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A. R. Olsen

July 1989

Prepared for
the U.S. Environmental Protection Agency
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Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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ABSTRACT

The focus of this report is on North American wet deposition temporal patterns from 1979 to 1986 and spatial patterns for 1986. The report provides statistical distribution summaries of annual precipitation-weighted average concentration and annual deposition for nine ion species: hydrogen, sulfate, nitrate, ammonium, calcium, chloride, sodium, potassium, and magnesium. The data in the report are from the Acid Deposition System (ADS) for the statistical reporting of North American deposition data. The data from ADS included in the report are from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN), the MAP3S precipitation chemistry network, the Utility Acid Precipitation Study Program (UAPSP), the Canadian Precipitation Monitoring Network (CAPMoN), and the daily and 4-weekly Acidic Precipitation in Ontario Study (APIOS-D and APIOS-C).

Isopleth maps, based on surface estimation using kriging, display concentration and deposition spatial patterns of pH, hydrogen, sulfate, nitrate, ammonium, and calcium ion species for 1986 annual, winter, and summer periods. The temporal pattern analyses use a subset of 30 sites over an 8-year (1979-1986) period and an expanded subset of 137 sites with greater spatial coverage over a 5-year (1982-1986) period. The 8-year period represents the longest period with wet deposition monitoring data available that has a sufficient number of sites with data of known quality to allow a descriptive summary of annual temporal patterns.

Annual total precipitation and annual precipitation weighted ion concentrations for 1979-1986 exhibit substantial inter-annual variation for the subset of 30 sites. The precipitation variation pattern is out of phase with the concentration patterns. Annual wet deposition patterns are weaker than those for annual concentration. Based on Sen's median trend estimate, all ion species, except nitrate and ammonium, show decreasing patterns for monthly concentration and deposition for more than 50% of the sites. Kendall seasonal tau (KST) tests found that all ion species have significantly decreasing trends at one or more sites. In particular, 33% and 26% of the 270 (30 sites by 9 ions) KST tests are significant (decreasing) for concentration and deposition, respectively. The percent decrease for concentration and

deposition is as great or greater for calcium, chloride, sodium, potassium and magnesium as for sulfate, hydrogen, nitrate and ammonium.

ACKNOWLEDGEMENTS

The preparation of this report has relied heavily on the expertise and assistance of many other individuals. Judy Sweeney, Trudy Erb, and Michelle Thomas of the Acid Deposition System (ADS) staff maintained the data and provided annual, seasonal, and monthly summaries for all networks from 1979 through 1986. Jeanne Simpson and Dave Engel completed all of the computing for the spatial analyses and associated isopleth map production. The final maps reflect the expertise of the Pacific Northwest Laboratory (PNL) graphics staff. Susan Cammann used her expertise to help organize the report, especially the preparation of the figures.

During the last several years a small group of researchers have worked together on related projects. This group knowingly and unknowingly have had a major impact on the structure of the report. Don Daly, Mike Kosorok, and Dan Carr previously developed temporal pattern analysis software that was adapted to the current study. Rich Barchet is a key member of our temporal and spatial pattern group of researchers. His experience in prior analyses and his lead contribution to preparation of related reports is reflected in the current study. This report would not be the same without these individuals.

This research has been funded as part of the National Acid Precipitation Assessment Program by the U.S. Environmental Protection Agency.

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1.0 SUMMARY

The focus of this report is on the temporal and spatial patterns of wet deposition occurring in North America. The report gives a descriptive statistical summary of wet deposition in terms of ion species concentration and deposition. Isopleth maps display 1986 annual and seasonal spatial patterns for pH, hydrogen, sulfate, nitrate, ammonium, and calcium wet deposition. The intent of the isopleth maps is to give a general overview of regional differences in wet deposition. Descriptive graphical displays of annual wet deposition at monitoring sites and Kendall seasonal tau statistical trend analyses provide information on the temporal patterns that existed during two time periods. The 1979-1986 time period is the longest continuous period when a reasonable number of wet deposition sites operated in the United States to enable temporal patterns to be examined. The shorter 5-year, 1982-1986, time period is used to expand the spatial coverage of the sites included in the temporal pattern analyses. Neither time period is sufficiently long to provide estimates of long-term trends.

The report consists of four sections and an extensive appendix. Section 1 gives an overall summary for the report and follows the general structure of the main body of the report. Section 2 describes the sources for the wet deposition data, the data quality and summarization procedures applied, the selection of sites included in the spatial and temporal pattern analyses, the spatial estimation methodology used, and the temporal pattern methodology used. Section 3 describes annual temporal patterns for nine ion species observed between 1979-1986 and their spatial pattern in 1986. Section 4 contains a detailed study of ion species temporal patterns based on a Kendall seasonal tau analysis of monthly data. Because the number of tables and figures used is large, they are placed together in an appendix.

1.1 SOURCE OF WET DEPOSITION DATA

Various federal, state, and local agencies and private industries support networks of sites for the collection and chemical analysis of precipitation samples. The data in this report are from networks that contribute data to the Acid Deposition System (ADS) (Watson and Olsen 1984) for statistical reporting of North American wet deposition data. Other networks collect precipitation chemistry data but did not contribute data to the ADS for the period 1979 to 1986.

The spatial and temporal pattern analyses include data from six regional networks. The Multi-State Atmospheric Pollution and Power Production study (MAP3S) initiated the MAP3S precipitation chemistry network (MAP3S/PCN) in 1976 with four sites in the northeastern United States to begin research on the scavenging of pollutants by precipitation. It was the first wet-deposition-only, event sampling network. In 1978 the National Atmospheric Deposition Program (NADP) established a weekly sampling network to monitor trends in precipitation chemistry in the United States. The NADP, in cooperation with federal, state, and private research agencies, was created to conduct research on atmospheric deposition and its effects. In 1982, the NADP assumed responsibility for coordinating the operation of the National Trends Network (NTN) of the federally-supported National Acid Precipitation Assessment Program (NAPAP). The merged networks now have the designation NADP/NTN. The daily sampling network designated as the Utility Acid Precipitation Study Program (UAPSP) began in 1978 as a nine-site research network initiated by the Electric Power Research Institute and expanded to twenty-two sites giving broader regional coverage in 1982 under the Utility Acid Precipitation Study Program. The daily Canadian federal network, CAPMoN, was formed using sites from two previous networks as a base: the Canadian Network for Sampling Acid Precipitation (1977-1985) and the Air and Precipitation Monitoring Network (1978-1985). In 1980, Ontario province established two networks as part of the Acidic Precipitation in Ontario Study (APIOS). The cumulative network, APIOS-C with a four-week sampling period, is designed to determine the long-term deposition pattern in Ontario. The daily network, APIOS-D, is designed to define the sector of origin of the ion species as well as the frequency and intensity of acidic deposition episodes.

Each network has its own set of specific protocols for selecting site locations, field operation procedures, laboratory analysis procedures, and data screening procedures. Networks implement these protocols to ensure the collection, laboratory analysis, and reporting of wet deposition data of consistent and known quality. Each network included in this report has and implements a quality assurance program. In addition, the current study uses the valid sample criteria and Unified Deposition Database Committee (UDDC) data completeness measures (Sweeney and Olsen 1987).

The selection of sites for the spatial pattern analyses applies the UDDC criteria. However, the selection of sites for the temporal pattern analyses uses a relaxed criteria for selection of a representative summary based on the data completeness measures. The criteria are a compromise between ensuring each summary is based on very complete data, i.e., meet UDDC criteria, and increasing the number of sites available for temporal pattern analysis. The criteria are that precipitation monitoring cover at least 90% of the annual period and that valid sample chemistry data represent at least 60% of the annual precipitation measured. Although the thresholds are the same as the UDDC criteria, the criteria do not apply the quarterly UDDC requirements or the other three UDDC data completeness measures. The number of sites meeting the relaxed criteria (Table 1.1) is substantially greater than those meeting the UDDC criteria. Over 94% of the sites operating a full year met the relaxed criteria each year.

For a site to be included in the 1979-1986 temporal pattern analysis a site must operate from 1979 to 1986 and meet the relaxed data completeness criteria in 1979 or before, and in at least 6 of the 8 years. Thirty sites meet this requirement: 17 from NADP/NTN, 8 from MAP3S, and 5 from UAPSP. This subset of sites is termed the 1979-1986 trend subset. Because of the

TABLE 1.1. Number of Sites by Network and Year Meeting the Relaxed Annual Summary Data Completeness Criteria for Sulfate

Network	1979	1980	1981	1982	1983	1984	1985	1986
NADP/NTN	20	35	66	82	97	133	168	175
MAP3S/PCN	8	8	9	9	9	9	9	9
UAPSP	18	6	6	21	20	19	21	24
CAPMoN	-	-	-	-	7	17	18	21
APIOS-D	-	-	7	16	16	14	14	13
APIOS-C	-	-	26	30	32	32	31	30
Total	46	49	114	158	181	224	261	272

limited number of sites in the 1979-1986 trend subset, the report includes secondary temporal pattern analyses for the 5-year period 1982 to 1986. Inclusion of sites for this 1982-1986 trend subset requires a site to operate from 1982 to 1986 and to meet the relaxed criteria in 1982 or before, and in at least 3 of the 5 years. A total of 137 sites meet this requirement: 71 from NADP/NTN, 9 from MAP3S, 18 from UAPSP, 12 from APIOS-D, and 27 from APIOS-C. The location of the sites in each of the subsets is presented in Figure 1.1 (see Tables A.1 and A.2 for site names).

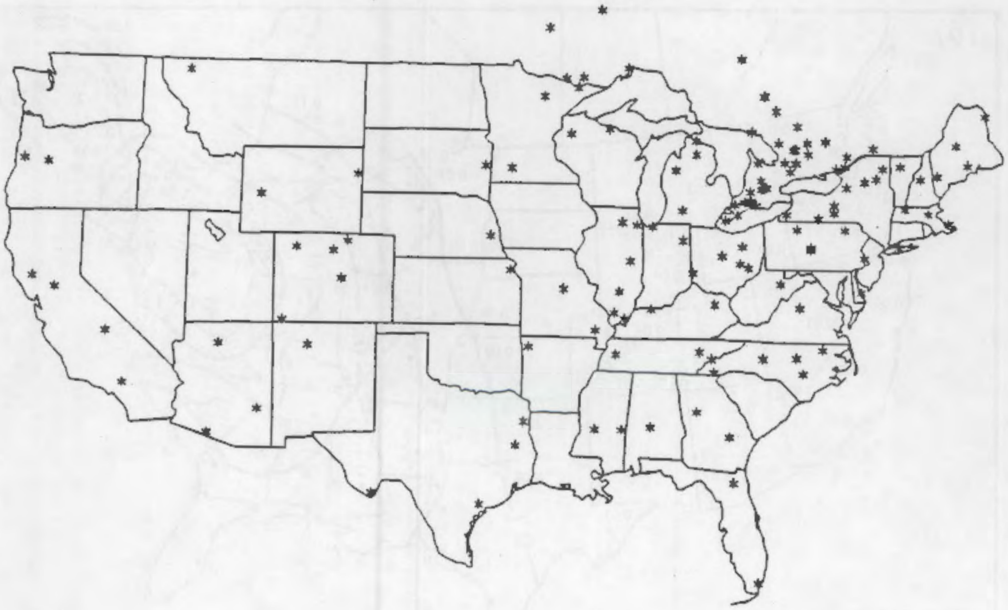
All monthly, seasonal and annual summary statistics are calculated using the procedures established by the Unified Deposition Database Committee. In particular, ion concentrations are precipitation-weighted means and total wet deposition is the product of the precipitation-weighted mean and the total precipitation measured during the summary period.

1.2 SPATIAL PATTERNS IN 1986

Annual, winter, and summer wet deposition spatial patterns for 1986 are presented in Section 3.0 for pH, hydrogen, sulfate, nitrate, ammonium, and calcium concentration and deposition. Figures 1.2, 1.3, and 1.4 present isopleth maps of annual deposition for these ions. The maps are the display of a spatial surface estimated by applying the spatial interpolation technique termed simple kriging.

The 1986 annual spatial pattern for pH (Figure 1.2) shows pH values below 5.0 for the eastern half of North America and pH values greater than 5.0 for western half, except for the Southwest and southern Rocky Mountain region. An ellipsoidal area of low pH (<4.2) extends from mid-Ohio and mid-West Virginia to western New York and from the southern Ontario border to eastern Pennsylvania and northern Virginia. Fourteen sites have a pH value below 4.2 and the lowest two values are 4.12 at Kane, Pennsylvania, and Parsons, West Virginia. The maximum pH is 5.66 at Las Animas in southeastern Colorado. The 1986 spatial pattern for hydrogen ion deposition (Figure 1.2) has the region of greatest deposition (>0.6 kg/ha) over West Virginia, eastern Ohio, Pennsylvania, New York, southern Vermont, western Massachusetts, and Connecticut. Although the isopleth lines give the impression of a reasonably smooth deposition surface, the spatial estimation procedure has smoothed the

1979-86 Trend Site Locations



1982-86 Trend Site Locations

FIGURE 1.1 Location of Sites Included in Trend Subsets for 1979-1986 and for 1982-1986.

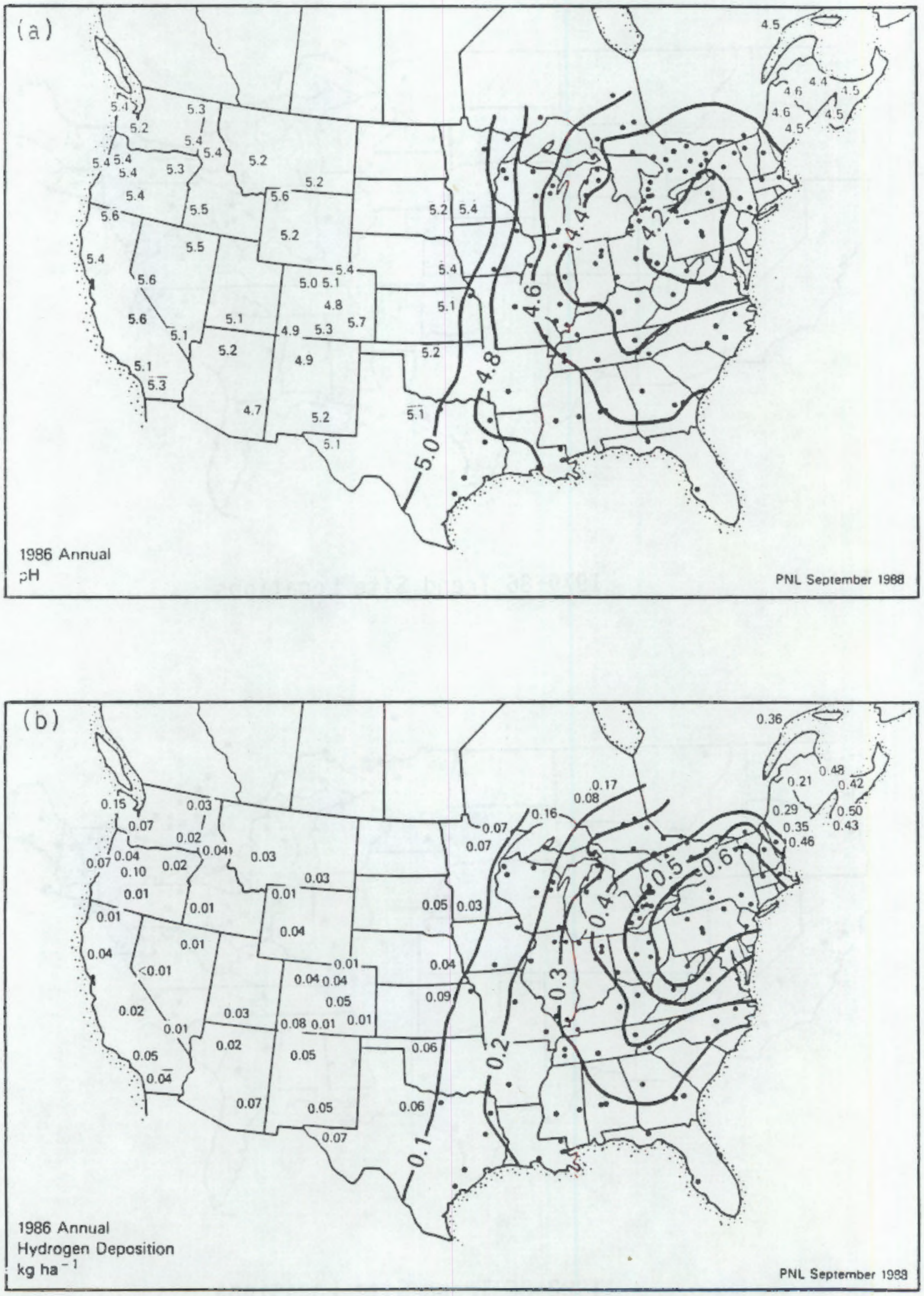


FIGURE 1.2. The 1986 Spatial Pattern of (a) Annual Precipitation-weighted pH and (b) Annual Hydrogen Ion Deposition

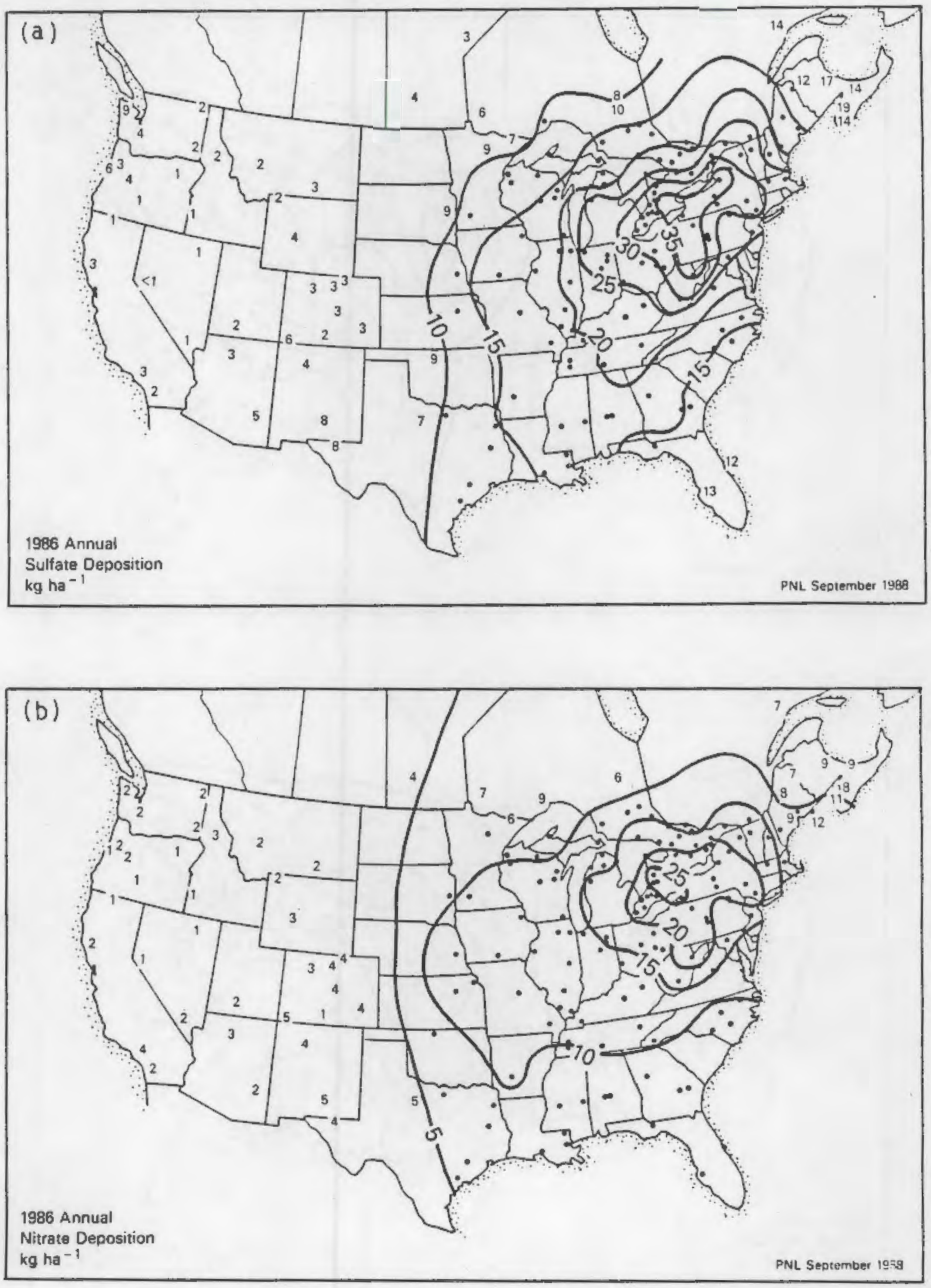


FIGURE 1.3. The 1986 Spatial Pattern of (a) Sulfate Annual Deposition and (b) Nitrate Annual Deposition

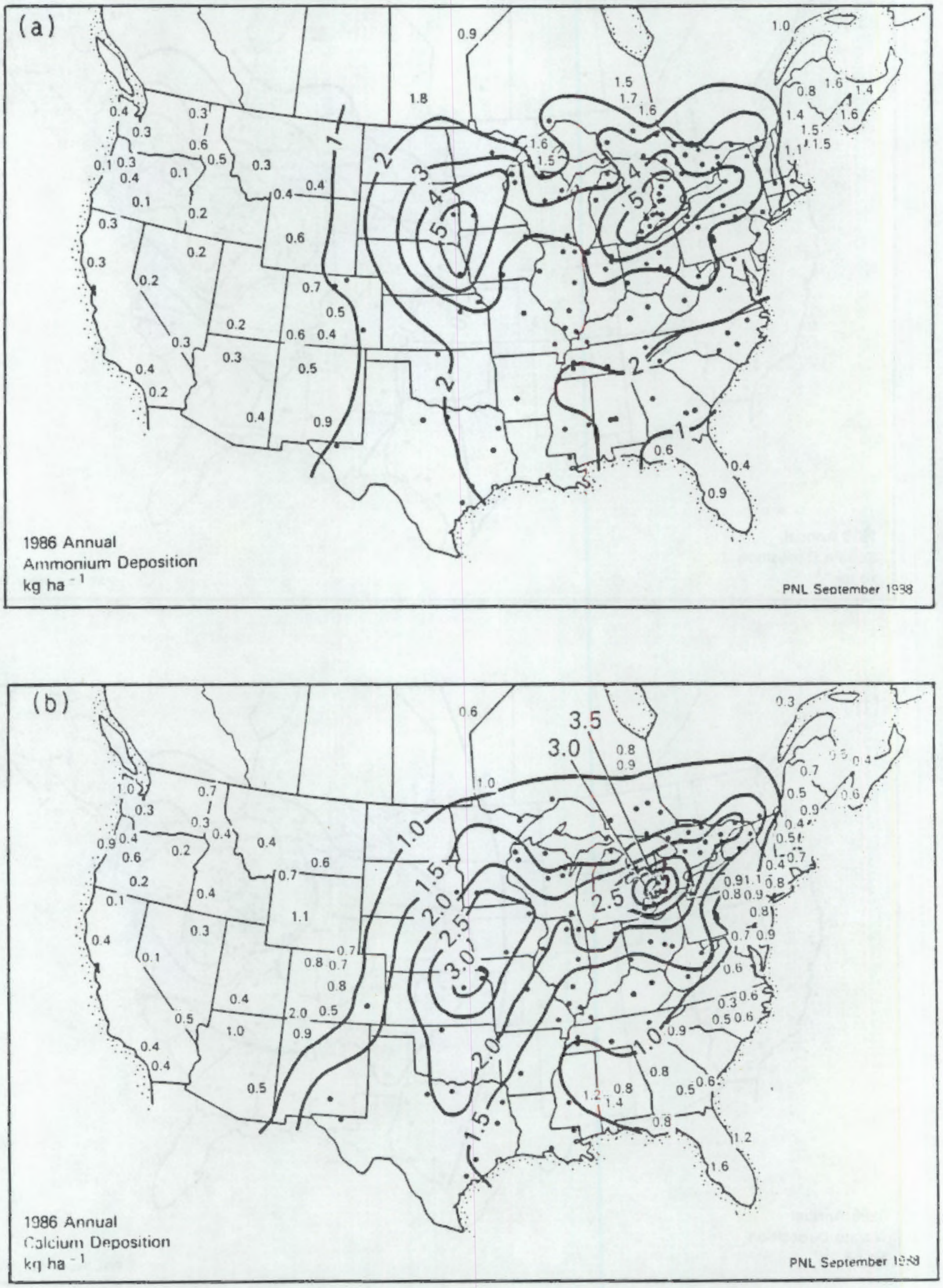


FIGURE 1.4. The 1986 Spatial Pattern of (a) Ammonium Annual Deposition and (b) Calcium Annual Deposition

substantial site-to-site variation in the high deposition area. For example, Sutton, Quebec has a deposition of 0.86 kg/ha and a number of sites with deposition greater than 0.6 kg/ha in southern Ontario are intermixed with sites with deposition less than 0.6 kg/ha. The maximum annual hydrogen deposition is 1.02 kg/ha at Parsons, West Virginia. Other sites with deposition greater than 0.8 kg/ha are Kane and Leading Ridge, Pennsylvania and Bennett Bridge, New York.

Sulfate annual deposition has a spatial pattern similar to sulfate concentration (Figure 1.3). A contour line for annual deposition greater than 35 kg/ha extends from northern West Virginia to north of Lake Ontario and from the eastern Ohio border to western Pennsylvania and western New York. The maximum measured deposition is 53.3 kg/ha at Parsons, West Virginia. Four additional sites have sulfate depositions between 40 and 45 kg/ha: Bennett Bridge and Chautauqua, New York; Kane, Pennsylvania; and Wellesley, Ontario. The 20 kg/ha contour line encloses an elliptical region from northern Alabama and Georgia to southern Ontario and New Hampshire and from eastern Wisconsin and central Illinois to eastern Virginia and Massachusetts. Annual nitrate deposition has a spatial pattern similar to sulfate deposition (Figure 1.3). A 25.0 kg/ha contour encloses a small region centered at the northern tip of Lake Ontario. Eight sites have nitrate depositions greater than 25.0 kg/ha: Bennett Bridge, Parsons, Chautauqua, and West Point in the United States, and Wellesley, North Easthope, Huron Park, and Sutton in Canada. Bennett Bridge has the maximum deposition at 32.4 kg/ha.

The annual ammonium deposition spatial pattern has two regions of high deposition (Figure 1.4). In addition, deposition is greater than 1.0 kg/ha east of the Rocky Mountains, except for Florida. The 2.0 kg/ha contour extends south to the Gulf Coast in Texas, Louisiana, and Mississippi. The maximum deposition is 7.50 kg/ha at Wellesley, Ontario. Five other sites in southern Ontario have depositions greater that 6.0 kg/ha. The highest deposition in the West is 5.39 kg/ha at Brookings, South Dakota. Mead, Nebraska, and Lamberton, Minnesota also are western sites with deposition greater than 5.0 kg/ha.

Annual calcium deposition has a spatial pattern with two maxima: one in southern Ontario, the location of the concentration maximum, and the other in eastern Kansas (Figure 1.4). The maximum deposition is 5.96 kg/ha at Longwoods, Ontario. Fourteen sites have annual deposition greater than 3.0 kg/ha, all but three sites located in southern Ontario. The three sites are El Verde, Puerto Rico; Konza Prairie, Kansas; and Parsons, West Virginia.

The 1986 spatial patterns have the same overall shape, pattern and contour line location as in previous years (Sweeney and Olsen 1987, Olsen and Slavich 1985, 1986). Maps for an ion from different years largely reflect differences in the sites available, measurement uncertainty, and natural year-to-year variability.

1.3 TEMPORAL PATTERN ANALYSIS

The temporal pattern analyses use a subset of 30 long-term sites over an 8-year (1979-1986) period and an expanded subset of 137 sites with greater spatial coverage over an 5-year (1982-1986) period. The 8-year period represents the longest period with wet deposition monitoring data available that has a sufficient number of sites with data of known quality to allow a descriptive summary of annual temporal patterns to be produced. The 5-year period restricts the number of years in order to increase the number of sites available with data of known quality. Boxplot displays (see Section 2.5 for definition) for annual precipitation amount, pH, hydrogen, sulfate, and calcium are used to provide an overview of the types of temporal patterns observed for all ions.

1.3.1 Annual Temporal Patterns

The 30 site 1979-1986 trend subset has a temporal pattern for annual pH that suggests a cyclic structure (Figure 1.5). The 25th and 75th percentile and the median annual pH were lowest in 1980, and tend to increase until 1983 and then decrease until 1985 and 1986. The lowest annual pH values are 4.07 and 4.09 in 1980 and 1981 at Caldwell. Caldwell, Zanesville, and Penn State consistently have among the lowest pH values in each year since they are located in the region of lowest pH. Similarly, four sites with the highest pH values in all years are Manitou, Mead, Lamberton, and Marcell. Although the specific rank order of sites varies from year-to-year, the order is generally consistent across years.

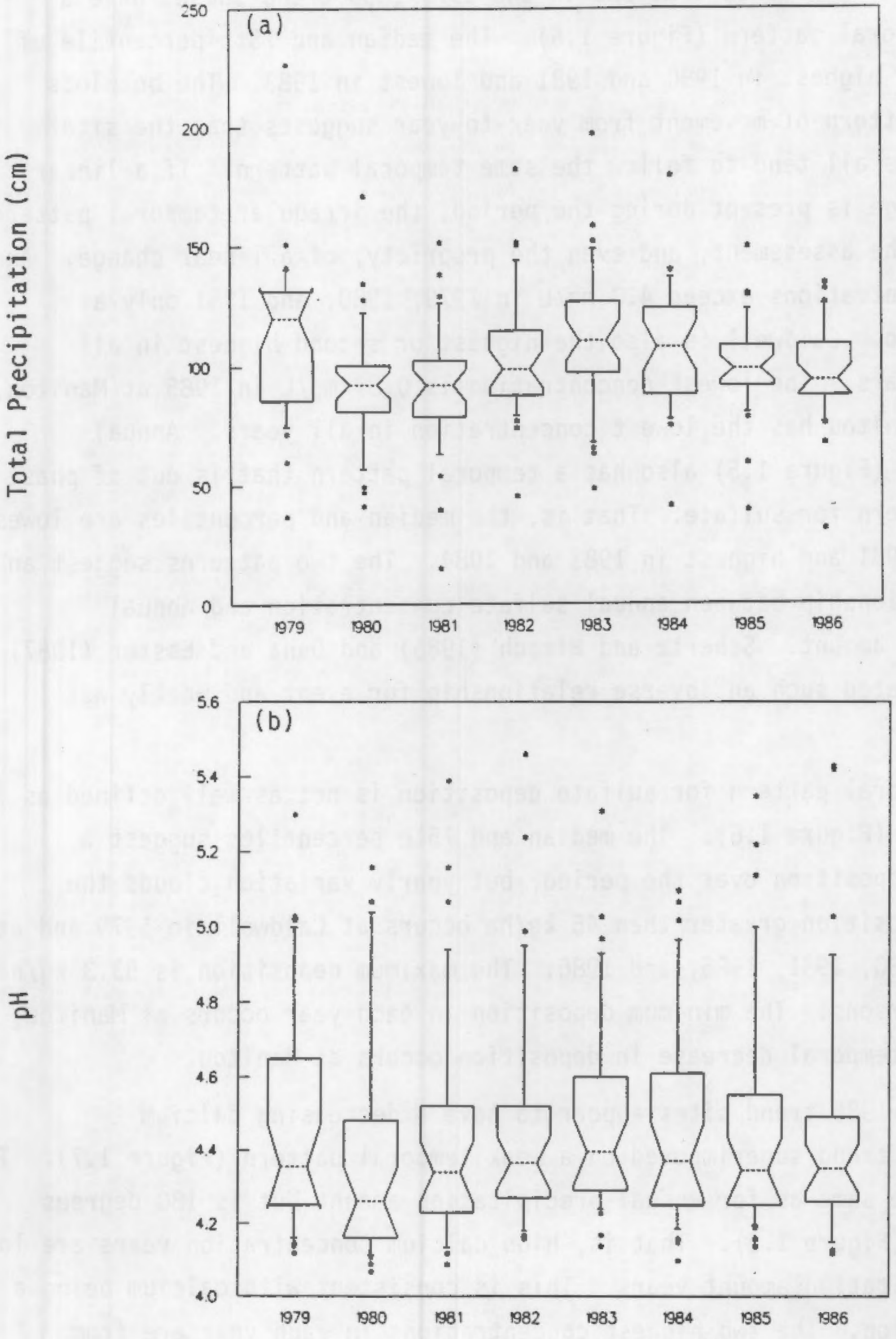
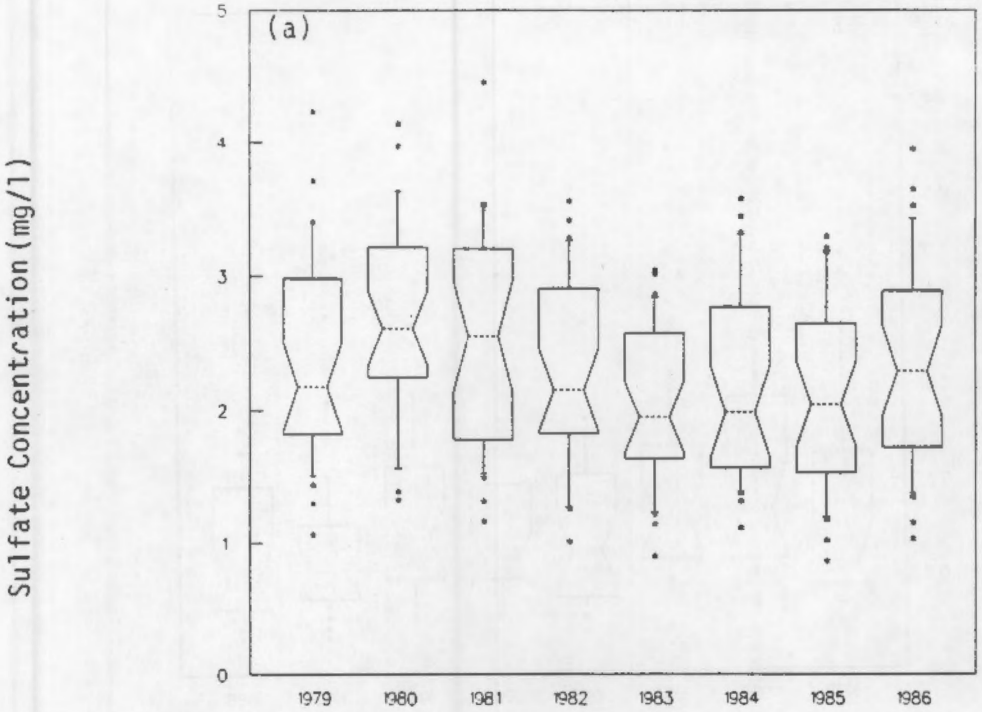


FIGURE 1.5. North American Temporal Pattern of (a) Annual Precipitation, and (b) Annual Precipitation-weighted pH. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-1986 Trend Subset.

Annual sulfate concentrations in the 1979-1986 trend subset have a definite temporal pattern (Figure 1.6). The median and 75th percentile of the sites are highest in 1980 and 1981 and lowest in 1983. The boxplots consistent pattern of movement from year-to-year suggests that the site concentrations all tend to follow the same temporal pattern. If a linear temporal change is present during the period, the irregular temporal pattern complicates the assessment, and even the propriety, of a linear change. Annual sulfate concentrations exceed 4.0 mg/L in 1979, 1980, and 1981 only at Caldwell, Ohio. Caldwell is also the highest or second highest in all subsequent years. The lowest concentration is 0.87 mg/L in 1985 at Manitou, Colorado. Manitou has the lowest concentration in all years. Annual precipitation (Figure 1.5) also has a temporal pattern that is out of phase with the pattern for sulfate. That is, the median and percentiles are lowest in 1980 and 1981 and highest in 1983 and 1984. The two patterns suggest an inverse relationship between annual sulfate concentration and annual precipitation amount. Schertz and Hirsch (1985) and Dana and Easter (1987) have demonstrated such an inverse relationship for event and weekly wet deposition.

The temporal pattern for sulfate deposition is not as well defined as for concentration (Figure 1.6). The median and 75th percentiles suggest a decrease in deposition over the period, but yearly variation clouds the picture. Deposition greater than 45 kg/ha occurs at Caldwell in 1979 and at Parsons in 1980, 1981, 1985, and 1986. The maximum deposition is 53.3 kg/ha in 1986 at Parsons. The minimum deposition in each year occurs at Manitou, Colorado. A temporal decrease in deposition occurs at Manitou.

The 1979-1986 trend sites appear to have a decreasing calcium concentration trend superimposed on a weak temporal pattern (Figure 1.7). The pattern is the same as for annual precipitation amount but is 180 degrees out of phase (Figure 1.5). That is, high calcium concentration years are low annual precipitation amount years. This is consistent with calcium being a soil-related ion. The two highest concentrations in each year are from Lamberton and Mead. Visual inspection of the extremes on the boxplots leads one to conclude that calcium concentrations have decreased at Lamberton and



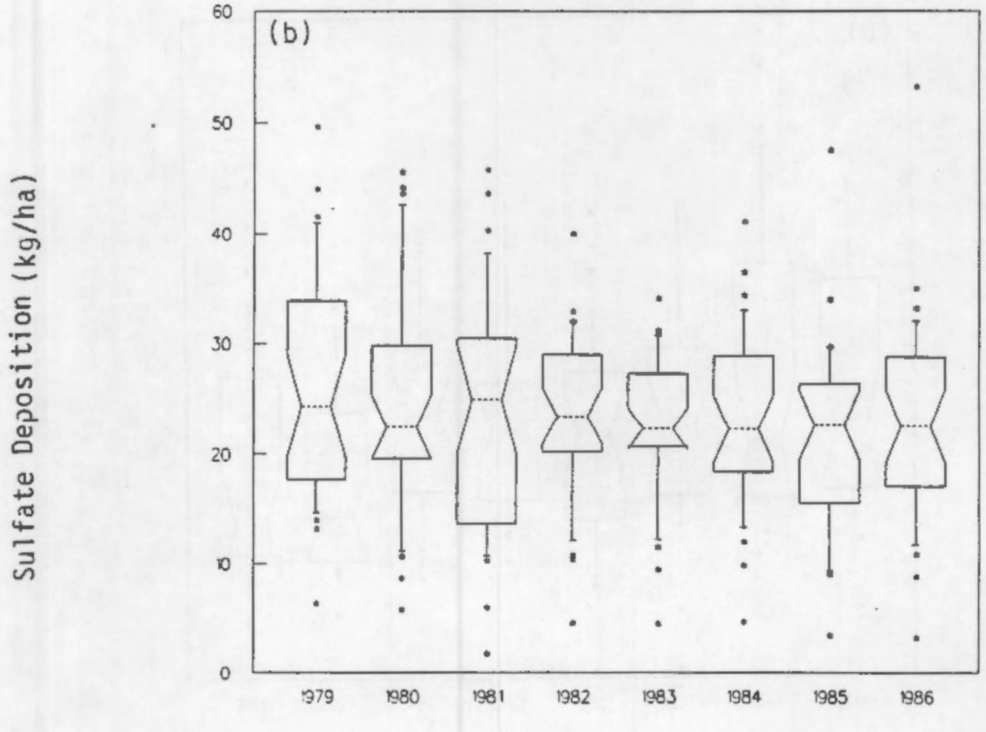


FIGURE 1.6. North American Temporal Pattern of (a) Annual Precipitation-weighted Sulfate Ion Concentration, and (b) Annual Sulfate Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-1986 Trend Subset.

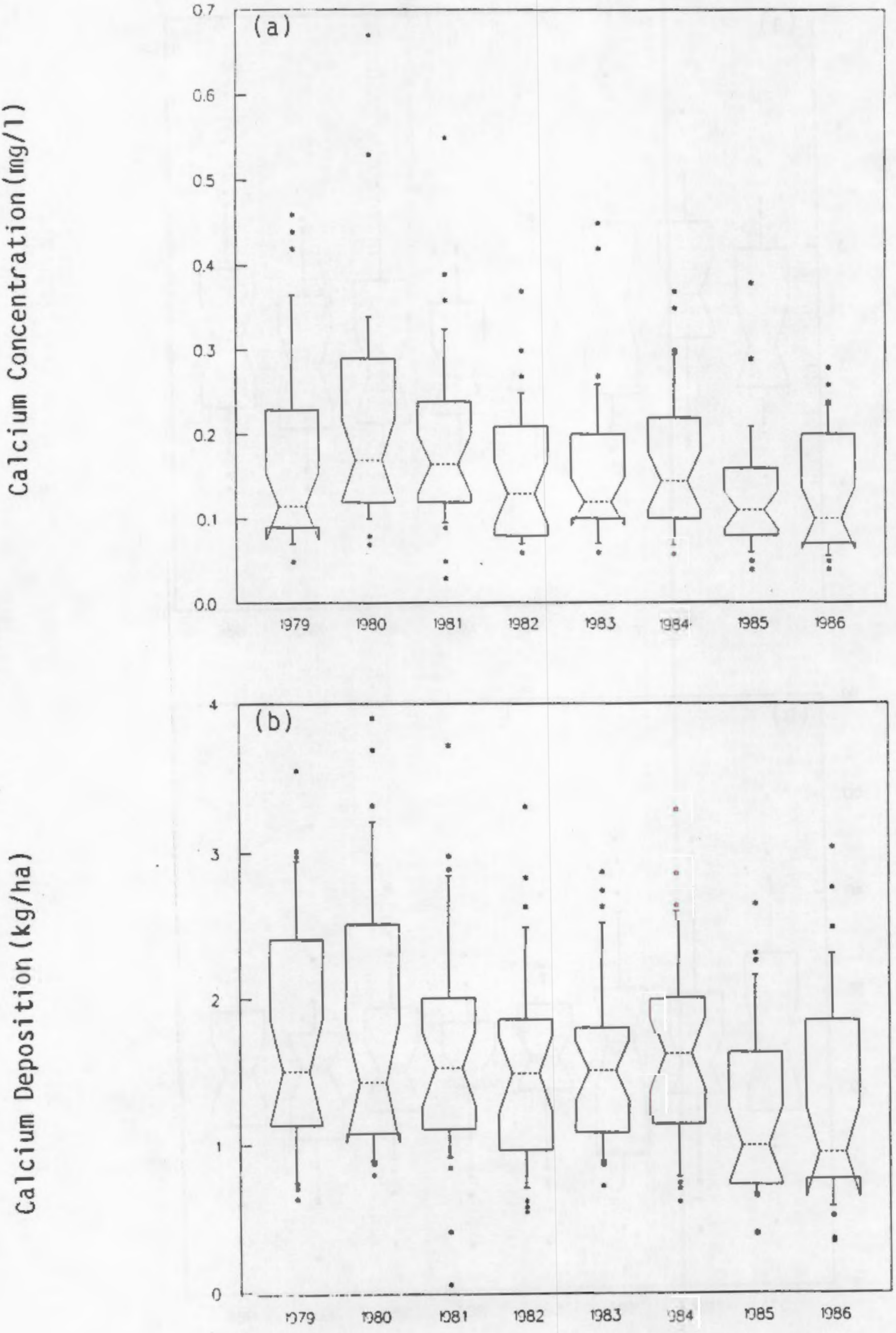


FIGURE 1.7. North American Temporal Pattern of (a) Annual Precipitation-weighted Calcium Ion Concentration, and (b) Annual Calcium Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-1986 Trend Subset.

Mead during the 8-years. The maximum concentration is 0.67 mg/L in 1980 at Mead, Nebraska.

Annual calcium deposition has a temporal pattern that is similar to, but weaker and more variable than calcium concentration (Figure 1.7). More site-to-site variation is present in 1979, 1980, 1985, and 1986 than in the middle four years. In addition, deposition appears to be substantially lower in 1985 and 1986 than in previous years. For example, two-thirds of the sites have an deposition less than 1.50 kg/ha in 1985 and 1986; while less than one-half of the sites do in any previous year. The three highest deposition sites in any year are usually Mead, Lamberton, and Parsons. However, the maximum deposition is 3.91 kg/ha at Zanesville in 1980.

1.3.2 Kendall Seasonal Tau Trend Analyses

The Kendall seasonal tau (KST) test provides a quantitative analysis for a linear temporal change in ion species concentration and deposition (see Section 2.0 for methodology). Hydrogen, sulfate, and calcium ion species are used to illustrate the general temporal change patterns found in concentration and deposition of ion species in precipitation. An overall summary is presented of the median and percent change trend estimates of all ion species concentration and deposition during 1979-1986 and during 1982-1986.

Based on the KST test, four 1979-1986 trend sites exhibit a significantly ($p < 0.05$) decreasing trend in hydrogen concentration: Raleigh, Ithaca, Scranton and Zanesville (Figure 1.8). One site, Bradford Forest, has a significantly increasing trend. The estimates appear to have no consistency in direction within any geographic region. Overall the median for 1979-1986 trend sites is -0.19 μeq/L/year and 25% of the sites have trend estimates of -1.10 μeq/L/year or less.

The Kendall seasonal tau test for linear trend identifies eight sites in the 1979-1986 trend sites with a significant ($p<0.05$) decreasing pattern for sulfate concentration (Figure 1.8). The presence of the previously identified cyclic pattern complicates, and may even invalidate, the interpretation of a linear trend. The eight sites are Lamberton, Mead, Manitou, Marcell, Piedmont Station, Oxford, Ithaca, and Hubbard Brook. No regional consistency appears to be present for sites with a statistically significant decreasing pattern.

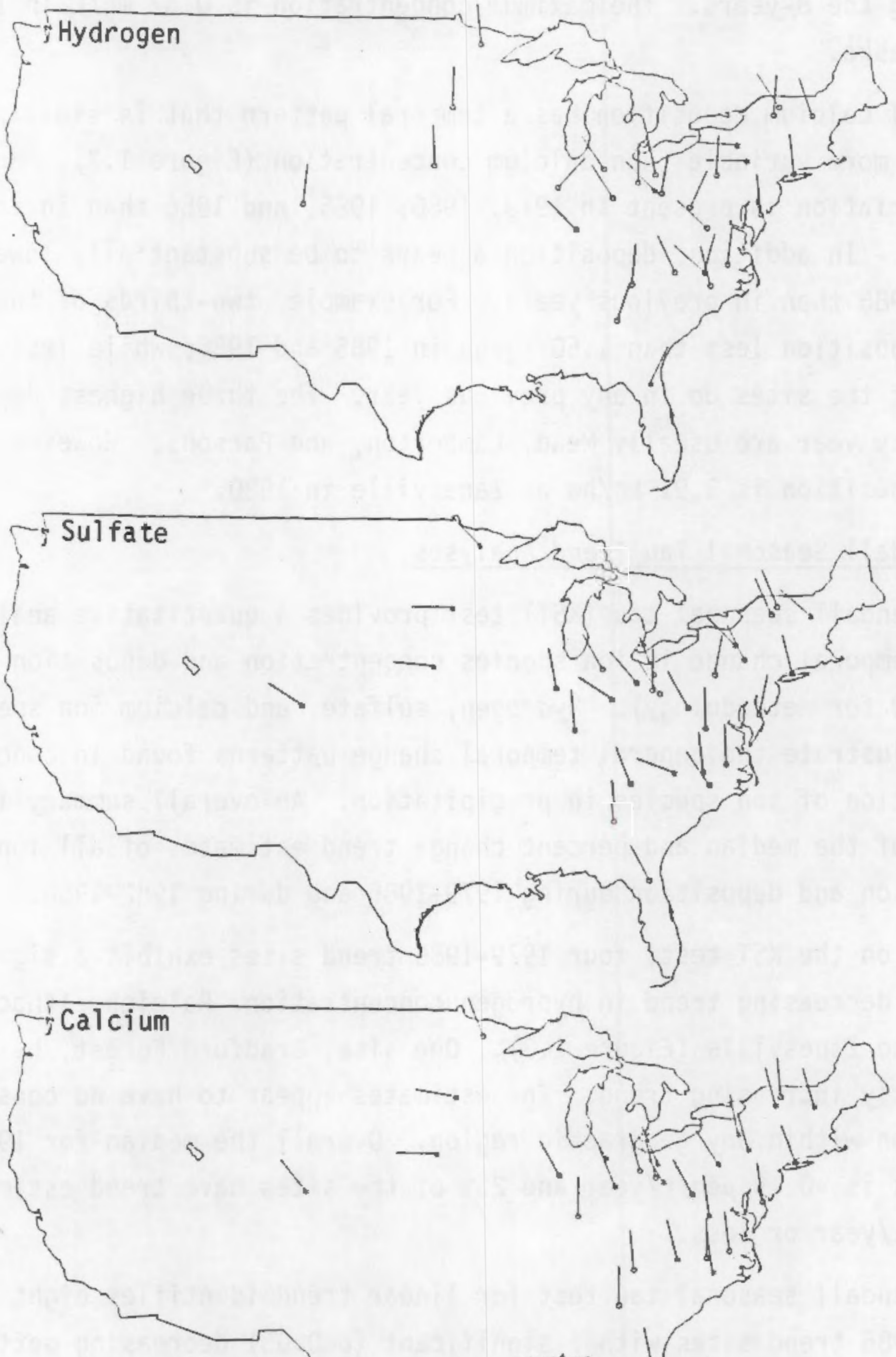


FIGURE 1.8. Hydrogen, Sulfate, and Calcium Concentration Trend for 1979-1986 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Solid Symbol Indicates Significant Trend ($p < 0.05$).

However, all but two of the thirty sites have decreasing trend estimates. The 25th, median, and 75th percentiles for trend estimates are -1.57, -0.91, and -0.23 μeq/L/year, respectively. The minimum trend estimate occurs at Mead (-3.15 μeq/L/year). The maximum occurs at Lewes (0.40 μeq/L/year).

All 1979-1986 trend sites have negative or zero calcium concentration trend estimates (decreasing or no trend) based on Sen's median trend estimate associated with the Kendall seasonal tau test (Figure 1.8). Seventeen of the thirty sites have significant ($p<0.05$) decreasing trends. Mead and Lamberton have the most extreme decreasing trend estimates of -1.96 and -1.74 μeq/L/year, respectively. The trend estimates also exhibit consistency among nearby sites. The 25th, 50th, and 75th percentiles for the thirty trend estimates are -0.61, -0.33, and -0.16 μeq/L/year. The maximum trend estimate is zero.

For 1979-1986 trend sites (Figure 1.9), significant hydrogen deposition decreases occur at four sites (Zanesville, Rockport, Raleigh, and Clinton Station) and increases occur at two sites (Brookhaven and Bradford Forest). The 25th percentile, median, and 75th percentile temporal changes are -1.32, -0.10, and 0.61 meq/m^2/year, respectively. The largest decrease (-5.64 meq/m^2/year) occurs at Zanesville.

Significant decreasing trends for sulfate deposition are found at eight sites in the 1979-1986 trend sites (Figure 1.9). The sites are Coweeta, Piedmont Station, Clinton Station, Finley, and Raleigh, North Carolina; Manitou, Colorado; Zanesville, Ohio; and Hubbard Brook, New Hampshire. The North Carolina sites demonstrated regional consistency. The trend direction at Zanesville is consistent with other sites in Ohio and West Virginia. The sites in New York nearest Hubbard Brook have increasing trend estimates, hence no regional consistency is apparent in this area. The 25th, 50th, and 75th percentiles for the 1982-1986 trend sites are -1.90, -0.62, and 0.00 meq/m^2/year, respectively. The minimum trend estimate is -3.75 meq/m^2/year occurring at Coweeta and Zanesville, and the maximum is 0.83 meq/m^2/year at Fort Wayne.

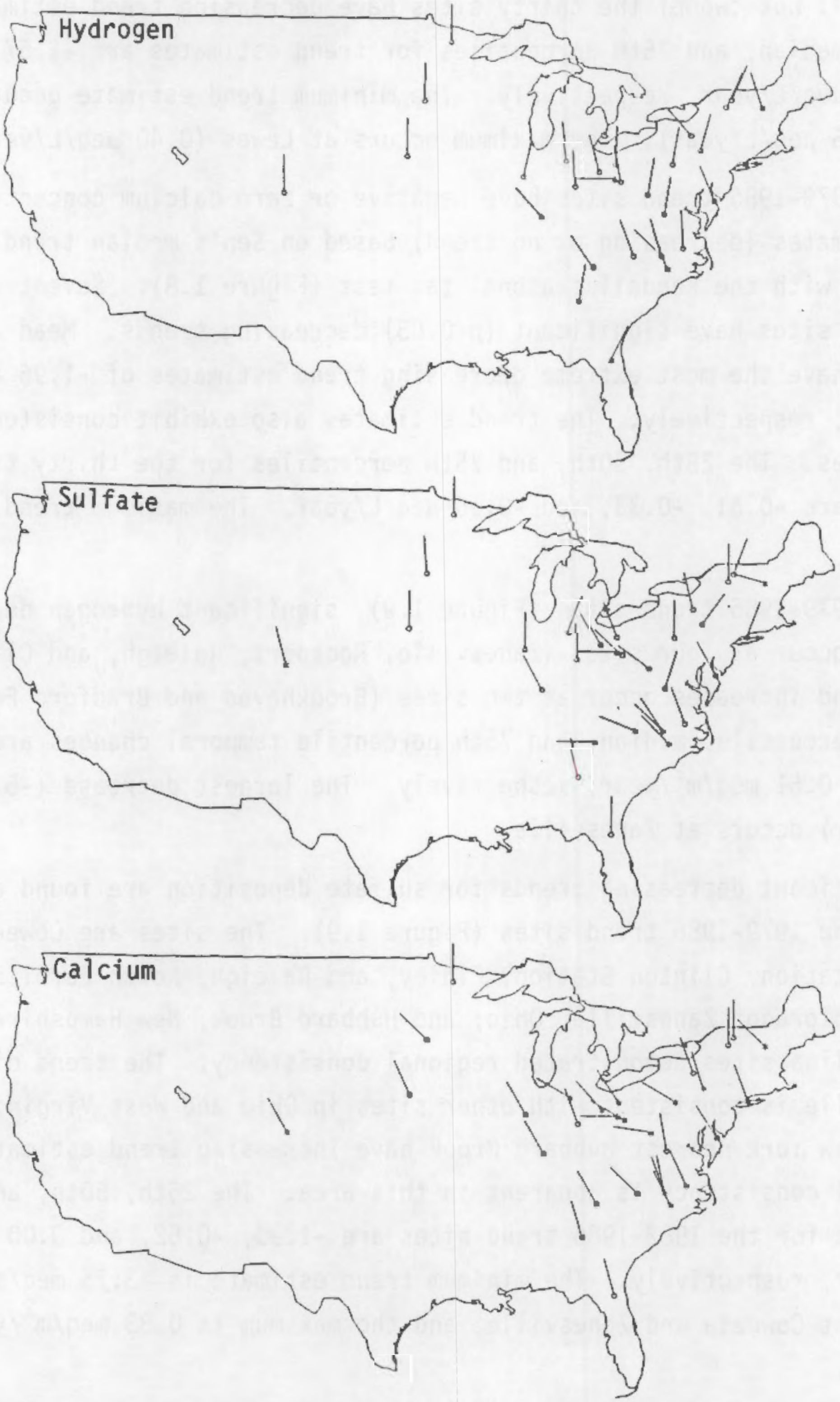


FIGURE 1.9. Hydrogen, Sulfate, and Calcium Deposition Trend for 1979-1986 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Seventeen of the 1979-1986 trend sites have significant decreasing trend estimates for calcium deposition (Figure 1.9). No site has an increasing trend estimate based on Sen's median slope estimate. The trend estimates are regionally consistent with nearby sites. The 25th, 50th, and 75th percentiles for the thirty trend sites are -0.45, -0.28, and -0.17 meq/m^2/year. The two most extreme decreasing trend estimates are -0.90 and -0.82 meq/m^2/year at Zanesville and Parsons, respectively.

Section 4.0 summarizes the multivariate aspects of the temporal patterns observed in wet deposition for the 1979-1986 and 1982-1986 trend site subsets. The discussion begins with an overview of the average annual concentrations for the ion species observed at the sites. This is followed by a comparison across the ion species of Sen's median slope trend estimate associated with the Kendall seasonal tau test. The trend estimate is then discussed in terms of percent change during the period. The reference for the percent change is the average 1982-1986 annual concentration for an ion species at a site. Individual year annual concentrations were also examined as a reference for percent change. The selection of the 5-year average from 1982-1986 provided a more stable reference base. The magnitude of the percent change does depend on the reference base at those sites with increasing or decreasing temporal patterns for an ion species. For example, using 1986 rather than 1982 as the reference, results in greater percent changes for cations at most sites.

An overview of the results of the Kendall seasonal tau test results can be obtained from Table 1.2 and Table 1.3. The most striking feature of the comparison across ion species for concentration is that the distributions are consistent, for all ion species except nitrate, ammonium, and chloride, in having over 50% of the estimates indicating a decreasing trend during the period. This is particularly the case for the 1979-1986 period. The concentration trend estimates for the 1982-1986 trend sites are more variable than those for the 1979-1986 trend sites. For the 1979-1986 trend sites, the median trend estimate is negative, i.e., decreasing trend, for all ion species (Table 1.2). At least 75% of the sites have decreasing trend estimates for sulfate, calcium, chloride, sodium, potassium, and magnesium. As discussed in Section 3.0, sodium concentrations decrease at all 1979-1986 trend sites. For the 1982-1986 trend sites, the median trend estimate indicates decreasing

TABLE 1.2. Median and Percent Change Trend Estimates of Ion Species Concentration and Deposition for 1979-1986 and 1982-1986 Trend Sites.

	Concentration (μeq/L/year)				Deposition (meq/m^2/year)			
	1979-1986		1982-1986		1979-1986		1982-1986	
Ion Species	Est.	%	Est.	%	Est.	%	Est.	%
Precipitation	-	-	-	-	-0.40	-0.4	-0.90	-1.0
Hydrogen	-0.19	-0.7	-0.37	-1.7	-0.10	-0.4	-0.40	-1.9
Sulfate	-0.91	-1.8	-0.62	-1.7	-0.62	-1.4	-0.62	-1.4
Nitrate	-0.01	-0.1	0.12	0.7	0.00	0.0	0.00	0.0
Ammonium	0.00	0.0	-0.09	-0.7	0.00	0.0	-0.33	-2.5
Calcium	-0.33	-4.6	-0.25	-3.6	-0.28	-4.0	-0.35	-5.2
Chloride	-0.13	-2.0	-0.05	-1.3	0.00	0.0	-0.06	-1.1
Sodium	-0.43	-9.9	-0.11	-2.7	-0.31	-8.8	-0.11	-3.8
Potassium	-0.03	-4.6	-0.03	-4.5	-0.03	-3.6	-0.03	-4.2
Magnesium	-0.08	-2.9	-0.11	-3.6	-0.06	-2.1	-0.10	-5.2

trends for all ion species except nitrate. Sulfate concentration has the most extreme median trend estimate of all ion species, -0.91 and -0.62 μeq/L/year in the 1979-1986 and 1982-1986 trend sites, respectively.

The concentration trend estimates may also be expressed in terms of percent change during the period. The reference for the percent change is the average 1982-1986 annual concentration for an ion species at a site. Sodium concentration for the 1979-1986 trend sites has the most extreme negative percent change. Potassium, calcium, and magnesium have the next most extreme distributions. The median percent changes at the 1979-1986 trend sites for sodium, potassium, calcium, and magnesium are -9.9, -4.6, -4.6, and -2.9 percent per year, respectively. Nitrate, ammonium, and, to a lesser extent, hydrogen, have distributions centered on zero percent change. Only sulfate and sodium have 90% or more of the sites with negative percent change. For the 1982-1986 trend sites, more consistency across ion species is present for the percent change distributions (Figure 4.4). The distributions are symmetric and have approximately the same variation (interquartile range) for all ion species. The only distinction among the ion species is the location of the center of the distribution. The increasing (most negative percent change) rank order of the medians for the distributions is potassium,

TABLE 1.3. Number and Percent of Significantly ($p<0.05$) Decreasing and Increasing Trend Estimates by Ion Species for Concentrations at 1979-1986 and 1982-1986 Trend Sites.

	1979-1986 Trend Sites Significance					1982-1986 Trend Sites Significance				
	Decrease		None	Increase		Decrease		None	Increase	
Ion Species	%	N	N	N	%	%	N	N	N	%
Precipitation	0	0	28	2	7	6	8	125	4	3
pH	3	1	25	4	13	3	4	124	9	7
Concentration										
Hydrogen	13	4	25	1	3	7	9	124	4	3
Sulfate	27	8	22	0	0	12	16	119	2	1
Nitrate	13	4	26	0	0	2	3	123	11	8
Ammonium	7	2	28	0	0	9	13	120	4	3
Calcium	57	17	13	0	0	15	20	111	6	4
Chloride	27	8	22	0	0	10	14	119	4	3
Sodium	83	25	5	0	0	9	13	124	0	0
Potassium	43	13	17	0	0	15	20	114	3	2
Magnesium	27	8	22	0	0	13	18	118	1	1
All Species	33	89	180	1	0	10	126	1072	35	3
Deposition										
Hydrogen	13	4	24	2	7	7	10	124	3	2
Sulfate	27	8	22	0	0	9	13	123	1	1
Nitrate	7	2	27	1	3	7	10	122	5	4
Ammonium	7	2	26	0	7	16	22	112	3	2
Calcium	30	9	21	0	0	25	34	97	6	4
Chloride	20	6	24	0	0	7	9	125	3	2
Sodium	73	22	8	0	0	12	16	121	0	0
Potassium	30	9	21	0	0	18	24	108	5	4
Magnesium	27	8	22	0	0	21	29	106	2	1
All Species	26	70	195	5	2	13	157	1038	28	2

magnesium, calcium, sodium, hydrogen, sulfate, chloride, and nitrate. Only nitrate has a median (0.7%) representing a positive percent change. Approximately 75% of the 1982-1986 trend sites have negative percent changes for calcium, potassium, magnesium, and sodium.

Kendall's seasonal tau tests for the significance of a trend in the temporal pattern. A significance level of $p=0.05$ is used as one way to summarize the test results. For each ion species and the two sets of trend sites, Table 1.3 summarizes the number and percent of the trend estimates that are significant, either decreasing or increasing. In the 1979-1986 trend sites, only one site (Bradford Forest, Florida) has a significant increasing trend in hydrogen ion concentration. In contrast, all ion species have significant decreasing trends at some sites, 89 sites for all ions. This represents 33% of the 270 tests calculated for the 30 sites and 9 ion species. Sodium, calcium, and potassium have significant decreasing trend estimates at the greatest number of sites, 25 (83%), 17 (57%), and 13 (43%), respectively. Sulfate has significant decreasing trend estimates at 8 (27%) sites. In the 1982-1986 trend sites, all ion species except sodium have significant increasing trend estimates at one or more sites, 35 sites for all ions. On average across all ions, this represents only 3% of the 1233 tests calculated for the 137 sites and 9 ion species. Ammonium and calcium have significant increasing trend estimates at the most sites, 11 and 6, respectively. Significant decreasing trend estimates occur for all ion species in 126 (10%) cases. Calcium, potassium, magnesium, and sulfate have significant decreasing trends at the most sites, 20 (15%), 20 (15%), 18 (13%), and 16 (12%), respectively.

Table 1.3 also summarizes the number of sites with significantly decreasing or increasing deposition trend estimates. In the 1979-1986 trend sites, only hydrogen, nitrate, and ammonium have a significant increasing deposition trend estimate, a total of 5 sites. Whiteface, New York has increasing trend estimates for nitrate and ammonium; Bradford Forest, Florida for hydrogen ion; Brookhaven, New York for hydrogen ion; and Scranton, Pennsylvania for ammonium (may be result of limited site operation in 1981). In contrast, 70 significant decreasing trend estimates are present for all ions. This number represents 26% of the 270 tests calculated for the 30 sites and 9 ion species. Sodium has significant decreasing trend estimates at 22 sites. Sulfate, calcium, chloride, potassium, and magnesium have significant decreasing trend estimates at 6-9 sites. For 1982-1986 trend sites, 28 (2%) significant increasing and 157 (13%) significant decreasing trend estimates occur for the 1233 tests calculated for the 137 sites. All ion species except sodium have a significant increasing trend estimate at one or more sites. If the tests are statistically independent and the normal distribution assumption is appropriate, approximately 2.5% of the tests would be expected to give significant increasing/decreasing results based on randomness above. Calcium, magnesium, potassium, and ammonium have significant decreasing trends at the most sites, 34 (25%), 29 (21%), 24 (18%), and 22 (16%), respectively. Precipitation amount has significant decreasing estimates at eight sites and significant increasing estimates at four sites.

2.0 INTRODUCTION

This report gives a descriptive statistical summary of wet deposition temporal and spatial patterns for the United States and eastern Canada. Isopleth maps display 1986 annual and seasonal spatial patterns for pH, hydrogen, sulfate, nitrate, ammonium, and calcium wet deposition. The intention of the isopleth maps is to give a general overview of regional differences in wet deposition. Graphical displays of annual wet deposition at monitoring sites and Kendall seasonal tau statistical trend analyses provide information on temporal patterns for 8-year (1979-1986) and 5-year (1982-1986) periods for wet deposition ion species.

The purpose of this section is to state the source of the wet deposition monitoring data, to describe the data summarization procedures, to define the site selection process for spatial and temporal pattern analyses, to describe the spatial pattern methodology, and to describe the temporal pattern methodology.

2.1 SOURCE OF WET DEPOSITION DATA

Various federal, state, and local agencies and private industries support networks of sites for the collection and chemical analysis of precipitation samples. The data in this report are from networks that contribute data to the Acid Deposition System (ADS) (Watson and Olsen 1984) for statistical reporting of North American wet deposition data. Other networks collect precipitation chemistry data but did not contribute data to the ADS for the period 1979 to 1986.

The spatial and temporal pattern analyses include data from six regional networks (Table 2.1). The Multi-State Atmospheric Pollution and Power Production study (MAP3S) initiated the MAP3S precipitation chemistry network (MAP3S/PCN) in 1976 with four sites in the northeastern United States to begin research on the scavenging of pollutants by precipitation. It was the first wet-deposition-only, event sampling network. In 1978 the National Atmospheric Deposition Program (NADP) established a weekly sampling network to monitor trends in precipitation chemistry in the United States. The NADP, in cooperation with federal, state, and private research agencies, was created

TABLE 2.1. Wet Deposition Monitoring Networks in the United States and Canada that Contributed Data to the ADS

Network Abbreviation	Network Name	Start Date	Number of Sites in 1986	Sample Type
MAP3S/PCN	MAP3S Precipitation Chemistry Network	1976	9	Daily,[a] Wet
NADP/NTN	National Atmospheric Deposition Program/ National Trends Network	1978	203	Weekly, Wet
UAPSP[b]	Utility Acid Precipitation Study Program	1978	25	Daily,[a] Wet
CAPMoN[c]	Canadian Air and Precipitation Monitoring Network	1983	24	Daily, Wet
APIOS-C	Acidic Precipitation in Ontario Study - Cumulative Network	1980	38	Monthly,[d] Wet
APIOS-D	Acidic Precipitation in Ontario Study - Daily Network	1980	17	Daily, Wet

[a] Daily or duration of precipitation event, whichever is greater.

[b] UAPSP sites prior to 1981 were part of the EPRI/SURE network. The UAPSP network is scheduled to cease operation on December 31, 1987; on January 1, 1988 the same sites or ones near to the present UAPSP sites will begin operation as the Operational Evaluation Network.

[c] Some CANSAP and APN sites were transferred to CAPMoN during the period 1983 to 1985; before 1983, CANSAP and APN operated as separate networks.

[d] Sampling interval changed from calendar month to 28 days in January 1982.

to conduct research on atmospheric deposition and its effects. In 1982, the NADP assumed responsibility for coordinating the operation of the National Trends Network (NTN) of the federally-supported National Acid Precipitation Assessment Program (NAPAP). The merged networks now have the designation NADP/NTN. The daily sampling network designated as the Utility Acid

Precipitation Study Program (UAPSP) began in 1978 as a nine-site research network initiated by the Electric Power Research Institute and expanded to twenty-two sites giving broader regional coverage in 1982 under the Utility Acid Precipitation Study Program. The daily Canadian federal network, CAPMoN, was formed using sites from two previous networks as a base: the Canadian Network for Sampling Acid Precipitation (1977-1985) and the Air and Precipitation Monitoring Network (1978-1985). In 1980, Ontario province established two networks as part of the Acidic Precipitation in Ontario Study (APIOS). The cumulative network, APIOS-C with a four-week sampling period, is designed to determine the long-term deposition pattern in Ontario. The daily network, APIOS-D, is designed to define the sector of origin of the ion species as well as the frequency and intensity of acidic deposition episodes.

A summary of the number of sites operated by each network from 1979 to 1986 is presented in Table 2.2. For the purposes of this table, a site is considered to have operated during a year if one or more samples were collected at some time during the year. Hence sites are included that operated only a portion of the year because they were either terminated or started during the year. The total number of sites increased from 65 in 1979 to 316 in 1985. The implementation of the NTN and expansion of the NADP network is responsible for most of the growth in the United States. In Canada, the initiation of the APIOS and CAPMoN networks in 1980 and 1983 accounts for all the increase. To consider only those sites that were operated for a full year, a definition of full year is required. A site is considered to have operated a full year if precipitation (not necessarily precipitation chemistry) is monitored more than 90% of the year. Only sites operating a full year can be considered as sites that may provide data for an annual summary. The number of sites operating a full year (Table 2.3) increased from 47 in 1979 to 290 in 1986. Figure 2.1 gives the location of sites operating a full year.

TABLE 2.2. Number of Sites by Network from 1979 to 1986

Network	1979	1980	1981	1982	1983	1984	1985	1986
NADP/NTN	39	82	97	110	142	177	195	203
MAP3S/PCN	8	8	9	9	9	9	9	9
UAPSP	18	10	21	22	25	27	26	25
CAPMoN	0	0	0	0	17	18	24	24
APIOS-D	0	8	16	16	16	17	15	17
APIOS-C	0	30	35	36	38	38	37	38
Total	65	138	178	193	247	286	306	316

TABLE 2.3. Number of Sites by Network that Monitored Precipitation more than 90 Percent of Year from 1979 to 1986

Network	1979	1980	1981	1982	1983	1984	1985	1986
NADP/NTN	21	36	73	89	102	139	179	187
MAP3S/PCN	8	8	9	9	9	9	9	9
UAPSP	18	6	6	21	20	19	21	24
CAPMoN	-	-	-	-	7	17	18	21
APIOS-D	-	-	12	16	16	14	14	13
APIOS-C	-	-	30	33	32	34	32	36
Total	47	50	130	168	186	232	273	290

2.2 DATA QUALITY AND SUMMARIZATION PROCEDURES

Each network has its own set of specific protocols for selecting site locations, field operation procedures, laboratory analysis procedures, and data screening procedures. Networks implement these protocols to ensure the collection, laboratory analysis, and reporting of wet deposition data of consistent and known quality. Each network included in this report has and implements a quality assurance program.

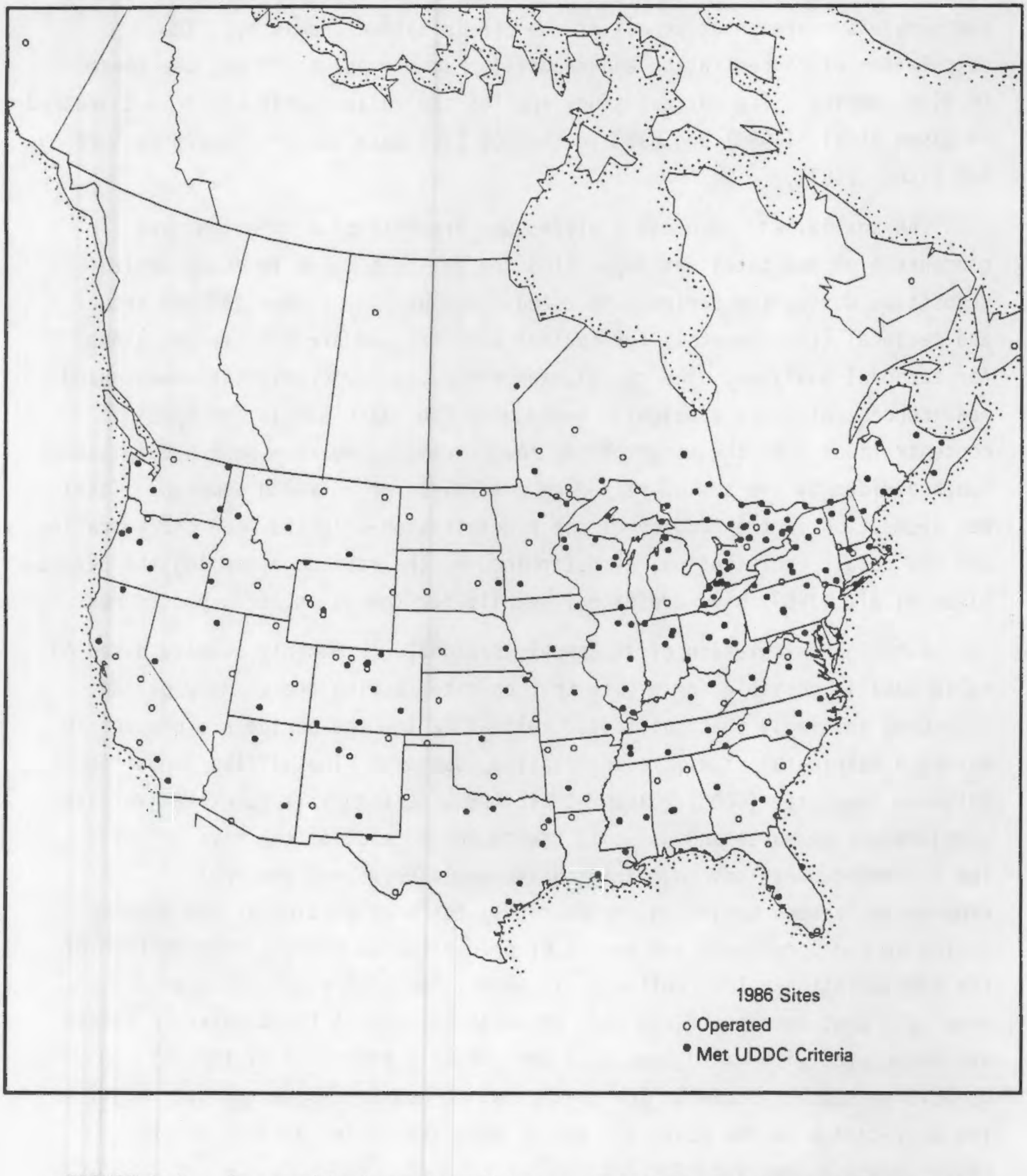


FIGURE 2.1. Geographic Location of Sites in 1986 Operating a Full Year and the Subset of Sites Meeting Annual UDDC Criteria for Sulfate.

However, even with careful attention, some samples must be declared invalid because of the violation of some aspect of the protocol that affects the sample's representativeness of the precipitation chemistry. The calculation of concentration and deposition summaries should not use these invalid samples. The current study applies the valid sample criteria discussed in Olsen et al. (1989) and used in the ADS 1985 Data Summary Report by Sweeney and Olsen (1987).

The spatial and temporal analyses use precipitation-weighted mean concentration and total wet deposition for an ion species to characterize wet deposition during the period summarized. The periods summarized are annual and seasonal (three months) for spatial analyses and are monthly and annual for temporal analyses. The calculation procedure for precipitation-weighted mean concentration is a weighted average of the valid sample ion species' concentrations with the weight being the corresponding rain gage precipitation depth divided by the sum of rain gage precipitation in valid samples. Total wet deposition is the product of the precipitation-weighted mean concentration and the total precipitation, as determined by the rain gage, during the period. Olsen et al. (1989) give additional details for the calculation procedures.

Although calculation of an annual, seasonal, or monthly summary based on valid data is possible for a site that operated during the summary period, this does not imply that sufficient valid data for the period are present to ensure a meaningful, i.e., representative, summary. The Unified Deposition Database Committee (UDDC) (Olsen et al. 1989) defined five quantitative data completeness measures and assigned thresholds to each of the five criteria. The following questions concerning data completeness and temporal representativeness motivated the measures: for what portion of the summary period is the occurrence and amount of precipitation known?; what portion of the precipitation volume collected is associated with valid deposition samples?; what percent of the time and what percent of the samples collected are associated with valid samples?; and, what is the ratio of the wet deposition sample volume to the precipitation measured by a standard gage? The application of the measures, and at what threshold, depends on the requirements appropriate for a study.

The ADS 1985 Data Summary Report by Sweeney and Olsen (1987) discusses the UDDC data completeness measures and criteria. Application of the UDDC criteria significantly reduces the number of sites available for defining the temporal or spatial pattern of wet deposition. Although the UDDC criteria are reasonable in the sense that a site meeting a network's protocol would be expected to exceed them, the criteria are in fact very strict. For example, an annual summary must meet minimum threshold levels for each of the five data completeness measures not only for the entire annual period but also for each of the four quarters within the annual period. The primary requirements are that precipitation monitoring cover at least 90% of the period (annual and each quarter) and that valid sample chemistry data be available for at least 60% (50%) of the annual (quarterly) precipitation measured.

Table 2.4 lists the number of sites by network that meet the UDDC data completeness criteria for annual summaries from 1979 to 1986. The number meeting the UDDC criteria increased from 37 in 1979 to 195 in 1986. Figure 2.1 shows the location of the subset of sites operating a full year that met the UDDC criteria for 1986 annual sulfate summaries. Of the sites failing the criteria, approximately one-half are west of the Mississippi River, one-fourth are in Canada, and one-fourth are in eastern United States. This distribution partially reflects where weekly and four-weekly networks operate sites. In 1986, 67% of sites operating a full year met the UDDC criteria. This is a reflection of the strictness of the criteria, especially for networks with a weekly or four-weekly sampling period. The number of invalid samples occurring during the first quarter (winter) is the most frequent cause for a site to fail the UDDC criteria, i.e., application of the quarterly criteria for all quarters.

The selection of sites for temporal pattern analyses uses a relaxed criteria for selection of a representative summary based on the data completeness measures. The criteria are a compromise between ensuring each summary is based on very complete data, i.e., meet UDDC criteria, and increasing the number of sites available for temporal pattern analysis. The criteria are that precipitation monitoring cover at least 90% of the annual period and that valid sample chemistry data represent at least 60% of the annual precipitation measured. Although the thresholds are the same as the

UDDC criteria, the criteria do not apply the quarterly UDDC requirements or the other three UDDC data completeness measures. The number of sites meeting the relaxed criteria (Table 2.5) is substantially greater than those meeting the UDDC criteria. Over 94% of the sites operating a full year met the relaxed criteria each year.

TABLE 2.4. Number of Sites by Network and Year Meeting the Unified Deposition Database Committee Annual Summary Data Completeness Criteria for Sulfate

Network	1979	1980	1981	1982	1983	1984	1985	1986
NADP/NTN	16	26	53	60	71	85	101	115
MAP3S/PCN	5	7	8	9	8	9	9	9
UAPSP	16	6	6	21	20	18	19	21
CAPMoN	-	-	-	-	4	11	11	17
APIOS-D	-	-	-	-	10	10	11	11
APIOS-C	-	-	10	14	22	21	20	22
Total	37	39	77	104	135	154	171	195

TABLE 2.5. Number of Sites by Network and Year Meeting the Relaxed Annual Summary Data Completeness Criteria for Sulfate

Network	1979	1980	1981	1982	1983	1984	1985	1986
NADP/NTN	20	35	66	82	97	133	168	175
MAP3S/PCN	8	8	9	9	9	9	9	9
UAPSP	18	6	6	21	20	19	21	24
CAPMoN	-	-	-	-	7	17	18	21
APIOS-D	-	-	7	16	16	14	14	13
APIOS-C	-	-	26	30	32	32	31	30
Total	46	49	114	158	181	224	261	272

2.3 SELECTION OF SITES FOR SPATIAL AND TEMPORAL PATTERN ANALYSES

Site selection is similar for the networks and is based on criteria for regional representativeness. A regionally representative site is one at which the precipitation chemistry is free from the influence of local pollution sources. One example of typical criteria specific to NADP/NTN is given by Robertson and Wilson (1985) and Bigelow (1984). Recently, a joint United States and Canadian committee, the Unified Deposition Database Committee, consolidated network criteria for regional representativeness into a single set of criteria (Olsen et al. 1989). In an analysis of sites with data in the ADS in 1985, the UDDC rated 80% of the sites as being regionally representative; 12% as marginally representative, often because those sites were in regions of high emission density where it is difficult to locate sites sufficiently far from all pollution sources; and 6% as unrepresentative of regional wet deposition. The current report does not exclude any site solely on the basis of being regionally unrepresentative as defined by the UDDC. For purposes of this report, adherence to individual network siting protocols was deemed a sufficient measure of representativeness.

2.3.1 Spatial Pattern Sites

The report presents 30 isopleth maps of hydrogen, sulfate, nitrate, ammonium, and calcium concentration and deposition spatial patterns for 1986 annual and seasonal (winter and summer) summary periods. Generation of the maps requires a separate selection of sites for each ion and summary period based on the UDDC data completeness criteria. As an example, the number of sites used to create the 1986 annual sulfate concentration and deposition isopleth maps is 195 (Table 2.4). The number for hydrogen, nitrate, ammonium, and calcium is similar since most reasons for an invalid value apply to the entire sample. Table A.3 gives the number of sites used for each map. Each isopleth map also gives the location of all sites used in its construction.

2.3.2 Temporal Pattern Sites

A primary consideration in selecting a site for temporal pattern analysis is the continuous operation of the site for the entire period of the analysis. Preferably, the site would also meet the UDDC data completeness criteria for each year during the period. Applying these criteria severely restricts the number of sites available for temporal pattern analysis covering five or more years. Since the objective of the data completeness criteria is to aid in identifying sites with good quality data, an alternative approach based on a relaxed criteria applied to multiple years was developed.

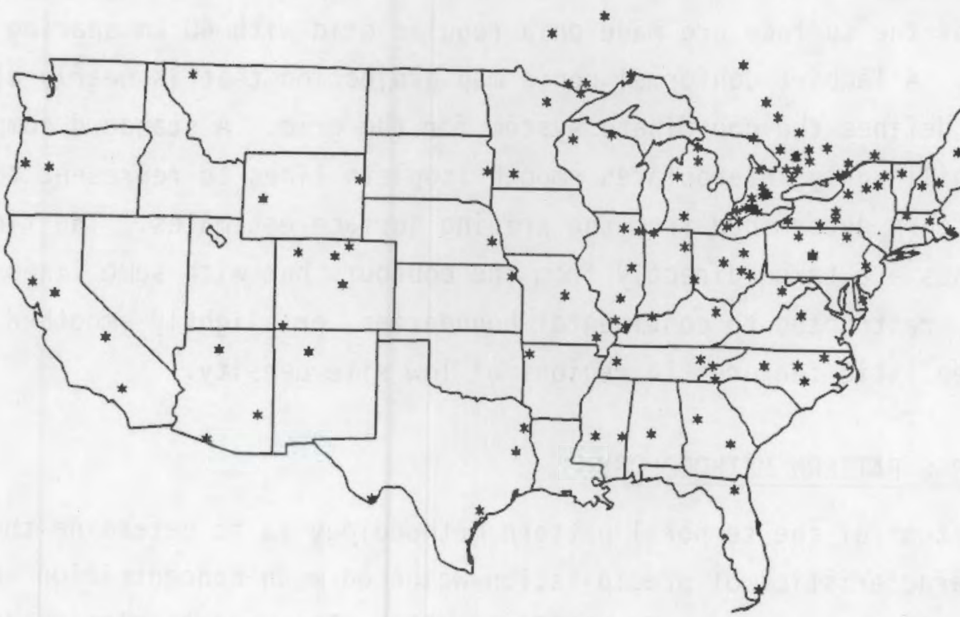
The alternative approach for a site to be included in the 1979-1986 temporal pattern analysis requires a site to operate from 1979 to 1986 and to meet the relaxed data completeness criteria in 1979 or before, and in at least 6 of the 8 years. Thirty sites meet this requirement: 17 from NADP/NTN, 8 from MAP3S, and 5 from UAPSP. This subset of sites is termed the 1979-1986 trend subset. Because of the limited number of sites in the 1979-1986 trend subset, the report includes secondary temporal pattern analyses for the 5-year period 1982 to 1986. Inclusion of sites for this 1982-1986 trend subset requires a site to operate from 1982 to 1986 and to meet the relaxed criteria in 1982 or before, and in at least 3 of the 5 years. A total of 137 sites meet this requirement: 71 from NADP/NTN, 9 from MAP3S, 18 from UAPSP, 12 from APIOS-D, and 27 from APIOS-C. The location of the sites in each of the subsets is presented in Figure 2.2.

2.4 SPATIAL ESTIMATION METHODOLOGY

Isopleth maps illustrate the spatial distribution of wet deposition for North America in 1986. Such maps provide a good visual display of the general features of the spatial distribution, e.g., location of spatial maxima and general orientation of regions with high concentration or deposition. The purpose of the maps is to convey the general features of the spatial pattern; not to interpolate monitoring data to non-monitored site locations. This section provides a description of the methodology used to prepare the maps.

Preparation of isopleth maps for wet deposition involves: (1) selection of sites that meet data quality criteria, (2) estimation of the spatial surface based on the selected sites, and (3) display of the estimated surface by use of isopleth lines. Using alternative procedures for any of the three steps can result in isopleth maps that differ. For reasonable alternatives, detailed features of the maps will differ but overall general features will remain similar. The maps presented should be viewed with this is mind.

1979-86 Trend Site Locations



1982-86 Trend Site Locations

FIGURE 2.2. Location of Sites Included in Trend Subsets for 1979-1986 and for 1982-1986.

Estimating the spatial distribution of an ion species' precipitation weighted average concentration or deposition (annual or seasonal) is a problem of estimating a spatial surface by interpolation between measurement sites. Since spatial correlation exists among the sites, the estimation technique should take this into consideration. One assumption that has been used is that spatial correlation is a function only of the distance between sites. Kriging is a surface estimation technique that explicitly utilizes this assumption. It uses the inter-site correlation in the data to determine the weights that are applied to measured site values in estimating the surface at any location. It also provides an estimate of the interpolation error, but this is not considered in this report. The term kriging actually designates a family of geostatistical estimation procedures. This application uses simple kriging. It has been used in environmental studies and has been used to analyze spatial patterns in acid precipitation data (Bilonick 1983; Eynon and Switzer 1983; Finkelstein and Seilkop 1981; Finkelstein 1984; Olsen and Slavich 1986).

Isopleth maps display the surface estimated by the kriging procedure. Estimates of the surface are made on a regular grid with 40 km spacing between grid nodes. A lambert conformal conic map projection that is nearly distance preserving defines the coordinate system for the grid. A standard computer algorithm bilinearly interpolates smooth isopleth lines to represent the spatial pattern determined from the kriging surface estimates. The final isopleth maps are taken directly from the contours but with some lines eliminated, restricted to continental boundaries, or slightly smoothed to remove unrealistic features in regions of low site density.

2.5 TEMPORAL PATTERN METHODOLOGY

The intent of the temporal pattern methodology is to determine the year-to-year characteristics of precipitation-weighted mean concentration and deposition temporal patterns from 1979 to 1986. The characteristics apply only to the period studied and do not imply either the presence or absence of any long-term trend. Eight years is not a sufficient number of years to address the issue of long-term trends.

The methodology includes two basic, and fundamentally different, approaches: graphical displays of annual data and statistical trend analysis of monthly data. Boxplots, defined below, graphically display percentiles of annual data from a subset of trend sites for each year in either the 1979-1986 or 1982-1986 period. Visual inspection of boxplots across years provides a visual assessment of annual temporal patterns during the period. Application of a Kendall seasonal tau trend statistical test and Sen's median slope estimation procedure to monthly data provide estimates of a general temporal pattern change over the entire period.

Analyses of temporal patterns use four related data sets differentiated by the time period covered and by the data summary period. The 1979-1986 period with 30 sites and the 1982-1986 period with 137 sites define two sets of years and sites. The 137 sites for the 5-year period include the 30 sites for the 8-year period. Hence, the two combinations of sites and years overlap. For each of these two sets of sites and years, the analyses use annual and monthly data summaries of precipitation-weighted mean concentration and deposition for each ion species. Annual analyses for a trend subset are based on all annual summaries, i.e., no site has a missing annual summary. Monthly analyses have missing monthly summaries for a site. A monthly data completeness requirement applied to each monthly summary requires precipitation monitoring to cover at least 75% of the month and at least 60% of the precipitation measured during the month must be associated with valid data.

Boxplots simultaneously display several features of a set of data. Figure 2.3 illustrates the definition of the boxplot as applied in this report. Each boxplot presents the 5th (or 10th), 25th, 50th (median), 75th, 95th (or 90th) percentiles of data from a set of sites. The plots display all sites more extreme than either the 5th (10th) or 95th (90th) percentiles. In some boxplots, very extreme data values are truncated and plotted at the plot boundary. Boxplots for the 1979-1986 trend subset of 30 sites report the 10th and 90th percentiles, while boxplots for the 1982-1986 subset of 137 sites report the 5th and 95th percentiles. Annual data boxplots are "notched" to display a 95% confidence interval for the median. The upper portion of a boxplot depicts the pattern of high concentration or deposition sites or of

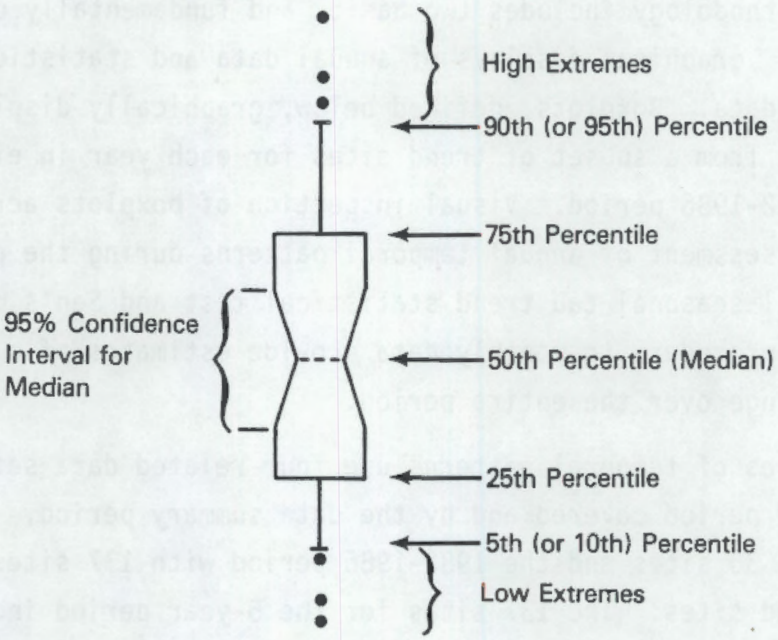


FIGURE 2.3. Definition of Boxplot Display

sites with most positive (or nearly positive) "trend", depending upon the type of data displayed by the boxplot. The lower portion of a boxplot depicts the pattern of low concentration or deposition sites or of sites with most negative "trend", depending upon the type of data displayed by the boxplot.

Hirsch et al. (1982) developed the Kendall seasonal tau (KST) test for trend detection in the presence of constant length cycles or seasonal effects. The test accommodates missing, tied, or limit of detection data values. Furthermore, the validity of the test does not require the data to be normally distributed. The test consists of computing the Mann-Kendall test statistic for year-to-year change separately for each season and combining these seasonal test statistics into a single statistic that can be tested for statistical significance. Here season refers to the twelve monthly precipitation-weighted mean concentrations or twelve monthly deposition values potentially available from each year. An estimate of the magnitude of the change is given by Sen's median slope (Gilbert 1987). The median slope of all possible seasonal pairs of values gives an estimate of the annual change. A nonparametric confidence interval is also computed and reported.

The number of applications of the Kendall seasonal tau test and slope estimate is large. For example, the 1979-1986 trend sites have the test applied to precipitation amount, pH and to each ion precipitation-weighted mean concentration and deposition for each of 30 sites. This results in the computation of 600 tests, slope estimates and confidence intervals. For the 1982-1986 trend subset of sites, the number of applications is 2740. Three different approaches summarize the KST results across sites in a trend subset for each ion.

The first approach gives percentile summaries across sites for the slope estimates for an ion concentration or deposition analysis. Boxplots display the percentiles in some situations. The text discusses sites with extreme slope estimates.

The second approach graphically displays the slope estimate for a site on a map of the United States (see upper plot Figure A.9). The angle of a constant length ray is proportional to the magnitude of the slope estimate. A vertical ray represents the median of all slope estimates on the map. Slopes smaller than the median tilt to the left and slopes greater than the median tilt to the right. A horizonal ray either to the left or the right represents the minimum or maximum slope depending on which one is farther from the median. The purpose of the map display is to display all slope estimates and to look for consistent regional patterns in temporal pattern changes by locating regions with consistent ray angles for all sites in the region. In addition to displaying the slope estimate, a solid symbol at the base of the ray identifies the sites with a KST test result significantly different from zero (at 5% level).

The third approach graphically displays the confidence interval for the slope estimate in a novel way and enhances the display by using a ray to identify the 1986 annual ion concentration or deposition for the site (see lower plot Figure A.9). The angle of the ray is proportional to the magnitude of the annual value. The confidence interval plot consists of plotting the center of the confidence interval versus the confidence interval half-width. Since the nonparametric confidence intervals are not necessarily symmetric the center need not equal the slope estimate. However, they are highly correlated so that the center may be interpreted as the slope estimate. Two

45 degree lines divide the plot into three regions. A confidence interval plotted in the left region has its upper limit less than zero indicating a significant decreasing temporal pattern. A confidence interval plotted in the right region has its lower limit greater than zero indicating a significant increasing temporal pattern. Confidence intervals in the middle region include zero indicating no significant temporal pattern. In addition to the ray displaying the annual value, a solid symbol at the base of the ray identifies the sites with a KST test result significantly different from zero (at 5% level). In most cases, the KST test result and the confidence interval significance result are consistent.

The confidence interval plots provide several types of information. It is easy to identify sites with significantly decreasing or increasing trends by the region in which the site is plotted or by a solid symbol. The horizonal pattern of confidence interval locations gives a general overview of the distribution of the slope estimates. The vertical pattern of confidence interval locations gives an overview of the within-site variation of monthly concentration or deposition; the larger the confidence interval half-width, the greater the within-site variation. The plot does not provide any direct information on geographic location of each site. However, the ray angle gives indirect information if the user knows the relationship between concentration or deposition annual magnitude and geographic location. For example, high sulfate concentrations occur in southern Ontario, western Pennsylvania, and Ohio; while the lowest concentrations occur west of the Mississippi River.

3.0 INDIVIDUAL ION SPATIAL AND TEMPORAL PATTERNS

This section contains descriptive statistical summaries and analyses of wet deposition spatial and temporal patterns for individual ions. For hydrogen, sulfate, nitrate, ammonium, and calcium ions, the description gives an overview of 1986 annual and seasonal (winter and summer) spatial patterns in terms of precipitation-weighted concentration and deposition. Temporal pattern information is presented using graphical displays of annual precipitation-weighted mean concentration and deposition and a statistical analysis based on the Kendall seasonal tau trend test. The temporal pattern analyses use a subset of 30 long-term sites over an 8-year (1979-1986) period and an expanded subset of 137 sets with greater spatial coverage over an 5-year (1982-1986) period. The 8-year period represents the longest period with wet deposition monitoring data available that has a sufficient number of sites with data of known quality to allow a descriptive summary of annual temporal patterns to be produced. The 5-year period restricts the number of years in order to increase the number of sites available with data of known quality. Section 2.0 describes the site selection and data summarization procedures, the spatial pattern methodology, and the temporal pattern graphical and statistical analysis methodology.

The section organization is as follows. First, a brief overview of emission trends is presented to provide a context for the interpretation of the deposition temporal patterns. Individual subsections describe spatial and temporal patterns for hydrogen (includes pH), sulfate, nitrate, ammonium, and calcium. The final subsection describes the temporal patterns for all other ions (sodium, chloride, potassium, and magnesium). Patterns that are common to multiple ions are discussed in Section 4.0. Rather than intersperse the many figures and tables used to support the descriptions, most figures and tables for this section appear in Appendix A.

3.1 EMISSION TEMPORAL PATTERNS 1979-1986

The temporal pattern of emissions for total suspended particulates (TSP), sulfur oxides, nitrogen oxides and volatile organic compounds provide a context for the interpretation of wet deposition temporal patterns. Nationwide temporal patterns in emissions indicate whether emissions are generally increasing or decreasing nationally, but give no information on regional differences in emission temporal patterns. This report makes no attempt to statistically relate national temporal patterns in emissions to national temporal patterns in wet deposition.

Nationwide TSP emissions (EPA 1988) decreased monotonically by approximately 25% from 1979 to 1986 (Figure 3.1). Emissions decreased from 8.9 to 6.8 million metric tons per year. The reduction is primarily from the industrial processes and fuel combustion source categories. Although the decrease is monotonic, TSP emissions were essentially constant from 1982 to 1986. Sulfur oxide emissions decreased by 16% between 1979 and 1986 (Figure 3.2) but remained essentially constant from 1982 to 1986. Emissions decreased from 25.1 to 21.2 million metric tons per year and is monotonic except for a small increase in 1984. The decrease reflects the installation of flue gas desulfurization controls at coal-fired electric generating stations and a reduction in the average sulfur content of fuels consumed. Nitrogen oxides emissions generally decreased from 1979 to 1983, slightly increased in 1984 and 1985, and then decreased again in 1986 (Figure 3.3). Emissions in 1979 and in 1986 are 21.0 and 19.3 million metric tons per year, respectively, an 8% decrease. Vehicle emissions (transportation) and fuel combustion are responsible for the temporal pattern of total nitrogen oxides emissions. Emissions of volatile organic compounds (VOC) decreased from 24.3 million metric tons per year in 1979 to 19.5 million metric tons per year in 1986, a 20% decrease (Figure 3.4). The decrease is monotonic except for small increases in 1983 and 1984.

3.2 pH AND HYDROGEN ION PATTERNS

3.2.1 Spatial Patterns in 1986

The 1986 annual spatial pattern for pH (Figure A.1) shows pH values below 5.0 for the eastern half of North America and pH values greater than 5.0 for the western half, except for the Southwest and southern Rocky Mountain regions. An ellipsoidal area of low pH (<4.2) extends from mid-Ohio and mid-West Virginia to western New York and from southern Ontario to eastern Pennsylvania and northern Virginia. Fourteen sites have a pH value below 4.2 and the

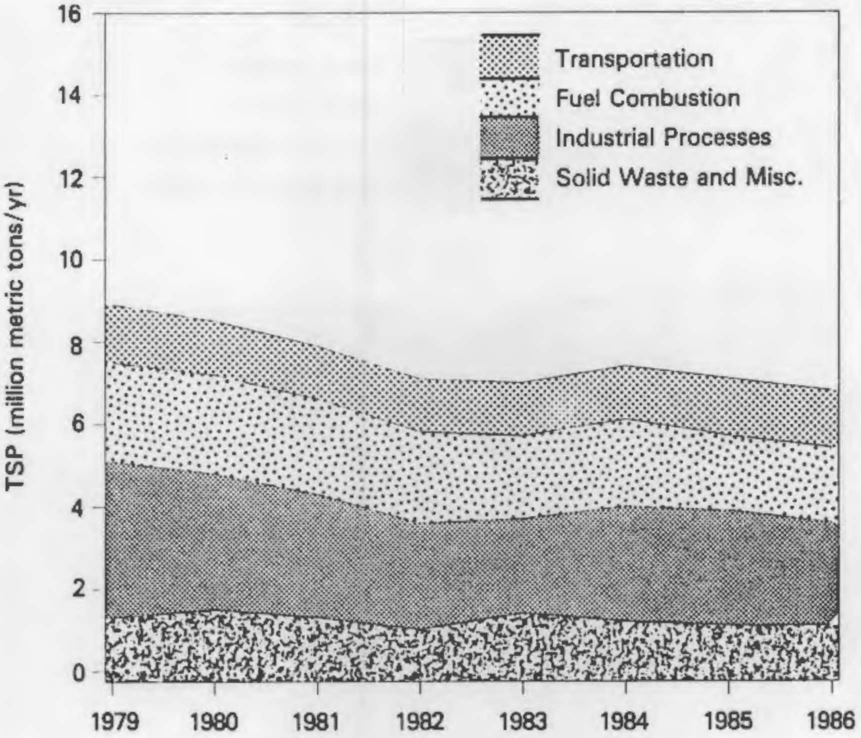


FIGURE 3.1. National Trend in Total Suspended Particulate Emissions, 1979-1986.

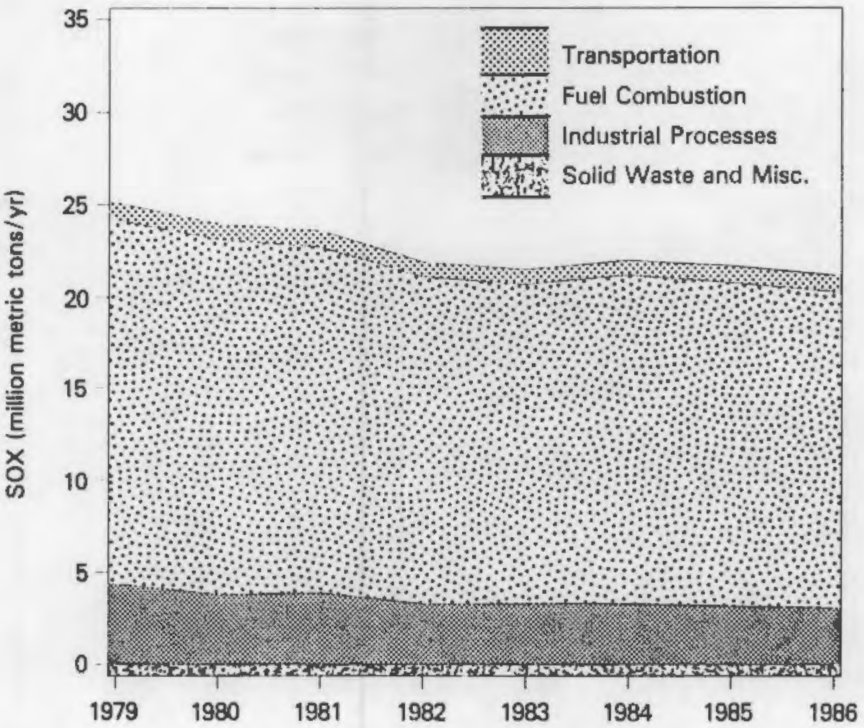


FIGURE 3.2. National Trend in Sulfur Oxide Emissions, 1979-1986.

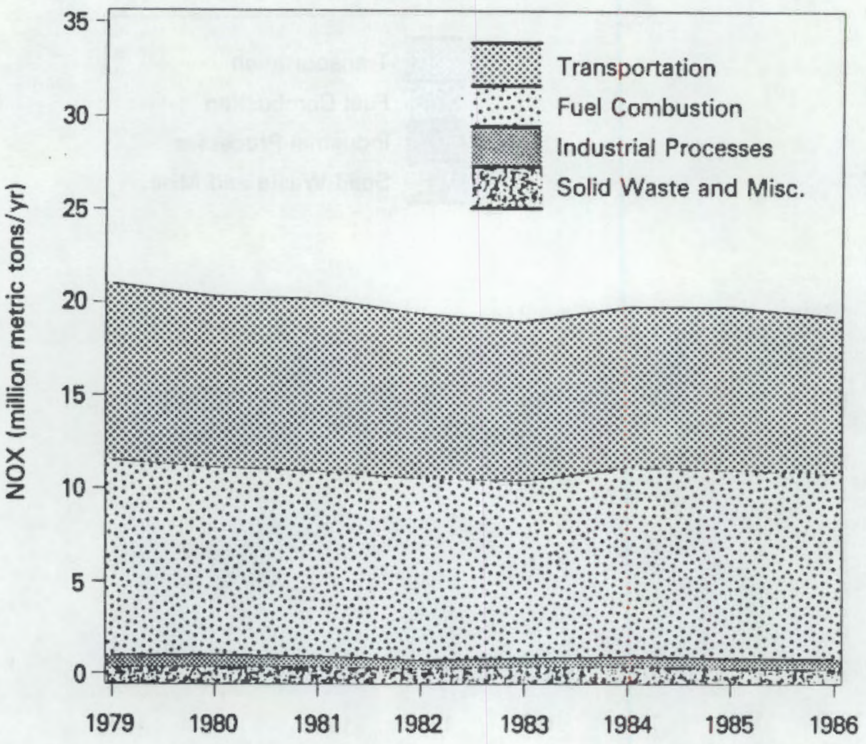


FIGURE 3.3. National Trend in Nitrogen Oxide Emissions, 1979-1986.

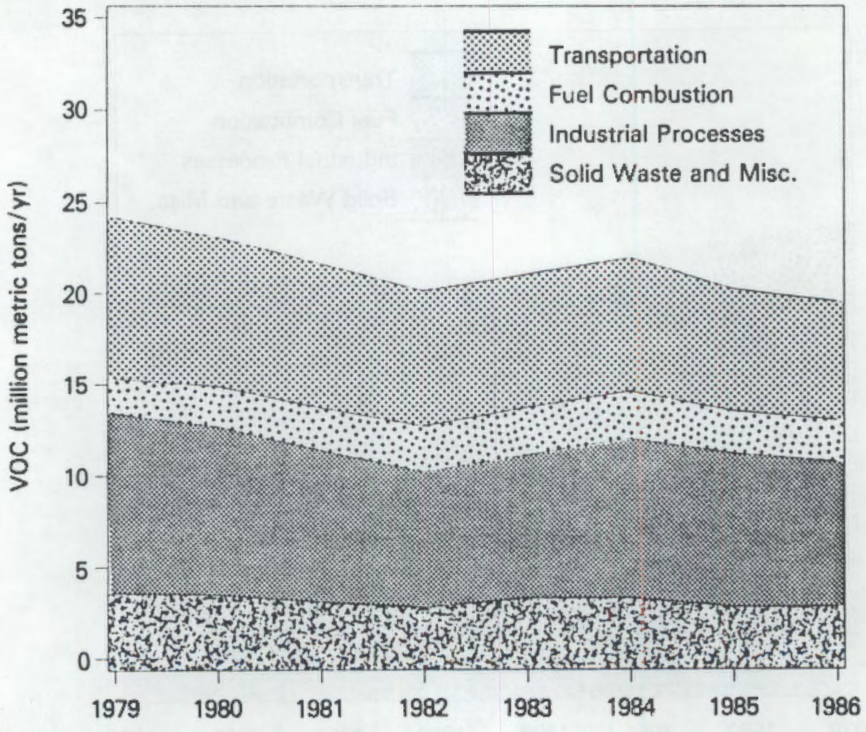


FIGURE 3.4. National Trend in Volatile Organic Compound Emissions, 1979-1986.

lowest two values are 4.12 at Kane, Pennsylvania and Parsons, West Virginia. The maximum pH is 5.66 at Las Animas in southeastern Colorado. The 1986 and 1985 (not shown) pH spatial patterns have very similar isopleth lines for 4.2 to 4.8. The 5.0 contour line differs but the change appears to be related to the availability of more sites in 1986, especially in Colorado.

The winter and summer spatial patterns for pH (Figures A.2 and A.3) are different and differ from the annual pattern. In winter, the area of low pH (<4.2) is larger than the annual region and extends from western Ohio to western New York and Pennsylvania and from northern Virginia to northern portion of southern Ontario. pH values below 5.0 occur east of a line from New Mexico to northern Minnesota. The location of the 5.0 isopleth line is difficult to determine due to the low density of sites in the area. The minimum winter pH is 4.04 at Chautauqua, New York, and the maximum winter value is 5.75 at Smith Valley, Nevada. In summer, the area of low pH (<4.2) extends from northeastern border of Kentucky to the western border of Maine and from mid-Virginia to northern New York. A region with pH below 4.0 extends from central Pennsylvania to northern West Virginia. Summer has a region of pH below 5.0 extending from southern Wyoming and western Colorado to the Mexican border, and from California to western New Mexico. The minimum summer pH is 3.91 at Penn State, Pennsylvania and the maximum summer value is 6.09 at Huron Well Field, South Dakota. Forty sites have summer pH values below 4.2.

The 1986 spatial pattern for hydrogen ion deposition (Figure A.1) has the region of greatest deposition (>0.6 kg/ha) over West Virginia, eastern Ohio, Pennsylvania, New York, southern Vermont, western Massachusetts, and Connecticut. The region with deposition greater than 0.6 kg/ha is larger than in 1985 (not shown). Although the isopleth lines give the impression of a reasonably smooth deposition surface, the spatial estimation procedure has smoothed the substantial site-to-site variation in the high deposition area. For example, Sutton, Quebec has a deposition of 0.86 kg/ha and a number of sites with deposition greater than 0.6 kg/ha in southern Ontario are intermixed with sites with deposition less than 0.6 kg/ha. The maximum annual hydrogen deposition is 1.02 kg/ha at Parsons, West Virginia. Other sites with deposition greater than 0.8 kg/ha are Kane and Leading Ridge, Pennsylvania and Bennett Bridge, New York.

The 1986 winter and summer hydrogen deposition spatial patterns (Figures A.2 and A.3) both have their highest deposition regions in western Pennsylvania. Approximately twice the deposition occurs in summer than in winter (Table A.4). Sites with the highest summer deposition are Parsons, West Virginia (0.46 kg/ha); Kane, Pennsylvania (0.41 kg/ha); Chautauqua, New York (0.36 kg/ha); Leading Ridge, Pennsylvania (0.36 kg/ha), and Bennett Bridge, New York (0.35 kg/ha). Sites with the highest winter deposition are Kane, Pennsylvania (0.19 kg/ha); Chautauqua, New York (0.17 kg/ha); Parsons, West Virginia (0.15 kg/ha); Priceville, Ontario (0.15 kg/ha), and West Point, New York (0.15 kg/ha).

3.2.2 Annual Temporal Patterns 1979-1986

The 30 site 1979-1986 trend subset shows a weak temporal pattern for annual pH (Figure A.4 and Table A.5). The 25th and 75th percentile and the median annual pH are lowest in 1980, increase until 1983 and then decrease until 1985 and 1986. The lowest annual pH values are 4.07 and 4.09 in 1980 and 1981 at Caldwell. Caldwell, Zanesville, and Penn State consistently have among the lowest pH values in each year since they are located in the region of lowest pH. Similarly, four sites with the highest pH values in all years are Manitou, Mead, Lamberton, and Marcell. Although the specific rank order of sites varies from year-to-year, the order is generally consistent across years. The hydrogen ion concentration temporal pattern is simply the reverse of the pH pattern (Figure A.6 and Table A.5).

Annual hydrogen deposition has a similar but less evident temporal pattern than hydrogen concentration (Figure A.6 and Table A.6). Sites and years with an annual hydrogen deposition greater than 0.90 kg/ha are Zanesville (1.06, 1.11, and 0.90 kg/ha in 1979, 1980, and 1981, respectively), Penn State (0.93 kg/ha in 1984); and Parsons (0.97 in 1985 and 1.02 in 1986). With the exception of the highest deposition each year at Penn State or Parsons, annual deposition is less than 0.75 kg/ha after 1981. At least four sites had annual deposition greater than 0.75 kg/ha from 1979 to 1981, suggesting that deposition may have decreased at many higher deposition sites.

3.2.3 Annual Temporal Patterns 1982-1986

The 137 site 1982-1986 trend subset has no discernable temporal pattern for annual pH (Figure A.5 and Table A.7). The 25th and 75th percentiles have a very weak tendency toward higher pH values during the period. The 1982 pH distribution appears shifted toward lower pH values. The hydrogen concentration temporal pattern reflects the pH pattern (Figure A.7 and Table A.7). The median annual hydrogen ion deposition for 1982-1986 sites is 0.42 kg/ha in 1982, 0.38 kg/ha in 1984, and 0.36 kg/ha in the other years (Figure A.7 and Table A.8). The maximum deposition (1.02 kg/ha) at a site occurs at Parsons, West Virginia in 1986. Kane, Pennsylvania ranks second or third in each year with a deposition range from 0.81 kg/ha to 0.93 kg/ha. Bennett Bridge, New York (0.94 kg/ha in 1986) and Penn State, Pennsylvania (0.94 kg/ha in 1984) are the only other sites with an annual deposition greater than 0.90 kg/ha.

3.2.4 Temporal Change Analysis

The Kendall seasonal tau (KST) test provides a quantitative analysis for a temporal change in pH and hydrogen ion concentration and deposition (see Section 2.0 for methodology). Based on the KST test, four 1979-1986 trend sites exhibit a significantly ($p < 0.05$) increasing trend in pH: Raleigh, Ithaca, Scranton and Zanesville (Figure A.10). The maximum estimated increasing trend is 0.02 pH units/year at Ithaca. One site, Bradford Forest, has a significantly decreasing pH trend (-0.02 pH units/year). Figure A.10 displays the trend estimates and confidence intervals for all 30 sites. The most notable feature of the estimates is that they have a symmetric distribution centered on zero change. Also, the estimates appear to have no consistency in direction within any geographic region. With the exception of Lamberton and Mead, the half-width of the confidence interval indicates that the month-to-month variation at the sites is similar.

The pH temporal pattern for the 1982-1986 trend sites is similar to the 1979-1986 trend site pattern (Figure A.11). Nine sites have a significantly increasing pH and four sites have a significantly decreasing pH ($p \leq 0.05$). Sites with decreasing pH are Great Smokey Mountains, Parsons, Coweeta, and Walker Branch. All sites with increasing pH are located in southern Ontario. If all confidence intervals are statistically independent and the KST significance levels are correct, then probabilistically one expects 3 or 4 decreasing and 3 or 4 increasing significant trends under a hypothesis of no temporal change. Temporal changes do show some consistency in several geographic regions. pH tends to increase in the Great Plains region and in the Ontario region.

The hydrogen concentration temporal pattern is characterized by the pH pattern, except increasing and decreasing changes are reversed (Figures A.12 and A.13). That is, hydrogen concentrations decrease significantly ($p<0.05$) at Ithaca, Scranton, Raleigh, and Zanesville. Overall the median for 1979-1986 trend sites is -0.19 μeq/L/year and 25% of the sites have trend estimates of -1.10 μeq/L/year or less. For the 137 1982-1986 trend sites, the median trend estimate is -0.37 μeq/L/year and the 25th percentile is -1.38 μeq/L/year. The largest decreasing trends occur in southern Ontario, Ithaca, Charlottesville, and Underhill Center. However, no geographic consistency in trend direction is apparent in the vicinity of the three United States sites nor are they statistically significant.

For 1979-1986 trend sites (Figure A.14), significant hydrogen deposition decreases occur at four sites (Zanesville, Rockport, Raleigh, and Clinton Station) and increases occur at two sites (Brookhaven and Bradford Forest). The 25th percentile, median, and 75th percentile temporal changes are -1.32, -0.10, and 0.61 meq/m^2/year, respectively. The largest decrease (-5.64 meq/m^2/year) occurs at Zanesville. For 1982-1986 trend sites (Figure A.15), the 25th percentile, median, and 75th percentile temporal changes are -1.85, -0.40, and 0.40 meq/m^2/year, respectively. The largest decreases are -7.34 and -6.82 meq/m^2/year at Raleigh, North Carolina, and Selma, Alabama, respectively. Sites with increases larger than 5 meq/m^2/year are Whiteface (8.28), Bennett Bridge (8.26), Chautauqua (5.56) and Kane (5.28). Ten sites have significant decreases and three sites have significant increases. No strong geographic pattern in trend direction is apparent.

3.3 SULFATE PATTERNS

3.3.1 Spatial Patterns in 1986

The spatial pattern for 1986 annual sulfate concentration (Figure A.16) is typified by high concentrations in central eastern North America and by low concentrations elsewhere. An elliptical region centered in eastern Ohio has concentrations greater than 3.5 mg/L. The highest concentration is 3.96 mg/L at Parsons (Table A.3). Eight other sites with concentrations greater than 3.5 mg/L are in southern Ontario, New York, Pennsylvania, and Ohio. A contour line for concentrations greater than 2.0 mg/L extends from western Illinois to the East Coast and from Tennessee to southern Ontario.

Sulfate annual deposition has a spatial pattern similar to sulfate concentration (Figure A.16). A contour line for annual deposition greater than 35 kg/ha extends from northern West Virginia to north of Lake Ontario and from the eastern Ohio border to western Pennsylvania and western New York. The maximum deposition is 53.3 kg/ha at Parsons, West Virginia (Table A.4). Four additional sites have sulfate depositions between 40 and 45 kg/ha: Bennett Bridge and Chautauqua, New York; Kane, Pennsylvania; and Wellesley, Ontario. The 20 kg/ha contour line encloses an elliptical region from northern Alabama and Georgia to southern Ontario and New Hampshire and from eastern Wisconsin and central Illinois to eastern Virginia and Massachusetts.

Winter sulfate concentrations have a spatial pattern with the highest concentration region shifted further west than for the annual (Figure A.17). The 3.0 mg/L contour line encloses two regions: one centered in northwestern Illinois and one centered in northern Ohio, southern Ontario, northwestern Pennsylvania, and western tip of New York. The maximum winter concentrations are 4.29 mg/L at Wilkesport, Ontario; 3.96 mg/L at Argonne, Illinois; 3.69 mg/L at Merlin, Ontario; and 3.51 mg/L at Port Stanley, Ontario. Parsons, the location of the highest annual concentration, has a winter concentration of 3.39 mg/L. Winter sulfate concentrations are lower in eastern New York and New England when compared to annual concentrations.

The spatial pattern for summer sulfate concentration differs markedly from the annual and winter patterns (Figure A.18). A primary difference is that summer concentrations are consistently higher compared to winter or annual. The region enclosed by the 2.0 mg/L contour for annual concentration is similar to the region enclosed by the 3.0 mg/L contour for summer concentration. The highest concentrations, within the 5.0 mg/L contour, occur in western Pennsylvania and northern West Virginia. The maximum summer concentration is 5.50 mg/L at Penn State (sampler operated by MAP3S network). Other concentrations greater than 5.0 mg/L are 5.42 mg/L at Penn State (NAOP/NTN sampler), 5.22 mg/L at Leading Ridge, 5.07 mg/L at Parsons, and 5.02 mg/L at Kane. Dana and Easter (1987) discuss the existence of a seasonal cycle for sulfate concentration. Although the concentration levels are lower, sulfate concentrations are also higher in summer than in winter in the western United States.

Winter sulfate deposition is highest (>5.0 kg/ha) in western Pennsylvania, eastern Ohio, and northern West Virginia (Figure A.17). This is the same region for high annual deposition, except it does not include western New York. Deposition is less than 2.0 kg/ha west of the Mississippi River. The maximum winter deposition is 8.7 kg/ha at Parsons (Table A.4). The next two highest deposition sites are Chautauqua (6.4 kg/ha) and Kane (5.8 kg/ha). Two sites among those with high winter deposition are American Samoa (5.6 kg/ha) and El Verde, Puerto Rico (5.4 kg/ha), both influenced by sea salt. The effect of sulfate associated with sea salt is apparent in the relatively high deposition along the eastern coast.

Summer sulfate deposition greater than 15.0 kg/ha occurs from southeastern Ontario and eastern Ohio to central West Virginia, central Pennsylvania and most of New York (Figure A.18). Deposition is less than 5.0 kg/ha west of the Mississippi River. The maximum summer deposition of 23.9 kg/ha occurs at Parsons, the same location as the winter maximum (Table A.4). Kane with 20.6 kg/ha is the only other site with summer deposition greater than 20.0 kg/ha. Only sites in West Virginia, Pennsylvania, New York, and southern Ontario have depositions greater than 15.0 kg/ha. To provide a contrast with winter, the summer sulfate depositions at American Samoa and El Verde are 2.0 and 7.0 kg/ha, respectively. The influence of sea salt is less noticeable during summer. Summer sulfate deposition is approximately three times winter sulfate deposition. The difference is partially a function of the higher summer concentrations and partially a function of greater summer precipitation amounts.

3.3.2 Annual Temporal Patterns 1979-1986

Annual sulfate concentrations in the 1979-1986 trend subset have a definite temporal pattern (Figure A.19 and Table A.5). The median and 75th percentile of the sites are highest in 1980 and 1981 and lowest in 1983.

The boxplots consistent pattern of movement from year-to-year suggests that the site concentrations all tend to follow the same pattern. The variability of the pattern precludes using a linear trend model to represent the temporal pattern. Annual sulfate concentrations exceed 4.0 mg/L in 1979, 1980, and 1981 only at Caldwell, Ohio. Caldwell is also the highest or second highest in all subsequent years. The lowest concentration is 0.87 mg/L in 1985 at Manitou, Colorado. Manitou has the lowest concentration in all years. Annual precipitation also has a temporal pattern that is out of phase with the pattern for sulfate. That is, the median and percentiles are lowest in 1980 and 1981 and highest in 1983 and 1984. The two patterns suggest an inverse relationship between annual sulfate concentration and annual precipitation amount. Schertz and Hirsch (1985) and Dana and Easter (1987) have demonstrated such an inverse relationship for event and weekly wet deposition.

The temporal pattern for sulfate deposition is not as well defined as for concentration (Figure A.19 and Table A.6). The median and 75th percentiles suggest a decrease in deposition over the period, but yearly variation clouds the picture. Deposition greater than 45 kg/ha occurs at Caldwell in 1979 and at Parsons in 1980, 1981, 1985, and 1986. The maximum deposition is 53.3 kg/ha in 1986 at Parsons. The minimum deposition in each year occurs at Manitou, Colorado. Note that the minimum reported for 1981 is actually Scranton but this is the result of Scranton only operating for part of 1981. A temporal decrease in deposition is apparent for Manitou.

3.3.3 Annual Temporal Patterns 1982-1986

The boxplots for the 1982-1986 trend sites do not show the same well defined pattern for sulfate concentration as the 1979-1986 trend sites (Figure A.20). Similarly, the boxplots for sulfate deposition show no discernable pattern. Deposition greater than 45 kg/ha occurs at Parsons in 1985 (47.6 kg/ha) and in 1986 (53.3 kg/ha), and at Colchester, Ontario in 1985 (45.6 kg/ha).

3.3.4 Temporal Change Analysis

The Kendall seasonal tau test for trend identifies eight sites in the 1979-1986 trend sites with a significant ($p<0.05$) decreasing pattern for sulfate concentration (Figure A.21). The presence of the previously identified temporal pattern complicates, and may even invalidate, the interpretation of a linear trend. The eight sites are Lamberton, Mead, Manitou, and Marcell west of the Mississippi River and Piedmont Station, Oxford, Ithaca, and Hubbard Brook east of the Mississippi River. Although not all are statistically significant, all but two of the thirty sites have decreasing trend estimates. The 25th, 50th, and 75th percentiles for trend estimates are -1.57, -0.91, and -0.23 μeq/L/year, respectively. The minimum trend estimate occurs at Mead (-3.15 μeq/L/year). The maximum occurs at Lewes (0.40 μeq/L/year).

The 1982-1986 trend sites have 16 sites with a significant decreasing trend and two sites with a significant increasing trend (Figure A.22). Sites with a significant increasing trend are Parsons and Great Smokey Mountain. Other sites near Great Smokey Mountain also have increasing trend estimates but are not significant. The increasing trend at Parsons appears to be a more local phenomenon. The greatest decreasing trends occur along the western side of the southern tip of Ontario and are from sites operated by the APIOS-C network. Sites in the United States with significant decreasing trends do not occur in any particular region. The 25th, 50th, and 75th percentiles for the 1982-1986 trend sites are -1.66, -0.62 and 0.49 μeq/L/year, respectively. The minimum trend estimate is -8.45 μeq/L/year at Colchester, Ontario and the maximum is 5.67 μeq/L/year at Parsons.

Significant decreasing trends for sulfate deposition are found at eight sites in the 1979-1986 trend sites (Figure A.23). The sites are Coweeta, Piedmont Station, Clinton Station, Finley, and Raleigh, North Carolina; Manitou, Colorado; Zanesville, Ohio; and Hubbard Brook, New Hampshire. The North Carolina sites have regional consistency. Zanesville is consistent with other sites in Ohio and West Virginia. The sites in New York nearest Hubbard Brook have increasing trend estimates, hence no regional consistency is apparent. The 25th, 50th, and 75th percentiles for the 1982-1986 trend sites are -1.90, -0.62, and 0.00 meq/m^2/year, respectively. The minimum trend estimate is -3.75 meq/m^2/year occurring at Coweeta and Zanesville, and the maximum is 0.83 meq/m^2/year at Fort Wayne.

Thirteen sites have significant decreasing sulfate deposition trends in the 1982-1986 trend sites (Figure A.24). Six sites are in the western United States where almost all sites have decreasing trends. Two sites are in

Mississippi and two sites are in Raleigh, North Carolina. Both sets are regionally consistent with nearby sites. The remaining sites are in southern Ontario and regional consistency, if any, is weak. The lack of regional consistency may be related to the sites in the region being operated as three different networks. Scranton is the only site with a significant increasing trend (this may be due in part to missing data during part of 1982) and no dominant regional trend pattern is apparent. The 25th, 50th, and 75th percentiles for the trend sites are -2.50, -0.62, and 0.00 $meq/m^2/year$, respectively. The minimum trend estimate is -6.45 $meq/m^2/year$ at Clinton, Mississippi, and the maximum is 5.41 $meq/m^2/year$ at Chautauqua, New York.

3.4 NITRATE PATTERNS

3.4.1 Spatial Patterns in 1986

The spatial pattern for 1986 annual nitrate concentration (Figure A.25) has a region of high concentration (> 2 mg/L) over southern Ontario. In contrast to the sulfate concentration pattern, a contour line at 1.0 mg/L extends across the northern Great Plains to Colorado. One explanation for the higher concentrations in this region is the influence of agricultural activities. The maximum concentration is 2.72 mg/L at Killarney, Ontario. In fact, the ten highest concentrations, all greater than 2.30 mg/L, occur in southern Ontario.

Annual nitrate deposition has essentially the same spatial pattern as nitrate concentration but locally is more variable (Figure A.25). A 25.0 kg/ha contour encloses a small region centered at the northern tip of Lake Erie. Eight sites have nitrate depositions greater than 25.0 kg/ha: Bennett Bridge, Parsons, Chautauqua, and West Point in the United States, and Wellesley, North Easthope, Huron Park, and Sutton in Canada. Bennett Bridge has the maximum deposition at 32.4 kg/ha.

Winter nitrate concentrations are greater than annual concentrations in the highest concentration region (Figure A.26). A 3.5 mg/L contour encloses a narrow region along Lake Erie extending from Detroit to the eastern tip of Ontario. Of eighteen sites with concentrations greater than 3.5 mg/L, only Chautauqua and Huntington, New York are in the United States. Charlottesville, Virginia is the only additional United States site among sites with concentrations greater than 3.0 mg/L, all other sites are in southern Ontario. The maximum winter nitrate concentration is 5.17 mg/L at Wilkesport, Ontario, located at the southern end of Lake Huron.

Summer nitrate concentrations are generally comparable to annual nitrate concentrations in magnitude and spatial pattern in high concentration regions (Figure A.27 and Table A.3). Differences in the spatial pattern occur for the 2.0 mg/L contour where the summer contour extends to the Atlantic coast and for the 1.0 mg/L contour where the summer contour extends further south to the coast of Louisiana and across the west to the Pacific Ocean. The maximum summer concentration is 3.04 mg/L at West Point. The next highest values are 2.91 mg/L at Tanbark Flat, California, northeast of Los Angeles; 2.79 mg/L at Balsam Lake, Ontario; and 2.71 mg/L at White Rock, Maryland. Winter nitrate concentrations are higher than summer concentrations in the Northeast but summer concentrations are higher than winter in the South and West.

The geographic region enclosed by the 3.0 kg/ha contour for winter nitrate deposition is comparable to the region enclosed by the 15.0 kg/ha contour for annual nitrate deposition. The region with maximum winter deposition (>6.0 kg/ha) is the same region as for maximum annual deposition. The maximum deposition is 9.4 kg/ha at Priceville, Ontario. Only Chautauqua (7.8 kg/ha), Kane (7.2 kg/ha), Parsons (6.6 kg/ha) in the United States have deposition greater than 6.0 kg/ha, all other sites are in southern Ontario. During the winter, deposition less than 1.0 kg/ha occurs west of the Mississippi River and in Florida.

Summer nitrate depositions are greater than winter nitrate depositions (Figure A.27 and Table A.4). The 5.0 kg/ha contour line for summer deposition encloses the same region as the 15.0 kg/ha contour line for annual deposition. Approximately one third of the annual deposition occurs during the summer. Summer nitrate deposition is greater than 1.0 kg/ha east of the Rocky Mountain region. In Florida, nitrate deposition during the summer accounts for most of the annual deposition. Greater summer deposition is related to higher summer concentrations and larger precipitation amounts compared to winter. The maximum summer nitrate deposition is 12.2 kg/ha at West Point. Other sites with deposition greater than 9.0 kg/ha are Chautauqua, Kane, and Parsons in the United States, and Uxbridge, Huron Park, Wellesley, and Waterloo in Ontario.

3.4.2 Annual Temporal Patterns 1979-1986

Annual nitrate concentrations in the 1979-1986 trend sites have a definite temporal pattern that is in phase with sulfate concentrations (Figure A.28 and Table A.5). The 25th, 50th, and 75th percentiles are high in 1980, 1981, and 1986, and are low in 1983 and 1984. The presence of the temporal pattern complicates the assessment of a linear trend, as with sulfate concentration. An inverse relationship appears to exist between annual nitrate concentration and annual precipitation amount. The maximum concentrations are 2.22 mg/L at Delaware in 1980, 2.15 mg/L at Penn State in 1982 and 1984, 2.21 mg/L at Ithaca in 1985, and 2.14 mg/L at Charlottesville, Virginia in 1986.

No temporal pattern or linear trend is visually obvious for nitrate deposition (Figure A.28). Parsons has the highest deposition in all years except 1983 and 1984. Sites and years with annual deposition greater than 20 kg/ha are Parsons, Caldwell, Zanesville, Delaware, and Penn State in 1979; Parsons, Delaware, Zanesville, and Caldwell in 1980; Parsons, Ithaca, and Zanesville in 1981; Penn State and Ithaca in 1984; Parsons in 1985; and Parsons in 1986. The maximum deposition is 27.7 kg/ha at Parsons in 1986. Manitou, Colorado has the minimum deposition each year (see note on Scranton in sulfate section) and deposition at Manitou consistently decreases with time.

3.4.3 Annual Temporal Patterns 1982-1986

The boxplots for the 1982-1986 trend sites show the nitrate concentration distributions in 1982 and 1983 to be at a lower average level than the 1984-1986 distributions (Figure A.29). The plot also suggests more site-to-site variation during the latter years, i.e., the interquartile range is greater. The nitrate deposition boxplots show no discernable trend but also exhibit the same increased variation during the latter years. The maximum depositions in each year are 26.5 kg/ha at Port Stanley, 28.4 kg/ha at Bennett Bridge, 29.6 kg/ha at Huron Park, 30.9 kg/ha at Bennett Bridge, and 32.4 kg/ha at Bennett Bridge in 1982 to 1986, respectively. The increased variation and the sequence of annual maximums suggest that nitrate deposition may increase in high deposition region over the 5-year period.

3.4.4 Temporal Change Analysis

The Kendall seasonal tau test for trend identifies four sites with a significant ($p<0.05$) decreasing trend for nitrate concentration (Figure A.30). The sites are Lamberton, Mead, Marcell, and Oxford. Based on only the four western sites available, a consistent regional decreasing trend is present in the upper Great Plains. Sites near Oxford also have decreasing trends but they are not as large and Zanesville in eastern Ohio has an increasing trend estimate. The northeast has no consistent pattern for nitrate concentration trend estimates. The 25th, 50th, and 75th percentiles for the 30 1979-1986 trend sites are -0.42, -0.01, and 0.30 μeq/L/year, respectively. The minimum trend estimate is -1.34 μeq/L/year at Lamberton and the maximum is 0.73 μeq/L/year at Finley. With the exception of the four western sites, the trend analysis shows no consistent pattern toward either increasing or decreasing trends.

Eleven 1982-1986 trend sites have significant increasing nitrate concentration trend estimates and three sites have significant decreasing trend estimates (Figure A.31). Alsea, Oregon; Sand Springs, Colorado, and Victoria, Texas are the sites with decreasing trend estimates (all are only marginally significant). Sites with increasing trend estimates are Wellesley, Nithgrove, Balsam Lake, and Melbourne, Ontario; Brookhaven, New York; Parsons, West Virginia; Zanesville, Ohio; Fernberg, Minnesota; Lewes, Delaware; Georgia Station, Georgia, and Coweeta, North Carolina. The maximum trend estimate is 3.17 μeq/L/year at Wellesley and the minimum is -2.86 μeq/L/year at Colchester, both in southern Ontario. Little geographic consistency is apparent in either the direction or magnitude of the linear trend estimates. However, the 25th, 50th, and 75th percentiles are -0.34, 0.12 and 0.81 μeq/L/year suggesting an overall tendency toward increasing trend estimates for nitrate concentration during the period.

The 1979-1986 trend subset has two sites with significant decreasing trends and one site with a significant increasing trend for nitrate deposition (Figure A.32). Hubbard Brook with a -1.37 meq/m^2/year trend estimate and Manitou with a -0.39 meq/m^2/year trend estimate are the sites with significant decreasing trends. Whiteface at 1.29 meq/m^2/year is the site with a significant increasing trend. No consistent regional pattern is apparent in the trend estimates. The 25th, 50th, and 75th percentiles for the 1979-1986 trend sites are -0.48, 0.00, and 0.33 meq/m^2/year, respectively. Nitrate deposition for the 1979-1986 trend sites has no overall linear trend pattern during the period.

Ten sites have significant decreasing nitrate deposition trend estimates and five sites have significant increasing trend estimates in the 1982-1986 trend sites (Figure A.33). Sites with significant increasing trend estimates are Wellesley, North Easthope, and Longwoods(B) in Ontario and Aurora and Whiteface, New York. Weak regional consistency may be present in upper New York. Sites with significant decreasing trends are Davis, Raleigh, Bridgton, Selma, Clinton, Everglades National Park, Hopland (Ukiah), Grand Canyon, Manitou, and Alsea. With the exception of Bridgton, the sites are in the West or the South. The 25th, 50th, and 75th percentiles are -0.65, 0.00, 0.60 meq/m^2/year for nitrate deposition trend estimates. Nitrate deposition for the 1982-1986 trend sites has no overall linear trend pattern during the 5-year period.

3.5 AMMONIUM PATTERNS

3.5.1 Spatial Patterns in 1986

Annual 1986 ammonium concentration has a spatial pattern dominated by a high concentration region centered in the northern Great Plains and another centered in the southern tip of Ontario (Figure A.34). Ammonium concentrations greater than 0.20 mg/L extend from the Rocky Mountains to Vermont and the Atlantic coast and from Nebraska, Arkansas, Tennessee, and North Carolina into Canada. Although the annual nitrate concentration spatial pattern is similar to that of ammonium, it does not have the second high concentration region in the west. The maximum concentration in the western region is 0.71 mg/L at Brookings, South Dakota, and in the Ontario region is 0.73 mg/L at Colchester. Other sites with annual ammonium concentrations greater than 0.50 mg/L are Lamberton, Minnesota; Pawnee, Colorado; Shawano, Wisconsin; and Mead, Nebraska in the west; and Palmerston, Wellesley, Huron Park, Waterloo, Merlin, Wilkesport, Melbourne, North Easthope, and Longwoods in Ontario.

The annual ammonium deposition spatial pattern also has two regions of high deposition (Figure A.34). In addition, deposition is greater than 1.0 kg/ha east of the Rocky Mountains, except for Florida. Note that the 2.0 kg/ha contour extends south to the Gulf Coast in Texas, Louisiana, and Mississippi. The maximum deposition is 7.50 kg/ha at Wellesley, Ontario (Table A.4). Five other sites in southern Ontario have depositions greater that 6.0 kg/ha. The highest deposition in the West is 5.39 kg/ha at Brookings, South Dakota. Mead, Nebraska, and Lamberton, Minnesota also are western sites with deposition greater than 5.0 kg/ha.

Winter ammonium concentrations are comparable to, or slightly less than, annual concentrations (Table A.3). The spatial pattern has high concentrations in northern Illinois, north of Lake Huron, and in southern Ontario (Figure A.35). No annual summaries meet the UDDC selection criteria to determine if a secondary maximum in South Dakota exists. The site-to-site variation in the winter suggests possible local influences for ammonium. The maximum winter concentration is 0.75 mg/L at Waterloo.

Summer ammonium concentrations show substantial geographic variation (Figure A.36). Although southern Ontario and the northern Great Plains remain high concentration regions, high concentrations also are present in Nevada, Colorado, western Kansas, and Mississippi (relative to nearby region). The maximum concentration is 0.97 mg/L at Colchester, Ontario. The next highest, and the maximum concentration in the United States, is 0.75 mg/L at Pawnee, Colorado. In Nevada, the maximum is 0.46 mg/L at Lehman Caves. This contrasts with a winter ammonium concentration of 0.04 mg/L at Lehman Caves.

The center of the region with the highest ammonium deposition in winter is the southern tip of Ontario (Figure A.35). Sites with deposition greater than 1.00 kg/ha are Priceville (1.40), Priceville 2 (1.11), Waterloo (1.06), and McKellar (1.01) in Ontario, and Chautauqua (1.14) in New York. Note that the two Priceville sites are co-located and the difference in the values demonstrates the variation possible in estimating annual deposition at a site. Winter accounts for less than one-fourth of the annual ammonium deposition.

Summer ammonium deposition is approximately three times greater than winter deposition (Table A.4 and Figure A.36). Approximately, one-third of the annual ammonium deposition occurs during the summer. Summer has two high deposition regions, similar to the annual spatial pattern. The maximum deposition is 4.06 kg/ha at Colchester. The next highest deposition is 2.78 at Huron Park. Other sites with deposition greater than 2.00 kg/ha are Palmerston, Waterloo, Merlin, Wellesley, Uxbridge, and North Easthope in Ontario, and Fort Wayne, Indiana. The secondary maximum is 1.59 kg/ha at Brookings, South Dakota.

3.5.2 Annual Temporal Patterns 1979-1986

Boxplots for annual ammonium concentrations at the 1979-1986 trend sites show higher concentrations for the middle 50% of the sites in 1980 and 1981 (Figure A.37 and Table A.5). Concentrations in 1986 are also larger but not by as much. These years correspond to years with lower annual precipitation amounts (Figure A.4). Ammonium concentrations greater than 0.50 mg/L occur at Lamberton in all years except 1985, at Mead in all years, at Fort Wayne in 1981, at Wooster in 1984, and at Delaware in 1985. The maximum concentration observed is 0.87 at Lamberton in 1983. The high concentrations at Mead and Lamberton is consistent with the secondary maximum in the 1986 spatial pattern. Since the 1979-1986 trend sites do not include any sites from Ontario, the analysis misses the maximum in southern Ontario.

Annual ammonium deposition temporal pattern is characterized by variation in the distributions for the years (Figure A.37 and Table A.6). The maximum deposition is 5.67 kg/ha in 1986 at Rockport. Other sites and years with deposition greater than 4.50 kg/ha are Lamberton in 1979, 1983, 1984, and 1986; Delaware in 1980; Fort Wayne in 1981, 1984, and 1986; and Mead in 1982, 1984, and 1986. Manitou has the lowest deposition in each year. A linear decrease superimposed on a weak temporal pattern may be present at Manitou.

3.5.3 Annual Temporal Patterns 1982-1986

The boxplots for annual ammonium concentration and deposition at the 1982-1986 trend sites show no definite temporal pattern over the 5-year period.

3.5.4 Temporal Change Analysis

The Kendall seasonal tau test for trend identifies two sites in the 1979-1986 trend sites with a significant ($p<0.05$) decreasing ammonium concentration trend (Figure A.39). Mead has a -2.74 μeq/L/year trend estimate and Lamberton has a -2.55 μeq/L/year trend estimate. With the exception of these two sites, no other temporal pattern is apparent. The 25th, 50th, and 75th trend estimate percentiles of the 1979-1986 trend sites are -0.41, 0.00, and 0.26 μeq/L/year.

The 1982-1986 trend sites have thirteen and four sites with significant decreasing and increasing trend estimates, respectively (Figure A.40). Sites with significant increasing trends are American Samoa, Parsons, Oak Ridge, Zanesville, and Brookhaven. Significant decreasing trend estimates occur at five western sites; five southern Ontario sites; Bridgton, Maine; Huntington, New York; and University Forest, Missouri. The three most extreme decreasing trend estimates are -3.52, -3.33, and -2.78 μeq/L/year at three southern Ontario sites. The most notable feature of the trend estimates, however, is the general lack of regional consistency and distribution pattern. The 25th, 50th, and 75th percentiles for the ammonium concentration trend estimates are -0.83, -0.09, and 0.28 μeq/L/year.

Ammonium deposition trend estimates for the 1979-1986 trend sites are significantly decreasing at Manitou and Zanesville, and significantly increasing at Whiteface and Scranton (Figure A.41). The trend direction at these sites appears to be in almost complete conflict with other nearby sites, lending little credence to any general region trend during the 5-year period. A notable feature of the confidence intervals for the estimates is that Lamberton and Mead have large half-widths indicating greater variation than for other sites. Manitou, in contrast, has a small half-width enabling a significant decreasing trend to be detected. The 25th, 50th, and 75th percentiles for the ammonium deposition trend estimates are -0.33, 0.00, and 0.22 meq/m^2/year.

Twenty-two sites have significant decreasing ammonium deposition trends in the 1982-1986 trend sites (Figure A.42). All but two of the sites are in the United States and are predominately located in the West, the South, and New England. Moreover, within nearby regions an indication of regional consistency is present. The two most extreme decreasing trend estimates are at Uxbridge, Ontario and Davis, California (not significant). Three sites have significant increasing trends: Whiteface, Chautauqua, and Scranton. Their trend direction is not consistent with nearby sites. Hence, local influences may be responsible for the increasing trends during the period.

The 25th, 50th, and 75th percentiles for the 1982-1986 ammonium deposition trend estimates are -0.88, -0.33, and 0.22 meq/m^2/year (Table A.8). The percentile distribution in conjunction with general regional consistency, except in southern Ontario and New York, suggests a general decreasing trend in ammonium deposition during the 5-year period.

3.6 CALCIUM PATTERNS

3.6.1 Spatial Patterns in 1986

The spatial pattern for 1986 annual calcium concentration differs from the corresponding spatial patterns for hydrogen, sulfate, nitrate and ammonium (Figure A.43). The high concentration region is still centered in the southern tip of Ontario, but the contour lines tend to extend southwest to the Mexican border. This is a reflection of calcium arising from a non-anthropogenic, soil related, source. The maximum concentration is 0.62 mg/L at Longwoods, Ontario (Table A.3). Other sites with concentrations greater than 0.40 mg/L are Wilkesport, Melbourne, Wellesley, and Merlin, all in Ontario. The maximum concentration in the United States is 0.38 mg/L at Red Rock Canyon, Nevada.

Annual calcium deposition has a spatial pattern with two maxima: one in southern Ontario, the location of the concentration maximum, and the other in eastern Kansas (Figure A.43). The maximum deposition is 5.96 kg/ha at Longwoods, Ontario (Table A.4). Of the fourteen sites that have annual deposition greater than 3.0 kg/ha, all but three sites are located in southern Ontario. The three sites are El Verde, Puerto Rico; Konza Prairie, Kansas; and Parsons, West Virginia.

The spatial pattern for winter calcium concentration has many of the same features as the annual pattern (Figure A.44). Differences in the sites used for contouring make a direct comparison difficult. The maximum concentration is 1.14 mg/L at Moonbeam in northern Ontario. Since a nearly co-located site has a concentration of 0.08 mg/L, the high concentration is likely to be due to local sources or contamination. The second highest concentration is 0.85 mg/L at Merlin, Ontario. The highest concentration in the United States is 0.67 mg/L at Indiana Dunes, Indiana, and in the western United States is 0.37 at Mesa Verde, Colorado.

Summer calcium concentrations are similar to annual concentrations in the high concentration area in southern Ontario and are substantially greater in southern California, Nevada, Arizona, and Texas (Figure A.45). The maximum concentration is 1.16 mg/L at Palomar Mountain, California. The second highest concentration is 0.89 mg/L at Longwoods, Ontario. In all, six sites in Ontario, six sites in the western and southwestern United States, and two in northern Illinois and Indiana have concentrations greater than 0.40 mg/L.

The spatial pattern for winter calcium deposition has a high deposition region centered in southern Ontario and extending to northern Illinois (Figure A.44). Within this region, deposition in winter accounts for 20% or less of the annual deposition. The maximum deposition, however, is 1.39 kg/ha at Parsons, West Virginia. Deposition greater than 0.50 kg/ha occurs at fifteen sites, ten located in Ontario. In addition to Parsons, the other sites not in Ontario are American Samoa; El Verde, Puerto Rico; and Indiana Dunes and Huntington, Indiana.

The summer calcium deposition spatial pattern is more variable and has several regions of secondary maxima (Figure A.45). The pattern is similar the annual pattern, in part due to the summer quarter contributing the most to the annual deposition in high deposition regions. The maximum deposition is 2.49 kg/ha at Longwoods, Ontario. The maximum in the United States is 1.43 kg/ha at Farlington, Kansas.

3.6.2 Annual Temporal Patterns 1979-1986

The 1979-1986 trend sites appear to have a decreasing calcium concentration trend superimposed on a weak temporal pattern (Figure A.46 and Table A.5). The temporal pattern is out of phase with the pattern for annual precipitation amount (Figure A.4). That is, high calcium concentration years are low annual precipitation amount years. This is consistent with calcium being a soil-related ion. The two highest concentrations in each year are from Lamberton and Mead. Visual inspection of the extremes on the boxplots leads one to conclude that calcium concentrations have decreased at Lamberton and Mead during the 8-years. The maximum concentration is 0.67 mg/L in 1980 at Mead, Nebraska.

Annual calcium deposition has a temporal pattern that is similar to, but weaker and more variable than calcium concentration (Figure A.46). More site-to-site variation is present in 1979, 1980, 1985, and 1986 than in the middle four years. In addition, deposition appears to be substantially lower in 1985 and 1986 than in previous years (Table A.6). For example, two-thirds of the sites have an deposition less than 1.50 kg/ha in 1985 and 1986; while less than one-half of the sites do in any previous year. The three highest deposition sites in any year are usually Mead, Lamberton, and Parsons. However, the maximum deposition is 3.91 kg/ha at Zanesville in 1980.

3.6.3 Annual Temporal Patterns 1982-1986

The boxplots for the 1982-1986 trend sites show a weak temporal pattern for calcium concentration and a possible decrease in calcium deposition in 1985 and 1985 (Figure A.47). The two highest concentrations are 0.91 mg/L in 1983 at Uxbridge, Ontario; and 0.89 mg/L in 1984 in Mesa Verde, Colorado. The two highest depositions are 7.88 kg/ha in 1984 at Huron Park, Ontario; and 6.83 kg/ha in 1983 at Uxbridge, Ontario.

3.6.4 Temporal Change Analysis

All 1979-1986 trend sites have negative or zero trend estimates (decreasing or no trend) based on Sen's median trend estimate associated with the Kendall seasonal tau test (Figure A.48). Seventeen of the thirty sites have significant ($p<0.05$) decreasing trends. Mead and Lamberton have the most extreme decreasing trend estimates of -1.96 and -1.74 μeq/L/year, respectively. The trend estimates also exhibit consistency among nearby sites. The 25th, 50th, and 75th percentiles for the thirty trend estimates are -0.61, -0.33, and -0.16 μeq/L/year. The maximum trend estimate is zero.

The temporal pattern for the 1982-1986 trend sites is not as consistent (Figure A.49). Significant decreasing trend estimates for calcium concentration occur at twenty sites and significant increasing trend estimates occur at six sites. The six sites with significant increasing trends are all sites operated by the APIOS-D network in Ontario. The increasing trend at the APIOS-D Fernberg site conflicts with the decreasing trend estimate for the co-located NADP/NTN sampler. The five sites with the most extreme decreasing trend estimates are in southern Ontario and are operated by the

APIOS-C network. The conflict arising from having sites with the most extreme increasing and decreasing trends being located in the same region but operated under two different sampling protocols requires further study. Sites in the United States with significant decreasing trend estimates are generally regionally consistent with trend estimates at nearby sites. The 25th, 50th, and 75th percentiles for the 137 trend estimates are -0.59, -0.25, and 0.00 μeq/L/year. Of the 31 sites with increasing trend estimates, only eight are located in the United States. In view of the general regional consistency in the trend estimates in the United States and the existence of 8 out of 98 sites in the United States with increasing trend estimates, the existence of an overall decreasing trend in calcium concentration during the 5-year period is postulated.

Seventeen of the 1979-1986 trend sites have significant decreasing trend estimates for calcium deposition (Figure A.50). No site has an increasing trend estimate based on Sen's median slope estimate. The trend estimates are regionally consistent with nearby sites. The 25th, 50th, and 75th percentiles for the thirty trend sites are -0.45, -0.28, and -0.17 meq/m^2/year. The two most extreme decreasing trend estimates are -0.90 and -0.82 meq/m^2/year at Zanesville and Parsons, respectively.

The calcium deposition temporal pattern for the 1982-1986 trend sites is similar in structure to the calcium concentration pattern (Figure A-51). Significant decreasing trend estimates for calcium deposition occur at thirty-four sites and significant increasing trend estimates occur at six sites. The six sites with significant increasing trends are all sites operated by the APIOS-D network in Ontario. Five sites with the most extreme decreasing trend estimates are in southern Ontario and are operated by the APIOS-C network. Sites in the United States with significant decreasing trend estimates are generally consistent regionally with trend estimates at nearby sites. The 25th, 50th, and 75th percentiles for the 137 trend estimates are -0.60, -0.35, and 0.00 meq/m^2/year. Of the twenty-two sites with increasing trend estimates, only four (none significant) are located in the United States: Whiteface, Chautauqua, and Big Moose, New York; and Parsons, West Virginia. In view of the general regional consistency in the trend estimates in the United States and the existence of 4 out of 98 sites in the United States with increasing trend estimates, the existence of an overall decreasing trend in calcium deposition during the 5-year period is postulated.

3.7 CHLORIDE, SODIUM, POTASSIUM AND MAGNESIUM PATTERNS

The temporal patterns for chloride, sodium, potassium, and magnesium concentration and deposition are discussed as a group. In part the decision to discuss them together is a result of their common presence in sea salt.

3.7.1 Annual Temporal Patterns 1979-1986

3.7.1.1 Chloride

Annual chloride concentrations in the thirty 1979-1986 trend sites have a general decreasing trend over the 8-year period (Figure A.52). The boxplots truncate all concentrations greater than 1.0 mg/L. Two sites, Lewes and Brookhaven heavily influenced by sea salt due to their location on the coast, have their concentrations truncated in each year. Lewes, located closest to the coast, has the maximum concentration each year, with the overall maximum of 2.70 mg/L occurring in 1980 (Table A.5). Annual chloride deposition has the same temporal pattern as concentration. The boxplots truncate the extreme deposition values for Lewes and Brookhaven. The maximum deposition is 38.45 kg/ha in 1983 at Lewes (Table A.6).

3.7.1.2 Sodium

The boxplots for annual sodium concentrations show the strongest decreasing trend during 1979-1986 of all the ions (Figure A.58). A step decrease appears to occur between 1980 and 1981. The decreasing pattern is substantially less for the years after 1980. In 1979 and 1980, the boxplots indicate more site-to-site variation excluding the sites influenced by sea salt. The increased variation, in addition to the higher concentrations suggests possible contamination during the sample collection process. Section 4.0 discusses this issue further. The maximum sodium concentration is 1.67 mg/L at Lewes (Table A.5). The boxplots for annual sodium deposition show the same large variation and high deposition pattern as concentration in 1979 and 1980. Subsequent years continue to show a decreasing trend for the lower deposition sites. The pattern for higher deposition sites is not as apparent. The maximum deposition is 24.22 kg/ha in 1983 at Lewes (Table A.6).

3.7.1.3 Potassium

Annual potassium concentrations tend to decrease during 1979-1986 (Figure A.64). However, the pattern is weaker than for sodium. The boxplots for potassium also provide an indication of a step decrease after 1980. The influence of sea salt is apparent in that Lewes has the highest concentration in 1982, 1983, 1985, and 1986. The boxplots for sodium deposition indicate an overall decreasing pattern superimposed on a variable temporal pattern. The maximum depositions for each year (Table A.6) are 1.03 kg/ha at Georgia Station in 1979, 1.54 kg/ha at Piedmont Station in 1980, 0.98 kg/ha at Penn State in 1981, 0.77 kg/ha at Bradford Forest in 1982, 0.99 kg/ha at Georgia Station in 1983, 1.23 kg/ha at Piedmont Station in 1984, 0.74 kg/ha at Lewes in 1985, and 0.81 kg/ha at Rockport in 1986.

3.7.1.4 Magnesium

Boxplots for annual magnesium concentration show a tendency for concentrations to decrease from 1979 to 1986 but the pattern is weaker than for chloride, sodium, or potassium (Figure A.70). Although sodium, chloride, and magnesium have sea salt as a source, the greater variation in 1979 and 1980 and the step change after 1980 that occurs in sodium, chloride, and potassium is not present for magnesium. If contamination is present, it does not appear to involve magnesium. The maximum concentration occurs at Lewes in all years, with the overall maximum being 0.18 mg/L in 1983. Magnesium deposition at the sites has a decreasing pattern, especially in 1985 and 1986. As expected the maximum deposition occurs at Lewes in all years. The overall maximum is 2.57 kg/ha in 1983.

3.7.2 Annual Temporal Patterns 1982-1986

The 1982-1986 trend sites have annual temporal patterns that are consistent with the temporal patterns present in the much small number of 1979-1986 trend sites. Figures A.53, A.59, A.65, and A.71 give boxplot displays for chloride, sodium, potassium, and magnesium, respectively. The 1982-1986 trend sites have more sites located near an ocean. Hence more sites have sea salt influenced concentrations and depositions resulting in distributions that are skewed and that have more extreme values. In order to display the general features of the distribution, the extreme values have been truncated in the boxplots.

3.7.3 Temporal Change Analysis

3.7.3.1 Chloride

The Kendall seasonal tau test for trend identifies seven sites with a significant ($p<0.05$) decreasing trend (Figure A.54). Only three sites, Rockport, Ithaca, and Lewes, have increasing trend estimates (none significant). Manitou, Marcell, Lamberton, and Mead in the West have consistent decreasing trend estimates of -0.39, -0.38, -0.35, and -0.21 μeq/L/year. The trend at Lewes is not consistent with Brookhaven, the other coastal site. The 25th, 50th, and 75th percentiles for the trend estimates at the sites are -0.25, -0.13, and 0.00 μeq/L/year. The trend estimates for the 1982-1986 trend sites are significantly decreasing at fourteen sites and significantly increasing at four sites (Figure A.55). Five sites that are not significant have decreasing trend estimates between -5.1 and -3.5 μeq/L/year and do not appear on the confidence interval plot. The sites are the coastal sites NACL, Massachusetts; Brookhaven, New York; Lewes, Delaware; Victoria, Texas; and American Samoa. With the exception of five sites in southern Ontario, the sites with significant decreasing trends do not cluster in any geographic region. The 25th, 50th, and 75th percentiles for the trend estimates are -0.59, -0.25, and 0.00 μeq/L/year. Although the trend estimates show substantial site-to-site variation and only a limited number are significantly decreasing estimates, the distribution of the estimates provides limited evidence that chloride concentration decreased during the 5-year period.

Chloride deposition trend estimates are significantly decreasing at six sites and significantly increasing at one site for the 1979-1986 trend sites (Figure A.56). Seven sites overall have increasing trend estimates, the most extreme being 0.55 meq/m^2/year at Brookhaven. The most extreme decreasing trend estimate sites are Piedmont Station, Clinton Station, Coweeta, Zanesville, and Hubbard Brook. Some regional consistency of the trend estimates is present in North Carolina. The 25th, 50th, and 75th percentiles for the deposition trend estimates are -0.22, 0.00, and 0.02 meq/m^2/year. The 1982-1986 trend sites have nine sites with significant decreasing and three sites with significant increasing trend estimates (Figure A-57). The sites do not cluster geographically. The sites with significant increasing trend estimates are Pickle Lake, Ontario; and Whiteface and Ithaca, New York. The most extreme decreasing trend estimates (<-2.0 meq/m^2/year) are sea salt influenced sites: NACL, Massachusetts; Brookhaven, New York; Lewes, Delaware; and Everglades National Park, Florida. Although the trend estimates are not significant, the estimates in the South and along the lower eastern coast do show a consistent pattern of decreasing deposition. The 25th, 50th, and 75th percentiles for the trend estimates are -0.34, -0.06, and 0.02 meq/m^2/year. The 10th percentile (-0.92 meq/m^2/year) is farther removed from the median of the distribution than the 90th percentile (0.25 meq/m^2/year) suggesting a possible decreasing pattern, especially with the cluster of consistent decreasing trend estimates in the South.

3.7.3.2 Sodium

Twenty-six of the thirty 1979-1986 trend sites have significant ($p<0.05$) sodium concentration trend estimates (Figure A.60). All trend estimates are for decreasing trends and their magnitudes are markedly consistent regionally. The 25th, 50th, and 75th percentiles for the trend estimates are -0.70, -0.43, and -0.30 μeq/L/year. The 1982-1986 trend sites have thirteen significant decreasing sodium concentration trend estimates and no significant increasing estimates (Figure A.61). Five sites with the most extreme decreasing trend estimates do not appear on the confidence interval plot: American Samoa; NACL, Massachusetts; Lewes, Delaware; Brookhaven, New York; and Victoria, Texas. Only Lewes and Brookhaven are significant. The 25th, 50th, and 75th percentiles for the estimates are -0.22, -0.11, and 0.02 μeq/L/year. In contrast to the definite decreasing trend for the 1979-1986 trend sites and 8-year period, only weak evidence of an overall decreasing trend exists for the 1982-1986 trend sites and 5-year period. The decrease for the 1979-1986 sites occurs mainly in the years prior to 1982. Hence it is likely that the analyses are consistent if both are restricted to the 1982-1986 period.

Sodium deposition trend estimates are significantly decreasing at twenty-two of the thirty 1979-1986 trend sites (Figure A.62). All trend estimates indicate decreasing trends and have regionally consistent magnitudes. The 25th, 50th, and 75th percentiles for the estimates are -0.69, -0.31, and

-0.18 meq/m^2/year. The 1982-1986 trend sites have sixteen significant decreasing sodium deposition trend estimates and no significant increasing estimates (Figure A.63). Three sites with the most extreme decreasing trend estimates do not appear on the confidence interval plot: NACL, Massachusetts; Brookhaven, New York; and Lewes, Delaware. American Samoa has the most extreme increasing trend estimate at 11.77 meq/m^2/year (not on plot). The 25th, 50th, and 75th percentiles for the estimates are -0.31, -0.11, and 0.01 meq/m^2/year. Most 1982-1986 trend sites in the South and along the lower eastern coast do show a consistent pattern of decreasing trends. Overall the sodium deposition trends during the 1979-1986 and 1982-1986 period are the same as for sodium concentration.

3.7.3.3 Potassium

Thirteen 1979-1986 trend sites have significant ($p<0.05$) decreasing trend estimates for potassium concentration (Figure A.66). The location of the sites encompasses the region from Minnesota to New York and from West Virginia to the Canadian border. Within the region, the trend estimates are consistent with nearby sites in trend direction but some inconsistency in magnitude is present. The most extreme decreasing trend estimate is -0.12 μeq/L/year at Brookhaven. Four sites have increasing trend estimates (none significant): Finley(A), Lewiston, and Coweeta, North Carolina and Delaware, Ohio. The 25th, 50th, and 75th percentiles for the estimates are -0.09, -0.03, and -0.01 μeq/L/year. Twenty 1982-1986 trend sites have significant decreasing potassium concentration trend estimates and three have significant increasing trend estimates (Figure A.67). Nine of the significant decreasing trend estimates are at sites in Ontario, including the five most extreme. Inspection of the map for regional consistency in the estimates reveals some regions, e.g., New England, that may have consistent estimates but the consistency tends to be weak. The 25th, 50th, and 75th percentiles for the trend estimates are -0.07, -0.03, and 0.00 μeq/L/year. Overall the potassium concentration trend estimates for the 1979-1986 and 1982-1986 trend periods are consistent. An overall estimate of the trend during the period is a decrease of 0.03 μeq/L/year (based on the median).

Potassium deposition at the 1979-1986 trend sites have significant ($p<0.05$) decreasing trend estimates at nine sites and no significantly increasing estimates (Figure A.68). Only two sites, Lewiston, North Carolina and Bradford Forest, Florida have increasing trend estimates. Regional consistency is present in most regions. However, consistency in the magnitude of the estimate does not always occur. The 25th, 50th, and 75th percentiles for the estimates are -0.05, -0.03, and -0.01 meq/m^2/year, respectively. The 1982-1986 trend sites have potassium deposition trend estimates that are significantly decreasing at twenty-four sites and significantly increasing at five sites (Figure A.69). The five sites with significant increasing trend estimates are Uvalda, Georgia; Charlottesville, Virginia; and Ithaca, Chautauqua, and Whiteface, New York. Note that the increasing trend at the sites is not consistent with nearby sites. The significant decreasing estimates occur throughout the United States and Ontario. The 25th, 50th, and 75th percentiles for the estimates are -0.06, -0.03, and 0.00 meq/m^2/year. Overall the potassium deposition trend estimates, just as for concentration, are consistent for the 1979-1986 and 1982-1986 periods. An overall estimate of the trend during the period is a decrease of 0.03 meq/m^2/year.

3.7.3.4 Magnesium

Eight 1979-1986 trend sites have significant ($p<0.05$) decreasing magnesium concentration trend estimates (Figure A-72). Only four sites have increasing, but not significant, trend estimates (Lewes, Delaware and Coweeta, Raleigh and Finley, North Carolina). The most extreme decreasing trend estimate is -0.32 μeq/L/year at Lamberton, Minnesota. The 25th, 50th, and 75th percentiles for the estimates are -0.17, -0.08, and -0.04 μeq/L/year, respectively. The 1982-1986 trend sites have eighteen sites with significant decreasing magnesium concentration trend estimates and one with a significant increasing trend estimate (Figure A.73). The significant increasing trend and the four most extreme significant decreasing trend estimates are at sites in Ontario. The remaining significant decreasing trend estimates occur at sites located throughout the United States. The 25th, 50th, and 75th percentiles for the estimates are -0.27, -0.11, and 0.01 μeq/L/year, respectively. The trend estimates from the 1979-1986 and the 1982-1986 periods agree in terms of the direction of the trend and are nearly consistent in terms of magnitude. An overall estimate of the magnesium concentration trend during the period is a decrease of approximately 0.08 to 0.11 μeq/L/year.

Eight 1979-1986 trend sites have a significant ($p<0.05$) decreasing magnesium deposition trend estimate (Figure A.74). Only three sites have increasing, but not significant, trend estimates (Lewes, Delaware and Huntington and Whiteface, New York). The most extreme decreasing trend estimate is -0.24 meq/m^2/year at Clinton Station, North Carolina. The 25th, 50th, and 75th percentiles for the estimates are -0.13, -0.06, and -0.02 meq/m^2/year, respectively. The 1982-1986 trend sites have twenty-nine sites with significant decreasing trend estimates and two sites with significant increasing trend estimates (Figure A.75). Whiteface, New York and North Easthope, Ontario are the two significant increasing trend estimate sites. The seven most extreme significant decreasing trends occur in southern Ontario. The other significant trend sites occur throughout the United States. The 25th, 50th, and 75th percentiles for the estimates are -0.31, -0.10, and 0.00 meq/m^2/year, respectively. The trend estimates and regional patterns are, in general, consistent for the 1979-1986 and 1982-1986 periods. An overall estimate of the magnesium deposition trend during the period is a decrease of approximately 0.06 to 0.10 meq/m^2/year.

4.0 MULTIVARIATE TEMPORAL PATTERNS

4.1 PRIOR TEMPORAL PATTERN STUDIES

Several recent studies have examined current temporal patterns. In a NRC (1986) study, data were obtained from 12 NADP/NTN sites from 1979 through 1982, and from several CANSAP and APN sites from 1980 to 1982. Schertz and Hirsch (1985) examined 19 sites from NADP/NTN over the period 1978 through 1983. Dana and Easter (1987) analyzed the precipitation chemistry data from 1978 through 1983 for the nine MAP3S/PCN sites. Barchet et al. (1987) undertook an analysis of the temporal patterns at sites in the NADP/NTN, UAPSP, MAP3S/PCN, APIOS, and CAPMoN (APN) networks for the period 1976 through 1984. Each of these studies used different data screening and statistical analysis techniques. A brief summary of the results and conclusions of each of these studies is presented below.

The data record available to the NRC (1986) study was too short to permit a quantitative analysis; only a qualitative description of temporal pattern tendencies is given. Despite the relatively sparse data available, their observations and conclusions have been largely supported by more recent and comprehensive studies. The NRC consistently noted the similar behavior of year-to-year changes in sulfate concentrate with that of ions not considered to have anthropogenic origins, e.g., calcium. Decreasing calcium concentrations were as prevalent as decreasing sulfate concentrations. For most of the 12 sites studied in the eastern United States, the concentration of most ions was higher in 1980 and 1981, and lower in 1979 and 1982. Canadian sites had similar multi-year patterns; however, year-to-year changes at some sites were different from neighboring sites.

Schertz and Hirsch (1985) used the Kendall seasonal test (Hirsch et al. 1982) to determine if a temporal change had occurred during the period covered in their study. They found decreases in ionic concentration were more common than increases for the 19 NADP/NTN sites. Of all the ions examined, sulfate had the largest decreases in concentration per year. Rates of change in concentration, significant at the 5% level, ranged from -0.20 to -0.48 mg/L/yr. When Schertz and Hirsch analyzed weekly ionic concentrations that were adjusted to account for variation in precipitation amount, even more sites showed significant decreases in concentration over the 1978 to 1983 period. All sites in Ohio, Pennsylvania, and West Virginia showed significant decreases in sulfate and nitrate concentration.

Dana and Easter (1987), analyzing the MAP3S/PCN data from 1976 through 1983, found that sulfate concentrations decreased about 2%/yr at most sites. However, these temporal changes were not statistically significant. Their analyses were based on a regression model applied to individual sample data that accounted for seasonal behavior through a phase-shifted sine wave and dependence on time assumed linear. The coefficient of the linear time term of the regression model is the parametric estimate of the rate of change in the period.

Barchet et al. (1987) present results for monthly changes in precipitation weighted mean concentration and deposition for seven ions. Their analyses are based on 48 sites that met a common data screening criteria applied to monthly summaries. The NADP/NTN weekly sampling network provided 28 sites; daily sampling networks provided 17 sites (eight from MAP3S/PCN, five from UAPSP, and four from CAPMoN/APN); three sites came from the APIOS monthly cumulative network. Most of the sites studied by Schertz and Hirsch and by Dana and Easter are among the sites selected. They used both the Kendall seasonal test and seasonal regression model in the statistical analysis. All estimates of change are based on the linear time coefficient of their regression model. Barchet et al. (1987) found that decreases in concentration dominate the temporal pattern during the period for most ions. The median annual change for sulfate concentration at all eastern sites (44 sites) was estimated as -0.11 mg/L/yr. Changes in concentration for the other ions tended to be downward but did not display as strong a pattern as for sulfate. Temporal changes in deposition over the period were not nearly as uniformly downward as for concentration. This appeared to be related to an increase in precipitation amount during the latter part of the period that counteracts the decrease in concentration at many sites.

4.2 MULTIVARIATE TEMPORAL PATTERN CONSISTENCY ACROSS ION SPECIES

Section 3 discusses the temporal patterns observed for each individual wet deposition ion species. The results suggest the sites have one or more ion species that either have similar increasing temporal patterns or have similar decreasing temporal patterns. Previous studies on wet deposition trends also support this view. The purpose of this section is to discuss the multivariate aspects of the temporal patterns observed in wet deposition for the 1979-1986 and 1982-1986 trend site subsets.

The approach will be first to discuss the multivariate temporal patterns for concentration and then to discuss deposition following the same approach. The discussion begins with an overview of the average annual concentrations for the ion species observed at the sites. This is followed by a comparison across the ion species of Sen's median slope trend estimate associated with the Kendall seasonal tau test. The trend estimate is then discussed in terms of percent change during the period. The reference for the percent change is the average 1982-1986 annual concentration for an ion species at a site. Individual year annual concentrations were also examined as a reference for percent change. The selection of the 5-year average from 1982-1986 provided a more stable reference base. The magnitude of the percent change depends on the reference base at those sites with increasing or decreasing temporal patterns for an ion species. For example, using 1986 rather than 1982 as the reference, results in greater percent changes for cations at most sites. The discussion for concentrations ends with a summary of the distributions and patterns of the z-values from the Kendall seasonal tau test statistic. All tables and figures appear at the end of Section 4.

4.2.1 Concentration Multivariate Temporal Patterns

The nine ion species routinely measured in wet deposition have differing annual precipitation-weighted average concentration levels at most sites. The nine ion species considered are hydrogen, sulfate, nitrate, ammonium, calcium, chloride, sodium, potassium, and magnesium. Boxplots in Figure 4.1 give distribution summaries of the nine ion species for 1979-1986 trend sites and 1982-1986 trend sites. The summaries use the 5-year average of an ion species' annual precipitation-weighted concentrations for the years 1982-1986. The 1979-1986 and 1982-1986 trend sites use the same 5-year average period. Hydrogen and sulfate ions have approximately the same relative concentration in precipitation. For the 5-year period examined and the sites in the two different trend subsets, the median concentrations are 40-45 μeq/L, approximately. Nitrate has median concentration levels of 20 μeq/L and ammonium median levels are 15 μeq/L, approximately. The median concentrations for the other ion species are below 10 μeq/L. Sodium and chloride are substantially higher than the median concentrations at several sites. These sites are coastal sites that have their precipitation chemistry influenced by sea salt.

4.2.1.1 Trend Estimates

The distributions of Sen's median slope trend estimates for each ion species are presented as boxplot displays in Figure 4.2 for the 1979-1986 and 1982-1986 trend site subsets. The most striking feature of the comparison is that the distributions are consistent, for all ion species except nitrate, ammonium, and chloride, in having over 50% of the estimates indicating a decreasing trend during the period. This is particularly the case for the 1979-1986 period. The trend estimates for the 1982-1986 trend sites are more variable than those for the 1979-1986 trend sites. The variation and magnitude of the estimates also appears to be a function of the site-to-site variation and magnitude of the ion species annual concentrations. For the 1979-1986 trend sites, the median trend estimate is negative, i.e., decreasing trend, for all ion species (Table 4.1). At least 75% of the sites have decreasing trend estimates for sulfate, calcium, chloride, sodium, potassium, and magnesium. As discussed in Section 3, sodium concentrations decrease at all 1979-1986 trend sites. For the 1982-1986 trend sites, the median trend estimate indicates decreasing trends for all ion species except nitrate. Sulfate concentration has the most extreme median trend estimate of all ion species, -0.91 and -0.62 μeq/L/year in the 1979-1986 and 1982-1986 trend sites, respectively.

In addition to providing Sen's median slope estimate, Kendall's seasonal tau provides a test of the significance for the temporal pattern. A significance level of $p=0.05$ is used as one way to summarize the test results. For each ion species and the two trend subsets, Table 4.3 summarizes the number and percent of the trend estimates that are significant, either decreasing or increasing. In the 1979-1986 trend sites, only hydrogen has a site with a significant increasing trend (Bradford Forest, Florida). In contrast, all ion species have sites with significant decreasing trends, 89 sites for all ions. This represents 33% of the 270 tests calculated for the 30 sites and 9 ion species. Sodium, calcium, and potassium have the greatest number of sites with significant decreasing trend estimates, with 25 (83%), 17 (57%), and 13 (43%), respectively. Sulfate has 8 (27%) sites with significant decreasing trend estimates. In the 1982-1986 trend sites, all ion species except sodium have one or more sites with significant increasing trend estimates, 35 sites for all ions. On average across all ions, this represents 3% of the 1233 tests calculated for the 137 sites and 9 ion species. Ammonium and calcium have the most sites with significant increasing trend estimates, 11 and 6, respectively. Significant decreasing trend estimates occur for all ion species in 126 (10%) cases. Calcium, potassium, magnesium, and sulfate have the most sites with significant decreasing trends, with 20 (15%), 20 (15%), 18 (13%), and 16 (12%), respectively.

Scatterplot matrices in Figure 4.3 provide one way to investigate the multivariate characteristics of ion species temporal patterns at trend sites. The scatterplot matrix is simply an organized collection of the scatterplots of all possible pairs of ion species and annual precipitation amount (TP). The purpose of the matrix is to examine the plots for relationships. The scatterplot of the concentration trend estimates for the 1979-1986 trend sites (Figure 4.3a) shows the presence of several sites with extreme trend estimates for ammonium, calcium, and chloride. The two extreme estimates for ammonium and calcium are at the same two sites: Lamberton, Minnesota and Mead, Nebraska. These sites also are the most extreme decreasing trend estimates for sulfate, nitrate, and magnesium (see NH4 column of plots). The extreme chloride trend estimate is not extreme for any other ion species. A strong positive association exists between sulfate and nitrate concentration trend estimates. No ion species, with the possible exception of magnesium exhibits an association with precipitation amount. Sodium, chloride, and magnesium trend estimates have positive associations, with the magnesium association being the weakest. The extreme chloride estimate negatively affects the ability to detect the presence of an association visually. Sulfate (see SO4 column) trend estimates also have weak positive associations with chloride, sodium, and magnesium. These positive associations are consistent with sea salt being a common source for these ions. However, the two most extreme decreasing sulfate trend estimates are from Lamberton and Mead.

The scatterplot matrix of the ion species concentration trend estimates for the 1982-1986 trend sites also shows multivariate extreme trend estimates (Figure 4.3b). Five sites, all with substantial sea salt influence, do not appear in the scatterplot matrix in order to enhance the visual display. The sites are American Samoa; NACL, Massachusetts; Brookhaven, New York; Lewes, Delaware, and Victoria, Texas. Sodium and chloride have a common site with an extreme increasing trend estimate, Everglades National Park. Magnesium has four sites (Palmerston, Colchester, Merlin and Huron Park, Ontario) with extreme decreasing trend estimates that are also extreme for calcium. The associations observed between sulfate and nitrate and between sodium and chloride in the 1979-1986 trend sites also exist for the 1982-1986 trend sites. Magnesium and calcium also have a positive association.

Table 4.4 gives the ranks by individual ion of the annual concentration trend estimates for the 1979-1986 trend sites.

4.2.1.2 Percent Change

The concentration trend estimates may also be expressed in terms of percent change during the period. The reference for the percent change is the average 1982-1986 annual concentration for an ion species at a site. Figure 4.4, Table 4.5, Table 4.6, and Figure 4.5 summarize the concentration percent change trend results. Sodium concentration for the 1979-1986 trend sites has the most extreme negative percent change. Potassium, calcium, and magnesium have the next most extreme distributions. The median percent changes at the 1979-1986 trend sites for sodium, potassium, calcium, and magnesium are -9.9, -4.6, -4.6, and -2.9 percent per year, respectively. Nitrate, ammonium, and, to a lesser extent, hydrogen, have distributions centered on zero percent change. Only sulfate and sodium have 90% or more of the sites with negative percent change. For the 1982-1986 trend sites, more consistency across ion species is present for the percent change distributions (Figure 4.4). The distributions are symmetric and have approximately the same variation (interquartile range) for all ion species. The only distinction among the ion species is the location of the center of the distribution. The increasing (most negative percent change) rank order of the medians for the distributions is potassium, magnesium, calcium, sodium, hydrogen, sulfate, chloride, and nitrate. Only nitrate has a median (0.7%) representing a positive percent change. Approximately 75% of the 1982-1986 trend sites have negative percent changes for calcium, potassium, magnesium, and sodium.

Scatterplot matrices presented in Figure 4.5 show fewer sites with extreme percent changes than the corresponding plots for the trend estimates. Sulfate and nitrate, sodium and chloride, and magnesium and calcium have a positive association for percent change, as before.

4.2.1.3 z-values

The Kendall seasonal tau test constructs a test statistic that is approximately normally distributed. The term z-value is used here for the test statistic value. Under the assumption that the sites from the 1979-1986 or the 1982-1986 trend sites represent a random sample from a common population for each ion species, the observed z-values have a distribution that is approximately normal. If the population for an ion species has a non-zero population mean trend, then the distribution of z-values will continue to be approximately normal but not be centered at zero. The assumption of a common population for the true trend associated with an ion species has several weak points. First, the sites are not a random sample from a population of sites. Second, spatial correlation among the sites may be present. Third, a common true mean trend in North America for an ion species seems unlikely. For example, in Section 3, pockets of regional consistency in trend estimates were identified. In view of these, and possibly other, limitations, the assumption appears inappropriate. However, the observed z-value distributions for both the 1979-1986 and 1982-1986 trend sites are surprisingly symmetric. No formal comparison to a normal distribution has been completed. For ion species that have a non-zero median trend estimate, e.g., sodium, the z-value distribution appears only to be shifted. If a decreasing trend was present in only a single region of the United States, the z-value distribution would reflect this by the present of a larger than expected (compared to normal distribution) lower tail of the distribution. A formal analysis of this has not been performed. If a general decreasing trend exists throughout the region covered by one of the two trend subsets, then we would expect the entire observed z-value distribution to be shifted. The latter appears more likely given the observed distributions for the 1979-1986 and the 1982-1986 trend sites.

Since a site has z-values associated with the Kendall seasonal tau test of each ion species, the z-values also provide information on the multivariate characteristics of the ion species trends during the period. In Figure 4.7, scatterplot matrices for the 1979-1986 and 1982-1986 trend sites show one aspect of the multivariate characteristics of the z-values across species. The most notable feature of the matrices is the lack of any clusters of extreme z-values for any ion or ion pair. The conclusion drawn is that no strong regional scale pattern of trends exist during the period. The scatterplot matrix provides no information on the presence of a general overall trend for all sites. Refer to Figures 4.2, 4.4 or 4.6 to assess overall trends for individual ion species. The z-values show positive associations for sulfate and nitrate, chloride and sodium, and magnesium and calcium. Precipitation amount does not appear to have any definite association with any ion species.

4.2.2 Deposition Multivariate Temporal Patterns

Figure 4.8 shows the average annual deposition for the nine ion species at the sites in the 1979-1986 and the 1982-1986 trend sites. The average annual deposition is obtained from the average of the annual deposition for the years 1982-1986 for both sets of trend sites. Sulfate and hydrogen ions have the highest deposition of the ions at most sites. The median sulfate deposition at the sites is approximately 45-50 meq/m^2/year for both sets of trend sites. The median hydrogen deposition is slightly less at 40-45 meq/m^2/year. The median depositions for nitrate and ammonium are approximately 20 and 15 meq/m^2/year. The other ion species have median depositions less than 10 meq/m^2/year. The inclusion of more sites located in the west and south in the 1982-1986 trend sites is reflected by the lower 25th percentiles for hydrogen and sulfate ions and by the additional high extreme deposition sites for chloride and sodium.

4.2.2.1 Trend Estimates

Boxplots of Sen's median slope trend estimates for deposition presented in Figure 4.9 show that approximately 75% or more of the sites in the 1979-1986 and 1982-1986 trend sites have decreasing trend estimates for sulfate, calcium, chloride, sodium, potassium, and magnesium. Hydrogen, nitrate, and ammonium have approximately an equal number of decreasing and increasing trend estimates. The variation of the trend estimates appears to be related to the relative deposition magnitude of the ion species, i.e., the greatest variation is associated with hydrogen and sulfate. The boxplots suggest the trend estimate distributions for some ion species may be negatively skewed. Tables 4.7 and 4.8 provide selected percentiles of the deposition trend estimates for each ion species. The common shift in the distributions for multiple ion species must be considered when interpreting the deposition trend estimates for any of the species. For example, why are the deposition trend estimates for sulfate, calcium, chloride, sodium, potassium, and magnesium all decreasing at most sites?

Table 4.9 summarizes the number of sites with significantly decreasing or increasing deposition trend estimates. A trend estimate is significant if the Kendall seasonal tau test has a p-value less than 0.05. In the 1979-1986 trend sites, only hydrogen, nitrate, and ammonium have sites (5 total) with a significant increasing deposition trend estimate. Whiteface, New York has increasing trend estimates for nitrate and ammonium; Bradford Forest, Florida for hydrogen ion; Brookhaven, New York for hydrogen ion; and Scranton, Pennsylvania for ammonium (may be result of limited site operation in 1981). In contrast, 70 significant decreasing trend estimates are present for all ions. This number represents 26% of the 270 tests calculated for the 30 sites and 9 ion species. Sodium has significant decreasing trend estimates at 22 sites. Sulfate, calcium, chloride, potassium, and magnesium have 6-9 sites with significant decreasing trend estimates. For 1982-1986 trend sites, 28 (2%) significant increasing and 157 (13%) decreasing trend estimates occur for the 1233 tests calculated for the 137 sites. All ion species except sodium have one or more sites with a significant increasing trend estimate. If the tests are statistically independent and the normal distribution assumption is appropriate, the 2% significant increases are what would be expected. Calcium, magnesium, potassium, and ammonium have the most sites with significant decreasing trends, with 34 (25%), 29 (21%), 24 (18%), and 22 (16%), respectively. Precipitation amount has significant decreasing estimates at eight sites and significant increasing estimates at four sites.

The scatterplot matrix in Figure 4.10a of the deposition trend estimates for the 1979-1986 trend sites shows a two-dimensional view of the multivariate characteristics of the trend estimates. Precipitation amount appears to have a positive association with hydrogen, sulfate, and nitrate; and to a lesser extent with chloride. A positive association also exists among sulfate, nitrate, and chloride; and between sodium and chloride. Any interpretation of trends for one of these ion species must also account for these positive associations. In Figure 4.10b, a scatterplot matrix for the 1982-1986 trend sites shows associations among the ion species more clearly (partly due to the increased number of sites). The previous positive associations of deposition trends with precipitation amount is not as clear however. This may be due to the increased number of western sites where an association might be expected to differ from an association that exists at eastern sites. Positive associations exist between sulfate and nitrate, sulfate and ammonium, sulfate and hydrogen, nitrate and ammonium, calcium and magnesium, and chloride and sodium.

Table 4.10 gives the ranks by individual ion of the deposition trend estimates for the 1979-1986 trend sites.

4.2.2.2 Percent Change

The deposition trend estimates may also be expressed in terms of percent change during the period studied. The reference for the percent change is the average 1982-1986 annual deposition for an ion species at a site. Boxplots of the distributions for percent change in ion deposition show that 75% or more of the 1979-1986 and 1982-1986 trend sites have a negative percent change in deposition for sulfate, calcium, chloride, sodium, potassium, and magnesium. Only hydrogen, nitrate, and ammonium have less than the 75% of the sites with a negative percent change. As for percent change in concentration, the distributions are surprisingly symmetric. The median percent changes (ordered by magnitude of change) of each ion species in the 1979-1986 trend sites are sodium (-8.8%), calcium (-4.0%), potassium (-3.6%), magnesium (-2.1%), sulfate (-1.4%), hydrogen (-0.4%), and nitrate, ammonium, and chloride (0%). For the 1982-1986 trend sites, the median percent changes are calcium(-5.2%), magnesium (-5.2%), potassium (-4.2%), sodium (-3.8%), ammonium (-2.5%), hydrogen (-1.9%), sulfate (-1.4%), chloride (-1.1%), and nitrate (0%). Tables 4.11 and 4.12 give other percentiles of the distributions.

In Figure 4.12, scatterplot matrices for percent change in deposition show several extreme multivariate outliers and pairwise positive associations for some ion species. Davis, California is a multivariate outlier for sulfate, nitrate, chloride, sodium, potassium, and magnesium in 1982-1986 trend site analysis. Zanesville, Ohio is an outlier for magnesium and potassium in 1979-1986 trend sites. Positive associations in percent change in deposition exist between sulfate and precipitation amount, sulfate and hydrogen, sulfate and nitrate, sulfate and ammonium, and nitrate and ammonium. Other associations are possible to interpret but they are weaker.

4.2.2.3 z-Values

The Kendall seasonal tau test statistic provides another way to examine the presence or absence of trend patterns in deposition. Section 4.2.1 gives further discussion on its use. The z-value distributions from the KST test for trend are given in Figure 4.13 for the ion species. The most notable feature of the distributions are that they are symmetric and resemble normal distributions and that the major difference across ion species is the difference in the location of the center of the distributions. In the 1979-1986 trend sites, the distributions for sodium, calcium, potassium, magnesium, sulfate, and chloride have medians that are negative, indicating an overall decreasing deposition trend estimate for the sites. The same pattern exists for the 1982-1986 trend sites but the shift toward negative z-values is not as great. However, a shift of the same magnitude occurs for the cations magnesium, potassium, calcium, and sodium. This suggests any interpretation in deposition trend must account for this multivariate shift. Figure 4.14 gives scatterplot matrices of the z-values for all ion species. Strong positive associations are seen for sulfate and nitrate, sodium and chloride, and calcium and magnesium. Other associations are discernable.

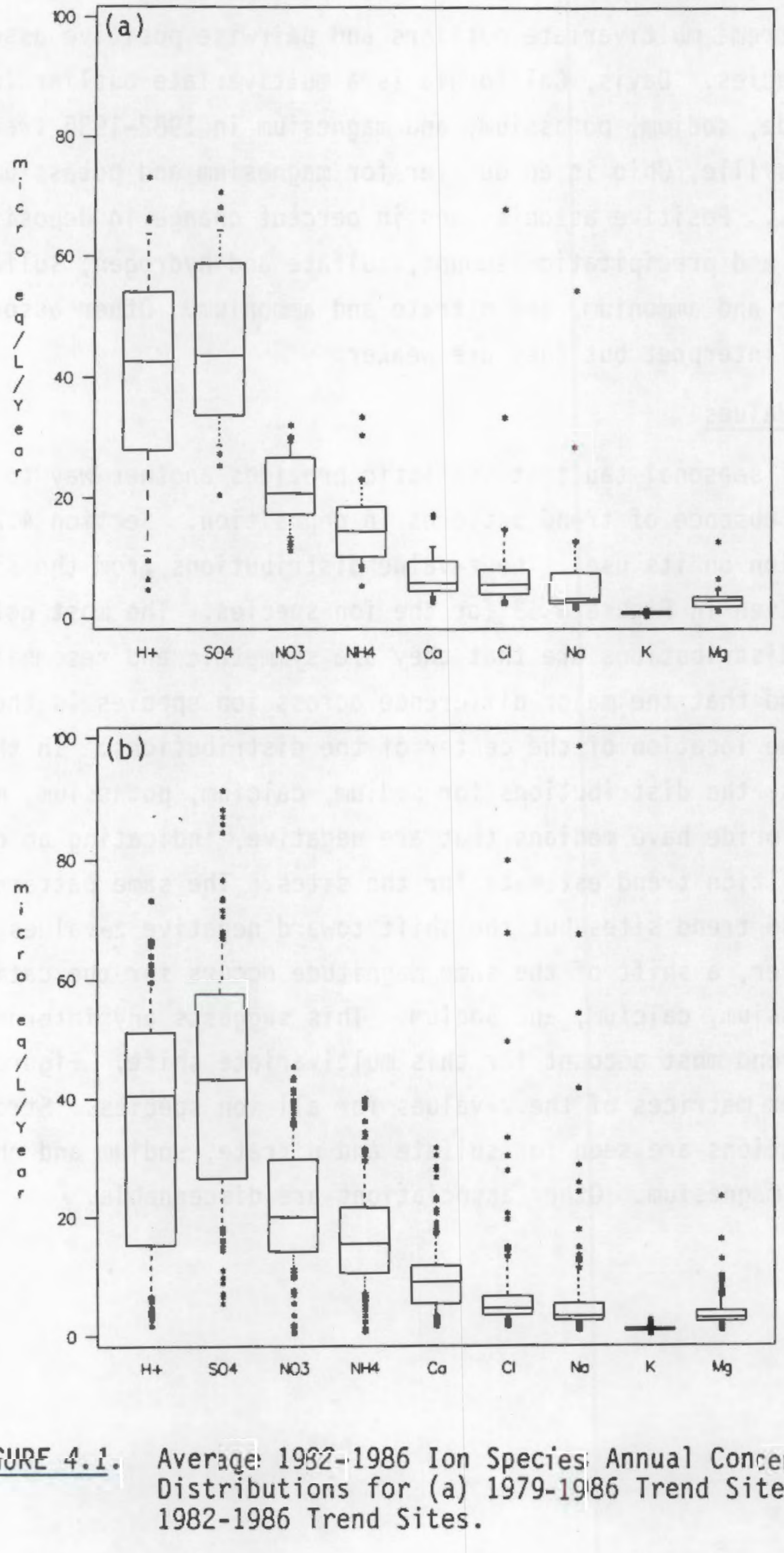
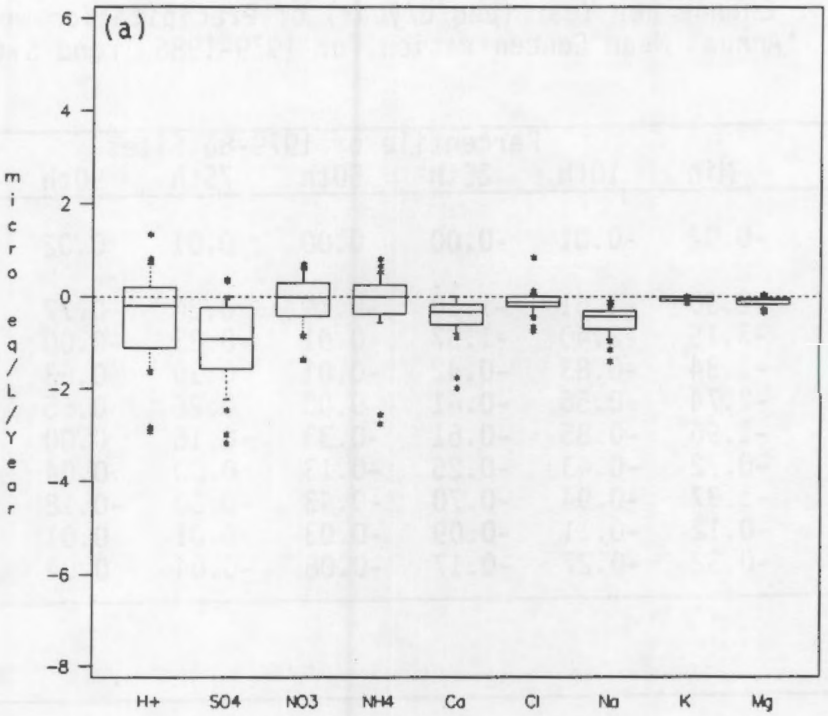


FIGURE 4.1. Average 1982-1986 Ion Species Annual Concentration Distributions for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.



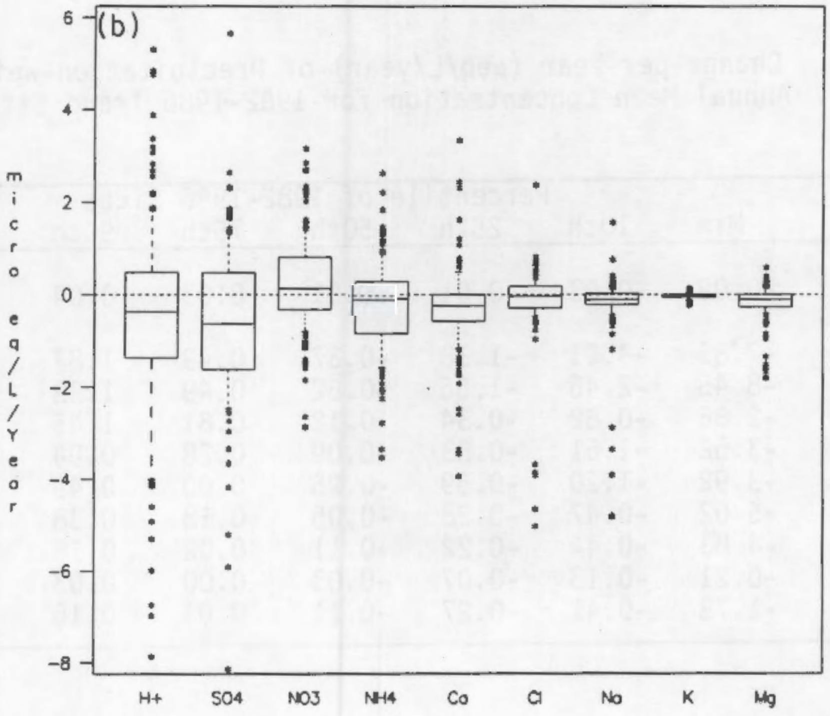


FIGURE 4.2. Ion Species Concentration Trend Estimate Distributions for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

TABLE 4.1. Change per Year (μeq/L/year) of Precipitation-weighted Annual Mean Concentration for 1979-1986 Trend Sites.

	Percentile of 1979-86 Sites						
Ion	Min	10th	25th	50th	75th	90th	Max
pH[a]	-0.02	-0.01	-0.00	0.00	0.01	0.02	0.02
Hydrogen	-2.90	-1.61	-1.10	-0.19	0.20	0.77	1.36
Sulfate	-3.15	-2.40	-1.57	-0.91	-0.23	-0.00	0.40
Nitrate	-1.34	-0.83	-0.42	-0.01	0.30	0.63	0.73
Ammonium	-2.74	-0.56	-0.41	0.00	0.26	0.55	0.83
Calcium	-1.96	-0.85	-0.61	-0.33	-0.16	0.00	0.00
Chloride	-0.72	-0.43	-0.25	-0.13	0.00	0.04	0.87
Sodium	-1.37	-0.94	-0.70	-0.43	-0.30	-0.18	-0.09
Potassium	-0.12	-0.11	-0.09	-0.03	-0.01	0.01	0.02
Magnesium	-0.32	-0.27	-0.17	-0.08	-0.04	0.04	0.06

[a] pH units

TABLE 4.2. Change per Year (μeq/L/year) of Precipitation-weighted Annual Mean Concentration for 1982-1986 Trend Sites.

	Percentile of 1982-1986 Sites						
Ion	Min	10th	25th	50th	75th	90th	Max
pH[a]	-0.09	-0.03	-0.01	0.01	0.03	0.04	0.08
Hydrogen	-7.85	-4.01	-1.38	-0.37	0.49	1.87	5.32
Sulfate	-8.45	-2.46	-1.66	-0.62	0.49	1.38	5.67
Nitrate	-2.86	-0.82	-0.34	0.12	0.81	1.45	3.17
Ammonium	-3.52	-1.61	-0.83	-0.09	0.28	0.94	2.64
Calcium	-3.92	-1.20	-0.59	-0.25	0.00	0.49	3.35
Chloride	-5.02	-0.47	-0.28	-0.05	0.18	0.38	2.39
Sodium	-4.83	-0.44	-0.22	-0.11	0.02	0.16	1.59
Potassium	-0.21	-0.13	-0.07	-0.03	0.00	0.03	0.16
Magnesium	-1.78	-0.41	-0.27	-0.11	0.01	0.16	0.60

[a] pH units

TABLE 4.3. Number and Percent of Significantly ($p<0.05$) Decreasing and Increasing Trend Estimates by Ion Species for Concentrations at 1979-1986 Trend Sites and 1982-1986 Trend Sites.

	1979-1986 Trend Sites Significance					1982-1986 Trend Sites Significance				
	Decrease		None	Increase		Decrease		None	Increase	
Ion Species	%	N	N	N	%	%	N	N	N	%
Precipitation	0	0	28	2	7	6	8	125	4	3
pH	3	1	25	4	13	3	4	124	9	7
Hydrogen	13	4	25	1	3	7	9	124	4	3
Sulfate	27	8	22	0	0	12	16	119	2	1
Nitrate	13	4	26	0	0	2	3	123	11	8
Ammonium	7	2	28	0	0	9	13	120	4	3
Calcium	57	17	13	0	0	15	20	111	6	4
Chloride	27	8	22	0	0	10	14	119	4	3
Sodium	83	25	5	0	0	9	13	124	0	0
Potassium	43	13	17	0	0	15	20	114	3	2
Magnesium	27	8	22	0	0	13	18	118	1	1
All Species	33	89	180	1	0	10	126	1072	35	3

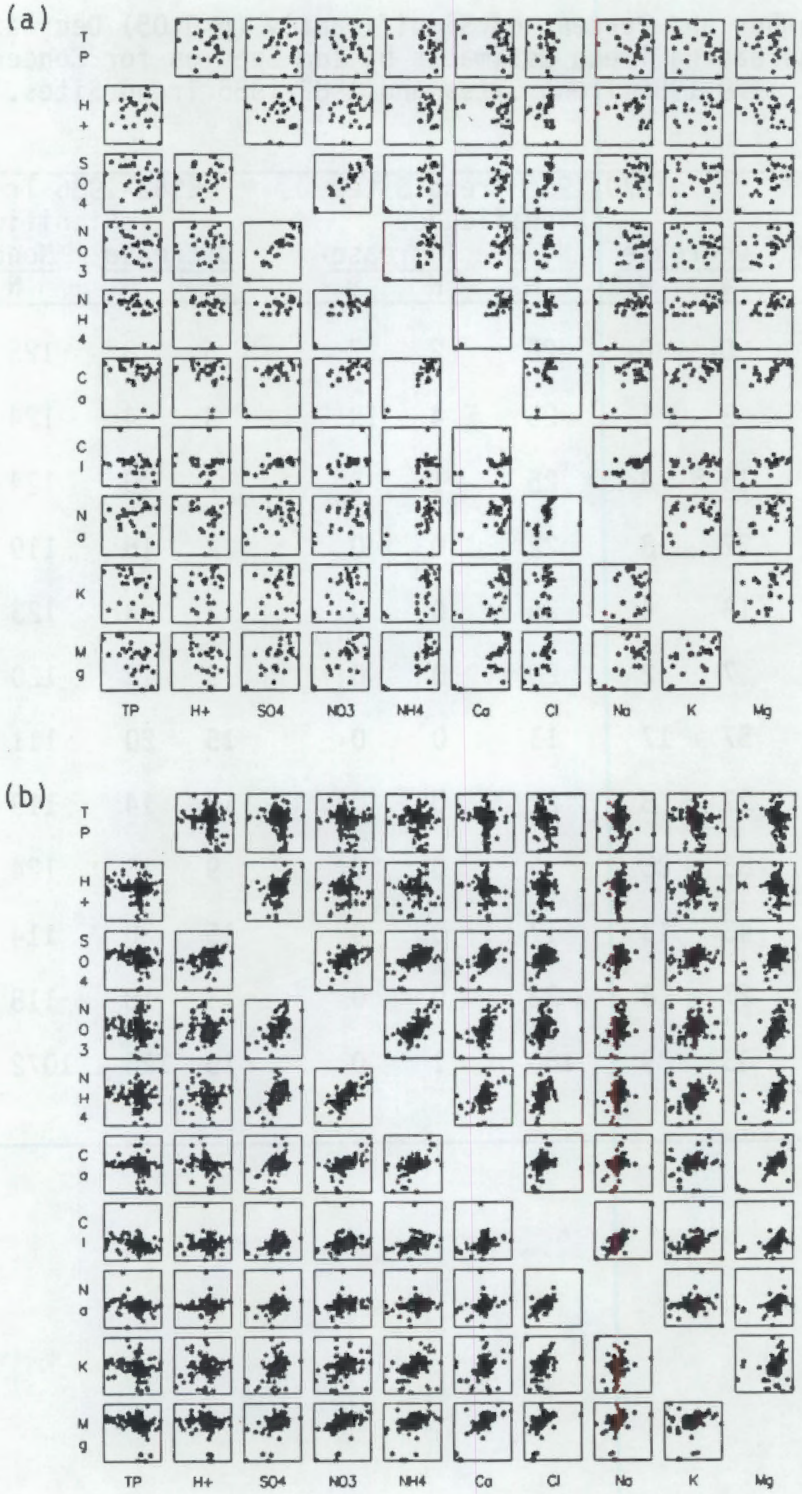


FIGURE 4.3. Scatterplot Matrix of Ion Species Concentration Trend Estimates for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

TABLE 4.4. Ranks by Individual Ion of Annual Concentration Temporal Change for 1979-1986 Trend Sites (Ordered by Average Rank).

	Individual Ion Rank										Ave
Sitename	pH	H+	SO4	NO3	CL	NH4	NA	K	CA	MG	Rank
Mead	20½	17	2	2	4	1	2	4	1	2	3.9
Lamberton	2	19	1	1	5	2	3	2	2	1	4.0
Manitou	4	23	5	6	7	4½	7	9½	3	4	7.7
Oxford	20½	9	4	4	11½	7	24	5½	10	9	9.3
Fort Wayne	20½	7	13	8	10	4½	21	5½	6½	9	9.4
Hubbard Brook	17	11	6	5	6	14	5	20	13	7	9.7
Marcell	20½	16	8	3	8	4½	18	12	10	15	10.5
Brookhaven	16	15	20	25	2	9	8	1	12	5	10.8
Clinton Station	24½	12	11	17	1	23	1	17	21	3	11.8
Piedmont Station	24½	10	3	9	3	19	6	25	18	15	12.0
Scranton	30	1	15	12	14	20	22½	13	15½	15	14.2
Caldwell	9	25	10	7	20	21	17	17	5	9	14.6
Bradford Forest	1	29	14	18	24½	8	4	21½	19½	6	16.1
Illinois	5	30	21	16	21	4½	27	8	6½	11	16.1
Wooster	15	13	19	13	9	29	15	21½	10	18	16.4
Ithaca	28½	2	7	10	29	14	30	19	19½	19	16.6
Penn State	12½	20	29	28	24½	14	16	3	4	12	16.7
Parsons	7	28	12	19	17	18	25	14	8	15	17.3
Huntington	12½	21	16	15	11½	11	22½	23½	15½	21	17.4
Zanesville	20½	6	28	29	14	27	20	7	15½	15	17.9
Delaware	12½	18	9	11	14	30	14	28	15½	23	18.1
Whiteface	26	8	17	20	24½	14	29	11	25½	25	19.3
Coweeta	8	22	18	21	18	14	11	27	25½	27	20.3
Lewes	12½	14	30	26	30	17	9	9½	22	30	20.8
Georgia Station	3	27	24	23	19	10	13	23½	28½	20	20.9
Raleigh	28½	3	23	24	24½	25	19	17	28½	28	21.3
Virginia	27	4	27	27	24½	22	26	15	23½	23	21.3
Lewiston	10	24	26	22	16	26	10	29	23½	23	22.2
Rockport	20½	5	25	14	28	28	28	26	28½	26	23.2
Finley (A)	6	26	22	30	24½	24	12	30	28½	29	25.1

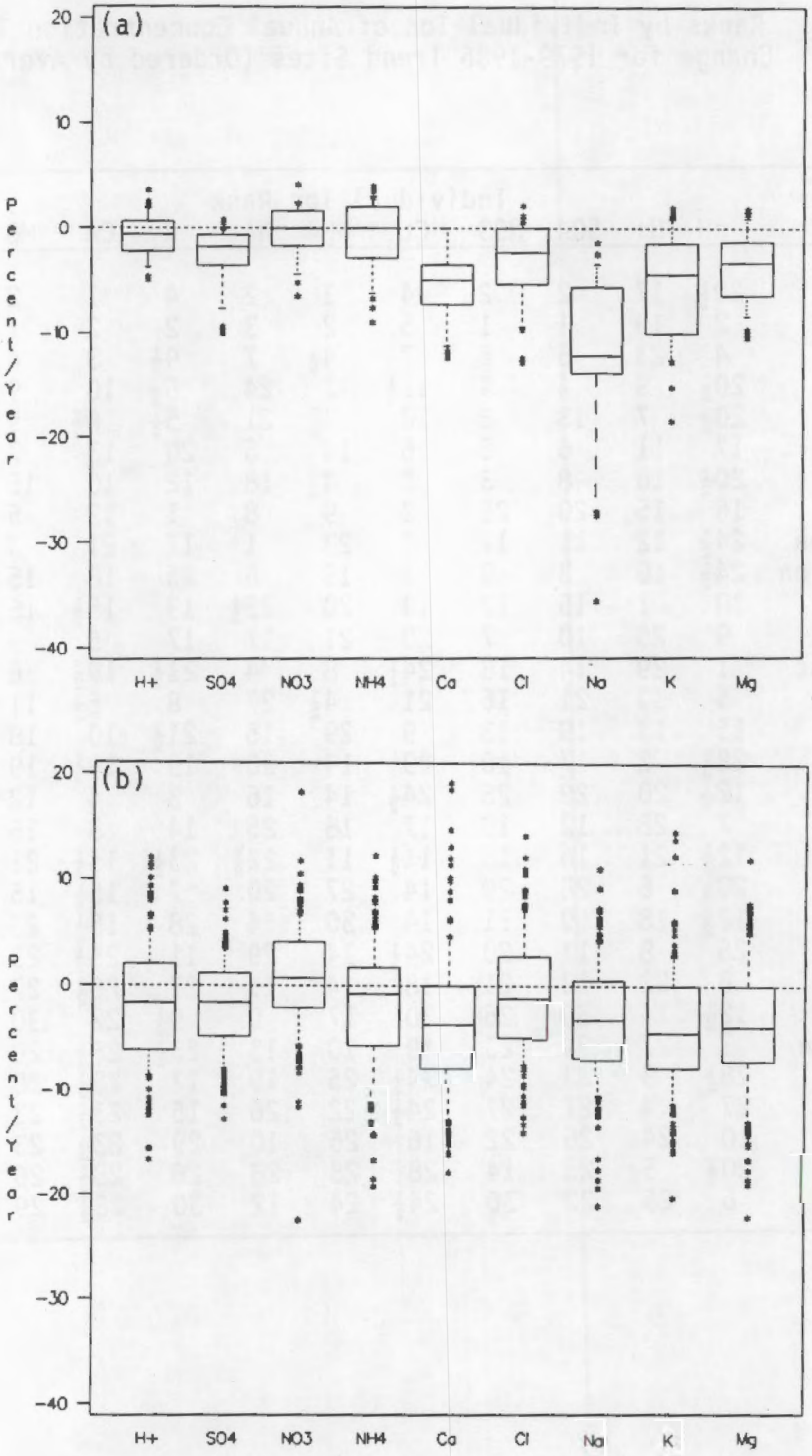


FIGURE 4.4. Distribution of Ion Species Concentration Trend Estimates as Percent of Average 1982-1986 Annual Concentration for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

TABLE 4.5. Percent Change per Year of Precipitation-weighted Annual Mean Concentration for 1979-1986 Trend Sites.

	Percentile of 1979-86 Sites						
Ion	Min	10th	25th	50th	75th	90th	Max
pH	-0.5	-0.3	-0.1	0.1	0.2	0.4	0.5
Hydrogen	-4.4	-3.7	-2.1	-0.7	0.8	2.3	4.1
Sulfate	-10.9	-9.1	-3.2	-1.8	-0.6	-0.0	0.9
Nitrate	-7.4	-5.2	-1.7	-0.1	1.9	2.6	4.5
Ammonium	-9.3	-6.6	-2.9	0.0	2.3	3.1	4.7
Calcium	-14.6	-9.4	-7.5	-4.6	-3.3	0.0	0.0
Chloride	-12.5	-11.1	-5.6	-2.0	0.0	0.6	2.6
Sodium	-33.2	-26.9	-13.4	-9.9	-5.9	-2.1	-0.7
Potassium	-15.4	-14.6	-10.0	-4.6	-1.1	1.2	3.0
Magnesium	-12.0	-8.6	-5.6	-2.9	-1.5	1.2	2.5

TABLE 4.6. Percent Change per Year of Precipitation-weighted Annual Mean Concentration for 1982-1986 Trend Sites.

	Percentile of 1982-86 Sites						
Ion	Min	10th	25th	50th	75th	90th	Max
pH	-1.7	0.6	-0.2	0.2	0.6	1.0	1.8
Hydrogen	-17.7	-7.5	-5.0	-1.7	2.1	6.2	25.5
Sulfate	-9.9	-7.5	-4.3	-1.7	1.2	3.8	21.0
Nitrate	-16.7	-5.3	-2.1	0.7	4.1	8.0	38.1
Ammonium	-16.7	-9.9	-5.5	-0.7	1.9	6.0	16.7
Calcium	-20.0	-10.6	-7.0	-3.6	0.0	5.6	28.5
Chloride	-16.7	-7.4	-4.5	-1.3	3.1	9.1	37.5
Sodium	-17.0	-10.5	-6.2	-2.7	0.7	5.1	20.7
Potassium	-30.0	-9.4	-7.2	-4.5	0.0	3.5	25.0
Magnesium	-22.6	-11.8	-7.0	-3.6	0.2	5.9	15.6

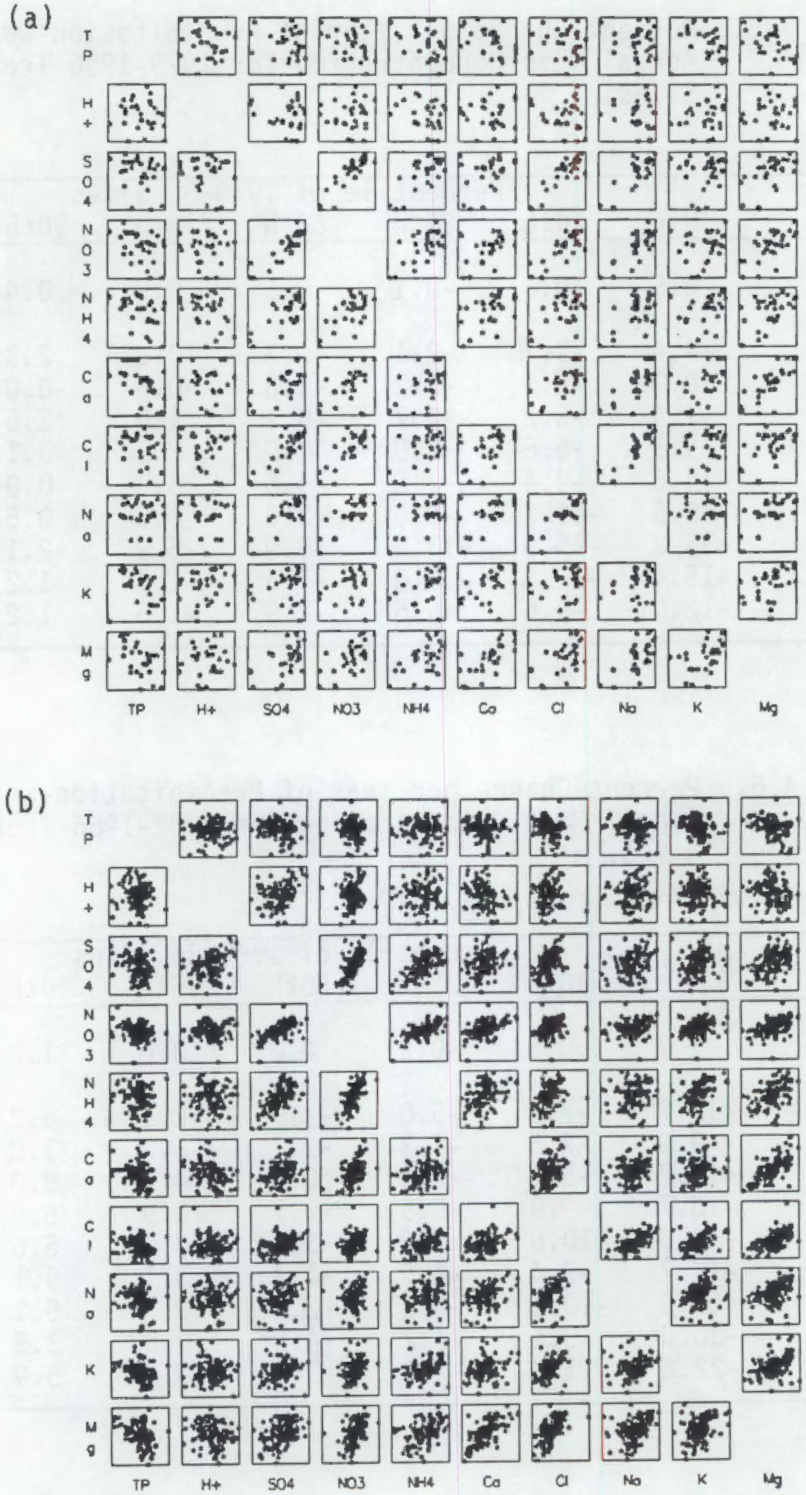


FIGURE 4.5. Scatterplot Matrix of Ion Species Concentration Trend Estimates as Percent of Average 1982-1986 Annual Concentration for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

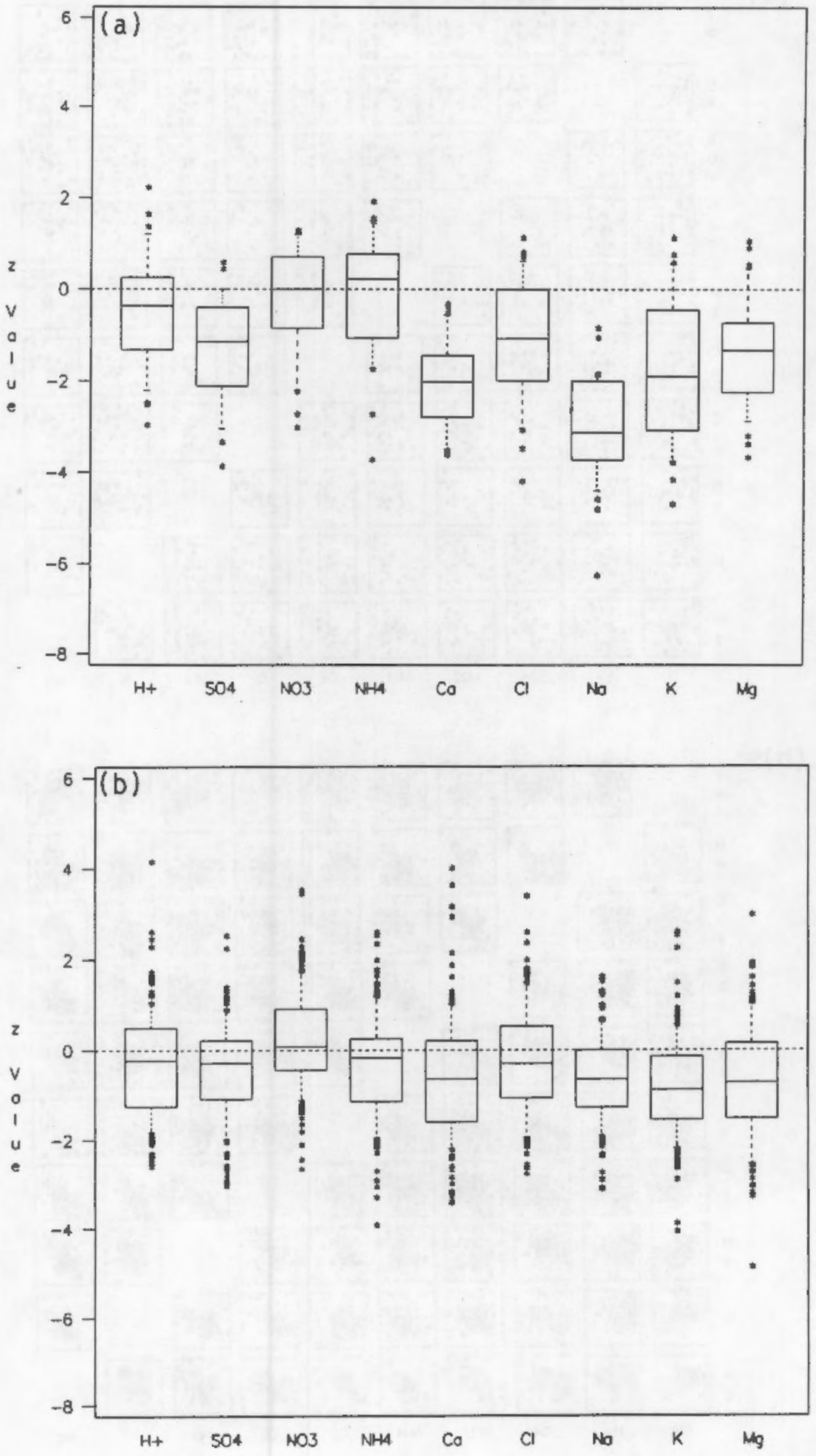


FIGURE 4.6. Distribution of Ion Species of Kendall Seasonal Tau Test Z-values for (a) 1979-1986 Trend Sites' Concentration and (b) 1982-1986 Trend Sites' Concentration.

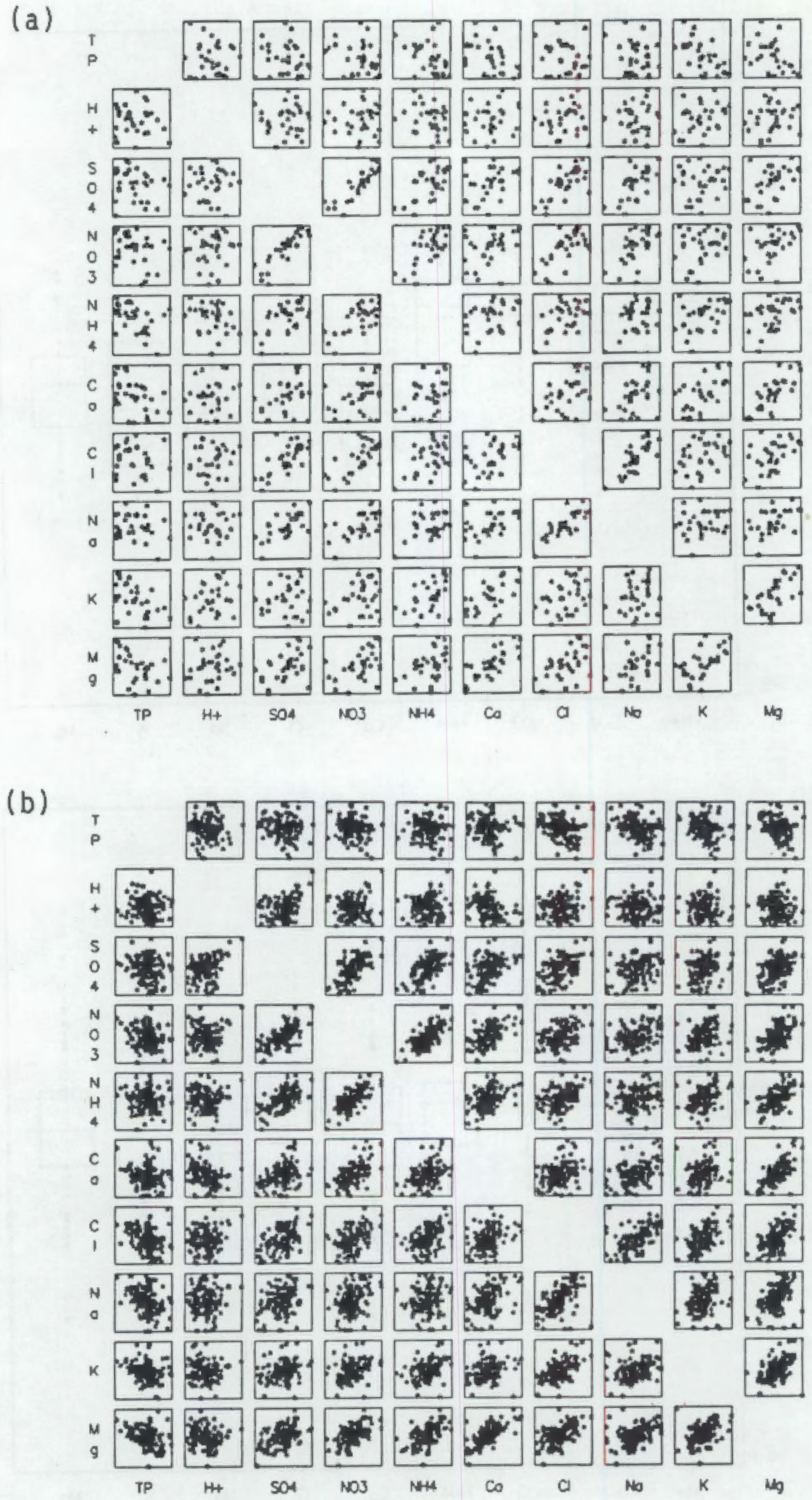


FIGURE 4.7. Scatterplot Matrix of Kendall Seasonal Tau Test Z-values for (a) 1979-1986 Trend Sites' Concentration and (b) 1982-1986 Trend Sites' Concentration.

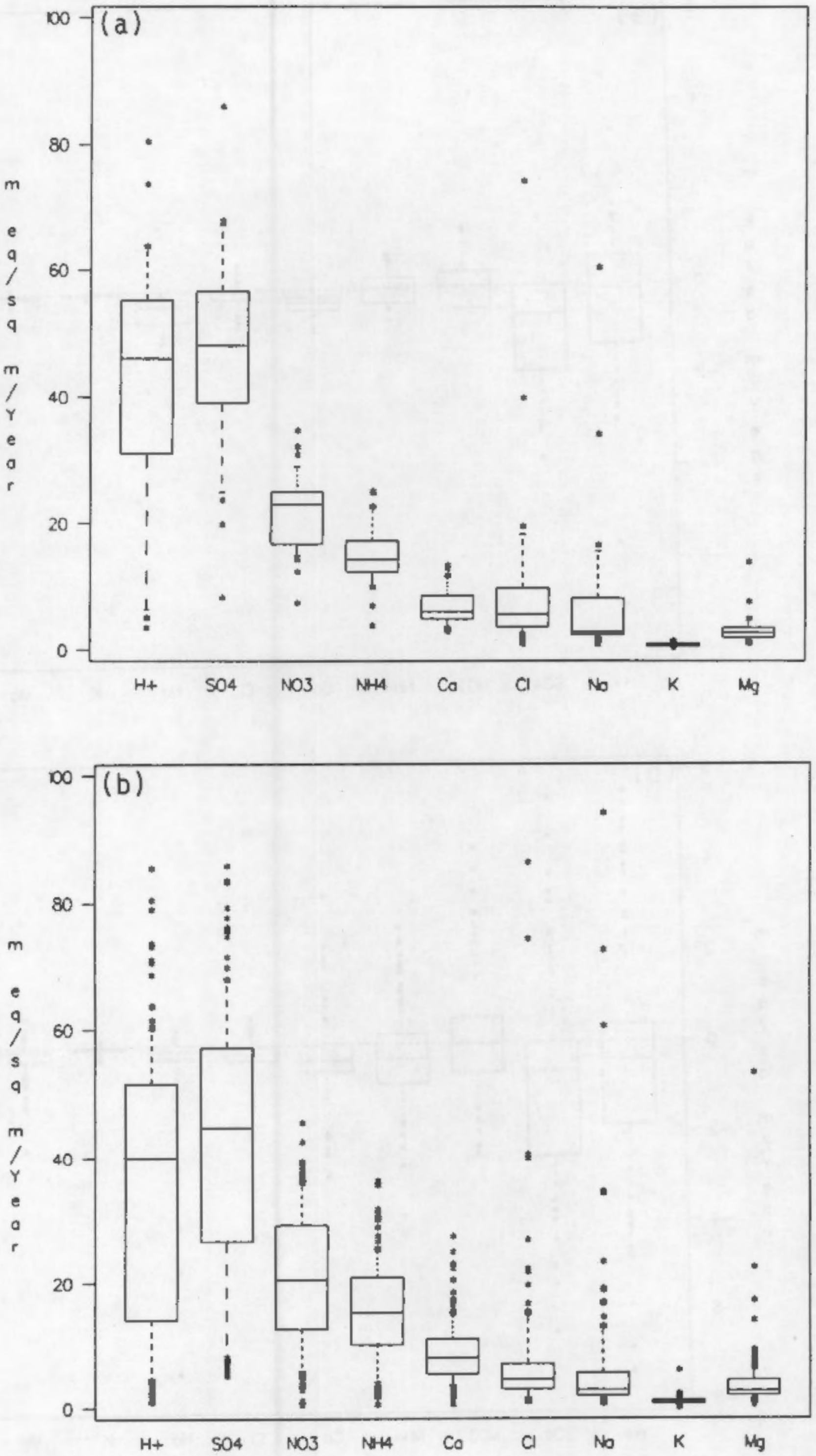


FIGURE 4.8. Average 1982-1986 Ion Species Annual Deposition Distributions for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

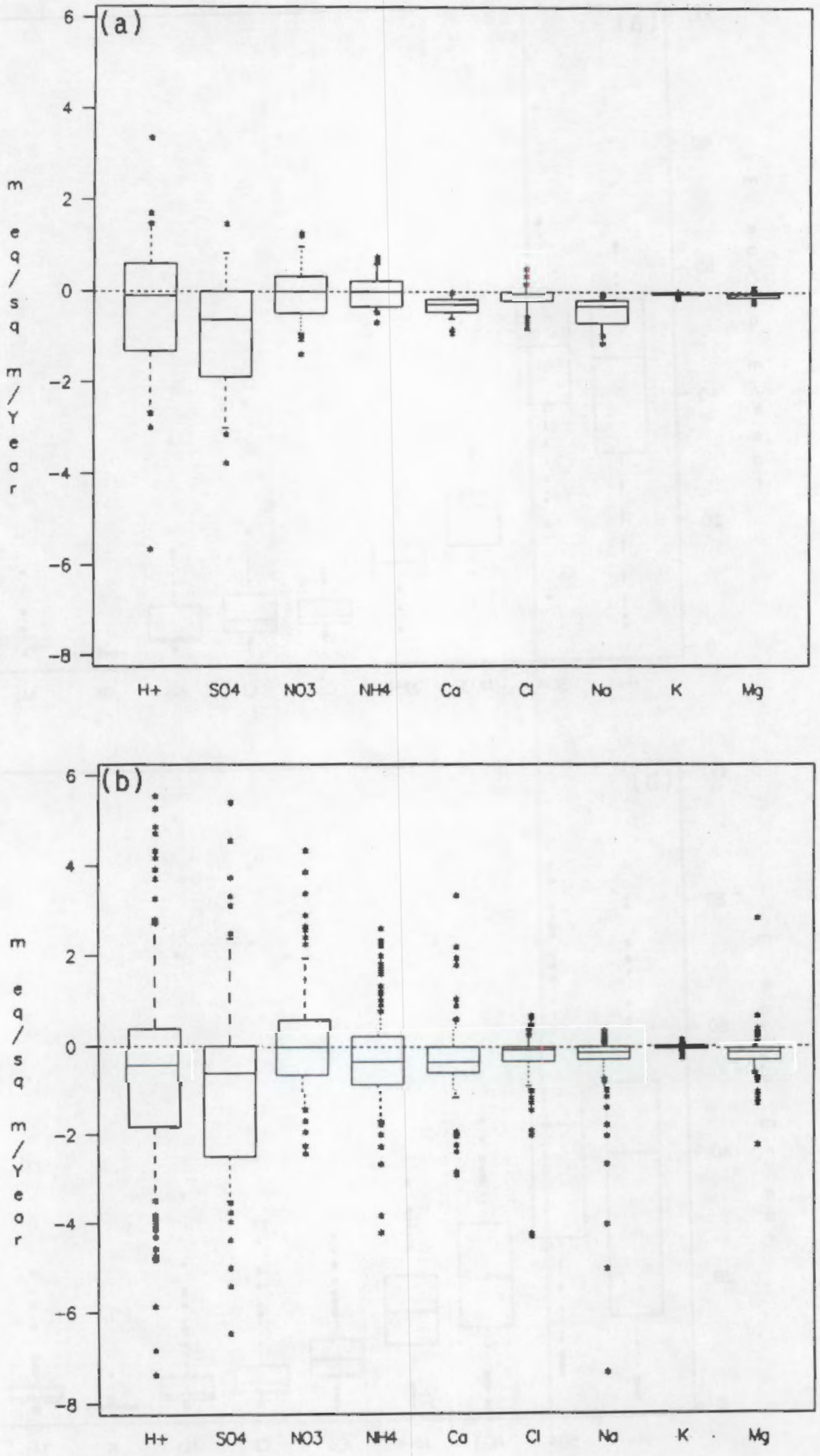


FIGURE 4.9. Ion Species Deposition Trend Estimate Distributions for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

TABLE 4.7. Change per Year (meq/sq m/year) of Annual Deposition for 1979-1986 Trend Sites.

		Percentile of 1979-1986 Sites					
Ion	Min	10th	25th	50th	75th	90th	Max
Precip[a]	-7.74	-4.46	-2.70	-0.40	1.97	3.67	4.77
Hydrogen	-5.64	-2.67	-1.32	-0.10	0.61	1.50	3.38
Sulfate	-3.75	-3.10	-1.90	-0.62	0.00	0.83	1.50
Nitrate	-1.37	-0.96	-0.48	0.00	0.33	0.97	1.29
Ammonium	-0.67	-0.44	-0.33	0.00	0.22	0.65	0.76
Calcium	-0.90	-0.60	-0.45	-0.28	-0.17	-0.01	0.00
Chloride	-0.79	-0.56	-0.22	0.00	0.02	0.22	0.55
Sodium	-1.11	-0.94	-0.69	-0.31	-0.18	-0.06	-0.04
Potassium	-0.12	-0.09	-0.05	-0.03	-0.01	0.00	0.02
Magnesium	-0.24	-0.20	-0.13	-0.06	-0.02	0.03	0.11

[a] Annual precipitation (cm)

TABLE 4.8. Change per Year (meq/sq m/year) of Annual Deposition for 1982-1986 Trend Sites.

		Percentile of 1982-1986 Sites					
Ion	Min	10th	25th	50th	75th	90th	Max
Precip[a]	-18.00	-9.77	-3.67	-0.90	1.20	4.40	17.10
Hydrogen	-7.34	-3.54	-1.85	-0.40	0.40	2.75	8.28
Sulfate	-6.45	-3.54	-2.50	-0.62	0.00	2.50	5.41
Nitrate	-2.42	-1.50	-0.65	0.00	0.60	1.94	4.35
Ammonium	-4.22	-1.72	-0.88	-0.33	0.22	0.81	2.61
Calcium	-2.92	-1.20	-0.60	-0.35	0.00	0.60	3.34
Chloride	-8.85	-0.92	-0.34	-0.06	0.02	0.25	14.87
Sodium	-7.33	-0.73	-0.31	-0.11	0.01	0.11	11.77
Potassium	-0.29	-0.11	-0.06	-0.03	-0.00	0.03	0.13
Magnesium	-2.25	-0.57	-0.31	-0.10	0.00	0.10	2.84

[a] Annual precipitation (cm)

TABLE 4.9. Number and Percent of Significantly ($p<0.05$) Decreasing and Increasing Trend Estimates by Ion Species for Deposition at 1979-1986 Trend Sites and 1982-1986 Trend Sites.

	1979-1986 Trend Sites Significance					1982-1986 Trend Sites Significance				
	Decrease		None	Increase		Decrease		None	Increase	
Ion Species	%	N	N	N	%	%	N	N	N	%
Precipitation	0	0	28	2	7	6	8	125	4	3
Hydrogen	13	4	24	2	7	7	10	124	3	2
Sulfate	27	8	22	0	0	9	13	123	1	1
Nitrate	7	2	27	1	3	7	10	122	5	4
Ammonium	7	2	26	0	7	16	22	112	3	2
Calcium	30	9	21	0	0	25	34	97	6	4
Chloride	20	6	24	0	0	7	9	125	3	2
Sodium	73	22	8	0	0	12	16	121	0	0
Potassium	30	9	21	0	0	18	24	108	5	4
Magnesium	27	8	22	0	0	21	29	106	2	1

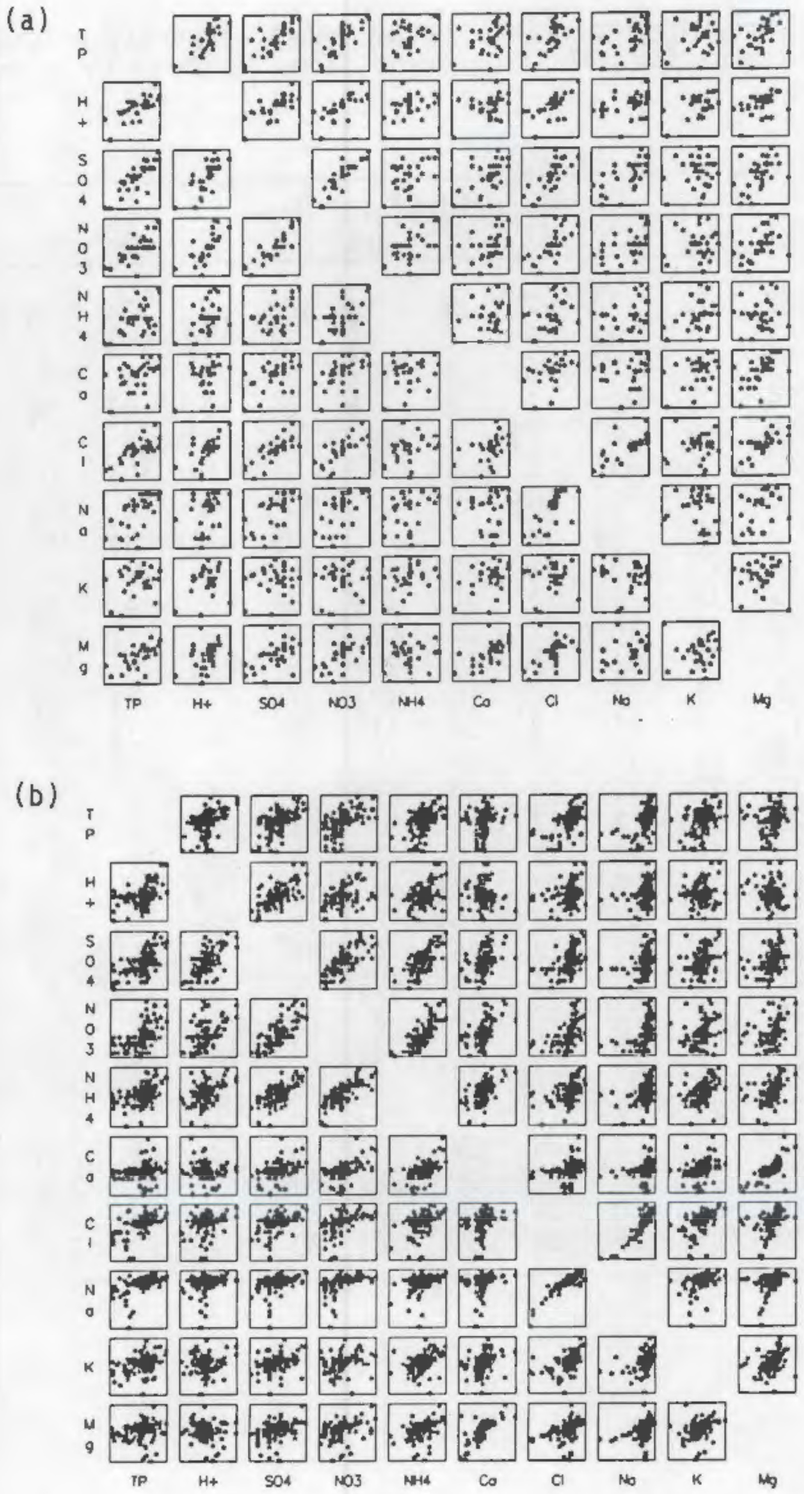


FIGURE 4.10. Scatterplot Matrix of Ion Species Deposition Trend Estimates for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

TABLE 4.10. Ranks by Individual Ion of Annual Deposition Temporal Change for 1979-1986 Trend Sites (Ordered by Average Rank).

		Individual Ion Rank									Ave
Sitename	Precip	H+	SO4	NO3	CL	NH4	NA	K	CA	MG	Rank
Zanesville	2	1	1½	2	4	1½	10	2	1	2½	2.8
Coweeta	1	5	1½	3	3	1½	3½	15½	13½	8	6.1
Piedmont Station	5	7	4	5	1	17½	2	20	8	4	7.6
Hubbard Brook	14	9	3	1	5	17½	6	19	6½	5½	8.1
Clinton Station	16	8	8	7½	2	22	1	21	16	1	9.6
Lewes	4	6	11	6	6	7	9	1	10½	30	9.6
Raleigh	6	2	9	10½	9	4	13	5	26	14½	10.3
Finley (A)	3	10	6½	12	7	7	3½	24½	20½	16	11.9
Parsons	24	22	5	17	12	12	16	9	2	13	12.0
Caldwell	9	12	6½	4	19	17½	18½	26	4	5½	12.6
Lamberton	20	19	18	17	19	7	14	11	6½	12	13.7
Delaware	7	11	11	7½	11	23½	15	27½	9	10½	14.0
Manitou	13	16	16	10½	10	17½	17	15½	13½	10½	14.1
Rockport	11	3	11	9	24	11	23	12	13½	25½	14.7
Georgia Station	8	22	16	17	19	13	7	10	23½	7	14.9
Penn State	12	22	22	26	19	17½	12	4	4	9	15.1
Fort Wayne	21	13	28	17	19	27	21	6	4	2½	15.3
Lewiston	10	14	14	17	8	17½	5	30	17	18	15.6
Mead	27	18	22	17	13½	5	11	15½	18	22½	15.8
Wooster	15	20	13	17	13½	28½	18½	22	10½	14½	17.5
Oxford	25	15	22	17	25	9	25	7	20½	20½	17.9
Illinois	22	25	22	24	19	17½	28	8	13½	19	19.6
Marcell	18	17	22	17	19	3	22	23	29	25½	19.7
Ithaca	17	4	16	22	27	25½	27	18	20½	20½	20.1
Brookhaven	28	30	28	27½	30	10	26	3	20½	17	21.3
Huntington	26	28	22	23	19	17½	20	27½	29	29	23.9
Bradford Forest	19	29	22	25	29	23½	8	29	25	25½	24.0
Virginia	23	24	26	27½	26	25½	30	13	27	22½	24.6
Scranton	29	26	30	30	19	30	24	15½	23½	25½	24.8
Whiteface	30	27	28	29	28	28½	29	24½	29	28	27.9

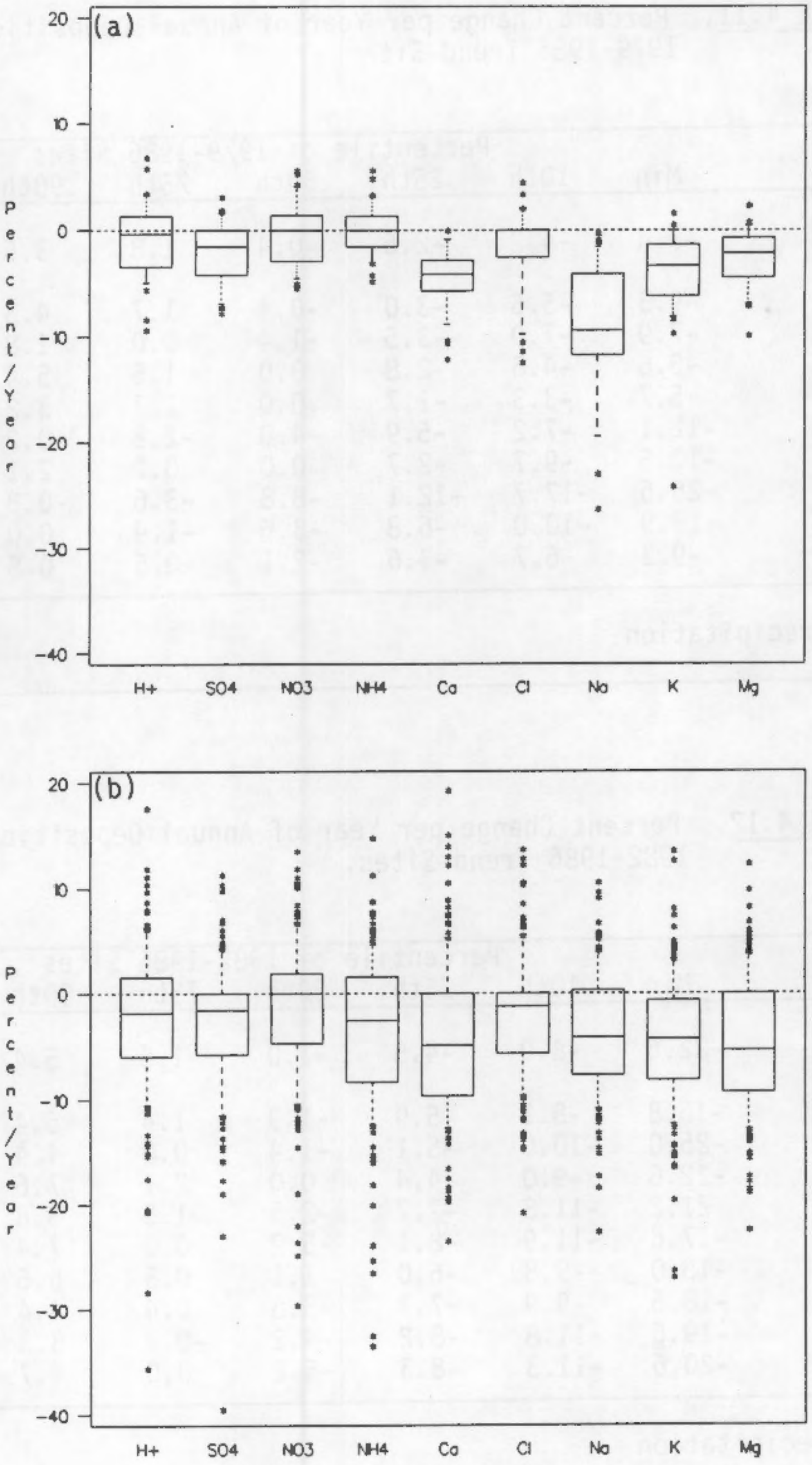


FIGURE 4.11. Distribution of Ion Species Deposition Trend Estimates as Percent of Average 1986 Annual Deposition for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

TABLE 4.11. Percent Change per Year of Annual Deposition for 1979-1986 Trend Sites.

Ion	Min	10th	25th	50th	75th	90th	Max
	Percentile of 1979-1986 Sites						
Precip[a]	-7.4	-4.2	-2.6	-0.4	1.8	3.5	5.9
Hydrogen	-9.3	-5.6	-3.0	-0.4	1.7	4.3	8.1
Sulfate	-7.9	-7.0	-3.5	-1.4	0.0	1.8	3.5
Nitrate	-5.6	-4.6	-2.8	0.0	1.5	5.4	8.1
Ammonium	-5.7	-3.3	-1.7	0.0	1.7	3.2	7.3
Calcium	-12.1	-7.2	-5.9	-4.0	-2.8	-0.2	0.0
Chloride	-10.5	-9.7	-2.7	0.0	0.2	2.1	10.5
Sodium	-26.6	-17.7	-12.1	-8.8	-3.6	-0.8	-0.2
Potassium	-19.9	-10.0	-6.8	-3.6	-1.9	0.0	1.2
Magnesium	-9.3	-6.7	-3.6	-2.1	-0.5	0.8	4.8

[a] Annual precipitation

TABLE 4.12. Percent Change per Year of Annual Deposition for 1982-1986 Trend Sites.

Ion	Min	10th	25th	50th	75th	90th	Max
	Percentile of 1982-1986 Sites						
Precip[a]	-22.6	-8.9	-4.5	-1.0	1.5	5.4	17.6
Hydrogen	-15.8	-8.1	-5.9	-1.9	1.4	6.4	24.3
Sulfate	-25.0	-10.0	-5.1	-1.4	0.D	4.4	13.9
Nitrate	-22.6	-9.0	-4.4	0.0	2.1	7.6	16.0
Ammonium	-21.2	-11.8	-7.7	-2.5	1.5	5.8	16.2
Calcium	-17.6	-11.9	-8.1	-5.2	0.0	7.4	28.6
Chloride	-18.0	-9.8	-6.0	-1.1	0.5	6.6	28.9
Sodium	-18.5	-9.9	-7.3	-3.8	0.4	4.4	13.7
Potassium	-19.6	-11.8	-8.2	-4.2	-0.4	3.3	23.6
Magnesium	-20.6	-11.3	-8.3	-5.2	0.0	4.7	16.2

[a] Annual precipitation

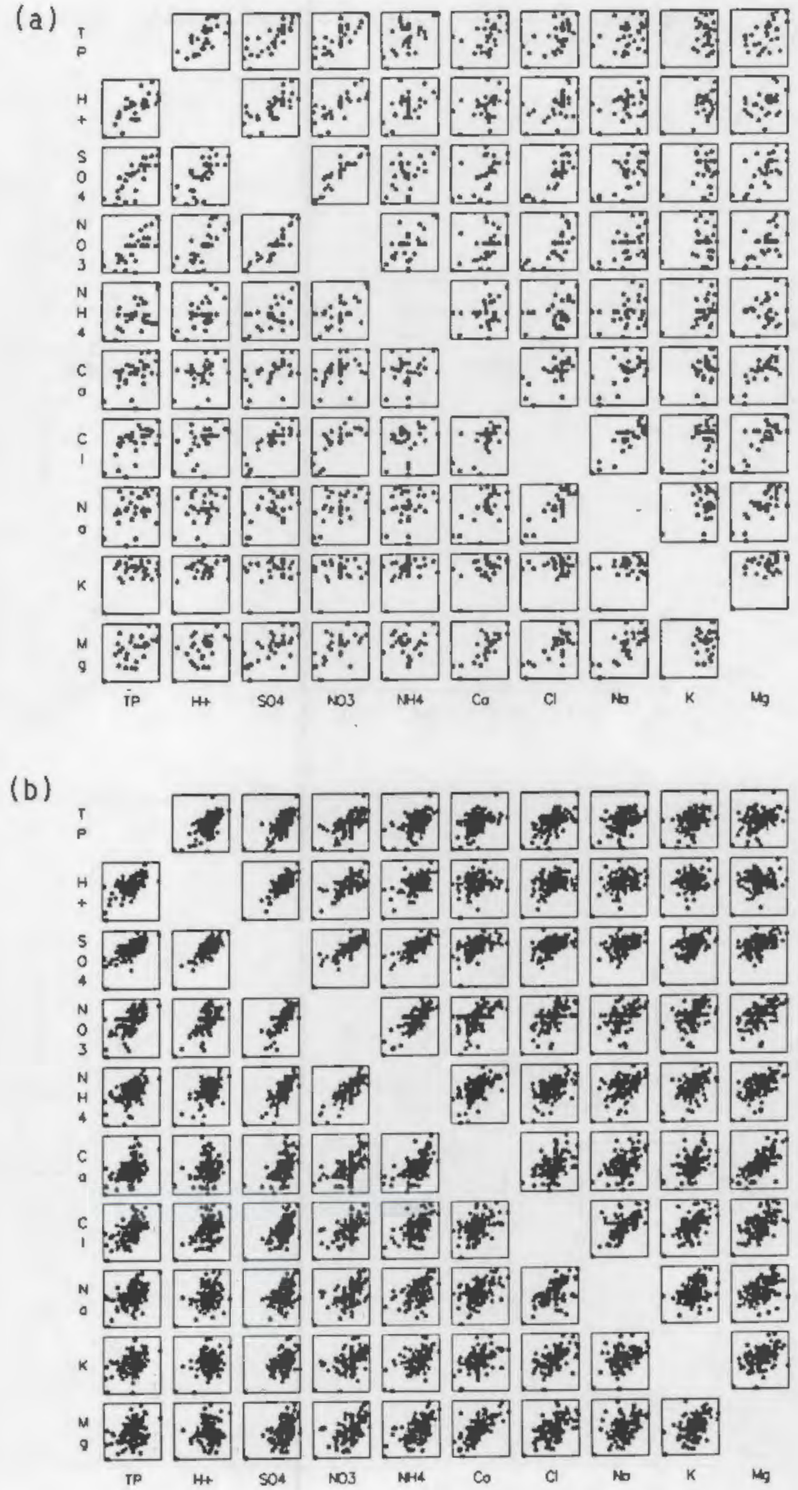


FIGURE 4.12. Scatterplot Matrix of Ion Species Deposition Trend Estimates as Percent of Average 1982-1986 Deposition for (a) 1979-1986 Trend Sites and (b) 1982-1986 Trend Sites.

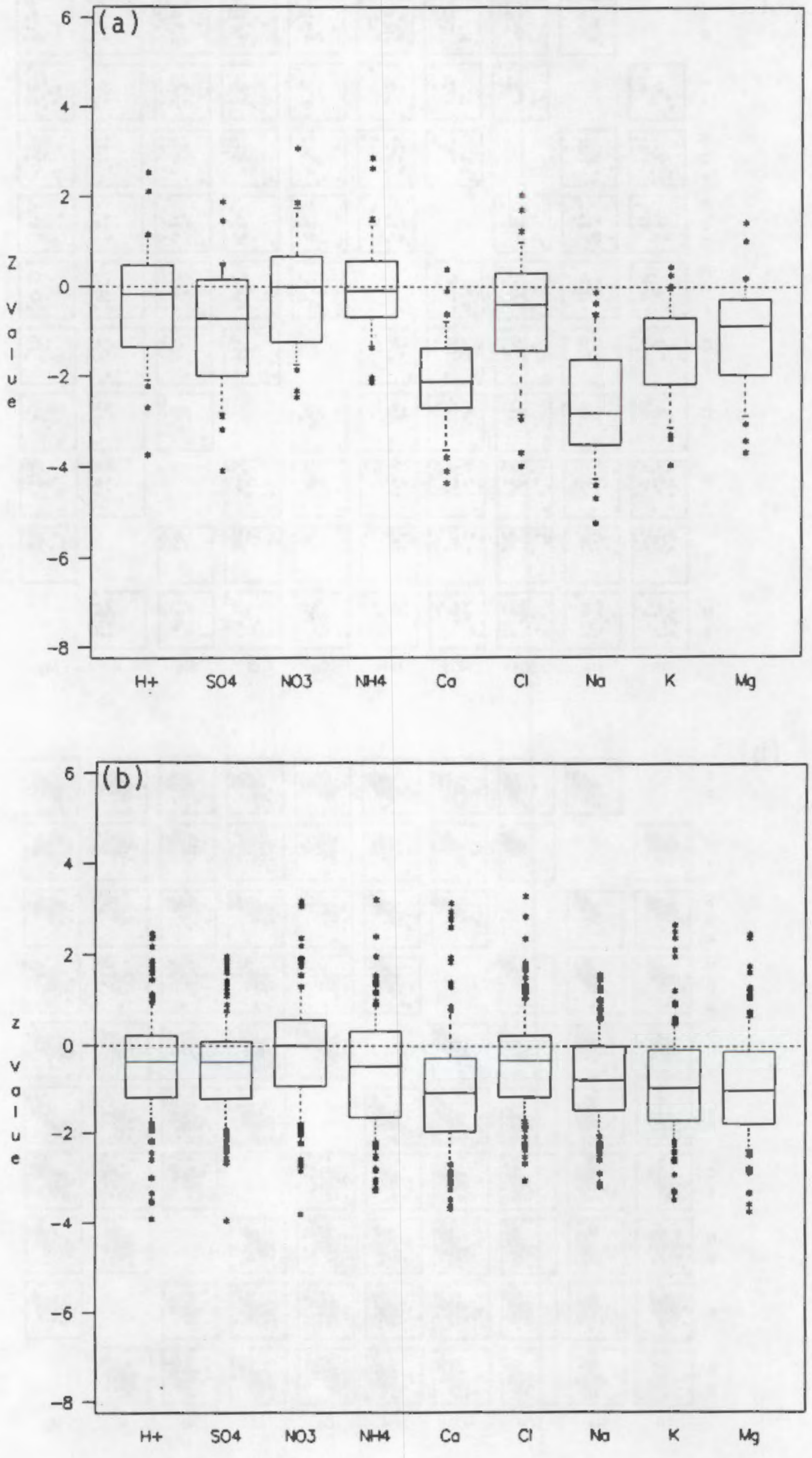


FIGURE 4.13. Distribution by Ion Species of Kendall Seasonal Tau Test z-values for (a) 1979-1986 Trend Sites' Deposition and (b) 1982-1986 Trend Sites' Deposition.

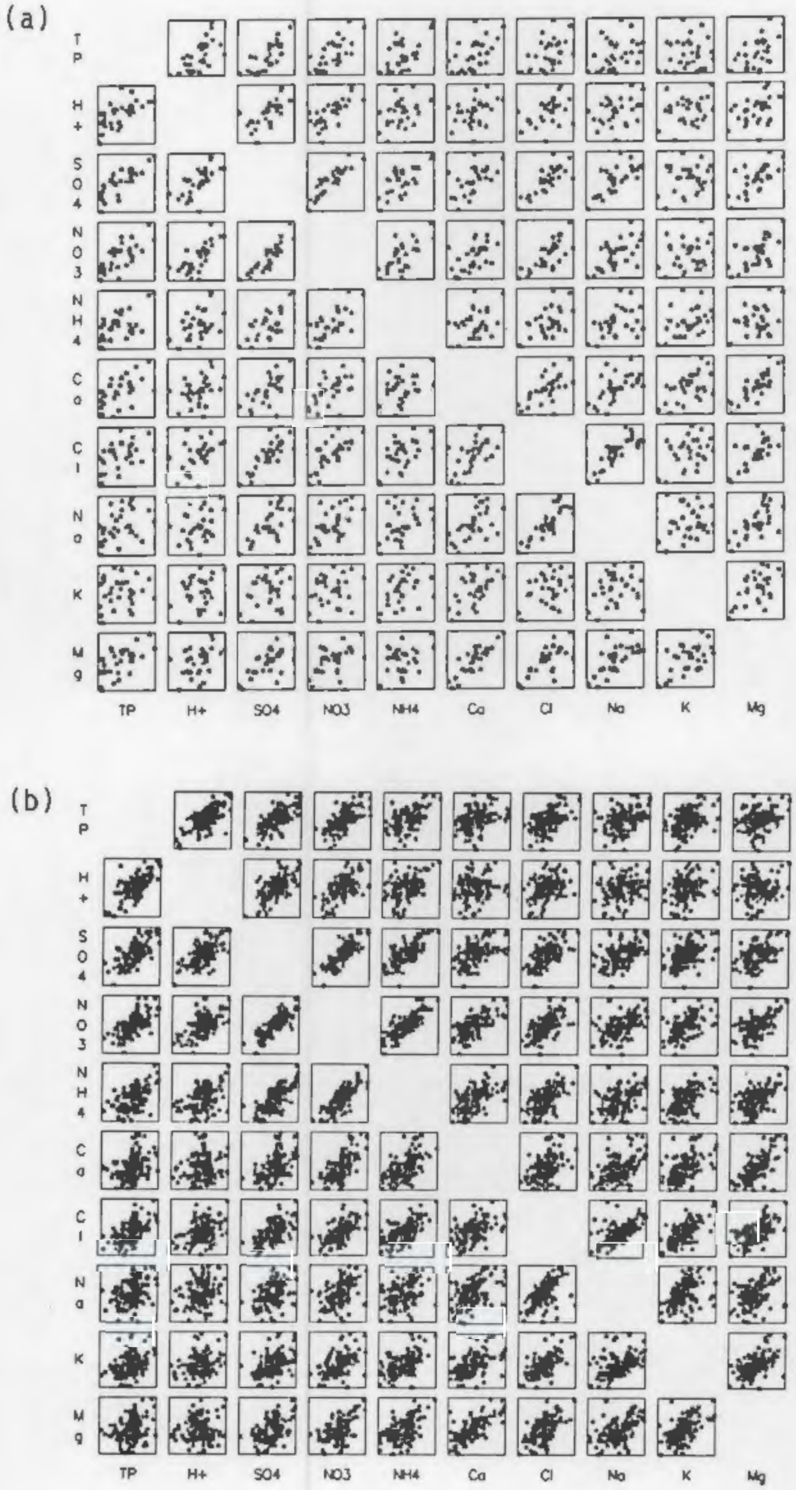


FIGURE 4.14. Scatterplot Matrix of Kendall Seasonal Tau Test Z-values for (a) 1979-1986 Trend Sites' Deposition and (b) 1982-1986 Trend Sites' Deposition.

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APPENDIX A

TABLES AND FIGURES FOR SPATIAL AND TEMPORAL ANALYSIS OF CONCENTRATION AND DEPOSITION

APPENDIX A

TABLES AND FIGURES FOR SPATIAL AND TEMPORAL ANALYSIS OF CONCENTRATION AND DEPOSITION

TABLE A.1. 1979-1986 Trend Sites Ordered by Sitename

Sitename	State	Network	ADSID	Latitude	Longitude
Bradford Forest	Florida	NADP/NTN	015a	29.97	-82.20
Brookhaven	New York	MAP3S	048a	40.87	-72.88
Caldwell	Ohio	NADP/NTN	056a	39.79	-81.53
Clinton Station	North Carolina	NADP/NTN	052a	35.02	-78.28
Coweeta	North Carolina	NADP/NTN	050a	35.06	-83.43
Delaware	Ohio	NADP/NTN	055a	40.35	-83.07
Finley (A)	North Carolina	NADP/NTN	053a	35.73	-78.68
Fort Wayne	Indiana	UAPSP	156a	41.04	-85.32
Georgia Station	Georgia	NADP/NTN	017a	33.18	-84.41
Hubbard Brook	New Hampshire	NADP/NTN	039a	43.94	-71.70
Huntington	New York	NADP/NTN	168a	43.97	-74.22
Illinois	Illinois	MAP3S	020b	40.05	-88.37
Ithaca	New York	MAP3S	044a	42.40	-76.65
Lamberton	Minnesota	NADP/NTN	035a	44.24	-95.30
Lewes	Delaware	MAP3S	013a	38.77	-75.00
Lewiston	North Carolina	NADP/NTN	049a	36.13	-77.17
Manitou	Colorado	NADP/NTN	D11a	39.10	-105.09
Marcell	Minnesota	NADP/NTN	034a	47.53	-93.47
Mead	Nebraska	NADP/NTN	038a	41.15	-96.49
Oxford	Ohio	MAP3S	057a	39.53	-84.72
Parsons	West Virginia	NADP/NTN	075a	39.09	-79.66
Penn State	Pennsylvania	MAP3S	065a	40.79	-77.95
Piedmont Station	North Carolina	NADP/NTN	051a	35.70	-80.62
Raleigh	North Carolina	UAPSP	053c	35.73	-78.68
Rockport	Indiana	UAPSP	154a	37.88	-87.13
Scranton	Pennsylvania	UAPSP	151a	41.58	-75.99
Virginia	Virginia	MAP3S	072a	38.04	-78.54
Whiteface	New York	MAP3S	043a	44.39	-73.86
Wooster	Ohio	NADP/NTN	058a	40.78	-81.93
Zanesville	Ohio	UAPSP	153a	39.98	-82.02

TABLE A.2. 1982-1986 Trend Sites Ordered by Sitename.

Sitename	State/Province	Network	ADSID	Latitude	Longitude
Alamo	Tennessee	UAPSP	246a	35.79	-89.13
Alsea	Oregon	NADP/NTN	059a	44.39	-123.62
Alvinston	Ontario	APIOS-C	180a	42.83	-81.83
American Samoa	American Samoa	NADP/NTN	172a	14.25	-170.56
Argonne	Illinois	NADP/NTN	021a	41.70	-87.99
Ashland	Missouri	NADP/NTN	252a	38.75	-92.20
Aurora	New York	NADP/NTN	040a	42.73	-76.66
Balsam Lake	Ontario	APIOS-D	225a	44.63	-78.86
Bear Island	Ontario	APIOS-C	198a	46.97	-80.08
Bennett Bridge	New York	NADP/NTN	046a	43.53	-75.95
Big Bend Nat'l Pk	Texas	NADP/NTN	070a	29.30	-103.18
Big Moose	New York	UAPSP	243a	43.82	-74.90
Bondville	Illinois	NADP/NTN	020a	40.05	-88.37
Bradford Forest	Florida	NADP/NTN	015a	29.97	-82.20
Bridgton	Maine	NADP/NTN	164a	44.11	-70.73
Brookhaven	New York	MAP3S	048a	40.87	-72.88
Brookings	South Dakota	UAPSP	245a	44.33	-96.83
Caldwell	Ohio	NADP/NTN	056a	39.79	-81.53
Caribou (a)	Maine	NADP/NTN	163a	46.87	-68.01
Charleston Lake	Ontario	APIOS-D	227a	44.50	-76.04
Chautauqua	New York	NADP/NTN	041a	42.30	-79.40
Clearfield	Kentucky	UAPSP	239a	38.14	-83.45
Clinton	Mississippi	UAPSP	242a	32.35	-90.29
Clinton Station	North Carolina	NADP/NTN	052a	35.02	-78.28
Colchester	Ontario	APIOS-C	176a	41.99	-82.93
Coldwater	Ontario	APIOS-C	190a	44.63	-79.54
Coweeta	North Carolina	NADP/NTN	050a	35.06	-83.43
Cuba	New Mexico	NADP/NTN	280a	36.04	-106.97
Dalhousie Mills	Ontario	APIOS-C	193a	45.32	-74.47
Davis	California	NADP/NTN	009a	38.53	-121.77
Delaware	Ohio	NADP/NTN	055a	40.35	-83.07
Dixon Springs	Illinois	NADP/NTN	023a	37.44	-88.67
Dorion	Ontario	APIOS-C	204a	48.84	-88.61
Dorset (b)	Ontario	APIOS-D	087b	45.22	-78.93
Dorset (c)	Ontario	APIOS-C	087c	45.22	-78.93
Douglas Lake	Michigan	NADP/NTN	031a	45.56	-84.68
Ear Falls	Ontario	APIOS-C	206a	50.64	-93.22
East	Massachusetts	NADP/NTN	277a	42.38	-71.21
Everglades Nat. Pa	Florida	NADP/NTN	016a	25.39	-80.70
Fayetteville	Arkansas	NADP/NTN	004a	36.10	-94.17
Fernberg	Minnesota	NADP/NTN	166a	47.95	-91.49
Fernberg	Minnesota	APIOS-D	166b	47.95	-91.49
Finley (A)	North Carolina	NADP/NTN	053a	35.73	-78.68
Forest Seed Ctr	Texas	NADP/NTN	254a	31.56	-94.86
Fort Wayne	Indiana	UAPSP	156a	41.04	-85.32
Gaylord	Michigan	UAPSP	241a	44.95	-84.64
Georgia Station	Georgia	NADP/NTN	017a	33.18	-84.41

TABLE A.2. (Continued)

Sitename	State/Province	Network	ADSID	Latitude	Longitude
Glacier Nat'l Park	Montana	NADP/NTN	037a	48.51	-114.00
Golden Lake	Ontario	APIOS-C	194a	45.61	-77.20
Gowganda	Ontario	APIOS-C	2D0a	47.65	-80.78
Grand Canyon	Arizona	NADP/NTN	068a	36.07	-112.15
Great Smoky Mts	Tennessee	NADP/NTN	028a	35.66	-83.59
Greenville Station	Maine	NADP/NTN	030a	45.49	-69.66
H.J. Andrews	Oregon	NADP/NTN	061a	44.22	-122.24
Hopland (Ukiah)	California	NADP/NTN	007a	39.00	-123.08
Hubbard Brook	New Hampshire	NADP/NTN	039a	43.94	-71.70
Huntington	New York	NADP/NTN	168a	43.97	-74.22
Huron Park	Ontario	APIOS-C	183a	43.29	-81.50
Illinois	Illinois	MAP3S	020b	40.05	-88.37
Indiana Dunes	Indiana	NADP/NTN	025a	41.63	-87.09
Ithaca	New York	MAP3S	044a	42.40	-76.65
Jasper	New York	NADP/NTN	047a	42.11	-77.54
Kane	Pennsylvania	NADP/NTN	063a	41.60	-78.77
Kellogg	Michigan	NADP/NTN	032a	42.41	-85.39
Killarney	Ontario	APIOS-C	196a	45.99	-81.49
Lac Le Croix (a)	Ontario	APIOS-C	208a	48.35	-92.21
Lamberton	Minnesota	NADP/NTN	035a	44.24	-95.30
Lancaster	Kansas	UAPSP	238a	39.57	-95.30
Leading Ridge	Pennsylvania	NADP/NTN	064a	40.66	-77.94
Lewes	Delaware	MAP3S	013a	38.77	-75.00
Lewiston	North Carolina	NADP/NTN	049a	36.13	-77.17
Longwoods (b)	Ontario	APIOS-D	143b	42.88	-81.48
Manitou	Colorado	NADP/NTN	011a	39.10	-105.09
Marcell	Minnesota	NADP/NTN	034a	47.53	-93.47
Marshall	Texas	UAPSP	247a	32.67	-94.42
Mattawa	Ontario	APIOS-C	197a	46.28	-78.82
Mauna Loa	Hawaii	NADP/NTN	018a	19.54	-155.58
McKellar	Ontario	APIOS-C	195a	45.52	-79.92
Mead	Nebraska	NADP/NTN	038a	41.15	-96.49
Melbourne	Ontario	APIDS-D	221a	42.79	-81.56
Meridian	Mississippi	NADP/NTN	036a	32.33	-88.74
Merlin	Ontario	APIDS-C	177a	42.25	-82.22
Mesa Verde	Colorado	NADP/NTN	029a	37.2D	-108.49
Moonbeam	Ontario	APIOS-C	201a	49.32	-82.15
NACL	Massachusetts	NADP/NTN	251a	41.97	-70.02
Newcastle	Wyoming	NADP/NTN	255a	43.87	-104.19
Nithgrove	Ontario	APIOS-D	224a	45.20	-79.07
North Easthope	Ontario	APIOS-D	222a	43.41	-80.89
Oak Ridge	Tennessee	MAP3S	171b	35.96	-84.29
Oliver Knoll	Arizona	NADP/NTN	054a	33.07	-109.86
Organ Pipe Mon.	Arizona	NADP/NTN	003a	31.95	-112.80
Oxford	Ohio	MAP3S	057a	39.53	-84.72
Palmerston	Ontario	APIOS-C	182a	43.80	-80.90
Parsons	West Virginia	NADP/NTN	075a	39.09	-79.66

TABLE A.2. (Continued)

Sitename	State/Province	Network	ADSID	Latitude	Longitude
Pawnee	Colorado	NADP/NTN	012a	40.81	-104.75
Penn State	Pennsylvania	MAP3S	065a	40.79	-77.95
Pickle Lake	Ontario	APIOS-C	207a	51.46	-90.20
Piedmont Station	North Carolina	NADP/NTN	051a	35.70	-80.62
Pinedale	Wyoming	NADP/NTN	284a	42.93	-109.79
Port Stanley	Ontario	APIOS-C	178a	42.67	-81.16
Quetico Centre	Ontario	APIOS-D	233a	48.41	-91.20
Railton	Ontario	APIOS-D	228a	44.38	-76.59
Raleigh	North Carolina	UAPSP	053c	35.73	-78.68
Raven Lake	Ontario	APIOS-D	226a	44.61	-78.91
Rockport	Indiana	UAPSP	154a	37.88	-87.13
Rocky Mt. Nat Park	Colorado	NADP/NTN	010a	40.36	-105.56
Salem	Illinois	NADP/NTN	161a	38.64	-88.97
Sand Spring	Colorado	NADP/NTN	173a	40.51	-107.70
Scranton	Pennsylvania	UAPSP	151a	41.57	-75.99
Selma	Alabama	UAPSP	235a	32.47	-87.08
Sequoia Nat. Park	California	NADP/NTN	008a	36.57	-118.78
Shabbona	Illinois	NADP/NTN	024a	41.84	-88.85
Shallow Lake	Ontario	APIOS-C	181a	44.58	-81.09
Smith's Falls	Ontario	APIOS-C	192a	44.94	-75.96
Southern Ill U	Illinois	NADP/NTN	022a	37.71	-89.27
Spooner	Wisconsin	NADP/NTN	077a	45.82	-91.88
Tanbark Flat	California	NADP/NTN	270a	34.21	-117.76
Trout Lake	Wisconsin	NADP/NTN	D76a	46.05	-89.65
Turners Falls	Massachusetts	UAPSP	150a	42.60	-72.55
Underhill Center	Vermont	UAPSP	248a	44.53	-72.87
University Forest	Missouri	NADP/NTN	253a	36.91	-90.32
Uvalda	Georgia	UAPSP	237a	32.05	-82.47
Uxbridge	Ontario	APIOS-C	187a	44.21	-79.21
Victoria	Texas	NADP/NTN	071a	28.84	-96.92
Virginia	Virginia	MAP3S	072a	38.04	-78.54
Walker Branch	Tennessee	NADP/NTN	171a	35.96	-84.29
Washington Xing	New Jersey	NADP/NTN	285a	40.31	-74.85
Waterloo	Ontario	APIOS-C	184a	43.48	-80.59
Wellesley	Ontario	APIOS-D	223a	43.47	-80.76
Wellston	Michigan	NADP/NTN	033a	44.22	-85.82
Whiteface	New York	MAP3S	043a	44.39	-73.86
Whitney	Ontario	APIOS-C	203a	45.54	-78.26
Wilberforce	Ontario	APIOS-C	188a	45.01	-78.22
Wilkesport	Ontario	APIOS-C	179a	42.70	-82.35
Winterport	Maine	UAPSP	240a	44.62	-68.97
Wooster	Ohio	NADP/NTN	058a	40.78	-81.92
Zanesville	Ohio	UAPSP	153a	39.98	-82.02

TABLE A.3. 1986 Frequency Distribution Percentiles Concentrations

		Number of Sites	Min	5	10	25	50	75	90	95	Max
pH											
	Annual	186	4.11	4.17	4.22	4.31	4.51	4.90	5.36	5.44	5.66
	Winter	188	4.04	4.11	4.17	4.39	4.52	4.90	5.33	5.46	5.75
	Spring	245	4.05	4.21	4.24	4.35	4.57	5.28	5.63	5.83	6.54
	Summer	243	3.91	4.07	4.12	4.25	4.50	4.96	5.37	5.56	6.09
	Fall	246	4.12	4.22	4.26	4.38	4.61	4.99	5.33	5.50	6.73
Sulfate											
	Annual	195	0.14	0.30	0.53	1.11	1.88	2.60	3.21	3.48	3.96
	Winter	188	0.01	0.16	0.24	0.63	1.30	2.06	2.75	3.30	4.96
	Spring	257	0.20	0.38	0.49	1.10	1.95	2.88	3.50	3.81	4.75
	Summer	243	0.30	0.55	0.70	1.03	1.84	3.19	4.02	4.50	5.50
	Fall	241	0.14	0.30	0.50	0.82	1.37	2.05	2.66	2.85	2.61
Nitrate											
	Annual	193	0.02	0.22	0.45	0.76	1.19	1.80	2.11	2.33	2.72
	Winter	189	0.01	0.09	0.24	0.54	1.31	2.40	3.49	3.92	5.17
	Spring	256	0.20	0.38	0.49	1.10	1.95	2.88	3.50	3.81	4.75
	Summer	245	0.30	0.55	0.70	1.03	1.84	3.19	4.02	4.50	5.50
	Fall	240	0.04	0.25	0.37	0.55	0.88	1.37	1.78	1.95	2.46
Ammonium											
	Annual	194	0.01	0.03	0.06	0.14	0.25	0.36	0.48	0.57	0.73
	Winter	187	0.01	0.01	0.02	0.06	0.17	0.30	0.43	0.55	0.75
	Spring	256	0.01	0.04	0.09	0.19	0.35	0.50	0.68	0.74	1.08
	Summer	245	0.01	0.04	0.06	0.13	0.23	0.37	0.50	0.58	0.97
	Fall	236	0.01	0.03	0.05	0.09	0.16	0.28	0.40	0.47	0.72
Calcium											
	Annual	198	0.02	0.04	0.04	0.09	0.14	0.22	0.30	0.34	0.62
	Winter	185	0.01	0.02	0.02	0.05	0.09	0.16	0.30	0.46	1.14
	Spring	252	0.01	0.05	0.08	0.13	0.21	0.32	0.45	0.60	0.97
	Summer	243	0.01	0.04	0.05	0.10	0.17	0.25	0.35	0.51	1.16
	Fall	240	0.01	0.02	0.03	0.05	0.09	0.14	0.21	0.28	1.11

TABLE A.4. 1986 Frequency Distribution Percentiles Depositions

		Number of Sites	Min	5	10	25	50	75	90	95	Max
Hydrogen											
	Annual	186	0.003	0.012	0.032	0.101	0.300	0.484	0.650	0.732	1.018
	Winter	188	0.000	0.002	0.006	0.024	0.070	0.114	0.168	0.188	0.274
	Spring	245	0.000	0.001	0.003	0.010	0.054	0.101	0.127	0.153	0.203
	Summer	243	0.000	0.002	0.005	0.024	0.091	0.178	0.254	0.305	0.459
	Fall	246	0.001	0.002	0.004	0.017	0.043	0.085	0.111	0.131	0.194
Sulfate											
	Annual	195	0.40	1.98	2.82	10.90	16.90	26.70	32.06	35.84	53.30
	Winter	188	0.00	0.41	0.70	1.90	3.90	5.70	8.00	8.89	12.80
	Spring	257	0.10	0.30	0.70	1.95	4.60	6.05	7.52	8.40	11.50
	Summer	243	0.00	0.34	0.80	2.30	5.40	10.00	13.90	16.08	23.90
	Fall	241	0.00	0.10	0.20	1.00	2.00	3.40	4.40	5.20	8.70
Nitrate											
	Annual	193	0.07	1.51	1.90	6.90	10.70	17.45	22.80	24.59	32.40
	Winter	189	0.00	0.40	0.50	1.20	2.40	3.60	5.29	6.00	8.80
	Spring	256	0.10	0.29	0.40	1.23	2.85	4.30	5.30	6.11	8.70
	Summer	245	0.00	0.33	0.80	2.00	3.30	5.40	7.04	8.17	12.20
	Fall	240	0.00	0.20	0.30	0.75	1.60	3.75	5.50	6.40	9.40
Ammonium											
	Annual	194	0.11	0.26	0.38	1.25	2.27	3.60	4.90	5.40	7.50
	Winter	187	0.01	0.06	0.08	0.21	0.40	0.73	1.13	1.36	2.55
	Spring	256	0.03	0.05	0.09	0.33	0.70	1.10	1.50	1.64	2.81
	Summer	245	0.00	0.05	0.11	0.28	0.63	1.12	1.64	1.92	4.06
	Fall	236	0.00	0.02	0.03	0.10	0.20	0.40	0.67	0.79	1.40
Calcium											
	Annual	196	0.12	0.34	0.41	0.75	1.27	1.97	2.74	3.46	5.96
	Winter	185	0.01	0.04	0.08	0.12	0.20	0.33	0.55	0.75	1.99
	Spring	252	0.01	0.09	0.14	0.23	0.42	0.65	0.91	1.12	2.00
	Summer	243	0.00	0.06	0.13	0.24	0.41	0.70	0.98	1.23	2.49
	Fall	240	0.00	0.03	0.04	0.07	0.13	0.23	0.40	0.73	1.39

TABLE A.5. Summary of 1979-1986 Trend Sites (n=30) Annual Precipitation-weighted Concentration by Year.

			Std		Percentile							
	Year	Mean	Dev	Min	5th	10th	25th	50th	75th	90th	95th	Max
pH	1979	4.47	0.31	4.12	4.13	4.15	4.25	4.36	4.65	5.02	5.15	5.30
	1980	4.38	0.32	4.07	4.08	4.11	4.16	4.28	4.48	5.06	5.11	5.16
	1981	4.43	0.32	4.09	4.09	4.12	4.23	4.34	4.52	5.04	5.26	5.39
	1982	4.46	0.32	4.16	4.17	4.18	4.28	4.37	4.53	4.98	5.34	5.46
	1983	4.48	0.28	4.14	4.16	4.23	4.29	4.39	4.62	4.96	5.18	5.31
	1984	4.46	0.28	4.10	4.13	4.16	4.25	4.37	4.61	5.01	5.08	5.10
	1985	4.46	0.31	4.14	4.15	4.19	4.24	4.35	4.56	5.11	5.28	5.35
	1986	4.44	0.33	4.12	4.13	4.15	4.25	4.35	4.50	5.01	5.42	5.43
Hydrogen (µg/l)	1979	41.7	21.6	5.0	7.4	9.6	22.2	44.0	57.5	69.9	74.3	76.1
	1980	50.6	23.4	7.0	7.8	8.7	33.0	55.0	68.7	78.0	83.7	85.6
	1981	44.3	21.1	4.1	5.6	9.2	30.2	46.1	59.5	74.9	81.3	81.8
	1982	41.8	19.2	3.5	4.7	10.4	29.3	43.4	54.9	66.6	68.4	68.6
	1983	38.7	17.9	4.9	7.4	10.8	23.7	40.5	51.3	59.2	70.0	73.0
	1984	40.5	19.5	7.9	8.2	9.7	24.6	42.3	56.5	68.4	74.2	78.7
	1985	41.9	19.3	4.5	5.4	7.8	28.0	45.4	57.3	64.3	70.7	72.0
	1986	43.5	19.4	3.7	3.8	9.8	31.5	44.5	56.4	71.2	74.8	75.7
Sulfate (mg/l)	1979	2.36	0.76	1.07	1.20	1.45	1.80	2.18	2.98	3.42	3.95	4.24
	1980	2.63	0.75	1.33	1.36	1.42	2.18	2.61	3.24	3.64	4.06	4.15
	1981	2.51	0.83	1.17	1.25	1.50	1.77	2.56	3.23	3.53	3.96	4.46
	1982	2.30	0.74	1.02	1.15	1.26	1.81	2.15	2.93	3.29	3.49	3.57
	1983	2.06	0.60	0.91	1.04	1.22	1.64	1.95	2.59	2.86	3.03	3.05
	1984	2.19	0.75	1.12	1.23	1.38	1.56	1.99	2.82	3.34	3.52	3.59
	1985	2.13	0.72	0.87	0.96	1.18	1.50	2.04	2.68	3.19	3.26	3.31
	1986	2.34	0.77	1.04	1.11	1.36	1.69	2.29	2.92	3.52	3.80	3.96
Nitrate (mg/l)	1979	1.29	0.41	0.47	0.55	0.67	0.98	1.48	1.61	1.77	1.83	1.90
	1980	1.59	0.41	0.68	0.77	0.93	1.26	1.72	1.92	2.04	2.13	2.22
	1981	1.41	0.38	0.83	0.84	0.85	1.07	1.47	1.77	1.94	1.97	2.00
	1982	1.31	0.40	0.59	0.63	0.73	0.99	1.23	1.67	1.84	2.01	2.15
	1983	1.23	0.33	0.63	0.64	0.66	1.07	1.17	1.58	1.69	1.73	1.73
	1984	1.37	0.44	0.76	0.78	0.79	1.01	1.29	1.78	2.07	2.13	2.15
	1985	1.33	0.40	0.74	0.76	0.80	0.93	1.40	1.63	1.81	2.11	2.21
	1986	1.44	0.38	0.82	0.88	0.94	1.17	1.39	1.80	2.00	2.10	2.14
Ammonium (mg/l)	1979	0.27	0.16	0.06	0.07	0.10	0.19	0.23	0.31	0.46	0.70	0.73
	1980	0.32	0.16	0.11	0.12	0.13	0.24	0.28	0.37	0.45	0.81	0.85
	1981	0.31	0.14	0.12	0.15	0.17	0.20	0.27	0.36	0.49	0.66	0.79
	1982	0.27	0.11	0.10	0.11	0.13	0.18	0.27	0.32	0.41	0.53	0.53
	1983	0.28	0.15	0.10	0.11	0.16	0.18	0.25	0.31	0.39	0.76	0.87
	1984	0.28	0.13	0.13	0.14	0.16	0.19	0.25	0.35	0.50	0.56	0.61
	1985	0.26	0.11	0.08	0.11	0.14	0.18	0.24	0.33	0.41	0.52	0.54
	1986	0.29	0.12	0.08	0.11	0.14	0.20	0.29	0.36	0.49	0.55	0.60
Calcium (mg/l)	1979	0.17	0.12	0.05	0.05	0.07	0.09	0.11	0.24	0.41	0.45	0.46
	1980	0.21	0.14	0.07	0.08	0.10	0.12	0.17	0.29	0.34	0.59	0.67
	1981	0.19	0.11	0.03	0.04	0.09	0.12	0.16	0.24	0.35	0.46	0.55
	1982	0.15	0.08	0.06	0.07	0.07	0.08	0.13	0.21	0.27	0.33	0.37
	1983	0.16	0.10	0.06	0.07	0.07	0.10	0.12	0.21	0.27	0.43	0.45
	1984	0.17	0.09	0.06	0.06	0.07	0.10	0.15	0.23	0.30	0.36	0.37
	1985	0.13	0.08	0.04	0.04	0.05	0.08	0.11	0.16	0.21	0.33	0.38
	1986	0.14	0.07	0.04	0.04	0.05	0.07	0.10	0.20	0.24	0.27	0.28

TABLE A.5. (Continued)

	Year	Mean	Std Dev	Min	Percentile							Max
					5th	10th	25th	50th	75th	90th	95th	
Magnesium (mg/l)	1979	0.04	0.02	0.02	0.02	0.02	0.03	0.04	0.05	0.07	0.10	0.13
	1980	0.04	0.03	0.01	0.02	0.02	0.03	0.04	0.05	0.08	0.13	0.16
	1981	0.05	0.03	0.01	0.01	0.02	0.03	0.05	0.08	0.08	0.10	0.13
	1982	0.04	0.03	0.01	0.01	0.02	0.02	0.03	0.04	0.08	0.12	0.15
	1983	0.04	0.03	0.01	0.01	0.01	0.02	0.04	0.05	0.08	0.13	0.18
	1984	0.04	0.03	0.01	0.01	0.02	0.03	0.04	0.05	0.07	0.11	0.15
	1985	0.04	0.03	0.01	0.01	0.02	0.02	0.03	0.04	0.06	0.11	0.15
	1986	0.03	0.03	0.01	0.01	0.02	0.02	0.03	0.04	0.05	0.12	0.16
Sodium (mg/l)	1979	0.31	0.22	0.05	0.06	0.08	0.14	0.26	0.45	0.61	0.83	1.07
	1980	0.26	0.28	0.05	0.05	0.06	0.10	0.18	0.32	0.53	1.09	1.42
	1981	0.17	0.17	0.02	0.03	0.04	0.08	0.11	0.17	0.52	0.69	0.76
	1982	0.19	0.29	0.04	0.04	0.05	0.07	0.09	0.15	0.54	1.12	1.47
	1983	0.20	0.32	0.03	0.04	0.05	0.06	0.08	0.22	0.45	1.17	1.67
	1984	0.17	0.24	0.04	0.04	0.05	0.05	0.08	0.17	0.37	0.88	1.25
	1985	0.14	0.18	0.03	0.03	0.04	0.05	0.06	0.15	0.37	0.69	0.86
	1986	0.13	0.20	0.03	0.03	0.04	0.05	0.06	0.15	0.27	0.79	1.01
Chloride (mg/l)	1979	0.35	0.33	0.11	0.12	0.12	0.15	0.23	0.39	0.73	1.34	1.68
	1980	0.40	0.50	0.13	0.14	0.16	0.20	0.23	0.40	0.66	2.01	2.70
	1981	0.30	0.30	0.08	0.09	0.10	0.15	0.19	0.32	0.72	1.28	1.39
	1982	0.35	0.55	0.06	0.06	0.09	0.15	0.21	0.28	0.48	2.11	2.98
	1983	0.36	0.51	0.09	0.10	0.11	0.13	0.16	0.40	0.78	1.92	2.65
	1984	0.34	0.45	0.10	0.11	0.11	0.17	0.20	0.32	0.65	1.73	2.44
	1985	0.30	0.36	0.07	0.08	0.10	0.13	0.18	0.32	0.68	1.38	1.94
	1986	0.30	0.39	0.07	0.08	0.08	0.11	0.20	0.31	0.49	1.58	2.08
Potassium (mg/l)	1979	0.05	0.02	0.02	0.02	0.02	0.03	0.04	0.07	0.08	0.09	0.10
	1980	0.05	0.03	0.02	0.02	0.02	0.03	0.04	0.06	0.12	0.14	0.15
	1981	0.04	0.02	0.01	0.01	0.02	0.02	0.03	0.05	0.08	0.10	0.11
	1982	0.03	0.02	0.02	0.02	0.02	0.02	0.03	0.04	0.05	0.07	0.07
	1983	0.04	0.02	0.02	0.02	0.02	0.02	0.03	0.05	0.07	0.09	0.09
	1984	0.04	0.02	0.02	0.02	0.02	0.02	0.03	0.04	0.06	0.10	0.11
	1985	0.03	0.01	0.01	0.01	0.01	0.02	0.03	0.04	0.05	0.06	0.07
	1986	0.04	0.02	0.01	0.01	0.02	0.02	0.03	0.05	0.06	0.08	0.09

TABLE A.6. Summary of 1979-1986 Trend Sites (n=30) Annual Deposition by Year.

			Std		Percentile							
	Year	Mean	Dev	Min	5th	10th	25th	50th	75th	90th	95th	Max
Hydrogen (kg/ha)	1979	0.48	0.27	0.04	0.04	0.06	0.30	0.45	0.68	0.86	0.96	1.06
	1980	0.49	0.26	0.03	0.04	0.05	0.36	0.51	0.62	0.86	0.98	1.11
	1981	0.43	0.25	0.02	0.04	0.06	0.23	0.44	0.55	0.85	0.89	0.90
	1982	0.42	0.18	0.03	0.04	0.06	0.34	0.44	0.58	0.61	0.68	0.74
	1983	0.44	0.20	0.03	0.04	0.07	0.33	0.46	0.59	0.71	0.75	0.77
	1984	0.43	0.21	0.05	0.06	0.07	0.32	0.43	0.58	0.69	0.82	0.94
	1985	0.43	0.21	0.04	0.04	0.06	0.29	0.45	0.57	0.68	0.83	0.97
	1986	0.43	0.22	0.03	0.04	0.05	0.32	0.42	0.56	0.71	0.85	1.02
Sulfate (kg/ha)	1979	26.14	10.21	6.40	10.14	14.13	17.65	24.35	34.15	41.48	46.62	49.70
	1980	24.57	10.30	5.80	7.39	10.78	19.18	22.45	29.95	43.48	44.83	45.60
	1981	23.87	10.85	1.80	4.11	10.37	13.48	24.95	30.85	39.97	44.65	45.80
	1982	23.38	7.56	4.60	7.79	11.05	20.10	23.50	29.03	32.08	36.19	40.10
	1983	22.73	6.98	4.50	7.25	11.63	20.50	22.35	27.97	30.95	32.60	34.20
	1984	23.08	7.98	4.70	7.56	12.25	18.22	22.30	29.02	34.21	38.67	41.20
	1985	21.28	8.77	3.50	6.58	9.31	15.30	22.65	26.50	29.78	40.17	47.60
	1986	22.87	9.49	3.30	6.38	11.06	16.98	22.55	28.97	33.06	43.29	53.30
Nitrate (kg/ha)	1979	13.76	4.58	5.90	6.06	8.67	10.05	12.80	16.80	21.36	22.61	23.00
	1980	14.27	4.96	6.20	6.53	8.34	11.00	13.85	17.28	23.44	25.11	26.10
	1981	13.01	5.41	1.90	4.05	6.96	8.58	12.60	16.55	20.35	24.12	24.40
	1982	12.94	3.51	5.20	6.63	8.77	10.52	13.10	15.85	18.24	19.32	19.70
	1983	13.20	3.08	5.20	6.63	8.30	11.52	13.80	15.58	17.06	17.86	18.30
	1984	14.17	4.35	5.20	6.63	8.83	10.96	13.90	16.80	19.20	23.35	25.60
	1985	12.98	4.43	4.10	6.08	7.82	9.00	14.15	15.70	18.81	21.55	24.30
	1986	13.88	4.64	4.20	5.96	7.89	10.12	13.80	17.25	19.76	23.41	27.70
Ammonium (kg/ha)	1979	2.73	1.10	0.79	0.80	1.03	1.90	2.95	3.34	4.00	4.62	5.31
	1980	2.71	1.01	1.10	1.16	1.38	2.00	2.55	3.72	4.23	4.54	4.67
	1981	2.74	1.11	0.18	0.59	1.36	1.96	2.64	3.70	4.22	4.60	4.98
	1982	2.64	0.93	0.77	1.15	1.53	2.06	2.57	3.04	4.06	4.75	5.09
	1983	2.84	0.88	0.67	1.27	1.75	2.34	2.86	3.27	3.79	4.92	5.52
	1984	2.84	0.96	0.91	1.28	1.73	2.17	2.63	3.56	4.50	4.57	4.58
	1985	2.48	0.85	0.60	0.83	1.63	1.79	2.40	3.25	3.44	3.93	4.23
	1986	2.82	1.27	0.49	0.67	1.55	1.86	2.65	3.61	5.15	5.50	5.87
Calcium (kg/ha)	1979	1.71	0.80	0.63	0.67	0.73	1.13	1.50	2.42	2.97	3.26	3.56
	1980	1.80	0.93	0.79	0.83	0.88	1.07	1.42	2.58	3.30	3.79	3.91
	1981	1.67	0.78	0.0S	0.25	0.85	1.10	1.52	2.01	2.88	3.32	3.73
	1982	1.51	0.70	0.54	0.56	0.63	0.96	1.49	1.92	2.60	3.05	3.31
	1983	1.55	0.57	0.71	0.79	0.67	1.08	1.50	1.62	2.61	2.80	2.87
	1984	1.65	0.70	0.60	0.65	0.74	1.11	1.62	2.03	2.64	3.06	3.30
	1985	1.22	0.60	0.39	0.40	0.64	0.71	0.99	1.64	2.25	2.47	2.65
	1986	1.28	0.73	0.34	0.35	0.52	0.75	0.94	1.86	2.45	2.89	3.04

TABLE A.6. (Continued)

	Year	Mean	Std Dev	Min	Percentile 5th	10th	25th	50th	75th	90th	95th	Max
Magnesium (kg/ha)	1979	0.45	0.30	0.16	0.17	0.19	0.27	0.40	0.51	0.76	1.23	1.77
	1980	0.39	0.26	0.11	0.13	0.18	0.24	0.36	0.49	0.55	1.15	1.47
	1981	0.42	0.20	0.01	0.07	0.18	0.27	0.42	0.55	0.73	0.75	0.75
	1982	0.42	0.30	0.08	0.11	0.15	0.23	0.36	0.49	0.89	1.22	1.50
	1983	0.47	0.46	0.13	0.13	0.15	0.26	0.32	0.51	0.77	1.87	2.57
	1984	0.43	0.28	0.13	0.13	0.17	0.24	0.37	0.52	0.69	1.23	1.53
	1985	0.35	0.28	0.11	0.12	0.15	0.21	0.28	0.39	0.62	1.14	1.61
	1986	0.31	0.25	0.12	0.13	0.15	0.18	0.27	0.32	0.41	1.12	1.38
Sodium (kg/ha)	1979	3.55	3.00	0.47	0.59	0.77	1.28	2.93	5.05	7.01	10.91	15.19
	1980	2.35	2.54	0.41	0.46	0.55	0.81	1.41	3.51	4.87	9.52	12.99
	1981	1.54	1.62	0.04	0.18	0.38	0.70	1.04	1.63	4.69	6.48	6.75
	1982	2.06	3.02	0.25	0.30	0.37	0.64	0.85	2.25	6.27	11.40	14.46
	1983	2.54	4.69	0.34	0.35	0.44	0.53	0.79	2.54	4.86	17.22	24.22
	1984	1.88	2.64	0.33	0.38	0.45	0.57	0.73	2.09	4.34	10.17	12.80
	1985	1.40	1.94	0.17	0.22	0.29	0.51	0.66	1.43	4.67	6.98	9.45
	1986	1.24	1.63	0.16	0.25	0.35	0.40	0.58	1.25	2.26	7.41	8.89
Chloride (kg/ha)	1979	4.14	4.54	0.52	0.67	1.03	1.39	2.98	4.90	8.71	16.84	23.78
	1980	3.68	4.57	0.82	0.89	1.10	1.40	2.32	3.98	8.04	17.62	24.74
	1981	2.80	2.80	0.14	0.35	0.68	1.21	2.06	2.99	8.43	11.85	12.41
	1982	3.70	5.56	0.49	0.51	0.67	1.46	1.94	3.35	8.68	21.47	29.20
	1983	4.61	7.51	0.45	0.64	0.88	1.33	1.78	4.70	9.04	28.37	38.45
	1984	3.82	5.01	0.46	0.64	1.02	1.57	1.99	3.80	7.58	19.99	24.95
	1985	3.07	3.99	0.41	0.52	0.63	1.26	1.95	2.95	7.88	14.61	21.29
	1986	2.85	3.55	0.26	0.50	0.77	1.20	2.17	2.85	4.08	14.70	18.27
Potassium (kg/ha)	1979	0.52	0.24	0.13	0.16	0.21	0.30	0.52	0.65	0.88	1.02	1.03
	1980	0.47	0.31	0.15	0.16	0.22	0.28	0.38	0.55	1.04	1.31	1.54
	1981	0.35	0.21	0.01	0.08	0.14	0.21	0.30	0.45	0.68	0.91	0.98
	1982	0.34	0.16	0.12	0.14	0.18	0.21	0.32	0.40	0.83	0.74	0.77
	1983	0.42	0.21	0.17	0.19	0.21	0.26	0.38	0.53	0.77	0.69	0.99
	1984	0.39	0.21	0.17	0.18	0.19	0.25	0.34	0.50	0.84	0.96	1.23
	1985	0.31	0.15	0.11	0.13	0.15	0.18	0.28	0.44	0.51	0.62	0.74
	1986	0.34	0.20	0.12	0.12	0.13	0.20	0.27	0.45	0.68	0.79	0.81

TABLE A.7. Summary of 1982-1986 Trend Sites (n=137) Annual Precipitation-weighted Concentration by Year.

			Std				Percentile					
	Year	Mean	Dev	Min	5th	10th	25th	50th	75th	90th	95th	Max
pH	1982	4.53	0.39	4.03	4.15	4.18	4.22	4.38	4.76	5.23	5.34	5.64
	1983	4.61	0.36	4.14	4.23	4.25	4.33	4.47	4.83	5.22	5.43	5.82
	1984	4.57	0.39	4.07	4.17	4.22	4.30	4.39	4.82	5.14	5.39	6.05
	1985	4.57	0.38	4.14	4.19	4.21	4.30	4.42	4.83	5.20	5.35	5.70
	1986	4.58	0.39	4.11	4.17	4.22	4.30	4.41	4.89	5.15	5.42	5.87
Hydrogen (µg/l)	1982	39.2	23.2	2.3	4.5	5.8	17.5	41.5	60.1	66.9	71.6	92.4
	1983	31.6	17.9	1.5	3.7	6.0	14.7	33.7	46.8	55.9	59.5	73.0
	1984	35.6	20.7	0.9	4.0	7.2	15.3	40.4	50.8	60.9	67.5	84.5
	1985	35.2	20.0	2.0	4.5	6.4	14.7	37.8	50.7	61.2	64.4	72.0
	1986	35.2	20.3	1.3	3.8	7.1	12.8	38.6	50.5	60.2	68.6	76.8
Sulfate (mg/l)	1982	2.19	1.02	0.27	0.57	0.91	1.38	2.22	2.91	3.53	3.80	5.00
	1983	1.97	0.94	0.32	0.55	0.85	1.21	1.89	2.60	3.03	3.78	4.64
	1984	2.13	0.98	0.31	0.70	0.98	1.38	2.01	2.78	3.36	3.81	4.72
	1985	2.03	0.93	0.24	0.64	0.75	1.21	2.08	2.77	3.16	3.55	4.20
	1986	2.05	0.95	0.18	0.47	0.77	1.31	2.08	2.76	3.39	3.55	3.96
Nitrate (mg/l)	1982	1.35	0.63	0.07	0.37	0.65	0.87	1.20	1.78	2.36	2.48	2.75
	1983	1.25	0.59	0.02	0.37	0.58	0.80	1.15	1.67	2.05	2.34	2.78
	1984	1.42	0.67	0.02	0.44	0.74	0.91	1.28	1.94	2.45	2.66	3.30
	1985	1.40	0.68	0.03	0.36	0.66	0.88	1.30	2.01	2.35	2.53	3.03
	1986	1.39	0.64	0.02	0.43	0.65	0.85	1.29	1.97	2.26	2.55	2.72
Ammonium (mg/l)	1982	0.31	0.17	0.02	0.07	0.12	0.19	0.29	0.40	0.53	0.65	0.85
	1983	0.29	0.17	0.02	0.06	0.10	0.17	0.27	0.37	0.52	0.64	0.87
	1984	0.32	0.16	0.01	0.06	0.13	0.19	0.30	0.41	0.55	0.64	0.79
	1985	0.29	0.17	0.01	0.07	0.12	0.17	0.27	0.40	0.51	0.58	1.24
	1986	0.30	0.16	0.01	0.06	0.09	0.18	0.28	0.39	0.53	0.60	0.73
Calcium (mg/l)	1962	0.20	0.14	0.02	0.05	0.07	0.11	0.18	0.24	0.37	0.51	0.78
	1983	0.21	0.16	0.03	0.05	0.07	0.11	0.17	0.26	0.42	0.56	0.91
	1984	0.22	0.14	0.03	0.07	0.08	0.12	0.19	0.28	0.40	0.51	0.89
	1985	0.19	0.12	0.03	0.05	0.07	0.10	0.18	0.26	0.36	0.42	0.82
	1986	0.17	0.11	0.01	0.04	0.05	0.09	0.16	0.22	0.32	0.38	0.62
Magnesium (mg/l)	1982	0.05	0.04	0.01	0.02	0.02	0.03	0.04	0.05	0.09	0.14	0.28
	1983	0.05	0.04	0.01	0.02	0.02	0.03	0.04	0.05	0.09	0.13	0.32
	1984	0.05	0.04	0.01	0.02	0.02	0.03	0.04	0.05	0.09	0.13	0.34
	1985	0.04	0.03	0.01	0.02	0.02	0.03	0.04	0.05	0.07	0.09	0.27
	1986	0.04	0.03	0.01	0.01	0.02	0.02	0.03	0.04	0.06	0.09	0.22
Sodium (mg/l)	1982	0.15	0.27	0.03	0.03	0.04	0.05	0.08	0.14	0.26	0.57	2.20
	1983	0.17	0.35	0.02	0.04	0.04	0.05	0.07	0.12	0.30	0.56	2.58
	1984	0.16	0.30	0.03	0.04	0.05	0.06	0.09	0.15	0.27	0.49	2.70
	1985	0.13	0.25	0.02	0.03	0.04	0.05	0.06	0.10	0.26	0.50	2.18
	1986	0.13	0.22	0.01	0.03	0.04	0.05	0.06	0.11	0.26	0.59	1.78
Chloride (mg/l)	1982	0.29	0.48	0.06	0.06	0.08	0.11	0.18	0.26	0.46	0.93	3.83
	1983	0.32	0.62	0.04	0.07	0.09	0.12	0.15	0.21	0.57	1.06	4.65
	1984	0.31	0.55	0.05	0.09	0.11	0.14	0.18	0.27	0.43	0.85	5.00
	1985	0.27	0.46	0.05	0.07	0.09	0.12	0.16	0.23	0.48	0.88	4.03
	1986	0.26	0.41	0.05	0.07	0.08	0.11	0.15	0.26	0.49	1.05	3.37
Potassium (mg/l)	1982	0.04	0.02	0.01	0.01	0.02	0.02	0.03	0.05	0.06	0.07	0.12
	1983	0.04	0.02	0.01	0.02	0.02	0.03	0.03	0.05	0.06	0.09	0.12
	1984	0.04	0.02	0.01	0.02	0.02	0.03	0.04	0.05	0.07	0.09	0.14
	1985	0.03	0.02	0.01	0.01	0.02	0.02	0.03	0.04	0.05	0.07	0.11
	1986	0.03	0.02	0.00	0.01	0.01	0.02	0.03	0.04	0.06	0.07	0.11

TABLE A.8. Summary of 1982-1986 Trend Sites (n=137) Annual Deposition by Year.

	Year	Mean	Std Dev	Min	Percentile							Max
					5th	10th	25th	50th	75th	90th	95th	
Hydrogen (kg/ha)	1982	0.38	0.23	0.01	0.03	0.05	0.17	0.42	0.58	0.66	0.73	0.89
	1983	0.33	0.21	0.01	0.03	0.05	0.16	0.36	0.48	0.59	0.68	0.83
	1984	0.36	0.23	0.00	0.03	0.04	0.11	0.38	0.52	0.62	0.70	0.94
	1985	0.35	0.22	0.01	0.02	0.04	0.14	0.36	0.52	0.61	0.69	0.97
	1986	0.35	0.23	0.01	0.02	0.04	0.11	0.38	0.51	0.63	0.72	1.02
Sulfate (kg/ha)	1982	21.14	10.39	2.20	3.86	5.42	12.15	22.20	28.85	34.34	37.42	43.80
	1983	19.91	9.73	0.80	3.70	4.98	12.90	21.70	26.65	31.42	35.46	40.50
	1984	20.68	10.60	1.80	3.49	4.54	12.20	21.10	28.40	34.80	37.70	44.30
	1985	19.80	11.03	1.20	2.40	3.46	11.75	20.60	27.36	33.22	38.97	47.60
	1986	19.87	11.13	1.50	2.79	3.80	11.45	19.60	27.85	34.26	38.59	53.30
Nitrate (kg/ha)	1982	12.88	6.11	0.90	2.90	3.56	8.15	12.90	16.90	21.08	23.02	26.50
	1983	12.37	5.89	0.10	2.77	3.98	7.85	12.70	16.70	20.04	21.71	28.40
	1984	13.43	7.02	0.20	2.50	3.46	7.95	13.40	19.00	22.80	25.24	29.60
	1985	13.36	7.75	0.30	1.79	2.68	7.75	13.00	19.15	24.32	28.10	30.90
	1986	13.13	7.48	0.30	1.90	2.96	7.55	11.80	19.85	23.80	25.74	32.40
Ammonium (kg/ha)	1982	2.87	1.49	0.33	0.51	0.76	1.89	2.80	3.93	4.78	5.59	7.44
	1983	2.82	1.42	0.03	0.51	0.73	1.76	2.80	3.61	4.64	5.54	7.17
	1984	2.93	1.51	0.08	0.52	0.71	1.82	2.99	3.88	5.08	5.56	6.28
	1985	2.75	1.69	0.06	0.27	0.51	1.61	2.59	3.68	4.87	6.21	8.67
	1986	2.75	1.68	0.08	0.31	0.51	1.63	2.54	3.88	5.24	5.72	7.50
Calcium (kg/ha)	1982	1.80	1.09	0.20	0.54	0.68	1.06	1.59	2.18	3.30	4.05	6.29
	1983	1.95	1.22	0.09	0.69	0.77	1.09	1.69	2.41	3.45	4.59	6.83
	1984	1.93	1.11	0.15	0.47	0.80	1.21	1.75	2.41	3.21	4.05	7.88
	1985	1.75	1.11	0.13	0.39	0.56	0.81	1.50	2.42	3.32	4.27	4.75
	1986	1.56	1.06	0.08	0.36	0.47	0.81	1.31	1.98	2.92	3.93	5.96
Magnesium (kg/ha)	1982	0.48	0.58	0.08	0.15	0.17	0.23	0.31	0.50	0.89	1.40	5.65
	1983	0.52	0.71	0.03	0.14	0.16	0.23	0.34	0.55	0.97	1.25	5.51
	1984	0.51	0.80	0.07	0.14	0.19	0.27	0.37	0.52	0.75	1.12	8.85
	1985	0.43	0.54	0.08	0.12	0.14	0.22	0.32	0.46	0.70	0.98	5.84
	1986	0.38	0.59	0.04	0.09	0.13	0.19	0.27	0.39	0.66	0.92	6.53
Sodium (kg/ha)	1982	1.82	4.52	0.19	0.23	0.30	0.42	0.73	1.37	3.40	6.82	44.33
	1983	2.27	5.94	0.14	0.25	0.34	0.45	0.65	1.24	4.57	7.90	44.06
	1984	2.03	6.50	0.19	0.33	0.42	0.49	0.73	1.38	3.41	5.39	69.98
	1985	1.50	4.38	0.14	0.18	0.27	0.42	0.60	0.99	2.21	4.93	46.93
	1986	1.53	4.84	0.11	0.24	0.29	0.38	0.59	0.95	2.19	5.67	53.33
Chloride (kg/ha)	1982	3.40	7.95	0.32	0.45	0.52	0.90	1.62	2.81	5.59	9.30	77.34
	1983	4.21	10.59	0.17	0.45	0.51	0.90	1.49	2.31	8.17	14.02	80.26
	1984	3.88	12.00	0.34	0.47	0.58	1.13	1.69	2.61	5.74	9.46	129.30
	1985	3.04	8.10	0.31	0.41	0.47	0.91	1.48	2.42	4.23	8.91	86.57
	1986	3.09	9.11	0.19	0.38	0.48	0.86	1.35	2.51	4.00	9.93	100.78
Potassium (kg/ha)	1982	0.37	0.24	0.06	0.12	0.15	0.22	0.32	0.44	0.64	0.80	1.89
	1983	0.40	0.28	0.02	0.13	0.17	0.23	0.34	0.46	0.72	0.99	1.99
	1984	0.40	0.35	0.02	0.12	0.17	0.23	0.34	0.46	0.66	0.89	3.59
	1985	0.32	0.25	0.02	0.08	0.11	0.17	0.27	0.39	0.52	0.74	2.30
	1986	0.32	0.26	0.03	0.08	0.11	0.18	0.25	0.42	0.58	0.77	2.25

TABLE A.9. Summary of 1979-1986 Trend Sites (n=30) and 1982-1986 Trend Sites (n=137) Annual Precipitation (cm) by Year.

			Std					Percentile				
	Year	Mean	Dev	Min	5th	10th	25th	50th	75th	90th	95th	Max
1979-86 Sites	1979	112.8	35.2	37.2	49.9	72.6	85.7	120.1	131.4	141.4	185.0	226.5
	1980	91.8	25.9	41.5	44.7	51.2	80.0	92.2	100.4	126.9	151.2	171.4
	1981	91.9	27.3	15.3	28.9	56.1	78.6	91.9	104.1	128.8	144.3	151.5
	1982	103.3	26.6	45.6	61.4	77.8	87.0	98.6	115.1	149.1	165.8	182.6
	1983	112.0	30.6	48.8	56.9	66.4	94.8	110.5	126.8	151.5	176.2	197.5
	1984	107.2	26.6	41.6	60.2	75.9	87.8	108.0	124.4	139.5	158.1	179.9
	1985	98.6	20.3	39.7	50.8	79.2	90.3	98.7	108.5	128.3	139.1	149.8
	1986	96.3	21.1	31.8	51.7	76.2	87.1	93.7	109.1	126.3	133.2	134.6
1982-86 Sites	1982	98.7	34.5	26.6	37.5	62.1	80.5	95.4	112.8	145.9	161.8	234.8
	1983	103.7	38.4	15.2	43.7	51.6	83.6	98.1	123.8	153.3	177.0	258.7
	1984	97.0	37.1	17.1	36.3	45.8	79.8	94.2	117.7	134.2	143.2	274.3
	1985	93.5	30.1	21.3	35.4	43.4	76.3	98.3	109.8	128.4	136.0	214.9
	1986	94.3	33.8	15.9	37.0	57.3	76.8	94.6	107.5	124.9	133.2	299.2

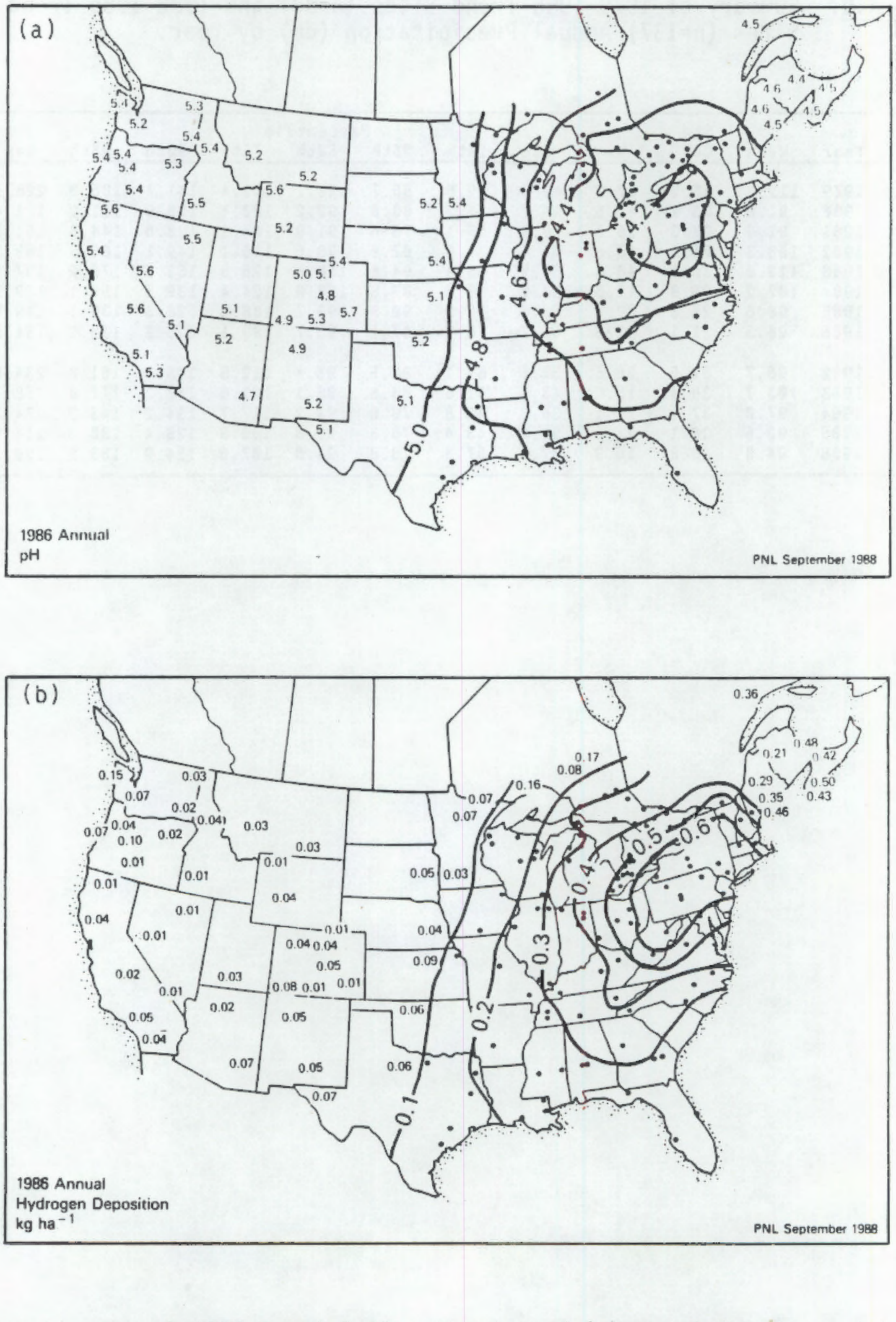


FIGURE A.1. Annual 1986 Spatial Distribution of (a) Precipitation-weighted pH and (b) Hydrogen Ion Deposition

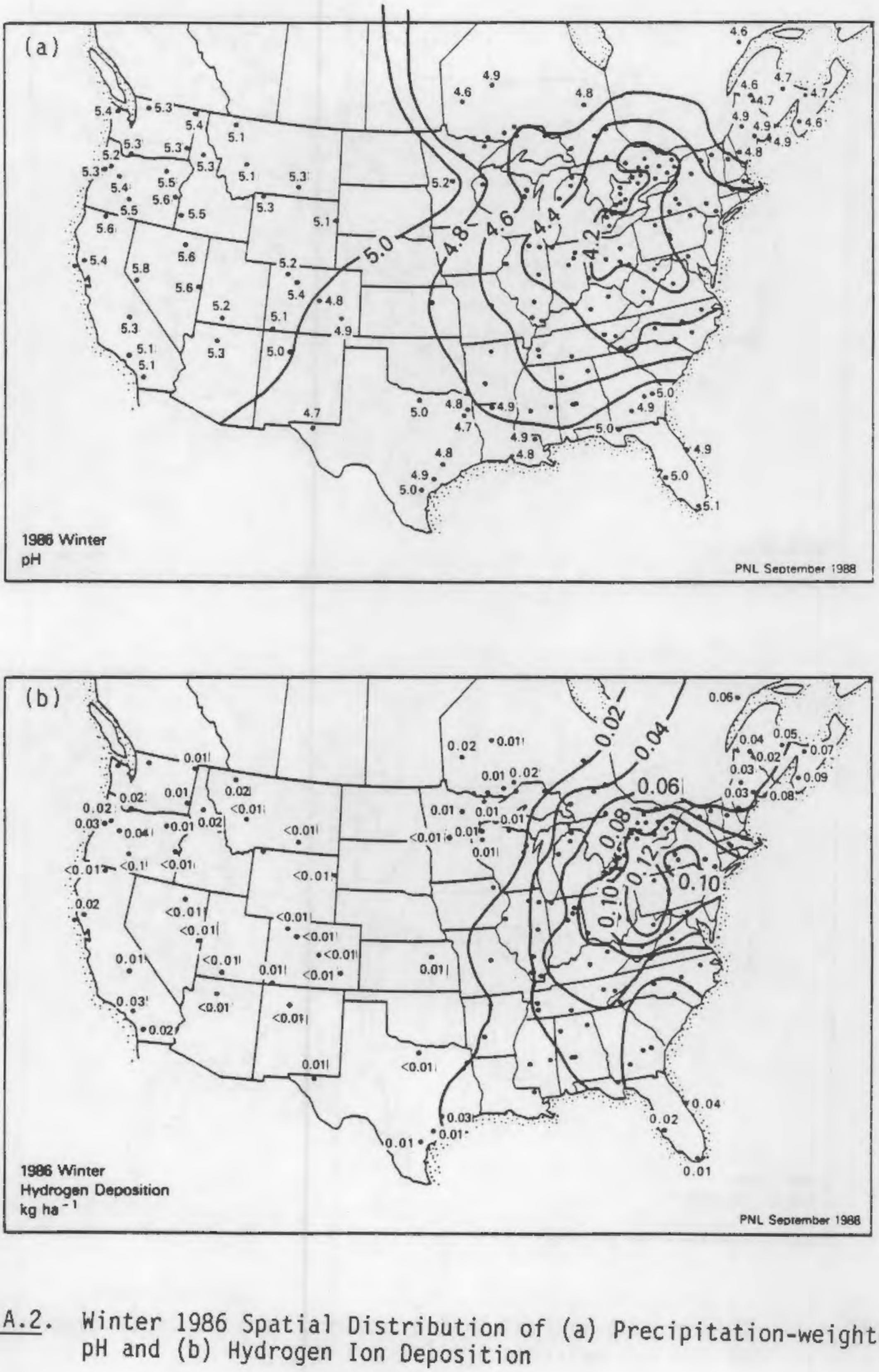


FIGURE A.2. Winter 1986 Spatial Distribution of (a) Precipitation-weighted pH and (b) Hydrogen Ion Deposition

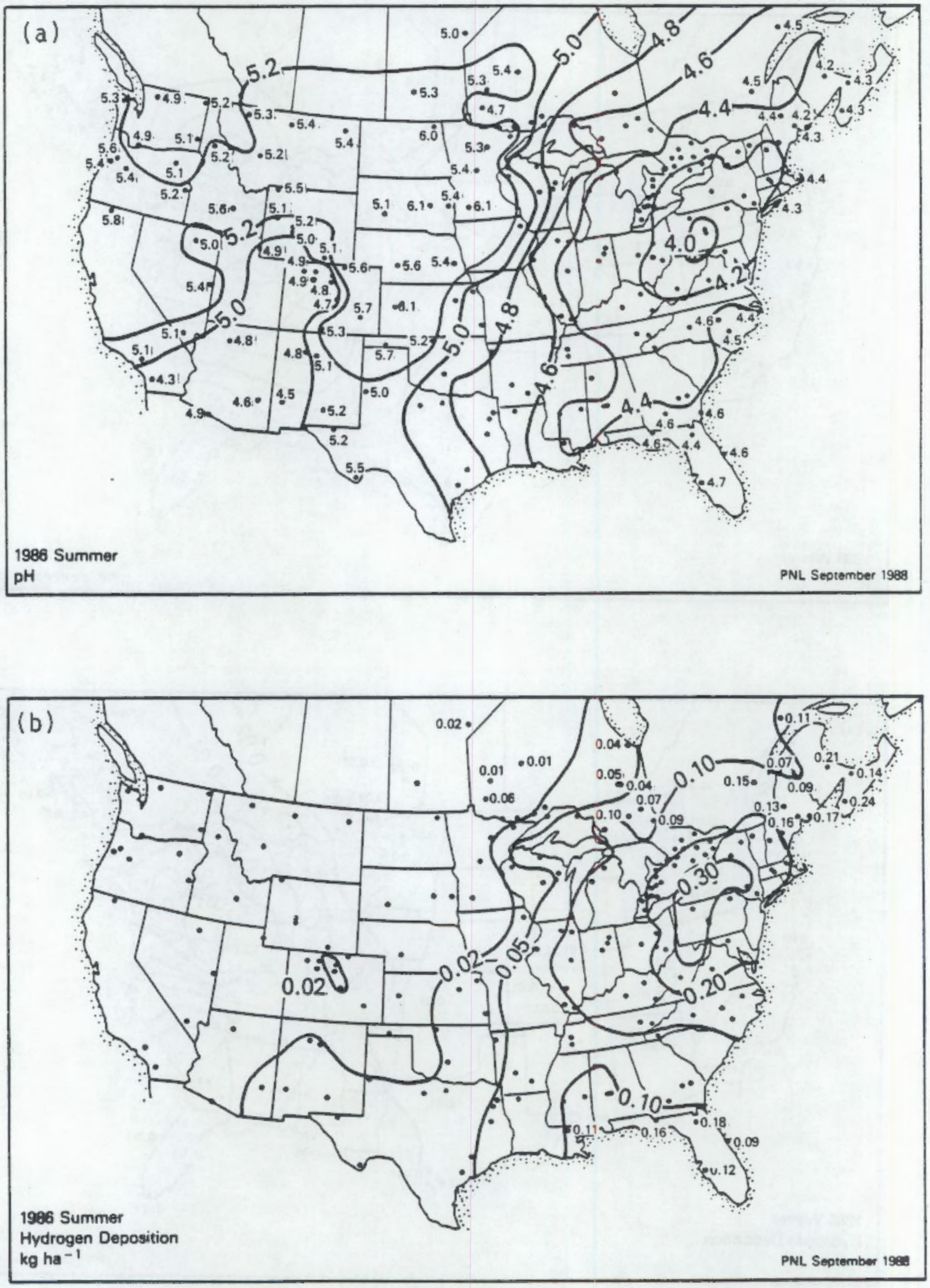
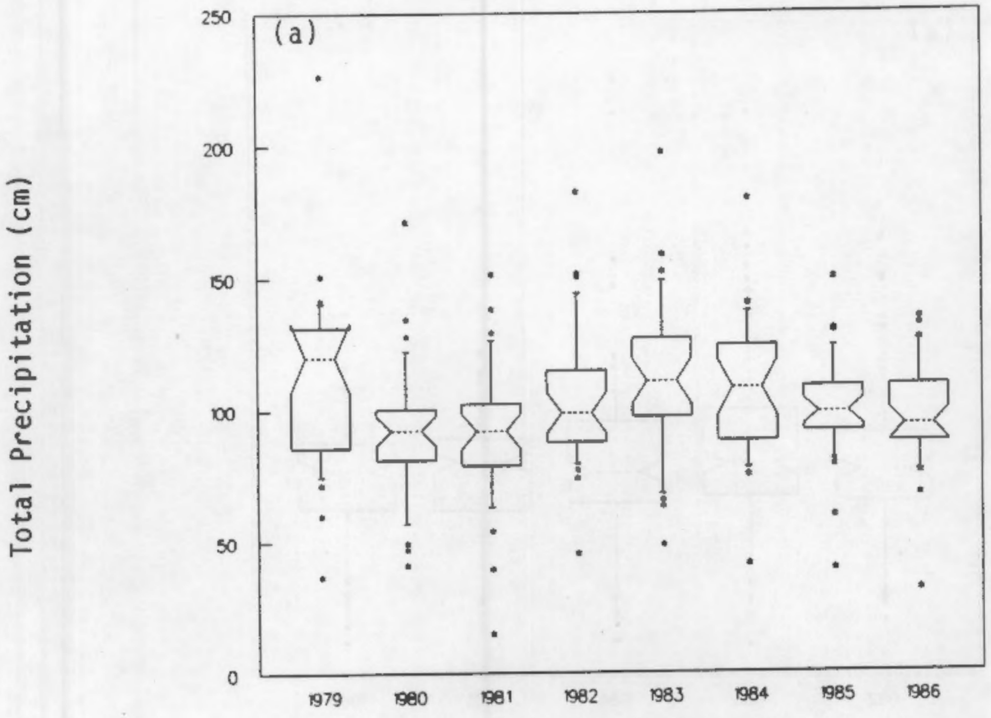


FIGURE A.3. Summer 1986 Spatial Distribution of (a) Precipitation-weighted pH and (b) Hydrogen Ion Deposition



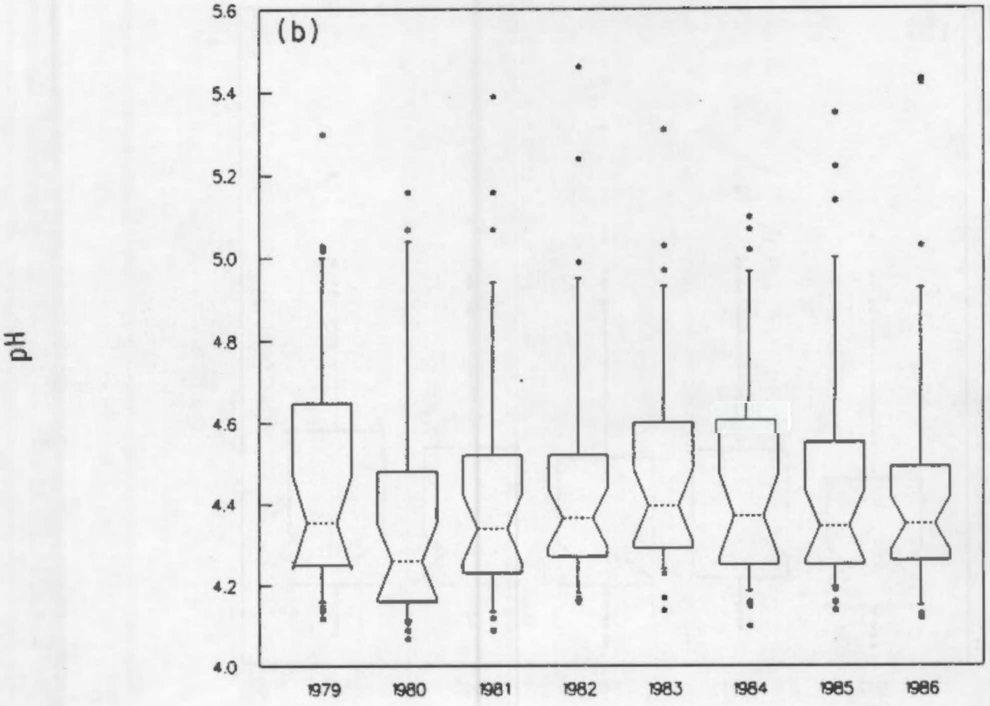


FIGURE A.4. North American Temporal Pattern of (a) Annual Precipitation, and (b) Annual Precipitation-weighted pH. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

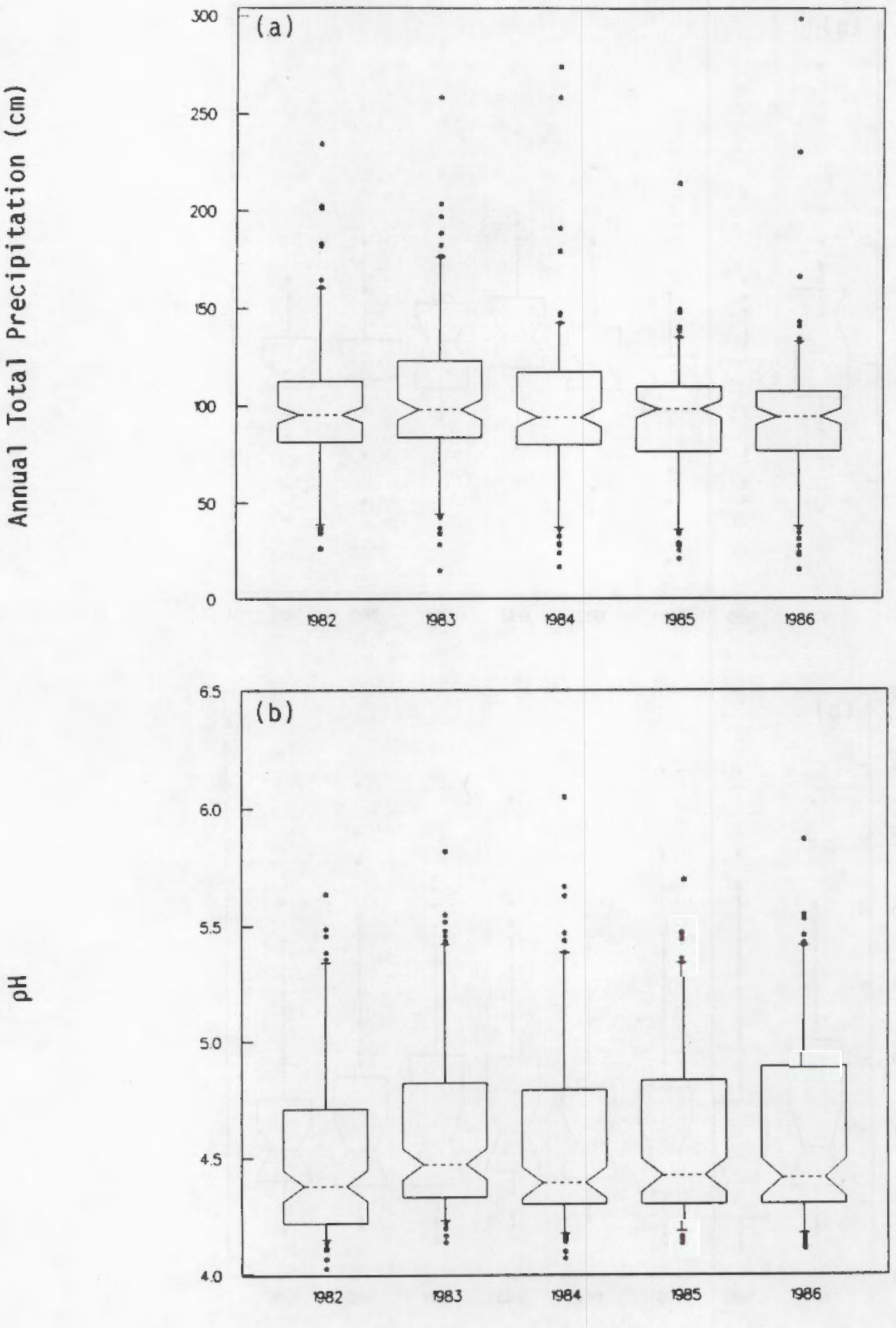


FIGURE A.5. North American Temporal Pattern of (a) Annual Precipitation, and (b) Annual Precipitation-weighted pH. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

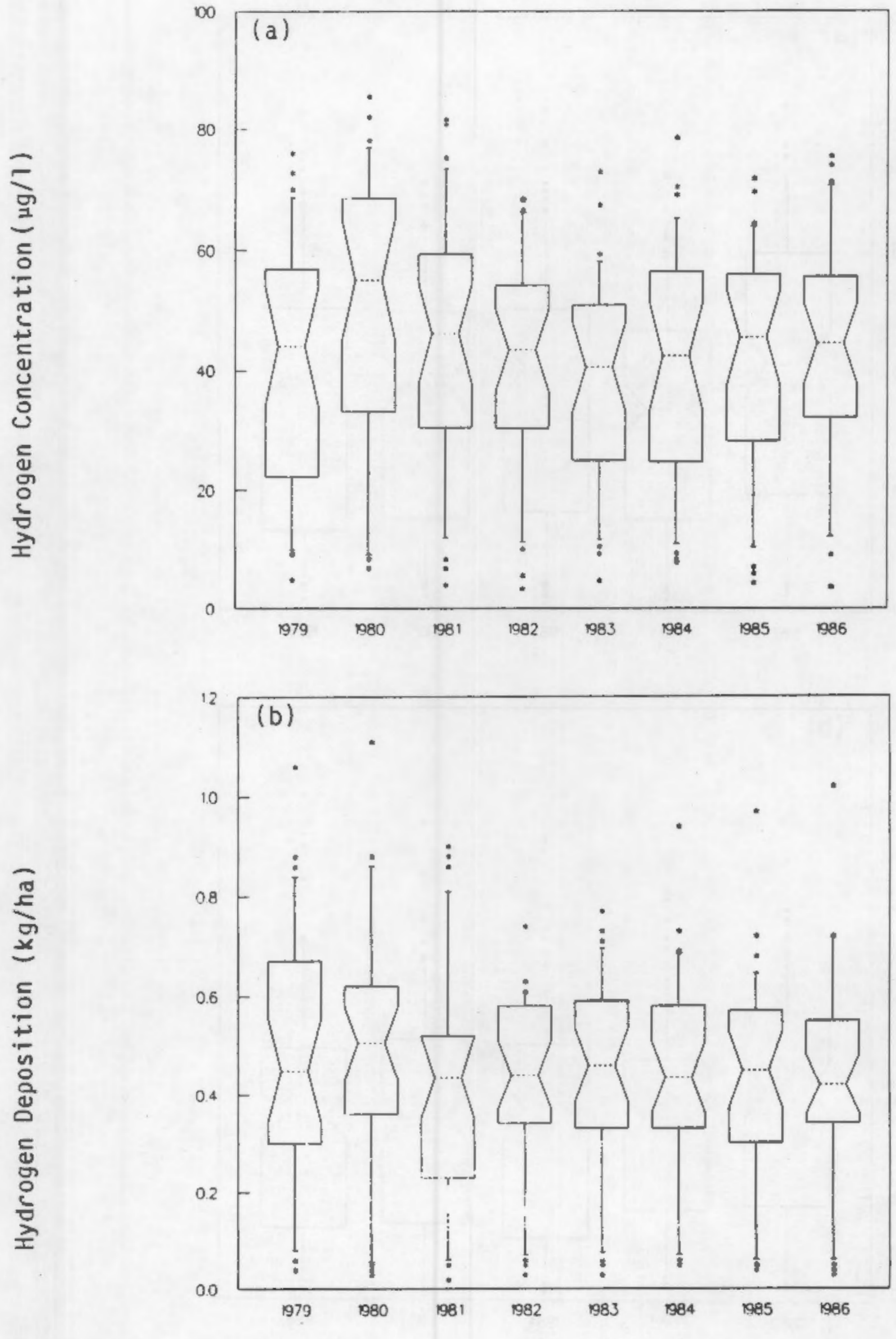


FIGURE A.6. North American Temporal Pattern of (a) Annual Precipitation-weighted Hydrogen Ion Concentration, and (b) Annual Hydrogen Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

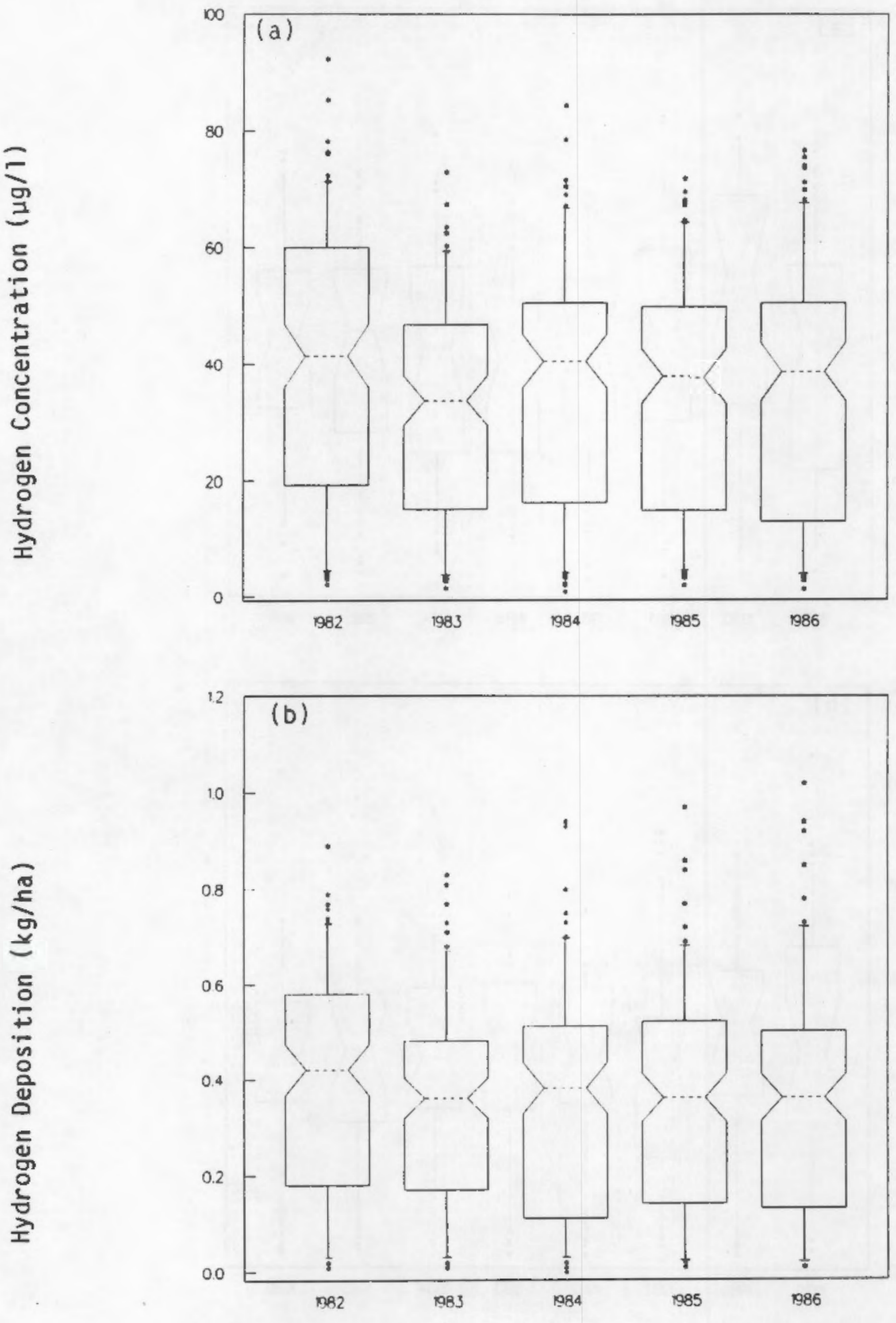


FIGURE A.7. North American Temporal Pattern of (a) Annual Precipitation-weighted Hydrogen Ion Concentration, and (b) Annual Hydrogen Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Total Precipitation (cm) Trend Estimate

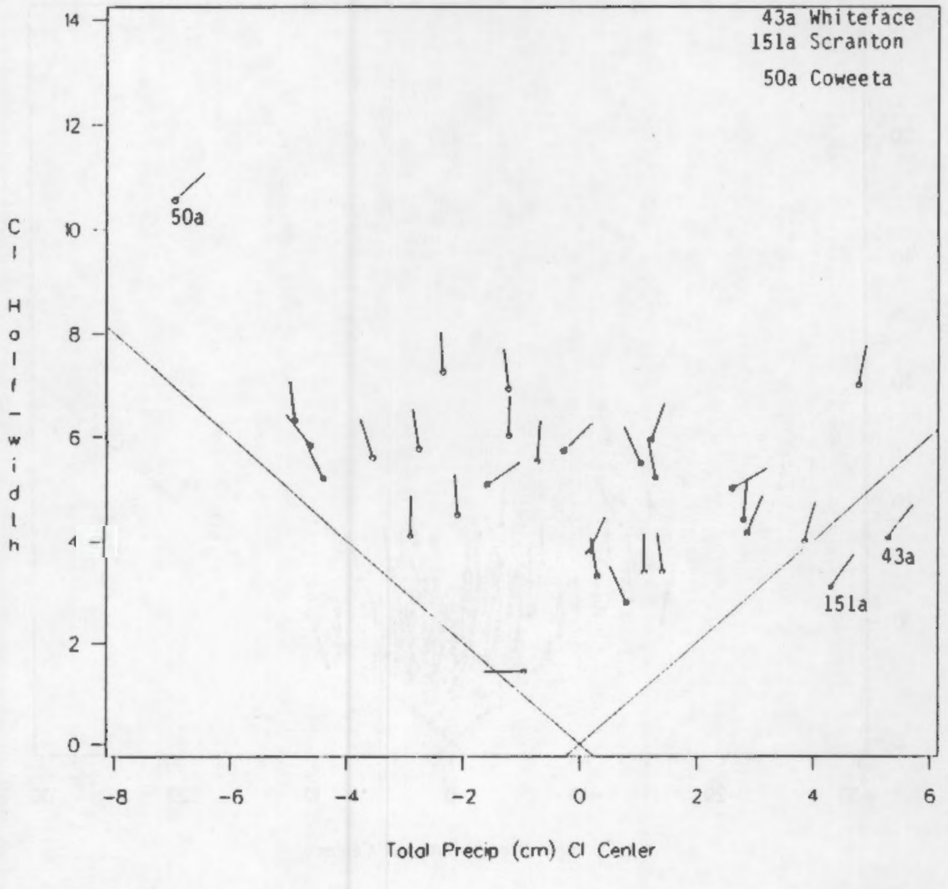
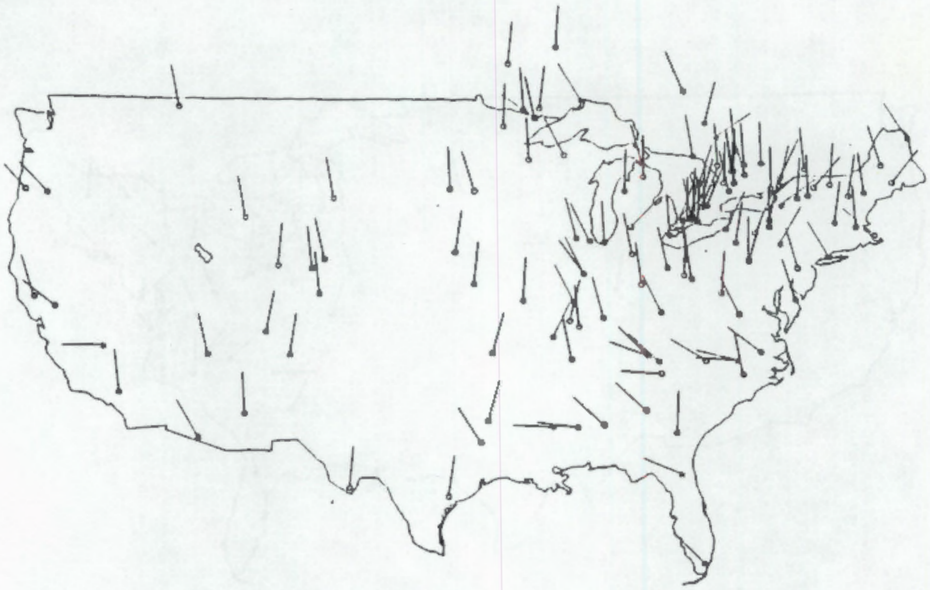


FIGURE A.8. Total Precipitation Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Precipitation. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Total Precipitation (cm) Trend Estimate

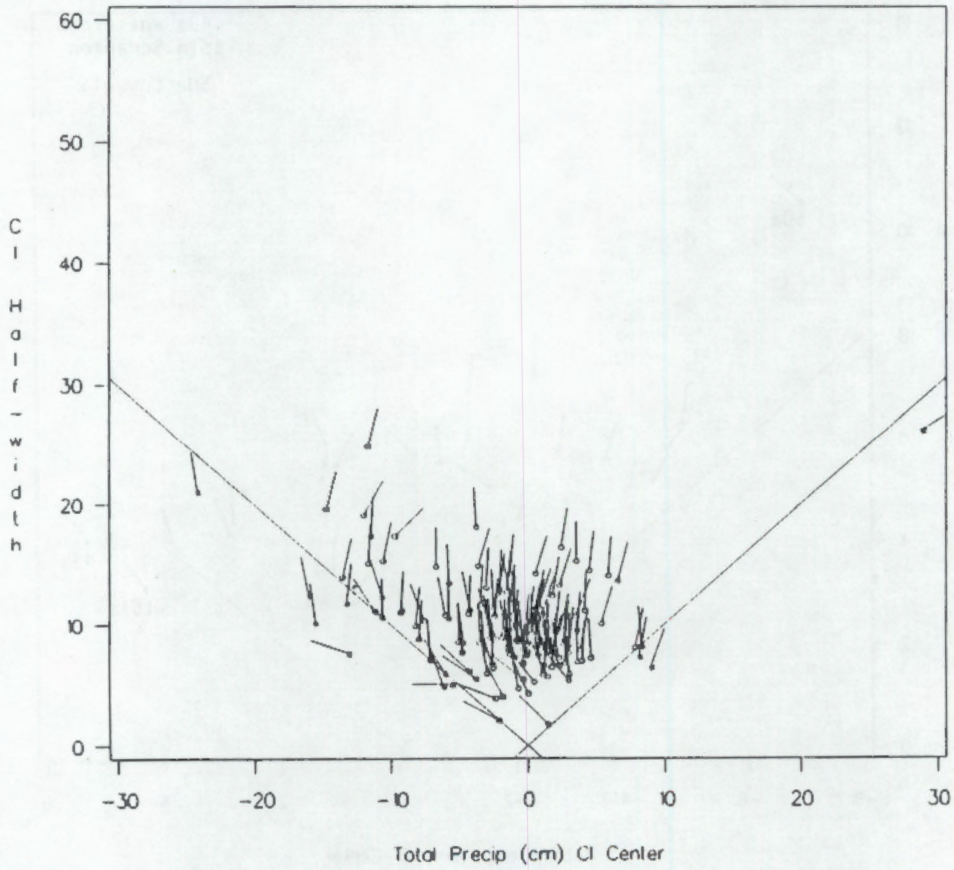


FIGURE A.9. Total Precipitation Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Precipitation. Solid Symbol Indicates Significant Trend ($p < 0.05$).

pH Trend Estimate

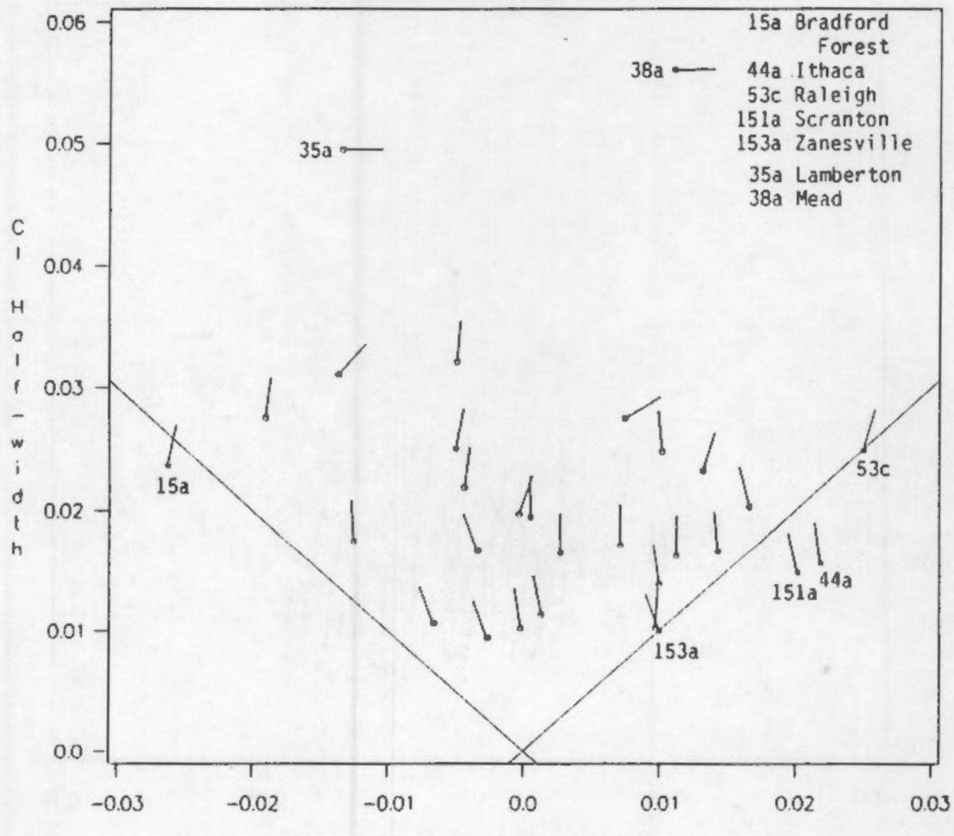


FIGURE A.10. pH Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual pH. Solid Symbol Indicates Significant Trend ($p < 0.05$).

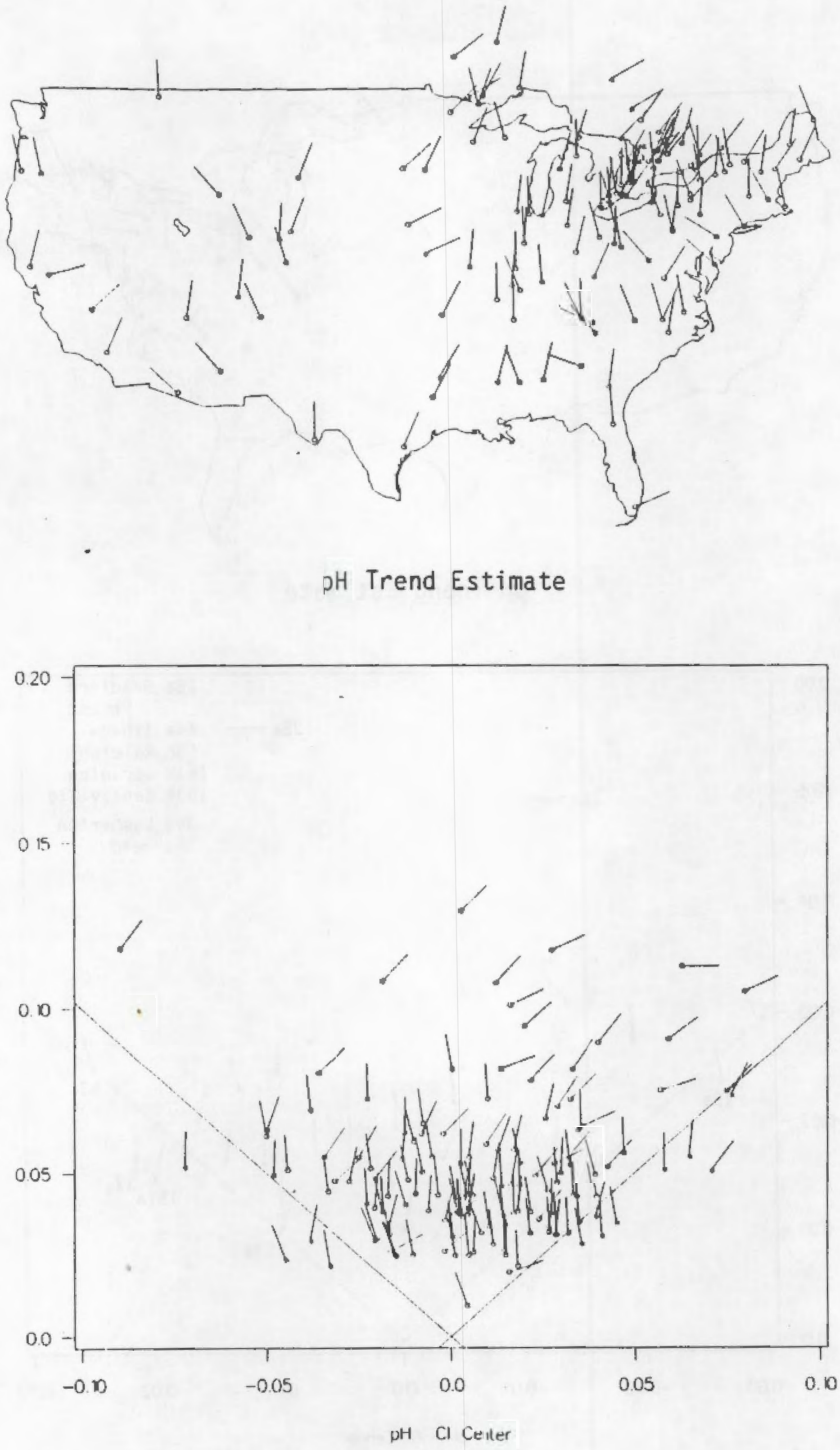


FIGURE A.11. pH Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual pH. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Hydrogen Concentration Trend Estimate (microeq/l)

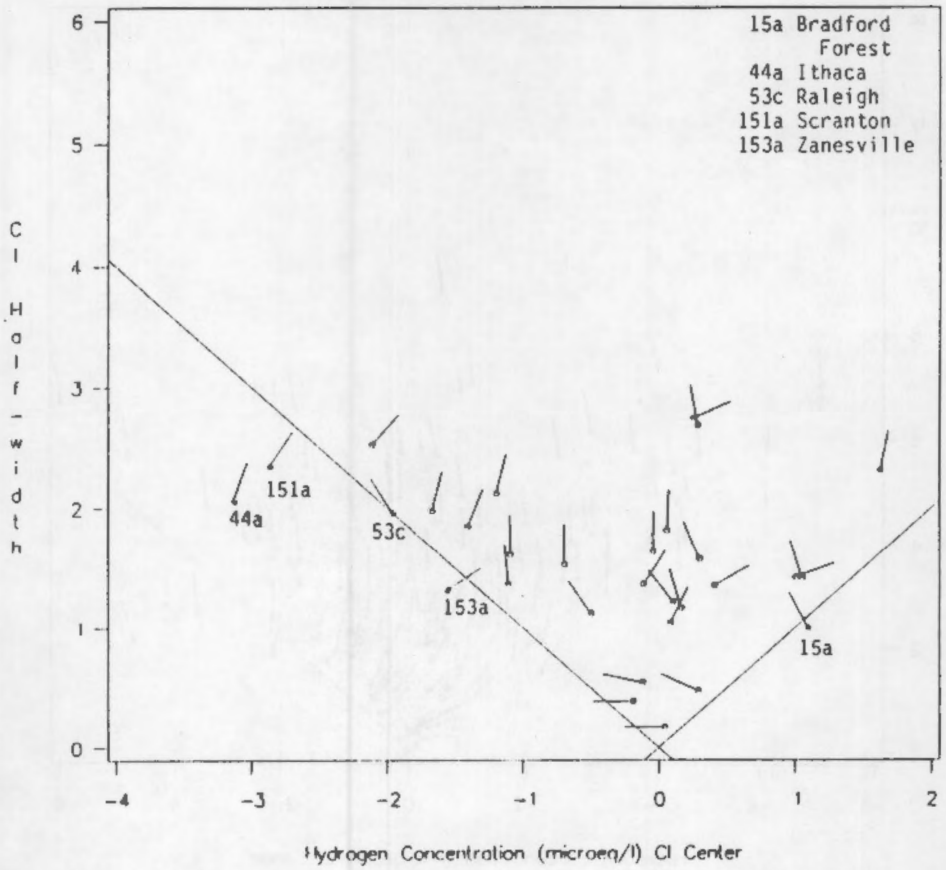
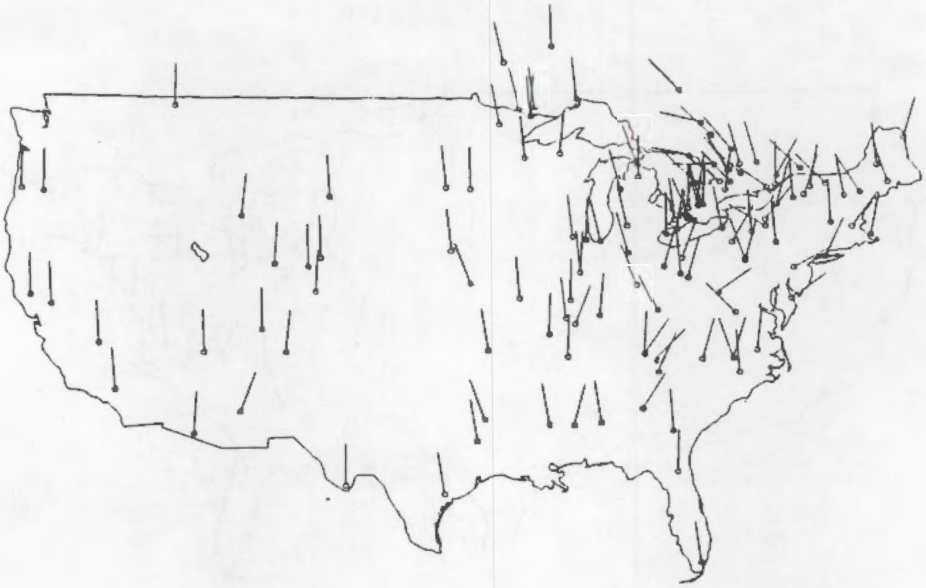


FIGURE A.12. Hydrogen Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Hydrogen Concentration Trend Estimate (microeq/l)

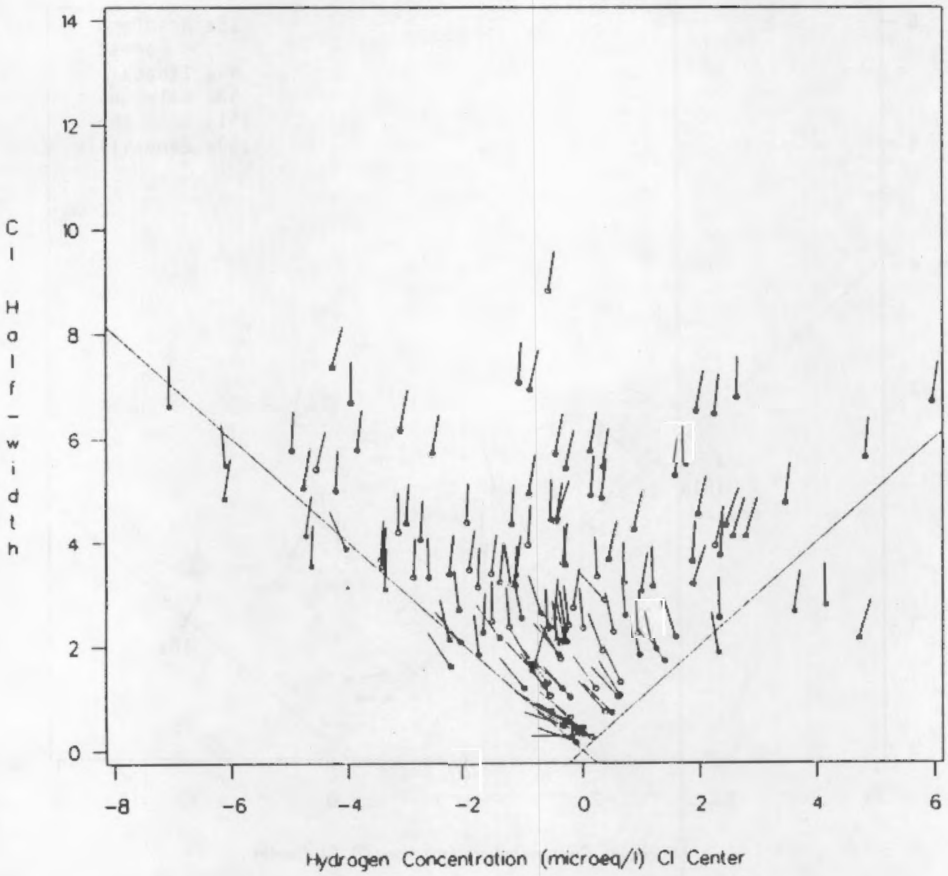
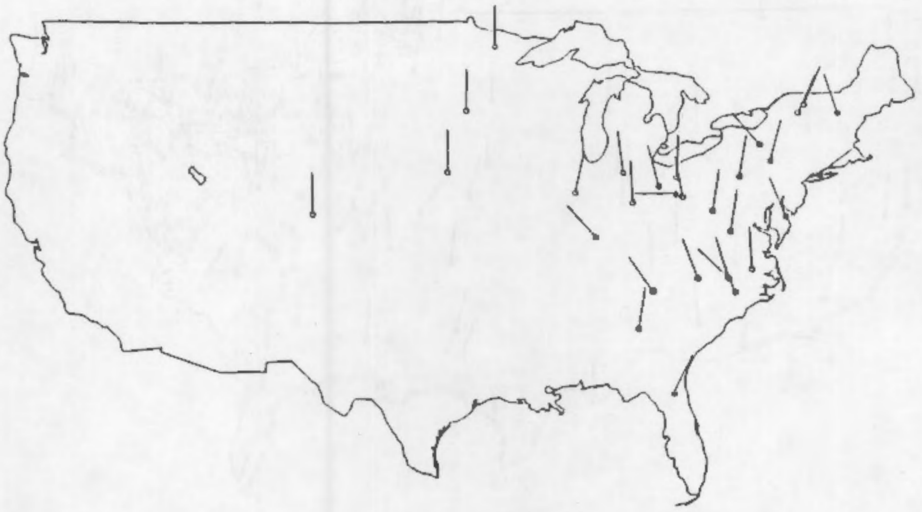


FIGURE A.13. Hydrogen Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Hydrogen Deposition Trend Estimate (meq/sq m)

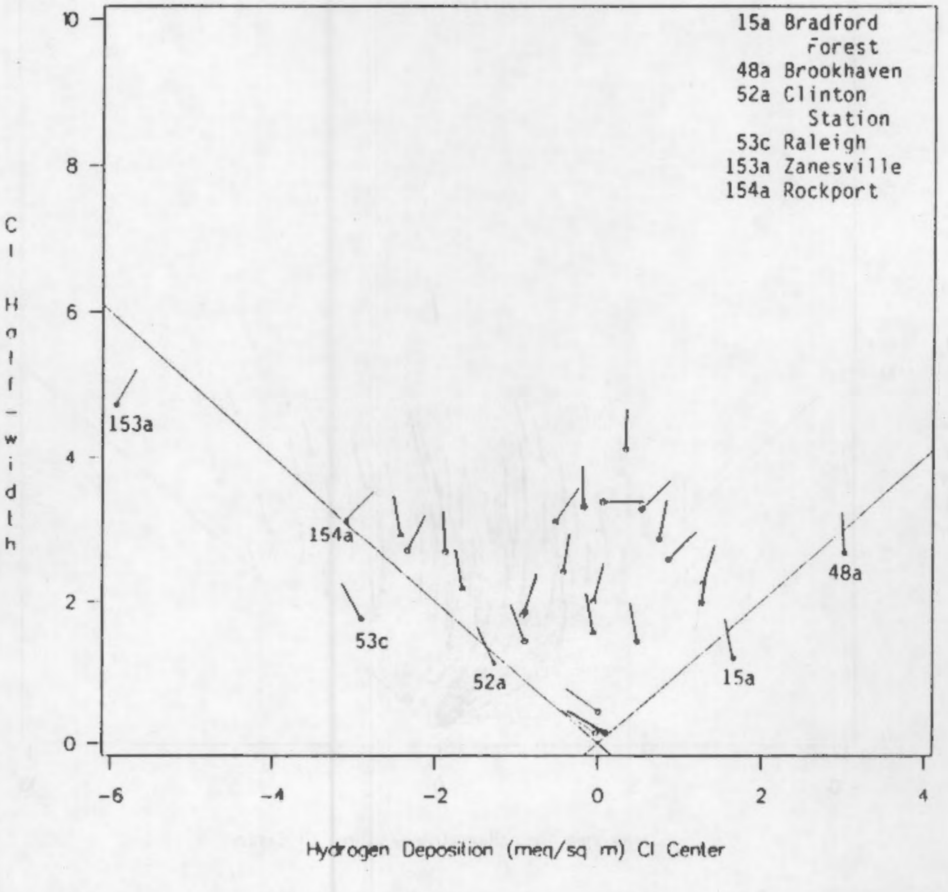


FIGURE A.14. Hydrogen Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Hydrogen Deposition Trend Estimate (meq/sq m)

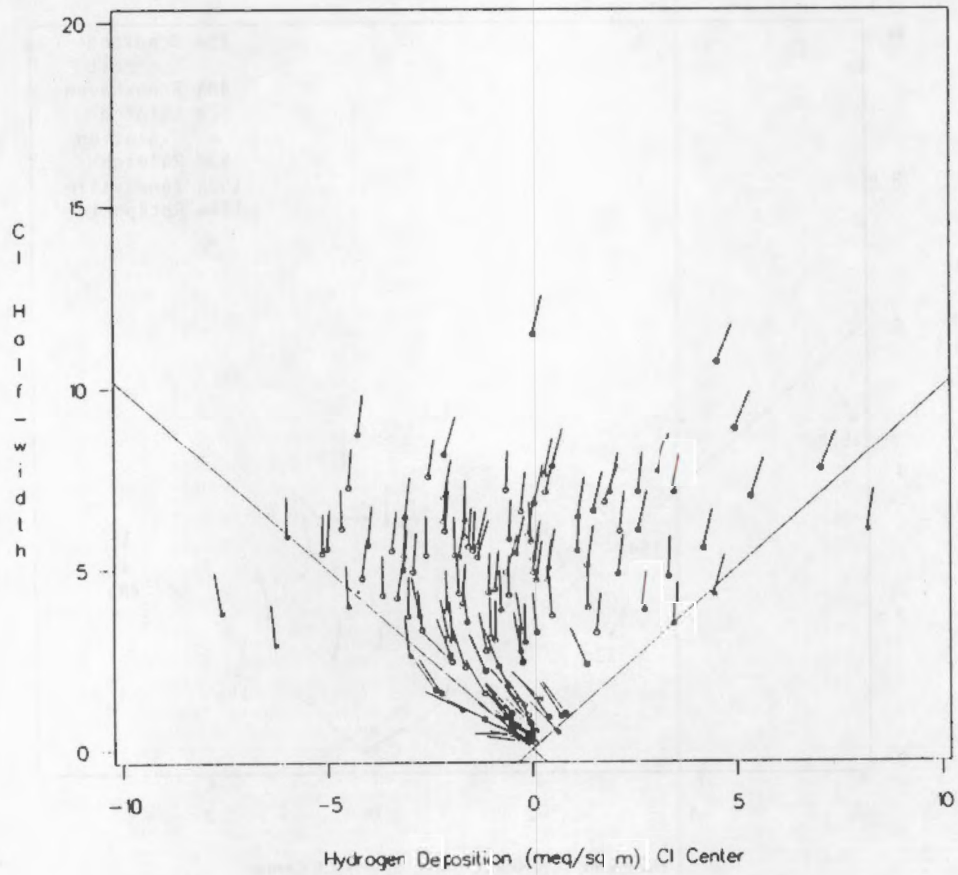


FIGURE A.15. Hydrogen Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

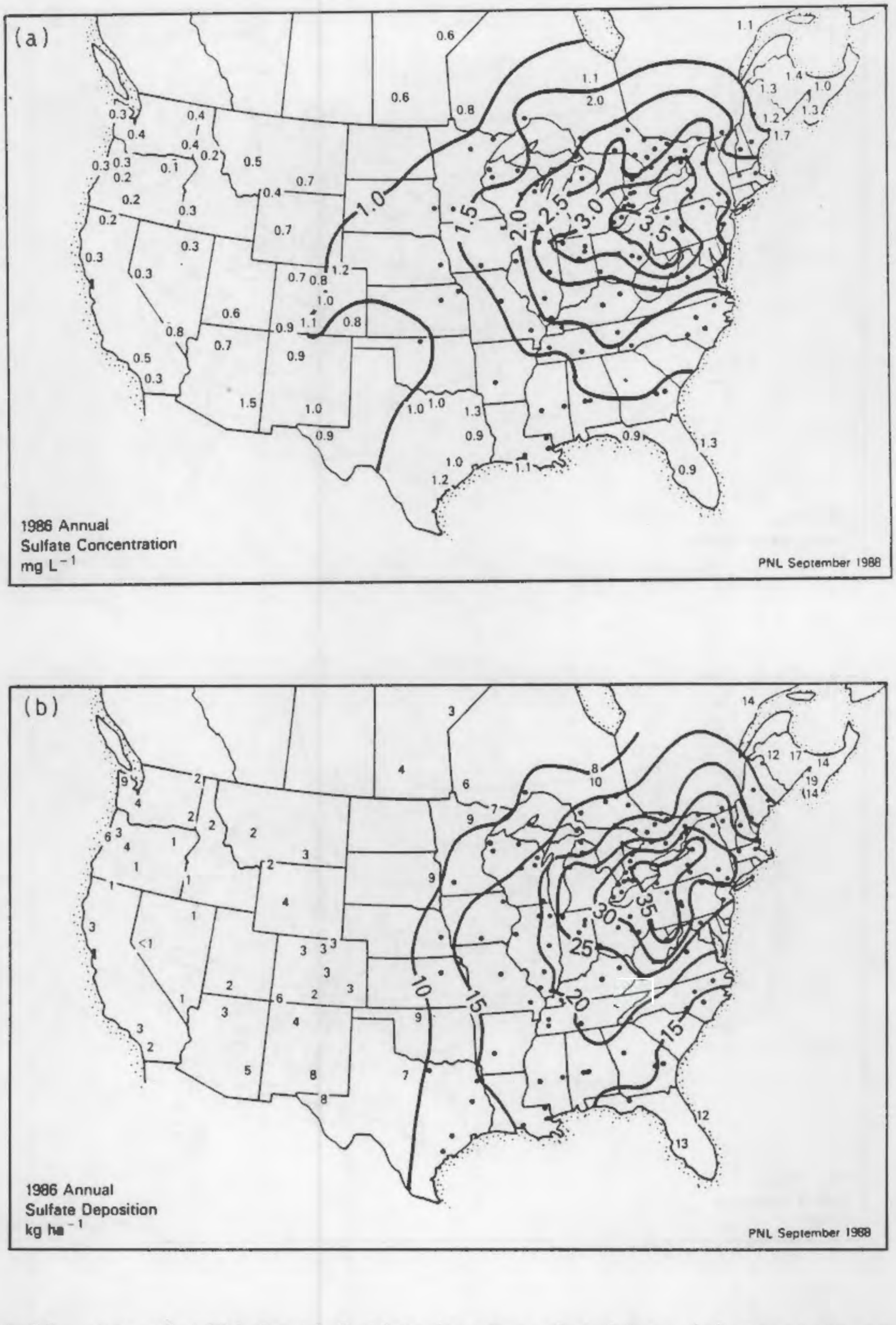
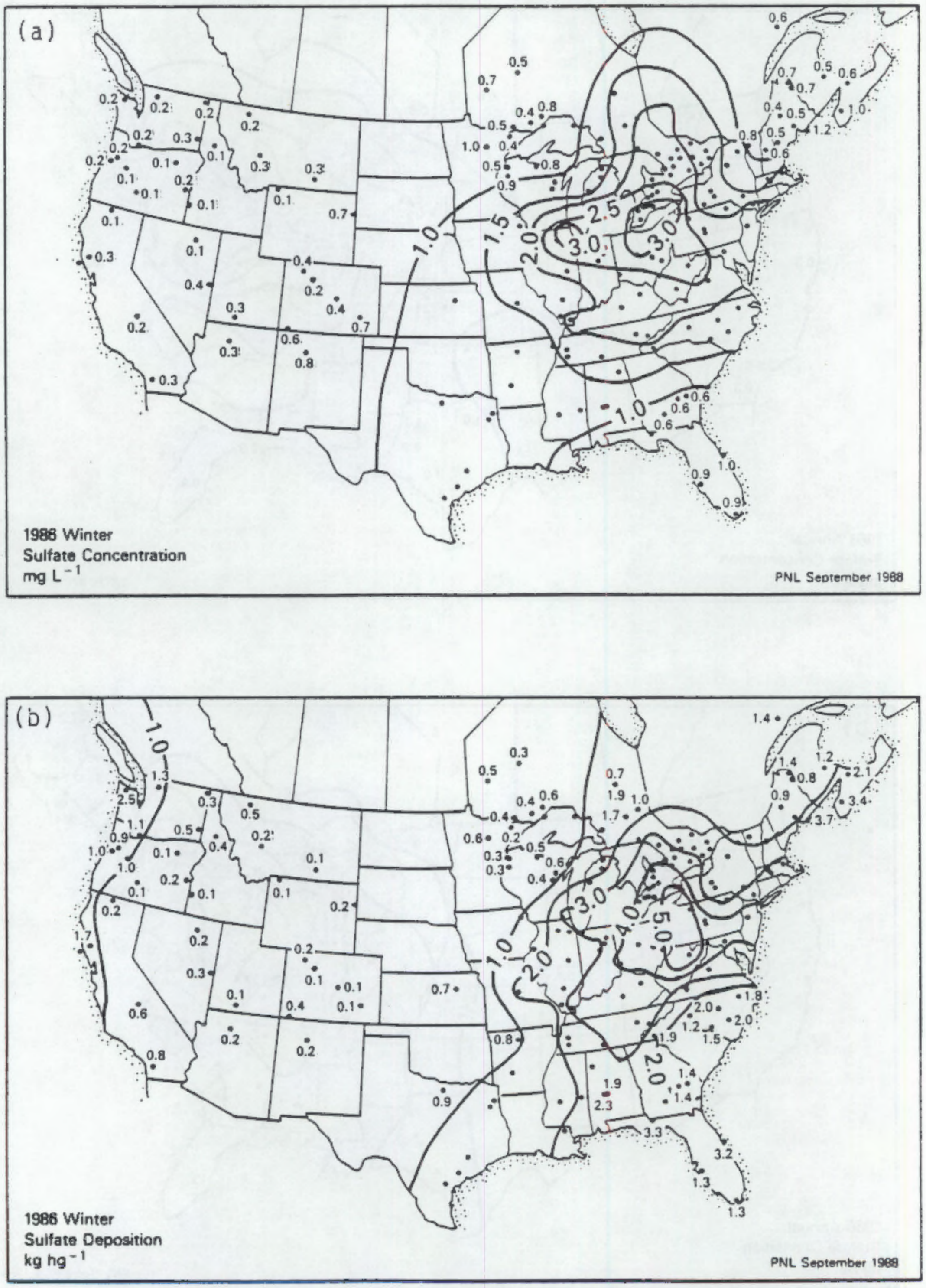


FIGURE A.16. Annual 1986 Spatial Distribution of Sulfate (a) Precipitation-weighted Concentration and (b) Deposition



1986 Winter
Sulfate Concentration
mg L^{-1}

PNL September 1988

1986 Winter
Sulfate Deposition
kg hg^{-1}

PNL September 1988

FIGURE A.17. Winter 1986 Spatial Distribution of Sulfate (a) Precipitation-weighted Concentration and (b) Deposition

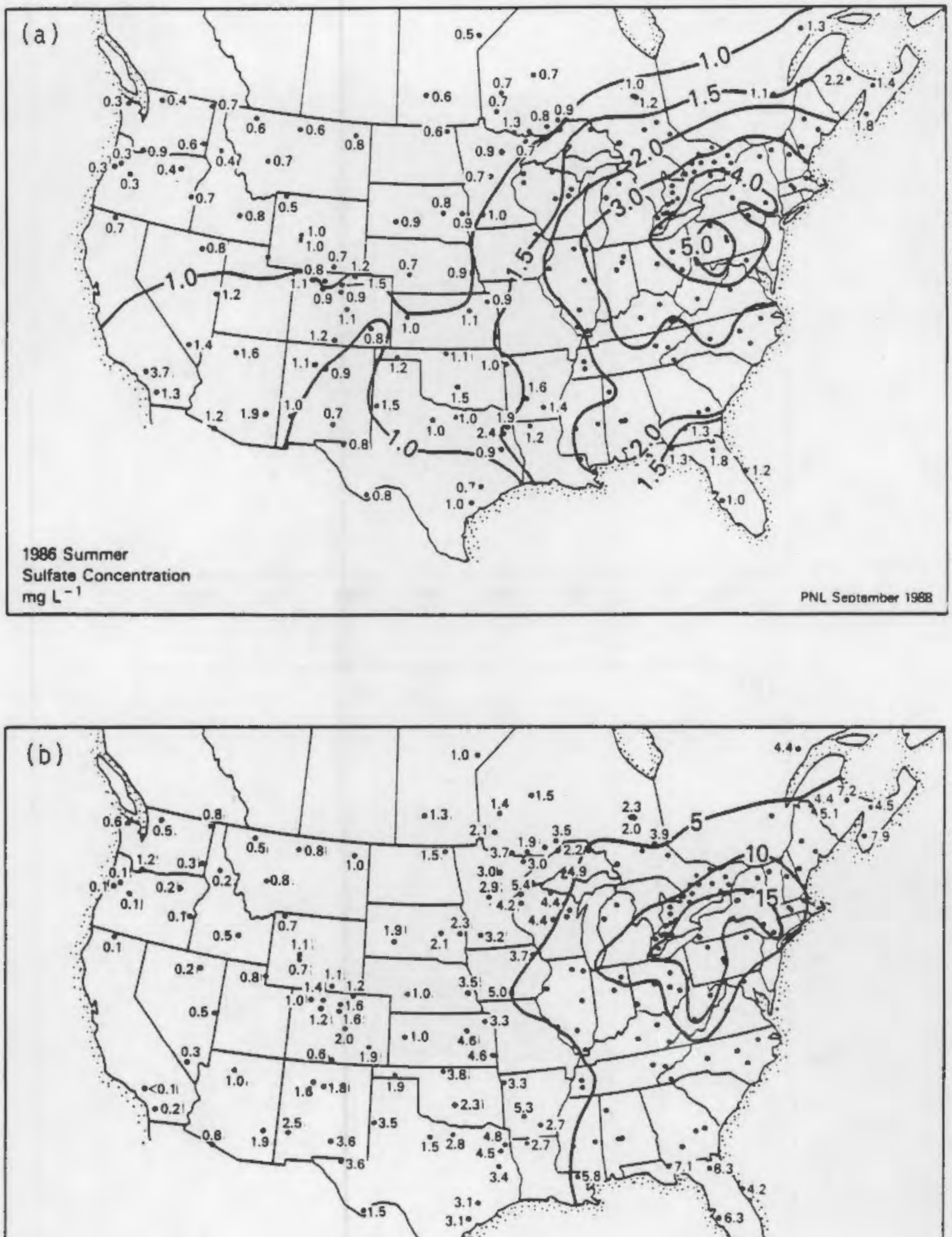


FIGURE A.18. Summer 1986 Spatial Distribution of Sulfate (a) Precipitation-weighted Concentration and (b) Deposition

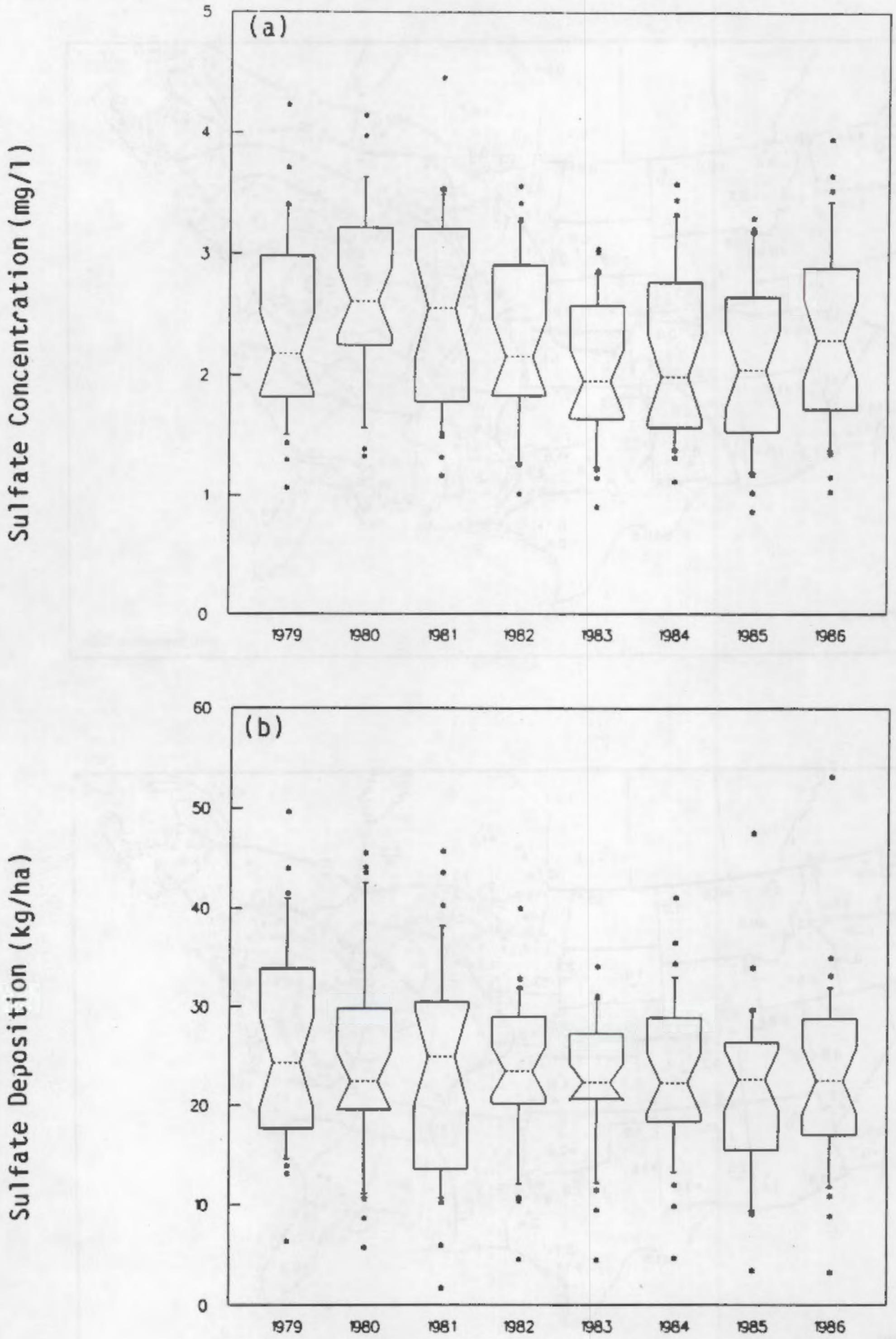


FIGURE A.19. North American Temporal Pattern of (a) Annual Precipitation-weighted Sulfate Ion Concentration, and (b) Annual Sulfate Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

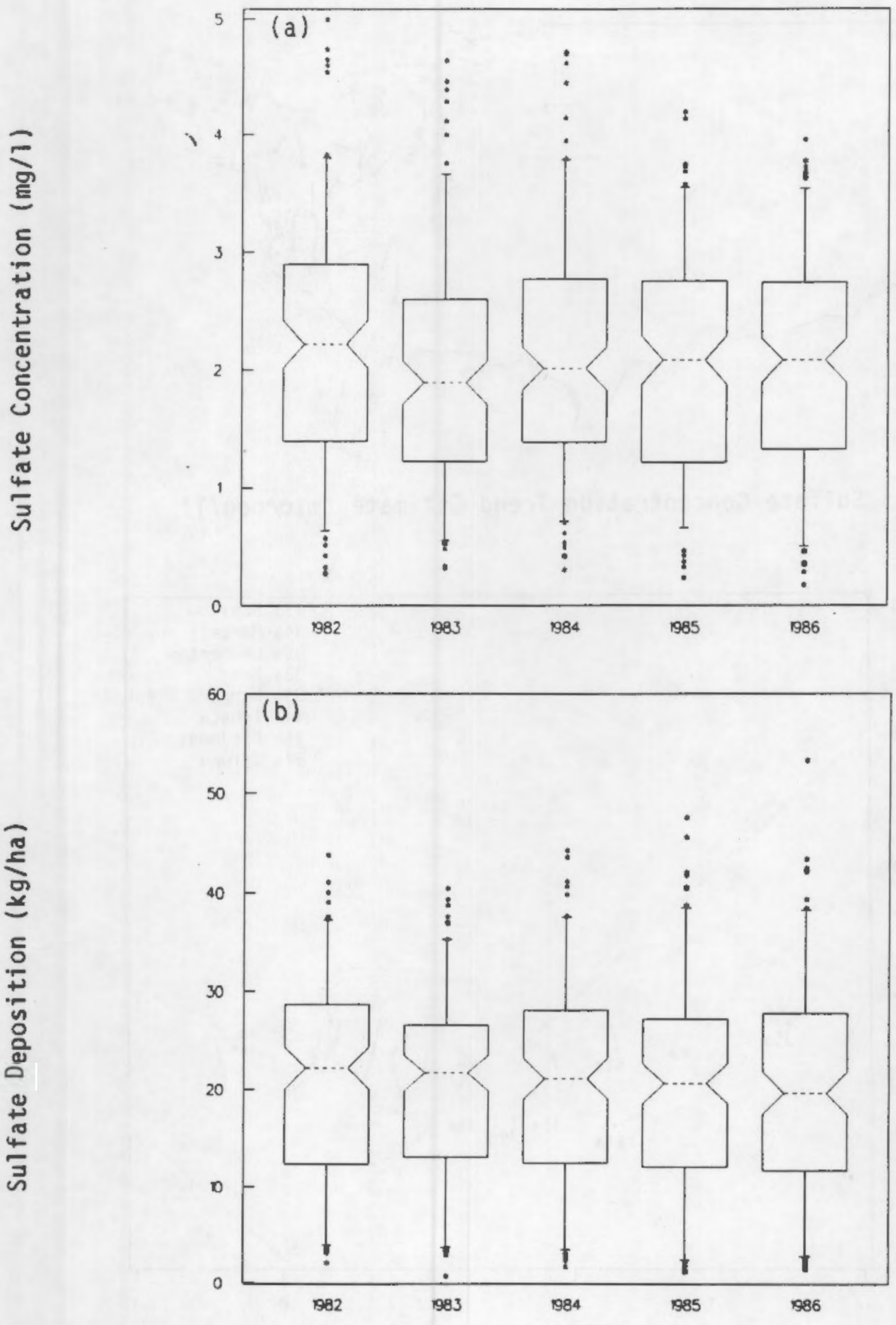


FIGURE A.20. North American Temporal Pattern of (a) Annual Precipitation-weighted Sulfate Ion Concentration, and (b) Annual Sulfate Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Sulfate Concentration Trend Estimate (microeq/l)

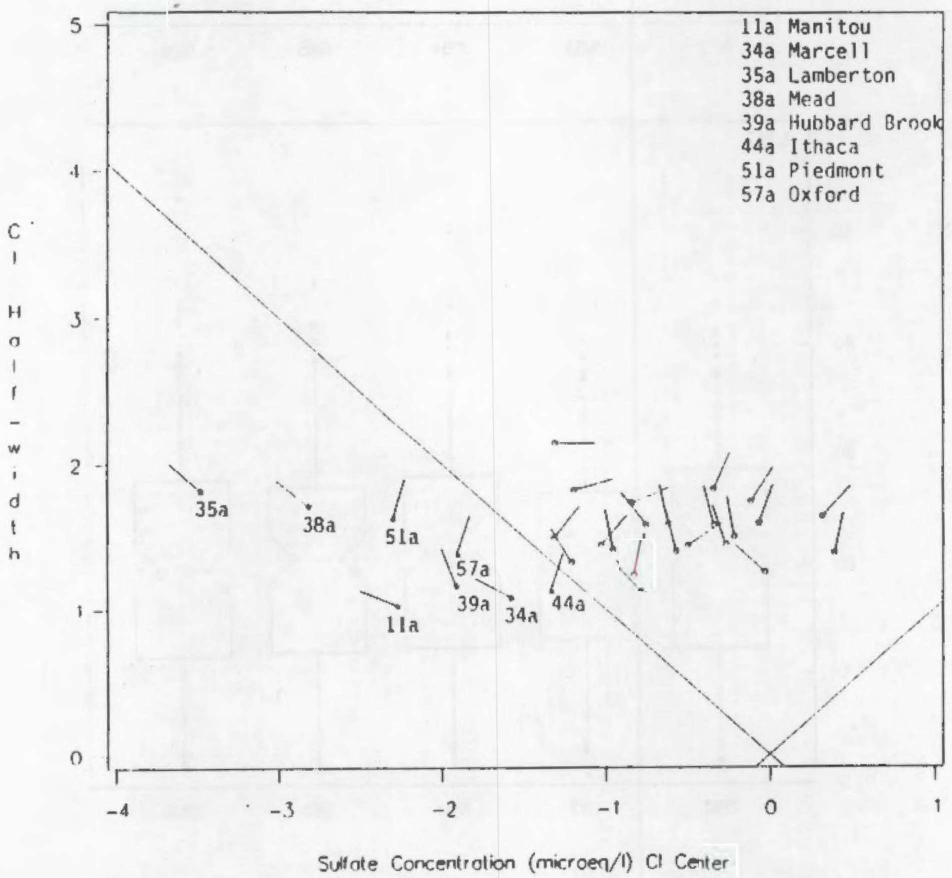
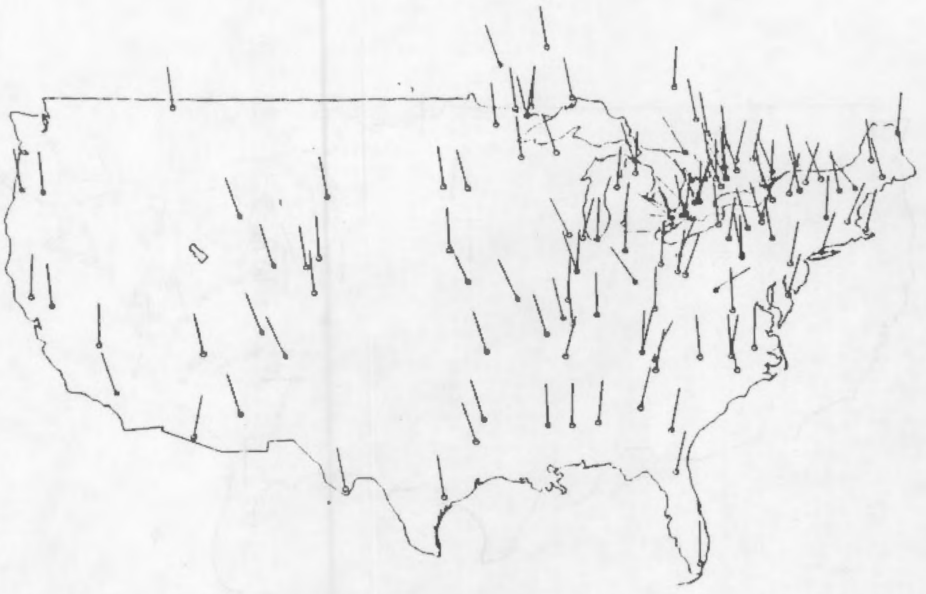


FIGURE A.21. Sulfate Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Sulfate Concentration Trend Estimate (microeq/l)

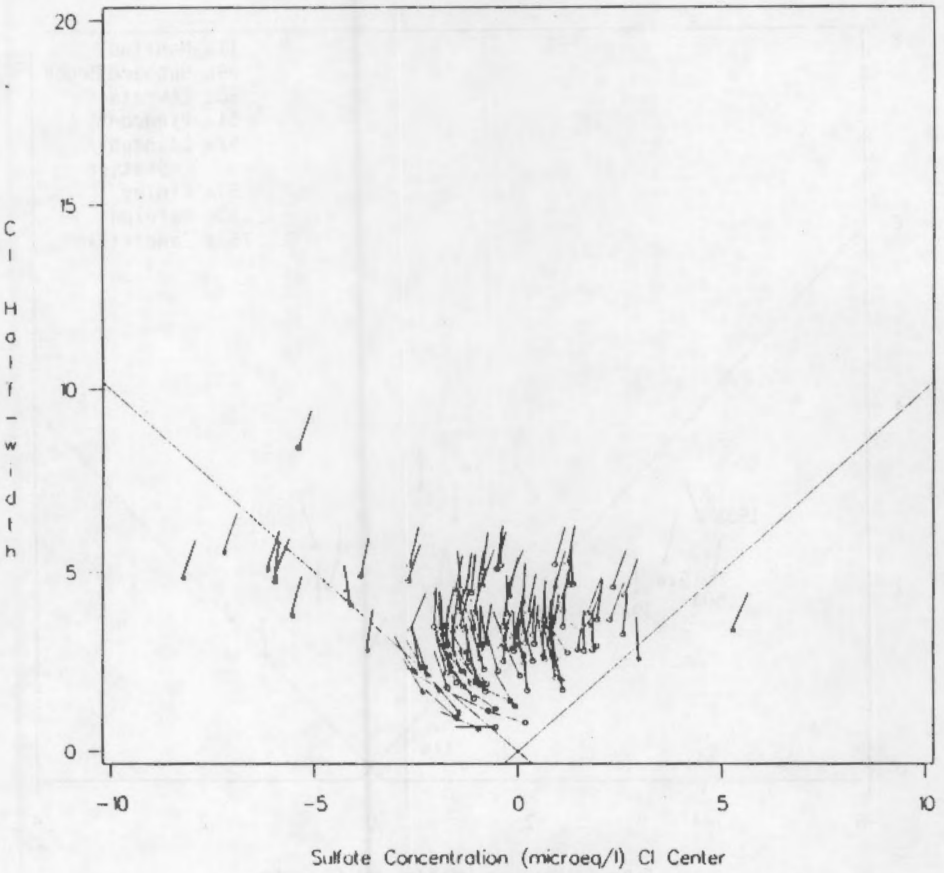


FIGURE A.22. Sulfate Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Sulfate Deposition Trend Estimate (meq/sq m)

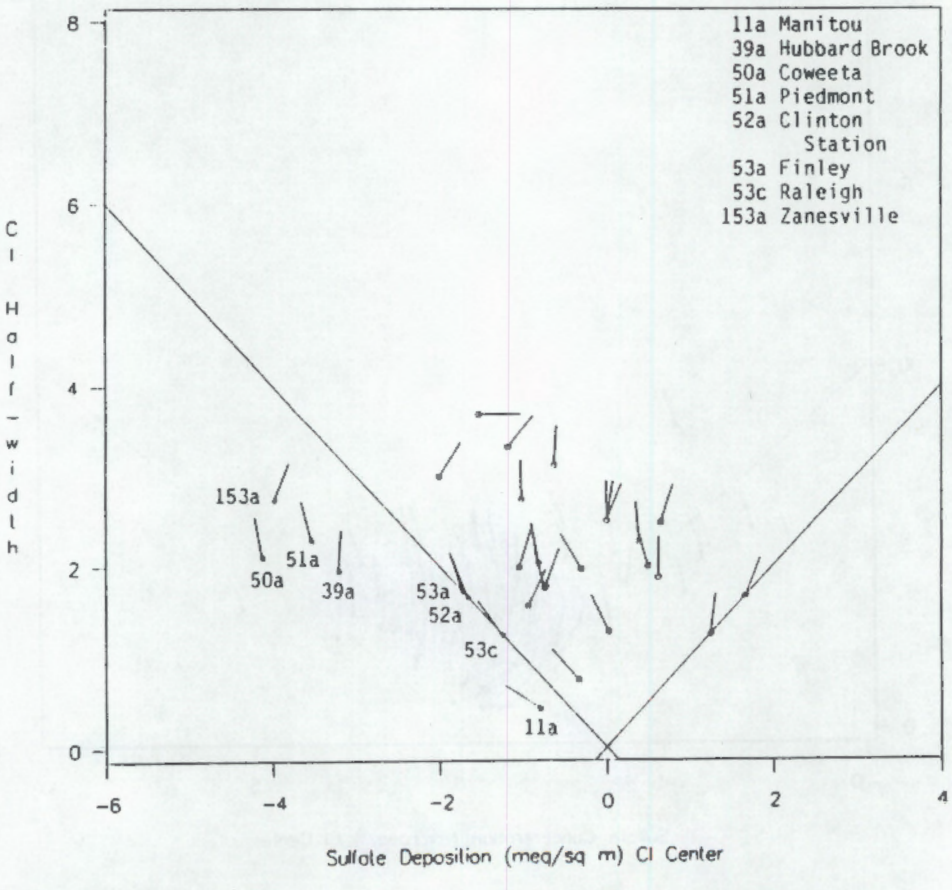
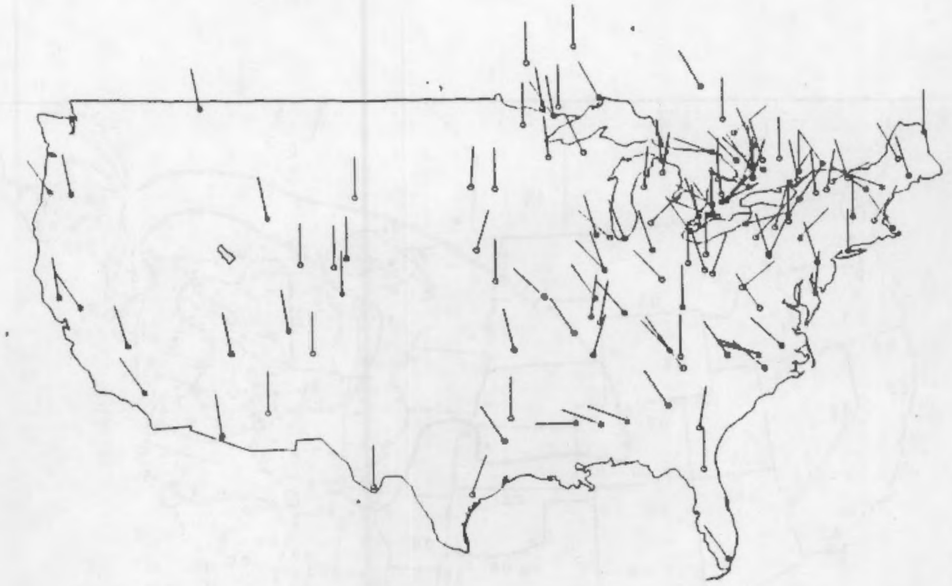


FIGURE A.23. Sulfate Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Sulfate Deposition Trend Estimate (meq/sq m)

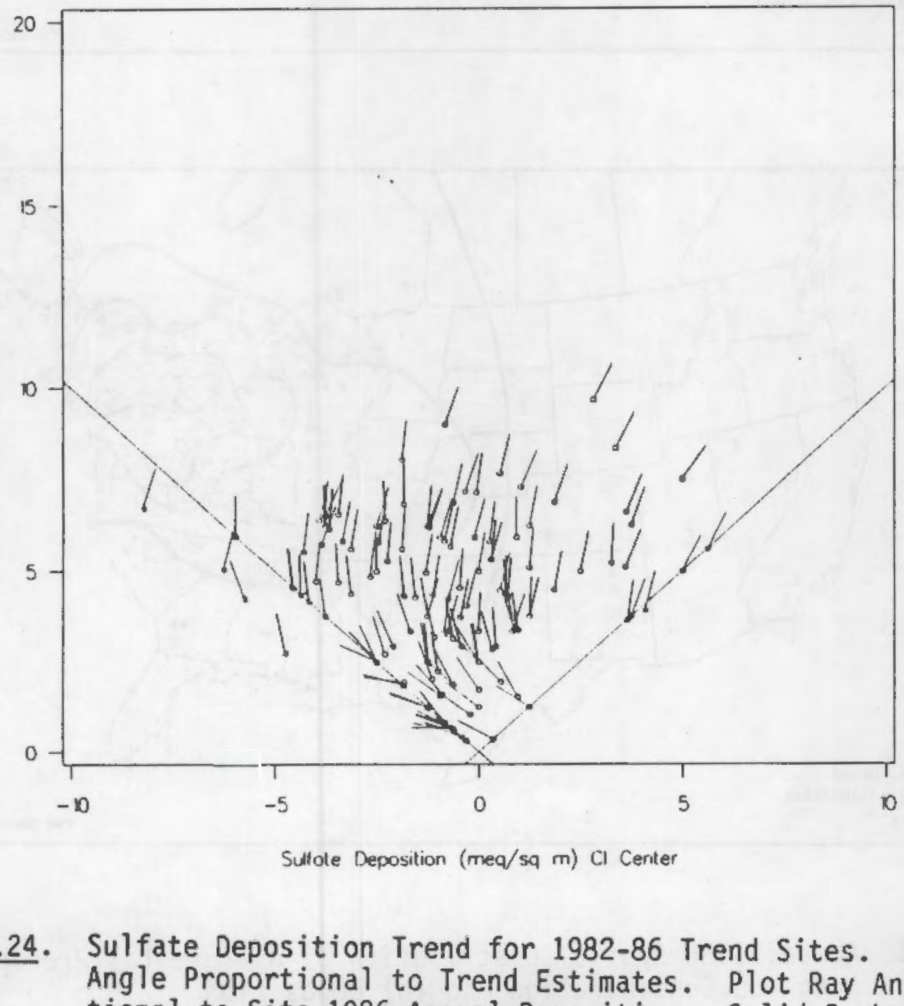
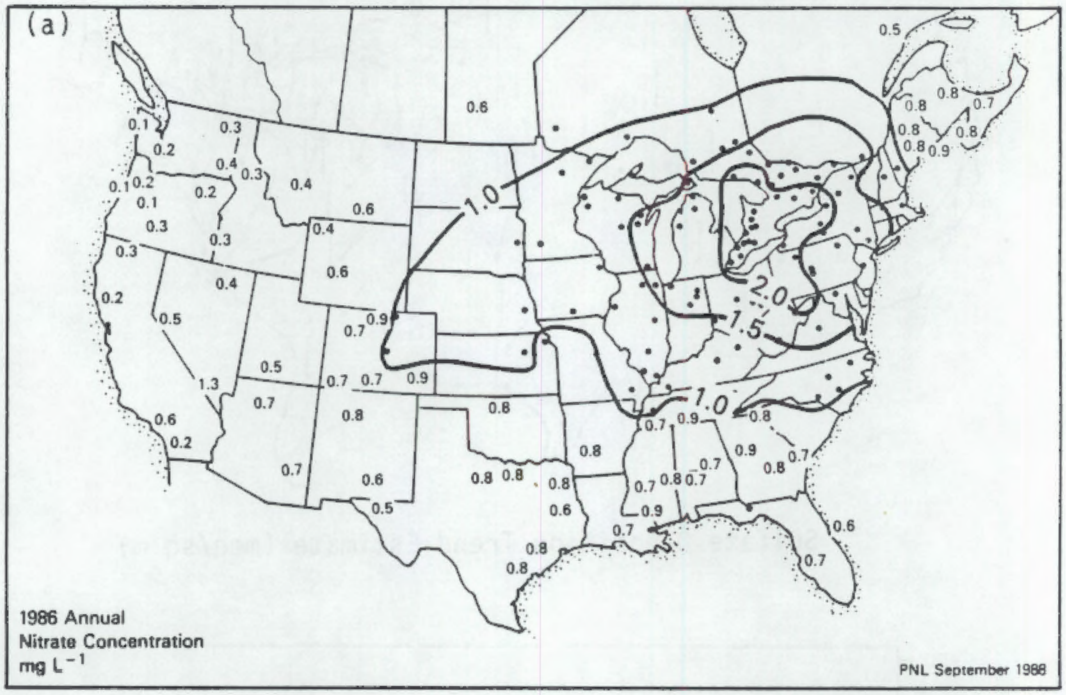


FIGURE A.24. Sulfate Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



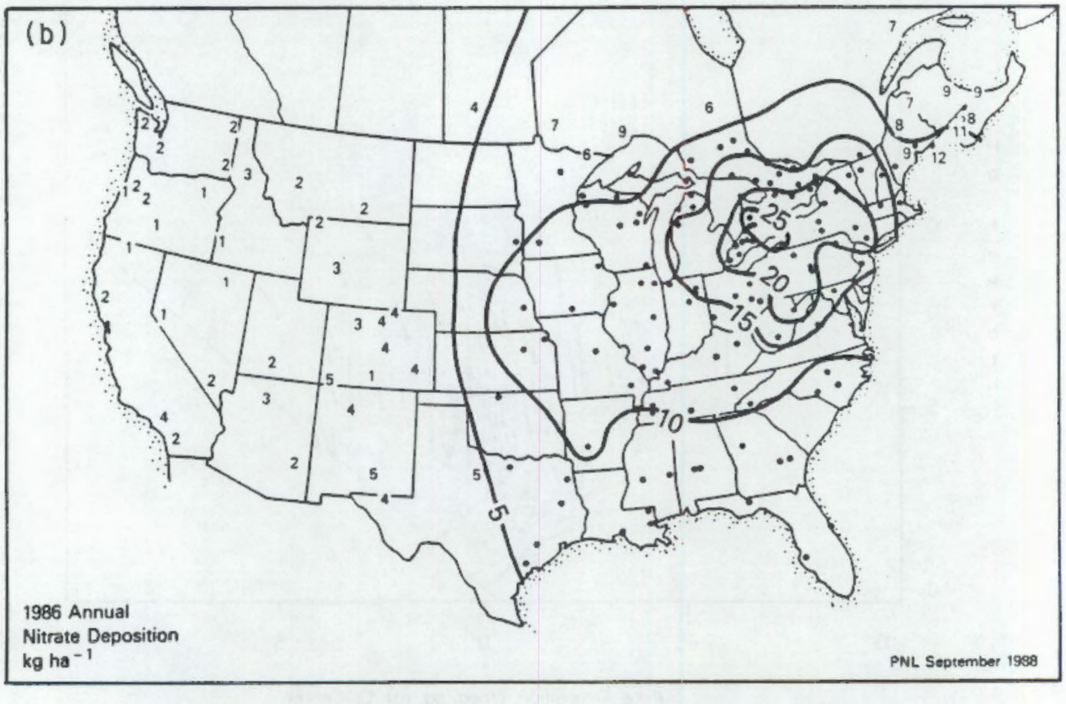


FIGURE A.25. Annual 1986 Spatial Distribution of Nitrate (a) Precipitation-weighted Concentration and (b) Deposition

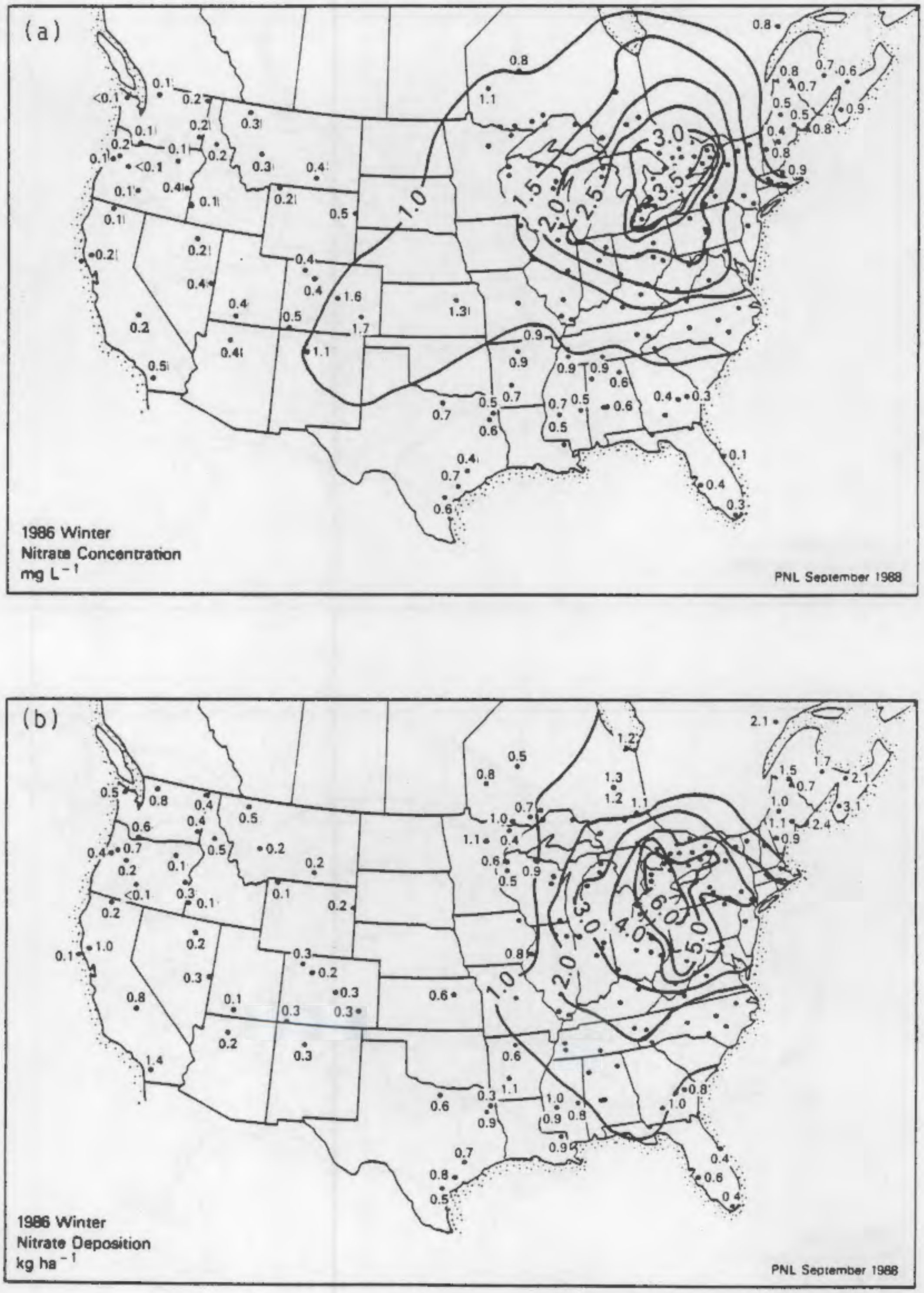


FIGURE A.26. Winter 1986 Spatial Distribution of Nitrate (a) Precipitation-weighted Concentration and (b) Deposition

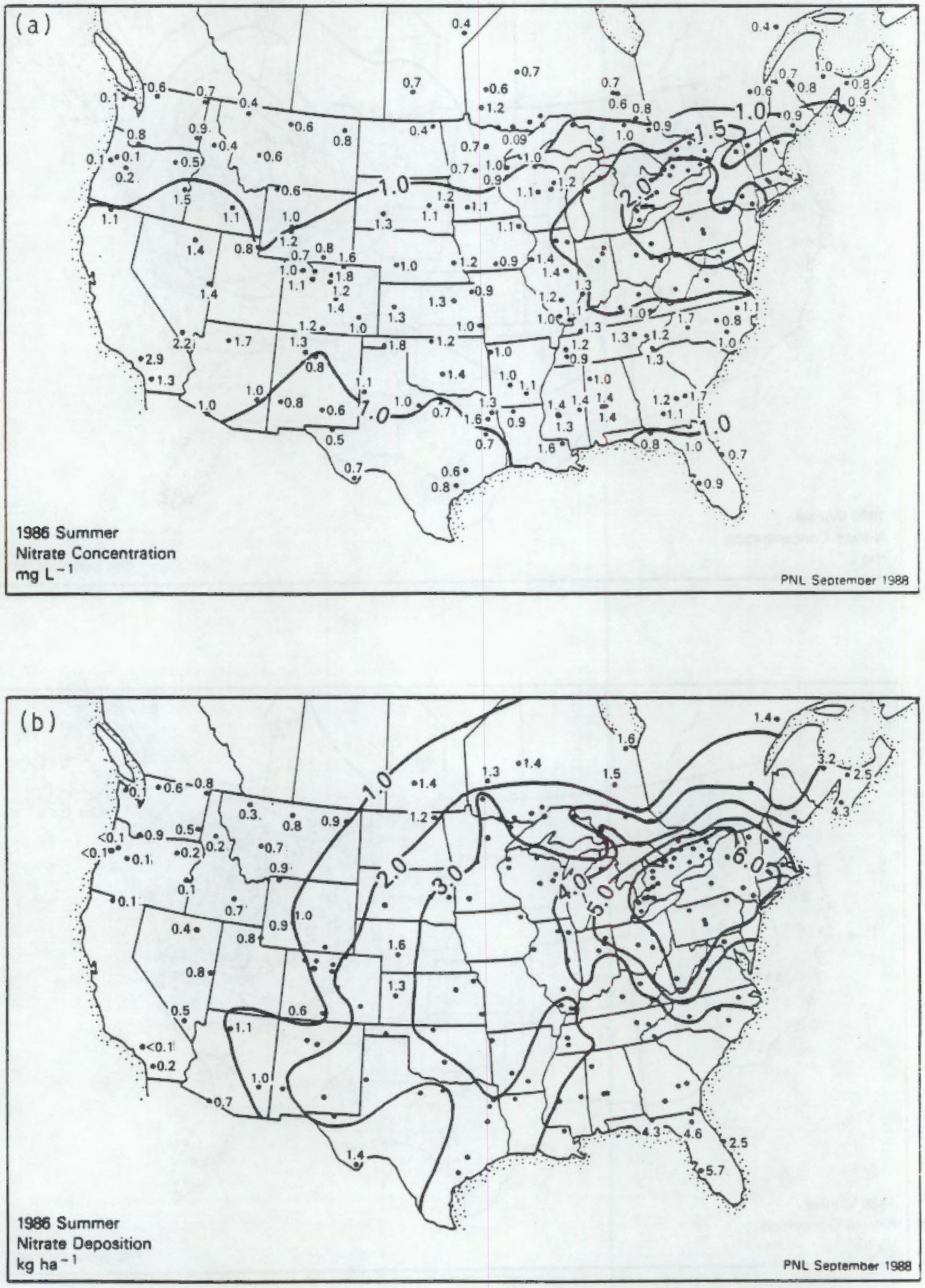


FIGURE A.27. Summer 1986 Spatial Distribution of Nitrate (a) Precipitation-weighted Concentration and (b) Deposition

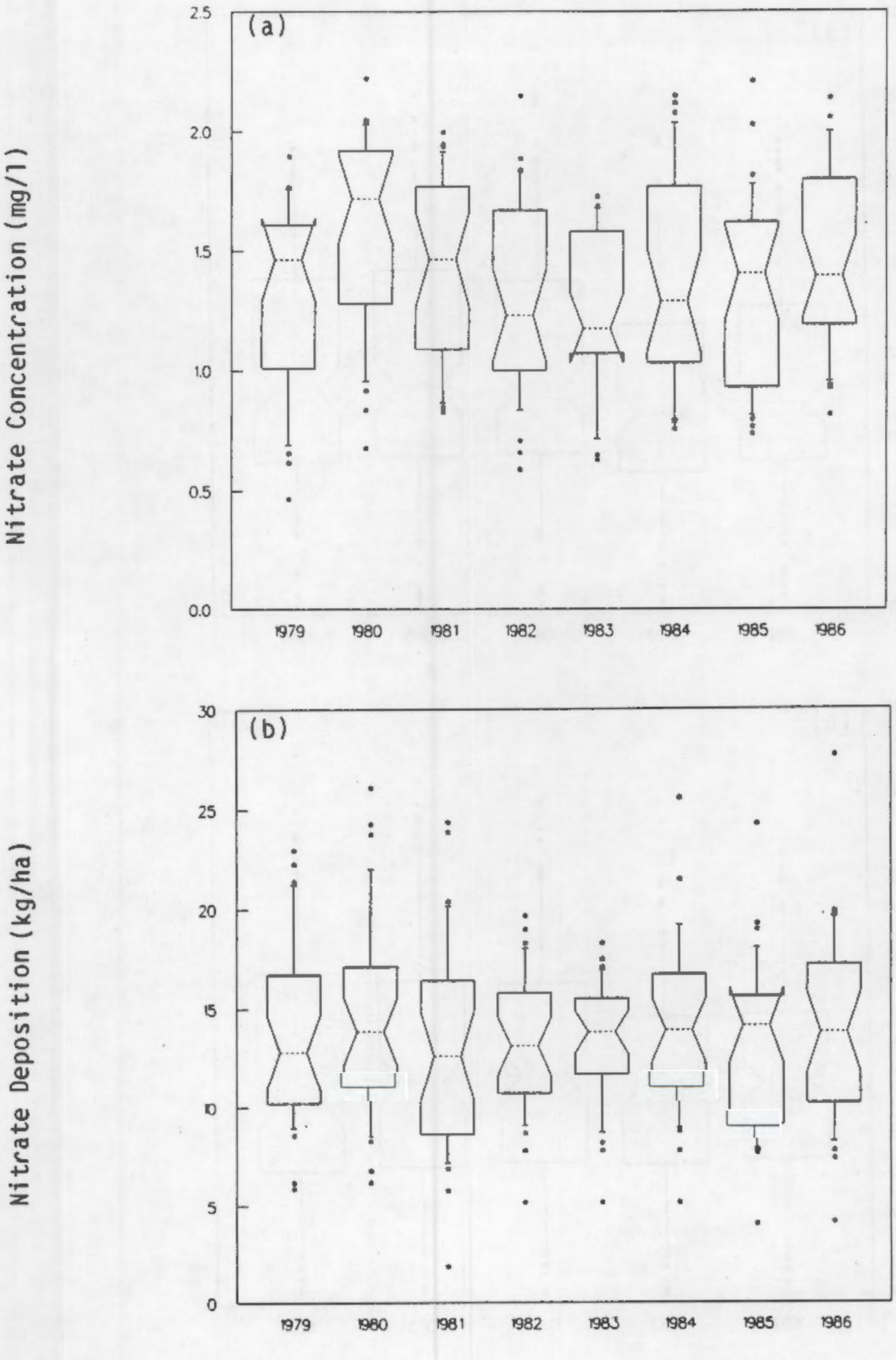


FIGURE A.28. North American Temporal Pattern of (a) Annual Precipitation-weighted Nitrate Ion Concentration, and (b) Annual Nitrate Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

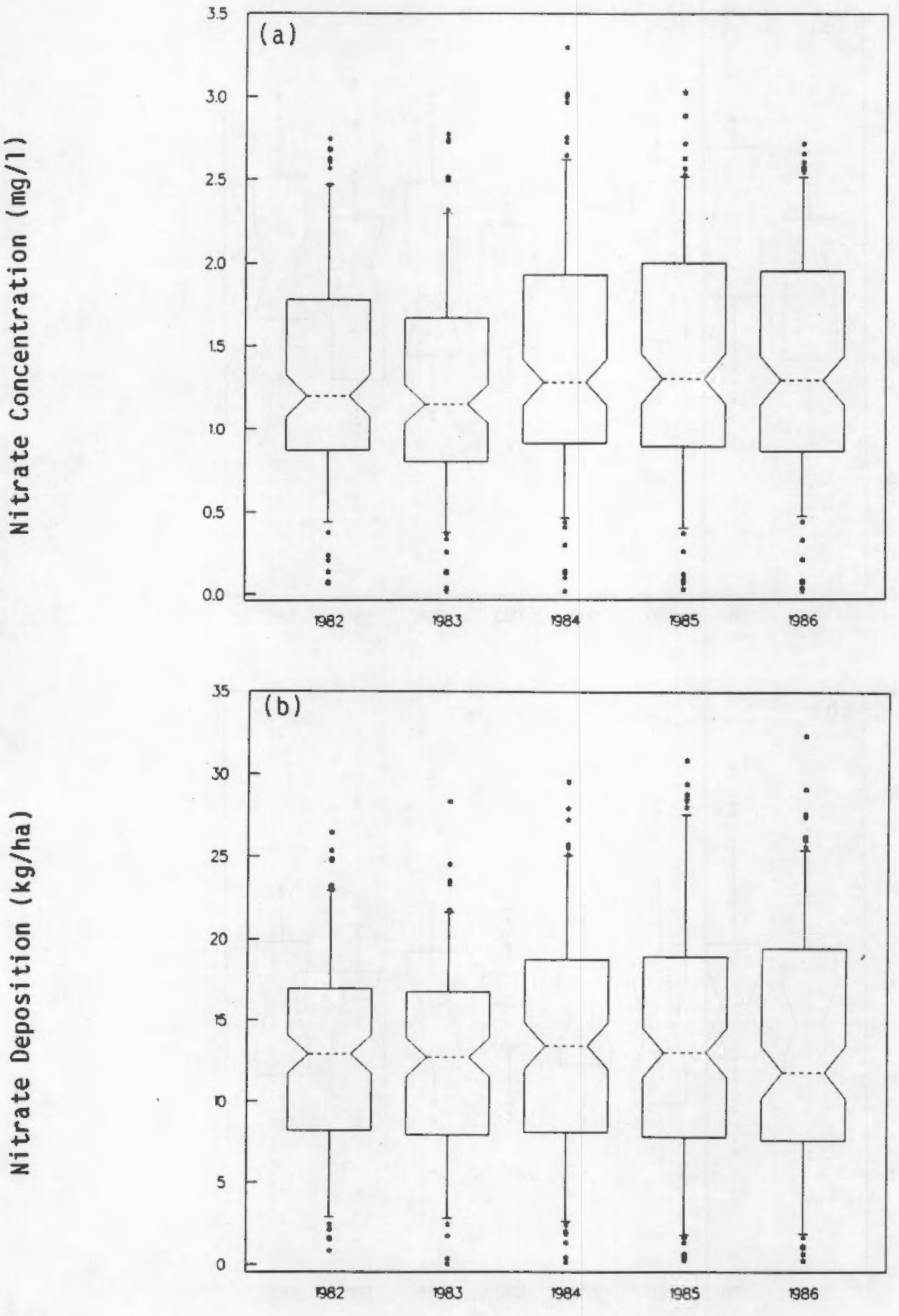


FIGURE A.29. North American Temporal Pattern of (a) Annual Precipitation-weighted Nitrate Ion Concentration, and (b) Annual Nitrate Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Nitrate Concentration Trend Estimate (microeq/l)

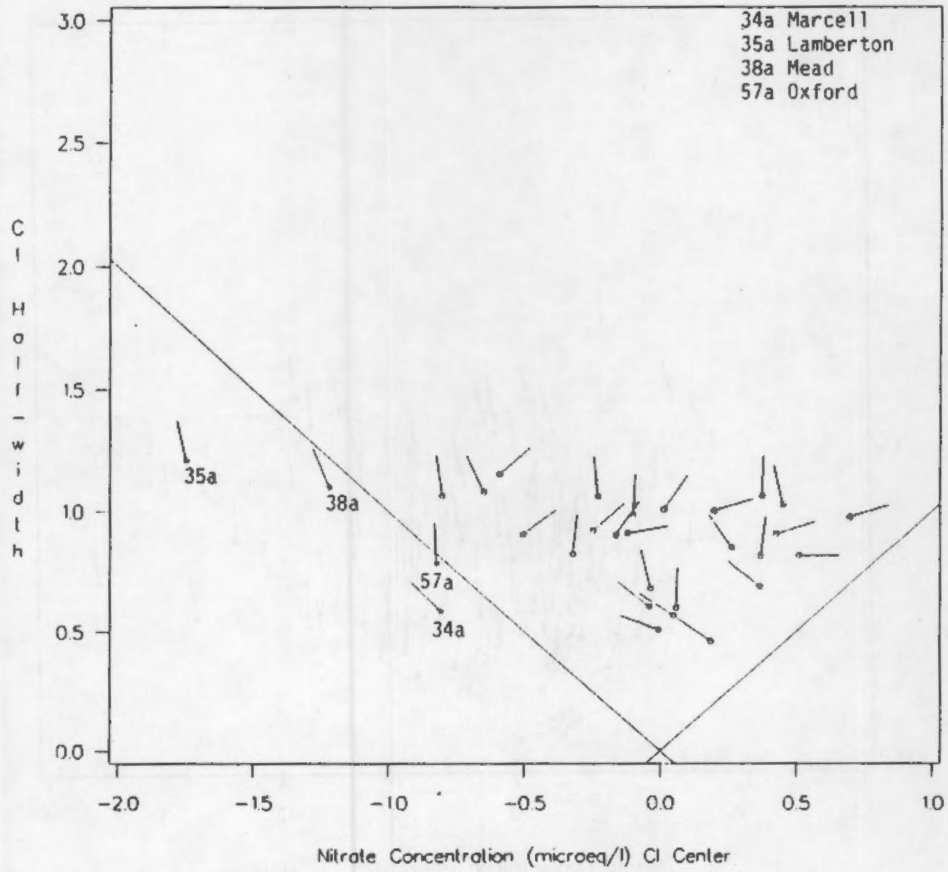


FIGURE A.30. Nitrate Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Nitrate Concentration Trend Estimate (microeq/l)

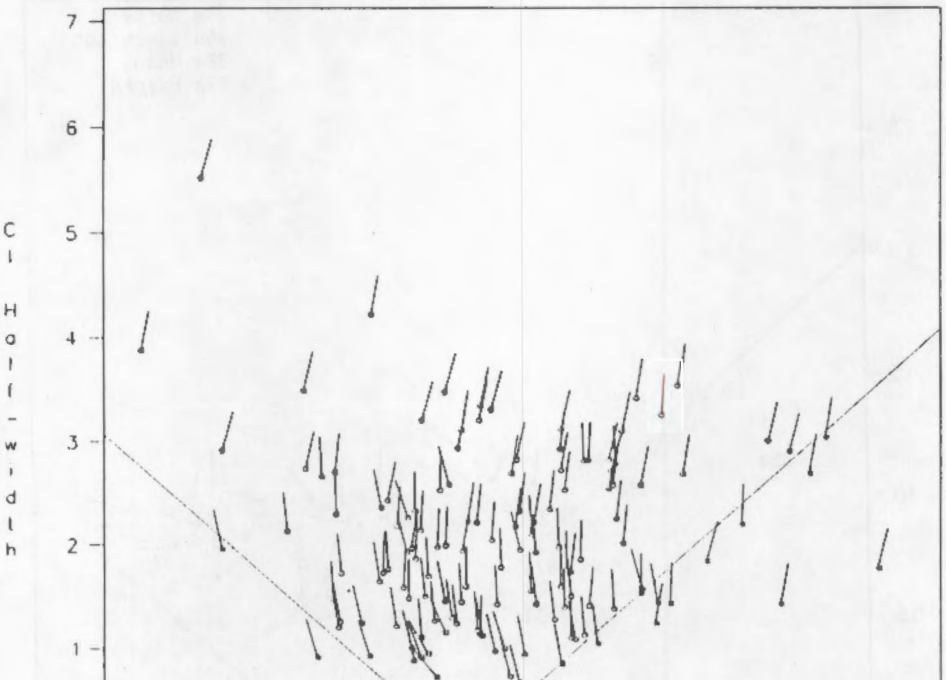


FIGURE A.31. Nitrate Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Nitrate Deposition Trend Estimate (meq/sq m)

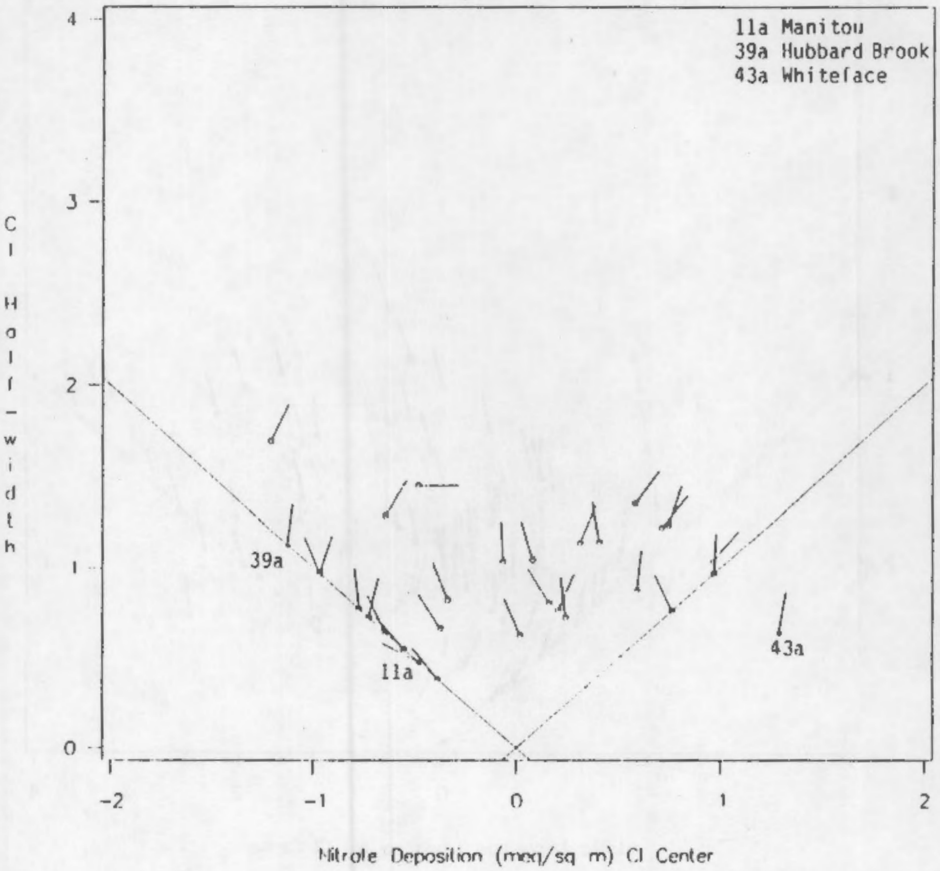
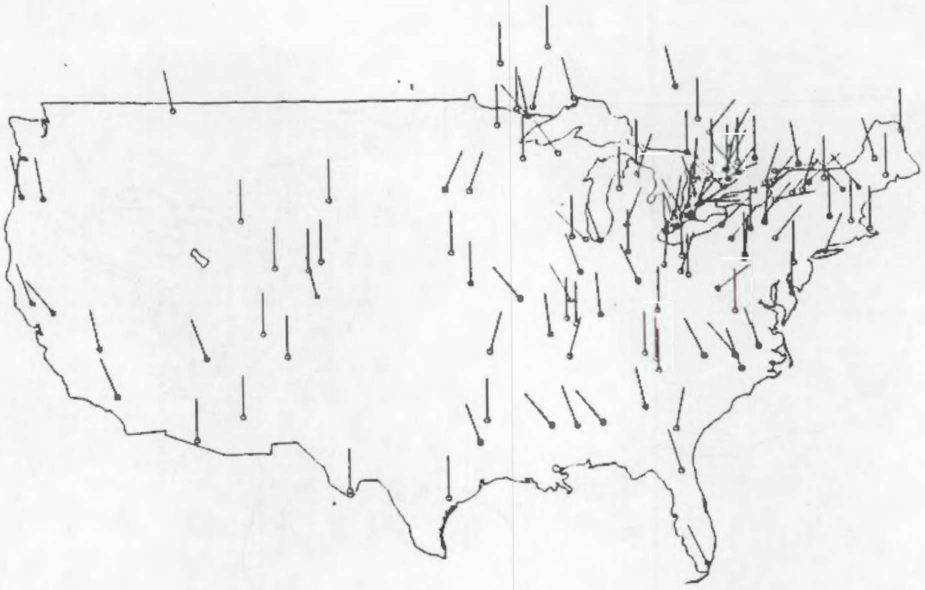


FIGURE A.32. Nitrate Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Nitrate Deposition Trend Estimate (meq/sq m)

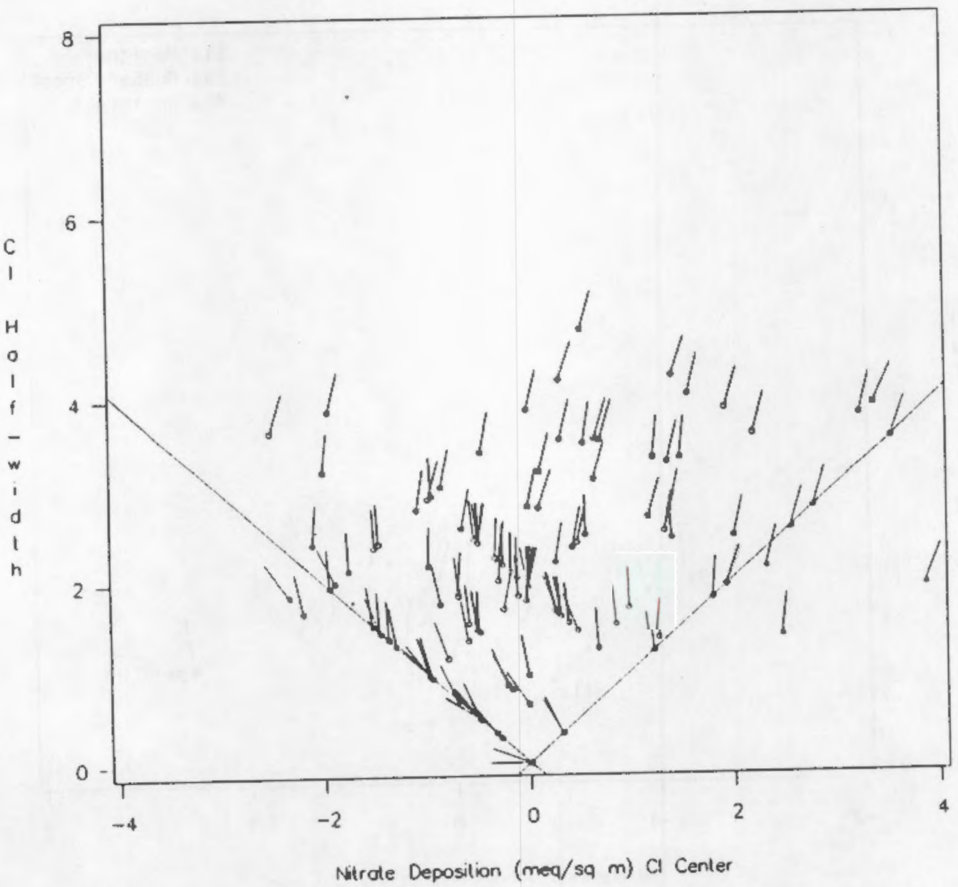
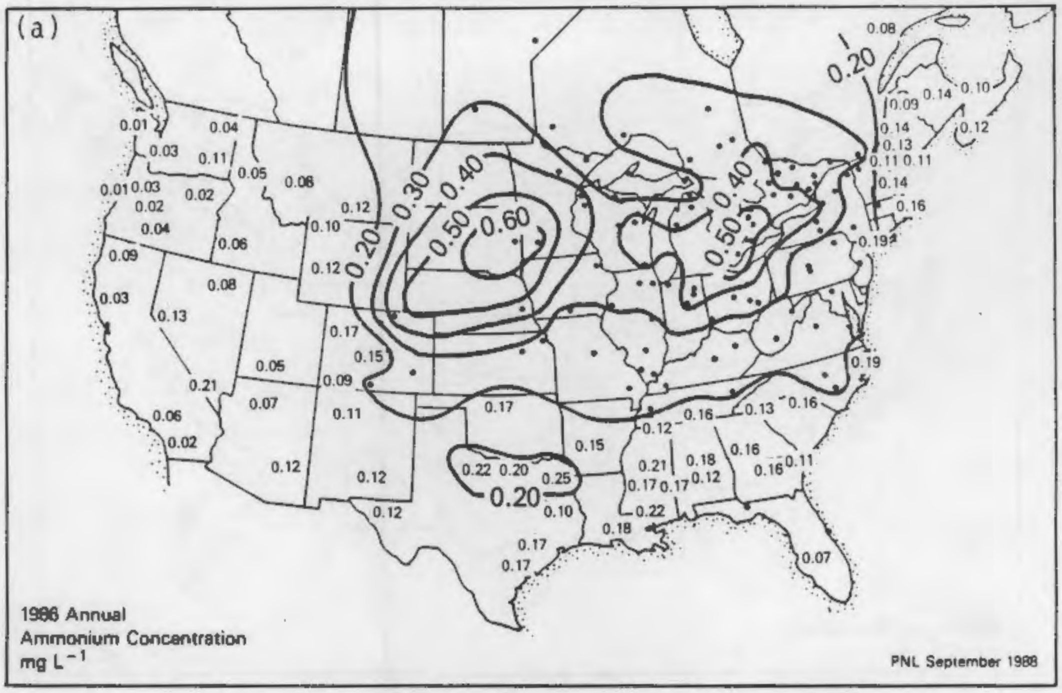


FIGURE A.33. Nitrate Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



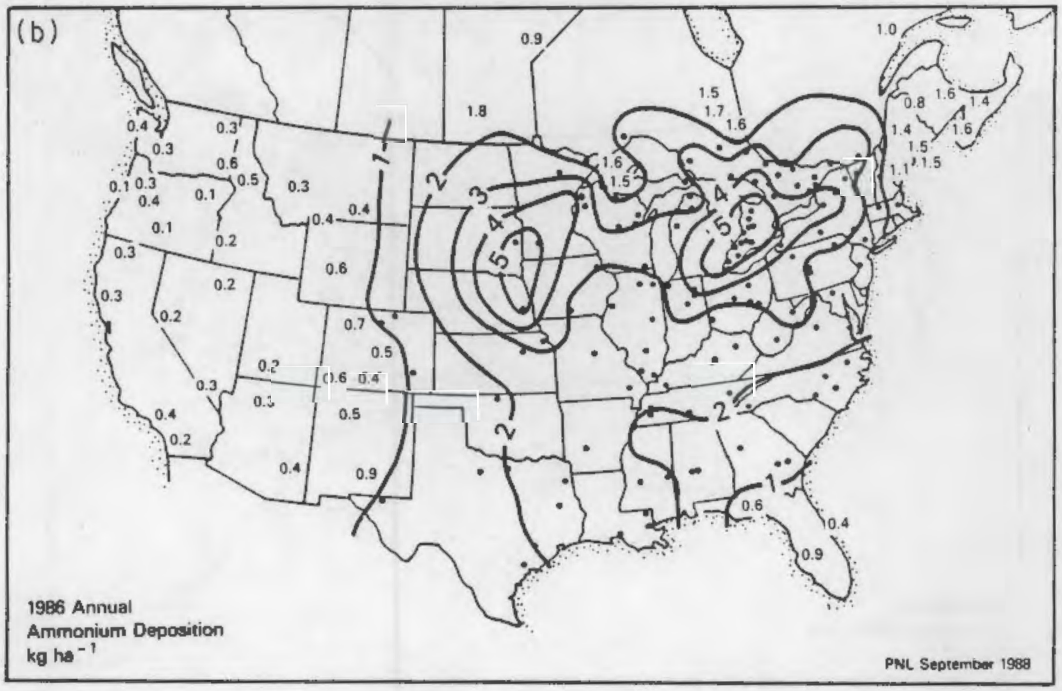


FIGURE A.34. Annual 1986 Spatial Distribution of Ammonium (a) Precipitation-weighted Concentration and (b) Deposition

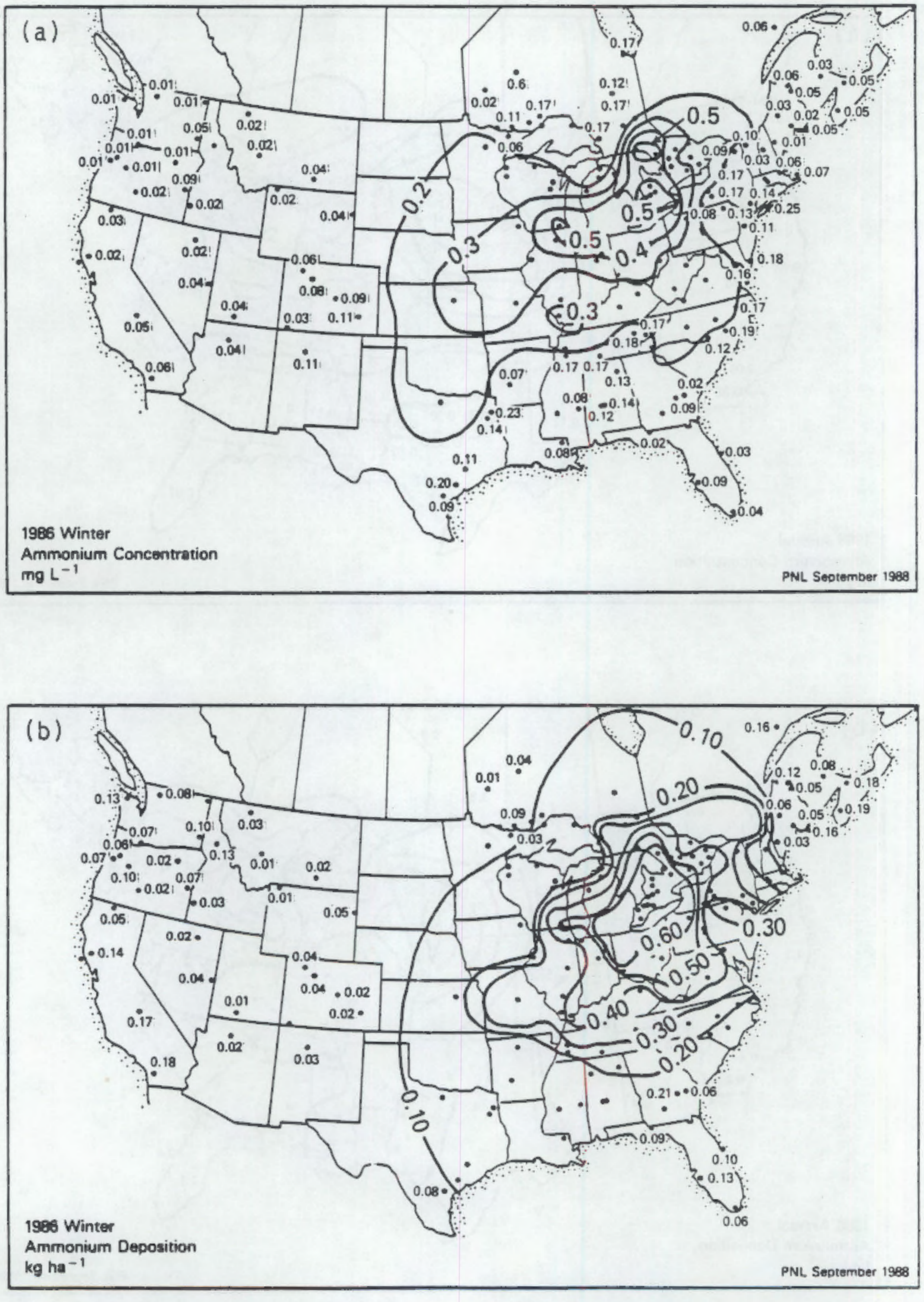


FIGURE A.35. Winter 1986 Spatial Distribution of Ammonium (a) Precipitation-weighted Concentration and (b) Deposition

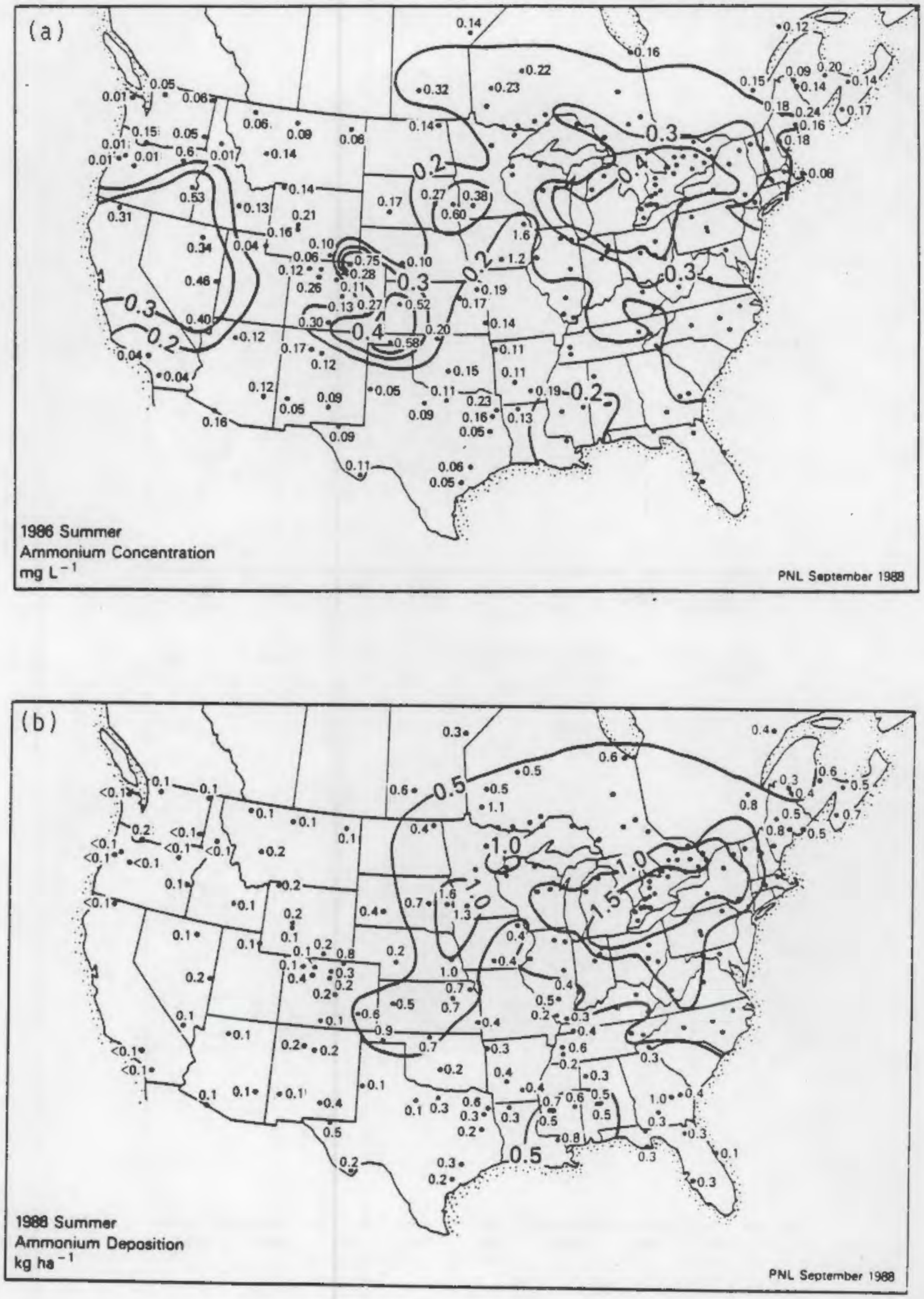


FIGURE A.36. Summer 1986 Spatial Distribution of Ammonium (a) Precipitation-weighted Concentration and (b) Deposition

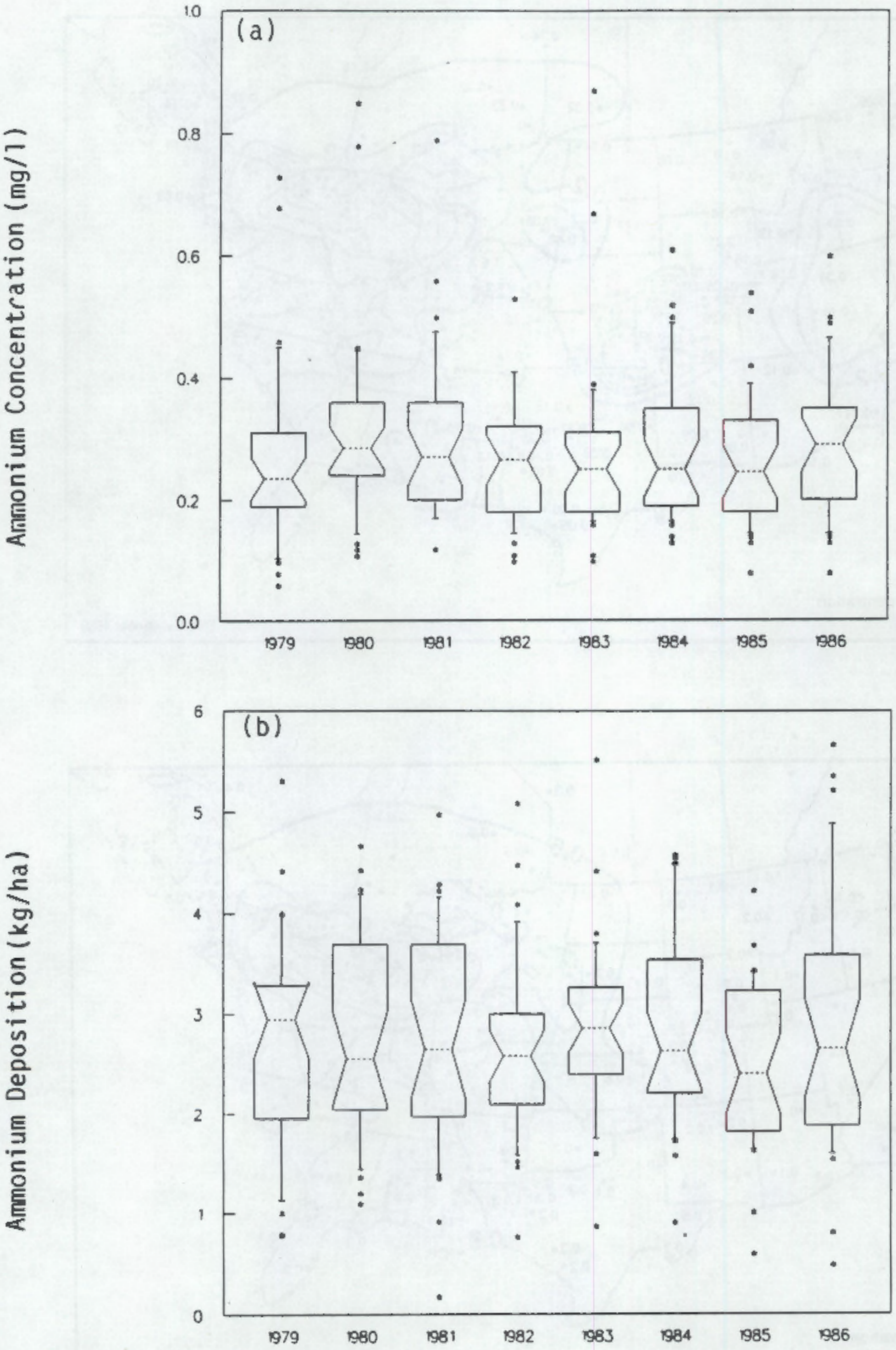


FIGURE A.37. North American Temporal Pattern of (a) Annual Precipitation-weighted Ammonium Ion Concentration, and (b) Annual Ammonium Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

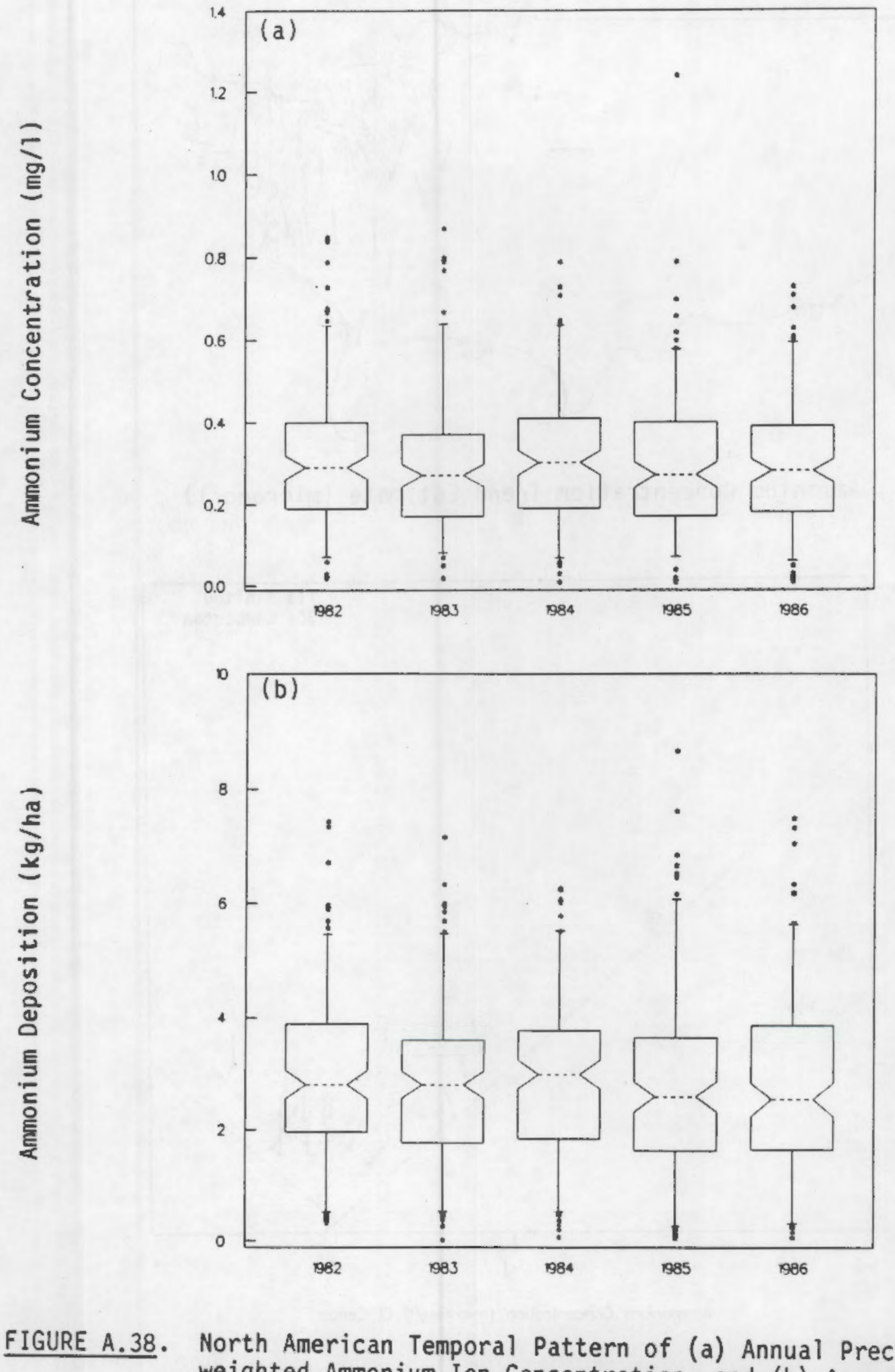


FIGURE A.38. North American Temporal Pattern of (a) Annual Precipitation-weighted Ammonium Ion Concentration, and (b) Annual Ammonium Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Ammonium Concentration Trend Estimate (microeq/l)

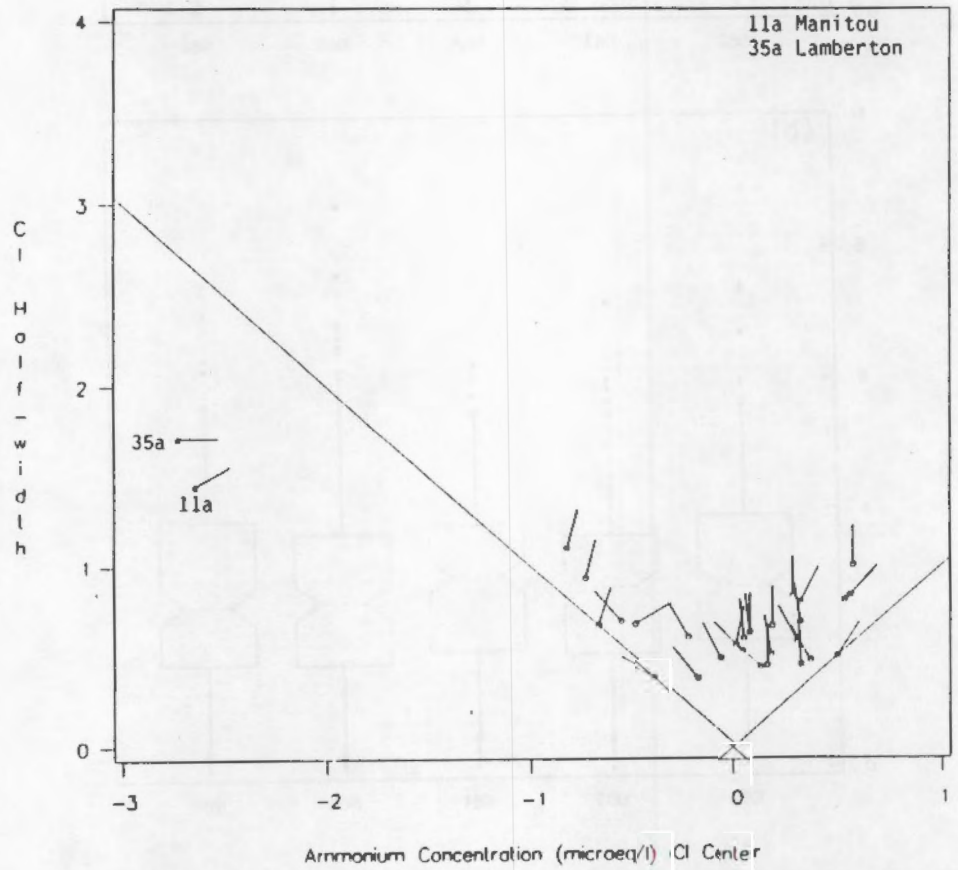
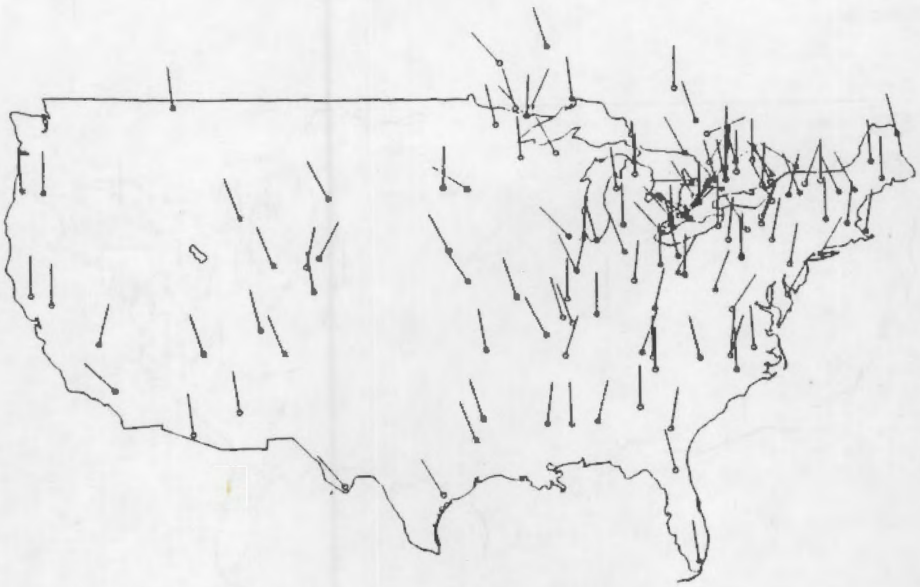


FIGURE A.39. Ammonium Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Ammonium Concentration Trend Estimate (microeq/l)

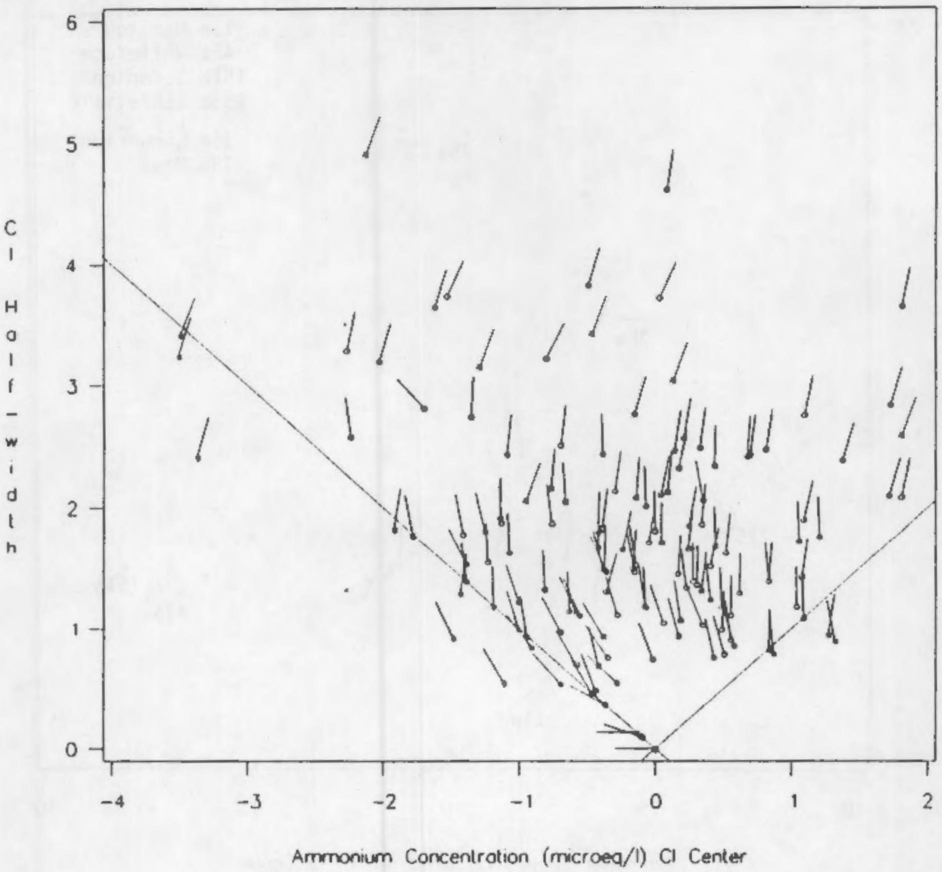


FIGURE A.40. Ammonium Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Ammonium Deposition Trend Estimate (meq/sq m)

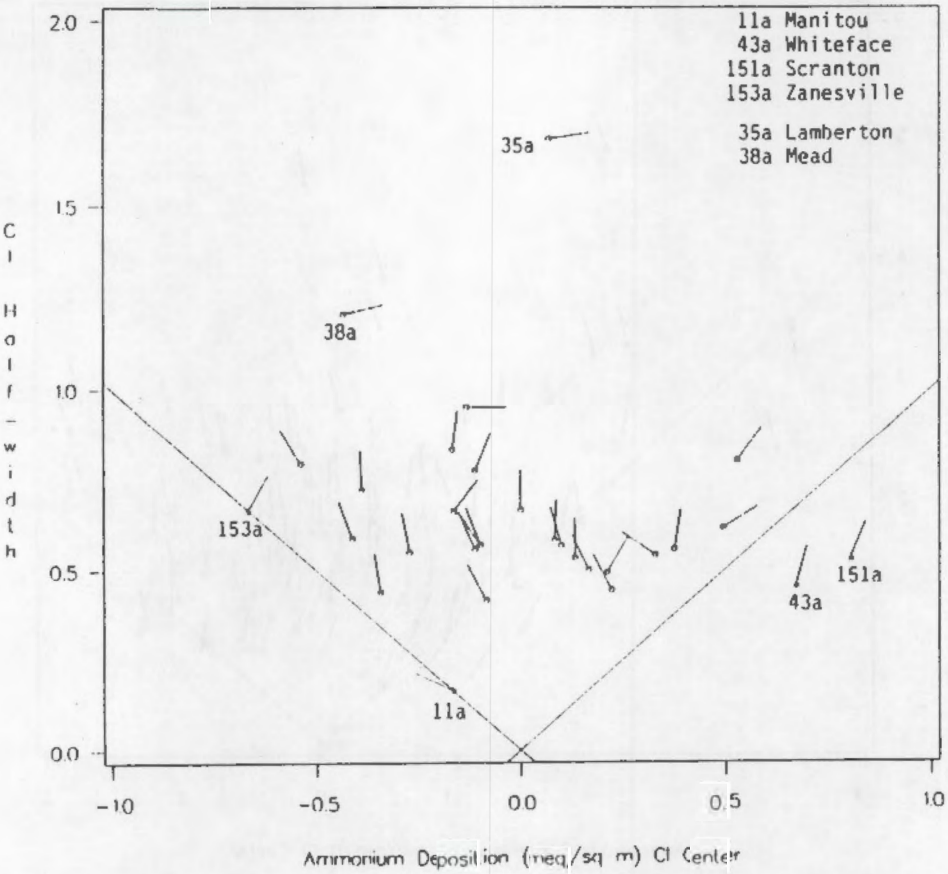


FIGURE A.41. Ammonium Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Ammonium Deposition Trend Estimate (meq/sq m)

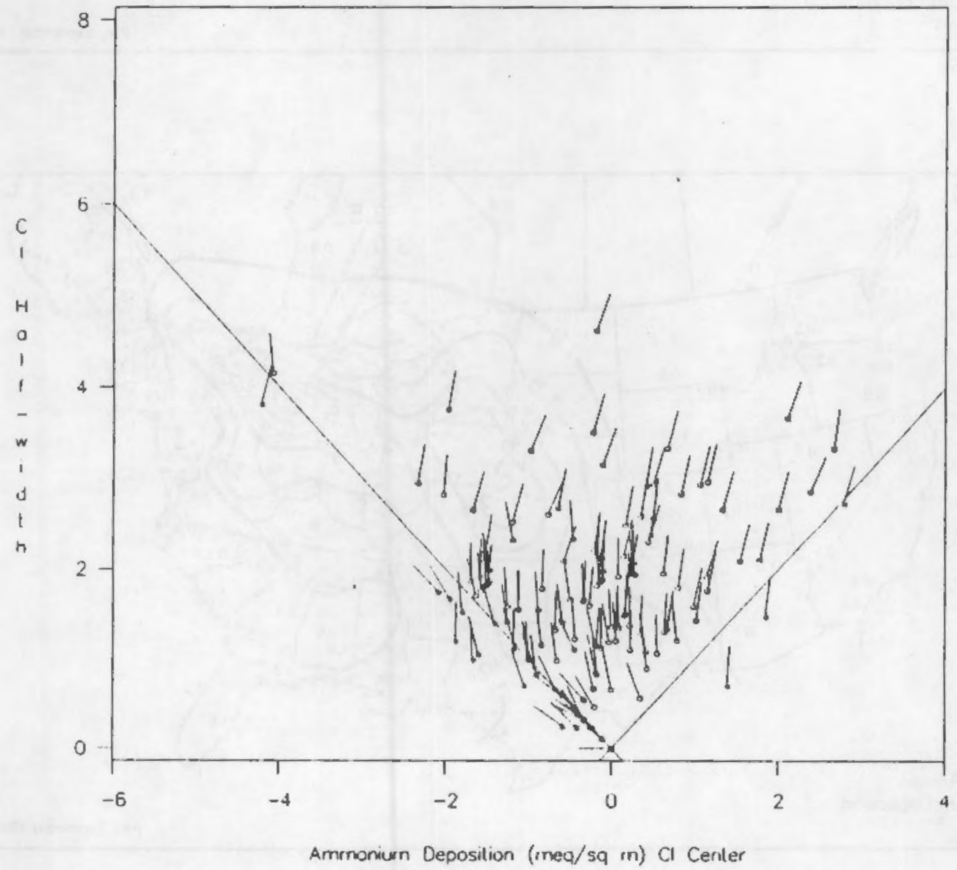


FIGURE A.42. Ammonium Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

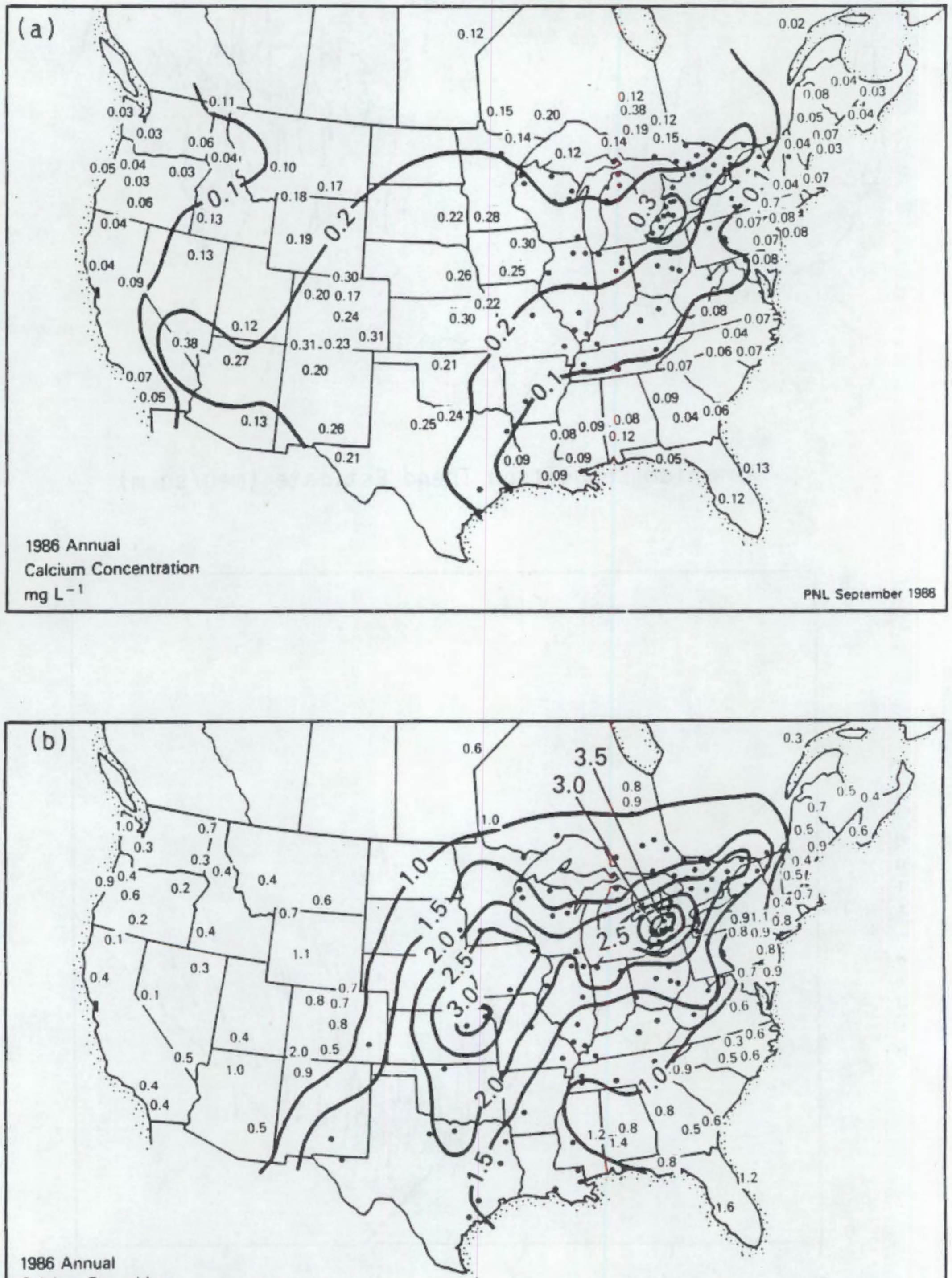


FIGURE A.43. Annual 1986 Spatial Distribution of Calcium (a) Precipitation-weighted Concentration and (b) Deposition

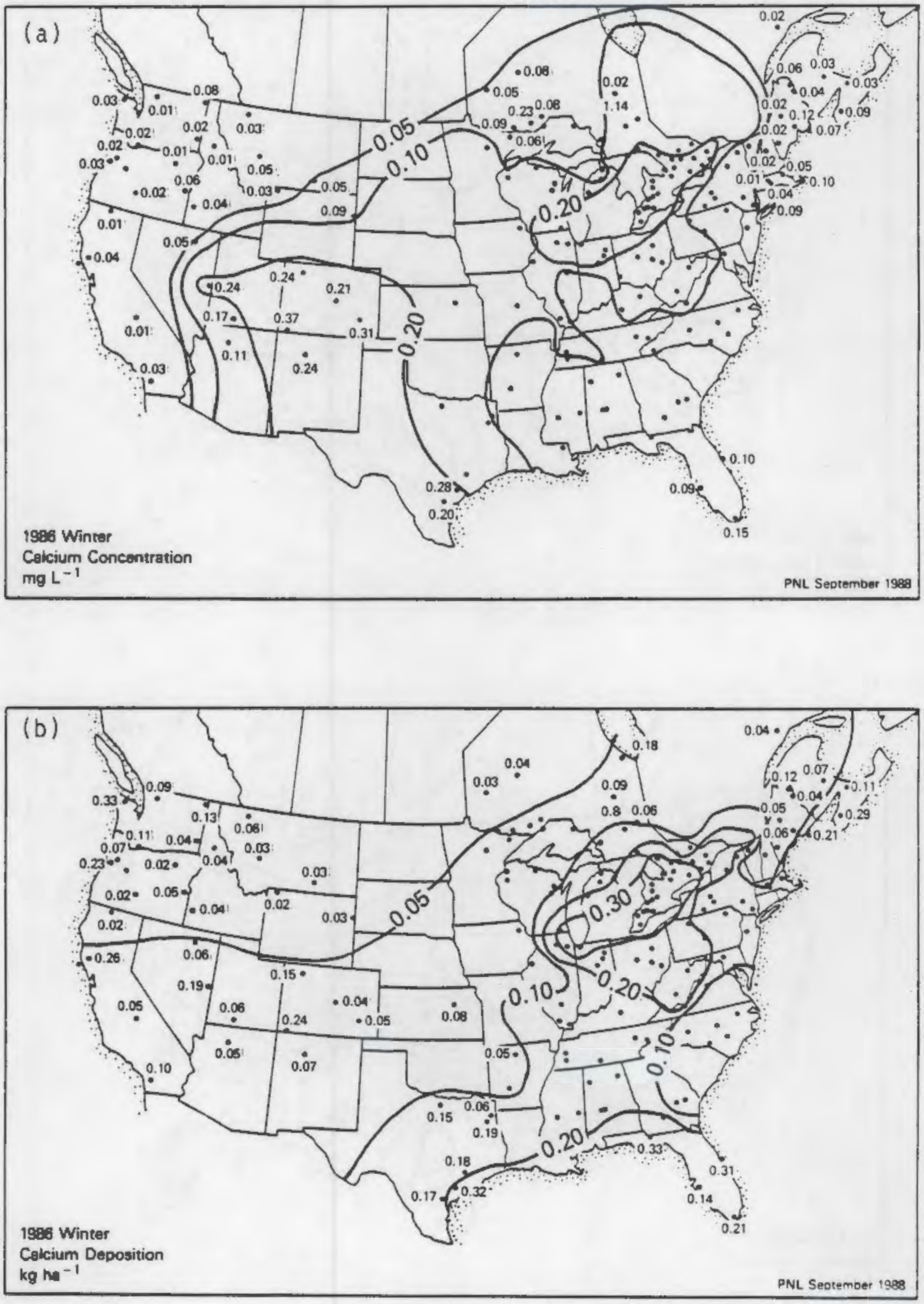


FIGURE A.44. Winter 1986 Spatial Distribution of Calcium (a) Precipitation-weighted Concentration and (b) Deposition

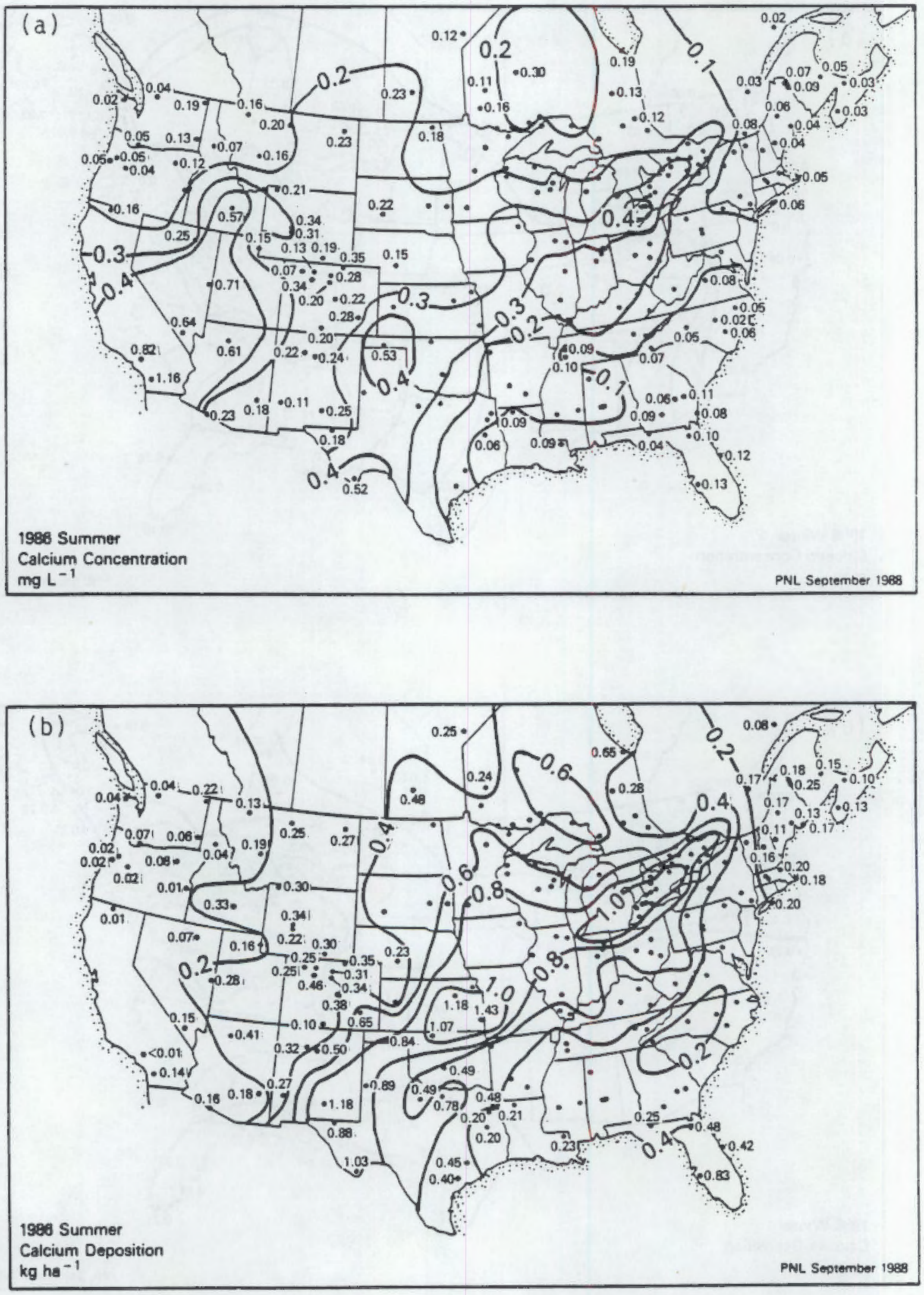


FIGURE A.45. Summer 1986 Spatial Distribution of Calcium (a) Precipitation-weighted Concentration and (b) Deposition

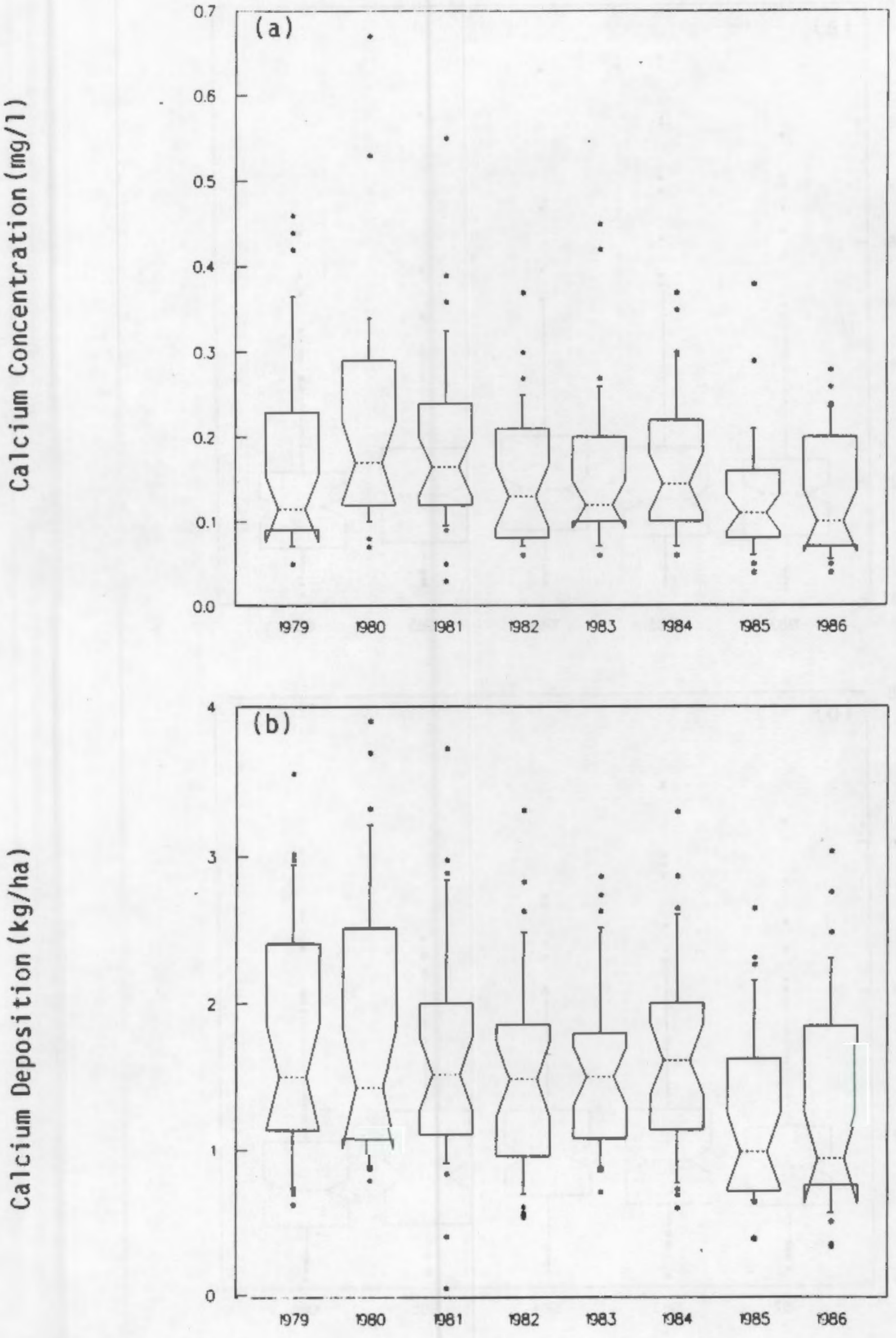


FIGURE A.46. North American Temporal Pattern of (a) Annual Precipitation-weighted Calcium Ion Concentration, and (b) Annual Calcium Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

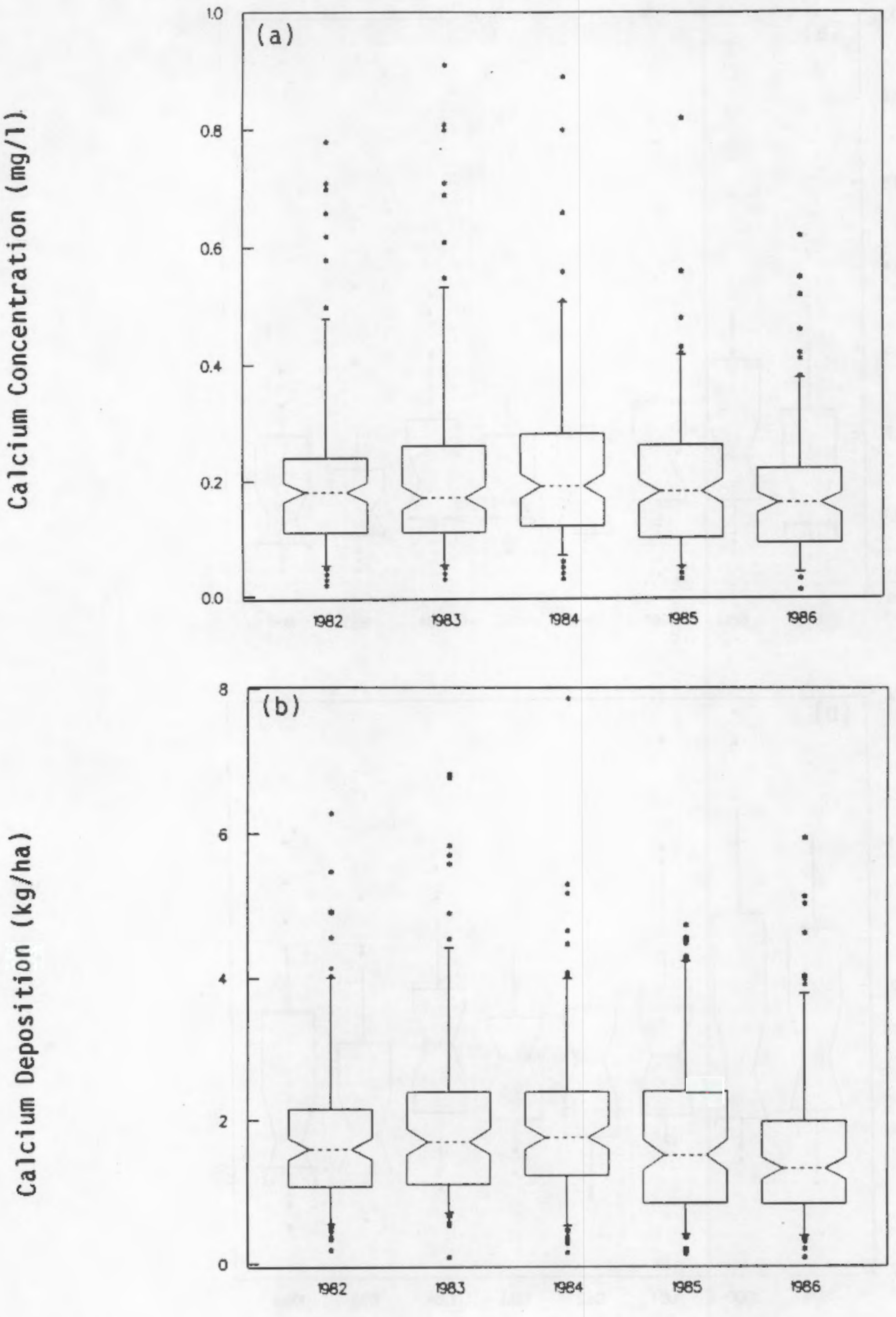


FIGURE A.47. North American Temporal Pattern of (a) Annual Precipitation-weighted Calcium Ion Concentration, and (b) Annual Calcium Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Calcium Concentration Trend Estimate (microeq/l)

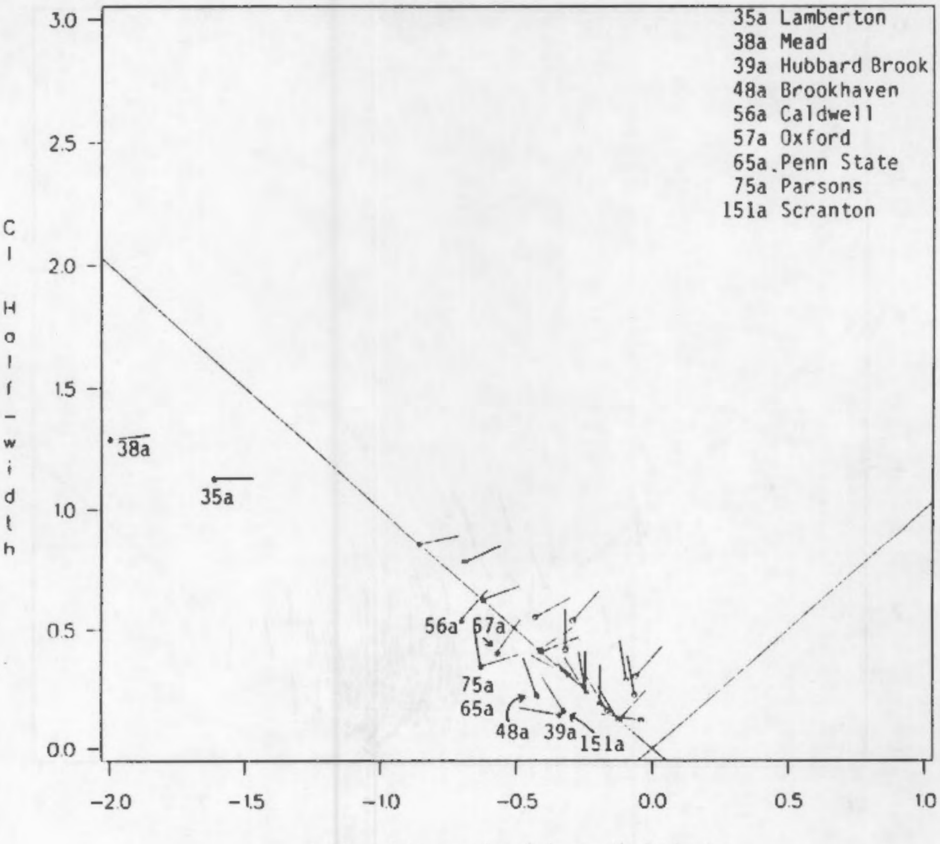


FIGURE A.48. Calcium Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Calcium Concentration Trend Estimate (microeq/l)

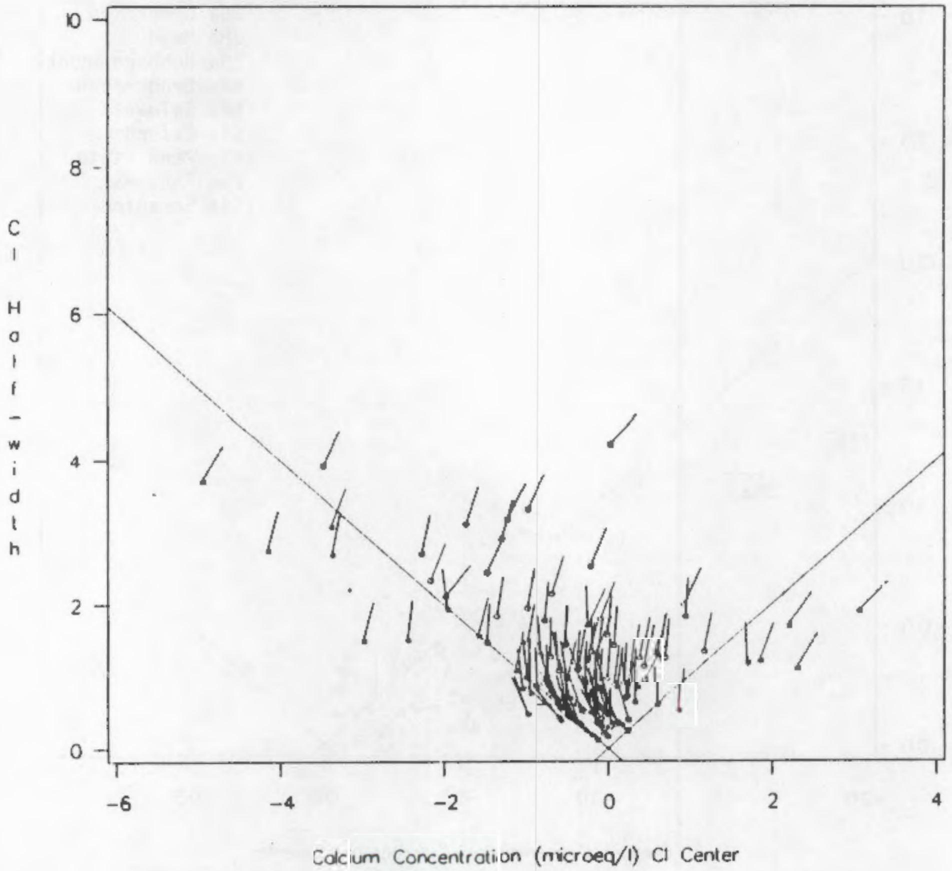


FIGURE A.49. Calcium Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Calcium Deposition Trend Estimate (meq/sq m)

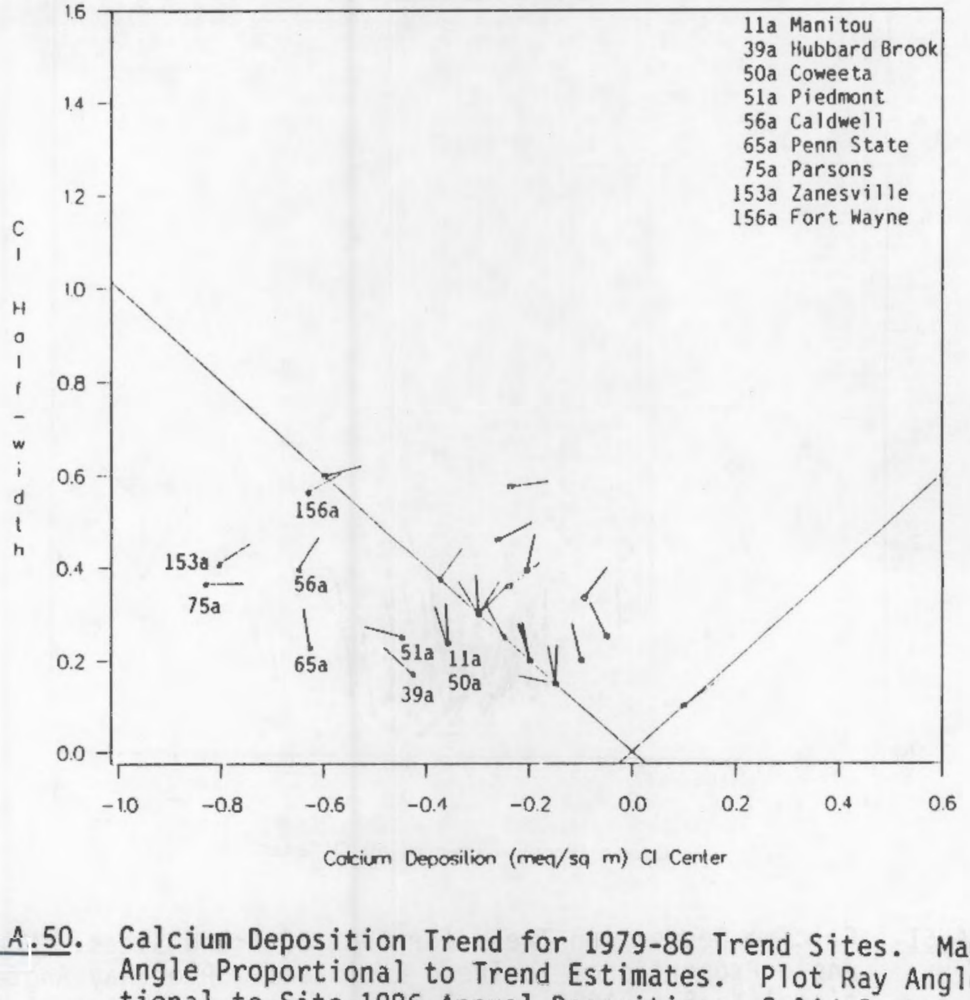


FIGURE A.50. Calcium Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Calcium Deposition Trend Estimate (meq/sq m)

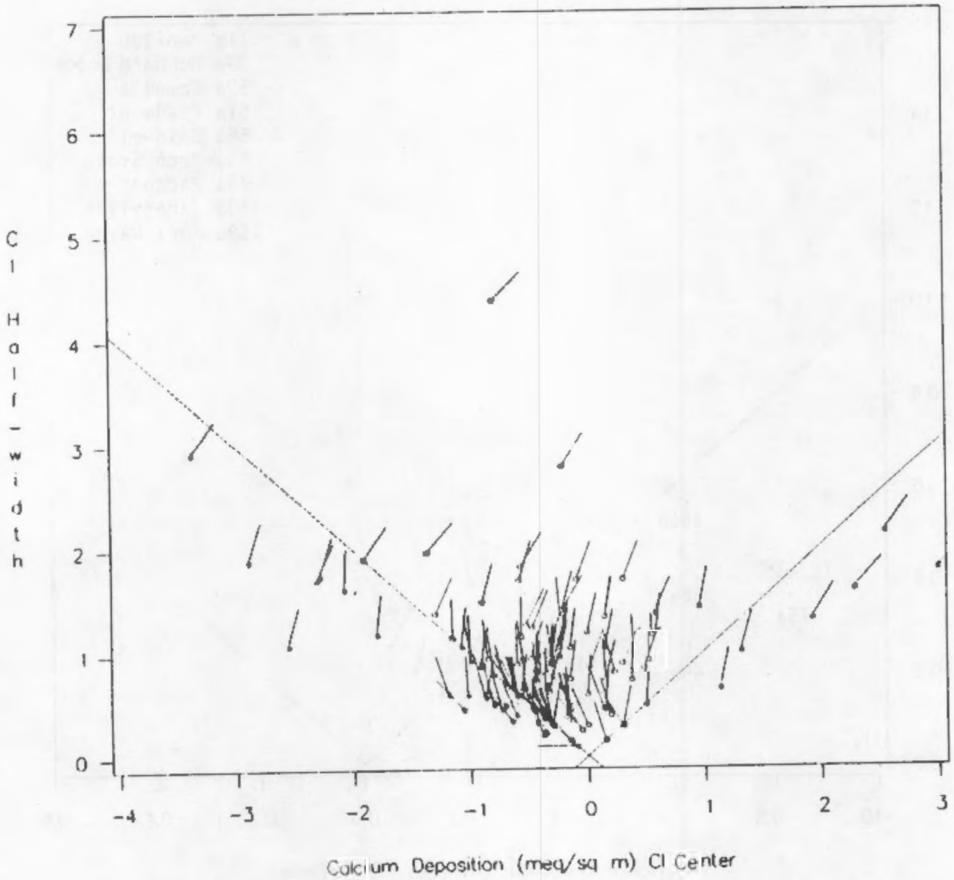


FIGURE A.51. Calcium Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

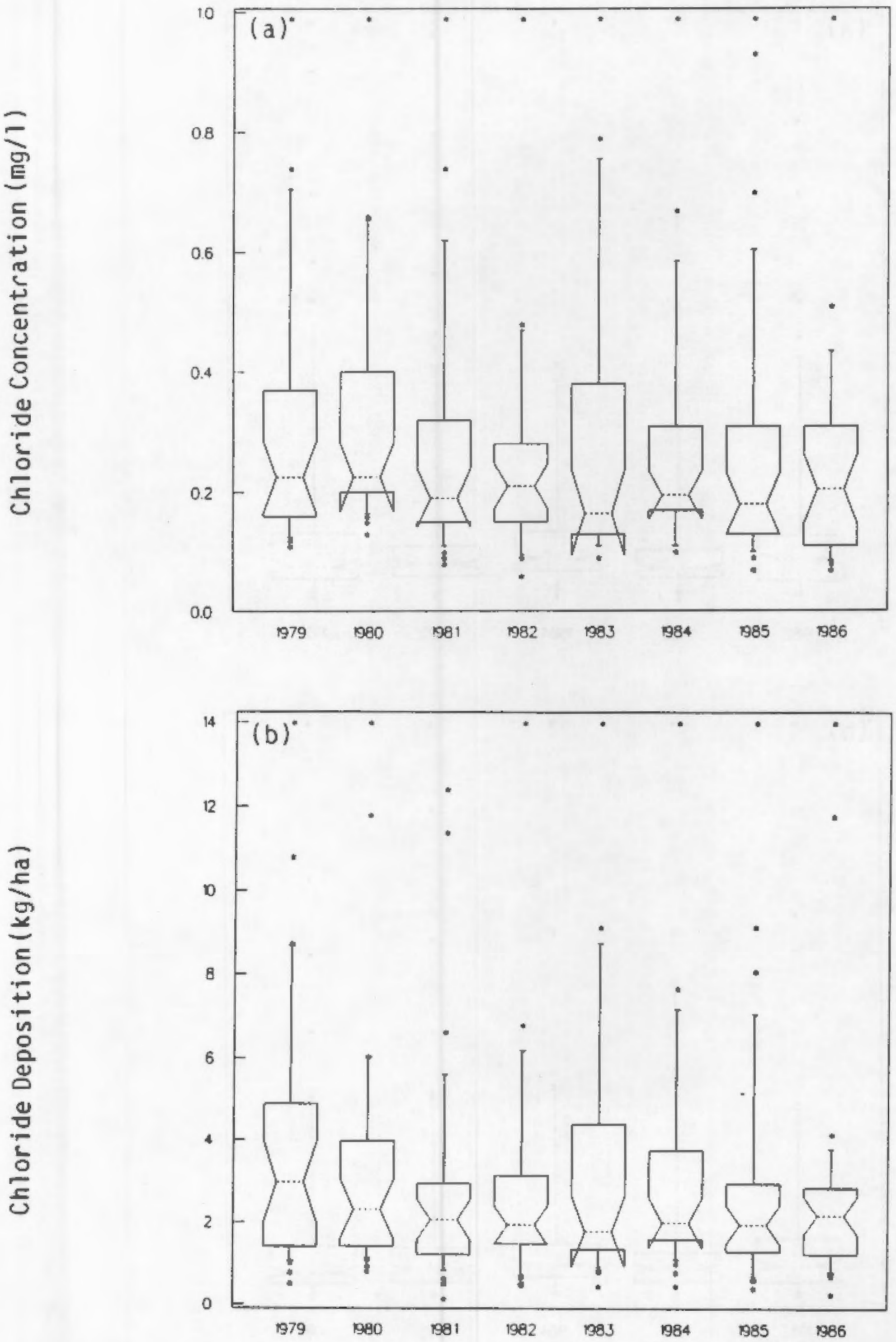


FIGURE A.52. North American Temporal Pattern of (a) Annual Precipitation-weighted Chloride Ion Concentration, and (b) Annual Chloride Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

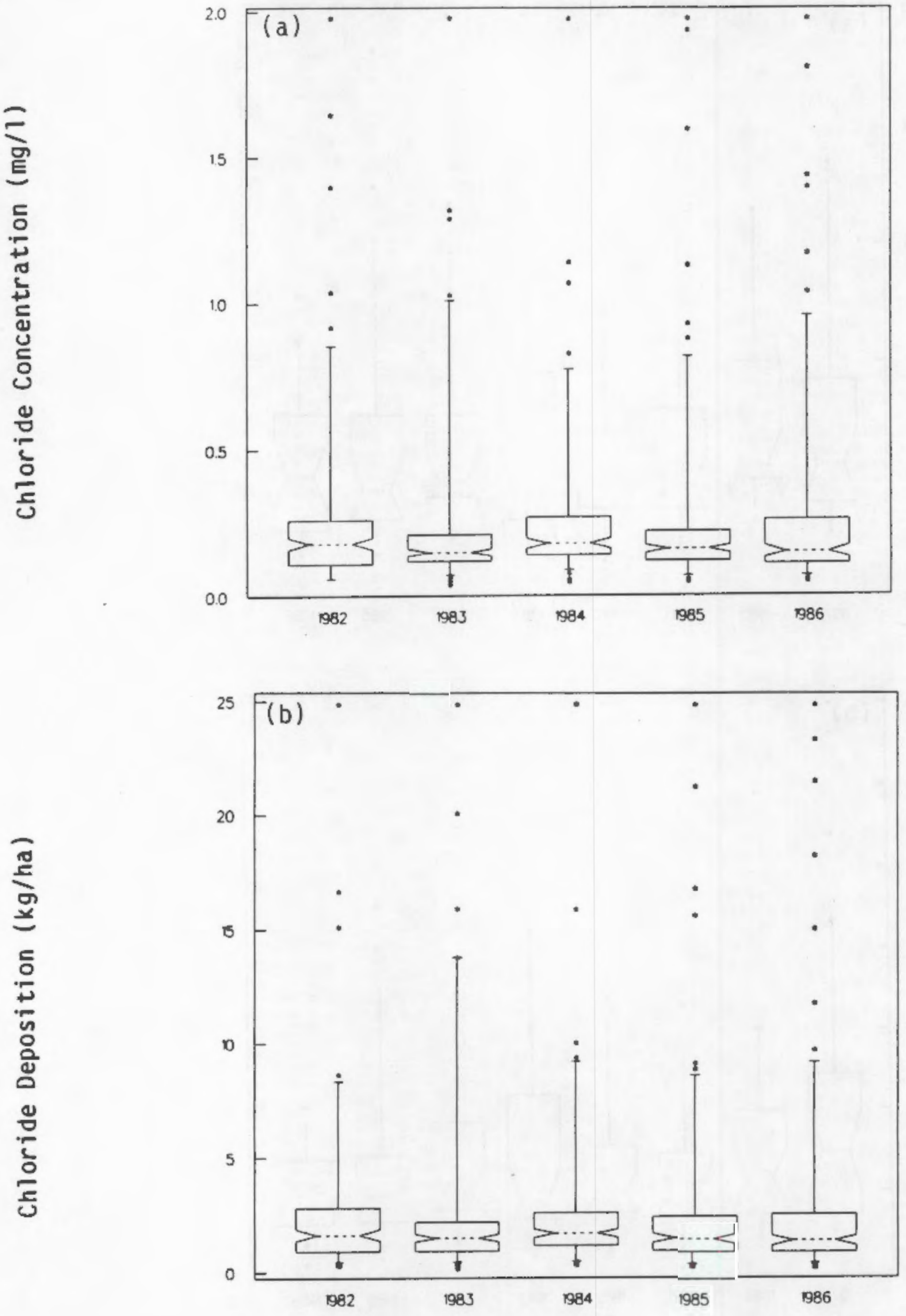


FIGURE A.53. North American Temporal Pattern of (a) Annual Precipitation-weighted Chloride Ion Concentration, and (b) Annual Chloride Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Chloride Concentration Trend Estimate (microeq/l)

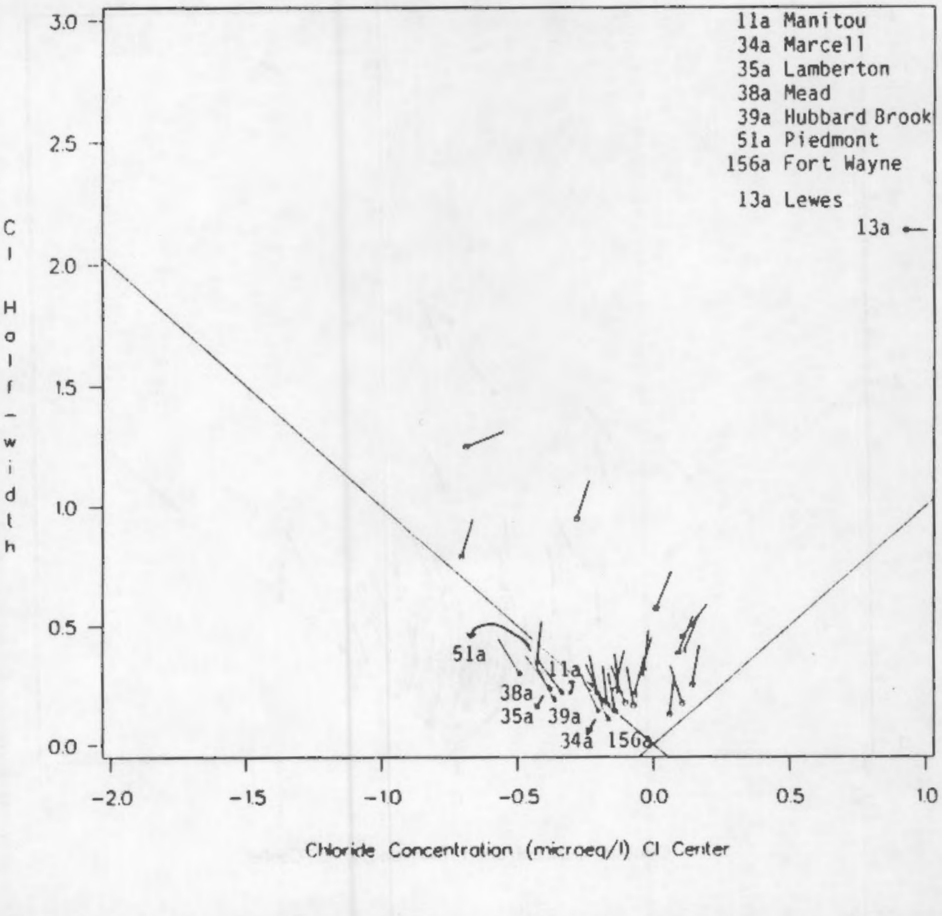
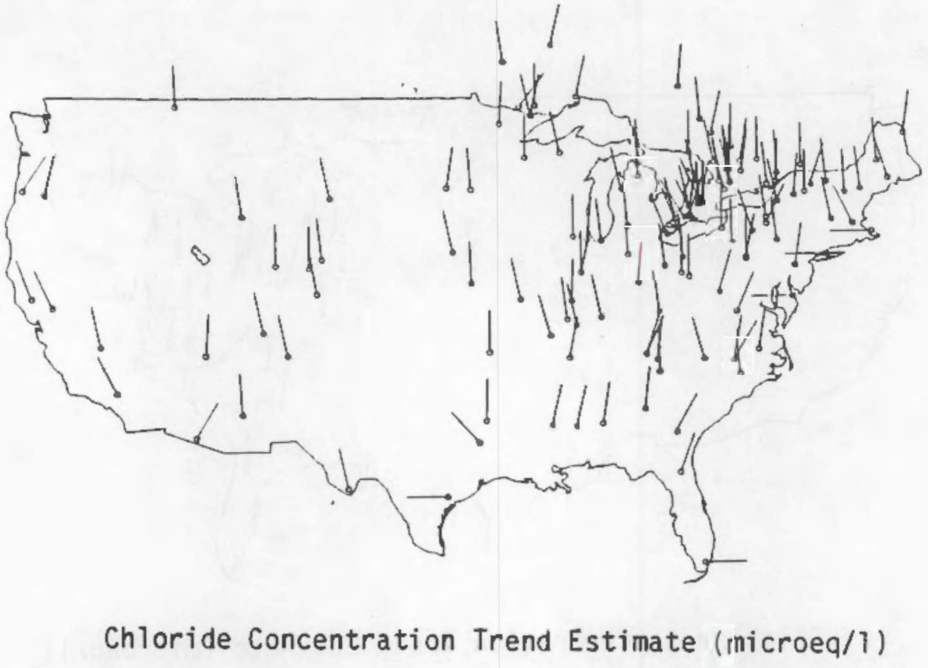


FIGURE A.54. Chloride Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



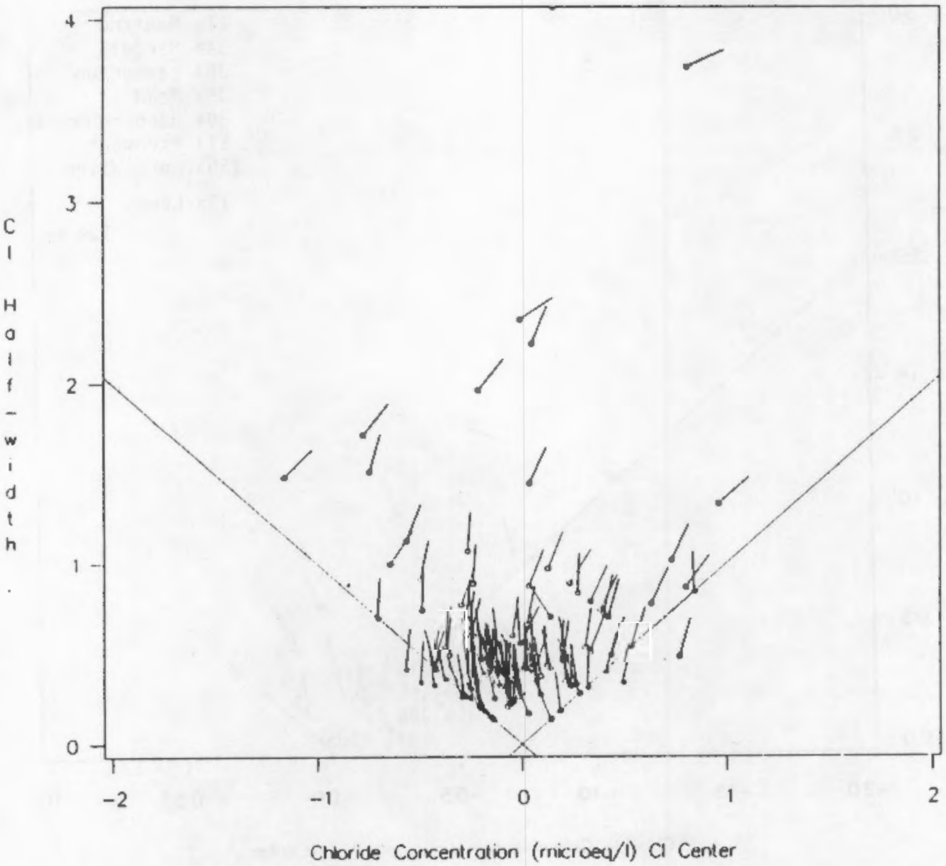


FIGURE A.55. Chloride Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Chloride Deposition Trend Estimate (meq/sq m)

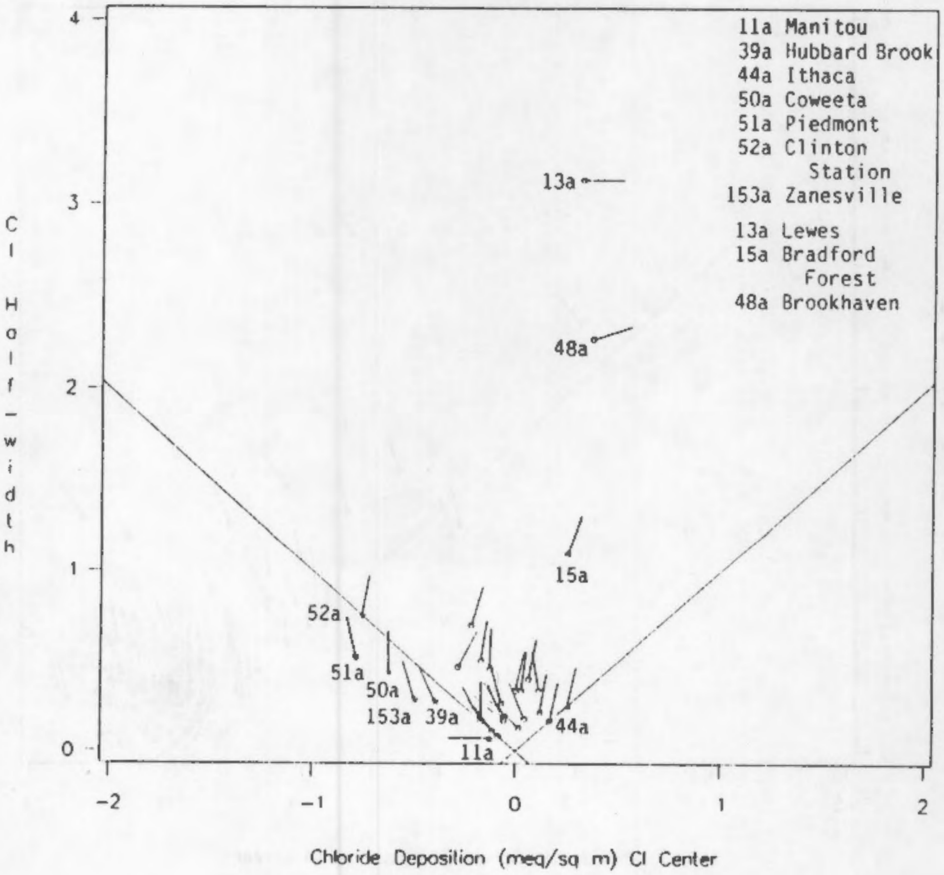
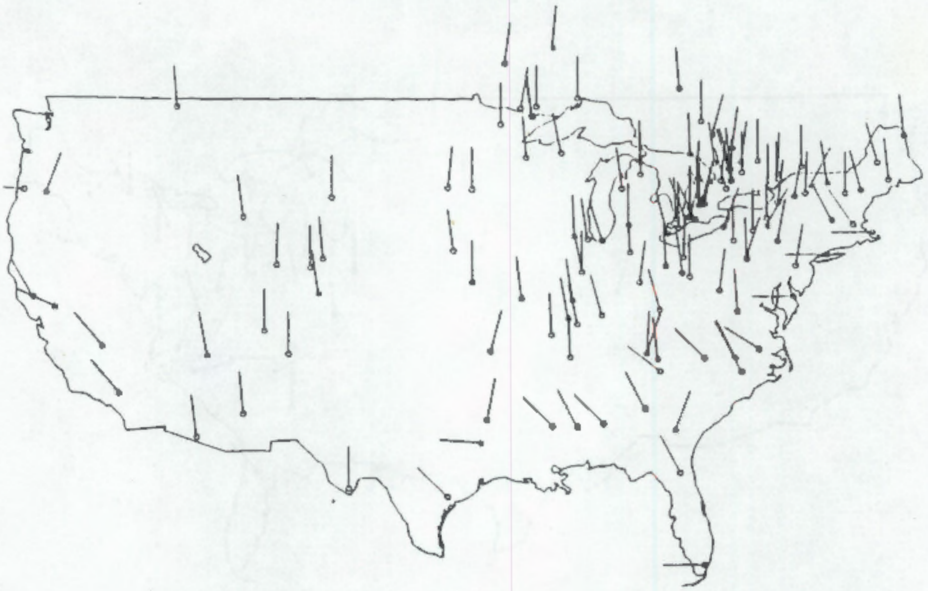


FIGURE A.56. Chloride Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Chloride Deposition Trend Estimate (meq/sq m)

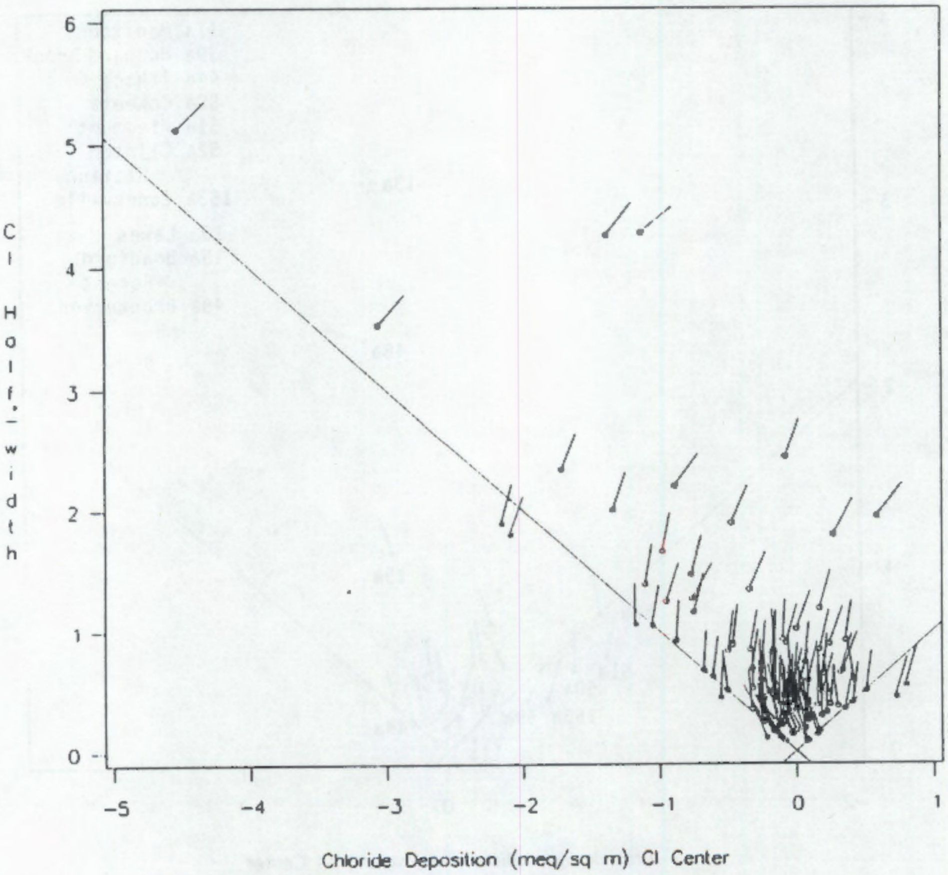


FIGURE A.57. Chloride Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

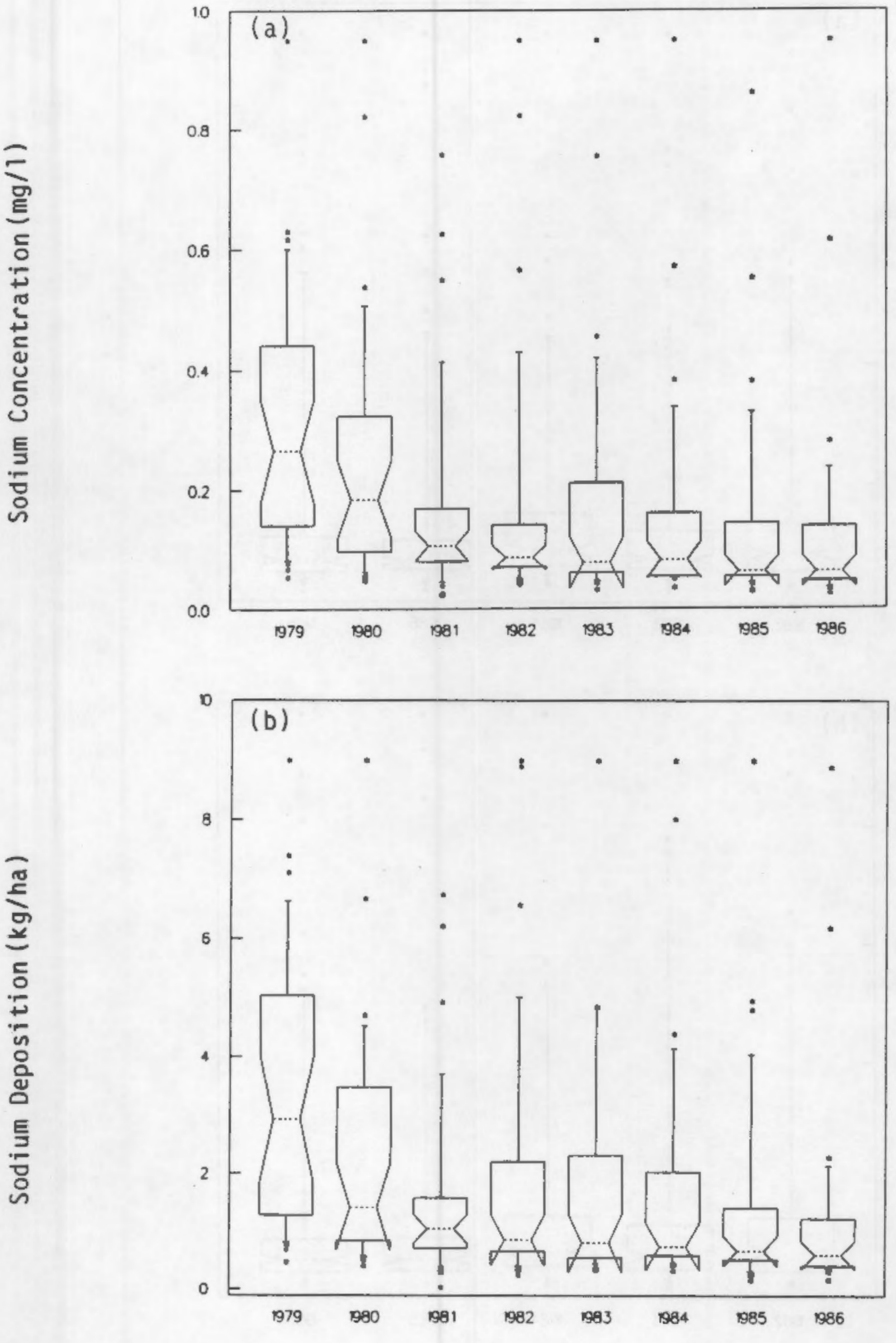


FIGURE A.58. North American Temporal Pattern of (a) Annual Precipitation-weighted Sodium Ion Concentration, and (b) Annual Sodium Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

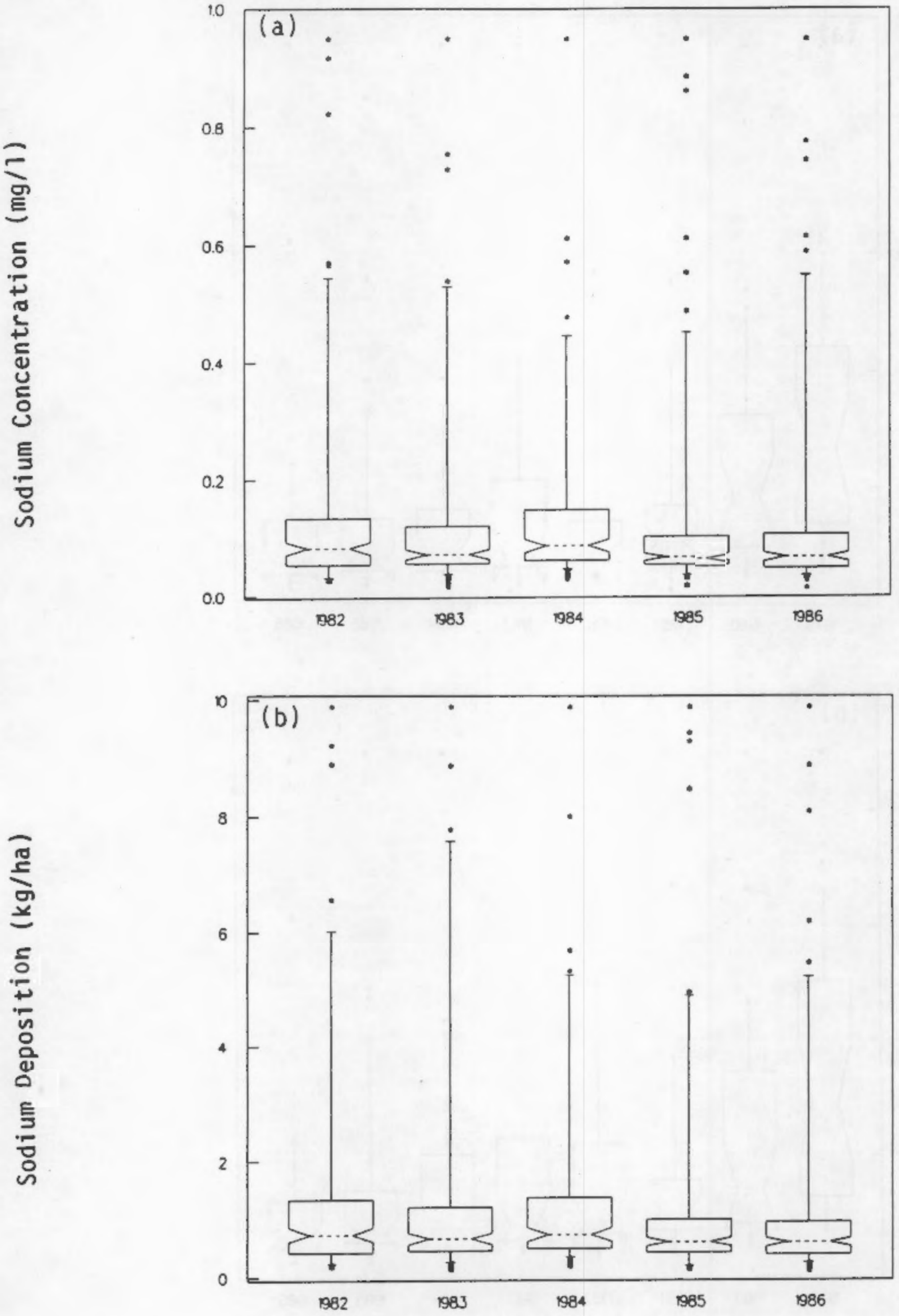


FIGURE A.59. North American Temporal Pattern of (a) Annual Precipitation-weighted Sodium Ion Concentration, and (b) Annual Sodium Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Sodium Concentration Trend Estimate (microeq/l)

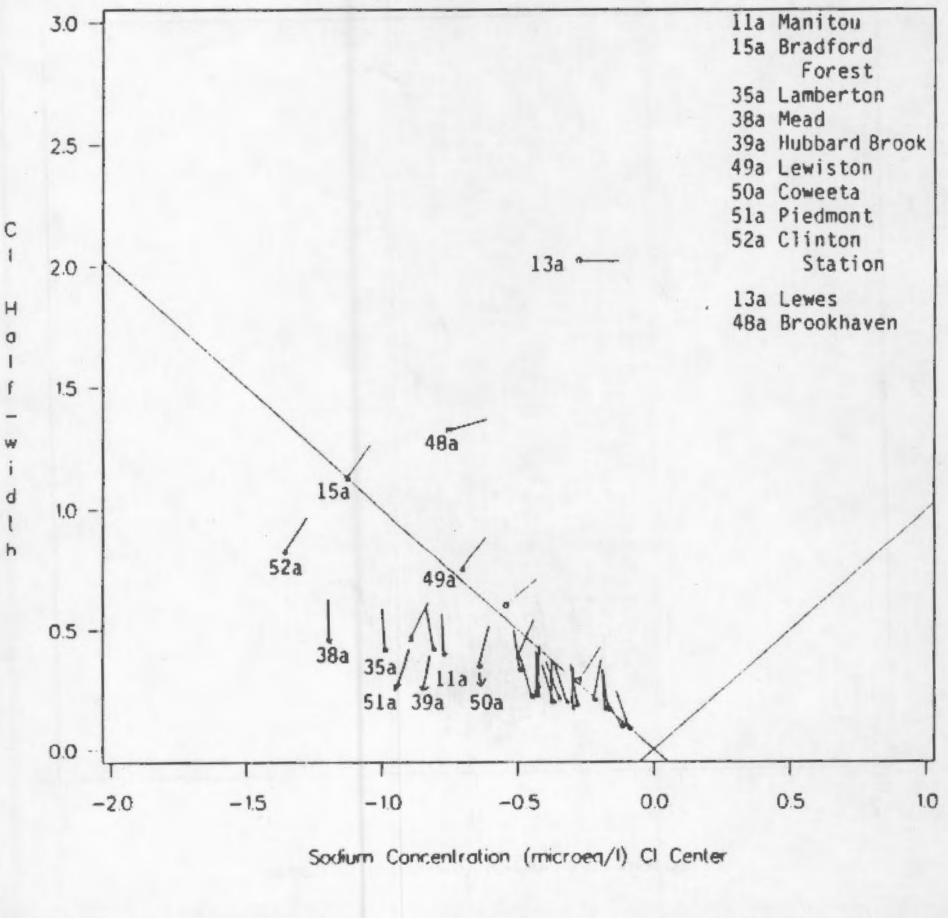
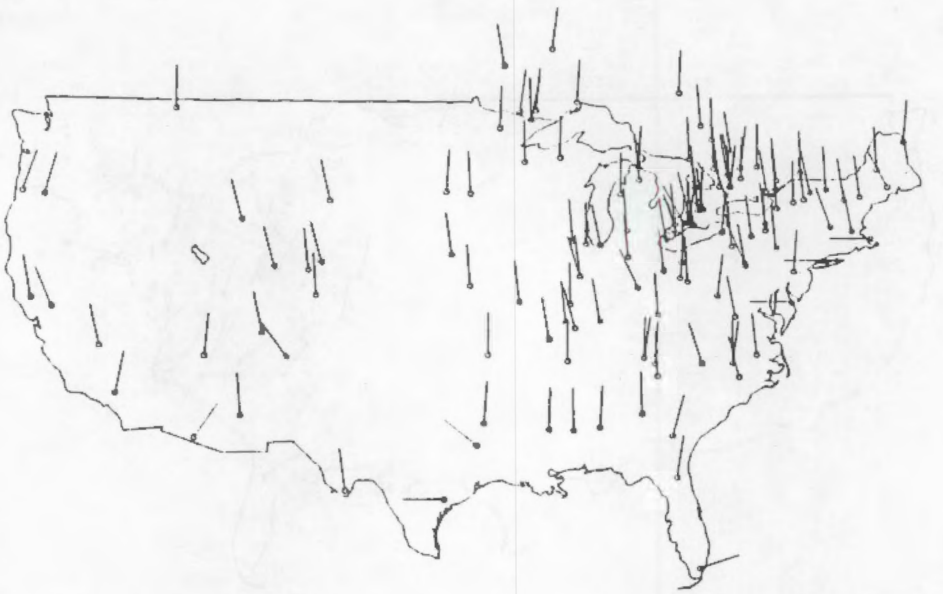


FIGURE A.60. Sodium Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Sodium Concentration Trend Estimate (microeq/l)

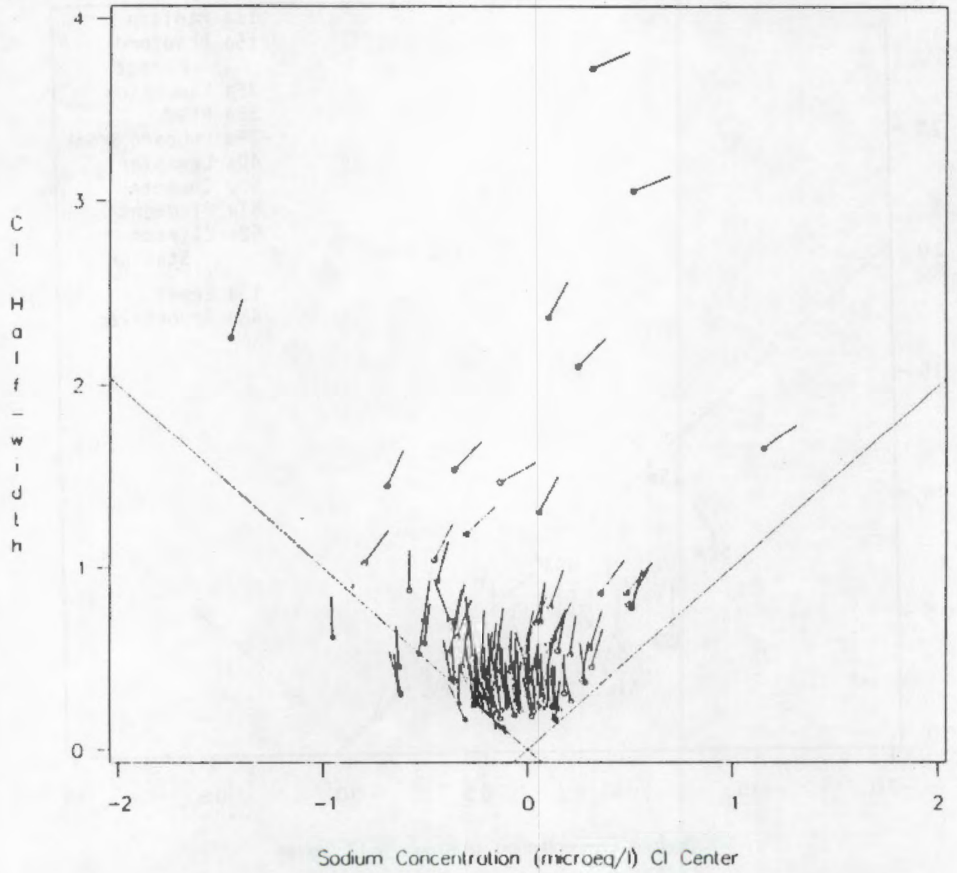


FIGURE A.61. Sodium Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Sodium Deposition Trend Estimate (meq/sq m)

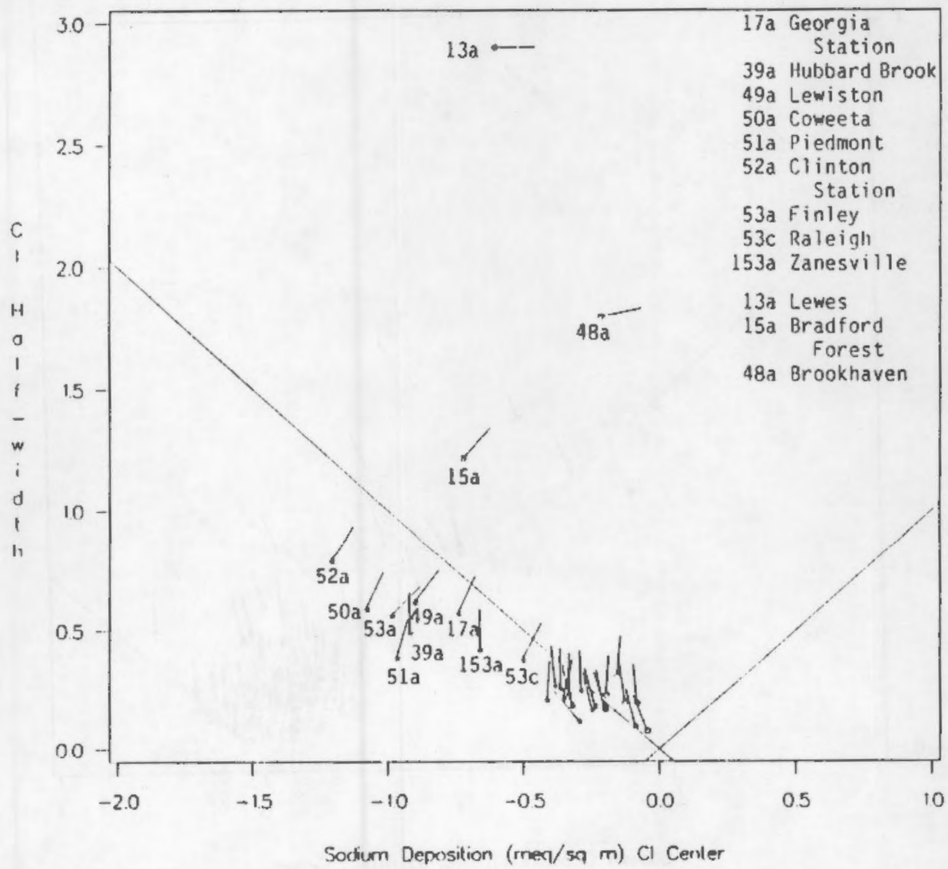
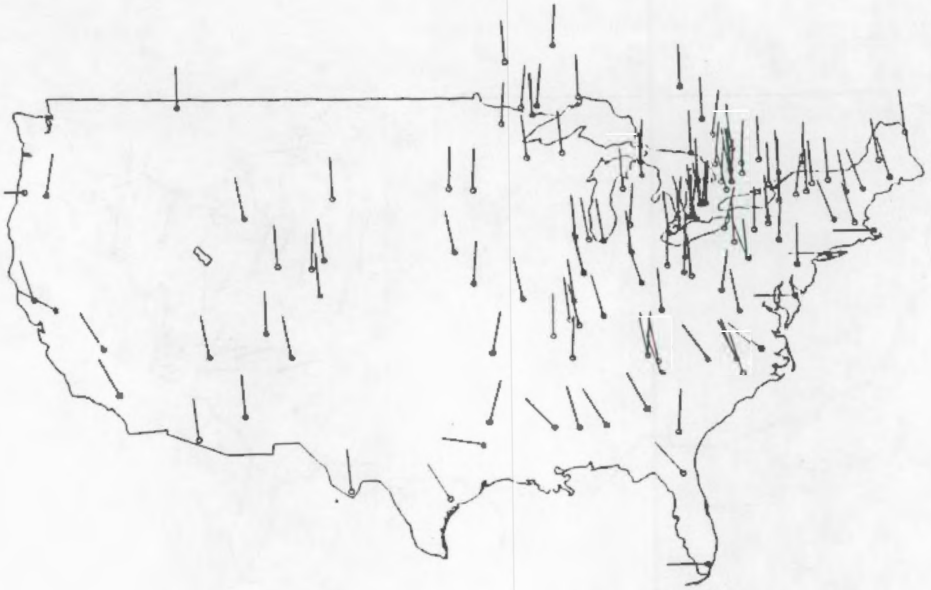


FIGURE A.62. Sodium Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Sodium Deposition Trend Estimate (meq/sq m)

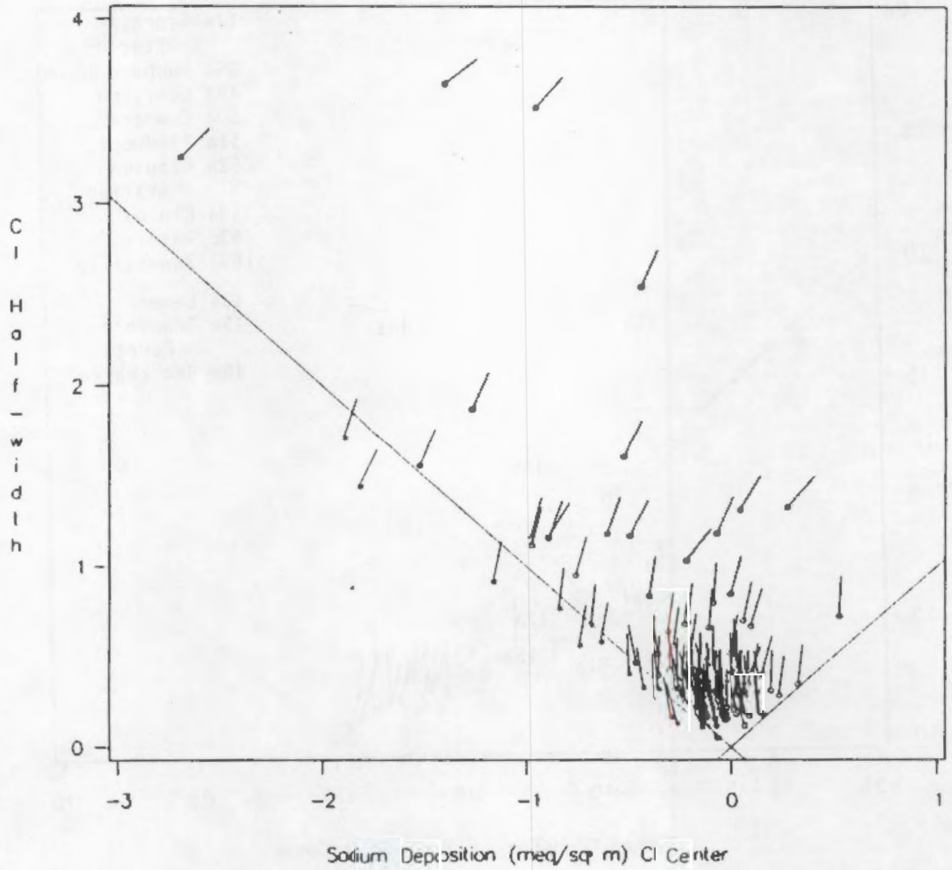


FIGURE A.63. Sodium Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

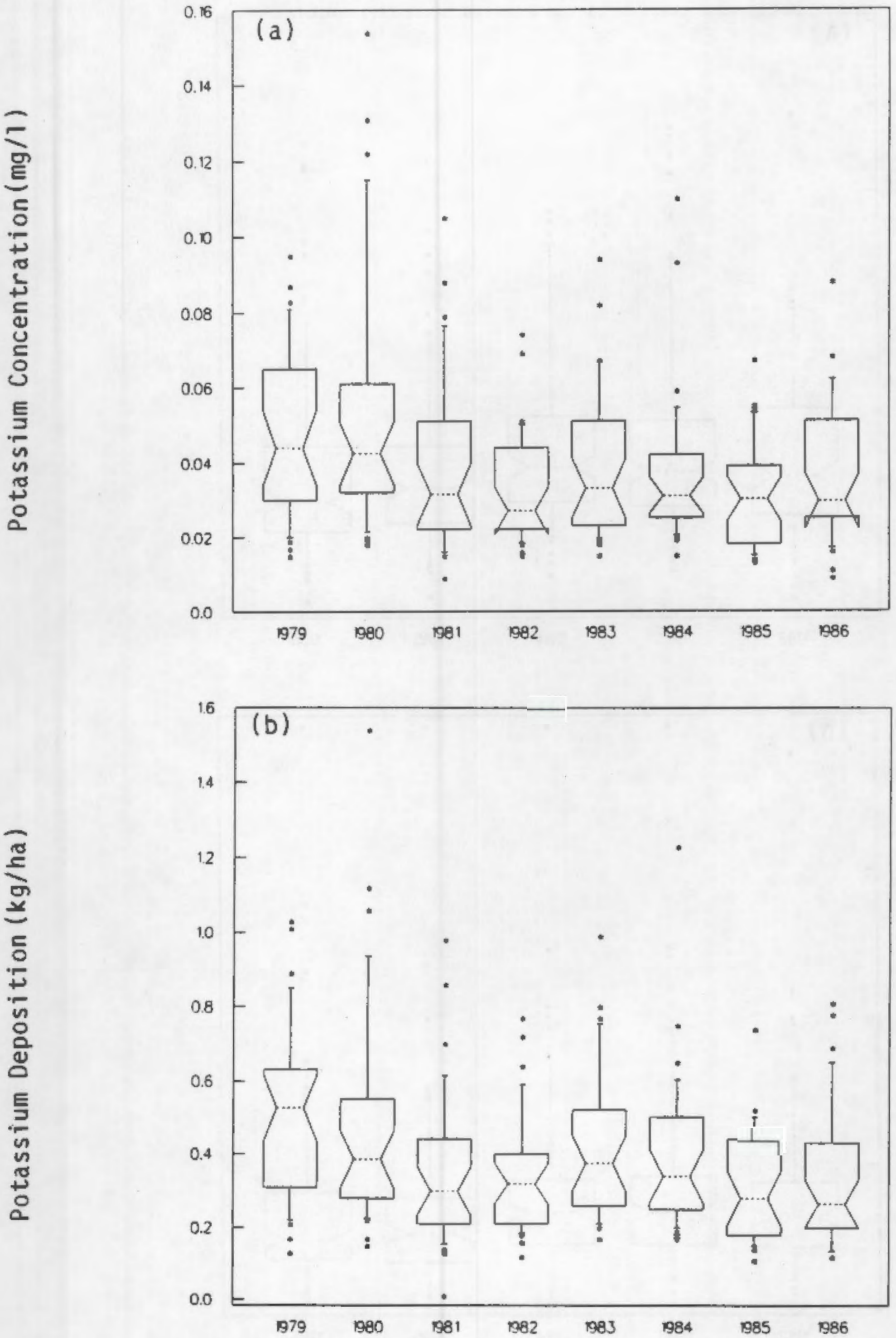


FIGURE A.64. North American Temporal Pattern of (a) Annual Precipitation-weighted Potassium Ion Concentration, and (b) Annual Potassium Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

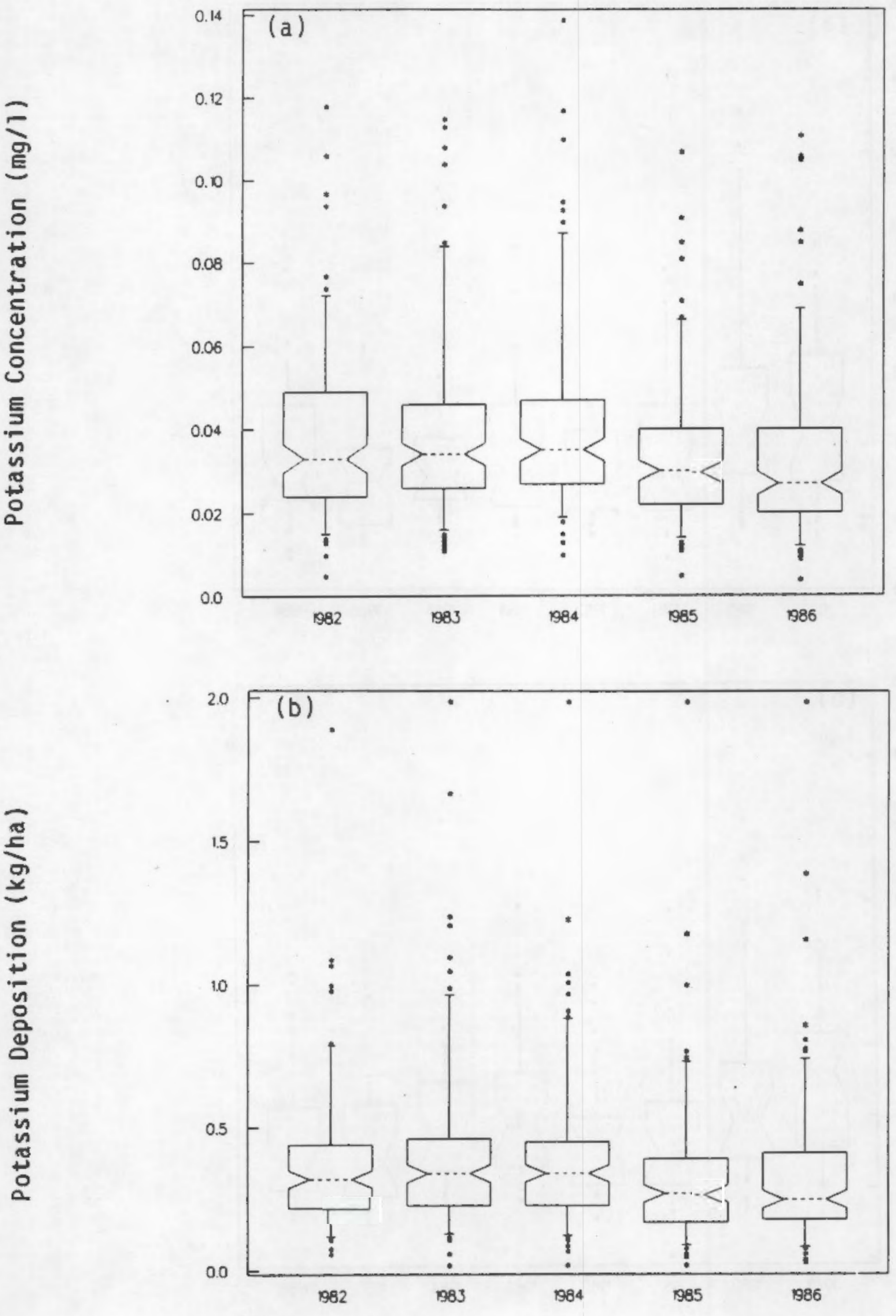


FIGURE A.65. North American Temporal Pattern of (a) Annual Precipitation-weighted Potassium Ion Concentration, and (b) Annual Potassium Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Potassium Concentration Trend Estimate (microeq/l)

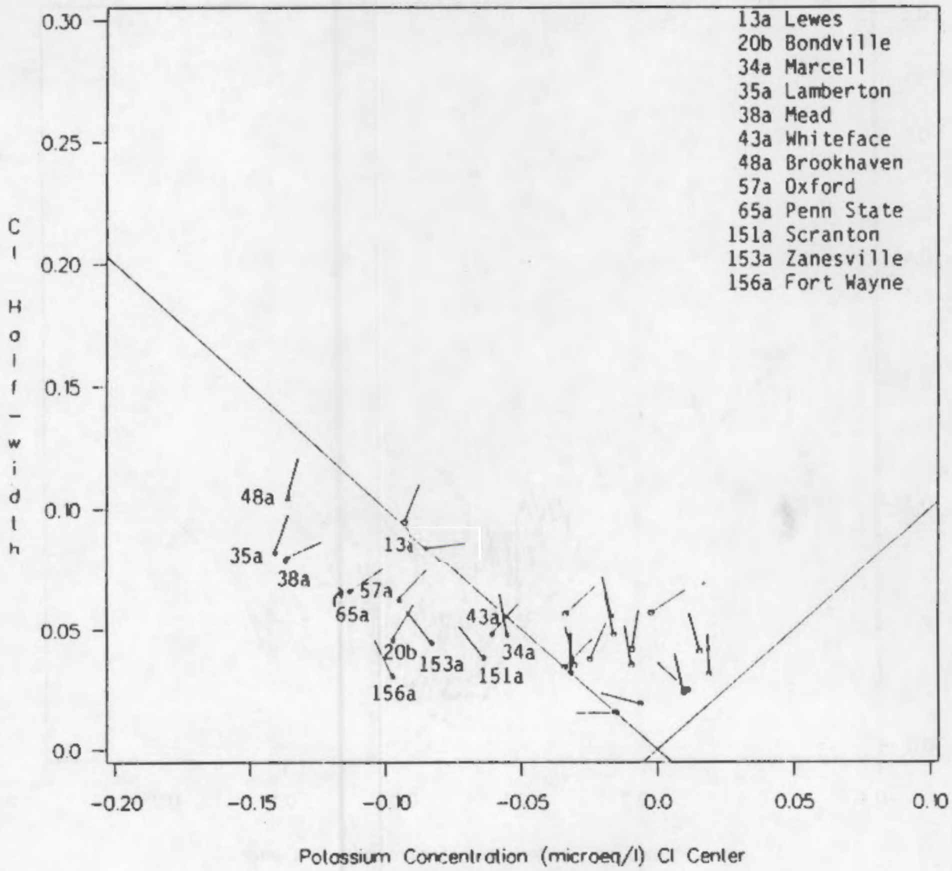


FIGURE A.66. Potassium Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Potassium Concentration Trend Estimate (microeq/l)

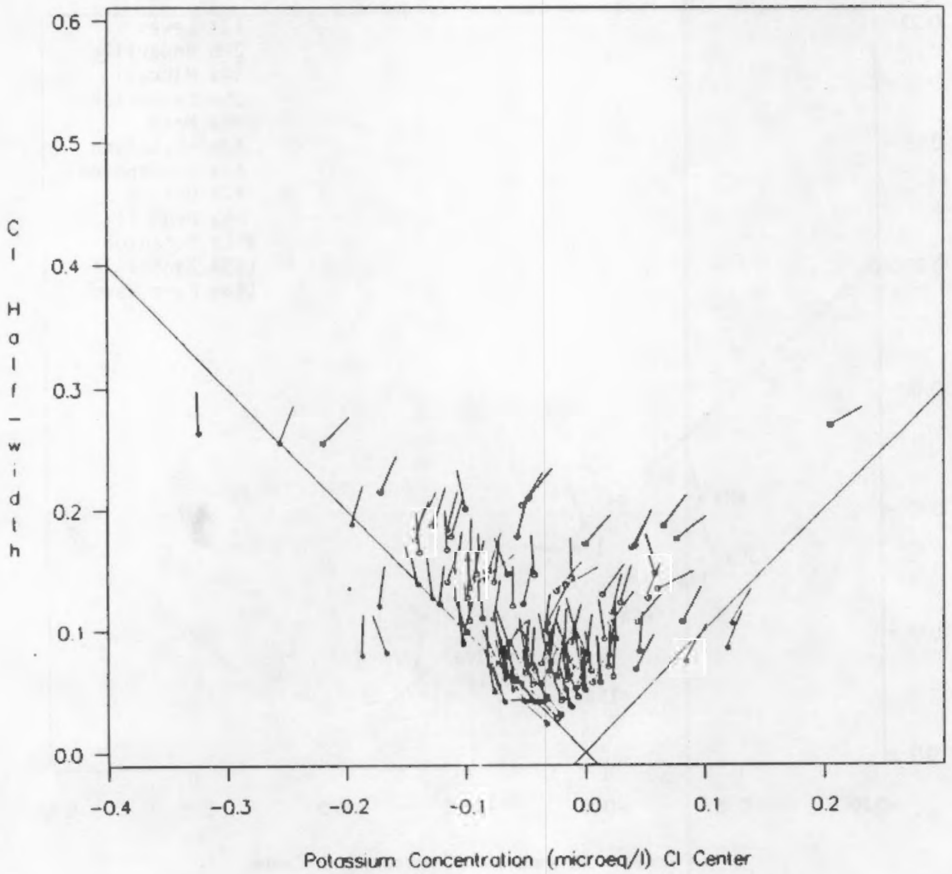


FIGURE A.67. Potassium Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Potassium Deposition Trend Estimate (meq/sq m)

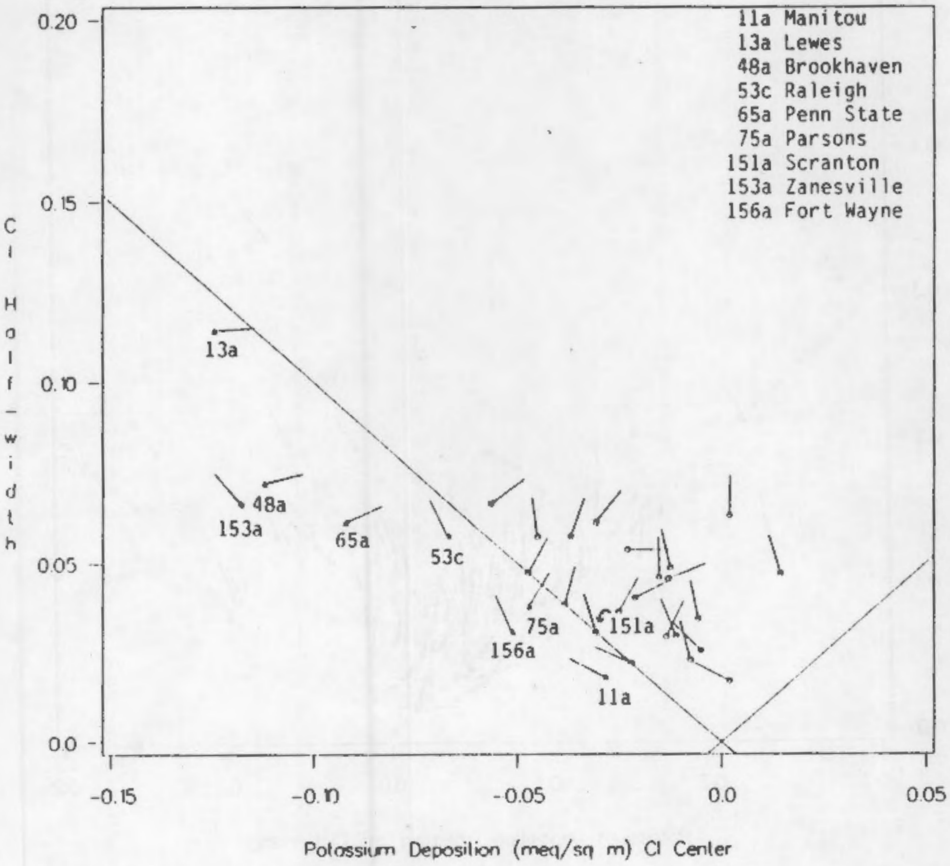


FIGURE A.68. Potassium Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

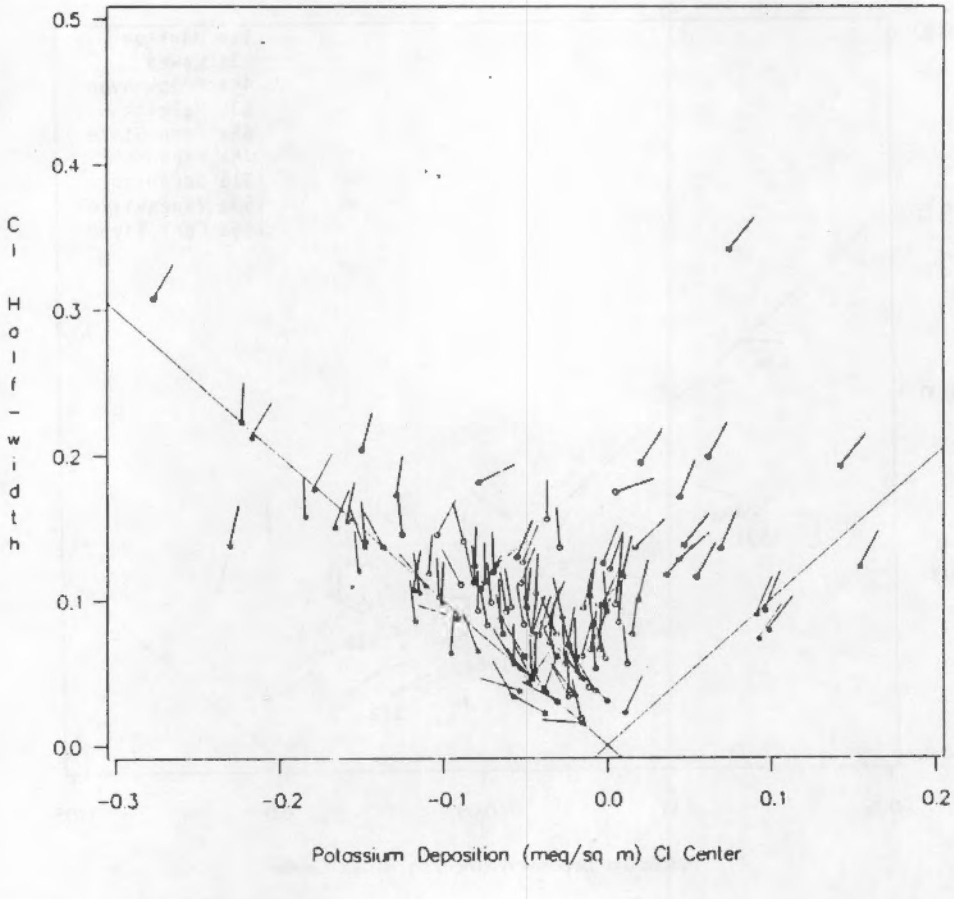


FIGURE A.69. Potassium Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

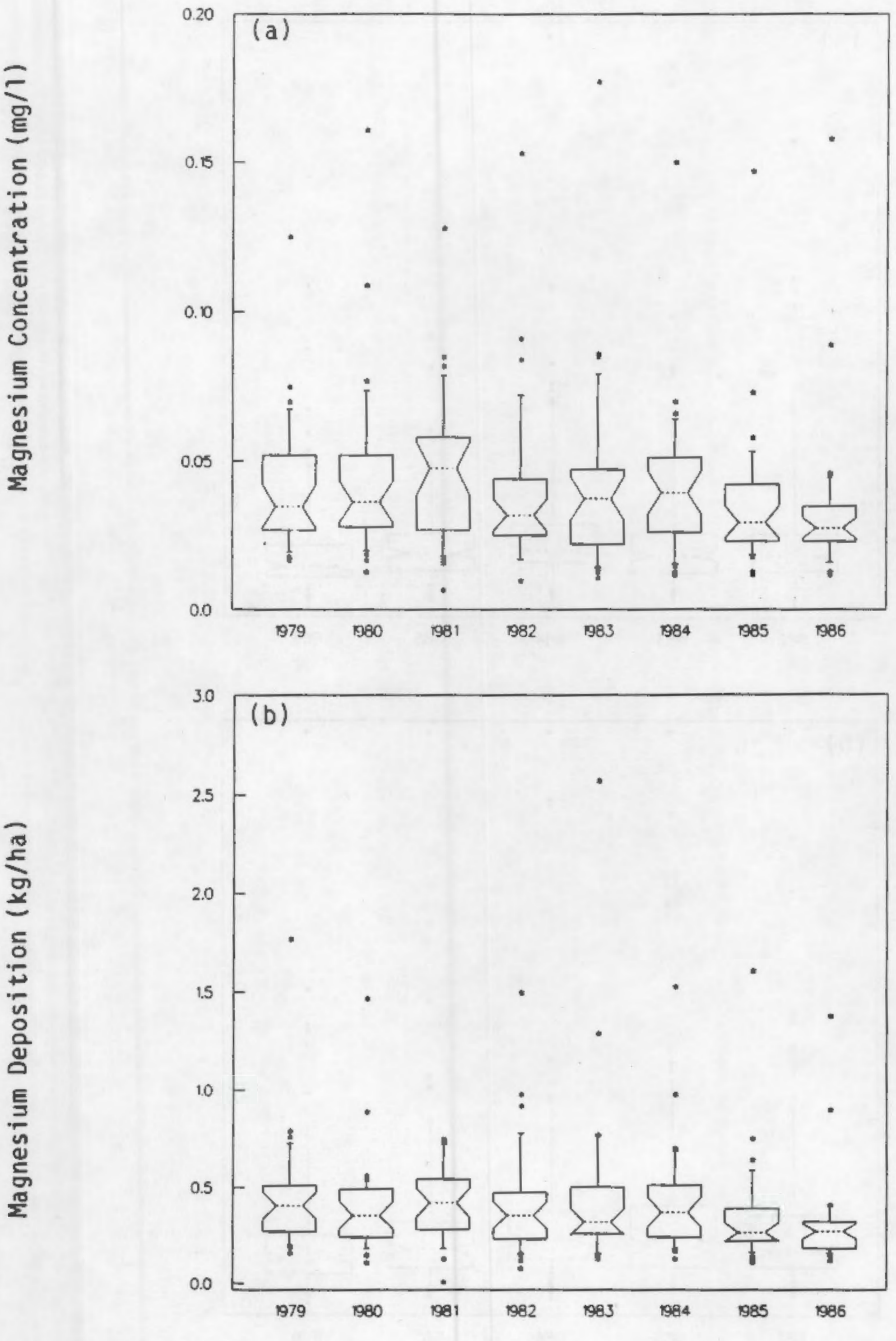


FIGURE A.70. North American Temporal Pattern of (a) Annual Precipitation-weighted Magnesium Ion Concentration, and (b) Annual Magnesium Ion Wet Deposition. Boxplots are the 10th, 25th, 50th, 75th, and 90th Percentiles of Sites in 1979-86 Trend Subset.

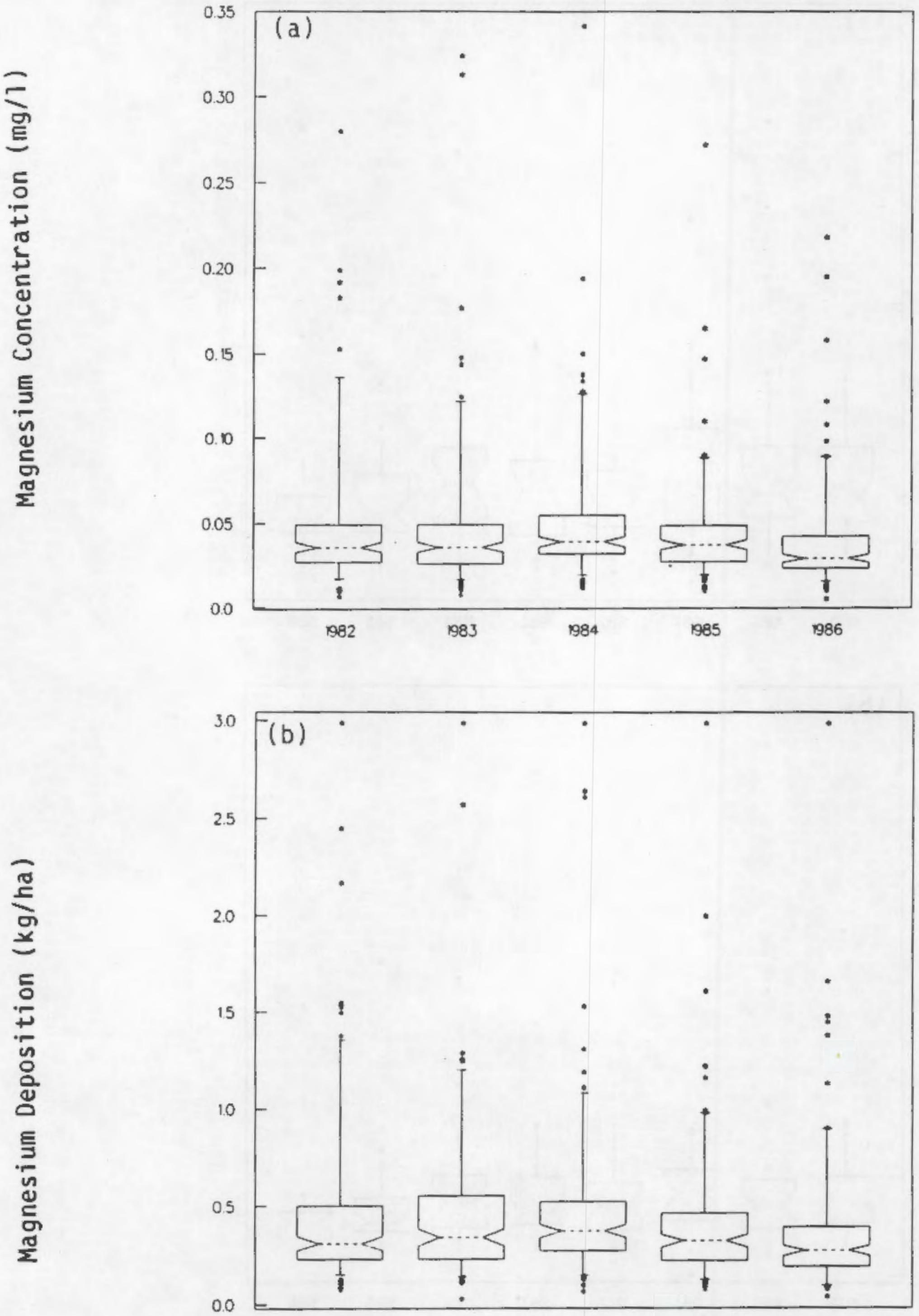


FIGURE A.71. North American Temporal Pattern of (a) Annual Precipitation-weighted Magnesium Ion Concentration, and (b) Annual Magnesium Ion Wet Deposition. Boxplots are the 5th, 25th, 50th, 75th, and 95th Percentiles of Sites in 1982-86 Trend Subset.

Magnesium Concentration Trend Estimate (microeq/l)

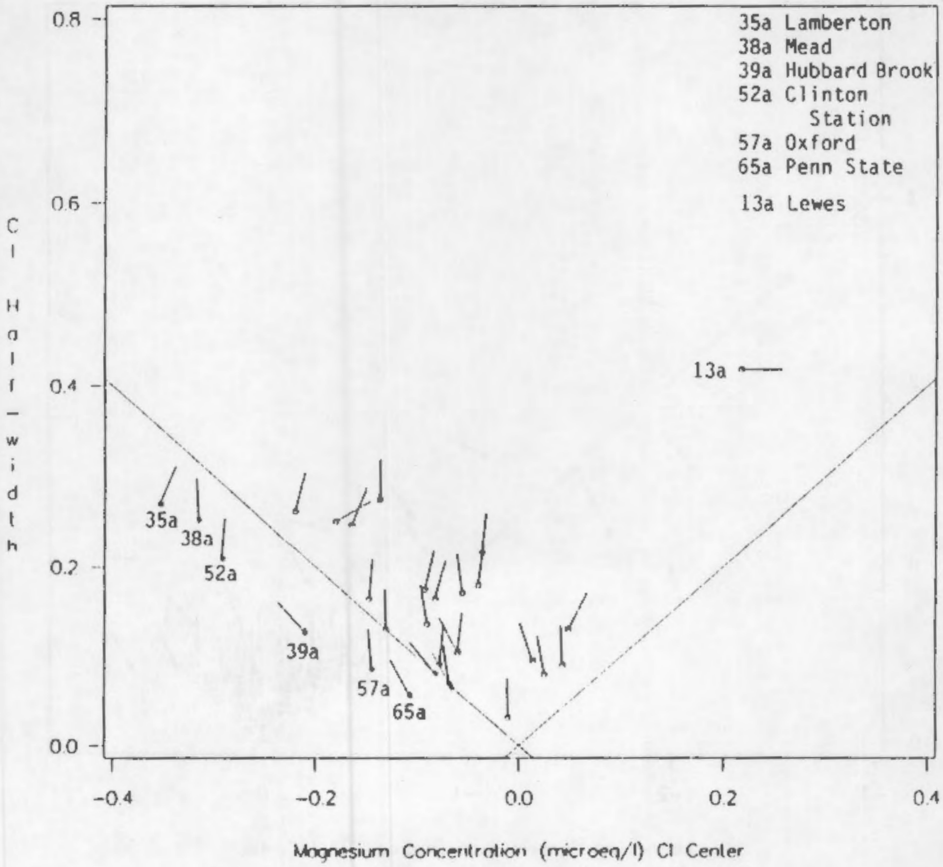
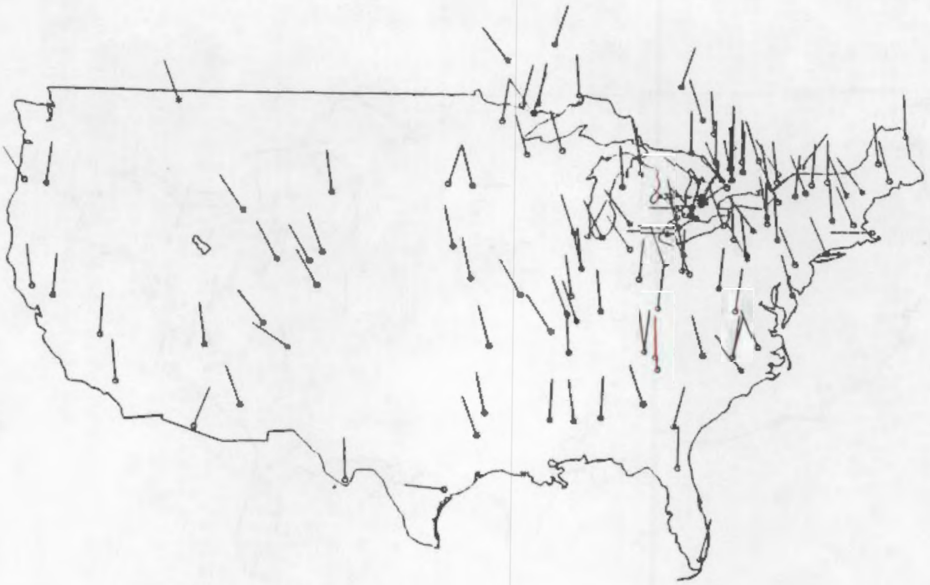


FIGURE A.72. Magnesium Concentration Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Magnesium Concentration Trend Estimate (microeq/l)

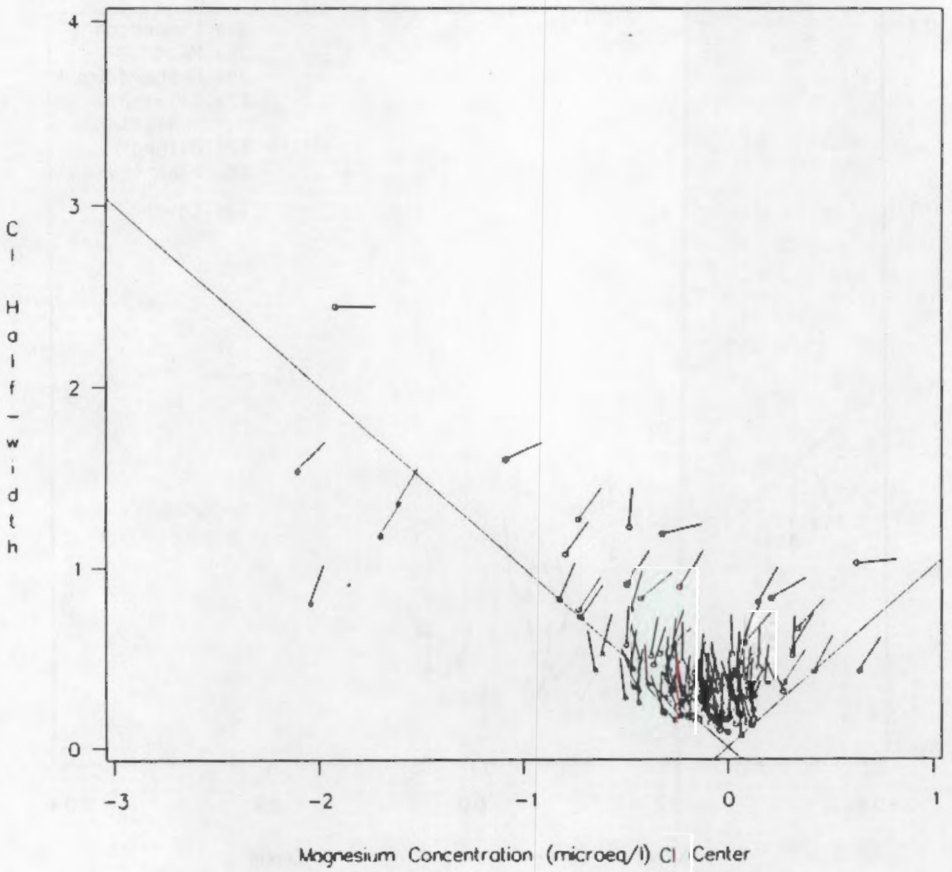


FIGURE A.73. Magnesium Concentration Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$).

Magnesium Deposition Trend Estimate (meq/sq m)

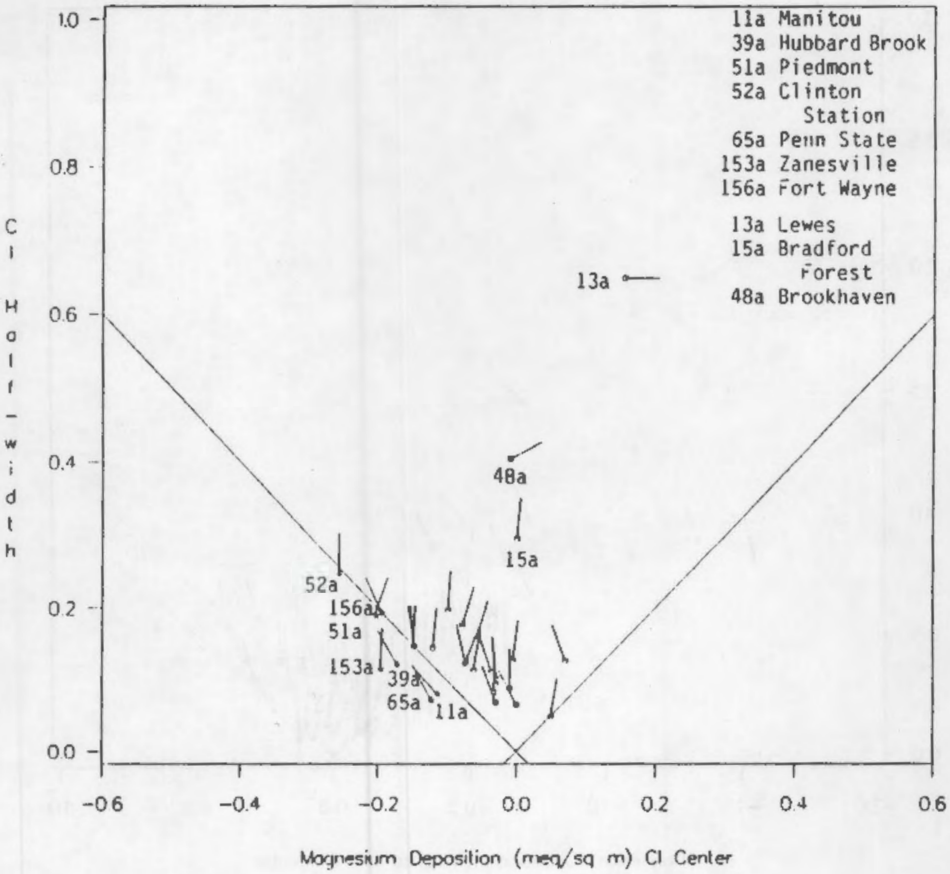
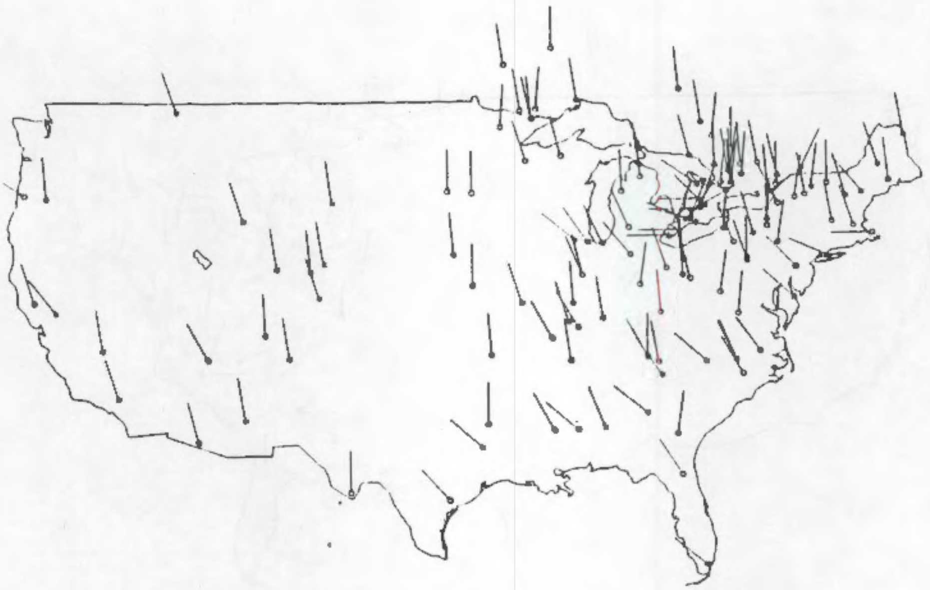


FIGURE A.74. Magnesium Deposition Trend for 1979-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).



Magnesium Deposition Trend Estimate (meq/sq m)

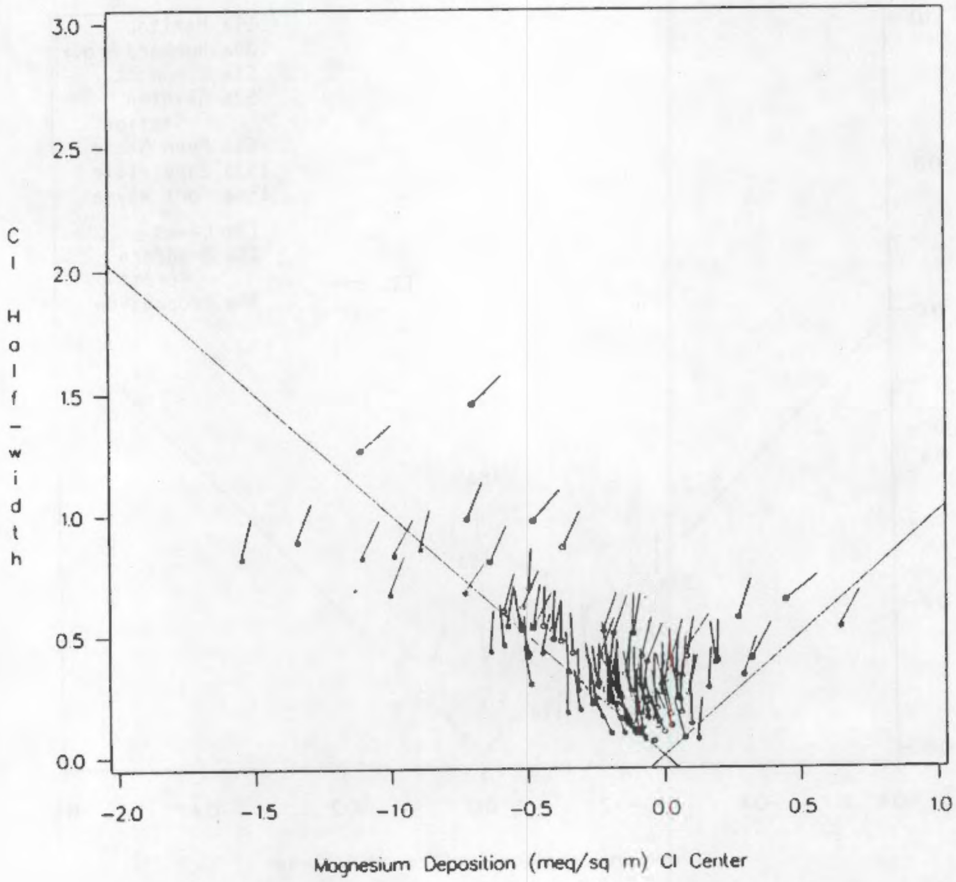


FIGURE A.75. Magnesium Deposition Trend for 1982-86 Trend Sites. Map Ray Angle Proportional to Trend Estimates. Plot Ray Angle Proportional to Site 1986 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$).

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