

1-1-1986

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Recommended Citation

Fernandez, I.J., L. Wortman, and S.A. Norton. 1986. Composition of precipitation at the National Atmospheric Deposition Program/ National Trends Network (NADP/NTN) site in Greenville, Maine. Maine Agricultural Experiment Station Technical Bulletin 118.

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Composition of Precipitation at the
National Atmospheric Deposition
Program/National Trends Network
(NADP/NTN) Site in Greenville, Maine

November 1979 to December 1982

by

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Cooperating Assistant Professor of Forest Resources

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National Atmospheric Deposition Program

Site Operator

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The NADP/NTN site at Greenville, Maine is supported by the Maine
Agricultural Experiment Station at the University of Maine at Orono.

INTRODUCTION

There has been a great deal of attention focused on the issue of "acid rain" in the last 10 to 15 years with regard to the possible effects that atmospherically derived chemicals may have on lakes, streams, crops, forests, and materials. The term "acid rain" is derived from the occurrence of sulfuric (H_2SO_4) and nitric (HNO_3) acids in all forms of precipitation as a result of emissions of sulfur (S) and nitrogen (N), largely from man's activities. Acid precipitation is only one component of our modern chemical climate which results in the deposition of acids in precipitation as well as base cations (e.g. calcium, magnesium, potassium, sodium), anions (e.g. phosphate, chloride, fluoride), trace metals (e.g. lead, copper, nickel, zinc, cadmium), photochemical oxidants (e.g. ozone) and organic compounds (e.g. hydrocarbons). Relatively poor information exists for the levels of most of these materials in deposition for rural areas across the nation with the exception of the monitoring of major cations and anions in precipitation carried out by the National Atmospheric Deposition Program (NADP).

Currently the NADP has the most extensive network for monitoring atmospheric deposition in the United States. This Federal/State program is carried out through the Cooperative State Research Service (U.S. Department of Agriculture) and involves a large number of Federal, State, University, and industry participants. The monitoring activity is highly organized with respect to siting and operational criteria for sampling sites, analytical methodology, and data handling and storage. The NADP was created in 1978 as Regional Project NC-141 which evolved to become Interregional Project IR-7 in 1982. This network has been well integrated with the Canadian Network for Sampling Precipitation (CANSAP), now called the Canadian Air and Precipitation Monitoring Network (CAPMON). As a part of the National Acid Precipitation Assessment Program (NAPAP), the National Trends Network (NTN) for deposition monitoring was created in 1982 (8). The NTN has

NADP/NTN MONITORING NETWORK

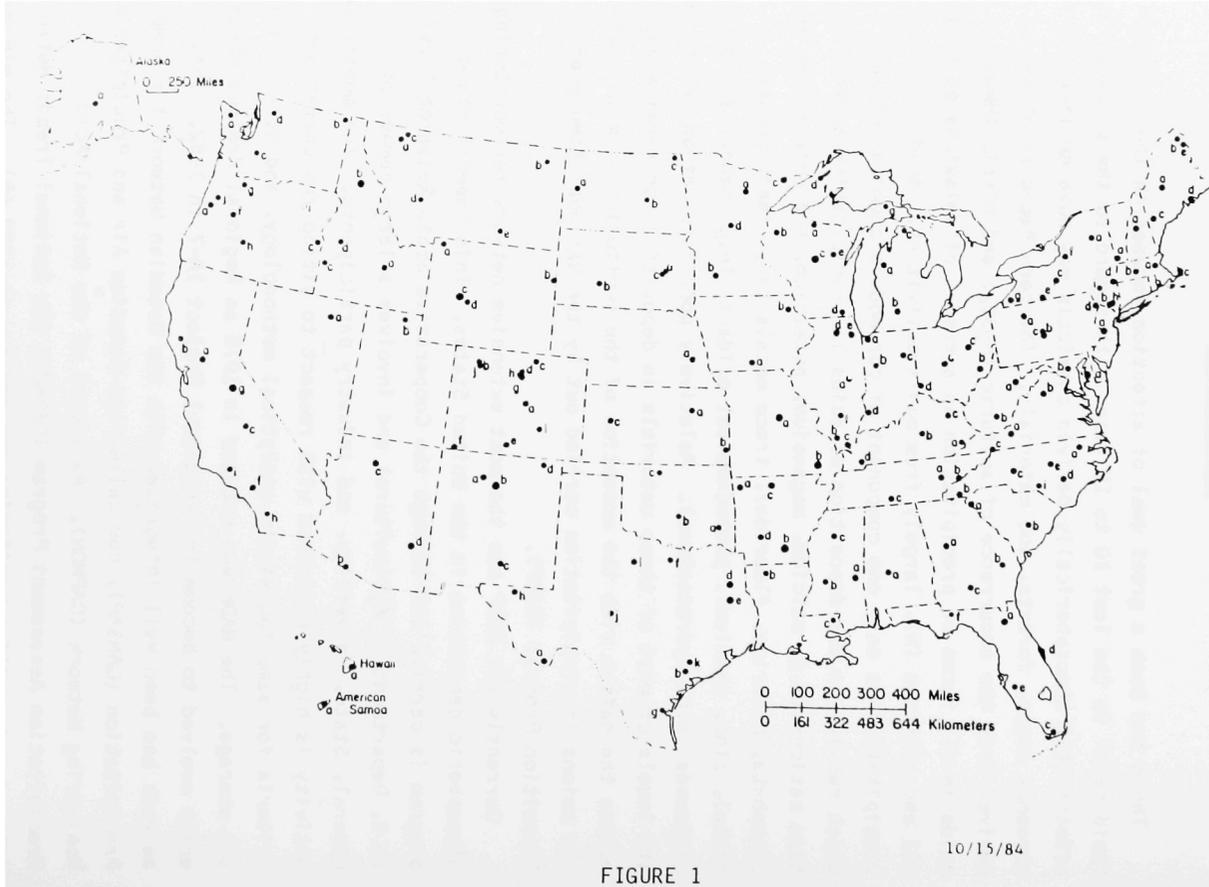


FIGURE 1

over 200 monitoring sites across the nation; the network includes a majority of the NADP sites, sites from other deposition monitoring networks, and some newly created sites to fulfill the objectives of the NTN. It is for this reason that in many instances sites are referred to as being part of the NADP/NTN network. Figure 1 shows a map of the location of sites in the NADP/NTN network as of 1984. The Greenville, Maine site is labelled "a". Maine sites b, c, d, and e locations and funding agencies are Caribou (National Oceanic and Atmospheric Administration), Bridgton (Maine Department of Environmental Protection and the National Park Service), Acadia National Park (Maine Department of Environmental Protection and the National Park Service), and Presque Isle (National Oceanic and Atmospheric Administration and the Maine Agricultural Experiment Station), respectively.

The activities of NADP extend beyond deposition monitoring, and it has as its objectives both (a) the deposition monitoring network research program and (b) an effects research program. Figure 2 illustrates the overall organization of the NADP and how the various activities are coordinated within the program.

Since 1979 the Maine Agricultural Experiment Station at the University of Maine at Orono has operated an NADP monitoring station (Station No. 200935) at Greenville, Maine. This is the first report summarizing data collected from work at the Greenville Station focusing on the period of collections from November 1979 to December 1982.

METHODOLOGY

This report presents results of the analyses of samples collected at the Greenville, Maine NADP station. The Greenville NADP/NTN station is in Piscataquis county at longitude 69°39'52" and latitude 45°29'23" at approximately 322 meters elevation. The site design is in conformance with NADP guidelines (2) and site operation is carried out according to standard NADP protocol (3). Samples are collected from the site each Tuesday morning and returned to the field laboratory

Figure 2 - Organizational Diagram of the National Atmospheric Deposition Program (IR-7)

EXECUTIVE COMMITTEE

Program Chairman
Program Vice Chairman
Program Secretary
Past Program Chairman
Quality Assurance Steering
Committee Chairman
Subcommittee Chairmen

OTHER PROGRAM OFFICERS

SAES Administrative Advisors
Southern Region
Northeastern Region
North Central Region
Western Region
Program Coordinator
Director - Central Analytical Lab.
USDA Representative
USGS Representative
NCSU Acid Precipitation Prog. Coord.

SUBCOMMITTEES

Network Site Criteria and Standards Committee
Methods Development and Quality Assurance Committee
Data Management and Analysis Committee
Effects Research Committee

Effects Research Committee Working Groups

Aquatics
Field and Horticultural Crops
Forestry
Materials

in Greenville for measurements of weight, conductance, and pH. In addition, the strip chart from a Belfort Recording Rain Gage and Event Recorder located adjacent to the deposition collector is retrieved. Samples are then shipped to the Central Analytical Laboratory (Illinois State Water Survey, Champaign, IL) for chemical analysis under a rigorous quality assurance program (6) which is administered by the U.S. Geological Survey.

The site is equipped with an Aerochem Metrics Model 301 Automatic Sensing Wet/Dry Precipitation Collector. We report here analyses from the wet deposition collector bucket only. Wet deposition includes rain, snow, sleet, hail, and heavy dew or fog events. This instrument consists of two plastic buckets with an automatic lid which covers the

wet side bucket except during precipitation events. At the onset of a precipitation event, a sensor causes the lid to automatically move from the wet side bucket to cover the dry side bucket. At the end of each event, the lid automatically returns to its original position.

The data reported here were obtained from the Acid Deposition System (ADS) data base operated by Batelle Pacific Northwest Laboratories funded as part of the National Acid Precipitation Assessment Program by the U.S. Environmental Protection Agency (9), which serves as the centralized data base facilities for NADP. The summary data for 1982 are derived from the NADP 1982 Annual Data Summary (7). All raw data from the Central Analytical Laboratory are submitted to ADS and a copy is kept on file at the University of Maine at Orono. Summary statistics and graphics were generated using SAS and SASGRAPH programs on the mainframe computing facilities at the University of Maine at Orono.

Precipitation collections are continuing at Greenville as part of the NADP/NTN network.

RESULTS AND DISCUSSION

Precipitation Characteristics

Descriptive statistics for pH, H^+ , SO_4^{2-} , NO_3^- and NH_4^+ concentrations ($mg\ l^{-1}$) and deposition ($g\ m^{-2}\ mo^{-1}$) are presented in Table 1. Monthly means are used in this paper throughout, because we found no significant differences in statistical relationships using weekly or quarterly values. The mean pH at Greenville from November 1979 to December 1982 was 4.55 and was dominated by the strong mineral acid anions SO_4^{2-} and NO_3^- . The ratio of SO_4^{2-}/NO_3^- was approximately 1.6/1. Precipitation averaged 90.6 cm annually for the period. Using mean monthly deposition data to calculate annual S and N loading, it appears that the Greenville site received, in precipitation alone, approximately 5.1 kg S $ha^{-1}\ yr^{-1}$ (15.4 kg $SO_4^{2-}\ ha^{-1}\ yr^{-1}$), 2.66 kg NO_3^-

ha⁻¹ yr⁻¹ (9.12 kg NO₃⁻ ha⁻¹ yr⁻¹), and 0.84 kg NH₄-N ha⁻¹ yr⁻¹ (1.08 kg NH₄⁺ ha⁻¹ yr⁻¹). Total N deposition in precipitation (as NO₃⁻ + NH₄⁺) was therefore 3.50 kg N ha⁻¹ yr⁻¹. Contributions of N and S from gaseous and particulate deposition would need to be included to determine total atmospherically derived N and S for the Greenville site. They are not currently measured in the standard NADP operation. Barrie et al. (1) state that dry deposition of S is less than or equal to wet deposition of S, with the importance of the dry deposition contribution decreasing with distance from the source. It is also worthy to note that at high elevations where forest canopies are present, cloud and fog droplet interception results in additional inputs of water and chemicals to the site. Lovett et al. (5) studied forest canopy interception of cloud droplets at 1220 m elevation in a balsam fir stand on Mt. Moosilauke, New Hampshire and determined water and chemical inputs from cloud droplets were 46 percent and 150 to 430 percent of bulk precipitation, respectively.

Table 1 - Descriptive statistics for monthly pH, hydrogen, sulfate, nitrate, and ammonium weighted concentrations and deposition at Greenville, Maine from November 1979 to December 1982.

	MEAN	RANGE	STANDARD DEVIATION
CONCENTRATIONS (mg l ⁻¹)			
pH	4.55	4.18 - 5.00	0.26
H ⁺	0.032	0.010 - 0.066	0.015
SO ₄ ²⁻	1.73	0.50 - 3.77	0.86
NO ₃ ⁻	1.09	0.44 - 3.15	0.56
NH ₄ ⁺	0.13	0.02 - 0.41	0.10
DEPOSITION (g m ⁻² mo ⁻¹)			
H ⁺	0.002	0.001 - 0.008	0.001
SO ₄ ²⁻	0.128	0.020 - 0.380	0.077
NO ₃ ⁻	0.076	0.020 - 0.190	0.037
NH ₄ ⁺	0.009	0.001 - 0.039	0.008
Precip. (cm)	7.55	1.90 - 25.90	4.38

MAINE AGRICULTURAL EXPERIMENT STATION TECHNICAL BULLETIN 118

Descriptive statistics were calculated for precipitation chemistry data from the 65 NADP sites reported in the 1982 annual summary (7) and are summarized in Table 2. A comparison between the mean monthly Greenville data (Table 1) and national annual averages (Table 2) suggests that the Greenville site receives precipitation slightly more acidic than the national average and well below the highest average pH reported in 1982 for the K-Bar site in Texas (pH=5.64). Sulfate and NO_3^- concentrations at Greenville were similar to national averages, but NH_4^+ was roughly half the national average.

Table 2 - Descriptive statistics for precipitation weighted annual average concentrations for pH, hydrogen, sulfate, nitrate, and ammonium for all NADP sites (n=63) reported for 1982 (7).

	MEAN	RANGE	STANDARD DEVIATION
CONCENTRATIONS (mg l^{-1})			
pH	4.70	4.16 - 5.64	0.41
H^+	0.028	0.002 - 0.069	0.019
SO_4^{2-}	1.78	0.27 - 3.51	0.85
NO_3^-	1.08	0.07 - 2.16	0.46
NH_4^+	0.24	0.02 - 0.53	0.12

In general, the highest precipitation SO_4^{2-} concentrations nationally occur in the Ohio, Pennsylvania, and New York regions with generally lower SO_4^{2-} concentrations west of the Mississippi River. Nitrate concentrations in precipitation are roughly similar in distribution to SO_4^{2-} , while the region of relatively high NH_4^+ concentrations is near Iowa and surrounding states. Overall, the Greenville site falls within the northeast region receiving acid precipitation dominated by SO_4^{2-} and NO_3^- anions, but is north of the most heavily acidified region based on annual average precipitation chemistry data for the region.

Table 3 illustrates detailed information on precipitation characteristics at the Greenville site for 1982 as summarized by the Acid Deposition System (ADS) for statistical reporting (6). Percentile ranks for concentrations of the various parameters provide an idea of the distribution of values which could be expected for the various parameters. Precipitation weighted means are calculated using the volume of precipitation collected in the rain gage along with the chemical analysis of samples from the wet-side bucket; geometric means are calculated as the arithmetic average of the natural logarithms of the data values followed by exponentiating the results; arithmetic means are calculated directly from wet-bucket sample concentration data. Deposition is calculated as the product of the weighted mean concentrations and the total measured concentration for the period under consideration.

Statistical Correlations

Because precipitation is a major process for transporting chemicals from the atmosphere to the ground, we might expect that greater precipitation amounts would result in a greater "washing out" of materials from the atmosphere but lower concentrations due to dilution. Table 4 supports this hypothesis using correlations between precipitation amount (cm) and ionic concentration and deposition data. Negative correlations between precipitation amount and ion concentrations reflect different processes involved in low precipitation events (such as drizzle) and high intensity precipitation events. The dilution effect appears most pronounced with NO_3^- . Positive correlations with deposition suggest that the net effect of greater precipitation amounts is greater total loading of chemicals to the landscape. The highly significant correlation with SO_4^{2-} deposition could be taken to suggest that the SO_4^{2-} source for precipitation diminishes less than NO_3^- with greater amounts of precipitation. The correlations with SO_4^{2-} and NO_3^- concentration support this concept.

Table 3 - Detailed summary of 1982 data for weekly precipitation chemistry at Greenville, Maine.

	Estimated Precip. (cm)	pH	Conductivity ($\mu\text{S cm}^{-1}$)	Equivalent Ratios		
				$\text{SO}_4^{2-}/\text{NO}_3^-$	$\frac{\text{SO}_4^{2-} + \text{NO}_3^-}{\text{H}^+}$	$\frac{\text{Cation}}{\text{Anion}}$
Precip. Weighted Mean		4.54				
Geometric Mean	0.91	4.66	16.1	1.72	2.22	0.92
Arithmetic Mean	1.65	4.68	20.4	3.63	4.89	0.98
Minimum Value	0.01	4.00	3.3	0.18	0.61	0.65
Percentile 10	0.11	4.08	5.7	0.33	1.24	0.72
Percentile 25	0.54	4.31	9.3	1.33	1.46	0.79
Percentile 50	1.38	4.53	17.4	2.22	1.72	0.88
Percentile 75	2.41	4.98	29.1	2.68	2.39	0.96
Percentile 90	3.90	5.26	41.1	3.50	4.04	1.19
Maximum Value	6.76	6.19	61.5	80.75	92.38	3.03

	Ion Concentrations (mg l^{-1})								
	Ca^{2+}	Mg^{2+}	K^+	Na^+	NH_4^+	NO_3^-	Cl^-	SO_4^{2-}	H^+
Precip. Weighted Mean	0.08	0.026	0.029	0.101	0.12	0.96	0.13	1.58	2.88×10^{-2}
Geometric Mean	0.09	0.031	0.033	0.078	0.08	0.96	0.12	1.27	2.08×10^{-2}
Arithmetic Mean	0.14	0.046	0.031	0.162	0.17	1.45	0.19	1.87	3.26×10^{-2}
No. Below Detection	1	0	1	0	11	1	3	2	0
Minimum Value	0.01	0.006	0.003	0.003	0.02	0.02	0.02	0.10	6.46×10^{-4}
Percentile 10	0.03	0.010	0.007	0.020	0.02	0.28	0.03	0.49	5.48×10^{-3}
Percentile 25	0.05	0.017	0.008	0.040	0.03	0.47	0.06	0.73	1.04×10^{-2}
Percentile 50	0.09	0.028	0.016	0.076	0.08	1.19	0.13	1.46	2.99×10^{-2}
Percentile 75	0.15	0.051	0.030	0.147	0.25	2.01	0.25	2.57	4.90×10^{-2}
Percentile 90	0.47	0.109	0.075	0.274	0.40	3.20	0.39	3.97	8.34×10^{-2}
Maximum Value	0.66	0.201	0.231	2.250	1.16	5.59	1.25	9.08	1.00×10^{-1}

Table 4 - Correlation coefficients (r) between monthly precipitation volume and the concentration and deposition of hydrogen, sulfate, nitrate and ammonium at Greenville, Maine from November 1979 to December 1982.

Precipitation		
	<u>r</u>	<u>Prob >r</u>
hydrogen concentration	-0.31	0.07
sulfate concentration	-0.28	0.10
nitrate concentration	-0.44	0.01
ammonium concentration	-0.28	0.11
hydrogen deposition	0.39	0.02
sulfate deposition	0.49	0.003
nitrate deposition	0.36	0.04
ammonium deposition	0.30	0.08

Correlation coefficients between pH, H^+ , SO_4^{2-} , NO_3^- and NH_4^+ (Table 5) show that higher precipitation pH values are associated with lower concentrations of the other ions, which would result when H_2SO_4 and HNO_3 were the dominant sources of acidity in solution. Both SO_4^{2-} and NO_3^- were highly correlated with H^+ , with SO_4^{2-} having the highest r value, yet SO_4^{2-} had the poorest correlation with NO_3^- . From the correlation coefficients with NH_4^+ , we could hypothesize that $(NH_4)_2SO_4$ aerosols contributed to SO_4^{2-} in precipitation as well as H_2SO_4 , and due to a more diverse source of SO_4^{2-} than for NO_3^- , a poor correlation between SO_4^{2-} and NO_3^- resulted. Gorham et al. (4) also reported a strong correlation between H^+ and SO_4^{2-} ($r^2 = 0.85$) and less so for H^+ and NO_3^- ($r^2 = 0.63$) for 33 stations in the eastern United States. However, these authors found good correlations between SO_4^{2-} and NO_3^- ($r^2 = 0.73$) which was not evident in the Greenville data.

Table 5 - Correlation coefficients (r) between mean monthly concentrations of hydrogen, sulfate, nitrate, ammonium and pH in precipitation at Greenville, Maine from November 1979 to December 1982.*

	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	H ⁺
pH	-0.82 (0.0001)	-0.69 (0.0001)	-0.79 (0.0001)	-0.97 (0.0001)
SO ₄ ²⁻		0.33 (0.05)	0.90 (0.0001)	0.83 (0.0001)
NO ₃ ⁻			0.37 (0.03)	0.70 (0.0001)
NH ₄ ⁺				0.83 (0.0001)

* Numbers in brackets indicate the significance level.

Monthly Precipitation Data

Figure 3 shows monthly precipitation weighted mean pH data from November 1979 to December 1982. Both yearly and seasonal variation are evident with all the parameters measured, and no statistically significant increasing or decreasing trend is evident for pH through the period covered. Values for pH ranged from 4.18 to 5.00. Figure 4 demonstrates the variation in monthly precipitation volumes with a tendency for higher volumes to occur in the fall and early summer, and lower volumes in the winter months.

Figures 5 through 12 illustrate precipitation weighted mean monthly values for the concentrations (mg/l) and deposition (g m⁻² mo⁻¹) of H⁺, SO₄²⁻, NO₃⁻ and NH₄⁺. The starting dates along the X axes represent the first collection day for each four week period included in each monthly average. In general, SO₄²⁻ concentrations were greater during the summer months, which coincides with typically greater amounts of precipitation during the summer season. The net results are fairly well defined peaks of SO₄²⁻ deposition (Figure 10) during summers. These data agree well with regional trends for the

northeast which generally show greater concentrations of SO_4^{2-} and lower pH in precipitation during the summer months (NADP 1985). Concentrations of NO_3^- appear much more variable than SO_4^{2-} , but tend to be greater in the winter and spring. Concentrations of NH_4^+ are distinctly greater in the spring and summer, which results in greater NH_4^+ deposition during these periods as well (Figure 12). If wind blown aerosols are a major source of NH_4^+ in precipitation, then frozen ground and snow covered soil conditions in the winter could explain the seasonal pattern of low NH_4^+ concentrations in precipitation during the winter months.

SUMMARY

Precipitation data for the National Atmospheric Deposition Program (NADP) station at Greenville, Maine are reported for the period from November 1979 to December 1982. Precipitation pH averaged 4.55 for the period and ranged from 4.18 to 5.00 on a monthly basis. Precipitation volume was negatively correlated with concentration (mg l^{-1}) and positively correlated with deposition quantity (g m^{-2}) for H^+ , SO_4^{2-} , NO_3^- and NH_4^+ . Significant positive correlations between H^+ and SO_4^{2-} , NO_3^- and NH_4^+ were evident. While the strongest positive correlation for ion pairs existed between NH_4^+ and SO_4^{2-} , a very weak correlation was evident between NH_4^+ and NO_3^- . A series of bar graphs is shown providing monthly means for precipitation volume and pH as well as concentrations and deposition of H^+ , SO_4^{2-} , NO_3^- and NH_4^+ .

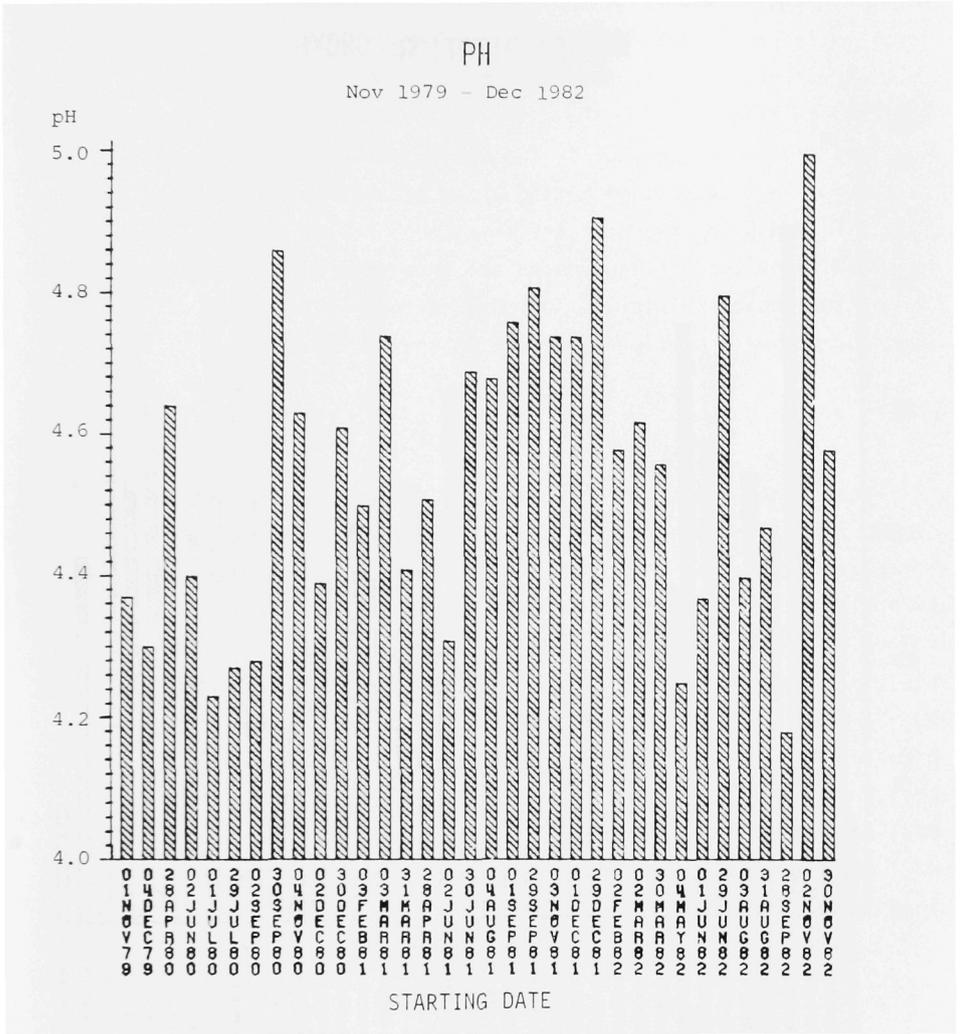


FIGURE 3

HYDROGEN CONCENTRATION

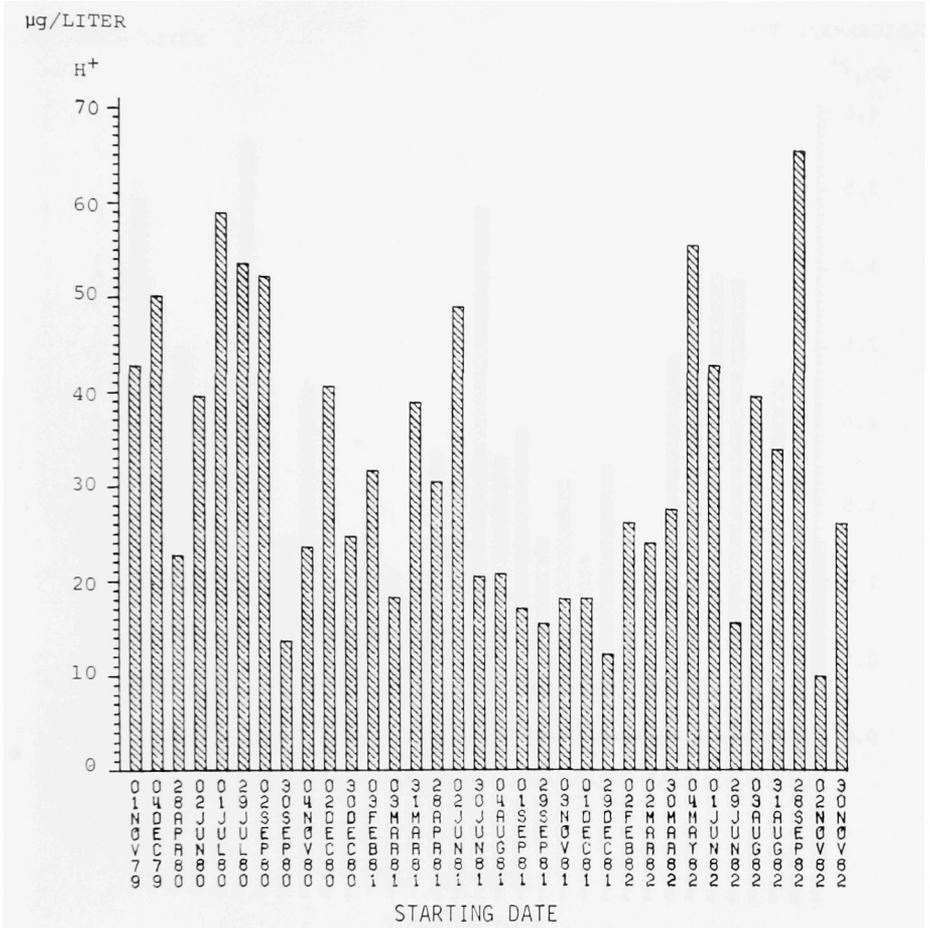


FIGURE 5

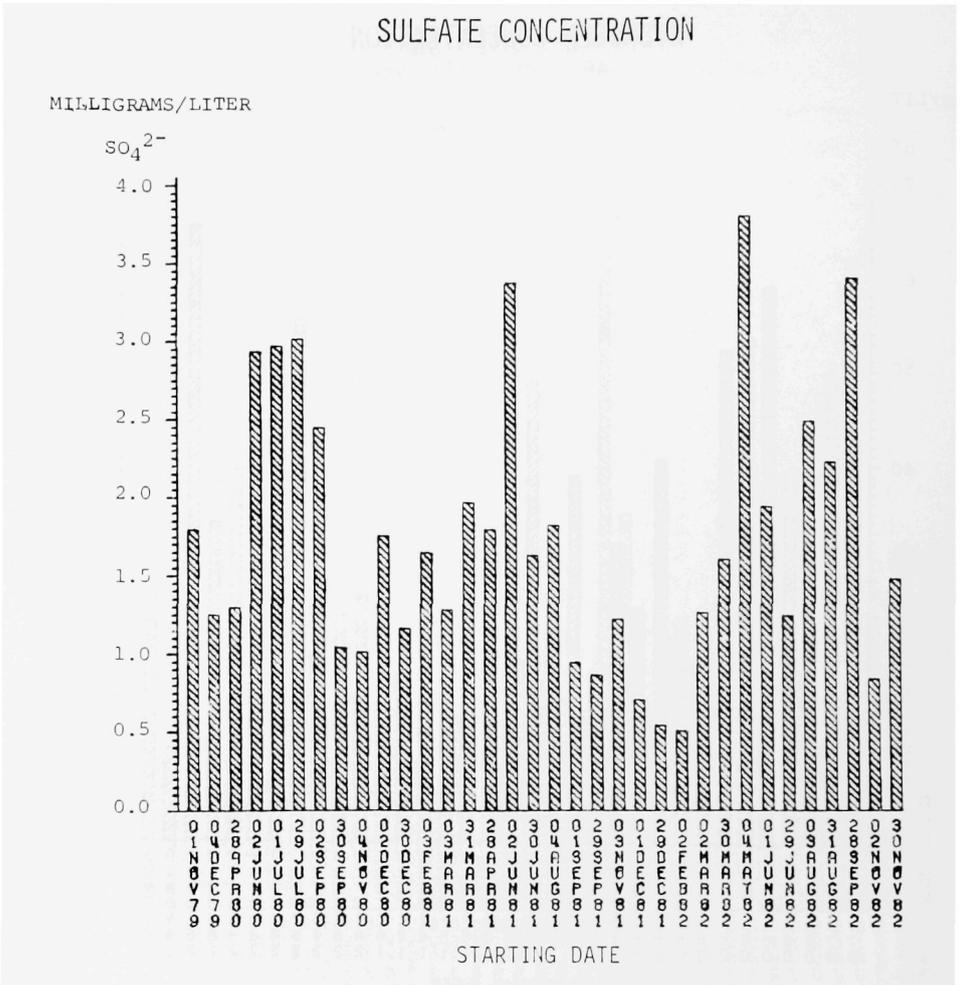


FIGURE 6

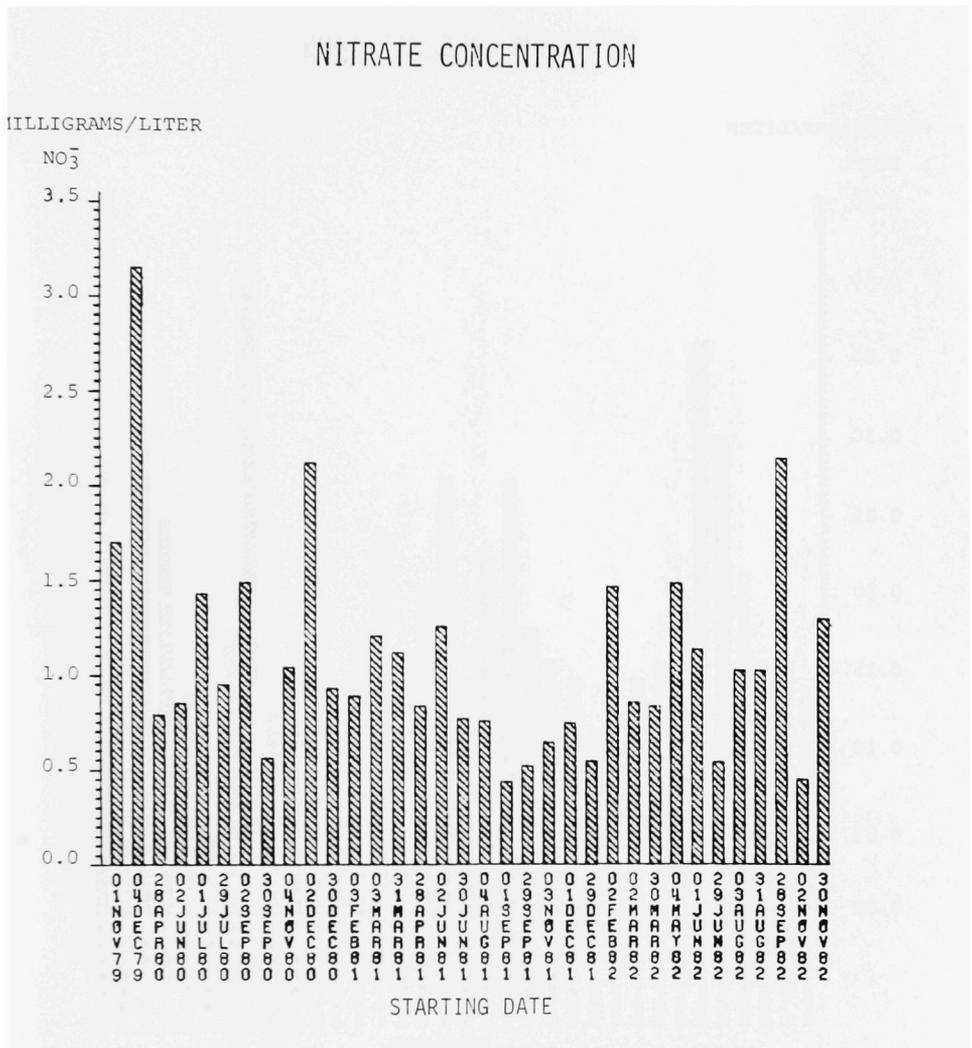


FIGURE 7

AMMONIUM CONCENTRATION

MILLIGRAMS/LITER

NH₄⁺

0.45

0.40

0.35

0.30

0.25

0.20

0.15

0.10

0.05

0.00

9-1-70
9-15-70
9-29-70
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10-27-70
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11-24-70
12-8-70
12-22-70
1-5-71
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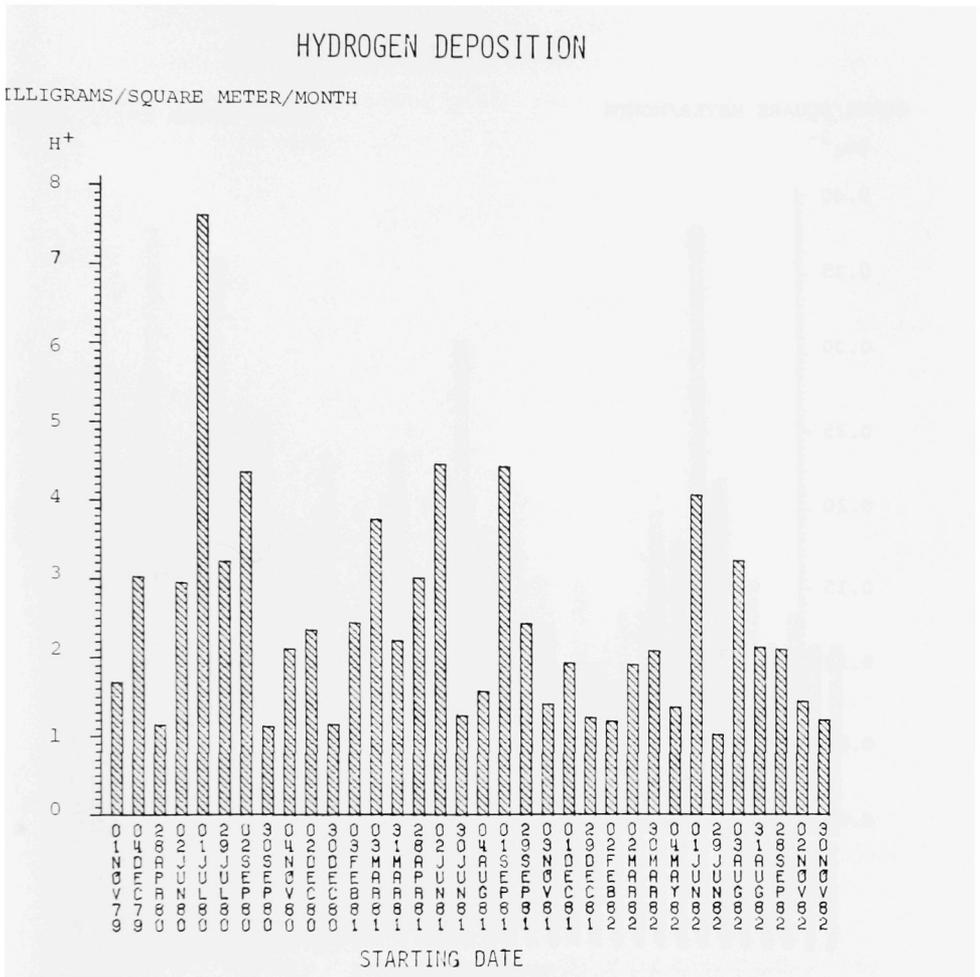


FIGURE 9

SULFATE DEPOSITION

GRAMS/SQUARE METER/MONTH

SO₄²⁻

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0.35
0.30
0.25
0.20
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0.10
0.05
0.00

01NOV79 004DEC79 002JAN80 000FEB80 002MARCH80 003APRIL80 000MAY80 003JUNE80 000JULY80 002AUG80 001SEPT80 003OCT80 002NOV80 000DEC80 001JAN81 003FEB81 002MARCH81 000APRIL81 003MAY81 001JUNE81 002JULY81 000AUG81 001SEPT81 003OCT81 002NOV81 000DEC81 001JAN82 003FEB82 002MARCH82 000APRIL82 001MAY82 003JUNE82 002JULY82 000AUG82 001SEPT82 003OCT82 002NOV82 000DEC82

STARTING DATE

FIGURE 10

NITRATE DEPOSITION

GRAMS/SQUARE METER/MONTH

NH₃

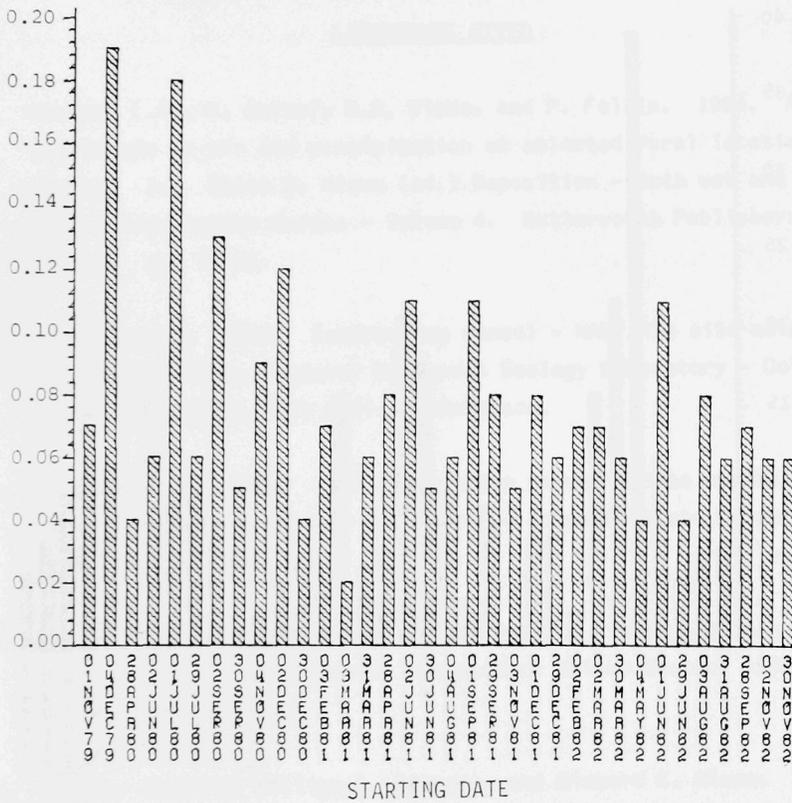


FIGURE 11

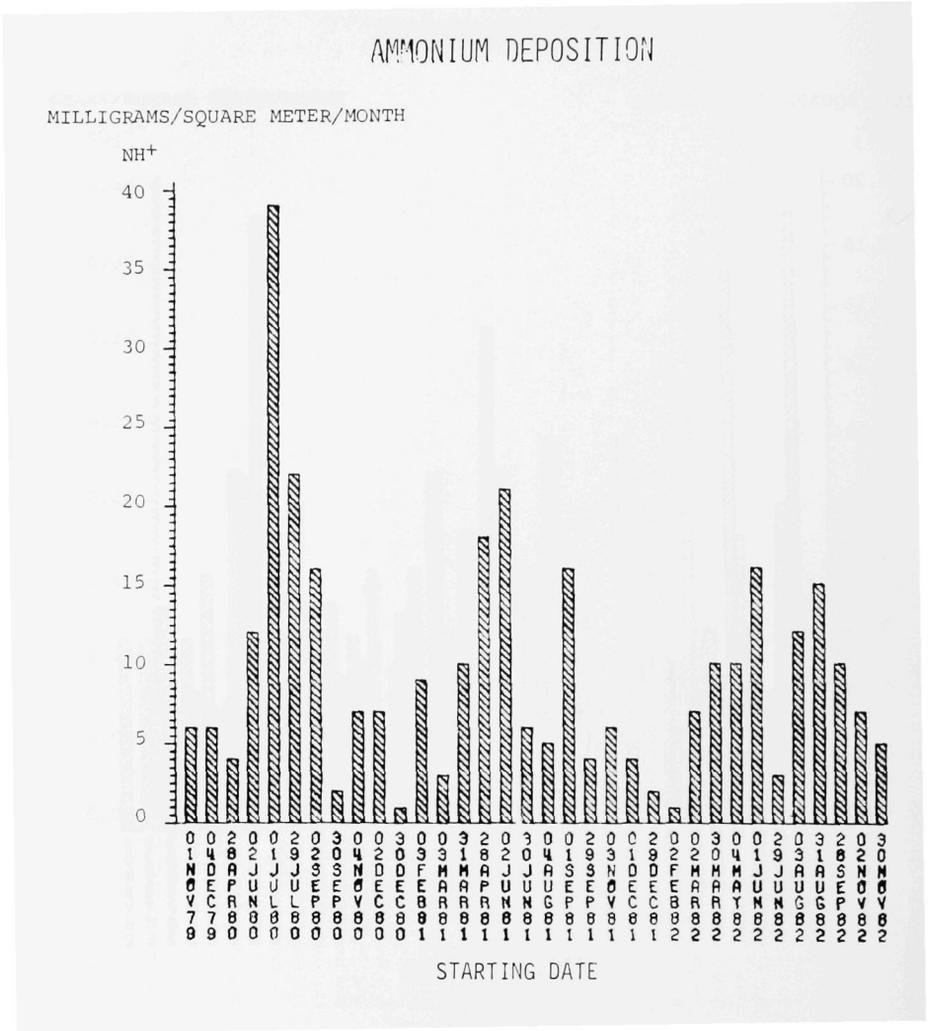


FIGURE 12

ACKNOWLEDGEMENTS

The authors wish to acknowledge the assistance of Mark Anderson, Eunice Padley and Mary Thibodeau in the ongoing NADP/NTN work and preparation of this manuscript.

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