1987 Wet Deposition Temporal and Spatial Patterns in North America

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March 1990

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ABSTRACT

The focus of this report is on North American wet deposition temporal patterns from 1979 to 1987 and spatial patterns for 1987. The report investigates the patterns 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 which includes 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).

Mosaic maps, based on surface estimation using kriging, display concentration and deposition spatial patterns of pH, hydrogen, sulfate, nitrate, ammonium, and calcium ion species for 1987 annual, winter, and summer periods. The temporal pattern analyses use a subset of 39 sites over a 9-year (1979-1987) period and an expanded subset of 140 sites with greater spatial coverage over a 6-year (1982-1987) period. The 9-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 exhibit substantial inter-annual variation during 1979-1987. 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 ($p < 0.05$) decreasing trends at one or more sites. In particular, 18% and 28% of the 39 sites have significant decreases for sulfate concentration and deposition, respectively. However, greater than 15% of the sites also have significant decreases for calcium,

chloride, sodium, potassium and magnesium concentration and deposition. Nitrate, ammonium and hydrogen ion have the fewest sites with significant trends. The most striking feature of the trend analysis is that over 75% of the sites have linear trend estimates for concentration and deposition indicating a decreasing trend during the 1979-87 period for sulfate, calcium, chloride, sodium, potassium, and magnesium. Nitrate, ammonium and hydrogen ion show little or no indication of a trend. Sulfate concentration had the most extreme median linear trend estimate, $-0.91 \mu\text{eq/l/year}$ (or -2.2% per year using 1987 as base year). Sulfate deposition also had the most extreme median linear trend estimate, $-0.88 \mu\text{eq/l/year}$ (or -3.0% per year using 1987 as base year).

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1.0 INTRODUCTION

The focus of this report is on the temporal and spatial patterns of wet deposition occurring in the United States and southeastern Canada. The report gives a descriptive statistical summary of wet deposition in terms of ion species concentration and deposition. Mosaic maps display 1987 annual and seasonal (winter and summer) spatial patterns for pH, hydrogen, sulfate, nitrate, ammonium, and calcium wet deposition. The intent of the mosaic 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 1979-1987 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 6-year time period, 1982-1987, 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 introduction to the report and reviews related studies. 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 during the 9-year and 6-year periods and the spatial pattern in 1987 for five of the ion species. In this section, the ion species are considered one-at-a-time. Because the number of tables and figures used is large, they are placed together in the appendix. Section 4 contains a study of ion species temporal patterns based on a modified Kendall seasonal tau analysis of four-weekly data. In this section, the nine ion species are considered simultaneously.

1.1 PRIOR SPATIAL PATTERN AND TREND STUDIES

Several studies have examined current spatial and temporal patterns. 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.

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. 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 concentration 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) examined 19 sites from NADP/NTN over the period 1978 through 1983. They 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/year. 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% per year at most all 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) 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. They present results from 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 for this study. 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/year. 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.

Endlich et al. (1988) investigated the spatial and temporal pattern of three daily sampling networks that included the UAPSP, the MAP3S, and the Wisconsin Acid Deposition Monitoring Program (WADMP). The study spanned the

period 1979-1983, with the combined UAPSP and MAP3S sites numbering 6 to 20 for any one year. In 1982, the WADMP added three more sites to the pooled data. Data were screened to create a single, uniform daily data set by examining data quality flags and codes, with the UAPSP screening program as the standard for data acceptance. Kriging was used for spatial analysis as discussed by Eynon (1988). On the basis of the fitted semi-variograms, kriging interpolation plots were produced for pH, sulfate, nitrate, and ammonium for 1979-1983. In the northeast where most monitoring sites were located, the region of highest acidity (lowest pH) for 1982 extended from approximately St. Louis, along the Ohio River Valley, across Pennsylvania and New York, and to New England. The patterns in other years were similar. For 1982 (and other years), sulfate and nitrate patterns were quite similar to those for hydrogen ion. The patterns for nitrate had the highest values in the northwest part of the region and extending southward toward the Ohio River Valley and eastward over the Great Lakes.

Seilkop and Finkelstein (1987) provided precipitation chemistry patterns and trend for eastern North America (roughly east of the Mississippi) for 1980-1984. Data were obtained from the ADS data summaries for the NADP/NTN, MAP3S, CANSAP, APN, UAPSP, and APIOS wet deposition networks. Data were screened using the data completeness measures described in Section 2 for annual summaries. However, no quarterly criteria were applied and a 75% criteria was required rather than 60%. Isopleth maps were used to provide characterizations of concentrations and wet deposition of sulfate, nitrate, and hydrogen ion. The authors used simple kriging to interpolate site values for isopleths. The individual annual sulfate spatial maps for the study period showed that the isopleths generally had a SW to NE major axis with a maximum centered between northern Ohio and southern Ontario, in the vicinity of Lake Erie. The spatial plots also showed a slight decrease in sulfate deposition between 1980 and 1983, with a slight increase in 1984. Both the peak levels and the generally higher levels around the peak decreased by about 15% in 1980-1983. The authors noted that levels far from the peak (considered background) did not seem to change from year to year. Except for 1984, the sulfate concentration patterns were quite consistent with deposition patterns

for all years. This consistency between deposition and concentration was also evident for hydrogen ion and nitrate. The nitrate patterns were generally quite similar to those of sulfate, but smoother. During the 1980-1983 period, nitrate also decreased by 15-20% in areas of high deposition and concentration. However, the nitrate maximum was displaced slightly east and north of the sulfate maximum. The hydrogen ion deposition maps showed the same general shape and temporal change as sulfate, but the maxima were shifted south and east of both the sulfate and nitrate maxima. The authors discussed the observed trend with regard to precipitation and emission trend. The absence of a precipitation-based explanation for deposition decreases does lend support to a hypothesized relationship between observed decreases in hydrogen ion, sulfate, and nitrate during 1980-1983 with reported reductions in emissions. However, the 15-20% reduction in ion concentrations must be viewed with caution, as the authors pointed out, because in a large part of the area of maximum deposition, the coefficient of variation (related to the location of isopleth lines) was approximately 15% in 1982 and 20% in 1980.

1.2 SPATIAL INTERPOLATION METHODOLOGY REVIEW

Wet deposition researchers use several different methods of spatial interpolation to investigate wet deposition spatial patterns. A major objective is the production of a map display that allows the visualization of the spatial pattern. Geographic mapping is one way to summarize data collected at geographically dispersed monitoring sites, to graphically "extend" the data, and to enhance qualitative assessment of the pattern. Three types of map display predominate in the literature for displaying the results: isopleth maps, mosaic maps, and 3-D perspective maps. The display choice is not dependent on the interpolation technique (with the possible exception of hand-drawn maps) but does involve a choice on what is the best display to convey the information on the spatial pattern. The choice depends on the intended purpose for the display.

Bilonick (1985) gives his view on criteria that may be expected for any reasonable spatial interpolation technique. "The method should (1) provide

estimates that are "close" to the actual (unknown) intersample values, (2) provide some idea of the reliability of its estimates, (3) incorporate as much objective information as possible (both directly and from the sample data and other sources) in producing the estimates, (4) be consistent (everyone using the technique with the same assumptions should produce essentially the same map) and (5) not be too costly." It is difficult, if not impossible, for any current technique to satisfy completely all of the criteria. These criteria should be kept in mind when techniques are discussed or are claimed to be optimal in some sense.

1.2.1 Categories of Alternative Techniques

The number of alternative techniques available for spatial interpolation is large and it is convenient to categorize them into three classes (following Burrough 1986): subjective or expert judgment, deterministic, and stochastic. Creutin and Obled (1982) use an alternative classification system. Their categories are named nearest neighbor, arithmetic mean, spline-surface fitting, so-called optimal interpolation, kriging method, and interpolation based on empirical orthogonal functions. In addition, they present terms that help distinguish essential characteristics of interpolation. These are statistical versus deterministic, spatial versus climatological, interpolation versus smoothing, and local versus global methods. Although these distinctions are useful, they are discussed only peripherally here.

Subjective or expert judgment interpolation results in generation of isopleth maps by visually interpolating point values on a mapped surface by generating lines associated with values of equal magnitude on the surface. Cartographers have used this procedure as early as in the 1920s. The "quality" of the interpolated surface depends upon the skill of the person generating the map and their ability to qualitatively incorporate their knowledge of concomitant information into the placement of the isopleth lines.

Deterministic techniques arose from surface fitting techniques, typically from the field of numerical analysis. The number of deterministic procedures is large and computer algorithms for their implementation are readily

available. Deterministic techniques fit the data exactly, implying that each datum is known exactly, i.e., no sampling or estimation error for datum is present. Techniques include assignment of nearest neighbor (also termed proximal or Thiessen polygon), bilinear interpolation, quadratic interpolation, spline fitting, local moving average, inverse distance-squared weighting (and other distance weighting functions, trend surface fitting, and many others. Typically, these procedures are easily computerized and require minimal computing resources. Hence they are convenient for quick preparation of interpolated surfaces and their subsequent display. Contouring algorithms rely mainly on one or more of these techniques. Trend surface fitting techniques do accommodate interpolation when data is known with error but they do not incorporate information on any spatial correlation structure, hence they are considered deterministic.

Stochastic techniques incorporate information on the stochastic structure of the data and typically use empirically determined spatial correlations (or their equivalent). Optimal interpolation developed by Gandin (1965) uses spatial covariance or correlation as the basis for interpolation. The fitted theoretical spatial correlation function is used to produce a weighting function for each point to be estimated such that the mean square error of the estimate is minimized. Optimal interpolation has been developed mainly by researchers in meteorology and is suggested for use by the World Meteorological Organization (1970). Additional research in this area has been done by Rodriguez-Iturbe and Mejia (1974) and Lenton and Rodriguez-Iturbe (1977). Kriging, developed by Matheron (1971), is a spatial interpolation technique used extensively in geostatistics and relies on the concept of spatial variance (equivalent in concept to spatial covariance). It results in a system of simultaneous equations whose solution produces interpolation weights based on a model of the covariance structure associated with the data. The spatial variance function is known as the semi-variogram and a unified, consistent methodology exists for fitting theoretical semi-variograms to empirical semi-variograms. In most cases, kriging is applied locally by using a globally defined semi-variogram and a local neighborhood of data points for the interpolation. Under assumptions that include the

"intrinsic hypothesis", the kriging estimator is the best linear unbiased estimator. The simple, linear kriging method has been extended to handle trends (universal kriging) and non-linear estimation problems (conditional expectation and disjunctive kriging). Details may be found in David (1977), Journel and Huijbregts (1978), Olea (1975), Ripley (1981), and Clark (1979).

1.2.2 Expert Judgment and Deterministic Technique Applications

The spatial patterns of wet deposition over areas of North America have been developed using expert judgment or deterministic techniques by several researchers. Cogbill and Likens (1974) produced a 1955-1956 map of volume-weighted, annual data on hydrogen ion concentration and a 1965-66 map of precipitation pH. Subsequently, Stensland and Semonin (1982) re-examined the data for 1955-56, prepared another isopleth map, and compared it with a map based on 1978-1980 data from the NADP network. Although not stated, it appears the latter maps were generated by a deterministic-based computer algorithm. Likens and Butler (1981) summarized prior work by preparing modified maps and compared it to the spatial pattern over eastern North America for 1975-76. They state the latter map was essentially the same whether based on computer drawn map using a surface fitting algorithm or based on subjectively drawn contours. Barrie and Hales (1984) developed hand-drawn isopleth maps of the mean annual concentration and deposition of hydrogen, sulfate, nitrate and ammonium ions in precipitation in North America for 1980 based on Canadian and U.S. monitoring network data. They describe the process used in developing the location of the hand-drawn contours and give a qualitative analysis of their uncertainty. Munger and Eisenreich (1983) produced computer interpolated maps of average sulfate concentration over the U.S. and Canada for 1970-80. No attempt was made to determine map reliability. Galloway and Whelpdale (1980) developed an isopleth map of 1977 average sulfate deposition for eastern Canada but did not state how the map was generated. Pack (1980) prepared hand-drawn contours for average sulfate concentration for the period August 1978 to June 1979. The isopleths are based on 17 sites from two networks (EPRI/SURE and MAP3S) and an attempt was made to statistically justify combining the two networks. Zemba, Golomb, and Fay (1988) consider the spatial pattern of four-year (1982-85) averaged

wet sulfate and nitrate deposition over eastern North America. Their patterns are depicted as contour maps produced by the NCAR computer graphics software (McArthur 1983). The algorithm interpolates irregularly spaced data to an internal grid from which it uses splines under tension to construct contours. The NADP network prepares an annual data summary report (e.g., NADP 1987) that includes isopleth maps prepared using a distance weighted interpolation algorithm. The algorithm, and its user-determined parameters, was selected only after the computer generated contours were qualitatively compared with hand-drawn contours prepared by network staff (Bowersox, personal communication).

It is apparent from the number of papers discussed, and many additional papers not discussed, that subjective and deterministic interpolation methods are widely used in presenting wet deposition patterns over North America. This may in part be attributed to the little to moderate effort required to prepare an isopleth map. Another factor may be related to the amount of information the authors desired to convey. Typically the discussion of the information content of the maps emphasized the broad general features of the spatial pattern displayed and not its detailed structure. Only broad qualitative comparisons among alternative maps were presented. Without direct discussions with the authors, it is not possible to determine if the level of information content was sufficient for their planned intentions or if the subjective quality of the maps limited their ability to give detailed spatial interpretations or comparisons.

1.2.3 Stochastic Technique Applications

Most applications of stochastic techniques to spatial interpolation of wet deposition data have used kriging. One exception is Gatz and Naiman (1980) who used the optimal method developed by Lenton and Rodriguez-Iturbe (1977). They examined the spatial variability of rain water impurities in mesoscale events. Finkelstein (1984) and Eynon and Switzer (1983) published two early applications of kriging methodology to wet deposition data.

Finkelstein used annual average precipitation-weighted concentrations from July 1979 to June 1980 to investigate the application of simple kriging

to wet deposition. Using polynomial functions as models for fitting semi-variograms to the annual data and addressing the behavior of the semi-variogram at distances near zero by analyzing a second small scale research network, isopleth maps for hydrogen and sulfate ions were presented. The uncertainty in the location of the isopleth line was expressed by presenting upper and lower "confidence isopleths" derived by adding and subtracting the estimated kriging variance from the estimates and contouring the results. Subsequently, Seilkop and Finkelstein (1987) applied the same simple kriging procedures in a study of wet deposition patterns and trends in eastern North America from 1980 to 1984. In both cases, kriging was applied locally with a globally determined semi-variogram. They state that estimation bias is not serious under moderate spatial trends (Journel and Huijbregts 1978).

Eynon and Switzer (1983) using daily data from the EPRI/SURE nine station network developed a spatio-temporal stochastic model that included deterministic components for spatial, temporal, and measurement variation. The fitted autocorrelation structure was then used as a semi-variogram in kriging to produce isopleth maps of seasonal and rainfall adjusted yearly average pH over the eastern United States.

Bilonick (1983, 1985) studied the spatial and temporal patterns of hydrogen and sulfate ions in the northeastern United States based on bulk precipitation samples collected by the U.S. Geological Survey from 1965-1979. His approach for hydrogen ion was to estimate semi-variograms for each month from 1965-1979 and then pool the monthly semi-variograms to produce average monthly semi-variograms. The latter was necessary due to limited data available each month. Rather than applying kriging to obtain point estimates to contour, he applied kriging to produce monthly block estimates for 80 km² blocks. These estimates and estimation variances are combined to form yearly block estimates and variance estimates. Although the possible impact of spatial anisotropy was recognized, insufficient data was available to segregate the data to produce directional semi-variograms. Theoretical spherical semi-variograms with a "nugget" effect were empirically fit. The block spatial estimation resulted in a rectangular array of 12 block estimates that were displayed by presenting a numerical value of the estimate.

This is a simplified version of a mosaic map display. The subsequent paper studied sulfate concentration and deposition space-time patterns by estimating a joint space-time semi-variogram. Because of limited number of sites, a three-dimensional semi-variogram that accommodated spatial anisotropy was simplified to a two-dimensional spatially isotropic model. The model included additive terms for a nugget effect, a cosine model for seasonal effect, a spherical model in time for small distances and a linear model in space. Simple kriging was then used to obtain block estimates as before. The spatial pattern was displayed using gray-scale shading of the block estimates superimposed over a gnomonic projection of the northeast. Using a second data set demonstrated the use of universal kriging when drift (spatial trend) is present. The resulting block estimates are displayed as a mosaic and qualitatively compared to a previously prepared hand-drawn isopleth map (Peters and Bonelli 1982). Bilonick concludes that the universal kriging and hand-drawn maps agree in broad outline with respect to the general level of sulfate deposition and tendency for deposition to increase from north to south. Differences are attributed to use of different map display techniques and different initial data screening. Additionally some smoothing appeared to also have been caused by kriging.

A series of reports (Olsen and Watson 1984; Olsen and Slavich 1985, 1986; Sweeney and Olsen 1987, 1989) from the Acid Deposition System (Watson and Olsen 1984) include annual precipitation-weighted concentration and deposition isopleth maps for hydrogen, sulfate, nitrate, ammonium, and calcium ions over North America. Each isopleth map is developed (1) using a common data screening criteria to determine which sites are included, (2) a theoretical isotropic semi-variogram (typically spherical with nugget effect) fitted to empirical semi-variogram, and (3) local simple kriging to produce point estimates on a dense regular grid (80 x 80 km for earlier years and 40 x 40 km in later years). The estimates on the regular grid were then contoured using a simple bilinear interpolation, with the final maps edited for unrealistic contouring edge effects. Isopleth maps presented by Barchet (1987) were prepared by the same group and followed the same procedures.

Endlich et al. (1986) and Eynon (1988) estimated the spatial pattern of precipitation chemistry over the eastern United States. Their studies relied on data from three daily networks: UAPSP, MAP3S, and WADMP (Wisconsin). To perform their kriging analysis, they used the availability of multiple years of data to first remove long-term spatial and temporal trends from year-to-year fluctuations. The least squares fitted function used was a quadratic spatial polynomial plus a linear trend versus time. The residuals from the fitted surface were then used in developing the kriging semi-variogram. They found that a simple linear semi-variogram with a nugget adequately described the spatial covariance structure and that an isotropic form was sufficient. The final estimated surface is constructed by adding the fitted trend surface to the estimated kriging surface from the residuals.

1.2.4 Issues Related To Spatial Interpolation

Prior studies lead to the identification of a number of issues that must be considered when wet deposition data are spatially interpolated and displayed. The issues are related to characteristics of wet deposition occurring in North America.

Fundamental to any application of an interpolation technique is the selection of data to be used in the analysis. This selection depends on the representativeness of the site and the completeness of the data for the period of interest. Criteria for selection have been developed by the Unified Deposition Database Committee (Olsen et al. 1990). When several maps based on the same time period of data are compared, a major reason for qualitative differences is the selection of different subsets of data by the researchers. Simpson and Olsen (1990) completed a study on the effect of using different data completeness criteria on isopleth maps. They concluded that the overall general regional patterns are reasonably insensitive to changes in criteria but that local features can be highly sensitive. In most cases, changes in criteria do not change the estimated concentration or deposition at a site but do impact whether a site is included in the analysis.

Simple kriging essentially requires an assumption of stationarity of the mean and spatial covariance. The spatial patterns of precipitation

chemistry in North America have a persistent spatial trend (non-stationary mean). Most researchers have used local application of simple kriging to address the issue. Venkatram (1988) questions the applicability of simple kriging to the spatial analysis of wet deposition data. He concludes that a primary assumption in kriging of a stationary mean is invalid when looking at wet deposition over eastern North America. Other researchers have also noted the violation and have concluded that the presence of non-stationarity, although limiting, does not preclude the use of simple kriging locally to obtain essentially unbiased estimates. Venkatram states that the local density of sites is likely to be insufficient for such local estimation. He proposes an alternative procedure to deal with non-stationary mean: application of a simple semi-empirical lagrangian model to estimate the non-stationary mean, followed by simple kriging of residuals from the model and observed data. In addition to removing non-stationarity of the mean, the spatial anisotropy observed by others is also likely to be removed. His preliminary study (Venkatram 1989) shows that the use of a deterministic model is likely to improve (smaller error variance) upon simple kriging without the model. Eynon (1988) removes non-stationarity of the mean by fitting spatial quadratic polynomials and then applying simple kriging to the residuals. Venkatram's approach of using other information to remove non-stationarity of the mean and other approaches such as Eynon's deserve additional study.

Annual mean (or median) precipitation chemistry values at a monitoring site are estimates that have an associated variance. Typically, applications of simple kriging have related this error to the nugget in the semi-variogram. Inclusion of a nugget does not explicitly incorporate the known measurement error at the monitoring site, since the kriging estimate at a monitoring site remains the measured value. If spatial correlation is present then nearby sites contain information that should be incorporated into the kriging estimate at the monitoring site. Monitoring site measurement error is included in kriging analyses used for preparing the spatial maps presented in Section 3.

Several researchers (e.g., Granat 1978, Bilonick 1985, Guertin et al. 1988, Vong et al. 1989, Wampler and Olsen 1987) have questioned the appropriateness of direct spatial interpolation of precipitation-weighted mean concentration and deposition. One issue centers on the non-additive character of precipitation-weighted means when the volume of precipitation is not constant over the area being estimated. Guertin et al. argue that deposition and precipitation amount should be spatially interpolated and then derive precipitation-weighted mean concentration by dividing the interpolated deposition by the interpolated precipitation amount. Others argue that the concentration field has smaller spatial variation than deposition and precipitation. Hence its spatial interpolation should have smaller variance estimates. The greater spatial variation of precipitation is addressed by using the greater number of precipitation monitoring stations, rather than relying solely on precipitation amounts from precipitation chemistry monitoring sites. Deposition is obtained by multiplying the interpolated concentration and precipitation amount. Vong et al. (1989), in a review of existing literature and available empirical evidence, conclude that the latter approach appears to be preferred when North American regional patterns are of interest. Further research is required, especially if local features in a spatial pattern are of interest.

Current state-of-the-science spatial interpolation methods applied to precipitation chemistry data have produced spatial isopleth or mosaic maps that have similar broad regional patterns but that differ in representation of local features. Knowledge is insufficient at present to identify a single best spatial interpolation procedure or to provide reliable quantitative associated variance estimates. Further studies are required to address the issues of data selection, non-stationarity of mean and variance, isotropy versus anisotropy, incorporation of measurement uncertainty, and direct versus indirect interpolation of deposition.

1.3 TREND ANALYSIS METHODOLOGY REVIEW

An objective of regional and national network monitoring in North America is the detection and assessment of wet deposition trends. The monitoring

network would, optimally, provide a long unbroken series of measurements, made using the same methods, at regionally representative sites. The record of data would be sufficiently long to cover the time period of interest. For wet deposition, the time period of interest starts in the early 1900s until the present. An extended time period would aid in drawing conclusions by minimizing the likelihood of being misled by short-term trends or drifts in the data series. Using the same methods for measurement, minimizes introduction of uncertainties at discrete time points due to changes in protocols. Documentation of changes and overlap of data collected under new and old procedures are essential. Inference that trends found at a site apply to a larger area require careful study, especially in assessing the regional representativeness of the site. Preferably, several regionally representative sites are available within the same region to enable an assessment of the consistency of their trends to be made.

Trend analyses have used a number of alternative methodologies and data sets. Investigators have used several alternative methods for trend analysis at a site, e.g., graphical plots, regression models, time series models, and non-parametric models. Those investigators who considered trends at multiple sites used several alternatives for combining information from the sites, e.g. comparison of spatial patterns across years, comparison of time plots combining sites, and empirical assessment of trend results. Investigators have used alternative measures for precipitation chemistry: concentration, deposition, or change in relationship between concentration and precipitation amount (all over some averaging period).

A difficulty in comparing available trend analyses occurs as a result of differences in how investigators selected sites for study, the time period included, the trend analysis methodology, and the selected measure for precipitation chemistry. This section gives an overview of some of the alternative trend analysis methodologies.

1.3.1 Trend Methods for a Single Site

Trend graphical plots are an important component of trend analysis for almost all investigators. Plots enable an investigator to visually assess the

temporal pattern of the data, either leading to a more formal model or to a qualitative statement about the presence of a trend. Trend plots may be based on individual wet deposition samples or averaged over a common time (monthly, quarterly, annual). Plots based on time averaged values reduce the visual impact of the typical large variation of individual samples, hence increasing the ability to see seasonal and longer term trends. An alternative to plotting averaged values is to enhance time series plots of individual samples by adding a smoothed model (e.g., lowess nonparametric smooth or regression model) to the plot to aid the visual detection of a pattern. Schertz and Hirsch (1985) make effective use of lowess plots to assess trend patterns at individual sites. Oehlert (1984), also summarized in NRC (1986), used deseasonalized time series plots of monthly volume-weighted concentration and deposition to investigate trends at Hubbard Brook from 1964 to 1979.

A widely used technique for trend analysis is linear regression modeling on individual samples, monthly averages, or annual averages. Typically, the regression model includes terms for a linear long term trend, a cyclic seasonal trend, and a term that adjusts for a postulated effect due to precipitation volume. Each investigator determines whether to include these effects and, if so, how complicated the model term will be. For example, Dana and Easter (1987) analyzed trends for the MAP3S sites from 1976-1983 using a simple linear and a simple linear plus a simple sine periodic function for seasonal effect. Their analyses are applied to individual sample concentrations which are transformed by taking logarithms, to account for the tendency toward log-normal distributions of individual sample concentrations. Linear trends are determined from the regression coefficient for long-term trends, with its significance based on the t-test for the coefficient. Likens et al. (1984) apply simple linear regression on annual volume-weighted averages to the Hubbard Brook data. The trend is computed as the change in annual weighted average, and standard errors for the trends are computed without accounting for autocorrelation between years. Oehlert (1985) notes that multiple regression analyses typically ignore autocorrelation, resulting in standard errors being calculated without appropriately modeling the error structure.

Time series techniques model the data as in regression, but the residuals are allowed to be autocorrelated. Hidy et al., (1984) estimated parametric autoregressive moving average (ARMA) models for lag twelve differences in monthly concentrations for trend analyses of the USGS precipitation chemistry network in New York. Differencing by lag twelve removes seasonality and the residuals are modeled as a constant plus an ARMA process residual, the constant being the average trend per year. Oehlert (1984) investigated trends for Hubbard Brook monthly volume-weighted average concentrations and deposition. Seasonal influences were eliminated by subtracting from each monthly value the mean of the time series for that month. Trends were determined (essentially) by ordinary least squares regression, but the standard errors of the trends are adjusted for the presence of autocorrelation in the residuals. Bilonick and Nichols (1983) approach trend analysis of the USGS data by concentrating on finding a stationary autoregressive integrated moving average model that adequately describes the data. If one can be found, the data are assumed to be stationary and the mean is determined to be a constant, i.e., no trend is present. Oehlert (1985) has criticized this approach.

Schertz and Hirsch (1985) introduced a nonparametric trend analysis method, termed Kendall seasonal tau (KST), in their analysis of trends in weekly NADP/NTN wet deposition data from 1978-1983. Hirsch et al. (1982) and Hirsch and Slack (1984) developed the method for examining trends in surface water quality. The method has two parts, a test for trend based on Kendall's tau and an estimate for trend based on Sen's median slope estimator. The trend estimate is very resistant to outliers and can be estimated in the presence of missing values. The original KST method does not account for autocorrelation, but the modified test does. The latter requires a longer time series (approximately 10 years of monthly data) and autocorrelation that is not too strong. Barchet (1987) presented a trend analysis based on KST for sites operating during 1978-1984 from several North American wet deposition networks. Recently, similar analyses have been completed by Olsen (1989) and forms the basis for the trend analysis in this report.

1.3.2 Trend Assessment for Networks of Sites

Determination of trends at individual sites is only one aspect of trend assessment for North America. Individual sites may reflect influences of local impacts (e.g., changes near the site in emissions or land use) and not reflect trends in surrounding regional area. Current wet deposition networks have time series data for many sites available for assessment of regional and national trends. Investigators have used several alternative methodologies to assess regional trends: changes in distribution of annual concentration for set of sites, comparison of annual spatial patterns, and qualitative analysis of trend results from set of individual sites.

Rodhe and Granat (1984) summarized more than 25 years of data on sulfate in precipitation from the European Air Chemistry Network for assessment of seasonal and long term trends. With data available from 50 to 100 stations, they assessed long term trends using annual values for individual sites and using time series plots that graphically presented summaries across groups of sites. For each group of sites, the 25th, 50th, and 75th percentile across sites for a five-year averaging period were plotted to visually assess presence of trends. Olsen (1989) uses a similar approach to present trends in annual concentration and deposition for subsets of North American sites. He constructs a time series of boxplots that summarize annual distribution for set of sites to visually assess changes (trends) in distribution from over time (see Appendix A for examples). Although this approach summarizes a large amount of data and presents it graphically, it only allows a qualitative assessment of patterns and is likely to be effective in detecting trends when the trends are substantial compared to interannual variation within a site and consistent across sites. The approach does not remove differences in mean level between sites, hence annual distributions reflect this variation.

Another approach to assessing trends, or changes, in annual concentration and deposition for network of sites is to compare the spatial patterns determined for different periods. Likens and Butler (1981) investigated temporal trends in precipitation acidity in eastern North America during 1955 to 1976. They determined the spatial pattern for three different time

periods: 1955-56, 1965-66, and 1975-76 and qualitatively assessed differences among the patterns. Having the data for the three periods collected by three different organizations and having the number and locations of the sites different for each period, limits the ability to assess, even qualitatively, the presence of trends. Using data available from North American networks operating during 1980-1984, Seilkop and Finkelstein (1987) compared changes in annual spatial patterns of sulfate, nitrate, and hydrogen ion concentration and deposition. They used a consistent methodology to calculate annual averages, to determine which sites to include each year, and to estimate and display the spatial pattern for each year. In addition to qualitatively assessing differences in patterns, they developed a method, based on kriging standard errors, to quantitatively determine regions of change between two years. Eynon (1988) completed a similar qualitative comparison across 1979-1983 for eastern North America using data from the UAPSP network. Spatial and temporal trends were estimated and subtracted from the annual values before the correlation structure of the residuals was used to estimate the semi-variogram for the kriging analysis. Bilonick (1985) estimated space-time semi-variograms of sulfate deposition data collected by the USGS during 1965-1979 in New York state and used them to determine the annual sulfate spatial patterns over New York. A qualitative approach was used to assess changes in patterns.

Trend assessment for multiple sites can also be approached by first completing a trend analysis for each site and then combining the trend results to determine regional or national consistency. Schertz and Hirsch (1985) used this approach to summarize their Kendall seasonal tau analyses of NADP/NTN sites. They tabulated and discussed the number of sites with increasing and decreasing trend estimates and with statistically significant increasing and decreasing trends. Olsen (1989) used boxplots of trend estimates for all sites and geographic displays of trend estimates to summarize trend analysis results. A qualitative assessment for regional consistency was included. Further research is needed in combining trend results from individual sites, or in determining an alternative approach.

2.0 DATA PREPARATION AND ANALYSIS METHODOLOGY

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, and to describe the spatial and temporal methodologies.

2.1 SOURCE OF WET DEPOSITION

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 regional and national networks that contribute data to the Acid Deposition System (ADS) (Watson and Olsen 1984) for statistical reporting of North American wet deposition data.

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. The National Atmospheric Deposition Program (NADP) established, in 1978, a weekly sampling network to monitor trends in precipitation chemistry in the United States. The NADP was created, in cooperation with federal, state, and private agencies, 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 22 sites in 1982 under the Utility Acid Precipitation Study Program, giving broader regional coverage. 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

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

<u>Network Abbreviation</u>	<u>Network Name</u>	<u>Start Date</u>	<u>Number of Sites in 1987</u>	<u>Sample Type</u>
MAP3S/PCN	MAP3S Precipitation Chemistry Network	1976	9	Daily ^(a)
NADP/NTN	National Atmospheric Deposition Program/ National Trends Network	1978	204	Weekly
UAPSP ^(b)	Utility Acid Precipitation Study Program	1978	25	Daily ^(a)
CAPMoN/APN ^(c)	Canadian Air and Precipitation Monitoring Network	1978	25	Daily
APIOS-C	Acidic Precipitation in Ontario Study - Cumulative Network	1980	37	Monthly ^(d)
APIOS-D	Acidic Precipitation in Ontario Study - Daily Network	1980	15	Daily

(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 ceased operation on December 31, 1987; on January 1, 1988 the same sites or ones near to the present UAPSP sites began 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.

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 1987 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 69 in 1979 to 315 in 1987. The implementation of the NTN and expansion of the NADP network in responsible for most of the growth in the United States. The initiation of the APIOS and CAPMoN networks in 1980 and 1983 accounts for most of the growth in Canada.

A site is considered to have operated a full year if precipitation (not necessarily precipitation chemistry) is monitored for at least 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 50 in 1979 to 283 in 1987. Figure 2.1 gives the location of sites operating a full year in 1987.

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 that the collection, laboratory analysis, and reporting of wet deposition data is consistent and of known quality. Each network included in this report has and implements a quality assurance program. 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

TABLE 2.2. Number of Sites by Network from 1979 to 1987

Network	1979	1980	1981	1982	1983	1984	1985	1986	1987
NADP/NTN	39	82	97	110	142	177	195	203	204
MAP3S/PCN	8	8	9	9	9	9	9	9	9
UAPSP	18	10	21	22	25	27	26	25	25
CAPMoN	4	6	6	8	17	18	24	24	25
APIOS-D	0	8	16	16	16	17	15	17	15
APIOS-C	0	30	35	36	38	38	38	37	37
Total	69	144	184	201	247	286	306	316	315

TABLE 2.3. Number of Sites by Network that Monitored Precipitation More than 90 Percent of Year from 1979 to 1987

Network	1979	1980	1981	1982	1983	1984	1985	1986	1987
NADP/NTN	22	36	73	89	102	139	179	187	182
MAP3S/PCN	8	8	9	9	9	9	9	9	9
UAPSP	18	6	6	21	20	19	21	24	23
CAPMoN	3	4	5	6	7	17	18	21	20
APIOS-D	0	0	12	16	16	14	14	13	14
APIOS-C	0	0	30	33	32	34	32	36	35
Total	50	54	135	174	186	232	273	290	283

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. (1990) and used in the ADS 1986 summary report by Sweeney and Olsen (1989).

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, December 1 to March 1 for winter and June 1 to September 1 for summer) for spatial analyses and are four-weekly and annual for temporal analyses. The precipitation-weighted mean concentration is calculated as a weighted average of the valid sample ion species' concentration with the weight being the corresponding rain gage precipitation depth divided by the sum of rain gage precipitation in valid samples.

FIGURE 2.1. Geographic Location of Sites in 1987 Operating a Full Year and the Subset of Sites Meeting Annual UDDC Criteria for Sulfate

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. (1990) give additional details of the calculation procedures.

Although calculation of an annual, seasonal, or four-weekly 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. 1990) 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 1986 data summary report by Sweeney and Olsen (1989) 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 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 1987. The number

TABLE 2.4. Number of Sites by Network Meeting the Unified Deposition Database Committee Annual Summary Data Completeness Criteria for Sulfate from 1979 to 1987

<u>Network</u>	<u>1979</u>	<u>1980</u>	<u>1981</u>	<u>1982</u>	<u>1983</u>	<u>1984</u>	<u>1985</u>	<u>1986</u>	<u>1987</u>
NADP/NTN	16	26	53	60	71	85	101	115	101
MAP3S/PCN	5	7	8	9	8	9	9	9	9
UAPSP	16	6	6	21	20	18	19	21	0
CAPMoN	2	2	3	4	4	11	11	17	13
APIOS-D	0	0	0	0	10	10	11	11	10
APIOS-C	0	0	10	14	22	21	20	22	21
Total	39	41	80	108	135	154	171	195	154

meeting the UDDC criteria increased from 39 in 1979 to 154 in 1987. Figure 2.1 shows the location of the subset of sites operating a full year that met the UDDC criteria for 1987 annual sulfate summaries. In 1987, 54% of sites operating a full year met the UDDC criteria for annual sulfate summaries. 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 (January 1 to April 1) is the most frequent cause for a site to fail the UDDC criteria, i.e., application of the quarterly criteria for all quarters. It should also be noted that in 1987 a number of sites failed to meet the UDDC criteria for annual sulfate summaries because of a laboratory analysis problem. It is reassuring that the quality assurance programs do indeed work and suspect data is removed. However, this additional loss of data points out one of the difficulties with long-term studies: over a long enough period of time, there will be gaps in the data which make the analysis of the data much more difficult.

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 the UDDC criteria, and increasing the number of sites available for temporal pattern analysis.

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. An 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. 1990). In an analysis of sites with data in the ADS in 1987, 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 mosaic maps of hydrogen, sulfate, nitrate, ammonium, and calcium wet deposition spatial patterns for 1987 annual and seasonal (winter and summer) summary periods. For each ion species, maps of both precipitation-weighted concentration and total wet deposition are presented, except hydrogen where precipitation-weighted pH is presented in place of concentration. The maps use 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 1987 annual sulfate concentration and deposition mosaic maps is 154 (Table 2.4). Tables A.3 and A.4 give the number of sites used for each map.

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 six or more years. Since the objective of the data criteria is to aid in identifying sites with good quality data, an alternative approach based on a relaxed criteria applied to multiple years is developed.

The modified Kendall seasonal tau statistical trend analyses uses the four-weekly summaries and the method allows missing data. Thus a site was only required to operate from 1979 (or 1982) to 1987 for inclusion in the nine (or six) year trend analyses. For the 9-year trend analyses there are 39 sites (Table A.1) and for the 6-year trend analyses there are 148 sites (Table A.2) that have been operating during the respective time periods. The location of the sites in each of the subsets is presented in Figure 2.2. A four-weekly data completeness is applied to each four-weekly summary for each of these sites. The criteria requires precipitation monitoring to cover at least 75% of the period and at least 60% of the precipitation measured during the period must be associated with valid data. For the 9-year subset, depending on the ion species, there are generally over 100 four-weekly periods, out of 117 total periods, included in the analyses for 31 of the 39 sites. Davis, California (ADS ID 009a) had the fewest valid four-weekly summaries, between 66 and 70, depending on the ion species. The lack of rain in the summer at Davis accounts for many of its incomplete summaries. For the 6-year subset, depending on the ion species, there are generally 65 or more four-weekly periods, out of 78 total periods, included in the analyses for 114 of the 148 sites.

The temporal pattern analyses also compare the annual summaries of the sites across years. These comparisons require that the sites have complete data for all the years. Therefore, a site is required to have valid samples in at least 10 four-weekly periods in every year and additionally at least 8 four-weekly periods in every year satisfy the four-weekly data completeness requirement described above. For the 39 nine-year trend sites, 17 sites have annual summaries for all ion species, 13 more sites have annual summaries for most of the ion species, and 9 sites have no annual summaries. For the

FIGURE 2.2. Locations of Sites Included in Trend Subsets for (a) 1979-1987 and (b) 1982-1987

148 six-year trend sites, 78 sites have annual summaries for all ion species, 41 more sites have annual summaries for most of the ion species, and 29 sites have no annual summaries. Tables A.1 and A.2 indicate which ion species for each site had annual summaries, for the 9-year and 6-year subsets, respectively.

2.4 SPATIAL ESTIMATION METHODOLOGY

Mosaic maps illustrate the spatial distribution of precipitation-weighted mean concentration (or pH) and total wet deposition for the United States and southeastern Canada in 1987. 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 mosaic maps involves: (1) selection of sites that meet data completeness criteria, (2) estimation of the spatial surface based on the selected sites, and (3) display of the estimated surface by use of gray-scale shading of hexagons. Using alternative procedures for any of the three steps can result in mosaic 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 in mind.

Estimating the spatial distribution of an ion species' concentration or deposition (annual or seasonal) is a problem of estimating a spatial surface by interpolation between measurement sites. It is assumed that spatial correlation exists among the sites, thus the estimation technique should take this into consideration. Additionally, it is assumed that the spatial correlation is a function only of the distance between sites. Kriging (a member of a family of geostatistical estimation procedures) is a surface estimation technique that explicitly utilizes this assumption. It uses the relationship between inter-site correlation and distance, modelled from 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. This application uses simple kriging, i.e., assumes that there is no deterministic drift. 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; Vong et al. 1989).

Often kriging uses a "nugget effect" to describe the within-site or short distance variability. This nugget appears as a jump discontinuity at the origin of the variogram, a device used to assess the relationship between inter-site correlation and distance and the overall variability of the phenomenon of interest. The consequence of using the nugget effect is to say that the annual summary at a site is measured without error and the kriging algorithm will interpolate the value at the site to be its annual summary ("exact" interpolation). An alternative approach is to estimate the within-site variability from information at the collocated sites and set the nugget effect to zero. When the within-site variability is used, kriging is no longer an exact interpolator. Thus the interpolated value at a site is not its annual summary. The kriging algorithm now takes into account the within-site variability and the values of nearby sites in interpolating at a location that has an annual summary. The alternative approach is used in this report.

Since western and eastern North America have distinctively different levels of wet deposition for many of the ion species considered, the estimation and display of the surface are done separately for these two regions. The regions are partitioned between Texas and Louisiana, in the south, and between the Dakotas and Minnesota, in the north. The variogram model used in simple kriging is estimated using sites only in the appropriate regions. However, the estimates produced by simple kriging used the eight closest sites to the hexagon's center, regardless of the regions.

Mosaic maps display the surface estimated by the kriging procedure. Estimates of the surface are made on a grid of hexagon centers. The hexagons

have an area of 2,000 squared kilometers. A lambert conformal conic map projection that is nearly distance preserving defines the coordinate system for the grid. The western region consists of 1875 hexagons while the eastern part consists of 1833 hexagons (1317 within the United States). The gray scale used in the maps are based on the percentiles from the frequency distribution of the estimates associated with the hexagons in the eastern and western regions of the United States.

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 total wet deposition temporal patterns from 1979 (or 1982) to 1987. The characteristics apply only to the period studied and do not imply either the presence or absence of any long-term trend. Nine (or six) 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 four-weekly data. Boxplots, defined below, graphically display percentiles of annual data from a subset of trend sites for each year in either the nine or six year period. Visual inspection of boxplots across years provides a visual assessment of annual temporal patterns during the period. Application of a modified Kendall seasonal tau trend statistical test and Sen's median slope estimation procedure to four-weekly data provide estimates of a general temporal pattern change over the entire period.

As discussed in Section 2.3.2, the statistical estimation procedures allow missing four-weekly summaries for a site, so 39 sites are used in the 1979-1987 period and 148 are used in the 1982-1987 period. However the graphical displays of annual data required complete and representative data for all years in the period, thus the number of sites used is reduced and different for each ion species.

Boxplots simultaneously display several features of a set of data. Figure 2.3 illustrates the definition of the boxplot as applied in this

FIGURE 2.3. Definition of Boxplot Display

report. Each boxplot presents the 5th (or 10th), 25th, 50th (median), 75th, 95th (or 90th) percentiles of the 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 above the plot boundary. Boxplots for the 1979-1987 period had at most 39 sites, so the 10th and 90th percentiles are shown, while the 1982-1987 period had at least 78 sites, so the 5th and 95th percentiles are shown. The boxplots include a shaded rectangle to display the 95% confidence interval for the median. The upper portion of a boxplot depicts the pattern of high concentration or deposition sites or of 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. A limitation of this test is that it, in essence, performs a number of univariate tests simultaneously at the same site without taking into account the problems of repeated tests (i.e., increase in false positive results).

Dietz and Killeen (1981) developed a similar multivariate test for monotone trend based on Kendall's tau statistic. They developed a covariance matrix for a multivariate KST. The incorporation of the covariance matrix alleviates the problem of repeated tests at a site. Dietz and Killeen's alternative hypothesis is that one or more seasons have a trend, but the trend need not be in the same direction for all seasons. Although they included ties in their statistic, missing values are not incorporated.

Hirsch and Slack (1984) extended the approach of Dietz and Killeen to include missing data. Additionally, their alternative hypothesis is that the trend for all seasons is either monotone increasing or decreasing. This change in alternative hypothesis probably accounts for the increased power that Hirsch and Slack observed for their test when compared to the Dietz and Killeen test. However, in their test, if some seasons are increasing and other seasons are decreasing, then the sum over all seasons would be zero and thus insignificant. The Hirsch and Slack approach is the modified KST test that is used in this report.

The seasons used in the modified KST test are the 13 four-weekly precipitation-weighted mean concentrations or total wet deposition values potentially available from each year. These periods are chosen to coincide with the four-weekly sampling schedule of the APIOS-C network. The NADP/NTN network sampling schedule is weekly and its week begins on the same day of the week that APIOS-C starts a new sampling period. The other networks are daily. Therefore, the four-weekly summaries are comparable across sites and networks. The modified KST test is still being repeated across a large number of sites, 39 sites in the 1979-1987 period and 148 sites in the

1982-1987 period, thus the possibility of false positives is still large. If there is no trend and the significance level of the tests is 5% (5% chance of a false positive result for one test), then for 39 sites there is a 4.4% chance of having 5 or more false positives and for 148 sites there is a 86.7% chance of having 5 or more false positives.

An estimate of the magnitude of the trend 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 can also be computed using the covariance matrix discussed above. Sen's median slope is related to the KST test, thus a significant median slope usually occurs when the KST test also indicates a significant trend.

Three different approaches summarize the modified KST test and Sen's median slope across sites in each trend period for each ion species. The first approach gives percentile summaries across sites for the slope estimates for an ion species' concentration or deposition analysis. Boxplots display the percentiles. The text discusses sites with extreme slope estimates.

The second approach displays the slope estimate for a site on a map of North America (see upper plot Figure A.20). The angle of a constant length ray is proportional to the magnitude of the slope estimate. A vertical ray represents a zero slope estimate. Positive slopes are represented by rays that point to the right and negative slopes are represented by rays that point to the left. A horizontal ray either to the left or the right represents the minimum or maximum slope depending on which has the largest absolute value. The direction of the rays are scaled to the largest absolute value. The purpose of the map display is to show all the slope estimates relative to their spatial location 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 modified KST test result significantly different from zero at the 5% significance level.

The third approach graphically displays the confidence interval for the slope estimate and enhances the display by using a ray which now identifies the estimated 1987 annual ion concentration or deposition for the site (see lower plot Figure A.20). The angle of the ray is proportional to the magnitude of the estimated annual value. A vertical ray represents the median estimate. Estimates larger than the median are represented by rays that point to the right and estimates smaller than the median are represented by rays that point to the left. A horizontal ray either to the left or the right represents the maximum deviation from the estimate, depending on which has the largest absolute difference. The direction of the rays are scaled to the largest absolute difference. 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 modified KST test result significantly different from zero at the 5% significance level.

The estimated 1987 annual values are obtained using kriging, as described in Section 2.4. The estimated 1987 annual values are used instead of the actual 1987 annual summaries because for many sites no annual summaries are calculated for one or more ion species because the data does not meet the UDDC completeness criteria.

The confidence interval plots provide several types of information. It is easy to identify sites with significantly decreasing or increasing trends. The horizontal pattern of confidence interval centers gives a general overview of the distribution of the slope estimates. The vertical

pattern of confidence interval half-widths gives an overview of the within-site variation of the four-weekly concentration or deposition; the larger the confidence interval half-width, the greater the within-site variation. The ray angle gives information about magnitude of trend and within-site variability relative to the magnitude of the estimated 1987 annual concentration or deposition. 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 1987 annual and seasonal (winter and summer) spatial patterns in terms of precipitation-weighted concentration (except hydrogen, where pH is used) and deposition. Temporal pattern information is presented for nine ions, the five ions above plus chloride, sodium, potassium and magnesium. Additionally, the annual total precipitation is considered. This information is presented using graphical displays of annual precipitation-weighted mean concentration and deposition and a statistical analysis based on a modified Kendall seasonal tau trend test. The modified Kendall seasonal tau trend test uses a subset of 39 long-term sites over a 9-year (1979-1987) period and an expanded subset of 148 sites with greater spatial coverage over a 6-year (1982-1987) period. The graphical displays of annual summaries uses subsets of the sites in the 9-year and 6-year periods. The 9-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 of annual temporal patterns to be produced. The 6-year period restricts the number of years in order to increase the number of sites available with data of known quality. Section 2 describes the site selection and data summarization procedures, the spatial pattern methodology, and the temporal pattern graphical and statistical analysis methodology.

This section is organized as follows. First, a brief overview of emission trends is presented to provide a context for the interpretation of the deposition temporal patterns. Then the 1987 annual and seasonal spatial patterns for precipitation-weighted concentration and deposition are presented. Finally, the temporal patterns for each of the ions are presented. 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 1978-1987

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 1989) decreased from approximately 9.1 to 7.0 million metric tons per year between 1978 and 1987 (Figure 3.1). This approximately 23% decrease is primarily from industrial processes and fuel combustion source categories. Sulfur oxide emissions decreased from approximately 24.8 to 20.0 million metric tons per year between 1978 and 1987 (Figure 3.2). This approximately 19% 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 oxide emissions decreased from approximately 21.1 to 19.5 million metric tons per year between 1978 and 1987 (Figure 3.3). This approximately 8% decrease reflects a decrease in vehicle emissions (transportation). Emissions of volatile organic compounds (VOC) decreased from approximately 23.7 to 19.6 million metric tons per year between 1978 and 1987 (Figure 3.4). This approximately 17.3% decrease is primarily from transportation and industrial processes source categories.

The TSP, sulfur oxide, nitrogen oxide and VOC temporal patterns are all decreasing. However all these patterns had an increase in 1984 and all but sulfur oxide increased in 1987. The increases in 1987 are at least partially attributed to the unusual meteorological conditions and forest fires. The increase in nitrogen oxide emissions is primarily due to an increase in stationary source fuel combustion.

FIGURE 3.1. National Trend in Total Suspended Particulate Emissions, 1978-1987

FIGURE 3.2. National Trend in Sulfur Oxide Emissions, 1978-1987

FIGURE 3.3. National Trend in Nitrogen Oxide Emissions, 1978-1987

FIGURE 3.4. National Trend in Volatile Organic Compound Emissions, 1978-1987

3.2 1987 SPATIAL PATTERNS

Mosaic maps, Figures A.1 through A.15, display the surface estimated by the kriging procedure described in Section 2.4. Estimates of the surface are made on a grid of hexagon centers. The hexagons have an area of 2,000 squared kilometers. The eastern and western parts of the maps are estimated and displayed separately because magnitude of the wet deposition is generally much larger in the east. The western part consists of 1875 hexagons while the eastern part consists of 1833 hexagons (1317 within the United States). The gray scale used in the maps are based on the percentiles from the frequency distribution of the estimates, within the United States, for each part.

The 1987 annual and seasonal precipitation-weighted concentration and total deposition used to generate the mosaic maps is summarized in Table A.3 and Table A.4, respectively. The frequency distribution percentiles of the estimated precipitation-weighted concentration and total deposition at the center of the hexagons in eastern and western United States are shown in Table A.9 and A.10, respectively. The relevant percentiles are also displayed on the individual mosaic maps. Finally, the location of the sites with the three highest and three lowest values, in the western and eastern United States and southeastern Canada (shown in the figures), are given in Tables A.11 through A.15 for each of the ions.

3.2.1 pH and Hydrogen

The estimated 1987 annual spatial pattern in the United States for pH (Figure A.1, Table A.3 and Table A.11) has a range of 4.12 to 5.07 in the east and a range of 4.64 to 6.09 in the west. Well over 50% of the east has pH values which are less than the lowest pH in the west. In the east, the lowest pH values occur in an ellipsoid area across Ohio and Pennsylvania and an additional area in western New York. The highest pH values in the east occur in Minnesota, Wisconsin and western Ontario. In the west, the lowest estimated pH values occur in two separate regions: eastern Kansas and Texas; and southeastern Arizona. The highest pH values in the west also occur in two

separate regions: western Kansas and eastern Colorado; and northern California and Nevada.

The estimated 1987 winter spatial pattern in the United States for pH (Figure A.2, Table A.3 and Table A.11) has a range of 4.21 to 5.02 in the east and a range of 4.59 to 6.00 in the west. Almost 50% of the east has pH values less than the lowest pH in the west, 4.59. In the east, the lowest pH values occur in a circular area centered over northeastern Ohio. The highest pH values in the east occur in three different regions: Minnesota; southern Louisiana; and Florida. In the west, the lowest pH values occur in eastern Kansas, Oklahoma and Texas. The highest pH values in the west also occur in two separate regions: eastern Colorado; and northern California and Nevada.

The estimated 1987 summer spatial pattern in the United States for pH (Figure A.3, Table A.3 and Table A.11) has a range of 3.96 to 5.68 in the east and a range of 4.53 to 5.96 in the west. Close to 75% of the pH values in the east are less than the lowest pH in the west. In the east, the lowest pH values occur in Pennsylvania, southern New York, New Jersey and Delaware. The highest pH values in the east occur in two regions: Minnesota, western Wisconsin and Ontario; and the Florida panhandle. In the west, the lowest pH values occur in two regions: eastern Kansas and Texas; and eastern Arizona and western New Mexico. The highest pH values in the west occur in two regions: western Utah; and the Great Plains from eastern North Dakota to eastern Colorado.

In both the east and west there are distinct differences in the summer and winter pH spatial patterns. In the east, the summer pH values are lower, the median in the summer is 4.44 while the winter median is 4.64, and the center of the lowest pH values has moved east from northeastern Ohio to eastern Pennsylvania. Additionally, in the east, the pH values in western Ontario decrease in the summer. In the west, the range of pH values are about the same in winter and summer, but the highest pH values in the summer are in the Great Plains which in the winter have much lower pH values. The highest pH values in the winter occur in northern California and western Nevada which have much lower pH values in the winter.

The estimated 1987 annual spatial pattern in the United States for hydrogen ion deposition (Figure A.1, Table A.4 and Table A.11) has a range of 0.053 kg/ha to 0.850 kg/ha in the east and a range of 0.010 kg/ha to 0.288 kg/ha in the west. Over 50% of the east has hydrogen deposition greater than the maximum value in the west. In the east, the highest deposition occurs in Pennsylvania, New Jersey and central New York. The lowest deposition in the east occurs in Minnesota, Wisconsin and western Ontario. In the west, the highest deposition occurs in eastern Kansas, Oklahoma, and Texas. The lowest deposition in the west occurs in two regions: western Kansas and eastern Colorado; and northern California, northern Nevada, and southern Idaho.

The estimated 1987 winter spatial pattern in the United States for hydrogen ion deposition (Figure A.2, Table A.4 and Table A.11) has a range of 0.005 kg/ha to 0.139 kg/ha in the east and a range of 0.002 kg/ha to 0.085 kg/ha in the west, with the median (0.008 kg/ha) in the west being almost eight times smaller than the median (0.062 kg/ha) in the east. In the east, the highest deposition occurs in two regions: southern New York and New Jersey; and northern Alabama, northern Georgia and eastern Tennessee. The lowest deposition in the east occurs in Minnesota and Wisconsin. In the west, the highest deposition occurs in eastern Kansas, eastern Oklahoma, and eastern Texas. The lowest deposition in the west occurs in three regions: eastern Colorado; northern Nevada and southern Idaho; and eastern Montana and the western Dakotas.

The estimated 1987 summer spatial pattern in the United States for hydrogen ion deposition (Figure A.3, Table A.4 and Table A.11) has a range of 0.005 kg/ha to 0.385 kg/ha in the east and a range of 0.002 kg/ha to 0.122 kg/ha in the west, with the median (0.010 kg/ha) in the west being over 11 times smaller than the median (0.116 kg/ha) in the east. In the east, the highest deposition occurs in Pennsylvania, New Jersey and central New York. In the west, the highest deposition occurs in eastern Kansas, Oklahoma, and Texas. The lowest deposition in the east occurs in Minnesota, Wisconsin and western Ontario. The lowest deposition in the west occurs in two regions:

northern California, northern Nevada, Oregon and southern Idaho; and the Dakotas.

The spatial patterns and the magnitudes of the summer and winter hydrogen ion deposition are generally the same in the west. The one major difference in the west is the increase in deposition along the west coast in the winter and the increase in deposition in Wyoming and Montana in the summer. In the east there is both a change in the spatial pattern and the magnitude between winter and summer. The median summer deposition (0.116 kg/ha) is almost twice the median winter deposition (0.062 kg/ha). The maximum estimate of summer deposition (0.385 kg/ha) is almost three times the maximum winter deposition (0.139 kg/ha). In the winter the highest deposition was in New Jersey and the south (Tennessee, Alabama and Georgia). However, in summer the high deposition in the northeast spreads to the west across Pennsylvania and central New York. Additionally, the south has changed from having the some of the highest deposition to having some of the lowest.

The estimated spatial patterns of pH and hydrogen deposition are similar for the annual and summer summaries. However, the estimated winter spatial patterns of pH and hydrogen deposition are different in the eastern United States. While the pH, in the winter, is highest in Ohio and eastern Pennsylvania, the hydrogen deposition is highest on the east coast in New Jersey and in the south.

3.2.2 Sulfate

The estimated 1987 annual spatial pattern in the United States for sulfate ion concentration (Figure A.4, Table A.3 and Table A.12) has a range of 0.92 mg/l to 4.29 mg/l in the east and a range of 0.28 mg/l to 1.66 mg/l in the west, with over 50% of the concentrations in the east being greater than the highest concentration in the west. In the east, the highest concentrations occur in southern Ontario, Ohio and western Pennsylvania. The lowest concentrations in the east occur in three regions: western Ontario; Nova Scotia and Maine; and Florida. In the west, the highest concentrations occur in two regions: eastern Kansas and Nebraska; and

southern Texas. The lowest concentrations in the west occur in northern California, Oregon, and Idaho.

The estimated 1987 winter spatial pattern in the United States for sulfate ion concentration (Figure A.5, Table A.3 and Table A.12) has a range of 0.38 mg/l to 2.69 mg/l in the east and a range of 0.17 mg/l to 1.64 mg/l in the west. The upper percentiles in the east are 50% greater than the upper percentiles in the west while the lower percentiles in the east are three times as large as the lower percentiles in the west. In the east, the highest concentrations occur in southern Ontario, Ohio and Indiana. The lowest concentrations in the east occur primarily in Maine and Nova Scotia. There are also a number of small regional lows throughout the southern coastal states. In the west, the highest concentrations occur in two regions: eastern South Dakota; and western Oklahoma and northern Texas. The lowest concentrations in the west occur in northern California, northern Nevada, and Oregon.

The estimated 1987 summer spatial pattern in the United States for sulfate ion concentration (Figure A.6, Table A.3 and Table A.12) has a range of 0.76 mg/l to 5.60 mg/l in the east and a range of 0.38 mg/l to 1.73 mg/l in the west. The lower percentiles in the east are two times larger than those in the west while the upper percentiles in the east are three times those in the west. In the east, the highest concentrations occur in southern Ontario, Pennsylvania, New Jersey, and Delaware. The lowest concentrations in the east occur in two regions: western Ontario and Minnesota; and the Florida panhandle, southern Alabama and southern Mississippi. In the west, the highest concentrations occur in two regions: eastern Kansas and Oklahoma; and southeastern Arizona. The lowest concentrations in the west occur in two regions: western Oregon; and northern Idaho.

In the east, the magnitude of the summer sulfate concentrations are nearly twice that of the winter sulfate concentrations. In the summer the maximum sulfate concentrations are centered over Pennsylvania, while in the winter the maximum sulfate concentrations are centered to the west, over Ohio. Additionally, in the east, during the summer, the lower concentrations are in the south, Minnesota and western Ontario, while in the winter the lower

concentrations are along the east coast. In the west, the magnitude of the higher concentrations increase slightly in the winter, but there is a large increase in the number of low concentrations (in the summer only 5% of the concentrations are less than 0.70 mg/l while in the winter close to 50% of the concentrations are less than that magnitude). In the west, while there is a relatively smooth spatial pattern in the winter, the summer spatial pattern shows a number of small regional highs and lows.

The estimated 1987 annual spatial pattern in the United States for sulfate ion deposition (Figure A.4, Table A.4 and Table A.12) has a range of 6.2 kg/ha to 42.2 kg/ha in the east and a range of 1.1 kg/ha to 18.7 kg/ha in the west, with the median in the east (18.4 kg/ha) almost five times larger than the median in the west (3.7 kg/ha). In the east, the highest deposition occurs in southern Ontario and eastern Pennsylvania. The lowest deposition in the east occurs in western Ontario, Minnesota and western Wisconsin. In the west, the highest deposition occurs in eastern Kansas, Oklahoma and Texas. The lowest deposition in the west occurs in western Idaho, eastern Oregon, and Nevada.

The estimated 1987 winter spatial pattern in the United States for sulfate ion deposition (Figure A.5, Table A.4 and Table A.12) has a range of 0.29 kg/ha to 6.71 kg/ha in the east and a range of 0.09 kg/ha to 4.75 kg/ha in the west, with the median in the east (2.79 kg/ha) five times larger than the median in the west (0.57 kg/ha). In the east, the highest deposition occurs in two regions: eastern Tennessee and northern Alabama; and southeastern Louisiana. The lowest deposition in the east occurs in western Ontario, Minnesota and Wisconsin. In the west, the highest deposition occurs in eastern Texas. The lowest deposition in the west occurs in northern Nevada, eastern Oregon, southern Idaho and southeastern Montana.

The estimated 1987 summer spatial pattern in the United States for sulfate ion deposition (Figure A.6, Table A.4 and Table A.12) has a range of 1.62 kg/ha to 18.90 kg/ha in the east and a range of 0.29 kg/ha to 6.12 kg/ha in the west, with the median in the east (6.64 kg/ha) over five times larger than the median in the west (1.22 kg/ha). In the east, the highest deposition occurs in southern Ontario, Pennsylvania, and New Jersey. The

lowest deposition in the east occurs in two regions: western Ontario and Minnesota; and Arkansas, Mississippi and Alabama. In the west, the highest deposition occurs in two regions: eastern Nebraska and Kansas; and Texas along the Gulf of Mexico. The lowest deposition in the west occurs in northern California and Oregon.

In the east, the median summer sulfate deposition (6.6 kg/ha) is just slightly greater than the maximum winter sulfate deposition. In the summer, the maximums occur in Pennsylvania and southern Ontario with the minimums occurring in the south, Minnesota and western Ontario. However, in the winter the maximums occur in the south, with the greatest changes occurring in Alabama. In the west, the summer sulfate deposition is also greater than the winter sulfate deposition, with the median summer deposition (1.2 kg/ha) being twice the median winter deposition (0.6 kg/ha). In the summer, the sulfate deposition is at a minimum in the Pacific Northwest, while in the winter, the west coast deposition increases and the minimums occur inland, as far west as eastern Montana.

The estimated annual spatial patterns of sulfate concentration and deposition are similar. The estimated summer spatial patterns of sulfate concentration and deposition are also similar in the east, but are different in the west. The summer sulfate deposition in the west has a smooth spatial pattern that generally decreases from east to west. The sulfate concentration spatial pattern is marked by numerous small regional highs and lows, with a very high concentration as far west as Arizona. The estimated winter spatial patterns of sulfate concentration and deposition are different in both the east and west. In the east, the high concentrations are in the north, centered in Ohio and the low concentrations are along the Atlantic and Gulf of Mexico. However, the deposition is reversed, Ohio has only median deposition while the coastal states have the high deposition. In the west, the winter concentrations are lowest in the Pacific Northwest, but the deposition is relatively high in the Pacific Northwest and the lowest deposition occurs inland, to the east.

3.2.3 Nitrate

The estimated 1987 annual spatial pattern in the United States for nitrate ion concentration (Figure A.7, Table A.3 and Table A.13) has a range of 0.56 mg/l to 2.87 mg/l in the east and a range of 0.20 mg/l to 1.41 mg/l in the west. Over 95% of the nitrate concentrations in the west are less than the median concentration in the east (1.23 mg/l). In the east, the highest concentrations occur in southern Ontario, northern Pennsylvania and northern New York. There are 17 sites in southern Ontario that have nitrate concentrations higher than Bennett Bridge, New York. The lowest concentrations in the east occur in two regions: New Brunswick, Nova Scotia and Maine; and Florida. In the west, the highest concentrations occur in two regions: eastern South Dakota and eastern Nebraska; and western Oklahoma, western Kansas and southeastern Colorado. The lowest concentrations in the west occur in Oregon, Washington and northern Idaho.

The estimated 1987 winter spatial pattern in the United States for nitrate ion concentration (Figure A.8, Table A.3 and Table A.13) has a range of 0.37 mg/l to 3.19 mg/l in the east and a range of 0.13 mg/l to 1.88 mg/l in the west, with the median in the east (1.10 mg/l) being almost twice the median in the west (0.67 mg/l). In the east, the highest concentrations occur in southern Ontario and Michigan. The lowest concentrations in the east occur in the south along the Gulf of Mexico. In the west, the highest concentrations occur in two regions: the eastern Dakotas; and southern Nevada. The lowest concentrations in the west occur in Oregon and Washington.

The estimated 1987 summer spatial pattern in the United States for nitrate ion concentration (Figure A.9, Table A.3 and Table A.13) has a range of 0.73 mg/l to 2.97 mg/l in the east and a range of 0.25 mg/l to 1.98 mg/l in the west. Over 95% of the western concentrations are less than the median concentration in the east (1.35 mg/l). In the east, the highest concentrations occur in two regions: southern Ontario; and eastern Pennsylvania and Delaware. The lowest concentrations in the east occur in two regions: Ontario and Quebec; and Florida. In the west, the highest concentrations occur primarily in three regions: Utah; eastern Colorado; and eastern Oregon. The lowest concentrations in the west occur in primarily

in Oregon, Washington, northern Idaho and western Montana. The concentrations in the west have a number of small regional highs and lows.

In both the east and west, there is not a significant difference between the magnitudes of the larger percentiles in the summer and winter nitrate concentrations, however the lower percentiles increase in magnitude in the summer for both parts of the United States. In the east, the nitrate concentrations increase from the south to the north in the winter, with the highest concentrations in southern Ontario. In the summer, the maximum nitrate concentrations move to the east to western Pennsylvania, and some of the lowest concentrations occur in the north. In the west, the largest nitrate concentrations in the winter occur in the eastern Dakotas and the concentrations decrease to the west. In the summer, the spatial pattern of the nitrate concentrations is dominated by a number of small regional highs and lows.

The estimated 1987 annual spatial pattern in the United States for nitrate ion deposition (Figure A.7, Table A.4 and Table A.13) has a range of 6.1 kg/ha to 27.6 kg/ha in the east and a range of 1.5 kg/ha to 11.9 kg/ha in the west, with almost all the deposition in the west less than the median deposition in the east (11.1 kg/ha). In the east, the highest deposition occurs in central New York, eastern Pennsylvania and southern Ontario. The lowest deposition in the east occurs in three regions: Ontario; Quebec and Maine; and Alabama and the Florida panhandle. In the west, the highest deposition occurs in eastern Nebraska, Kansas and Oklahoma. The lowest deposition in the west occurs in two regions: eastern Oregon, western Idaho, and northern Nevada; and southeastern Arizona.

The estimated 1987 winter spatial pattern in the United States for nitrate ion deposition (Figure A.8, Table A.4 and Table A.13) has a range of 0.55 kg/ha to 6.14 kg/ha in the east and a range of 0.17 kg/ha to 2.57 kg/ha in the west, with more than 95% of the western concentrations being less than the median concentration in the east (2.2 kg/ha). In the east, the highest deposition occurs in two regions: southern Ontario; and upstate New York and Massachusetts. The lowest deposition in the east occurs in Minnesota, Iowa and Wisconsin. In the west, the highest deposition occurs

in eastern Texas. The lowest deposition in the west occurs in northern Nevada, eastern Oregon, southern Idaho and southeastern Montana.

The estimated 1987 summer spatial pattern in the United States for nitrate ion deposition (Figure A.9, Table A.4 and Table A.13) has a range of 1.80 kg/ha to 9.14 kg/ha in the east and a range of 0.17 kg/ha to 5.32 kg/ha in the west. The median in the east (3.90 kg/ha) is over three times larger than the median in the west (1.20 kg/ha). In the east, the highest deposition occurs in southern Ontario, Pennsylvania and New Jersey. The lowest deposition in the east occurs in two regions: Ontario, Quebec, Nova Scotia and Maine; and Alabama and Arkansas. In the west, the highest deposition occurs in two regions: eastern Nebraska and Kansas; and the Texas coast. The lowest deposition in the west occurs in Oregon and Washington.

In both the east and the west, the summer nitrate deposition is nearly twice the winter nitrate deposition. In the east, the summer maximums occur in Pennsylvania, while in the winter, the maximums move north and west to New York and southern Ontario. In the west, the minimum summer nitrate deposition occurs in the Pacific Northwest. However, in the winter, the minimum nitrate deposition moves inland as far east as eastern Montana. The high nitrate summer deposition in eastern Kansas and Nebraska is not present in the winter.

The estimated annual spatial patterns for nitrate concentration and deposition are the same for most of the United States and southeastern Canada except for a high concentration in southeastern Colorado that has only a moderately high deposition. The estimated winter spatial patterns for nitrate concentration and deposition are different in both the east and west. In the east, the concentrations are very high over southern Ontario and diminish to the east and south, while the deposition is lowest in Minnesota and increases toward the east. In the west, the nitrate concentrations are highest in the eastern Dakotas and decrease toward the south, while the nitrate deposition is highest in eastern Texas and decreases toward the north. Additionally, in the winter, the Pacific Northwest has the lowest concentrations, while the deposition is high along the coast in the Pacific Northwest.

3.2.4 Ammonium

The estimated 1987 annual spatial pattern in the United States for ammonium ion concentration (Figure A.10, Table A.3 and Table A.14) has a range of 0.080 mg/l to 0.725 mg/l in the east and a range of 0.035 mg/l to 0.651 mg/l in the west, with the concentrations in the west being only slightly less than those in the east. In the east, the highest concentrations occur in two regions: southern Ontario and Ohio; and Minnesota. The lowest concentrations in the east occur in two regions: Maine; and Florida. In the west, the highest concentrations occur in two regions: eastern South Dakota and Nebraska; and western Oklahoma and Kansas. The lowest concentrations in the west occur in Oregon, western Washington and central Idaho.

The estimated 1987 winter spatial pattern in the United States for ammonium ion concentration (Figure A.11, Table A.3 and Table A.14) has a range of 0.032 mg/l to 0.575 mg/l in the east and a range of 0.017 mg/l to 0.671 mg/l in the west. The east has a larger median (0.17 mg/l) than the west (0.12 mg/l), but the range is much greater in the west. In the east, the highest concentrations occur in southern Ontario and Michigan. The lowest concentrations in the east occur primarily in Quebec, New Brunswick, Maine and Massachusetts, with a number of small regional lows along the Atlantic coast. In the west, the highest concentrations occur in two regions: eastern South Dakota; and western Kansas and Oklahoma. The lowest concentrations in the west occur in western Washington, northern Oregon, southern Idaho and western Wyoming.

The estimated 1987 summer spatial pattern in the United States for ammonium ion concentration (Figure A.12, Table A.3 and Table A.14) has a range of 0.081 mg/l to 0.874 mg/l in the east and a range of 0.031 mg/l to 0.748 mg/l in the west, with the east having a higher median (0.31 mg/l) than the west (0.21 mg/l). In the east, the highest concentrations occur in two regions: southern Ontario and northern Ohio; and the Michigan peninsula. The lowest concentrations in the east occur in the deep south. In the west, there are numerous small regional highs and lows.

The summer ammonium concentration is greater than the winter concentration, in both the east and the west. The spatial pattern in the east does not change significantly between the winter and summer. The spatial pattern in the west changes between the summer and winter. The winter spatial pattern is relatively smooth with high concentrations in eastern South Dakota and eastern Colorado and a decrease in concentrations toward the Pacific Northwest. The summer spatial pattern is not smooth, being dominated by numerous small regional highs and lows.

The estimated 1987 annual spatial pattern in the United States for ammonium ion deposition (Figure A.10, Table A.4 and Table A.14) has a range of 0.81 kg/ha to 6.09 kg/ha in the east and a range of 0.33 kg/ha to 4.28 kg/ha in the west. The median deposition in the east (2.62 kg/ha) is over four times greater than the median deposition in the west (0.62 kg/ha). In the east, the highest deposition occurs in two regions: southern Ontario and central New York; and northern Illinois. The lowest deposition in the east occurs in two regions: Maine, New Brunswick and Nova Scotia; and South Carolina and Florida. In the west, the highest deposition occurs in eastern South Dakota, Nebraska, Kansas and Oklahoma. The lowest deposition in the west occurs in two regions: Oregon; and Arizona, Utah and Wyoming.

The estimated 1987 winter spatial pattern in the United States for ammonium ion deposition (Figure A.11, Table A.4 and Table A.14) has a range of 0.06 kg/ha to 1.40 kg/ha in the east and a range of 0.02 kg/ha to 0.87 kg/ha in the west, with the median in the east (0.30 kg/ha) over two times the median in the west (0.11 kg/ha). In the east, the highest deposition occurs in two regions: southern Ontario; and southern Mississippi and Louisiana. The lowest deposition in the east occurs in two regions: western Ontario and northern Minnesota; and Maine, New Brunswick and Nova Scotia. The east also has a number of small regional highs and lows. In the west, the highest deposition occurs in eastern Oklahoma and Texas. The lowest deposition in the west occurs in eastern Oregon, northern Nevada, southern Idaho and western Wyoming.

The estimated 1987 summer spatial pattern in the United States for ammonium ion deposition (Figure A.12, Table A.4 and Table A.14) has a range

of 0.22 kg/ha to 3.47 kg/ha in the east and a range of 0.03 kg/ha to 1.32 kg/ha in the west, with the median in the east (0.83 kg/ha) almost four times the median in the west (0.22 kg/ha). In the east, the highest deposition occurs in two regions: western Ontario; and southeastern Ontario, Ohio, and Pennsylvania. The lowest deposition in the east occurs in the Alabama, Mississippi and Arkansas. The east also has a number of small regional highs and lows. In the west, the highest deposition occurs in eastern South Dakota, Nebraska, western Kansas and Oklahoma. The lowest deposition in the west occurs in northern California, western Oregon and Washington.

Ammonium deposition in the summer is more than twice the deposition in winter. The spatial pattern of the ammonium deposition in the east is dominated by small regional highs and lows. In the east, the minimum deposition in the summer generally occurs in the south, while in the winter the minimums occur in the north. In the west, as with the other ions, the summer minimums occur in the Pacific Northwest, while in the winter the minimums move inland.

The estimated annual spatial patterns for ammonium concentration and deposition are generally the same in the east and west. The estimated summer spatial patterns for ammonium concentration and deposition are the same in the east, but differ in the west. In the west, the summer deposition has a smooth spatial pattern, while the summer concentrations are dominated by a number of small regional highs and lows. The estimated winter spatial patterns for ammonium concentration and deposition are different in both the east and west. In the west, the concentrations are highest in South Dakota and the Oklahoma panhandle and decrease toward the Pacific Northwest. In the east, the concentrations are highest in the north and decrease toward the Atlantic Ocean, while the deposition is higher along the Gulf of Mexico and Atlantic Ocean and decrease toward Minnesota.

3.2.5 Calcium

The estimated 1987 annual spatial pattern in the United States for calcium ion concentration (Figure A.13, Table A.3 and Table A.15) has a range of 0.030 mg/l to 0.553 mg/l in the east and a range of 0.037 mg/l to 0.377

mg/l in the west, with the west having a higher median (0.18 mg/l) than the east (0.12 mg/l). In the east, the highest concentrations occur in two regions: southern Ontario and Minnesota; and Iowa. The lowest concentrations in the east occur in two regions: New Brunswick, Nova Scotia and New England; and the Carolinas, Georgia and Alabama. In the west, the highest concentrations occur in western Kansas, Oklahoma, northern Texas, and southeastern Colorado. The lowest concentrations in the west occur in northern California, western Oregon and Washington.

The estimated 1987 winter spatial pattern in the United States for calcium ion concentration (Figure A.14, Table A.3 and Table A.15) has a range of 0.008 mg/l to 0.527 mg/l in the east and a range of 0.029 mg/l to 0.459 mg/l in the west, with the west having a larger median (0.14 mg/l) than the east (0.08 mg/l). In the east, the highest concentrations occur in two regions: southeastern Ontario, Michigan and northern Indiana; and western Ontario. The lowest concentrations in the east occur in two regions: New England; and North Carolina. In the west, the highest concentrations occur in three regions: the Dakotas; southern Nevada; and southwestern Texas. The lowest concentrations in the west occur along the coast of the Pacific Ocean.

The estimated 1987 summer spatial pattern in the United States for calcium ion concentration (Figure A.15, Table A.3 and Table A.15) has a range of 0.033 mg/l to 0.609 mg/l in the east and a range of 0.030 mg/l to 0.826 mg/l in the west, with the west having a larger median (0.24 mg/l) than the east (0.15 mg/l). In the east, the highest concentrations occur in two regions: southern Ontario; and southern Minnesota and western Iowa. The lowest concentrations in the east occur in two regions: New Brunswick, Nova Scotia, Quebec, and New England; and the Carolinas. In the west, the highest concentrations occur in Utah and in several other small regions. The lowest concentrations in the west occur in western Oregon and Washington.

In both the east and west, the calcium concentration in the summer is approximately twice the winter concentration. In the east, the summer and winter spatial patterns are generally similar. The spatial pattern of the calcium concentration in the west is dominated by the high concentrations

in the summer in Utah, and high concentrations in the Dakotas and southern Nevada in the winter.

The estimated 1987 annual spatial pattern in the United States for calcium ion deposition (Figure A.13, Table A.4 and Table A.15) has a range of 0.30 kg/ha to 4.58 kg/ha in the east and a range of 0.20 kg/ha to 2.49 kg/ha in the west, with the median in the east (1.2 kg/ha) larger than the median in the west (0.7 kg/ha). In the east, the highest deposition occurs in three regions: southern Ontario; northern Illinois; and southern Iowa. The lowest deposition in the east occurs in three regions: New Brunswick, Nova Scotia, Quebec, and New England; North Carolina; and Alabama. In the west, the highest deposition occurs in northcentral Texas, central Oklahoma and eastern Kansas. The lowest deposition in the west occurs in two regions: central California and western Nevada; and eastern Washington.

The estimated 1987 winter spatial pattern in the United States for calcium ion deposition (Figure A.14, Table A.4 and Table A.15) has a range of 0.023 kg/ha to 0.626 kg/ha in the east and a range of 0.034 kg/ha to 0.483 kg/ha in the west, with the median in the east (0.14 kg/ha) greater than the median in the west (0.08 kg/ha). In the east, the highest deposition occurs in two regions: southern Ontario; and Louisiana. The lowest deposition in the east occurs in two regions: New England; and Minnesota, Wisconsin, Iowa and Illinois. In the west, the highest deposition occurs in eastern Texas. The lowest deposition in the west occurs in two regions: eastern Colorado; and California, northern Nevada, and southern Oregon.

The estimated 1987 summer spatial pattern in the United States for calcium ion deposition (Figure A.15, Table A.4 and Table A.15) has a range of 0.06 kg/ha to 2.21 kg/ha in the east and a range of 0.02 kg/ha to 1.28 kg/ha in the west, with the median deposition in the east (0.41 kg/ha) greater than the median deposition in the west (0.28 kg/ha). In the east, the highest deposition occurs in three regions: southern Ontario; Iowa; and northern Indiana. The lowest deposition in the east occurs in three regions: New Brunswick, Nova Scotia, Quebec, and New England; North Carolina; and Alabama. In the west, the highest deposition occurs in Kansas, central

Oklahoma, northern Texas. The lowest deposition in the west occurs in northern California, Oregon and Washington.

In both the east and west, calcium deposition in the summer is approximately three times the deposition in the winter. In the east, the spatial pattern of the calcium deposition are almost opposites in the summer and winter. In the summer, the minimum deposition is along the Atlantic coast, while in the winter, the maximum deposition is primarily along the Atlantic coast and the Gulf of Mexico. In the west, the summer minimums again occur in the Pacific Northwest. However, in the winter, the Pacific Northwest has relatively high calcium deposition.

The estimated spatial patterns for calcium concentration and deposition are similar for both the annual and summer summaries. The estimated winter spatial patterns for calcium concentration and deposition are different. The concentrations in the east are highest in Ontario and decrease toward the south, while the deposition is highest in the south along the Gulf of Mexico and decreases toward Minnesota. In the west, the concentration is highest in the Dakotas and the deposition is highest in eastern Texas. As before, the concentrations are lowest along the Pacific coast and the deposition is lowest inland.

3.3 TEMPORAL PATTERNS

The temporal patterns are examined using two approaches described in Section 2.5. First, the annual temporal patterns are examined using boxplots of the percentiles of the annual summaries for each year in the two trend subsets, 1979-1987 and 1982-1987. Then the temporal changes are examined using rayplots for each of the two trend subsets. The temporal patterns examine both concentration and deposition for nine ion species: hydrogen, sulfate, nitrate, ammonium, calcium, chloride, sodium, potassium and magnesium. pH and precipitation are also examined.

3.3.1 Hydrogen, pH and Precipitation

3.3.1.1 Annual Temporal Patterns

The 30 site 1979-1987 trend subset for annual total precipitation (Table A.6 and Figure A.16) has a maximum median in 1979 of 117.83 cm and minimum median of 89.39 cm in 1987. The 118 site 1982-1987 trend subset for annual total precipitation (Table A.8 and Figure A.17) has a maximum median of 101.28 cm in 1983 and a minimum median of 87.05 cm in 1987. While the 9-year subset has inter-annual variation that suggests an almost cyclic appearance, the 6-year subset has no distinctive inter-annual pattern. The 95% confidence intervals for the median are a measure of the variability for the entire region that the trend subsets cover, thus they are not sensitive indicators of a temporal trend. Additionally, the confidence intervals are relatively large for the 1979-1987 trend subset because of the small number of sites in the subset. However, in spite of these limitations, the 95% confidence intervals for the median do not overlap for several years in both subsets. The 1980 median precipitation is significantly less than the 1983 median precipitation in the 1979-1987 trend subset and the 1987 median precipitation is significantly less than the 1983 and 1985 median precipitations in the 1982-1987 trend subset.

The 23 site 1979-1987 trend subset for annual pH (Table A.5 and Figure A.16) has approximately equal maximum medians in 1979, 1983, 1984, and 1987 of 4.43. 1980 has the lowest median pH of 4.28. The 108 site 1982-1987 trend subset for annual pH (Table A.7 and Figure A.17) has a maximum median pH of 4.33 in 1983 and a minimum median pH of 4.21 in 1982. The 9-year trend subset appears to have a minimum in 1980-1981, but the 95% confidence intervals are large enough that the difference is not significant. The 6-year trend subset appears to have no significant trend.

The annual hydrogen concentration for the 1979-1987 trend subset (Table A.5 and Figure A.18) has a minimum median of 36.77 $\mu\text{g/l}$ in 1983 and a maximum median of 53.06 $\mu\text{g/l}$ in 1980. The 1982-1987 trend subset (Table A.7 and Figure A.19) has a minimum median of 33.57 $\mu\text{g/l}$ in 1983 and a maximum median of 44.27 $\mu\text{g/l}$ in 1982, with the 1987 median of 43.50 $\mu\text{g/l}$ being only

slightly smaller than the maximum median. The 95% confidence intervals for the medians indicate that there are no significant differences for either the 9-year or 6-year trend subsets.

The annual hydrogen deposition (Table A.6 and Figure A.18) has approximately equal minimum medians in 1979, 1984 and 1987 of 0.43 kg/ha. 1980 has the highest annual hydrogen median deposition of 0.50 kg/ha. The annual hydrogen deposition (Table A.8 and Figure A.19) has a maximum median of 0.43 kg/ha in 1982 and a minimum median of 0.35 kg/ha in 1986. The 95% confidence intervals for the medians indicate that there are no significant differences for either the 9-year or 6-year trend subsets.

The annual hydrogen deposition pattern appears to be a damped version of the annual hydrogen concentration pattern. The damping of the deposition pattern may be associated with the annual hydrogen concentration and total precipitation patterns which are acting in opposite directions. Since deposition is a product of concentration and precipitation, the opposite patterns have a damping effect on the product. The opposite point of view may be taken with this association, the annual hydrogen concentration pattern is the result of the precipitation since the concentration is the ratio of deposition to precipitation and the deposition changes are small compared to those of precipitation.

3.3.1.2 Temporal Change Analysis

Based on the modified KST test, two of the 39 1979-1987 trend sites (Figure A.20) exhibit a significantly ($p < 0.05$) decreasing trend for total annual precipitation: Delaware, OH (-3.9 cm/year), and Wooster, OH (-1.4 cm/year). One of the 148 1982-1987 trend sites (Figure A.21) had a significantly decreasing trend: Salem, IL (-7.3 cm/year). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Except the sites in or near the Great Plains which show no precipitation trend, the spatial pattern of the trends show only small local areas of consistency. The magnitudes of the trends (the confidence interval center) appear unrelated to the 1987 estimated precipitation while the variability of the trend estimates (the

confidence interval half-width) appear to increase with the magnitude of the 1987 estimated precipitation. The lack of significant trends for precipitation is not surprising when the cyclic nature of the precipitation (see Figure A.16) is considered.

Three of the 39 1979-1987 trend sites exhibit a significantly ($p < 0.05$) increasing trend in pH: Ithaca, NY (0.018 pH units/year); Fort Wayne, IN (0.011 pH units/year); and Zanesville, OH (0.009 pH units/year). Four of the 148 1982-1987 trend sites exhibit a significantly increasing trend for pH: Brookings, SD (0.068 pH units/year); Killarney, ON (0.040 pH units/year); Ear Falls, ON (0.039 pH units/year); and Shallow Lake, ON (0.024 pH units/year). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Davis, CA, has the largest increasing pH trend for the 9-year subset (0.048 pH units/year), and the second largest trend for the 6-year subset (0.065 pH units/year). Davis also has the highest pH for both subsets. Figure A.22 and Figure A.23 display the trend estimates and confidence intervals for the 1979-1987 and 1982-1987 trend subsets, respectively. For both trend subsets, the spatial pattern of the trends show only small local areas of consistency. The confidence interval plots show no relationship between the magnitude of the estimated 1987 pH and the trend estimate while the variability of the trend estimate increases with the magnitude of the 1987 pH.

Since pH is a transformation of hydrogen concentration, the same sites have significant decreasing hydrogen concentrations (since the higher the pH, the lower the hydrogen concentration): Ithaca, NY ($-2.50 \mu\text{eq/l/year}$); Fort Wayne, IN ($-1.21 \mu\text{eq/l/year}$); and Zanesville, OH ($-1.14 \mu\text{eq/l/year}$), in the 1979-1987 trend subset; and Killarney, ON ($-5.52 \mu\text{eq/l/year}$); Shallow Lake, ON ($-2.66 \mu\text{eq/l/year}$); Ear Falls, ON ($-0.91 \mu\text{eq/l/year}$); and Brookings, SD ($-0.59 \mu\text{eq/l/year}$), in the 1982-1987 trend subset. With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.24 and Figure A.25 display the trend estimates and confidence intervals for the 1979-1987 and 1982-1987 trend subsets, respectively. Unlike pH, the hydrogen concentration

trends are consistently small throughout the entire region west of the Mississippi River. The eastern part of the United States and Canada show only a small local area of consistency. The confidence interval plots show that the sites with the smaller estimated 1987 hydrogen concentrations have smaller trend estimates and also smaller variability, in the 1982-1987 subset two of the four significant trends were less than $-1 \mu\text{eq/l/year}$.

For 1979-1987 trend sites, significant hydrogen deposition decreases occur at three sites: Zanesville, OH ($-4.00 \text{ meq/m}^2/\text{year}$, maximum for all sites); Rockport, IN ($-3.75 \text{ meq/m}^2/\text{year}$); and Raleigh, NC ($-2.65 \text{ meq/m}^2/\text{year}$), and increases occur at two sites: Bradford Forest, FL ($1.43 \text{ meq/m}^2/\text{year}$); and Chalk River, ON ($1.21 \text{ meq/m}^2/\text{year}$). As will be seen later, Chalk River, ON, is an unusual site for a number of different ion species. Only one site, Zanesville, OH, had significant decreases in both concentration and deposition. For 1982-1987 trend sites, significant hydrogen deposition decreases occur at two sites: Selma, AL ($-4.23 \text{ meq/m}^2/\text{year}$); Coldwater, ON ($-3.12 \text{ meq/m}^2/\text{year}$); and Alsea, OR ($-0.28 \text{ meq/m}^2/\text{year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.26 and Figure A.27 display the trend estimates and confidence intervals for the 1979-1987 and 1982-1987 trend subsets, respectively. The spatial pattern of the trend estimates for hydrogen deposition look very similar to those for hydrogen concentration. The confidence interval plots show the same behavior as concentration.

3.3.2 Sulfate

3.3.2.1 Annual Temporal Patterns

The 26 site 1979-1987 trend subset for annual sulfate concentration (Table A.5 and Figure A.28) has maximum medians in 1980 (2.62 mg/l) and 1981 (2.65 mg/l). 1987 has the lowest median, 1.90 mg/l . The annual sulfate deposition (Table A.6 and Figure A.28) has a maximum median in 1979 of 26.82 kg/ha and a minimum median in 1987 of 20.86 kg/ha . While median annual concentrations appear to have inter-annual variation that is opposite of precipitation amount, deposition appears to decrease consistently. The

extreme lows for both concentration and deposition occur at Sand Springs, CO, and ELA (b), ON. The extreme highs for both concentration and deposition occur at Parsons, WV; Caldwell, OH; Penn State, PA; and Lewes, DE. The extremely high deposition in 1985 and 1986 was at Parsons, WV.

The 102 site 1982-1987 trend subset for annual sulfate concentration (Table A.7 and Figure A.29) has maximum medians of 1.23 mg/l in 1982 and 1.22 mg/l in 1984 and minimum medians of 0.97 mg/l in 1985 and 1986. The annual sulfate deposition (Table A.8 and Figure A.29) has a maximum median in 1982 of 24.07 kg/ha and a minimum median of 21.01 kg/ha in 1987. The deposition appears to have a small decreasing trend while the concentration has no noticeable trend.

3.3.2.2 Temporal Change Analysis

Based on the modified KST test, seven of the 39 1979-1987 trend sites exhibit a significantly ($p < 0.05$) decreasing trend in sulfate concentration: Chalk River, ON ($-2.87 \mu\text{eq/l/year}$, maximum for all sites); Manitou, CO ($-2.27 \mu\text{eq/l/year}$); Sand Spring, CO ($-2.07 \mu\text{eq/l/year}$); Dixon Springs, IL ($-1.62 \mu\text{eq/l/year}$); Davis, CA ($-1.42 \mu\text{eq/l/year}$); Illinois, IL ($-1.38 \mu\text{eq/l/year}$); and Hubbard Brook, NH ($-1.27 \mu\text{eq/l/year}$). Figure A.30 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates appear fairly consistent in the western United States, all decreasing. The eastern United States is inconsistent with the most inconsistencies occurring along the Appalachian Mountains where the trends are increasing. The confidence interval plot shows no relationship between the magnitude of the 1987 estimated concentration and either the trend estimate or variability.

For 1982-1987 trend sites (Figure A.31), only three sites exhibit significantly decreasing trend in sulfate concentration: Huron Park, ON ($-4.03 \mu\text{eq/l/year}$); Acadia, ME ($-1.96 \mu\text{eq/l/year}$); and Cuba, NM ($-1.72 \mu\text{eq/l/year}$); and one site has a significantly increasing trend, Wooster, OH ($2.28 \mu\text{eq/l/year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. While the trend estimates are almost all negative for the 1979-1987 trend sites, the trend estimates for the 1982-1987 trend sites are positive

for many of the sites. The spatial pattern of the trend estimates are only consistent in small local areas, although all the estimates west of the Mississippi River are negative. The confidence interval plot shows that the greater the magnitude of the 1987 sulfate concentration, the greater the magnitude of the variation of the trend estimate, but there appears to be no relationship between the 1987 concentration and the trend estimate.

For 1979-1987 trend sites (Figure A.32), significant sulfate deposition decreases occur at 11 sites: Zanesville, OH ($-4.00 \text{ meq/m}^2/\text{year}$, maximum for all sites); Rockport, IN ($-3.42 \text{ meq/m}^2/\text{year}$); Piedmont Station, NC ($-3.36 \text{ meq/m}^2/\text{year}$); Coweeta, NC ($-3.30 \text{ meq/m}^2/\text{year}$); Delaware, OH ($-2.27 \text{ meq/m}^2/\text{year}$); Hubbard Brook, NH ($-2.24 \text{ meq/m}^2/\text{year}$); Chalk River, ON ($-2.12 \text{ meq/m}^2/\text{year}$); Caldwell, OH ($-1.29 \text{ meq/m}^2/\text{year}$); Georgia Station, GA ($-1.14 \text{ meq/m}^2/\text{year}$); ELA (b), ON ($-0.87 \text{ meq/m}^2/\text{year}$); Marcell, MN ($-0.80 \text{ meq/m}^2/\text{year}$); and Manitou, CO ($-0.49 \text{ meq/m}^2/\text{year}$). Only three sites, Chalk River, Manitou and Hubbard Brook, had significant decreases in both concentration and deposition. As seen in Figure A.32(b), except for the three sites in Ohio, the sites with significantly decreasing trends have estimated 1987 sulfate deposition below the median. The sites with increasing trend estimates for deposition occur in the east along the Appalachian and Adirondack Mountains and in the Midwest in Indiana and Illinois.

For 1982-1987 trend sites (Figure A.33), significant sulfate deposition decreases occur at eight sites: Tanbark Flat, CA ($-3.04 \text{ meq/m}^2/\text{year}$); Illinois, IL ($-2.33 \text{ meq/m}^2/\text{year}$); ELA (b), ON ($-1.71 \text{ meq/m}^2/\text{year}$); Dorion, ON ($1.22 \text{ meq/m}^2/\text{year}$); Fernberg, MN ($-1.13 \text{ meq/m}^2/\text{year}$); Sequoia National Park, CA ($-0.86 \text{ meq/m}^2/\text{year}$); Craters of Moon, ID ($-0.49 \text{ meq/m}^2/\text{year}$); and Alamosa, CO ($-0.43 \text{ meq/m}^2/\text{year}$). As seen in Figure A.33(b) all the sites with significantly decreasing sulfate deposition also have relatively small 1987 estimated sulfate deposition. The spatial pattern of the sulfate deposition trends has only small local consistences in the east, while west of the Mississippi River, all the trend estimates are decreasing. Although only eight sites have significant trends and would not be considered unusual given that 148 sites are being tested simultaneously, the fact that all the

significant results are in the same direction increases the probability that this is not a spurious result.

3.3.3 Nitrate

3.3.3.1 Annual Temporal Patterns

The 29 site 1979-1987 trend subset for annual nitrate concentration (Table A.5 and Figure A.34) has maximum median in 1980 of 1.28 mg/l. The lowest median is 0.92 mg/l in 1985. The annual nitrate deposition (Table A.6 and Figure A.34) has a maximum median in 1985 of 14.62 kg/ha and a minimum median in 1987 of 12.88 kg/ha. Annual concentration has substantial inter-annual variation, while the deposition variation is smaller.

The 119 site 1982-1987 trend subset for annual nitrate concentration (Table A.7 and Figure A.35) has a maximum median in 1987 of 1.54 mg/l. The lowest median is 1.25 mg/l in 1983. The annual nitrate deposition (Table A.8 and Figure A.35) has a maximum median in 1984 of 14.72 kg/ha and a minimum median in 1986 of 12.83 kg/ha. Again, deposition does not appear to have a clear trend, while concentration appears to have an increasing trend.

3.3.3.2 Temporal Change Analysis

Based on the modified KST test, two of the 39 1979-1987 trend sites exhibit a significantly ($p < 0.05$) decreasing trend in nitrate concentration: Sand Spring, CO ($-0.84 \mu\text{eq/l/year}$); and Manitou, CO ($-0.71 \mu\text{eq/l/year}$). The maximum decreasing trend was $-0.87 \mu\text{eq/l/year}$ at Chalk River, ON. With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.36 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates appear fairly consistent in the west of the Mississippi River, all decreasing. The eastern United States is inconsistent except along the eastern coast where the trends are increasing.

There are 11 sites in the 1982-1987 trend subset that have significantly increasing nitrate concentration: Dorset (b), ON ($2.87 \mu\text{eq/l/year}$); Nithgrove, ON ($2.40 \mu\text{eq/l/year}$); Parsons, WV ($2.22 \mu\text{eq/l/year}$); Golden Lake, ON ($1.93 \mu\text{eq/l/year}$); Pawnee, CO ($1.51 \mu\text{eq/l/year}$); Yosemite, CA (1.49

$\mu\text{eq/l/year}$); Sequoia National Park, CA ($1.39 \mu\text{eq/l/year}$); Washington Xing, NJ ($1.27 \mu\text{eq/l/year}$); Walker Branch, TN ($1.23 \mu\text{eq/l/year}$); Oak Ridge, TN ($1.03 \mu\text{eq/l/year}$); and Bradford Forest, FL ($0.35 \mu\text{eq/l/year}$). Figure A.37 displays the trend estimates and confidence intervals for all 148 sites. The spatial pattern of the trend estimates shows only small local areas of consistency. The magnitude of the estimated 1987 nitrate concentration appears to be unrelated to the trend estimate, but is related to the variability of the trend estimate.

For 1979-1987 trend sites (Figure A.38), significant nitrate deposition decreases occur at three sites: Hubbard Brook, NH ($-0.93 \text{ meq/m}^2/\text{year}$); Coweeta, NC ($-0.85 \text{ meq/m}^2/\text{year}$); and Marcell, MN ($-0.51 \text{ meq/m}^2/\text{year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. The maximum decreasing trend was $-0.98 \text{ meq/m}^2/\text{year}$ at Zanesville OH. While all the trends were increasing in New York, Pennsylvania and Virginia, they were decreasing in West Virginia and North Carolina. The mid-western states had no consistent trend. The sites with significant deposition trends were not the same sites with significant concentration trends.

Two of 1982-1987 trend sites had significantly increasing nitrate deposition: Bennett Bridge, NY ($3.01 \text{ meq/m}^2/\text{year}$), and Whiteface, NY ($1.74 \text{ meq/m}^2/\text{year}$); and Quabbin Reservoir, MA ($-2.64 \text{ meq/m}^2/\text{year}$) had a significant decreasing trend. With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.39 displays the trend estimates and confidence intervals for all 148 sites. The trends west of the Mississippi River are fairly consistent, while the trends in the east show only small local areas of consistency. Note that the three significant sites were in the same area, New England, but were not all in the same direction. As with most of the other ion species, the magnitude of the estimated 1987 nitrate deposition is not related to the trend but is related to the magnitude of the variation of the trend estimate.

3.3.4 Ammonium

3.3.4.1 Annual Temporal Patterns

The 30 site 1979-1987 trend subset for annual ammonium concentration (Table A.5 and Figure A.40) has approximately equal maximum medians in 1980 and 1981 of 0.30 mg/l. 1979 and 1985 has approximately equal lowest medians, 0.25 mg/l. The annual ammonium deposition (Table A.6 and Figure A.40) has a maximum median in 1983 of 2.86 kg/ha and a minimum median in 1985 of 2.46 kg/ha. Neither concentration nor deposition exhibits a clear increasing or decreasing trend.

The 119 site 1982-1987 trend subset for annual ammonium concentration (Table A.7 and Figure A.41) has a maximum median in 1987 of 0.32 mg/l and minimum medians of 0.28 mg/l in 1983 and 1985. The annual ammonium deposition (Table A.8 and Figure A.41) has a maximum median of 3.06 kg/ha in 1984 and a minimum median of 2.64 kg/ha in 1986. There does not appear to be a trend in either the concentration or deposition for this trend subset.

3.3.4.2 Temporal Change Analysis

Based on the modified KST test, one of the 39 1979-1987 trend sites exhibits a significantly ($p < 0.05$) decreasing trend in ammonium concentration: Dixon Springs, IL ($-0.65 \mu\text{eq/l/year}$); and one site has a significantly increasing trend: Zanesville, OH ($0.57 \mu\text{eq/l/year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. The maximum decreasing trend was $-1.35 \mu\text{eq/l/year}$ at Mead, NE. Figure A.42 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates appear fairly consistent in the eastern United States, all decreasing with the magnitude decreasing to the east. The western United States also was consistent with decreasing trends. The transition between increasing and decreasing trends occurs in the Midwest (Illinois and Indiana).

The 1982-1987 trend sites have three sites with significantly decreasing trends: Alamosa, CO ($-2.76 \mu\text{eq/l/year}$); Craters of Moon, ID ($-1.14 \mu\text{eq/l/year}$); and Everglades National Park, FL ($-0.39 \mu\text{eq/l/year}$); and two

sites with significantly increasing trends: Nithgrove, ON ($1.28 \mu\text{eq/l/year}$), and Uvalda, ON ($0.68 \mu\text{eq/l/year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.43 displays the trend estimates and confidence intervals for all 148 sites. The spatial pattern of the trend estimates show only small local areas of consistency. Some areas, such as southeastern Ontario, have sites with large increasing and decreasing trends situated next to each other. The magnitude of the trend estimate is not related to the estimated 1987 ammonium concentration.

For 1979-1987 trend sites (Figure A.44), significant ammonium deposition decreases occur at two sites: Marcell, MN ($-0.79 \text{ meq/m}^2/\text{year}$, maximum across all sites); and Chalk River, ON ($-0.60 \text{ meq/m}^2/\text{year}$). There are three sites with significant increasing trends: Whiteface, NY ($0.66 \text{ meq/m}^2/\text{year}$); Scranton, PA ($0.62 \text{ meq/m}^2/\text{year}$); and Virginia, VA ($0.61 \text{ meq/m}^2/\text{year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. There do not appear to be any consistent regional trends in the eastern United States. The sites with significant deposition trends were not the same sites with significant concentration trends.

The 1982-1987 trend sites have six sites with significantly decreasing trends: Alvinston, ON ($-1.81 \text{ meq/m}^2/\text{year}$); Craters of Moon, ID ($-0.70 \text{ meq/m}^2/\text{year}$); Fernberg, MN ($-0.69 \text{ meq/m}^2/\text{year}$); Hubbard Brook, NH ($-0.54 \text{ meq/m}^2/\text{year}$); Big Bend National Park, TX ($-0.29 \text{ meq/m}^2/\text{year}$); and Pinedale, WY ($-0.29 \text{ meq/m}^2/\text{year}$); and two sites with significantly increasing trends: Bennett Bridge, NY ($2.30 \text{ meq/m}^2/\text{year}$), and Whiteface, NY ($1.32 \text{ meq/m}^2/\text{year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.45 displays the trend estimates and confidence intervals for all 148 sites. The spatial pattern of the trend estimates show only small local areas of consistency. Some areas, such as southeastern Ontario, have sites with large increasing and decreasing trends situated next to each other. The magnitude of the trend estimate is not related to the estimated 1987 ammonium deposition.

3.3.5 Calcium Patterns

3.3.5.1 Annual Temporal Patterns

The 30 site 1979-1987 trend subset for annual calcium concentration (Table A.5 and Figure A.46) has a maximum median in 1980 of 0.21 mg/l. 1985 has the lowest median, 0.13 mg/l. The annual calcium deposition (Table A.6 and Figure A.46) has a maximum median in 1984 of 1.70 kg/ha and a minimum median in 1987 of 0.99 kg/ha. Except for 1979, annual concentration appears to decrease. Additionally, the upper range (median to maximum) of the concentration has decreased over time, with the largest change between 1981 and 1982. The deposition has no trend prior to 1985 and has decreased since then.

The 114 site 1982-1987 trend subset for annual calcium concentration (Table A.7 and Figure A.47) has a maximum median in 1984 of 0.20 mg/l and a minimum median of 0.16 mg/l in 1986. The calcium deposition (Table A.8 and Figure A.47) has a maximum median in 1984 of 1.85 kg/ha and a minimum median of 1.27 kg/ha in 1987. The three highest median concentrations are in the first three years and the three smallest medians are in the last three years suggesting a decrease. Deposition does show a significant trend. The 1984 median deposition is significantly greater than the 1986 and 1987 median deposition.

3.3.5.2 Temporal Change Analysis

Based on the modified KST test, 13 of the 39 1979-1987 trend sites exhibits a significantly ($p < 0.05$) decreasing trend in calcium concentration: Chalk River, ON (-1.63 $\mu\text{eq/l/year}$, maximum among all sites); Sand Spring, CO (-1.53 $\mu\text{eq/l/year}$); Fort Wayne, IN (-0.78 $\mu\text{eq/l/year}$); Manitou, CO (-0.76 $\mu\text{eq/l/year}$); Mead, NE (-0.75 $\mu\text{eq/l/year}$); Dixon Springs, IL (-0.61 $\mu\text{eq/l/year}$); Caldwell, OH (-0.61 $\mu\text{eq/l/year}$); Marcell, MN (-0.56 $\mu\text{eq/l/year}$); Parsons, WV (-0.52 $\mu\text{eq/l/year}$); Kane, PA (-0.50 $\mu\text{eq/l/year}$); Hubbard Brook, NH (-0.34 $\mu\text{eq/l/year}$); Huntington, NY (-0.31 $\mu\text{eq/l/year}$); and Lewiston, NC (-0.28 $\mu\text{eq/l/year}$). Figure A.48 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates appear fairly

consistent with the largest decreases in the west and the smallest decreases on the east coast.

The 1982-1987 trend sites have nine sites with significantly decreasing concentrations: Dalhousie Mills, ON (-2.49 $\mu\text{eq/l/year}$); Victoria, TX (-1.75 $\mu\text{eq/l/year}$); Fort Wayne, IN (-1.16 $\mu\text{eq/l/year}$); Manitou, CO (-1.10 $\mu\text{eq/l/year}$); Sand Spring, CO (-0.94 $\mu\text{eq/l/year}$); Everglades National Park, FL (-0.61 $\mu\text{eq/l/year}$); Lewiston, NC (-0.49 $\mu\text{eq/l/year}$); East, MA (-0.49 $\mu\text{eq/l/year}$); and Kane, PA (-0.47 $\mu\text{eq/l/year}$). There are also three sites with significantly increasing concentrations: Longwoods (b), ON (3.25 $\mu\text{eq/l/year}$); North Easthope, ON (1.67 $\mu\text{eq/l/year}$); and Charleston Lake, ON (1.39 $\mu\text{eq/l/year}$). Figure A.49 displays the trend estimates and confidence intervals for all 148 sites. The spatial pattern of the calcium concentration trends are relatively consistent for a number of large regions. However, in southeastern Ontario, and upstate New York, the trends are not consistent, with neighboring sites having large increasing and decreasing trends.

For 1979-1987 trend sites (Figure A.50), significant calcium deposition decreases occur at 17 sites: Chalk River, ON (-1.52 $\text{meq/m}^2/\text{year}$, maximum across all sites); Parsons, WV (-0.87 $\text{meq/m}^2/\text{year}$); Zanesville, OH (-0.75 $\text{meq/m}^2/\text{year}$); Caldwell, OH (-0.59 $\text{meq/m}^2/\text{year}$); Fort Wayne, IN (-0.57 $\text{meq/m}^2/\text{year}$); Kane, PA (-0.46 $\text{meq/m}^2/\text{year}$); Hubbard Brook, NH (-0.43 $\text{meq/m}^2/\text{year}$); Coweeta, NC (-0.43 $\text{meq/m}^2/\text{year}$); Penn State, PA (-0.42 $\text{meq/m}^2/\text{year}$); Piedmont Station, NC (-0.41 $\text{meq/m}^2/\text{year}$); Wooster, OH (-0.40 $\text{meq/m}^2/\text{year}$); Oxford, OH (-0.31 $\text{meq/m}^2/\text{year}$); ELA (b), ON (-0.29 $\text{meq/m}^2/\text{year}$); Lewiston, NC (-0.24 $\text{meq/m}^2/\text{year}$); Finley (A), NC (-0.21 $\text{meq/m}^2/\text{year}$); and Manitou, CO (-0.20 $\text{meq/m}^2/\text{year}$). The trend estimates appear fairly consistent with the smallest increases on the east coast and the largest decreases in Ohio, West Virginia and Indiana.

The 1982-1987 trend sites have 13 sites with significantly decreasing trends: Dalhousie Mills, ON (-2.47 $\text{meq/m}^2/\text{year}$); Dixon Springs, IL (-1.52 $\text{meq/m}^2/\text{year}$); Ashland, MO (-1.17 $\text{meq/m}^2/\text{year}$); Chalk River, ON (-1.11 $\text{meq/m}^2/\text{year}$); University Forest, MO (-1.04 $\text{meq/m}^2/\text{year}$); Walker Branch, TN (-0.98 $\text{meq/m}^2/\text{year}$); Forest Seed Center, TX (-0.93 $\text{meq/m}^2/\text{year}$); Oak Ridge,

TN ($-0.67 \text{ meq/m}^2/\text{year}$); Caldwell, OH ($-0.62 \text{ meq/m}^2/\text{year}$); Huntington, NY ($-0.48 \text{ meq/m}^2/\text{year}$); East, MA ($-0.47 \text{ meq/m}^2/\text{year}$); Newcastle, WY ($-0.39 \text{ meq/m}^2/\text{year}$); and Fernberg, MN ($-0.37 \text{ meq/m}^2/\text{year}$). There is one site with a significantly increasing trend: Charleston Lake, ON ($1.50 \text{ meq/m}^2/\text{year}$). Figure A.51 displays the trend estimates and confidence intervals for all 148 sites. As with the concentration trend estimates, the spatial pattern of the deposition estimates are consistent for a number of large regions, except for southeastern Ontario and upstate New York. All the significantly decreasing trends are for sites with estimated 1987 deposition magnitudes near the median. As with the other ion species, the magnitude of the variation in the trend estimate increases with the estimated 1987 deposition.

3.3.6 Chloride

3.3.6.1 Annual Temporal Patterns

The 28 site 1979-1987 trend subset for annual chloride concentration (Table A.5 and Figure A.52) has approximately equal maximum medians in 1979 and 1980 of 0.22 mg/l . 1983 has the lowest median, 0.15 mg/l . The 1980 and 1983 medians are significantly different. The annual chloride deposition (Table A.6 and Figure A.52) has a maximum median in 1979 of 2.69 kg/ha and a minimum median in 1987 of 1.72 kg/ha . There does not appear to be any consistent trends for either the concentration or deposition. The distribution of both the concentration and deposition are highly skewed and, except for 1981, there are extremely large values that are not included on Figure A.52.

The 115 site 1982-1987 trend subset for annual chloride concentration (Table A.7 and Figure A.53) has approximately equal maximum medians in 1982 and 1984 of 0.18 mg/l . 1983 and 1985 have approximately equal minimum medians of 0.15 mg/l . The 1983 and 1984 medians are significantly different. The annual chloride deposition (Table A.8 and Figure A.53) has a maximum median of 1.74 kg/ha in 1983 and a minimum median of 1.47 kg/ha in 1987. There does not appear to be any consistent trends for either the concentration or deposition. As with the 1979-1987 trend subset, the

concentration and deposition are highly skewed with extremely large values that are not included on Figure A.53.

3.3.6.2 Temporal Change Analysis

Based on the modified KST test, eight of the 39 1979-1987 trend sites exhibits a significantly ($p < 0.05$) decreasing trend in chloride concentration: Chalk River, ON ($-1.15 \mu\text{eq/l/year}$, maximum among all sites); ELA (b), ON ($-0.81 \mu\text{eq/l/year}$); Brookhaven, NY ($-0.62 \mu\text{eq/l/year}$); Dixon Springs, IL ($-0.36 \mu\text{eq/l/year}$); Mead, NE ($-0.33 \mu\text{eq/l/year}$); Sand Spring, CO ($-0.23 \mu\text{eq/l/year}$); Fort Wayne, IN ($-0.22 \mu\text{eq/l/year}$); and Wellston, MI ($-0.19 \mu\text{eq/l/year}$). Figure A.54 displays the trend estimates and confidence intervals for all 39 sites. The spatial pattern of the trend estimates has small local area which fairly consistent except along the east coast.

The 1982-1987 trend subset has five sites with significantly decreasing trends for chloride concentration: Yellowstone, WY ($-0.46 \mu\text{eq/l/year}$); Cuba, NM ($-0.38 \mu\text{eq/l/year}$); McKellar, ON ($-0.28 \mu\text{eq/l/year}$); Clearfield, KY ($-0.25 \mu\text{eq/l/year}$); and Underhill Center, VT ($-0.20 \mu\text{eq/l/year}$). There are also three sites with significantly increasing trends: Lac Le Croix (a), ON ($0.70 \mu\text{eq/l/year}$); Whiteface, NY ($0.43 \mu\text{eq/l/year}$); and Dorion, ON ($0.36 \mu\text{eq/l/year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.55 displays the trend estimates and confidence intervals for all 148 sites. The trends appear fairly consistent except for the large, both increasing and decreasing, trends along the coast of both the Pacific and Atlantic Oceans and the Gulf of Mexico.

For 1979-1987 trend sites (Figure A.56), significant chloride deposition decreases occur at eight sites: Chalk River ON ($-0.96 \text{ meq/m}^2/\text{year}$, maximum across all sites); Piedmont Station, NC ($0.52 \text{ meq/m}^2/\text{year}$); Coweeta, NC ($-0.44 \text{ meq/m}^2/\text{year}$); ELA (b), ON ($-0.44 \text{ meq/m}^2/\text{year}$); Zanesville, IL ($-0.33 \text{ meq/m}^2/\text{year}$); Hubbard Brook, NH ($-0.32 \text{ meq/m}^2/\text{year}$); Caldwell, OH ($-0.21 \text{ meq/m}^2/\text{year}$); and Wellston, MI ($-0.15 \text{ meq/m}^2/\text{year}$). The spatial pattern of the trend estimates has small local area which fairly consistent except along the east coast.

The 1982-1987 trend sites have eight sites with significantly decreasing trends for chloride deposition: Forest Seen Center, TX (-1.62 meq/m²/year); Selma, AL (-0.94 meq/m²/year); Clinton, MS (-0.81 meq/m²/year); Sequoia National Park, CA (-0.67 meq/m²/year); ELA (b), ON (-0.60 meq/m²/year); Argonne, IL (-0.26 meq/m²/year); Underhill Center, VT (-0.17 meq/m²/year); and Newcastle, WY (-0.11 meq/m²/year). There is also one site with a significantly increasing trend: Whiteface, NY (0.43 meq/m²/year). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.57 displays the trend estimates and confidence intervals for all 148 sites. The trends appear fairly consistent except for the large, almost all decreasing, trends along the coast of both the Pacific and Atlantic Oceans and the Gulf of Mexico. The trends do not appear to be related with the magnitude of the 1987 estimated chloride deposition, while the variation in the trend estimate does appear to increase with the magnitude of the 1987 estimated deposition.

3.3.7 Sodium

3.3.7.1 Annual Temporal Patterns

The 28 site 1979-1987 trend subset for annual sodium concentration (Table A.5 and Figure A.58) has a maximum median in 1979 of 0.14 mg/l. 1987 has the lowest median, 0.05 mg/l. The annual sodium deposition (Table A.6 and Figure A.58) has a maximum median in 1979 of 2.87 kg/ha and a minimum median in 1987 of 0.52 kg/ha. Both the concentration and deposition decreased dramatically between 1980 and 1981 and has continued to decrease since then, but at a slower pace. The concentration and deposition medians for 1982 through 1987 are all significantly different from the 1979 concentration and deposition median, respectively.

The 111 site 1982-1987 trend subset for annual sodium concentration (Table A.7 and Figure A.59) has a maximum median in 1984 of 0.09 mg/l and a minimum median concentration of 0.06 mg/l in 1986. The annual sodium deposition (Table A.8 and Figure A.59) has maximum medians of 0.73 kg/ha in 1982 and 1983 and a minimum median of 0.54 kg/ha in 1987. While the

deposition appears to be decreasing, the concentration does not appear to have any trend.

3.3.7.2 Temporal Change Analysis

Based on the modified KST test, 22 of the 39 1979-1987 trend sites exhibit a significantly ($p < 0.05$) decreasing trend in sodium concentration: Chalk River, ON ($-1.23 \mu\text{eq/l/year}$, maximum among all sites); Clinton Station, NC ($-0.98 \mu\text{eq/l/year}$); Brookhaven, NY ($-0.89 \mu\text{eq/l/year}$); ELA (b), ON ($-0.80 \mu\text{eq/l/year}$); Mead, NE ($-0.75 \mu\text{eq/l/year}$); Manitou, CO ($-0.75 \mu\text{eq/l/year}$); Piedmont Station, NC ($-0.71 \mu\text{eq/l/year}$); Horton's Station, VA ($-0.63 \mu\text{eq/l/year}$); Lamberton, MN ($-0.58 \mu\text{eq/l/year}$); Sand Spring, CO ($-0.58 \mu\text{eq/l/year}$); Hubbard Brook, NH ($-0.49 \mu\text{eq/l/year}$); Penn State, PA ($-0.46 \mu\text{eq/l/year}$); Delaware, OH ($-0.43 \mu\text{eq/l/year}$); Coweeta, NC ($-0.38 \mu\text{eq/l/year}$); Fort Wayne, IN ($-0.33 \mu\text{eq/l/year}$); Dixon Springs, IL ($-0.31 \mu\text{eq/l/year}$); Caldwell, OH ($-0.30 \mu\text{eq/l/year}$); Virginia, VA ($-0.27 \mu\text{eq/l/year}$); Scranton, PA ($-0.27 \mu\text{eq/l/year}$); Oxford, OH ($-0.26 \mu\text{eq/l/year}$); Kane, PA ($-0.17 \mu\text{eq/l/year}$); and Whiteface, NY ($-0.10 \mu\text{eq/l/year}$). Figure A.60 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates appear fairly consistent with the smallest decreases occurring in the Midwest and south of the Great Lakes. The sites with the largest estimated 1987 sodium concentration, did not have significant decreases in the sodium concentration trend.

Only six of the 148 1982-1987 trend sites have significantly decreasing trends for sodium concentration: Dixon Springs, IL ($-0.49 \mu\text{eq/l/year}$); Penn State, PA ($-0.46 \mu\text{eq/l/year}$); Oxford, OH ($-0.37 \mu\text{eq/l/year}$); Virginia, VA ($-0.30 \mu\text{eq/l/year}$); Fort Wayne, IN ($-0.26 \mu\text{eq/l/year}$); and Clearfield, KY ($-0.20 \mu\text{eq/l/year}$). Additionally, one of the sites has a significantly increasing trend: Uvalda, ON ($0.61 \mu\text{eq/l/year}$). With the number of sites being tested, a small number of sites with significant trends would be expected even if no trend actually existed. Figure A.61 displays the trend estimates and confidence intervals for all 148 sites. The spatial pattern of the trends show the largest, although not significant, decreasing trends were along the north Atlantic Coast.

For 1979-1987 trend sites (Figure A.62), significant sodium deposition decreases occur at 18 sites: Chalk River, ON (-1.19 meq/m²/year); Coweeta, NC (-0.90 meq/m²/year); Piedmont Station, NC (-0.78 meq/m²/year); Georgia Station, GA (-0.67 meq/m²/year); Horton's Station, VA (-0.59 meq/m²/year); Hubbard Brook, NH (-0.55 meq/m²/year); ELA (b), ON (-0.51 meq/m²/year); Zanesville, OH (-0.46 meq/m²/year); Delaware, OH (-0.38 meq/m²/year); Kane, PA (-0.33 meq/m²/year); Caldwell, OH (-0.33 meq/m²/year); Mead, NE (-0.33 meq/m²/year); Penn State, PA (-0.31 meq/m²/year); Fort Wayne, IN (-0.20 meq/m²/year); Huntington, NY (-0.17 meq/m²/year); Sand Spring, CO (-0.15 meq/m²/year); Wellston, MI (-0.14 meq/m²/year); and Ithaca, NY (-0.11 meq/m²/year). The maximum decrease, -1.28 meq/m²/year, occurred at Davis, CA. The trend estimates appear fairly consistent with the largest decreases in the southeast and the decreases becoming smaller towards the Great Lakes.

The 1982-1987 trend sites have 11 sites with significantly decreasing trends for sodium deposition: Forest Seed Center, TX (-1.36 meq/m²/year); Sequoia National Park, CA (-0.71 meq/m²/year); Clinton, MS (-0.71 meq/m²/year); Meridian, MS (-0.53 meq/m²/year); Argonne, IL (-0.43 meq/m²/year); Penn State, PA (-0.42 meq/m²/year); Alvinston, ON (-0.28 meq/m²/year); Fort Wayne, IN (-0.22 meq/m²/year); Palmerston, ON (-0.16 meq/m²/year); Fernberg, MN (-0.11 meq/m²/year); and Gaylord, MI (-0.07 meq/m²/year). Figure A.63 displays the trend estimates and confidence intervals for all 148 sites. The largest decreasing trends, although not significant, were along the north Atlantic and Pacific coasts and in Texas.

3.3.8 Potassium

3.3.8.1 Annual Temporal Patterns

The 26 site 1979-1987 trend subset for annual potassium concentration (Table A.5 and Figure A.64) has approximately equal maximum medians in 1979, 1980, 1981 and 1983 of 0.03 mg/l. All the other years have approximately equal lowest medians, 0.02 mg/l. The annual potassium deposition (Table A.6 and Figure A.64) has a maximum median in 1979 of 0.52 kg/ha and a minimum median in 1987 of 0.23 kg/ha. There do not appear to be any consistent trends for the concentration and deposition.

The 110 site 1982-1987 trend subset for annual potassium concentration (Table A.7 and Figure A.65) has the largest median in 1984 and smallest median in 1986. The 1984 median is significantly larger than the medians for 1985 through 1987. The annual potassium deposition (Table A.8 and Figure A.65) has the largest median in 1983, 0.36 kg/ha and the smallest median, 0.23 kg/ha, in 1987, with the 1983 and 1984 medians being significantly greater than the 1986 and 1987 medians. Both concentration and deposition appear to have decreasing trends.

3.3.8.2 Temporal Change Analysis

Based on the modified KST test, 10 of the 39 1979-1987 trend sites exhibit a significantly ($p < 0.05$) decreasing trend in potassium concentration: Chalk River, ON (-0.43 $\mu\text{eq/l/year}$, maximum among all sites); ELA (b), ON (-0.24 $\mu\text{eq/l/year}$); Mead, NE (-0.10 $\mu\text{eq/l/year}$); Lamberton, MN (-0.10 $\mu\text{eq/l/year}$); Fort Wayne, IN (-0.08 $\mu\text{eq/l/year}$); Marcell, MN (-0.06 $\mu\text{eq/l/year}$); Bondville, IL (-0.05 $\mu\text{eq/l/year}$); Dixon Springs, IL (-0.04 $\mu\text{eq/l/year}$); Parsons, WV (-0.04 $\mu\text{eq/l/year}$); and Kane, PA (-0.03 $\mu\text{eq/l/year}$). Figure A.66 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates appear fairly consistent with the decreases occurring in the Midwest to the west coast, except for the two highest values in Canada.

The 1982-1987 trend sites have 12 significantly decreasing trends for potassium concentration: Chalk River, ON (-0.31 $\mu\text{eq/l/year}$); ELA (b), ON (-0.18 $\mu\text{eq/l/year}$); Kejimikujik (b), NS (-0.17 $\mu\text{eq/l/year}$); Pickle Lake, ON (-0.15 $\mu\text{eq/l/year}$); University Forest, MO (-0.11 $\mu\text{eq/l/year}$); Nithgrove, ON (-0.11 $\mu\text{eq/l/year}$); Meridian, MS (-0.10 $\mu\text{eq/l/year}$); Alsea, OR (-0.07 $\mu\text{eq/l/year}$); Organ Pipe Monument, AZ (-0.05 $\mu\text{eq/l/year}$); Shabbona, IL (-0.04 $\mu\text{eq/l/year}$); Kane, PA (-0.04 $\mu\text{eq/l/year}$); and Turner Falls, MA (-0.03 $\mu\text{eq/l/year}$). There is also one site with a significantly increasing trend: Uvalda, ON (0.15). Of the 11 largest increasing trends for potassium concentration, 9 of them are from the MAP3S/PCN network. The MAP3S/PCN sites had 50% to 100% increases in annual potassium concentrations between 1986 and 1987. Figure A.67 displays the spatial pattern and confidence intervals for the potassium concentration. The consistency of the spatial pattern of

the trends in the east is primarily disturbed by the increasing trends at the MAP3S/PCN sites.

For 1979-1987 trend sites (Figure A.68), significant potassium deposition decreases occur at seven sites: Chalk River, ON (-0.37 meq/m²/year, maximum across all sites); ELA (b), ON (-0.13 meq/m²/year); Zanesville, IL (-0.07 meq/m²/year); Parsons, WV (-0.05 meq/m²/year); Fort Wayne, IN (-0.05 meq/m²/year); Raleigh, NC (-0.05 meq/m²/year); and Bondville, IL (-0.03 meq/m²/year). The trend estimates appear fairly consistent except along the east coast. The trend estimates for the deposition have the same pattern as the concentration.

There are 18 sites in the 1982-1987 trend site with significantly decreasing potassium deposition trends: Kejimikujik (b), NS (-0.23 meq/m²/year); Chalk River, ON (-0.23 meq/m²/year); Cambellford, ON (-0.13 meq/m²/year); Selma, AL (-0.12 meq/m²/year); Tanbark Flat, CA (-0.12 meq/m²/year); McKellar, ON (-0.11 meq/m²/year); ELA (b), ON (-0.10 meq/m²/year); Alvinston, ON (-0.10 meq/m²/year); Alsea, OR (-0.09 meq/m²/year); Ear Falls, ON (-0.08 meq/m²/year); University Forest, MO (-0.08 meq/m²/year); Coldwater, ON (-0.07 meq/m²/year); Bondville, IL (-0.07 meq/m²/year); Dixon Springs, IL (-0.07 meq/m²/year); Palmerston, ON (-0.07 meq/m²/year); Cuba, NM (-0.03 meq/m²/year); Craters of Moon, ID (-0.03 meq/m²/year); and Dorion, ON (-0.03 meq/m²/year). Additionally, there was one site with a significantly increasing potassium deposition: Whiteface, NY (0.23 meq/m²/year). The MAP3S/PCN sites had nine of the 14 largest increasing potassium deposition trends (the other five are in Ontario). Figure A.69 displays the spatial pattern and confidence intervals for the potassium deposition. The consistency of the spatial pattern of the trends in the east is primarily disturbed by the increasing trends at the MAP3S/PCN sites and in Ontario.

3.3.9 Magnesium

3.3.9.1 Annual Temporal Patterns

The 30 site 1979-1987 trend subset for annual magnesium concentration (Table A.5 and Figure A.70) has a maximum median in 1981 of 0.04 mg/l. 1983, 1985, 1986 and 1987 have approximately equal lowest medians, 0.02 mg/l. The 1986 and 1987 medians are significantly smaller than the 1980 and 1981 medians. The annual magnesium deposition (Table A.6 and Figure A.70) has a maximum median in 1981 of 0.42 kg/ha and a minimum median in 1987 of 0.23 kg/ha. The 1986 and 1987 medians are significantly smaller than the all medians prior to 1985 except 1983. Both the concentration and deposition appear to have decreased, especially since 1984.

The 112 site 1982-1987 trend subset for annual magnesium concentration (Table A.7 and Figure A.71) has the smallest medians in 1986 and 1987. These medians are significantly smaller than the 1984 median and the 1986 median is also significantly smaller than the 1982 and 1983 medians. The annual magnesium deposition (Table A.8 and Figure A.71) has a maximum median of 0.38 kg/ha in 1984 and minimum medians of 0.27 kg/ha in 1986 and 1987. The 1986 and 1987 medians are significantly smaller than the 1982 through 1984 medians. Again, both the concentration and deposition appear to have decreased, especially since 1984.

3.3.9.2 Temporal Change Analysis

Based on the modified KST test, six of the 39 1979-1987 trend sites exhibits a significantly ($p < 0.05$) decreasing trend in magnesium concentration: Sand Spring, CO (-0.43 $\mu\text{eq/l/year}$, maximum among all sites); Chalk River, ON (-0.41 $\mu\text{eq/l/year}$); Fort Wayne, IN (-0.27 $\mu\text{eq/l/year}$); Clinton Station, NC (-0.26 $\mu\text{eq/l/year}$); Hubbard Brook, NH (-0.20 $\mu\text{eq/l/year}$); and Kane, PA (-0.09 $\mu\text{eq/l/year}$). Figure A.72 displays the trend estimates and confidence intervals for all 39 sites. The trend estimates are inconsistent, with the greatest inconsistencies along the east coast.

There are nine 1982-1987 trend sites with significantly decreasing magnesium concentration trends: Manitou, CO (-0.54 $\mu\text{eq/l/year}$); Clinton Station, NC (-0.44 $\mu\text{eq/l/year}$); Shallow Lake, ON (-0.42 $\mu\text{eq/l/year}$);

University Forest, MO (-0.40 $\mu\text{eq/l/year}$); Chalk Creek, ON (-0.39 $\mu\text{eq/l/year}$); East, MA (-0.35 $\mu\text{eq/l/year}$); Georgia Station, GA (-0.29 $\mu\text{eq/l/year}$); Huntington, NY (-0.18 $\mu\text{eq/l/year}$); and Kane, PA (-0.12 $\mu\text{eq/l/year}$). There is also one site with a significantly increasing trend: Uvalda, ON (0.23 $\mu\text{eq/l/year}$). Figure A.73 displays the spatial pattern and confidence intervals for the magnesium concentration trend estimates. The estimates are somewhat consistent in small local areas but there are several areas, such as southeastern Ontario, where the trends are not consistent.

For 1979-1987 trend sites (Figure A.74), significant magnesium deposition decreases occur at seven sites: Chalk River, ON (-0.40 $\text{meq/m}^2/\text{year}$, maximum across all sites); Fort Wayne, IN (-0.22 $\text{meq/m}^2/\text{year}$); Hubbard Brook, NH (-0.22 $\text{meq/m}^2/\text{year}$); Caldwell, OH (-0.19 $\text{meq/m}^2/\text{year}$); Delaware, OH (-0.18 $\text{meq/m}^2/\text{year}$); Zanesville, IL (-0.11 $\text{meq/m}^2/\text{year}$); and Oxford, OH (-0.07 $\text{meq/m}^2/\text{year}$). The trend estimates appear fairly consistent except along the east coast. The trend estimates for the deposition appear fairly consistent with most of the significant decreases in Ohio and Indiana.

There are 14 sites in the 1982-1987 trend subset with significantly decreasing magnesium deposition trends: Huron Park, ON (-0.98 $\text{meq/m}^2/\text{year}$); Merlin, ON (-0.75 $\text{meq/m}^2/\text{year}$); Alsea, OR (-0.54 $\text{meq/m}^2/\text{year}$); Shenandoah National Park, VA (-0.54 $\text{meq/m}^2/\text{year}$); University Forest, MO (-0.50 $\text{meq/m}^2/\text{year}$); Washington Xing, NJ (-0.43 $\text{meq/m}^2/\text{year}$); Meridian, MS (-0.39 $\text{meq/m}^2/\text{year}$); Kellogg, MI (-0.33 $\text{meq/m}^2/\text{year}$); Clinton, MS (-0.25 $\text{meq/m}^2/\text{year}$); Caldwell, OH (-0.22 $\text{meq/m}^2/\text{year}$); Fernberg, MN (-0.18 $\text{meq/m}^2/\text{year}$); Newcastle, WY (-0.16 $\text{meq/m}^2/\text{year}$); Cuba, NM (-0.14 $\text{meq/m}^2/\text{year}$); and Gowanda, ON (-0.11 $\text{meq/m}^2/\text{year}$). There is also one site with a significantly increasing trend: Whiteface, NY (0.09 $\text{meq/m}^2/\text{year}$). Figure A.75 displays the spatial pattern and confidence intervals for the magnesium concentration trend estimates. The estimates somewhat consistent in small local areas. Southeastern Ontario again is an unusual area.

4.0 MULTIVARIATE TEMPORAL PATTERNS

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 1979-1987 and 1982-1987 trend site subsets.

4.1 MULTIVARIATE TEMPORAL PATTERN CONSISTENCY ACROSS ION SPECIES

This section simultaneously considers the nine ion species: hydrogen, sulfate, nitrate, ammonium, calcium, chloride, sodium, potassium, and magnesium. To facilitate the simultaneous use of nine ions, the units used are in terms of equivalents instead of grams. The equivalents are obtained by dividing the number of grams by the formula weight and multiplying by the valance. The approach will be first to discuss the multivariate temporal patterns for concentration and then to discuss deposition. The discussion begins with an overview of the estimated 1987 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, a trend estimate associated with the modified Kendall seasonal tau test. The trend estimate is then discussed in terms of percent change during the period. The reference is the estimated 1987 annual concentration for an ion species at a site. The estimated annual concentration is used because not all the sites have annual summaries, due to missing or invalid samples during a significant portion of at least one season. Additionally, the estimates are more stable than the actual values, since the estimates take into account within-site variability and spatial correlation with nearby sites. 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 1987 rather than 1982 as the reference, results in greater percent changes for cations at most sites, since the cations have decreased markedly over that period of time. The

discussion for concentrations ends with a summary of the distributions and patterns of the z-values from the modified seasonal tau test statistic. All tables and figures appear at the end of Section 4.

4.1.1 Concentration Multivariate Patterns

Boxplots in Figure 4.1 give the distribution summaries of the estimated 1987 annual precipitation-weighted concentrations of all the ions for the 1979-1987 and 1982-1987 trend sites. Note that the boxplots use the 10th and 90th percentile for the 1979-1987 subset and the 5th and 95th percentile for the 1982-1987 subset. Hydrogen and sulfate ions have about the same relative median concentration in precipitation, approximately 40 $\mu\text{eq/l}$. Nitrate has a median concentration level of 20 $\mu\text{eq/l}$ and ammonium median level is 15 $\mu\text{eq/l}$, approximately. The median concentration for the other ion species are below 10 $\mu\text{eq/l}$. Sodium and chloride have a skewed distribution due to large concentrations at several coastal sites that have their precipitation chemistry influenced by sea salt. Calcium, potassium and magnesium also have skewed distributions, due to a number of sites with relatively high concentrations.

4.1.1.1 Trend Estimates

The distribution of Sen's median slope trend estimates for each ion species concentration are presented as boxplot displays in Figure 4.2 and the percentiles are shown in Tables 4.1 and 4.2 for the 1979-1987 trend set and the 1982-1987 trends sets, respectively. For the 1979-1987 subset only ammonium has more than 50% of the slope estimates greater than zero (increasing trends). All the sodium trends are negative (decreasing), approximately 90% of the sulfate, calcium and magnesium trends are negative and over 75% of the chloride and potassium trends are negative. Hydrogen and nitrate are approximately centered at zero trend. For the 1982-1987 subset the percentage of negative slope estimates are reduced for all the ions, except ammonium. The most striking differences are nitrate, with almost 75% of the trends increasing, and sodium with almost 50% of the trends increasing. This shift toward more positive slope estimates is not due entirely to the additional sites in the 1982-1987 subset. For all the ions,

over 50% of the 39 sites in both subsets had positive increases in their slope estimates (smaller negative slopes).

In addition to providing Sen's median slope estimate, the modified Kendall 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. For the 1979-1987 trend sites, only ammonium concentration has a site with a significant increasing trend. In contrast, all ion species have sites with significant decreasing trends. However, because there are 39 sites being tested simultaneously, it is not unusual for a small number of sites to be significant even when there is no trend (false positive). Assuming the tests are independent, for 39 simultaneous tests five or more significant results occur only 4.4% of the time when there is no trend, thus the number of significant decreasing slopes for sulfate, calcium, chloride, sodium, potassium and magnesium are greater than what would be expected if there is no trend.

For the 1982-1987 trend sites, there are fewer significant decreasing trends and more significant increasing trends. For 148 simultaneous tests five or more significant results occur over 86% of the time when there is no trend, nine or more significant results occur over 20% of the time when there is no trend and 12 or more significant results occur only 3.5% of the time when there is no trend. Therefore, only the number of significantly increasing slopes for nitrate and decreasing slopes for potassium are greater than what would be expected if there is no trend.

For hydrogen, sulfate, nitrate, ammonium and chloride, none of the sites which have significant trends in the 1979-1987 subset have significant trends in the 1982-1987 subset. For potassium and magnesium, only three sites with significant trends in the 1979-1987 subset have significant trends in the 1982-1987 subset. For calcium and sodium, only five and six sites, respectively, with significant trends in the 1979-1987 subset have significant trends in the 1982-1987 subset. The reduction in the number of significant trends for the 39 sites in both subsets can be attributed to two

factors: first, there is a decrease in power (the ability to detect a significant trend, when indeed one exists) because of the shorter time span in the 1982-1987 subset; and second, more than 50% of the slopes had positive increases (either became less negative or changed sign and became positive). As noted in Section 3, for many of the ions, the maximum concentrations occurred in 1980 and 1981, prior to the 1982-1987 subset, thus the slope estimates and significance of those slopes in the 1979-1987 subset are a reflection of the decreases from those 1980 and 1981 maximums.

Table 4.4 gives the ranks, by individual ion, of the concentration trend estimates for the 1979-1986 trend sites, ordered by the average rank of the nine ions. The rank of the precipitation trend estimate is also included. All the ions, except hydrogen behave in a somewhat consistent fashion within the sites. Generally, the sites with the greatest decreasing trend estimate for hydrogen concentration, have the greatest increasing trend estimates for the other ions, and vice versa. None of the trend estimates for the ion concentration appear to behave like the trend estimate for total precipitation. One site, Chalk River, ON (ADS ID 141a), is unusually consistent. For the nine ions, it has the largest decreasing trend for six of the ions (sulfate, nitrate, chloride, sodium, potassium, and calcium), the second most decreasing trend for magnesium, the second most increasing trend for hydrogen and the third most decreasing trend for nitrate.

4.1.1.2 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 estimated 1987 annual concentration for an ion species at a site. Table 4.5, Table 4.6 and Figure 4.3 summarize the concentration percent change results. For the 1979-1987 trend subset, sodium has the largest median percent change, -9.5%. Extrapolating this change back to 1979, the 1979 median sodium concentration is 85.5% larger in 1979 than in 1987. Calcium, (-6.1%), magnesium (-4.1%), potassium (-3.8%), chloride (-2.8%) and sulfate (-2.2%) also have relatively large median percent changes. For the 1982-1987 trend subset, the median percent changes are approximately the same as the 1979-1987 trend subset for only hydrogen, potassium and magnesium. The

percent change decreases by more than a half for sulfate, ammonium, calcium, chloride, and sodium. For nitrate, the median change in the 1979-1987 trend subset is approximately zero, while the 1982-1987 trend subset has a 1.7% median change.

4.1.1.3 z-Values

The modified 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 1979-1987 or the 1982-1987 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 sites may be present. Third, a common true mean trend in 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. If a general decreasing trend exists throughout the United States and southeastern Canada, then we would expect the entire observed z-value distribution to be shifted. If a decreasing trend is present in only a single region, the z-value distribution would reflect this by the presence of a larger than expected (compared to the normal distribution) lower tail of the distribution. As seen in Figure 4.4, the observed z-values are generally symmetric for most ions, especially in the 1982-1987 trend subset, which has far more sites than the 1979-1987 subset. Additionally, a number of ions have a significant negative shift, except nitrate that has a significant positive shift in the 1982-1987 trend subset. The symmetry of the distributions appears to indicate that if there are decreasing trends, then they are occurring throughout the region covered by the trend subsets, not in isolated regions.

4.1.2 1987 Deposition Multivariate Patterns

Boxplots in Figure 4.5 give the distribution summaries of the estimated 1987 annual total deposition of all the ions for the 1979-1987 and 1982-1987 trend sites. Sulfate has a median deposition of approximately 40 meq/m^2 . Hydrogen has a median deposition of approximately 35 meq/m^2 . Nitrate has a median deposition level of 20 meq/m^2 and ammonium median deposition level is 14 meq/m^2 , approximately. The median deposition for the other ion species are below 10 meq/m^2 . Sodium and chloride have a skewed distribution due to high deposition at several coastal sites that have their precipitation chemistry influenced by sea salt. Calcium, potassium and magnesium also have skewed distributions, due to a number of sites with relatively high deposition. The total deposition has less variation (a smaller interquartile range) than the precipitation-weighted concentration, see Figure 4.1, but the total deposition has larger extremes for hydrogen.

4.1.2.1 Trend Estimates

The distribution of Sen's median slope trend estimates for each ion species total deposition are presented as boxplot displays in Figure 4.6 and the percentiles are shown in Table 4.7 and Table 4.8 for the 1979-1986 and the 1982-1987 trend subsets, respectively. For the 1979-1987 subset no ion has more than 50% of the slope estimates greater than zero (increasing trend). All the sodium and calcium trends are negative (decreasing trends), over 90% of the potassium and magnesium trends are negative and over 75% of the sulfate and chloride trends are negative. Only hydrogen, nitrate and ammonium have less than 75% of their trend estimates decreasing. For the 1982-1987 subset, again no ion has more than 50% of the slope estimates greater than zero. Except for hydrogen, nitrate and ammonium, over 75% of the trend estimates are negative. Unlike the trend estimates for the concentration (see Section 4.1.1.1), the percentiles do not change markedly between the two trend subsets, except for calcium and sodium. For the 39 sites that are in both trend subsets, only sodium has more than 50% of the deposition trends estimates which are more positive (smaller decreasing trend) in 1982-1987 subset than in the 1979-1987 subset.

Table 4.9 summarizes the number of sites with significantly decreasing or increasing deposition trend estimates. As discussed in Section 4.1.1.1, because a large number of sites are being tested simultaneously, a few significant trends can be expected even when there is no change in the trend. However, for the 1979-1987 subset, sulfate, calcium, chloride, sodium, potassium and magnesium all have more significant decreasing trends than chance alone would usually explain. For the 1982-1987 subset, calcium, sodium, potassium and magnesium all have more significant decreasing trends than chance alone would usually explain. Additionally, sulfate has eight sites with decreasing trends and no sites with increasing trends. While eight or more significant trends can occur 32% of the time when there is no trend, these significant trends would be expected to be both negative and positive if there is no trend. Thus the decreasing sulfate deposition trend also appears to be significant. Few of the sites that had significant trends in the 1979-1987 subset, had significant trends in the 1982-1987 subset; one site each for sulfate, chloride, calcium and magnesium; two sites for sodium and three sites for potassium.

Table 4.10 gives the ranks, by individual ion, of the deposition trend estimates for the 1979-1986 trend sites, ordered by the average rank of the nine ions. The rank of the precipitation trend estimate is also included. Sulfate and nitrate trends behave consistently within sites. Hydrogen deposition trends, unlike hydrogen concentration trends, appear to behave similarly to the sulfate and nitrate deposition trends (except at Chalk River, ON, which has already been pointed out to be an unusual site). Sodium and chloride trends also appear consistent within sites. The other ions, all cations, do not generally appear to behave consistently within sites. The total precipitation trend estimate does appear to be consistent, within sites, with the deposition trends of hydrogen, sulfate and nitrate, which was not the case with the concentration trend estimates. The ordering of the sites by the average rank of the deposition trends appears to be unrelated to the ordering of the sites by the average rank of the concentration trends (see Table 4.4).

4.1.2.2 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 estimated 1987 annual deposition for an ion species at a site. Table 4.11, Table 4.12 and Figure 4.7 summarize the deposition percent change results. For the 1979-1987 trend subset, sodium has the largest median percent change, -10.4%. Extrapolating this change back to 1979, the median sodium deposition in 1979 is almost double the deposition in 1987. Calcium, (-5.5%), magnesium (-4.3%), potassium (-4.3%), chloride (-3.7%) and sulfate (-3.0%) also have relatively large median percent changes. For the 1982-1987 trend subset, the median percent changes are larger (more negative) than the median percent changes in the 1979-1987 subset for most ions. Only nitrate and sodium have smaller median percent changes and the median percent change for chloride remains about the same. This is unlike concentration, where the median percent change decreases (becomes less negative) for most of the ion species in the 1982-1987 subset.

4.1.2.3 z-Values

Figure 4.8 shows the distribution of Kendall seasonal tau test z-values for the 1979-1987 and 1982-1987 trend subsets. These distributions generally show a shift, in the negative direction, with the distributions remaining symmetric, with the possible exception of the cations (calcium, sodium, potassium and magnesium) in the 1982-1987 trend subsets. The shift, with symmetry maintained, indicates the trends are occurring throughout the region covered by the trend subsets, not in isolated regions. The asymmetry of the distributions for the cations indicate that the deposition trends may have some regionalization, which is discussed in Section 3.

FIGURE 4.1. Estimated 1987 Ion Species Concentration for (a) 1979-1987 Trend Sites and (b) 1982-1987 Trend Sites

TABLE 4.1. Change per Year ($\mu\text{eq/l/year}$) of Precipitation-weighted Annual Mean Concentration for 1979-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>10th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>90th</u>	<u>Max</u>
pH (a)	-0.04	-0.01	-0.01	0.00	0.01	0.01	0.05
Hydrogen	-2.51	-1.40	-0.73	-0.05	0.24	0.93	1.53
Sulfate	-2.87	-2.07	-1.52	-0.91	-0.44	0.04	0.66
Nitrate	-0.87	-0.71	-0.40	-0.01	0.29	0.47	0.92
Ammonium	-1.35	-0.65	-0.18	0.12	0.28	0.54	0.98
Calcium	-1.63	-0.78	-0.56	-0.35	-0.16	-0.08	0.25
Chloride	-1.15	-0.62	-0.28	-0.14	-0.05	0.12	0.64
Sodium	-1.28	-0.98	-0.70	-0.38	-0.22	-0.14	-0.10
Potassium	-0.43	-0.10	-0.06	-0.03	-0.01	0.01	0.06
Magnesium	-0.43	-0.26	-0.18	-0.09	-0.05	-0.01	0.30

(a) pH units

TABLE 4.2. Change per Year ($\mu\text{eq/l/year}$) of Precipitation-weighted Annual Mean Concentration for 1982-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>5th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>95th</u>	<u>Max</u>
pH (a)	-0.04	-0.03	-0.02	0.00	0.02	0.04	0.07
Hydrogen	-7.41	-3.00	-0.79	-0.04	0.90	2.27	4.26
Sulfate	-4.68	-3.12	-1.20	-0.28	0.44	2.33	4.59
Nitrate	-1.18	-0.72	-0.05	0.42	0.92	2.23	2.90
Ammonium	-2.76	-1.11	-0.42	0.05	0.55	1.40	2.29
Calcium	-3.00	-1.69	-0.50	-0.14	0.11	1.18	3.25
Chloride	-3.43	-0.71	-0.16	-0.03	0.09	0.45	1.12
Sodium	-2.94	-0.58	-0.19	-0.08	0.00	0.21	1.07
Potassium	-0.31	-0.13	-0.06	-0.03	-0.00	0.13	0.30
Magnesium	-1.25	-0.54	-0.23	-0.10	0.01	0.23	0.62

(a) pH units

FIGURE 4.2. Ion Species Concentration Trend Estimates for (a) 1979-1987 Trend Sites and (b) 1982-1987 Trend Sites

TABLE 4.3. Number and Percent of Significantly ($p < 0.05$) Decreasing and Increasing Trend Estimates by Ion Species for Concentrations at 1979-1987 and 1982-1987 Trend Sites

Ion Species	1979-1987 Trend Sites					1982-1987 Trend Sites				
	Significance					Significance				
	Decrease %	N	None N	Increase N	%	Decrease %	N	None N	Increase N	%
pH	0	0	36	3	8	0	0	144	4	3
Hydrogen	8	3	36	0	0	3	4	144	0	0
Sulfate	18	7	32	0	0	2	3	144	1	1
Nitrate	5	2	37	0	0	0	0	137	11	7
Ammonium	3	1	37	1	3	2	3	143	2	1
Calcium	33	13	26	0	0	6	9	136	3	2
Chloride	21	8	31	0	0	3	5	140	3	2
Sodium	56	22	17	0	0	4	6	141	1	1
Potassium	26	10	29	0	0	8	12	135	1	1
Magnesium	15	6	33	0	0	6	9	138	1	1

TABLE 4.4. Ranks by Individual Ion of Annual Concentration Temporal Change for 1979-1987 Trend Sites (Ordered by Average Rank)

SITENAME	Precip	H	SO4	NO3	CL	NH4	NA	K	CA	MG	Ave Rank
Chalk River	16	38	1	1	1	3	1	1	1	2	5.4
Mead	34	24	3	6	8	1	7	3	3	11	7.3
Manitou	15	31	2	4	10	8	8	8.5	5	7	9.3
Sand Spring	24	21	4	2	12	6	13	28	2	1	9.9
Fort Wayne	25	6	19	11	13	9	22	6	4	3	10.3
Dixon Springs	7	11	8	12	6	4	23	15	8.5	13	11.2
Lamberton	22	25	12	7	14	2	12	4	7	23	11.8
Hubbard Brook	12	8	17	5	11	15	15	22	21	9	13.7
Brookhaven	36	15	23	30	4	16	5	11	18	5	14.1
Davis	21	18	13	13.5	3	11	2	26	26	16	14.3
Piedmont Station	8	17	5	18	7	22	9	20	19	12	14.3
ELA (b)	23	30	16	16	2	29	6	2	6	24.5	14.6
Marcell	14	20	22	9	17	12	35	10	10	14	16.6
Bondville	30	26	7	3	25	5	36	12	25	18	17.4
Oxford	28	10	6	17	32	7	28	27	11	19.5	17.5
Bradford Forest	35	37	24	27	9	18	3	13.5	23	6	17.8
Scranton	38	1	9	20	16	19	27	17	27	30	18.4
Wellston	37	16	15	13.5	15	21	34	18.5	16	24.5	19.3
Huntington	33	14	10	10	19	14	31	29	22	27	19.6
Clinton Station	5	22	27	32	5	33	4	22	29	4	19.8
Caldwell	10	23	20	8	21	34	24	30	8.5	10	19.8
Lewiston	19	19	21	23	18	25	10	33	24	8	20.1
Illinois	31	12	14	19	37	17	32	24	20	17	21.3
Kane	29	36	26	15	30	13	33	18.5	13	19.5	22.7
Wooster	13	28	18	29	20	39	21	22	14.5	15	22.9
Georgia Station	20	27	30	22	28	10	17	25	36	22	24.1
Parsons	18	34	25	21	31	30	30	16	12	26	25.0
Zanesville	4	7	29	37	26	37	25	7	28	34	25.6
Raleigh	6	9	36	34	22	26	19	13.5	37	37	25.9
Penn State	17	35	37	39	35	31	16	8.5	14.5	21	26.3
Ithaca	27	2	11	31	38	23	38	36	30	33	26.9
Delaware	3	29	32	26	23	38	18	32	17	28	27.0
Finley (A)	9	33	28	28	24	28	14	35	31	29	27.8
Coweeta	1	32	33	24	27	20	20	31	34	31	28.0
Lewes	11	13	39	35	39	24	29	5	38	39	29.0
Whiteface	39	5	31	25	33	27	39	38	32	36	29.6
Virginia	32	3	35	36	36	32	26	39	35	32	30.4
Horton's Station	26	39	38	38	29	35	11	34	33	35	32.4
Rockport	2	4	34	33	34	36	37	37	39	38	32.4

TABLE 4.5. Percent Change per Year of Precipitation-weighted Annual Mean Concentration for 1979-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>10th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>90th</u>	<u>Max</u>
pH	-0.76	-0.28	-0.12	0.04	0.21	0.32	0.78
Hydrogen	-5.66	-3.98	-1.65	-0.36	0.82	2.55	5.39
Sulfate	-16.87	-6.87	-3.70	-2.22	-0.91	0.10	1.25
Nitrate	-6.10	-2.94	-1.81	-0.03	1.34	2.30	4.02
Ammonium	-8.19	-3.70	-1.62	0.71	1.91	3.44	5.16
Calcium	-25.41	-11.80	-7.40	-6.14	-2.85	-2.17	2.23
Chloride	-38.82	-11.67	-6.21	-2.83	-1.00	1.90	5.40
Sodium	-80.03	-26.97	-17.29	-9.50	-6.70	-5.27	-0.96
Potassium	-66.30	-12.75	-7.69	-3.77	-1.12	0.56	2.12
Magnesium	-23.77	-10.24	-6.24	-4.08	-2.39	-1.02	3.03

TABLE 4.6. Percent Change per Year of Precipitation-weighted Annual Mean Concentration for 1982-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>5th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>95th</u>	<u>Max</u>
pH	-0.99	-0.68	-0.32	0.03	0.38	0.92	1.32
Hydrogen	-14.77	-8.27	-3.34	-0.22	2.54	6.25	9.41
Sulfate	-11.99	-9.28	-3.53	-0.98	0.82	4.13	9.47
Nitrate	-9.67	-4.39	-0.28	1.79	3.96	8.23	12.74
Ammonium	-18.67	-9.37	-3.17	0.28	2.86	7.60	12.76
Calcium	-34.56	-17.61	-8.39	-2.62	1.42	7.94	16.77
Chloride	-44.75	-13.18	-3.42	-0.74	1.77	9.49	34.66
Sodium	-69.88	-14.07	-6.22	-2.86	0.03	6.25	14.34
Potassium	-48.83	-20.37	-10.41	-4.64	-0.56	8.05	20.08
Magnesium	-40.21	-19.35	-8.60	-3.76	0.42	5.33	16.04

FIGURE 4.3. Distribution of Ion Species Concentration Trend Estimates as Percent of Estimated 1987 Annual Concentration for (a) 1979-1987 Trend Sites and (b) 1982-1987 Trend Sites

FIGURE 4.4. Distribution of Kendall Seasonal Tau Test z-values for (a) 1979-1987 Trend Sites' Concentration and (b) 1982-1987 Trend Sites' Concentration

FIGURE 4.5. Estimated 1987 Ion Species Deposition for (a) 1979-1987 Trend Sites and (b) 1982-1987 Trend Sites

TABLE 4.7. Change per Year ($\text{meq/m}^2/\text{year}$) of Annual Deposition for 1979-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>10th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>90th</u>	<u>Max</u>
Precip ^(a)	-5.49	-3.38	-1.75	-0.04	1.10	2.38	4.07
Hydrogen	-4.00	-2.23	-1.17	-0.09	0.08	1.21	1.96
Sulfate	-3.42	-2.27	-1.64	-0.88	-0.18	0.45	0.64
Nitrate	-0.98	-0.65	-0.51	-0.20	0.19	0.52	0.84
Ammonium	-0.79	-0.45	-0.24	-0.07	0.10	0.45	0.66
Calcium	-1.52	-0.75	-0.43	-0.29	-0.20	-0.11	0.00
Chloride	-0.96	-0.44	-0.32	-0.13	-0.06	0.15	0.26
Sodium	-1.22	-0.79	-0.59	-0.32	-0.15	-0.07	-0.04
Potassium	-0.37	-0.07	-0.04	-0.03	-0.02	0.01	0.03
Magnesium	-0.40	-0.22	-0.11	-0.08	-0.05	0.00	0.04

(a) Annual precipitation (cm)

TABLE 4.8 Change per Year ($\text{meq/m}^2/\text{year}$) of Annual Deposition for 1982-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>5th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>95th</u>	<u>Max</u>
Precip ^(a)	-15.30	-9.28	-4.11	-1.85	-0.28	2.60	5.27
Hydrogen	-6.70	-4.04	-1.64	-0.50	0.02	1.76	5.04
Sulfate	-5.32	-3.54	-1.89	-1.08	-0.23	1.61	4.82
Nitrate	-2.64	-1.40	-0.66	-0.10	0.41	1.79	4.09
Ammonium	-2.71	-1.36	-0.67	-0.25	0.14	0.97	2.30
Calcium	-2.47	-1.48	-0.52	-0.32	-0.08	0.66	1.95
Chloride	-2.25	-1.18	-0.30	-0.11	0.00	0.20	0.87
Sodium	-2.54	-0.94	-0.34	-0.13	-0.06	0.04	0.40
Potassium	-0.23	-0.13	-0.07	-0.03	-0.02	0.09	0.27
Magnesium	-0.98	-0.56	-0.25	-0.13	-0.05	0.10	0.48

(a) Annual precipitation (cm)

FIGURE 4.6. Ion Species Deposition Trend Estimates for (a) 1979-1987 Trend Sites and (b) 1982-1987 Trend Sites

TABLE 4.9. Number and Percent of Significantly ($p < 0.05$) Decreasing and Increasing Trend Estimates by Ion Species for Deposition at 1979-1987 and 1982-1987 Trend Sites

Ion Species	1979-1987 Trend Sites					1982-1987 Trend Sites				
	Significance					Significance				
	Decrease %	N	None N	Increase N	%	Decrease %	N	None N	Increase N	%
Precipitation	5	2	37	0	0	1	1	147	0	0
Hydrogen	8	3	34	2	5	2	3	145	0	0
Sulfate	28	11	28	0	0	5	8	140	0	0
Nitrate	8	3	36	0	0	1	1	145	2	1
Ammonium	5	2	34	3	8	4	6	140	2	1
Calcium	44	17	22	0	0	9	13	134	1	1
Chloride	21	8	31	0	0	5	8	138	2	1
Sodium	46	18	21	0	0	7	11	137	0	0
Potassium	18	7	32	0	0	12	18	129	1	1
Magnesium	18	7	32	0	0	9	14	133	1	1

TABLE 4.10. Ranks of Individual Ion of Annual Deposition Temporal Change for 1979-1987 Trend Sites (Ordered by Average Rank)

SITENAME	Precip	H	SO4	NO3	CL	NH4	NA	K	CA	MG	Ave Rank
Coweeta	1	5	3	3	4.5	4	3	21	10	8	6.8
Chalk River	16	36	6	11	1	3	2	1	1	1	6.9
Zanesville	4	1	1	1	9	17	14	4	4	13	7.1
Piedmont Station	8	8	2	5	3	16	5	12	12	4	7.4
Hubbard Brook	12	9	5	2	10	14	12	22	9	4	9.7
Delaware	3	6	4	4	19	10	16	25.5	7	7	10.9
Clinton Station	5	10	12	12	2	27	4	16.5	13	4	11.2
Parsons	18	23	7	13	18	22	22	5.5	2	13	13.9
Caldwell	10	12	9	7	14	28	18.5	28.5	5	6	14.2
Finley (A)	9	14	8	10	8	12	6	33.5	27.5	13	14.7
Davis	21	24	15	16	6	8.5	1	16.5	27.5	18.5	14.8
Dixon Springs	7	11	10	9	13	2	32.5	23.5	3	29	14.8
Raleigh	6	3	13	14	11	5	15	8	36	29	14.9
Rockport	2	2	11	6	23	13	24	11	18	34	15.8
ELA (b)	23	19	21	18	4.5	24	13	2	20	29	16.7
Fort Wayne	25	13	31	23	22	24	26	5.5	6	2	16.9
Lewes	11	7	20	24	7	26	7	3	29	39	18.0
Marcell	14	21	22	8	24	7	28	16.5	16.5	20	18.1
Lamberton	22	27	18	19.5	25.5	1	23	8	19	22	18.1
Georgia Station	20	34	16	26	12	8.5	9	23.5	22.5	13	18.3
Wooster	13	16	14	28	21	36	20	28.5	14	13	21.2
Penn State	17	15	25	32	31	29	21	10	11	18.5	21.4
Lewiston	19	22	19	25	16	32	11	36	24	13	22.0
Mead	34	29	30	33	20	6	18.5	13	30	29	23.2
Wellston	37	20	26	15	17	24	32.5	28.5	33	17	23.7
Bondville	30	32	33	17	33	11	38	16.5	15	22	24.2
Horton's Station	26	30	23	21	15	30.5	10	33.5	32	25	24.4
Kane	29	39	39	31	25.5	33	17	16.5	8	13	24.7
Bradford Forest	35	38	36	35	28	30.5	8	20	22.5	9	25.2
Oxford	28	18	24	29	34	19	34	28.5	16.5	24	25.2
Manitou	15	26	27	19.5	27	20	28	25.5	31	26	25.6
Sand Spring	24	33	29	22	29.5	18	30	33.5	21	22	26.4
Huntington	33	35	28	27	29.5	15	28	31	25	29	27.5
Ithaca	27	4	17	30	35	34	35	37	34.5	33	28.8
Brookhaven	36	37	34	38	38	21	39	8	26	38	31.0
Scranton	38	17	38	36	32	38	31	16.5	37	35.5	31.2
Illinois	31	28	32	34	37	35	37	33.5	34.5	32	33.7
Virginia	32	31	35	37	36	37	25	38.5	38	35.5	34.8
Whiteface	39	25	37	39	39	39	36	38.5	39	37	36.6

TABLE 4.11. Percent Change per Year of Annual
Deposition for 1979-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>10th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>90th</u>	<u>Max</u>
Precip	-7.21	-3.78	-1.94	-0.09	1.25	2.31	3.87
Hydrogen	-9.35	-6.29	-3.15	-0.65	0.26	2.73	5.32
Sulfate	-34.26	-7.02	-5.11	-2.99	-0.71	0.70	1.44
Nitrate	-8.06	-3.95	-2.91	-1.20	0.61	1.96	3.73
Ammonium	-9.67	-4.32	-2.05	-0.59	1.09	2.23	4.37
Calcium	-30.30	-13.56	-8.26	-5.52	-3.54	-1.23	0.00
Chloride	-41.33	-9.90	-6.06	-3.70	-0.96	2.85	6.35
Sodium	-96.81	-24.79	-16.38	-10.37	-5.15	-3.14	-0.21
Potassium	-75.19	-11.30	-7.36	-4.27	-1.81	0.95	2.26
Magnesium	-29.59	-9.61	-7.67	-4.25	-2.40	0.00	1.71

TABLE 4.12. Percent Change per Year of Annual
Deposition for 1982-1987 Trend Sites

<u>Ion</u>	<u>Min</u>	<u>5th</u>	<u>25th</u>	<u>50th</u>	<u>75th</u>	<u>95th</u>	<u>Max</u>
Precip	-38.47	-11.01	-5.45	-2.39	-0.54	2.98	5.47
Hydrogen	-32.32	-18.70	-6.98	-2.91	0.25	4.46	6.99
Sulfate	-58.02	-17.38	-6.55	-3.46	-0.83	3.66	8.74
Nitrate	-37.16	-11.98	-4.23	-0.76	1.80	6.38	10.83
Ammonium	-90.60	-13.80	-6.43	-1.95	1.13	5.41	8.83
Calcium	-40.44	-19.91	-11.47	-6.27	-1.03	7.37	12.89
Chloride	-94.61	-21.25	-7.18	-3.34	0.05	5.11	15.08
Sodium	-73.73	-24.78	-10.52	-5.22	-2.27	2.08	9.40
Potassium	-78.59	-26.59	-12.48	-7.58	-3.11	10.11	15.94
Magnesium	-48.10	-24.42	-12.33	-6.70	-2.35	3.97	8.36

FIGURE 4.7. Distribution of Ion Species Deposition Trend Estimates as Percent of Estimated 1987 Annual Deposition for (a) 1979-1987 Trend Sites and (b) 1982-1987 Trend Sites

FIGURE 4.8. Distribution of Kendall Seasonal Tau Test z-values for (a) 1979-1987 Trend Sites' Wet Deposition and (b) 1982-1987 Trend Sites' Wet Deposition

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APPENDIX A

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

TABLE A.1. 1979-1987 Trend Sites Ordered by Sitename

Sitename	State/Province	Network	ADS ID	Latitude	Longitude	Comments
Bondville	Illinois	NADP/NTN	020a	40.05	-88.37	
Bradford Forest	Florida	NADP/NTN	015a	29.97	-82.20	0
Brookhaven	New York	MAP3S	048a	40.87	-72.08	1
Caldwell	Ohio	NADP/NTN	058a	39.79	-81.53	
Chalk River	Ontario	CAPMON/APN	141a	40.07	-77.40	4,5
Clinton Station	North Carolina	NADP/NTN	052a	35.03	-78.28	
Coveeta	North Carolina	NADP/NTN	050a	35.00	-83.43	
Davis	California	NADP/NTN	009a	38.54	-121.78	0
Delaware	Ohio	NADP/NTN	055a	40.30	-83.07	
Dixon Springs	Illinois	NADP/NTN	023a	37.44	-88.67	
ELA (b)	Ontario	CAPMON/APN	135b	49.67	-93.72	3,4,5
Finley (A)	North Carolina	NADP/NTN	053a	35.73	-78.88	0
Fort Wayne	Indiana	UAPSP	150a	41.04	-85.32	2,6
Georgia Station	Georgia	NADP/NTN	017a	33.18	-84.41	
Horton's Station	Virginia	NADP/NTN	073a	37.33	-80.56	0
Hubbard Brook	New Hampshire	NADP/NTN	039a	43.94	-71.70	
Huntington	New York	NADP/NTN	108a	43.97	-74.22	
Illinois	Illinois	MAP3S	020b	40.05	-88.37	0
Ithaca	New York	MAP3S	044a	42.40	-76.65	1
Kane	Pennsylvania	NADP/NTN	003a	41.00	-78.77	0
Lamberton	Minnesota	NADP/NTN	035a	44.24	-95.30	
Leves	Delaware	MAP3S	013a	38.77	-75.00	1
Lewiston	North Carolina	NADP/NTN	049a	36.13	-77.17	
Manitou	Colorado	NADP/NTN	011a	39.10	-105.09	0
Marcell	Minnesota	NADP/NTN	034a	47.63	-93.47	
Mead	Nebraska	NADP/NTN	030a	41.15	-96.49	
Oxford	Ohio	MAP3S	057a	39.53	-84.72	1
Parsons	West Virginia	NADP/NTN	075a	39.00	-79.66	
Penn State	Pennsylvania	MAP3S	005a	40.79	-77.95	1
Piedmont Station	North Carolina	NADP/NTN	051a	35.70	-80.02	
Raleigh	North Carolina	UAPSP	053c	35.73	-78.88	2,6
Rockport	Indiana	UAPSP	154a	37.88	-87.13	2,6
Sand Spring	Colorado	NADP/NTN	173a	40.51	-107.70	
Scranton	Pennsylvania	UAPSP	151a	41.68	-75.99	0
Virginia	Virginia	MAP3S	072a	38.04	-78.54	1
Wellston	Michigan	NADP/NTN	033a	44.22	-85.82	0
Whiteface	New York	MAP3S	043a	44.39	-73.86	1
Wooster	Ohio	NADP/NTN	050a	40.78	-81.92	
Zanesville	Ohio	UAPSP	153a	39.98	-82.02	2,6

Comments:

0 No annual averages

2 No annual average for sulfate

4 No annual average for chloride

6 No annual average for potassium

1 No annual average for hydrogen

3 No annual average for nitrate

5 No annual average for sodium

TABLE A.2. 1982-1987 Trend Sites Ordered by Sitename

Sitename	State/Province	Network	ADS ID	Latitude	Longitude	Comments
Acadia > 11/81	Maine	NADP/NTN	257a	44.37	-68.28	0
Alamo	Tennessee	UAPSP	248a	35.79	-89.13	0
Alamosa	Colorado	NADP/NTN	168a	37.44	-105.87	
Alsea	Oregon	NADP/NTN	059a	44.39	-123.82	
Alvinston	Ontario	APIOSC	188a	42.83	-81.83	
Argonne	Illinois	NADP/NTN	021a	41.78	-88.88	
Ashland	Missouri	NADP/NTN	252a	38.75	-92.28	
Aurora	New York	NADP/NTN	040a	42.73	-78.88	
Balsam Lake	Ontario	APIOSD	225a	44.63	-78.88	6,8
Bear Island	Ontario	APIOSC	198a	46.97	-80.88	0
Bennett Bridge	New York	NADP/NTN	046a	43.53	-75.95	
Bennington	Vermont	NADP/NTN	249a	42.88	-73.18	0
Big Bend Nat'l Pk	Texas	NADP/NTN	070a	29.38	-103.18	0
Big Moose	New York	UAPSP	243a	43.82	-74.98	2
Bondville	Illinois	NADP/NTN	020a	40.85	-88.37	
Bradford Forest	Florida	NADP/NTN	015a	29.97	-82.20	
Bridgton	Maine	NADP/NTN	164a	44.11	-78.73	
Brookhaven	New York	MAP3S	048a	40.87	-72.88	1
Brookings	South Dakota	UAPSP	245a	44.33	-98.83	2
Caldwell	Ohio	NADP/NTN	058a	39.79	-81.53	
Campbellford	Ontario	APIOSC	189a	44.29	-77.79	
Caribou (a)	Maine	NADP/NTN	163a	46.87	-68.81	
Chalk River	Ontario	CAPMON/APN	141a	46.87	-77.48	4,5
Charleston Lake	Ontario	APIOSD	227a	44.58	-76.84	6,8
Chautauqua	New York	NADP/NTN	041a	42.38	-79.48	
Clearfield	Kentucky	UAPSP	239a	38.14	-83.45	2
Clinton	Mississippi	UAPSP	242a	32.31	-90.32	2
Clinton Station	North Carolina	NADP/NTN	052a	35.83	-78.28	
Colchester	Ontario	APIOSC	178a	41.99	-82.93	
Coldwater	Ontario	APIOSC	190a	44.63	-79.54	
Coweeta	North Carolina	NADP/NTN	050a	35.88	-83.43	
Craters of Moon	Idaho	NADP/NTN	019a	43.48	-113.55	0
Cuba	New Mexico	NADP/NTN	288a	36.84	-106.97	
Dalhousie Mills	Ontario	APIOSC	193a	45.32	-74.47	
Davis	California	NADP/NTN	009a	38.54	-121.78	0
Delaware	Ohio	NADP/NTN	055a	40.38	-83.87	
Dixon Springs	Illinois	NADP/NTN	023a	37.44	-88.87	
Dorion	Ontario	APIOSC	204a	48.84	-88.81	
Dorset (b)	Ontario	APIOSD	087b	45.22	-78.93	

Comments:

- 0 No annual averages
- 2 No annual average for sulfate
- 5 No annual average for sodium
- 7 No annual average for calcium

- 1 No annual average for hydrogen
- 4 No annual average for chloride
- 6 No annual average for potassium
- 8 No annual average for magnesium

TABLE A.2. (Continued)

Sitename	State/Province	Network	ADS ID	Latitude	Longitude	Comments
Dorset (c)	Ontario	APIOSC	007c	45.22	-78.93	
Douglas Lake	Michigan	NADP/NTN	031a	45.58	-84.88	
Ear Falls	Ontario	APIOSC	200a	50.64	-93.22	5-8
Exst	Massachusetts	NADP/NTN	277a	42.38	-71.21	
ELA (b)	Ontario	CAPMON/APN	135b	49.67	-93.72	4, 6
Everglades Nat. Pa	Florida	NADP/NTN	018a	25.39	-80.60	0
Fayetteville	Arkansas	NADP/NTN	004a	36.10	-94.17	
Fernberg	Minnesota	APIOSD	168b	47.95	-91.49	0
Fernberg	Minnesota	NADP/NTN	168a	47.95	-91.50	
Finley (A)	North Carolina	NADP/NTN	053a	35.73	-78.68	
Forest Seed Ctr	Texas	NADP/NTN	254a	31.50	-94.86	
Fort Wayne	Indiana	UAPSP	160a	41.04	-85.32	2
Gaylord	Michigan	UAPSP	241a	44.95	-84.84	2
Georgia Station	Georgia	NADP/NTN	017a	33.18	-84.41	
Glacier Nat'l Park	Montana	NADP/NTN	037a	48.51	-114.00	0
Golden Lake	Ontario	APIOSC	194a	45.61	-77.20	6
Gowganda	Ontario	APIOSC	200a	47.65	-80.78	4, 6, 8
Grand Canyon	Arizona	NADP/NTN	068a	36.07	-112.15	0
Great Smoky Mts	Tennessee	NADP/NTN	028a	35.60	-83.59	0
Greenville Station	Maine	NADP/NTN	030a	45.49	-69.68	
H.J. Andrews	Oregon	NADP/NTN	001a	44.21	-122.26	
Hopland (Ukiah)	California	NADP/NTN	007a	39.00	-123.08	0
Horton's Station	Virginia	NADP/NTN	073a	37.33	-80.50	0
Hubbard Brook	New Hampshire	NADP/NTN	039a	43.94	-71.70	
Huntington	New York	NADP/NTN	168a	43.97	-74.22	
Huron Park	Ontario	APIOSC	183a	43.29	-81.50	
Illinois	Illinois	MAP3S	020b	40.05	-88.37	1
Indiana Dunes	Indiana	NADP/NTN	025a	41.63	-87.00	
Ithaca	New York	MAP3S	044a	42.40	-76.65	1
Jasper	New York	NADP/NTN	047a	42.11	-77.54	
Kane	Pennsylvania	NADP/NTN	003a	41.60	-78.77	
Kejimikujik (b)	Ontario	CAPMON/APN	103b	44.43	-85.20	5
Kellogg	Michigan	NADP/NTN	032a	42.41	-85.39	
Killarney	Ontario	APIOSC	190a	45.99	-81.49	
Lac Le Croix (a)	Ontario	APIOSC	200a	48.35	-92.21	0
Lamberton	Minnesota	NADP/NTN	035a	44.24	-95.30	
Lancaster	Kansas	UAPSP	238a	39.57	-95.30	2
Leading Ridge	Pennsylvania	NADP/NTN	004a	40.60	-77.94	
Leves	Delaware	MAP3S	013a	38.77	-75.00	1

Comments:

0 No annual averages

2 No annual average for sulfate

5 No annual average for sodium

7 No annual average for calcium

1 No annual average for hydrogen

4 No annual average for chloride

6 No annual average for potassium

8 No annual average for magnesium

TABLE A.2. (Continued)

Sitename	State/Province	Network	ADS ID	Latitude	Longitude	Comments
Lewiston	North Carolina	NADP/NTN	049a	36.13	-77.17	
Longwoods (b)	Ontario	APIOSD	143b	42.88	-81.48	
Manitou	Colorado	NADP/NTN	011a	39.10	-105.09	0
Marcell	Minnesota	NADP/NTN	034a	47.53	-93.47	
Marshall	Texas	UAPSP	247a	32.87	-94.42	2
Mattawa	Ontario	APIOSC	197a	48.28	-78.82	
McKellar	Ontario	APIOSC	195a	45.52	-79.92	
Mead	Nebraska	NADP/NTN	038a	41.16	-96.49	
Melbourne	Ontario	APIOSD	221a	42.79	-81.58	
Meridian	Mississippi	NADP/NTN	036a	32.33	-88.74	
Merlin	Ontario	APIOSC	177a	42.25	-82.22	
Mesa Verde	Colorado	NADP/NTN	029a	37.20	-108.49	
Montmorency	Quebec	CAPMON/APN	144a	47.32	-71.15	4,5,7
Moonbeam	Ontario	APIOSC	201a	49.32	-82.15	
Newcastle	Wyoming	NADP/NTN	255a	43.87	-104.19	
Nithgrove	Ontario	APIOSD	224a	45.20	-79.07	1,5-8
North Easthope	Ontario	APIOSD	222a	43.41	-80.89	
Oak Ridge	Tennessee	MAP3S	171b	35.96	-84.29	1
Oliver Knoll	Arizona	NADP/NTN	054a	33.07	-109.86	
Organ Pipe Mon.	Arizona	NADP/NTN	003a	31.95	-112.80	0
Oxford	Ohio	MAP3S	057a	39.53	-84.72	1
Palmerston	Ontario	APIOSC	182a	43.81	-80.90	1
Parsons	West Virginia	NADP/NTN	075a	39.89	-79.66	
Pawnee	Colorado	NADP/NTN	012a	40.81	-104.75	
Penn State	Pennsylvania	MAP3S	005a	40.79	-77.95	1
Pickle Lake	Ontario	APIOSC	207a	51.48	-90.20	0
Piedmont Station	North Carolina	NADP/NTN	051a	35.70	-80.82	
Pinedale	Wyoming	NADP/NTN	204a	42.93	-109.79	
Port Stanley	Ontario	APIOSC	178a	42.87	-81.17	
Quabbin Reservoir	Massachusetts	NADP/NTN	276a	42.39	-72.34	0
Quetico Centre	Ontario	APIOSD	233a	48.41	-91.20	5-8
Railton	Ontario	APIOSD	228a	44.38	-76.59	0
Raleigh	North Carolina	UAPSP	053c	35.73	-76.08	2
Raven Lake	Ontario	APIOSD	226a	44.81	-78.91	5-8
Rockport	Indiana	UAPSP	154a	37.08	-87.13	2
Salem	Illinois	NADP/NTN	181a	38.64	-88.97	
Sand Spring	Colorado	NADP/NTN	173a	40.51	-107.70	
Scranton	Pennsylvania	UAPSP	151a	41.58	-75.99	2
Selma	Alabama	UAPSP	235a	32.47	-87.08	2

Comments:

0 No annual averages

2 No annual average for sulfate

5 No annual average for sodium

7 No annual average for calcium

1 No annual average for hydrogen

4 No annual average for chloride

6 No annual average for potassium

8 No annual average for magnesium

TABLE A.2. (Continued)

Sitename	State/Province	Network	ADS ID	Latitude	Longitude	Comments
Sequoia Nat. Park	California	NADP/NTN	008a	36.57	-118.78	0
Shabbona	Illinois	NADP/NTN	024a	41.84	-88.85	
Shallow Lake	Ontario	APIOSC	181a	44.58	-81.89	
Shenandoah Nat'l	Virginia	NADP/NTN	258a	38.52	-78.44	0
Smith's Falls	Ontario	APIOSC	192a	44.94	-75.98	8
Southern Ill U	Illinois	NADP/NTN	022a	37.71	-89.27	
Spooner	Wisconsin	NADP/NTN	077a	45.82	-91.87	
Tanbark Flat	California	NADP/NTN	270a	34.21	-117.78	0
Teddy Roosevelt NP	North Dakota	NADP/NTN	082a	47.88	-103.28	0
Trout Lake	Wisconsin	NADP/NTN	076a	48.05	-89.85	
Turners Falls	Massachusetts	UAPSP	158a	42.88	-72.55	2
Underhill Center	Vermont	UAPSP	248a	44.53	-72.87	2
University Forest	Missouri	NADP/NTN	253a	36.91	-90.32	
Uvalde	Georgia	UAPSP	237a	32.06	-82.47	2
Uxbridge	Ontario	APIOSC	187a	44.21	-79.21	0
Victoria	Texas	NADP/NTN	071a	28.85	-98.92	
Virginia	Virginia	MAPSS	072a	38.84	-78.54	1
Walker Branch	Tennessee	NADP/NTN	171a	35.98	-84.29	
Washington Xing	New Jersey	NADP/NTN	285a	40.31	-74.85	
Waterloo	Ontario	APIOSC	184a	43.48	-80.59	
Wellesley	Ontario	APIOSC	223a	43.47	-80.78	8
Wellston	Michigan	NADP/NTN	033a	44.22	-85.82	0
Whiteface	New York	MAPSS	043a	44.39	-73.88	1
Whitney	Ontario	APIOSC	203a	45.54	-78.26	0
Wilberforce	Ontario	APIOSC	188a	45.81	-78.22	
Wilkesport	Ontario	APIOSC	179a	42.78	-82.35	
Winterport	Maine	UAPSP	240a	44.82	-68.97	2
Wooster	Ohio	NADP/NTN	058a	40.78	-81.92	
Yellowstone	Wyoming	NADP/NTN	078a	44.92	-110.42	0
Yosemite	California	NADP/NTN	157a	37.86	-119.86	0
Zanesville	Ohio	UAPSP	153a	39.98	-82.82	2

Comments:

0 No annual averages
 2 No annual average for sulfate
 5 No annual average for sodium
 7 No annual average for calcium

1 No annual average for hydrogen
 4 No annual average for chloride
 6 No annual average for potassium
 8 No annual average for magnesium

TABLE A.3. 1987 Frequency Distribution Percentiles for Annual and Seasonal Ion Species Concentration

		Number of									
		Sites	Min	5	10	25	50	75	90	95	Max
pH	Annual	173	4.076	4.198	4.221	4.291	4.486	4.802	5.209	5.503	6.128
	Winter	217	4.124	4.304	4.338	4.425	4.618	4.845	5.250	5.484	6.085
	Spring	233	4.083	4.185	4.221	4.348	4.557	4.945	5.310	5.583	6.263
	Summer	240	3.901	4.049	4.093	4.233	4.508	4.985	5.318	5.569	6.045
	Fall	245	4.045	4.220	4.261	4.391	4.658	4.974	5.337	5.517	6.392
Sulfate (mg/l)	Annual	154	0.208	0.449	0.708	1.136	1.849	2.773	3.244	3.621	4.774
	Winter	210	0.133	0.273	0.394	0.785	1.168	1.533	2.029	2.532	2.926
	Spring	230	0.279	0.489	0.678	1.220	1.938	2.951	3.757	4.227	6.000
	Summer	217	0.274	0.601	0.709	0.948	1.856	3.458	4.392	4.735	6.288
	Fall	224	0.139	0.349	0.539	0.881	1.404	2.278	2.839	3.319	5.392
Nitrate (mg/l)	Annual	173	0.040	0.378	0.620	0.870	1.318	1.839	2.509	2.878	3.274
	Winter	210	0.060	0.304	0.410	0.548	1.101	1.944	2.665	3.063	4.508
	Spring	231	0.081	0.422	0.558	0.899	1.378	2.122	2.890	3.358	4.930
	Summer	239	0.132	0.543	0.665	0.948	1.310	1.842	2.428	2.738	3.450
	Fall	241	0.016	0.351	0.453	0.649	1.034	1.642	2.257	2.499	3.258
Ammonium (mg/l)	Annual	172	0.013	0.061	0.089	0.162	0.278	0.427	0.550	0.640	0.885
	Winter	212	0.011	0.026	0.035	0.082	0.150	0.266	0.402	0.477	0.975
	Spring	231	0.026	0.087	0.117	0.192	0.384	0.582	0.754	0.871	1.631
	Summer	238	0.011	0.077	0.119	0.170	0.310	0.444	0.624	0.742	1.305
	Fall	243	0.010	0.032	0.063	0.117	0.223	0.348	0.515	0.621	1.416
Calcium (mg/l)	Annual	170	0.016	0.048	0.051	0.081	0.159	0.236	0.315	0.450	1.159
	Winter	200	0.003	0.026	0.034	0.053	0.079	0.149	0.276	0.466	0.934
	Spring	229	0.032	0.053	0.068	0.107	0.200	0.322	0.493	0.591	1.512
	Summer	240	0.023	0.039	0.057	0.105	0.171	0.279	0.374	0.522	0.935
	Fall	240	0.009	0.020	0.036	0.056	0.129	0.223	0.317	0.371	2.184

TABLE A.4. 1987 Frequency Distribution Percentiles for Annual and Seasonal Ion Species Deposition

		Number of									
		Sites	Min	5	10	25	50	75	90	95	Max
Hydrogen											
(kg/ha)	Annual	173	0.003	0.018	0.037	0.123	0.326	0.434	0.515	0.646	0.945
	Winter	217	0.000	0.003	0.005	0.016	0.049	0.073	0.093	0.105	0.179
	Spring	233	0.000	0.003	0.005	0.017	0.051	0.088	0.124	0.143	0.223
	Summer	240	0.001	0.003	0.006	0.020	0.081	0.158	0.209	0.250	0.417
	Fall	245	0.000	0.001	0.003	0.013	0.055	0.099	0.139	0.154	0.254
Sulfate											
(kg/ha)	Annual	154	0.822	2.282	3.179	9.235	18.598	24.625	29.270	32.700	44.025
	Winter	210	0.061	0.162	0.303	0.901	2.026	3.426	4.802	5.139	8.044
	Spring	230	0.203	0.573	0.798	1.805	3.057	5.510	8.031	7.784	10.300
	Summer	217	0.202	0.434	0.798	1.793	4.674	9.149	12.005	13.905	21.329
	Fall	224	0.028	0.200	0.393	1.009	3.279	5.139	7.075	8.340	11.334
Nitrate											
(kg/ha)	Annual	173	0.291	2.210	3.171	7.185	11.179	16.014	21.050	23.153	29.259
	Winter	210	0.024	0.273	0.440	0.930	1.912	2.040	4.155	4.992	7.124
	Spring	231	0.057	0.621	0.793	1.475	2.573	3.687	4.770	5.814	7.081
	Summer	239	0.000	0.581	0.883	1.760	3.340	5.061	6.897	7.210	10.411
	Fall	241	0.021	0.204	0.433	0.897	1.972	3.613	5.547	6.292	10.695
Ammonium											
(kg/ha)	Annual	172	0.092	0.399	0.597	1.414	2.539	3.525	4.327	5.135	8.757
	Winter	212	0.005	0.032	0.059	0.129	0.268	0.413	0.622	0.822	1.501
	Spring	231	0.022	0.139	0.172	0.312	0.644	0.979	1.270	1.535	2.408
	Summer	238	0.007	0.103	0.152	0.317	0.785	1.217	1.659	1.940	5.905
	Fall	243	0.001	0.031	0.070	0.198	0.409	0.717	1.110	1.337	3.810
Calcium											
(kg/ha)	Annual	170	0.075	0.302	0.421	0.755	1.107	1.798	2.840	3.331	8.734
	Winter	209	0.000	0.028	0.049	0.079	0.130	0.205	0.453	0.500	1.290
	Spring	229	0.023	0.107	0.133	0.189	0.325	0.501	0.792	0.902	2.022
	Summer	240	0.013	0.060	0.110	0.213	0.401	0.603	1.012	1.243	2.900
	Fall	240	0.000	0.034	0.055	0.110	0.208	0.365	0.629	0.740	4.250

TABLE A.5. Summary of 1979-1987 Trend Sites Annual
Precipitation-weighted Concentration by Year

	Year	N	Mean	Std Dev	Min	5th	10th	Percentile						Max
								25th	50th	75th	90th	95th		
pH	1979	23	4.53	0.33	4.12	4.12	4.19	4.26	4.42	4.66	5.14	5.28	5.38	
	1980	23	4.42	0.33	4.07	4.07	4.09	4.16	4.28	4.51	5.04	5.14	5.16	
	1981	23	4.48	0.37	4.09	4.09	4.10	4.22	4.34	4.54	5.12	5.34	5.39	
	1982	23	4.52	0.35	4.17	4.17	4.20	4.27	4.38	4.67	5.16	5.41	5.46	
	1983	23	4.54	0.30	4.23	4.23	4.25	4.31	4.43	4.76	5.04	5.28	5.31	
	1984	23	4.53	0.32	4.15	4.15	4.18	4.28	4.43	4.85	5.09	5.14	5.15	
	1985	23	4.53	0.36	4.16	4.16	4.19	4.27	4.37	4.60	5.19	5.32	5.35	
	1986	23	4.52	0.39	4.12	4.13	4.15	4.26	4.36	4.56	5.31	5.43	5.43	
	1987	23	4.50	0.33	4.15	4.15	4.17	4.22	4.43	4.67	5.04	5.20	5.24	
Hydrogen (µg/l)	1979	23	38.76	20.68	4.98	5.21	7.43	21.88	38.18	54.84	64.85	75.34	76.64	
	1980	23	47.55	24.87	6.97	7.29	9.06	31.23	53.06	88.52	88.57	84.89	85.59	
	1981	23	42.64	23.82	4.10	4.66	7.55	29.14	46.00	59.75	78.65	81.58	81.76	
	1982	23	37.66	19.40	3.48	3.93	7.11	21.29	42.16	53.33	62.89	67.78	68.32	
	1983	23	34.39	17.14	4.91	5.71	9.11	17.56	36.77	49.49	58.45	58.98	59.42	
	1984	23	36.32	19.68	7.08	7.25	8.13	22.20	37.47	52.43	65.87	70.15	70.38	
	1985	23	37.92	20.62	4.50	4.82	8.53	25.05	42.22	54.20	65.16	68.89	69.78	
	1986	23	39.58	21.05	3.74	3.76	5.21	27.48	44.30	54.66	70.73	74.80	75.66	
	1987	23	39.09	21.24	5.81	6.41	9.08	21.58	37.11	60.90	67.58	71.11	71.27	
Sulfate (mg/l)	1979	26	2.48	0.76	1.44	1.48	1.64	1.96	2.23	3.28	3.66	4.06	4.24	
	1980	26	2.65	0.85	0.99	1.02	1.25	2.18	2.62	3.35	3.79	4.09	4.15	
	1981	26	2.57	0.83	1.12	1.19	1.52	1.81	2.65	3.29	3.54	4.14	4.46	
	1982	26	2.27	0.75	0.95	1.06	1.27	1.89	2.16	2.93	3.34	3.52	3.56	
	1983	26	2.06	0.82	1.02	1.03	1.17	1.62	2.05	2.61	2.91	3.04	3.05	
	1984	26	2.12	0.71	1.00	1.10	1.30	1.58	1.99	2.63	3.38	3.55	3.59	
	1985	26	2.09	0.75	0.72	0.78	0.96	1.50	2.04	2.59	3.20	3.28	3.31	
	1986	26	2.25	0.83	0.71	0.74	1.05	1.67	2.16	2.74	3.57	3.85	3.96	
	1987	26	2.18	0.86	0.80	0.81	1.02	1.56	1.90	3.01	3.59	3.63	3.64	
Nitrate (mg/l)	1979	29	1.32	0.41	0.62	0.84	0.72	1.01	1.40	1.59	1.77	2.06	2.10	
	1980	29	1.63	0.41	0.84	0.86	0.92	1.28	1.77	1.93	2.05	2.19	2.22	
	1981	29	1.46	0.39	0.85	0.85	0.88	1.10	1.50	1.79	1.95	2.07	2.13	
	1982	29	1.32	0.41	0.59	0.65	0.80	0.98	1.26	1.71	1.89	2.03	2.15	
	1983	29	1.26	0.33	0.63	0.64	0.78	1.04	1.18	1.59	1.71	1.73	1.73	
	1984	29	1.37	0.42	0.76	0.78	0.80	1.07	1.30	1.75	1.99	2.14	2.15	
	1985	29	1.35	0.43	0.68	0.71	0.77	0.92	1.38	1.64	2.03	2.16	2.21	
	1986	29	1.42	0.41	0.61	0.72	0.94	1.07	1.38	1.61	2.00	2.10	2.14	
	1987	29	1.48	0.45	0.64	0.70	0.87	1.04	1.53	1.68	2.07	2.11	2.12	
Ammonium (mg/l)	1979	30	0.28	0.15	0.08	0.09	0.12	0.19	0.25	0.36	0.46	0.70	0.73	
	1980	30	0.33	0.16	0.11	0.12	0.15	0.25	0.30	0.39	0.48	0.81	0.85	
	1981	30	0.32	0.14	0.17	0.17	0.17	0.21	0.30	0.36	0.49	0.66	0.79	
	1982	30	0.28	0.11	0.11	0.13	0.16	0.19	0.28	0.33	0.41	0.53	0.53	
	1983	30	0.29	0.15	0.11	0.14	0.17	0.20	0.27	0.31	0.39	0.76	0.87	
	1984	30	0.29	0.12	0.13	0.14	0.17	0.19	0.27	0.36	0.49	0.56	0.81	
	1985	30	0.27	0.10	0.10	0.12	0.14	0.20	0.25	0.33	0.41	0.53	0.54	
	1986	30	0.30	0.12	0.09	0.11	0.14	0.20	0.29	0.38	0.48	0.55	0.60	
	1987	30	0.32	0.14	0.12	0.12	0.13	0.21	0.28	0.39	0.54	0.82	0.64	

TABLE A.5. (Continued)

	Year	N	Mean	Std Dev	Min	5th	10th	Percentile					Max
								25th	50th	75th	90th	95th	
Calcium (mg/l)	1979	30	0.26	0.14	0.05	0.05	0.07	0.09	0.17	0.28	0.41	0.52	0.59
	1980	30	0.23	0.14	0.07	0.08	0.10	0.12	0.21	0.30	0.45	0.50	0.67
	1981	30	0.21	0.11	0.05	0.07	0.10	0.12	0.19	0.25	0.30	0.51	0.66
	1982	30	0.17	0.09	0.06	0.06	0.07	0.10	0.16	0.23	0.30	0.33	0.37
	1983	30	0.17	0.10	0.07	0.07	0.07	0.10	0.16	0.22	0.27	0.43	0.45
	1984	30	0.18	0.09	0.06	0.06	0.07	0.10	0.16	0.25	0.30	0.36	0.37
	1985	30	0.14	0.08	0.04	0.04	0.06	0.08	0.13	0.18	0.21	0.33	0.38
	1986	30	0.14	0.07	0.04	0.04	0.05	0.07	0.14	0.20	0.23	0.27	0.28
1987	30	0.15	0.08	0.04	0.04	0.05	0.07	0.14	0.21	0.25	0.32	0.36	
Magnesium (mg/l)	1979	30	0.04	0.02	0.02	0.02	0.02	0.03	0.03	0.06	0.07	0.10	0.12
	1980	30	0.05	0.03	0.01	0.02	0.02	0.03	0.04	0.05	0.08	0.13	0.16
	1981	30	0.05	0.02	0.02	0.02	0.02	0.04	0.04	0.06	0.08	0.11	0.13
	1982	30	0.04	0.03	0.01	0.01	0.02	0.03	0.03	0.05	0.08	0.12	0.15
	1983	30	0.04	0.03	0.01	0.01	0.01	0.02	0.03	0.04	0.08	0.13	0.18
	1984	30	0.04	0.03	0.01	0.01	0.02	0.03	0.04	0.05	0.07	0.11	0.15
	1985	30	0.04	0.02	0.01	0.02	0.02	0.02	0.03	0.04	0.05	0.11	0.15
	1986	30	0.03	0.03	0.01	0.01	0.02	0.02	0.03	0.03	0.04	0.12	0.16
1987	30	0.04	0.04	0.01	0.01	0.02	0.02	0.03	0.04	0.06	0.15	0.22	
Sodium (mg/l)	1979	28	0.34	0.31	0.06	0.06	0.08	0.14	0.24	0.43	0.68	1.26	1.39
	1980	28	0.24	0.20	0.05	0.05	0.08	0.09	0.14	0.26	0.57	1.15	1.42
	1981	28	0.16	0.16	0.03	0.04	0.06	0.08	0.11	0.17	0.31	0.70	0.76
	1982	28	0.20	0.30	0.04	0.04	0.05	0.07	0.09	0.15	0.59	1.18	1.47
	1983	28	0.19	0.33	0.03	0.04	0.05	0.06	0.08	0.16	0.49	1.20	1.67
	1984	28	0.16	0.24	0.04	0.04	0.05	0.05	0.08	0.14	0.32	0.96	1.25
	1985	28	0.13	0.18	0.03	0.04	0.05	0.06	0.08	0.11	0.31	0.73	0.88
	1986	28	0.13	0.21	0.03	0.03	0.04	0.05	0.08	0.12	0.23	0.83	1.01
1987	28	0.16	0.32	0.03	0.03	0.03	0.05	0.07	0.11	0.33	1.18	1.85	
Chloride (mg/l)	1979	28	0.34	0.34	0.11	0.11	0.12	0.16	0.22	0.32	0.78	1.40	1.60
	1980	28	0.39	0.52	0.13	0.14	0.16	0.17	0.22	0.33	0.73	2.14	2.70
	1981	28	0.29	0.29	0.08	0.09	0.12	0.15	0.19	0.31	0.57	1.28	1.39
	1982	28	0.35	0.57	0.06	0.08	0.09	0.14	0.20	0.29	0.57	2.27	2.98
	1983	28	0.35	0.53	0.11	0.11	0.11	0.13	0.15	0.31	0.85	2.05	2.85
	1984	28	0.33	0.46	0.10	0.11	0.11	0.15	0.19	0.29	0.57	1.85	2.44
	1985	28	0.28	0.37	0.07	0.08	0.10	0.13	0.18	0.27	0.66	1.49	1.94
	1986	28	0.29	0.41	0.07	0.07	0.09	0.11	0.17	0.30	0.44	1.87	2.68
1987	28	0.33	0.56	0.07	0.09	0.10	0.14	0.18	0.28	0.65	1.97	2.82	
Potassium (mg/l)	1979	26	0.06	0.04	0.01	0.02	0.02	0.03	0.04	0.07	0.11	0.15	0.16
	1980	28	0.06	0.05	0.02	0.02	0.02	0.03	0.05	0.10	0.14	0.17	0.18
	1981	26	0.05	0.03	0.02	0.02	0.02	0.03	0.04	0.06	0.09	0.12	0.13
	1982	28	0.04	0.02	0.02	0.02	0.02	0.02	0.03	0.04	0.06	0.10	0.12
	1983	26	0.04	0.02	0.02	0.02	0.02	0.03	0.03	0.05	0.07	0.08	0.08
	1984	26	0.04	0.02	0.01	0.02	0.02	0.02	0.03	0.05	0.07	0.10	0.11
	1985	26	0.03	0.01	0.01	0.01	0.02	0.02	0.03	0.04	0.05	0.06	0.07
	1986	26	0.03	0.02	0.01	0.01	0.01	0.02	0.03	0.05	0.06	0.08	0.09
1987	28	0.05	0.05	0.01	0.01	0.01	0.02	0.03	0.09	0.13	0.16	0.17	

TABLE A.6. Summary of 1979-1987 Trend Sites Annual Deposition by Year

	Year	N	Mean	Std Dev	Min	5th	10th	25th	Percentile 50th	75th	90th	95th	Max
Precipitation (cm)													
1979	30	108.88	37.24	28.17	42.29	61.48	80.49	117.83	138.73	141.24	184.87	228.51	
1980	30	98.38	27.17	30.67	39.79	50.21	70.49	91.90	100.24	126.90	151.21	171.40	
1981	30	95.80	23.75	41.75	48.67	63.45	79.34	93.79	118.78	128.78	144.29	151.55	
1982	30	101.65	27.70	37.18	54.66	77.84	84.50	97.15	113.06	150.22	180.15	142.57	
1983	30	100.57	32.12	42.74	51.85	63.82	87.65	106.86	126.84	146.42	172.61	197.49	
1984	30	106.55	28.35	46.95	54.83	75.39	87.40	104.63	127.22	146.32	181.68	179.90	
1985	30	98.38	20.36	43.82	52.61	79.20	87.19	98.18	106.09	128.32	142.84	149.83	
1986	30	94.48	20.82	39.18	52.74	68.85	81.49	92.57	107.24	126.31	133.26	134.61	
1987	30	88.53	24.70	32.37	41.38	52.13	75.58	89.39	104.38	116.28	139.06	145.87	
Hydrogen (kg/ha)													
1979	23	0.42	0.28	0.02	0.02	0.05	0.28	0.41	0.54	0.84	1.01	1.05	
1980	23	0.47	0.29	0.03	0.04	0.04	0.31	0.50	0.67	0.87	1.06	1.11	
1981	23	0.43	0.27	0.02	0.03	0.05	0.23	0.47	0.62	0.82	0.89	0.90	
1982	23	0.40	0.20	0.03	0.03	0.04	0.34	0.43	0.50	0.62	0.72	0.74	
1983	23	0.37	0.19	0.03	0.03	0.05	0.18	0.43	0.48	0.61	0.82	0.83	
1984	23	0.39	0.21	0.03	0.04	0.06	0.31	0.40	0.55	0.66	0.69	0.69	
1985	23	0.39	0.24	0.04	0.04	0.04	0.24	0.43	0.57	0.66	0.92	0.97	
1986	23	0.39	0.25	0.03	0.03	0.04	0.23	0.38	0.52	0.69	0.96	1.02	
1987	23	0.34	0.10	0.03	0.03	0.05	0.23	0.40	0.46	0.55	0.59	0.60	
Sulfate (kg/ha)													
1979	26	20.10	10.71	6.42	7.41	11.99	17.64	26.02	34.11	41.54	47.75	49.71	
1980	26	24.16	10.81	3.20	4.04	7.70	20.21	23.44	29.91	42.17	44.95	45.64	
1981	26	24.83	10.49	6.60	6.79	9.28	10.69	25.63	30.88	41.43	45.08	45.61	
1982	26	22.83	8.45	3.53	5.95	10.63	18.82	23.77	28.99	33.58	38.25	40.08	
1983	26	22.33	8.14	4.38	5.82	8.50	18.64	23.01	30.22	31.07	33.17	34.21	
1984	26	22.49	8.57	4.64	5.78	9.31	18.25	22.32	27.94	34.57	38.93	41.17	
1985	26	21.19	9.64	3.17	4.46	8.44	15.88	21.80	26.55	34.19	43.02	47.64	
1986	26	21.70	9.92	2.80	3.58	7.72	17.10	22.07	26.26	31.61	46.32	53.32	
1987	26	19.27	7.38	2.70	3.12	7.24	14.40	20.00	24.46	28.09	29.61	30.38	
Nitrate (kg/ha)													
1979	29	14.02	4.75	2.51	5.57	9.29	10.39	13.73	17.09	21.27	22.68	23.02	
1980	29	14.77	5.07	2.73	5.52	8.89	11.17	14.40	17.90	23.79	25.20	26.09	
1981	29	14.05	5.05	4.10	5.49	7.51	10.07	13.88	17.58	20.44	24.15	24.42	
1982	29	13.14	3.78	2.99	5.40	8.60	10.70	13.89	15.93	18.34	19.35	19.65	
1983	29	13.31	3.22	3.89	5.80	8.21	11.83	13.80	15.29	17.13	17.93	18.34	
1984	29	14.43	4.37	4.24	6.02	8.77	11.22	14.14	17.12	19.25	23.54	25.57	
1985	29	13.23	4.55	2.99	5.35	7.72	8.99	14.62	15.92	19.02	21.82	24.34	
1986	29	13.72	4.89	2.40	4.91	7.85	10.29	14.22	16.93	19.37	23.74	27.09	
1987	29	12.87	3.89	2.82	4.84	7.99	10.80	12.88	15.33	17.75	18.58	19.09	
Ammonium (kg/ha)													
1979	30	2.81	1.09	0.41	0.74	1.25	2.07	2.91	3.50	4.18	4.82	5.30	
1980	30	2.81	1.04	0.40	0.96	1.51	2.12	2.60	3.72	4.34	4.54	4.87	
1981	30	2.95	1.04	0.73	1.07	1.43	2.22	2.77	3.88	4.25	4.60	4.97	
1982	30	2.75	0.95	0.60	1.07	1.60	2.16	2.61	3.29	4.23	4.75	5.09	
1983	30	2.89	0.88	0.60	1.27	1.70	2.45	2.88	3.28	3.97	4.92	5.52	
1984	30	2.91	0.90	0.79	1.31	1.77	2.17	2.83	3.68	4.50	4.57	4.58	
1985	30	2.54	0.80	0.44	1.09	1.64	2.01	2.40	3.20	3.44	3.92	4.23	
1986	30	2.82	1.23	0.34	1.00	1.66	1.94	2.65	3.60	5.15	5.50	5.67	
1987	30	2.65	0.86	0.30	0.93	1.69	2.07	2.66	3.23	3.68	4.42	4.83	

TABLE A.6. (Continued)

	Year	N	Mean	Std Dev	Min	5th	10th	Percentile					Max
								25th	50th	75th	90th	95th	
Calcium (kg/ha)	1979	30	1.83	0.85	0.63	0.67	0.73	1.16	1.84	2.63	3.81	3.48	3.56
	1980	30	1.98	1.01	0.79	0.81	0.88	1.11	1.83	2.78	3.88	4.13	4.39
	1981	30	1.82	0.74	0.41	0.65	1.02	1.34	1.88	2.42	2.88	3.32	3.73
	1982	30	1.67	0.82	0.54	0.58	0.77	1.07	1.54	2.15	2.81	3.64	4.04
	1983	30	1.61	0.68	0.88	0.88	0.91	1.15	1.59	1.91	2.69	2.88	2.87
	1984	30	1.72	0.88	0.66	0.65	0.78	1.25	1.78	2.15	2.84	3.88	3.29
	1985	30	1.29	0.61	0.39	0.48	0.78	0.82	1.28	1.68	2.32	2.54	2.65
	1986	30	1.29	0.71	0.34	0.35	0.52	0.78	1.01	1.74	2.45	2.89	3.84
	1987	30	1.15	0.58	0.44	0.58	0.61	0.77	0.99	1.59	1.95	2.19	2.25
Magnesium (kg/ha)	1979	30	0.45	0.38	0.16	0.17	0.19	0.28	0.48	0.51	0.78	1.23	1.76
	1980	30	0.39	0.28	0.11	0.11	0.18	0.24	0.35	0.49	0.55	1.15	1.47
	1981	30	0.43	0.17	0.13	0.16	0.18	0.31	0.42	0.54	0.78	0.75	0.75
	1982	30	0.42	0.29	0.08	0.12	0.18	0.23	0.38	0.49	0.89	1.22	1.58
	1983	30	0.45	0.46	0.13	0.13	0.15	0.26	0.31	0.48	0.76	1.87	2.57
	1984	30	0.42	0.28	0.13	0.13	0.19	0.28	0.37	0.58	0.67	1.23	1.53
	1985	30	0.34	0.27	0.12	0.14	0.18	0.22	0.28	0.39	0.52	1.08	1.81
	1986	30	0.31	0.25	0.14	0.14	0.15	0.18	0.28	0.31	0.41	1.12	1.38
	1987	30	0.33	0.37	0.11	0.13	0.15	0.18	0.23	0.28	0.48	1.47	2.12
Sodium (kg/ha)	1979	28	3.51	3.01	0.47	0.59	0.76	1.29	2.87	4.79	7.14	11.89	15.19
	1980	28	2.18	2.88	0.38	0.39	0.49	0.88	1.27	2.93	4.98	10.15	12.99
	1981	28	1.49	1.51	0.38	0.39	0.59	0.73	1.04	1.54	2.87	6.51	8.76
	1982	28	2.11	3.18	0.29	0.32	0.38	0.88	0.85	2.35	6.82	11.95	14.48
	1983	28	2.46	4.84	0.35	0.38	0.44	0.53	0.73	2.82	5.53	18.49	24.22
	1984	28	1.81	2.68	0.42	0.43	0.45	0.55	0.72	1.95	4.29	18.65	12.88
	1985	28	1.31	1.88	0.27	0.27	0.35	0.52	0.68	1.32	3.44	7.41	9.45
	1986	28	1.28	1.87	0.27	0.29	0.34	0.42	0.58	1.15	2.28	7.88	8.89
	1987	28	1.54	3.85	0.24	0.25	0.28	0.35	0.52	1.27	3.59	11.48	15.72
Chloride (kg/ha)	1979	28	4.81	4.89	0.69	0.74	1.08	1.31	2.89	4.45	8.93	17.94	23.78
	1980	28	3.53	4.71	0.51	0.71	1.08	1.41	2.19	3.45	6.82	18.91	24.74
	1981	28	2.78	2.75	0.58	0.61	0.98	1.26	2.11	2.92	5.26	11.95	12.41
	1982	28	3.75	5.73	0.38	0.43	0.65	1.42	1.92	3.77	6.57	22.87	29.28
	1983	28	4.48	7.75	0.47	0.62	0.84	1.28	1.77	4.84	18.22	38.28	38.45
	1984	28	3.72	5.13	0.58	0.67	0.97	1.44	1.99	3.58	7.88	28.89	24.95
	1985	28	2.89	3.98	0.44	0.52	0.62	1.25	1.87	2.92	6.27	15.41	21.29
	1986	28	2.75	3.87	0.37	0.52	0.75	1.09	1.81	2.78	4.18	15.35	18.27
	1987	28	3.14	4.88	0.39	0.45	0.57	0.97	1.72	2.88	6.82	19.13	24.88
Potassium (kg/ha)	1979	26	0.55	0.29	0.11	0.12	0.18	0.28	0.52	0.77	1.02	1.11	1.18
	1980	26	0.54	0.41	0.08	0.09	0.18	0.28	0.44	0.68	1.24	1.68	1.75
	1981	26	0.42	0.25	0.14	0.16	0.19	0.28	0.35	0.58	0.89	1.18	1.18
	1982	26	0.38	0.21	0.08	0.11	0.17	0.28	0.35	0.48	0.73	0.89	0.97
	1983	26	0.48	0.28	0.13	0.16	0.21	0.28	0.35	0.48	0.73	0.91	0.99
	1984	26	0.48	0.22	0.15	0.18	0.19	0.28	0.34	0.58	0.67	1.08	1.23
	1985	26	0.32	0.15	0.18	0.12	0.15	0.19	0.28	0.44	0.52	0.68	0.74
	1986	26	0.33	0.19	0.08	0.18	0.12	0.15	0.27	0.45	0.63	0.74	0.77
	1987	26	0.48	0.48	0.18	0.18	0.12	0.18	0.23	0.81	1.23	1.48	1.82

TABLE A.7. Summary of 1982-1987 Trend Sites Annual Precipitation-weighted Concentration by Year

	Year	N	Mean	Std Dev	Min	5th	10th	25th	Percentile					Max
									50th	75th	90th	95th		
pH	1982	108	4.49	0.36	4.03	4.13	4.17	4.21	4.35	4.67	5.05	5.29	5.68	
	1983	108	4.57	0.31	4.20	4.23	4.25	4.33	4.47	4.71	5.02	5.29	5.57	
	1984	108	4.52	0.33	4.07	4.16	4.20	4.27	4.38	4.68	5.03	5.17	5.51	
	1985	108	4.52	0.33	4.16	4.19	4.21	4.29	4.40	4.65	5.15	5.27	5.37	
	1986	108	4.53	0.34	4.11	4.16	4.22	4.28	4.43	4.62	5.06	5.38	5.43	
	1987	108	4.51	0.33	4.08	4.18	4.20	4.26	4.36	4.70	5.06	5.23	5.45	
Hydrogen (µg/l)	1982	108	41.61	22.50	2.19	5.16	9.01	21.59	44.37	61.07	67.66	74.10	92.35	
	1983	108	32.79	18.63	2.67	5.16	9.59	19.33	33.57	47.15	56.99	59.38	63.58	
	1984	108	37.61	20.25	3.08	6.79	9.35	20.68	41.33	53.92	63.76	68.50	84.48	
	1985	108	38.95	18.76	4.22	5.39	7.15	22.64	39.64	51.14	61.98	65.26	69.70	
	1986	108	37.08	19.50	3.72	4.19	6.75	23.97	37.39	52.47	60.43	69.17	76.84	
	1987	108	38.30	20.15	3.52	5.82	8.70	20.13	43.50	54.57	63.34	68.09	83.98	
Sulfate (mg/l)	1982	102	2.43	0.98	0.29	0.96	1.23	1.62	2.47	3.10	3.63	4.43	5.00	
	1983	102	2.15	0.91	0.32	0.93	1.04	1.45	2.07	2.81	3.21	4.06	4.64	
	1984	102	2.33	0.96	0.31	1.06	1.22	1.48	2.35	3.00	3.66	4.12	4.72	
	1985	102	2.21	0.90	0.24	0.75	0.97	1.47	2.39	2.84	3.26	3.72	4.20	
	1986	102	2.23	0.91	0.16	0.79	0.97	1.50	2.25	2.97	3.50	3.85	3.96	
	1987	102	2.33	1.06	0.21	0.78	1.02	1.38	2.50	3.19	3.63	4.10	5.55	
Nitrate (mg/l)	1982	119	1.43	0.61	0.14	0.64	0.70	0.96	1.31	1.88	2.38	2.61	2.75	
	1983	119	1.32	0.56	0.13	0.53	0.63	0.92	1.25	1.70	2.14	2.34	2.78	
	1984	119	1.51	0.66	0.10	0.74	0.79	0.98	1.34	1.90	2.46	2.83	3.30	
	1985	119	1.49	0.64	0.07	0.68	0.73	0.94	1.40	2.08	2.37	2.53	2.99	
	1986	119	1.45	0.60	0.07	0.66	0.76	0.94	1.38	1.98	2.25	2.46	2.55	
	1987	119	1.62	0.74	0.13	0.71	0.76	1.00	1.54	2.13	2.70	2.95	3.61	
Ammonium (mg/l)	1982	119	0.33	0.16	0.02	0.11	0.13	0.20	0.31	0.40	0.54	0.68	0.85	
	1983	119	0.31	0.17	0.02	0.09	0.11	0.20	0.28	0.38	0.50	0.64	0.87	
	1984	119	0.33	0.16	0.03	0.13	0.16	0.20	0.31	0.42	0.60	0.64	0.79	
	1985	119	0.30	0.15	0.01	0.10	0.13	0.19	0.28	0.39	0.51	0.60	0.79	
	1986	119	0.31	0.15	0.01	0.09	0.12	0.19	0.29	0.40	0.53	0.60	0.73	
	1987	119	0.35	0.19	0.02	0.10	0.13	0.21	0.32	0.46	0.65	0.71	0.89	

TABLE A.7. (Continued)

	Year	N	Mean	Std Dev	Min	5th	10th	Percentile						Max
						25th	50th	75th	90th	95th				
Calcium (mg/l)	1982	114	0.21	0.14	0.03	0.06	0.08	0.11	0.19	0.27	0.38	0.52	0.71	
	1983	114	0.22	0.16	0.03	0.06	0.07	0.11	0.19	0.27	0.43	0.56	0.81	
	1984	114	0.23	0.15	0.03	0.07	0.08	0.13	0.20	0.29	0.41	0.53	0.89	
	1985	114	0.19	0.12	0.04	0.06	0.07	0.10	0.17	0.26	0.37	0.42	0.66	
	1986	114	0.18	0.11	0.03	0.04	0.06	0.09	0.16	0.23	0.33	0.39	0.62	
	1987	114	0.21	0.17	0.03	0.04	0.05	0.09	0.17	0.26	0.43	0.51	1.16	
Magnesium (mg/l)	1982	112	0.05	0.03	0.01	0.02	0.02	0.03	0.04	0.05	0.09	0.14	0.28	
	1983	112	0.05	0.03	0.01	0.01	0.02	0.03	0.04	0.05	0.09	0.12	0.18	
	1984	112	0.05	0.03	0.01	0.02	0.02	0.03	0.04	0.06	0.09	0.13	0.15	
	1985	112	0.04	0.02	0.01	0.02	0.02	0.03	0.04	0.05	0.07	0.09	0.15	
	1986	112	0.04	0.03	0.01	0.01	0.02	0.02	0.03	0.04	0.08	0.08	0.22	
	1987	112	0.04	0.03	0.01	0.02	0.02	0.02	0.03	0.05	0.09	0.10	0.22	
Sodium (mg/l)	1982	111	0.13	0.10	0.03	0.03	0.04	0.05	0.08	0.14	0.23	0.38	1.47	
	1983	111	0.14	0.22	0.02	0.04	0.04	0.06	0.07	0.12	0.28	0.48	1.67	
	1984	111	0.14	0.17	0.03	0.05	0.05	0.06	0.09	0.15	0.27	0.35	1.25	
	1985	111	0.11	0.13	0.02	0.03	0.04	0.05	0.07	0.10	0.20	0.33	0.88	
	1986	111	0.10	0.14	0.01	0.03	0.03	0.04	0.06	0.10	0.18	0.31	1.01	
	1987	111	0.12	0.18	0.02	0.03	0.04	0.05	0.07	0.10	0.22	0.34	1.65	
Chloride (mg/l)	1982	115	0.26	0.35	0.06	0.06	0.08	0.12	0.18	0.26	0.39	0.59	2.98	
	1983	115	0.26	0.37	0.04	0.08	0.09	0.12	0.15	0.21	0.50	1.07	2.65	
	1984	115	0.27	0.32	0.05	0.09	0.11	0.14	0.18	0.26	0.39	0.75	2.44	
	1985	115	0.23	0.27	0.05	0.07	0.09	0.12	0.16	0.22	0.40	0.74	1.94	
	1986	115	0.22	0.26	0.05	0.07	0.08	0.11	0.15	0.25	0.35	0.72	2.08	
	1987	115	0.25	0.30	0.08	0.08	0.10	0.13	0.17	0.26	0.49	0.76	2.62	
Potassium (mg/l)	1982	110	0.04	0.02	0.01	0.02	0.02	0.02	0.03	0.05	0.08	0.07	0.12	
	1983	110	0.04	0.02	0.01	0.02	0.02	0.03	0.03	0.05	0.08	0.07	0.11	
	1984	110	0.04	0.02	0.01	0.02	0.02	0.03	0.04	0.05	0.08	0.08	0.11	
	1985	110	0.03	0.02	0.01	0.01	0.02	0.02	0.03	0.04	0.05	0.06	0.11	
	1986	110	0.03	0.02	0.01	0.01	0.01	0.02	0.03	0.04	0.05	0.07	0.11	
	1987	110	0.04	0.04	0.01	0.01	0.02	0.02	0.03	0.04	0.10	0.13	0.27	

TABLE A.8. Summary of 1982-1987 Trend Sites Annual Deposition by Year

	Year	N	Mean	Std Dev	Min	5th	10th	25th	50th	75th	90th	95th	Max
Precipitation (cm)	1982	119	98.28	32.34	12.47	41.43	64.94	81.83	95.36	112.50	137.40	152.72	246.81
	1983	119	105.84	30.34	18.48	43.76	63.58	85.87	101.28	122.95	148.58	178.95	258.87
	1984	119	100.15	32.70	18.04	40.86	69.31	81.24	98.36	118.88	133.68	141.92	274.31
	1985	119	98.74	25.29	24.96	37.32	64.66	84.16	100.24	111.82	129.52	133.85	149.83
	1986	119	98.88	28.84	20.83	44.94	84.81	88.87	98.72	110.24	129.80	141.87	230.88
	1987	119	88.17	28.68	18.83	32.37	53.90	71.87	87.85	102.92	118.37	138.88	148.53
Hydrogen (kg/m)	1982	108	0.41	0.22	0.00	0.03	0.07	0.24	0.43	0.59	0.68	0.73	0.89
	1983	108	0.35	0.19	0.00	0.03	0.07	0.20	0.37	0.47	0.58	0.64	0.83
	1984	108	0.38	0.21	0.01	0.04	0.07	0.20	0.40	0.53	0.63	0.70	0.93
	1985	108	0.37	0.21	0.01	0.04	0.05	0.23	0.38	0.52	0.61	0.71	0.97
	1986	108	0.37	0.23	0.01	0.03	0.07	0.21	0.35	0.53	0.65	0.76	1.02
	1987	108	0.34	0.19	0.01	0.03	0.05	0.20	0.36	0.46	0.53	0.63	0.94
Sulfate (kg/m)	1982	102	23.17	9.97	1.75	4.13	8.28	16.38	24.87	30.15	36.18	38.95	43.81
	1983	102	21.63	9.33	2.57	4.28	7.84	14.33	22.58	28.32	32.50	37.84	40.47
	1984	102	22.89	10.07	2.00	4.87	7.70	15.72	22.99	30.45	34.98	39.81	44.28
	1985	102	21.95	10.70	1.87	3.19	5.79	14.07	23.05	29.31	35.87	40.61	47.64
	1986	102	21.28	10.70	1.89	4.18	5.54	13.90	21.71	28.00	35.34	41.58	53.32
	1987	102	19.85	9.80	1.26	2.75	5.81	12.96	21.81	28.48	31.96	37.28	44.82
Nitrate (kg/m)	1982	119	13.54	5.82	0.98	3.16	5.79	9.88	13.97	17.88	21.28	23.24	28.53
	1983	119	13.26	5.56	1.78	3.34	5.47	9.29	13.75	17.88	20.24	21.75	28.36
	1984	119	14.63	6.84	1.43	3.50	6.38	9.20	14.72	19.47	23.80	25.80	29.81
	1985	119	14.51	7.27	0.70	2.58	6.24	8.74	14.38	19.30	24.58	28.11	30.93
	1986	119	13.89	6.84	1.23	2.40	5.13	8.89	12.83	19.77	23.33	24.97	32.42
	1987	119	13.62	6.53	1.19	2.39	5.07	8.78	13.30	18.37	22.73	24.66	29.26
Ammonium (kg/m)	1982	119	3.83	1.48	0.40	0.60	1.05	2.15	2.83	3.97	4.79	5.70	7.44
	1983	119	3.00	1.38	0.39	0.84	1.31	2.86	2.99	3.88	5.02	5.70	7.17
	1984	119	3.15	1.42	0.38	0.68	1.36	2.15	3.06	4.16	5.20	5.78	8.28
	1985	119	2.92	1.59	0.12	0.44	1.16	1.87	2.80	3.77	4.90	6.18	8.87
	1986	119	2.98	1.59	0.18	0.44	1.07	1.74	2.84	3.84	5.36	5.92	7.50
	1987	119	2.89	1.50	0.28	0.38	1.04	1.68	2.78	3.89	4.99	6.38	8.77

TABLE A.8. (Continued)

	Year	N	Mean	Std Dev	Min	5th	10th	Percentile					Max
								25th	50th	75th	90th	95th	
Calcium (kg/m)	1982	114	1.98	1.14	0.34	0.56	0.72	1.11	1.66	2.24	3.59	4.36	6.29
	1983	114	2.04	1.22	0.53	0.78	0.88	1.22	1.72	2.52	3.75	4.93	6.78
	1984	114	2.06	1.11	0.32	0.73	0.98	1.31	1.85	2.55	3.36	4.14	7.88
	1985	114	1.81	1.15	0.36	0.56	0.83	0.87	1.47	2.34	3.69	4.35	5.35
	1986	114	1.68	1.04	0.34	0.48	0.63	0.81	1.41	2.05	3.08	3.74	5.96
	1987	114	1.65	1.29	0.27	0.41	0.59	0.88	1.27	2.09	3.28	3.99	8.73
Magnesium (kg/m)	1982	112	0.45	0.34	0.07	0.13	0.17	0.24	0.36	0.52	0.91	1.24	2.17
	1983	112	0.48	0.41	0.10	0.14	0.17	0.26	0.35	0.57	0.94	1.18	3.02
	1984	112	0.47	0.33	0.13	0.16	0.20	0.29	0.38	0.56	0.79	1.12	2.64
	1985	112	0.46	0.25	0.07	0.12	0.17	0.23	0.34	0.47	0.71	0.97	1.61
	1986	112	0.34	0.26	0.05	0.12	0.15	0.19	0.27	0.39	0.63	0.82	1.69
	1987	112	0.36	0.36	0.04	0.11	0.14	0.18	0.27	0.42	0.66	0.99	2.12
Sodium (kg/m)	1982	111	1.42	2.32	0.18	0.27	0.33	0.49	0.73	1.42	2.92	4.18	16.86
	1983	111	1.73	3.56	0.15	0.34	0.36	0.46	0.67	1.29	4.17	6.93	24.22
	1984	111	1.51	2.49	0.19	0.42	0.44	0.54	0.73	1.61	3.24	4.78	21.08
	1985	111	1.89	1.45	0.13	0.28	0.27	0.45	0.62	1.04	2.12	3.93	9.45
	1986	111	1.84	1.83	0.11	0.21	0.28	0.37	0.57	0.93	2.04	4.06	12.42
	1987	111	1.12	1.98	0.16	0.23	0.27	0.37	0.54	0.93	2.44	3.68	15.72
Chloride (kg/m)	1982	115	2.79	4.48	0.17	0.42	0.54	1.09	1.68	2.88	5.35	7.38	36.25
	1983	115	3.38	6.83	0.28	0.47	0.59	0.99	1.58	2.18	8.05	13.79	43.74
	1984	115	3.81	4.72	0.37	0.58	0.68	1.19	1.74	2.92	5.88	9.52	39.38
	1985	115	2.48	3.81	0.25	0.43	0.56	1.07	1.59	2.54	4.32	8.38	21.29
	1986	115	2.38	3.18	0.21	0.39	0.51	0.98	1.48	2.44	3.68	7.85	23.35
	1987	115	2.42	3.36	0.28	0.38	0.51	0.99	1.47	2.26	4.75	8.18	24.88
Potassium (kg/m)	1982	118	0.37	0.21	0.06	0.12	0.15	0.22	0.34	0.44	0.63	0.85	1.89
	1983	118	0.48	0.22	0.06	0.14	0.18	0.25	0.36	0.48	0.72	0.89	1.24
	1984	118	0.39	0.26	0.11	0.15	0.18	0.24	0.34	0.50	0.64	0.88	1.23
	1985	118	0.31	0.18	0.06	0.09	0.11	0.19	0.27	0.40	0.52	0.74	1.88
	1986	118	0.38	0.28	0.06	0.09	0.12	0.16	0.25	0.40	0.54	0.72	1.39
	1987	118	0.36	0.37	0.05	0.08	0.12	0.18	0.23	0.36	0.78	1.22	2.26

TABLE A.9. Frequency Distribution Percentiles of Estimated Concentrations (mg/l) at the Center of 2000 Squared Kilometer Hexagons in Eastern (n=1317) and Western (n=1875) United States

			Min	1	5	10	25	50	75	90	95	99	Max
pH	Eastern	Annual	4.12	4.17	4.22	4.25	4.35	4.55	4.68	4.82	4.97	5.01	5.07
		Winter	4.21	4.29	4.38	4.48	4.51	4.84	4.75	4.83	4.88	4.96	5.02
		Summer	3.98	3.99	4.07	4.13	4.23	4.44	4.58	4.98	5.23	5.48	5.68
	Western	Annual	4.64	4.70	4.80	4.88	5.00	5.18	5.34	5.53	5.63	5.88	6.09
		Winter	4.59	4.64	4.80	4.78	4.95	5.10	5.31	5.49	5.57	5.84	6.00
		Summer	4.53	4.66	4.79	4.86	5.00	5.14	5.28	5.52	5.65	5.81	5.96
Sulfate	Eastern	Annual	0.92	0.98	1.04	1.16	1.34	1.72	2.38	3.02	3.33	3.91	4.29
		Winter	0.38	0.47	0.84	0.96	1.08	1.23	1.50	1.94	2.24	2.54	2.69
		Summer	0.76	0.89	1.17	1.31	1.58	2.14	3.28	4.64	4.35	4.82	5.68
	Western	Annual	0.28	0.33	0.36	0.40	0.55	0.88	1.20	1.31	1.35	1.52	1.68
		Winter	0.17	0.19	0.22	0.26	0.36	0.75	1.16	1.32	1.35	1.45	1.64
		Summer	0.38	0.55	0.64	0.70	0.80	0.90	1.07	1.19	1.28	1.49	1.73
Nitrate	Eastern	Annual	0.56	0.58	0.63	0.74	0.82	1.23	1.55	1.92	2.15	2.61	2.87
		Winter	0.37	0.42	0.47	0.49	0.62	1.10	1.67	2.04	2.34	3.01	3.19
		Summer	0.73	0.63	0.95	1.01	1.15	1.35	1.64	2.12	2.31	2.65	2.97
	Western	Annual	0.20	0.25	0.36	0.43	0.64	0.76	0.94	1.16	1.23	1.34	1.41
		Winter	0.13	0.15	0.23	0.31	0.49	0.67	0.95	1.23	1.42	1.73	1.88
		Summer	0.25	0.52	0.64	0.75	0.88	1.04	1.18	1.25	1.33	1.49	1.98
Ammonium	Eastern	Annual	0.08	0.09	0.10	0.12	0.18	0.25	0.39	0.46	0.51	0.61	0.73
		Winter	0.03	0.04	0.08	0.08	0.10	0.17	0.28	0.33	0.35	0.50	0.57
		Summer	0.08	0.11	0.13	0.15	0.20	0.31	0.40	0.46	0.51	0.67	0.87
	Western	Annual	0.04	0.05	0.08	0.10	0.13	0.19	0.28	0.42	0.49	0.58	0.65
		Winter	0.02	0.02	0.03	0.04	0.07	0.12	0.24	0.40	0.47	0.60	0.67
		Summer	0.03	0.10	0.12	0.14	0.17	0.21	0.28	0.42	0.47	0.55	0.75
Calcium	Eastern	Annual	0.03	0.03	0.06	0.06	0.08	0.12	0.21	0.27	0.32	0.39	0.55
		Winter	0.01	0.02	0.04	0.05	0.06	0.08	0.13	0.18	0.19	0.28	0.53
		Summer	0.03	0.04	0.06	0.07	0.09	0.15	0.24	0.32	0.35	0.43	0.61
	Western	Annual	0.04	0.04	0.05	0.07	0.12	0.18	0.23	0.28	0.30	0.34	0.38
		Winter	0.03	0.03	0.04	0.04	0.06	0.12	0.18	0.22	0.24	0.29	0.46
		Summer	0.03	0.04	0.08	0.11	0.18	0.24	0.28	0.35	0.40	0.62	0.83

TABLE A.10. Frequency Distribution Percentiles of Estimated Deposition (kg/ha) at the Center of 2000 Squared Kilometer Hexagons in Eastern (n=1317) and Western (n=1875) United States

			Min	1	5	10	25	50	75	90	95	99	Max
Hydrogen	Eastern	Annual	0.05	0.06	0.07	0.13	0.25	0.30	0.42	0.50	0.61	0.71	0.85
		Winter	0.00	0.01	0.01	0.01	0.03	0.06	0.08	0.09	0.09	0.10	0.14
		Summer	0.00	0.01	0.02	0.04	0.07	0.12	0.16	0.21	0.26	0.33	0.38
	Western	Annual	0.01	0.01	0.02	0.02	0.03	0.04	0.06	0.15	0.20	0.26	0.29
		Winter	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.04	0.05	0.07	0.08
		Summer	0.00	0.00	0.00	0.00	0.01	0.01	0.02	0.03	0.05	0.08	0.12
Sulfate	Eastern	Annual	6.23	7.35	8.77	10.53	15.16	18.42	21.92	27.17	29.91	36.20	42.19
		Winter	0.29	0.46	0.58	0.70	1.24	2.79	4.33	4.97	5.47	6.23	6.71
		Summer	1.62	2.32	2.95	3.42	4.55	6.84	8.86	12.21	13.43	16.10	18.96
	Western	Annual	1.07	1.27	1.46	1.77	2.41	3.69	6.67	12.88	14.77	17.66	18.68
		Winter	0.09	0.11	0.16	0.22	0.36	0.57	1.09	2.58	3.41	4.37	4.75
		Summer	0.29	0.34	0.39	0.48	0.72	1.22	2.18	3.38	4.26	5.42	6.12
Nitrate	Eastern	Annual	0.10	0.49	7.15	7.81	9.29	11.12	13.18	17.85	20.16	22.90	27.58
		Winter	0.56	0.72	0.83	0.94	1.30	2.15	2.47	3.16	3.53	4.26	6.14
		Summer	1.80	2.09	2.48	2.72	3.30	3.90	4.79	6.13	6.80	7.57	9.14
	Western	Annual	1.48	1.58	1.79	2.07	2.43	3.13	5.95	9.08	10.08	11.17	11.91
		Winter	0.17	0.20	0.26	0.31	0.43	0.58	0.81	1.63	1.95	2.25	2.57
		Summer	0.17	0.31	0.46	0.53	0.79	1.20	2.11	3.02	3.66	4.19	5.32
Ammonium	Eastern	Annual	0.81	0.97	1.25	1.39	1.93	2.62	3.13	3.57	3.94	4.69	6.09
		Winter	0.06	0.09	0.13	0.15	0.21	0.30	0.41	0.54	0.73	1.00	1.40
		Summer	0.22	0.29	0.34	0.38	0.54	0.83	1.16	1.43	1.63	2.37	3.47
	Western	Annual	0.33	0.38	0.41	0.43	0.47	0.62	1.98	2.83	3.01	3.68	4.28
		Winter	0.02	0.03	0.03	0.04	0.07	0.11	0.21	0.41	0.46	0.63	0.87
		Summer	0.03	0.05	0.09	0.10	0.14	0.22	0.56	0.87	1.03	1.20	1.32
Calcium	Eastern	Annual	0.30	0.35	0.58	0.67	0.88	1.24	1.65	2.16	2.52	3.08	4.58
		Winter	0.02	0.05	0.06	0.07	0.09	0.14	0.21	0.29	0.34	0.37	0.83
		Summer	0.06	0.09	0.14	0.17	0.23	0.41	0.68	1.01	1.13	1.23	2.21
	Western	Annual	0.20	0.24	0.29	0.34	0.49	0.71	1.48	1.94	2.12	2.31	2.49
		Winter	0.03	0.04	0.04	0.05	0.06	0.08	0.15	0.32	0.35	0.39	0.48
		Summer	0.02	0.02	0.04	0.08	0.18	0.28	0.47	0.78	0.80	1.04	1.28

**TABLE A.11. Minimum and Maximum Annual, Winter, and Summer
Precipitation-weighted pH and Hydrogen Total
Deposition Observed at ADS Sites in 1987 by Region**

pH	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	pH	Sitename	State	pH	Sitename	Prov.	pH
ANNUAL	Marshall	TX	4.63	Leading Ridge	PA	4.68	Charleston Lake	ON	4.28
	Longview	TX	4.65	Penn State	PA	4.18	Melbourne	ON	4.28
	Farlington	KS	4.71	Zanesville	OH	4.15	Alvinston	ON	4.21
	Scott Lake	KS	5.72	Lamberton	MN	5.02	Quetico Centre	ON	4.93
	Goodwell Research	OK	5.73	Spooner	WI	5.18	Expt. Lake Area	ON	4.96
	Davis	CA	6.13	Fernberg	MN	5.13	Pickle Lake	ON	5.18
WINTER	Longview	TX	4.54	Caldwell	OH	4.21	Colchester	ON	4.12
	Attwater Prairie C	TX	4.59	Delaware	OH	4.24	Killarney	ON	4.14
	Farlington	KS	4.68	Zanesville	OH	4.28	Longwoods (a)	ON	4.28
	Smith Valley	NY	5.69	Camp Ripley	MN	5.02	Harcourt	NB	4.81
	Davis	CA	5.75	Verna Well Field	FL	5.06	Jackson	NS	4.82
	Las Animas	CO	6.09	Iberia	LA	5.09	Palmerston	ON	5.01
SUMMER	Marshall	TX	4.51	Leading Ridge	PA	3.98	Huron Park	ON	4.06
	Oliver Knoll	AZ	4.65	Lewes	DE	3.92	Wilmer	ON	4.06
	Farlington	KS	4.66	Scranton	PA	3.93	Wilberforce	ON	4.07
	Woodworth	ND	5.88	Fernberg	MN	5.21	Pickle Lake	ON	5.31
	Green River	UT	5.90	Spooner	WI	5.41	Expt. Lake Area	ON	5.37
	Huron Well Field	SD	6.04	Camp Ripley	MN	5.81	Exr Falls	ON	5.51
Deposition	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	kg/ha	Sitename	State	kg/ha	Sitename	Prov.	kg/ha
ANNUAL	Davis	CA	0.0027	Fernberg	MN	0.0459	Pickle Lake	ON	0.0378
	Smith Valley	NY	0.0048	Lamberton	MN	0.0496	Expt. Lake Area	ON	0.0506
	Reynolds Creek	ID	0.0048	Spooner	WI	0.0502	Quetico Centre	ON	0.0634
	Farlington	KS	0.2203	Washington Xing	NJ	0.7474	Wilberforce	ON	0.5254
	Marshall	TX	0.2924	Leading Ridge	PA	0.8341	Wilmer	ON	0.5499
	Longview	TX	0.3056	Kane	PA	0.9449	Charleston Lake	ON	0.5971
WINTER	Las Animas	CO	0.0003	Camp Ripley	MN	0.0019	Exr Falls	ON	0.0109
	Brookings	SD	0.0006	Round Lake	WI	0.0039	Palmerston	ON	0.0113
	Custer Battlefield	MT	0.0007	Gaylord-2	MI	0.0046	Geraldton	ON	0.0119
	Attwater Prairie C	TX	0.0735	West Point	NY	0.1177	Longwoods (a)	ON	0.0929
	Marshall	TX	0.0877	Bennett Bridge	NY	0.1270	Priceville 2	ON	0.1025
	Longview	TX	0.0977	Walker Branch	TN	0.1787	Killarney	ON	0.1057
SUMMER	Green River	UT	0.0007	Camp Ripley	MN	0.0020	Exr Falls	ON	0.0000
	Vines Hill	OR	0.0008	Spooner	WI	0.0122	Expt. Lake Area	ON	0.0124
	Huron Well Field	SD	0.0016	Lamberton	MN	0.0153	ELA (b)	ON	0.0139
	Konza Prairie	KS	0.0494	Leading Ridge	PA	0.3767	McKellar	ON	0.2044
	Marshall	TX	0.0641	Washington Xing	NJ	0.4090	Colchester	ON	0.2062
	Farlington	KS	0.0653	Kane	PA	0.4171	Wilberforce	ON	0.2349

TABLE A.12. Minimum and Maximum Annual, Winter, and Summer Sulfate Precipitation-Weighted Concentrations and Total Deposition Observed at ADS Sites in 1987 by Region

Concentration	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	mg/l	Sitename	State	mg/l	Sitename	Prov. mg/l	
ANNUAL	H.J. Andrews	OR	0.21	Greenville Station	ME	0.86	Pickle Lake	ON	0.74
	Starkey Experiment	OR	0.23	Quincy	FL	0.92	Jackson	NS	0.98
	Headquarters	ID	0.28	Fernberg	MN	1.08	Harcourt	NB	0.95
	Konza Prairie	KS	1.45	Delaware	OH	3.58	Palmerston	ON	4.18
	Wad	NE	1.58	Wooster	OH	3.82	Colchester	ON	4.52
	Oliver Knoll	AZ	1.59	Leading Ridge	PA	3.78	Port Stanley	ON	4.77
WINTER	Saval Ranch	NV	0.13	Greenville Station	ME	0.32	Port Cartier	PQ	0.27
	Starkey Experiment	OR	0.14	Winterport	ME	0.43	Harcourt	NB	0.43
	Montague	CA	0.15	Bridgton	ME	0.45	Jackson	NS	0.47
	Longview	TX	1.63	Indiana Dunes	IN	2.78	Merlin	ON	2.88
	Huron Well Field	SD	1.72	Caldwell	OH	2.82	Palmerston	ON	2.88
	Brookings	SD	1.98	Delaware	OH	2.93	Port Stanley	ON	2.91
SUMMER	Alsea	OR	0.27	Quincy	FL	0.88	Pickle Lake	ON	0.63
	Headquarters	ID	0.42	Camp Ripley	MN	0.93	ELA (b)	ON	0.64
	H.J. Andrews	OR	0.44	Marcell	MN	0.94	Ear Falls	ON	0.71
	Konza Prairie	KS	1.43	Penn State	PA	4.89	Port Stanley	ON	5.13
	Farlington	KS	1.81	Leading Ridge	PA	6.04	Wilkesport	ON	5.68
	Oliver Knoll	AZ	1.72	Lewes	DE	8.29	Huron Park	ON	5.78

Deposition	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	kg/ha	Sitename	State	kg/ha	Sitename	Prov. kg/ha	
ANNUAL	Starkey Experiment	OR	0.82	Fernberg	MN	8.22	Pickle Lake	ON	4.25
	Smith Valley	NV	0.91	Spooner	WI	8.12	Expt. Lake Area	ON	5.19
	Reynolds Creek	ID	1.14	Marcell	MN	8.87	Quetico Centre	ON	8.98
	Konza Prairie	KS	12.92	Bennett Bridge	NY	33.48	Merlin	ON	32.83
	Farlington	KS	15.71	Leading Ridge	PA	37.51	Port Stanley	ON	35.98
	Longview	TX	19.19	Kane	PA	44.83	Colchester	ON	48.84
WINTER	Saval Ranch	NV	0.88	Round Lake	WI	0.15	Geraldton	ON	0.36
	Silver Lake Ranger	OR	0.88	Gaylord-2	MI	0.19	Port Cartier	PQ	0.41
	Custer Battlefield	MT	0.18	Shawano	WI	0.27	Bonner Lake	ON	0.48
	Attwater Prairie C	TX	4.17	Sand Mountain Exp	AL	7.18	Killarney	ON	3.98
	Marshall	TX	4.51	Southeast	LA	7.28	Port Stanley	ON	4.14
	Longview	TX	5.51	Walker Branch	TN	8.84	Merlin	ON	4.88
SUMMER	Alsea	OR	0.28	Camp Ripley	MN	1.24	ELA (b)	ON	1.51
	Vines Hill	OR	0.28	Blackbelt	AL	2.13	Ear Falls	ON	1.83
	Hyslop Farm	OR	0.27	Caddo Valley	AR	2.45	Warsaw Caves	ON	1.94
	Great Plains Apiar	OK	4.81	Leading Ridge	PA	18.14	Merlin	ON	13.95
	Konza Prairie	KS	4.73	Washington Xing	NJ	18.96	Wilkesport	ON	18.81
	Farlington	KS	4.78	Kane	PA	21.33	Colchester	ON	19.48

TABLE A.13. Minimum and Maximum Annual, Winter, and Summer Nitrate Precipitation-Weighted Concentrations and Total Deposition Observed at ADS Sites in 1987 by Region

Concentration	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	mg/l	Sitename	State	mg/l	Sitename	Prov.	mg/l
ANNUAL	H.J. Andrews	OR	0.15	Quincy	FL	0.55	Jackson	NS	0.53
	La Grande	WA	0.32	Greenville Station	ME	0.58	Harcourt	NB	0.58
	Bull Run	OR	0.32	Kennedy Space Cent	FL	0.60	Port Cartier	PQ	0.58
	Brookings	SD	1.28	Penn State	PA	2.19	Palmerston	ON	2.98
	Mead	NE	1.41	Leading Ridge	PA	2.44	Alvinston	ON	2.97
	Las Animas	CO	1.45	Bennett Bridge	NY	2.49	Port Stanley	ON	3.27
WINTER	H.J. Andrews	OR	0.07	Everglades Nat. Pa	FL	0.35	Jackson	NS	0.47
	Olympic Nat. Park	WA	0.08	Newton	MS	0.38	Kejikujukik 2	NS	0.52
	Hyslop Farm	OR	0.15	Tifton, ARS	GA	0.38	Harcourt	NB	0.54
	Huron Well Field	SD	1.88	Wellston	MI	2.67	Port Stanley	ON	3.42
	Red Rock Canyon	NV	1.88	Bennett Bridge	NY	2.77	Palmerston	ON	3.55
	Brookings	SD	1.92	Gaylord-2	MI	3.22	Killarney	ON	4.51
SUMMER	Alsea	OR	0.14	Quincy	FL	0.81	Port Cartier	PQ	0.51
	Hyslop Farm	OR	0.38	Bridgton	ME	0.88	Pickle Lake	ON	0.53
	La Grande	WA	0.44	Blackbelt	AL	0.86	Bonner Lake	ON	0.81
	Las Animas	CO	1.72	Wilford	PA	3.00	Wilkesport	ON	3.14
	Vines Hill	OR	1.75	Leading Ridge	PA	3.04	Alvinston	ON	3.17
	Green River	UT	2.09	Lewes	DE	3.22	Huron Park	ON	3.45
Deposition	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	kg/ha	Sitename	State	kg/ha	Sitename	Prov.	kg/ha
ANNUAL	Smith Valley	NV	1.27	Greenville Station	ME	5.85	Pickle Lake	ON	4.87
	Starkey Experiment	OR	1.29	Selma	AL	5.91	Port Cartier	PQ	5.81
	Reynolds Creek	ID	1.54	Fernberg	MN	5.94	Quetico Centre	ON	5.77
	Mead	NE	10.72	Kane	PA	23.81	Priceville	ON	24.77
	Konza Prairie	KS	10.78	Leading Ridge	PA	24.28	Wilmer	ON	26.48
	Farlington	KS	11.23	Bennett Bridge	NY	29.26	Colchester	ON	26.58
WINTER	Saval Ranch	NV	0.10	Gaylord-2	MI	0.31	Ear Falls	ON	0.87
	Brookings	SD	0.14	Round Lake	WI	0.33	Geraldton	ON	0.74
	Silver Lake Ranger	OR	0.15	Shawano	WI	0.55	Port Cartier	PQ	1.06
	Farlington	KS	2.39	Virginia	VA	5.38	Coldwater	ON	5.41
	Longview	TX	2.49	Turners Falls	MA	6.42	Killarney	ON	6.57
	Marshall	TX	2.78	Bennett Bridge	NY	7.12	Priceville 2	ON	8.59
SUMMER	Alsea	OR	0.11	Selma	AL	0.98	Warsaw Caves	ON	1.22
	Hyslop Farm	OR	0.22	Greenville Station	ME	1.57	Jackson	NS	1.53
	La Grande	WA	0.28	Blackbelt	AL	1.82	Pickle Lake	ON	1.70
	Farlington	KS	3.97	Leading Ridge	PA	9.14	Wilkesport	ON	8.88
	Mead	NE	4.26	Kane	PA	9.48	Uxbridge	ON	9.37
	Konza Prairie	KS	4.34	Washington Xing	NJ	10.41	Colchester	ON	10.08

TABLE A.14. Minimum and Maximum Annual, Winter, and Summer Ammonium Precipitation-Weighted Concentrations and Total Deposition Observed at ADS Sites in 1987 by Region

Concentration	Western United States			Eastern United States			Southeastern Canada	
	Sitename	State	mg/l	Sitename	State	mg/l	Sitename	Prov. mg/l
ANNUAL	H.J. Andrews	OR	0.021	Greenville Station	ME	0.066	Jackson	NS 0.092
	La Grande	WA	0.051	Quincy	FL	0.077	Kejimikujik (b)	NS 0.097
	Headquarters	ID	0.060	Kennedy Space Cent	FL	0.077	Harcourt	NB 0.102
	Mead	NE	0.609	Rockport	IN	0.528	Merlin	ON 0.773
	Goodwell Research	OK	0.642	Zanesville	OH	0.545	Coldwater	ON 0.816
	Brookings	SD	0.673	Lamberton	MN	0.640	Palmerston	ON 0.866
WINTER	H.J. Andrews	OR	0.011	Greenville Station	ME	0.021	Harcourt	NB 0.022
	Starkey Experiment	OR	0.011	Turners Falls	MA	0.026	Port Cartier	PQ 0.022
	Olympic Nat. Park	WA	0.014	Bridgton	ME	0.027	Geraldton	ON 0.027
	Huron Well Field	SD	0.654	Bennett Bridge	NY	0.411	Coldwater	ON 0.494
	Brookings	SD	0.719	Spooner	WI	0.497	Killarney	ON 0.720
	Goodwell Research	OK	0.746	Gaylord-2	MI	0.759	Palmerston	ON 0.975
SUMMER	Alsea	OR	0.011	Quincy	FL	0.044	Port Cartier	PQ 0.120
	Buffalo Pass	CO	0.045	Clinton	MS	0.071	Jackson	NS 0.122
	H.J. Andrews	OR	0.047	Bradford Forest	FL	0.112	Bonner Lake	ON 0.162
	Pawnee	CO	0.677	Lamberton	MN	0.541	Merlin	ON 1.055
	Goodwell Research	OK	0.706	Penn State	PA	0.664	Dawson Creek	ON 1.226
	Green River	UT	0.846	Lewes	DE	0.823	Whitney	ON 1.306
Deposition	Western United States			Eastern United States			Southeastern Canada	
	Sitename	State	mg/l	Sitename	State	mg/l	Sitename	Prov. mg/l
ANNUAL	Starkey Experiment	OR	0.22	Greenville Station	ME	0.67	Pickle Lake	ON 0.92
	H.J. Andrews	OR	0.31	Bridgton	ME	0.70	Harcourt	NB 1.11
	Oliver Knoll	AZ	0.34	Quincy	FL	0.87	Kejimikujik (b)	NS 1.20
	Goodwell Research	OK	3.330	Shabbona	IL	4.58	Colchester	ON 0.36
	Brookings	SD	3.817	Argonne	IL	4.68	Palmerston	ON 0.36
	Mead	NE	4.035	Bennett Bridge	NY	4.82	Coldwater	ON 0.76
WINTER	Starkey Experiment	OR	0.01	Greenville Station	ME	0.04	Geraldton	ON 0.02
	Saval Ranch	NY	0.01	Bridgton	ME	0.04	Port Cartier	PQ 0.03
	Gypsum Creek	WY	0.02	Round Lake	WI	0.06	Harcourt	NB 0.05
	Farlington	KS	0.51	Bennett Bridge	NY	1.06	Killarney	ON 1.05
	Forest Seed Ctr	TX	0.55	Clinton	MS	1.15	Pricerville 2	ON 1.10
	Marshall	TX	0.61	Iberia	LA	1.58	Palmerston	ON 1.14
SUMMER	Alsea	OR	0.01	Selma	AL	0.13	Warsaw Caves	ON 0.28
	Hyslop Farm	OR	0.03	Quincy	FL	0.15	Jackson	NS 0.30
	La Grande	WA	0.04	Clinton	MS	0.19	Kejimikujik 2	NS 0.37
	Goodwell Research	OK	1.32	Illinois	IL	1.92	Colchester	ON 3.23
	Scott Lake	KS	1.49	Kane	PA	1.95	Merlin	ON 3.41
	Brookings	SD	1.52	Big Moose	NY	2.08	Dawson Creek	ON 5.91

Table A.15. Minimum and Maximum Annual, Winter, and Summer Calcium Precipitation-Weighted Concentrations and Total Deposition Observed at ADS Sites in 1987 by Region

Concentration	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	mg/l	Sitename	State	mg/l	Sitename	Prov.	mg/l
ANNUAL	H.J. Andrews	OR	0.030	Winterport	ME	0.029	Harcourt	NB	0.038
	Bull Run	OR	0.043	Greenville Station	ME	0.033	Jackson	NS	0.039
	La Grande	WA	0.044	Turners Falls	MA	0.039	Port Cartier	PQ	0.040
	Goodwell Research	OK	0.337	Indiana Dunes	IN	0.315	Merlin	ON	0.543
	Scott Lake	KS	0.355	McNay Research Sta	IA	0.310	Colchester	ON	0.604
	Las Animas	CO	0.401	Lamberton	MN	0.345	Port Stanley	ON	1.100
WINTER	Tanbark Flat	CA	0.022	Turners Falls	MA	0.003	Port Cartier	PQ	0.015
	La Grande	WA	0.022	Raleigh	NC	0.015	Jackson	NS	0.027
	Sequoia Nat. Park	CA	0.025	Winterport	ME	0.016	Harcourt	NB	0.032
	Big Bend Nat'l Pk	TX	0.430	Parsons	WV	0.227	Palmerston	ON	0.713
	Red Rock Canyon	NV	0.502	Wellston	MI	0.202	Port Stanley	ON	0.803
	Brookings	SD	0.929	Indiana Dunes	IN	0.562	Huron Park	ON	0.934
SUMMER	Hyslop Farm	OR	0.028	Winterport	ME	0.031	Jackson	NS	0.024
	Alsea	OR	0.030	Turners Falls	MA	0.036	Port Cartier	PQ	0.030
	La Grande	WA	0.036	Bridgton	ME	0.037	Kejimikujik 2	NS	0.035
	Lancaster	KS	0.524	Monmouth	IL	0.401	Huron Park	ON	0.557
	Murphy Ridge	UT	0.709	Surging-NADP	WI	0.423	Longwoods (b)	ON	0.819
	Green River	UT	0.935	Lamberton	MN	0.423	Colchester	ON	0.709
Deposition	Western United States			Eastern United States			Southeastern Canada		
	Sitename	State	kg/ha	Sitename	State	kg/ha	Sitename	Prov.	kg/ha
ANNUAL	Smith Valley	NV	0.10	Winterport	ME	0.27	Port Cartier	PQ	0.30
	Starkey Experiment	OR	0.22	Greenville Station	ME	0.34	Harcourt	NB	0.42
	Palouse Conservati	WA	0.31	Bridgton	ME	0.30	Jackson	NS	0.52
	Farlington	KS	2.10	Indiana Dunes	IN	2.00	Merlin	ON	4.32
	Mead	NE	2.25	Argonne	IL	3.00	Colchester	ON	5.43
	LBJ National Grass	TX	2.00	McNay Research Sta	IA	3.22	Port Stanley	ON	0.73
WINTER	Silver Lake Ranger	OR	0.03	Turners Falls	MA	0.01	Port Cartier	PQ	0.02
	Manitou	CO	0.03	Gaylord-2	MI	0.01	Bear Island	ON	0.00
	Las Animas	CO	0.03	Round Lake	WI	0.03	Harcourt	NB	0.07
	LBJ National Grass	TX	0.50	Indiana Dunes	IL	0.40	Merlin	ON	0.90
	Big Bend Nat'l Pk	TX	0.54	Parsons	WV	0.49	Port Stanley	ON	1.14
	Great Plains Apiar	OK	0.60	Lilley Cornett Woo	KY	0.54	Huron Park	ON	1.29
SUMMER	Hyslop Farm	OR	0.02	Winterport	ME	0.05	Jackson	NS	0.00
	Alsea	OR	0.02	Greenville Station	ME	0.00	Kejimikujik 2	NS	0.00
	La Grande	WA	0.02	Selma	AL	0.00	Harcourt	NB	0.11
	Lancaster	KS	1.22	Big Springs Fish H	IA	1.24	Wellesley	ON	1.53
	Muleshoe Nat'l Wil	TX	1.20	McNay Research Sta	IA	1.20	Uxbridge	ON	2.00
	Great Plains Apiar	OK	1.44	Indiana Dunes	IN	1.20	Colchester	ON	2.74

FIGURE A.1 Annual 1987 Spatial Distribution of (a) Precipitation-weighted pH and (b) Hydrogen Ion Deposition

FIGURE A.2. Winter 1987 Spatial Distribution of (a) Precipitation-weighted pH and (b) Hydrogen Ion Deposition

FIGURE A.3. Summer 1987 Spatial Distribution of (a) Precipitation-weighted pH and (b) Hydrogen Ion Deposition

FIGURE A.4. Annual 1987 Spatial Distribution of Sulfate (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.5. Winter 1987 Spatial Distribution of Sulfate (a) Precipitation-weighted Concentration (b) Deposition

FIGURE A.6. Summer 1987 Spatial Distribution of Sulfate (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.7. Annual 1987 Spatial Distribution of Nitrate (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.8. Winter 1987 Spatial Distribution of Nitrate (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.9. Summer 1987 Spatial Distribution of Nitrate (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.10. Annual 1987 Spatial Distribution of Ammonium (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.11. Winter 1987 Spatial Distribution of Ammonium (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.12. Summer 1987 Spatial Distribution of Ammonium (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.13. Annual 1987 Spatial Distribution of Calcium (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.14. Winter 1987 Spatial Distribution of Calcium (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.15. Summer 1987 Spatial Distribution of Calcium (a) Precipitation-weighted Concentration and (b) Deposition

FIGURE A.16. 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 the 1979-87 Trend Subset.

FIGURE A.17. 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 the 1982-87 Trend Subset.

FIGURE A.18. 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 the 1979-87 Trend Subset.

FIGURE A.19. 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 the 1982-87 Trend Subset.

FIGURE A.20. Total Precipitation Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Precipitation. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.21. Total Precipitation Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Precipitation. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.22. pH Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual pH. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.23. pH Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual pH. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.24. Hydrogen Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.25. Hydrogen Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.26. Hydrogen Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.27. Hydrogen Deposition Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.28. 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 the 1979-87 Trend Subset.

FIGURE A.29. 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 the 1982-87 Trend Subset.

FIGURE A.30. Sulfate Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.31. Sulfate Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.32. Sulfate Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.33. Sulfate Deposition Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.34. 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 the 1979-87 Trend Subset.

FIGURE A.35. 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 the 1982-87 Trend Subset.

FIGURE A.36. Nitrate Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.37. Nitrate Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.38. Nitrate Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.39. Nitrate Deposition Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.40. 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 the 1979-87 Trend Subset.

FIGURE A.41. 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 the 1982-87 Trend Subset.

FIGURE A.42. Ammonium Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.43. Ammonium Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.44. Ammonium Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.45. Ammonium Deposition Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.46. 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 the 1979-87 Trend Subset.

FIGURE A.47. 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 the 1982-87 Trend Subset.

FIGURE A.48. Calcium Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.49. Calcium Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.50. Calcium Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.51. Calcium Deposition Trend for 1982-87 Trend Sites.
 (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition.
 Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.52. 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 the 1979-87 Trend Subset.

FIGURE A.53. 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 the 1982-87 Trend Subset.

FIGURE A.54. Chloride Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.55. Chloride Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.56. Chloride Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.57. Chloride Deposition Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.58. 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 the 1979-87 Trend Subset.

FIGURE A.59. 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 the 1982-87 Trend Subset.

FIGURE A.60. Sodium Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.61. Sodium Concentration Trend for 1982-87 Trend Sites.
 (a) Ray Angle Proportional to Trend Estimate. (b) Ray
 Angle Proportional to Estimated 1987 Annual Concentration.
 Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.62. Sodium Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.63. Sodium Deposition Trend for 1982-87 Trend Sites.
 (a) Ray Angle Proportional to Trend Estimate. (b) Ray
 Angle Proportional to Estimated 1987 Annual Deposition.
 Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.64. 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 the 1979-87 Trend Subset.

FIGURE A.65. 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 the 1982-87 Trend Subset.

FIGURE A.66. Potassium Concentration Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.67. Potassium Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.68. Potassium Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.69. Potassium Deposition Trend for 1982-87 Trend Sites.
 (a) Ray Angle Proportional to Trend Estimate. (b) Ray
 Angle Proportional to Estimated 1987 Annual Deposition.
 Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.70. 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 the 1979-87 Trend Subset.

FIGURE A.71. 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 the 1982-87 Trend Subset.

FIGURE A.72. Magnesium Concentration Trend for 1979-87 Trend Sites.
 (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration.
 Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.73. Magnesium Concentration Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Concentration. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.74. Magnesium Deposition Trend for 1979-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

FIGURE A.75. Magnesium Deposition Trend for 1982-87 Trend Sites. (a) Ray Angle Proportional to Trend Estimate. (b) Ray Angle Proportional to Estimated 1987 Annual Deposition. Solid Symbol Indicates Significant Trend ($p < 0.05$)

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