TREND ANALYSIS OF WEEKLY ACID RAIN DATA -- 1978–83

By Terry L. Schertz and Robert M. Hirsch

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CONTENTS

ILLUSTRATIONS

TABLES

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ABSTRACT

There are 19 stations in the National Atmospheric Deposition Program which operated over the period 1978-83 and were subsequently incorporated into the National Trends Network in 1983. The precipitation chemistry data for these stations for this period were analyzed for trend, spatial correlation, seasonality, and relationship to precipitation volume. The intent of the analysis was to provide insights on the sources of variation in precipitation chemistry and to attempt to ascertain what statistical procedures may be most useful for ongoing analysis of the National Trends Network data. The Seasonal Kendall test was used for detection of trends in raw concentrations of dissolved constituents, pH and specific conductance, and residuals of these parameters from regression analysis. Forty-one percent of the trends detected in the raw concentrations were downtrends, 4 percent were uptrends, and 55 percent showed no trends at $\alpha = 0.2$. At a more restrictive significance level of $\alpha = 0.05$, 24 percent of the trends detected were downtrends, 2 percent were uptrends, and 74 percent showed no trends. The two constituents of greatest interest in terms of human generated emissions and environmental effects, sulfate and nitrate, showed only downtrends, and sulfate showed the largest decreases in concentration per year of all the ions tested. The relationship between dissolved concentration and precipitation volume was expressed as:

$$\ln(C) = a + b * \ln(P)$$

and residuals from regression analysis of the data with this model were also tested for trends. Of the 133 tests for trends in residuals, 49 percent showed downtrends, 7 percent showed uptrends and 44 percent showed no detectable trends at $\alpha = 0.2$. For $\alpha = 0.05$, 38 percent of the tests showed downtrends, 2 percent showed uptrends and 60 percent showed no detectable trend.

Further examination of the residuals showed evidence of a seasonal pattern in the data for most constituents from most stations. For example, sulfate and nitrate residuals generally were higher in the summer, and chloride residuals generally were higher in the winter. Characteristics from the regression model above and the same regression model with a seasonal term added were compared among all of the stations for all parameters, and no evidence of geographical patterns was detected.

Smoothed residual series of sodium over time for all 19 stations showed a decline in 1980, some of which were very large. For example, the concentrations at Wellston, Michigan, at the beginning of 1980 were about 7.4 times the concentrations at the end of 1980. The simultaneity of the changes over the Nation shown by these plots suggests the possibility that some trends may have been the result of changes in procedures or materials used in sample collection, shipment or analysis.

Five of the 19 stations were located in North Carolina which provided an opportunity to explore the spatial relationship of the records and a few of the possibilities for spatial analysis. The cross-correlation coefficients for the sulfate residuals from the seasonal model for all possible pairs of records and the interstation distance showed a strong relationship ($R^2 = 0.86$). The Seasonal Kendall test was applied to all five stations simultaneously, which indicated a downward trend in sulfate that was significant at $p = 0.018$.

INTRODUCTION

The National Trends Network (NTN) was established in 1982 to provide a nationwide, long-term, continuous set of data to be used in documenting and understanding changes in the chemistry of atmospheric deposition. Prior to the establishment of the NTN program, the only nationwide program for collecting such data was conducted as part of the National Atmospheric Deposition Program (NADP) under the sponsorship of the State Agricultural Experiment Stations. The NADP monitoring program, established in 1978, utilizes a Program Coordination Office located at Colorado State University for operational coordination and a contract Central Analytical Laboratory operated by the Illinois State Water Survey for sample analysis. The NADP sample collection procedures, chemical analysis laboratory and analytical procedures, and many of the stations operated by the NADP have been incorporated into the NTN. Coordination for what is now the NADP/NTN network is provided by NADP. Data from 19 stations established by NADP in 1978 and subsequently incorporated into the NADP/NTN network constitute the data base for this study (see fig. 1). The available data record from each of these stations covers more than a 5-year period from late 1978 through the end of 1983.

Purpose and Scope

The purpose of this study is to examine the chemical quality of precipitation from the 19 NADP/NTN stations that have 5 or more years of weekly data. This examination is intended to describe some of the characteristics of these data--for example, the relationship among chemical concentrations, precipitation volume, and time of year, the spatial correlations of the data, and the distributional properties of the data. These data also are examined for evidence of temporal trends to begin the effort to meet the established goals of the NTN. This evaluation also serves to supplement the current quality assurance program. If trends are found to be rather abrupt, all in the same direction and contemporaneous in time at many of the stations, this would suggest that there was a change in sample collection, shipment, or analysis, or possibly, a large-scale climatic variable. Whereas, if trends are gradual, or are abrupt but occurring at different times at different stations, this would suggest that the trends in precipitation chemistry may be real. If the trends at certain stations are very abrupt, this would suggest that the precipitation chemistry at that station may be strongly influenced by a particular source of the chemical in question rather than by a multitude of sources spread over a wide area. The description provided here is just a first exploratory step in evaluating the actual trends in precipitation chemistry and possible problems with the data-collection network. In combination with emission data or meteorological data, the method of analysis presented in this report could lead to significant interpretation of the processes. The explanation of the causes of the observed trends is beyond the scope of this report. Based on the results of the data analysis, some suggestions are made concerning fruitful approaches to ongoing exploratory data analysis and future ongoing trend analysis to be used as the NTN matures.

The 19 stations are scattered across the United States, but a majority are located in the east. Five of the 19 stations are located in North Carolina. These stations are examined as a group as well as individually to investigate the spatial correlation structure of the data and its possible impact on regional trend analysis.

Data Base Description

The location and identification of the 19 stations used in this study are shown in figure 1. The methods of collection of wet-deposition data for stations in the NADP/NTN programs are described by Bigelow (1982). The samples collected weekly at each site are sent to the Central Analytical Laboratory for analysis. The chemical and physical parameters determined by the laboratory are pH, specific conductance, and dissolved concentrations of sulfate, nitrate, calcium, chloride, ammonia, sodium, potassium, and magnesium. The laboratory flags outliers based on cation/anion balances and historical percentile categories of values as well as visual inspection of collectors for nonstandard samples as part of the data-validation and quality-assurance procedures. Analyses flagged due to discrepancies in charge balances are rerun, but the values are allowed to stand without error flags if similar results are obtained. Otherwise, the values are replaced. Values from analyses that are flagged as nonstandard samples retain the error flags. The Central Analytical Laboratory analytical methods have been documented by Stensland and others (1980) and the quality-assurance procedures have been documented by Bowersox (1984). The data from the laboratory are sent to the NADP/NTN Coordination Office at Colorado State University for further quality-assurance screening. The analyses from the Central Analytical Laboratory that bear error flags are reviewed to determine whether they should be included in the final data set. The approved data are sent to the Pacific Northwest Laboratory (PNL) for storage in a common data bank for acid deposition data collected through most of the larger deposition-monitoring programs in the United States. The data used in this study was obtained from PNL after it had been processed through all existing quality assurance steps.

The data were edited further by the authors. The additional editing procedures were: (1) All samples flagged as bulk samples were deleted; (2) all samples that had measured components reported, but volume of precipitation missing, were deleted; and (3) values that were flagged as "less than the limit of detection" were used in the statistical analyses of this study as one-half of the reported value. This was an arbitrary decision and constituted less than 5 percent of the samples for any given constituent, but comparison tests were run using one-quarter of the reported values and three-quarters of the reported values without a detectable difference in the results. Verification of the validity of the approved data set, by performing chemical logic checks or other screening procedures, is not within the scope of this study. The purpose is to analyze the data as reported by PNL, because these are the data which are widely available to the scientific community. In the process of data analysis, a number of highly suspect values were identified (on the basis of being extreme outliers in regression analyses), and they are listed in an appendix of the report. A statistical summary of the data from each station is shown in table 1.

Some individuals associated with the NADP/NTN programs have expressed concern over a possible contamination problem from the lids of the sample containers (buckets). The cause of this alleged problem is the possibility that the sample may react with an O-ring in the bucket lid if the bucket is inverted during shipment. Bowersox (written commun., February 1985) summarized the results of some studies done concerning the problem and concluded the following:

> Bucket blank tests have been done to determine the potential for biases in the inorganic ion concentrations of wet-deposition samples

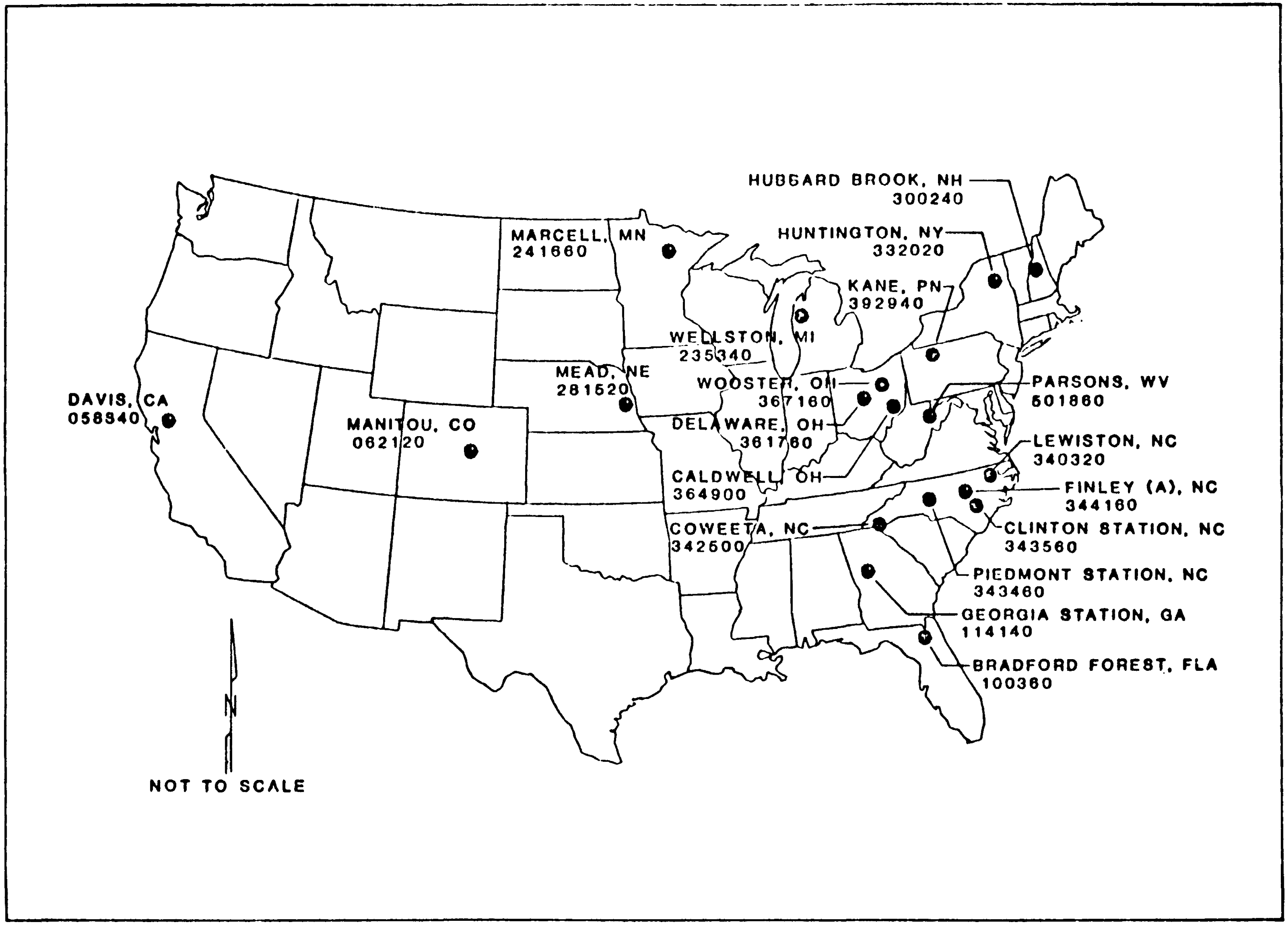


Figure 1.--Location of National Trends Network stations included in the study.

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK

[UMHOS, micromho per centimeter at 25° Celsius; MG/L, milligram per liter; ML, milliliter]

STATION NUMBER: 058840 STATION NAME: DAVIS, CALIFORNIA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM NOV. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	122	526.50	3.10	21.92	84.53	17.12	10.05	6.70	4.60
PH (LAB)	129	7.43	3.91	5.92	6.95	6.27	5.93	5.61	4.86
SULFATE (MG/L)	115	18.75	0.10	1.46	5.96	1.40	0.80	0.51	0.30
NITRATE (MG/L)	115	20.31	0.22	1.56	4.14	2.08	1.03	0.59	0.37
CHLORIDE (MG/L)	115	6.63	0.05	0.64	2.09	0.72	0.37	0.20	0.08
AMMONIUM (MG/L)	115	5.50	0.02	0.68	1.75	0.77	0.52	0.32	0.15
SODIUM (MG/L)	115	23.41	0.02	0.87	4.00	0.48	0.23	0.12	0.05
POTASSIUM (MG/L)	115	0.59	0.00	0.05	0.20	0.04	0.03	0.02	0.01
CALCIUM (MG/L)	115	3.80	0.01	0.23	1.15	0.19	0.09	0.04	0.02
VOLUME OF PRECIPITATION (ML)	138	8504.90	0.10	1534.67	6193.06	2175.45	768.25	92.72	2.95
CATION/ANION BALANCE (PERCENT)	114	40.90	-11.13	10.50	35.14	17.18	8.86	1.72	-6.00

STATION NUMBER: 062120 STATION NAME: MANITOU, COLORADO

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	167	66.40	2.90	15.23	35.70	19.70	12.80	8.80	4.45
PH (LAB)	173	7.40	4.18	5.36	6.78	5.82	5.18	4.80	4.44
SULFATE (MG/L)	160	18.47	0.10	1.76	4.61	2.09	1.28	0.77	0.29
NITRATE (MG/L)	160	7.29	0.02	1.66	4.36	2.19	1.31	0.81	0.20
CHLORIDE (MG/L)	160	1.22	0.02	0.21	0.62	0.27	0.15	0.07	0.03
AMMONIUM (MG/L)	160	1.91	0.02	0.26	0.67	0.31	0.20	0.08	0.02
SODIUM (MG/L)	160	4.27	0.01	0.30	1.42	0.27	0.11	0.05	0.02
POTASSIUM (MG/L)	160	1.12	0.00	0.09	0.32	0.10	0.05	0.02	0.01
CALCIUM (MG/L)	160	6.89	0.03	0.55	1.85	0.61	0.31	0.17	0.06
VOLUME OF PRECIPITATION (ML)	191	4025.20	0.30	618.47	2260.52	933.70	326.80	60.60	3.68
CATION/ANION BALANCE (PERCENT)	160	52.89	-22.85	2.54	33.59	6.15	-0.58	-6.35	-15.04

STATION NUMBER: 100360 STATION NAME: BRADFORD FOREST, FLORIDA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	183	497.30	2.80	24.87	66.30	23.40	14.30	9.50	5.52
PH (LAB)	185	6.84	3.54	4.98	6.22	5.24	4.91	4.59	4.07
SULFATE (MG/L)	181	28.92	0.10	2.21	6.67	2.10	1.35	0.90	0.52
NITRATE (MG/L)	181	6.60	0.02	1.03	3.24	1.29	0.72	0.42	0.16
CHLORIDE (MG/L)	181	52.62	0.05	1.37	4.21	1.08	0.57	0.30	0.14
AMMONIUM (MG/L)	181	1.92	0.02	0.17	0.69	0.19	0.09	0.03	0.02
SODIUM (MG/L)	181	29.30	0.04	1.01	3.40	0.76	0.38	0.19	0.09
POTASSIUM (MG/L)	181	3.81	0.00	0.10	0.25	0.08	0.04	0.02	0.01
CALCIUM (MG/L)	181	6.37	0.02	0.33	1.32	0.28	0.15	0.09	0.04
VOLUME OF PRECIPITATION (ML)	203	7657.70	0.30	1771.20	5645.04	2593.70	1228.90	388.30	22.96
CATION/ANION BALANCE (PERCENT)	179	25.49	-32.68	-0.39	11.55	3.49	-1.17	-4.41	-10.45

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK--CONTINUED

STATION NUMBER: 114140 STATION NAME: GEORGIA STATION, GEORGIA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	209	197.20	4.60	24.44	59.15	26.40	17.10	11.00	6.50
PH (LAB)	212	7.56	3.51	4.81	6.29	4.98	4.68	4.40	4.03
SULFATE (MG/L)	208	28.30	0.19	2.71	7.11	2.96	1.79	1.20	0.61
NITRATE (MG/L)	208	6.74	0.02	1.09	2.87	1.42	0.83	0.47	0.18
CHLORIDE (MG/L)	208	10.37	0.02	0.51	1.65	0.47	0.29	0.20	0.08
AMMONIUM (MG/L)	208	13.80	0.01	0.29	0.86	0.27	0.13	0.08	0.02
SODIUM (MG/L)	208	29.29	0.01	0.61	1.72	0.43	0.18	0.09	0.04
POTASSIUM (MG/L)	208	4.57	0.00	0.10	0.36	0.06	0.03	0.02	0.00
CALCIUM (MG/L)	208	6.66	0.01	0.24	0.91	0.25	0.11	0.06	0.03
VOLUME OF PRECIPITATION (ML)	232	8340.80	0.50	1768.01	6257.28	2427.15	1224.45	364.80	4.26
CATION/ANION BALANCE (PERCENT)	207	60.16	-29.96	-0.78	18.02	1.86	-2.67	-6.15	-12.57

STATION NUMBER: 235340 STATION NAME: WELLSTON, MICHIGAN

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	190	139.20	5.30	30.73	64.89	36.75	24.80	17.87	10.90
PH (LAB)	191	6.80	3.67	4.59	6.13	4.66	4.45	4.26	3.98
SULFATE (MG/L)	189	24.80	0.42	3.46	9.29	4.07	2.78	1.73	0.73
NITRATE (MG/L)	189	16.30	0.02	2.62	5.95	3.35	2.05	1.46	0.71
CHLORIDE (MG/L)	189	1.99	0.02	0.27	0.96	0.30	0.17	0.11	0.05
AMMONIUM (MG/L)	189	3.02	0.02	0.52	1.46	0.72	0.39	0.18	0.03
SODIUM (MG/L)	189	9.47	0.00	0.45	1.80	0.21	0.08	0.04	0.02
POTASSIUM (MG/L)	189	0.85	0.00	0.05	0.13	0.04	0.03	0.02	0.01
CALCIUM (MG/L)	189	5.55	0.02	0.48	1.67	0.51	0.26	0.14	0.06
VOLUME OF PRECIPITATION (ML)	223	5495.00	0.30	1073.30	3351.92	1551.40	762.80	290.60	24.34
CATION/ANION BALANCE (PERCENT)	189	44.47	-20.42	-0.50	12.70	2.77	-1.51	-5.24	-10.55

STATION NUMBER: 241660 STATION NAME: MARCELL, MINNESOTA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM JULY 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	226	91.60	2.50	16.12	37.36	18.00	12.60	9.15	5.90
PH (LAB)	231	7.48	3.95	5.31	6.43	5.71	5.20	4.83	4.41
SULFATE (MG/L)	220	15.00	0.10	1.93	6.28	2.18	1.50	0.88	0.44
NITRATE (MG/L)	220	15.69	0.02	1.76	4.55	2.08	1.30	0.84	0.38
CHLORIDE (MG/L)	220	8.48	0.02	0.24	0.68	0.23	0.12	0.07	0.04
AMMONIUM (MG/L)	220	5.78	0.02	0.48	1.50	0.62	0.32	0.11	0.02
SODIUM (MG/L)	220	11.72	0.00	0.35	1.68	0.25	0.07	0.04	0.02
POTASSIUM (MG/L)	220	2.47	0.00	0.08	0.24	0.09	0.04	0.02	0.01
CALCIUM (MG/L)	220	3.10	0.04	0.41	1.30	0.48	0.23	0.15	0.07
VOLUME OF PRECIPITATION (ML)	246	9071.50	1.20	942.45	3268.35	1298.50	503.10	165.02	4.73
CATION/ANION BALANCE (PERCENT)	220	45.15	-25.88	2.80	23.92	7.27	0.63	-4.42	-10.63

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK--CONTINUED

STATION NUMBER: 281520 STATION NAME: MEAD, NEBRASKA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM JULY 1978 TO DEC. 1983

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	185	131.00	4.80	24.03	57.82	28.85	17.00	10.95	7.40
PH (LAB)	192	7.64	3.73	5.78	6.97	6.44	5.86	5.12	4.46
SULFATE (MG/L)	180	16.35	0.27	3.00	8.17	3.82	2.21	1.38	0.75
NITRATE (MG/L)	180	21.76	0.02	2.50	7.49	2.77	1.73	1.10	0.66
CHLORIDE (MG/L)	179	3.66	0.02	0.38	1.28	0.42	0.20	0.11	0.06
AMMONIUM (MG/L)	180	8.14	0.02	1.12	2.99	1.32	0.84	0.48	0.13
SODIUM (MG/L)	180	7.08	0.01	0.48	2.13	0.43	0.18	0.07	0.03
POTASSIUM (MG/L)	180	2.45	0.01	0.15	0.51	0.17	0.07	0.04	0.01
CALCIUM (MG/L)	180	11.90	0.05	0.87	2.60	1.14	0.50	0.22	0.10
VOLUME OF PRECIPITATION (ML)	213	9107.50	0.10	1055.77	4038.12	1576.85	439.00	96.85	1.34
CATION/ANION BALANCE (PERCENT)	179	74.23	-21.98	10.66	31.89	18.90	7.69	0.79	-5.06

STATION NUMBER: 300240 STATION NAME: HUBBARD BROOK, NEW HAMPSHIRE

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM JULY 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	249	175.50	2.80	29.30	70.20	36.30	24.60	14.45	7.20
PH (LAB)	252	6.68	3.50	4.47	5.52	4.61	4.35	4.14	3.90
SULFATE (MG/L)	248	15.88	0.10	2.66	6.50	3.69	2.09	1.11	0.55
NITRATE (MG/L)	248	12.00	0.02	1.95	5.07	2.54	1.49	0.93	0.37
CHLORIDE (MG/L)	248	15.96	0.02	0.36	1.05	0.29	0.16	0.09	0.04
AMMONIUM (MG/L)	248	1.64	0.02	0.20	0.61	0.28	0.13	0.05	0.02
SODIUM (MG/L)	248	4.32	0.01	0.28	1.34	0.22	0.09	0.05	0.01
POTASSIUM (MG/L)	247	0.67	0.00	0.03	0.08	0.03	0.02	0.01	0.00
CALCIUM (MG/L)	248	3.58	0.01	0.20	0.64	0.19	0.10	0.05	0.02
VOLUME OF PRECIPITATION (ML)	263	6076.10	1.70	1510.40	3986.40	2117.00	1198.60	561.00	48.26
CATION/ANION BALANCE (PERCENT)	247	43.28	-32.80	-2.53	10.84	1.04	-3.70	-7.22	-13.45

STATION NUMBER: 332020 STATION NAME: HUNTINGTON, NEW YORK

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM NOV. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	224	163.80	4.40	29.23	66.12	37.12	25.20	15.12	6.80
PH (LAB)	224	6.77	3.54	4.44	5.50	4.59	4.35	4.16	3.90
SULFATE (MG/L)	224	21.73	0.22	2.85	7.32	3.78	2.22	1.18	0.47
NITRATE (MG/L)	224	11.02	0.03	2.15	5.55	2.62	1.74	1.05	0.50
CHLORIDE (MG/L)	224	1.12	0.02	0.18	0.54	0.21	0.14	0.07	0.03
AMMONIUM (MG/L)	224	2.34	0.02	0.31	0.82	0.41	0.23	0.09	0.02
SODIUM (MG/L)	224	2.83	0.00	0.17	0.62	0.17	0.07	0.03	0.01
POTASSIUM (MG/L)	224	0.45	0.00	0.03	0.09	0.03	0.02	0.01	0.00
CALCIUM (MG/L)	224	2.10	0.01	0.23	0.83	0.26	0.12	0.07	0.02
VOLUME OF PRECIPITATION (ML)	237	6221.00	0.10	1346.98	3476.44	1980.90	1134.00	422.90	61.83
CATION/ANION BALANCE (PERCENT)	224	28.03	-48.32	-3.96	7.58	-0.53	-4.14	-7.64	-14.72

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK--CONTINUED

STATION NUMBER: 340320 STATION NAME: LEWISTON, NORTH CAROLINA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM NOV. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	235	275.70	3.20	29.84	88.38	33.20	22.20	13.20	6.78
PH (LAB)	243	7.21	3.28	4.66	5.81	4.85	4.56	4.30	3.90
SULFATE (MG/L)	231	37.61	0.19	3.30	10.49	3.43	2.08	1.21	0.60
NITRATE (MG/L)	231	27.37	0.02	1.76	5.10	2.08	1.03	0.58	0.23
CHLORIDE (MG/L)	231	10.38	0.03	0.82	3.32	0.88	0.39	0.22	0.08
AMMONIUM (MG/L)	231	5.22	0.02	0.31	1.05	0.36	0.15	0.07	0.02
SODIUM (MG/L)	231	30.90	0.01	0.78	2.22	0.68	0.28	0.13	0.04
POTASSIUM (MG/L)	231	0.55	0.00	0.06	0.18	0.06	0.03	0.02	0.01
CALCIUM (MG/L)	231	6.42	0.01	0.34	1.68	0.28	0.12	0.07	0.04
VOLUME OF PRECIPITATION (ML)	254	8624.00	0.70	1390.33	4383.72	2007.52	1013.80	274.82	3.65
CATION/ANION BALANCE (PERCENT)	230	27.31	-28.05	-1.92	7.51	0.86	-2.05	-5.06	-10.62

STATION NUMBER: 342500 STATION NAME: COWEETA, NORTH CAROLINA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM JULY 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	238	122.10	2.80	20.08	52.13	24.97	16.05	10.10	5.89
PH (LAB)	240	6.83	3.53	4.69	5.84	4.89	4.61	4.41	4.09
SULFATE (MG/L)	235	13.53	0.10	2.16	5.32	2.60	1.63	1.06	0.54
NITRATE (MG/L)	235	5.16	0.02	1.08	3.12	1.40	0.81	0.48	0.19
CHLORIDE (MG/L)	235	3.17	0.02	0.26	0.84	0.30	0.17	0.10	0.05
AMMONIUM (MG/L)	235	1.86	0.01	0.18	0.62	0.22	0.11	0.05	0.02
SODIUM (MG/L)	235	3.21	0.00	0.25	0.99	0.25	0.11	0.05	0.02
POTASSIUM (MG/L)	235	0.39	0.00	0.03	0.12	0.04	0.02	0.01	0.00
CALCIUM (MG/L)	235	3.35	0.01	0.19	0.69	0.18	0.08	0.04	0.02
VOLUME OF PRECIPITATION (ML)	259	11280.00	1.80	2363.25	7502.30	3143.70	1745.80	598.60	11.60
CATION/ANION BALANCE (PERCENT)	235	36.22	-33.42	-2.49	9.56	1.03	-2.60	-6.60	-14.11

STATION NUMBER: 343460 STATION NAME: PIEDMONT STATION, NORTH CAROLINA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	220	216.50	4.50	33.68	84.26	43.30	26.60	16.42	7.33
PH (LAB)	228	7.47	3.26	4.55	6.04	4.70	4.41	4.14	3.81
SULFATE (MG/L)	219	25.86	0.34	3.70	9.52	4.49	2.71	1.64	0.85
NITRATE (MG/L)	219	10.21	0.22	1.83	4.42	2.53	1.36	0.82	0.37
CHLORIDE (MG/L)	219	10.33	0.04	0.56	1.73	0.55	0.29	0.16	0.07
AMMONIUM (MG/L)	219	10.00	0.02	0.49	1.55	0.50	0.25	0.12	0.02
SODIUM (MG/L)	219	7.70	0.01	0.38	1.32	0.42	0.19	0.07	0.03
POTASSIUM (MG/L)	219	8.78	0.00	0.12	0.24	0.07	0.04	0.02	0.01
CALCIUM (MG/L)	219	3.12	0.01	0.26	0.97	0.28	0.13	0.08	0.03
VOLUME OF PRECIPITATION (ML)	253	8163.10	0.10	1452.22	4602.68	2219.05	952.20	226.75	2.47
CATION/ANION BALANCE (PERCENT)	219	35.24	-35.82	-2.50	8.60	-0.12	-2.76	-5.92	-12.75

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK--CONTINUED

STATION NUMBER: 343560 STATION NAME: CLINTON STATION, NORTH CAROLINA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	226	154.40	2.60	27.31	73.37	35.10	20.90	12.90	6.47
PH (LAB)	231	7.36	3.76	4.70	5.96	4.93	4.58	4.30	3.96
SULFATE (MG/L)	221	24.77	0.10	2.92	7.80	3.62	1.98	1.20	0.58
NITRATE (MG/L)	221	10.56	0.02	1.54	5.27	1.89	1.06	0.53	0.24
CHLORIDE (MG/L)	221	14.60	0.07	0.93	3.14	1.03	0.44	0.23	0.10
AMMONIUM (MG/L)	221	2.53	0.01	0.29	1.13	0.38	0.15	0.07	0.02
SODIUM (MG/L)	221	6.89	0.02	0.68	3.40	0.68	0.28	0.14	0.05
POTASSIUM (MG/L)	221	0.49	0.00	0.06	0.21	0.06	0.03	0.02	0.01
CALCIUM (MG/L)	221	3.24	0.01	0.27	1.37	0.25	0.12	0.06	0.03
VOLUME OF PRECIPITATION (ML)	256	11519.90	0.30	1543.05	4822.83	2139.92	1027.05	263.25	3.22
CATION/ANION BALANCE (PERCENT)	221	58.92	-26.36	-2.06	11.17	5.75	0.78	-5.82	-13.10

STATION NUMBER: 344160 STATION NAME: FINLEY (A), NORTH CAROLINA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	215	193.10	2.10	28.33	76.24	34.80	21.80	12.80	5.56
PH (LAB)	223	7.36	3.37	4.66	6.01	4.88	4.56	4.27	3.89
SULFATE (MG/L)	213	26.16	0.10	3.09	9.03	3.43	2.06	1.31	0.60
NITRATE (MG/L)	213	10.70	0.02	1.68	4.24	2.32	1.23	0.66	0.29
CHLORIDE (MG/L)	213	10.85	0.04	0.60	2.10	0.53	0.28	0.16	0.08
AMMONIUM (MG/L)	213	4.72	0.02	0.39	1.11	0.51	0.23	0.11	0.03
SODIUM (MG/L)	213	22.93	0.01	0.57	1.70	0.44	0.18	0.09	0.03
POTASSIUM (MG/L)	213	1.22	0.00	0.05	0.18	0.05	0.03	0.02	0.01
CALCIUM (MG/L)	213	2.18	0.01	0.21	0.76	0.23	0.11	0.06	0.02
VOLUME OF PRECIPITATION (ML)	247	6583.40	0.10	1371.80	4380.30	1947.00	945.90	276.00	3.34
CATION/ANION BALANCE (PERCENT)	213	32.81	-31.52	-2.40	10.46	0.27	-2.70	-5.96	-15.01

STATION NUMBER: 361760 STATION NAME: DELAWARE, OHIO

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM OCT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	248	252.00	6.90	40.22	99.44	46.42	30.40	22.52	15.29
PH (LAB)	248	6.89	3.45	4.34	4.93	4.44	4.30	4.09	3.79
SULFATE (MG/L)	243	24.86	0.22	4.13	10.94	4.82	3.22	2.26	1.35
NITRATE (MG/L)	243	25.32	0.10	2.93	8.38	3.25	2.13	1.40	0.76
CHLORIDE (MG/L)	243	2.26	0.02	0.28	0.98	0.31	0.19	0.12	0.05
AMMONIUM (MG/L)	243	6.62	0.02	0.51	1.38	0.58	0.33	0.18	0.02
SODIUM (MG/L)	243	3.28	0.00	0.23	1.01	0.25	0.10	0.05	0.02
POTASSIUM (MG/L)	243	0.61	0.00	0.04	0.14	0.05	0.03	0.02	0.00
CALCIUM (MG/L)	243	7.13	0.02	0.49	1.83	0.55	0.25	0.14	0.04
VOLUME OF PRECIPITATION (ML)	260	8069.10	0.20	1276.36	3947.33	1843.12	816.90	286.27	11.62
CATION/ANION BALANCE (PERCENT)	243	44.95	-29.43	-2.22	7.15	0.66	-2.66	-5.49	-10.70

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK--CONTINUED

STATION NUMBER: 364900 STATION NAME: CALDWELL, OHIO

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM SEPT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	248	409.50	10.20	45.99	99.61	52.90	37.70	26.90	18.06
PH (LAB)	250	7.78	3.42	4.24	4.83	4.35	4.19	4.04	3.75
SULFATE (MG/L)	245	25.39	0.36	4.65	12.36	5.48	3.75	2.55	1.48
NITRATE (MG/L)	245	13.61	0.22	2.70	6.72	3.24	2.12	1.36	0.75
CHLORIDE (MG/L)	245	33.32	0.04	0.44	0.99	0.36	0.21	0.14	0.07
AMMONIUM (MG/L)	245	2.83	0.02	0.37	1.11	0.47	0.27	0.13	0.03
SODIUM (MG/L)	245	22.53	0.01	0.33	0.97	0.19	0.09	0.05	0.02
POTASSIUM (MG/L)	245	2.34	0.00	0.06	0.18	0.05	0.03	0.01	0.01
CALCIUM (MG/L)	245	6.58	0.02	0.47	1.84	0.56	0.26	0.12	0.06
VOLUME OF PRECIPITATION (ML)	259	6495.60	0.80	1415.29	3963.70	1985.30	1111.50	354.50	21.70
CATION/ANION BALANCE (PERCENT)	245	30.02	-24.14	-3.36	3.65	-0.48	-3.35	-6.39	-10.76

STATION NUMBER: 367160 STATION NAME: WOOSTER, OHIO

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM SEPT. 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	240	247.30	9.10	38.89	84.13	45.85	31.15	23.95	15.52
PH (LAB)	243	6.58	3.50	4.36	5.29	4.46	4.29	4.10	3.85
SULFATE (MG/L)	237	26.12	0.73	4.46	10.66	5.19	3.70	2.36	1.48
NITRATE (MG/L)	237	20.56	0.10	2.79	7.06	3.15	2.11	1.41	0.69
CHLORIDE (MG/L)	237	2.30	0.02	0.31	0.94	0.36	0.19	0.13	0.05
AMMONIUM (MG/L)	237	5.95	0.01	0.55	1.71	0.64	0.37	0.21	0.07
SODIUM (MG/L)	237	10.13	0.01	0.36	1.25	0.25	0.09	0.05	0.02
POTASSIUM (MG/L)	237	0.66	0.00	0.05	0.18	0.05	0.03	0.02	0.00
CALCIUM (MG/L)	237	5.99	0.02	0.46	1.45	0.53	0.26	0.14	0.05
VOLUME OF PRECIPITATION (ML)	260	10812.00	1.00	1156.68	3341.47	1756.00	828.60	246.25	7.75
CATION/ANION BALANCE (PERCENT)	237	29.20	-23.32	-3.14	8.20	-0.14	-2.98	-6.49	-13.34

STATION NUMBER: 392940 STATION NAME: KANE, PENNSYLVANIA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM JULY 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	184	181.40	8.40	40.81	80.37	49.40	35.15	25.70	13.85
PH (LAB)	185	7.22	3.47	4.24	4.82	4.35	4.17	3.99	3.81
SULFATE (MG/L)	182	36.35	0.68	3.99	7.60	4.97	3.45	2.20	1.06
NITRATE (MG/L)	182	8.21	0.02	2.39	5.60	2.91	2.05	1.40	0.65
CHLORIDE (MG/L)	182	6.26	0.02	0.36	1.11	0.28	0.17	0.11	0.05
AMMONIUM (MG/L)	182	1.37	0.02	0.32	0.77	0.45	0.27	0.13	0.02
SODIUM (MG/L)	182	36.95	0.01	0.42	0.82	0.14	0.07	0.04	0.02
POTASSIUM (MG/L)	182	0.76	0.00	0.04	0.09	0.04	0.02	0.01	0.01
CALCIUM (MG/L)	182	2.75	0.02	0.25	0.65	0.29	0.16	0.08	0.04
VOLUME OF PRECIPITATION (ML)	220	5836.30	2.90	1584.17	4005.08	2345.32	1352.25	559.57	47.86
CATION/ANION BALANCE (PERCENT)	181	18.14	-31.03	-2.96	7.21	-0.17	-3.55	-5.96	-10.89

TABLE 1.--STATISTICAL SUMMARY OF SELECTED PRECIPITATION CHEMISTRY DATA FROM SITES IN THE NATIONAL TRENDS NETWORK--CONTINUED

STATION NUMBER: 501860 STATION NAME: PARSONS, WEST VIRGINIA

SUMMARY OF SELECTED WATER QUALITY DATA COLLECTED AT WEEKLY INTERVALS FROM JULY 1978 TO JAN. 1984

	DESCRIPTIVE STATISTICS				PERCENT OF SAMPLES IN WHICH VALUES WERE LESS THAN OR EQUAL TO THOSE SHOWN				
PRECIPITATION CONSTITUENT	SAMPLE SIZE	MAXIMUM	MINIMUM	MEAN	95	75	MEDIAN 50	25	5
SPECIFIC CONDUCTANCE (UMHOS)	276	134.60	3.60	34.32	77.21	43.30	31.00	19.97	9.28
PH (LAB)	277	6.30	3.49	4.33	5.01	4.48	4.28	4.10	3.85
SULFATE (MG/L)	275	17.44	0.25	3.68	8.34	4.40	3.27	2.00	0.92
NITRATE (MG/L)	275	11.16	0.18	2.15	5.30	2.71	1.85	1.11	0.52
CHLORIDE (MG/L)	275	1.66	0.02	0.20	0.50	0.24	0.14	0.09	0.05
AMMONIUM (MG/L)	275	2.89	0.01	0.29	0.85	0.36	0.22	0.10	0.02
SODIUM (MG/L)	275	4.74	0.00	0.19	0.75	0.14	0.06	0.03	0.02
POTASSIUM (MG/L)	275	0.38	0.00	0.05	0.13	0.05	0.03	0.02	0.01
CALCIUM (MG/L)	275	3.29	0.01	0.35	1.14	0.40	0.22	0.13	0.05
VOLUME OF PRECIPITATION (ML)	283	7489.50	1.20	1620.88	4424.60	2345.10	1237.00	555.10	140.12
CATION/ANION BALANCE (PERCENT)	275	20.31	-20.58	-4.30	5.11	-0.78	-4.36	-8.07	-13.06

> from the collection buckets, which are also used to contain and seal the samples for shipment to the Central Analytical laboratory. Analysis of blanks from inverted buckets have indicated that the O-ring in the lid of the bucket collectors is a source of measureable ion concentrations in almost all cases. For typical wet deposition samples and for averages, the potential for O-ring contamination is much less than 10 percent for all ions except magnesium. For sulfate, the average contribution is 1.3 percent. The results from inverted bucket blank tests are probably a worst-case situation since NADP samples are not likely to be kept in an inverted position for long periods. Although inverted bucket blanks were not routinely taken prior to the summer of 1981, there is no evidence at the Central Analytical Laboratory that the O-ring effect has changed during the time frame of the program.

Based on these conclusions, the authors have elected not to examine magnesium.

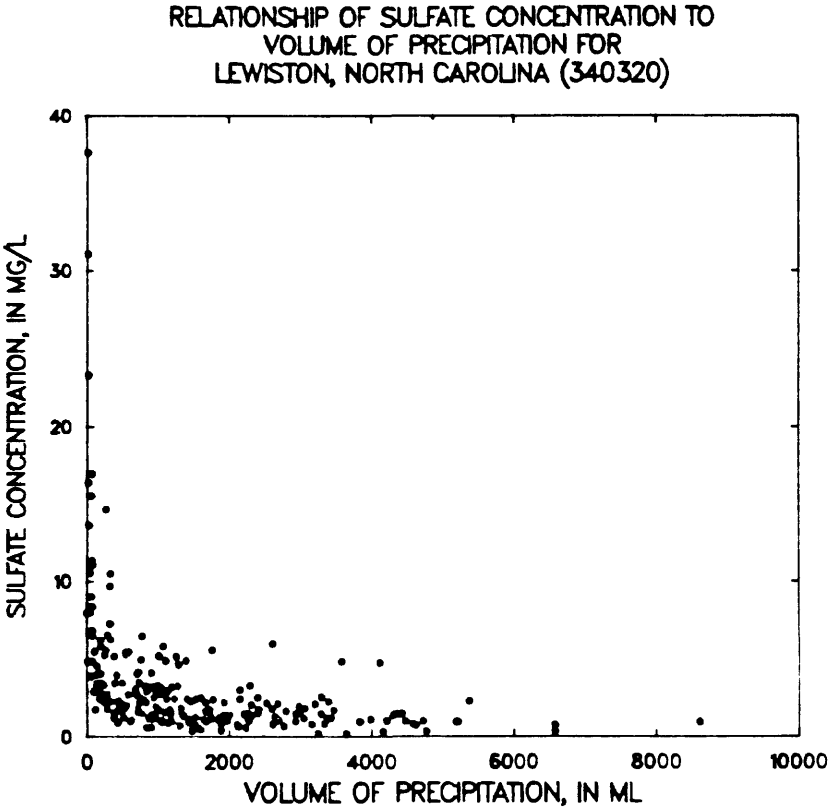
MODELING VARIATIONS IN CONCENTRATION

Regression analysis is usually applied to a sample from a population with the idea that inferences about the population parameters may be made based on analysis of the sample. The simplest form of regression deals with one dependent variable Y and one independent variable X. The first objective of regression analysis is to find the curve that best fits the data in such a way as to approximate the relationship between X and Y. For precipitation data, some relationship may exist between the concentration of a dissolved constituent and the volume of precipitation. Some example plots of concentration against volume are shown in figure 2. These examples, which were typical of the relationships found between the other dissolved constituents and precipitation volume, indicate a non-linear relationship with residuals which are heteroscedastic and positively skewed. The basic equation to fit a curve to the data would be:

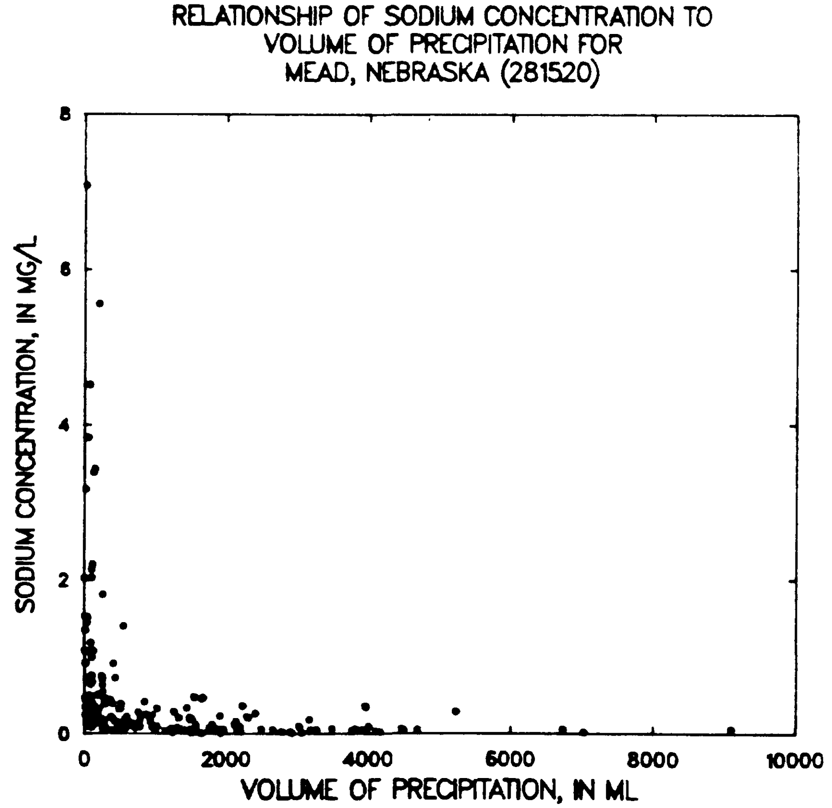
$$C = a + b * f(P) \qquad (1)$$

where C is concentration,
a is the intercept,
b is the slope, and
f(P) is some function of precipitation such as a hyperbolic function.

However, there are problems with the use of regression to predict these concentration values. In order to make inferences from the sample about the population, certain statistical assumptions must be made. Two of the assumptions are that the residuals are approximately normally distributed and homoscedastic with respect to the independent variable. When one or more of the underlying assumptions of regression analysis are not satisfied, the original data can be transformed to new data for which the standard regression assumptions are more closely satisfied. Log transformation of data can (1) stabilize the variance if the original data is heteroscedastic, (2) normalize the dependent variable if the distribution of the dependent variable is positively skewed, and (3) linearize the regression model when the relationship of the dependent variable with an independent variable suggests a model with a consistently increasing slope (Kleinbaum and Kupper, 1978). Figure 3 shows the same data used in figure 2 plotted on a logarithmic scale. The log transformed data exhibit the properties necessary for regression analysis. An equation to

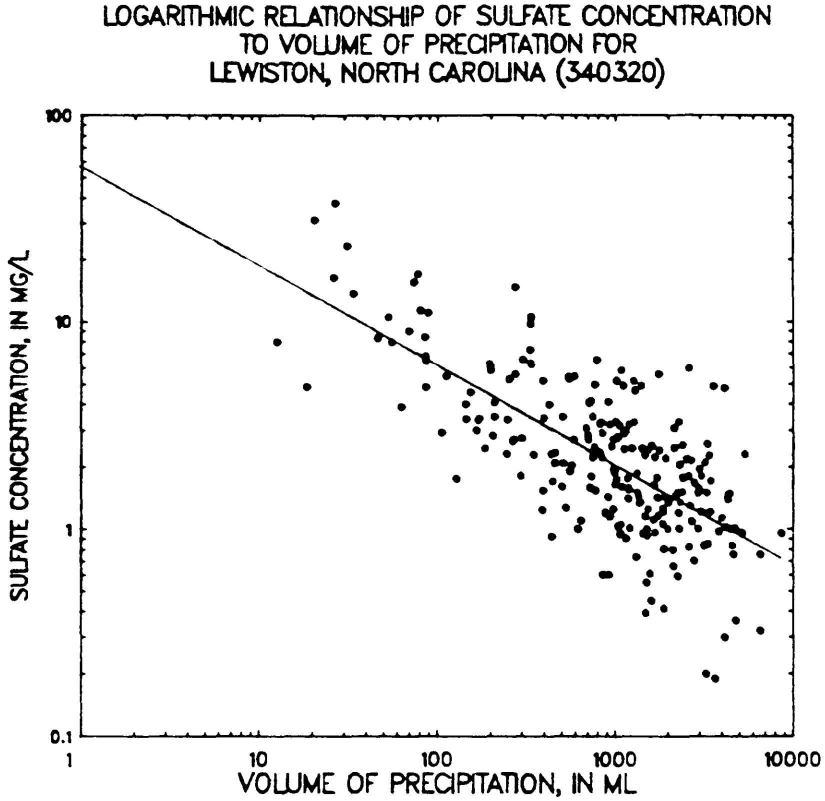


RELATIONSHIP OF SULFATE CONCENTRATION TO VOLUME OF PRECIPITATION FOR LEWISTON, NORTH CAROLINA (340320)



RELATIONSHIP OF SODIUM CONCENTRATION TO VOLUME OF PRECIPITATION FOR MEAD, NEBRASKA (281520)

Figure 2.--Examples of the relationship of dissolved concentrations to the volume of precipitation.



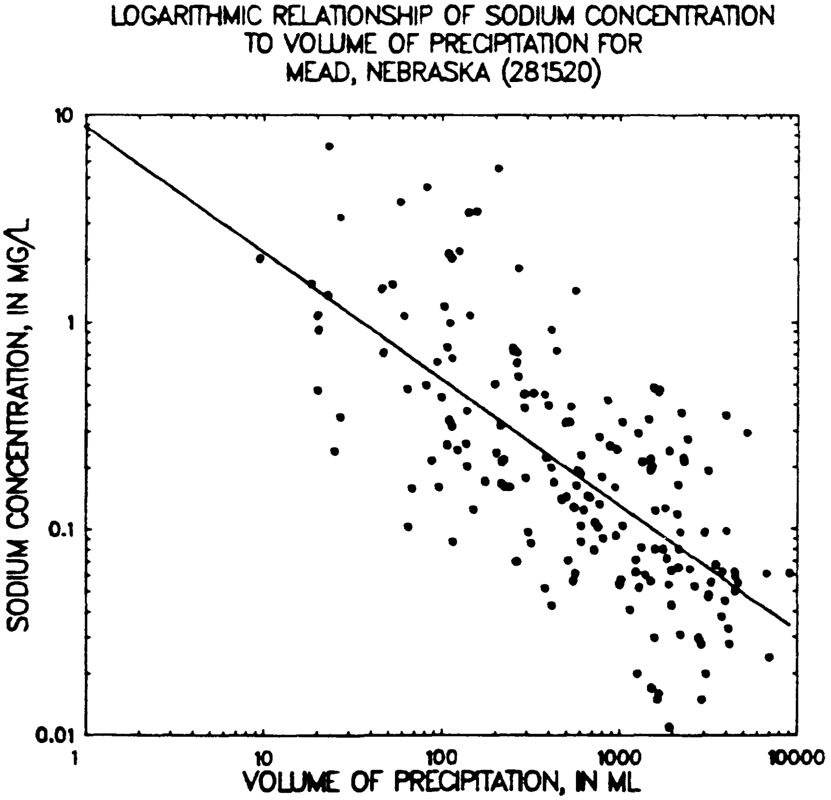


Figure 3.--Examples of the relationship of dissolved concentrations to the volume of precipitation on logarithmic scales.

express the relationship exhibited between the log of the concentration and the log of the volume is referred to here as the non-seasonal model:

$$\ln(C) = a + b * \ln(P) \qquad (2)$$

where C is concentration,
a is the intercept,
b is the slope,
P is the precipitation, and
ln is the natural log.

Residuals from a model, which are defined as the difference between the observed value and the predicted value, represent the error or amount of discrepancy between observed and predicted values from regression analysis. The regression model is appropriate for describing the relationship between ln(C) and ln(P) because the residuals appear to be approximately homoscedastic, symmetrically distributed and independent of ln(P). Boxplots showing the distribution of residuals for several constituents are shown in figures 4a-c. Tukey (1977, p. 27-49) and Velleman and Hoaglin (1981, p. 65-70) describe the drawing and interpretation of boxplots. The box covers the middle 50 percent of the data. Its top and bottom are the upper and lower quartiles, respectively. The bar in the middle of the box represents the median. All outliers which are 1.5 to 3 times the interquartile range beyond the quartiles are shown as an '*'. All outliers which are more than 3 times the interquartile range beyond the quartiles are shown as an '0'.

The boxplots of sulfate and nitrate residuals show an excess of negative outliers although the central portion of the distribution appears quite symmetrical. Nitrate is more extreme than sulfate in the tendency to have an excess of negative outliers. Ammonia residuals show a similar distribution to the sulfate residuals. Sodium residuals show a relatively symmetrical distribution with both positive and negative outliers. The distribution of residuals of the other ions most closely resemble the sodium distribution. Ignoring the outliers, all cases show relative symmetry, with a median generally close to the mean (the mean of the residuals is zero by definition) and centered between the upper and lower quartiles. But there are far more outliers than one would expect from a normal distribution. For a normal distribution, outliers designated with '*' should occur less than 1 percent of the time and those designated with '0' should occur less than 0.01 percent of the time. Outliers of both levels generally were between 2 and 4 percent of the residual values. The excessive numbers of outliers shows that the distribution of residuals is more "heavy-tailed" than a normal distribution. This fact is important in terms of selecting a trend testing procedure which will be powerful and resistant to the effects of these outliers. It is often argued that by selection of an appropriate transformation the data can be rendered approximately normal, and that after such transformations, the parametric tests (such as the test for a significant slope in a regression) can be used. Transformations may resolve problems of asymmetry, but not problems of heavy tails as exhibited in these data. In general, the non-parametric tests are more powerful (have a higher probability of detecting a significant trend if one exists) than their parametric alternatives when the data are heavy tailed (Bradley, 1968).

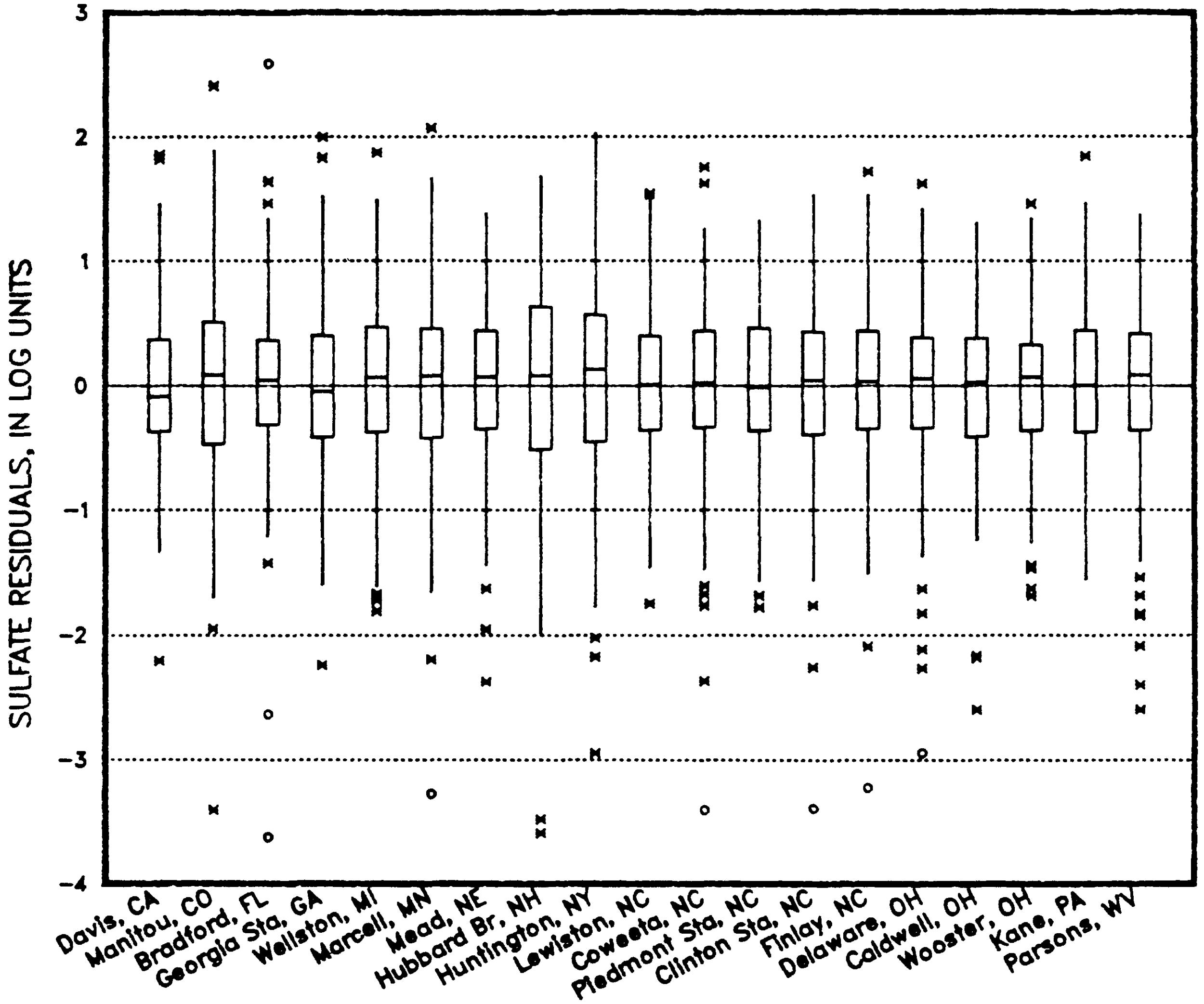


Figure 4a.--Examples of the distribution of sulfate residuals from the non-seasonal regression. The boxes cover the middle 50 percent of the data. The bar in the middle of the box is the median. Individual outliers are shown as '*' or '0', depending on their distance from the box. The vertical lines above and below the box extend to the most extreme values not designated as outliers.

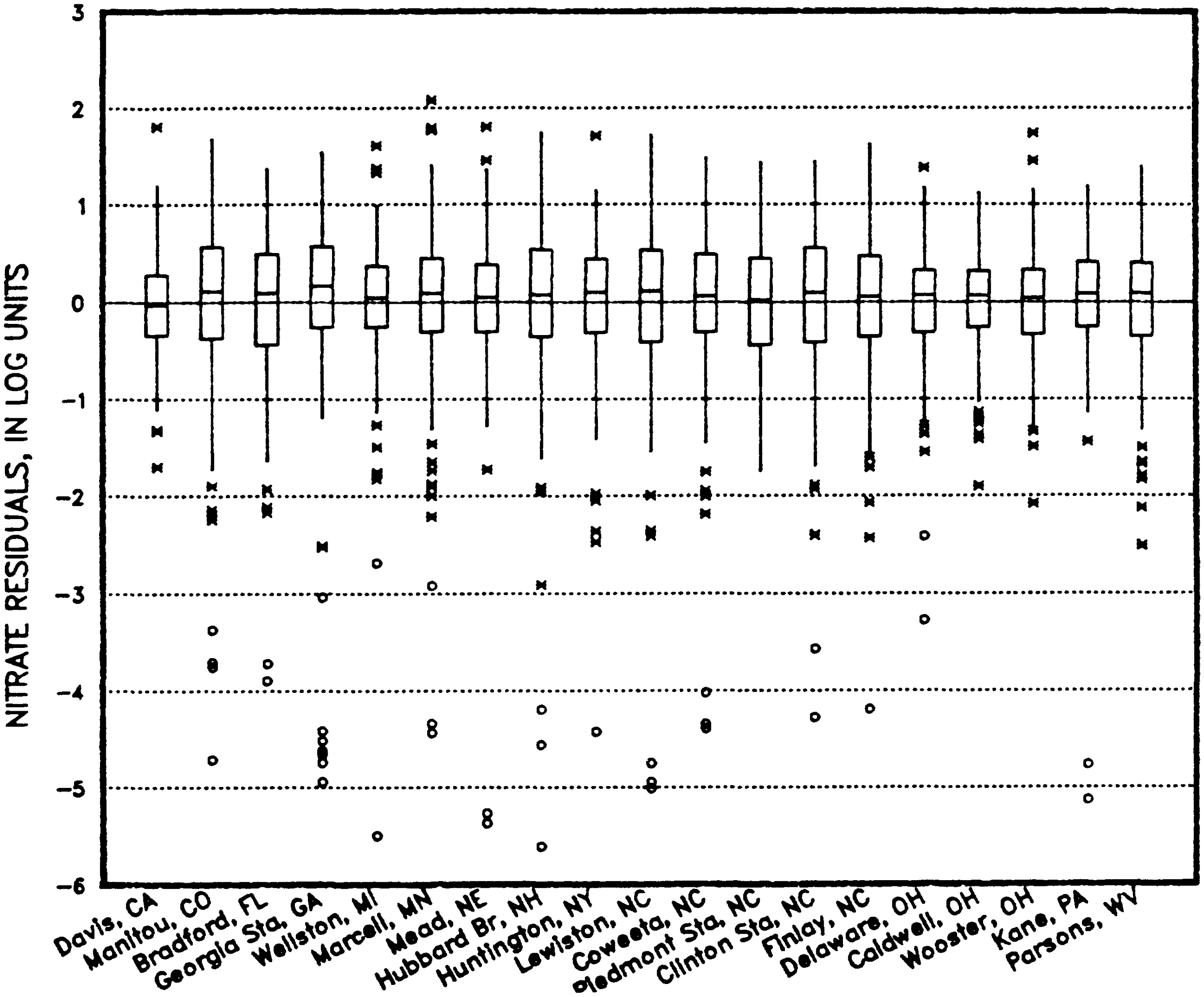


Figure 4b.--Examples of the distribution of nitrate residuals from the non-seasonal regression. The boxes cover the middle 50 percent of the data. The bar in the middle of the box is the median. Individual outliers are shown as '*' or '0', depending on their distance from the box. The vertical lines above and below the box extend to the most extreme values not designated as outliers.

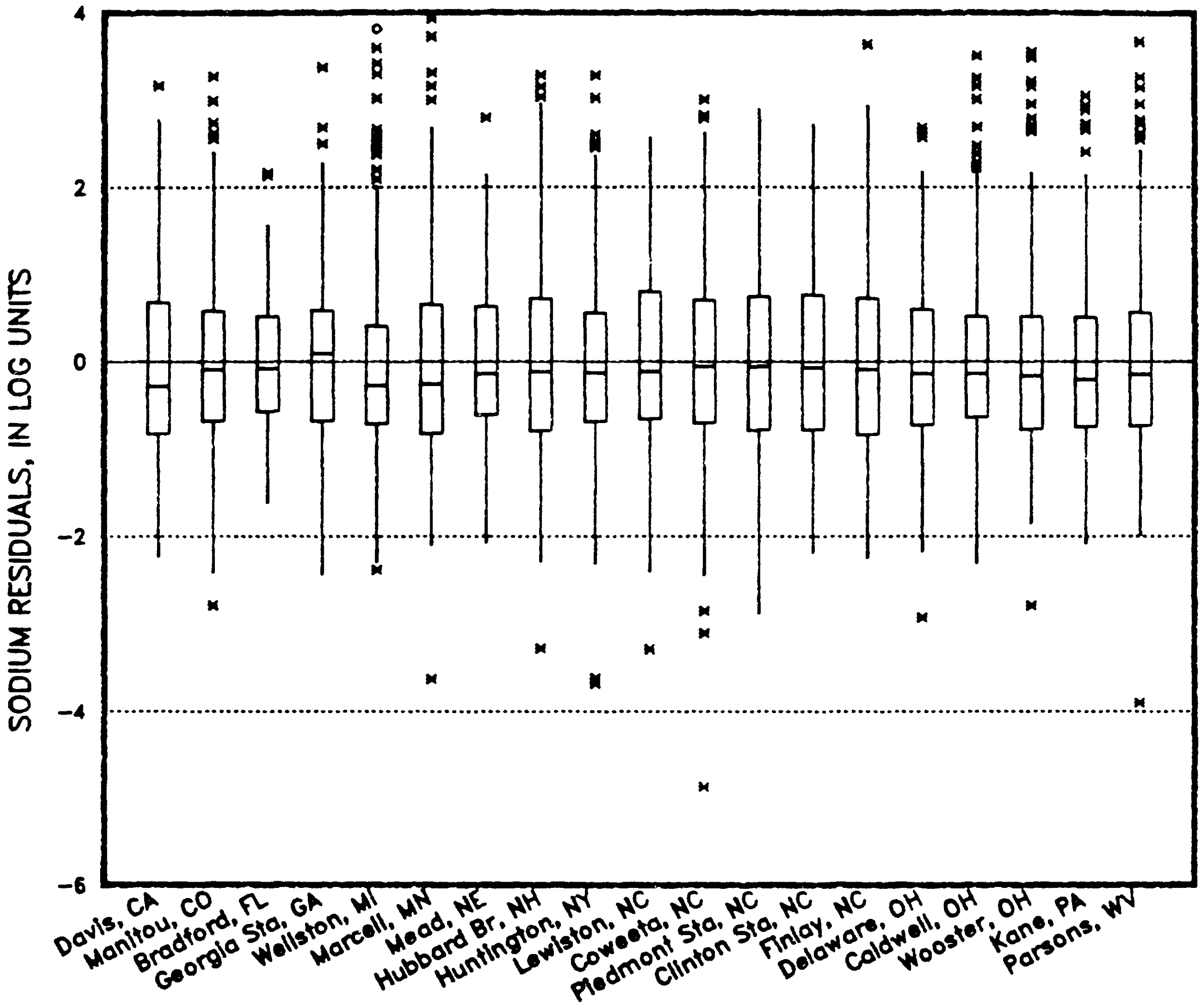


Figure 4c.--Examples of the distribution of sodium residuals from the non-seasonal regression. The boxes cover the middle 50 percent of the data. The bar in the middle of the box is the median. Individual outliers are shown as '*' or '0', depending on their distance from the box. The vertical lines above and below the box extend to the most extreme values not designated as outliers.

Residuals can indicate more than whether a model is appropriate or inappropriate. A pattern in the residuals can indicate a need to add parameters to the model. Examination of the residuals from equation 2, the non-seasonal model, shows evidence of a seasonal pattern at most stations and for most constituents (figs. 5a,b). A seasonal term added to the non-seasonal model to explain a fluctuating mean concentration during the period of a year that resembles a sine wave pattern, results in the following seasonal model:

$$\ln(C) = a + b * \ln(P) + c * \sin(2\pi T) + d * \cos(2\pi T) \qquad (3)$$

where C is the concentration,
P is the volume of precipitation,
ln is the natural log,
a is the intercept,
b is the slope,
c and d are regression coefficients, and
T is time, in years.

Residuals from the seasonal model that lay more than three standard deviations from the mean of the residuals (zero) were tabled in the Appendix at the end of the report. These values could be considered highly suspect, on the basis that they are extreme outliers in the regression analysis. Although they are suspect values, they are not necessarily invalid values, so they were not deleted from the regression analyses or trend analyses.

After fitting both the non-seasonal model and the seasonal model to the data for each station and constituent, there are two hypothesis tests (both at $\alpha = 0.2$) that are made. The first is to determine if log concentration is dependent on log volume (that is, to reject the null hypothesis that $b = 0$ in the equation). The second is to determine if season has a significant influence. This is done by an F-test comparing the seasonal model to the non-seasonal model if the nonseasonal model was found to be significant in the previous test, or by an overall F-test for the seasonal model if the non-seasonal model was not found to be significant. The fitted coefficients and other descriptors of these models are given in tables 2a-h for most constituents. The table entries give the attained significance levels (p-values) for these two tests and five of the characteristics of the seasonal model if it is significant, or the non-seasonal model if the seasonal model is not significant.

The slope is an indicator of how concentrations vary with volume. The non-seasonal and seasonal models are actually power functions with respect to precipitation volume. The non-seasonal model, $\ln(C) = a + b*\ln(P)$, translates to $C = e^{a} * P^{b}$. The seasonal model, $\ln(C) = a + b*\ln(P) + c*\sin(2\pi T) + d*\cos(2\pi T)$, translates to $C = e^{(a + c*\sin(2\pi T) + d*\cos(2\pi T))} * P^{b}$. The slope is also an indicator of how deposition (denoted here as D) varies with volume. Given that $D \propto C * P$, and that $C \propto P^{b}$, then it follows that $D \propto P^{b} * P$ or, $D \propto P^{(b+1)}$.

For all of the variables considered, the slopes (equivalent to the exponent in the power function relating concentration to volume) were negative, indicating lower values of concentration are associated with higher values of precipitation volume. The steepest negative slopes are those for calcium, in the range -0.45 to -0.72. Flatter slopes were found for sulfate, nitrate, ammonia, and specific conductance (from -0.10 to -0.47). An intermediate group consisted

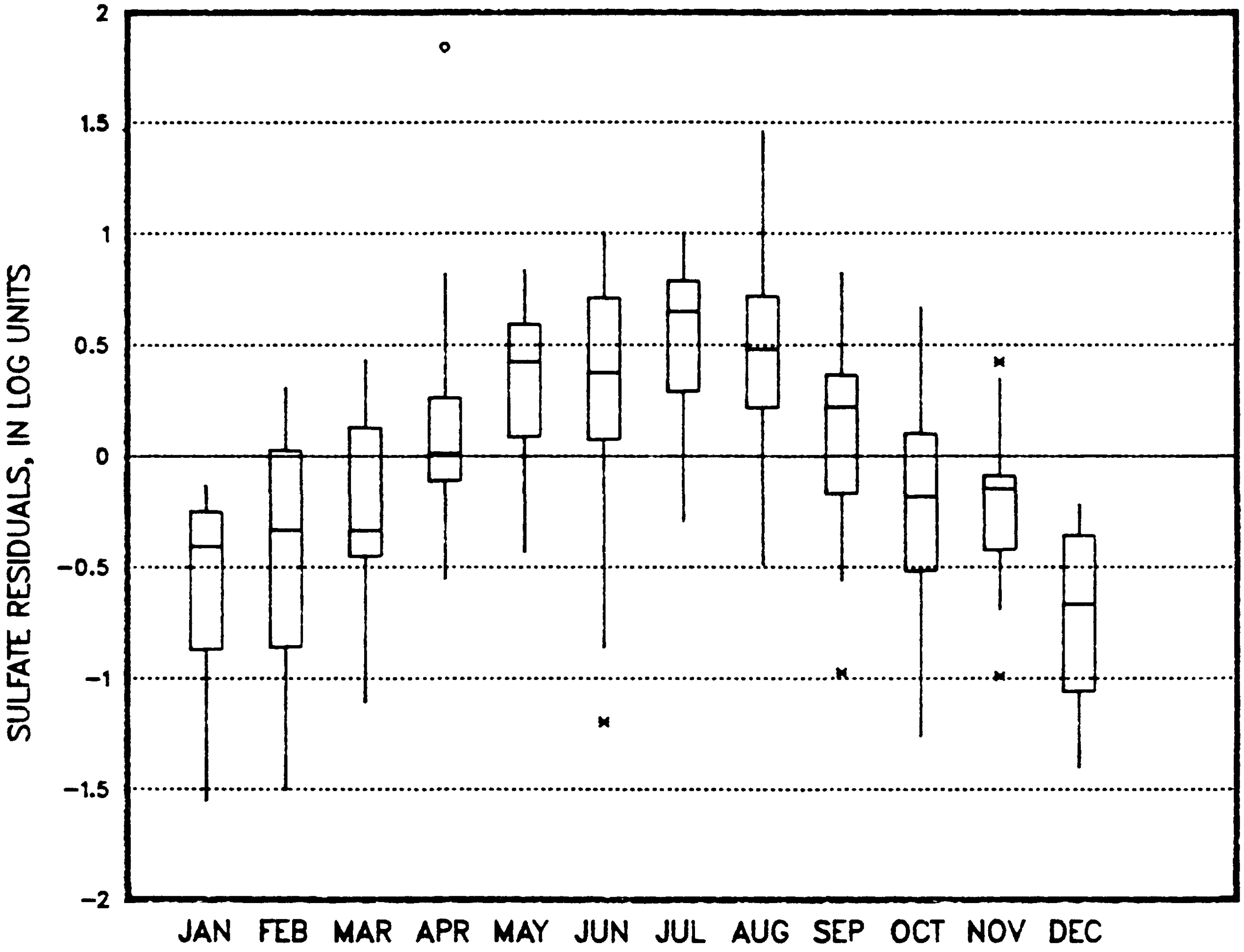


Figure 5a.--Examples of the seasonal variation in the distribution of non-seasonal sulfate residuals by month at Kane, PA (392940). The boxes cover the middle 50 percent of the data. The bar in the middle of the box is the median. Individual outliers are shown as '*' or '0', depending on their distance from the box. The vertical lines above and below the box extend to the most extreme values not designated as outliers.

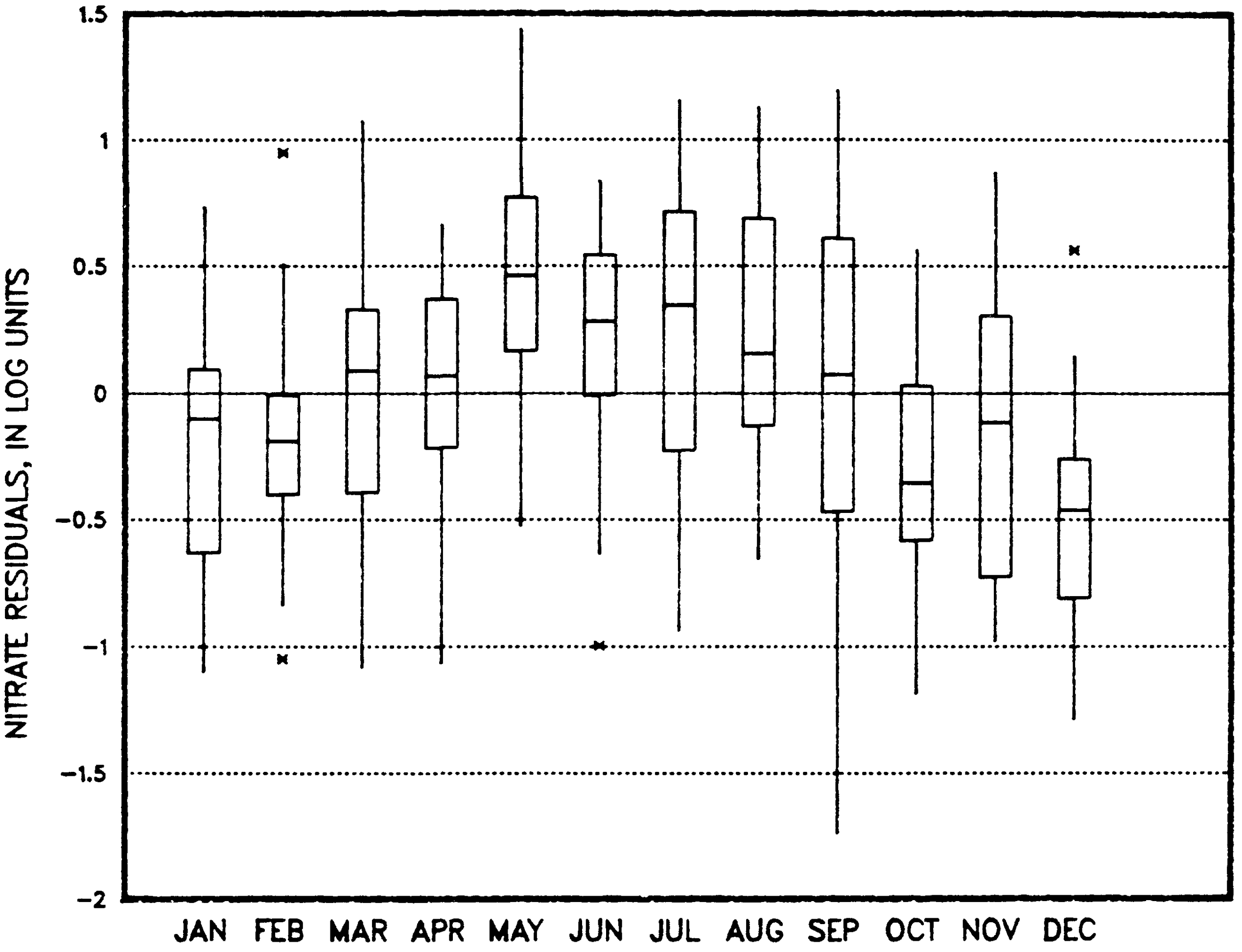


Figure 5b.--Examples of the seasonal variation in the distribution of non-seasonal nitrate residuals by month at Piedmont Station, NC (343460). The boxes cover the middle 50 percent of the data. The bar in the middle of the box is the median. Individual outliers are shown as '*' or '0', depending on their distance from the box. The vertical lines above and below the box extend to the most extreme values not designated as outliers.

Table 2a.--Characteristics of statistical models for sulfate data collected with the National Trends Network during 1978-83

Slope - Slope of the regression line from significant model.
Standard error (percent) - The ratio of standard deviation of the concentration to the mean concentration.
R^2 (percent) - The percent of the total variation in the data that is explained by the model.
Peak day - The day of the year when the sine wave component of a seasonal model is at its peak.
Amplitude - The amount of variation in the data associated with seasonality, expressed as half of the distance from peak to trough of the sine wave component of the seasonal model.
p - The significance level.

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.40	69.4	48.8	250	0.18	0.000	0.094	
062120	Manitou, Colorado	-.34	73.7	37.5	168	.85	.012	.000	
100360	Bradford Forest, Florida	-.39	75.5	37.8	--	.0	.000	.500	A
114140	Georgia Station, Georgia	-.40	65.9	40.1	150	.23	.000	.001	
235340	Wellston, Michigan	-.27	65.7	32.3	163	.48	.000	.000	
241660	Marcell, Minnesota	-.23	83.1	14.0	186	.29	.000	.001	
281520	Mead, Nebraska	-.27	68.1	33.1	31	.14	.000	.140	
300240	Hubbard Brook, New Hampshire	-.19	79.6	31.8	177	.62	.000	.000	
332020	Huntington, New York	-.21	73.6	36.7	168	.66	.000	.000	
340320	Lewiston, North Carolina	-.47	63.7	54.8	146	.24	.000	.000	
342500	Coweeta, North Carolina	-.23	65.5	34.3	154	.33	.000	.000	
343460	Piedmont Station, North Carolina	-.35	56.8	49.3	146	.38	.000	.000	
343560	Clinton Station, North Carolina	-.39	70.9	45.0	161	.28	.000	.000	
344160	Finley (A), North Carolina	-.30	69.1	47.2	150	.35	.000	.000	
361760	Delaware, Ohio	-.19	56.7	33.4	169	.45	.000	.000	
364900	Caldwell, Ohio	-.24	51.7	40.0	166	.47	.000	.000	
367160	Wooster, Ohio	-.23	50.9	36.0	165	.40	.000	.000	
392940	Kane, Pennsylvania	-.27	49.5	46.9	167	.57	.000	.000	
501860	Parsons, West Virginia	-.29	59.5	36.7	164	.53	.000	.000	

See footnote at end of table.

Table 2b.--Characteristics of statistical models for nitrate data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.39	57.0	54.5	311	0.34	0.000	0.000	
062120	Manitou, Colorado	-.21	113.6	13.7	143	.58	.209	.000	B
100360	Bradford Forest, Florida	-.44	83.7	42.2	164	.43	.000	.000	
114140	Georgia Station, Georgia	-.29	133.0	13.3	143	.29	.000	.012	
235340	Wellston, Michigan	-.24	79.0	17.1	56	.12	.000	.000	
241660	Marcell, Minnesota	-.25	95.5	14.7	--	.0	.000	.550	A
281520	Mead, Nebraska	-.29	92.0	22.7	--	.0	.000	.307	A
300240	Hubbard Brook, New Hampshire	-.26	104.0	12.1	267	.15	.000	.149	
332020	Huntington, New York	-.30	78.5	22.3	86	.12	.000	.189	
340320	Lewiston, North Carolina	-.43	108.4	33.0	141	.34	.000	.000	
342500	Coweeta, North Carolina	-.29	88.9	28.9	162	.29	.000	.000	
343460	Piedmont Station, North Carolina	-.39	58.2	51.9	142	.35	.000	.000	
343560	Clinton Station, North Carolina	-.43	87.0	43.0	156	.39	.000	.000	
344160	Finley (A), North Carolina	-.46	76.1	46.2	142	.34	.000	.000	
361760	Delaware, Ohio	-.35	59.1	45.1	146	.30	.000	.000	
364900	Caldwell, Ohio	-.40	50.3	50.9	130	.22	.000	.000	
367160	Wooster, Ohio	-.39	55.4	49.3	137	.33	.000	.000	
392940	Kane, Pennsylvania	-.26	82.2	12.8	--	.0	.000	.023	A
501860	Parsons, West Virginia	-.42	61.4	34.4	143	.28	.000	.000	

See footnote at end of table.

Table 2c.--Characteristics of statistical models for calcium data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.61	76.4	67.5	181	0.43	0.000	0.022	
062120	Manitou, Colorado	-.53	87.3	42.1	164	.76	.000	.000	
100360	Bradford Forest, Florida	-.60	73.9	59.5	148	.24	.000	.003	
114140	Georgia Station, Georgia	-.64	86.0	51.2	116	.30	.000	.000	
235340	Wellston, Michigan	-.49	92.3	36.7	164	.36	.000	.000	
241660	Marcell, Minnesota	-.45	83.0	36.4	187	.47	.000	.000	
281520	Mead, Nebraska	-.48	92.0	43.1	159	.46	.000	.000	
300240	Hubbard Brook, New Hampshire	-.59	92.8	46.6	157	.44	.000	.000	
332020	Huntington, New York	-.64	76.5	61.1	157	.66	.000	.000	
340320	Lewiston, North Carolina	-.72	70.3	69.5	145	.23	.000	.000	
342500	Coweeta, North Carolina	-.59	85.3	58.2	121	.38	.000	.000	
343460	Piedmont Station, North Carolina	-.61	73.2	61.8	120	.43	.000	.000	
343560	Clinton Station, North Carolina	-.64	65.9	70.8	137	.38	.000	.000	
344160	Finley (A), North Carolina	-.67	71.7	65.3	133	.41	.000	.000	
361760	Delaware, Ohio	-.60	85.5	56.5	148	.50	.000	.000	
364900	Caldwell, Ohio	-.65	97.5	58.8	156	.45	.000	.000	
367160	Wooster, Ohio	-.63	75.5	58.9	153	.52	.000	.000	
392940	Kane, Pennsylvania	-.57	78.5	44.6	154	.37	.000	.000	
501860	Parsons, West Virginia	-.62	78.5	42.8	118	.28	.000	.000	

See footnote at end of table.

Table 2d.--Characteristics of statistical models for chloride data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.28	110.6	17.6	--	0.0	0.000	0.803	A
062120	Manitou, Colorado	-.54	87.9	42.5	164	.47	.000	.000	
100360	Bradford Forest, Florida	-.51	85.2	48.3	357	.20	.000	.038	
114140	Georgia Station, Georgia	-.37	93.8	23.6	358	.18	.000	.070	
235340	Wellston, Michigan	-.39	73.4	42.3	349	.25	.000	.002	
241660	Marcell, Minnesota	-.49	84.6	44.3	232	.28	.000	.006	
281520	Mead, Nebraska	-.51	76.8	54.5	--	.0	.000	.498	A
300240	Hubbard Brook, New Hampshire	-.36	122.7	17.1	340	.16	.000	.196	
332020	Huntington, New York	-.44	84.1	33.2	--	.0	.000	.498	A
340320	Lewiston, North Carolina	-.39	116.1	29.0	350	.50	.000	.000	
342500	Coweeta, North Carolina	-.25	104.5	13.5	--	.0	.000	.280	A
343460	Piedmont Station, North Carolina	-.41	98.0	33.0	2	.38	.000	.000	
343560	Clinton Station, North Carolina	-.32	111.4	28.6	354	.47	.000	.000	
344160	Finley (A), North Carolina	-.37	104.4	26.4	354	.33	.000	.000	
361760	Delaware, Ohio	-.40	67.9	48.5	363	.20	.000	.003	
364900	Caldwell, Ohio	-.43	68.3	46.1	12	.22	.000	.001	
367160	Wooster, Ohio	-.47	72.5	49.3	5	.19	.000	.014	
392940	Kane, Pennsylvania	-.47	98.6	26.9	--	.0	.000	.869	A
501860	Parsons, West Virginia	-.45	75.3	30.1	--	.0	.000	.429	A

See footnote at end of table.

Table 2e.--Characteristics of statistical models for ammonia data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.23	94.6	22.8	329	0.66	0.002	0.000	
062120	Manitou, Colorado	-.29	146.6	26.5	151	1.07	.931	.000	B
100360	Bradford Forest, Florida	-.41	204.4	16.7	102	.35	.000	.000	
114140	Georgia Station, Georgia	-.17	198.0	7.7	124	.42	.038	.000	
235340	Wellston, Michigan	-.11	167.2	5.5	159	.39	.428	.015	B
241660	Marcell, Minnesota	-.21	229.5	7.6	173	.58	.305	.001	B
281520	Mead, Nebraska	-.24	120.8	16.9	52	.29	.000	.012	
300240	Hubbard Brook, New Hampshire	-.14	185.0	14.9	170	.70	.073	.000	
332020	Huntington, New York	-.34	139.4	27.1	154	.75	.000	.000	
340320	Lewiston, North Carolina	-.40	183.7	23.2	152	.53	.000	.000	
342500	Coweeta, North Carolina	-.10	178.1	10.1	136	.50	.030	.000	
343460	Piedmont Station, North Carolina	-.42	108.8	26.8	115	.52	.000	.000	
343560	Clinton Station, North Carolina	-.43	159.5	26.5	159	.46	.000	.000	
344160	Finley (A), North Carolina	-.43	123.3	26.8	148	.32	.000	.002	
361760	Delaware, Ohio	-.29	144.8	20.2	153	.61	.000	.000	
364900	Caldwell, Ohio	-.34	127.0	19.9	148	.49	.000	.000	
367160	Wooster, Ohio	-.38	106.9	28.5	154	.55	.000	.000	
392940	Kane, Pennsylvania	-.25	147.1	9.2	160	.38	.007	.007	
501860	Parsons, West Virginia	-.40	160.4	--	155	.61	.000	.000	

See footnote at end of table.

Table 2f.--Characteristics of statistical models for sodium data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.31	156.2	14.8	--	0.0	0.000	0.980	A
062120	Manitou, Colorado	-.61	145.4	33.2	164	.43	.000	.019	
100360	Bradford Forest, Florida	-.50	89.9	45.7	363	.24	.000	.014	
114140	Georgia Station, Georgia	-.50	124.9	27.1	353	.21	.000	.093	
235340	Wellston, Michigan	-.64	182.0	30.2	--	.0	.000	.869	A
241660	Marcell, Minnesota	-.55	177.1	26.9	--	.0	.000	.373	A
281520	Mead, Nebraska	-.61	119.1	48.3	--	.0	.000	.878	A
300240	Hubbard Brook, New Hampshire	-.44	173.6	15.3	--	.0	.000	.795	A
332020	Huntington, New York	-.47	147.6	22.0	133	.22	.000	.105	
340320	Lewiston, North Carolina	-.46	135.0	26.8	360	.36	.000	.001	
342500	Coweeta, North Carolina	-.33	162.3	13.9	--	.0	.000	.861	A
343460	Piedmont Station, North Carolina	-.41	140.3	22.9	16	.36	.000	.003	
343560	Clinton Station, North Carolina	-.39	136.3	26.4	352	.39	.000	.001	
344160	Finley (A), North Carolina	-.45	156.8	21.3	9	.23	.000	.124	
361760	Delaware, Ohio	-.48	125.7	32.1	--	.0	.000	.764	A
364900	Caldwell, Ohio	-.53	138.4	27.8	--	.0	.000	.980	A
367160	Wooster, Ohio	-.58	150.1	31.3	--	.0	.000	.698	A
392940	Kane, Pennsylvania	-.56	130.4	25.5	--	.0	.000	.363	A
501860	Parsons, West Virginia	-.53	144.8	18.5	173	.19	.000	.115	

See footnote at end of table.

Table 2g.--Characteristics of statistical models for specific-conductance data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (percent)	R^2 (percent)	Peak day	Amplitude (log units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.39	53.6	66.6	306	0.02	0.000	0.015	
062120	Manitou, Colorado	-.27	50.8	38.4	164	.58	.000	.000	
100360	Bradford Forest, Florida	-.41	62.1	49.7	189	.23	.000	.001	
114140	Georgia Station, Georgia	-.34	59.7	39.7	157	.23	.000	.000	
235340	Wellston, Michigan	-.18	53.4	20.5	132	.21	.000	.000	
241660	Marcell, Minnesota	-.19	55.2	22.1	--	.0	.000	.844	A
281520	Mead, Nebraska	-.32	49.6	52.4	125	.12	.000	.052	
300240	Hubbard Brook, New Hampshire	-.17	70.4	17.5	136	.05	.000	.000	
332020	Huntington, New York	-.20	66.5	21.0	155	.32	.000	.000	
340320	Lewiston, North Carolina	-.33	62.0	42.6	126	.17	.000	.005	
342500	Coweeta, North Carolina	-.20	57.1	35.6	161	.30	.000	.000	
343460	Piedmont Station, North Carolina	-.30	59.5	41.3	152	.35	.000	.000	
343560	Clinton Station, North Carolina	-.21	58.9	43.4	163	.22	.000	.000	
344160	Finley (A), North Carolina	-.36	65.5	41.5	148	.27	.000	.000	
361760	Delaware, Ohio	-.19	49.5	35.3	156	.35	.000	.000	
364900	Caldwell, Ohio	-.24	43.0	42.9	153	.33	.000	.000	
367160	Wooster, Ohio	-.17	45.6	31.0	155	.34	.000	.000	
392940	Kane, Pennsylvania	-.18	46.6	32.7	160	.38	.000	.000	
501860	Parsons, West Virginia	-.27	54.4	35.1	161	.46	.000	.000	

See footnote at end of table.

Table 2h.--Characteristics of statistical models for pH data collected with the National Trends Network during 1978-83

Station	Station name	Slope	Standard error (ph units)	R^2 (percent)	Peak day	Amplitude (pH units)	p (non-seasonal)	p (seasonal)	Remarks 1/
058840	Davis, California	-0.09	0.57	9.9	--	0.0	0.000	0.353	A
062120	Manitou, Colorado	-.21	.60	28.4	60	.15	.000	.077	
100360	Bradford Forest, Florida	-.05	.60	4.0	54	.14	.092	.098	
114140	Georgia Station, Georgia	-.09	.68	2.8	--	.0	.015	.613	A
235340	Wellston, Michigan	-.14	.57	11.5	249	.12	.000	.144	
241660	Marcell, Minnesota	-.21	.54	30.8	179	.43	.000	.000	
281520	Mead, Nebraska	-.21	.73	19.2	126	.22	.000	.019	
300240	Hubbard Brook, New Hampshire	-.10	.51	6.0	--	.0	.000	.246	A
332020	Huntington, New York	--	.47	--	--	.0	.608	.242	C
340320	Lewiston, North Carolina	-.09	.58	9.5	274	.11	.000	.120	
342500	Coweeta, North Carolina	-.12	.46	16.2	355	.15	.000	.002	
343460	Piedmont Station, North Carolina	-.10	.62	10.2	348	.19	.000	.007	
343560	Clinton Station, North Carolina	-.08	.59	5.1	--	.0	.001	.684	A
344160	Finley (A), North Carolina	-.08	.58	7.8	332	.13	.001	.058	
361760	Delaware, Ohio	-.07	.44	12.2	352	.14	.000	.003	
364900	Caldwell, Ohio	-.07	.42	9.6	334	.12	.000	.007	
367160	Wooster, Ohio	-.11	.41	17.1	313	.12	.000	.009	
392940	Kane, Pennsylvania	-.09	.42	15.4	5	.17	.000	.001	
501860	Parsons, West Virginia	-.02	.34	19.9	344	.23	.009	.000	

1/

Remark	Explanation
blank	The seasonal model is significant when compared to the non-seasonal model. All components in the table are from the regression of the seasonal model against the data. The p (seasonal) value is from the F test between the seasonal and non-seasonal models.
A	The non-seasonal model is significant, but the seasonal model is not significant when compared to the non-seasonal model. All components in the table are from the regression of the non-seasonal model against the data. The p (seasonal) value is from the F test between the non-seasonal model and the seasonal model.
B	The seasonal model is significant, but the non-seasonal model is not significant. All components in the table are from the regression of the seasonal model against the data. The p (seasonal) value is from the F test between the seasonal model and no model.
C	Neither the seasonal model nor the non-seasonal model is significant. The p (seasonal) value is from the F test between the seasonal model and no model. The sum of squares, corrected total divided by the degrees of freedom, is used in the calculation of the standard error instead of the mean square, error.

of sodium and chloride with slopes of -0.25 to -0.61. The equation for pH can be reexpressed as a relationship between hydrogen ion concentration ([H+]) and precipitation volume:

$$[H+] \propto p^{B}$$

where B is -2.303 * b, where b is the slope reported in the table. These B values range from a low of 0.16 to a high of 0.48. Thus hydrogen ion concentration increases with precipitation volume.

No geographical patterns were evident in the slopes of the regressions which suggests that there are no geographical patterns in the relationship of concentration of dissolved constituents to volume of precipitation or rate of deposition to rate of precipitation. The standard errors of the slopes from the regressions generally were smaller than the difference between the largest slope and the smallest slope for each constituent. This indicates that, even though a geographic pattern was not in evidence, some significant differences existed between the slopes of some stations and a single regression model for all of the stations would be inappropriate.

The standard error is an indicator of the amount of variation in the data that is not explained by components of the models. The standard error in percent is the ratio (multiplied by 100) of the conditional standard deviation of the concentration to the conditional mean concentration. The conditioning is on the particular volume of precipitation and time of year, assuming that the full seasonal model is used. If the non-seasonal is used then the conditioning is only on the volume of precipitation. If no model is used, then it is the ratio of the unconditional standard deviation to the unconditional mean. In any of these cases, the calculation is based on the assumption that the residuals from the regression (or in the latter case, the log concentrations) are normally distributed. Sodium and ammonia show the largest standard errors for all stations, but all of the parameters have standard errors that generally exceed 50 percent. These high standard errors indicate that in any given week the regressions described in the tables would provide rather poor predictions of concentrations, given the precipitation volume and time of year. The data are inherently very noisy and the regressions remove only a modest fraction of the noise. However, most of the regression models are highly significant: indicating that the relationship between concentration and volume and time of year are much stronger than might occur by chance alone.

R-square (Kleinbaum and Kupper, 1978) is a measure of the strength of the relationship between the dependent variable and the set of explanatory variables but is not a measure of the appropriateness of the model. The highest R-square values were consistently for the calcium regressions and the lowest were consistently the pH regressions. The R-square values for all parameters at each station showed no evidence of a single station or geographical location that had a much better or much worse relationship of the dependent to the independent variables. The R-square is the fraction of the variance in the data which is removed by the regression. Thus, if there has been a change in concentration not related to volume or time of year (presumably it would be related to a change in emissions, natural sources, or transport patterns), the potential for being able to identify the change (distinguish it from noise) would be enhanced by removing the influence of volume and time of year. The ratio of the standard deviation of the residuals from a regression to the standard deviation of the raw data is $\sqrt{(1-R^2)}$. Thus, the use of any statistically significant regression, no matter how low the R-square, will decrease the amount of apparent noise in the data and make the changes that are of interest more noticeable, either to the eye or to a formal statistical test (both of which are considered below).

The seasonal term in the seasonal model can be converted from:

$$y = c * \sin(2\pi T) + d * \cos(2\pi T)$$

to:

$$y = f * \sin(2\pi(T + T_0))$$

where f is the amplitude and
T_0 is the phase shift of the seasonal term.
The amplitude and phase shift can be calculated from the following equations:

$$f = \sqrt{(c^2 + d^2)}$$

$$T_0 = (1/2\pi) * \arctan(c/d)$$

The amplitude represents the amount of the variation in the constituent concentration, in log units, that is due to seasonal influences. The phase shift can be converted to a peak day using the following method:

$$T_{max} = 1/4 - T_0.$$

$$\text{if } T_{max} < 0, \text{ let } T_{max} = T_{max} + 1$$

$$\text{if } T_{max} > 1, \text{ let } T_{max} = T_{max} - 1$$

where T_{max} is the peak of the sine wave, in years.
To convert from years (T_{max}) to day of the year (D_{max}):

$$D_{max} = T_{max} * 365$$

The peak day represents the day of the year when the sine wave component of the seasonal model is at its peak and the greatest positive contribution to the concentration of a constituent from seasonal variation occurs. Approximately 60 percent of the peak days for volume-adjusted concentrations of constituents and stations where significant seasonal models were found were between 100 and 200, which falls in mid-April to mid-July. The peak days for sulfate and nitrate, for example, were almost exclusively in the summer months. Chloride is the main constituent that has peak days outside of the "summer" peak days. At 11 out of the 13 stations where significant seasonality was found for chloride, the peak volume-adjusted concentrations occur between day 340 and day 20, which falls in December and January. Sodium had a weaker seasonal component than chloride, but at six of the nine stations where significant seasonality was found, the peak days for sodium were also in the December to January period. Marine environments are a common source of sodium and chloride in the atmosphere. So, winter peak days for these two constituents could be due to the activity of cyclonic storms that are most vigorous and numerous during the winter months and capable of pumping moist air from the Gulf of Mexico or the Atlantic Ocean far inland into precipitation systems.

The peak days for pH (lowest acidity) also occur in the winter months. Since pH is the -log of the hydrogen ion concentration, the peak for the hydrogen ion concentration would occur opposite the peak for pH, or during the summer months when most of the other constituents peak.

Anomalies exist in the seasonal components of the regressions. For example, the peak day for sulfate concentrations at Mead, Nebraska, is considerably different from the peak days for sulfate concentrations at the other stations. But the level of significance (p value) is relatively high and the amplitude relatively low, indicating that the seasonal variation in the sulfate concentrations at Mead are not well defined, and the peak day might not be a reliable value. The same behavior of seasonal components are evident for the ammonia concentrations at Mead, Nebraska, and the nitrate concentrations at Wellston, Michigan, and Huntington, New York. The data at Davis, California, show consistently later peak days or no significant seasonal model at all for all the constituents when compared to the other stations. The difference could be attributed to the sparse nature of the data from this station. In the 5-year period of record, there was no rainfall recorded with enough volume for chemical analysis in either July or August and only one recorded volume with analysis in June.

Generally, the seasonal model is least significant in the sodium regressions and relatively insignificant in the pH regressions. Other than these two constituents, there does not appear to be a particular station or constituent that has a significantly stronger or weaker seasonal component in its data.

TREND ANALYSIS

Trend Test Results

The classical parametric methods of trend detection are often inappropriate when applied to water-quality data. Precipitation chemistry data, for example, is characterized by missing data and values reported as less than the limit of detection. These features present no computational or theoretical problems for the Seasonal Kendall test for trends (Hirsch and others, 1982), a non-parametric test developed for use with data exhibiting seasonality. Nonparametric tests ignore magnitude in favor of assignment of a plus or minus value from comparison of pairs of data values (Crawford and others, 1983). This technique is most suitable for detecting monotonic trends during some interval of time.

The Seasonal Kendall test for trends was applied to the raw concentrations for each constituent at each station. The year is divided into 12 "seasons" (months) of exactly equal length. All concentration values for a particular season of a particular year are summarized for trend-testing purposes, by their median. The test is then carried out on these seasonal medians. Thus, while a given data set may contain about 260 values (5 years, 52 weeks per year), the test is performed on a set of about 60 values (5 years, 12 seasons per year) which summarize the original data. This procedure is described in detail by Crawford and others (1983). The use of the medians in the testing procedure makes the test especially resistant to outliers because they generally have no effect on the median. It also provides some protection against the effects of serial correlation in the data.

The null hypothesis for the test is that the probability distribution of the random variable, concentration, does not change over time. To avoid the problem of seasonality, observations from 1 month are only compared to other observations from the same month over the study period. If a later concentration (in time) is higher, a plus is recorded; if the later concentration is lower, a minus is recorded. A greater number of pluses or minuses indicates a trend in the time series of concentrations. The magnitude of the trend can be determined using the Seasonal Kendall slope estimator (Hirsch and others, 1982). The median of the slopes of the ordered pairs of data values compared in the Seasonal Kendall is the value used as the estimated slope and results in a milligrams per liter change per year in concentration.

The results of the trend analysis are shown in table 3. Of the 133 tests for trends in concentrations (7 constituents for 19 stations), 41 percent were downtrends, 4 percent were uptrends, and 55 percent showed no trends at $\alpha = 0.2$. At a more restrictive significance level of $\alpha = 0.05$, the breakdown was 24 percent downtrends, 2 percent uptrends, and 74 percent with no trend. The two constituents of greatest interest in terms of human generated emissions and environmental effects, sulfate and nitrate, showed only downtrends and sulfate showed the largest decreases in concentration per year of all the ions tested.

While not of great environmental interest, sodium shows downtrends at 17 of the 19 stations at $\alpha = 0.2$ (10 stations at $\alpha = 0.05$). In the absence of any evidence to suggest that emissions of sodium have been reduced nationwide or that a large-scale change in the circulation of ocean-derived sodium has occurred, the detection of these downtrends would certainly raise questions about the possibility that the trends in sodium were an artifact of changes in sampling or laboratory procedures or materials.

Since some relationship has been shown to exist between dissolved concentrations and volume of precipitation, it may be desirable to remove the variations in concentration that are due to variation in precipitation volume. The removal of this source of variation has the effect of increasing the power of the test procedure. Residuals from the non-seasonal regression, which are the difference between the predicted and the observed log concentrations, are log concentration values that have been adjusted to remove the influence of precipitation volume. The Seasonal Kendall test is applied to the residuals in the same manner as the raw concentrations. The residuals from the non-seasonal model are used instead of the residuals from the seasonal model since the Seasonal Kendall test is designed to compensate for seasonality in the data. The residuals are in logarithmic units, so the results from the Seasonal Kendall slope estimator are in percent change per year. The results of the trend analysis of the residuals (or the log of the concentrations if the non-seasonal model was insignificant) are given in table 4.

Of the 133 tests for trends in residuals, 49 percent showed downtrends, 7 percent showed uptrends, and 44 percent showed no detectable trend for $\alpha = 0.2$. For $\alpha = 0.05$, 38 percent of the tests showed downtrends, 2 percent showed uptrends, and 60 percent showed no detectable trends. For both significance levels, the number of trends detected in residuals was higher than the number of trends detected in the dissolved concentrations. Sulfate showed downtrends at 14 stations (all significant at the $\alpha = 0.05$ level), nitrate showed downtrends at 12 stations, chloride showed downtrends at 12 stations, and sodium showed a downtrend at 18 stations. Sulfate and nitrate showed significant

Table 3.--Trends in unadjusted concentrations for selected stations in the National Trends Network during 1978-83

[μmhos, micromhos per centimeter at 25° Celsius]

Station	Station name	Sulfate	Nitrate	Calcium	Chloride	Ammonia	Sodium	Potassium	Specific conduct-ance (μmhos per year)	pH (pH units per year)
		Milligrams per liter per year								
058840	Davis, California	-0.17 (.016)	-0.06 (.146)	--	--	--	-0.06 (.036)	--	-1.55 (.005)	--
062120	Manitou, Colorado	-.15 (.003)	--	--	--	--	-.02 (.083)	--	--	--
100360	Bradford Forest, Florida	-.10 (.165)	--	--	--	--	-.03 (.113)	--	--	-0.07 (.100)
114140	Georgia Station, Georgia	-.15 (.060)	--	--	--	--	-.04 (.033)	--	--	--
235340	Wellston, Michigan	-.48 (.001)	-.31 (.001)	-0.03 (.022)	-0.02 (.003)	-0.05 (.006)	-.05 (.000)	-0.004 (.065)	-1.88 (.121)	--
241660	Marcell, Minnesota	--	--	--	-.01 (.006)	--	-.02 (.019)	--	--	--
281520	Mead, Nebraska	-.37 (.002)	-.24 (.013)	-.10 (.042)	-.02 (.150)	-.06 (.036)	-.06 (.000)	-.012 (.067)	-1.35 (.065)	--
300240	Hubbard Brook, New Hampshire	-.21 (.013)	-.15 (.018)	--	-.03 (.002)	--	-.02 (.000)	--	-2.28 (.128)	.04 (.008)
332020	Huntington, New York	--	--	--	--	.02 (.148)	-.01 (.071)	--	--	.04 (.050)
340320	Lewiston, North Carolina	--	--	--	--	.03 (.007)	--	.005 (.007)	--	--
342500	Coweeta, North Carolina	-.14 (.065)	-.06 (.086)	--	-.01 (.030)	--	-.03 (.000)	--	--	.06 (.111)
343460	Piedmont Station, North Carolina	-.09 (.151)	--	--	--	--	-.04 (.090)	--	--	--
343560	Clinton Station, North Carolina	--	--	--	--	.02 (.087)	-.04 (.088)	--	--	--
344160	Finley (A), North Carolina	--	--	--	-.02 (.111)	--	-.04 (.086)	--	--	--
361760	Delaware, Ohio	--	--	--	--	--	--	.001 (.041)	--	--
364900	Caldwell, Ohio	-.23 (.046)	--	--	--	--	-.02 (.006)	--	--	--
367160	Wooster, Ohio	-.43 (.000)	--	-.02 (.106)	-.01 (.046)	--	-.02 (.001)	--	-1.22 (.053)	.04 (.025)
392940	Kane, Pennsylvania	-.20 (.047)	-.15 (.095)	--	-.02 (.095)	--	-.07 (.177)	--	-2.20 (.021)	.04 (.001)
501860	Parsons, West Virginia	-.24 (.012)	-.11 (.043)	-.01 (.074)	-.01 (.027)	--	-.02 (.000)	--	-1.19 (.190)	.02 (.024)

NOTE: Significance level is in parentheses below the trend value. If the significance level is greater than 0.20, nothing is listed.

Table 4.--Trends in volume-adjusted concentrations for selected stations in the National Trends Network during 1978-83

Station	Station name	Sulfate	Nitrate	Calcium	Chloride	Ammonia	Sodium	Potassium	Specific conductance	pH (pH units per year)
		Percent per year								
058840	Davis, California	-14.0 (.000)	-9.6 (.076)	--	--	--	-19.5 (.024)	--	-9.3 (.001)	--
062120	Manitou, Colorado	-10.9 (.002)	--	--	-11.9 (.083)	--	-20.6 (.004)	--	--	--
100360	Bradford Forest, Florida	--	4.7 (.130)	--	--	23.5 (.100)	--	--	-5.2 (.100)	-0.07 (.075)
114140	Georgia Station, Georgia	-9.4 (.012)	--	--	--	5.7 (.060)	-15.1 (.033)	--	--	--
235340	Wellston, Michigan	-14.9 (.000)	-7.8 (.010)	-4.2 (.161)	-9.6 (.006)	-13.7 (.022)	-26.3 (.006)	--	-5.6 (.065)	--
241660	Marcell, Minnesota	--	--	-7.2 (.090)	-10.9 (.009)	--	-21.1 (.019)	--	--	-.06 (.192)
281520	Mead, Nebraska	-11.8 (.004)	-3.6 (.050)	--	-6.4 (.050)	-7.5 (.050)	-28.4 (.000)	--	--	--
300240	Hubbard Brook, New Hampshire	-9.9 (.004)	-9.6 (.009)	--	-13.1 (.001)	--	-22.1 (.000)	--	-6.5 (.078)	.04 (.013)
332020	Huntington, New York	--	-3.1 (.130)	--	-6.6 (.097)	--	-14.7 (.051)	--	-5.7 (.071)	.04 (.050)
340320	Lewiston, North Carolina	--	--	--	--	27.3 (.000)	-11.2 (.068)	15.0 (.000)	--	--
342500	Coweeta, North Carolina	-9.7 (.013)	-6.9 (.086)	--	-11.1 (.021)	7.6 (.086)	-20.9 (.000)	--	--	.05 (.143)
343460	Piedmont Station, North Carolina	-10.1 (.004)	-4.7 (.192)	--	--	--	-19.3 (.003)	--	--	--
343560	Clinton Station, North Carolina	--	6.7 (.152)	--	--	17.5 (.035)	-13.4 (.035)	8.1 (.088)	-5.3 (.117)	--
344160	Finley (A), North Carolina	-7.5 (.026)	--	--	-9.5 (.111)	--	-18.2 (.018)	--	-5.6 (.065)	--
361760	Delaware, Ohio	-5.6 (.013)	-3.7 (.026)	--	-5.9 (.086)	--	-11.9 (.036)	--	--	--
364900	Caldwell, Ohio	-5.7 (.009)	-5.6 (.025)	--	--	--	-19.0 (.004)	--	--	--
367160	Wooster, Ohio	-10.1 (.000)	-6.1 (.062)	-9.3 (.034)	-10.3 (.006)	--	-24.2 (.000)	-9.0 (.135)	-5.4 (.004)	--
392940	Kane, Pennsylvania	-6.7 (.021)	-10.5 (.047)	--	-7.8 (.014)	--	-14.2 (.014)	-7.1 (.131)	-4.8 (.095)	.04 (.021)
501860	Parsons, West Virginia	-9.9 (.003)	-7.3 (.032)	-8.0 (.006)	-7.4 (.009)	--	-21.1 (.000)	-7.6 (.003)	--	.03 (.074)

NOTE: Significance level is in parentheses below the trend value. If the significance level is greater than 0.20, nothing is listed.

downtrends at all stations in Ohio, Pennsylvania, and West Virginia, a geographical area that has often been singled out as an area of concern with respect to acid deposition. Nitrate, ammonia and potassium were the only constituents to show evidence of uptrends. Only 33 percent of the uptrends detected were significant at the $\alpha = 0.05$ level. As might be expected for precipitation data with a predominance of downtrends in constituent concentration, pH showed a predominance of uptrends and specific conductance showed a predominance of downtrends. Sodium consistently showed the largest percent per year decreases (an average of 19 percent per year) even though in milligrams per liter, they were among the smallest trends. Sulfate and chloride trends generally were larger than all other constituents except sodium. A summary of the trends detected for raw concentrations and volume-adjusted concentrations in all constituents at all 19 stations is given below in table 5.

Table 5.--Summary of trends detected in all dissolved constituents at all 19 stations

	Unadjusted concentrations		Volume-adjusted concentrations	
	$\alpha = 0.05$	$\alpha = 0.20$	$\alpha = 0.05$	$\alpha = 0.20$
Downtrends	32	54	50	65
Uptrends	3	5	3	9
No trends	98	74	80	59

Interpretation of Trend Test Results

The attained significance level (p) for the Seasonal Kendall test is a measure of risk. It is the probability that a random arrangement of data would result in an indication of trend, as measured by the Seasonal Kendall test statistic, which is as strong or stronger than that observed in the data set at hand. In hypotheses testing, one selects a significance level (denoted here as α) that is defined as the risk (a probability) that one would reject the null hypothesis when it was, in fact, true. Rather than simply stating whether the null hypothesis is rejected or not, based on the author's own selected α value, this report gives an attained significance level (p). This allows the readers to make their own determination about rejection. A reader who preferred to keep the risk of false rejection very low, say, $\alpha = 0.01$, would reject only when $p < 0.01$. Another reader who was less risk adverse might pick $\alpha = 0.20$ and would reject only when $p < 0.20$.

A null hypothesis can only be developed on the basis of a particular mathematical model of the process. In the case of the Seasonal Kendall test, the null hypothesis is that for each and every season, the probability distribution does not change from one year to the next (stationary superimposed on a seasonal cycle) and that the process is serially independent. When the test results in a particularly low value of p, it is necessary to consider the possible stochastic models which could give rise to this result and to evaluate their plausibility in terms of the knowledge of the properties of the time series and the physical processes involved. The classes of possible models are the following.

(It is assumed in this discussion that any of these may be superimposed on a regular seasonal cycle.)

(1) The process is indeed stationary and independent.

(2) The process is stationary, with short memory with $0 < \rho_1 < 0.4$, where ρ_1 is the lag 1 week autocorrelation coefficient. A common example is the autoregressive lag 1 model (AR(1)) (see Box and Jenkins, 1970).

(3) The process is stationary with long memory with $0 < \rho_1 < 0.4$. An example would be the autoregressive-moving average model (ARMA (1,1)) with $\phi > 0.4$, where ϕ is the autoregressive parameter of the ARMA (1,1) model.

(4) The process is non-stationary.

Any other processes, such as ones with ρ_1 substantially greater than 0.4 or negative can be effectively ruled out by an examination of the sample autocorrelation function (ACF) of the time series of residuals from the seasonal regression model. The first 12 sample autocorrelations $r_1,\ldots,r_{12}$ of the time series for SO_4, NO_3, and Ca from each of the 19 stations were computed. Because of the many missing values (due to weeks of no rain), these autocorrelation coefficients were computed as cross correlation coefficients of lagged series and the unlagged series. The number of data pairs used in computing each of the correlation coefficients is about 200. With this sample size, the standard error of any of the r_j values is about 0.07, assuming independence (Kendall, 1975).

Table 6 summarizes some of the characteristics of these ACFs. About the only conclusions that can be drawn from these sample ACFs is that at many of the stations the processes may not be independent but in no case are the sample ACFs consistent with processes with ρ_1 values greater than about 0.4 or substantially less than zero. Because the r_j values for $j > 1$ are predominantly in the "noise" range close to zero, it is impossible to argue that they are more consistent with any particular one of the following: a short memory, a long memory, or a non-stationary process. The formal procedures of model identification and estimation described by Box and Jenkins (1970) cannot be applied because of the missing data.

Table 6.--Summary of the sample autocorrelation functions for the non-seasonal residuals at all 19 stations

		Constituent		
		SO_4	NO_3	Ca
For the 19 stations	maximum r_1	0.35	0.18	0.33
	median r_1	.10	.01	.13
	minimum r_1	-.06	-.05	.33
Number of stations where only 0, 1, or 2 of the r_j, $j=1,2,\ldots,12$ values were significantly different from zero ($\alpha = 0.1$)		12	17	9

Given this knowledge, it is possible to evaluate the plausibility of and implications of, the four classes of models described above when the attained significance level (p) is low.

(1) The process could, in fact, be stationary and independent. However, in this case, the reported p value provides an accurate assessment of the risk of erroneously concluding that there is trend if this were, in fact, the case.

(2) The process could be stationary with short memory with $0 < \rho_1 < 0.4$. Based on Monte Carlo simulations with record lengths of 5 years, it can be concluded that the reported p value provides an accurate assessment of the risk of concluding that there is trend if this were, in fact, the case. One physical mechanism which might lead to this kind of stochastic process would be the tendency for particular weather patterns to persist for periods from a few days to a few weeks. For example, the storms occurring during a few consecutive weeks at a station may all have moved along a particular storm track which carries them across an area which is a major source for some particular constituent. The tendency for this kind of phenomenon to occur, from time to time, could be modeled as a short memory stationary process. The word "accurate" with regard to the p values is intended to imply that where p is reported to be, say, 0.05, the true risk is in the range of about 0.04 to 0.065, or where p is reported to be 0.01, the true risk is in the range of about 0.01 to 0.015.

(3) The process could be stationary with a long memory but with $0 < \rho_1 < 0.4$. If this were the case, then the true risk of concluding that a trend exists could be substantially higher than p. One physical mechanism which might lead to this kind of stochastic behavior is that there are long-lasting (several months to years in duration) climatic phenomena which cause a sustained tendency for positive (or negative) departures from the long-term mean. It should be recognized that these departures are above and beyond those which can be "explained" by a regression relationship on precipitation volume or by the normal seasonal cycle. Positive departures in atmospheric deposition could be associated with increases in the amount of wind erosion which would accompany protracted and widespread drought conditions. Positive (or negative) departures could also be associated with protracted episodes of greater (or lesser) emissions of constituents from sources such as power plants. These episodes could be related to episodes of high economic activity or changes in supply and demand conditions in markets for various types of fuels. Whatever the cause, these phenomena could lead to records which have the appearance of trend--that is, prolonged episodes of higher or lower than normal deposition. They may create an impression of gradual monotonic change, abrupt monotonic change, or a series of fluctuations with a general tendency toward decreasing or increasing values. The difference between these fluctuations and "nonstationarity" is a mathematically meaningful difference but probably not a difference that is of practical significance. In either event, there is a real difference in the concentrations observed in the early part of the record and those observed in the late parts of the record.

(4) The other possibility is that the process is indeed non-stationary and this is, of course, precisely the condition one would like the test to identify. The practical question one would like to evaluate is what might be the cause of the observed trend. The use of some graphical procedures applied to a set of stations can be quite useful here.

If one were to plot the raw concentrations or logs of the raw concentrations at a station versus time, the plots would be difficult to interpret because of the confounding effects of season and precipitation volume variability. Plotting the residuals from the seasonal regression model would surmount these problems but would still not be completely satisfactory because the human eye tends to be sensitive to the extremes of the data rather than to the drift of the bulk of the data. To surmount this problem, one can smooth the time series with robust smoothing techniques which minimize the influence of the extremes and track the overall drift of the data.

By plotting logical groups of smoothed series together, it may be possible to develop more refined hypotheses about the trends that have been observed. Some clear-cut situations where such plotting would be very useful are these:

(a) All stations in a region show gradual trends in the same direction. The plausible hypotheses about the cause might include: widespread changes in the operation of many facilities which are sources of the chemical in question (for example, technological change, change in fuel, or changes in levels of activity driven by economic forces or installation of emission control equipment required by law); or the changes are driven by long-lasting climatic phenomena, in particular those related to drought and wind erosion.

(b) All stations in a region show rather abrupt changes, at about the same time and in the same direction. Some plausible hypotheses in this case include both the climatically and economically driven ones mentioned above. Another possibility is a methodological artifact associated with a change in data collection protocol, material used, shipment method, or analytical technique. The analysis of data from the entire network should be useful for consideration of this hypothesis. The hypothesis that emission controls are the cause would not be particularly plausible because the controls are likely to be instituted at different times at each of the many facilities which determine the precipitation quality of the region.

(c) Some or all of the stations show abrupt changes which occur at different times, in the same or different directions. The hypotheses that this would suggest are that, at each station which showed abrupt change, the change was brought on by a modification of emissions at a particular source which has a major influence on atmospheric deposition at the station, or that some condition or practice at the station which would lead to sample contamination had substantially changed at that time.

Every one of these hypotheses suggested by the trend analysis and by graphical inspection should lead to an investigation of the history of the network, the station, and the factors suggested in the hypothesis. The investigation may, or may not, rely on statistical arguments to attempt to confirm or deny the hypothesis. Some of the data preparation approaches used in the present study may be useful in the exploration of these hypotheses. The following section shows examples of the kind of graphical exploration and hypothesis building described above.

GRAPHICAL INTERPRETATION OF TRENDS

Scattter plots of the regression residuals versus time are useful tools for determining if changes in the record are gradual or abrupt, and if they are abrupt, for determining the time of the shift. The usefulness of a scatterplot can be enhanced by the use of smoothing which graphically summarizes the pattern of the data with respect to time. In particular, when one would like to evaluate several time series simultaneously (one constituent at several stations or several constituents at one station) one can plot many of these smooth curves without the actual data and determine the similarities and differences in their histories.

The specific smoothing technique used here is called "lowess" (locally weighted scatterplot smoothing). It is described by Cleveland (1979) and Cleveland and McGill (1984). The term lowess refers to a general catagory of techniques within which there are numerous specific options one may select for any given application. In general terms the technique relies on repeated use of weighted least squares regressions to make a robust estimate of the mean value of the series at any given time in its history. The procedure is iterative in that an initial smoothed series is calculated, residuals from this smoothed series are calculated, and then the smoothed series is recalculated giving reduced weight to those observations which had large (in absolute value) residuals in the previous iteration.

The precise algorithm used is described below. Define, for all i=1,...,n (where n is the number of observations in the record):

y_i = the data to be smoothed (in this case the residuals of from the seasonal regression model)

$\hat{y}_i$ = the smoothed estimate of y_i

t_i = the time of the observation of y_i (in years)

Also, define h as the half width of the smoothing time-window. Thus any observation more than h years from t_i has no influence on the estimate $\hat{y}_i$. By trial and error, it was determined that 0.5 year is an appropriate value for h for these data.

Step 1. For each i, compute a weight to apply to each of the n observations, based on their distance in time from t_i. This weight is computed for j = 1,....,n, by the bisquare weight function (Mosteller and Tukey, 1977), according to the following formula.

$$d_j = | t_i - t_j |$$

$$w_j = \begin{cases} (1.0 - (d_j/h)^2)^2 & \text{if } d_j \leq h \\ 0.0 & \text{if } d_j > h \end{cases}$$

Step 2. Fit a regression to the points close to i by using a weighted least squares fit of the y_j on t_j using the weights w_j. The smoothed value at t_i is $\hat{y}_i$, the y value of the fitted regression at $t = t_i$. The residual at t_i is $r_i = y_i - \hat{y}_i$.

Step 3. Repeat steps 1 and 2 for all i, i=1,....,n.

This is the first iteration, it is a locally weighted regression but is not robust against the influence of outliers. The next two iterations use the residuals to compute "robustness weights" for each observation which are then multiplied by the "distance weights" from step 1 to provide a new set of combined weights.

Step 4. Define s as the median of the absolute values of the n r_i values. Compute the robustness weights, u_i, for all n observations from a bisquare weight function, as follows:

$$u_i = \begin{cases} (1.0 - (r_i/(6*s))^2)^2 & \text{if } |r_i| \leq 6*s \\ 0.0 & \text{if } |r_i| > 6*s \end{cases}$$

Step 5. For each value of i compute the distance weight, w_j, for each j according to the rules given in step 1. Compute the combined weight, z_j, for each j as:

$$z_j = w_j * u_j.$$

Step 6. Fit a regression to the data points close to i by using a weighted least squares fit of the y_j on t_j, using the weights z_j. The smoothed value at t_i is $\hat{y}_i$, the y value of the fitted regression at $t = t_i$. The residual at t_i is $r_i = y_i - \hat{y}_i$.

Step 7. Repeat steps 5 and 6 for all i, i=1,...,n.

Step 8. Using these new residuals iterated through steps 4 through 7 again to arrive at the final smoothed series $\hat{y}_i$, i=1,...,n. What is plotted are the $\hat{y}_i$ values versus t_i, with linear interpolation between the computed values.

Figures 6 and 7 show examples of such smoothed series for sulfate and for sodium at the Coweeta, North Carolina, station. The trend test results shown in table 4 for these time series showed downward trends in both. For sulfate the trend slope was -9.7 percent per year ($p = 0.013$). For sodium the trend slope was -20.9 percent per year ($p < 0.0005$). The plot for sulfate shows rather consistent behavior through 1979 and 1980, a small rise in the last half of 1981 and an abrupt fall-off in early 1982 with the low values persisting through the remainder of 1982 and 1983. The pattern for sodium is quite different: high values in 1978 and 1979, an abrupt drop in early to middle 1980 followed by generally low levels through the end of 1983, with only a short-duration rise and fall in late 1981.

In the case of sulfate, the average levels typical of 1979 and 1980 are about 1.5 times the levels of late 1982 through 1983. Such ratios are computed as the antilog of the difference in the smoothed residual values (note that the logs are base e, so the antilog is the exponential function). For sodium the smoothed values reach a sustained high of about 0.7 in 1979 and a sustained low of about -0.4 in 1982 and 1983. This represents a level of concentrations in 1979 that is three times that of 1982-83. If the trends detected in sodium were an artifact of changes in laboratory or sampling changes, it does not appear that these changes affected sulfate.

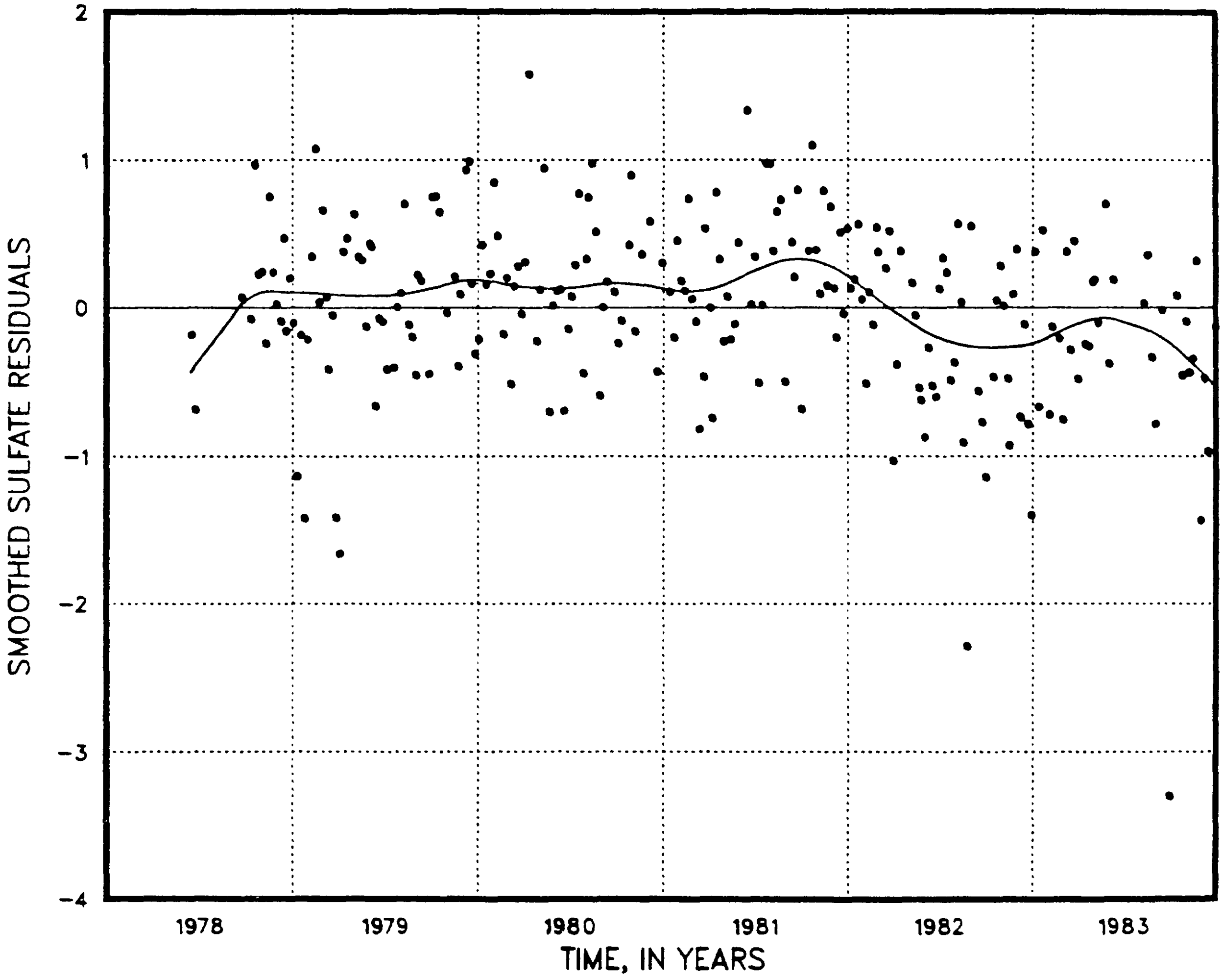


Figure 6.--Smoothed sulfate residuals from Coweeta, NC, for 1978-83.

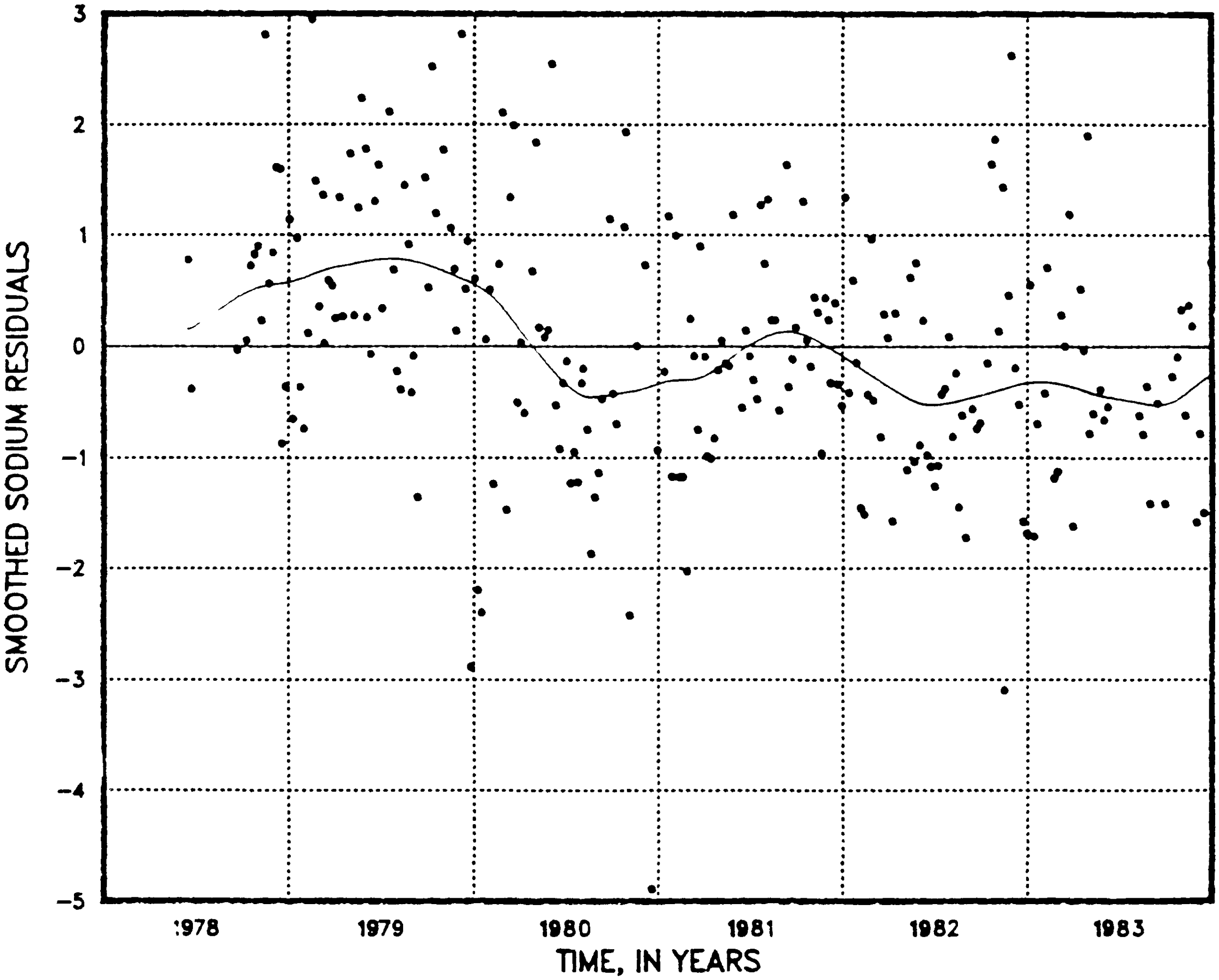


Figure 7.--Smoothed sodium residuals from Coweeta, NC, for 1978-83.

These smoothed curves, without the scattered data, are shown in figures 8 and 9 for each of the 19 stations. The plots show that sulfate changes are much less pronounced than changes in sodium. Sulfate data do show some tendencies which are shared by almost all of the stations east of the Mississippi River. They show small rises in late 1981 and declines in 1982, with persistent low levels in 1983. All 19 of the sodium records show a decline from the begining of 1979 to the end of 1980. Most stations show a rise and fall in 1981, followed by a general tendency to stay low for the remainder of the record, although some stations show an increase at the end of 1983. Some of the changes over a single year are very large. For example, the concentrations at Wellston, Michigan, at the begining of 1980 are about 7.4 times the concentrations at the end of 1980.

It is beyond the scope of this study to suggest the reasons for such changes, or to catalog and display all of the records. However, the simultaneity of the changes over the Nation shown by these plots suggests the possibility that some trends may have been the result of changes in procedures or materials used in sample collection, shipment or analysis. In addition to evaluating the history of the network's operations, analysis of other atmospheric deposition data sets (collected with different procedures and equipment and analyzed in different laboratories) as well as stream chemistry data sets from pristine areas, would be useful for investigating the origins of these observed trends. If other independent data sources show similar patterns, this would help to confirm that these widespread trends are real. If similar patterns are not found, this would tend to suggest that the trends are a result of changes in the operation of the network.

MULTIPLE STATION ANALYSES

In addition to analyses of trends and computation of summary statistics at individual stations, another approach to data analysis deals with geographical groupings of stations. It is possible to compute block average of variables over an area such as a state using Kriging (Finklestein, 1984) or other spatial data analysis methods. It is also possible to do trend analyses on collections of stations.

The existance of five stations in the state of North Carolina provides an opportunity to explore the spatial relationships of records and a few of the possibilities for spatial analysis. The seasonal model residuals for sulfate were computed at these five stations and then the product-moment (Pearson) cross correlation coefficients computed between all possible pairs of records. Each of these records contain from 213 to 235 values. The 10 cross correlation coefficients computed are based on 179 to 191 data pairs each. The reason that the number of data pairs is smaller than the number of values at a station is because the weeks of missing values at the stations were not always coincident in time.

The cross correlation coefficients ranged from a low of 0.11 for the Lewiston and Coweeta stations (579 km apart), to highs in the range 0.56 to 0.64 for the three pairs made up from the three nearby stations, Lewiston, Clinton, and Finley (ranging from 86 to 158 km apart).

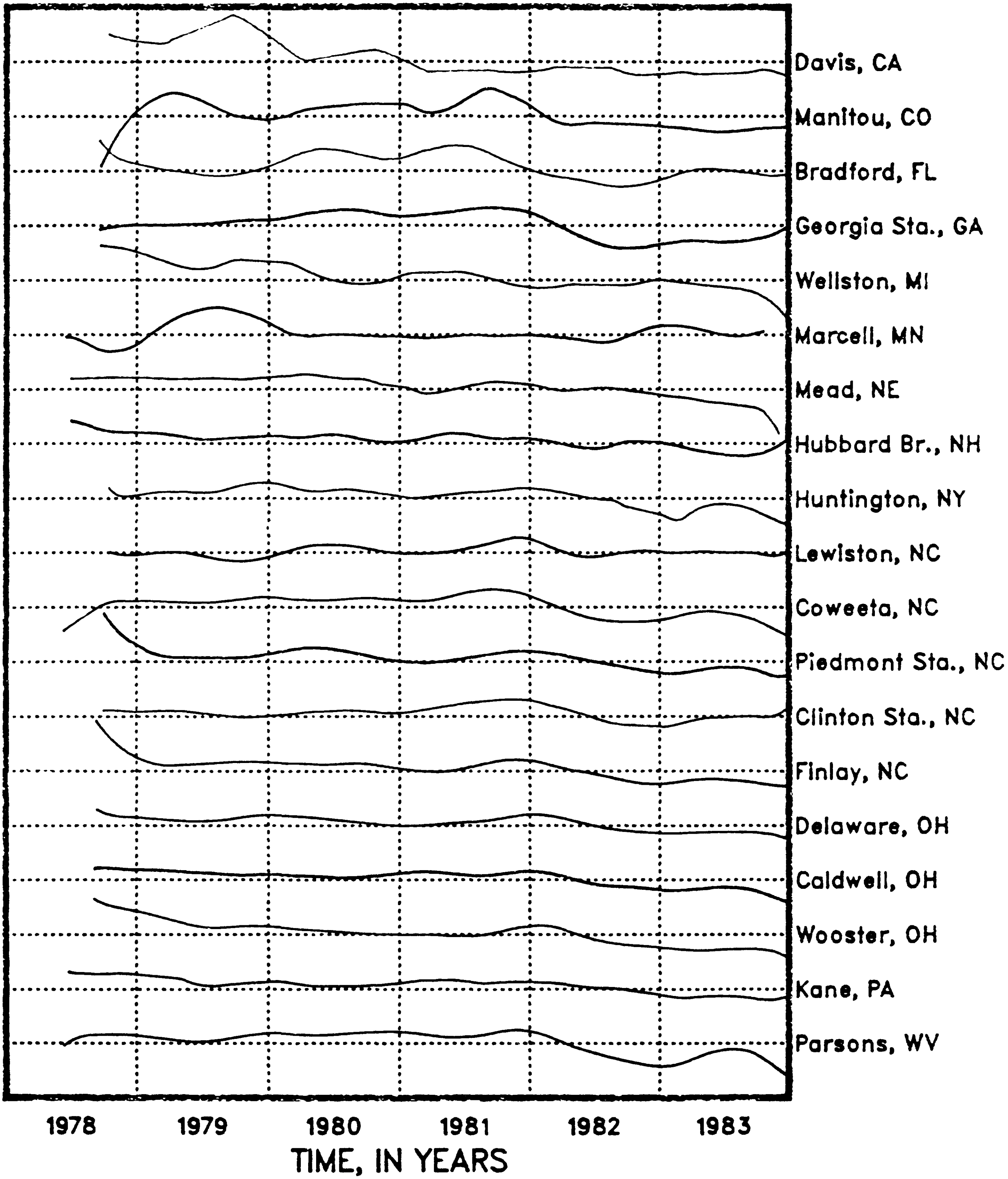


Figure 8.--Smoothed sulfate residuals for each of the 19 stations during 1978-83.

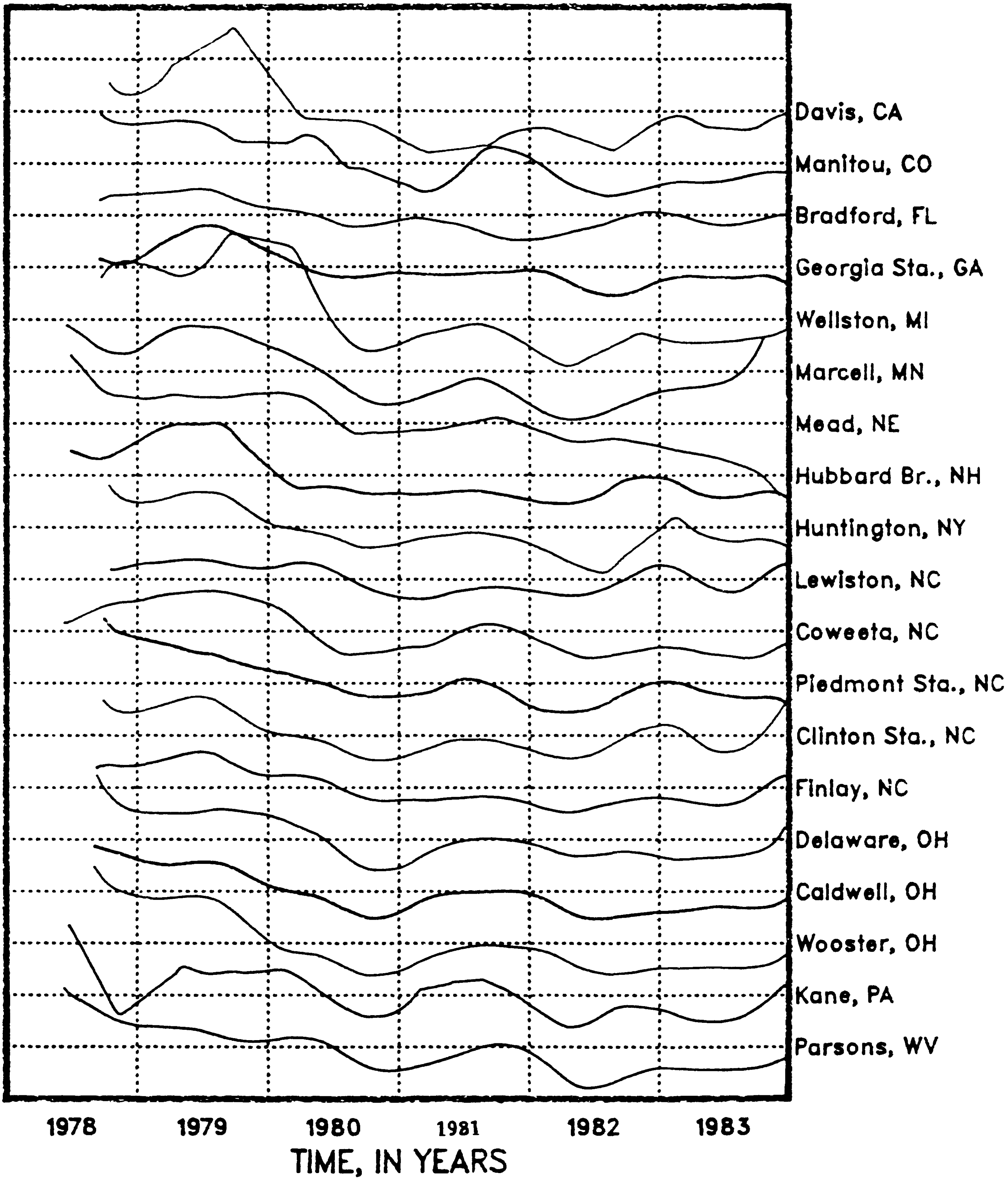


Figure 9.--Smoothed sodium residuals for each of the 19 stations during 1978-83.

Figure 10 shows the relationship between these correlation coefficients and the interstation distance for all station pairs. The curve was fitted as a linear regression with zero intercept:

$$\ln(\rho) = b * k$$

where ρ is the correlation coefficient,
ln is the natural logarithm,
k is the interstation distance in kilometers, and
b is the slope.

The fitted value of b is -0.00404. The equation can be transformed by taking antilogs, resulting in the equation:

$$\rho = 0.9960^{k}$$

An "equivalent R-square" for this equation, computed as the fraction of the variance in the ρ values explained by the regression is 0.86. The particular functional form of the curve was selected because of two desirable properties: ρ takes on a value of 1 at zero distance, and ρ decreases asymptotically to zero with increasing distance. This curve can be used to determine optimal weights for computing statewide averages, the possible gain in information that might be achieved by adding stations to the network, or the loss of information from dropping stations. (See Hirsch and Gilroy, 1985).

It is possible to apply the Seasonal Kendall test to all five stations simultaneously. This is accomplished by summing the 60 Mann-Kendall S statistics (one for each season - station combination) to form a regional test statistic. A naive approach to this kind of test would be to assume independence between stations so that the variance of the sum of the S statistics would be assumed to be the sum of the 60 variances, which are known. Using this approach the test would indicate a downwards trend in sulfate for the whole collection of stations with a p-value of 0.0004. This result is clearly rather extreme in light of the fact that two of the five stations show virtually no indication of a trend.

If the S statistics are treated as correlated random variables then the variance of their sum can be calculated correctly, by including estimates of all of the covariances between the S statistics. The formula for estimating these covariances was developed by Dietz and Killeen (1981) and expanded to consider missing values by Hirsch and Slack (1984). Taking this approach, the data from the five stations indicate a downward trend, with a p-value of 0.018. This type of regional trend analysis offers an attractive alternative to single station trend analysis.

In addition to such regional trend tests, regional weighted averages can be computed for various periods (weeks, seasons, or years) by generalized least squares or Kriging procedures. These procedures also allow for the calculation of the error variance of these regional averages.

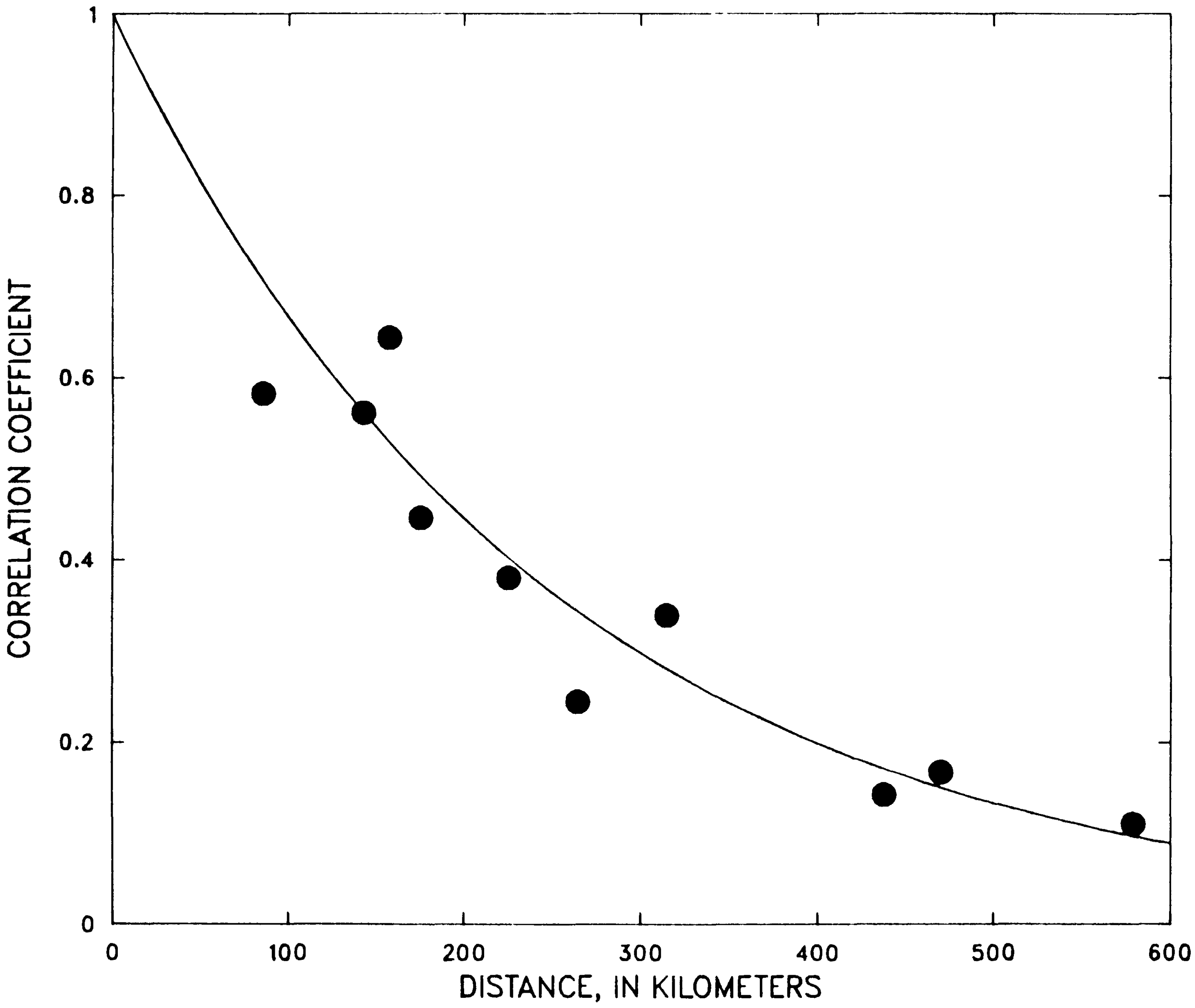


Figure 10.--Cross correlation coefficients versus distance for sulfate data from the five stations in North Carolina.

SUGGESTIONS

The analyses performed and plots examined in this study are only a first-look at the precipitation data available from NTN. As the period of record increases at these stations, these analyses should continually be redone. More data over a longer period will only enhance the reliability of any trends detected, and the data will become more suitable for various other avenues of analyses. As more stations reach a period of record that is suitable for trend analysis, the national picture for precipitation chemistry obtained from studies such as this will become more clear and more detailed. Besides continuation of the analyses done in this study, it might prove beneficial to examine trends in the ratios of various constituent concentrations or trends in residuals from a regression of log monthly deposition against log monthly volume. Trend tests on weekly data or residuals may not be wise because of the effects of serial correlation at this short time interval. On the other hand, summarization to annual totals would substantially decrease the ability to detect a trend because of the greatly reduced sample size.

SUMMARY OF CONCLUSIONS

The data for seven chemical constituents (sulfate, nitrate, calcium, chloride, ammonia, sodium, and potassium) from 19 NADP/NTN stations across the United States were examined for evidence of spatial and temporal trends in 5 years of record. Scatter plots of dissolved-constituent concentrations and volume of precipitation from these sites typically showed a positively skewed, non-linear relationship. Log transformation of the data resulted in a more normally distributed, linear relationship. The regression model:

$$\ln(C) = a + b * \ln(P)$$

called the non-seasonal model, was used to describe the relationship between ln(C) and ln(P). The model was shown to be appropriate for describing the relationship through examination of the residuals from the regression. The residuals were approximately homoscedastic, symmetrically distributed and independent of ln(P). The residuals typically exhibited too many outliers to be called a normal distribution.

Further examination of the residuals from the non-seasonal model showed evidence of a seasonal pattern in most dissolved constituents at most stations. A seasonal term added to the non-seasonal model resulted in the following seasonal model:

$$\ln(C) = a + b * \ln(C) + c * \sin(2\pi T) + d * \cos(2\pi T).$$

The results from regression analysis of both models against the data from each of the 19 stations were examined for detectable patterns in the characteristics of the models. The slopes for all of the variables considered were negative (lower values of concentration associated with higher values of precipitation volume). The exception was hydrogen ion concentration (derived from the equation for pH) which increases with precipitation volume. Standard errors for the regressions were generally high (greater than 50 percent), indicating that a large amount of variation in the data is not explained by the components of the models. R-square values, which indicate the strength of the relationship between the dependent and independent variables, were consistently highest for the calcium regressions and lowest for the pH regressions. If the seasonal model proved to be significant, then the amplitude and phase shift of the seasonal term were examined. The peak day represents the day of the year when the amplitude is at its peak. Approximately 60 percent of the peak days for the residuals were between mid-April to mid-July. Chloride consistently peaked in December and January. Sodium and pH had weaker seasonal components than chloride but also tended to peak in the winter months. The higher concentrations of sodium and chloride in the winter could be due to the influence of cyclonic storms in the Gulf of Mexico or the Atlantic Ocean that are most vigorous during the winter months. Generally, the seasonal model is least significant in the sodium regressions and relatively insignificant in the pH regressions. No patterns were detected, geographical or otherwise, in the characteristics exhibited by either the seasonal or the non-seasonal regressions.

The non-parametric Seasonal Kendall test was used for trend detection due to its ability to handle missing data, values reported as less than the limit of detection, and symmetrical, but non-normal, distribution of the data. The test was applied to both the raw concentrations and the residuals from the non-seasonal model for each constituent at each station. Of the 133 tests for trends in concentrations, 41 percent were downtrends, 4 percent were uptrends, and 55 percent showed no trends at $\alpha = 0.2$. At a more restrictive significance level of $\alpha = 0.05$, the breakdown was 24 percent downtrends, 2 percent uptrends, and 74 percent with no trend detected. The two constituents of greatest interest in terms of human generated emissions and environmental effects, sulfate and nitrate, showed only downtrends and sulfate showed the largest decreases in concentration per year of all the ions tested. Sodium showed downtrends at 17 of the 19 stations at $\alpha = 0.2$ (10 stations at $\alpha = 0.05$), which could be an artifact of changes in sampling or laboratory procedures or materials. Of the 133 tests for trends in residuals, 49 percent showed downtrends, 7 percent showed uptrends, and 44 percent showed no detectable trend for $\alpha = 0.2$. For $\alpha = 0.05$, 38 percent of the tests showed downtrends, 2 percent showed uptrends, and 60 percent showed no detectable trends. For both significance levels, the number of trends detected in residuals was higher than the number of trends detected in the dissolved concentrations.

Plots of smoothed residual series over time for sulfate and sodium were examined for evidence of temporal patterns. The plots show that sulfate changes were much less pronounced than changes in sodium. Sulfate data showed some tendencies which were shared by almost all of the stations east of the Mississippi River. They showed small rises in late 1981 and declines in 1982, with persistent low levels in 1983. All 19 of the sodium records showed a decline from the beginning of 1980 to the end of 1980. Most stations showed a rise and fall in 1981, followed by a general tendency to stay low for the remainder of the record. Some of the changes over a single year were very large. Although it is beyond the scope of the study to suggest the reasons for such changes, the simultaneity of the changes over the Nation shown by these plots suggests the possibility that some trends may have been the result of changes in procedures or materials used in sample collection, shipment or analysis.

The existence of five stations in the state of North Carolina provided an opportunity to explore the spatial relationships of records and a few of the possibilities for spatial analysis. The seasonal model residuals for sulfate were computed at these five stations and then the product-moment (Pearson) cross-correlation coefficients computed between all possible pairs of records.

A curve fitted to the relationship between the correlation coefficients and the interstation distance for all station pairs showed a strong relationship ($R^2 = 0.86$). A curve of this type could be used to determine optional weights for computing statewide averages, the possible gain in information that might be achieved by adding stations to the network, of the loss of information from dropping stations. The Seasonal Kendall test was applied to all five stations simultaneously and showed a downward trend with a p-value of 0.018.

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APPENDIX

Seasonal residuals of sulfate that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
811110	058840	DAVIS, CALIFORNIA	-2.3647	1.85521
821207	062120	MANITOU, COLORADO	-2.9282	1.95791
800722	100360	BRADFORD FOREST, FLORIDA	-2.6968	2.00186
810929	100360	BRADFORD FOREST, FLORIDA	2.5135	2.00186
820601	100360	BRADFORD FOREST, FLORIDA	-3.6353	2.00186
810922	114140	GEORGIA STATION, GEORGIA	1.9843	1.78912
821221	114140	GEORGIA STATION, GEORGIA	-2.0377	1.78912
810915	235340	WELLSTON, MICHIGAN	-1.8066	1.78192
790605	241660	MARCELL, MINNESOTA	-3.4134	2.15901
791106	281520	MEAD, NEBRASKA	-2.3084	1.83537
830308	300240	HUBBARD BROOK, NEW HAMPSHIRE	-3.0523	2.08870
831220	300240	HUBBARD BROOK, NEW HAMPSHIRE	-3.0130	2.08870
800205	332020	HUNTINGTON, NEW YORK	-2.5184	1.96051
820914	342500	COWEETA, NORTH CAROLINA	-2.2861	1.78001
831018	342500	COWEETA, NORTH CAROLINA	-3.2983	1.78001
790612	343460	PIEDMONT STATION, NORTH CAROLINA	-1.5864	1.57732
830726	343460	PIEDMONT STATION, NORTH CAROLINA	-1.9832	1.57732
781226	343560	CLINTON STATION, NORTH CAROLINA	-3.1355	1.90059
800722	343560	CLINTON STATION, NORTH CAROLINA	-2.0271	1.90059
830614	343560	CLINTON STATION, NORTH CAROLINA	-2.5482	1.90059
790925	344160	FINLEY (A), NORTH CAROLINA	-2.0677	1.85932
791120	344160	FINLEY (A), NORTH CAROLINA	-2.9280	1.85932
790102	361760	DELAWARE, OHIO	-1.6776	1.57490
800226	361760	DELAWARE, OHIO	-2.7385	1.57490
830419	361760	DELAWARE, OHIO	-2.4427	1.57490
800129	364900	CALDWELL, OHIO	-1.8615	1.45186
800226	364900	CALDWELL, OHIO	-2.3877	1.45186
830208	364900	CALDWELL, OHIO	-1.8095	1.45186
800226	367160	WOOSTER, OHIO	-1.5188	1.42961
800429	392940	KANE, PENNSYLVANIA	1.4712	1.39349
820601	392940	KANE, PENNSYLVANIA	-1.5385	1.39349
791120	501860	PARSONS, WEST VIRGINIA	-1.9834	1.64202
800129	501860	PARSONS, WEST VIRGINIA	-1.7549	1.64202
810519	501860	PARSONS, WEST VIRGINIA	-1.7772	1.64202
821228	501860	PARSONS, WEST VIRGINIA	-2.2554	1.64202
830927	501860	PARSONS, WEST VIRGINIA	-1.7816	1.64202
831011	501860	PARSONS, WEST VIRGINIA	-1.7499	1.64202

Seasonal residuals of nitrate that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
800122	058840	DAVIS, CALIFORNIA	1.6298	1.57131
781121	062120	MANITOU, COLORADO	-3.0967	2.70599
810721	062120	MANITOU, COLORADO	-4.9237	2.70599
811020	062120	MANITOU, COLORADO	-3.3362	2.70599
830913	062120	MANITOU, COLORADO	-3.7847	2.70599
820921	100360	BRADFORD FOREST, FLORIDA	-3.9814	2.16762
830208	100360	BRADFORD FOREST, FLORIDA	-3.3275	2.16762
781205	114140	GEORGIA STATION, GEORGIA	-4.3595	3.00533
790724	114140	GEORGIA STATION, GEORGIA	-4.7646	3.00533
791009	114140	GEORGIA STATION, GEORGIA	-4.6452	3.00533
791204	114140	GEORGIA STATION, GEORGIA	-4.4021	3.00533
800429	114140	GEORGIA STATION, GEORGIA	-5.1805	3.00533
810602	114140	GEORGIA STATION, GEORGIA	-4.7275	3.00533
811110	235340	WELLSTON, MICHIGAN	-2.6099	2.07203
820427	235340	WELLSTON, MICHIGAN	-5.5920	2.07203
791211	241660	MARCELL, MINNESOTA	-2.9571	2.40254
820928	241660	MARCELL, MINNESOTA	-4.4325	2.40254
830628	241660	MARCELL, MINNESOTA	-4.3051	2.40254
820921	281520	MEAD, NEBRASKA	-5.2644	2.32637
830419	281520	MEAD, NEBRASKA	-5.4172	2.32637
790612	300240	HUBBARD BROOK, NEW HAMPSHIRE	-5.5337	2.55297
810630	300240	HUBBARD BROOK, NEW HAMPSHIRE	-4.5092	2.55297
820427	300240	HUBBARD BROOK, NEW HAMPSHIRE	-4.0484	2.55297
830308	300240	HUBBARD BROOK, NEW HAMPSHIRE	-2.7844	2.55297
790612	332020	HUNTINGTON, NEW YORK	-4.4831	2.06419
790703	332020	HUNTINGTON, NEW YORK	-2.3791	2.06419
801021	332020	HUNTINGTON, NEW YORK	-2.3530	2.06419
800916	340320	LEWISTON, NORTH CAROLINA	-4.6768	2.62686
810324	340320	LEWISTON, NORTH CAROLINA	-5.0208	2.62686
820126	340320	LEWISTON, NORTH CAROLINA	-4.7845	2.62686
790417	342500	COWEETA, NORTH CAROLINA	-4.3676	2.27395
790612	342500	COWEETA, NORTH CAROLINA	-2.3858	2.27395
810609	342500	COWEETA, NORTH CAROLINA	-4.6300	2.27395
820119	342500	COWEETA, NORTH CAROLINA	-3.7844	2.27395
790911	343460	PIEDMONT STATION, NORTH CAROLINA	-1.7440	1.60496
781226	343560	CLINTON STATION, NORTH CAROLINA	-3.9054	2.23757
790911	343560	CLINTON STATION, NORTH CAROLINA	-2.5197	2.23757
810609	343560	CLINTON STATION, NORTH CAROLINA	-2.7980	2.23757
830614	343560	CLINTON STATION, NORTH CAROLINA	-2.3022	2.23757
831115	343560	CLINTON STATION, NORTH CAROLINA	-3.2871	2.23757
790626	344160	FINLEY (A), NORTH CAROLINA	-2.0374	2.01359
790911	344160	FINLEY (A), NORTH CAROLINA	-2.4420	2.01359
791120	344160	FINLEY (A), NORTH CAROLINA	-3.8849	2.01359
790911	361760	DELAWARE, OHIO	-2.3713	1.63073
800226	361760	DELAWARE, OHIO	-3.2588	1.63073
810317	361760	DELAWARE, OHIO	-1.6738	1.63073
790710	364900	CALDWELL, OHIO	-1.4396	1.41621
790911	364900	CALDWELL, OHIO	-1.8395	1.41621

800226	364900	CALDWELL, OHIO	-1.9201	1.41621
781107	367160	WOOSTER, OHIO	1.7040	1.54256
790911	367160	WOOSTER, OHIO	-1.9709	1.54256
810609	367160	WOOSTER, OHIO	-1.7867	1.54256
800429	392940	KANE, PENNSYLVANIA	-4.8182	2.14533
830628	392940	KANE, PENNSYLVANIA	-5.1852	2.14533
791120	501860	PARSONS, WEST VIRGINIA	-1.8518	1.68647
821228	501860	PARSONS, WEST VIRGINIA	-2.3484	1.68647
831011	501860	PARSONS, WEST VIRGINIA	-1.6924	1.68647

Seasonal residuals of calcium that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
830315	058840	DAVIS, CALIFORNIA	2.0374	2.00680
811201	062120	MANITOU, COLORADO	2.7307	2.23760
810929	100360	BRADFORD FOREST, FLORIDA	3.0485	1.96395
811027	100360	BRADFORD FOREST, FLORIDA	1.9698	1.96395
810210	114140	GEORGIA STATION, GEORGIA	-2.2970	2.21512
810317	114140	GEORGIA STATION, GEORGIA	2.3300	2.21512
810922	114140	GEORGIA STATION, GEORGIA	3.2082	2.21512
811110	235340	WELLSTON, MICHIGAN	2.9764	2.33610
820427	235340	WELLSTON, MICHIGAN	2.4582	2.33610
811201	241660	MARCELL, MINNESOTA	2.5380	2.15658
791106	281520	MEAD, NEBRASKA	-2.9498	2.32892
820511	300240	HUBBARD BROOK, NEW HAMPSHIRE	2.6369	2.34917
800205	340320	LEWISTON, NORTH CAROLINA	-2.1910	1.88936
820223	340320	LEWISTON, NORTH CAROLINA	-1.9884	1.88936
781024	343460	PIEDMONT STATION, NORTH CAROLINA	2.0501	1.95133
820706	343460	PIEDMONT STATION, NORTH CAROLINA	2.1062	1.95133
811124	343560	CLINTON STATION, NORTH CAROLINA	1.8987	1.78975
790102	361760	DELAWARE, OHIO	-2.7908	2.20816
831220	361760	DELAWARE, OHIO	2.2342	2.20816
780926	364900	CALDWELL, OHIO	2.2876	1.99144
801104	364900	CALDWELL, OHIO	2.2809	1.99144
780926	367160	WOOSTER, OHIO	2.0750	2.00212
790925	367160	WOOSTER, OHIO	-2.6243	2.00212
830301	392940	KANE, PENNSYLVANIA	2.6581	2.06107

Seasonal residuals of chloride that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
800318	058840	DAVIS, CALIFORNIA	-2.8388	2.66440
830517	062120	MANITOU, COLORADO	-2.2681	2.24749
791204	100360	BRADFORD FOREST, FLORIDA	-2.6739	2.19755
800123	100360	BRADFORD FOREST, FLORIDA	-2.7818	2.19755
810929	100360	BRADFORD FOREST, FLORIDA	2.3648	2.19755
810922	114140	GEORGIA STATION, GEORGIA	3.0472	2.36600
820302	114140	GEORGIA STATION, GEORGIA	-3.3811	2.36600
800429	235340	WELLSTON, MICHIGAN	-2.2880	1.95391
820427	235340	WELLSTON, MICHIGAN	2.0823	1.95391
830712	235340	WELLSTON, MICHIGAN	-2.7242	1.95391
810414	241660	MARCELL, MINNESOTA	-2.6136	2.18876
811201	241660	MARCELL, MINNESOTA	2.5261	2.18876
811229	241660	MARCELL, MINNESOTA	-2.2689	2.18876
820119	241660	MARCELL, MINNESOTA	-2.3767	2.18876
831101	241660	MARCELL, MINNESOTA	2.2814	2.18876
810428	281520	MEAD, NEBRASKA	-2.1829	2.02875
820706	281520	MEAD, NEBRASKA	-2.2533	2.02875
820803	281520	MEAD, NEBRASKA	2.5255	2.02875
780808	300240	HUBBARD BROOK, NEW HAMPSHIRE	3.5787	2.85811
780815	300240	HUBBARD BROOK, NEW HAMPSHIRE	3.6431	2.85811
811222	300240	HUBBARD BROOK, NEW HAMPSHIRE	-2.9003	2.85811
830621	300240	HUBBARD BROOK, NEW HAMPSHIRE	-2.9713	2.85811
830412	332020	HUNTINGTON, NEW YORK	2.4168	2.18161
800205	340320	LEWISTON, NORTH CAROLINA	-3.0552	2.75277
820119	342500	COWEETA, NORTH CAROLINA	-2.6539	2.55805
820302	342500	COWEETA, NORTH CAROLINA	-2.7430	2.55805
800401	343460	PIEDMONT STATION, NORTH CAROLINA	2.5880	2.44355
830329	343560	CLINTON STATION, NORTH CAROLINA	2.7481	2.67711
831220	343560	CLINTON STATION, NORTH CAROLINA	2.7427	2.67711
800205	344160	FINLEY (A), NORTH CAROLINA	-2.6548	2.55820
801223	344160	FINLEY (A), NORTH CAROLINA	2.8235	2.55820
791211	361760	DELAWARE, OHIO	-2.4333	1.83553
810421	361760	DELAWARE, OHIO	-2.7547	1.83553
790206	364900	CALDWELL, OHIO	2.9566	1.84558
791211	364900	CALDWELL, OHIO	-2.1794	1.84558
800318	364900	CALDWELL, OHIO	-2.1355	1.84558
820119	364900	CALDWELL, OHIO	2.3686	1.84558
810421	367160	WOOSTER, OHIO	2.1245	1.93661
830322	367160	WOOSTER, OHIO	-2.8238	1.93661
830517	367160	WOOSTER, OHIO	-2.5128	1.93661
780725	392940	KANE, PENNSYLVANIA	3.0130	2.46391
780801	392940	KANE, PENNSYLVANIA	3.8396	2.46391
780808	392940	KANE, PENNSYLVANIA	3.4271	2.46391
780815	392940	KANE, PENNSYLVANIA	2.5183	2.46391
811027	392940	KANE, PENNSYLVANIA	-2.7465	2.46391
820119	392940	KANE, PENNSYLVANIA	-2.6478	2.46391
830301	392940	KANE, PENNSYLVANIA	2.7726	2.46391
810519	501860	PARSONS, WEST VIRGINIA	-2.7118	2.00074
830524	501860	PARSONS, WEST VIRGINIA	-2.4332	2.00074

Seasonal residuals of ammonia that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
790109	058840	DAVIS, CALIFORNIA	-2.7047	2.36620
790410	058840	DAVIS, CALIFORNIA	-2.7380	2.36620
791009	058840	DAVIS, CALIFORNIA	-3.1604	2.36620
800318	058840	DAVIS, CALIFORNIA	-3.6577	2.36620
790313	114140	GEORGIA STATION, GEORGIA	4.3179	3.75944
790306	235340	WELLSTON, MICHIGAN	-3.5945	3.43752
790919	235340	WELLSTON, MICHIGAN	-3.8406	3.43752
800318	235340	WELLSTON, MICHIGAN	-3.5200	3.43752
800422	235340	WELLSTON, MICHIGAN	-3.6324	3.43752
810721	235340	WELLSTON, MICHIGAN	-4.0194	3.43752
820427	235340	WELLSTON, MICHIGAN	-3.7258	3.43752
791204	281520	MEAD, NEBRASKA	-4.5943	2.82185
820921	281520	MEAD, NEBRASKA	-4.1192	2.82185
830419	281520	MEAD, NEBRASKA	-4.7340	2.82185
830621	281520	MEAD, NEBRASKA	-4.0050	2.82185
790612	332020	HUNTINGTON, NEW YORK	-4.0596	3.09589
800624	332020	HUNTINGTON, NEW YORK	-3.5498	3.09589
800617	340320	LEWISTON, NORTH CAROLINA	-4.2142	3.62020
810428	342500	COWEETA, NORTH CAROLINA	-3.6124	3.56270
790410	343460	PIEDMONT STATION, NORTH CAROLINA	-3.3800	3.20897
790626	343460	PIEDMONT STATION, NORTH CAROLINA	-3.6341	3.20897
830628	343460	PIEDMONT STATION, NORTH CAROLINA	-4.1635	3.20897
830726	343460	PIEDMONT STATION, NORTH CAROLINA	-3.9385	3.20897
810609	343560	CLINTON STATION, NORTH CAROLINA	-4.1491	3.35121
790424	344160	FINLEY (A), NORTH CAROLINA	-3.1085	2.86430
790626	344160	FINLEY (A), NORTH CAROLINA	-3.4571	2.86430
800401	344160	FINLEY (A), NORTH CAROLINA	-4.2260	2.86430
830705	344160	FINLEY (A), NORTH CAROLINA	-3.7679	2.86430
790612	361760	DELAWARE, OHIO	-4.7862	3.16960
790918	361760	DELAWARE, OHIO	-3.4728	3.16960
791127	361760	DELAWARE, OHIO	-3.2189	3.16960
800226	361760	DELAWARE, OHIO	-3.5677	3.16960
820810	361760	DELAWARE, OHIO	-4.4024	3.16960
830419	361760	DELAWARE, OHIO	-4.1242	3.16960
800129	364900	CALDWELL, OHIO	-3.5056	2.92186
800226	364900	CALDWELL, OHIO	-3.4151	2.92186
800701	364900	CALDWELL, OHIO	-3.8660	2.92186
830329	364900	CALDWELL, OHIO	-3.1025	2.92186
830920	364900	CALDWELL, OHIO	-3.1860	2.92186
830927	364900	CALDWELL, OHIO	-4.0804	2.92186
790911	367160	WOOSTER, OHIO	-2.6382	2.60190
791204	367160	WOOSTER, OHIO	-3.4771	2.60190
800108	367160	WOOSTER, OHIO	-2.7172	2.60190
800318	367160	WOOSTER, OHIO	-3.0957	2.60190
810609	367160	WOOSTER, OHIO	-4.2840	2.60190
810901	367160	WOOSTER, OHIO	-3.0436	2.60190
790828	392940	KANE, PENNSYLVANIA	-3.7405	3.19318
800722	392940	KANE, PENNSYLVANIA	-3.3064	3.19318
830628	392940	KANE, PENNSYLVANIA	-3.2423	3.19318
800624	501860	PARSONS, WEST VIRGINIA	-3.5520	3.36660
810519	501860	PARSONS, WEST VIRGINIA	-4.0102	3.36660

Seasonal residuals of sodium that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
790327	062120	MANITOU, COLORADO	3.5099	3.16664
810609	100360	BRADFORD FOREST, FLORIDA	2.3007	2.28862
791204	114140	GEORGIA STATION, GEORGIA	3.2337	2.88736
781121	235340	WELLSTON, MICHIGAN	3.7870	3.61363
791211	241660	MARCELL, MINNESOTA	3.9938	3.54894
800527	241660	MARCELL, MINNESOTA	3.7163	3.54894
800812	241660	MARCELL, MINNESOTA	-3.7161	3.54894
800708	332020	HUNTINGTON, NEW YORK	-3.7831	3.20397
800902	332020	HUNTINGTON, NEW YORK	3.2874	3.20397
810127	332020	HUNTINGTON, NEW YORK	-3.5440	3.20397
800205	340320	LEWISTON, NORTH CAROLINA	-3.5989	3.03585
810106	342500	COWEETA, NORTH CAROLINA	-4.8941	3.39674
800401	344160	FINLEY (A), NORTH CAROLINA	3.5330	3.31796
801118	361760	DELAWARE, OHIO	-2.9611	2.91093
790206	364900	CALDWELL, OHIO	3.1574	3.09455
791106	364900	CALDWELL, OHIO	3.3080	3.09455
791113	364900	CALDWELL, OHIO	3.5552	3.09455
781017	367160	WOOSTER, OHIO	3.4382	3.24830
791211	367160	WOOSTER, OHIO	3.4773	3.24830
791113	392940	KANE, PENNSYLVANIA	3.0797	2.96531
800205	392940	KANE, PENNSYLVANIA	3.1029	2.96531
791030	501860	PARSONS, WEST VIRGINIA	3.2193	3.17419
791127	501860	PARSONS, WEST VIRGINIA	3.3554	3.17419
800415	501860	PARSONS, WEST VIRGINIA	3.6309	3.17419
801118	501860	PARSONS, WEST VIRGINIA	-3.7551	3.17419

Seasonal residuals of potassium that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
791120	058840	DAVIS, CALIFORNIA	2.7497	2.14541
830315	058840	DAVIS, CALIFORNIA	2.5118	2.14541
830510	062120	MANITOU, COLORADO	3.1287	2.90061
781017	100360	BRADFORD FOREST, FLORIDA	3.2790	2.40535
820921	100360	BRADFORD FOREST, FLORIDA	3.1594	2.40535
830920	100360	BRADFORD FOREST, FLORIDA	2.6576	2.40535
790313	114140	GEORGIA STATION, GEORGIA	4.5385	3.46792
800429	114140	GEORGIA STATION, GEORGIA	-3.7684	3.46792
801028	114140	GEORGIA STATION, GEORGIA	3.5159	3.46792
810728	114140	GEORGIA STATION, GEORGIA	3.5348	3.46792
831115	114140	GEORGIA STATION, GEORGIA	3.8864	3.46792
810210	235340	WELLSTON, MICHIGAN	-2.9566	2.20863
811110	235340	WELLSTON, MICHIGAN	2.5077	2.20863
820427	235340	WELLSTON, MICHIGAN	3.0228	2.20863
791002	241660	MARCELL, MINNESOTA	2.3227	2.15039
831101	241660	MARCELL, MINNESOTA	2.3120	2.15039
791106	281520	MEAD, NEBRASKA	-2.9804	2.34440
810331	281520	MEAD, NEBRASKA	2.4584	2.34440
790508	332020	HUNTINGTON, NEW YORK	2.7184	2.28384
790703	332020	HUNTINGTON, NEW YORK	-2.3139	2.28384
790821	332020	HUNTINGTON, NEW YORK	-2.5865	2.28384
810203	332020	HUNTINGTON, NEW YORK	-2.4966	2.28384
790926	340320	LEWISTON, NORTH CAROLINA	-2.3873	2.16397
821019	340320	LEWISTON, NORTH CAROLINA	3.0727	2.16397
790828	342500	COWEETA, NORTH CAROLINA	2.7290	2.58304
810407	342500	COWEETA, NORTH CAROLINA	-3.1171	2.58304
800401	343460	PIEDMONT STATION, NORTH CAROLINA	4.5158	2.80989
800408	343460	PIEDMONT STATION, NORTH CAROLINA	3.5221	2.80989
801112	343460	PIEDMONT STATION, NORTH CAROLINA	3.9099	2.80989
830524	343460	PIEDMONT STATION, NORTH CAROLINA	3.0774	2.80989
830719	343460	PIEDMONT STATION, NORTH CAROLINA	2.9584	2.80989
800930	343560	CLINTON STATION, NORTH CAROLINA	2.4913	2.16870
830322	344160	FINLEY (A), NORTH CAROLINA	3.1739	2.13604
790911	361760	DELAWARE, OHIO	-2.5893	2.47920
810203	361760	DELAWARE, OHIO	-2.6886	2.47920
810210	361760	DELAWARE, OHIO	-2.5931	2.47920
831011	364900	CALDWELL, OHIO	2.5043	2.49702
790710	367160	WOOSTER, OHIO	-2.4522	2.37191
810210	367160	WOOSTER, OHIO	-2.7020	2.37191
781205	392940	KANE, PENNSYLVANIA	-2.4657	2.23401
801021	392940	KANE, PENNSYLVANIA	-2.4730	2.23401
821026	392940	KANE, PENNSYLVANIA	3.4205	2.23401
830301	392940	KANE, PENNSYLVANIA	2.3996	2.23401
831108	392940	KANE, PENNSYLVANIA	2.4429	2.23401
800429	501860	PARSONS, WEST VIRGINIA	2.5062	2.08078
810303	501860	PARSONS, WEST VIRGINIA	-2.0974	2.08078
810519	501860	PARSONS, WEST VIRGINIA	-3.3181	2.08078

Seasonal residuals of specific conductance that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
791211	058840	DAVIS, CALIFORNIA	1.8409	1.48777
811201	062120	MANITOU, COLORADO	1.6508	1.42347
800429	100360	BRADFORD FOREST, FLORIDA	2.4011	1.69933
810929	100360	BRADFORD FOREST, FLORIDA	2.1429	1.69933
811027	100360	BRADFORD FOREST, FLORIDA	1.7468	1.69933
810922	114140	GEORGIA STATION, GEORGIA	1.7852	1.64373
811201	241660	MARCELL, MINNESOTA	1.8438	1.54171
830802	281520	MEAD, NEBRASKA	1.4183	1.39486
830329	300240	HUBBARD BROOK, NEW HAMPSHIRE	-1.9541	1.89079
790612	332020	HUNTINGTON, NEW YORK	-2.0360	1.80419
790703	332020	HUNTINGTON, NEW YORK	-1.9359	1.80419
820907	342500	COWEETA, NORTH CAROLINA	-1.7117	1.58308
831018	342500	COWEETA, NORTH CAROLINA	-1.5935	1.58308
790925	344160	FINLEY (A), NORTH CAROLINA	-1.7912	1.77998
800226	361760	DELAWARE, OHIO	-1.5392	1.39529
800226	364900	CALDWELL, OHIO	-1.4727	1.22883
781031	367160	WOOSTER, OHIO	1.3186	1.29511
781107	367160	WOOSTER, OHIO	1.5047	1.29511
791120	501860	PARSONS, WEST VIRGINIA	-1.5960	1.52021
821228	501860	PARSONS, WEST VIRGINIA	-2.1431	1.52021
830927	501860	PARSONS, WEST VIRGINIA	-1.8470	1.52021
831011	501860	PARSONS, WEST VIRGINIA	-1.7912	1.52021

Seasonal residuals of pH that are greater than three standard deviations (±) from the mean

DATE	STATION	STATION NAME	RESIDUAL	3*STANDARD DEVIATION
800122	058840	DAVIS, CALIFORNIA	-2.2508	1.84011
800311	058840	DAVIS, CALIFORNIA	-2.0580	1.84011
811110	058840	DAVIS, CALIFORNIA	-2.8056	1.84011
821207	062120	MANITOU, COLORADO	-2.5681	1.86532
800722	100360	BRADFORD FOREST, FLORIDA	-1.8848	1.88440
820601	100360	BRADFORD FOREST, FLORIDA	-2.3362	1.88440
790313	114140	GEORGIA STATION, GEORGIA	2.5553	2.02748
790529	114140	GEORGIA STATION, GEORGIA	2.0951	2.02748
800429	114140	GEORGIA STATION, GEORGIA	2.0454	2.02748
820511	114140	GEORGIA STATION, GEORGIA	2.0465	2.02748
800415	235340	WELLSTON, MICHIGAN	2.2620	1.70186
800701	235340	WELLSTON, MICHIGAN	1.7577	1.70186
820427	235340	WELLSTON, MICHIGAN	1.9330	1.70186
790605	241660	MARCELL, MINNESOTA	-2.5077	1.75334
800108	241660	MARCELL, MINNESOTA	-2.6732	1.75334
791106	281520	MEAD, NEBRASKA	-2.8545	2.31522
790612	300240	HUBBARD BROOK, NEW HAMPSHIRE	1.9859	1.55306
800624	300240	HUBBARD BROOK, NEW HAMPSHIRE	1.6904	1.55306
810825	300240	HUBBARD BROOK, NEW HAMPSHIRE	1.8019	1.55306
820810	300240	HUBBARD BROOK, NEW HAMPSHIRE	1.6526	1.55306
830308	300240	HUBBARD BROOK, NEW HAMPSHIRE	-1.7150	1.55306
830719	300240	HUBBARD BROOK, NEW HAMPSHIRE	1.7921	1.55306
831220	300240	HUBBARD BROOK, NEW HAMPSHIRE	-1.6757	1.55306
790612	332020	HUNTINGTON, NEW YORK	1.6878	1.42263
800205	332020	HUNTINGTON, NEW YORK	-2.4030	1.42263
800429	332020	HUNTINGTON, NEW YORK	1.6810	1.42263
800624	332020	HUNTINGTON, NEW YORK	1.5485	1.42263
810707	332020	HUNTINGTON, NEW YORK	1.4696	1.42263
830208	332020	HUNTINGTON, NEW YORK	2.3346	1.42263
790703	340320	LEWISTON, NORTH CAROLINA	1.9493	1.72228
790814	340320	LEWISTON, NORTH CAROLINA	-1.8767	1.72228
800729	340320	LEWISTON, NORTH CAROLINA	1.9531	1.72228
800909	340320	LEWISTON, NORTH CAROLINA	1.7276	1.72228
810623	340320	LEWISTON, NORTH CAROLINA	2.1402	1.72228
810825	340320	LEWISTON, NORTH CAROLINA	1.9792	1.72228
790612	342500	COWEETA, NORTH CAROLINA	1.9505	1.50772
820914	342500	COWEETA, NORTH CAROLINA	-2.4695	1.50772
831018	342500	COWEETA, NORTH CAROLINA	-1.8633	1.50772
831025	342500	COWEETA, NORTH CAROLINA	1.6292	1.50772
800401	343460	PIEDMONT STATION, NORTH CAROLINA	2.8041	1.85729
800408	343460	PIEDMONT STATION, NORTH CAROLINA	2.0371	1.85729
830524	343460	PIEDMONT STATION, NORTH CAROLINA	2.6085	1.85729
830719	343460	PIEDMONT STATION, NORTH CAROLINA	2.2050	1.85729
781226	343560	CLINTON STATION, NORTH CAROLINA	-2.1875	1.80298
790313	343560	CLINTON STATION, NORTH CAROLINA	2.2603	1.80298
810428	343560	CLINTON STATION, NORTH CAROLINA	1.9452	1.80298
810623	343560	CLINTON STATION, NORTH CAROLINA	2.2397	1.80298
790911	344160	FINLEY (A), NORTH CAROLINA	1.8095	1.74476

820504	344160	FINLEY (A), NORTH CAROLINA	2.3742	1.74476
790904	361760	DELAWARE, OHIO	1.4107	1.32039
801216	361760	DELAWARE, OHIO	1.3437	1.32039
810310	361760	DELAWARE, OHIO	2.3633	1.32039
810317	361760	DELAWARE, OHIO	2.1589	1.32039
820420	361760	DELAWARE, OHIO	2.2626	1.32039
820810	361760	DELAWARE, OHIO	1.3281	1.32039
831220	361760	DELAWARE, OHIO	1.9273	1.32039
780926	364900	CALDWELL, OHIO	1.68844	1.26122
790904	364900	CALDWELL, OHIO	1.31649	1.26122
800429	364900	CALDWELL, OHIO	3.33919	1.26122
801104	364900	CALDWELL, OHIO	2.09231	1.26122
810317	364900	CALDWELL, OHIO	1.73040	1.26122
811027	364900	CALDWELL, OHIO	1.78375	1.26122
790227	367160	WOOSTER, OHIO	1.52575	1.23046
790724	367160	WOOSTER, OHIO	1.83625	1.23046
790918	367160	WOOSTER, OHIO	1.65159	1.23046
810310	367160	WOOSTER, OHIO	1.50938	1.23046
811006	367160	WOOSTER, OHIO	1.79926	1.23046
811013	367160	WOOSTER, OHIO	1.34872	1.23046
800429	392940	KANE, PENNSYLVANIA	2.32879	1.23990
820427	392940	KANE, PENNSYLVANIA	2.50813	1.23990
830301	392940	KANE, PENNSYLVANIA	1.90050	1.23990
791016	501860	PARSONS, WEST VIRGINIA	1.69146	1.01491
791226	501860	PARSONS, WEST VIRGINIA	1.63128	1.01491
810825	501860	PARSONS, WEST VIRGINIA	1.24563	1.01491
821116	501860	PARSONS, WEST VIRGINIA	1.34026	1.01491
821228	501860	PARSONS, WEST VIRGINIA	1.31005	1.01491