimkujik, NS) generally record lower average concentrations in precipitation throughout the year than their southern and western counterparts.

Examination of the MDN Mercury and NADP Ion Chemistry Data

The MDN network operates on the same time schedule as the co-located NADP network. This allows for a course comparison of the two data sets. The NADP and MDN samplers integrate precipitation samples over a period of one week, which potentially may include several precipitation events. This sampling frequency may obscure relationships that might be more apparent in event-based sampling. A correlation coefficient (r) matrix for the two data sets (Table

16) highlights potential relationships between mercury and other contaminants in precipitation. The highest correlations exhibited by mercury are with sulfate ($r = 0.675$) and precipitation pH (laboratory), the latter exhibiting a fairly strong negative correlation of $r = -0.640$. More pollutant and acidity laden precipitation tends to also contain more mercury. Plots of sulfate and pH versus mercury are shown in Figures 53 and 54.

Mercury Deposition and Precipitation at MDN Sites in the Northeast

It is useful to compare seasonal mercury deposition (1988 data) observed among these MDN sites and this is done in Table 17. During the summer and fall months, and on an annual basis, the highest mercury deposition across the sites occurs at Freeport. In general, mercury deposition was greatest during the summer and least during the winter. The annual (1998) and seasonal precipitation amounts are shown in Table 18. Mercury concentrations in precipitation and mercury deposition show similar seasonal patterns. Deposition and precipitation exhibit a decidedly inverse relationship at several of the sites during the summer months. The southern Maine sites (Freeport, Bridgton) and northern inland site (Greenville) record the greatest mercury deposition and precipitation depths during the summer months. Conversely, while deposition is also greatest during the summer months at the more northerly sites of Acadia and Kejimkujik, these are the driest months at these two sites.

At six of the seven sites, mercury deposition was found to be highest during the summer months. Throughout the year, atmospheric mercury is predominantly in the gas phase, as elemental mercury. Elemental mercury is very insoluble and must be oxidized to the more soluble Hg^{+2} form which tends to be in particulate form. The particulate form of mercury is efficiently scavenged in precipitation. Atmospheric gases such as ozone, and chlorine and bromine compounds generated from the ocean, play an important role in mercury oxidation along the coast and inland. Ozone concentrations are higher during the summer months and some of the highest ozone levels are recorded along the coast. Factors such as higher temperatures (higher volatility of mercury from all surfaces) and higher oxidant levels (more rapid conversion to soluble mercury) play an important role in the removal of mercury from the atmosphere in precipitation during the summer.

In general, mercury deposition at the inland sites was considerably lower than at the coastal New Hampshire and Maine sites. This suggests that, as in the case of the IMPROVE aerosol data, the coastal environment is influenced more by pollutants along this corridor. Complex factors interact to influence the atmospheric input of pollutants such as mercury to both terrestrial and marine ecosystems. Seasonal variations in factors such as mercury chemistry, meteorological flow patterns, and emissions all affect the pollutant concentrations in precipitation. Inter-annual variations in precipitation amount and timing, and other factors result in significant changes in year-to-year deposition patterns.

The relationship of precipitation amount and mercury deposition at the seven sites for the calendar year 1998 is plotted in Figure 55. The data exhibit a good correlation of $r = 0.813$ ($r^2 = 0.660$). In general, higher precipitation rates lead to greater mercury deposition. Seasonal variations in the precipitation/mercury deposition relationship are presented in Figure 56. Wet deposition is thought to be the dominant mechanism that removes mercury from the atmosphere. The winter and summer data form the upper and lower boundaries of the scatter plot, with spring and fall values falling in-between. During the relatively dry winter months this relationship is

relatively steep. During the wetter summer months this relationship is much more flat, with moderate increases in precipitation producing significantly greater increases in mercury deposition.

Very low precipitation and deposition values were recorded at the St. Andrews site during the summer of 1998. As one moves north along the coast of the Gulf of Maine, the incidence of fog increases. Fog is especially prevalent during the summer months and it may be that an increase in fog events during the summer at the northerly coastal site results in significant scavenging of mercury (and other pollutants) which may partially explain the low deposition rate at St. Andrews. Many other factors play a role in determining Hg in deposition along the coast. Certainly coastal areas that remain bathed in fog also experience lower temperatures and lower solar radiation, two factors which play a role in revolatilization and mercury transformation chemistry. All things considered, however, the importance of fog in the removal of pollutants in the coastal environment is not well understood at present.

Atmospheric Inputs of Mercury to Casco Bay

The Freeport MDN data can provide a semi-quantitative estimate of the magnitude of the input of atmospheric mercury into Casco Bay. The predominant form of Hg found in the atmosphere is Hg^0 gas, a form which is not readily soluble in precipitation. Dry deposition of mercury gases and particles is generally not considered as important as wet deposition. Estimates of dry deposition of mercury vary. No consensus on the importance of dry deposition has been reached, although many studies estimate that as much as 1/3 of the total direct deposition of mercury to water surfaces may be from dry deposition. The MDN wet deposition values may be used to provide a "back-of-the -envelope"calculation of the direct input of total mercury to the surface of Casco Bay. Assuming an area for Casco Bay of 518 km^2), the annual input is calculated as 4.7 kg/Hg to the surface of the Bay (Table 19). This is a conservative estimate of atmospherically derived mercury input, since mercury is also deposited to the watershed and a small amount of this deposition certainly finds its way into the Bay in the form of runoff and riverine flow. Input from the surrounding watershed is probably most important during the spring melt when accumulated snowfall and attendant pollutants are flushed into the Bay, having little interaction with the frozen soil.

Estimates of point source inputs from NOAA for the year 1991 allow us to put the atmospheric flux into perspective. We can see that direct input from the atmosphere is roughly 25-30% that derived from known water pollution point sources (18.2 kg/yr). As these point sources come under increasing regulation, it is evident that input of mercury from the atmosphere to Casco Bay will become even more significant.

D. PAH Data

This section of the Casco Bay Air Deposition Data Report provides information on 16 dry deposition and 12 wet deposition PAH samples collected at the Freeport site between March 17, 1997 and February 15, 1998. PAH collection and analysis was conducted by Dr. Dan Golomb and his colleagues from the University of Massachusetts at Lowell. The reader is referred to the PAH section of the Casco Bay Estuary Project QAPP for details on the sampling system and

analytical procedures used in the collection and analysis of the PAH samples collected at Wolfe's Neck.

PAH Concentrations in Dry and Wet Deposition at Freeport, ME

Sixteen PAH compounds were measured in dry and wet deposition samples collected at Freeport ME monitoring site. These data are presented in Tables 20 and 21.

Dry and Wet Deposition

Sixteen (16) separate weekly composite dry deposition samples were collected over the twelve month sampling period. The average total PAH deposition rate was 8.7 ng m^{-2} hr^{-1} with values ranging from 2.7 to 20.1 ng m^{-2} hr^{-1}. The major PAH components (by weight) that were measured in the dry deposition samples at Freeport were fluoranthene (25%), pyrene (21%), and fluorene (8%) (Figure 57 and Table 22). Dry deposition rates were lowest in the summer and highest during the winter months. Combustion sources of all kinds (the burning of wood, coal, oil, municipal and solid waste, forest fires, and motor vehicle emissions) are know to be sources of PAHs and thus increased fuel use during the winter months may partially explain the higher concentrations of the PAHs found in dry deposition in the winter. Golomb et al (1999) found similar dry deposition rates at a remote monitoring site in Truro, MA (Cape Cod), another coastal site 150 miles south of Freeport. Much higher PAH deposition rates were found at Nahant MA, however, another coastal monitoring site located downwind of Boston and Logan International Airport.

Twelve (12) wet deposition samples were collected during the period March 1997 through February 1998. Four of the sixteen sampling weeks contained no sample because no precipitation fell. The PAH content of the wet deposition is shown in Table 21. The average weekly wet deposition rate was 2,234 ng/m^2 and samples ranged from 587 to 5664 ng/m^2. Unlike the dry deposition data, there was no clear seasonal variation in the wet deposition rates for total PAHs at Freeport. Major PAHs found in the wet deposition were phenanthrene (21%), fluorene, (17%) and fluoranthene (11%) (Figure 58 and Table 22). The relationship between PAH concentration and precipitation depth is shown in Figure 59. The amount of precipitation does not appear to influence the PAH concentration in precipitation collected at Freeport.

The sixteen PAHs that were analyzed in the samples from Wolfe's Neck represent a subset of organic compounds from a family of more than a hundred compounds composed of two or more aromatic rings. In general, as the molecular weight of PAH compounds increases, their atmospheric volatility decreases, their solubility in water decreases, and their solubility in fatty tissues increases. As distances from source(s) increase, the original composition of PAHs from emissions becomes fractionated as the effects of solubility and volatility work to separate the various PAH compounds in the atmosphere.

Potential Sources of PAHs in Dry and Wet Deposition at Freeport

Many combustion sources emit PAHs into the atmosphere. The PAH data from Freeport were compared to the PAH distribution in the wet and dry samples from literature derived source profiles in an effort to learn which sources might be important PAH contributors. Khalli et al. (1995) measured a suite of PAH compounds in several single and mixed combustion sources. These authors attempted to determine the PAH source fingerprints for coke ovens, diesel and

gasoline engines, highway tunnels, and wood combustion emissions. We have selected four of these profiles (diesel, tunnel, gasoline, and wood) and compared them with the PAH profiles found in the wet and dry deposition samples collected at Freeport. Typically, individual PAH concentrations are normalized to those of benz(e)pyrene or benz(a)pyrene to facilitate the comparison between different test results. We first normalized the PAH concentrations in each sample to the sample benzo(a)pyrene concentration.

The average ratios found for the 16 dry and 8 wet deposition samples collected at Freeport are presented in Table 23 along with the source profile ratios reported by Khalli et al. (1995). This comparison does not allow unambiguous conclusions concerning the source or sources of PAHs found in wet and dry deposition at Freeport, even though some of the wet and dry samples agree well with one or more of the source profiles. For example, the anthracene/ benzo(a) pyrene found in the Freeport samples (0.78 and 0.88 for dry and wet samples respectively) seems to agree well with the source ratio found in diesel exhaust (0.831). However, no definitive source could be identified using this approach in the majority of cases. This may be because the wet and dry samples are collected for a seven day period, thus integrating inputs from many sources. In addition these comparisons are made between mean annual wet and dry ratios with point source literature data. Multiple local and regional emission sources emit PAHs and contribute to the wet and dry deposition measured at Freeport. Therefore, it is not unusual that no definitive source(s) can be identified.

PAH Wet and Dry Deposition to Casco Bay

Using the results of the weekly wet and dry deposition sampling at Freeport, we may estimate the annual atmospheric input of total PAHs to Casco Bay. An average dry deposition rate of 8.7 ng m^{-2} hr^{-1} was determined for the 16 weekly samples collected over the period of March 1997 to February 1998. Scaling this figure to an annual basis (the area of Casco Bay 518 km^2), dry deposition to the Bay surface was calculated as 39 kg total PAHs/yr.

Wet deposition inputs to Casco Bay were estimated using the 10 wet deposition samples collected in 1998 and the NADP precipitation depths reported for the sampling intervals. This scaling process yielded a wet deposition input of approximately 25 kg PAHs/yr to Casco Bay via wet deposition. As shown in Table 12, direct atmospheric deposition to the surface of Casco Bay introduces approximately 64 kg PAHs/yr, with roughly 60% of the deposition input as dry deposition. It is difficult to put this rough estimate of atmospheric deposition into context with other sources of PAH to the Bay due to a lack of estimates of inputs from other sources to sediment PAH burdens. Previous studies have found that PAH sediment concentrations in close proximity to Portland do exceed levels believed to produce toxic responses in benthic organisms (Kennicutt et al., 1992). The general concentration pattern of PAHs in sediments observed by these authors, higher in-shore concentrations and lowest values in the outer Bay, suggests that riverine and local runoff inputs are certainly important for PAHs to Casco Bay sediments.

Table 1. Summary Statistics for Measured Elements and Compounds - IMPROVE Data Freeport, ME.

	Mean	Standard Deviation	Median	Range	Minimum	Maximum	Number
Species	Species Concentration (ng/m^3)						
Arsenic	0.360	0.267	0.249	0.806	0.868	0.062	27
Bromine	1.61	1.22	1.39	5.82	0.184	6.00	64
Calcium	15.9	13.9	12.4	56.9	1.99	58.9	54
Copper	1.26	1.26	0.812	5.16	0.122	5.28	41
Chlorine[a]	123	220	51.8	658	5.56	664	8
Chlorine[b]	60.1	185	8.44	752	2.61	754	16
Chloride ion	138	231	56.0	1630	0.816	1630	67
Iron	15.3	16.8	8.79	101	0.276	101	68
Gallium	0.261	0.218	0.266	0.450	0.031	0.481	4
H	144	105	119	412	29.2	441	44
Fine Mass	5050	5450	3530	41300	160	41500	89
Nickel	1.93	3.77	1.12	24.3	0.0614	24.3	45
Aluminum	24.3	15.9	20.7	62.2	1.72	63.9	34
Gold	4.41	3.50	5.20	6.87	0.580	7.45	3
Lead	2.15	1.71	2.13	8.52	0.153	8.67	64
Chromium	3.47	2.53	2.74	9.80	0.367	10.2	30
Potassium	23.2	17.4	17.7	65.7	2.79	68.4	60
Magnesium	36.3	34.4	26.2	140	2.12	142	23
Manganese	3.79	2.67	3.90	13.4	0.249	13.7	38
Mercury	0.958	0.401	1.07	1.09	0.195	1.28	6
Sodium	116	186	69.4	1070	6.76	1070	37
Phosphorus	12.9	3.96	13.9	9.28	7.28	16.6	4
Sulfur	327	258	254	1030	4.69	1030	61
Silicon	30.8	19.5	25.4	103	4.52	107	59

Species	Mean	Standard Deviation	Median	Range	Minimum	Maximum	Number
Titanium	4.70	3.39	3.96	12.2	0.553	12.8	53
Rubidium	0.677	0.446	0.717	1.63	0.114	1.74	20
Vanadium	5.48	4.36	3.95	15.7	0.397	16.1	48
Selenium	0.432	0.427	0.268	1.81	0.0305	1.84	36
Strontium	0.468	0.432	0.247	1.32	0.0307	1.34	20
Yttrium	0.527	0.604	0.217	2.07	0.0307	2.10	16
Zinc	4.41	4.25	3.24	22.0	0.276	22.3	63
Zirconium	0.693	0.639	0.448	2.14	0.0307	2.17	30
Nitrite ion (NO_2)	18.8	20.5	10.4	93.1	0.286	93.4	35
Nitrate ion (NO_3)	532	530	353	2790	2.92	2790	74
Sulfate ion (SO_4)	2330	2350	1450	13900	153	14000	72

summary statistics for the period 1/23/98 - 5/30/99 with the exception of MF values which cover the period 1/23/98- 9/3/99
all values reported as ng/m^3
values rounded to three significant figures
[a] = determined by XRF
[b] = determined by PIXE

	As	Br	Ca	Cu	Fe	H	MF	Ni	Al	Pb	Cr	K	Mg	Mn	Na	Si	Ti	V	Rb	Se	Sr	Y	Zn	Zr	Cl	NO2	NO3	SO4	EC	OC
As	1.00																													
Br	0.63	1.00																												
Ca	0.38	0.72	1.00																											
Cu	0.73	0.38	0.36	1.00																										
Fe	0.56	0.54	0.56	0.63	1.00																									
H	0.59	0.60	0.50	0.21	0.35	1.00																								
MF	0.28	0.73	0.53	0.16	0.32	0.77	1.00																							
Ni	0.68	0.28	0.34	0.55	0.13	0.89	0.22	1.00																						
Al	0.52	0.57	0.35	0.14	0.38	0.68	0.34	0.03	1.00																					
Pb	0.84	0.75	0.60	0.65	0.47	0.73	0.50	0.45	0.45	1.00																				
Cr	0.19	0.37	0.59	0.44	0.23	0.07	0.06	0.10	0.31	0.55	1.00																			
K	0.61	0.56	0.33	0.09	0.33	0.86	0.66	0.14	0.59	0.38	-0.25	1.00																		
Mg	0.03	0.74	0.52	-0.11	-0.02	0.43	0.72	-0.11	0.46	0.17	-0.39	0.52	1.00																	
Mn	0.53	0.28	0.44	0.57	0.18	0.52	0.30	0.64	0.08	0.41	0.69	0.17	-0.14	1.00																
Na	0.20	0.64	0.50	-0.12	0.02	0.27	0.55	0.02	0.68	0.17	0.51	0.31	0.86	0.22	1.00															
Si	0.71	0.42	0.62	0.36	0.49	0.61	0.26	-0.01	0.64	0.50	0.30	0.44	-0.11	0.21	0.03	1.00														
Ti	0.50	0.31	0.40	0.61	0.25	0.44	0.07	0.52	0.22	0.40	0.72	0.14	-0.11	0.78	0.07	0.38	1.00													
V	0.46	0.45	0.51	0.59	0.53	0.69	0.47	0.81	0.34	0.62	0.60	0.42	-0.08	0.59	0.13	0.53	0.52	1.00												
Rb	0.47	0.82	0.72	0.80	0.53	0.45	0.46	0.29	0.13	0.86	0.72	0.05	-0.31	0.75	0.51	0.25	0.67	0.86	1.00											
Se	0.67	0.78	0.71	0.55	0.56	0.64	0.53	0.78	0.35	0.85	0.82	0.46	0.22	0.53	0.23	0.53	0.43	0.63	0.95	1.00										
Sr	-0.33	0.13	0.33	-0.38	-0.37	-0.04	-0.19	-0.10	-0.33	0.26	0.55	-0.11	0.52	0.31	0.71	-0.47	0.12	-0.30	0.80	0.20	1.00									
Y	-0.16	0.44	0.94	0.11	0.08	0.47	0.18	0.55	0.48	0.75	0.74	0.01	-0.16	0.41	0.71	0.31	0.89	0.44	0.99	0.86	0.69	1.00								
Zn	0.74	0.72	0.60	0.61	0.68	0.78	0.72	0.32	0.41	0.74	0.24	0.61	0.16	0.33	0.13	0.41	0.26	0.67	0.62	0.81	-0.47	0.31	1.00							
Zr	0.47	0.56	0.73	0.49	0.53	0.08	0.22	0.23	0.38	0.70	0.62	-0.01	-0.08	0.85	0.13	0.42	0.70	0.75	0.88	0.85	0.56	0.91	0.44	1.00						
Cl	-0.11	0.38	0.25	-0.07	-0.09	0.10	0.28	-0.08	0.23	-0.07	-0.25	0.16	0.79	0.01	0.92	-0.15	-0.15	-0.07	-0.17	0.02	0.42	0.08	-0.04	-0.08	1.00					
NO2	-0.43	-0.21	-0.10	-0.07	-0.24	-0.62	0.17	0.33	-0.45	-0.17	-0.37	-0.28	-0.51	-0.20	-0.25	-0.36	-0.41	-0.14	0.32	-0.30	0.94	0.05	-0.14	-0.09	-0.04	1.00				
NO3	0.09	0.38	0.15	0.08	0.09	0.58	0.63	0.13	0.17	0.21	-0.16	0.41	0.35	0.26	0.35	0.10	0.15	0.35	0.17	0.25	-0.20	0.03	0.41	0.17	0.27	-0.12	1.00			
SO4	0.14	0.19	0.02	0.09	0.01	0.31	0.30	0.01	0.17	0.32	0.14	0.07	0.37	0.27	0.37	0.12	0.29	0.31	0.37	0.46	0.35	0.67	0.31	0.35	0.09	-0.08	0.54	1.00		
EC	0.38	0.22	0.01	0.09	0.23	0.73	0.46	0.03	0.35	0.20	-0.13	0.52	0.02	0.32	-0.02	0.22	0.17	0.53	-0.27	0.40	-0.53	-0.01	0.53	-0.10	-0.11	-0.16	0.51	0.63	1.00	
OC	0.17	0.22	0.04	0.11	0.21	0.64	0.41	0.08	0.33	0.17	-0.13	0.44	-0.02	0.40	-0.01	0.16	0.19	0.48	-0.22	0.46	-0.46	0.02	0.48	-0.01	-0.05	-0.16	0.56	0.66	0.92	1.00

Table 2. Correlation Matrix (r) for the IMPROVE Freeport, ME Data.

Table 3.
Seasonal Summary Statistics - IMPROVE Data, Freeport, ME.

Species	Winter	Spring	Summer	Fall
Arsenic	0.469 ± 0.253 (7)	0.249 ± 0.272 (8)	0.173 ± 0.051 (3)	0.436 ± 0.275 (9)
Bromine	2.08 ± 1.13 (17)	1.68 ± 1.48 (26)	1.07 ± 0.637 (8)	1.18 ± 0.791 (13)
Calcium	17.2 ± 6.18 (14)	18.4 ± 19.8 (21)	7.52 ± 3.81 (7)	15.0 ± 10.3 (12)
Copper	0.973 ± 0.531(13)	1.23 ± 1.44 (11)	0.950 ± 1.07 (7)	1.89 ± 1.73 (10)
Chlorine[a]	89.1 (1)	142 ± 257 (6)	45.2 (1)	nd
Chlorine[b]	14.2 ± 16.4 (4)	103 ± 263 (8)	6.72 (1)	25.2 ± 11.6 (3)
Chloride ion	114 ± 151 (20)	190 ± 347 (24)	104 ± 116 (13)	101 ± 73.7 (10)
Iron	15.4 ± 8.51 (18)	14.1 ± 15.4 (26)	5.29 ± 2.68 (11)	26.0 ± 27.3 (13)
Gallium	0.123 (1)	0.031 (1)	0.446 (1)	nd
H	257 ± 96.1 (13)	51.4 ± 37.4 (12)	101 ± 36.4 (6)	136 ± 71.5 (13)
Hg	0.195 (1)	1.18 (2)	1.07 ± 0.200 (3)	nd
Fine Mass	5830 ± 2600 (20)	4190 ± 2880 (30)	6880 ± 9190 (25)	2510 ± 755 (14)
Nickel	3.60 ± 5.93 (15)	1.22 ± 2.01 (18)	0.869 ± 0.560 (8)	0.997 ± 0.339 (4)
Aluminum	30.5 ± 16.2 (13)	21.6 ± 16.0 (17)	nd	15.0 ± 6.22 (4)
Gold	7.45 (1)	2.89 (2)	nd	nd
Lead	2.56 ± 1.18 (18)	2.36 ± 2.29 (24)	1.99 ± 1.27 (9)	1.31 ± 1.07 (13)
Chromium	2.61 ± 1.85 (9)	5.14 ± 2.96 (11)	2.62 ± 2.26 (3)	2.31 ± 1.36 (7)
Potassium	39.7 ± 19.4 (19)	13.9 ± 9.57 (22)	11.6 ± 5.01 (6)	20.1 ± 9.32 (13)
Magnesium	39.2 ± 41.6 (7)	38.7 ± 58.2 (5)	31.9 ± 22.0 (4)	34.0 ± 9.07 (7)
Manganese	4.33 ± 3.06 (15)	3.10 ± 2.83 (13)	3.77 (1)	3.89 ± 1.77 (9)
Sodium	142 ± 96.1 (13)	164 ± 335 (10)	33.8 ± 40.7 (3)	64.6 ± 59.6 (11)
Phosphorous	7.28 (1)	13.7 (1)	14.1 (1)	16.6 (1)
Sulfur	465 ± 267 (19)	293 ± 282 (23)	223 ± 84.9 (6)	232 ± 173 (13)
Silicon	35.7 ± 18.0 (18)	30.7 ± 25.6 (23)	24.4 ± 9.04 (6)	27.1 ± 9.33 (12)
Titanium	5.59 ± 3.24 (17)	3.88 ± 3.28 (20)	4.37 ± 2.17 (5)	4.95 ± 3.72 (11)
Vanadium	6.68 ± 3.79 (17)	3.72 ± 5.17 (13)	3.61 ± 2.48 (6)	6.61 ± 4.40 (12)

Species	Winter	Spring	Summer	Fall
Rubidium	0.558 ± 0.369 (6)	1.07 ± 0.362 (6)	0.540 ± 0.426 (6)	0.248 ± 0.136 (2)
Selenium	0.485 ± 0.346 (10)	0.386 ± 0.552 (13)	0.596 ± 0.450 (5)	0.337 ± 0.281 (8)
Strontium	0.259 ± 0.199 (8)	0.923 ± 0.615 (4)	0.782 ± 0.124 (4)	0.118 ± 0.069 (4)
Yttrium	0.237 ± 0.171 (5)	0.606 ± 0.704 (4)	0.888 ± 0.796 (5)	0.191 ± 0.0957 (2)
Zinc	6.50 ± 4.31 (18)	3.53 ± 4.77 (25)	2.42 ± 0.589 (7)	4.26 ± 3.31 (13)
Zirconium	0.324 ± 0.268 (8)	0.919 ± 0.805(13)	0.773 ± 0.504 (7)	0.426 ± 0.314 (2)
Nitrite ion (NO_2)	21.2 ± 19.0 (8)	24.3 ± 23.5 (18)	7.11 ± 6.76 (5)	4.04 ± 2.84 (4)
Nitrate ion (NO_3)	866 ± 644 (19)	432 ± 477 (30)	533 ± 452 (13)	256 ± 246 (12)
Sulfate ion (SO_4)	1910 ± 1140 (19)	2290 ± 2320 (29)	4130 ± 3670 (12)	1300 ± 1200 (12)

[a] = determined by XRF
[b] = determined by PIXE
seasonal summary statistics compiled for the period 1/23/98 - 5/30/99 with the exception of Fine Mass where data period is 1/23/98 - 9/3/99
Winter = December, January and February
Spring = March, April and May
Summer = June, July and August
Fall = September, October and November
nd = no data
all values reported as ng/m^3, rounded to three significant figures.

Table 4.
Comparison of Freeport and Acadia National Park (ME) and St Andrews, Canada IMPROVE Trace Metal Data.

Species	Freeport	Acadia National Park	St Andrews, Canada
Arsenic	0.360 ± 0.267 (27)	0.288 (980)	0.400 ± 0.029
Chromium	3.47 ± 2.53 (30)	0.678 (980)	0.107 ± 0.0068
Copper	1.26 ± 1.26 (41)	0.95 (980)	0.666 ±0.141
Lead	2.14 ± 1.71 (64)	2.79 (980)	1.58 ± 0.098
Nickel	1.93 ± 3.77 (45)	1.04 (980)	0.669 ± 0.050
Selenium	0.432 ± 0.427 (36)	0.400 (980)	0.332 ± 0.027
Vanadium	5.48 ± 4.36 (48)	2.76 (980)	1.65 ± 0.104
Zinc	4.41 ± 4.25 (63)	5.54 (980)	4.89 ± 0.443

all concentrations reported as ng/m^3
(n) = number of samples.
n = 125 for all St Andrews data, collection period 15 May, 1996 - 26 July, 1997.
St. Andrews data courtesy of Dr. J. L. Campbell, University of Guelph, Ontario, Canada, for internal use only, do not publish.
Acadia National Park data from IMPROVE (1988 through 1997).

Table 5.
IMPROVE Fine Mass Composite Variables.

IMPROVE Composite Variables		
Abbreviation	**Component**	**Composition**
KNON	Nonsoil Potassium	[K] - 0.6[Fe] a qualitative smoke tracer
NHSO	Ammonium Sulfate $(NH_4)_2SO_4$	4.125[S]†
SOIL	Fine Soil Component	2.20[Al] + 2.49[Si] + 1.63[Ca] + 2.42[Fe] + 1.94[Ti]
NHNO	Ammonium Nitrate $(NH_4)_2NO_3$	1.29[NO_3]
OC	Total Organic Carbon	[OC1] + [OC2] +[OC3] + [OC4] + [OP]
OMC	Organic Mass by Carbon	1.4[OC]
LAC	Light Absorbing Carbon	[EC1] + [EC2] + [EC3] - [OP]
TC	Total Carbon	[OC1] + [OC2] + [OC3] + [OC4] + [EC1] + [EC2] + [EC3]
RCMC	Reconstructed Mass without Nitrate	[NHSO]† + [SOIL] + [OMC] + [LAC] + 1.4[KNON] + 2.5[NA]
∑Species	Sum of Species	[Trace Elements] + [Ions] + [LAC]+ [OC]†*
Sea Salt	Marine Component	[Chloride ion] + [Na]

†sulfate-sulfur used
*chloride ion used for chlorine

Table 6.
Summary Statistics for Composite Variables - IMPROVE Data Freeport, ME.

Variable	Mean	Standard Deviation	Median	Range	Minimum	Maximum	Number
KNON	15.8	15.6	8.60	56.9	0.849	57.8	55
NHSO	1350	1060	1050	4230	19.3	4250	61
SOIL	154	120	114	440	7.28	447	70
Sea Salt	190	354	89.1	2700	0.816	2700	71
NHNO	687	684	455	3600	3.76	3600	74
OC	1840	1180	1500	5260	388	5650	71
OMC	2580	1650	2110	7360	544	7910	71
LAC	511	326	412	1600	93.0	1690	71
TC	2350	1480	1920	6610	503	7110	71
RCMC	4270	2690	3690	12500	51.2	12600	76
RCFM	4720	3100	4030	14400	47.0	14500	77

summary statistics for the period 1/23/98 - 5/30/99
all values reported as ng/m^3
values rounded to three significant figures

Table 7.
Typical source profile categories for the IMPROVE Aerosol Freeport, ME data.

Source of Aerosol Component	Important Species
Naturally Produced Aerosol	
Soil	Fe, Al, Ca, Ti, Si, Mn
Marine	Na, Cl
Anthropogenic Aerosol	
Industrial	V, Ni, Hg, As, Se, Pb, Cu, Zn, Cr, SO_4, NO_3
Residential	SO_4, NO_3, K
Carbon	Organic carbon (OC), Elemental Carbon (LAC)
Mobile	NO_2, NO_3, Rb
Mixed	K, Br

Table 8.
Major aerosol components and their contribution to the $PM_{2.5}$ fine mass at the Freeport, ME site for the period 1/23/98 - 5/30/99.

Component	All samples	Winter	Spring	Summer	Fall
Soil	7.9 ± 17 (66)	5.3 ± 3.0 (17)	6.5 ± 7.2 (26)	1.6 ± 0.94 (11)	11± 10 (11)
Marine (Sea Salt)	4.3 ± 6.0 (71)	3.2 ± 2.7 (20)	5.2 ± 7.7 (25)	1.5 ± 2.6 (13)	7.1 ±7.6 (13)
Organic Carbon (OC)	35 ± 13 (71)	38 ± 11 (18)	31 ± 11 (30)	37 ± 16 (13)	41 ± 10 (10)
Sulfate	40 ± 14 (72)	31 ± 8.4 (19)	45 ± 11 (29)	49 ± 16 (12)	34 ± 15 (12)

(n) = number of samples
no data reported for summer months, see text for discussion.

Table 9.
Major Aerosol Events at the Freeport Me, IMPROVE Site.

Date	Ranking - Species	Source Region - Trajectory
January 29, 1998	1: Ni	coastal Maine, Nova Scotia - **Figure 37**
February 22, 1998	2: Cr, Pb, Ti 4: Mn	northern New England, Quebec, Montreal - **Figure 38**
March 18, 1998	1: Br, MF, Pb, Rb, Se, Zn, Zr, 2: Ca, Cu, Fe, Ni, V, 4: NO_3	New England - **Figure 39**
April 23, 1998	1: Cr 4: Rb	Coastal New England - **Figure 40**
May 29, 1998	2: SO_4 4: OC	Northern Mid-west, Toronto Detroit - **Figure 41**
July 16, 1998	1: SO_4, OC, Sum Species, TC 2: LAC, Se	no meteorological data available
July 22, 1998	3: SO_4 4: Mg	northern New England , southern Ontario, Toronto - **Figure 42**
July 28, 1998	5: SO_4, Cl, Cu	Ohio River Valley - **Figure 43**
December 7, 1998	1: NO_3 2: Marine 3: OC, Na, Cl, Na, Sum Species 4: SO_4, TC, LAC, Ti 5: H	Pennsylvania, Ohio River Valley - **Figure 44**
December 31, 1998	1: K, , KNON, As	Ottawa, Montreal - **Figure 45**
January 6, 1999	1: V, LAC 2: OC, H, MF, TC 3: Zn 5: Al, Sum Species	southern New Hampshire, western Massachusetts - **Figure 46**
January 12, 1999	1: H 2: Zn 3: NO_3, MF 5: Ni	Ohio River Valley, US east coast - **Figure 47**
March 25, 1999	2: NO_3	Montreal - **Figure 48**

sampling period 1/23/98 - 5/30/99.
species ranking: 1 = highest level of species observed during sampling period, etc.

Table 10.
Volume Weighted Mean Ion Concentrations (mg/L) at the Freeport NADP Site for the period 12/30/97 - 12/29/98.

Species/Season	All Samples	Winter	Spring	Summer	Fall
Ca	0.06	0.02	0.07	0.05	0.09
Mg	0.067	0.029	0.037	0.085	0.174
K	0.026	0.011	0.017	0.03	0.064
Na	0.602	0.259	0.271	0.754	1.74
Cl	1.01	0.45	0.49	1.31	2.72
NH_4	0.1	0.03	0.14	0.11	0.15
NO_3	0.85	0.37	0.89	0.8	1.84
SO_4	1.11	0.5	0.95	1.34	2.17
pH	4.61	4.91	4.69	4.54	4.29

Table 11.
Annual Volume Weighted Mean Ion Concentrations (mg/L) at Four Maine NADP Sites.

Species/Site	Freeport	Acadia	Bridgton	Greenville
Ca	0.06	0.05	0.04	0.04
Mg	0.067	0.063	0.015	0.01
K	0.026	0.039	0.012	0.013
Na	0.602	0.533	0.093	0.049
NH_4	0.1	0.09	0.12	0.1
NO_3	0.85	0.79	0.88	0.75
Cl	1.01	0.94	0.18	0.12
SO_4	1.11	1.2	1.0	0.84
pH	4.61	4.58	4.60	4.65

Table 12.
Annual Wet Deposition (kg/ha) at four Maine NADP sites for the period 12/30/97 - 12/29/98.

Species/Site	Freeport	Acadia	Bridgton	Greenville
Ca	0.72	0.79	0.46	0.40
Mg	0.86	0.93	0.17	0.11
K	0.33	0.57	0.13	0.14
Na	7.75	7.83	1.03	0.56
NH_4	1.32	1.32	1.32	1.08
NO_3	10.9	11.6	9.76	8.52
Inorganic N	3.49	3.64	3.23	2.76
Cl	13.1	13.8	2.01	1.33
SO_4	14.2	17.7	11.0	9.53
H^+	0.32	0.39	0.28	0.25

H^+ - from pH measured in lab

Table 13.
Annual Precipitation Depth at four Maine NADP sites for the period 12/30/97 - 12/29/98.

Site/Annual Precipitation	Precipitation Depth (mm)	Precipitation Depth (inches)
Freeport	1286.7	50.7
Acadia	1467.4	57.8
Bridgton	1103.9	43.5
Greenville	1131.3	44.5

Table 14.
Estimates of Atmospheric and Point Source Nitrogen Inputs to Casco Bay.

Source	Annual Input to Casco Bay (10^5 kg N/yr)
Wet deposition	1.8
Dry Deposition	1.8
Total Atmospheric	3.6
Point Source	5.4

point source estimate from NOAA (1994) for the year 1991
dry deposition estimated as equal to wet.

Table 15.
Statistical Summary of Mercury Volume Weighted Mean Concentrations in Precipitation at Eight MDN Sites

Site/Season	All	Winter	Spring	Summer	Fall
Laconia				7.12	8.92
New Castle	7.69	4.12	7.29	11.2	7.21
Bridgton	6.56	4.60	6.30	8.07	6.24
Greenville	5.87	2.55	4.08	9.53	5.42
Freeport	7.78	4.52	6.56	11.2	7.08
Acadia	6.06	3.29	6.57	10.4	6.26
St Andrews, NB	6.54	3.57	8.77	8.81	5.61
Kejimkujik, NS	5.33	3.35	4.95	11.2	4.24

concentration units = volume weighted mean - ng/l
sampling dates - 12/30/97- 12/29/98 except Freeport (1/6/98 - 12/29/98) and Laconia (4/28/98 - 12/29/98)

Table 16.
Correlation Matrix (r) - MDN and NADP Data Freeport, ME.

	Hg	Ca	Mg	K	Na	NH4	NO3	Cl	SO4	Lab pH	Field pH	Lab Cond	Field Cond
Hg	1												
Ca	0.482	1											
Mg	0.064	0.593	1										
K	0.361	0.684	0.467	1									
Na	-0.014	0.476	0.983	0.319	1								
NH4	0.595	0.796	0.175	0.666	0.041	1							
NO3	0.529	0.793	0.452	0.554	0.365	0.704	1						
Cl	-0.003	0.471	0.986	0.345	0.997	0.040	0.346	1					
SO4	0.675	0.771	0.398	0.587	0.306	0.834	0.878	0.298	1				
Lab pH	-0.640	-0.610	-0.377	-0.406	-0.318	-0.550	-0.840	-0.310	-0.842	1			
Field pH	-0.587	-0.591	-0.282	-0.413	-0.217	-0.505	-0.831	-0.213	-0.803	0.978	1		
Lab Cond	0.574	0.737	0.576	0.515	0.515	0.615	0.939	0.500	0.925	-0.896	-0.856	1	
Field Cond	0.552	0.708	0.469	0.609	0.381	0.612	0.947	0.376	0.924	-0.875	-0.858	0.988	1

Table 17.
Annual and Seasonal Mercury Wet Deposition (ng/m^2) at Seven MDN Sites.

Site	Annual	Winter	Spring	Summer	Fall
New Castle	7786	852 (11)	2272 (29)	3067 (39)	1594 (20)
Freeport	12069	1270 (11)	2318 (19)	5402 (45)	3080 (26)
Bridgton	6897	816 (12)	1661 (24)	2716 (39)	1705 (25)
Acadia	8963	1677 (19)	2606 (29)	2743 (31)	1936 (22)
Greenville	6714	589 (9)	1192 (18)	3639 (54)	1294 (19)
St Andrews	7322	1023 (14)	2913 (40)	1587 (22)	1799 (25)
Kejimkujik	6406	1090 (17)	1453 (23)	2239 (35)	1624 (25)
Mean/S.D.	8022 ± 1975	1045 ± 353	2059 ± 635	3056 ± 1217	1862 ± 573

(#) = % of annual total
highest annual and seasonal deposition amounts (ng/m^2) are shown in red
sampling dates - 12/30/97- 12/29/98 except Freeport (1/6/98 - 12/29/98)

Table 18.
Annual and Seasonal Mercury Precipitation Depths (cm) at Seven MDN Sites.

Site	Annual	Winter	Spring	Summer	Fall
New Castle	101.25	20.67 (20)	31.17 (31)	27.30 (27)	22.12 (22)
Freeport	155.15	28.10 (18)	35.33 (23)	48.21 (31)	43.50 (28)
Bridgton	105.09	17.74 (17)	26.37 (25)	33.66 (32)	27.32 (26)
Acadia	147.94	51.04 (35)	39.66 (27)	26.30 (18)	30.93 (21)
Greenville	114.37	23.08 (20)	29.24 (26)	38.20 (33)	23.85 (21)
St. Andrews	111.98	28.65 (26)	33.21 (30)	18.01 (16)	32.10 (29)
Kejimkujik	120.24	32.51 (27)	29.39 (24)	20.07 (17)	38.26 (32)

(#) = % of annual total
highest annual and seasonal amounts are shown in red
sampling dates - 12/30/97- 12/29/98 except Freeport (1/6/98 - 12/29/98)

Table 19.
Estimates of the Annual Atmospheric and Point Source Mercury Inputs to Casco Bay.

Source	Annual Input to Casco Bay (kg Hg/yr)
Direct Wet Deposition	4.7
Point Source	18.2

point source estimate from NOAA (1994) for the year 1991.

Table 20.
PAH Concentrations in Dry Deposition Samples at Freeport, ME.

Dry - ng/m²/hr	17-Mar	7-Apr	14-Apr	21-Apr	5-May	12-May	2-Jun	9-Jun	14-Jul	11-Aug	8-Sep	13-Oct	10-Nov	18-Dec	12-Jan	9-Feb
Acenaphthylene	0.31	0.16	0.47	0.81	0.22	0.15	0.27	0.11	0.00	0.24	0.24	0.00	0.71	1.81	0.44	0.35
Fluorene	0.95	0.00	0.00	0.00	0.35	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.87	0.00
Phenanthrene	0.31	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2.85	0.00
Anthracene	0.93	0.04	0.14	0.05	0.22	0.22	0.24	0.32	0.00	0.28	0.43	0.00	0.25	0.00	0.22	0.13
Fluoranthene	3.57	0.76	1.94	1.78	0.85	0.83	1.20	1.91	2.67	2.33	0.94	1.88	2.55	3.77	2.71	4.50
Pyrene	1.71	0.43	1.46	1.00	0.55	0.64	0.75	1.15	8.38	2.97	1.50	4.85	1.92	2.20	1.78	3.72
Benz(a)anthracene	0.49	0.14	0.54	0.38	0.14	0.12	0.15	0.14	0.14	0.08	0.29	0.19	0.13	0.22	0.62	1.07
Chrysene	0.56	0.25	0.96	0.85	0.40	0.19	0.30	0.22	0.42	0.26	0.54	0.06	0.41	0.51	1.01	2.12
Benzo(b)fluoranthene	0.82	0.22	1.07	0.94	0.83	0.28	0.24	0.22	0.00	0.18	0.74	0.00	0.26	0.46	1.89	3.20
Benzo(k)fluoranthene	0.70	0.22	0.60	0.99	0.00	0.00	0.00	0.00	0.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Benzo(a)pyrene	0.99	0.12	0.73	0.61	0.45	0.00	0.00	0.00	0.00	0.00	0.41	0.00	0.07	0.19	0.77	1.36
Perylene	0.39	0.29	0.23	0.22	0.24	0.11	0.29	0.30	0.61	0.05	0.34	0.12	0.04	0.09	0.68	0.34
Indeno(1,2,3-c,d)pyrene	0.58	0.21	0.87	0.70	0.20	0.09	0.04	0.05	0.06	0.00	0.39	0.18	0.12	0.17	0.80	1.19
Dibenz(a,h)anthracene	0.40	0.00	0.27	0.28	0.00	0.00	0.00	0.00	0.00	0.00	0.18	0.00	0.03	0.00	0.31	0.38
Benzo(g,h,i)perylene	0.50	0.18	0.74	0.74	0.22	0.10	0.04	0.07	0.00	0.02	0.33	0.12	0.00	0.13	0.70	0.99
Coronene	0.27	0.16	0.49	0.62	0.00	0.00	0.00	0.00	0.00	0.00	0.26	0.20	0.02	0.25	0.92	0.78
Total	13.48	3.18	10.51	9.95	4.68	2.73	3.53	4.49	12.30	6.41	6.59	7.59	6.50	9.80	17.57	20.12

total 16 samples = 139.4 ng/m²/hraverage 16 samples = 8.7 ng/m²/hr

Table 21.
PAH Concentrations in Wet Deposition Samples at Freeport, ME.

Wet - ng/m²	17-Mar	7-Apr	14-Apr	21-Apr	5-May	12-May	2-Jun	9-Jun	14-Jul	11-Aug	8-Sep	13-Oct	10-Nov	18-Dec	12-Jan	9-Feb
Precipitation (cm)	0.0	1.60	0.0	6.10	3.25	5.72	3.43	7.11	0.66	0.70	0.0	29.72	0.0	0.84	2.92	31.50
Acenaphthylene	0.0	13.5	0. 0	39.3	15.0	15.5	44.8	109.0	68.7	0.0	0.0	119.6	0.0	122.7	312.3	97.6
Fluorene	0. 0	0.0	0. 0	0.0	0.0	149.3	0.0	1338.2	202.4	564.5	0.0	290.7	0.0	0.0	332.8	0.0
Phenanthrene	0. 0	0.0	0. 0	144.6	0.0	280.0	0.0	2194.0	168.4	1490.7	0.0	989.6	0.0	577.8	1805.8	381.1
Anthracene	0. 0	0.0	0. 0	14.9	38.4	0.0	33.7	217.9	63.8	126.6	0.0	91.5	0.0	0.0	40.7	17.3
Fluoranthene	0. 0	76.2	0. 0	329.6	121.7	89.5	88.2	130.4	87.2	163.6	0.0	221.7	0.0	635.9	992.5	348.2
Pyrene	0. 0	50.7	0. 0	209.2	91.5	78.1	84.0	196.8	106.4	63.4	0.0	168.7	0.0	1848.4	534.9	352.8
Benz(a)anthracene	0. 0	0.0	0. 0	64.2	32.2	29.0	32.6	19.6	22.1	24.9	0.0	40.6	0.0	77.6	130.5	130.3
Chrysene	0. 0	39.0	0. 0	148.7	50.0	13.4	65.2	14.4	31.7	0.0	0.0	72.7	0.0	98.9	238.5	97.6
Benzo(b)fluoranthene	0. 0	54.2	0. 0	170.7	123.2	19.4	142.5	0.0	0.0	9.7	0.0	93.4	0.0	178.0	401.6	295.8
Benzo(k)fluoranthene	0. 0	54.2	0. 0	127.2	0.0	0.0	45.5	0.0	0.0	16.8	0.0	0.0	0.0	40.2	0.0	0.0
Benzo(a)pyrene	0. 0	40.1	0. 0	110.9	45.7	8.6	40.6	0.0	0.0	0.0	0.0	30.4	0.0	74.7	127.4	121.5
Perylene	0. 0	85.3	0. 0	40.6	63.7	0.0	151.9	0.0	148.3	94.5	0.0	30.1	0.0	84.7	15.3	0.0
Indeno(1,2,3-c,d)pyrene	0. 0	60.4	0. 0	171.8	25.2	35.7	31.2	25.3	9.3	0.0	0.0	82.6	0.0	78.0	198.1	108.0
Dibenz(a,h)anthracene	0. 0	0.0	0. 0	56.3	0.0	0.0	0.0	0.0	0.0	21.4	0.0	67.2	0.0	88.1	93.6	75.2
Benzo(g,h,i)perylene	0. 0	56.8	0. 0	155.8	19.6	35.1	16.3	41.3	5.9	0.0	0.0	64.6	0.0	57.8	155.7	48.5
Coronene	0. 0	56.7	0. 0	157.4	0.0	0.0	10.5	0.0	0.0	0.0	0.0	75.8	0.0	108.5	283.9	87.0
Total	0. 0	586.9	0. 0	1941.2	626.3	753.6	787.1	4286.7	914.0	2576.1	0.0	2438.9	0.0	4071.2	5663.5	2160. 9

total 12 samples = 26,806 ng/m² average 12 samples = 2,234 ng/m²

Table 22.
Average PAH Composition for Wet and Dry Deposition at Freeport, ME.

Wet Deposition		Dry Deposition	
	%		%
phenanthrene	21.5	fluoranthene	25.1
fluorene	16.6	pyrene	21.5
fluoranthene	11.2	fluorene	8.3
pyrene	9.2	benzo(b)fluoranthene	7.7
coronene	7.5	phenanthrene	6.1
benzo(b)fluoranthene	5.2	chyrsene	5.7
benzo(a)pyrene	3.8	acenaphthylene	4.7
chyrsene	3.8	benzo(a)pyrene	4.6
perylene	3.3	benzo(k)fluoranthene	3.5
acenaphthylene	3.3	benz(a)anthracene	3.1
indeno(1,2,3-c,d)pyrene	3.2	indeno(1,2,3-c,d)pyrene	3.0
anthracene	3.0	coronene	2.9
benzo(k)fluoranthene	2.9	anthracene	2.9
benz(a)anthracene	2.5	benzo(g.h.i)perylene	2.8
benzo(g,h,i)perylene	2.5	perylene	2.8
dibenz(a,h)anthracene	2.1	dibenz(a,h)anthracene	1.9

geometric means

Table 23.
Mean Species/benzo(a)pyrene measured in wet and dry deposition at Freeport and in source profile samples.

Species	S/b(a)p-dry	S/b(a)p-wet	Diesel/ b(a)p	Tunnel/ b(a)p	Gasoline/ b(a)p	Wood/ b(a)p
acenaphthylene	2.5 ± 3.9 (10)	1.4 ± 1.2 (9)	1.54	7.11	2.62	9.01
fluorene	1.4 ± 0.91 (3)	9.8 ± 7.4 (3)	2.16	6.49	4.56	0.631
phenanthrene	2.0 ± 2.4 (2)	15 ± 14 (6)	1.56	4.79	1.47	1.08
anthracene	0.78 ± 1.0 (9)	0.88 ± 1.1 (6)	0.831	2.83	1.44	1.72
fluoranthene	8.3 ± 11 (10)	5.2 ± 3.3 (9)	0.267	1.87	1.65	0.472
pyrene	5.8 ± 8.2 (10)	6.0 ± 7.4 (9)	0.162	3.08	2.66	0.493
benz(a)anthracene	0.86 ± 0.44 (10)	1.2 ± 0.89 (8)	0.824	1.44	0.217	0.0921
chyrsene	2.7 ± 2.3 (10)	1.4 ± 0.49 (9)	0.474	1.24	1.08	0.162
benzo(b)fluoranthene	2.0 ± 0.60 (10)	2.5 ± 0.72 (9)	0.454	0.697	1.22	0.115
benzo(k)fluoranthene	1.2 ± 0.56 (4)	1.0 ± 0.35 (4)	0.324	0.658	0.944	0.220
perylene	0.70 ± 0.64 (10)	1.4 ± 1.2 (7)				
indeno(1,2,3-c,d)pyrene	1.1 ± 0.42 (10)	1.6 ± 1.1 (9)	0.828	3.19		
dibenz(a,h)anthracene	0.40 ± 0.060 (7)	1.0 ± 0.70 (5)				
benzo(g,h,i)perylene	0.87 ± 0.33 (9)	1.4 ± 1.1 (9)	0.358	0.272	0.340	
coronene	0.81 ± 0.42 (9)	1.2 ± 0.88 (8)	0.0828		0.537	

source profiles from Khalli et al. (1995).

Table 24.
Estimation of Annual Atmospheric Input of PAHs to Casco Bay.

Source	Annual Input to Casco Bay (kg PAH)
Wet deposition	25
Dry deposition	39
Total Atmospheric Input	64

Figure 1.
Aluminum - IMPROVE Data Freeport, Me

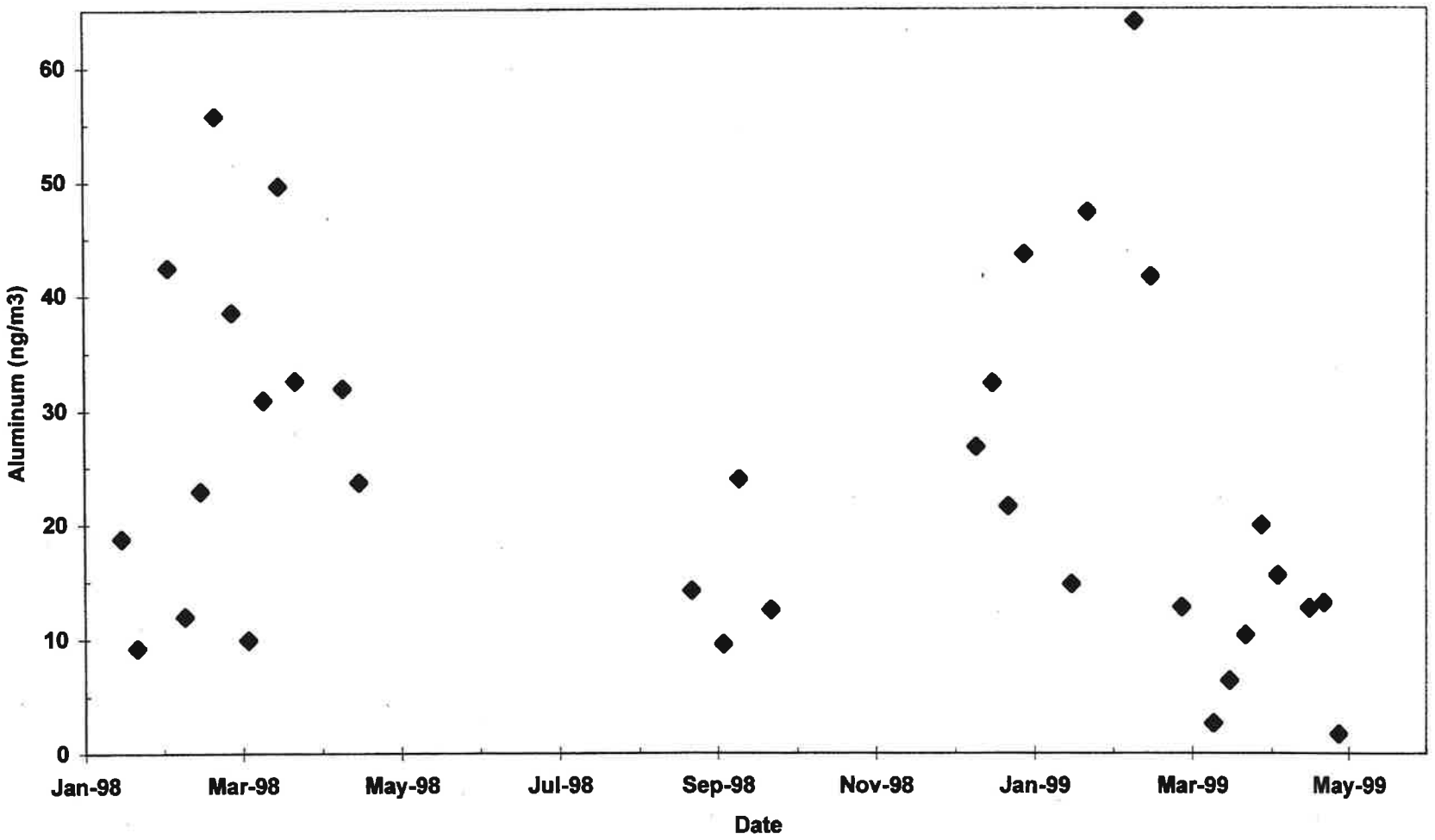


Figure 2.
Calcium - IMPROVE Data Freeport, Me

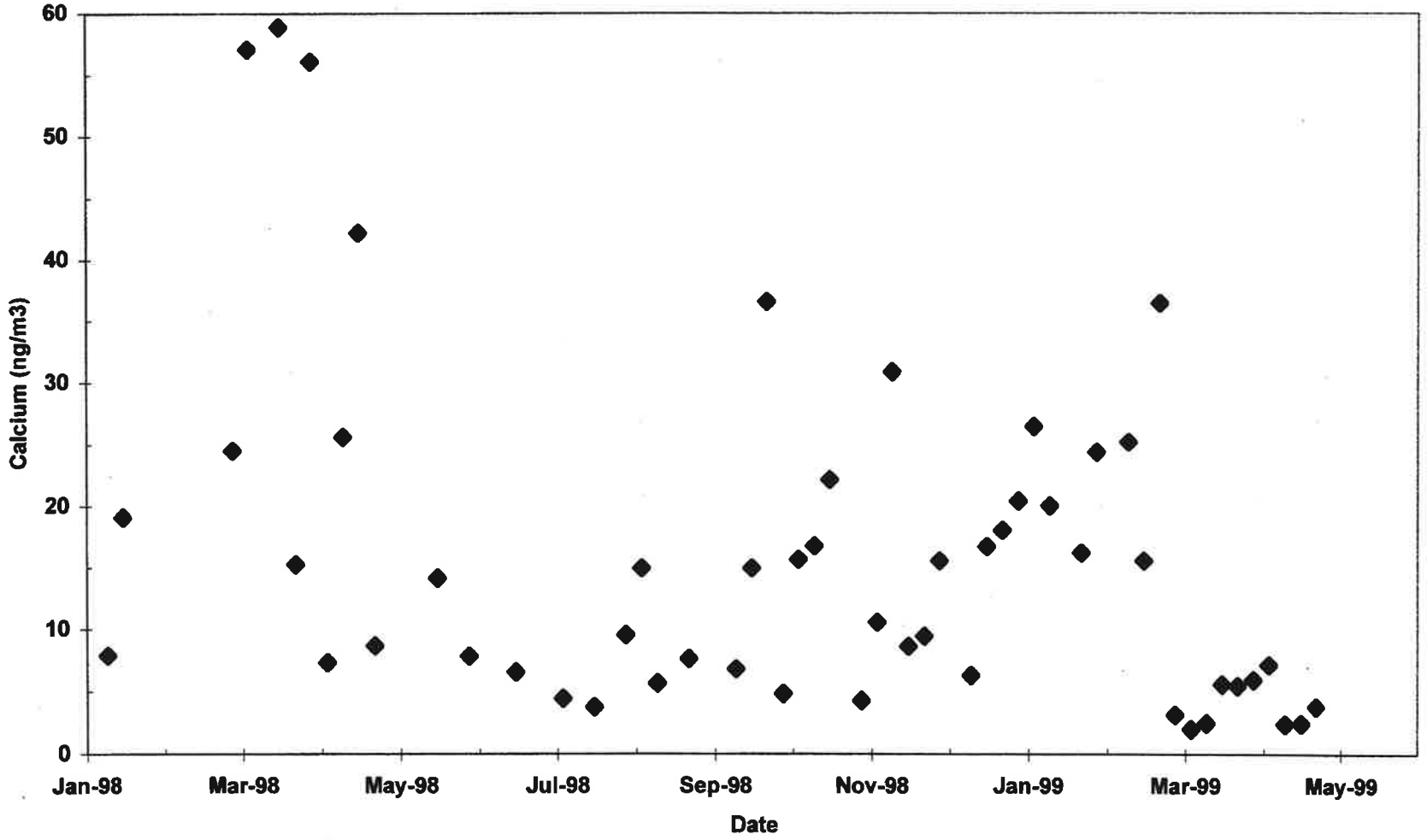


Figure 3.
Iron - IMPROVE Data Freeport, Me

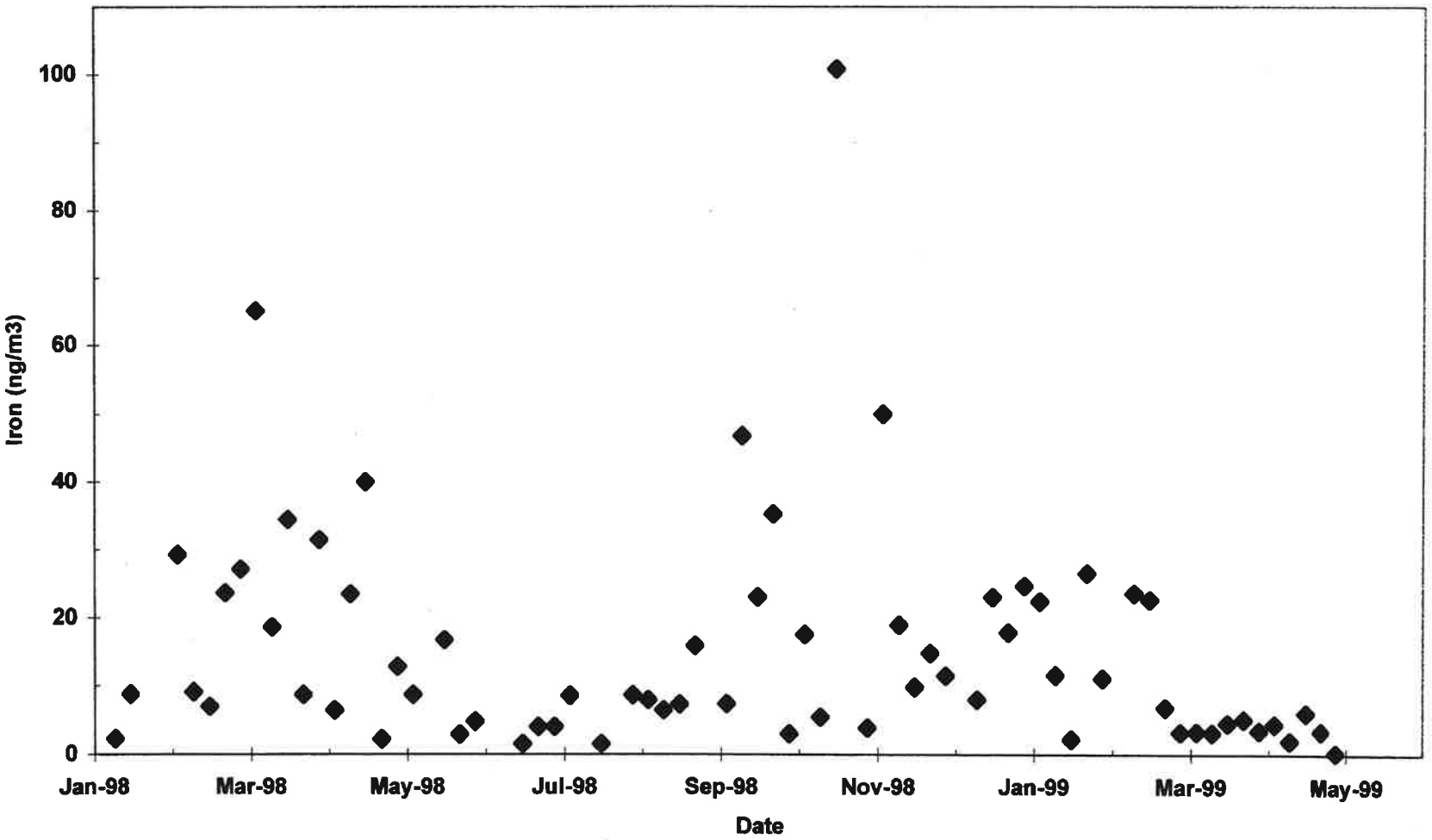


Figure 4.
Silicon - IMPROVE Data Freeport, Me

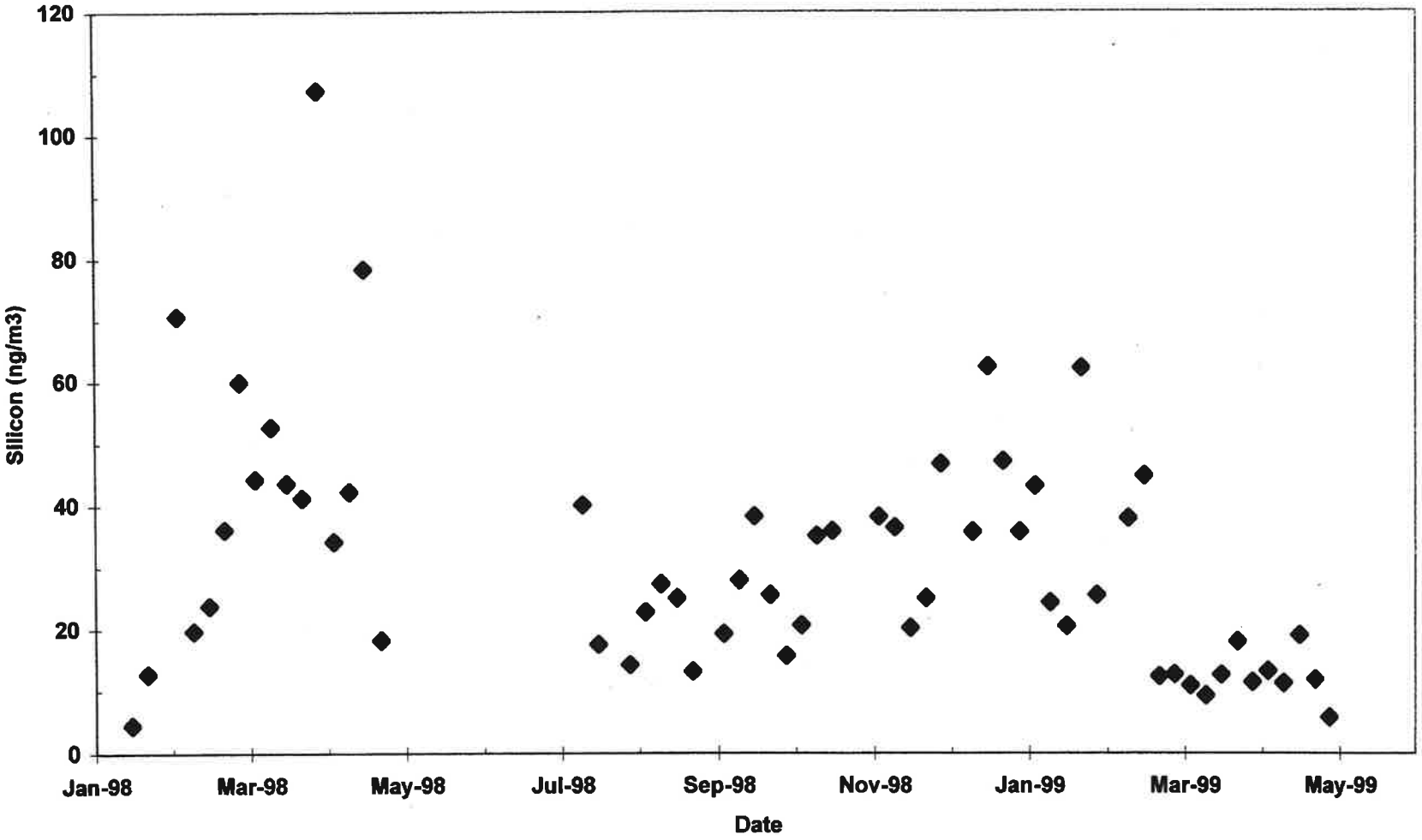


Figure 5.
Titanium - IMPROVE Data Freeport, Me

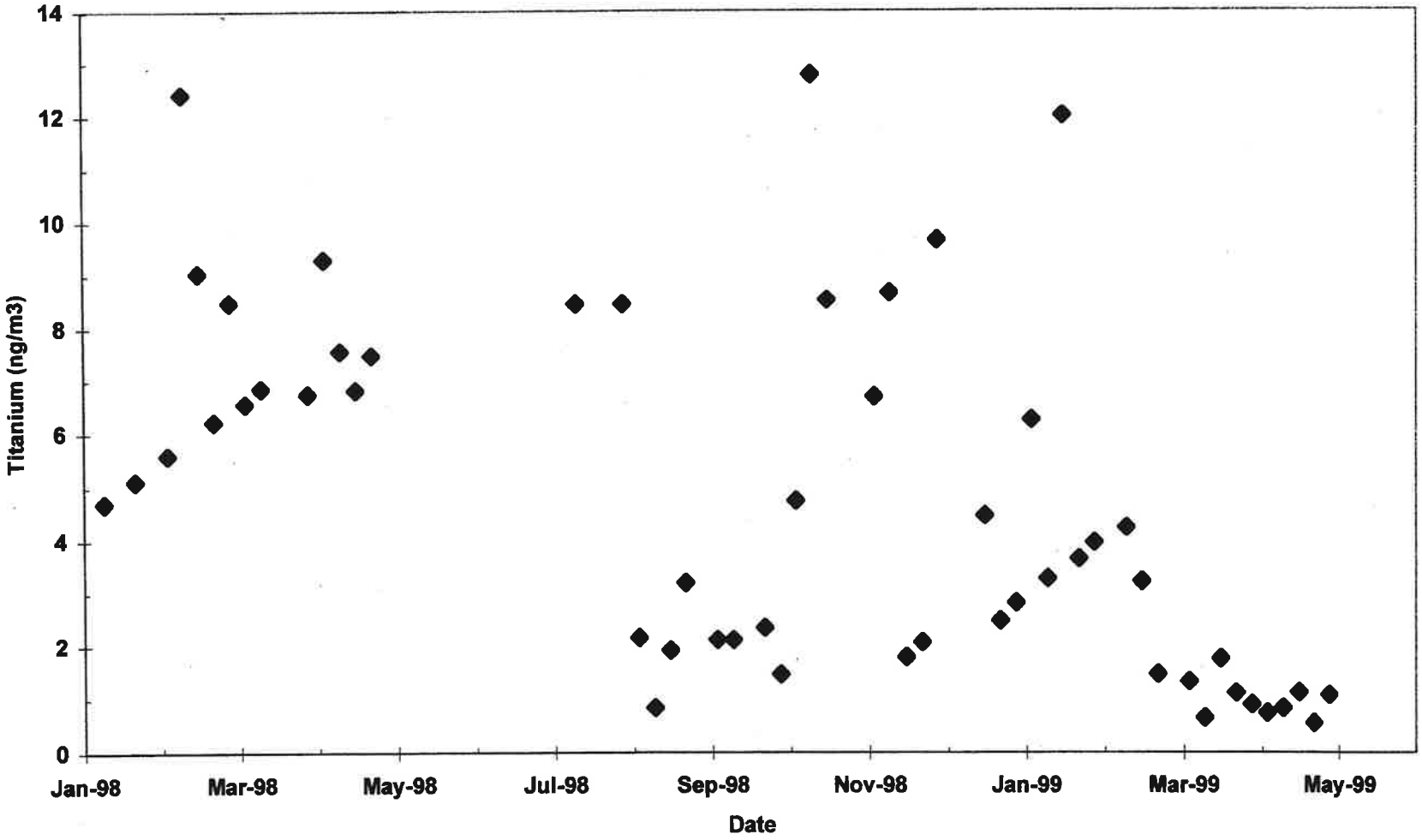


Figure 6.
Sodium - IMPROVE Data Freeport, Me

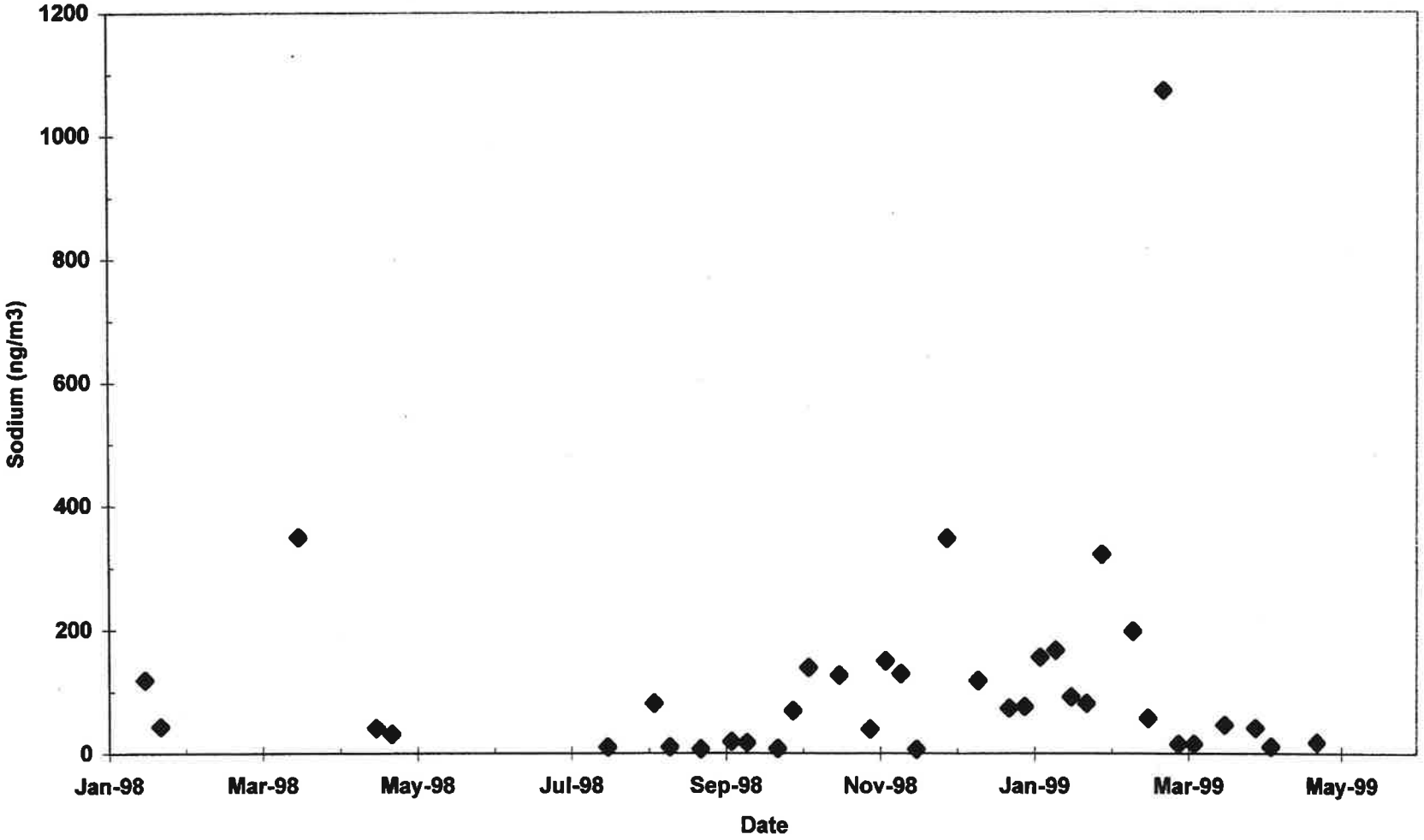


Figure 7.
Chloride ion - IMPROVE Data Freeport, Me

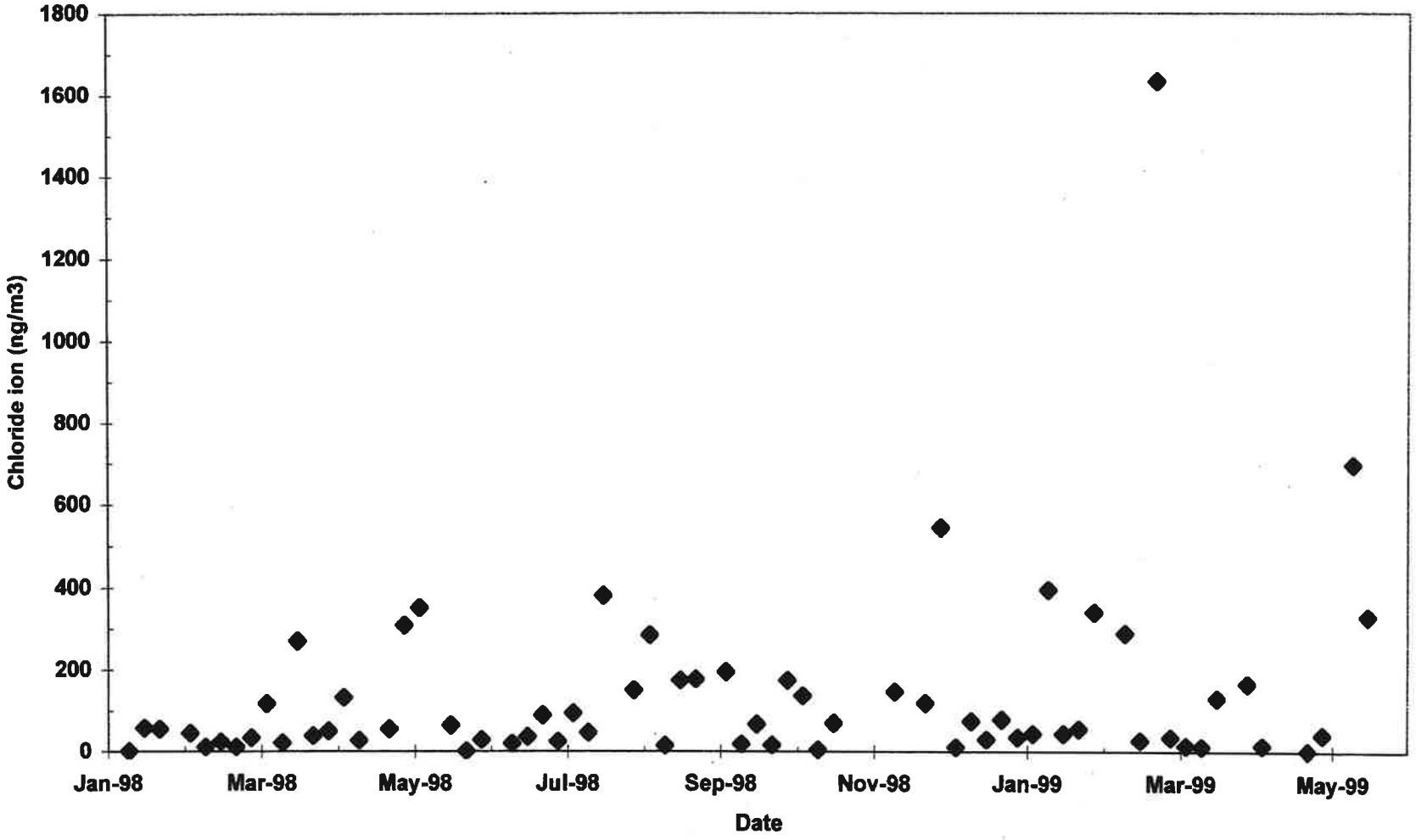


Figure 8.
Arsenic - IMPROVE Data Freeport, Me

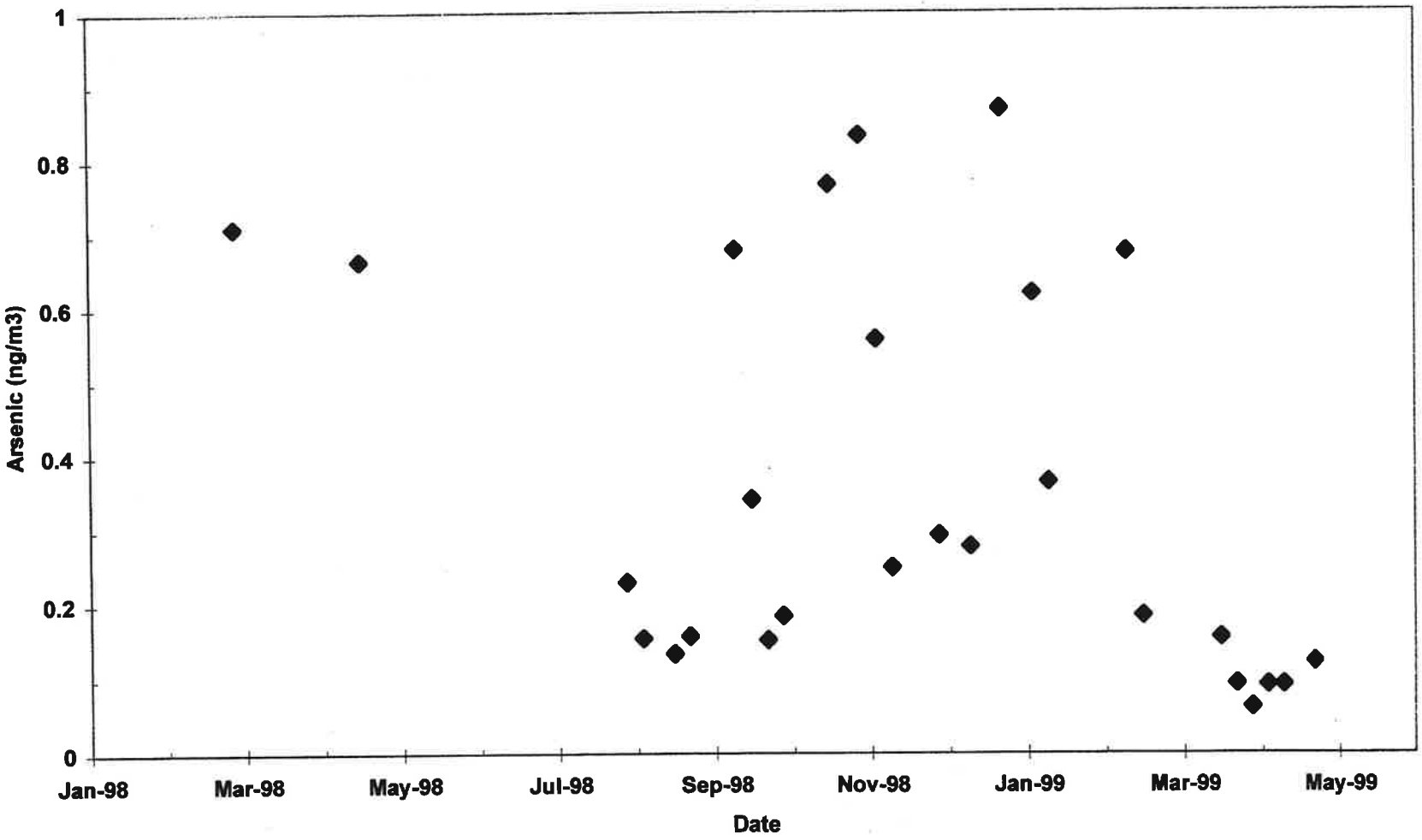


Figure 9.
Chromium - IMPROVE Data Freeport, Me

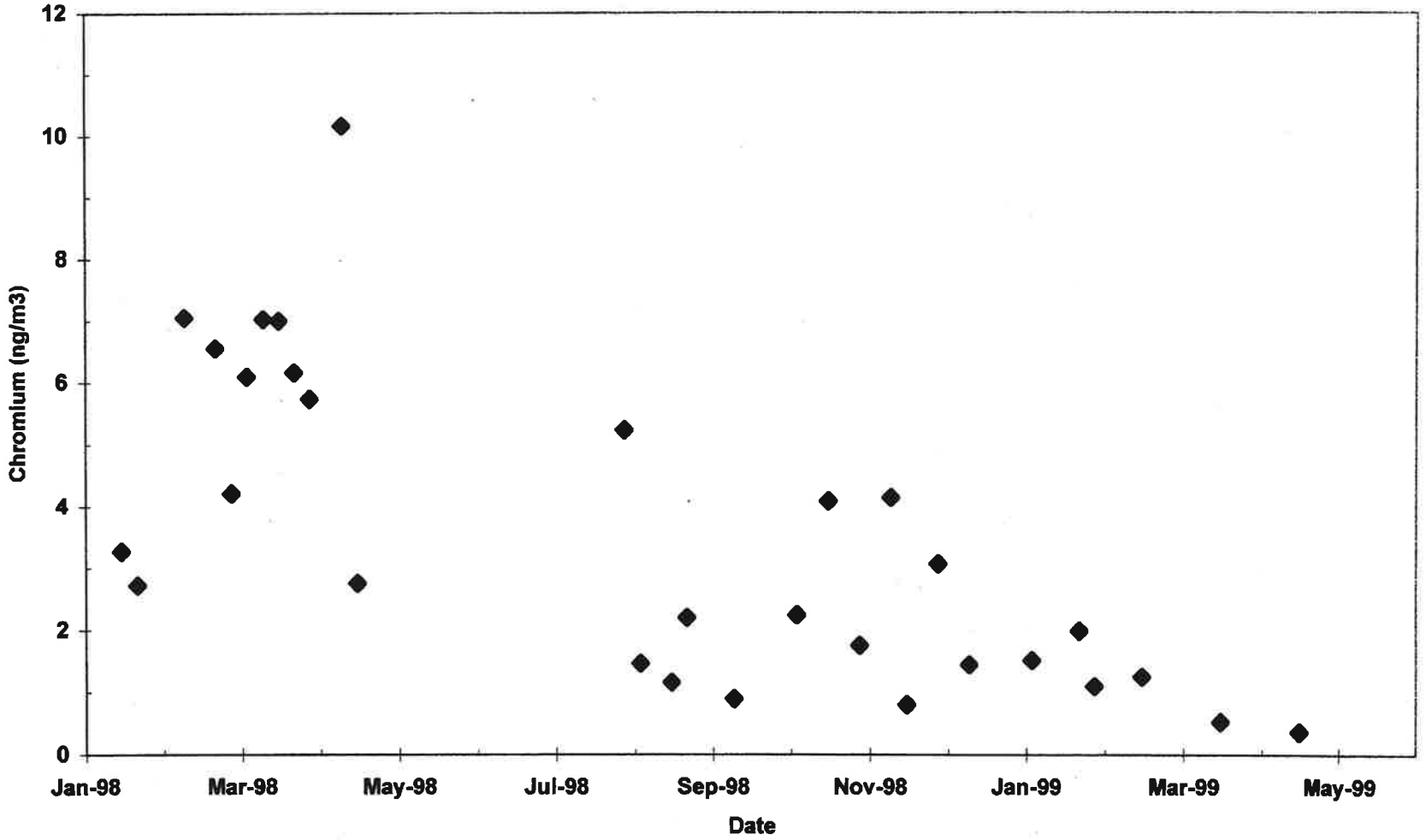


Figure 10.
Copper- IMPROVE Data Freeport, Me

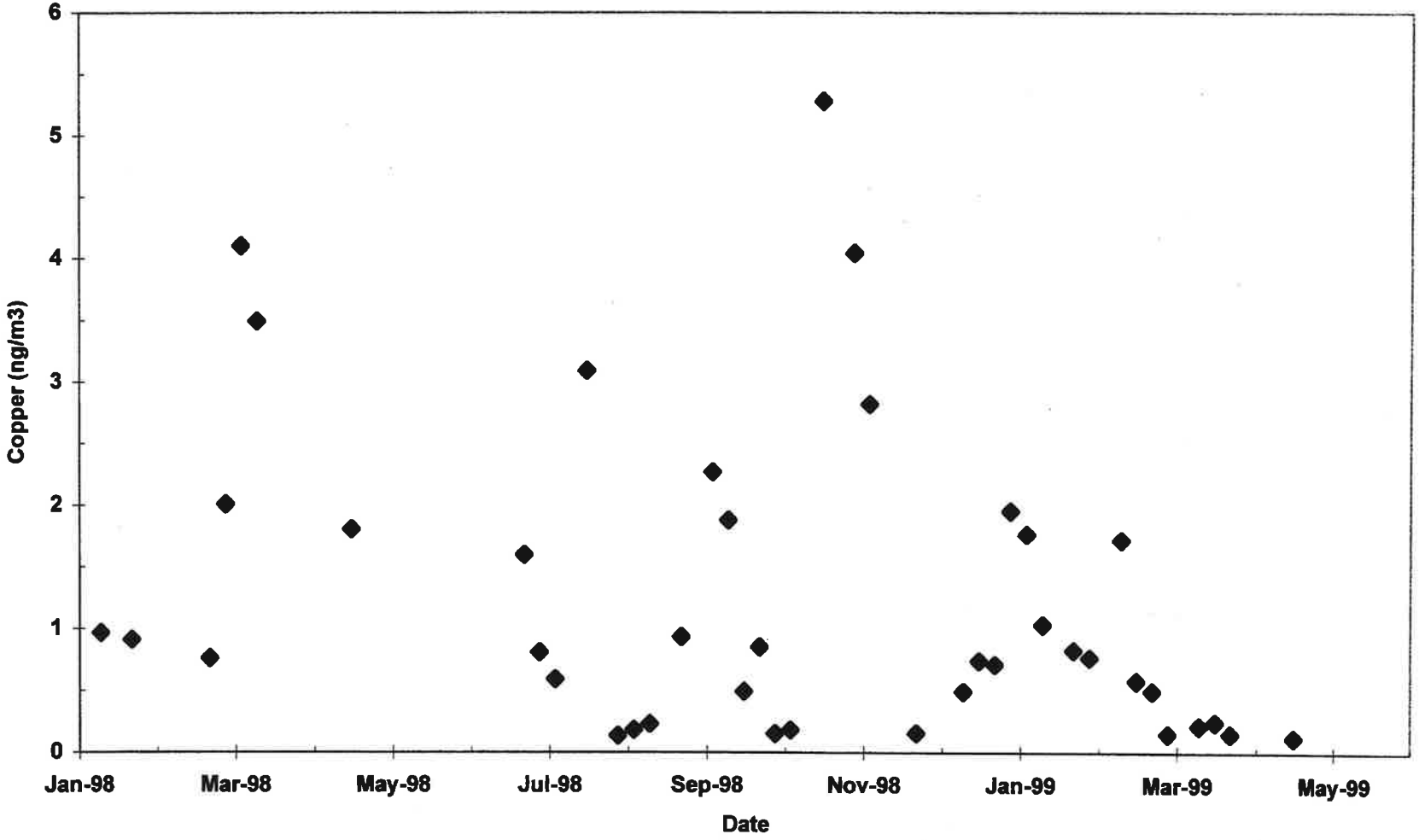


Figure 11.
Lead - IMPROVE Data Freeport, Me

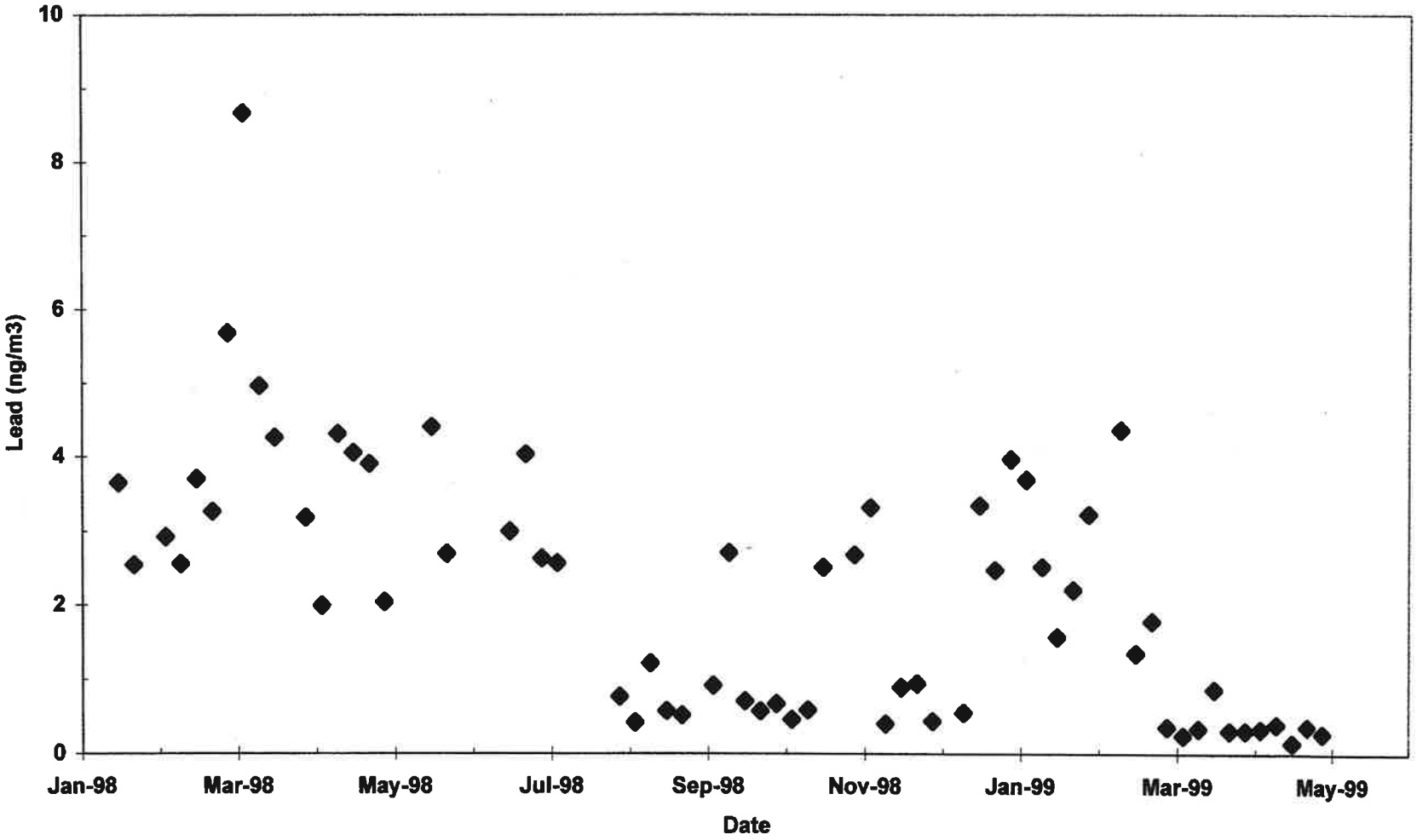


Figure 12.
Nickel - IMPROVE Data Freeport, Me

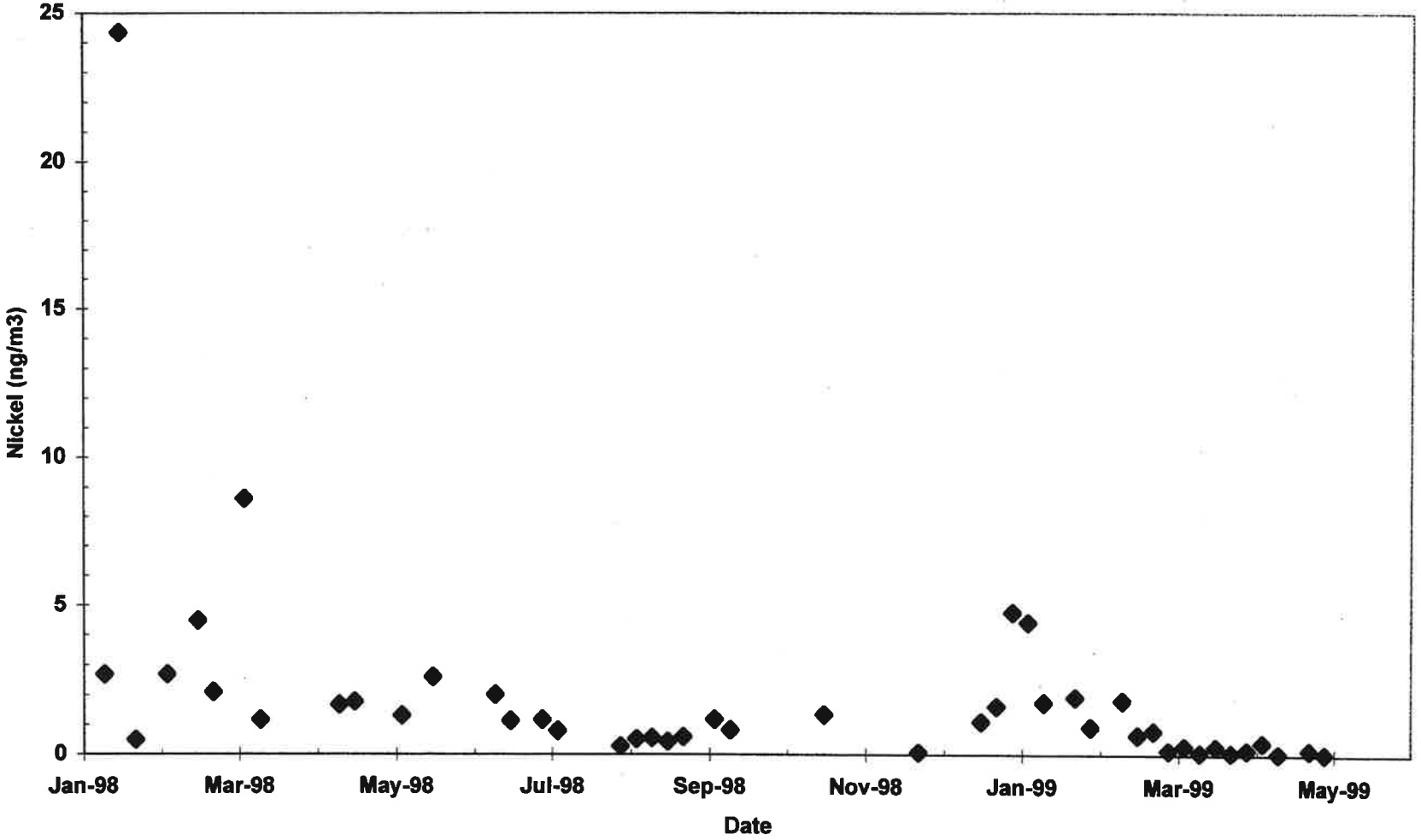


Figure 13.
Selenium - IMPROVE Data Freeport, Me

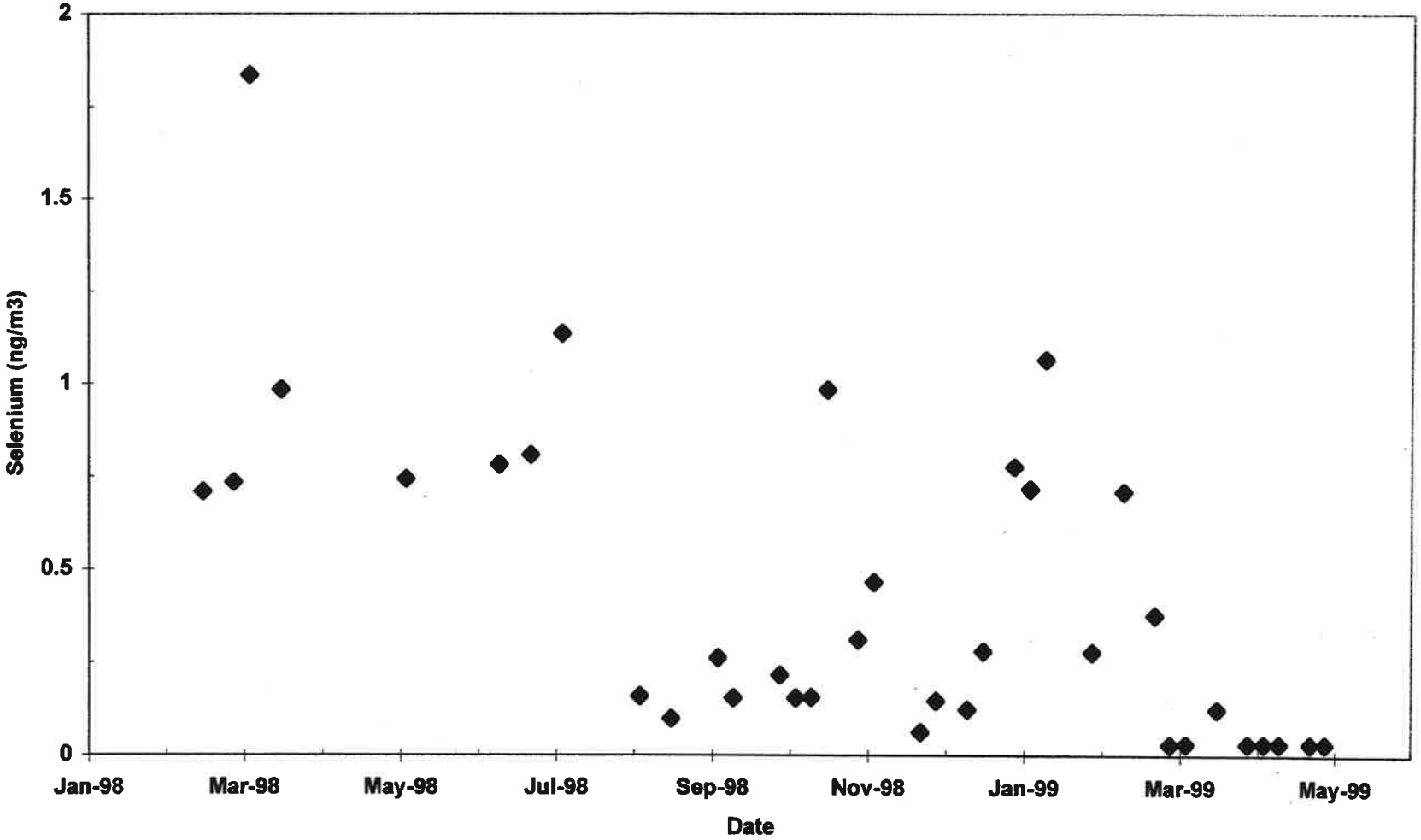


Figure 14.
Vanadium - IMPROVE Data Freeport, Me

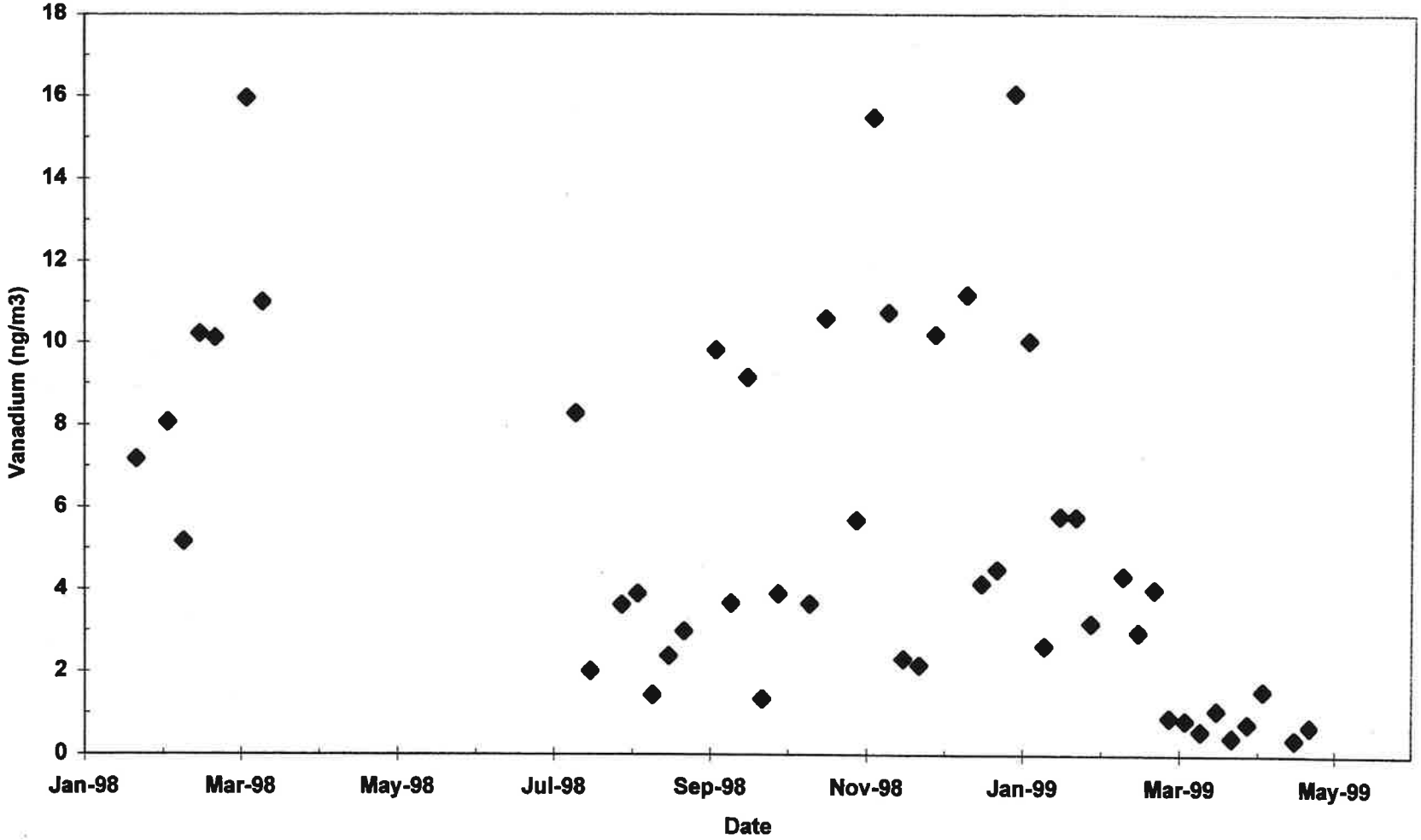


Figure 15.
Zinc - IMPROVE Data, Freeport, ME

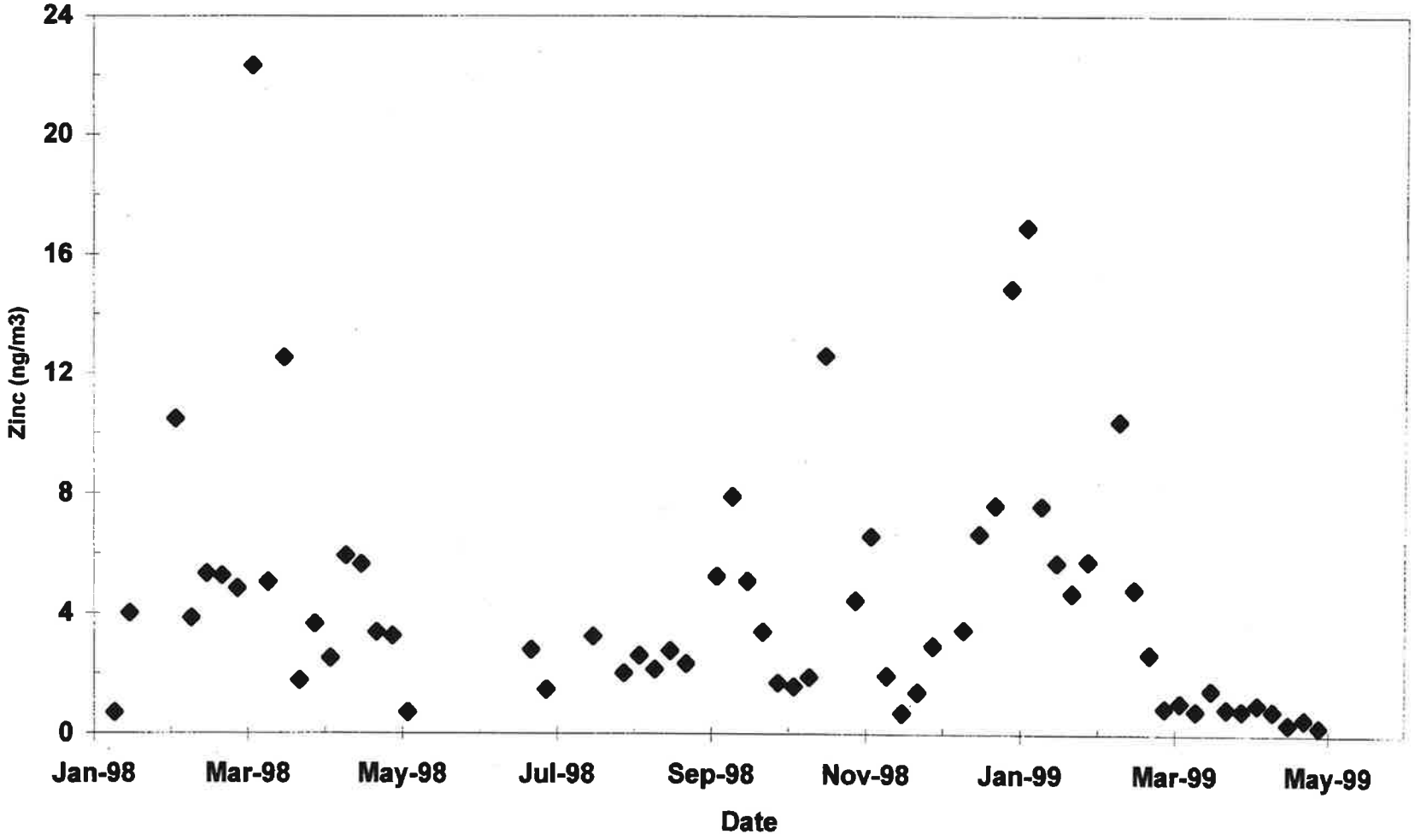


Figure 16.
Bromine - IMPROVE Data Freeport, Me

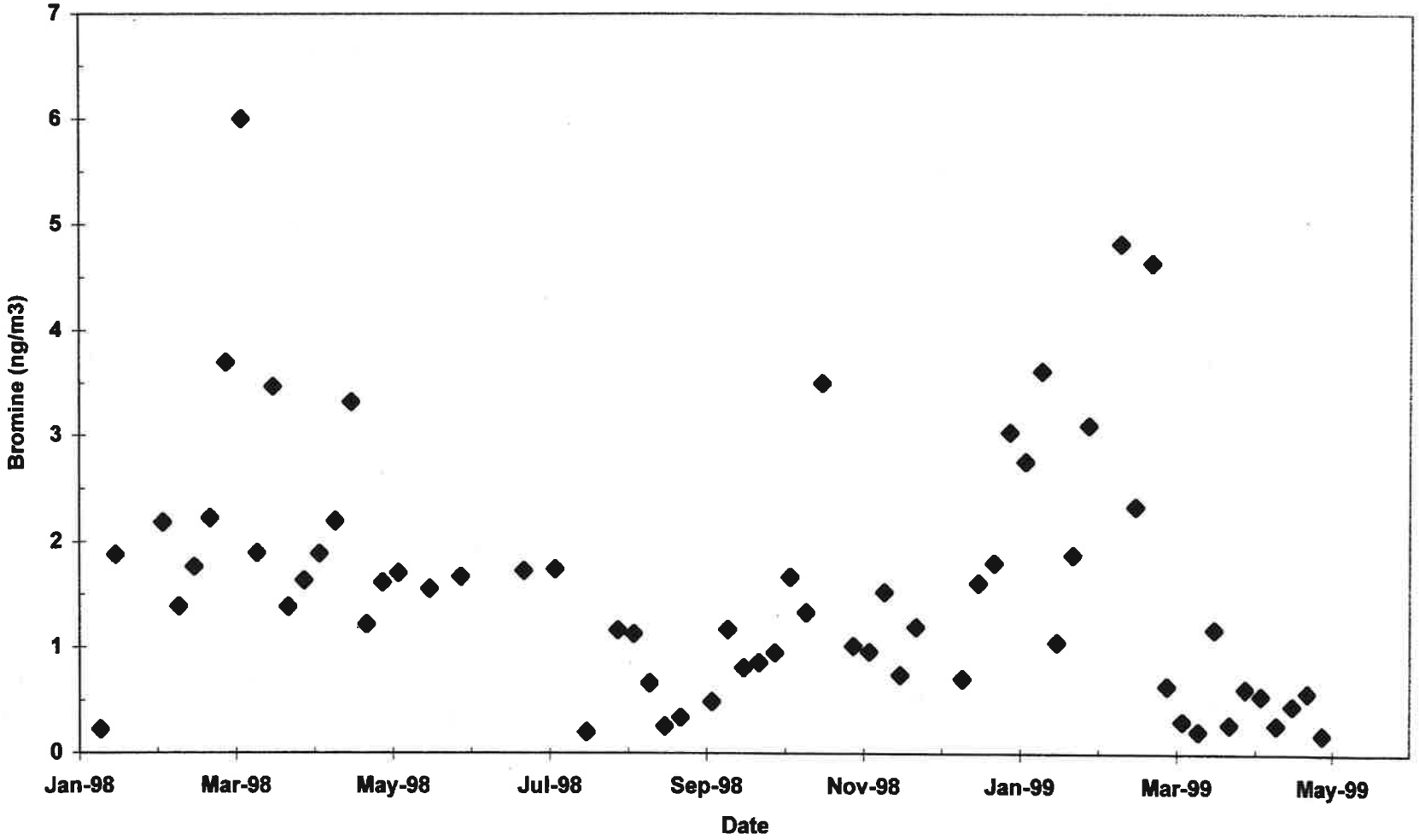


Figure 17.
Bromine/Lead - IMPROVE Data Freeport, Me

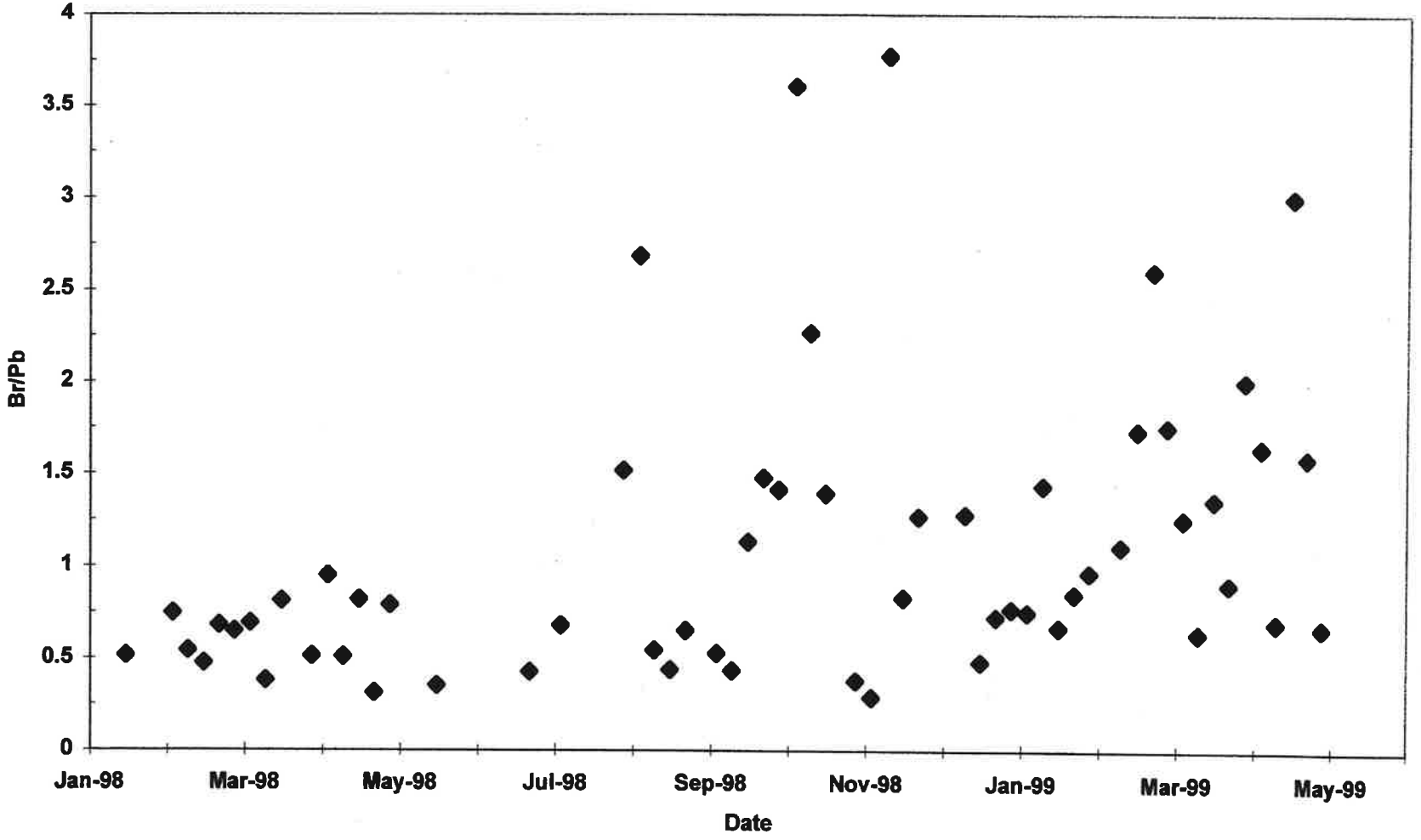


Figure 18.
Sulfate ion (SO4) - IMPROVE Data Freeport, Me

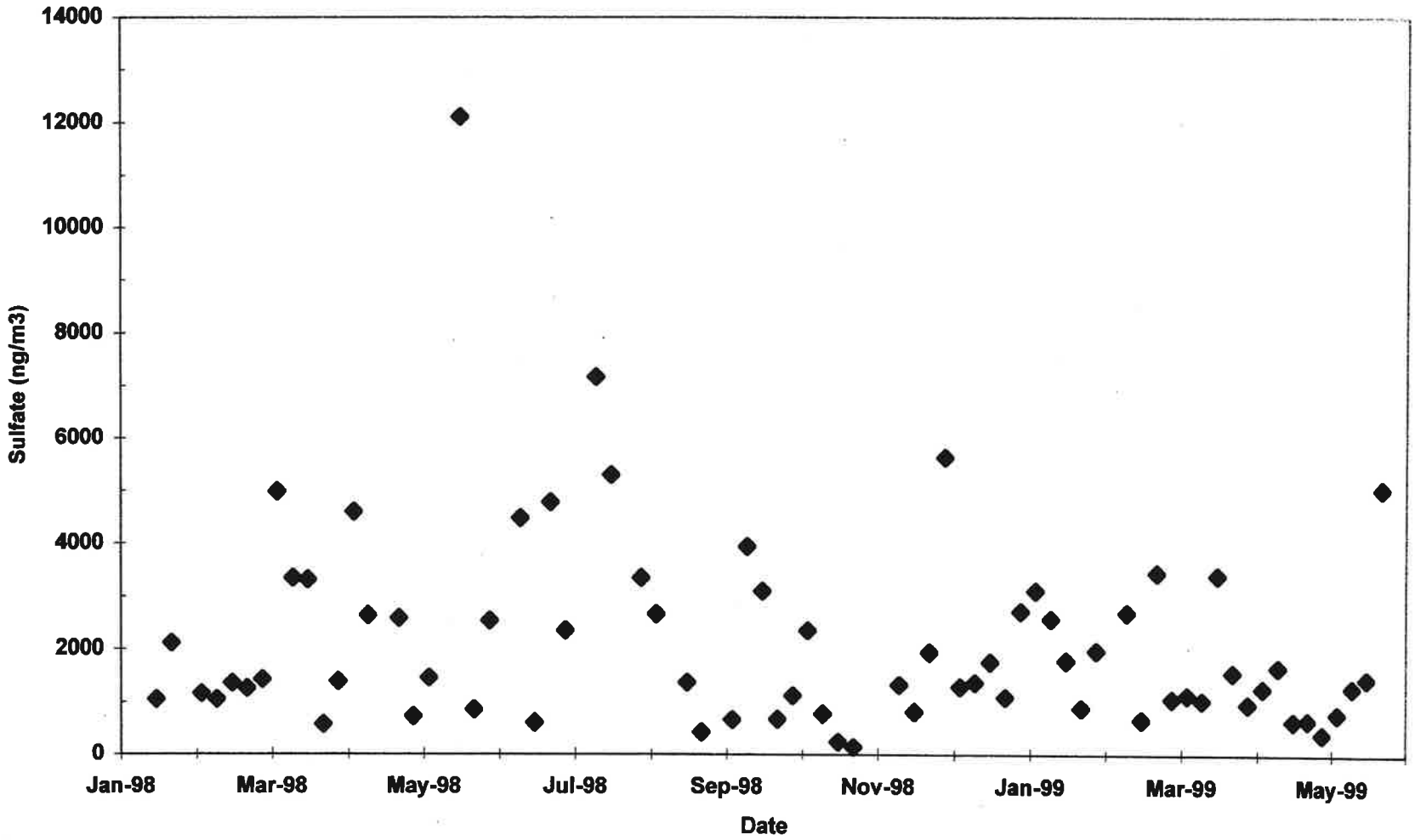


Figure 19.
Nitrite ion (NO2) - IMPROVE Data Freeport, Me

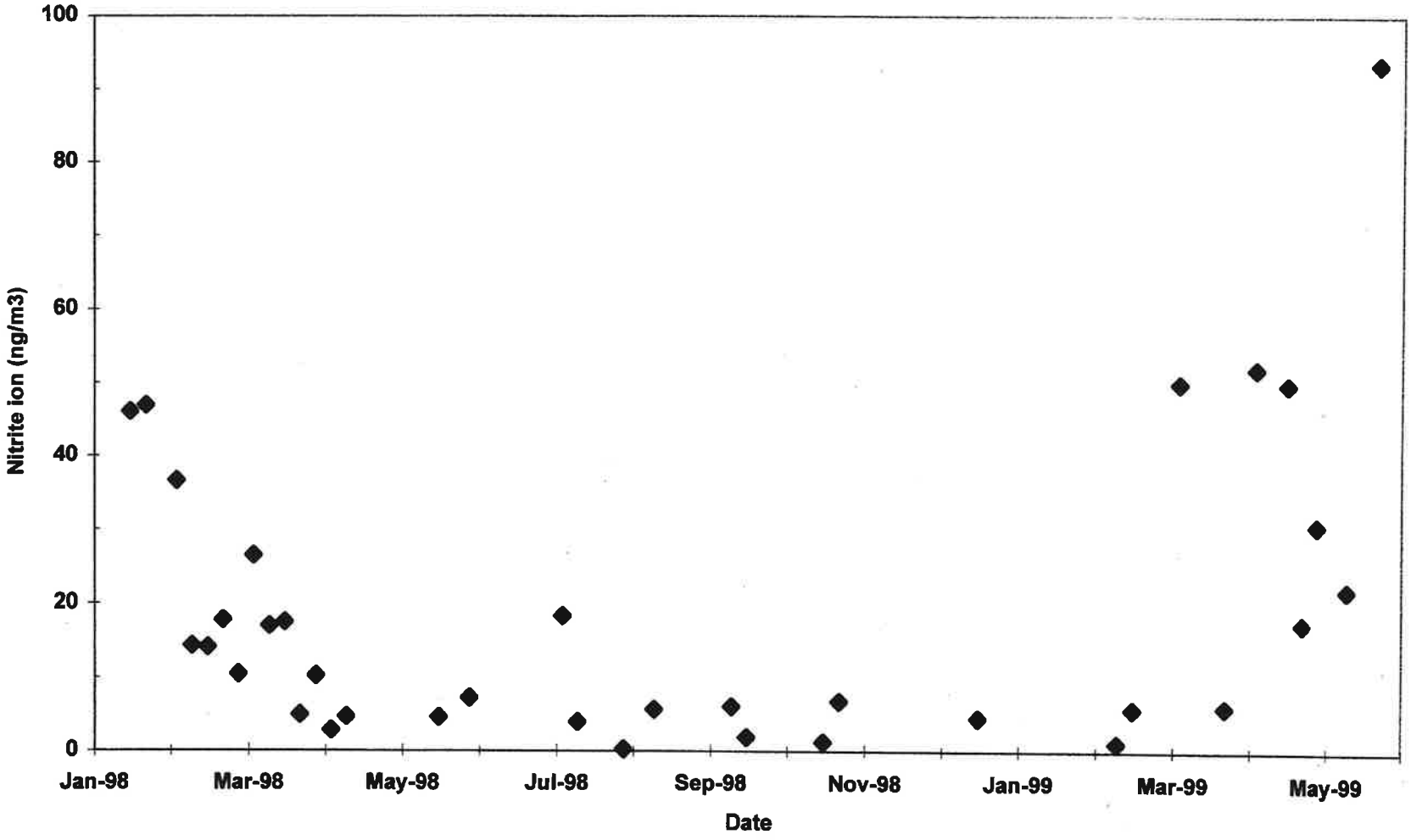


Figure 20.
Nitrate ion (NO3) - IMPROVE Data Freeport, Me

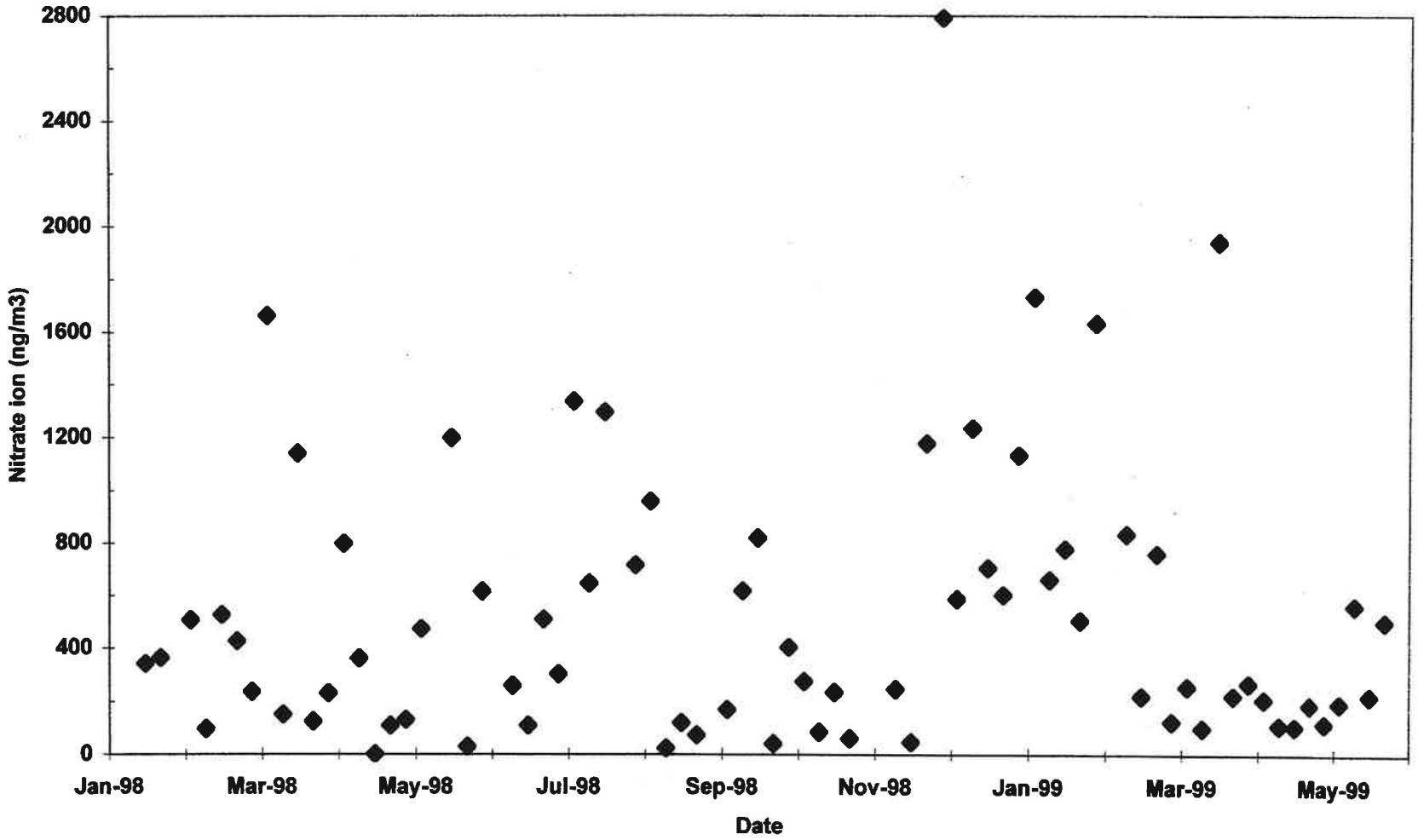


Figure 21.
Hydrogen Ion - IMPROVE Data Freeport, Me

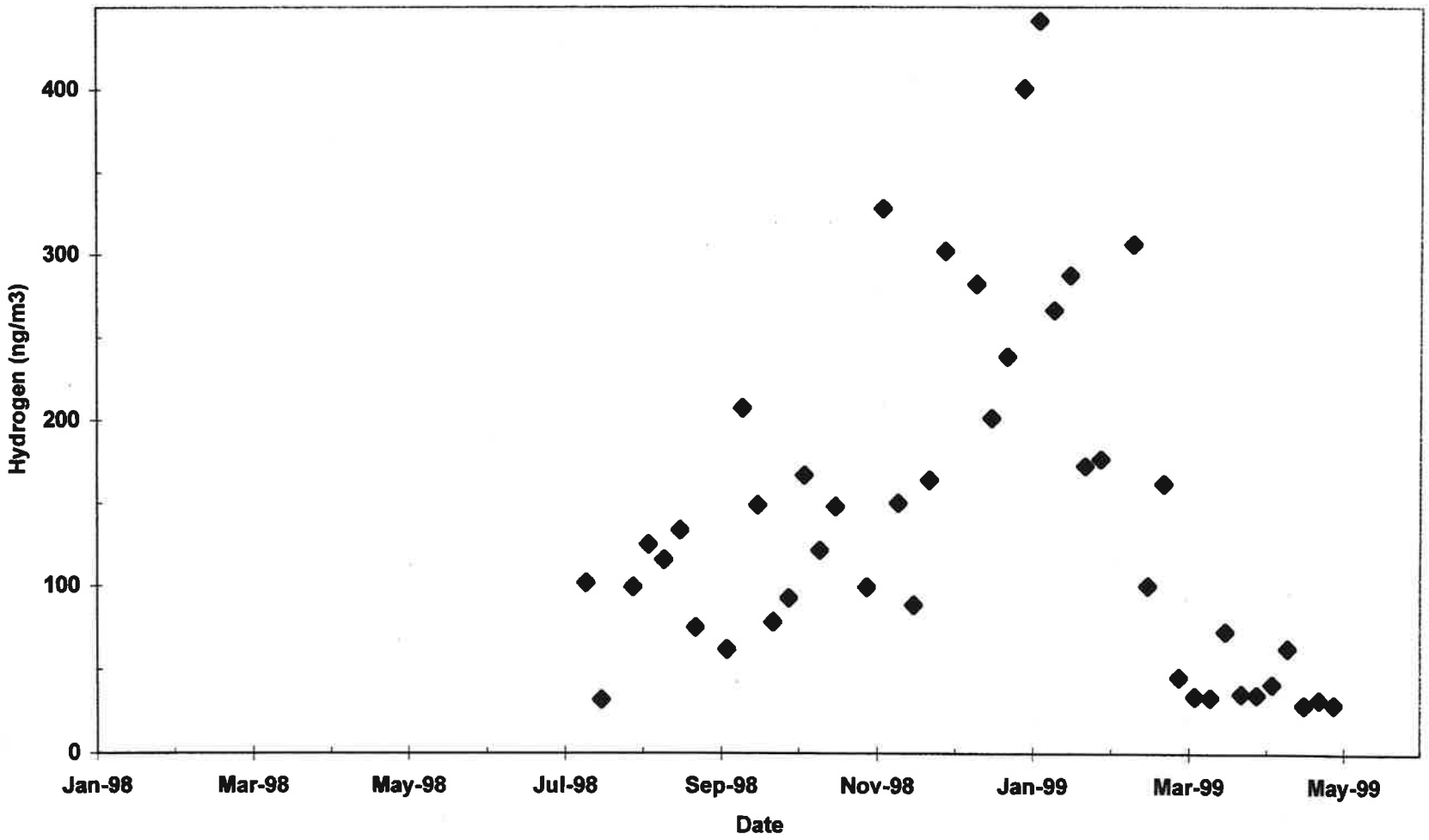


Figure 22.
Potassium - IMPROVE Data Freeport, Me

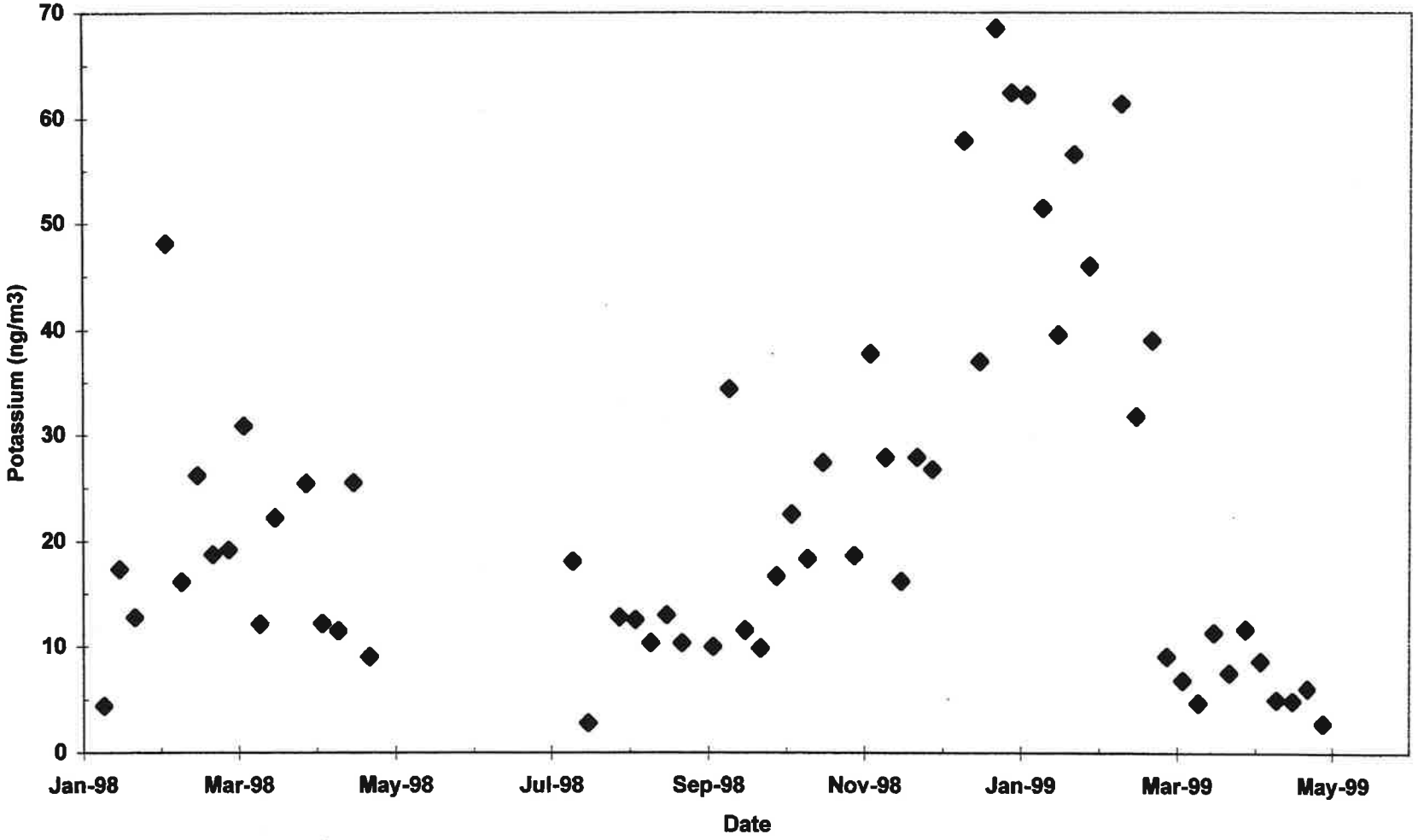


Figure 23.
Magnesium - IMPROVE Data Freeport, Me

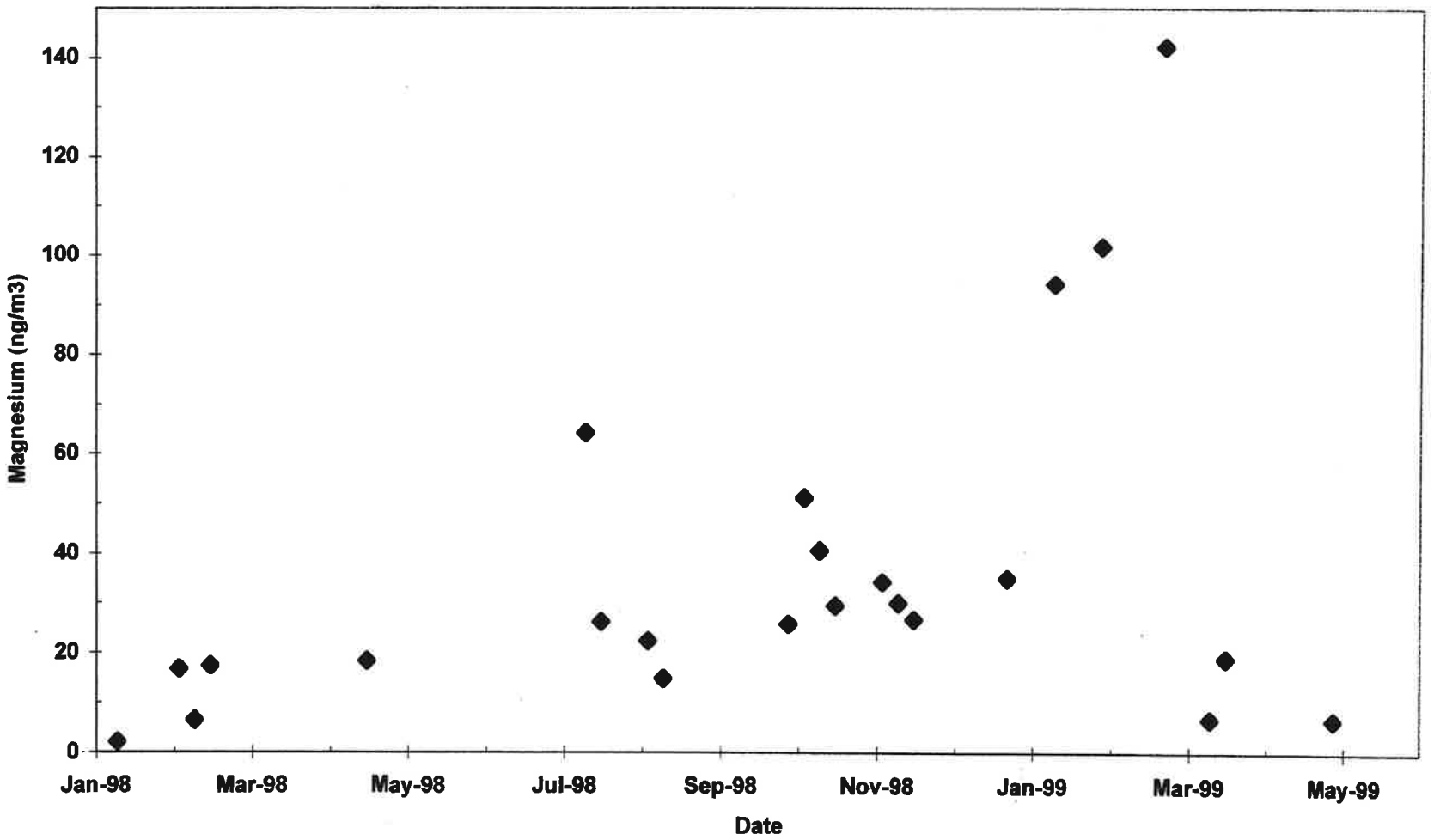


Figure 24.
Manganese - IMPROVE Data Freeport, Me

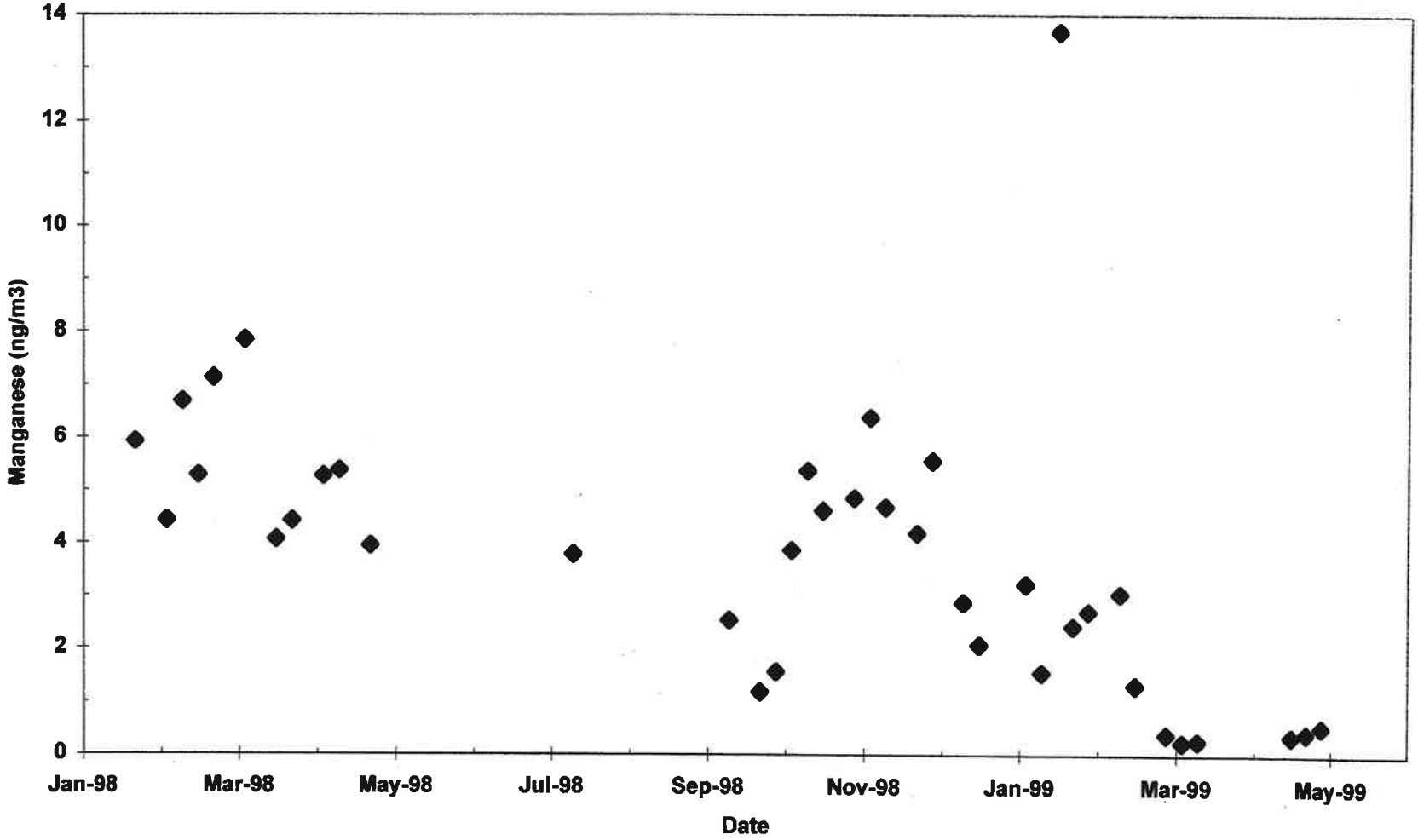


Figure 25.
Rubidium - IMPROVE Data Freeport, Me

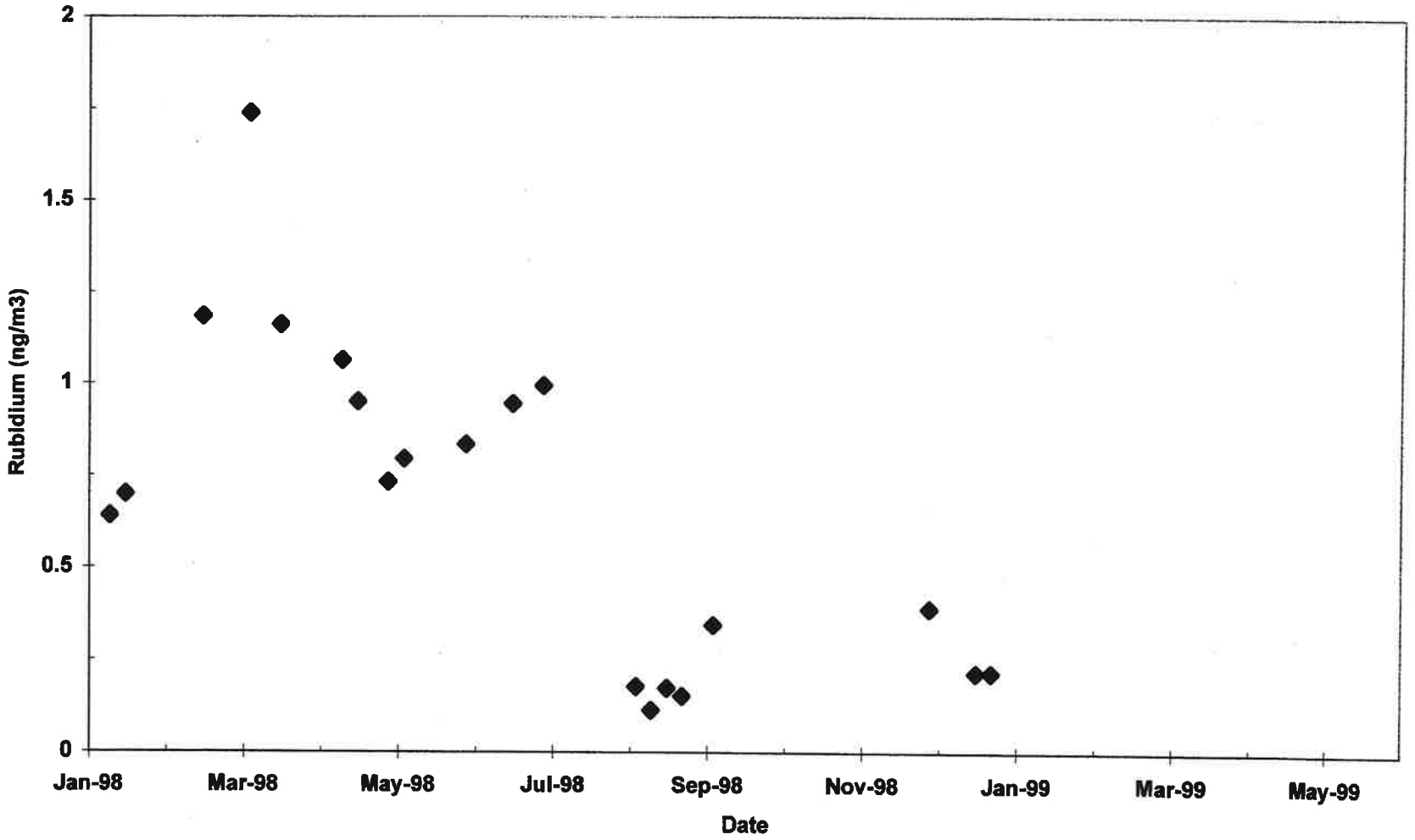


Figure 26.
Strontium - IMPROVE Data Freeport, Me

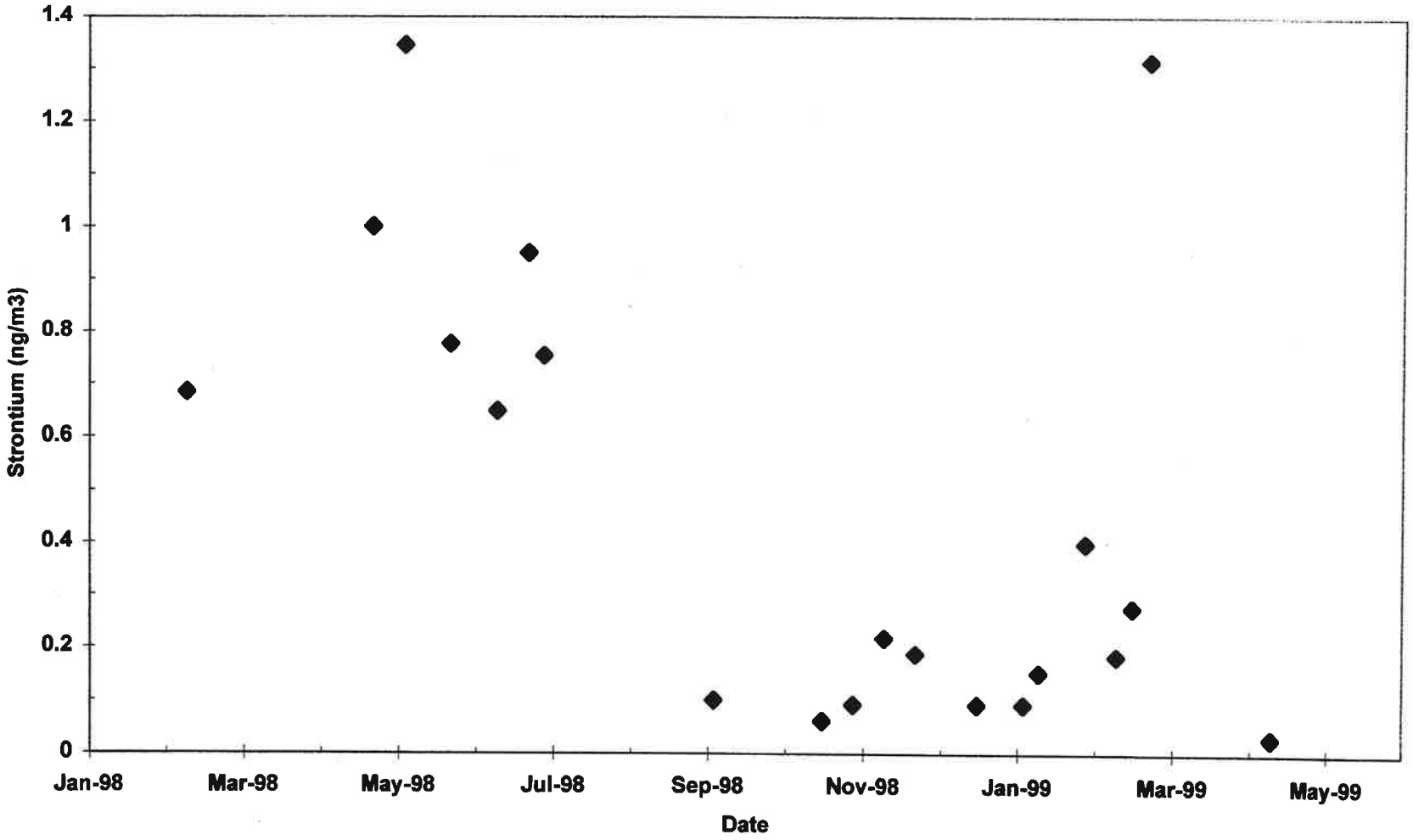


Figure 27.
Yttrium - IMPROVE Data Freeport, Me

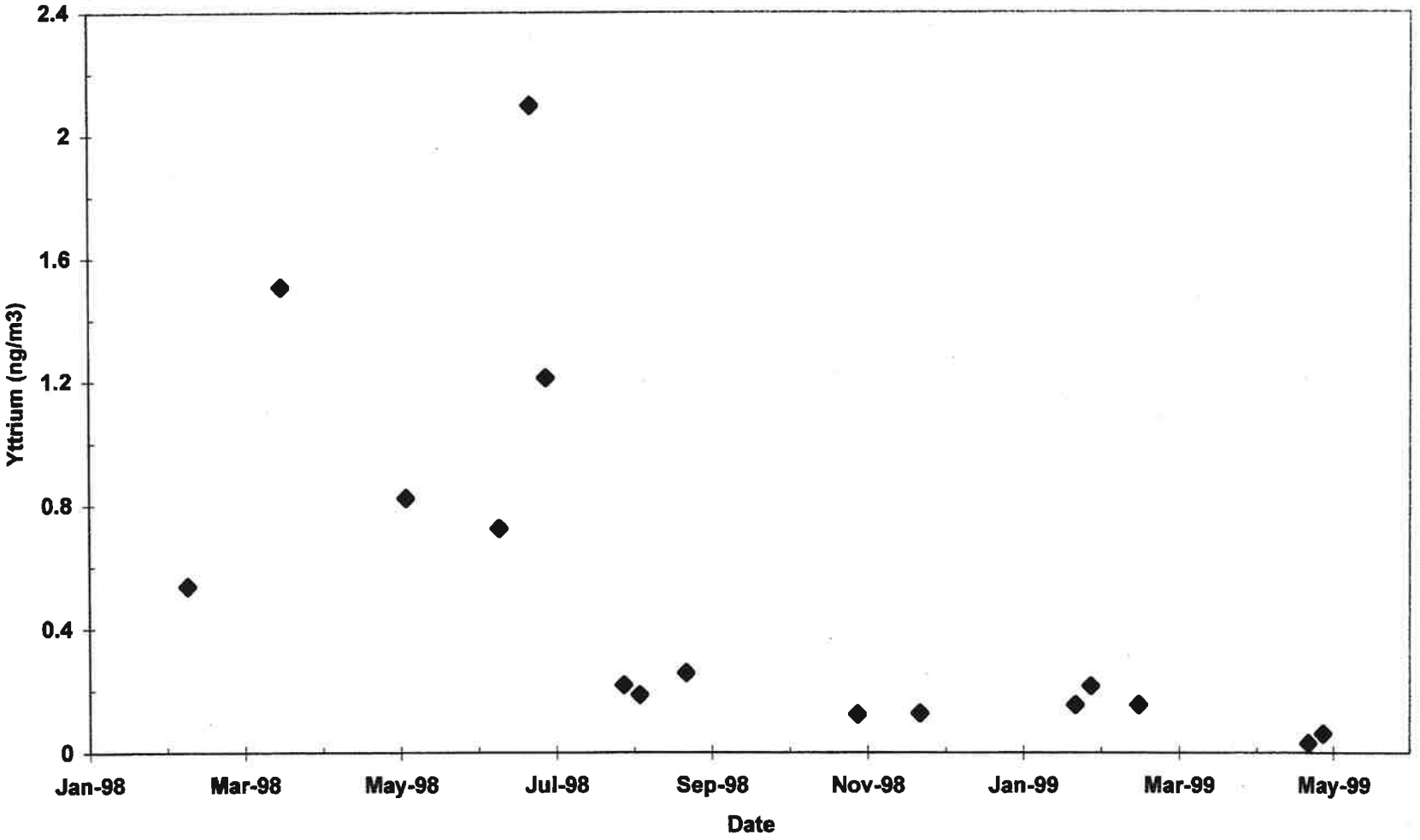


Figure 28.
Zirconium - IMPROVE Data, Freeport, ME

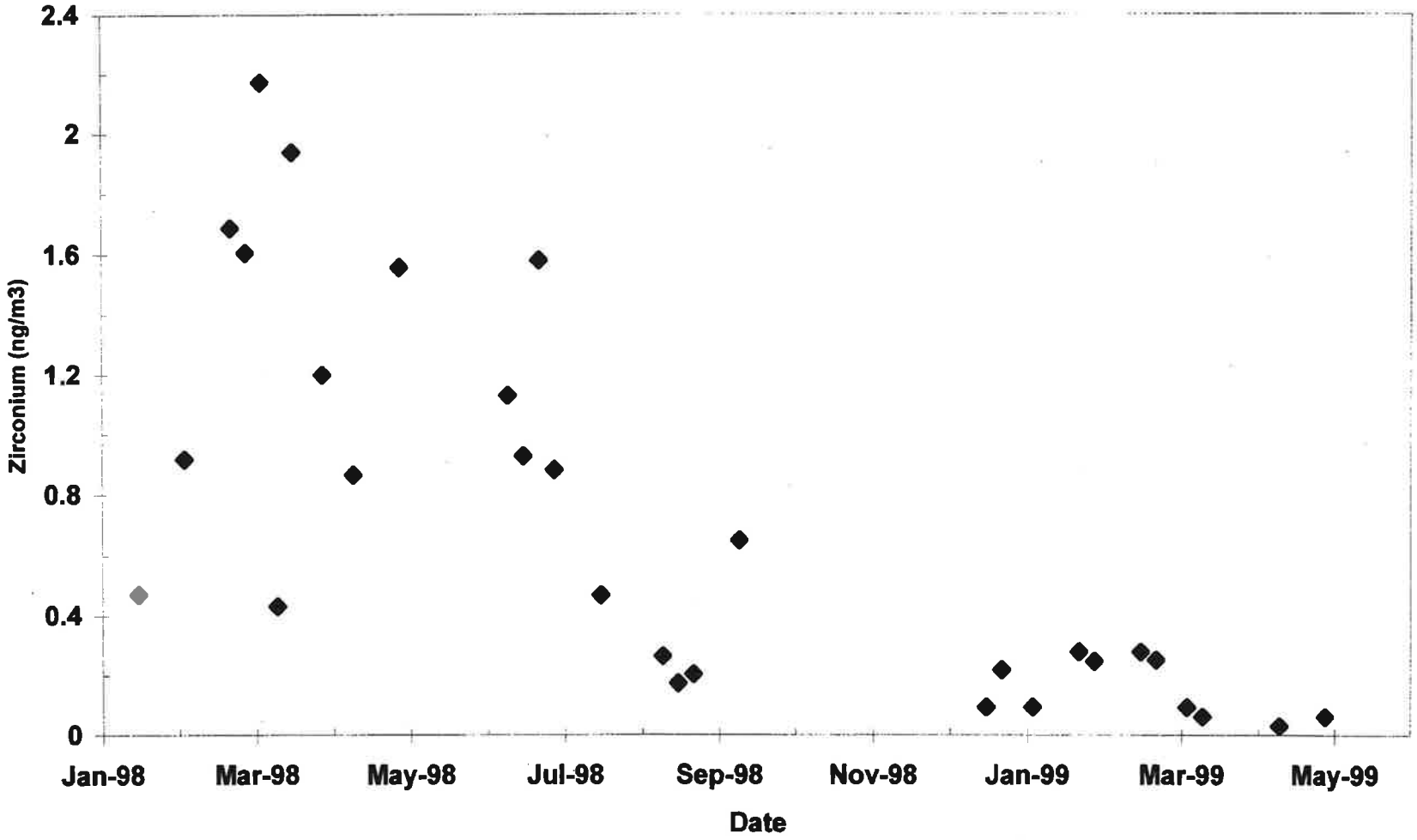


Figure 29.
Elemental Carbon - IMPROVE Data Freeport, Me

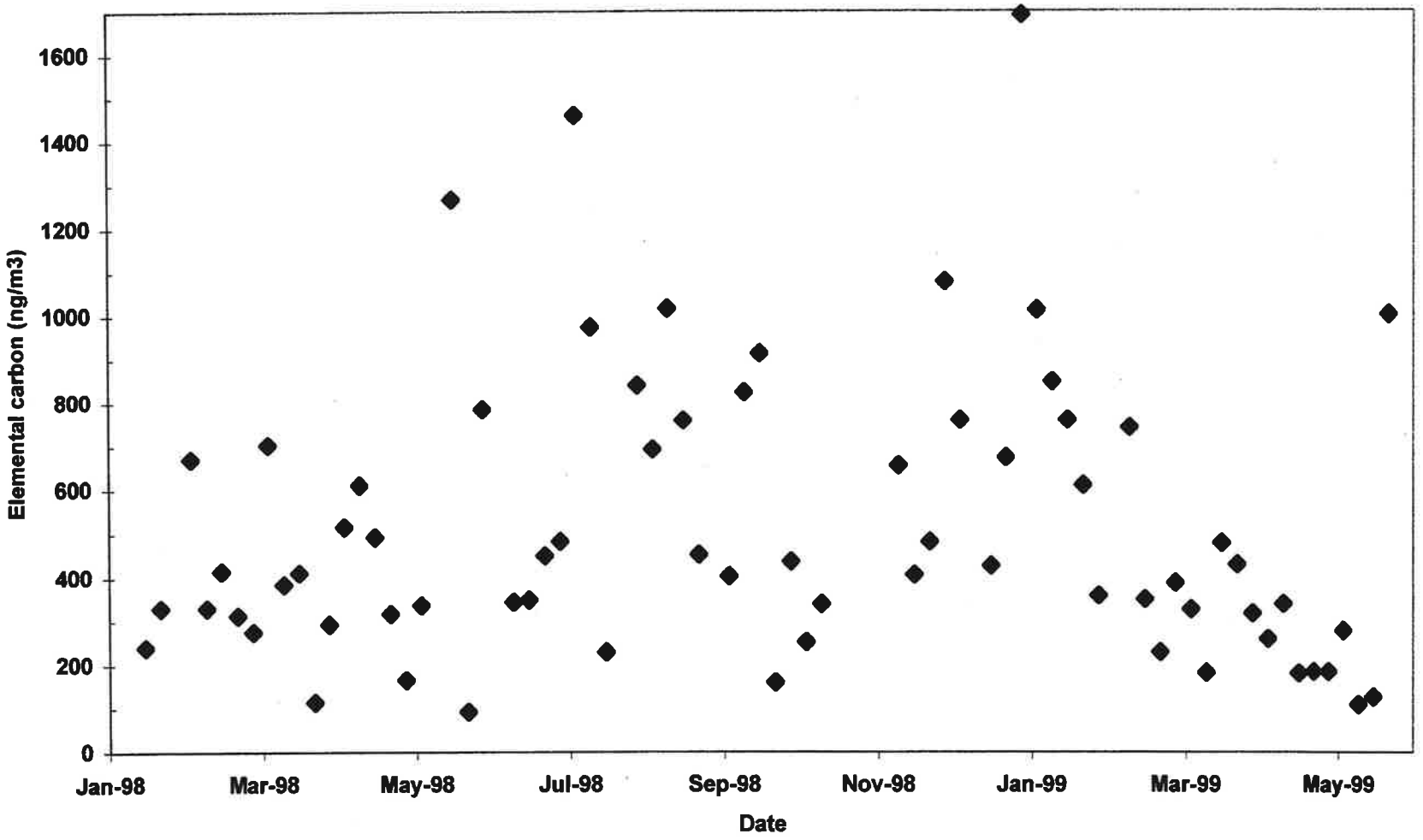


Figure 30.
Organic Carbon - IMPROVE Data Freeport, Me

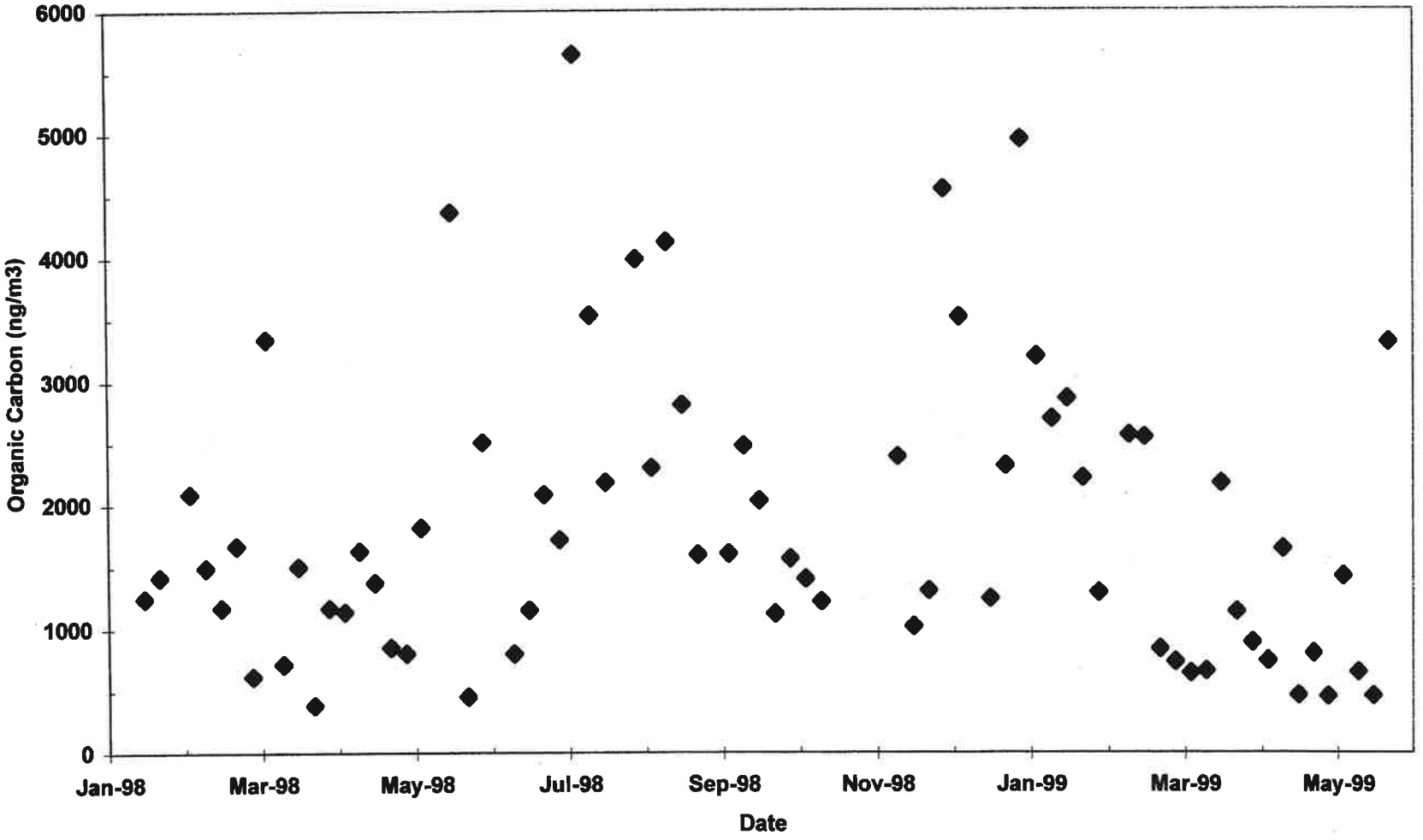


Figure 31.
Organic/Elemental Carbon - IMPROVE Data Freeport, Me

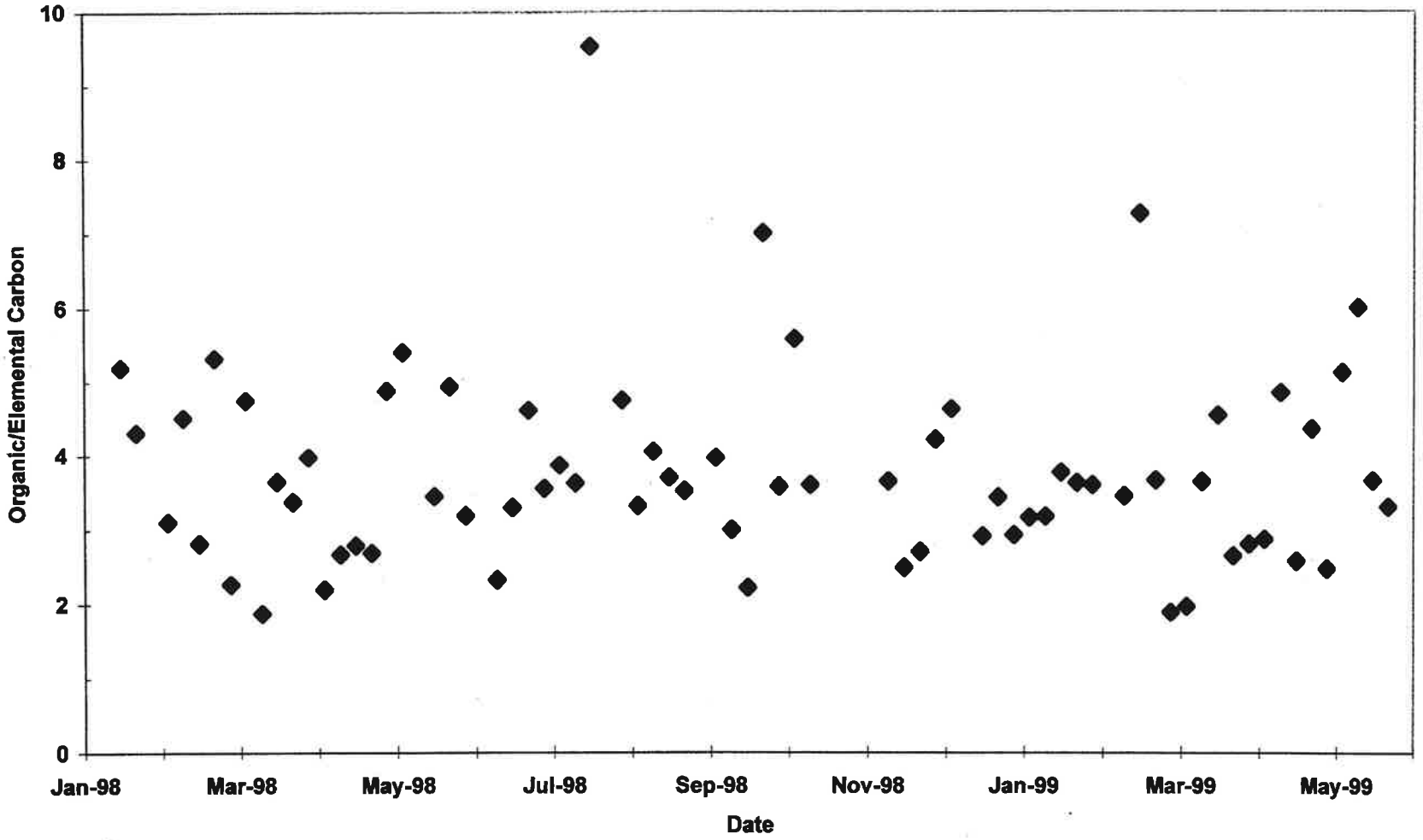


Figure 32.
Non Soil Potassium (KNON) - IMPROVE Data, Freeport, ME

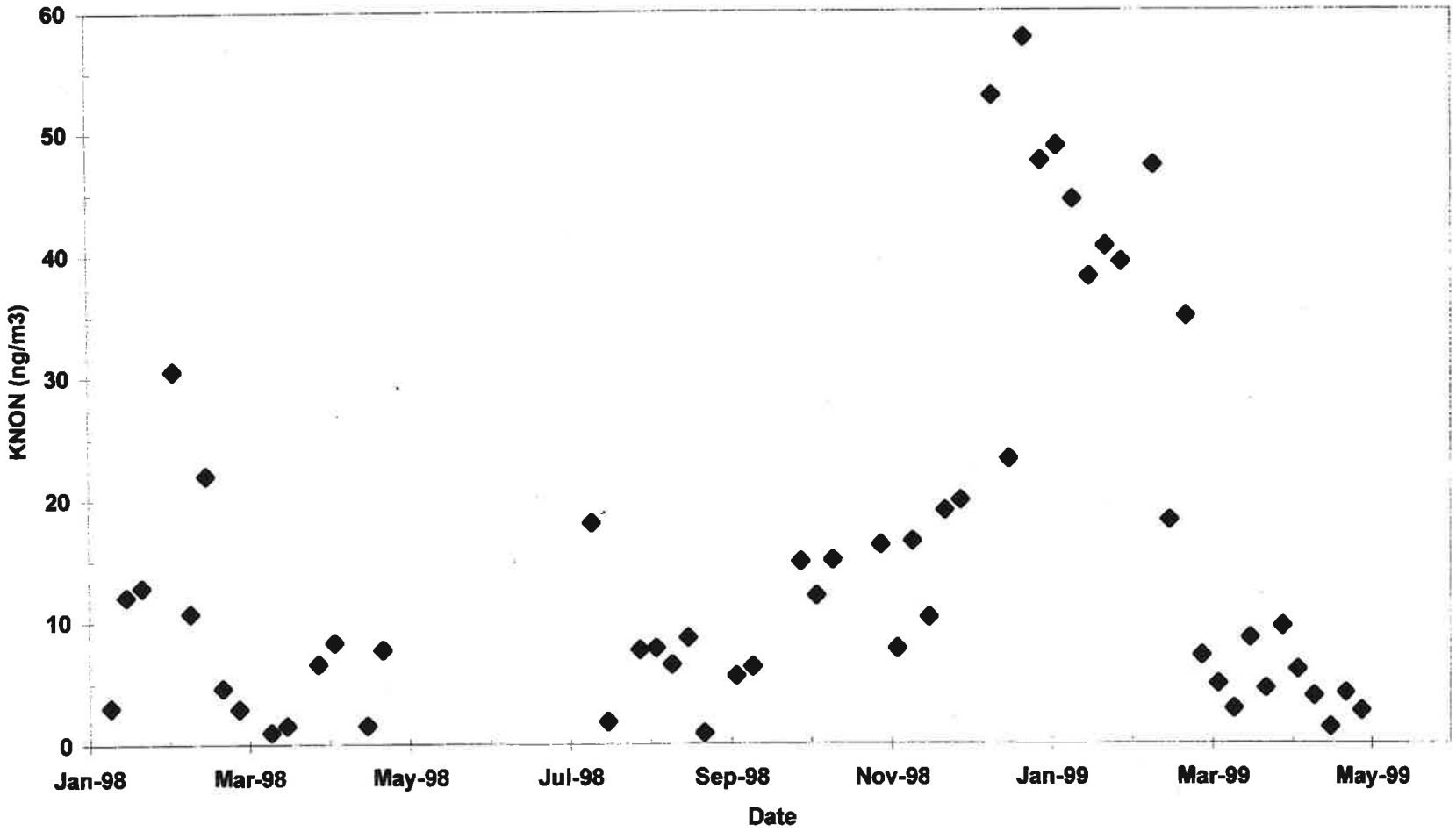


Figure 33.
Ammonium Sulfate [NHSO, (NH4)2SO4] - IMPROVE Data, Freeport, ME

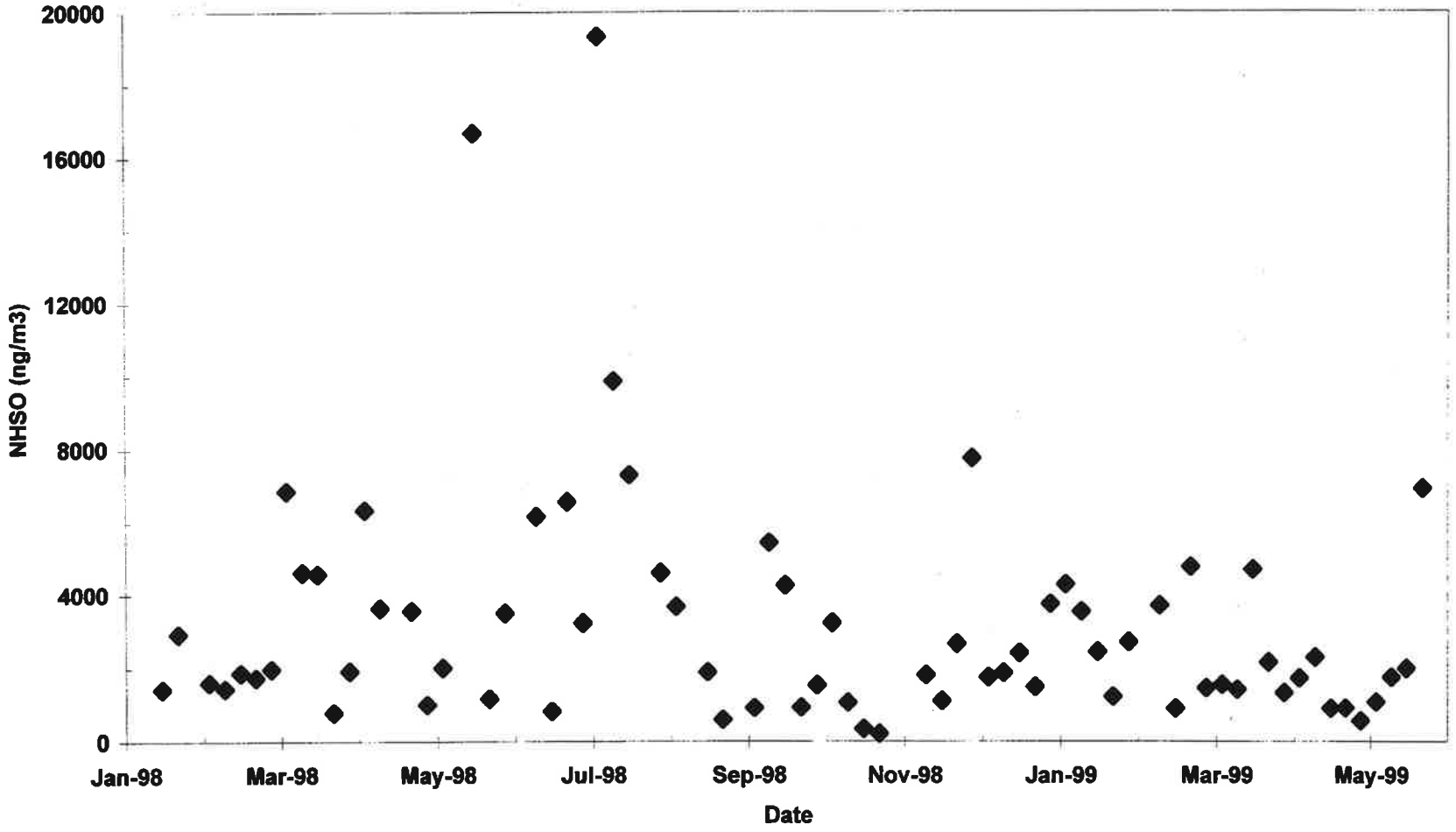


Figure 34.
Ammonium Nitrate [NHNO, (NH4)2NO3)] - IMPROVE Data, Freeport, ME

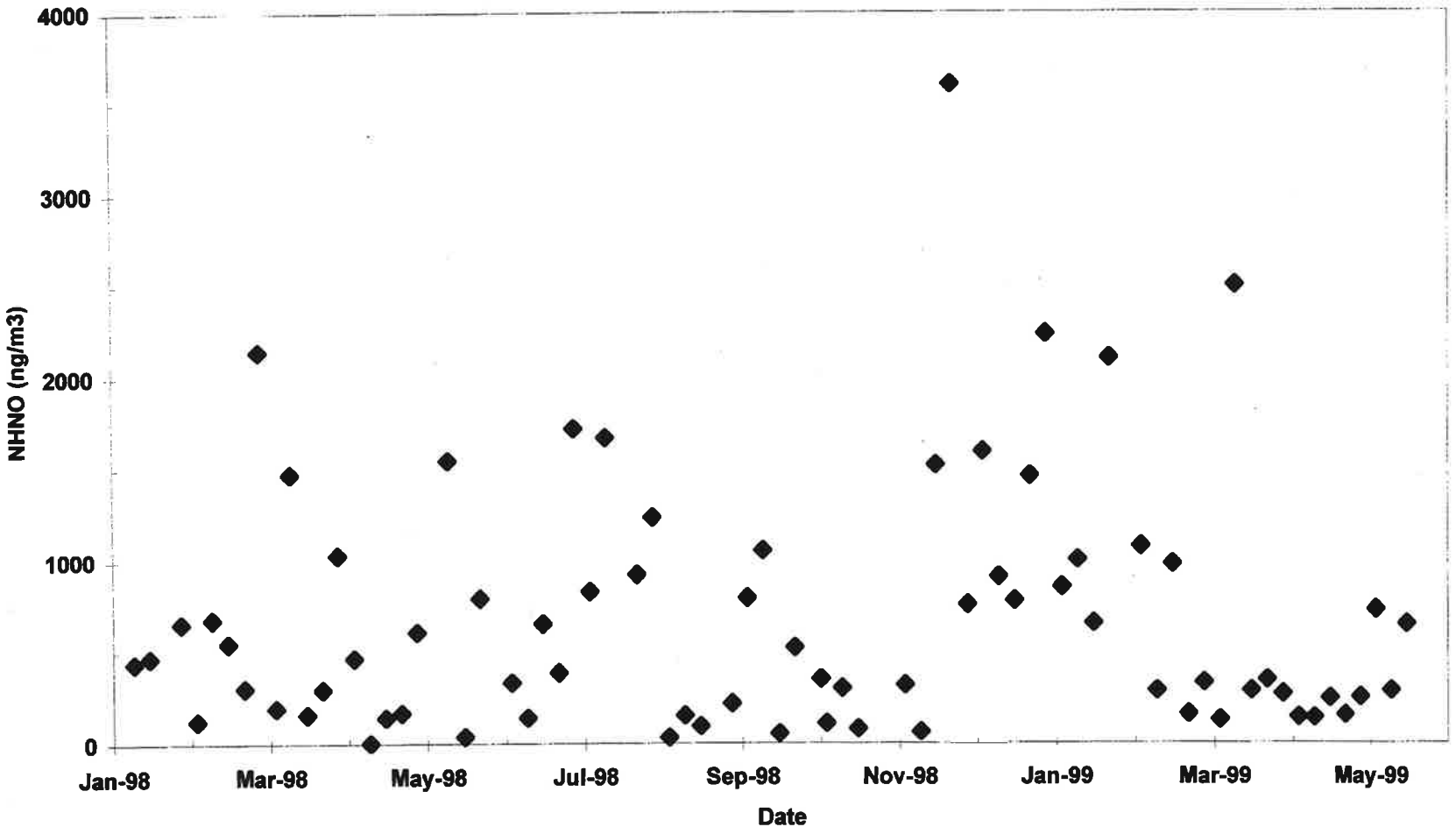


Figure 35.
Reconstructed Mass without Nitrate (RCMC) - IMPROVE Data, Freeport, ME

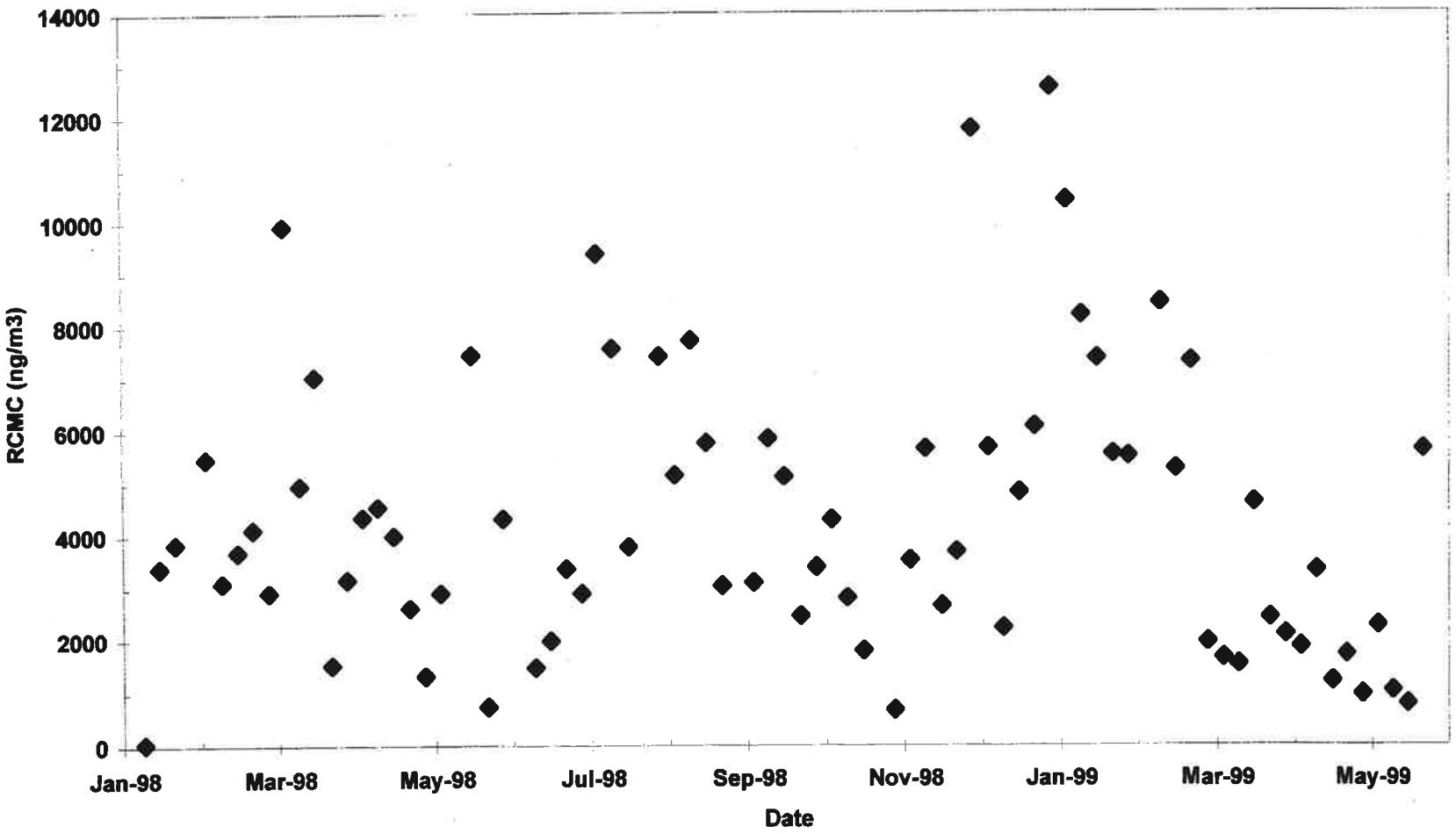


Figure 36.

Average Aerosol Composition
Improve Data, Freeport, ME

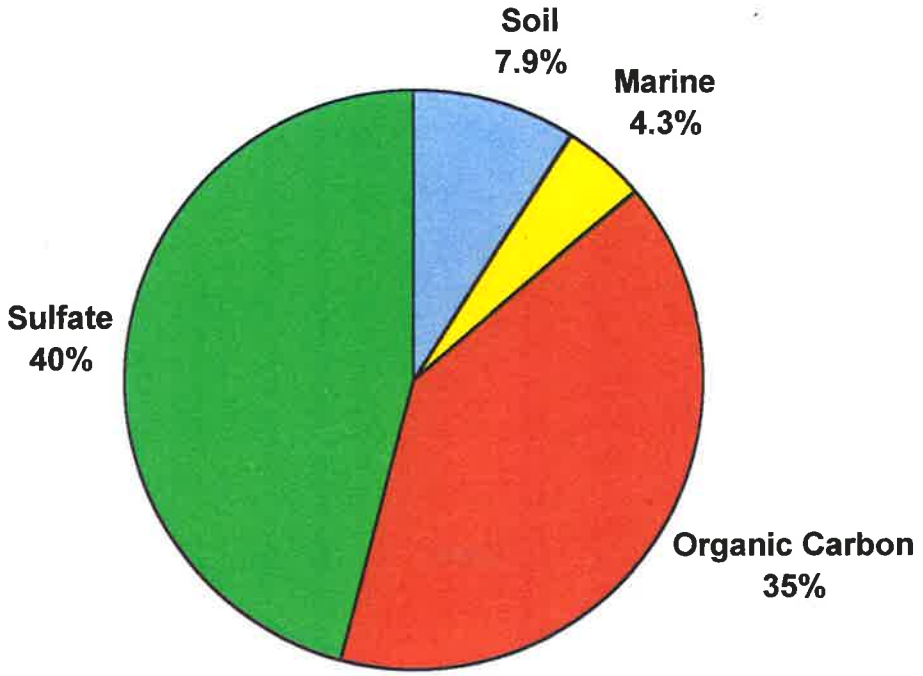


Figure 37.

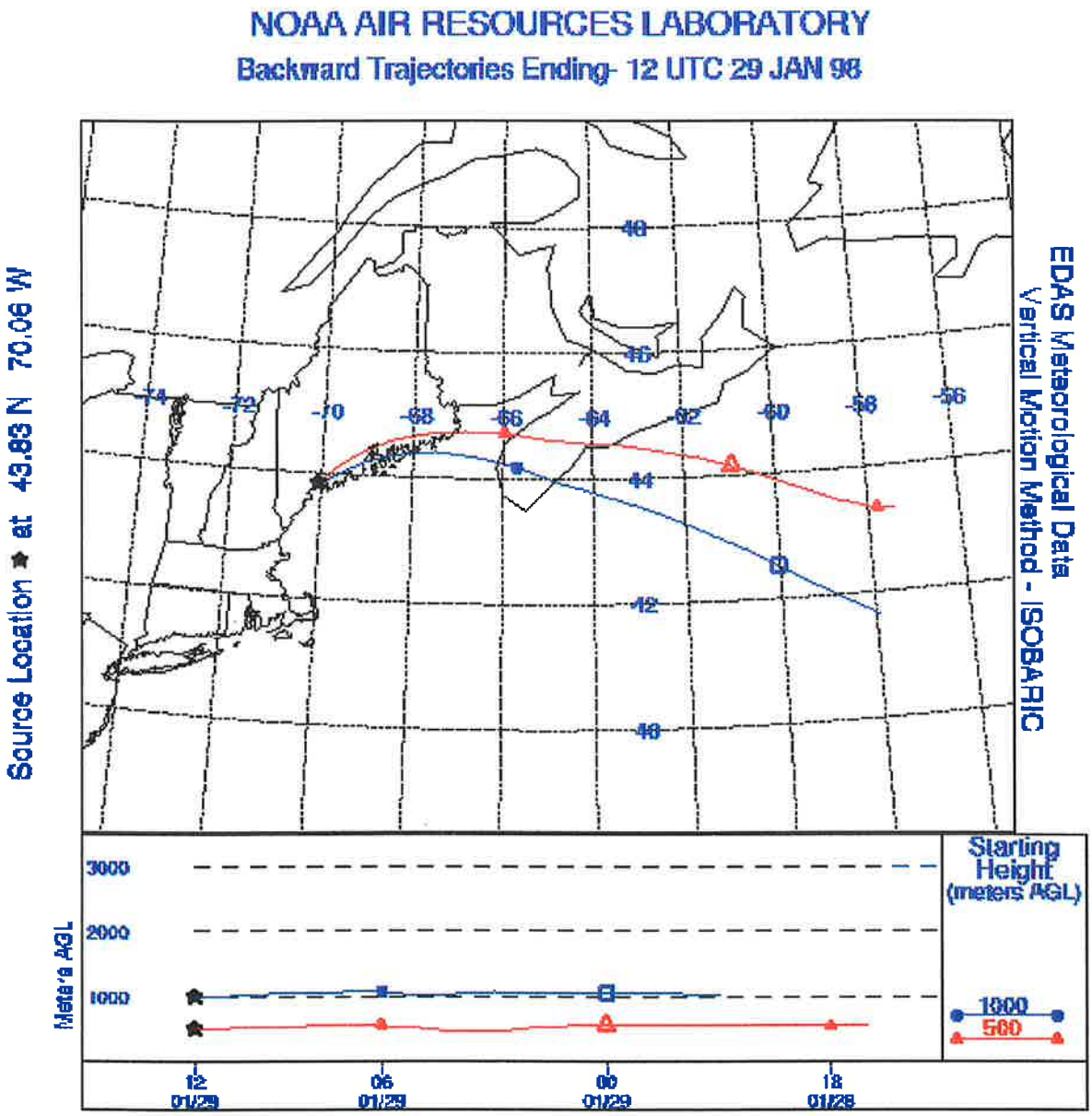


Figure 38.

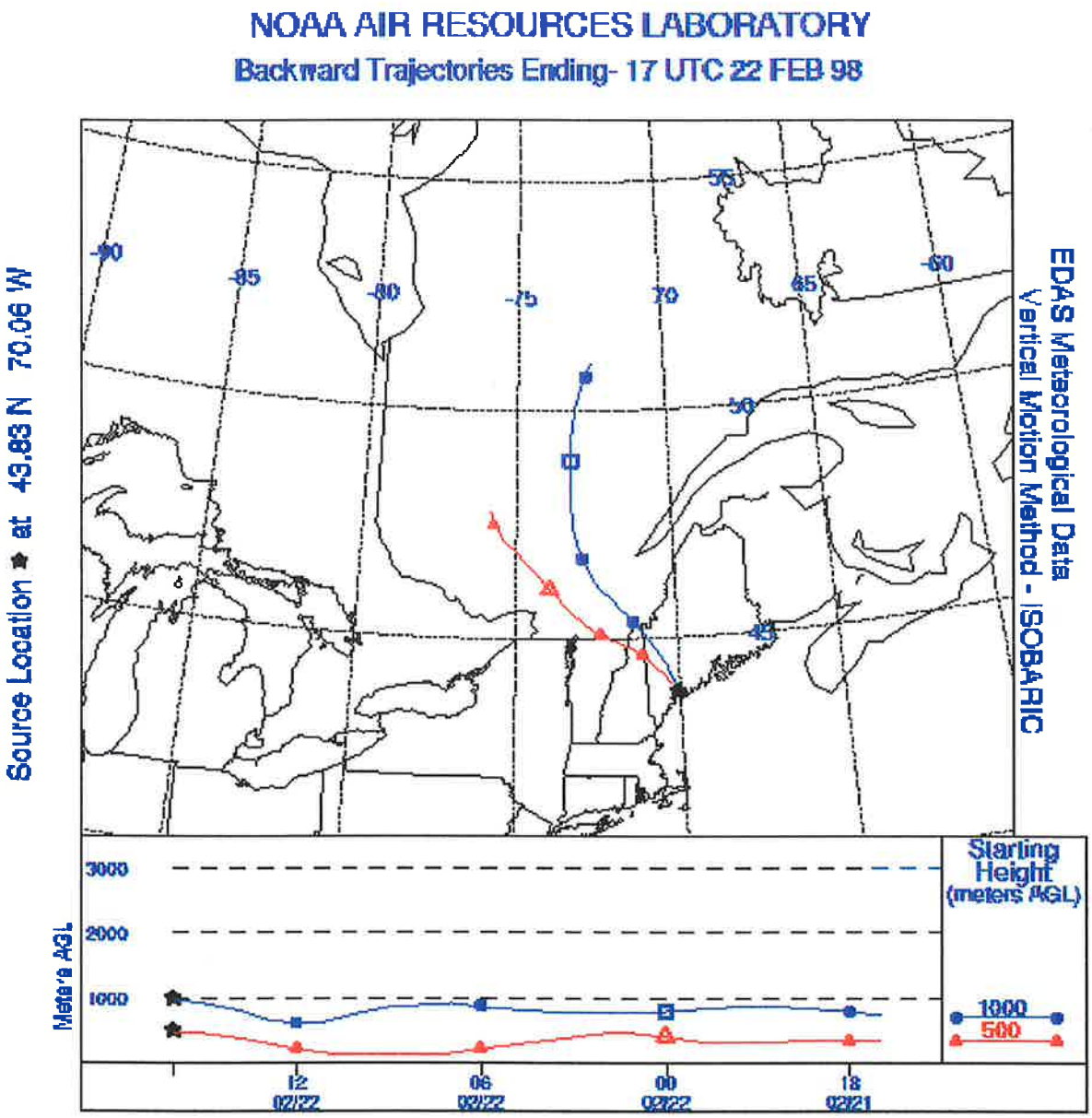


Figure 39.

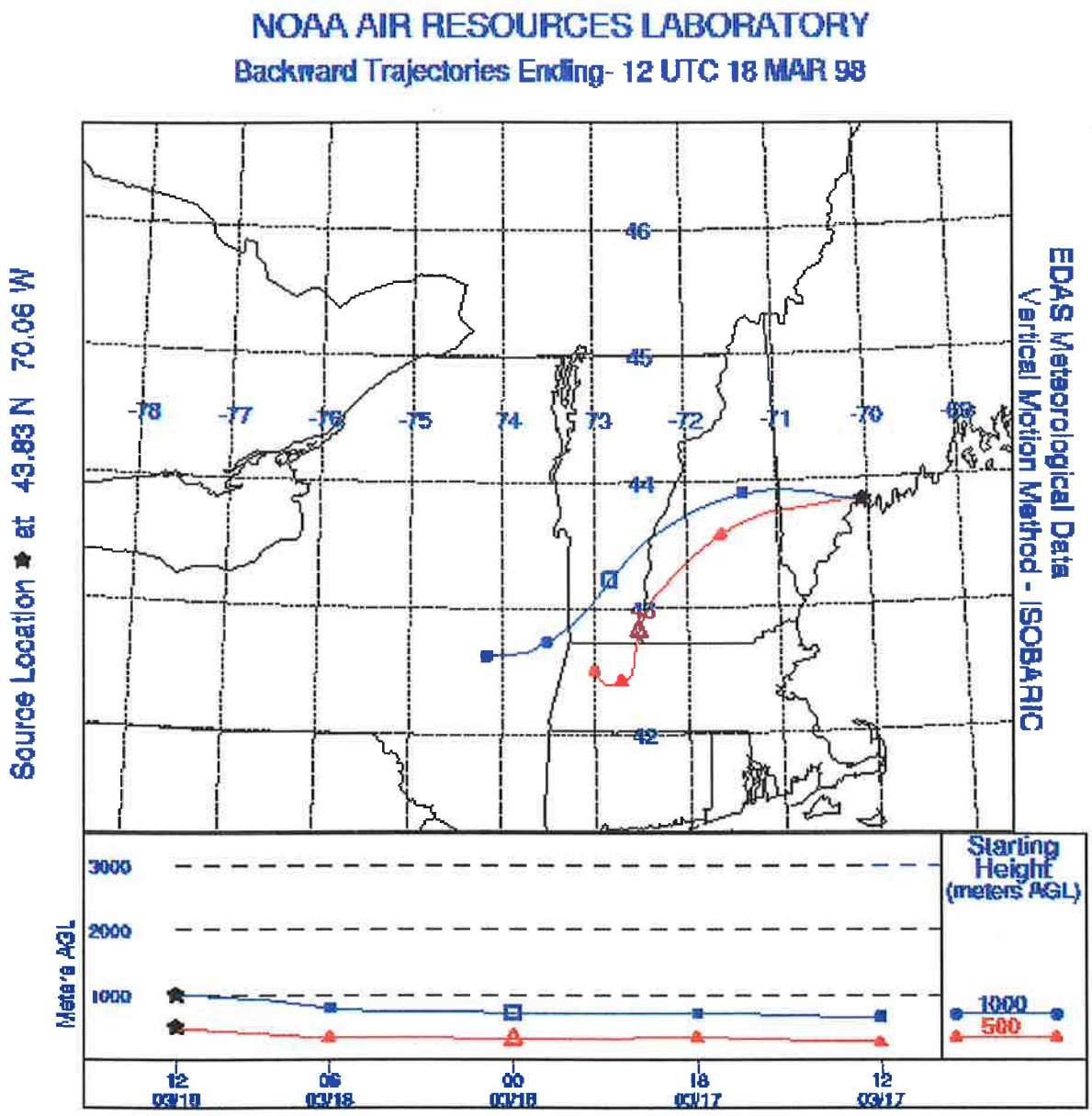


Figure 40.

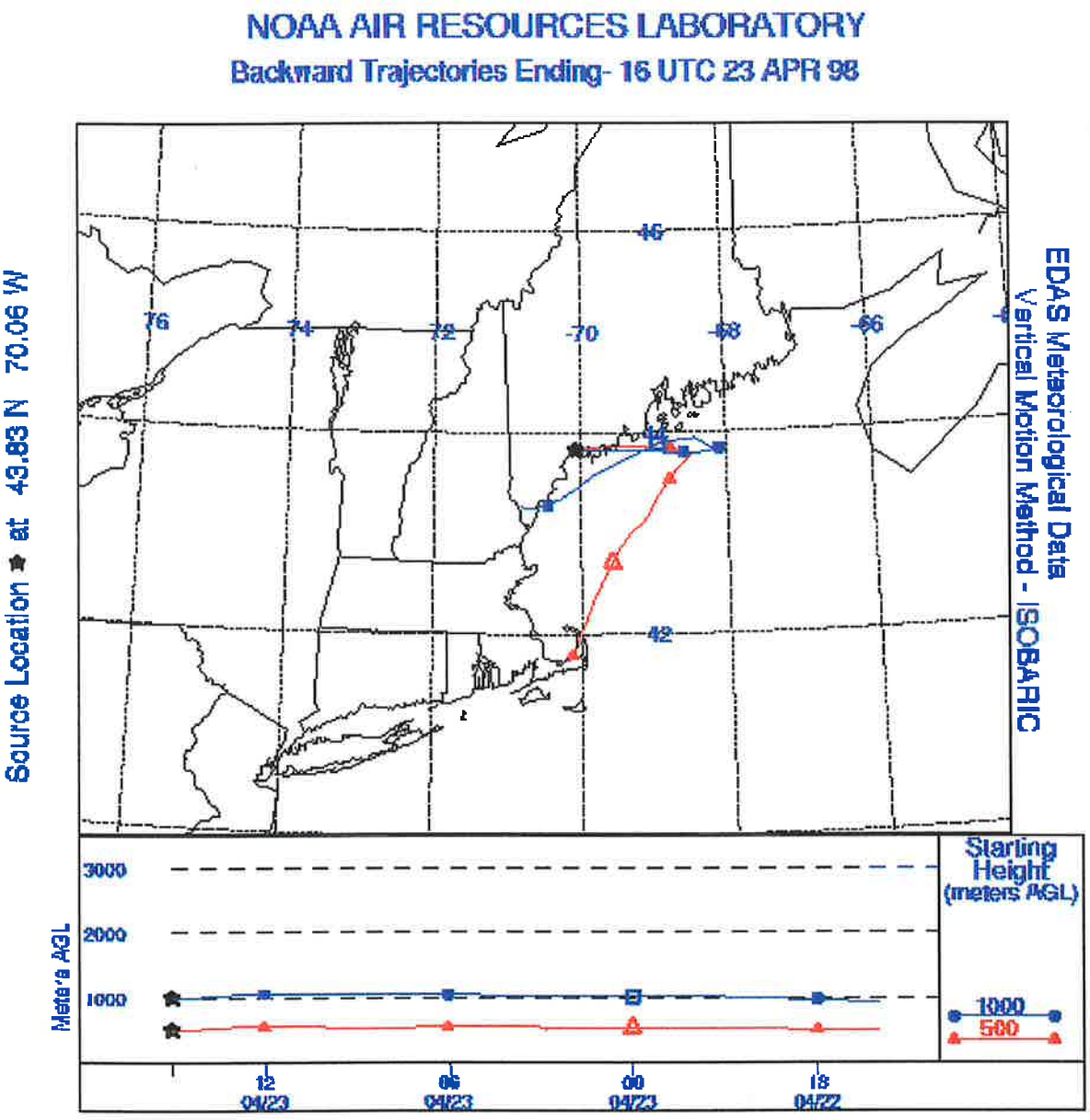


Figure 41.

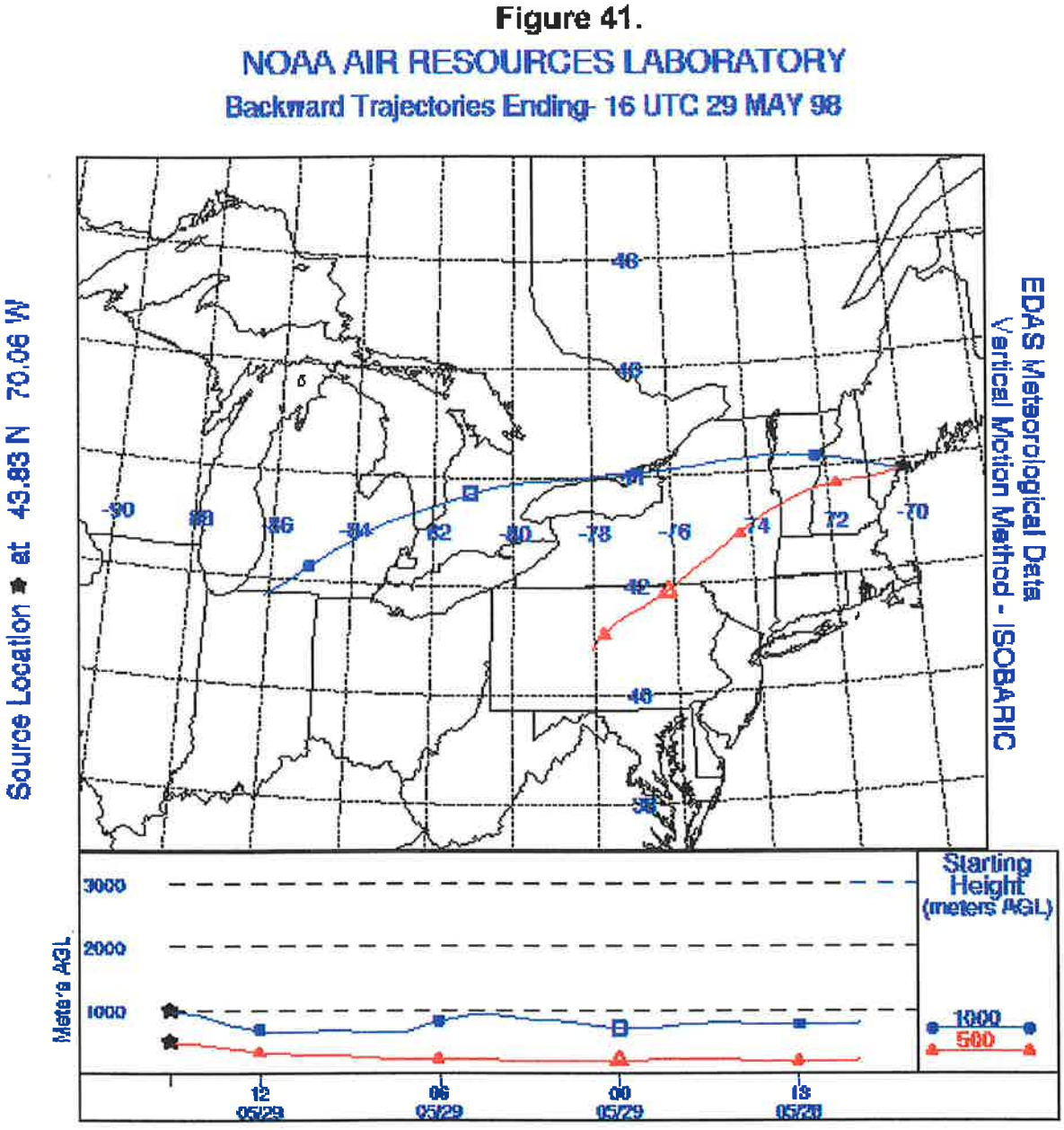
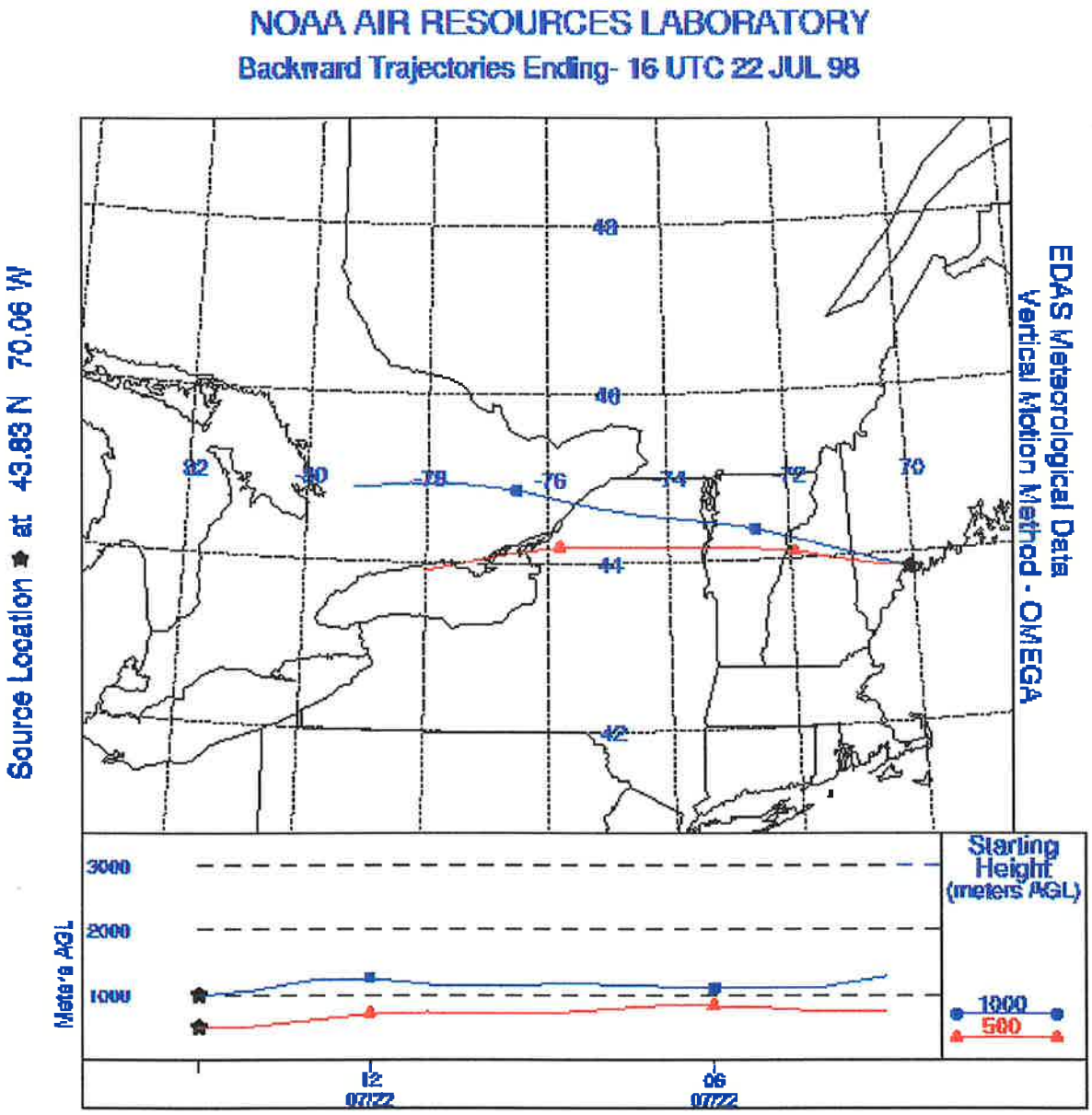


Figure 42.



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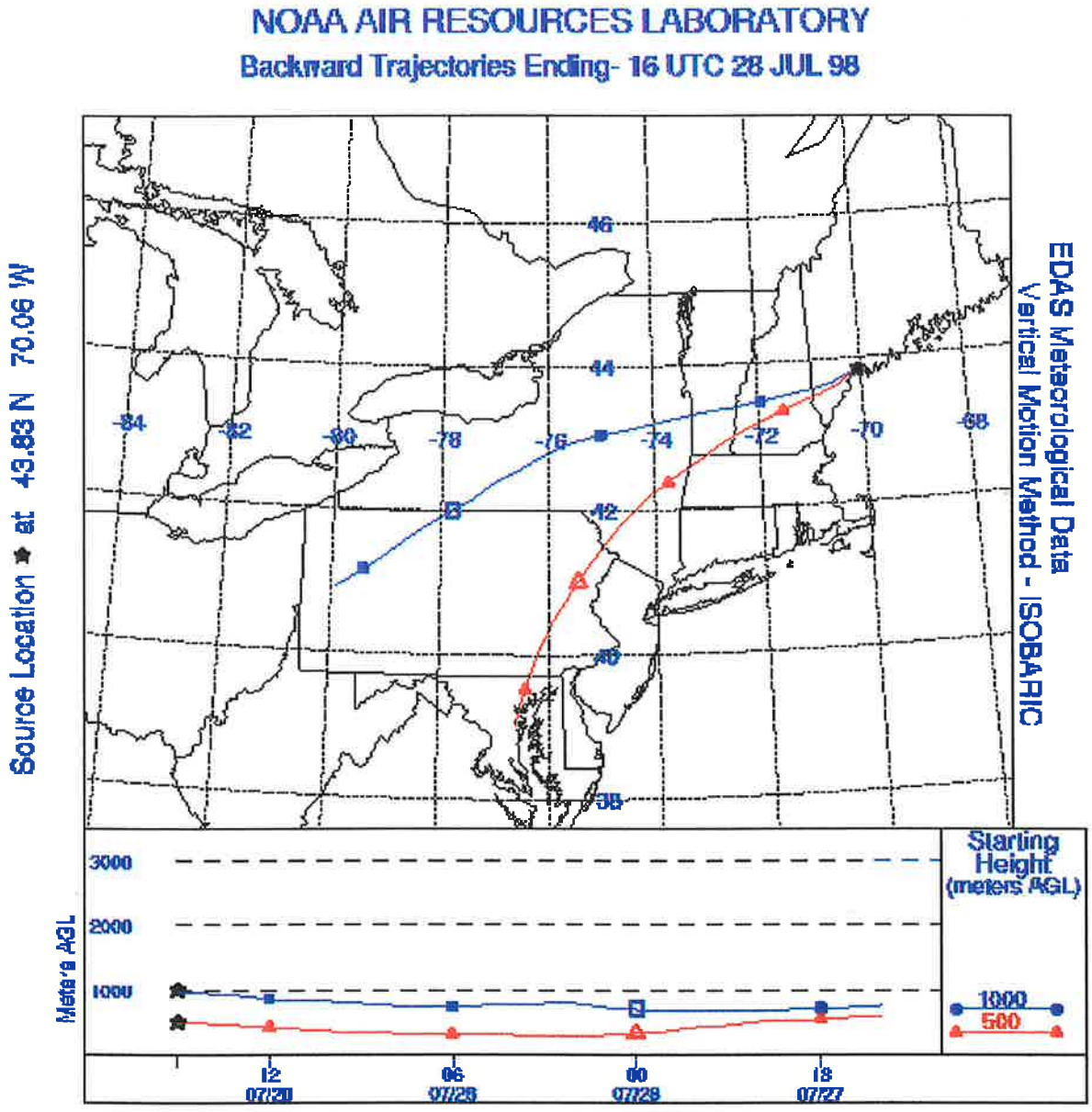

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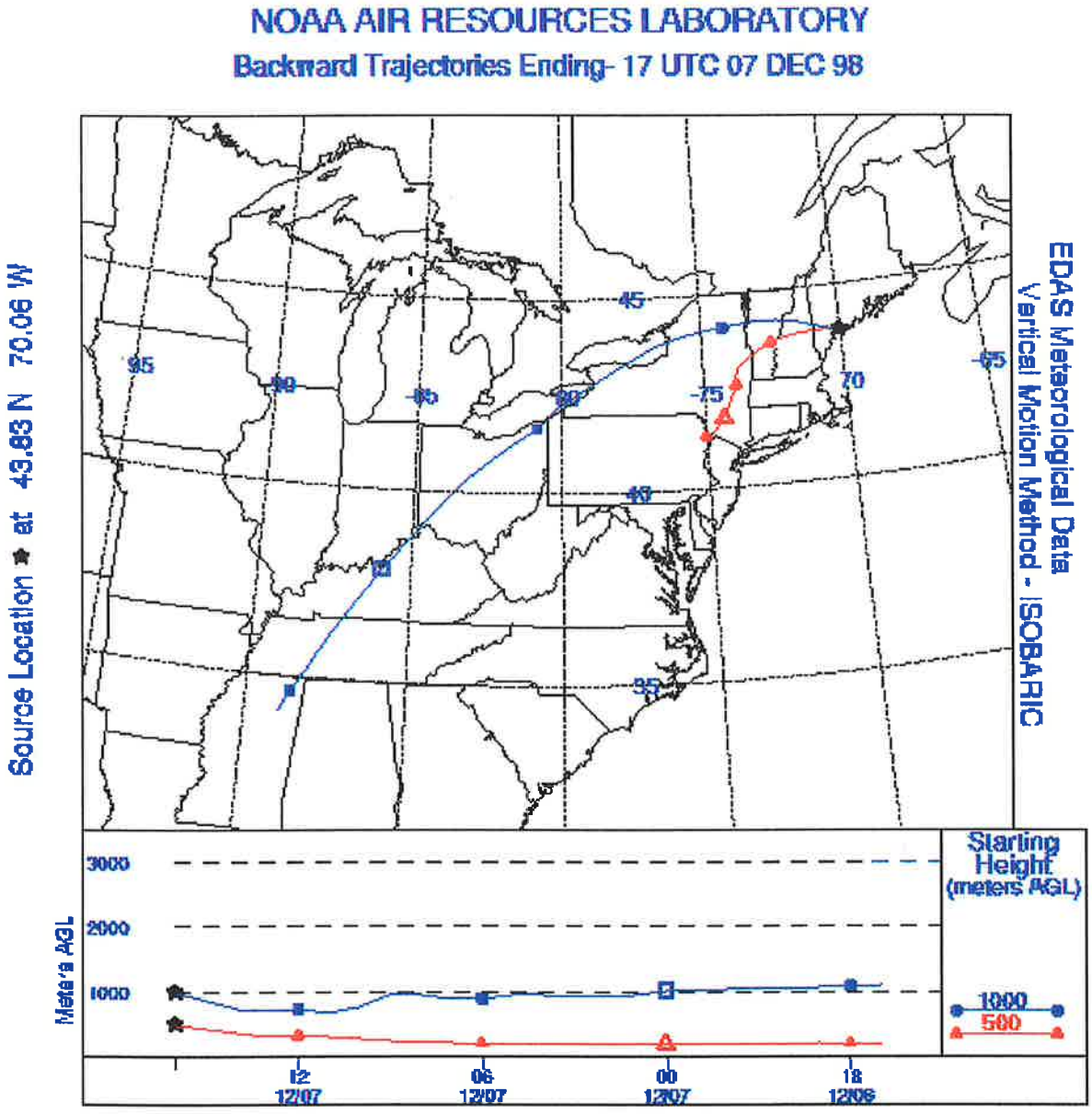

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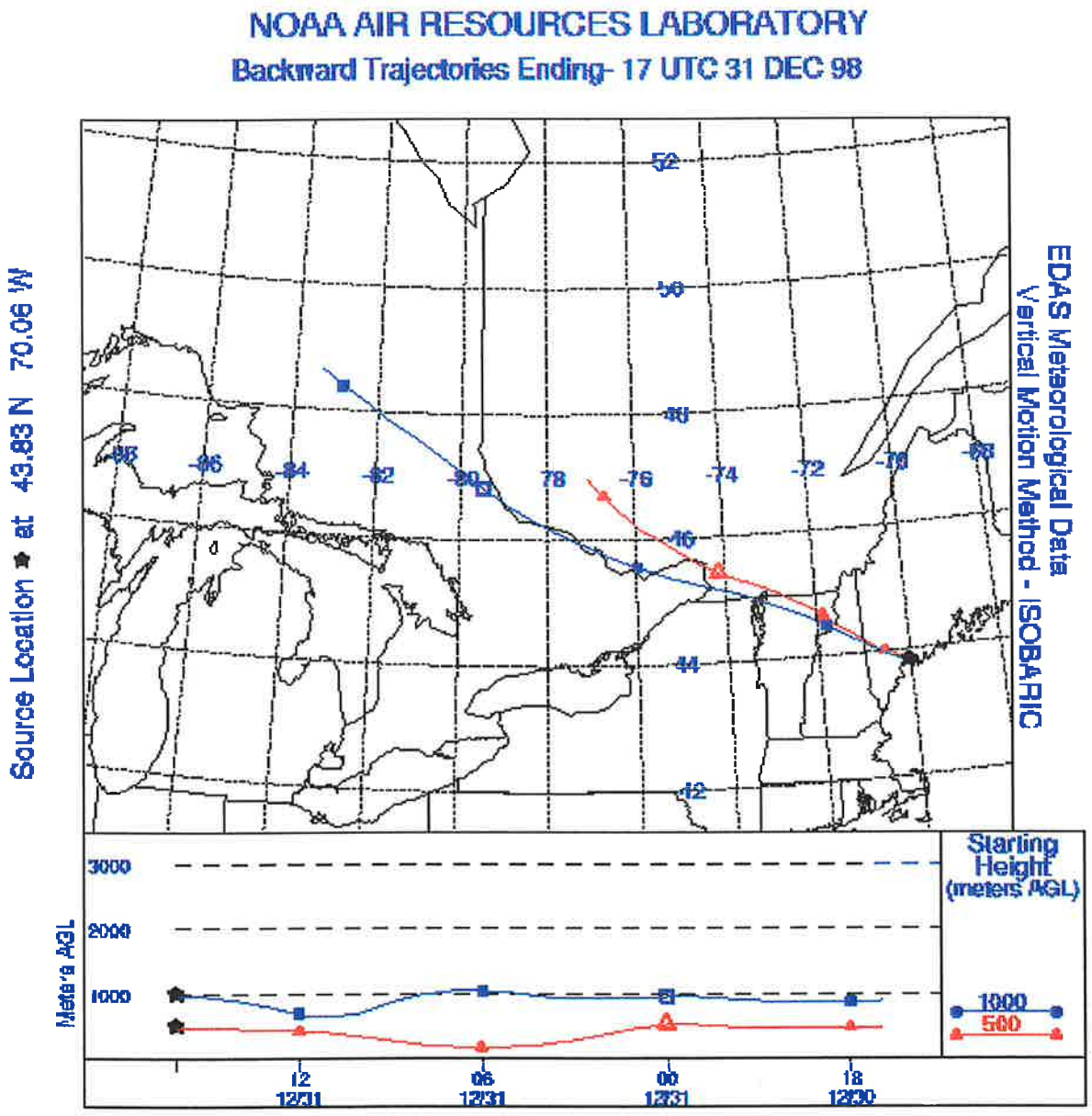


Figure 46.

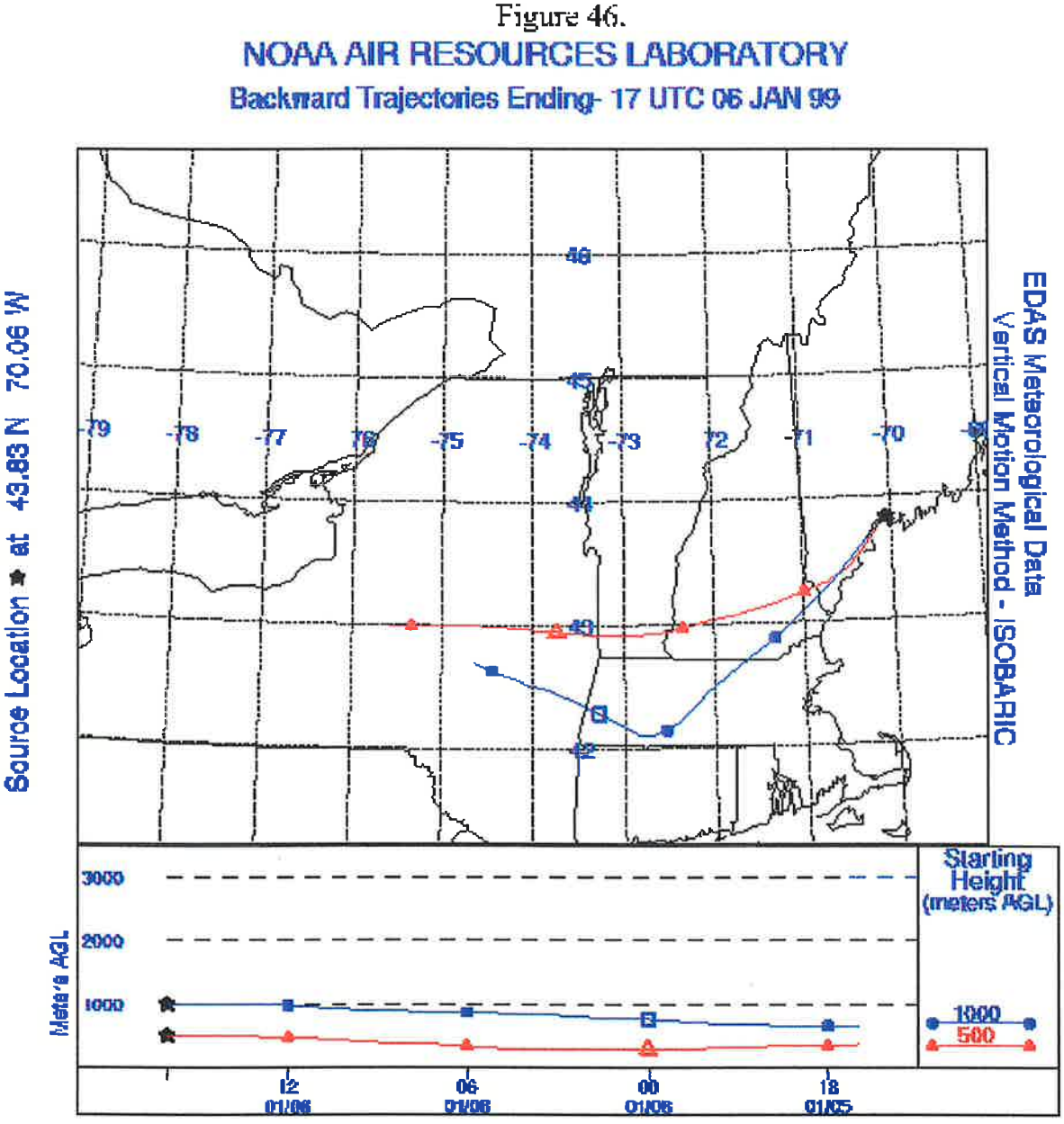


Figure 47.

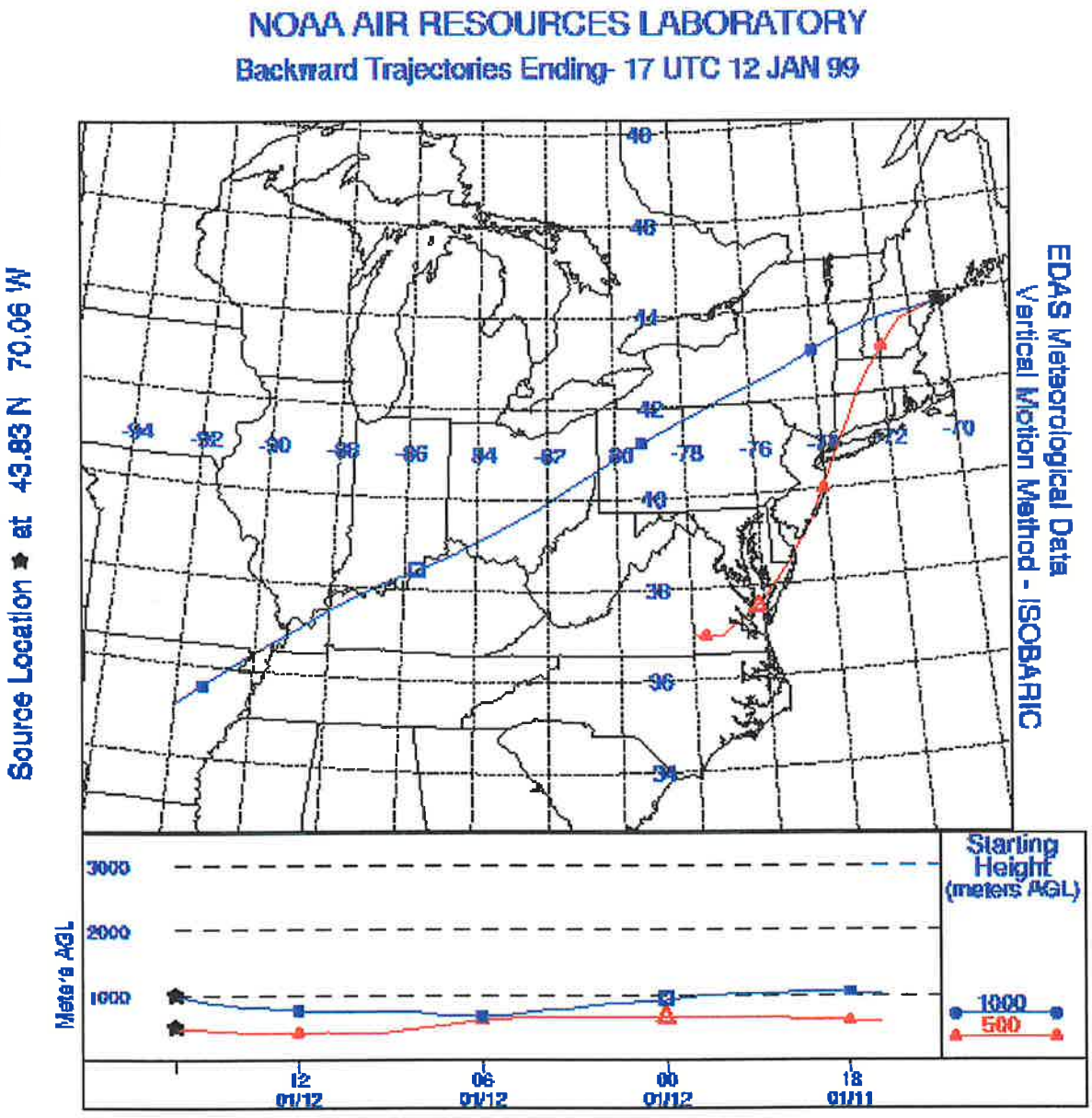


Figure 48.

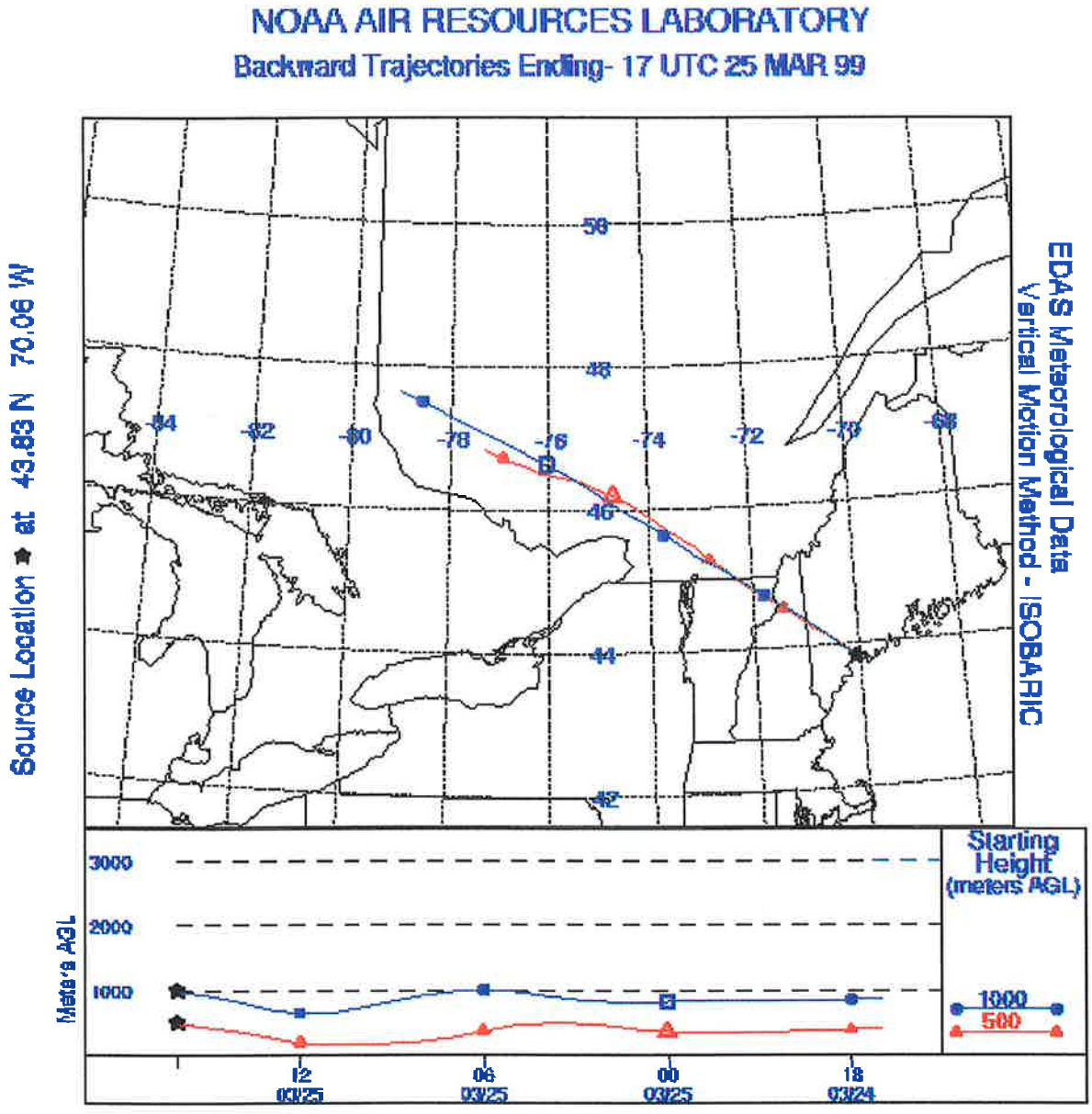


Figure 50.
Average Precipitation Composition NADP Data - 1998 Freeport, ME
(volume weighted mean, mg/l)

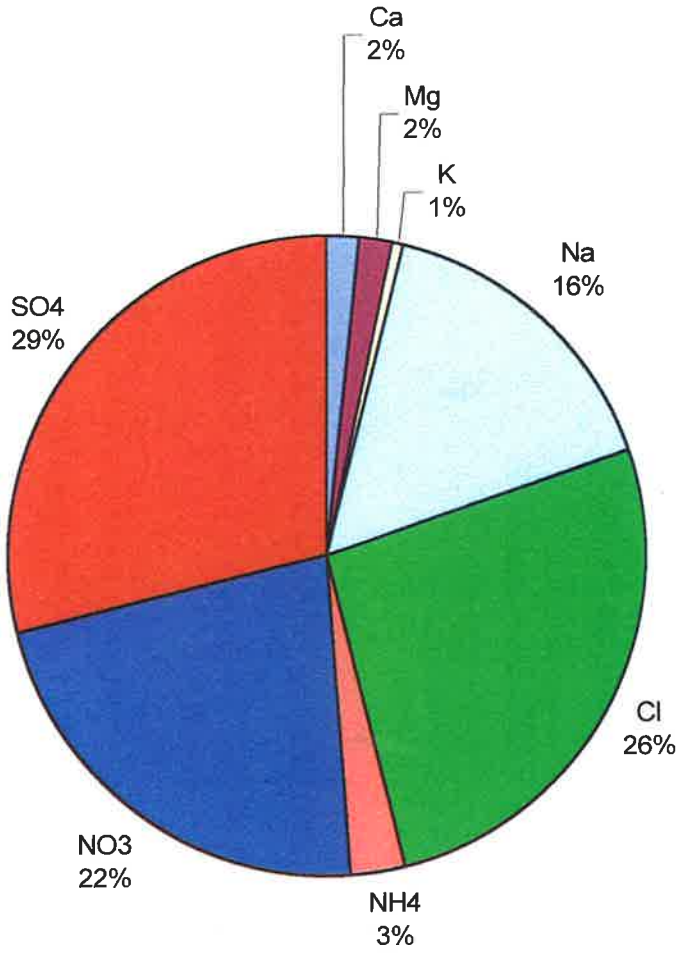


Figure 52.
Mercury Concentration and Deposition
MDN Data, Freeport, ME

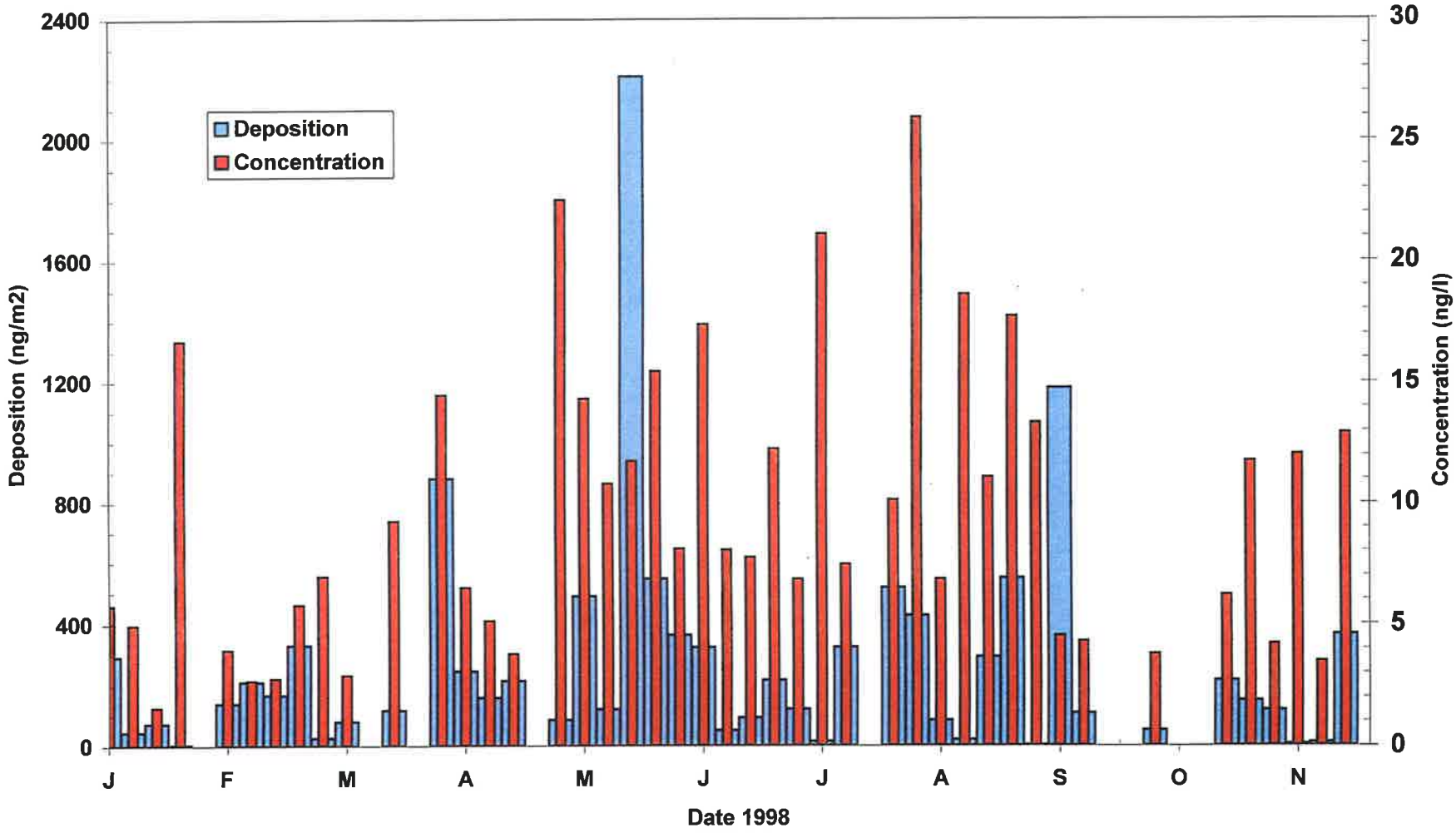


Figure 53.
Hg vs SO4 MDN and NADP Data, Freeport, ME

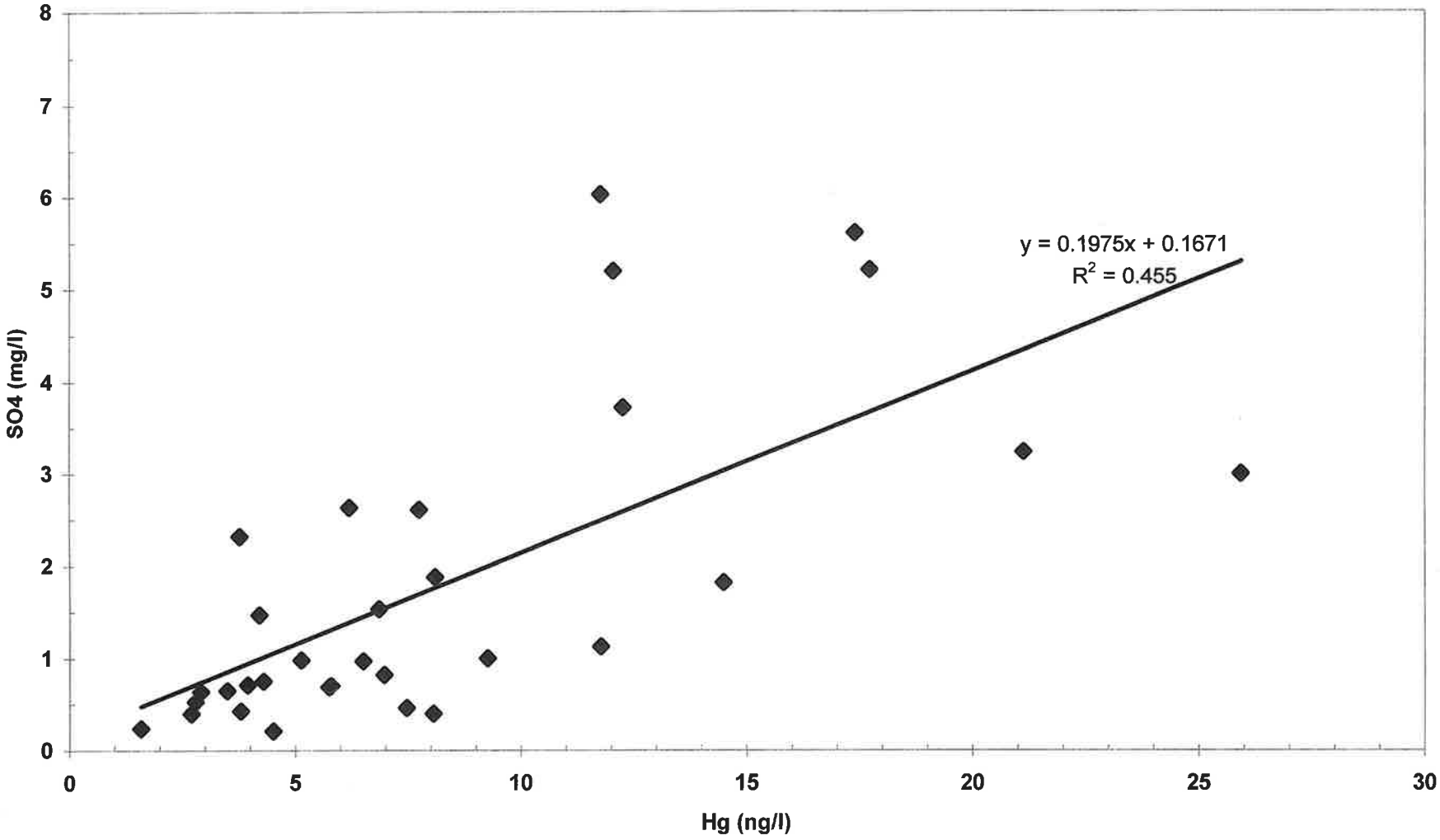


Figure 54.
Hg vs pH MDN and NADP Data, Freeport, ME

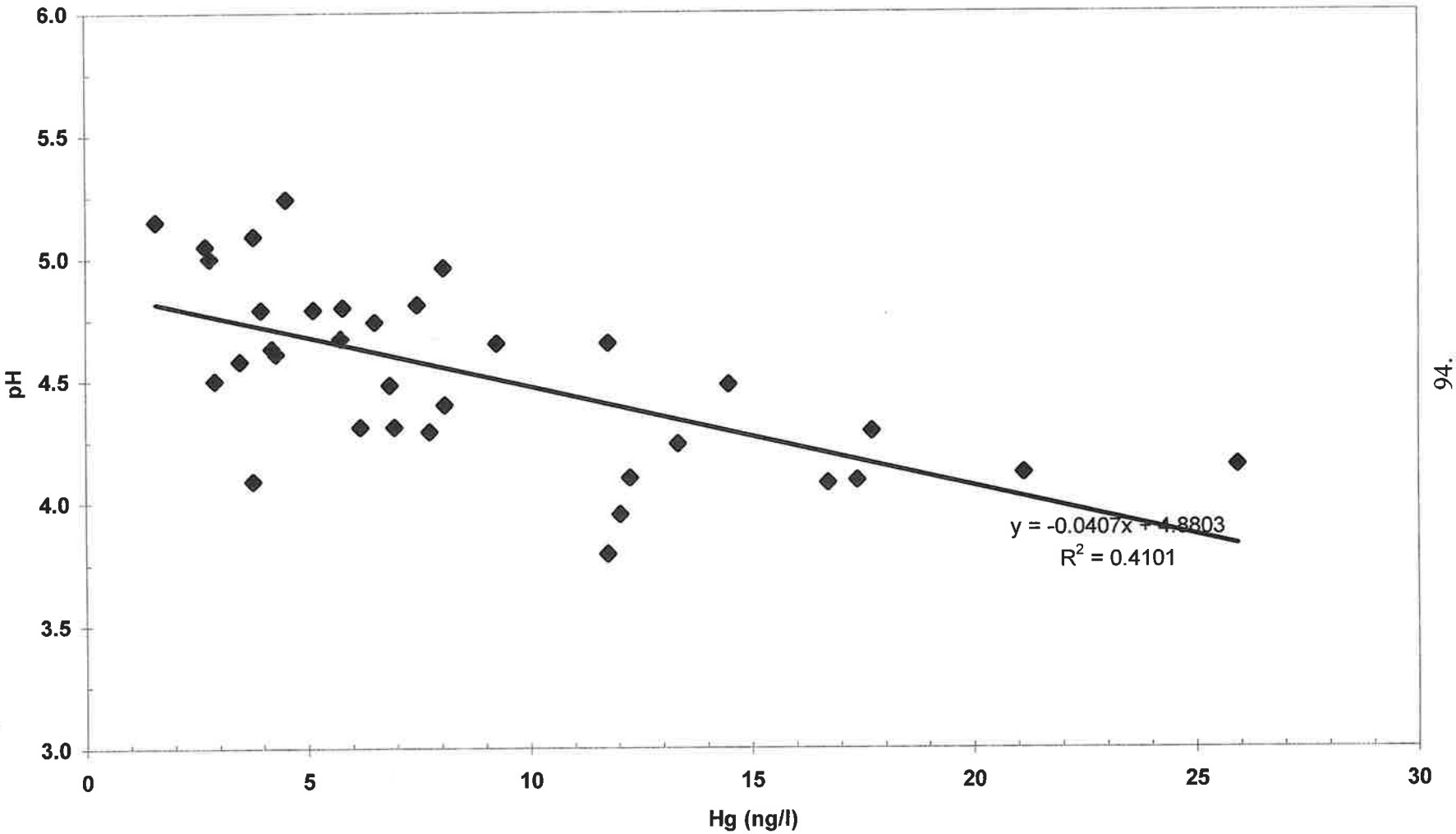


Figure 55.
Hg vs Precipitation Depth MDN Data, Freeport, ME

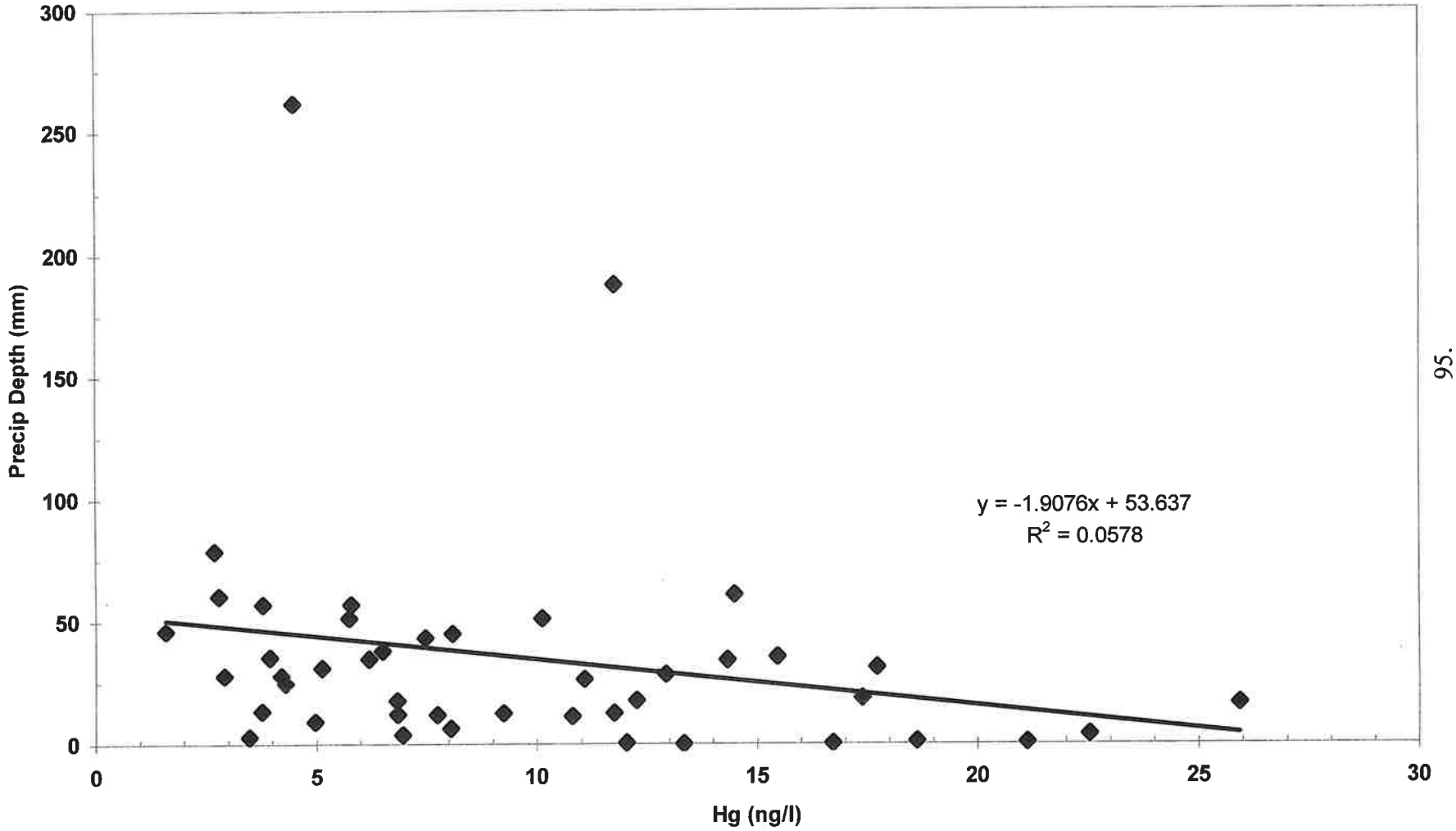


Figure 56.
Seasonal Mercury Deposition vs Precipitation Depth

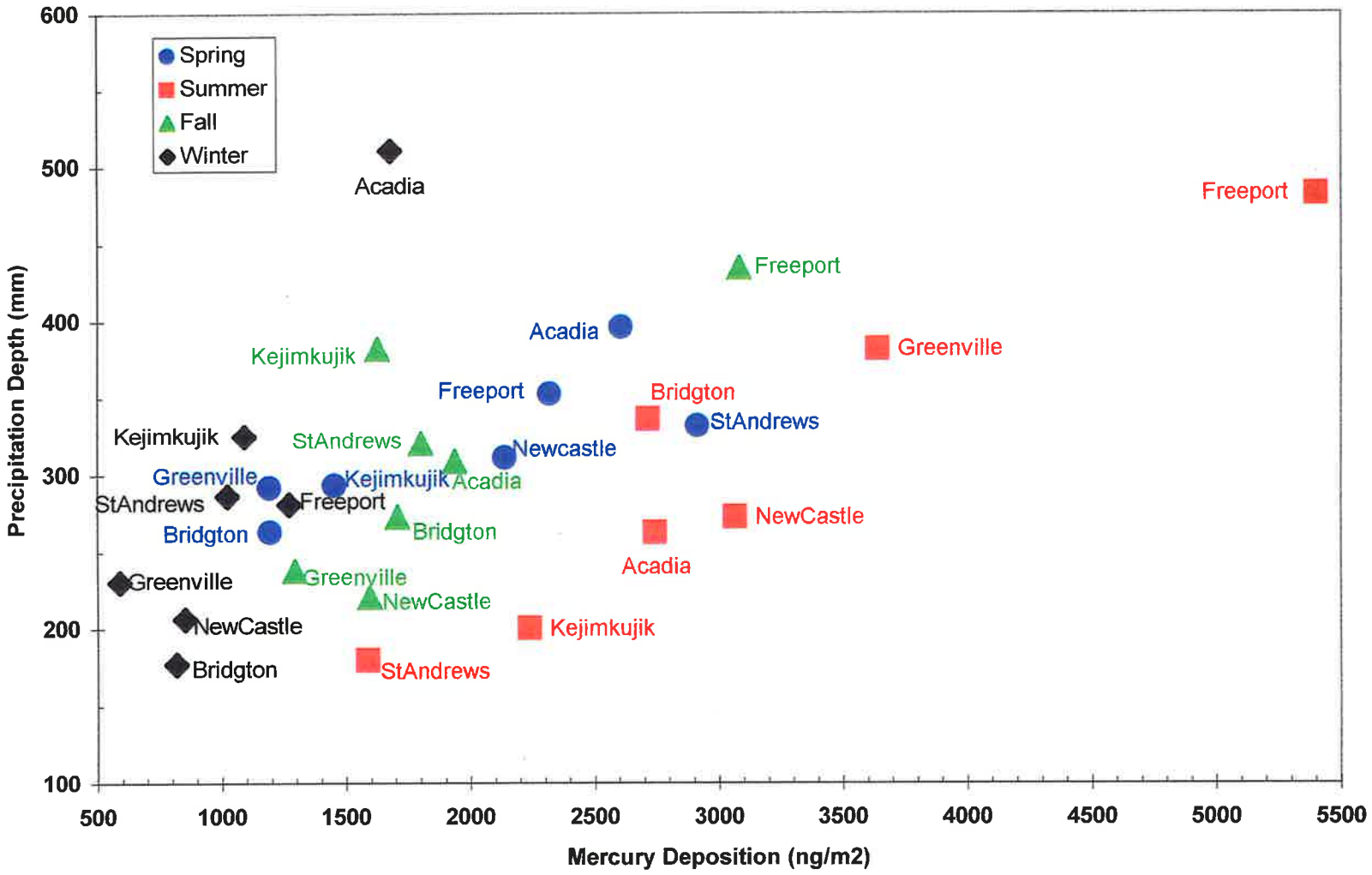


Figure 57.
Average PAH Composition for Dry Deposition at Freeport, ME

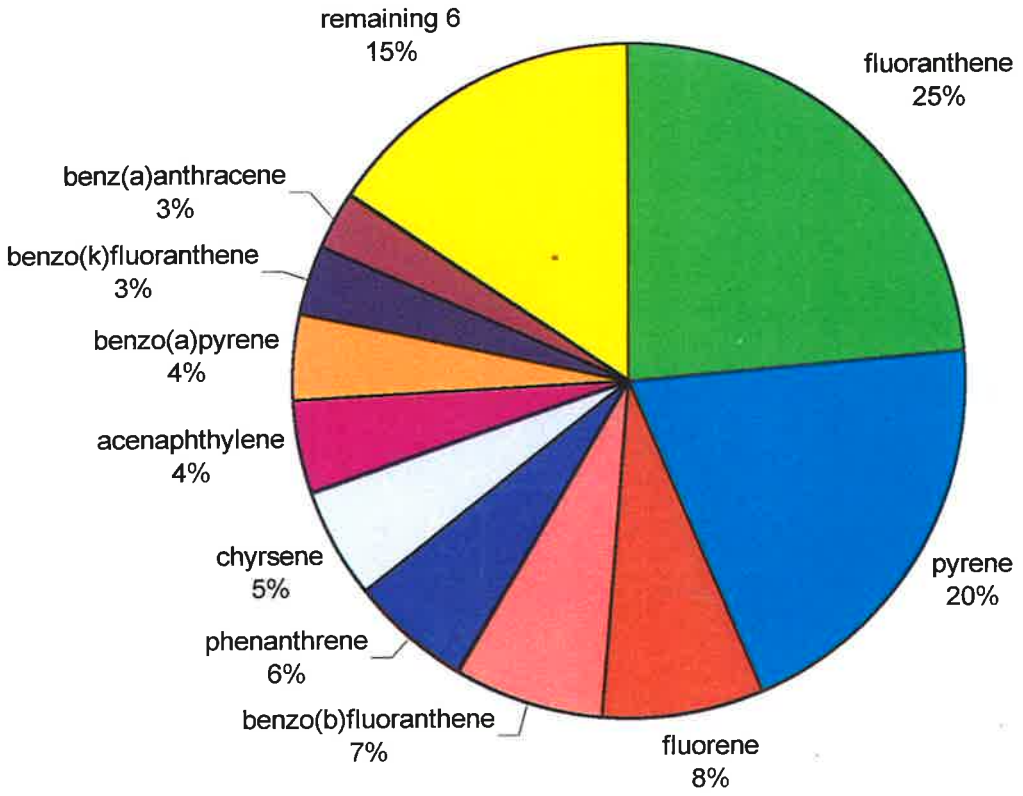


Figure 58.
Average PAH Composition in Wet Deposition at Freeport, ME

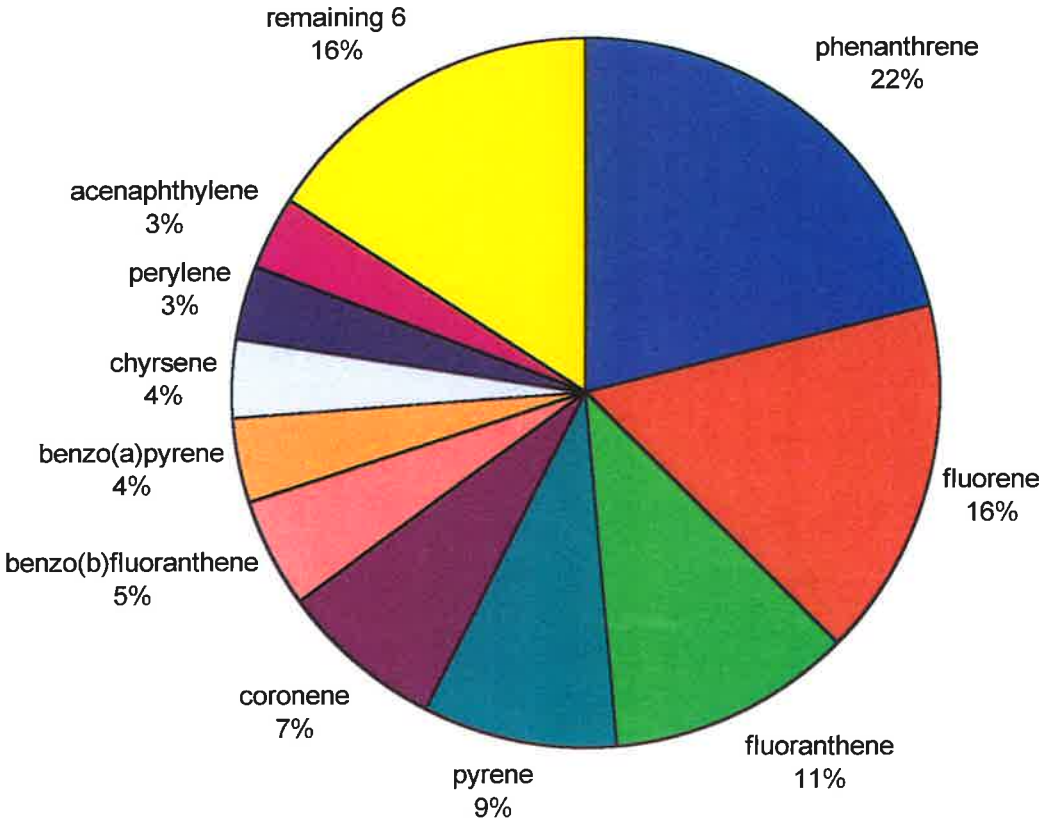
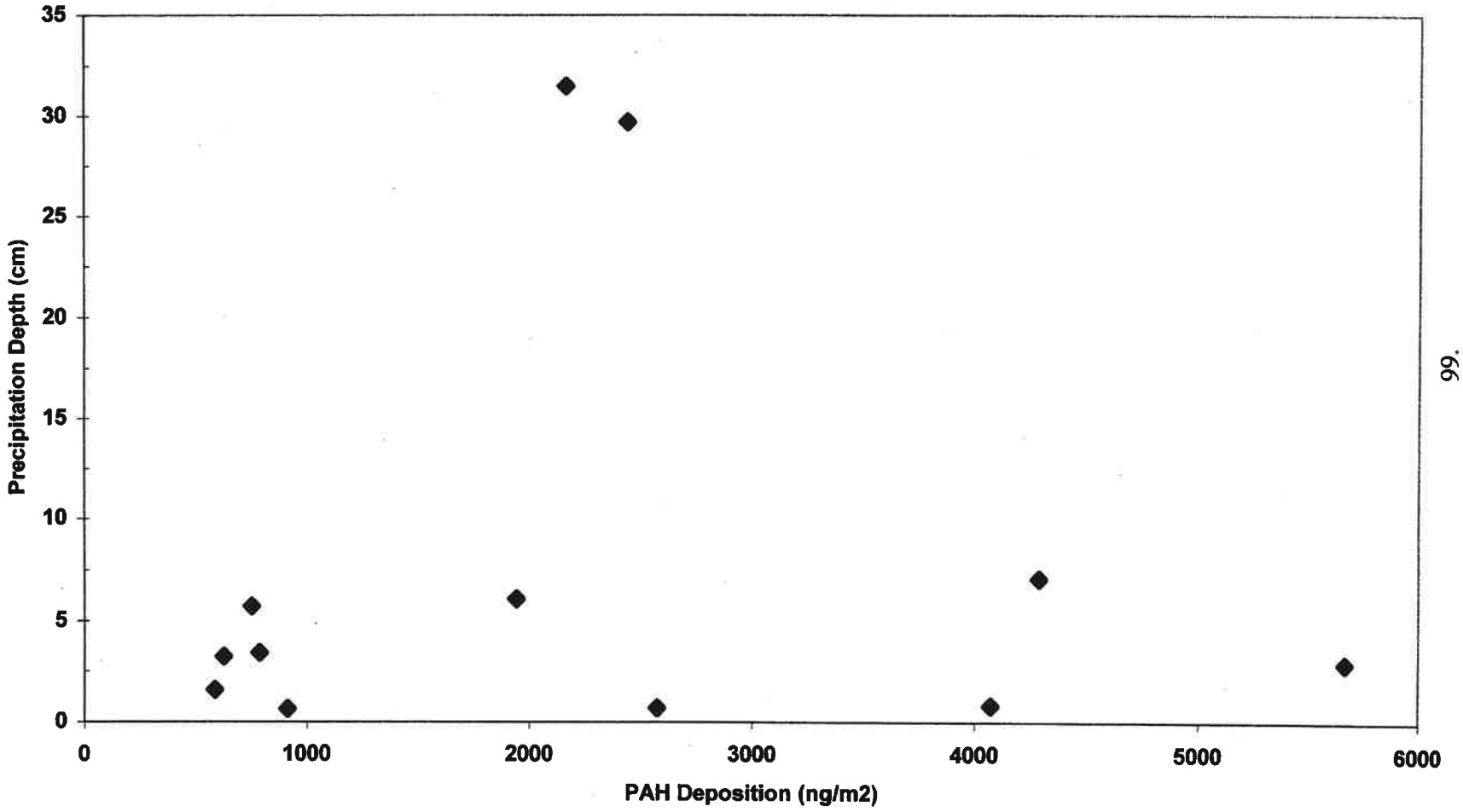


Figure 59.
PAH Wet Deposition vs Precipitation Depth



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