

CHEMICAL QUALITY OF PRECIPITATION AT GREENVILLE, MAINE

By Joseph A. Smath and Thomas L. Potter

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DONALD PAUL HODEL, Secretary

GEOLOGICAL SURVEY

Dallas L. Peck, Director

For additional information
write to:

U.S. Geological Survey, WRD
26 Ganneston Drive
Augusta, ME 04330

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CONVERSION FACTORS AND ABBREVIATIONS

For the convenience of readers who may prefer to use inch-pound units rather than the metric (International System) units used in this report, values may be converted by using the following factors:

Multiply metric units	by	To obtain inch-pound units
<u>Length</u>		
millimeter (mm)	0.03937	inch (in)
centimeter (cm)	0.3937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
<u>Area</u>		
square meter (m ²)	10.76	square foot (ft ²)
hectare (ha)	2.471	acre
<u>Volume</u>		
liter (L)	0.2642	gallon (gal)
milliliter (ml)	0.0610	cubic inch (in. ³)
<u>Mass</u>		
gram (g)	0.03527	ounce (oz)
<u>Temperature</u>		
degree Celsius (°C)	$F = 1.8 \times \text{°C} + 32$	degree Fahrenheit (°F)

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ABSTRACT

Weekly composite precipitation samples were collected at a rural site located in Greenville, Maine for analysis of trace metals and organic compounds. Samples collected during February 1982, through May 1984, were analyzed for cadmium, chromium, copper, lead, mercury, nickel, and zinc and during February 1982, through March 1983, for chlorinated hydrocarbon pesticides, phthalate ester plasticizers, and polychlorinated biphenyls. Deposition rates were computed. Data reported by the NADP (National Atmospheric Deposition Program) was used to evaluate the general chemical quality of the precipitation.

The precipitation had relatively high concentrations of hydrogen ions, sulfate, and nitrate, compared to other constituents. Of the trace metals included for analysis, only copper, lead, and zinc were consistently detected. Lead concentrations exceeded the U.S. Environmental Protection Agency recommended limit for domestic water supply in three samples. High deposition rates for some of the metals were episodic. Alpha-hexachlorocyclohexane was the only organic compound that was consistently detected (maximum 120 nanograms per liter). The beta, gamma, and delta isomers were detected less frequently. None of the other organic compounds were detected in any of the samples.

INTRODUCTION

Concern has been mounting over pollutants deposited from the atmosphere in the northeastern United States and their potential impact on terrestrial and aquatic ecosystems, water quality, and public health (Bridge and Fairchild, 1981). The major focus of attention has been the deposition of acid-forming constituents commonly termed "acid rain." Studies have also shown that the deposition of toxic metals, such as lead and mercury, and synthetic organic chemicals may also be occurring over this region.

Pollutants deposited from the atmosphere seem to be distributed over broad geographic areas. This indicates that pollutants emitted into the upper atmosphere may travel thousands of miles before they are deposited (National Academy of Sciences, 1983). The problems associated with the atmospheric deposition of pollutants and the potential solutions are, therefore, national and international in scope.

In the eastern United States, the general patterns of atmospheric circulation suggest that pollutants are transported into the northeast from urban and industrialized regions to the south and west. The pollutants are deposited by precipitation (wet deposition) which scours them from the atmosphere or as particles and gases (dry deposition) that reach the earth's surface by gravitational settling and surface impaction.

In response to concern about the possible consequences of atmospheric deposition in the United States, the NADP was created in 1978 by the North-Central Regions of the State Agricultural Experiment Stations. The Program is designed to exist for many years in order to collect long-term information on the amount and extent of deposition of major chemical constituents and mineral acidity over the United States. Currently, there are 110 stations operating in the nationwide sampling network. The stations were sited to avoid local sources of air pollution and are operated in cooperation with numerous State, Federal, and local agencies, industry research groups, and private citizens.

There is no similar effort to monitor the deposition of other pollutants, such as trace metals and synthetic organic chemicals. Direct measurements of these classes of pollutants are limited even though their deposition may be harmful to the environment.

Purpose and Scope

Atmospheric deposition is a highly charged political and scientific issue in Maine. The great interest in "acid" precipitation may obscure the fact that other components of atmospheric deposition may be as harmful to the environment. In 1982, the U.S. Geological Survey and the Maine Department of Environmental Protection undertook this study to investigate the occurrence of trace metals and organic compounds in the precipitation of Maine.

The scope of this study includes sampling and analysis of precipitation at one of the NADP sites in the State for trace metals and organics, evaluation of the NADP data collected at the site to supplement the information on trace metals and organics, and a discussion of the possible direction of future atmospheric deposition sampling efforts in Maine.

Previous Studies

Paleolimnologic studies in remote high elevation lakes and ponds in the northeastern United States have revealed a marked increase in the lead and zinc concentration in sediments, beginning 30 to 80 years ago (Galloway and Likens, 1979; Norton and others, 1981). Lead and zinc have also been reported to be accumulating in undisturbed forest soils in alpine and subalpine areas of New England (Reiners and others, 1975; Siccama and Smith, 1978; Siccama and others, 1980) and in mosses and lichens collected in New Hampshire's White Mountains (Groet, 1976). The principal source of these metals is presumed to be atmospheric deposition. The broad geographic area over which these observations have been made also indicates that atmospheric deposition of lead and other potentially toxic metals is of regional scale. Estimates of lead deposition based on these historical records range from 200 to 1000 (g/ha)/yr (gram per hectare per year).

Direct measurements of the occurrence and the rate of deposition of certain trace metals in precipitation have been published for several locations in northern New England. Unfortunately, they are limited in scale and scope. The data reported are for bulk deposition with composite periods of 1 to 3 months spanning a period from 1966 through 1981. The monitoring periods ranged from 3 months to 4 years and none of the sample collection periods for the different studies coincided. Sampling and analysis techniques were not standardized and investigators chose different suites of trace metals for analysis. The one metal common to all was lead.

The earliest reported data is from Lazrus and others (1970). They sampled precipitation at a site in Caribou, Maine from September 1966 to January 1967 and found deposition rates for lead, zinc, copper, and nickel of 37, 53, 23, and 2 (g/ha)/mo (grams per hectare per month), respectively. Mean concentration values in precipitation could not be computed because the precipitation record for this station was not reported.

At the Hubbard Brook Experimental Forest in New Hampshire, Smith and Siccama (1981) have reported a monthly mean lead concentration in precipitation of 21.6 ug/L (micrograms per liter) for the period January 1975 to December 1978. They also note that the monthly mean lead concentration computed on an annual basis decreased steadily from 25.3 ug/L in 1975 to 14.8 ug/L in 1978. They have suggested that this decline represents a measurable effect of regulatory controls placed on the use of lead in gasoline which were initiated in 1975. However, the limited data base makes this interpretation suspect. The average lead deposition reported for this site was 266 (g/ha)/yr, ranging from a high of 318 in 1975 to a low of 156 in 1978.

Peters and Bonelli (1982) examined data from bulk precipitation samples collected at 179 sites in the north-central and northeastern United States. The samples were composites for the period December 1980 through February 1981. Ten of the sites were located in Maine and four each in New Hampshire and Vermont. The median concentrations for lead, copper, zinc, nickel, and cadmium for all sites was 5, less than 10 (detection limit), 13, 1, and less than 1 (detection limit) $\mu\text{g/L}$, respectively. The Maine sites had a median concentration of 5, less than 10, 5, 1, and 2 $\mu\text{g/L}$, respectively. The median lead deposition rate for all sites was 0.44 (g/ha)/day. For the Maine sites alone, it was 0.3 (g/ha)/day.

The most recent information on trace-metal deposition in northern New England was reported by Kahl and Norton (1983). They sampled precipitation at two sites in eastern Maine during the period September 1980 to December 1981 and found lead concentrations ranging from 0.8 to 8.7 $\mu\text{g/L}$ and zinc concentrations from 6 to 42 $\mu\text{g/L}$. They also reported that samples collected beneath the tree canopy had higher metals concentrations, suggesting a significant contribution from dry deposition. The bulk deposition rate reported for these sites for 1981 was 60 and 300 (g/ha)/yr for lead and zinc, respectively.

These data reported for northern New England demonstrates that lead and zinc and possibly copper, nickel, and cadmium are ubiquitous components of bulk precipitation in rural areas in this region. The data also suggest that there has been a decline in lead deposition commencing in 1975 and that dry deposition may be a significant source of lead and zinc in bulk deposition. More data are needed to establish geographic and temporal trends and to clearly define the modes of deposition.

Organic chemical pollutants enter the atmosphere by way of numerous industrial and agricultural sources. The widespread use of chlorinated hydrocarbon pesticides and industrial chemicals such as PBC's (polychlorobiphenyls) over many decades has apparently established them as a ubiquitous part of the global atmosphere and, likewise, precipitation. This conclusion is supported by a report that a wide range of organic pollutants have been found in air and precipitation samples collected on Enewetak, a remote coral island in the northern Pacific Ocean (Atlas and Giam, 1981). The compounds detected included PCB's, DDT, dieldrin, chlordane, and phthalate ester plasticizers.

Research on the content of organic pollutants in North American precipitation has been focused in the Great Lakes region. In Canada, analyses of samples collected at 24 sites in an area adjacent to the Great Lakes often found PCB's, lindane, alpha-BHC, DDT residues, alpha- and beta-endosulfan, dieldrin, and methoxychlor (Strachan and Huneault, 1979). For those samples in which they were detected, the mean concentrations of these compounds were 21, 5, 12, 3, 2, 5, 1, and 8 ng/L (nanograms per liter), respectively. Sanderson and Lavelle (1979) analyzed monthly bulk composites of precipitation collected at six sites in southern Ontario from June 1975 through June 1977. They found an average PCB concentration of 100 ng/L with a maximum of 320 ng/L.

The Canadian government has maintained a network of 12 precipitation sampling stations in Canada since 1977. Monthly composites of wet deposition have been collected and analyzed for chlorinated hydrocarbon pesticides and PCBs (A. O. Davis, Environment Canada, written commun., 1984). The results from the stations in the Great Lakes Basin are similar to those reported by Strachan and Huneault (1979). At two sites in eastern Canada, St. Hubert, Quebec and Truro, Nova Scotia, where conditions may be similar to those in northern New England, alpha- and gamma-BHC and alpha- and beta-endosulfan have routinely been detected. The concentration of these compounds was in the 1 to 50 ng/L range (A. O. Davis, Environment Canada, unpublished data).

In the United States, Bidleman and others (1981) have reported on the presence of chlorinated hydrocarbon pesticides in precipitation collected at a South Carolina coastal site from 1976 through 1978. Toxaphene occurred most frequently and at levels in the range of less than 8 to 195 ng/L. Wingender and Williams (1984) have reported the occurrence of PCBs and polychlorinated terphenyls in precipitation at a station on Lake Huron. The concentration range in four precipitation samples collected from June to October 1980 was 3.8 to 1300 ng/L for the PCBs and 3.4 to 960 ng/L for the polychlorinated terphenyls.

No information on organic compounds in precipitation has been reported for sites in northern New England.

The National Atmospheric Deposition Program in Maine

There are four NADP sites located in Maine (fig. 1): Bridgton, in the southwestern part of the State; Acadia, a coastal location in eastern Maine; Greenville in the north-central part of the State; and Caribou in extreme northern Maine. Each site is instrumented with a recording rain gage and an automated wet-and dry-deposition collector. The collectors operate in a manner whereby the wet and dry components of the precipitation are collected separately in individual plastic buckets. Weekly wet-composite samples are removed from the collectors by field observers who determine the pH and conductivity before shipping the sample to the Central Analytical Laboratory in Illinois. The volume of precipitation collected and information from the rain gage are also recorded. Samples of dry deposition have been collected and analyzed on a bimonthly basis. Several problems associated with the collection of dry deposition have been identified, and the NADP has proposed that collection of these samples be discontinued until a more effective method is developed.

The composited wet-precipitation samples are analyzed at the Central Analytical Laboratory for sodium, potassium, magnesium, chloride, sulfate, nitrate, ammonia, phosphate, pH, and specific conductance.

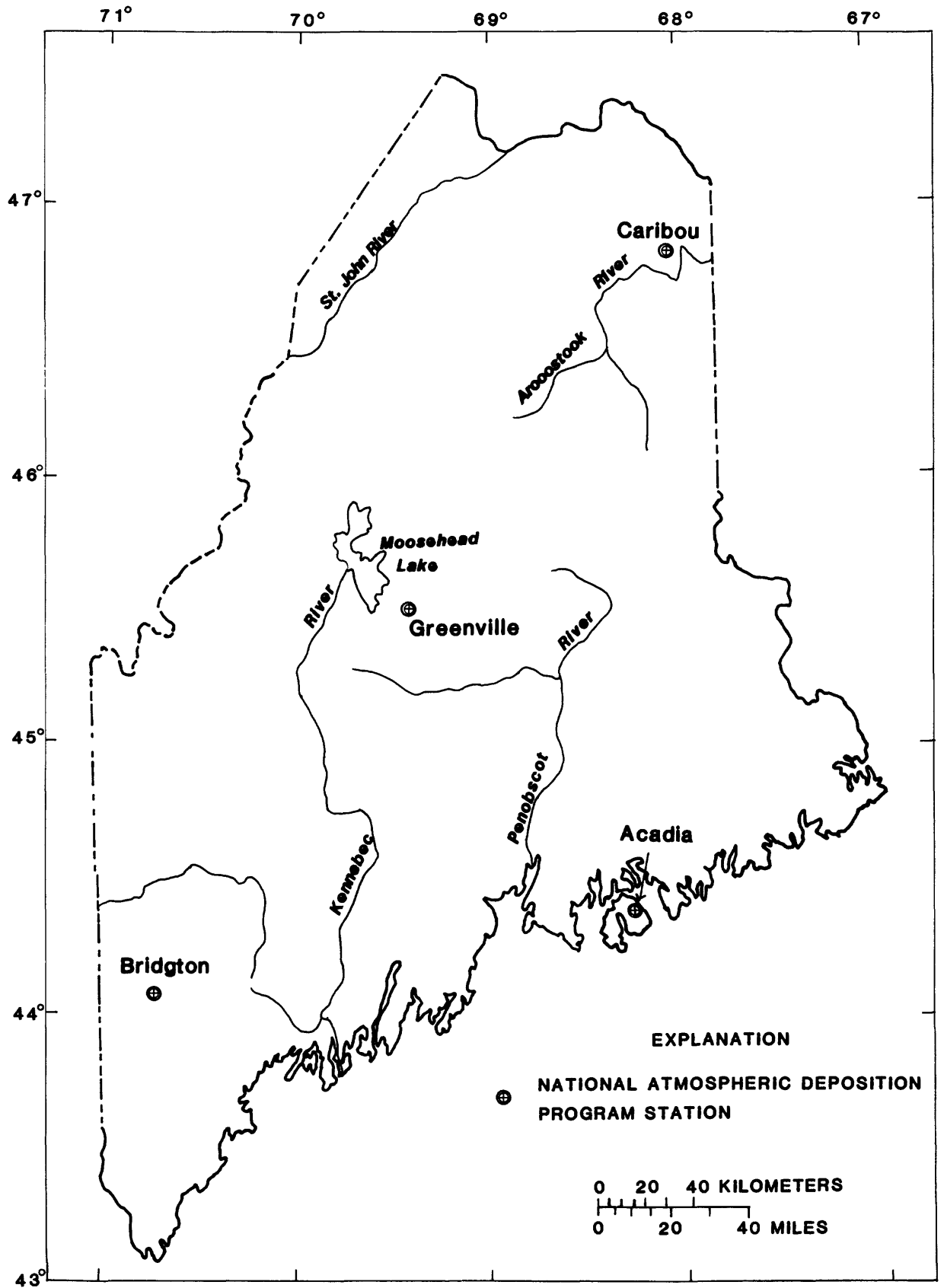


Figure 1.--Location of Maine National Atmospheric Deposition Program stations.

Acknowledgments

The authors wish to express gratitude to their field observer, Llew Wortman of Greenville, Maine, for his untiring efforts and concerned interest that were so important to the completion of this study. Thanks also go to Charles Elvin of the Maine Department of Environmental Protection for performing the metals analyses.

LOCATION AND DESCRIPTION OF STUDY AREA

The samplers used for the collection of precipitation samples for trace metals and organics analysis were installed adjacent to the NADP sampling site in Greenville, Maine. The location of the sampling site is shown in figure 2. Greenville (which includes Greenville Junction) has a population of 1,839 people (U.S. Bureau of the Census, 1984) located on the extreme southern shoreline of Moosehead Lake, the largest lake in Maine. The economic base of the town is the lumber industry and recreation.

The precipitation samplers were located approximately 8 kilometers northwest of the center of Greenville. The general location is rural and heavily forested. The surrounding terrain is fairly rugged. Within 5 kilometers of the site is the ridge of Little Squaw Mountain and the summit of Big Squaw Mountain (elevation 974 meters).

The climate is that of the temperate northern latitudes. Winters are long and snowy. The snow-free months are generally May through October. The mean annual temperature is near 4.4 °C. The mean annual precipitation is approximately 84 centimeters and is generally evenly distributed throughout the year.

The samplers were located on the grounds of a State Forest Service installation, which is at an elevation of approximately 320 m above sea level. They were positioned at the margin of a clearing which serves as access to buildings used for storage of vehicles and equipment. The samplers were about 9 m from the right bank of Upper Squaw Brook, a small perennial stream. It is heavily forested on the opposite bank. State Route 15, a paved road that serves as the only northwestward route out of Greenville, is located within 300 m of the samplers. The road handles seasonal recreational travel including summer tourists and winter skiers and is used by pulp trucks.

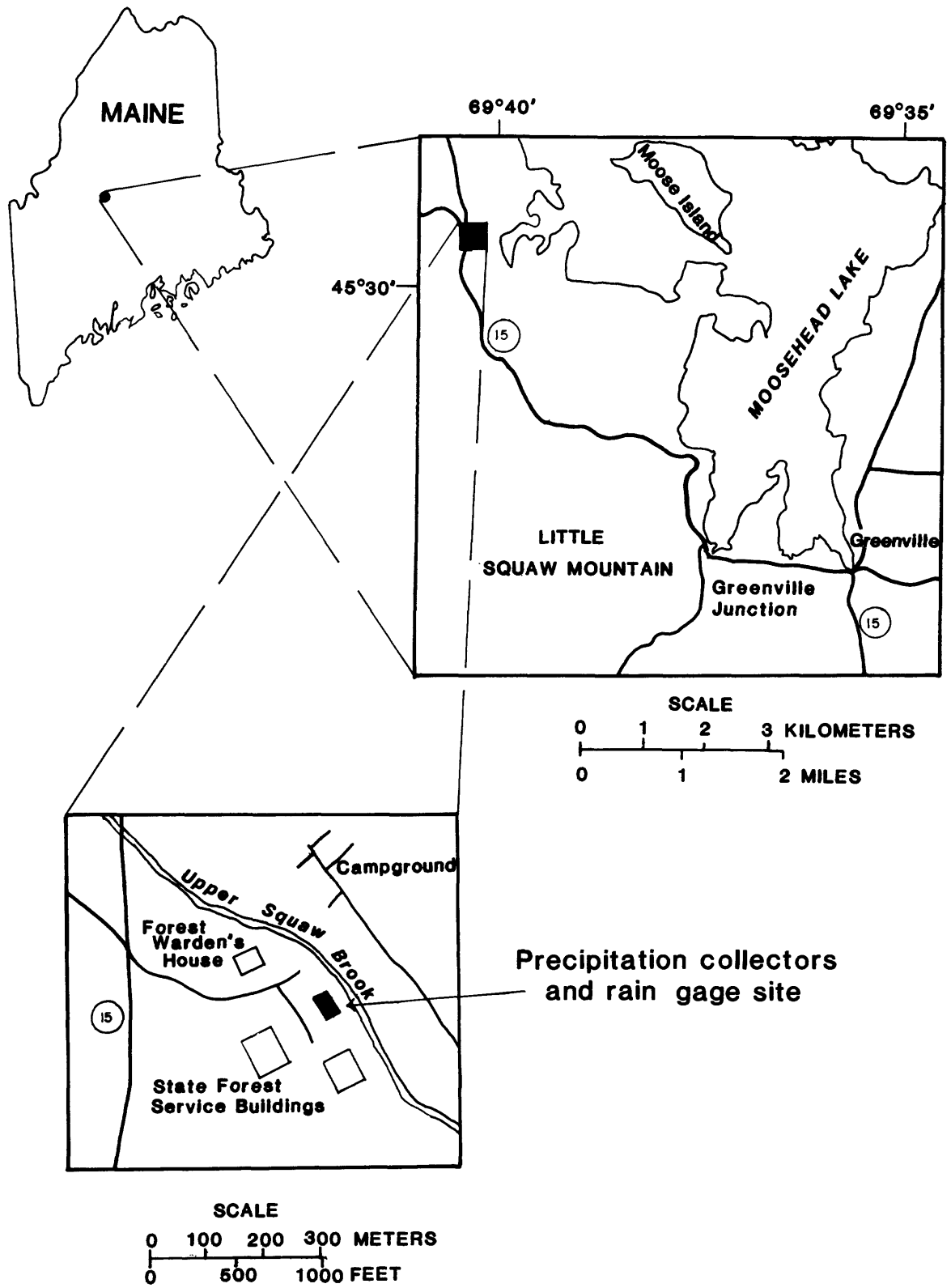


Figure 2.--Location of the precipitation collectors and rain gage.

METHODS OF STUDY

Design of Collectors

The primary precipitation samplers used in this study were Geotech¹ models 0600 and 0650 automated wet- and dry-deposition collectors, which are of a design similar to the NADP collectors. The samplers consist of two buckets that are mounted on an aluminum-frame platform. Also mounted on the frame is a motorized cover which can be moved from one bucket to the other. This cover is controlled by a moisture-sensing device such that during periods of precipitation the designated wet-only bucket is uncovered to receive a sample while the dry-bucket is covered to exclude precipitation. The sensor is heated and when it dries after the precipitation event, the wet-bucket is once again covered and the dry-bucket is uncovered. The heated sensor also serves to melt frozen precipitation which then activates the cover motor. In this study, dry-deposition samples were not collected.

Because of special precautions that must be followed to collect precipitation samples for trace metals and organic analysis, separate samplers were used to collect samples for each. The sampler used for trace metals was outfitted with the polyethylene buckets normally supplied with the unit. These buckets were cleaned with a hot hydrochloric-acid solution and rinsed with deionized water prior to installation in the sampler. The other sampler was outfitted with stainless steel buckets and was used only for collection of samples for organics analysis. The stainless steel buckets were rinsed with methylene chloride prior to each use.

During periods when the automated samplers were malfunctioning (approximately 60 percent of the time), the samplers were left in the wet-only mode continuously and collected a bulk sample (bulk samples are those which have components of both wet-fall and dry-fall). When it was apparent that an extended period of bulk collection would be required during seasons of high evaporation (approximately 20 percent of the time), bulk samples were collected using a simple glass funnel-type collector.

The NADP sampling equipment was located next to the metals and organics samplers. It consisted of an automated wet- and dry-deposition sampler and a weighing bucket rain gage with a 7-day recorder. Data from the rain gage were used to provide a record of precipitation at the site.

The sampling site is shown in figure 3. The organics and metals sampler are to the right in the figure and the NADP sampler and rain gage are to the left.

¹ The use of brand names in this report is for identification purposes only and does not imply endorsement by the U.S. Geological Survey or Maine Department of Environmental Protection.



Figure 3.—The precipitation sampling site showing (left to right) NADP (National Atmospheric Deposition Program) rain gage, NADP sampler, organics sampler and metals sampler.

Field Methods

Sample collection protocol was designed to coincide with the NADP procedure. The field observer for this study (also the NADP observer) visited the sampling site every Tuesday to remove the sample buckets and replace them with clean buckets. The sample buckets were covered and brought to a wastewater-treatment plant laboratory for processing.

The buckets were weighed to determine sample volume. An aliquot of sample was removed from the plastic (metals) bucket and used to determine pH and specific conductance. The remainder was transferred to acid-rinsed plastic bottles and preserved with highly purified nitric acid. The sample from the stainless steel (organics) bucket was transferred to 1-liter amber glass bottles which had been cleaned with laboratory detergent, triple rinsed with distilled deionized water, and baked in a muffle furnace at 450 °C for 2 hours. The bottle caps were lined with precleaned teflon. All samples were placed in a cooler with ice-packs and shipped to the laboratory of the Maine Department of Environmental Protection for analysis. Samples were received within 24 hours of shipment and stored at 4 °C until analysis.

Selection of Analytes

The metals and organic chemicals selected for monitoring in this study were chosen on the basis of their potential for human, plant, or animal toxicity, reported occurrence in atmospheric deposition, and availability of sensitive analytical chemical techniques which could be applied routinely. The metals included cadmium, chromium, copper, lead, mercury, nickel, and zinc. The organic chemicals included the chlorinated hydrocarbon pesticides and pthalate ester plasticizers identified as priority pollutants (U.S. Environmental Protection Agency, 1982) and the PCBs.

Laboratory Methods

Unfiltered precipitation samples were analyzed for "total recoverable metals" using atomic-absorption techniques (U.S. Environmental Protection Agency, 1983). Graphite furnace techniques were used for lead, copper, and nickel, flame techniques for zinc, cadmium, and chromium, and the manual cold vapor technique for mercury. Samples were prepared for metals analysis by digestion of an aliquot in either boiling nitric acid or boiling nitric acid/hydrochloric acid. The mercury analyses were performed on separate aliquots. All sample preparation was performed as specified in the analytical protocols cited (U.S. Environmental Protection Agency, 1983).

Reagent blanks were analyzed with each set of samples. "Blind" field blanks were routinely analyzed for the acid-washed buckets. The field blanks were prepared by randomly selecting a bucket and rinsing with three 100 ml aliquots of distilled deionized water. The rinsate was then transferred to a field sample bottle, acidified with nitric acid, and submitted "blind" to the laboratory for analysis. Zinc was detected infrequently in the field blanks at the 1 to 5 µg/L range. None of the other metals were detected in any of the blanks.

The unfiltered precipitation samples were prepared for "organics" analysis by sequential extraction with three 60 ml aliquots of dichloromethane in a separatory funnel. The solvents used in the preparation of these samples were Baker Pesticide Residue Analysis grade. The entire composite sample, up to a maximum of 2 liters, was extracted. The extract was concentrated to 10 ml using Kuderna-Danish apparatus. The solvent was then exchanged by the addition of 50 ml of n-hexane and concentrated to 1 ml using macro and micro Kuderna-Danish apparatus.

Laboratory reagent blanks were extracted and analyzed for each set of sample extracts prepared. Additionally, "field blanks" were prepared on three occasions by rinsing sample buckets with three 200 ml aliquots of "blank" water. The rinsate was then submitted to the laboratory as a sample.

Prior to use, all glassware was carefully rinsed with methanol followed by dichloroethane and n-hexane. The final 10 ml of n-hexane rinse was collected and analyzed by gas chromatography with electron-capture detection. The column used was a 1.83 m long, 2 mm I.D. (inside diameter) glass column packed with 180/100 Chromasorb WHP coated with 1.5 percent OV-17 and 1.95 percent QF1 (Supelco). The column was operated at 200 °C isothermally with a nitrogen carrier gas flow rate of 30 ml per minute. The instrument was calibrated such that 30 picograms of α-BHC gave a full scale deflection. Glassware was not used unless the final 10 mm of n-hexane rinse gave no peaks. All glassware used was reserved exclusively for this analysis.

The concentrated sample extracts and blanks were analyzed by HRGC-ECD (high resolution gas chromatography with electron capture detection) using a Hewlett-Packard Model 5880 GC (gas chromatograph). The chromatograph was equipped with an Ultra no. 2 (Hewlett-Packard) fused silica capillary column (25 m X 0.33 mm I.D.). The GC oven was temperature programmed from 50 °C to 100 °C at 25 °C per minute with an initial hold time of 1 minute and then from 100 °C to 250 °C at 4 °C per minute with a final hold time of 10 minutes. The helium carrier gas flow rate was set at 5 ml per minute. A "splitless" injection technique was used. Under the described conditions, baseline resolution of the most difficult to separate pair of the common chlorinated hydrocarbon pesticides, dieldrin and pp'-DDE, could be achieved.

Prior to performing any HRGC-ECD analysis, 50 picograms of pp'-DDT was injected. If the retention time matched previously determined values within ± 0.05 minutes and decomposition was less than 5 percent, analysis of samples proceeded. Otherwise, corrective action was taken.

Tentative identification and quantification of analytes in the extracts was made by comparing retention times and response factors to those of standards. For confirmation, all sample extracts were pooled and concentrated to 1 ml. This concentrate was analyzed by high-resolution gas-chromatography mass spectrometry, using a Hewlett-Packard Model 5985 system. The gas-chromatographic conditions used were identical to those described previously for the HRGC-ECD analysis. The mass spectrometer was operated in the selected ion monitoring mode. The ions monitored included 183, 181, and 109. These ions are principal fragments produced when hexachlorocyclohexanes are ionized by electron impact at 70 EV (U.S. Environmental Protection Agency, 1982).

CHEMICAL QUALITY OF PRECIPITATION

Major Constituents

Data from the Greenville NADP site indicates the general chemical quality of precipitation in the vicinity of the trace-metals and organic-compounds samplers. Volume-weighted mean concentrations of the chemical constituents, which are included in the NADP analysis (except phosphate which was only rarely detected), are presented in table 1. The sampling period for the computations coincides with the sampling period for the collection of trace metals and organics for this study. The data are consistent with those reported for other NADP sites in the northeastern United States and may be considered to be included in the general category of "acidified precipitation." The low pH (volume-weighted mean 4.6) and the proportionately high concentrations of sulfate and nitrate indicate that atmospheric pollutants are a dominant factor influencing the general chemical quality of precipitation at this site.

The mean concentrations reported in table 1 were converted to microequivalents and are presented in table 2. An activity coefficient of 1 was assumed for the conversion of pH to hydrogen-ion concentration. The sum of the microequivalent concentrations of the cations is equal to the sum for the anions, indicating a good chemical balance. The hydrogen ion is the dominant cation, accounting for 58 percent of the total cation equivalents. Sulfate and nitrate are the dominant anions, accounting for 63 and 28 percent, respectively, of the total anion equivalents. The predominance of these three ions indicates that the Greenville precipitation is influenced by sulfuric and nitric acids. The hydrogen-ion concentration was most strongly correlated with the sum of the concentrations of nitrate and sulfate (r^2 or coefficient of determination = 0.67). The correlations between the hydrogen-ion concentrations and the nitrate ($r^2 = 0.51$) and sulfate ($r^2 = 0.50$) concentrations were not as great. These relations are presented in figures 4 through 6.

Table 1.--Volume-weighted mean concentrations of dissolved chemical constituents for National Atmospheric Deposition Program precipitation samples at Greenville, Maine

(Sampling period: Feb. 16, 1982, through Jan. 24, 1984;
sample values not detected set equal to zero.)

CONCENTRATIONS (milligrams per liter)									
Number of observations	Field pH (units)	Calcium	Magnesium	Potassium	Sodium	Ammonia	Nitrate	Chlorine	Sulfate
87	4.6	0.072	0.022	0.026	0.084	0.152	0.72	0.13	1.32

Table 2.--Cation-anion balance of volume-weighted mean concentrations of dissolved chemical constituents for National Atmospheric Deposition Program precipitation samples at Greenville, Maine

(Sampling period: Feb. 16, 1982, through Jan. 24, 1984;
sample values not detected set equal to zero.)

CONCENTRATIONS (microequivalents per liter)										
Hydrogen ion	Calcium	Magnesium	Potassium	Sodium	Ammonia	Nitrate	Chlorine	Sulfate	Cation sum	Anion Sum
28	3.6	1.8	0.7	3.7	8.4	12	3.7	27	43	43

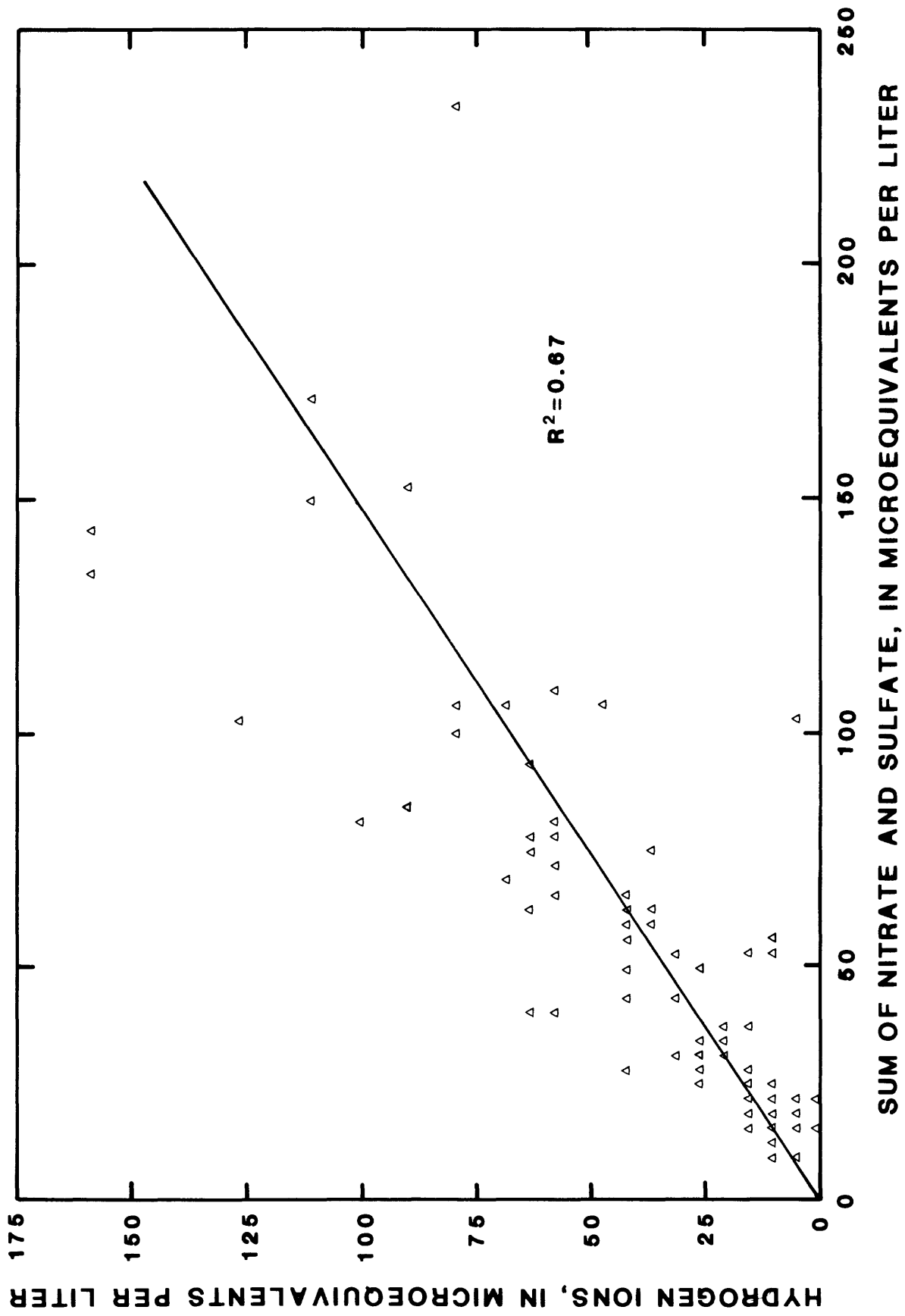


Figure 4.—Relation of the hydrogen ion to the sum of nitrate and sulfate concentrations for National Atmospheric Deposition Program samples.

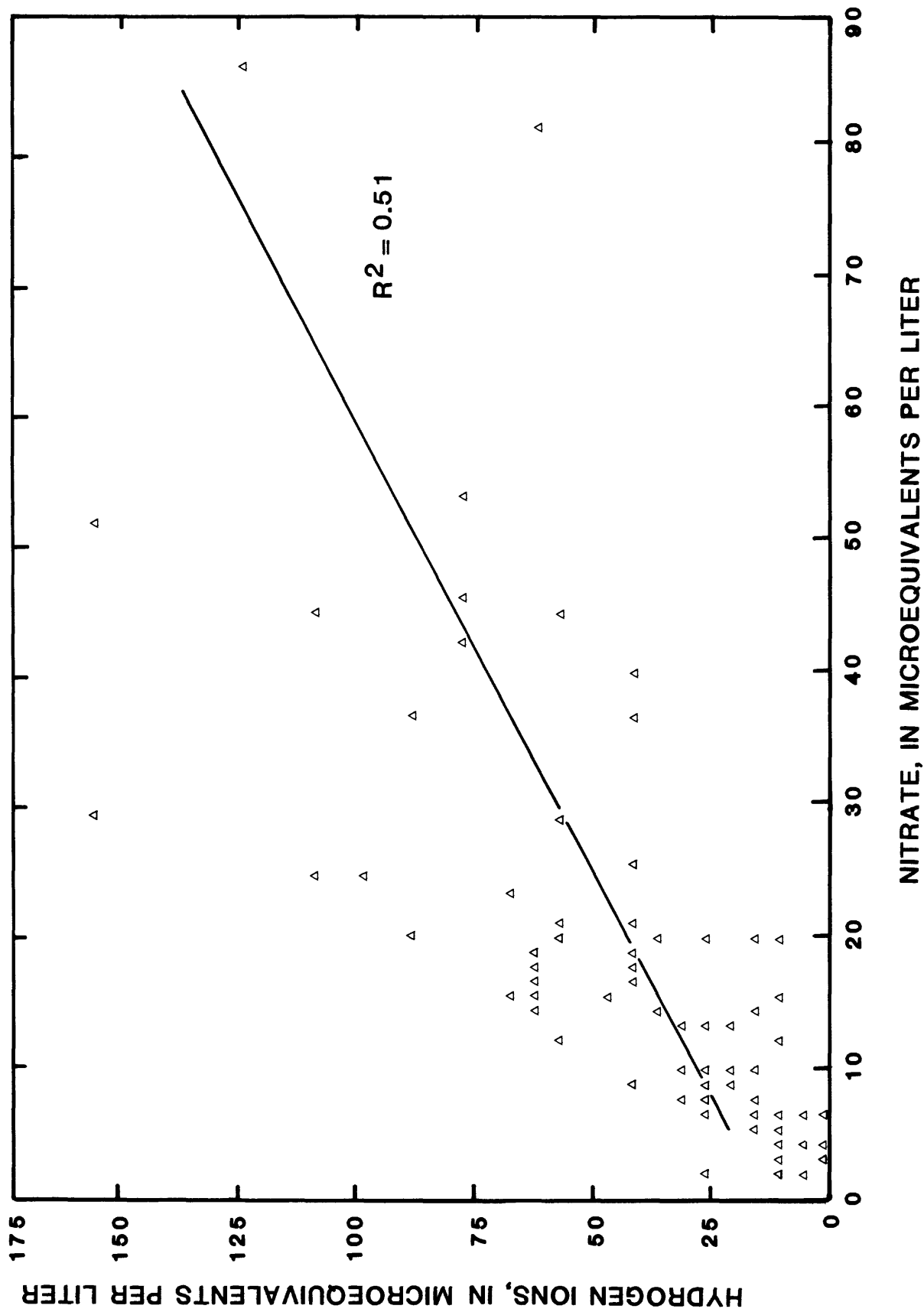


Figure 5.--Relation of the hydrogen ion to the nitrate concentration for National Atmospheric Deposition Program samples.

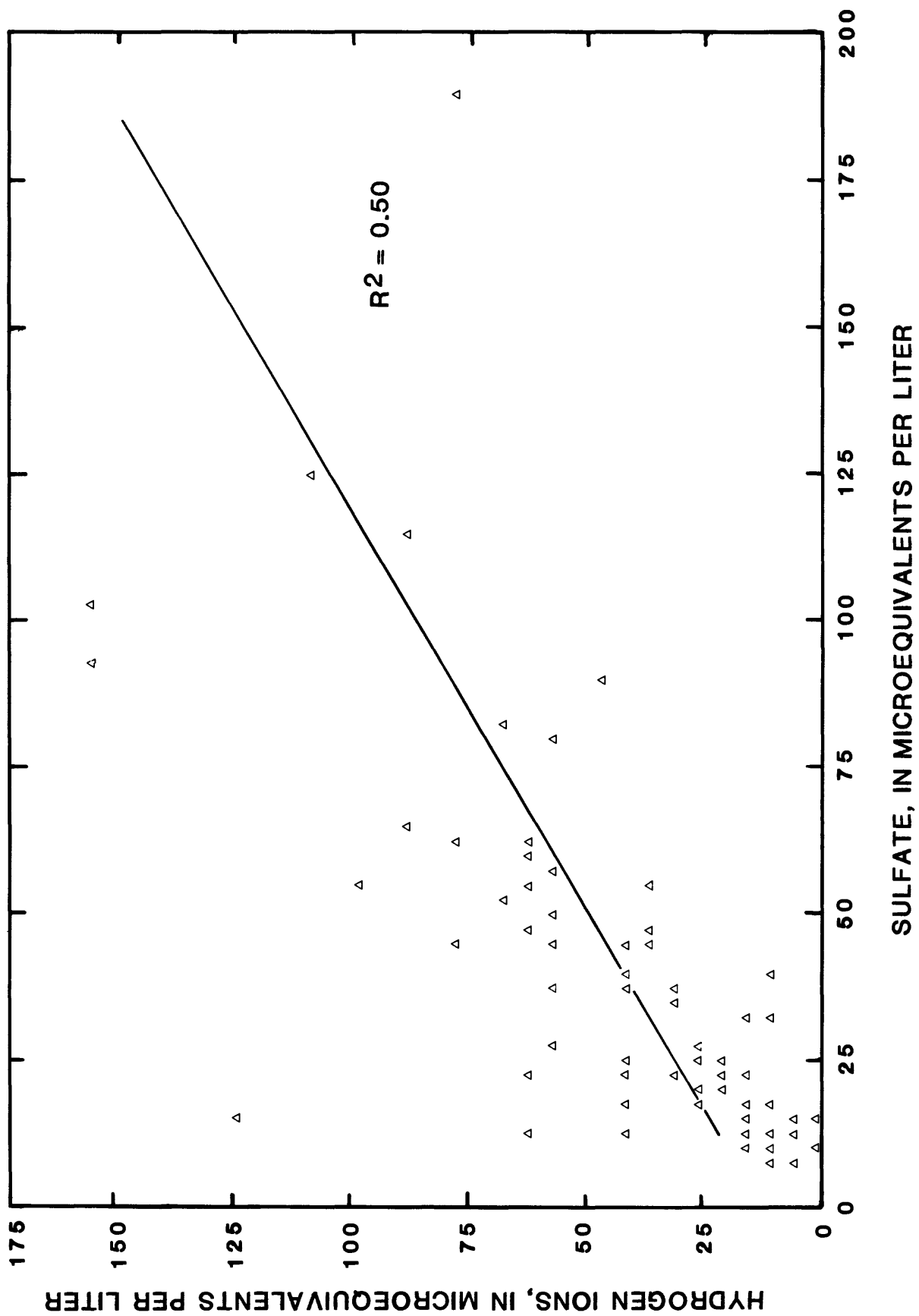


Figure 6.--Relation of the ion to the sulfate concentration for National Atmospheric Deposition Program samples.

In contrast, Gorham and others (1984) found that at the NADP sites east of longitude 95° W for 1980 and 1981, the hydrogen-ion concentration was more strongly correlated with the sulfate concentration alone ($r^2 = 0.85$) than with the sum of the nitrate and sulfate ($r^2 = 0.73$) or the nitrate concentrations alone ($r^2 = 0.63$). This difference suggests that nitric acid is a more significant component of precipitation acidity at the Greenville site than at many other NADP sites in the eastern United States. This finding is consistent with the data presented by Galloway and Likens (1981) for the Hubbard Brook Experimental Forest for the period 1964 through 1979 which showed that the relative contribution of nitric acid to precipitation acidity increased with time.

The ammonium and calcium in the precipitation are presumably derived from neutralization of precipitation acidity by atmospheric ammonia (NH_3) and by calcareous particles from soil and other sources. The correlation between the sum of the calcium, ammonium, and hydrogen-ion concentrations and the sum of the concentrations of sulfate and nitrate ($r^2 = 0.71$) supports this contention. The relation is shown in figure 7.

Sea spray is the likely source of the sodium and chloride in the Greenville precipitation. Sodium and chloride concentrations, however, are relatively low in comparison to data from coastal locations. Sea spray probably does not contribute a significant amount of sulfate. In seawater, the concentration of sulfate has been found to be approximately 14 percent of the concentration of chloride, by weight (Goldberg, 1963). Assuming that all of the chloride in Greenville precipitation is derived from sea spray, the contribution of sulfate from sea spray at this site is less than 1 percent of the total sulfate concentration.

The volume of precipitation sample multiplied by the concentration of a constituent yields the deposition of that constituent for the sample-collection interval. The annual deposition rates for sulfate, nitrate, and the hydrogen ion at Greenville were calculated and are 14,000, 7,600, and 310 (g/ha)/yr, respectively.

The calculated wet deposition of sulfate at Greenville is less than the target loading maximum of 20 (kg/ha)/yr (kilograms per hectare per year), proposed in several acid-rain control strategies (United States-Canada Memorandum of Intent on Transboundary Air Pollution, 1983, as cited by Gorham and others, 1984). The volume-weighted mean pH at Greenville of 4.6, however, has been reported as a damage threshold for sensitive aquatic ecosystems (Henriksen, 1979; Evans and others, 1981). A factor that may account for this discrepancy is that nitric acid, rather than sulfuric acid, may be a more significant component of precipitation acidity at Greenville than in the study areas used to establish the target loadings. Also, year to year variation in sulfate deposition was not accounted for at this site.

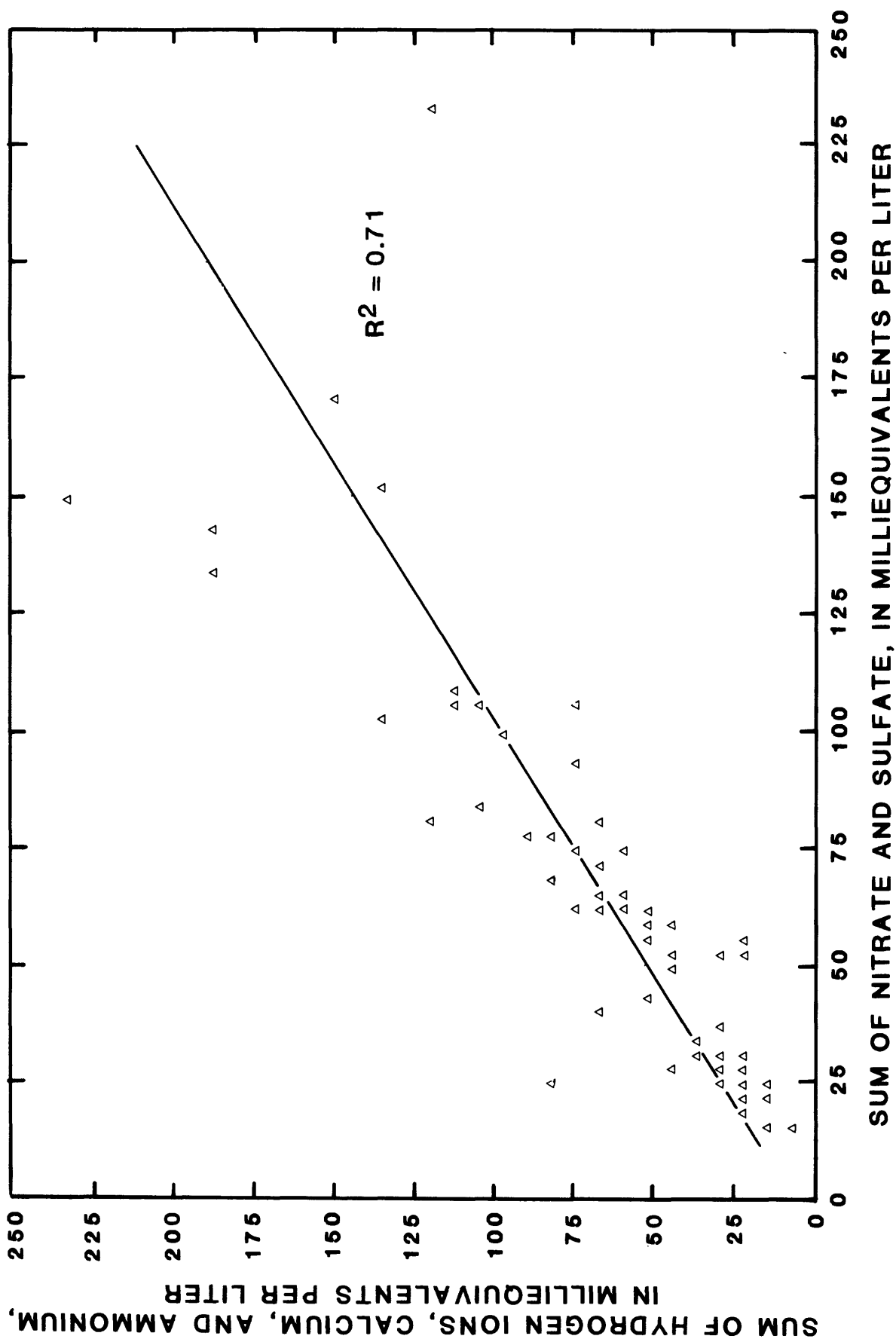


Figure 7.--Relation of the sum of hydrogen ion, calcium, and ammonium to the sum of nitrate and sulfate concentrations for National Atmospheric Deposition Program samples.

Trace Metals

Table 12 (at back of report) describes the wet- and bulk-deposition samples that were collected for trace-metals analysis. The majority of the samples were 1-week composites. During weeks when only a trace volume of sample was collected (insufficient for sample analysis), the bucket was left in the sampler for an additional week. By doing this, low-volume samples were not excluded from calculations of long-term volume-weighted mean concentrations or deposition. In table 12, precipitation volume represents the total precipitation for the composite period as measured by the rain gage and calculated to represent an equivalent volume that would have been collected in one of the sample buckets. The sample type indicates wet-only for a sample collected during a period when the automated sampler was functioning and bulk for periods when it was not operating and left in the "open-bucket" position or when another bulk sampler was used.

Ideally, a study of this kind should be consistent with respect to the type of samples collected. However, as previously discussed, problems with the automated sampler necessitated the collection of wet-only samples during part of the collection period and bulk samples for the remainder. Because wet-only samples exclude dry fallout, there may be some qualitative and quantitative discrepancies between the two types of samples collected in this study. In an attempt to determine the magnitude of the possible discrepancy, a correlation analysis was performed between the specific conductance values reported for the NADP samples (which were all wet-only) and those determined for the two types of trace-metals samples. The correlation coefficient during periods when the trace-metals sampler was collecting wet-only samples is 0.734. During periods when bulk samples were collected, the correlation coefficient is 0.733. The agreement between these two values suggests that any differences between the bulk and wet-only samples is minor.

Table 13 (at back of report) presents the results of the trace-metals analysis for each of the composite periods. The detection limit is entered in the table for those constituents which were not detected in a given sample. Chromium was not detected in any of the precipitation samples. Mercury and cadmium were rarely detected; but, when detected, they were found at surprisingly high concentrations, suggesting contamination. The mercury values also are suspect because elemental and organic forms of mercury are volatile, and the trapping efficiency of the collection apparatus was probably poor for this metal.

Nickel was detected in 26 percent of the samples. Its concentration was generally found to be close to the detection limit. Lead, copper, and zinc were consistently detected in the samples. In three samples, the concentration of lead exceeded the recommended limit for domestic water supply (U.S. Environmental Protection Agency, 1986) of 50 $\mu\text{g/L}$. In one sample, the concentration of copper was nearly one-half the recommended limit of 1,000 $\mu\text{g/L}$.

A statistical summary of the concentrations of lead, copper, and zinc in the precipitation samples is presented in table 3. In computing statistics for such data, an analyst is given the problem of how to treat values below the detection limit of the respective analytical procedures. Such values may either be considered to be zero or may be set equal to the detection limit. The difference between these two computational methods will increase as the proportion of values not detected increases. In table 3, statistics for the sample concentration of dissolved lead, copper, and zinc are presented for both methods and, thus, represent a range of values that should bracket the probable value.

The volume-weighted mean concentration of lead in the Greenville samples is in the range 3.6 to 3.7 $\mu\text{g/L}$. The median of the values is 3.0. The mean concentration of copper is 5.1 to 5.3 $\mu\text{g/L}$ and the median is 1.0. For zinc, the mean concentration is from 13 to 14 $\mu\text{g/L}$ with a median of 12. The skew coefficients for all three metals are highly positive. These coefficients and the medians being less than the means demonstrate that the values of concentration for these metals are all skewed populations with the majority of the values residing in the low range.

The potential impact of a precipitation-borne contaminant on a given area is a function of both the concentration of the contaminant and the volume of precipitation which falls in that area. A commonly used parameter which accounts for both concentration and volume is deposition, the units of which are mass per unit area. Deposition is the total amount of a constituent that is deposited in a given area during a given time period.

Central to the computation of deposition is a measure of precipitation volume. In our study, there were two sources of information on precipitation volume -- the rain gage and the sampling bucket. Generally, an approved rain gage is taken as the standard of measurement of precipitation quantity. Using either of these for the computation of deposition will generate varying degrees of uncertainty for different reasons.

If the precipitation sampler were 100 percent efficient (no loss from evaporation or misadjustment/malfunction of the automated sampler), either precipitation volume from the rain gage or the precipitation sampler would yield identical results for computing deposition. In our study, the sampler had an average efficiency of 91 percent when compared to the rain gage. Figure 8 is a graphical comparison of the composite sample volume to the given "standard" of precipitation volume as measured by the rain gage.

In computing deposition, using sample volume (as opposed to precipitation volume measured by a rain gage) will give an inaccurate value if the sampler is not 100 percent efficient and if the constituent is evenly distributed throughout the event. If the sampler is less than 100 percent efficient, some of the total constituent load will be unaccounted for and the deposition will be underestimated. This method would only be accurate in such a case if the constituent is not evenly distributed throughout the event but the sampler collects that portion of the event that includes all of the constituent load. An advantage of this method is that it is not sensitive to changes in constituent concentration because of evaporation from the sampler.

Table 3.--Statistical summary of concentrations of dissolved lead, copper, and zinc in precipitation samples
(All concentrations in micrograms per liter.)

Constituent	Number of observations	Number of observations above detection limit	Range	Concentrations below detection limit set equal to zero					
				Volume-weighted mean concentration	Median	Skew	Volume-weighted mean concentration	Median	Skew
Lead	93	87	< 0.5 - 80	3.6	3.0	4.6	3.7	3.0	4.7
Copper	89	63	< .2 - 430	5.1	1.0	9.0	5.3	1.0	9.0
Zinc	90	79	< 5 - 280	13	12	4.7	14	12	4.8

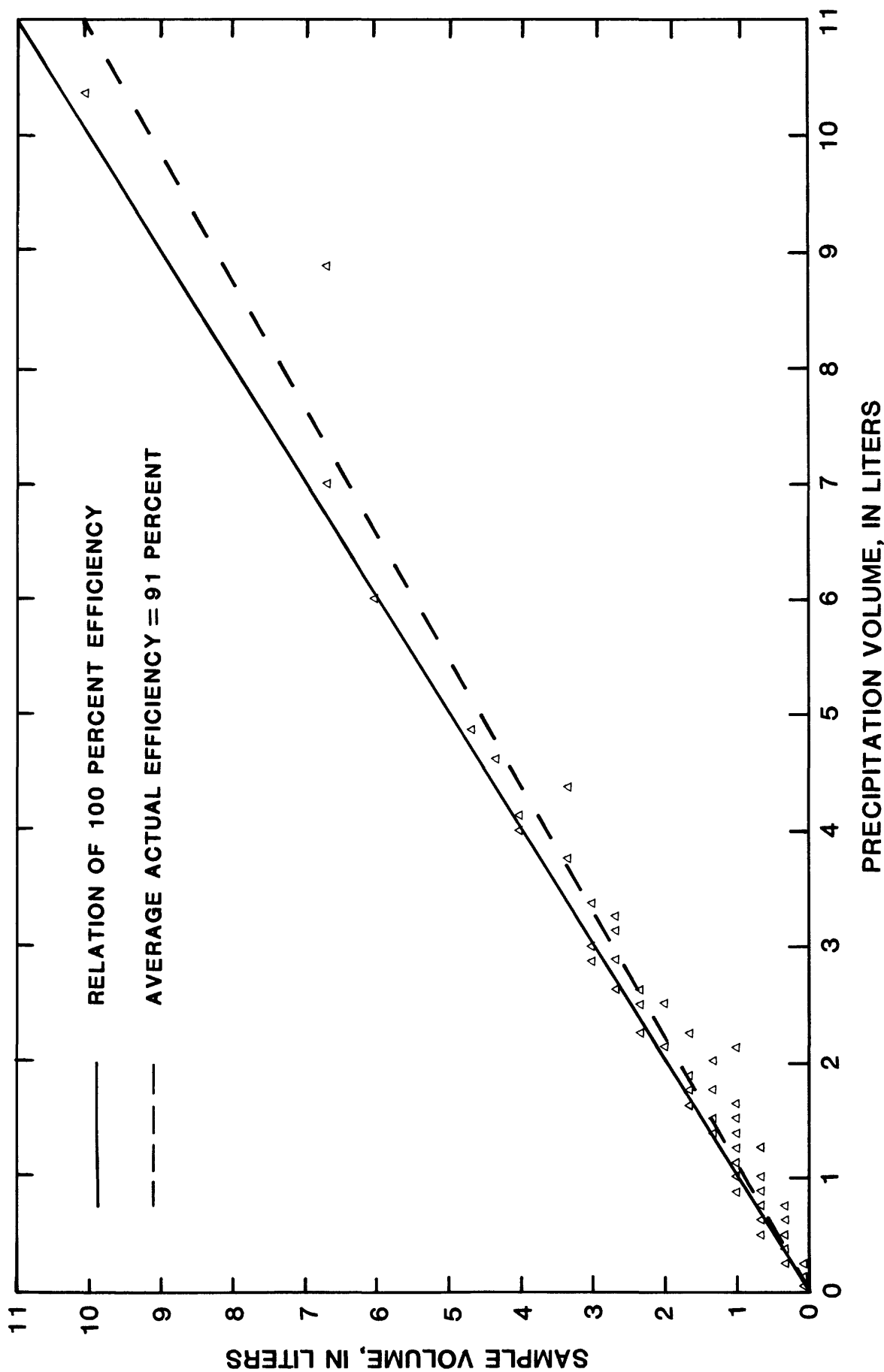


Figure 8.--Comparison of precipitation quantity recorded by rain gage to sample volume collected by trace-metal collector.

Using precipitation volume in the calculation assumes that the constituent is uniformly distributed throughout the event. In this way, even if some of the event is "missed" by the sampler, the calculation of deposition will not be affected. If the constituent is not evenly distributed throughout the event and the sampler collects that portion of the event that includes all of the constituent but excludes other portions, this method will overestimate the deposition. This method is sensitive to changes in constituent concentration because of sample evaporation.

As previously discussed, computations involving constituent concentrations also are influenced by the manner in which analytical results below the detection limit are interpreted. This results in four possible methods of computing deposition depending on whether sample volume or precipitation volume is used and if concentrations not detected are set to zero or are set to the detection limit of the analysis. Table 4 illustrates the possible discrepancy between these various computational methods. In the example, computed mean yearly deposition for the lead data varies as much as 21 percent, depending on the method of computation.

Statistics on the deposition of lead, copper, and zinc are presented in table 5. Statistics on the constituents were computed using the four methods previously discussed. Maximum lead deposition for a composite period was 1,000 $\mu\text{g}/\text{m}^2$ (micrograms per square meter) and the mean deposition is within 95 to 121 $\mu\text{g}/\text{m}^2$. The mean annual deposition was computed as the mean daily deposition multiplied by 365.25 days and is within 45 to 58 (g/ha)/yr. Copper deposition had a maximum value of 7,170 $\mu\text{g}/\text{m}^2$ and a mean from 140 to 170 $\mu\text{g}/\text{m}^2$. The mean yearly copper deposition was from 66 to 81 (g/ha)/yr. The maximum zinc deposition was 3,310 $\mu\text{g}/\text{m}^2$ with a mean of from 366 to 462 $\mu\text{g}/\text{m}^2$. Yearly mean deposition was from 173 to 221 (g/ha)/yr. As was the case with sample concentrations, the deposition values are positively skewed and the median values are lower than the mean values.

Figures 9 through 11 illustrate the distribution of copper, lead, and zinc deposition, respectively. The deposition amount used in these illustrations, as well as all other illustrations in this report, were computed using precipitation volume. Samples with no detectable levels of copper, lead, or zinc were excluded. For comparison, figure 12 is a plot of the precipitation quantity that was recorded during each of the sample composite periods.

The distribution of copper shows that for three samples collected in 1983, the concentration of copper was much greater than that of the other samples (10 to 70 times greater than the mean). It is possible that this is the result of sample contamination, but no evidence of contamination or analytical error could be found. If the results are representative, they indicate that copper deposition is extremely episodic.

Table 4.--Difference between methods of calculation of mean yearly lead deposition. Percentage is difference between value calculated using precipitation volume and concentrations below detection limit set to detection limit.

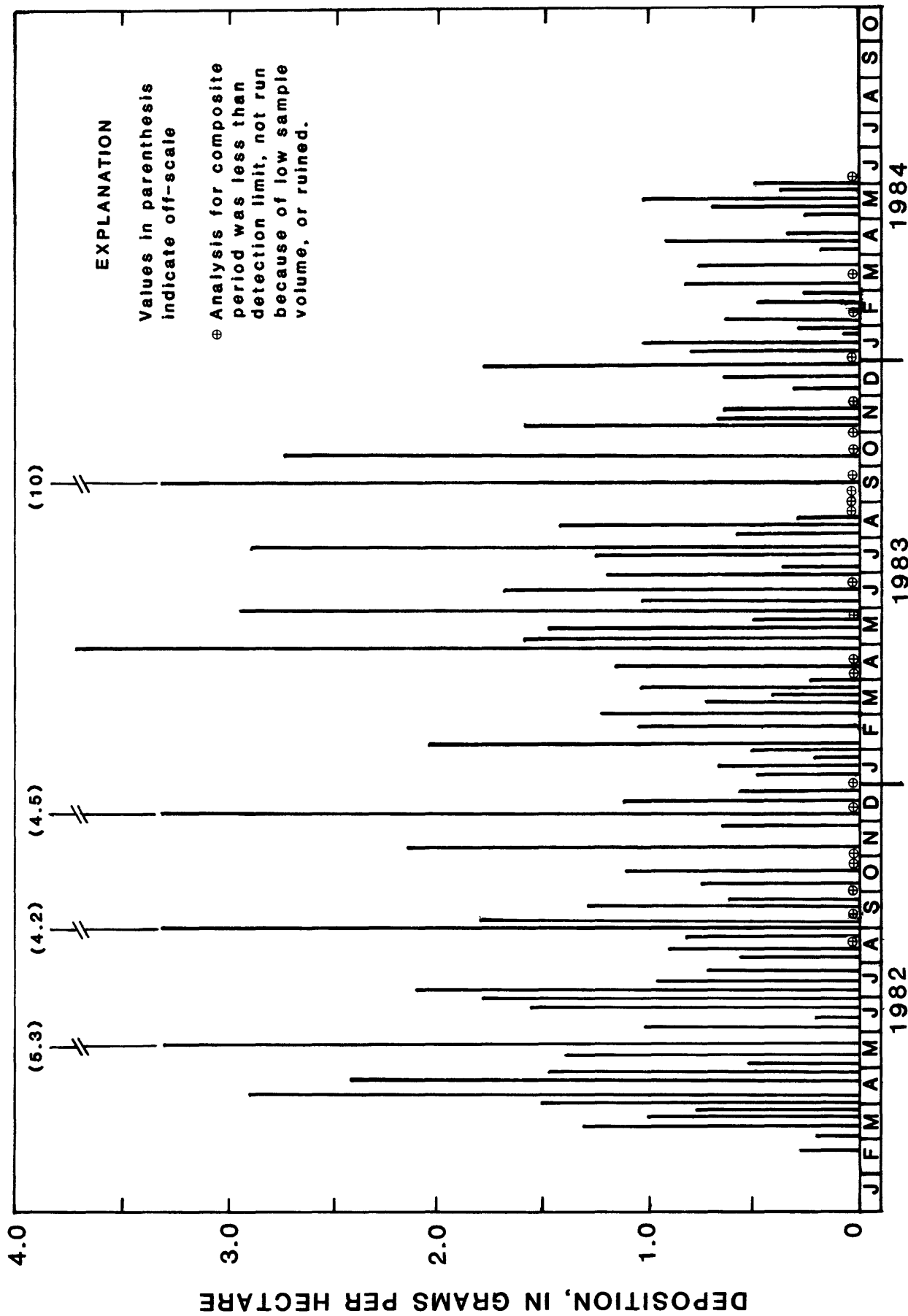
(All data are reported in grams per hectare per year)

	Calculated using precipitation volume	Calculated using sample volume
Concentrations below detection limit set to detection limit	58(0 percent)	47(-18 percent)
Concentrations below detection limit set equal to zero	56(-5.3 percent)	47(-21 percent)

Table 5.--Summary of deposition of lead, copper and zinc in precipitation samples

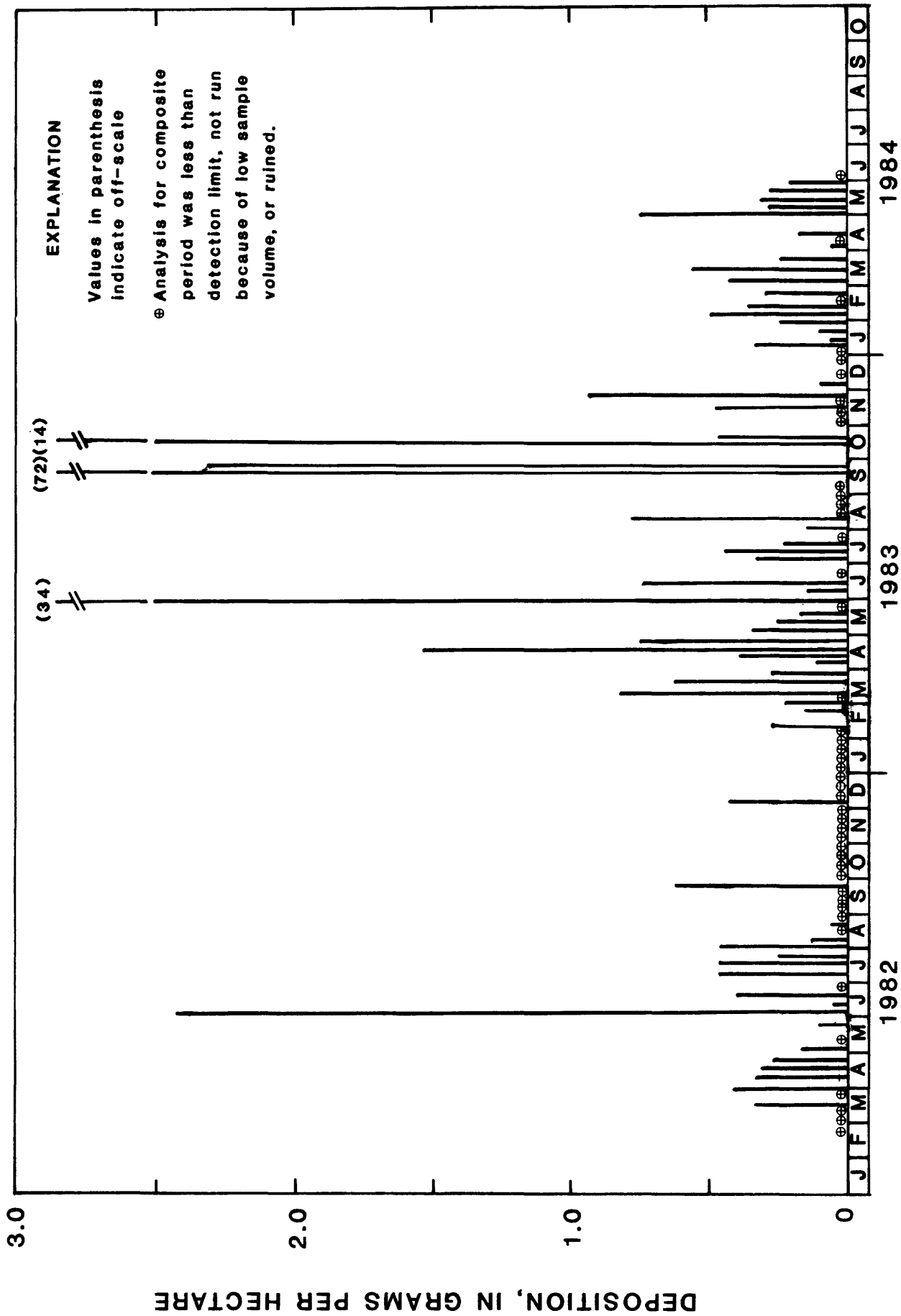
[Values in parenthesis are computed based upon sample volume;
all others are computed based upon precipitation volume;
ug/m is micrograms per square meter, g/ha is grams
per hectare, and (g/ha)/yr is grams per hectare per year]

Constituent	Number of obser- vations	Range ($\mu\text{g}/\text{m}^2$)	Composite-period deposition				Skew	Yearly deposition (mean) (g/ha)/yr
			Mean	Median	Mean	Median		
			$\mu\text{g}/\text{m}^2$		g/ha			
Concentrations below detection limit are set equal to zero								
Lead	92 (93)	0 - 1,000 (0 - 906)	116 (95)	76 (66)	1.16 (0.95)	0.76 (0.66)	3.7 (4.2)	56 (45)
Copper	88 (89)	0 - 7,170 (0 - 6,490)	165 (140)	23 (16)	1.65 (1.40)	0.23 (0.16)	7.3 (7.7)	79 (66)
Zinc	89 (90)	0 - 3,310 (0 - 3,270)	438 (366)	311 (236)	4.38 (3.66)	3.11 (2.36)	3.0 (3.5)	210 (173)
Concentrations below detection limit are set equal to the detection limit of the constituent								
Lead	92 (93)	8 - 1,000 (3 - 906)	121 (99)	76 (66)	1.22 (0.99)	0.76 (0.66)	3.8 (4.4)	58 (47)
Copper	88 (89)	4 - 7,170 (1 - 6,490)	170 (145)	23 (16)	1.70 (1.45)	0.23 (0.16)	7.3 (7.7)	81 (68)
Zinc	89 (90)	40 - 3,310 (13 - 3,270)	462 (392)	311 (236)	4.62 (3.92)	3.11 (2.36)	3.2 (3.7)	221 (186)



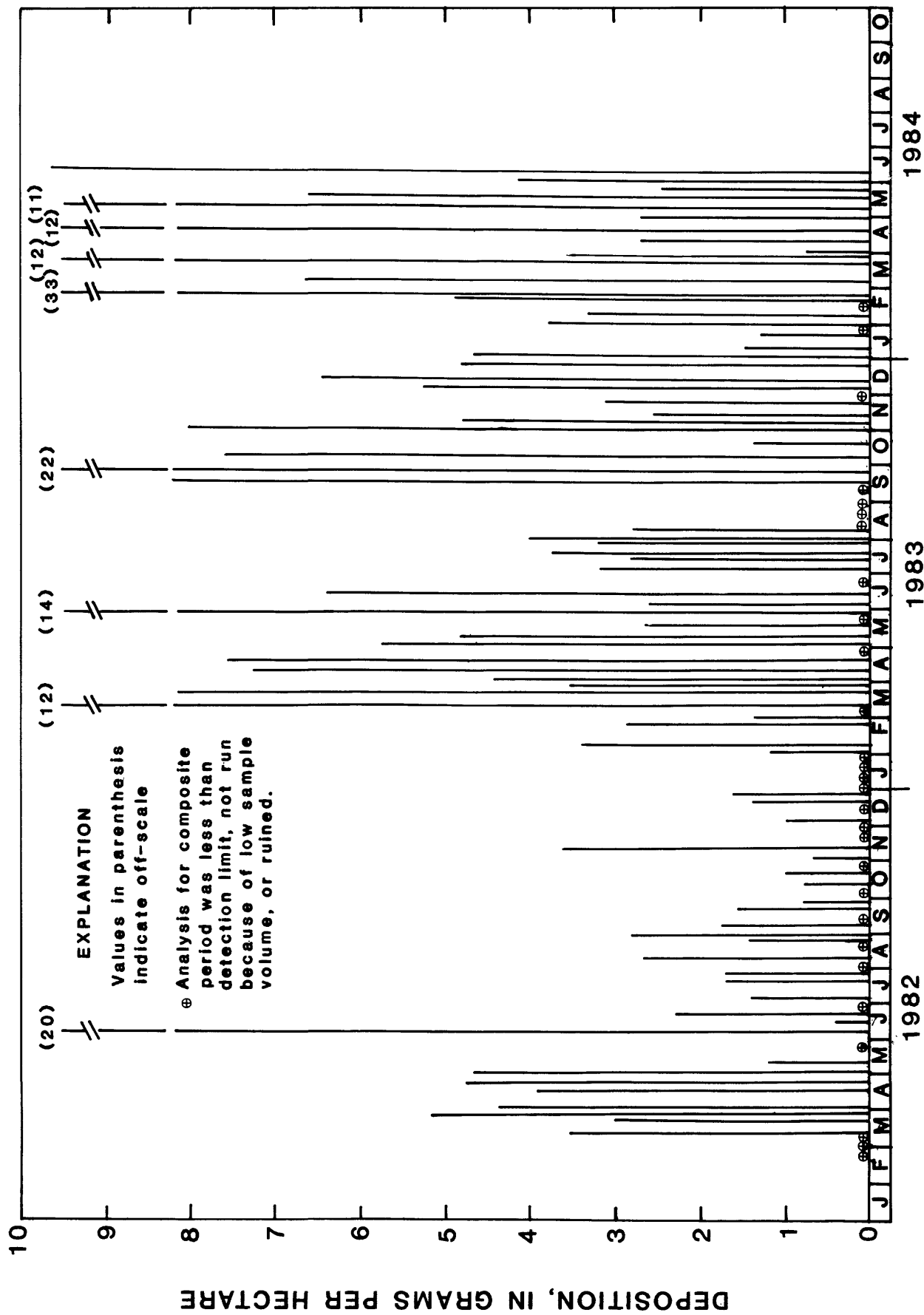
DATA PLOTTED ON LAST DAY OF COMPOSITE SAMPLING PERIOD

Figure 9.--Distribution of lead deposition.



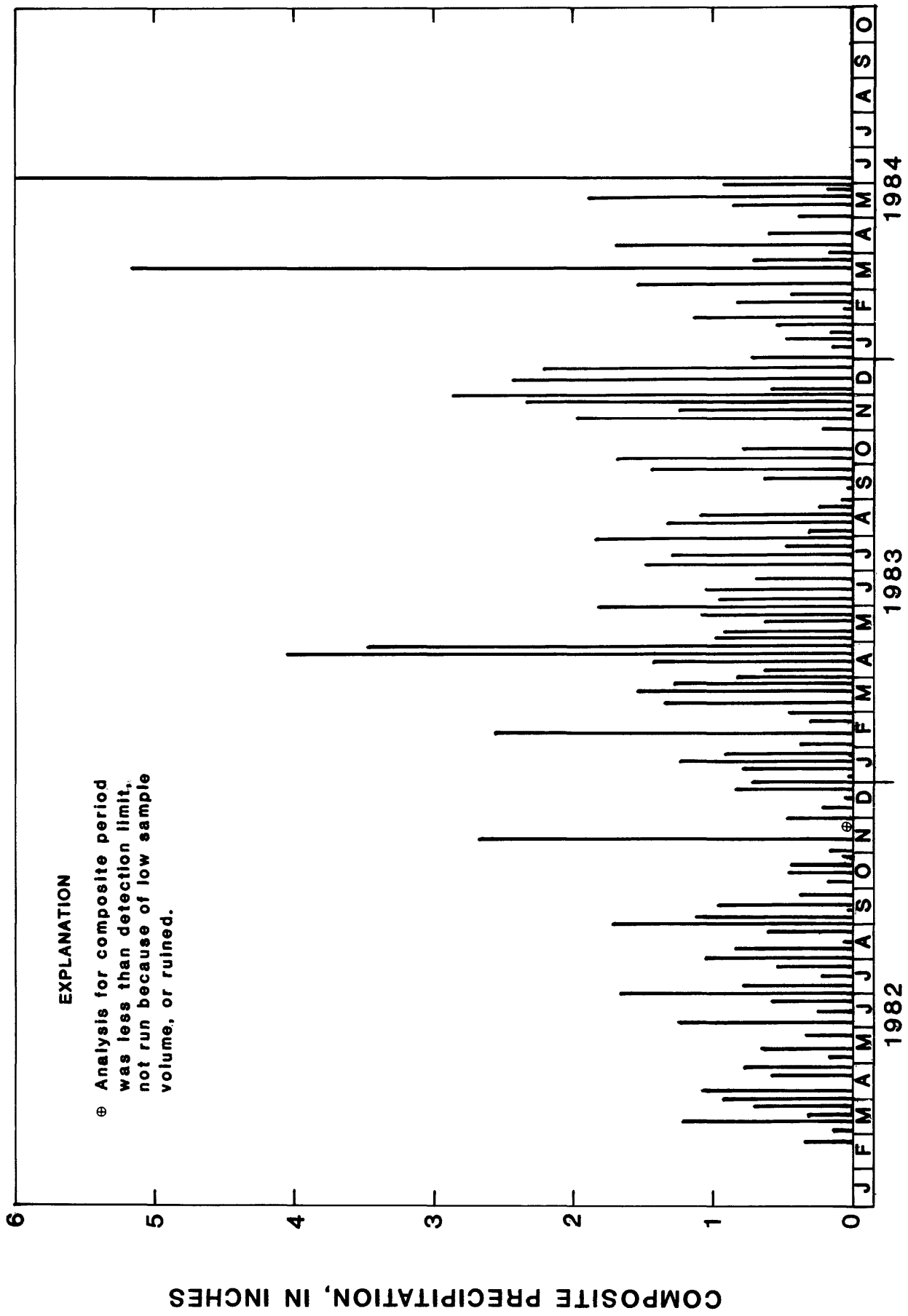
DATA PLOTTED ON LAST DAY OF COMPOSITE SAMPLING PERIOD

Figure 10.--Distribution of copper deposition.



DATA PLOTTED ON LAST DAY OF COMPOSITE SAMPLING PERIOD

Figure 11.--Distribution of zinc deposition.



EXPLANATION

⊕ Analysis for composite period was less than detection limit, not run because of low sample volume, or ruined.

DATA PLOTTED ON LAST DAY OF COMPOSITE SAMPLING PERIOD

Figure 12.--Precipitation quantity for sample composite periods.

A cross-correlation analysis was performed between the copper, lead, zinc, and hydrogen-ion deposition. For this analysis, extreme outliers (greater than 6 times the standard deviation from the mean for lead and zinc and greater than 6 times the mean for copper) were excluded. It was assumed that these outliers are not representative of the general tendency for the data. The correlation coefficients are presented in table 6. The correlation between lead and copper and between copper and zinc were the only correlations that indicated possible relations between the deposition of one metal with that of another. These relations are shown in figures 13 and 14. This would infer that the sources or processes that influence the deposition of these metals may be related. The deposition of hydrogen ions is related only to the lead deposition. The deposition of hydrogen ions is related only to the lead deposition. This relation is shown in figure 15. The correlation indicates that the sources of acidic deposition may be similar to the sources of atmospheric lead.

Organic Compounds

Forty-two composite precipitation samples were analyzed for organic chemical pollutants. The target analytes in these samples included 13 chlorinated hydrocarbon pesticides, polychlorinated biphenyls, and 6 phthalate ester plasticizers. The compounds and their estimated detection limits are reported in table 7. The detection limit range for each compound is a function of its electron capture detector response, sample volume, and the relative amount of interfering substances found in the reagent blanks.

Table 14 (at back of report) summarizes the presence of the alpha, gamma, beta, and delta isomers of hexachlorocyclohexane in each of the composite precipitation samples. None of the other target analytes were detected in any samples within the range of the reported detection limits. N-butyl phthalate and bis-2-(ethyl hexyl) phthalate were found routinely in precipitation extracts, but they were also found in a similar concentration range in the reagent blanks.

The identity of the alpha isomer of hexachlorocyclohexane, detected in 34 (81 percent) of the 42 samples analyzed, was confirmed by GC/MS analysis of a pooled concentrate of all sample extracts. The gamma, beta, and delta isomers of hexachlorocyclohexane were detected in 3 (7 percent), 12 (29 percent), and 5 (12 percent) of the samples, respectively. However, their identity could not be confirmed by GC/MS. Their concentration in the pooled concentrate was below the limit of detection of the mass spectrometer (100 pg/ μ L, picograms per liter).

Table 6.--Correlation coefficients (r) for lead, copper, zinc,
and hydrogen-ion deposition

	Copper deposition	Zinc deposition	Hydrogen-ion deposition
Lead deposition	0.42	0.06	0.64
Copper deposition	1.00	.31	.24
Zinc deposition	.31	1.00	.16

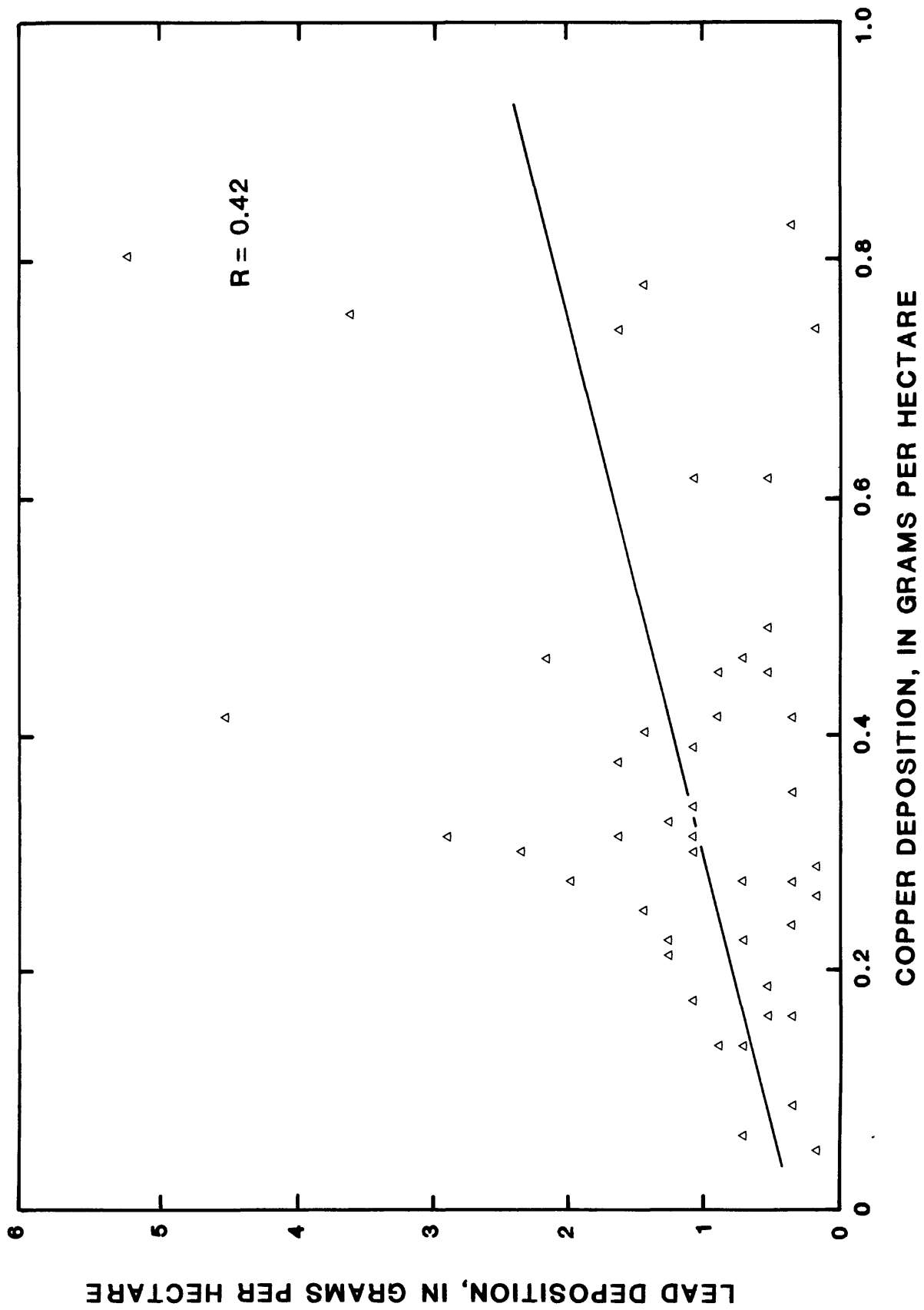


Figure 13.--Relation of lead deposition to copper deposition for composite samples.

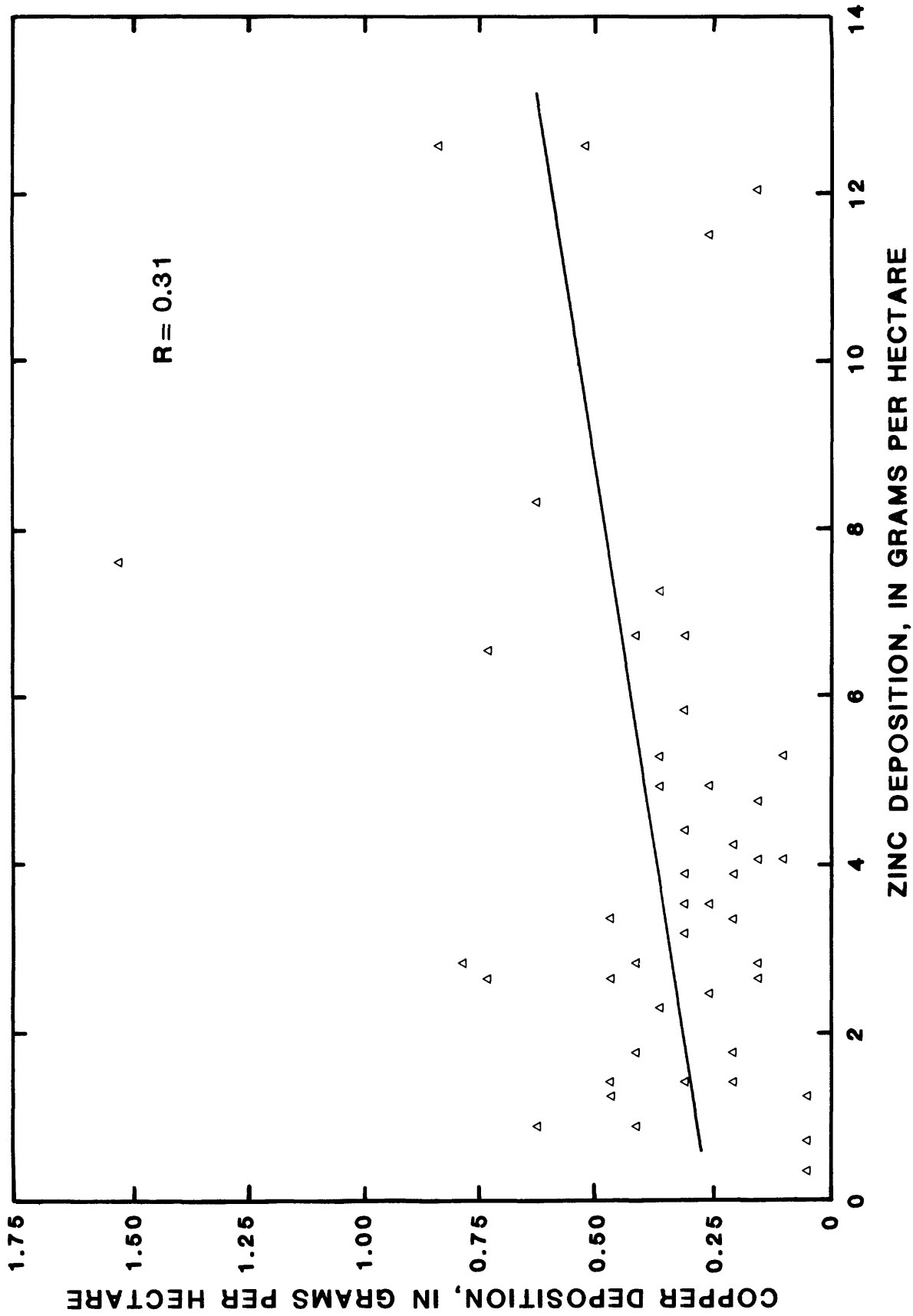
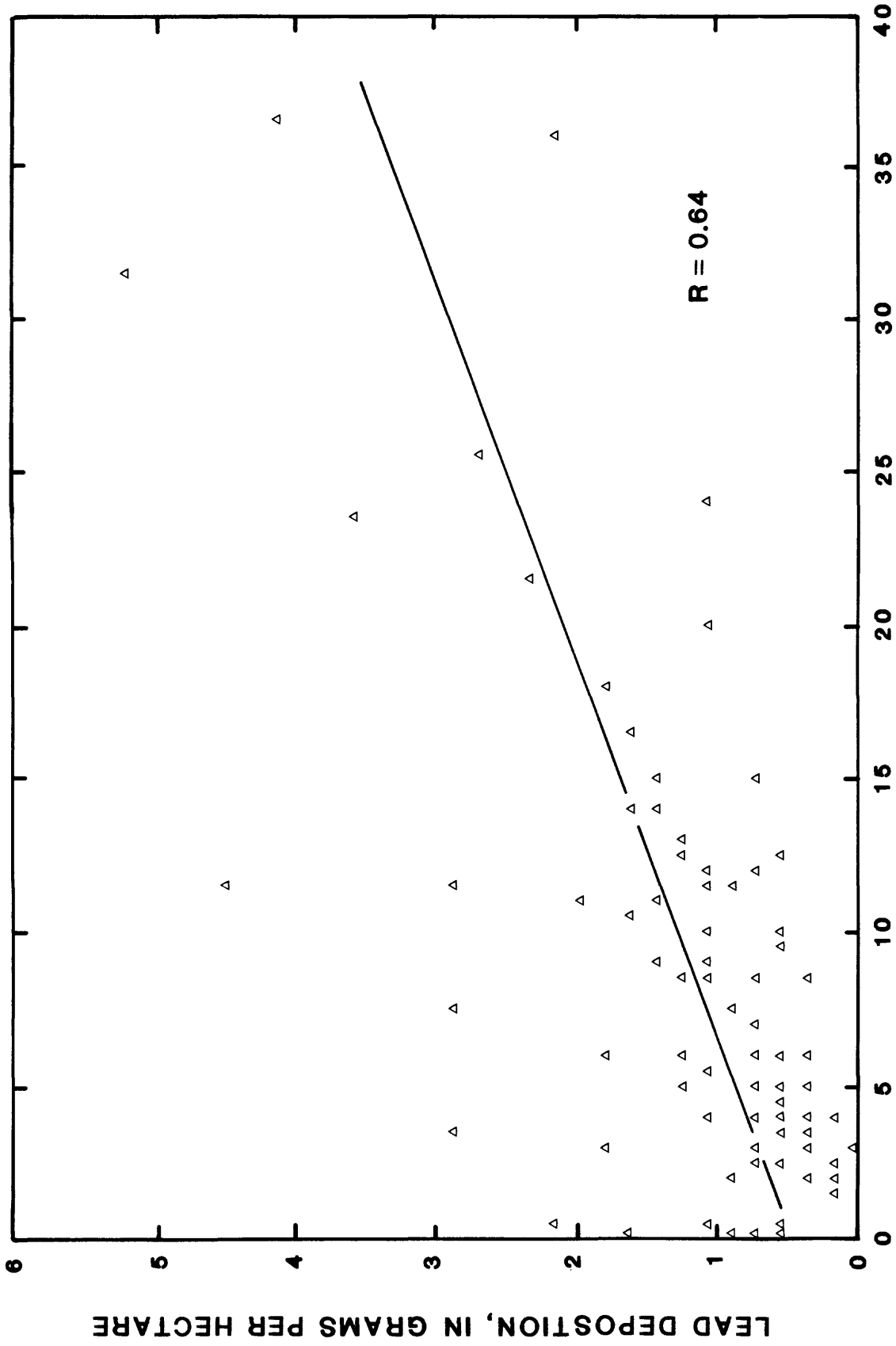


Figure 14.--Relation of copper deposition to zinc deposition for composite samples.



HYDROGEN ION DEPOSITION, IN GRAMS PER HECTARE

Figure 15.--Relation of lead deposition to hydrogen ion deposition for composite samples.

Table 7.--Detection limits for analysis of organic compounds in precipitation samples

Compound	Minimum detectable quantity (picograms)	Detection limit range (nanograms per liter)
Pesticides		
α-BHC	1	< 1 - < 24
γ-BHC	2	< 2 - < 48
β-BHC	1	< 1 - < 24
Δ-BHC	2	< 2 - < 48
Dieldrin	2	< 2 - < 48
Heptaclor	1	< 1 - < 24
Aldrin	2	< 2 - < 48
Heptaclor epoxide	1	< 1 - < 24
Endosulfan I	4	< 4 - 200
Endosulfan II	4	< 4 - 200
pp' DDE	4	< 4 - 200
pp' DDD	4	< 4 - 200
pp' DDT ¹	4	< 4 - 200
PCB's (Aroclor 1242)	40	< 40 - < 960
Endrin	4	< 4 - 200
Phthalates		
Dimethyl phthalate	300	< 300 - < 7,200
Diethyl phthalate	250	< 250 - < 6,000
Di-n-butyl-phthalate ²	40	< 2,000 - < 48,000
Bis-2-(ethyl hexyl) phthalate ²	400	< 4,000 - < 96,000
Butyl-benzyl phthalate ²	20	< 200 - < 4,800
Di-n-octyl-phthalate	500	< 500 - < 12,000

¹ Interference.

² Di-n-butyl phthalate, butyl-benzyl phthalate, and bis-2-ethyl hexyl phthalate were consistently found in the solvent blanks. Concentration of 150 milliliters methylene chloride and 50 milliliters hexane to 10 milliliters yielded a solution containing 1, 0.1, and 0.2 nanograms per microliter of di-n-butyl phthalate, butyl-benzyl phthalate, and bis-2-ethyl hexyl phthalate, respectively. The quantity of these compounds recovered in the solvent blank was used to establish the range of detection limits.

Examination of the concentrations of the hexachloro-cyclohexane isomers in the precipitation samples shows that the alpha isomer was present in the greatest concentration in all samples. This is reflected in the volume-weighted mean values reported in table 8. These values are consistent with mean values reported for the alpha and gamma isomers in precipitation samples collected at St. Hubert, Quebec and Truro, Nova Scotia (tables 9 and 10). These stations are operated as part of a national network of precipitation chemistry monitoring stations for organic chemical pollutants (CANSOC) by Environment Canada and are in relatively close geographic proximity to the Greenville site (A. O. Davis, Environment Canada, written commun., 1984).

Hexachlorocyclohexanes have been reported in precipitation in a similar concentration range at all of the other CANSOC sites at several sites in the Great Lakes basin (Strachan and Huneault, 1979), and in the Hawaiian Islands (Benevise and others, 1972). They also have been reported at lower concentrations in precipitation collected at Enewetak, a remote coral island in the north Pacific Ocean (Atlas and Giam, 1981). At the Greenville site, as at the other sites, the alpha isomer of hexachlorocyclohexane was found to be present at concentrations greater than the other isomers.

On the basis of the data collected in this study and the numerous published measurements, it seems that hexachlorocyclohexanes, in particular the alpha isomer, are ubiquitous components of precipitation in the Northern Hemisphere. This is apparently because of the widespread use of the hexachlorocyclohexane pesticides, BHC, and lindane. BHC was introduced in 1942 and used in the United States until voluntary cancellation of its registration in 1977. Commercial products sold under the trade name BHC were a mixture of five stereoisomers of hexachlorocyclohexane in the following concentration ranges: alpha, 55 to 70 percent; beta, 6 to 8 percent; gamma, 10 to 18 percent; delta, 3 to 4 percent; and epsilon, <1 percent (U.S. Environmental Protection Agency, 1980).

Products containing greater than 99 percent of the gamma isomer were introduced after the discovery that nearly all of the insecticidal activity of hexachlorocyclohexane was attributed to the gamma isomer (Windholz, 1983). These were marketed under the trade name Lindane. Lindane currently is registered for use in greenhouses and for topical applications. Registration for large scale areal application in agriculture and forestry was cancelled in 1978 (Roy Clark, U.S. Environmental Protection Agency, oral commun., 1984). Five years after the cancellation, this study detected hexachlorocyclohexane in precipitation samples, indicating the relatively long atmospheric residence time of this compound.

The fact that alpha-hexachlorocyclohexane is the principal isomer found in precipitation samples reflects the relative composition of BHC. It also may be because of isomerization of the gamma isomer to the alpha isomer by photolysis in direct sunlight (Malaiyandi and Shah, 1984).

Table 8 .--Statistical summary of concentrations of BHC-isomers in weekly composite precipitation samples

Isomer (in ng/L)	Number of obser- vations	Number of obser- vations above de- tection limit	Range	Concentrations below detection limit set equal to zero					
				Volume-weighted mean concentration	Median	Skew	Volume-weighted mean concentration	Median	Skew
Alpha	42	34	< 2 - 120	21	12	2.3	21	14	2.5
Gamma	42	3	< 1 - 25	1.3	0	3.7	5.5	4	2.1
Beta	42	12	< 1 - 42	2.8	0	3.5	4.7	5	2.7
Delta	42	5	< 1 - 24	1.2	0	3.2	5.2	4	2.2

Table 9.--Concentration of alpha-BHC in precipitation
at selected sites

Site	Observation period	Concentrations (Nanograms per liter)			
		Maximum	Minimum	Mean	Median
Greenville, Maine ^{1/}	Feb. 1982 to Mar. 1983	120	< 2	21	12
St. Hubert, Quebec ^{2/}	May 1976 to May 1980	72	< 1	31	31
Truro, Nova Scotia ^{2/}	Sep. 1977 to June 1980	43	< 1	17	17
Enewetak Atoll ^{3/}	Apr. 1979 to Aug. 1979	7	1	3	--

^{1/} Mean concentration for Greenville, Maine computed using sample concentrations below detection limit set equal to zero.

^{2/} Unpublished data, A. O. Davis, Environment Organization, Canada, written commun., 1984.

^{3/} Atlas and Giam, 1981.

Table 10.--Concentration of gamma-BHC in precipitation
at selected sites

Site	Observation period	Concentrations (Nanograms per liter)			
		Maximum	Minimum	Mean	Median
Greenville, Maine ^{1/}	Feb. 1982 to Mar. 1983	25	< 1	1.3	< 4
St. Hubert, Quebec ^{2/}	May 1976 to May 1980	11	< 1	5	4
Truro, Nova Scotia ^{2/}	Mar. 1977 to June 1980	14	< 1	5	5
Enewetak Atoll ^{3/}	Apr. 1979 to Aug. 1979	1.6	.3	.5	--

^{1/} Mean concentration for Greenville, Maine computed using sample concentrations below detection limit set equal to zero.

^{2/} Unpublished data, A. O. Davis, Environment Organization, Canada, written commun., 1984.

^{3/} Atlas and Giam, 1981.

The U.S. Environmental Protection Agency (1980) has recommended a threshold concentration of 80 ng/L (nanograms of hexachlorocyclohexane per liter) for protection of aquatic life. The concentration levels found in this study were generally well below this level. The U.S. Environmental Protection Agency has set a threshold level of 10 to 20 ng/L for drinking water. The volume-weighted mean concentration of alpha-hexachlorocyclohexane found at the Greenville site falls within this range.

The rate of deposition of hexachlorocyclohexane measured at the Greenville site was approximately 0.3 (g/ha)/yr (table 11).

SUGGESTIONS FOR FUTURE STUDIES

Results from the analyses of trace metals and organics in precipitation samples collected at the Greenville site indicate the importance of looking further than pH and major chemical constituents when attempting to assess the impact of pollutant deposition in precipitation. This study also indicates that precipitation quality, including trace metals and organics, may be a cause of concern in more rural areas. Maine is geographically removed from the major industrial centers that produce a large part of the atmospheric "load" of manmade pollutants. Because of atmospheric circulation, however, Maine is a recipient of these materials and is subject to their impacts.

Based on the results of this study, the collection and analysis of trace metals and organic compounds should be expanded in Maine. Additional samplers should be located at the other three NADP sites in Maine to complement a continued sampling program at Greenville.

Further expansion of this effort should include sampling for trace metals and organic compounds at other NADP sites in the Northeast and Midwest. An expanded program could be used to gather baseline data and to evaluate the sources of these constituents and their transport mechanisms.

SUMMARY AND CONCLUSIONS

Results from the analysis of precipitation samples by the NADP indicate that the general chemical quality of precipitation in the Greenville area is similar to other sites in the northeastern United States. The precipitation is characterized by relatively high concentrations of hydrogen ions, sulfate, and nitrate indicating that the precipitation at Greenville is affected by sulfuric and nitric acids in the atmosphere. Nitric acid seems to be a more significant component of precipitation acidity at Greenville than at many other NADP sites in the eastern United States.

Table 11.--Statistical summary of deposition of BHC-isomers in precipitation samples

[Values in parenthesis are computed based upon sample volume;
all others are computed based upon precipitation volume.]

Isomer	Number of obser- vations	Range (ng/m ³)	Composite-period deposition				Skew	Yearly deposition (mean) mg/ha/yr
			Mean	Median	Mean	Median		
			ng/m		mg/ha			
Concentrations below detection limit are set equal to zero								
Alpha	41 (42)	0 - 4,300 (0 - 3,710)	501 (488)	274 (247)	5.01 (4.88)	2.74 (2.47)	3.5 (3.2)	230 (223)
Gamma	41 (42)	0 - 920 (0 - 745)	38.7 (31.7)	0 (0)	0.39 (0.32)	0 (0)	4.8 (4.7)	18 (14)
Beta	41 (42)	0 - 740 (0 - 596)	79.7 (67.0)	0 (0)	0.80 (0.67)	0 (0)	2.5 (2.4)	37 (31)
Delta	41 (42)	0 - 490 (0 - 418)	35.2 (28.6)	0 (0)	0.35 (0.29)	0 (0)	3.3 (3.3)	16 (13)
Concentrations below detection limit are set equal to the detection limit of the constituent								
Alpha	41 (42)	38 - 4,300 (35 - 3,710)	524 (503)	274 (247)	5.24 (5.03)	2.74 (2.47)	3.6 (3.2)	240 (230)
Gamma	41 (42)	32 - 925 (28 - 745)	143 (130)	95 (95)	1.43 (1.30)	0.95 (0.95)	3.7 (3.3)	67 (59)
Beta	41 (42)	19 - 740 (19 - 596)	125 (110)	66 (59)	1.25 (1.10)	0.66 (0.59)	2.6 (2.5)	58 (50)
Delta	41 (42)	26 - 491 (22 - 499)	137 (124)	99 (100)	1.37 (1.24)	0.99 (1.00)	1.9 (2.4)	64 (57)

Of the trace metals included for analysis of the Greenville precipitation, only lead, copper, and zinc were consistently found. Nickel, mercury, cadmium, and chromium were either not found in the samples or were found in concentrations very close to the detection limit of the analysis. In three samples, the concentration of lead exceeded the recommended limit for domestic water supply established by the U.S. Environmental Protection Agency. One sample contained nearly one-half of the recommended limit for copper.

The volume-weighted mean concentration for lead in the Greenville precipitation was in the range 3.6 to 3.7 $\mu\text{g/L}$ depending on the method of calculation. The mean concentration for copper was from 5.1 to 5.3 $\mu\text{g/L}$ and for zinc was from 13 to 14 $\mu\text{g/L}$. Mean deposition rates for the composite periods were from 95 to 121 $\mu\text{g/m}^2$ for lead, 140 to 170 $\mu\text{g/m}^2$ for copper, and 173 to 217 $\mu\text{g/m}^2$ for zinc. The distribution of the composite-period deposition of these metals, especially copper, was found to be uneven, suggesting that their occurrence in atmospheric deposition is episodic.

The deposition of the metals in the precipitation samples was poorly correlated with each other. The best correlations were between lead and copper and between copper and zinc. The deposition of hydrogen ions was best correlated with lead deposition indicating that there may be a link between the sources of acidic deposition and atmospheric lead.

Alpha-hexachlorocyclohexane was the only organic chemical pollutant consistently detected in the precipitation samples. Because the use of hexachlorocyclohexane pesticides in the United States has been limited since 1978, detection at the Greenville site indicates that this chemical has a relatively long atmospheric residence time and may be transported over long distances. The concentration level detected and the rate of deposition measured were small. The beta, gamma, and delta isomers of hexachlorocyclohexane were found less frequently in the precipitation samples than the alpha isomer. None of the other target organic analytes were detected in any of the samples.

Trace metals and organic compounds should be considered as important aspects of the chemical quality of precipitation in Maine, as elsewhere. Future studies should include the analysis of these compounds to improve an understanding of their distribution, possible sources, and transport mechanisms. The study of precipitation chemistry should not be limited to industrialized regions. Rural areas, as typified by Maine, are often the recipients of and are subject to possible harmful effects from atmospheric pollutants.

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Table 12.--Descriptions of precipitation samples for trace-metals analysis

[Precipitation type: R, rain; S, snow; M, mixed.
 Sample type: B, bulk; W, wet only.
 ml, milliliter; $\mu\text{S}/\text{cm}$ at 25 °C, microsiemens per
 centimeter at 25 degrees Celsius.]

Start composite	End composite	Precipitation type	Precipitation volume (ml)	Sample type	Sample volume (ml)	Specific conductance ($\mu\text{S}/\text{cm}$) 25 °C)	pH units
16FEB82	23FEB82	S	590	W	285	19	4.4
23FEB82	02MAR82	S	240	W	209	34	4.0
02MAR82	09MAR82	M	2,090	B	2,030	7	4.8
09MAR82	16MAR82	M	530	B	363	50	3.8
16MAR82	23MAR82	S	1,210	B	1,190	18	4.5
23MAR82	30MAR82	M	1,590	B	1,418	16	4.4
30MAR82	06APR82	M	1,840	B	1,624	18	4.4
06APR82	20APR82	M	970	B	577	36	3.8
20APR82	27APR82	M	1,340	B	937	31	4.2
27APR82	04MAY82	M	280	B	123	48	4.0
04MAY82	11MAY82	R	1,120	B	1,127	47	4.2
11MAY82	25MAY82	R	570	B	112	206	3.4
25MAY82	08JUN82	R	2,160	B	1,470	7	5.8
08JUN82	15JUN82	R	430	B	397	8	4.6
15JUN82	22JUN82	R	1,000	B	593	78	6.1
22JUN82	29JUN82	R	2,860	B	2,364	15	4.4
29JUN82	06JUL82	R	1,340	B	1,198	10	5.7
06JUL82	20JUL82	R	380	B	350	62	5.6
20JUL82	27JUL82	R	910	B	754	4	6.5
27JUL82	03AUG82	R	1,810	B	1,530	5	6.2
03AUG82	10AUG82	R	1,430	B	1,278	13	5.0
10AUG82	17AUG82	R	120	B	166	25	5.2
17AUG82	24AUG82	R	1,030	B	910	11	4.8
24AUG82	31AUG82	R	2,970	B	2,450	15	4.1
31AUG82	07SEP82	R	1,930	B	1,567	7	5.0
07SEP82	14SEP82	R	30	B	42	--	--
14SEP82	21SEP82	R	1,660	B	1,422	15	4.3
21SEP82	28SEP82	R	660	B	602	10	5.4
28SEP82	05OCT82	R	290	B	274	25	4.2
05OCT82	12OCT82	R	780	B	655	37	3.9
12OCT82	19OCT82	R	710	B	634	39	4.0
19OCT82	26OCT82	R	140	B	116	31	6.6
26OCT82	02NOV82	R	280	B	286	14	4.2
02NOV82	09NOV82	R	4,590	B	4,211	7	4.3
09NOV82	23NOV82	M	--	B	4,984	3	4.5

Table 12.--Descriptions of precipitation samples for trace-metals analysis--Continued

Start composite	End composite	Precipitation type	Precipitation volume (ml)	Sample type	Sample volume (ml)	Specific conductance ($\mu\text{S}/\text{cm}$) 25 °C)	pH units
23NOV82	30NOV82	M	810	B	854	19	4.1
30NOV82	07DEC82	R	360	B	312	34	3.7
07DEC82	14DEC82	R	90	B	34	--	--
14DEC82	21DEC82	M	1,450	B	1,378	15	4.4
21DEC82	28DEC82	M	1,220	B	1,144	15	4.5
28DEC82	04JAN83	S	30	B	48	--	--
04JAN83	11JAN83	M	3,090	B	2,783	6	5.0
11JAN83	18JAN83	M	2,120	B	1,942	3	5.1
18JAN83	25JAN83	M	1,550	B	1,570	5	5.0
25JAN83	01FEB83	S	640	B	604	8	4.6
01FEB83	08FEB83	M	4,400	B	3,478	11	4.8
08FEB83	22FEB83	S	520	B	324	21	4.2
22FEB83	01MAR83	S	780	B	552	31	4.2
01MAR83	08MAR83	M	2,290	B	1,586	4	5.2
08MAR83	15MAR83	M	2,640	B	2,542	6	5.2
15MAR83	22MAR83	R	2,190	B	2,108	30	4.6
22MAR83	29MAR83	M	1,410	B	1,028	13	5.0
29MAR83	05APR83	M	1,100	W	1,083	6	5.0
05APR83	12APR83	M	2,450	W	325	65	4.2
12APR83	19APR83	R	6,970	W	6,887	6	5.0
19APR83	26APR83	M	5,970	W	6,062	13	4.6
26APR83	03MAY83	R	1,690	B	1,570	62	4.2
03MAY83	10MAY83	R	1,570	W	1,419	56	4.2
10MAY83	17MAY83	M	1,070	W	904	19	4.6
17MAY83	24MAY83	R	1,860	W	1,514	26	4.4
24MAY83	31MAY83	R	3,120	W	2,365	15	4.8
31MAY83	07JUN83	R	1,640	W	1,319	32	4.4
07JUN83	14JUN83	R	1,810	W	462	30	4.3
14JUN83	21JUN83	R	1,170	-	--	--	--
21JUN83	05JUL83	R	2,570	W	2,546	22	4.5
05JUL83	12JUL83	R	2,220	W	2,213	10	4.6
12JUL83	19JUL83	R	790	W	767	24	4.3
19JUL83	26JUL83	R	2,310	W	2,241	5	5.0
26JUL83	02AUG83	R	530	B	529	75	3.8
02AUG83	09AUG83	R	2,260	B	1,920	21	4.4
09AUG83	16AUG83	R	1,860	W	1,864	6	4.8
16AUG83	23AUG83	R	400	B	7	--	--
23AUG83	30AUG83	R	120	B	106	77	5.4
30AUG83	13SEP83	R	760	B	822	19	5.1
13SEP83	20SEP83	R	1,070	W	968	19	4.5

Table 12.--Descriptions of precipitation samples for trace-metals analysis--Continued

Start composite	End composite	Precipitation type	Precipitation volume (ml)	Sample type	Sample volume (ml)	Specific conductance ($\mu\text{S}/\text{cm}$) 25 °C)	pH units
20SEP83	27SEP83	R	2,480	W	2,379	6	5.0
27SEP83	11OCT83	R	2,900	W	2,666	25	4.2
11OCT83	18OCT83	R	1,330	W	905	6	5.0
18OCT83	01NOV83	M	340	W	223	--	--
01NOV83	08NOV83	R	3,380	W	2,655	12	4.7
08NOV83	15NOV83	R	2,120	W	1,766	10	4.6
15NOV83	22NOV83	M	3,980	W	3,736	6	4.7
22NOV83	29NOV83	M	4,910	W	4,738	3	5.2
29NOV83	06DEC83	M	970	B	908	6	4.9
06DEC83	13DEC83	M	4,160	B	4,188	6	5.1
13DEC83	27DEC83	M	3,790	B	3,624	6	5.0
27DEC83	03JAN84	M	1,240	B	1,280	10	4.7
03JAN84	10JAN84	S	240	B	236	50	4.0
10JAN84	17JAN84	S	790	B	776	15	4.5
17JAN84	24JAN84	S	260	W	104	31	4.2
24JAN84	31JAN84	S	930	W	550	25	4.4
31JAN84	07FEB84	M	1,950	W	1,472	6	5.0
07FEB84	14FEB84	M	90	W	33	--	--
14FEB84	21FEB84	M	1,410	W	979	16	4.6
21FEB84	28FEB84	M	760	W	750	7	4.9
28FEB84	06MAR84	M	2,640	B	2,404	10	4.8
06MAR84	20MAR84	M	8,880	B	7,270	9	5.1
20MAR84	27MAR84	M	1,210	B	727	18	4.5
27MAR84	03APR84	S	280	B	48	--	--
03APR84	10APR84	M	2,910	B	2,693	15	4.6
10APR84	17APR84	R	1,020	W	1,018	12	4.5
17APR84	01MAY84	R	660	W	522	12	4.8
01MAY84	08MAY84	M	1,470	W	1,350	19	4.5
08MAY84	15MAY84	R	3,260	W	2,668	19	4.4
15MAY84	22MAY84	R	290	W	307	38	4.2
22MAY84	29MAY84	R	1,550	W	1,484	19	4.4
29MAY84	05JUN84	R	10,380	W	9,402	3	5.0

Table 13.--Concentrations of total recoverable trace-metals in precipitation samples
[A dash indicates missing data]

Start composite	End composite	Concentration (micrograms per liter)						
		Pb	Cu	Zn	Ni	Hg	Cd	Cr
16FEB82	23FEB82	3	--	--	40	1.4	< 4	--
23FEB82	02MAR82	5	--	--	< 20	--	< 4	--
02MAR82	09MAR82	4	< 2.0	--	< 20	< .2	< 4	--
09MAR82	16MAR82	12	4.0	42	--	--	< 4	--
16MAR82	23MAR82	4	< 1.0	16	--	< .2	< 4	--
23MAR82	30MAR82	6	1.6	21	--	< .2	< 4	--
30MAR82	06APR82	10	1.1	15	--	< .2	< 4	--
06APR82	20APR82	16	2.0	26	--	< .2	< 4	--
20APR82	27APR82	7	1.2	23	--	< .2	< 4	--
27APR82	04MAY82	12	3.8	110	--	--	< 4	--
04MAY82	11MAY82	8	< .6	7	< 2	< .2	< 4	--
11MAY82	25MAY82	60	9.0	--	--	--	< 4	--
25MAY82	08JUN82	3	7.2	60	8	< .2	< 4	--
08JUN82	15JUN82	3	.8	6	< 2	--	< 4	--
15JUN82	22JUN82	10	2.4	15	2	< .2	< 4	--
22JUN82	29JUN82	4	< .6	< 5	< 2	< .2	< 4	--
29JUN82	06JUL82	10	2.2	6	< 2	< .2	< 4	--
06JUL82	20JUL82	16	7.6	29	3	< .2	< 4	--
20JUL82	27JUL82	5	1.6	12	< 2	< .2	< 4	--
27JUL82	03AUG82	2	1.6	< 5	< 2	< .2	< 4	--
03AUG82	10AUG82	4	.6	12	< 2	< .2	< 4	--
10AUG82	17AUG82	--	--	--	--	--	--	--
17AUG82	24AUG82	5	.4	8	< 2	< .2	< 4	--
24AUG82	31AUG82	9	< .4	6	< 2	< .2	< 4	--
31AUG82	07SEP82	6	< .4	6	< 3	< .2	< 4	--
07SEP82	14SEP82	--	--	--	--	--	--	--
14SEP82	21SEP82	5	< .4	6	< 3	< .2	< 4	--
21SEP82	28SEP82	6	6.0	8	< 3	< .2	< 4	--
28SEP82	05OCT82	--	--	--	< 3	< .2	--	--
05OCT82	12OCT82	6	--	7	< 3	< .2	< 4	< 10
12OCT82	19OCT82	10	< 0.4	9	< 3	< 0.2	< 4	< 10
19OCT82	26OCT82	--	--	--	< 3	--	--	--
26OCT82	02NOV82	--	--	17	< 3	--	< 4	< 10
02NOV82	09NOV82	3	< .4	5	< 3	< .2	< 4	< 10
09NOV82	23NOV82	2	< .4	< 5	< 3	< .2	< 4	< 10

Table 13.--Concentrations of total recoverable trace-metals in precipitation samples
 --Continued

Start composite	End composite	Concentration (micrograms per liter)						
		Pb	Cu	Zn	Ni	Hg	Cd	Cr
23NOV82	30NOV82	5	< 0.4	< 5	< 2	< 0.2	< 4	< 10
30NOV82	07DEC82	80	7.2	17	< 2	--	< 4	< 10
07DEC82	14DEC82	--	--	--	--	--	--	--
14DEC82	21DEC82	5	< .4	6	< 2	3.4	< 4	< 10
21DEC82	28DEC82	3	< .4	9	< 1	< .2	< 4	< 10
28DEC82	04JAN83	--	--	--	--	--	--	--
04JAN83	11JAN83	1	< .4	< 5	< 1	< .2	< 4	< 10
11JAN83	18JAN83	2	< .4	< 5	< 1	< .2	< 4	< 10
18JAN83	25JAN83	1	< .4	< 5	< 1	.2	< 4	< 10
25JAN83	01FEB83	5	< .4	11	< 1	< .2	< 4	< 10
01FEB83	08FEB83	3	.4	5	< 1	< .2	< 4	< 10
08FEB83	22FEB83	13	2.2	35	1	< .2	4	< 10
22FEB83	01MAR83	10	1.8	12	< 1	--	< 4	< 10
01MAR83	08MAR83	2	< .4	< 5	< 1	--	< 4	< 10
08MAR83	15MAR83	1	2.0	30	< 1	--	< 4	< 10
15MAR83	22MAR83	3	1.8	24	< 1	--	< 4	< 10
22MAR83	29MAR83	1	1.2	16	1	--	< 4	< 10
29MAR83	05APR83	< 1	.6	23	< 1	--	< 4	< 10
05APR83	12APR83	3	1.0	19	1	--	< 4	< 10
12APR83	19APR83	< 1	1.4	7	< 1	--	< 4	< 10
19APR83	26APR83	4	.8	< 5	< 1	--	< 4	< 10
26APR83	03MAY83	6	1.2	22	< 1	--	< 4	< 10
03MAY83	10MAY83	6	1.0	20	< 1	--	< 4	< 10
10MAY83	17MAY83	3	1.0	16	< 1	--	< 4	< 10
17MAY83	24MAY83	--	--	--	--	--	--	--
24MAY83	31MAY83	6	70	28	< 1	--	< 4	< 10
31MAY83	07JUN83	4	0.7	10	< 1	--	< 4	< 10
07JUN83	14JUN83	6	2.6	23	< 1	--	< 4	< 10
14JUN83	21JUN83	--	--	--	--	--	--	--
21JUN83	05JUL83	3	.8	8	2	--	< 4	< 10
05JUL83	12JUL83	1	1.2	8	< 1	--	< 4	< 10
12JUL83	19JUL83	10	1.8	31	1	--	< 4	< 10
19JUL83	26JUL83	8	---	9	< 1	--	< 4	< 10
26JUL83	02AUG83	8	1.6	48	3	--	< 4	10
02AUG83	09AUG83	4	2.2	8	2	--	< 4	< 10

Table 13.--Concentrations of total recoverable trace-metals in precipitation samples
--Continued

Start composite	End composite	Concentration (micrograms per liter)						
		Pb	Cu	Zn	Ni	Hg	Cd	Cr
09AUG83	16AUG83	1	< 0.4	< 5	< 1	--	< 4	< 10
16AUG83	23AUG83	--	--	--	--	--	--	--
23AUG83	30AUG83	--	--	--	--	--	--	--
30AUG83	13SEP83	--	--	--	--	--	--	--
13SEP83	20SEP83	60	430	130	3	--	< 4	< 10
20SEP83	27SEP83	< 1	6.0	25	< 1	--	< 4	< 10
27SEP83	11OCT83	6	30	17	2	--	< 4	< 10
11OCT83	18OCT83	< 1	2.2	7	< 1	--	< 4	< 10
18OCT83	01NOV83	--	---	150	--	--	< 4	< 10
01NOV83	08NOV83	3	< .4	9	< 1	--	< 4	< 10
08NOV83	15NOV83	2	1.4	8	2	--	< 4	< 10
15NOV83	22NOV83	1	< .4	5	< 1	--	< 4	< 10
22NOV83	29NOV83	--	1.2	< 5	< 1	--	< 4	< 10
29NOV83	06DEC83	2	.6	35	< 1	--	< 4	< 10
06DEC83	13DEC83	1	< .4	10	< 1	--	< 4	< 10
13DEC83	27DEC83	3	< .4	8	< 1	--	< 4	< 10
27DEC83	03JAN84	4	< .4	24	< 1	--	< 4	< 10
03JAN84	10JAN84	27	8.2	37	1	--	< 4	< 10
10JAN84	17JAN84	1	.4	10	< 1	--	< 4	< 10
17JAN84	24JAN84	2	2.4	--	2	--	--	--
24JAN84	31JAN84	2	1.6	26	1	--	< 4	< 10
31JAN84	07FEB84	2	1.6	11	< 1	--	12	< 10
07FEB84	14FEB84	--	--	--	--	--	--	--
14FEB84	21FEB84	2	1.6	22	5	--	< 4	< 10
21FEB84	28FEB84	2	2.4	280	3	--	< 4	< 10
28FEB84	06MAR84	2	1.0	16	3	--	< 4	< 10
06MAR84	20MAR84	< 1	.4	9	< 1	--	< 4	< 10
20MAR84	27MAR84	4	1.2	18	< 1	--	< 4	< 10
27MAR84	03APR84	4	1.2	18	< 1	--	< 4	< 10
03APR84	10APR84	2	< .4	6	< 1	--	< 4	< 10
10APR84	17APR84	2	1.0	75	1	--	< 4	< 10
17APR84	01MAY84	2	7.2	26	2	--	< 4	< 10
01MAY84	08MAY84	3	1.2	50	< 1	--	< 4	< 10
08MAY84	15MAY84	2	.6	13	< 1	--	< 4	< 10
15MAY84	22MAY84	7.5	6.0	53	17	--	--	--
22MAY84	29MAY84	2.0	.8	17	< 1	--	--	--
29MAY84	05JUN84	< .5	< .2	6	< 1	--	--	--

Table 14.--Concentration of BHC-isomers in precipitation samples

[ml is milliliters; dashes
indicate missing data]

Start composite	End composite	Sample volume (ml)	Concentration (Nanograms per liter)			
			alpha	gamma	beta	delta
16FEB82	23FEB82	314	--	--	--	--
23FEB82	02MAR82	189	--	--	--	--
02MAR82	09MAR82	2099	3	< 2	< 1	< 2
09MAR82	16MAR82	451	--	--	--	--
16MAR82	23MAR82	1219	9	< 2	< 1	< 2
23MAR82	30MAR82	1632	12	< 4	< 2	< 4
30MAR82	06APR82	1892	9	< 4	< 2	< 4
06APR82	20APR82	629	14	< 13	< 7	< 13
20APR82	27APR82	1105	13	< 4	< 2	< 4
27APR82	04MAY82	98	--	--	--	--
04MAY82	11MAY82	1261	--	--	--	--
11MAY82	25MAY82	163	< 24	< 48	< 24	< 48
25MAY82	08JUN82	1609	5	3	2	< 3
08JUN82	15JUN82	433	25	< 9	< 5	< 9
15JUN82	22JUN82	708	21	< 4	16	24
22JUN82	29JUN82	2437	16	< 1	6	11
29JUN82	06JUL82	1198	14	< 2	7	14
06JUL82	20JUL82	371	12	< 12	< 6	< 12
20JUL82	27JUL82	751	< 3	< 6	< 3	< 6
27JUL82	03AUG82	1493	31	< 2	5	< 2
03AUG82	10AUG82	1237	16	< 6	< 3	< 6
10AUG82	17AUG82	--	--	--	--	--
17AUG82	24AUG82	906	10	< 2	4	8
24AUG82	31AUG82	2388	93	20	16	< 1
31AUG82	07SEP82	1571	26	< 4	11	< 4
07SEP82	14SEP82	35	--	--	--	--
14SEP82	21SEP82	1421	49	15	6	< 1
21SEP82	28SEP82	592	31	< 4	21	< 4
28SEP82	05OCT82	275	22	< 20	< 10	< 20
05OCT82	12OCT82	653	52	< 12	< 6	< 12
12OCT82	19OCT82	754	120	25	42	14
19OCT82	26OCT82	123	--	--	--	--
26OCT82	02NOV82	316	75	< 22	< 11	< 22
02NOV82	09NOV82	4665	51	< 1	6	< 1
09NOV82	23NOV82	7993	15	< 4	< 2	< 4

Table 14.--Concentration of BHC-isomers in precipitation samples
 --Continued

Start composite	End composite	Sample volume (ml)	Concentration (Nanograms per liter)			
			alpha	gamma	beta	delta
23NOV82	30NOV82	884	22	< 10	< 5	< 10
30NOV82	07DEC82	373	30	< 14	< 7	< 14
07DEC82	14DEC82	53	--	--	--	--
14DEC82	21DEC82	1586	19	< 4	< 2	< 4
21DEC82	28DEC82	1238	< 2	< 4	< 2	< 4
28DEC82	04JAN83	79	--	--	--	--
04JAN83	11JAN83	2975	11	< 2	< 1	< 2
11JAN83	18JAN83	1900	20	< 4	< 2	< 4
18JAN83	25JAN83	1676	--	--	--	--
25JAN83	01FEB83	763	< 9	< 18	< 9	< 18
01FEB83	08FEB83	3579	4	< 4	< 2	< 4
08FEB83	22FEB83	399	< 16	< 32	< 16	< 32
22FEB83	01MAR83	553	< 10	< 20	< 10	< 20
01MAR83	08MAR83	1908	11	< 2	< 1	< 2
08MAR83	15MAR83	2848	12	< 2	< 1	< 2
15MAR83	22MAR83	1984	6	< 2	< 1	< 2
22MAR83	29MAR83	1375	< 3	< 6	< 3	< 6
29MAR83	05APR83	1122	12	< 6	< 3	< 6