

ACID PRECIPITATION: ORIGINS AND DISTRIBUTION IN THE UNITED STATES

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We have often heard the saying, "What goes up must come down", but all too frequently have not given the concept too much thought. It now has become a very important issue not only in this country, but everywhere on the planet earth and is the center focus of the acid rain debate. The debate today does not question the statement given above, but rather addresses the tougher problem of "When something goes up how, when and where does it come down?". A related question might be "When it comes down, is it beneficial or harmful to its final resting place?". A further important question is "Is the acidity of precipitation increasing and is it spreading to larger areas in the U.S. and beyond its borders?". The answers to these questions are central to the establishment of policy regarding how to deal with the national and international concerns about acidic deposition.

To begin to address these and other questions, it is appropriate to very briefly examine what little is known about previous studies of the chemistry of precipitation. It must be borne in mind, however, that while there are some indications of great interest in atmospheric chemistry over many years, the technology for providing analyses of both the atmosphere and its precipitation is still improving. An implication of this is that it is most difficult to compare more recently acquired data with those of the relatively distant past to determine the extent and trend of changing precipitation chemistry.

It seems central to the on-going debates, both scientific and political, that the local and regional trend of important chemical ions in precipitation be determined in order that their contribution to the chemical cycle of local ecosystems can be more accurately assessed. Equally important to such an assessment is the quantification of the magnitude of the wet and dry deposition from the atmosphere and their ultimate disposition in the identified cycles.

In this brief paper, we will not attempt to address all of the complex biochemical and geochemical sciences, but we will attempt to describe the chemistry of precipitation as a single, and frequently minor, input to those chemical budgets of importance to the environment. We will also not attempt a comprehensive description of dry deposition as there is no single agreed-upon method for its measurements. Experience tells us that it does not precipitate all of the time, and in the eastern mid-latitude of the U.S. no precipitation is observed about 90% of the time during any

given year. However, experience also tells us that material from the atmosphere is continuously returning to the earth in dry form. Some preliminary data obtained from exposed bucket collections will be discussed as a "poor man's" estimate of a portion of the dry deposition.

PRECIPITATION CHEMISTRY BACKGROUND

There is early documentation of interest in the chemical composition of rain dating back to 1727, followed by a fairly thorough study of the subject in the mid-1800's (1). These early efforts focused on sulfur in rain, snow, and dew, and some of the perceived impacts on health and agriculture. The modern interest began in the post-World War II years when some monitoring of precipitation chemistry was initiated in the Scandinavian countries (2). A particular effort was put forth in Sweden with agriculturalists and meteorologists expressing equal interest in the chemistry of precipitation, but for entirely different reasons. The agricultural interests focused on the quantity of nutrients being deposited by precipitation as an aid to plant growth while the atmospheric scientists were attempting to use the chemistry to further their knowledge of the origin of precipitation water and to trace atmospheric motions. The national commitment of Sweden to continue the network operation placed them in the forefront of the emerging issues related to acid rain.

During the early quarter of this century, agriculturalists in the U.S. showed interest in the amount of nutrient falling on productive soils as evidenced by the literature in the 1920's (3,4). Following the lead of Sweden, some interest in the chemistry of precipitation was expressed by meteorologists in the U.S. which culminated in the establishment of a national network in 1955 (5,6). This network was operated for a single year (1955-1956) and the data have been used by some as the baseline data for demonstrating that acid rain is worsening and spreading in the eastern U.S. (7,8,9,).

A short time after the demise of this initial network a similar one was begun by the U.S. Public Health Service (PHS) in 1960. This network was maintained until 1966 with management responsibilities eventually falling upon the National Center for Atmospheric Research (NCAR) (10). No national network was operated after the closing of the PHS/NCAR stations in the 1960's although several isolated measurement programs were carried out in various parts of the country (11,12). An excellent summary of sampling activities in North America is available for additional information (13).

The First International Symposium on Acid Precipitation and the Forest Ecosystem in 1975 concluded with workshops addressing various needs to properly research the topic of acidic deposition in the U.S. (14). Among the recommendations from one of the workshops was that a network be established for the purpose of long-term monitoring of the precipitation chemistry across the U.S. and its territories. This currently operating network extends from Alaska to Puerto Rico, and from Maine to American

Samoa with 190 sampling sites. Many of the stations have been identified as National Trends Network (NTN) sites - the monitoring network of the National Acid Precipitation Assessment Program (NAPAP).

PRECIPITATION CHEMISTRY QUALITY

The chemistry of precipitation is characterized by trace quantities of most substances found in the atmosphere. Concentrations are typically measured in parts per million, parts per billion, and even parts per trillion. When considered as an ionic solution, about 95% of the total ionic strength is accounted for by the analysis of calcium, magnesium, ammonium, sodium, potassium, hydrogen, chloride, nitrate, and sulfate. The hydrogen ion is usually determined from measurements of the sample pH.

The acidity of precipitation is commonly presented as pH although more and more evidence is becoming available to show that organic acids also contribute to the total acidity of a sample (15,16). However, it is important to also examine all of the other ions in a sample to understand the effects on the environment. There has been a suggestion put forth to limit the sulfate deposition without regard to the precipitation acidity. It is presumed that a sulfate reduction will automatically result in higher pH values, and, thus, a double environmental benefit will result. But without better quantification of all sources of sulfate, any reduction strategy may not bring about the desired result.

A discussion of the quality of precipitation chemistry must include the analytical methods used as well as a description of the device used to collect samples. Most importantly, the quality of the data must be assured when consideration is given to the impacts on the precipitation receiving system. Students of precipitation chemistry have used everything from glass bottles to baby bottle plastic liners to collect samples for analysis. Laundry baskets, staked to the ground, as well as fence-post mounted plastic bottles have also been used in network operations (17). One of the most interesting collectors was the entire roof of a wood frame building covered with polyethylene sheeting and special gut-tering. A very large sample could be collected in a very short period of time even during the lightest rainfalls.

A number of other devices have been built directed toward acquiring one sample after another during a single rain event to obtain fine detail of the chemical structure of precipitation. There are various means used to control the sample collection by either the volume per sample or time interval between sample collections. Owing to the rapid collection of numbers of samples in a very short period of time, none of these sequential collection devices have been used in a regional network.

The most widely used sampler in the United States networks today is comprised of two buckets, and a rain-activated switch to operate a movable cover. During non-precipitating periods, the cover remains tightly sealed on one bucket. Precipitation falling on the sensitive switch completes an

electrical circuit activating a motor which lifts the cover from one side and places it on the opposite bucket. The sampler thus provides a dry sample as well as a wet sample. This is the standard instrument used throughout the NADP/NTN network.

Anyone wishing to collect rain or snow for chemical analysis is cautioned to first check the collection vessel for the chemical of interest to see if, in fact, the analysis will be contaminated. For example, it would be unwise to collect samples in a weighing-bucket raingage for zinc analysis when the bucket is zinc-coated and leaches into the sample.

A second serious consideration is whether one wishes to collect a bulk sample as opposed to a wet-only sample. A bulk sample is one which is directly exposed to the atmosphere and remains open throughout a prescribed interval of time. This is not a very satisfactory way of collecting precipitation samples because of the natural tendency of birds to perch on the rim of the collector always facing outward contributing to the debris deposited inside the container. Equally important, dust, leaves, and other natural wind-blown materials are likely to enter the sampler and contaminate the precipitation in an unpredictable manner.

The interval between the collection of samples is largely determined by the goals for the sampling program. If one wants to study the effects of precipitation chemistry on the forest, for example, it is highly unlikely that it is necessary to collect samples on intervals of anything less than a one-week period and perhaps even one month may suffice for the majority of biological effects studies. On the other hand, if one wants to study the variability of precipitation chemistry in convective storms during the warm season, a sequential sampler may be necessary to obtain samples as frequently as one or more per minute. So in establishing a sampling program, it is most important to carefully consider the goal of that program and then determine the need for event sampling, as opposed to less costly longer periods, to achieve that goal. The NADP/NTN weekly collection network is an arbitration between event samples and monthly samples, but was chosen to address the program goals of determining 1) the long-term trend of precipitation chemistry and 2) atmospheric deposition effects on the environment.

Once a sample has been confined within the collecting vessel, the safest thing is to immediately seal that vessel and carry it or ship it to the analytical laboratory. However, it is a practice in some operations to allow prior handling of the sample such as withdrawal of aliquots for the local determination of a particular parameter. For example, the NADP/NTN allows extraction of a few milliliters for the field determination of pH and conductivity. Immediately after the aliquot has been withdrawn, the sample is sealed and then shipped to a central laboratory for further chemical analysis. Shipment of the sample is an important consideration for any type of sampling program since one must be sure that the collecting vessel does not leak in transit.

Written documentation of everything concerning the sample up to this point should be provided for the laboratory staff as the analysis of precipitation chemistry proceeds. Certainly, any laboratory, whether it is adjacent to the sampling site or several thousands of kilometers distant, should have certain analytical capabilities for the determination of trace materials in precipitation. The analysts must be trained to recognize the expected concentrations in precipitation and detect contamination in a sample. Contamination can originate from either natural causes or handling of the sample.

Finally, one must be alert that even though a determination may be perfectly accurate and within statistically allowable errors of the instrumentation, the value may, in fact, be excluded from a data set for other reasons. For example, a loose covering over the collection vessel can allow crustal dust to enter into the collector during non-precipitating intervals and can artificially raise the concentrations of those materials. A "leaky" seal results in values that are not representative of precipitation but are more representative of a bulk sample. The major point is that the sample quality control does not begin or end in the laboratory, but must be extended to include everything from the sample collection in the field to the point of preparing the data for dissemination or further interpretation and archiving.

Concern has been expressed about the chemical integrity of samples collected less frequently than the duration of a single storm. There is reason for some scientific inquiry on this matter, but the available data suggest that any chemical changes in a sample will occur in a relatively brief period after the precipitation has ended (18). However, event samples may not be any more stable than weekly samples if the delay between collection of the sample and its analysis is of the order of one or more days. Consequently, until real-time chemical analysis can be performed in the field, all currently available data contain largely unknown errors from chemical changes that occur between the end of an event and the analysis.

SELECTED INTERPRETATIVE ANALYSES

There are at least two obvious ways of viewing the chemistry data from precipitation samples. The first is the concentration of samples and the second is the deposition (or loading) of ions of interest to the surface. From a simple perspective, the concentration is of interest to atmospheric chemists while the deposition is of interest to effects research scientists. It should be kept in mind that the concentration and precipitation are the observables and the deposition is a derived quantity. The deposition is calculated by multiplying the observed concentration by the amount of precipitation associated with the sample thereby obtaining a value of the mass deposited per unit area. In the following discussion, both the concentration and deposition will be shown and described. Only the major ions calcium, magnesium, potassium, ammonium, sodium, chloride, nitrate, sulfate, hydrogen, and sample volume will be shown. There are many ways

that the data can be displayed, but for this discussion median values from the entire data set were selected for each site and maps were hand-drawn to illustrate national patterns for each ion and the precipitation.

Sample Volume

The median sample volume is shown in Figure 1. The unit of measure in Figure 1 is the liter since that is the field observed quantity and is shown instead of converting to inches or millimeters of precipitation. It should be remembered that the values shown are median values from the site history of sampling locations with more than 50 samples (that is approximately one year).

The first comment to make is that the astute student of climatology will notice some differences between this map and the long-term climate values commonly presented. The station data are of variable length, with some stations only having the most recent 50 weeks while others may extend back to July 1978. Nonetheless, these are the data which represent the precipitation associated with the deposition maps to be shown later.

The heaviest precipitation is noted in the Pacific Northwest in an area with a high frequency of cyclonic storms entering the coastline. The second area of relatively high precipitation is in the mid-Gulf Coast region and is possibly a reflection of the natural variability of precipitation in a short-term record. The last obvious area of high precipitation is along the northeastern coastline from northern Virginia to Maine. Similar to the Northwest high, this region of the U.S. is well known for cyclonic systems affecting the precipitation over the region.

The dry region extending south from Canada through the Great Plains to New Mexico is characteristic of that area. In fact, the dry western states are pretty obvious in this figure with less than 0.5 liter samples obtained from the Dakotas south to the Big Bend area of Texas and west to the Sierra-Nevada and Cascade mountains. From the Gulf and East coasts, the precipitation decreases inland to the north and west, respectively.

The pattern of precipitation, as revealed by the median sample volume, is important for further discussion of ionic deposition. The deposition, recall, is the product of the concentrations and the precipitation and, therefore, some relationships will be seen in the following figures.

Calcium

The calcium contribution to the ionic strength of a sample is thought to be mainly due to the incorporation of soil aerosol into the precipitation before it is collected. Owing to agricultural practices and the semi-aridness of the Great Plains, it is not surprising to find the highest concentrations in that region (Figure 2). There is seen a large area of relatively high concentration extending from Montana-North Dakota south through central and western Texas. A secondary area is seen extending

northwest into southern Idaho from the Four Corners area where Utah, Colorado, New Mexico, and Arizona join. The coast lines of the U.S. are areas of relatively low concentration with values 3 to 6 times less than in the interior continent.

The deposition, expressed in kilograms per hectare per year, also shown in Figure 2 maximizes slightly to the southeast from the Center of maximum concentration and is found over southeast South Dakota, southwest Minnesota, northwest Iowa, and northeast Nebraska area. This slight shift is due to the somewhat greater precipitation toward the southeast while maintaining high concentrations. The isolated maximum in the Four Corners area is a direct result of an isolated maximum in precipitation and the maximum concentration. The same is true of the maximum in southeastern Louisiana. Note the excursions of high deposition into northern Illinois, western Kentucky, and even a small maximum in northern West Virginia. During periods of drought and attendant dust storms, such excursions can be more severe and cover much larger areas leading to misinterpretation of the meaning of the chemistry regarding trends (19).

Magnesium

The magnesium concentration pattern bears some similarities to the calcium distribution, but also shows some distinct differences (Figure 3). The greatest similarity is the area of maximum concentration in the northern Great Plains extending into the Midwest. There is also a lesser maximum over Arizona northward to southern Utah. These maxima are influenced by the soil composition in those areas, but beyond these two likenesses to the calcium pattern the remainder of the magnesium concentration distribution appears to be related to an oceanic source. Obviously, between these two regions of high concentrations, relatively low values along the mountains of the eastern states and along the lower Ohio and Mississippi River valleys are observed.

The coastal deposition pattern reflects the precipitation pattern faithfully with general decreased deposition toward the continental interior (the bottom of Figure 3). The ratios of calcium of magnesium in proximity to the coastal areas simulates that of seawater with values less than one and frequently less than 0.5. However, the central Plains ratios and most of the interior sites have ratios exceeding one and at many locations greater than 10.

Potassium

The potassium concentration, again, suggests a soil source, particularly in the northern Great Plains (Figure 4). The maximum over northwest Iowa and central Nebraska is in the same location as the calcium maximum and in a relative high magnesium concentration area. As with magnesium, however, a seawater component can be observed along the coast lines of the country. Also similar to the magnesium is the relatively low potassium concentration region along the mountains of the east and westward across Tennessee, Arkansas, and Texas. There is a ridge of high concentrations observed

across eastern Colorado into northern New Mexico which corresponds to similar high concentrations of calcium and magnesium seen in Figures 2 and 3.

The deposition of potassium shown at the bottom in Figure 4 is almost an exact duplicate of the magnesium. A high deposition area is seen in Figure 4 to extend northward from the Mississippi River delta into western Iowa. The Rocky mountains exhibit little deposition primarily due to the low precipitation in the region. The coastal areas reflect the seawater potassium component as well as the high precipitation in those areas shown in Figure 1.

Ammonium

The previous three ions likely appear in precipitation as the result of natural causes. All three are in relative abundance in the natural sources of the oceans and the earth's crust. The source strength has not been quantified although some have attempted adjustments for seawater contributions to observed concentrations (20).

The ammonium ion source is also largely undetermined. The concentration pattern shown in Figure 5 suggests a relationship to the large feed lots associated with the cattle industry of the central Plains region. However, this possible source has not been measured, but only surmised from the geographical relationship between the maximum concentration and the known feed lot distribution. In one sense, the distribution of ammonium in precipitation has a natural source, but somewhat controlled by man.

Unlike some of the three previously described ions, the ammonium maximum is over the center of the interior U.S. decreasing outward in all directions with a few small, isolated high values scattered in other areas. For the most part, the coastline precipitation contains the lowest observed ammonium concentrations.

The deposition of ammonium is dominated by the concentration pattern as modulated slightly by the precipitation (Figure 5). The maximum deposition of greater than 4 kilograms per hectare per year is centered over the identical area of the maximum concentration. Since the deposition is influenced greatly by the regional weather (winds, storm systems and movement), it is interesting to observe the close relationship between the presumed source, the concentration, and the deposition. This distributional relationship certainly suggests a region for the testing of long-range transport and transformation models or it suggests something about the atmospheric chemistry of ammonium.

Sodium and Chloride

The distribution of these two ions is acknowledged to be controlled by the proximity of a sampling site to the oceans. This is borne out by the concentration patterns shown in Figures 6 and 7. The ratio of sodium to chloride at many of the coastal sites is very close to that for seawater of 0.86.

There are two features of the concentration patterns that are interesting and cause for some speculation. Both ions show a relative maximum extending from the Gulf coast northward across Texas into the upper Great Plains states. The seawater ratio, however, does not hold beyond northern Texas and the ratio becomes one or greater further north. This observation suggests either an inland source of sodium (soil?) or a selective decrease of chloride during the precipitation process. There is also a low concentration area observed for both ions over the Smokey Mountains, but the seawater ratio seems to be sustained. One could interpret this observation as due to the simple reduction of the seasalt component in the atmosphere with increased altitude and distance from the coast. There is also a tongue of high concentration extending from the central Gulf coast northeastward to Ohio with a strong possibility of a seawater influence as evidenced by the nearness to that ratio. This may reflect the meteorologists notion that the source of atmospheric water vapor for precipitation in the Midwest originates in the Gulf of Mexico, particularly during the warm season.

The deposition of sodium and chloride, obviously, is a mirror image of the precipitation pattern showing decreasing values with distance from the U.S. coastline (lower half of Figures 6 and 7). These two ions are worthy of additional study because of their domination by natural sources, and their obvious relationship to coastal influences.

Nitrate

The nitrate concentration in Figure 8 shows the highest values over central New York and Pennsylvania and an equal maximum over southwestern Michigan. The extension of these isolated maxima to the west as far as South Dakota and Nebraska appears to be associated with the ammonium concentration maximum in the same area indicating the possible presence of ammonium nitrate.

The major source for atmospheric nitrate is attributed to vehicular traffic and certainly the small center of high concentration over southwest California seems to confirm that relationship. It is not so easy to relate to other areas of high concentration to such high density mobile sources. This pattern is not easily traceable to the currently identified sources and considerable research is needed to explain the distribution of nitrate in precipitation.

The deposition pattern in Figure 8 is almost identical to the concentrations. The one exception to the simple correlation between concentration and deposition is in southeast Louisiana. The heavy precipitation noted in Figure 2 in that area explains the relative maximum in deposition extending from the Delta region northward into central Arkansas.

Sulfate

The sulfate concentration is shown in Figure 9. The most obvious feature in this figure is the large area of high concentration in the eastern U.S. The values decrease outward from maxima over southwestern Pennsylvania and

south-central New York. It is interesting to note that the east-west axis of the maximum lies to the north of the Ohio River frequently presumed to be the major source region for sulfur dioxide in the east.

The west coast population centers of San Francisco and Los Angeles appear with minor maxima associated with them. The other two small maxima over the central Washington and Oregon border and over the southern Arizona and New Mexico border are difficult to explain. Equally noticeable is the lack of high concentrations in the Four Corners region.

The deposition shown in Figure 9 is a good visual representation of the product of the concentration and precipitation. The axis of the major high deposition area is shifted to the south and oriented northeast-southwest. This slightly shifted pattern from the concentration maximum is due to the gradual increase of precipitation from Illinois southeastward. The pattern, then, gives the appearance of little long-range transport from the primary source region, that is, the heaviest deposition occurs directly over the highest sulfur dioxide emission area.

pH and Hydrogen Ion

The median pH distribution shown in Figure 10 reveals values less than 5.0 over almost the entire eastern half of the U.S. It has been argued that this pattern has not changed significantly since prior to the early 1950's (19). The major features to be noted are the low pH values in the east, the high values in the Great Plains from the Canadian border south to the Texas border, and the more variable pattern in the mountainous west.

The hydrogen ion deposition, also shown in Figure 10, exceeds 20 grams per hectare per year over most of Michigan, Illinois, eastern Missouri and Arkansas, Mississippi, Alabama, and the northern half of Georgia. The greatest deposition of over 60 grams per hectare per year is observed over western Pennsylvania.

SUMMARY AND DISCUSSION

The author has recounted briefly the history of scientific interest in precipitation chemistry and focused attention on some of the important problems dealing with the quality assurance of analyzed samples. These considerations lead the author to conclude that comparisons between data collected prior to the late 1970's and those more recently acquired should not be used to establish trends. The variety of sampling methods used, frequently unknown analytical procedures used, and the absence of measured key chemistry variables do not permit objective interpretation of the older data. These inadequacies of the available data prior to the establishment of the NADP/NTN network gave rise to controversy concerning the reality of a presumed trend toward greater precipitation acidity in the Northeast U.S. and areal spreading to the Midwest and Southeast (9,19). The trend issue is of importance as national policy is gradually emerging from the work of the National Acid Precipitation Assessment Program

(NAPAP). The pH measurements from the oldest, continuous operating site under the NAPAP is shown in Figure 11. This site is located at Parsons, West Virginia in the northeast part of that state. Three observations are apparent in the figure. First, there is great variability in the week-to-week pH with the range of values extending from about 3.5 to 5.7. The median value for the station is 4.27. Second, there is a distinct seasonal signal in the data. Lower pH measurements are obtained in the summer months, in general, and higher values in the winter. This point will be emphasized further in the discussion. Third, if there is a trend in these data it is well disguised in the variability although a slight overall improvement in pH may be seen.

An additional example of the seasonal variability of pH and sulfate is shown in Figure 12. The data shown are from a site in rural, east-central Illinois. The individual weekly samples were used to generate 12 week moving averages which emphasizes the seasonality of the sulfate in precipitation. A linear fit to the concentration data shows a marked decline of sulfate over the period of record (not shown on the figure). The peak in concentration during the summer months is readily seen in this figure although there are year-to-year differences in the maximum value. The middle part of Figure 12 depicts the precipitation moving average. The seasonality of the Illinois precipitation is also quite noticeable with summertime peaks. The next curve, moving up on the figure, is the sulfate deposition. The seasonal peaks are emphasized for the deposition because the concentration and the precipitation are in phase. Note that the deposition, however, is dominated by the precipitation. For example, the greatest concentration was observed in the 1980 summer, but the greatest deposition was observed in the 1981 summer due to the higher summer rainfall in that year. Finally, the pH moving average short-term trend is shown at the top of Figure 12. While there appears to be a direct correlation between the sulfate concentration and the pH, it is by no means perfect. One of the most obvious discrepancies appears in 1984 where the sulfate peak concentration occurs with a relative maximum pH although there is a decline noticeable shortly thereafter.

One final point can be made from the data in Figure 12. If we assume that a reduction by 50 percent of the peak concentration (90 microequivalents per liter) in the 1983 summer, the resulting concentration would be approximately that observed in the previous winter. Yet, the pH change over those two seasons was observed to be from about 4.3 to 4.5 or 0.2 pH units. Clearly, the acid rain issue is a complex one with no easy solutions.

It was stated at the outset that no dry deposition monitoring method has been decided and approved, but the dry bucket data from the precipitation network are available and, perhaps, are a source of some information. The total deposition was calculated at five sites in the NADP/NTN network and the percentage that was observed wet and dry was determined. The sites are located in east-central Illinois, northeast Ohio, southeast New York, central North Carolina, and extreme southwest North Carolina. All sites are rural in character with the southwest North Carolina site standing in

a forest clearcut area. The data are shown in Figure 13. Recall the point made earlier that the first three ions shown in the figure are related to crustal dust. At all but one of the sites, these ions are deposited primarily dry while those associated with the oceans and anthropogenic sources are deposited wet. The one exception is the forest clearcut site in North Carolina where wet deposition accounts for almost all of the deposition for all ions but potassium.

In conclusion, as the data base for precipitation chemistry grows so does our knowledge of its variability and trend. It seems rather clear, that there is not a rapidly declining quality of precipitation and any changes in recent years are going to be difficult to quantify in the presence of the observed great variability. It is equally clear, that to try to estimate a trend using data prior to the implementation of the NADP/NTN network opens the door to controversy since those data were not collected for trend analysis and did not include some of the important ions necessary to address the precipitation acidity issue. The total deposition must be measured if the insult to the biosphere is to be fully assessed. In the absence of a dry deposition monitoring network, the bucket estimates suggest that for some ions dry deposition is most important while for others wet deposition dominates. In any event, there does not appear to be a 50-50 split between the two forms. Damage mitigation strategies must consider this imbalance between wet and dry deposition and its regionality. It must be obvious from the foregoing data and discussion, that additional work needs to be accomplished before answers to many key questions will be forthcoming.

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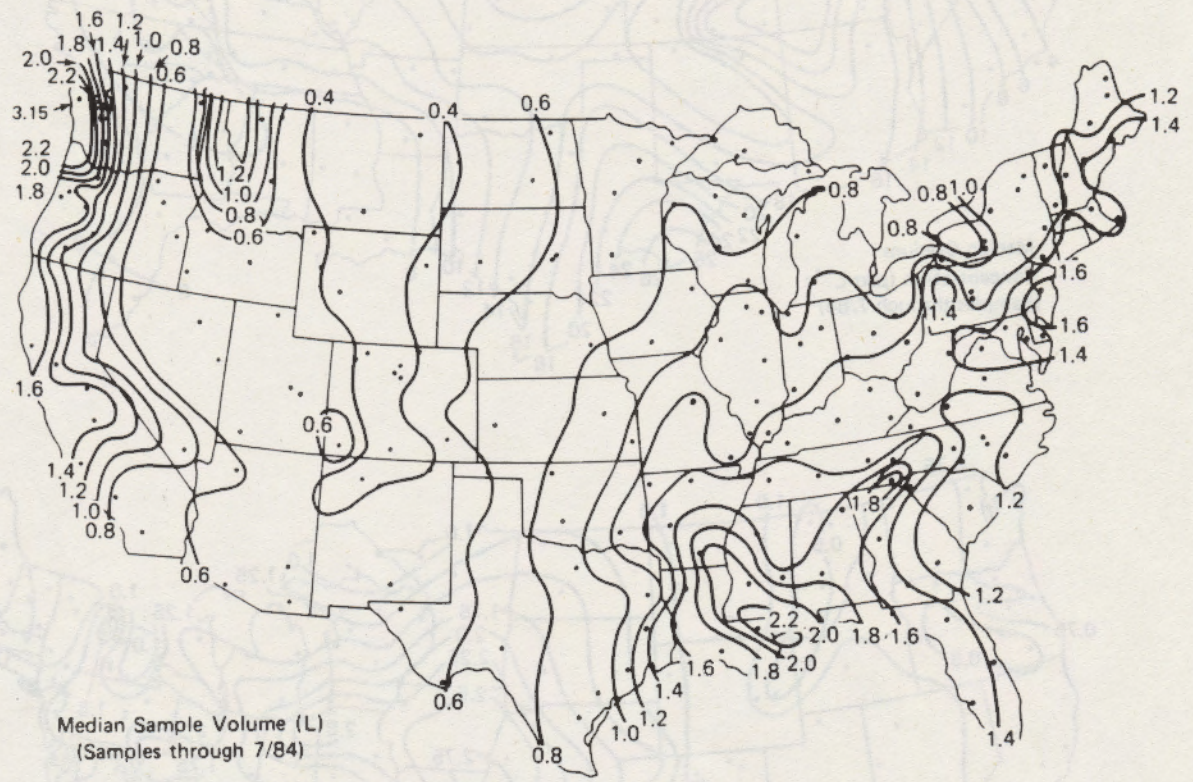


Figure 1. The median sample volume in liters for samples collected at each NADP/NTN site through July, 1984.

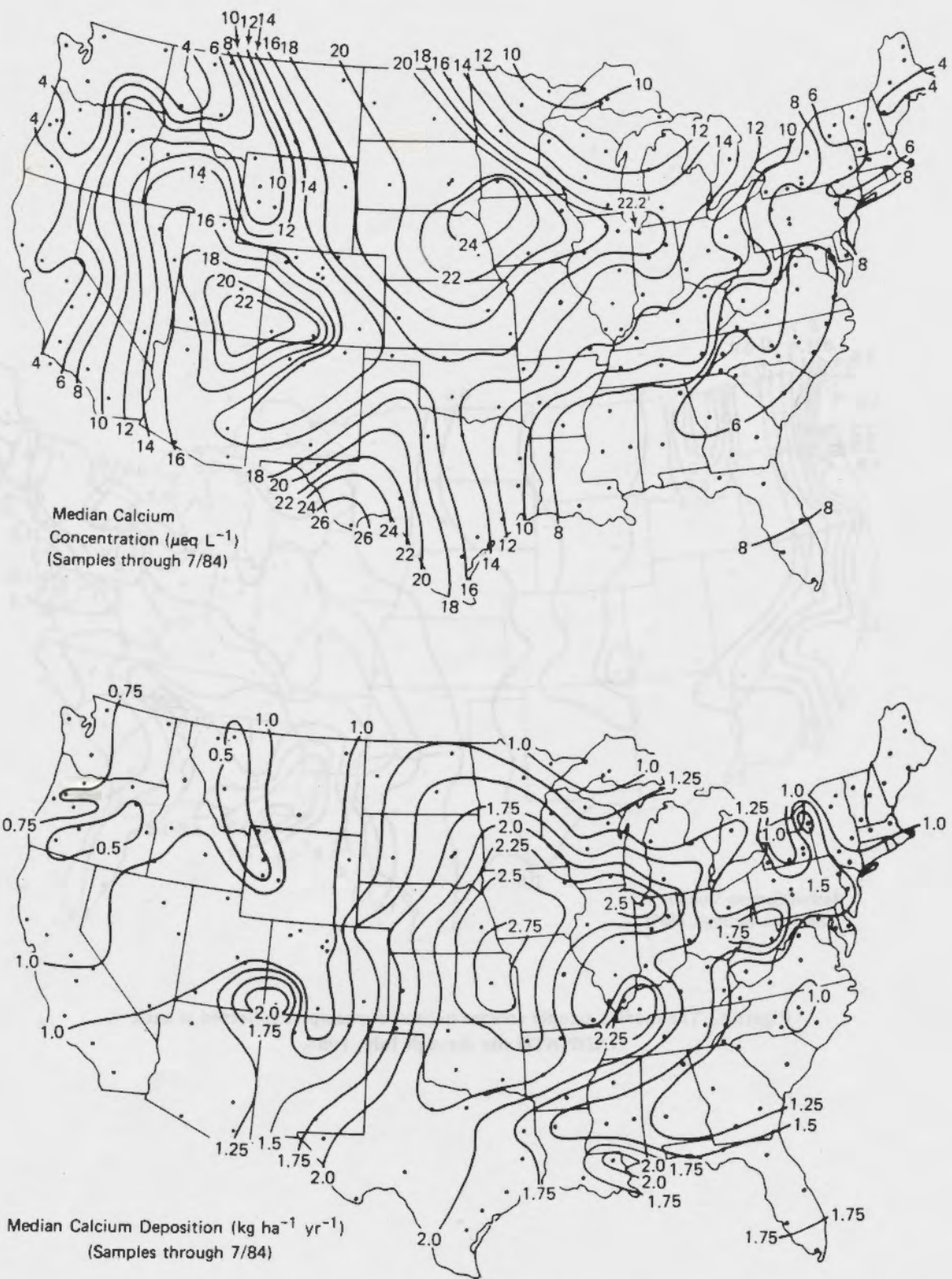


Figure 2. The median calcium concentration in microequivalents per liter from the NADP/NTN network for samples through July, 1984 (top) and the deposition of calcium in kilograms per hectare per year (bottom). Unless indicated, all units are the same for the following figures.

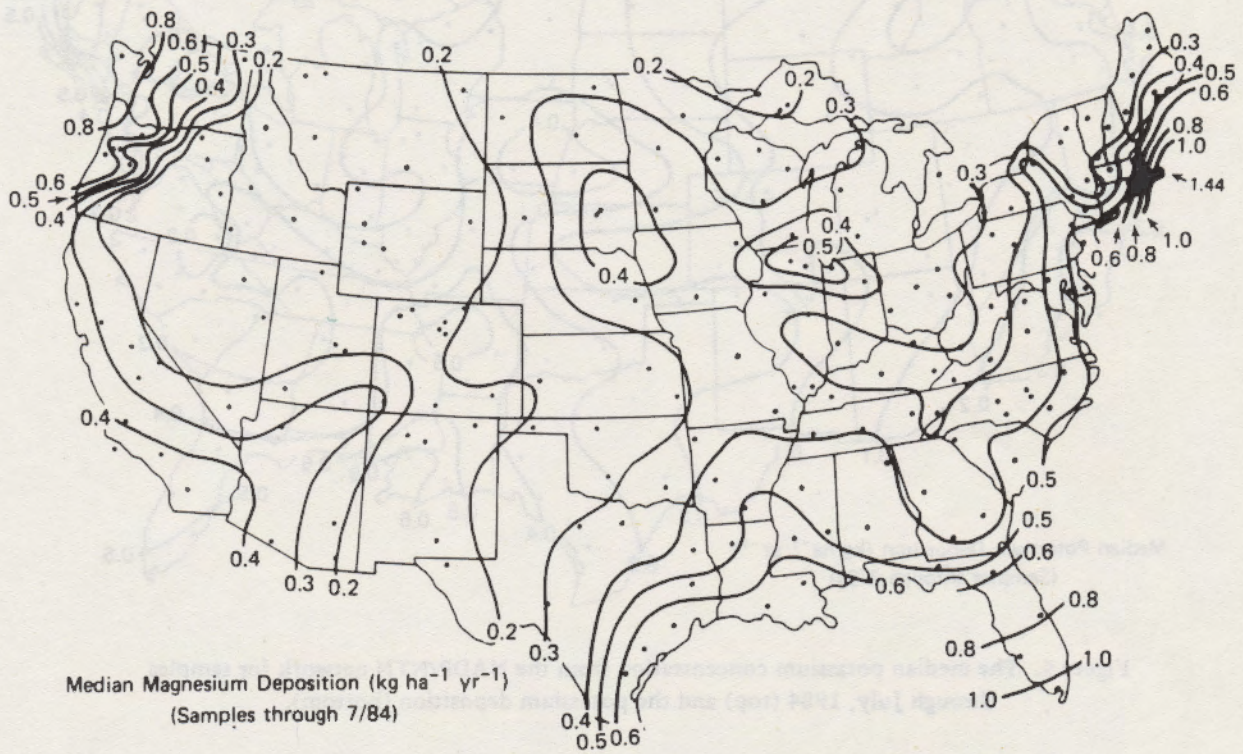
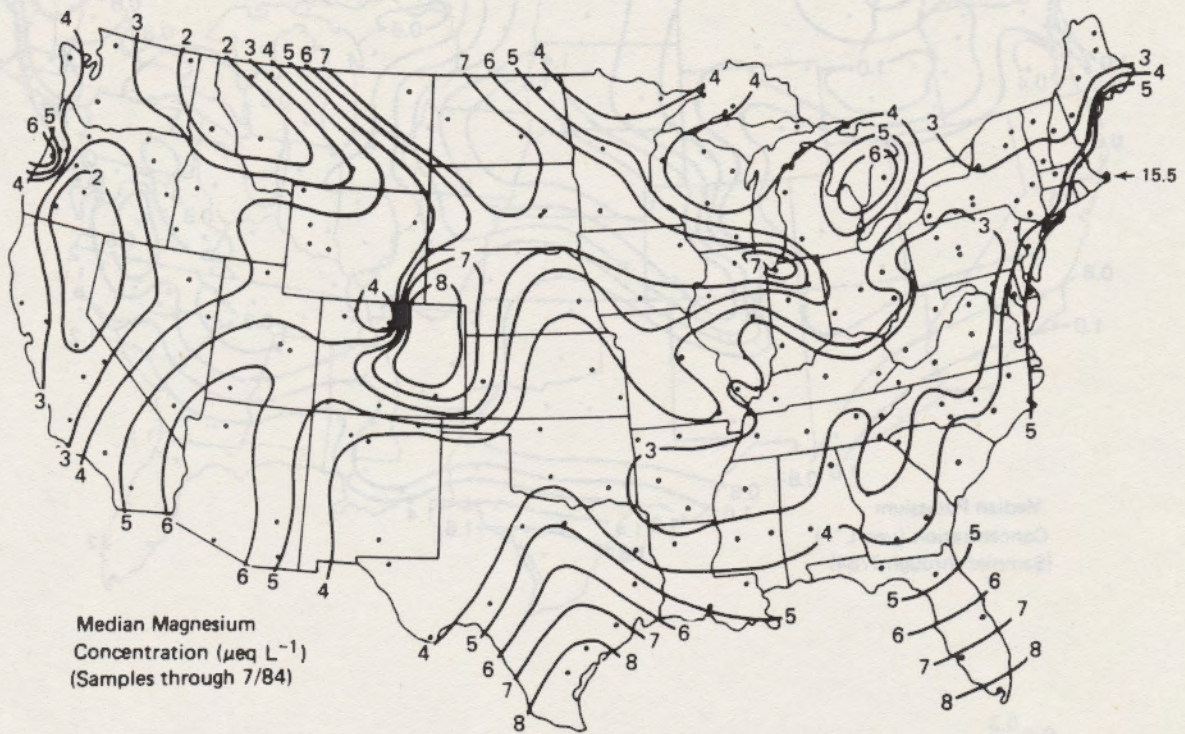


Figure 3. The median magnesium concentration from the NADP/NTN network for samples through July, 1984 (top) and the magnesium deposition (bottom).

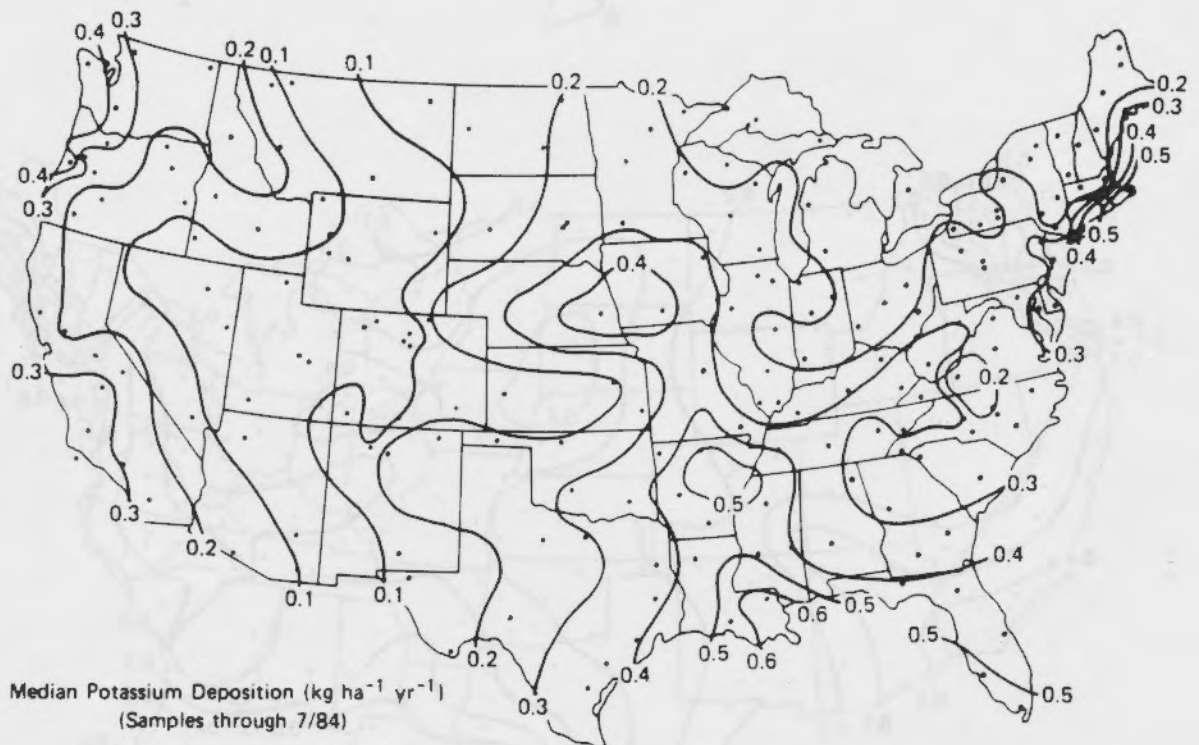
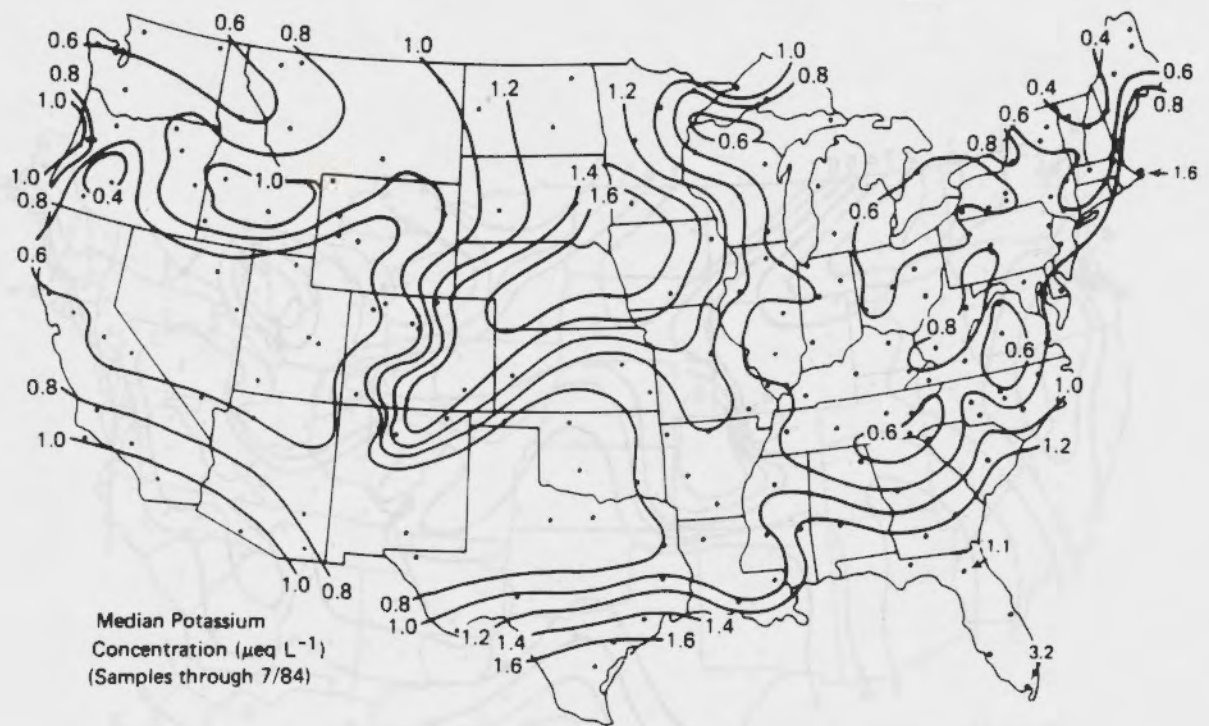


Figure 4. The median potassium concentration from the NADP/NTN network for samples through July, 1984 (top) and the potassium deposition (bottom).

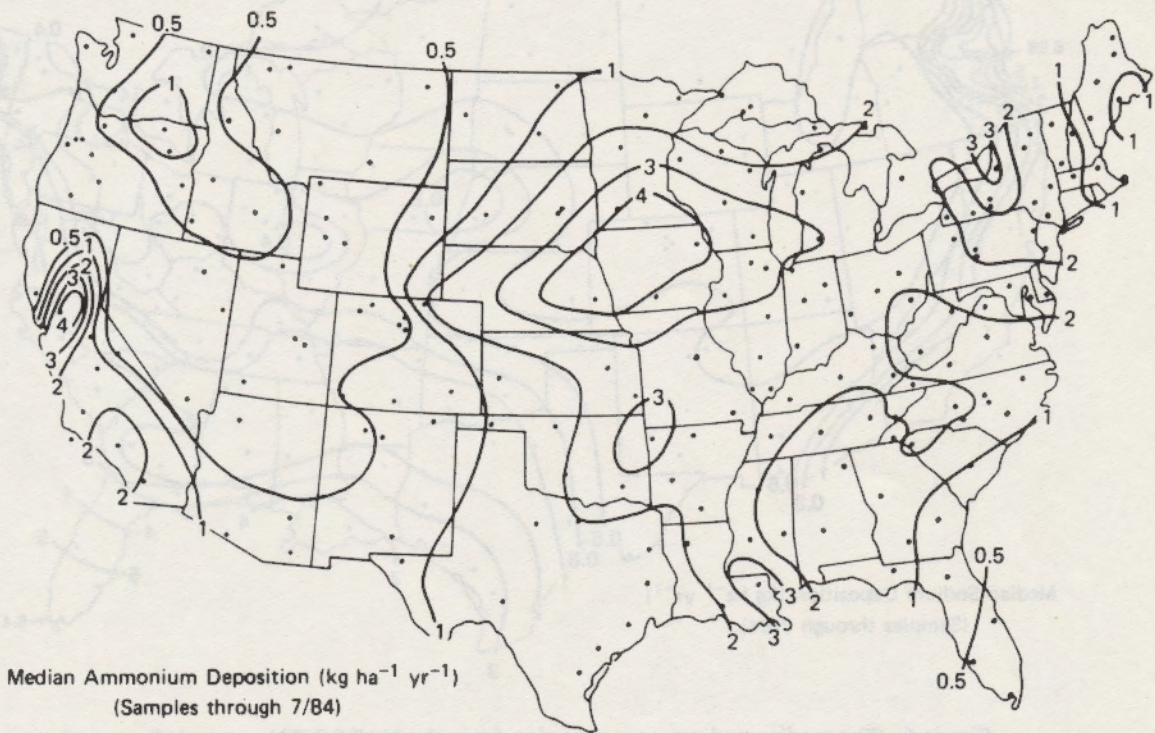
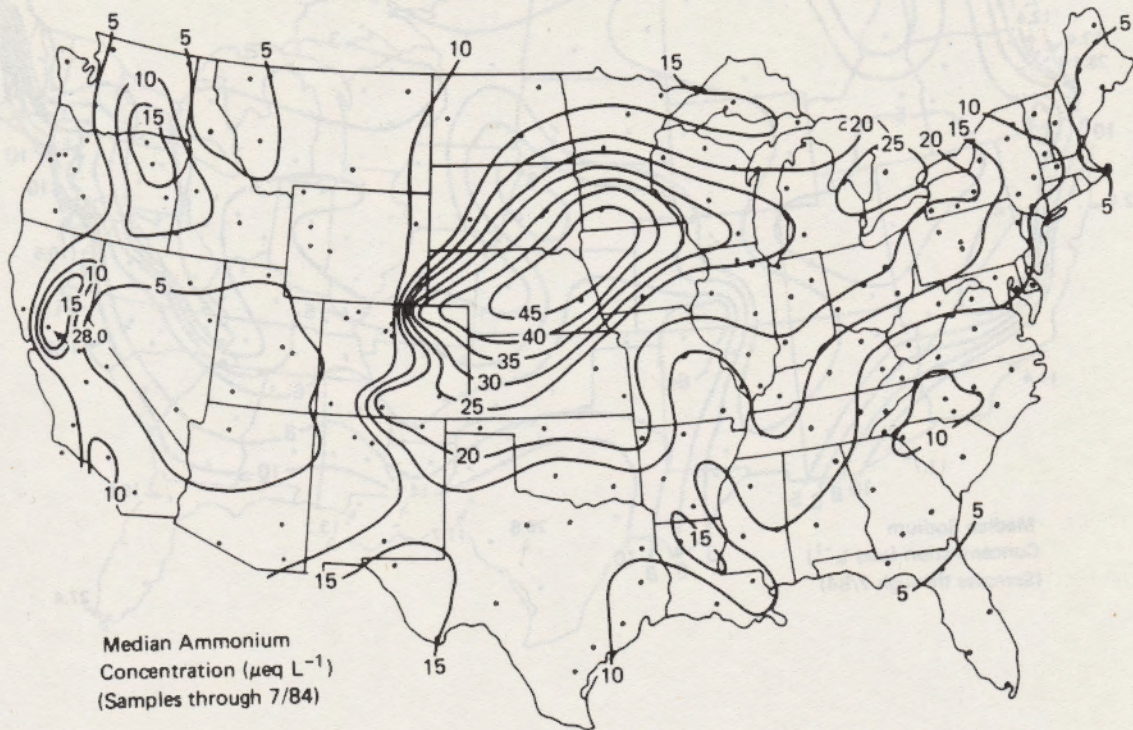


Figure 5. The median ammonium concentration from the NADP/NTN network for samples through July, 1984 (top) and the ammonium deposition (bottom).

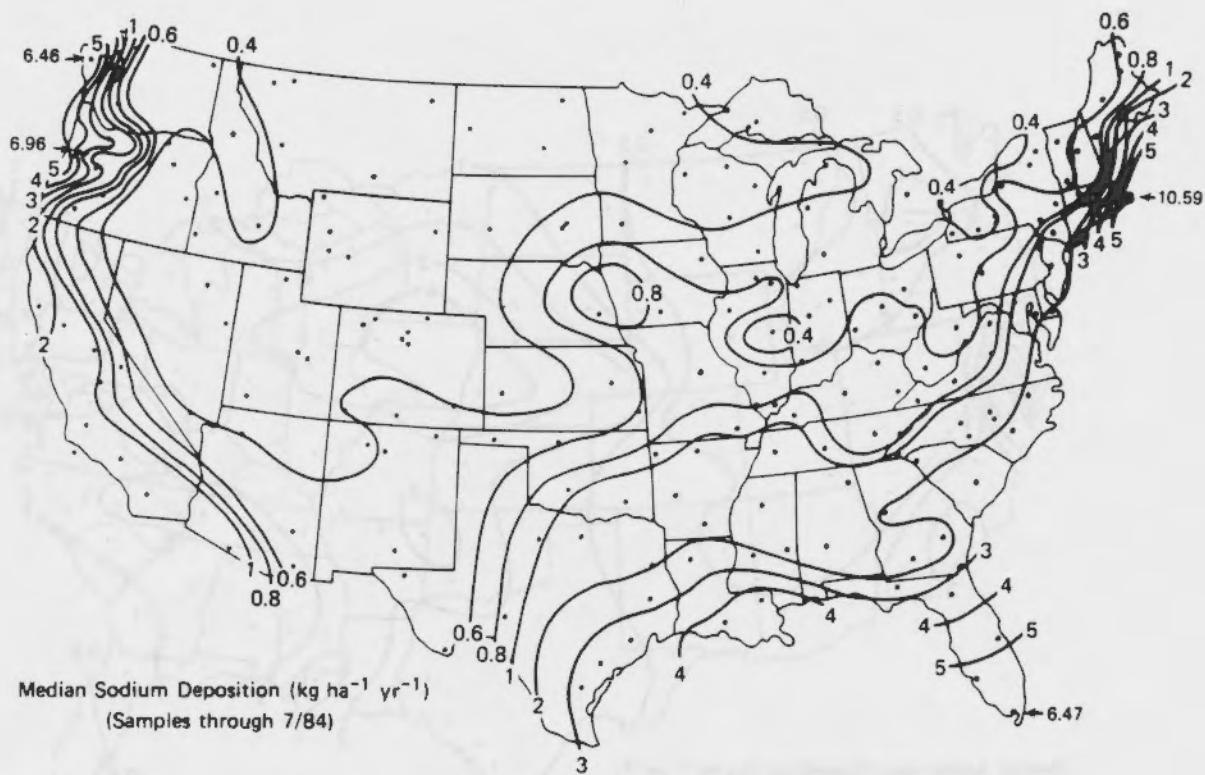
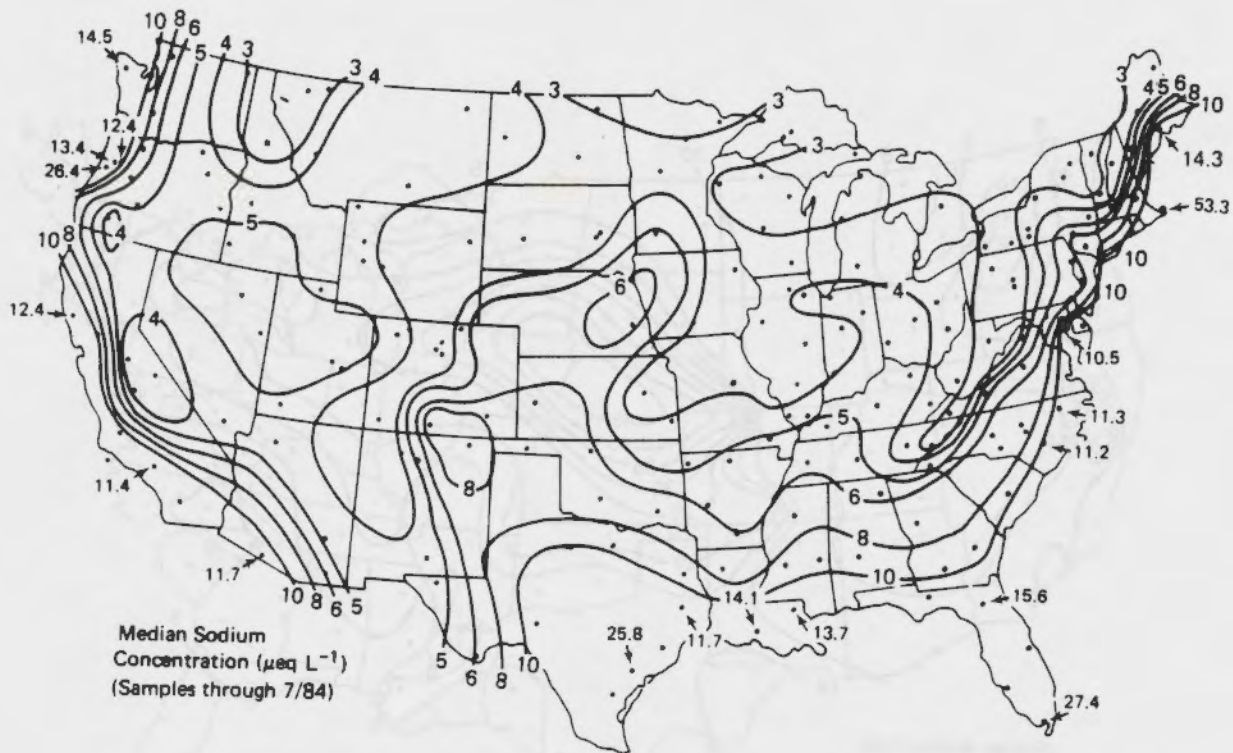


Figure 6. The median sodium concentration from the NADP/NTN network for samples through July, 1984 (top) and the sodium deposition (bottom).

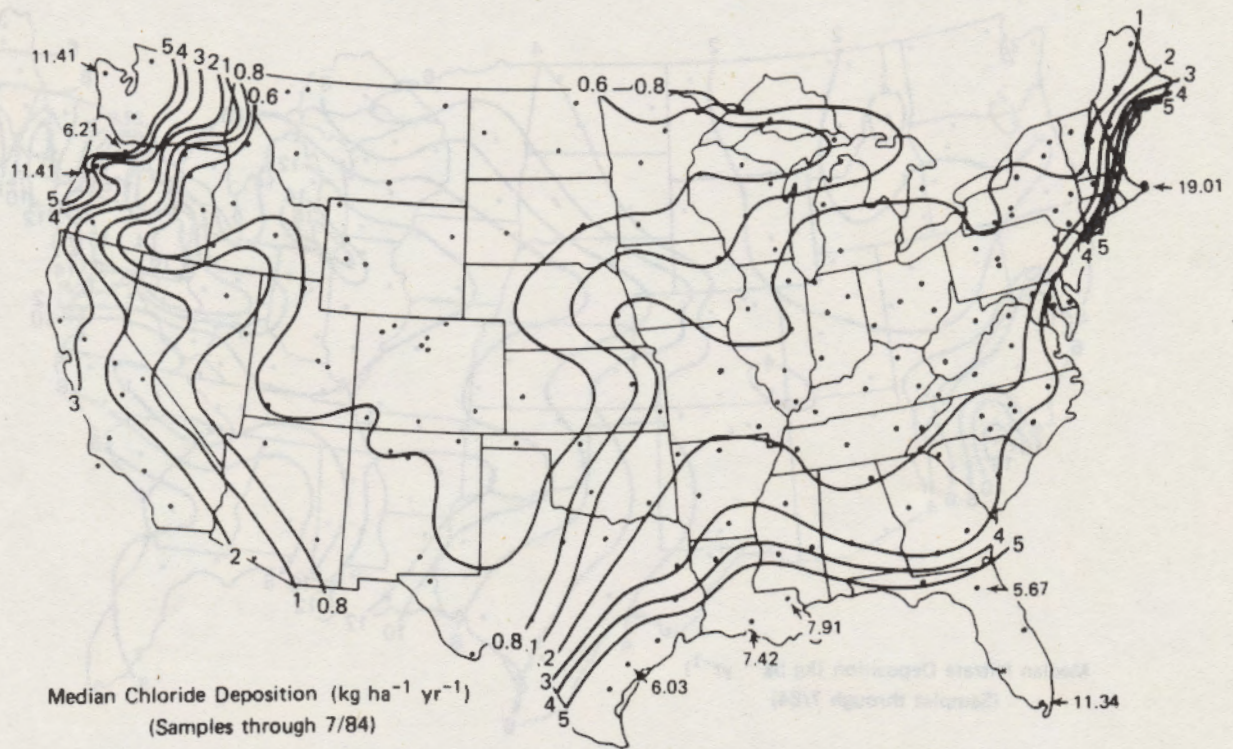
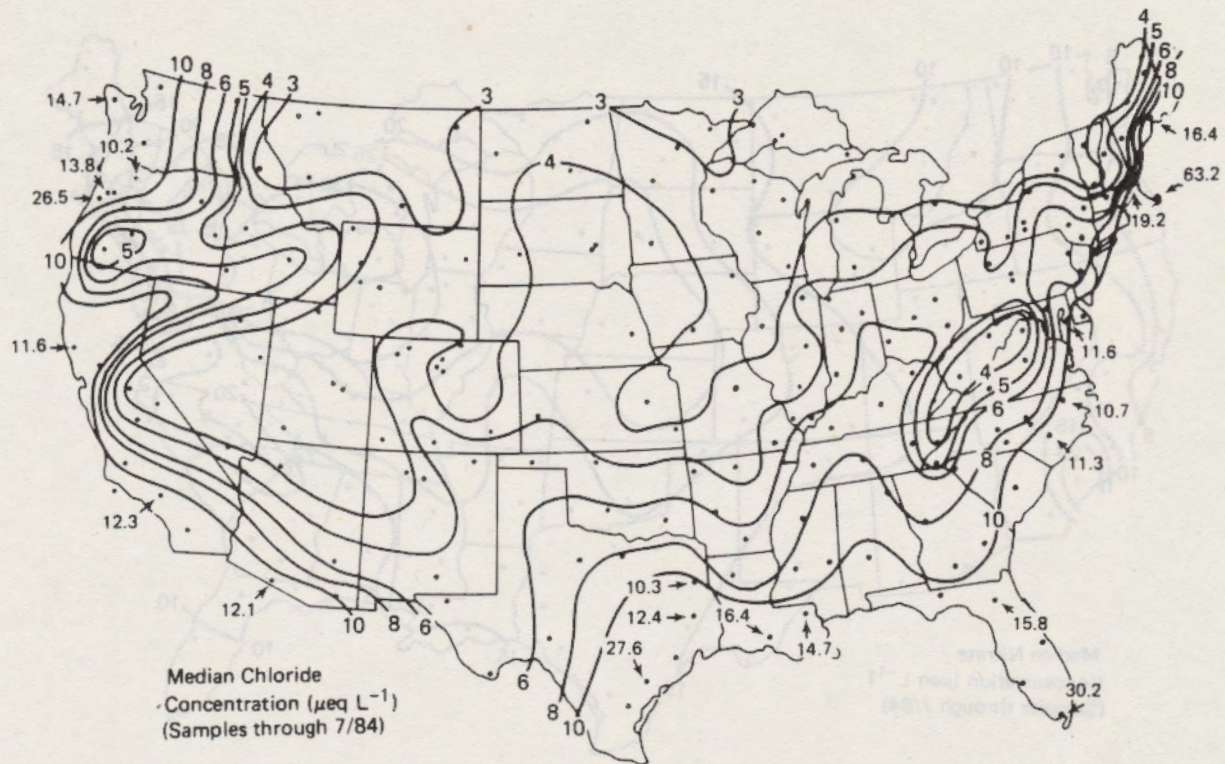


Figure 7. The median chloride concentration from the NADP/NTN network for samples through July, 1984 (top) and the chloride deposition (bottom).

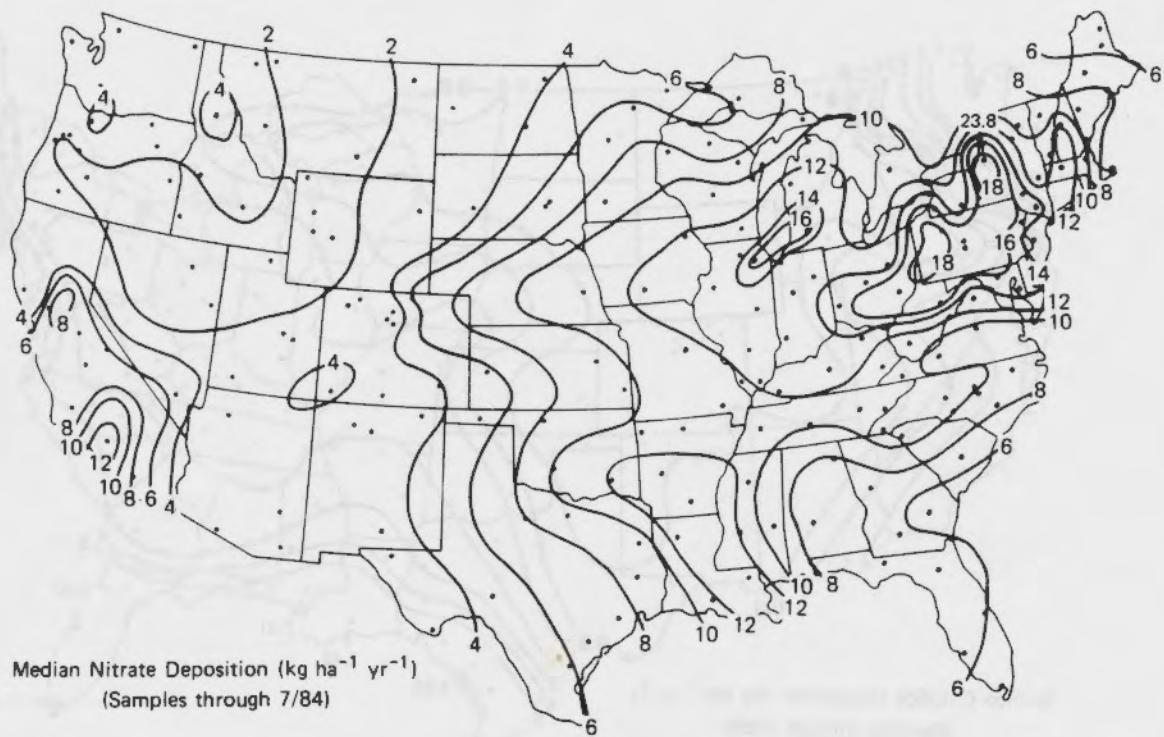
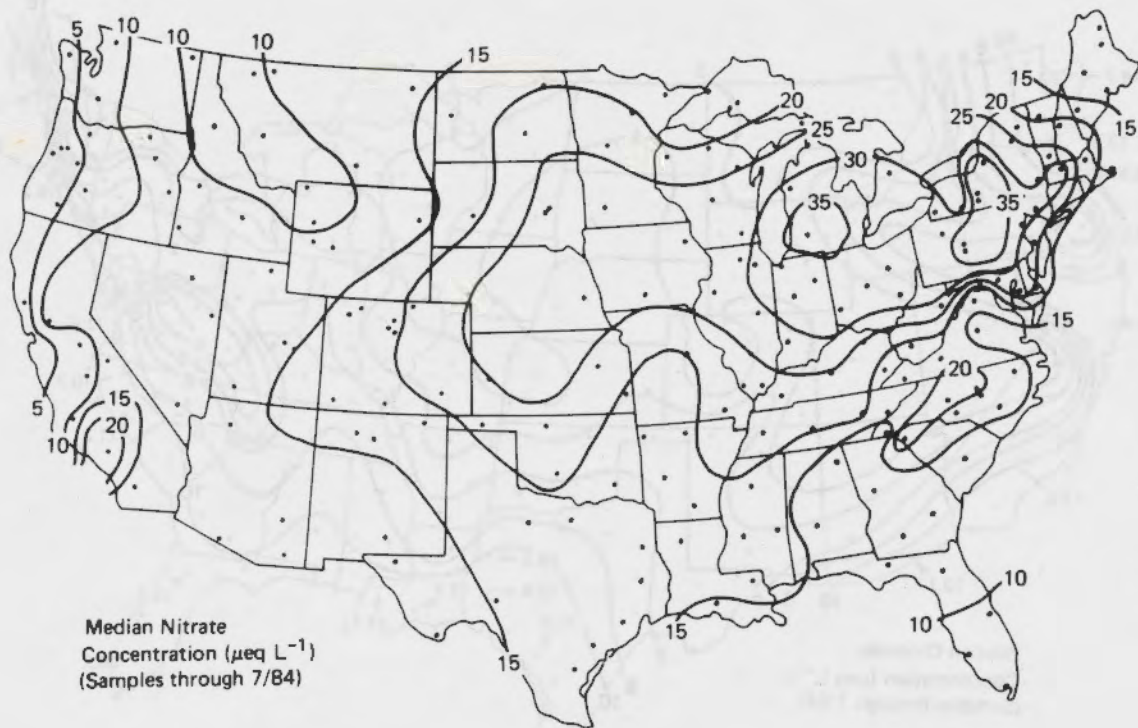


Figure 8. The median nitrate concentration from the NADP/NTN network for samples through July, 1984 (top) and the nitrate deposition (bottom).

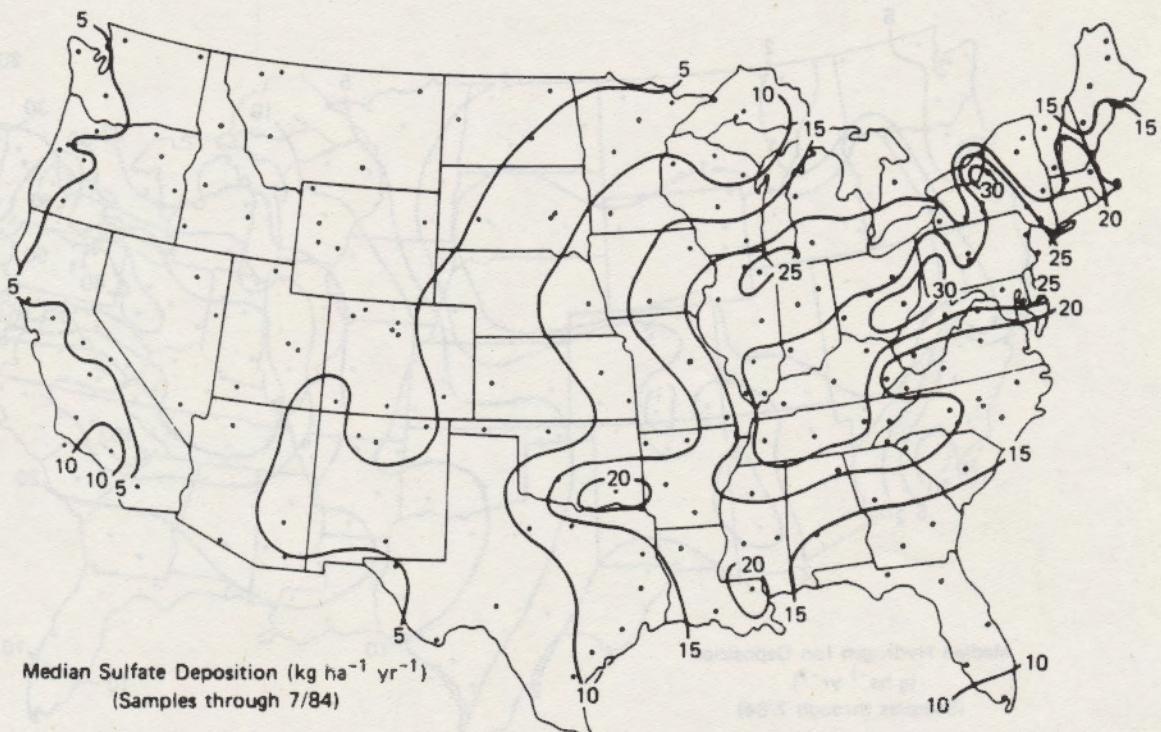
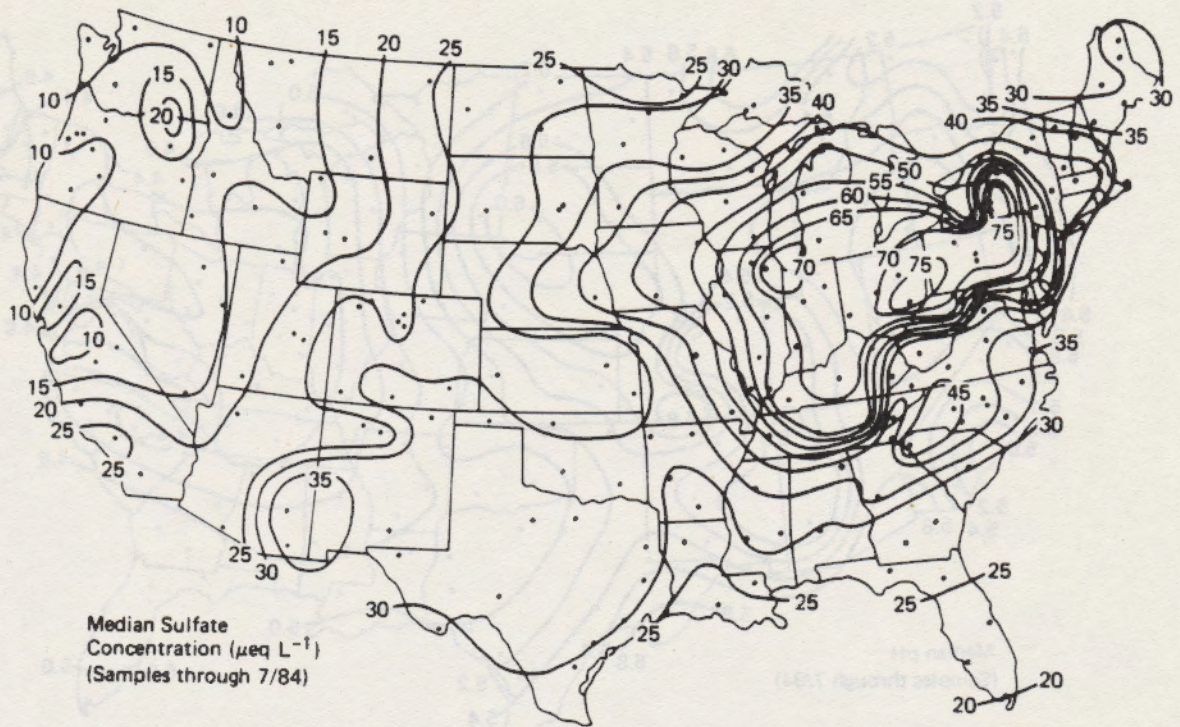


Figure 9. The median sulfate concentration from the NADP/NTN network for samples through July, 1984 (top) and the sulfate deposition (bottom).

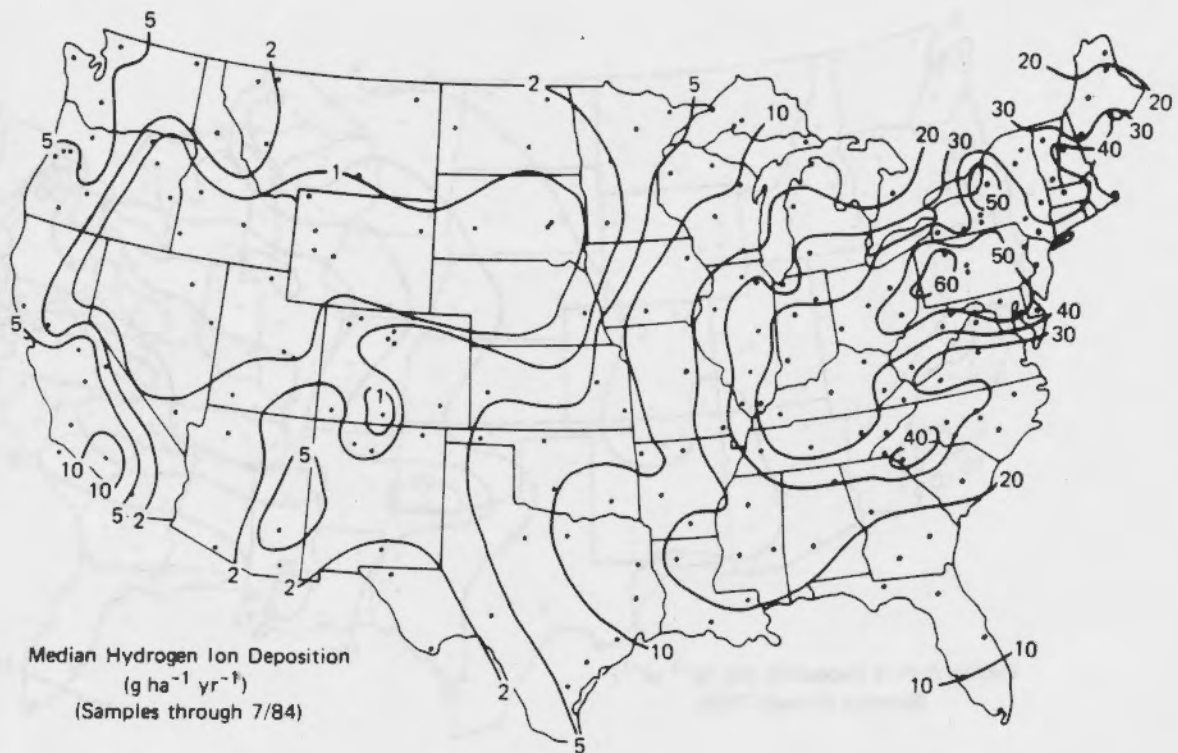
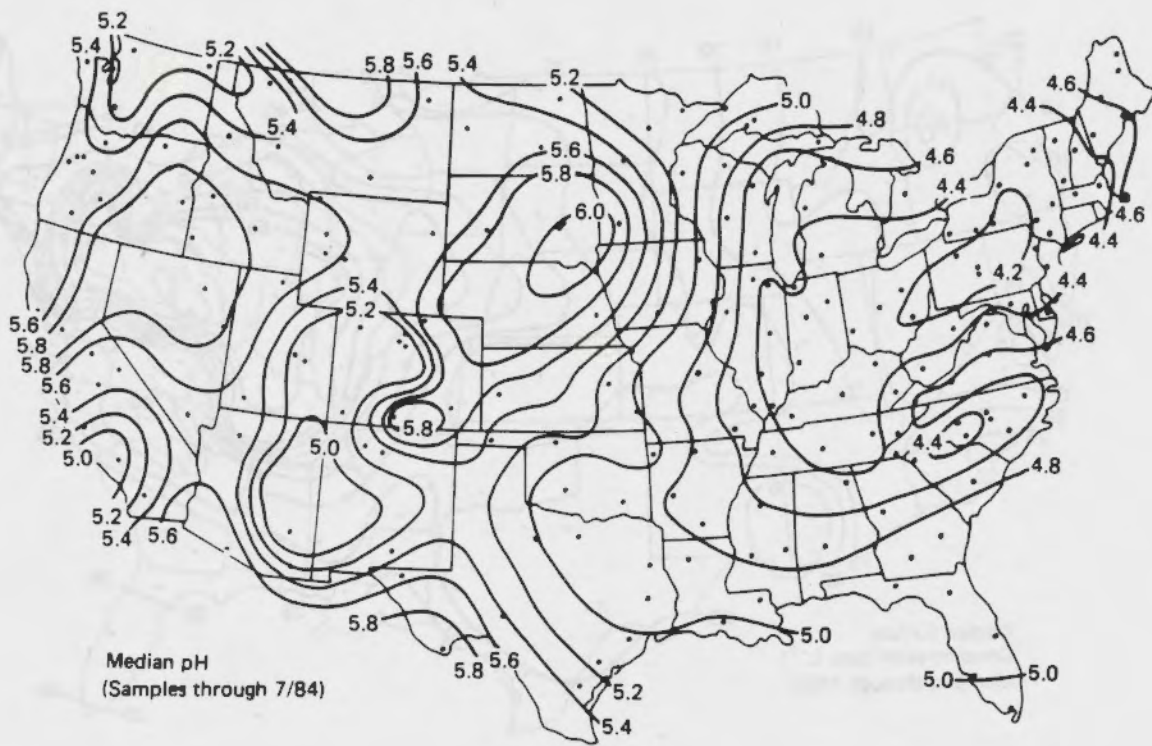


Figure 10. The median pH from the NADP/NTN network for samples through July, 1984 (top) and the hydrogen ion deposition in grams per hectare per year (bottom)

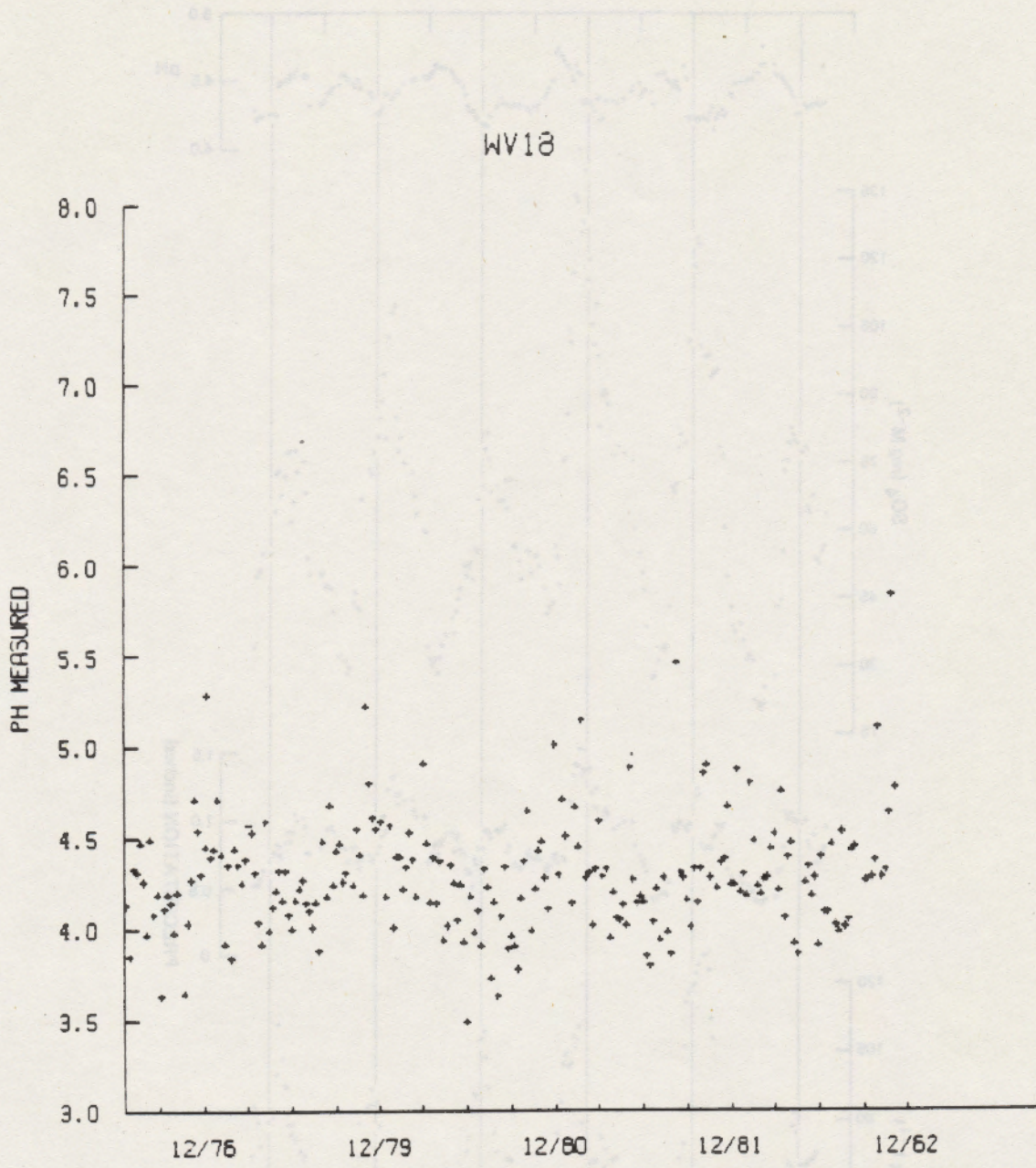


Figure 11. Individual sample pH values for the NADP/NTN site at Parsons, West Virginia. This site is in the center of the heaviest hydrogen ion deposition area in the United States. Note that there is no discernible trend for the period of record shown between July, 1978 and December, 1982. However, a distinct seasonal variation can be seen as well as isolated events of both high and low pH. The median value is 4.27.

Figure 12. The pH (top), sulfate deposition (middle) and precipitation (bottom) from the NADP/NTN site. The yearly values were used to construct the 11 week moving averages shown. Note the strong seasonal signal present in the data with generally higher values of sulfate concentration and deposition and precipitation. Only the pH shows an inverse relation to the season with higher values in winter and lower in summer.

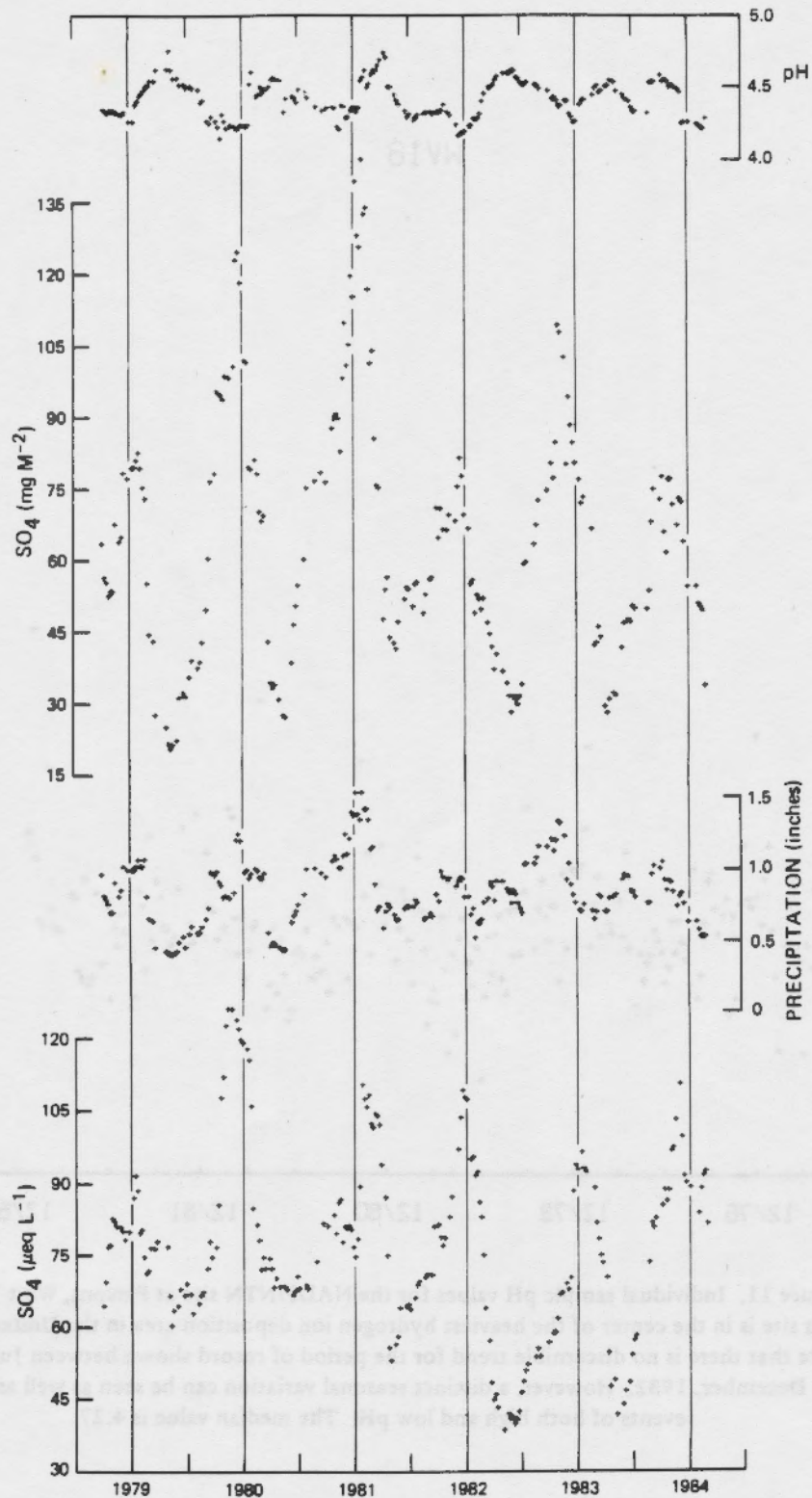


Figure 12. The pH (top), sulfate deposition (first down from the top), precipitation (first up from the bottom), and the sulfate concentration (bottom) from the Bondville, Illinois NADP/NTN site. The weekly values were used to construct the 12 week moving averages shown. Note the strong seasonal signal present in the data with generally higher values of sulfate concentration and deposition, and precipitation. Only the pH shows an inverse relation to the season with higher values in winter and lower in summer.

TOTAL DEPOSITION AT SELECTED NADP SITES

(mg M⁻²)

		Ca	Mg	K	Na	NH ₄	NO ₃	Cl	SO ₄	Total
IL 11 (17)	T	11.2	1.7	1.3	1.0	6.6	35.2	2.5	62.4	121.8
	%D	69.2	70.5	74.2	25.4	17.7	26.0	24.0	27.9	31.6
	%W	30.8	29.5	25.8	74.6	82.3	74.0	76.0	72.1	68.4
OH 71 (14)	T	13.1	1.9	1.4	1.6	5.2	32.0	3.5	71.0	129.7
	%D	78.5	67.5	77.7	31.5	10.9	20.7	31.5	30.9	33.4
	%W	21.5	32.5	22.3	68.5	89.1	79.3	68.5	69.1	66.6
NY 51 (13)	T	4.8	1.9	0.6	3.7	3.0	31.4	4.4	69.5	109.4
	%D	55.5	58.9	38.8	36.5	11.0	10.3	16.7	17.2	18.3
	%W	44.5	41.1	61.2	63.5	89.0	89.7	83.3	82.8	81.7
NC 41 (12)	T	3.8	1.2	1.4	3.8	6.7	22.8	5.5	44.8	89.9
	%D	58.6	53.9	73.1	40.4	53.0	23.0	32.1	30.8	33.1
	%W	41.4	46.1	26.9	59.6	47.0	77.0	67.9	69.2	66.9
NC 25 (10)	T	2.5	0.7	0.9	2.6	3.2	22.1	3.6	45.6	81.1
	%D	18.7	16.0	52.5	12.7	9.3	1.7	3.2	7.5	6.9
	%W	81.3	84.0	47.5	87.3	90.7	98.3	96.8	92.5	93.1

Figure 13. The total wet and dry deposition at selected NADP/NTN network sites. The analyses of the dry bucket of the two bucket collector was used as the indicator of dry deposition. The natural crustal dust components are deposited mostly dry while the oceanic and anthropogenic substances are largely deposited wet. The exception to this sample view is the NC 25 station at the bottom of the figure which is located in southwestern North Carolina in a forest clearcut area.