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PATTERNS OF ACID DEPOSITION VARIABILITY
IN THE EASTERN UNITED STATES, 1981-84

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Patterns of Acid Deposition Variability
in the Eastern United States, 1981-84

by

Harry F. Lins^{1/}, Kenneth J. Lanfear^{1/}, and Terry L. Schertz^{2/}

ABSTRACT

An increase in pH and a decrease in sulfate concentration of precipitation were recorded at National Atmospheric Deposition Program and National Trends Network (NADP/NTN) monitoring sites in the Eastern United States between 1981 and 1984. The decline in acidity, however, was not spatially or temporally uniform. The range in acidity and sulfate concentrations decreased during the 4-year period. Variations in the area of constant pH surfaces take the general form of area reductions in both the lower (pH 4.01-4.40) and upper (pH 4.91-5.40) range of values with concomitant area increases in the middle (pH 4.41-4.90) range. The pattern for sulfate is simpler, with area increases occurring in the lower (1.0-1.9 milligrams per liter) range, decreases in the upper (2.5-4.4 milligrams per liter) range, with approximate stability in the middle (2.0-2.4 milligrams per liter) range of values.

INTRODUCTION

Several studies in recent years have characterized spatial and temporal trends in precipitation chemistry data. Most of these studies have focused on producing contour maps of annual mean concentrations of a variety of acid-forming constituents in precipitation. Qualitative comparisons of the maps are made to identify regional patterns that appear to change from year to year. In all such studies, the contour or isopleth maps are derived from

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interpolation of a field of data points corresponding to the sampling network. In some instances, sophisticated geostatistical models, such as those based on Kriging, have been used to estimate the spatial field.

An alternative assessment of trend in acid precipitation data has been performed by Schertz and Hirsch (1985). Rather than extrapolating point values to an area by contouring, and then subjectively looking for interannual changes in the position of specific contour lines, Schertz and Hirsch evaluated trend direction and magnitude at each sampling point by using a nonparametric statistical procedure. No attempt was made to estimate precipitation chemistry trends beyond the individual sampling points.

Each of the above trend assessment procedures has strengths and weaknesses. The contouring approach provides a more complete synoptic picture of precipitation chemistry spatial patterns and does so for each period of interest. The procedure, however, is subject to the vagaries and inaccuracies that result from extrapolation and generalization of a limited number of discrete point values to a continuous spatial field. It also lacks the objectivity of a robust statistical procedure. The purely statistical approach to trend assessment, on the other hand, provides a robust estimate of trend at a specified level of confidence, but does not provide spatial information beyond the points where data have been collected. Conceptually, the two approaches are complementary. Their complementarity is enhanced if more detailed areal information (beyond what the eye perceives) can be extracted from the spatial generalization or contouring approach.

A method for providing such an enhancement, vis-a-vis spatially generalized precipitation sulfate and pH data, is demonstrated in this report. The system makes use of some of the more rudimentary capabilities of geographic information systems (GIS) technology.

APPROACH

The basic procedure begins with the preparation of contour maps of precipitation volume-weighted means of annual sulfate concentration and pH for the Eastern United States. The contours are fit to a rectangular grid of values estimated by a distance-weighted linear interpolation scheme. At this point, which is traditionally where investigators make subjective interpretations and comparisons of maps, the procedure is taken another step. The field of estimated grid values of precipitation SO_4 and pH is put into an interactive geographic information system (GIS). The GIS is used to produce the contour maps and, more importantly, to produce tabular summaries of the areas within constant isopleth surfaces or within specified isopleth intervals. Thus, the spatial extent of precipitation acidity is quantified and areal changes, both absolute and relative, between time periods can be measured.

Data Base

The atmospheric deposition data used in this study were obtained from the Environmental Protection Agency's Acid Deposition System (ADS) at Pacific Northwest Laboratories. This system was established to provide a single location for deposition data from various networks in North America and to make these data accessible to the general research community. The majority of the data used in this study came from sites operated by the National Atmospheric Deposition Program (NADP) and/or the National Trends Network (NTN), but a few sites were from the Canadian Network for Sampling Acid Precipitation (CANSAP). NADP was established in 1978 under the sponsorship of the State Agricultural Experiment Stations. NTN was established in 1982 by the U.S. Geological Survey to provide a long-term, continuous set of data to

be used in documenting and understanding changes in the chemistry of atmospheric deposition. The NADP chemical analysis laboratory, sample collection procedures, analytical procedures, and many of the stations operated by the NADP were incorporated into the NTN. The methods of collection of wet deposition data for stations in the NADP/NTN programs are described by Bigelow (1982). The processing of each sample and the quality-assurance used on the resulting analysis are described by Schertz and Hirsch (1985). The period of study was from 1980 to 1984, but data from the Canadian sites were not available for 1984. Site selection for that year was done from the NTN and NADP networks only.

Site Selection and Data Evaluation

The selection of sites for each year of analysis was based on an overall quality level assigned to the data from each site by ADS. The overall data quality level is derived from the site representativeness and data completeness levels. Three levels of evaluation are used for site representativeness. Level one sites are regionally representative sites that are not affected by any known interference. Level two is the level assigned to a large portion of the existing sites for which site representativeness is difficult to assess. Level three sites are regionally unrepresentative sites with local influences known or strongly suspected to have a significant effect on concentration and deposition. There are four levels of data completeness. A level one site has the best information and has come the closest to meeting the planned program of data acquisition. The least confidence is given to a level four site, which is not generally viewed as providing representative data for the time period. Five data completeness measures are used to determine the data completeness level. Criteria are set for (1) the percent of precipitation coverage length, (2) valid sample length, (3) samples with measured

precipitation that are valid samples, (4) total precipitation associated with valid samples, and (5) collection efficiency. The combination of the levels of the measures are used to determine the level assigned to the site for data completeness. Each of these measures is described in detail by Olsen and Slavich (1985). The data from any site that had an overall quality level of one or two (the highest quality), was included in the estimation of the contour location for that year.

Some additional screening of the data from the selected sites was done by the authors before the precipitation volume-weighted means were calculated. All samples flagged as bulk samples were deleted and all samples that had measured components reported, but volume of precipitation missing, were deleted.

SPATIAL INTERPOLATION OF POINT DATA

Surface II* Graphics System is a computer software system for creation of displays of spatially distributed data. The basis for all operations in Surface II is a rectangular grid of values which is a numerical representation of the surface to be displayed. Contour lines are laced through the grid by linearly interpolating between grid nodes to locate the points where a contour line of specified value will cross the edge of a grid cell. The elements in the grid matrix are estimated by a nearest neighbor search of the sample data points around each element. The search procedure calculates the straight-line distance from the location of a grid node to the surrounding data points. The closest ten data points are used to estimate the value of the grid node.

* Mention of trade names is for identification purposes only and is not an endorsement by the U.S. Geological Survey.

For this study, annual precipitation volume-weighted means of SO_4 concentration and pH were calculated for each site. The latitude and longitude coordinates of each site were converted to rectangular coordinates for plotting. The projection used for the conversion was a Lambert conformal conic projection. The sites used varied for each year of the study, so corner points were used to keep the conversion to plot units uniform for all of the maps.

The Surface II Graphics System produced computer files for making plots of SO_4 and pH contours, along with reference points of known latitude and longitude. Geographical reference of the contours to the earth's surface was required so that measurements of area could be made. Each plot file consisted of pairs of x,y coordinates (in plotter units) and pen movement instructions. After a simple format conversion, the plot files were read into the ARC/INFO Geographic Information System to form digital maps, or coverages. With the interactive graphics editor, ARCEDIT, the reference points were identified and each contour line coded with its proper SO_4 or pH value. The coverages were then registered to the reference points and projected into a standard Albers equal-area projection for the United States.

Closure lines were added so that the contours formed the boundaries of polygons. The minimum and maximum SO_4 and pH values of the enclosing contour lines were determined for each polygon, and saved as polygon attributes. Finally, all polygons were "clipped" to the base map of the conterminous United States (derived from Domaratz and others, 1983) shown in figures 1 and 2, so that all calculations would be made on the same basis. Tables 1 and 2 were computed by summing the polygon areas within each specified contour.

pH

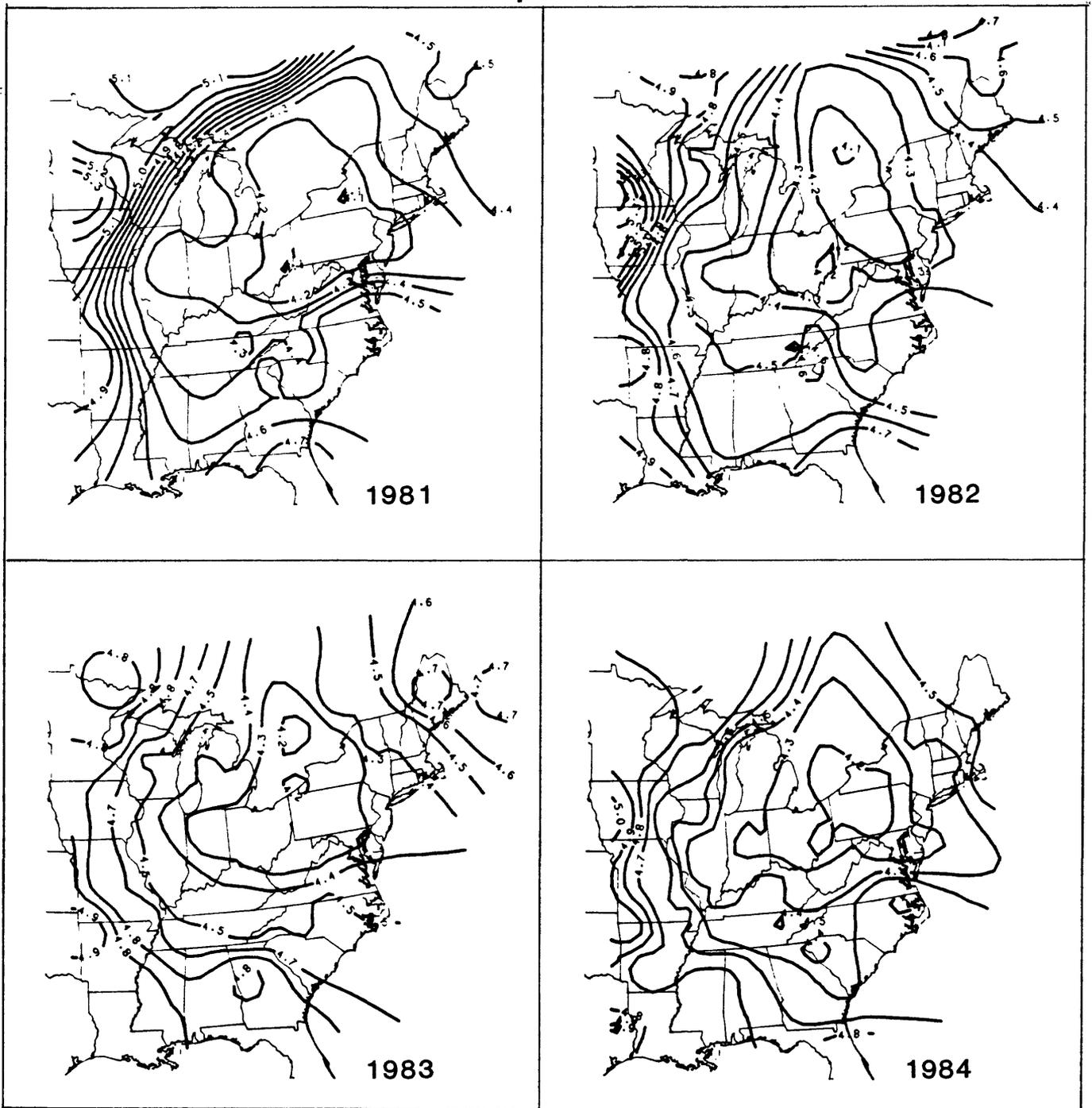


Figure 1.--Mean annual pH of precipitation for the Eastern United States and Southeastern Canada, 1981-84.

SO₄

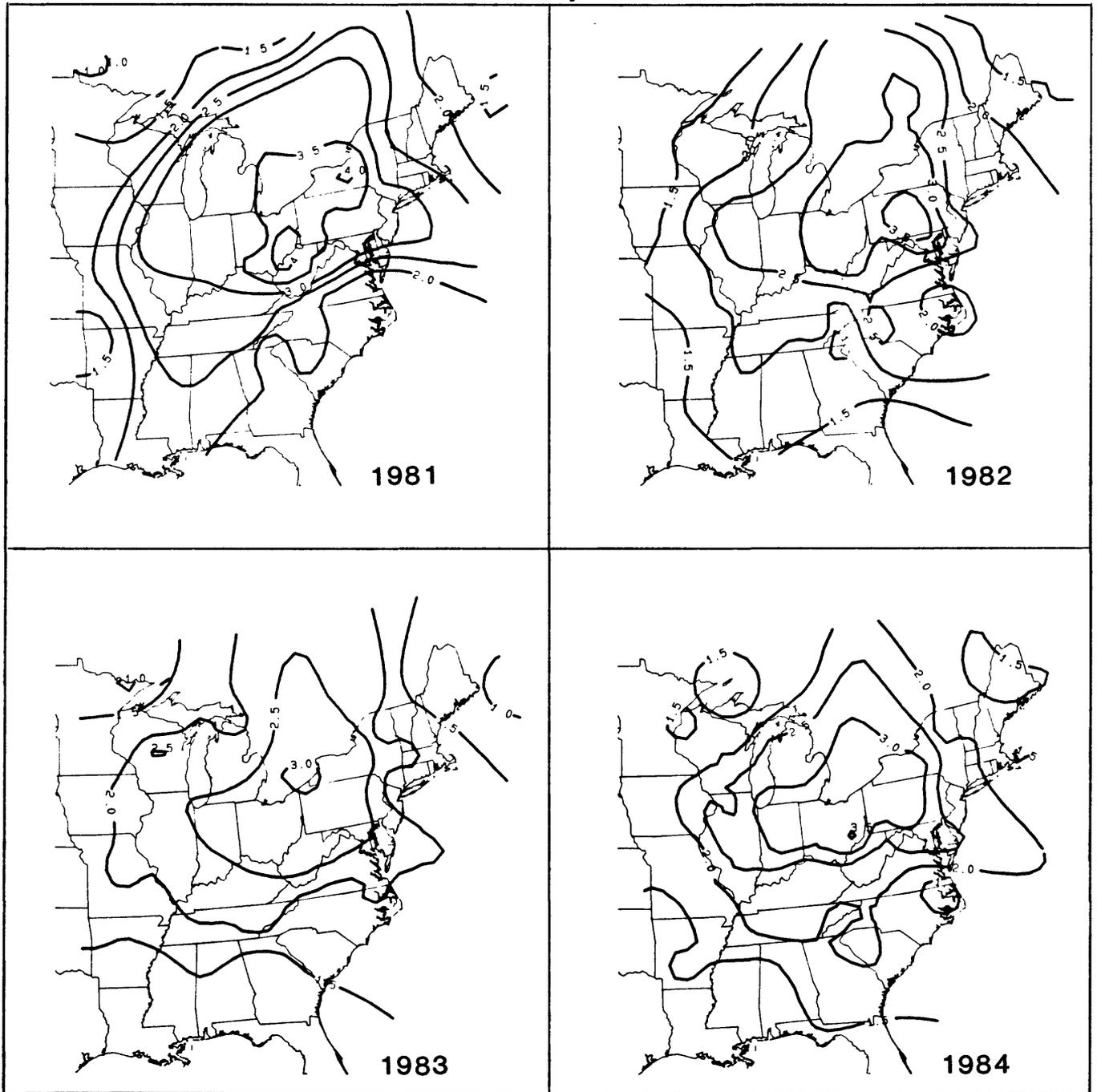


Figure 2.--Mean annual sulfate concentration of precipitation for the Eastern United States and Southeastern Canada, 1981-84.

Table 1. Area values of constant pH surfaces depicted in Figure 1, by year.

pH Surface	Area (km ²)			
	1981	1982	1983	1984
4.01 - 4.10	1,538	0	0	0
4.11 - 4.20	341,112	125,169	4,039	126,125
4.21 - 4.30	401,579	299,907	592,809	274,265
4.31 - 4.40	525,000	415,077	318,758	467,922
4.41 - 4.50	370,822	493,615	522,783	451,570
4.51 - 4.60	392,125	590,855	371,147	464,924
4.61 - 4.70	180,856	322,165	38,318	362,366
4.71 - 4.80	154,032	188,580	851,638	394,528
4.81 - 4.90	109,982	173,009	375,913	258,820
4.91 - 5.00	124,103	150,397	0	64,742
5.01 - 5.10	130,430	35,961	0	8,145
5.11 - 5.20	68,078	30,160	0	0
5.21 - 5.30	44,034	25,824	0	0
5.31 - 5.40	31,715	24,596	0	0

Table 2. Area values of constant SO₄ surfaces depicted in Figure 2, by year.

SO ₄ (mg/L) Surface	Area (km ²)			
	1981	1982	1983	1984
1.0 - 1.4	151,123	460,754	752,375	468,097
1.5 - 1.9	782,647	941,647	896,422	1,185,835
2.0 - 2.4	641,893	774,780	777,673	602,645
2.5 - 2.9	510,589	393,595	442,119	319,193
3.0 - 3.4	567,645	255,245	6,812	298,883
3.5 - 3.9	191,198	49,380	0	755
4.0 - 4.4	30,012	0	0	0

SPATIAL AND TEMPORAL PATTERNS OF ACID PRECIPITATION

During the period 1981-1984 an increase in the pH and decrease in the sulfate concentration of precipitation was recorded at NADP/NTN monitoring sites across the Eastern United States. The pattern is manifest most apparently in pH and SO_4 isopleth maps (figs. 1 and 2) as a "shrinking bull's-eye" of the higher acidity contours. This decline in acidity was not, however, uniform spatially or temporally. Moreover, it was not consistent over all ranges of acidity. We focus on describing these nonuniformities using graphical display and tabular summaries of the precipitation chemistry data.

We consider first the spatial characteristics of precipitation pH and SO_4 (figs. 1 and 2). The general pattern for both variables, within any one year is very similar. The lowest pH and highest sulfate values are centered on the region encompassing the Upper Ohio River Valley and Lower Great Lakes. The highest pH and lowest sulfate values are generally found in the Upper Mississippi River Valley. The area of most acidic conditions for both variables tends to be rather broad with gradient increases occurring with proximity to the Upper Great Lakes, Middle Mississippi Valley, and Mid-Atlantic Coastal Plain regions. The steepest gradient axis is generally aligned northwest-southeast with the shallowest running northeast-southwest.

Notably, this spatial pattern of acidity maximum, minimum, and gradient alignments conforms very closely to the distributional pattern of sulfur dioxide emission loads. In analyzing a new set of monthly state SO_2 emissions data, Lins (in press) calculated area-normalized emission loads that revealed a pattern with an emissions maximum centered on the Upper Ohio River Valley and Lower Great Lakes, low relative emissions in the Upper Mississippi River Valley as well as in the Lower Mississippi Valley and in much of New England,

and gradients that were steepest in the northwest-southeast direction and shallowest in the northeast-southwest direction. The spatial correspondence between SO₂ emissions and the two acid-deposition variables is suggestive of an association that, in a longer-term (i.e. annual) average context, exists principally at the mesoscale.

From a temporal perspective the patterns of change in the isopleth maps are less distinct. For example, in looking at the core region of highest acidity, the area within the most acid isopleths appears to decrease between 1981 and 1984. This decrease is not uniform, however, in that the decrease only occurs between 1981 and 1983. The areas within the relatively high acid contours (e.g. ≤ 4.2 pH surface, ≥ 3.0 SO₄ surface) actually increase in 1984, although the increase leaves the areas of these contours short of their 1982 values. The more moderate and low ranges of acidity values vary differently from this pattern making generalizations about the overall variability difficult to infer from the isopleth maps.

Characteristics of Temporal Variability

To overcome the problems of subjectivity and inaccuracy associated with visually discriminating interannual changes in contours, area values of constant pH and SO₄ surfaces were calculated digitally using the ARC/INFO geographic information system. The area values, calculated for the conterminous United States east of the 95th meridian and north of the 28th parallel, appear in tables 1 and 2. Graphically, these same data are plotted as ogives in figures 3 and 4. Variations in pH (table 1, figure 3) through the 4-year period take the general form of area reductions in both the lower (4.01-4.40) and upper (4.91-5.40) range of values with concomitant increases in the middle (4.41-4.90) range. The pattern with SO₄ (table 2, figure 4) is somewhat

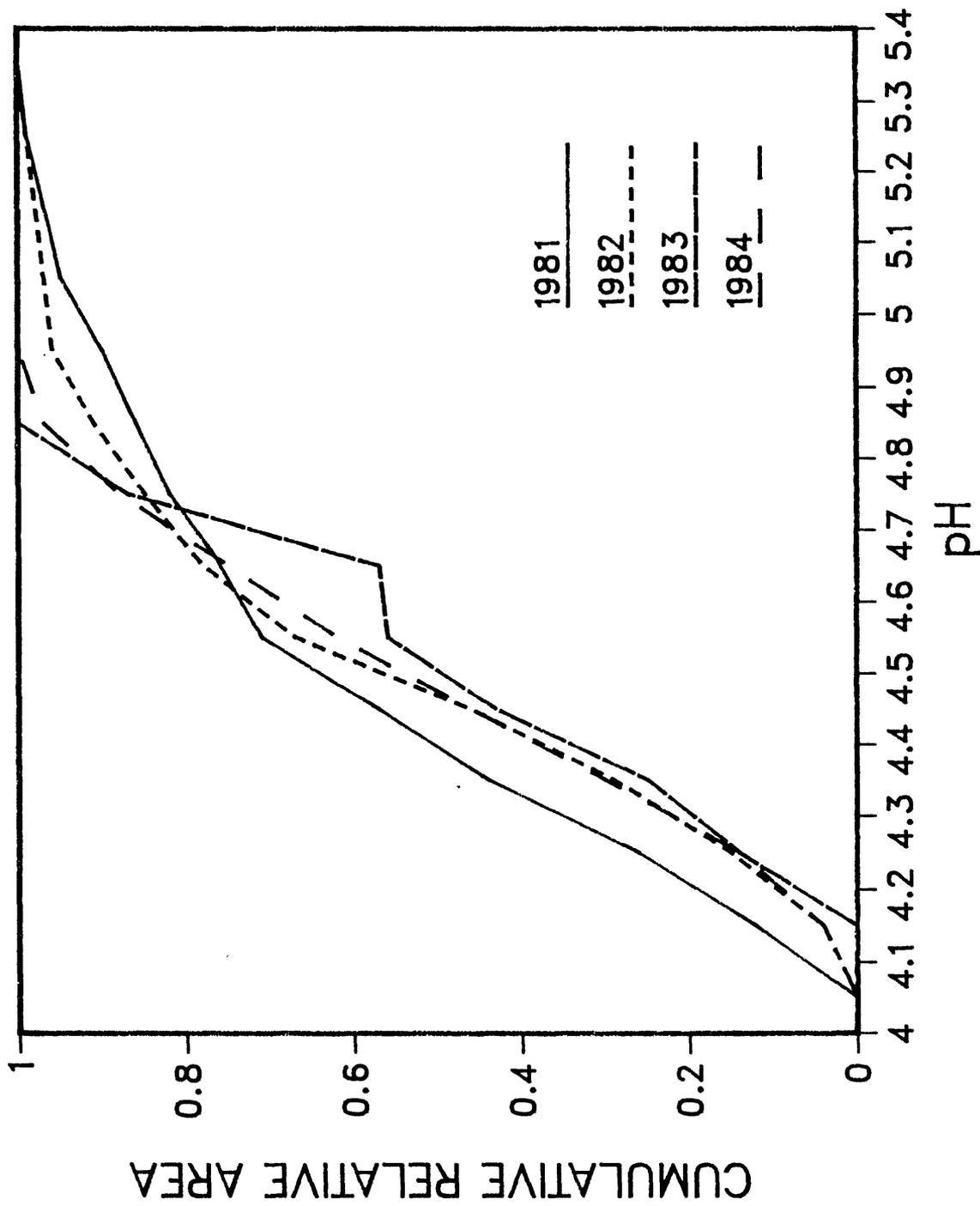


Figure 3.--Ogive of precipitation pH versus area for the Eastern United States during the years 1981-84.

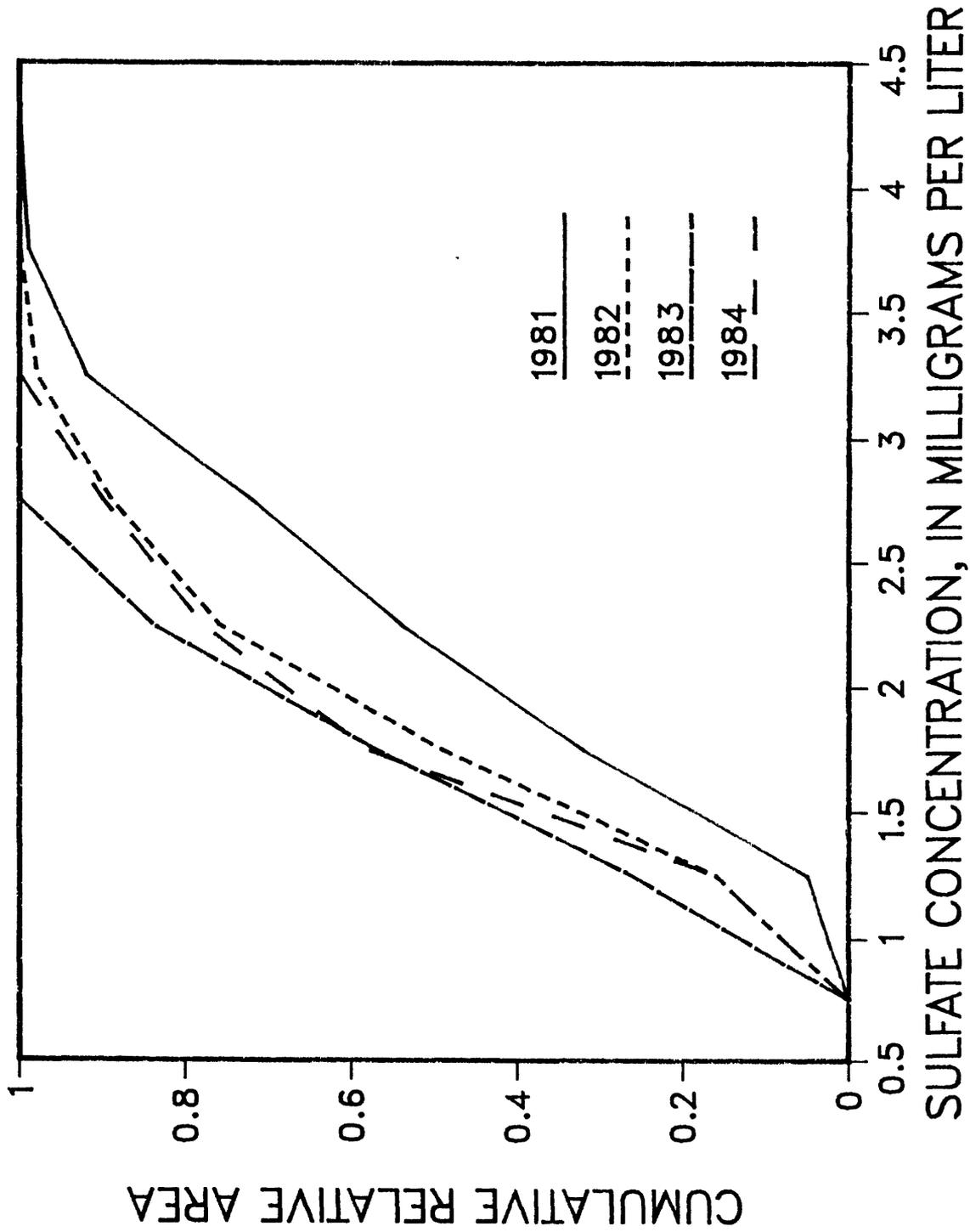


Figure 4,--Ogive of the concentration of SO_4 in precipitation versus area for the Eastern United States during the years 1981-84.

simpler; area increases occurred in the lower (1.0-1.9) range, decreases in the upper (2.5-4.4), with approximate stability in the middle (2.0-2.4) range of values. A distinct reduction in the range of both variables is evident in 1983, followed by a minor range expansion in 1984. Also, the decrease in the area of the most acid pH and SO₄ surfaces through 1983 followed by an increase in 1984, apparent in figures 1 and 2, is confirmed in the area summaries and graphical plots.

In looking at variations recorded at only four points in time, it is difficult to infer anything substantial about how and why the system is behaving in a particular manner. It is noteworthy, however, that sulfur dioxide emissions in the Eastern United States exhibited an almost identical temporal pattern of variation during the same 4-year period. Lins (in press) reported that the "national" SO₂ emissions pattern during the period 1975-1984 was one characterized by a slight emission increase through 1975 and 1976, followed by a consistent decline through early 1983, culminating in an emission increase in late 1983 and 1984. Although this general emission pattern was derived from data for each of the 48 States, Lins also included a description of the individual State patterns. For the 30 States east of the 95th meridian, all but 6 had SO₂ emission patterns that generally conformed to the deposition model. Although at this time the data used herein remain insufficient to statistically establish the form of a relation between SO₂ emissions and acid deposition, the data indicate that on State to multi-State spatial scales and over annual time scales, the direction (if not the magnitude) of variation in SO₂ emissions and acid deposition appear to parallel each other.

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