The Changing Illinois Environment: Critical Trends

*Technical Report of the
Critical Trends
Assessment Project
Volume 1: Air Resources*

ENR
Illinois Department of
Energy and Natural Resources

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Volume 1: Air Resources

Volume 2: Water Resources

Volume 3: Ecological Resources

Volume 4: Earth Resources

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ABOUT THE CRITICAL TRENDS ASSESSMENT PROJECT

The Critical Trends Assessment Project (CTAP) is an on-going process established to describe changes in ecological conditions in Illinois. The initial two-year effort involved staff of the Illinois Department of Energy and Natural Resources (ENR), including the Office of Research and Planning, the Geological, Natural History and Water surveys and the Hazardous Waste Research and Information Center. They worked with the assistance of the Illinois Environmental Protection Agency and the Illinois departments of Agriculture, Conservation, Mines and Minerals, Nuclear Safety, Public Health, and Transportation (Division of Water Resources), among other agencies.

CTAP investigators adopted a "source-receptor" model as the basis for analysis. Sources were defined as human activities that affect environmental and ecological conditions and were split into categories as follows: manufacturing, transportation, urban dynamics, resource extraction, electricity generation and transmission, and waste systems. Receptors included forests, agro-ecosystems, streams and rivers, lakes, prairies and savannas, wetlands, and human populations.

The results are contained in a seven-volume technical report, *The Changing Illinois Environment: Critical Trends*, consisting of *Volume 1: Air Resources, Volume 2: Water Resources, Volume 3: Ecological Resources, Volume 4: Earth Resources, Volume 5: Waste Generation and Management, Volume 6: Sources of Environmental Stress*, and *Volume 7: Bibliography*. Volumes 1-6 are synopsized in a summary report.

The next step in the CTAP process is to develop, test, and implement tools to systematically monitor changes in ecological and environmental conditions in Illinois. Given real-world constraints on budgets and human resources, this has to be done in a practical and cost-effective way, using new technologies for monitoring, data collection and assessments.

As part of this effort, CTAP participants have begun to use advanced geographic information systems (GIS) and satellite imagery to map changes in Illinois' ecosystems and to develop ecological indicators (similar in concept to economic indicators) that can be evaluated for their use in long-term monitoring. The intent is to recruit, train, and organize networks of people — high school science classes, citizen volunteer groups — to supplement scientific data collection to help gauge trends in ecological conditions.

Many of the databases developed during the project are available to the public as either spreadsheet files or ARC-INFO files. Individuals who wish to obtain additional information or participate in CTAP programs may call 217/785-0138, TDD customers may call 217/785-0211, or persons may write:

Critical Trends Assessment Project
Office of Research and Planning
Illinois Department of Energy and Natural Resources
325 West Adams Street, Room 300
Springfield, IL 62704-1892

Copies of the summary report and volumes 1-7 of the technical report are available from the ENR Clearinghouse at 1/800/252-8955. TDD customers call 1/800/526-0844, the Illinois Relay Center. CTAP information and forum discussions can also be accessed electronically at 1/800/528-5486.

FOREWORD

"If we could first know where we are
and whither we are tending,
we could better judge what we do
and how to do it..."

Abraham Lincoln

Imagine that we knew nothing about the size, direction, and composition of our economy. We would each know a little, i.e., what was happening to us directly, but none of us would know much about the broader trends in the economy — the level or rate of housing starts, interest rates, retail sales, trade deficits, or unemployment rates. We might react to things that happened to us directly, or react to events that we had heard about — events that may or may not have actually occurred.

Fortunately, the information base on economic trends is extensive, is updated regularly, and is easily accessible. Designed to describe the condition of the economy and how it is changing, the information base provides the foundation for both economic policy and personal finance decisions. Typical economic decisions are all framed by empirical knowledge about what is happening in the general economy. Without it, we would have no rational way of timing these decisions and no way of judging whether they were correct relative to trends in the general economy.

Unfortunately, this is not the case with regard to changes in environmental conditions. Environmental data has generally been collected for regulatory and management purposes, using information systems designed to answer very site-, pollutant-, or species-specific questions. This effort has been essential in achieving the many pollution control successes of the last generation. However, it does not provide a systematic, empirical database similar to the economic database which describes trends in the general environment and provides a foundation for both environmental policy and, perhaps more importantly, personal decisions. The Critical Trends Assessment Project (CTAP) is designed to begin developing such a database.

As a first step, CTAP investigators inventoried existing data to determine what is known and not known about historical ecological conditions and to identify meaningful trends. Three general conclusions can be drawn from CTAP's initial investigations:

Conclusion No. 1: The emission and discharge of regulated pollutants over the past 20 years has declined, in some cases dramatically. Among the findings:

- Between 1973 and 1989, air emissions of particulate matter from manufacturing have dropped 87%, those of sulfur oxides 67%, nitrogen oxides 69%, hydrocarbons 45%, and carbon monoxide 59%.

- Emissions from cars and light trucks of both carbon monoxide and volatile organic compounds were down 47% in 1991 from 1973 levels.

- Lead concentrations were down substantially in all areas of the state over the 1978-1990 period, reflecting the phase-out of leaded gasoline.

- From 1987 to 1992, major municipal sewage treatment facilities showed reductions in loading of biological/carbonaceous oxygen demand, ammonia, total suspended solids and chlorine residuals that ranged from 25 to 72%.

- Emissions into streams of chromium, copper, cyanide, and phenols from major non-municipal manufacturing and utility facilities (most of them industrial) also showed declines over the years 1987-1992 ranging from 37% to 53%.

Conclusion No. 2: Existing data suggest that the condition of natural ecosystems in Illinois is rapidly declining as a result of fragmentation and continual stress. Among the findings:

- Forest fragmentation has reduced the ability of Illinois forests to maintain biological integrity. In one Illinois forest, neotropical migrant birds that once accounted for more than 75% of breeding birds now make up less than half those numbers.

- In the past century, one in seven native fish species in Lake Michigan was either extirpated or suffered severe population crashes and exotics have assumed the roles of major predators and major forage species.

- Four of five of the state's prairie remnants are smaller than ten acres and one in three is smaller than one acre — too small to function as self-sustaining ecosystems.

- Long-term records of mussel populations for four rivers in east central Illinois reveal large reductions in numbers of all species over the last 40 years, apparently as suitable habitat was lost to siltation and other changes.

- Exotic species invasions of Illinois forests are increasing in severity and scope.

Conclusion No. 3: Data designed to monitor compliance with environmental regulations or the status of individual species are not sufficient to assess ecosystem health statewide. Among the findings:

- Researchers must describe the spatial contours of air pollutant concentrations statewide using a limited number of sampling sites concentrated in Chicago and the East St. Louis metro area.

- Much more research is needed on the ecology of large rivers, in particular the effects of human manipulation.

- The length of Illinois' longest stream gaging records is generally not sufficient to identify fluctuations that recur less frequently than every few decades.

- The Sediment Benchmark Network was set up in 1981 with some 120 instream sediment data stations; by 1990 the network had shrunk to 40 stations, the majority of which have data for only one to three years.

CTAP is designed to begin to help address the complex problems Illinois faces in making environmental policy on a sound ecosystem basis. The next edition of the Critical Trends Assessment Project, two years hence, should have more answers about trends in Illinois' environmental and ecological conditions to help determine an effective and economical environmental policy for Illinois.

CONTENTS

AIR RESOURCES: VOLUME SUMMARY

REPORT OVERVIEW

Although considerable progress has been made in improving the environment, national and state goals for environmental quality still have not been completely met, and additional effort and expenditures will be necessary. To use public funds as effectively as possible, Illinois environmental officials must set priorities so that the most serious problems receive the most attention. The priorities should be based on an assessment of the current status of Illinois' environment. This technical report on air resources is one of a series aimed at documenting and assessing the status of Illinois' environment. It includes comprehensive information about Illinois' climate and air quality and the deposition of atmospheric constituents on the earth's surface. In each subject area, currently available data have been assembled to provide a picture of how air resources have changed over time and how they vary spatially from place to place around the state.

SUMMARY AND SIGNIFICANT RESULTS

Climate Trends

This report presents an analysis of climate trends since the late nineteenth century in Illinois. This work is based on a variety of climate data and information from Illinois sites. Long-term daily temperature and precipitation data available from 41 locations formed the basis for much of the analysis of temperature and precipitation in this report. More detailed observations from National Weather Service office sites (including Chicago, Springfield, Peoria, Moline, St. Louis, and Evansville) were used to assess trends in cloud cover, freezing precipitation, and visibility. Citizen reports were used to assess trends in tornado frequency.

Although long-term trends can be identified in various climate parameters, the dominant characteristic of Illinois' climate is the presence of variability on all time scales from decades to a few years or less. That is, the magnitude of long-term trends is in general considerably less than the changes that can occur from one year or a few years to the next. With that being said, however, some identifiable long-term features can be noted.

The most persistent and extreme period of summertime drought and heat occurred during the 1930s, the Dust Bowl era. This mirrors the long-term average temperatures in Illinois, which increased by 4 to 5°F from the mid- to late 1800s to the 1930s, and then cooled by about half that amount to the present. This trend in temperature is consistent with trends found in global records. Consistent with the cooling trend after about 1930, generally benign and moderate summers occurred during the 1960s and 1970s. However, since that time, the 1980s were characterized by more frequent hot and dry summers, similar to the first half of the century. In fact, the severity of the 1988 growing-season drought in Illinois has only been equaled in two other years since the turn of the century.

In general, the frequency of extreme cold events has increased from about 1930 to the present. This upward trend reached its peak in the late 1970s and early 1980s, with an unprecedented string of extremely cold winters. However since the early 1980s, winters have generally been on the benign side.

Tornado frequency records exhibit large variability. Reliable records of tornado frequencies are limited to the past three decades. In that time, no upward or downward trend in frequencies has been identified.

It is clear that climate has not been stable in Illinois during the last 150 years, nor should it have been so anticipated. Many of the changes, although abrupt, were of relatively small magnitude. Yet those relatively small-scale changes levied a substantial toll on the inhabitants of the state through discomfort, lost income, increased costs, and impediments to commerce and agricultural production, in spite of major strides. The past gives little hint as to the direction and magnitude of any possible future changes, but it does suggest the degree to which climate may change in the future, and demonstrate the magnitude of that impact on human activities.

Air Quality

Air quality can have direct effects on human and ecosystem health. Thus it is necessary to examine current air pollutant concentrations and their spatial variations over the state. We also need to know whether concentrations are increasing, decreasing, or remaining constant, especially in the major population centers. Finally, we seek to identify gaps in air quality data and research that need to be filled to permit wise planning for the future.

This assessment of air quality is based primarily on Illinois Environmental Protection Agency (IEPA) measurements of seven pollutants for which state or national standards have been set, plus eight additional pollutants for which standards have not been set. These data cover the time period from 1978-1990. Summary data published in IEPA annual reports were tabulated in computer spreadsheet and Geographic Information System (GIS) files and are available to others.

Temporal changes in pollutant concentrations are depicted graphically using box plots, which concisely show several features of observed distributions of concentrations at sampling sites within various geographic areas. Spatial variations of average pollutant concentrations in the Chicago area are shown by plots of concentration contours. Trends in concentrations over time were tested for statistical significance using the nonparametric Spearman Rank Correlation Coefficient. Statistical tests were also carried out to identify significant differences in concentrations between geographic regions.

For the criteria pollutants tested for time trend in any geographic region of the state, results indicated only decreasing trends or no significant (5 percent) trends. No increasing trends in criteria pollutants were detected. For the state as a whole, seven of twelve pollutant/averaging time datasets tested for time trend showed significant trends (5 percent or better) toward decreasing concentrations over the 1978-1990 period. After accounting for temperature effects, O_3 showed a significant decreasing trend (2 percent level), rather than no trend.

In the Chicago area, eight of the twelve datasets showed decreasing trends at the 5 percent level (all but one of these were at the 1 or 2 percent level). After accounting for temperature, O_3 (ozone) in the Chicago area showed a decreasing trend (2 percent level, table 3 of *Air Quality Trends in Illinois* chapter), rather than no trend. In the MetroEast area, only four pollutant/averaging time data sets were measured at enough sites to warrant testing for time trend. Only Pb (lead) showed a significant (5 percent) trend toward decreasing concentrations. After accounting for temperature, O_3 showed a decreasing trend at the 6 percent level, but not the 5 percent level. Three of nine datasets tested for time trend in the "remainder" region showed decreasing trends at the 5 percent level or better.

The eight noncriteria pollutants were all tested for trend in all four areas mentioned above, except for the MetroEast region, where the data were not adequate to test Cr (chromium) and Ni (nickel). Of the 30 trend tests, 20 showed no significant (5 percent) trend. Over the state as a whole, and in the Chicago area, two species showed significant decreases—SO_4^{2-} (sulfaate ion) and As (arsenic). In the MetroEast area, Fe (iron) and Mn (manganese) showed significant increases (the only increasing trends found in this study). In the "remainder" region, SO_4^{2-}, As, Cd, and Mn showed significant (5 percent or better) decreases.

Comparison of median and maximum pollutant concentrations within geographic regions, from the yearly box plots in figures 1-30 (*Air Quality Trends in Illinois* chapter), indicates which geographic areas of the state experienced the highest concentrations of air pollutants. Chicago generally had higher median regional values of annual mean NO_3^- (nitrate ion), and annual mean and 24-hour Cr. It also experienced higher median annual mean Ni and the highest individual 24-hour Ni concentrations. On the other hand, the Chicago area generally experienced lower concentrations of 3-hour and 24-hour SO_2 than the rest of the state.

The MetroEast area generally experienced higher concentrations than the rest of the state for annual mean Pb, annual mean TSP (total suspended particulate matter), and both 24-hour and annual mean As, Cd, Fe, and Mn. The Chicago and MetroEast areas experienced higher concentrations than the rest of the state for 1-hour maximum O_3 and annual mean SO_2.

The analyses of spatial distribution of pollutant concentrations in the Chicago area showed that only one location stands out for its high concentrations of multiple pollutants. This is the industrial area of southeast Chicago around Lake Calumet. This area has persistent, relatively high annual mean or 24-hour concentrations, or both, of SO_4^{2-}, Fe, Mn, and Pb, and possibly Cd, Cr, and Mn. The evidence for the latter three is somewhat weaker than for the others, however. Other locations in the Chicago area appeared to have persistent high concentrations of only one or two pollutants.

Atmospheric Deposition

Atmospheric deposition is an ensemble of environmental processes by which airborne pollutants from various sources are delivered to receptor systems at the earth's surface. Among the six natural environmental receptors treated by the Critical Trends project, atmospheric deposition is considered for two, forest ecosystems and lakes and impoundments, because research has shown possible damage to these receptors from certain kinds and amounts of atmospheric deposition. The character-

istics of atmospheric deposition in Illinois, and how it varies across the state and throughout the year are described in this chapter. Where there are data sufficient for an analysis, changes over several years are calculated and trends are inferred, if these changes are significant. Also shown are maps of deposition loadings, which together with the concentration data provide information necessary for assessments of the exposure of Illinois' natural environment to atmospheric deposition. While an explicit description of the source-receptor relationships for major pollutants, such as sulfur dioxide (SO_2) and nitrogen oxides (NO_x), is not considered, the sources of these pollutants in Illinois and surrounding states are compared to their occurrence in atmospheric deposition. Finally, additional work is discussed that is needed to improve our assessment of atmospheric deposition in Illinois and over Lake Michigan.

Atmospheric deposition includes gases and aerosols that are solid, liquid, or mixed phase. It includes both primary pollutants, which retain their chemical identity between source and receptor, and secondary pollutants, which undergo transformation during transport in the atmosphere. Deposition of pollutants from the atmosphere is a continuous process, though there are large temporal variations in the deposition rate or flux. These variations relate to the kind of deposition that is occurring and to surface and atmospheric conditions. There are two kinds of atmospheric deposition, wet and dry. Wet deposition is defined as the delivery of pollutants to the surface by precipitation. Dry deposition is the delivery of gases and aerosols to the surface by mass transfer processes other than precipitation. In principle, dry deposition occurs continuously, while wet deposition occurs episodically, e.g., when it rains.

Wet deposition is measured by chemically analyzing precipitation. For this project, wet deposition data from the national network, the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) were used. The NADP/NTN reports the concentrations of dissolved calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), ammonium (NH_4^+), sulfate (SO_4^{2-}), nitrate (NO_3^-), chloride (Cl^-), orthophosphate (PO_4^{3-}), and free hydrogen ion (H^+), measured in pH units. No trace metals or organics are reported by the NADP/NTN. Samples are filtered to remove insoluble materials, so NADP/NTN provides data for the soluble major inorganic ions found in precipitation, those chemicals that result in acidic deposition or "acid rain," which occurs over all of Illinois.

Dry deposition includes the mass transfer of pollutants to the surface by a variety of physicochemical processes: turbulent diffusion, diffusion followed by surface sorption of gases, gravitational settling of large particles, impaction, and interception of solid and liquid particles. Dry deposition fluxes are strongly affected by atmospheric factors, which influence the rate at which pollutants are delivered to a receptor surface; and by surface factors, which influence the efficiency with which pollutants "stick" to a receptor surface. Among the atmospheric factors is wind speed and turbulence, air temperature, solar radiation, and relative humidity. Among the surface factors are roughness, wetness, surface-to-air temperature difference, and type of surface, which includes whether the surface is animate (plant) or inanimate.

The relative importance of these factors in determining the dry deposition rate depends also on the physical and chemical nature of the pollutant. For example, factors that affect the mass transfer of carbonaceous soot, an unreactive, insoluble particle, are much different than the ones affecting nitric acid, a highly reactive, soluble gas. The dry deposition of gases and submicron aerosols involves highly complex processes, and direct measurements are intractable on a spatial domain the size and complexity of Illinois. For this reason, an indirect method is applied to infer, rather than measure, dry deposition fluxes.

This inferential method employs a conceptual model that estimates an atmosphere-to-surface coupling parameter known as the "deposition velocity" V_d. The dry deposition flux is the product of V_d and the measured air concentration of a particular pollutant. Model inputs, including the atmospheric and surface factors discussed earlier, are measured at a network of sites sponsored by the National Oceanic and Atmospheric Administration's Atmospheric Turbulence and Diffusion Laboratory (NOAA/ATDD). Land-use and vegetation type and status are also reported at these sites, along with the airborne concentrations of Cl^-, $SO_4^=$, particulate NO_3^-, nitric acid vapor (HNO_3), and SO_2. The NOAA/ATDD sampling system is especially designed to *exclude* large particles, since the inferential method of calculating dry deposition applies specifically to gases and submicron aerosols.

The dry deposition of large particles, which have an aerodynamic diameter greater than 1 micrometer (μm), is typically estimated from an analysis of the mass of a pollutant accumulated on a surrogate surface. To estimate the dry deposition of large particles (i.e., sedimentation or dryfall) for CTAP, data from the

NADP/NTN were used. NADP/NTN measures dryfall in samples taken from the same collector used for precipitation. This device, a wet/dry collector, has two identical containers; it discriminates between wet and dry conditions, exposing the wet deposition container during precipitation and the dryfall container at all other times. Dryfall samples are sent for analysis of the same analytes measured in precipitation.

Wet deposition in Illinois has been monitored for ten or more years at eight NADP/NTN sites. NADP/NTN reports the concentrations of ten separate chemical pollutants in precipitation. Just five of these are needed to account for about 90 percent of the chemical composition that causes Illinois precipitation to be acidic. They are, in order of importance: sulfate (SO_4^{2-}) > hydrogen ion (H^+) > nitrate (NO_3^-) > ammonium (NH_4^+) > calcium (Ca^{2+}). Illinois precipitation is most simply described as a dilute solution of mineral sulfuric and nitric acids, partly neutralized by ammonium and calcium. Based on statistical tests of time series data alone, there is no unambiguous trend that applies to all of the important pollutants causing acid rain in Illinois. Based on a "weight of the evidence" analysis, however, several points can be made about Illinois precipitation chemistry changes during the 1980s:

1. Sulfate has decreased 2 to 4 percent per year in the southern third of the state, with smaller decreases elsewhere.

2. Calcium decreased by 3 to 7 percent per year, except at Argonne (suburban Chicago), where it remained steady.

3. Nitrogen species, ammonium and nitrate, remained unchanged.

4. pH increased, but the increase is too small and too variable to be quantified.

5. Sulfur dioxide and NO_x emissions have decreased slightly.

Dry deposition in Illinois tends to be somewhat higher in the Chicago area, both due to higher airborne concentrations of most pollutants and higher deposition velocities. For sulfate, nitrate, sulfur dioxide, ozone, arsenic, and manganese, the differences are on the order of 10 to 30 percent. For cadmium, chromium, iron, nickel, lead, and total suspended particles, dry deposition in the Chicago area exceeds the rest of the state by 200 to 400 percent, which is primarily caused by the differences in air quality (see *Air Quality Trends in Illinois* chapter). Temporal trends in dry deposition generally follow air quality trends, although additional variability is introduced into the time series data by

interannual variation in deposition velocities. More important for ecological impacts is the seasonal nature of dry deposition loadings, with higher deposition velocities for many pollutants occuring during the warm season, when biological impacts may also be the greatest.

The total deposition of sulfur in the Chicago area is about 15 percent higher than in the rest of the state. For sulfur (sulfate *plus* sulfur dioxide), the ratio of wet to dry deposition is about 1 part wet to 1.5 parts dry. The deposition of nitrogen in the Chicago area is about 30 percent higher than in the rest of the state. For nitrogen (nitrate *plus* ammonium *plus* nitric acid vapor), the ratio of wet to dry deposition is about 1 part wet to 3 parts dry.

The spatial and temporal variation information is most useful in describing the coupling of the atmosphere to receptors that are also distributed in space and have temporally-varying sensitivity to the depositing pollutants. Acid deposition to forests for example, is most likely to have an effect during the growing season, and much less likely to be harmful in the dormant season. Toxic deposition to forests, however, may act through a *cumulative* effect, where the temporal variation is less important to understanding the impact on the receptor system.

Acid deposition to Lake Michigan presents a special difficulty in this analysis, since neither wet nor dry deposition is measured over the lake, although the refinement of estimates based on shoreline measurements is an ongoing research topic.

Agricultural systems have been shown to be relatively insensitive to current levels of acid deposition, but the impact of toxic deposition and dry deposition of many pollutants is unknown. Ozone has been demonstrated to have negative impacts on yield and quality of cash crops in several areas of the United States. The role of wet and dry toxics deposition as a contributor to nonpoint source pollution in surface and ground-water supplies for human consumption is also unknown at this time. The impact of atmospheric deposition (acid rain, toxic pollutants, and biological nutrients) to lakes and streams in Illinois (i.e., other than Lake Michigan waters) has not been documented, although consideration of the magnitude of deposition for many chemicals would indicate that significant impacts are possible. Finally, the impacts of SO_4^{2-}, SO_2, acids, and NO_3^- deposition, both in precipitation and dry deposition, has been demonstrated in recent federally sponsored research in many areas of the United States. Materials impacts in Illinois are as yet unquantified, but they are potentially large.

INTRODUCTION

CRITICAL TRENDS ASSESSMENT PROGRAM (CTAP) OVERVIEW

Large amounts of money have been and are being spent on pollution control efforts to protect human health and the environment. Although these efforts have had considerable benefits, many environmental goals still have not been met nationally and in Illinois. Thus, we may expect that the expenditures for environmental improvement will continue and perhaps increase.

It is incumbent upon state officials and environmental policy makers to use public funds for environmental improvement wisely. To use funds wisely in this context means to effect the greatest possible environmental improvement from the available resources. This implies that priorities must be set for the use of public funds for environmental improvement. What criteria should govern the setting of priorities? The U.S. EPA's Science Advisory Board (1990) stated that "policy affecting the environment must become more integrated and more focused on opportunities for environmental improvement than it has in the past. Integration in this case means that government agencies should assess the range of environmental problems of concern and then target protective efforts at the problems that seem to be the most serious."

The first step in addressing these issues is to assess the state of the environment. By executive order, Illinois' Governor Edgar directed the Department of Energy and Natural Resources (ENR) and the Governor's Science Advisory Committee (GSAC) to "conduct an environmental trends analysis and to report to the public on the state of the Illinois environment." The goals of this effort are:

1) to document techniques for searching, organizing, analyzing, interpreting, and evaluating data for geographic and temporal presentation,

2) to document assessment methodologies for analyzing environmental and human health information,

3) to identify critical data gaps, for use in prioritizing future research,

4) to establish an integrated data structure that will provide the means to judge trade-offs associated with anticipated changes in the state's natural environment or infrastructure, and

5) to produce a final report that documents past conditions and assesses the present status of the Illinois environment.

RELATIONSHIP OF AIR RESOURCES TO CTAP PROJECT GOALS

The atmospheric environment is an important component of the overall environment of the state. Weather and climate affect a broad range of ecosystem and human health issues. For example, temperature and moisture conditions and their seasonal changes determine to a great extent the geographical distribution of plant and animal species. Droughts and floods have great impacts on agriculture and transportation. Similarly, extremes of temperature and precipitation and severe weather are major hazards to human health.

Exposure to airborne contaminants also has impacts on the environment and human health, as evidenced by the use of human and environmental health as criteria in setting air pollution standards. Cases of injury to plants and human health caused by extreme concentrations of airborne pollutants are well documented in the literature.

Atmospheric deposition of airborne nutrients and contaminants is more important to ecosystem health than human health at current levels. Benefits may be realized from the atmospheric deposition of nutrients to agricultural lands, although excess nutrients in water bodies can lead to problems. Deposition of acidifying substances can lead to acidification of water bodies and ecosystem damage in certain situations.

REPORT COMPONENTS AND STRUCTURE

An assessment of the status of the atmospheric environment must include statements related to both its physical features (which over the short term we call weather, and over the long term, climate), its chemical composition, primarily as related to the concentrations of pollutants in air and precipitation, and the deposition of airborne materials back to the earth's surface. An assessment of the state of Illinois' atmospheric environment should include representations of both the spatial distributions and the time history of important features of weather and climate, as well as pollutant concentrations and deposition.

This report is composed of three major sections: *Climate Trends in Illinois*, *Air Quality Trends in Illinois*, and *Atmospheric Deposition Trends in Illinois*. Each presents information on the current status and temporal and spatial trends of important features of the Illinois environment, based on currently available data.

CLIMATE TRENDS IN ILLINOIS

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INTRODUCTION

This report presents an analysis of climate trends since the late nineteenth century in Illinois. This work was pursued under the Critical Trends Assessment Project (CTAP) of the Illinois Department of Energy and Natural Resources. Weather and climate events have significant impacts on many socioeconomic and environmental aspects of Illinois. Therefore, an analysis of climate trends provides the necessary background for understanding trends in other aspects of the Illinois environment.

Significant climate variability can occur at virtually all time scales. Therefore, a variety of climate data and information from Illinois sites are assembled here in order to investigate the temporal variability of various climate parameters. In addition to studying statewide changes in annual temperature, precipitation, and other climate parameters since the turn of the century, changes by month and by season and in the frequency of extreme events will be investigated over the period of record.

CAUSES OF CLIMATE

Climate is a description of the overall character of daily weather conditions that is manifested from season to season and from year to year. It is the sum of all statistical weather information that describes the average and variability of weather at a given place. Climate does not include information on the weather of a particular day.

We investigated changes in the frequency and magnitude of several climatic parameters. Some of these yield a clear indication of a trend. For example, all stations indicate a common temperature trend over the past 100 or more years, with associated changes in related climatic parameters such as degree-day totals, cloud cover, frequency of frozen precipitation, etc. It is not yet possible to definitively identify the specific causes of these trends. However, it is useful to discuss in a general way some of the possible causes of climate variability to provide a perspective on the analyses presented in this report.

Ultimately, the climate of the earth is driven by energy received from the sun. Although some of that energy is reflected back to space, most of it remains in the atmosphere to provide the fuel for global circulation patterns. The single most important variable affecting climate at a particular point is latitude because this affects the total amount and seasonal distribution of solar radiation reaching the surface at that point. The mid-latitude location of Illinois results in a substantial variation in the amount of solar radiation received between winter and summer. Thus, the seasonal variation in climate is relatively large.

It is known that variations in the orbit of the earth around the sun result in cyclic variations in solar radiation on time scales of greater than 10,000 years. Many scientists believe this to be a major factor in the periodic occurrences and retreat of ice ages. However, on shorter time scales (100 years or less), it is not known with certainty whether significant variations in solar radiation occur. Measurements of solar radiation are not accurate enough nor of long enough duration to identify significant fluctuations.

Another major factor affecting climate is the distribution of continents and oceans. It is known that temperature differences between the ocean surface and the land surface are a major factor in establishing certain circulation patterns in the atmosphere. For instance, during the summertime, the Atlantic Ocean is cooler than the North American continent. This results in a pressure difference (high pressure over the ocean, low pressure over the continent) that forces a southerly flow of air over eastern North America, transporting significant moisture from the Gulf of Mexico into Illinois. This gives us our humid summertime climate with generally abundant rainfall.

It is known that fluctuations in ocean surface temperatures can occur and that these can cause fluctuations in climate that are of significance. The El Niño phenomenon is one example, where a warm pool of water develops aperiodically over the eastern equatorial Pacific. This warm pool of water and the associated disruptions in equatorial wind patterns are known to have effects on climate in many other parts of the globe. It is possible, even probable, that fluctuations in ocean surface temperatures in the Pacific and the Atlantic have effects on climates thousands of miles

away and may be responsible for some significant fluctuations that have occurred in the past. However, it is very difficult to identify such ocean temperature changes as the clear cause of any particular climate fluctuation experienced in Illinois.

The radiation balance and energy transfer within the atmosphere are influenced by the composition of the atmosphere. Some constituents of the atmosphere, notably carbon dioxide, methane, chlorofluorocarbons, and water vapor, limit the loss of infrared radiation by the earth to space. These have a net warming effect on surface temperatures. It is well known that the concentration of many of these gases are increasing because of human activities, such as the burning of fossil fuels. If all other causes of climate were to remain constant, we would expect that the temperature at the surface of the earth would increase as the concentration of these gases increased. However, it is still too early to ascribe observed changes in Illinois climate solely to this cause.

Other factors may also play a role in modulating Illinois' climate. For instance, volcanic eruptions can inject large amounts of dust into the earth's stratosphere. This can decrease the amount of solar radiation reaching the earth's surface for a year or more in some cases. It is likely that past volcanic eruptions have caused significant changes in the earth's climate. For instance, the recent interruption of Mt. Pinatubo is believed to have resulted in a temporary decrease in global surface temperature of 1°F. In large urban areas such as Chicago, the change from a natural or agricultural land surface to large percentages of paved areas and buildings can cause a local change in climate. Urban areas tend to be warmer than nearby rural areas.

Since there are multiple factors that can affect Illinois' climate, it is difficult, if not impossible, to cite a specific cause for any particular feature or trend in Illinois' climate.

DATA

Data from two major categories of climate stations are analyzed in this report. The first category includes the National Weather Service (NWS) First Order Sites (FOS). They provide data from at least hourly measurements taken at major airports. The data are derived from continuous observation of cloud cover, visibility, pressure, temperature, dew-point temperature, winds, hydrometeors, and other obstructions to visibility. This very detailed set of observations is archived in digital form for only five sites in Illinois.

The second major category of observing sites are those of the NWS Cooperative Observer Network (CON). This network is composed primarily of individual and institutional volunteers who obtain measurements on a daily basis for the NWS. The NWS provides the measurement equipment, and for the most part, the observers are not paid. The observations include daily maximum and minimum temperatures, daily precipitation, daily snowfall and snow depth, and in a few cases soil temperature and pan evaporation. Although these observations are not as detailed as those for the FOS network (they provide daily rather than hourly observations), the network is much denser and therefore offers a much-used, quality dataset. For this report, 41 CON stations have been selected, each with long-term data extending back to the turn of the century.

For purposes of climate applications, the NWS has divided the state into nine regions, known as "climate divisions." They are shown in figure 1. The National Climatic Data Center has developed a special dataset that consists of temperature and precipitation values averaged for all stations, as available in each of these climate divisions. These values are available monthly from 1895 through the present. Because this is a relatively compact dataset and incorporates information for all available stations, these data were used to assess overall trends in precipitation and temperature.

Data from individual stations were used to assess trends in more detailed aspects of temperature and precipitation that are not available by climate divisions. Although climate division values are available for monthly average temperature and monthly total precipitation, no such data are available on a daily time scale. Therefore, trends in the following climatic elements were assessed from the data from individual stations:

Temperature
- *Monthly maximum and minimum temperatures.*
- *Number of days with daily mean temperature above 50°F.* This threshold is examined because the rate of growth and development of corn and soybeans becomes significant when the daily mean temperature is above 50°F.
- *Extreme daily maximum temperature.* This is the highest single daily value occurring in a month.
- *Extreme minimum daily temperature.* This is the lowest single daily value occurring in a month.
- *Number of days with daily minimum temperature above 70°F.* This is a somewhat arbitrary threshold that is meant to represent summer nights of abnormal warmth.

Figure 1. Illinois climate divisions, designated by the National Weather Service.

- *Number of days with daily minimum temperature below 0°F.* This threshold is examined as a representation of particularly "cold" nights.
- *Number of days with daily minimum temperature below 32°F.* This threshold is examined because the freezing point affects many processes.
- *Number of days with daily maximum temperature above 86°F.* This threshold is examined because the rate of corn and soybean growth and development typically reach a plateau near this temperature threshold. That is, above this threshold, the rate of crop growth and development no longer increases with rising temperature.
- *Number of days with daily maximum temperature above 90°F.* This is a somewhat arbitrary threshold, representing particularly warm days of discomfort to many people.
- *Number of days with daily maximum temperature above 100°F.* This threshold is examined as a representation of the frequency of days of great discomfort to many people.

Precipitation
- *Number of days with precipitation.*
- *Number of days with precipitation of 1 inch or more.* This threshold is examined as a somewhat arbitrary representation of rather heavy rain days.
- *Total monthly snowfall.*
- *Number of days with snowfall.*

METHODOLOGY - STATISTICAL TECHNIQUES

Trends in Means

In order to detect potential trends in climate, several variables were subjected to a mathematical process to remove the seasonal cycle in the data. This process was performed on climate division values of monthly temperature and monthly precipitation and on station values of monthly minimum and maximum temperatures. Specifically, the data were "standardized" with the following formula:

$$x_{ij} = (x_{ij} - \overline{x}_j)/s_j$$

where x_{ij} is the value for year i and month j; $\overline{x}_j$ is the average over all years of the values in month j; and s_j is the corresponding standard deviation for those values. These converted values are referred to as "standard-

ized" temperature and precipitation. They express the values in terms of the number of standard deviations above or below the mean.

The climate division time series consisted of 1,164 data values for each element (97 years x 12 months). Overall trends were assessed by subjecting the time series to a linear regression; the statistical significance of the resulting trends was tested using a t-test for significance of slope of the regression (Draper and Smith, 1981). Trends in individual seasons were also assessed. The meteorological definition of seasons was used:

- *Winter* (December 1 through February 28/29)
- *Spring* (March 1 through May 31)
- *Summer* (June 1 through August 31)
- *Fall* (September 1 through November 30)

Seasonal values were obtained by averaging temperature over the three months and totaling precipitation over the three months. Thus, the resulting seasonal time series each consisted of 97 data points. For the time series of individual stations, only 91 years of data were available.

For variables involving the number of exceedances in temperature and precipitation and for monthly snowfall totals, the monthly values were summed for each year. The resulting data (91 data points) were subjected to the regression analysis for trend indicated above. The extreme monthly maximum temperatures were analyzed by taking the highest value in each year and subjecting each of the 91 values to the regression analysis for trend. The lowest extreme monthly minimum temperature value for each year was also assessed for trend in this manner.

In all of the above regressions, the trends were tested for significance at the 10 percent level using a two-sided t-test.

Variance Assessments

In addition to testing the time series for trends in the mean, the data were also analyzed for changes in the variability of temperature and precipitation over the period of record. A subjective analysis of the time series suggested that certain periods of time, a decade or more in length, were characterized by significantly less or more variability than the rest of the time series. To assess this, the climate division values of temperature and precipitation were broken into six 15-year segments, beginning in 1901. For each 15-year subset

of data, the standard deviation of the time series was calculated. The resulting data were plotted and subjectively analyzed.

Temperature and Precipitation Index

An additional objective of this study was to identify certain periods of time characterized by specific climate conditions—in particular, multiyear periods characterized as:

- Warm and wet
- Warm and dry
- Cool and wet
- Cool and dry

To assess these conditions, two combined temperature/precipitation indices were defined and calculated as follows:

- Index 1 = standardized temperature + standardized precipitation
- Index 2 = standardized temperature - standardized precipitation

The association between these indices and the above scenarios is:

- Warm and wet (large positive values of Index 1)
- Warm and dry (large positive values of Index 2)
- Cold and wet (large negative values of Index 2)
- Cold and dry (large negative values of Index 1)

Modifications to this rather straightforward categorization scheme were required, as will be explained as part of the section *Anomalous Episodes from the Illinois Climate Record*. Time series of the index were plotted and examined to identify multiyear periods with the above climate conditions.

TRENDS IN PRECIPITATION

Trends in Average Precipitation

Mean annual statewide precipitation, averaged for all available stations since 1840 is shown in figure 2. The data are considered to well represent statewide conditions since about 1890, by which time the annual values were derived from at least 113 stations in the state. The "statewide" totals during the 1880s were

based on 18 stations; on six or seven stations from 1852 to 1880; and on only one station (Athens, Sangamon County) from 1840 to 1851. Because of the paucity of data prior to about 1890, some care must be exercised in its interpretation. If representative, they indicate a slight decline in annual precipitation from the mid-1800s to about the turn of the century, with an increase thereafter. However, as shown, the year-to-year variation is the dominant characteristic of statewide precipitation, as with an individual station's record.

The statewide mean annual precipitation record based on 150 to 200 sites for each year (figure 3) has varied widely from 25 to 50 inches. (Figure 3 duplicates the more recent portion of figure 2. This time frame facilitates comparison with other figures in this report, most of which cover the period from 1901 to 1991.) Occasionally, consecutive years exhibit similar anomalies, but in the vast majority of cases, one year's precipitation is unrelated to that of the next. The statewide average is about 37 inches, with about 5 inches less in northern counties and about 5 inches more in the south.

No obvious discontinuity in statewide annual precipitation has occurred since 1901. Changnon (1984), in an analysis of data through 1980, also noted that year-to-year variability was the dominant characteristic of the time series. He noted a slight tendency for lower values during the period 1910 to 1940 compared to values both before and after that 30-year period. A close examination of figure 3 does suggest this. In addition, an analysis of trends of individual stations (reported later) also suggests a general upward trend in precipitation during this century. However, this trend is weak compared to the short-term variability.

The number of days per year with precipitation (averaged for all long-term stations) exhibits a somewhat stronger upward trend since 1901 (figure 4) than does annual precipitation. The number of days per year with precipitation appears to have increased by about 10 during the 90 years of record, although variability appears to increase as well.

The annual number of days per year with precipitation trended upward with time at nearly all the 41 long-term stations; only Carbondale (Jackson County) and Walnut (Bureau County) digressed. Thus, the trend has been rather ubiquitous throughout the state. However, several of these upward trends were rather weak and not statistically significant.

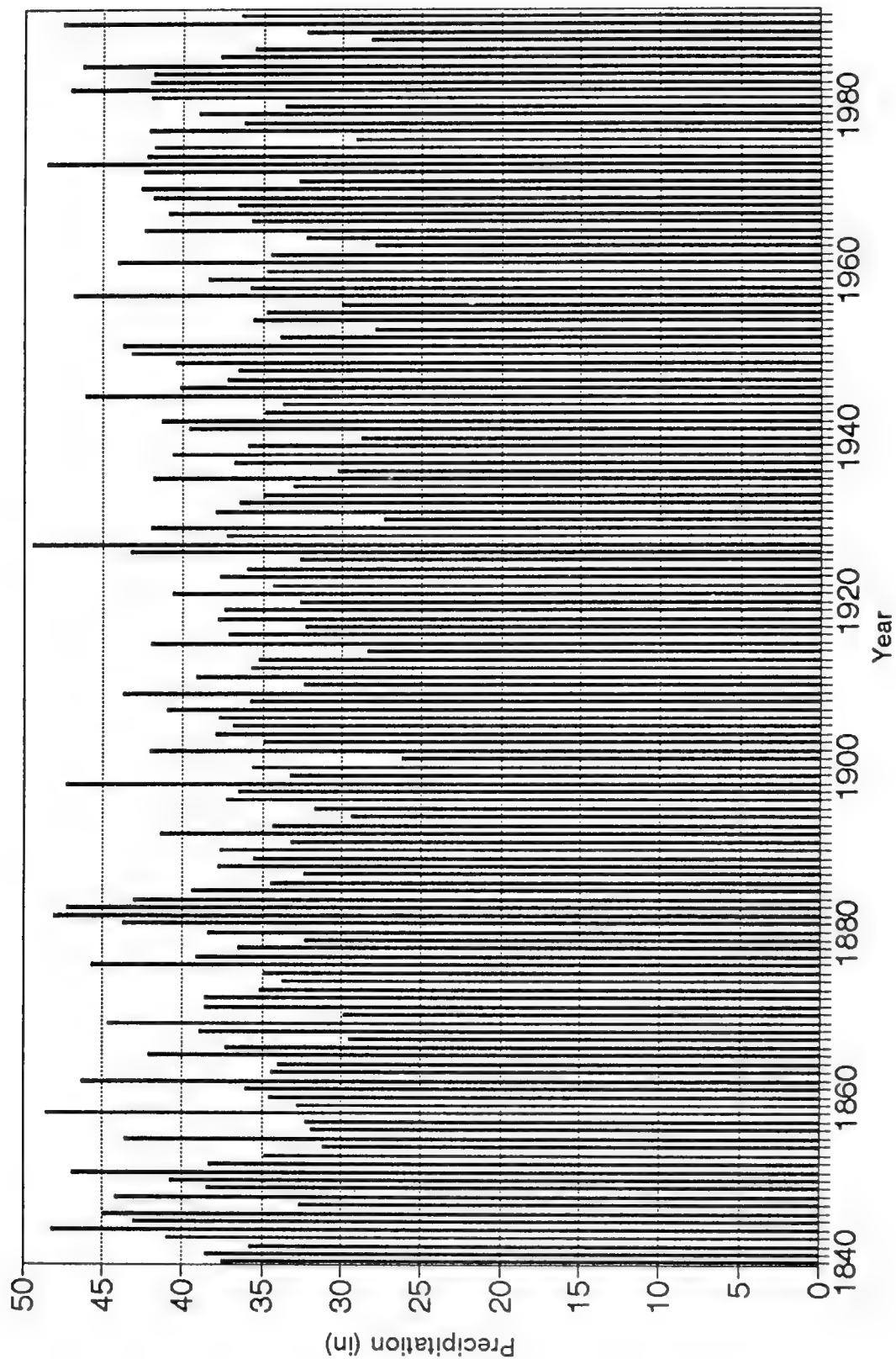The number of days per year with at least 1 inch of precipitation (with a present recurrence frequency of

Figure 2. Statewide average annual Illinois precipitation composed of available sites, 1840-1992 (inches).
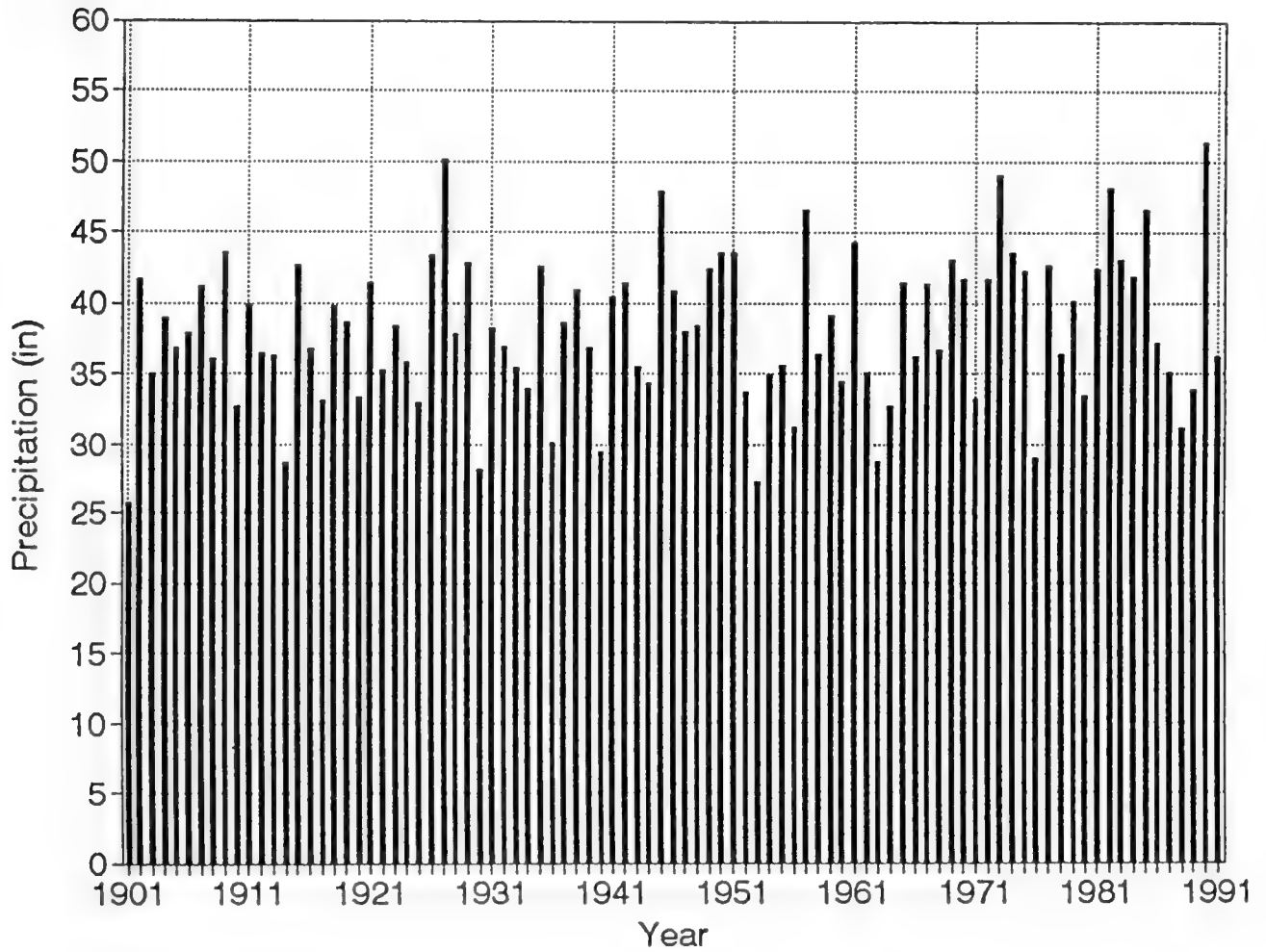
Figure 3. Statewide average annual Illinois precipitation composed of available sites, 1901-1991 (inches).
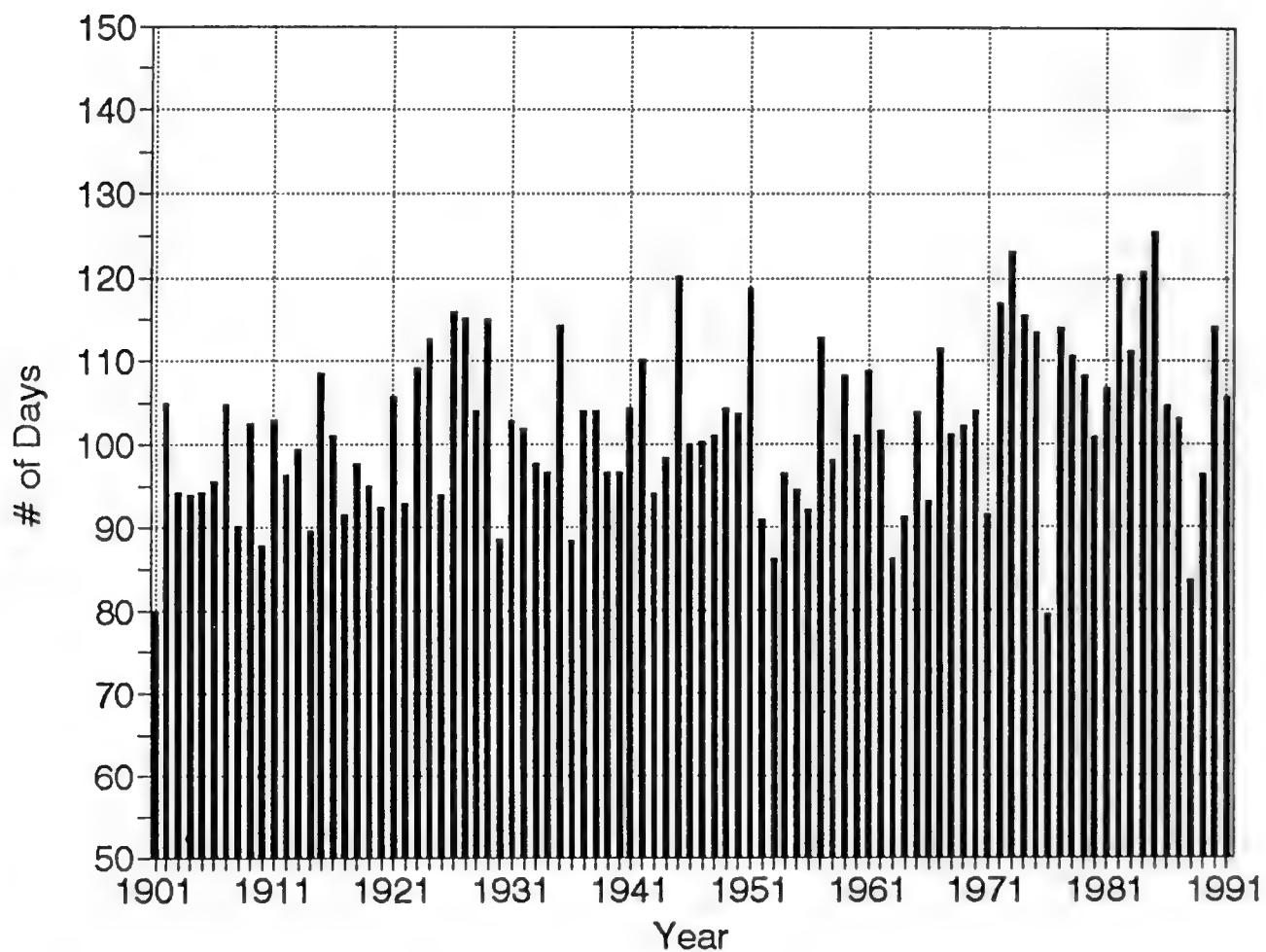
Figure 4. Average annual number of days per year in Illinois with precipitation.

about once every other month in Illinois) is shown in figure 5. As with the above two precipitation measures, identification of trends is difficult. Nonetheless, a slight decline appears during the first few decades with an increase during recent decades. The days per year with a least 1 inch of precipitation at individual stations in the state all exhibit upward trends, except for White Hall (in Greene County). Changnon (1984) found a general increase in the number of days with rains of 2 inches or more during the warm season. In addition, Huff and Angel (1989) found that the frequency of very heavy rainstorms had increased from the early part of this century to the latter part.

To summarize temporal trends in precipitation over the state, magnitude (figure 2) appears to decline from 1840 to about the 1930s, increasing thereafter. The frequency of precipitation events appears to have increased slightly in the 90 years since 1901, paralleling the trend in annual precipitation. Much more significant is the substantial year-to-year variation.

Trends significant at the 10 percent level, were identified for 9 to 21 of the 41 sites for the five precipitation parameters. Trends for individual stations are listed in appendix table A.1, and a summary is given in table 1. Interestingly, nearly all these trends were positive, except for the number of days with measurable snowfall. Sites in northern and east-central Illinois were somewhat more likely to have experienced significant trends than sites in other parts of the state.

Trends in Snowfall

Annual snowfall for the state since 1901 (figure 6) shows some rather clear characteristics:

1. Decline from 1901 to the 1930s.
2. Increase from the 1930s to the 1980s.
3. Decline in the last decade.
4. Continuity from year-to-year over episodes of three to six years. For example, note similar snowfall amounts from 1903-1906, 1919-1923, 1939-1945, 1952-1959, 1961-1965, and 1987-1991.

Because the year-to-year variation is so large, five-year running averages of statewide annual snowfall were calculated (figure 7). This figure exhibits a slight increase with time, but much more apparent is the fluctuating nature, as with annual precipitation. Fluctuations in annual snowfall exhibit secular episodes of about 13 years, varying from 7 to 17 years since 1901.

In general, years with greater than average snowfall also experienced a greater than average number of days with snowfall (figure 8). A decline in the number of days with snowfall is apparent from the turn of the century to the early 1930s (coincident with the warming shown in figure 11), with a possible increase thereafter. More prominent than trends are the several years with relatively high frequencies of days with snowfall, notably, 1914, 1924, 1951, 1960, 1977, 1978, and 1980.

Of 16 Illinois locations with statistically significant trends in annual snowfall (table A.1, p.72), five (four in east-central Illinois) exhibited declines, whereas 11 indicated increases in annual snowfall from 1901 to 1991 (table 2). Such differences in trend are not likely within such a relatively small area, emphasizing a problem with snowfall observations, i.e., they are extremely sensitive to the exposure of the site, as well as the method used to determine snowfall (precipitation in a gage, average of several depth measurements, or density of snow cores).

Trends in Variability

Trends in year-to-year variability in precipitation were examined by separating the precipitation data into 15-year segments, beginning with the period 1901-1915. For each 15-year segment, the interannual standard deviation of precipitation was calculated for each season (figure 9).

For the summer, a notable feature is the low variability during the period 1961-1975, as opposed to much higher variability from 1901 to 1945 and 1976 to 1990. Figure 10 shows summer precipitation for this century. The low variability during 1961-1975 is the result of an absence of severe summer droughts during the 1960s and 1970s. More frequent droughts during the period 1976-1990 have made variability similar to that during the earlier part of the century.

A period of relatively low variability also occurred during the fall in the 1940s, 1950s, and 1960s. By contrast, the period 1916-1945 was characterized by the highest variability. For the winter season, the first half of the century exhibited very low variability compared to the second half. The greatest variability was exhibited from 1946 to 1960, virtually opposite to the trends for summer. Finally, during the spring, high variability was experienced between 1916 and 1945, with low values before and after, essentially the same trends as those of fall.
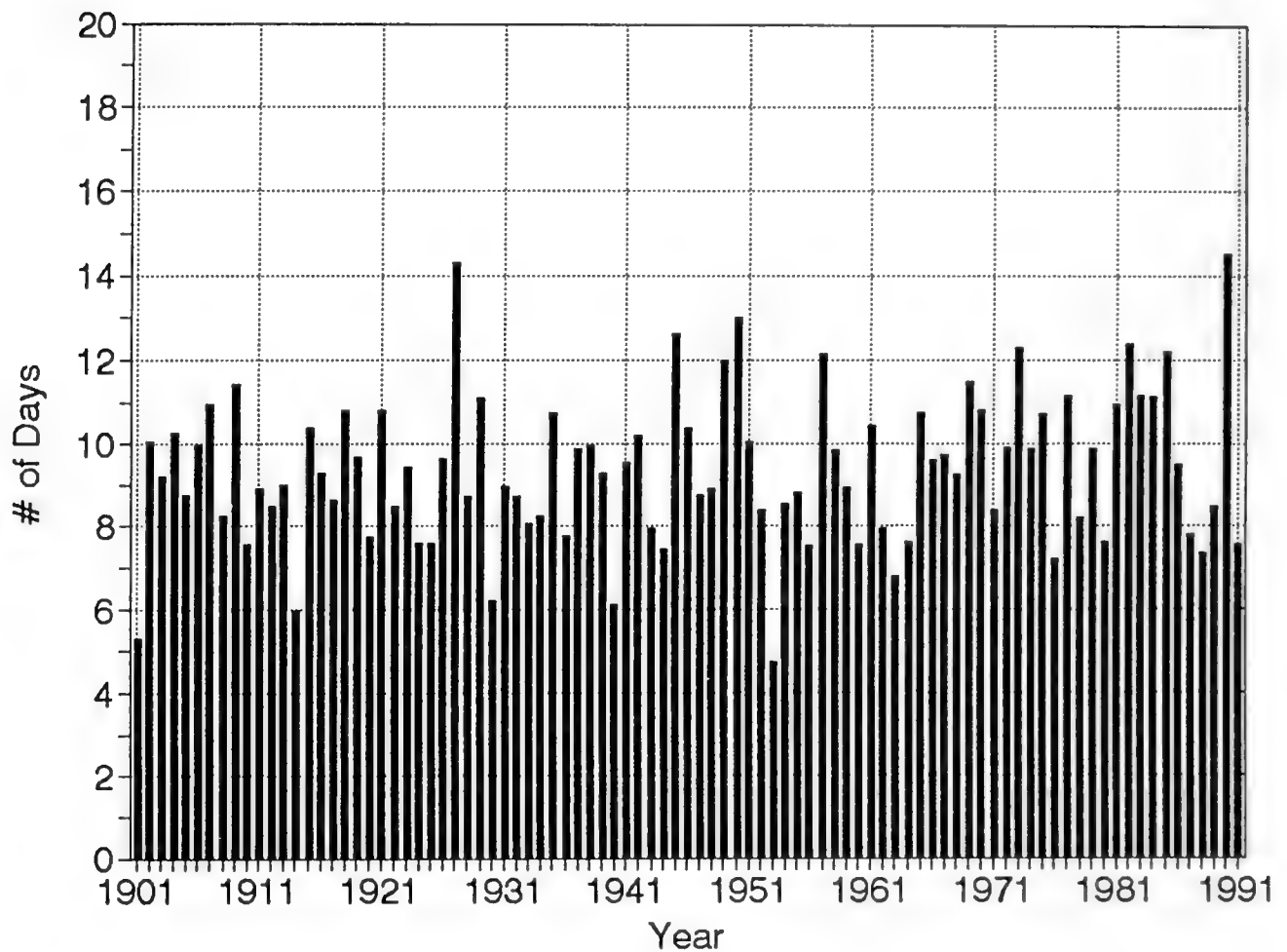
Figure 5. Average number of days per year in Illinois with at least 1 inch of precipitation.

Table 1. Summary of Precipitation Parameter Trends

Precipitation parameter	Number of stations with statistically significant upward trends	Number of stations with statistically significant downward trends
Days with precipitation	19	2
Days with precipitation >1 inch	8	1
Total precipitation	11	0
Total snowfall	11	5
Days with snowfall	10	11
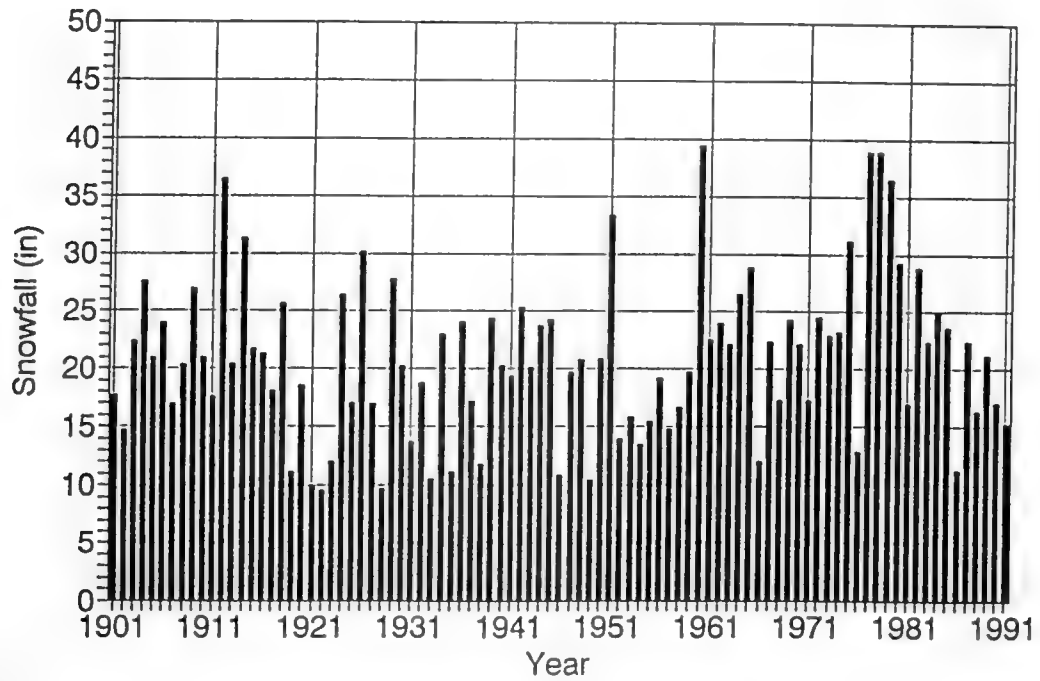
Figure 6. Average annual snowfall in Illinois (inches).
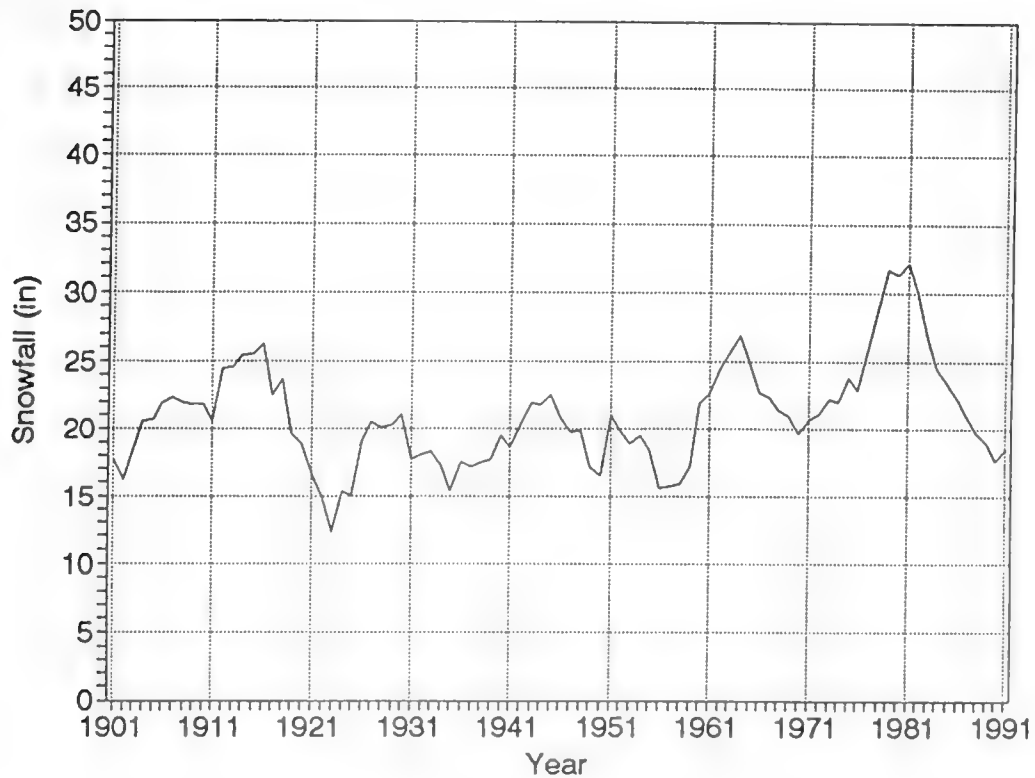
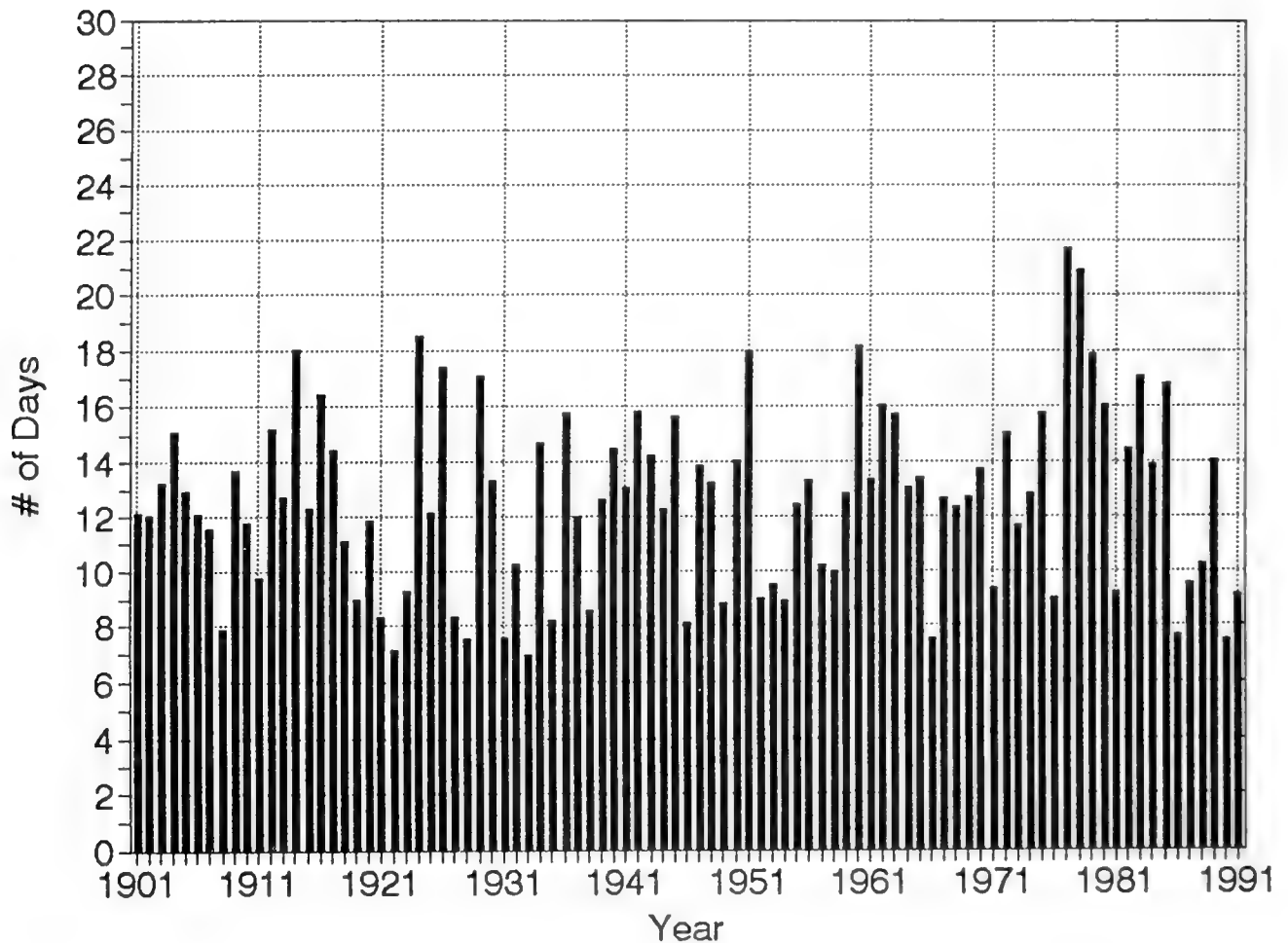*Figure 7. Running five-year averages of Illinois annual snowfall (inches).*

Figure 8. Statewide average number of days per year in Illinois with snowfall.

Table 2. Stations where Annual Snowfall Significantly Increased or Decreased (Linear Regression Analysis), 1901-1991

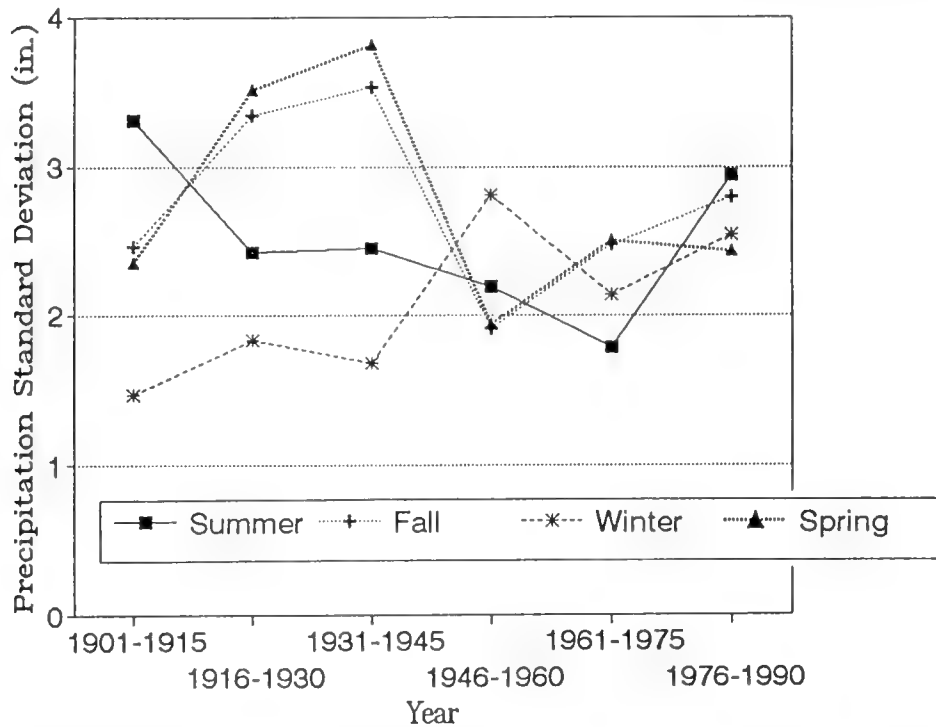| Increased		Decreased
Aledo	Palestine	Flora
Anna	Pana	Hoopeston
Dixon	Paris	Olney
Effingham	Urbana	Pontiac
Griggsville	Walnut	White Hall
Marengo		

Note: Significant at the 10 percent level.

Figure 9. Standard deviation of 15-year segments of Illinois statewide precipitation by season, 1901-1915 to 1976-1990.
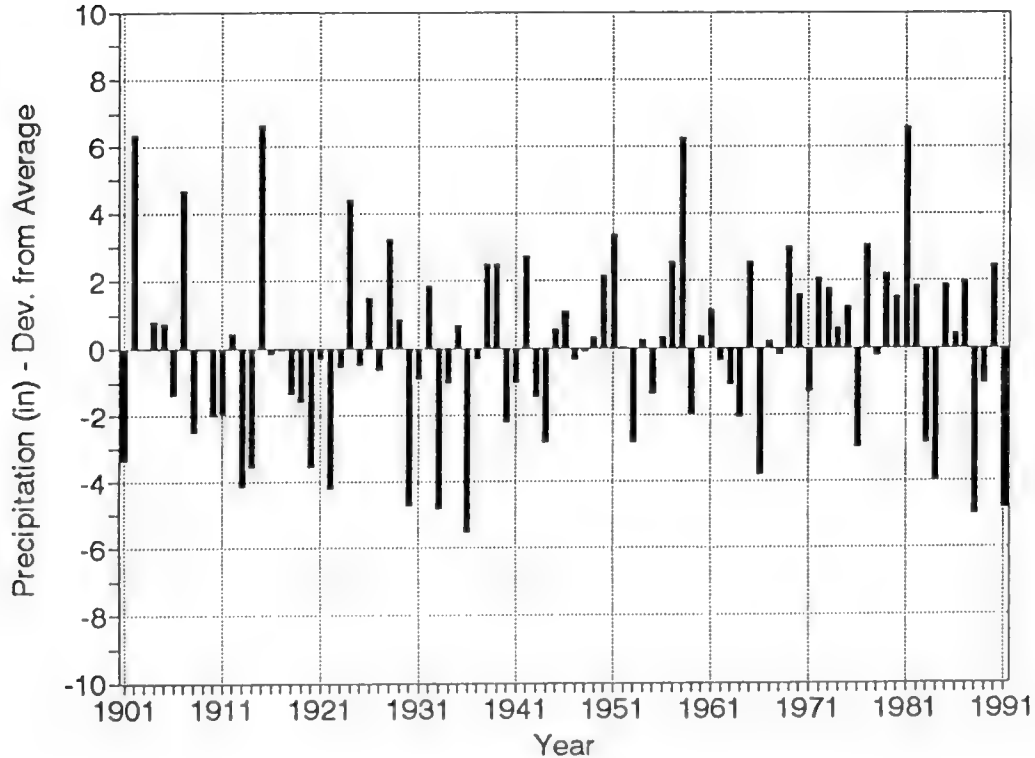

Figure 10. Statewide summer precipitation (inches) for Illinois, 1901-1991, expressed as the deviation from the period average.

TRENDS IN TEMPERATURE

Trends in Average Temperature

Changnon (1984) presented an analysis of trends in statewide average annual temperatures for Illinois for the period 1940 to 1980. His analysis is extended here to 1991 and presented in figure 11. The number of observing sites prior to 1890 is limited, as they were for statewide precipitation. Temperatures in Illinois follow a similar trend to those of the nation, i.e., rather stable temperatures during the first few decades (with a statewide mean of about 50°F). A warming trend follows until about 1930 (to about 53°F), after which cooling reduces the mean to about 52°F by 1980, with warming suggested thereafter. Cool temperatures during the first few decades reflect an expression of what has been called the "Little Ice Age" (Lamb, 1966). It is important to recognize that temperatures cooled substantially from the 1930s to 1980 or after (a similar trend for the Northern Hemisphere), five decades during which carbon dioxide was increasing!

Mean annual Illinois maximum and minimum temperatures since 1901 (figures 12 and 13, respectively) both exhibit similar trends, i.e., warming of 2 to 3°F from 1901 to the 1930s, followed by an equivalent cooling through about 1980, and the suggestion of warming again during the last few years. These trends are not surprising, since they are found in temperature records from the entire United States, the Northern Hemisphere, and the world, although the magnitudes of change decrease with ever-increasing areas of interest (e.g., Wigley and Barnett, 1990).

The average maxima and minima exhibit extremes during the same years, i.e., those much warmer or colder than the long-term average, e.g., 1917, 1921, 1924, 1931, 1978, 1979, etc., throughout the period of record. There is no evidence of periodicities.

The interseasonal fluctuations of average statewide summer maxima (figure 14) and those of average statewide winter maxima (figure 15), are quite different from each other. This is not unexpected, since temperature anomalies seldom continue longer than just a few months, and, meteorologically, one would not expect similar temperature characteristics to prevail from one season to another. Similar comments apply to statewide winter and summer minima (not shown), i.e., the interseasonal fluctuations need not parallel each other, and they do not.

The mean number of days (averaged over all stations) with temperatures above 100°F (figure 16) are indeed

few, particularly during the most recent decades, which are exhibiting cooling. Several years, namely 1901, 1913, 1930, 1934, and particularly 1936, exhibited high frequencies of such days, all during the first four decades of the record. Years with a high frequency of days with temperature greater than 100°F tended to occur in episodes composed of a few sequential years, e.g., 1913-1914, 1930-1936, 1939-1941, and 1952-1954, all of which represented major droughts!

The statewide average number of days per year with maximum temperatures above 86°F (figure 17) exhibits trends similar to those shown in figure 16, although the absolute frequencies are greater. The 1930s experienced more days with higher temperatures than any other decade on record.

Figures 18 and 19, showing the number of days per year with maximum temperatures below 32° and below 0°F, respectively, exhibit inverse trends to those found in figures 16 and 17. More frequent cooler maxima have occurred since about 1960 than before. This trend suggests that the cooler 1960s, 1970s, and early 1980s comprised more frequent cool days, as opposed to cooler extremes only. Note the relatively high frequency of cold maxima in 1912, 1924, and 1936 in both figures 18 and 19. The relatively high frequency of maxima below 32°F in the late 1960s, 1970s, and early 1980s is also seen in the frequency of maxima below 0°F (figure 19).

Figure 20 presents the statewide average number of days per year when the minimum temperature exceeded 70°F. Not surprisingly, the trend follows that of average temperature and maximum and minimum temperatures, with increasing frequency from 1901 until the 1930s, a decline thereafter, with a possible increase again during the last few years. It should be noted that the high frequencies of 1913, 1931, 1934, and 1936 have not been equaled in Illinois since.

Figure 21 shows the statewide average number of days per year when the minimum temperature was below 32°F. A declining trend may be suggested from 1901 to perhaps the mid-1940s, with either stable frequencies or perhaps a slight increase thereafter.

Figure 22 presents the statewide average number of days per year when the minimum temperature was below 0°F. Such frequencies were decidedly minimal during the 1940s and 1950s, which were warm decades. The highest frequency of such days occurred in 1936, 1963, 1977-1979, and 1985.
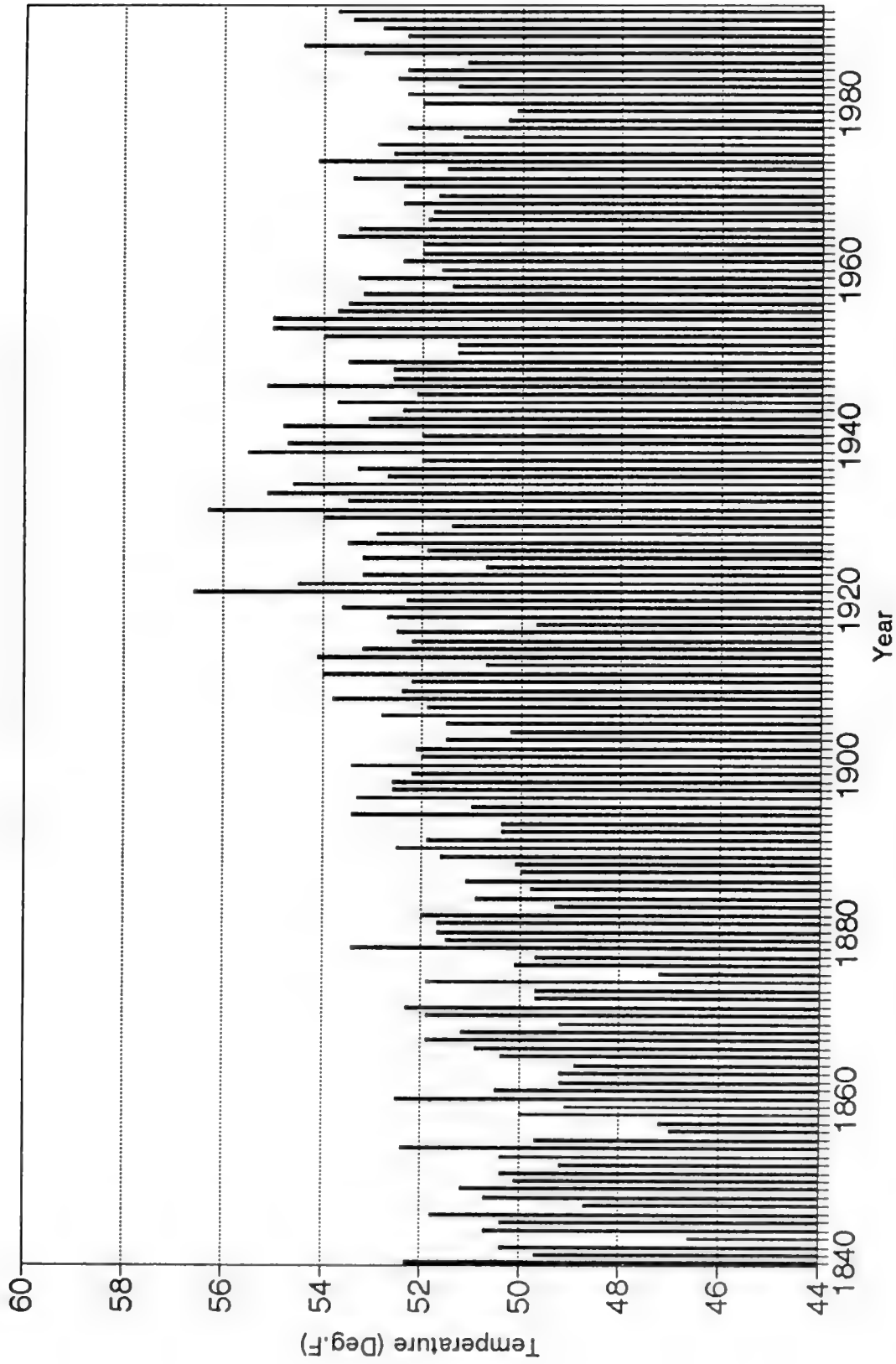
Figure 11. Statewide average annual temperatures for Illinois (°F) composed of available sites, 1840-1992.
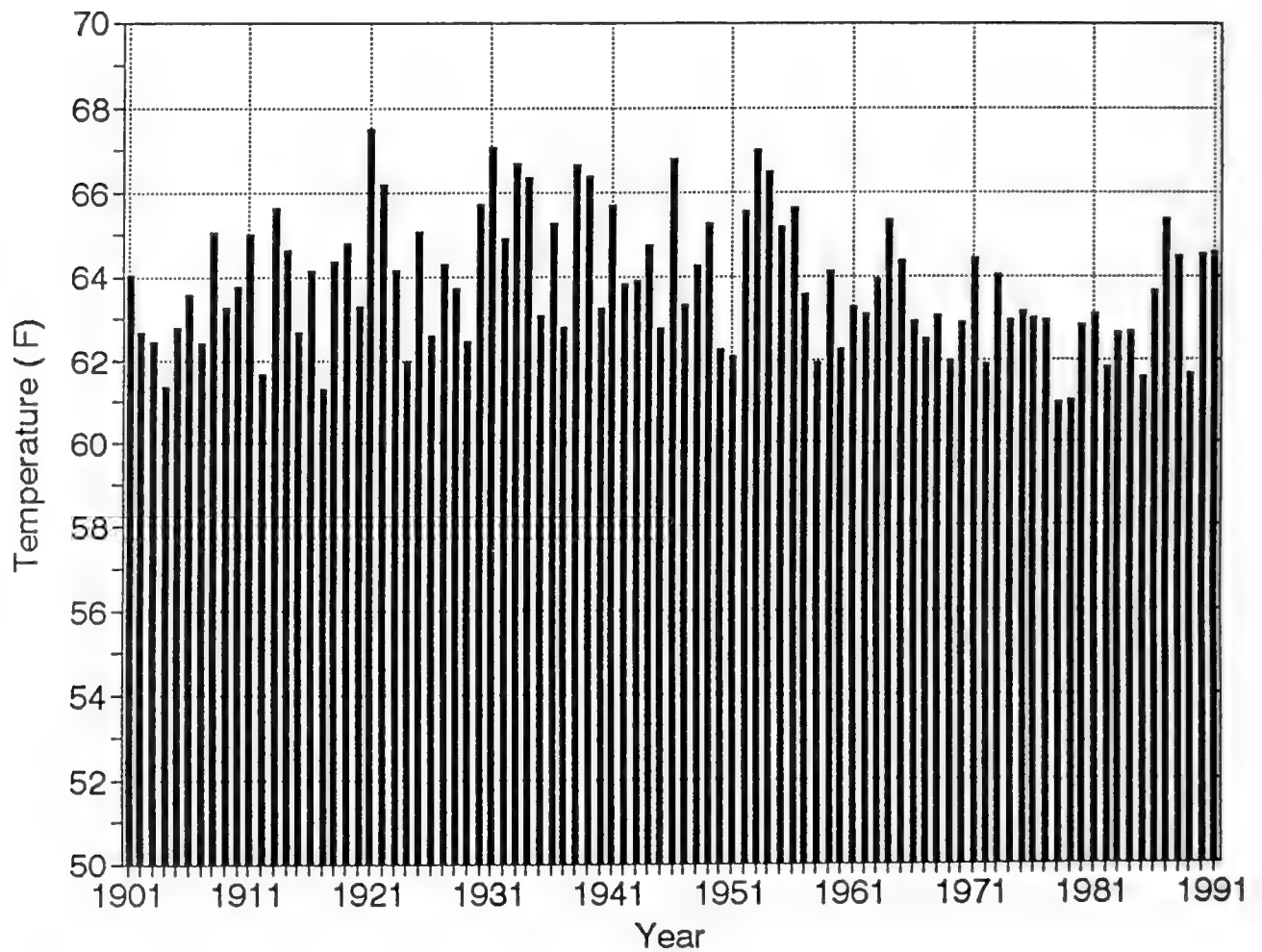
Figure 12. Statewide average annual maximum temperatures for Illinois (°F), 1901-1991.
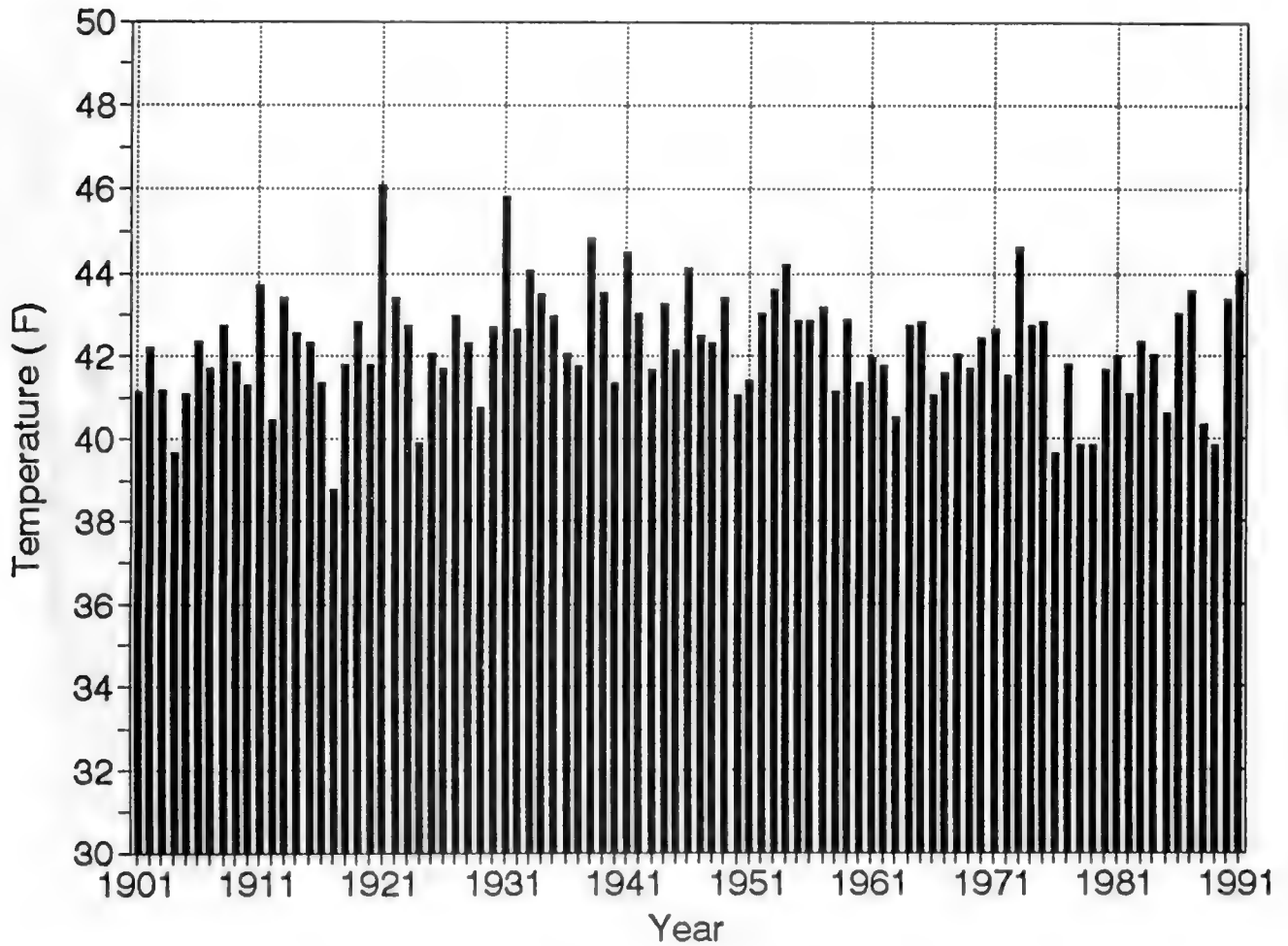
Figure 13. Statewide average annual minimum temperatures for Illinois (°F), 1901-1991.
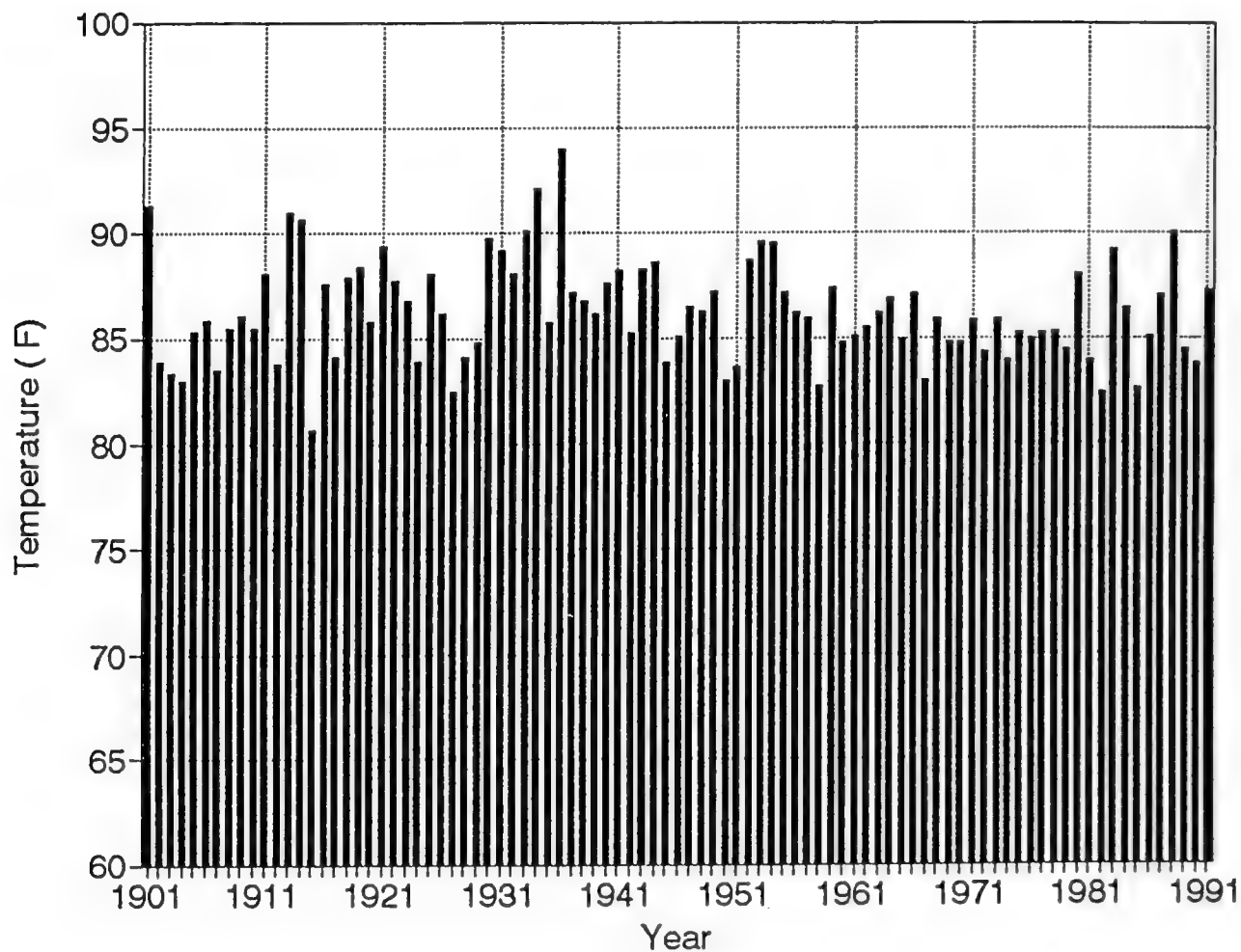
Figure 14. Statewide average maximum summer temperatures for Illinois (°F), 1901-1991.
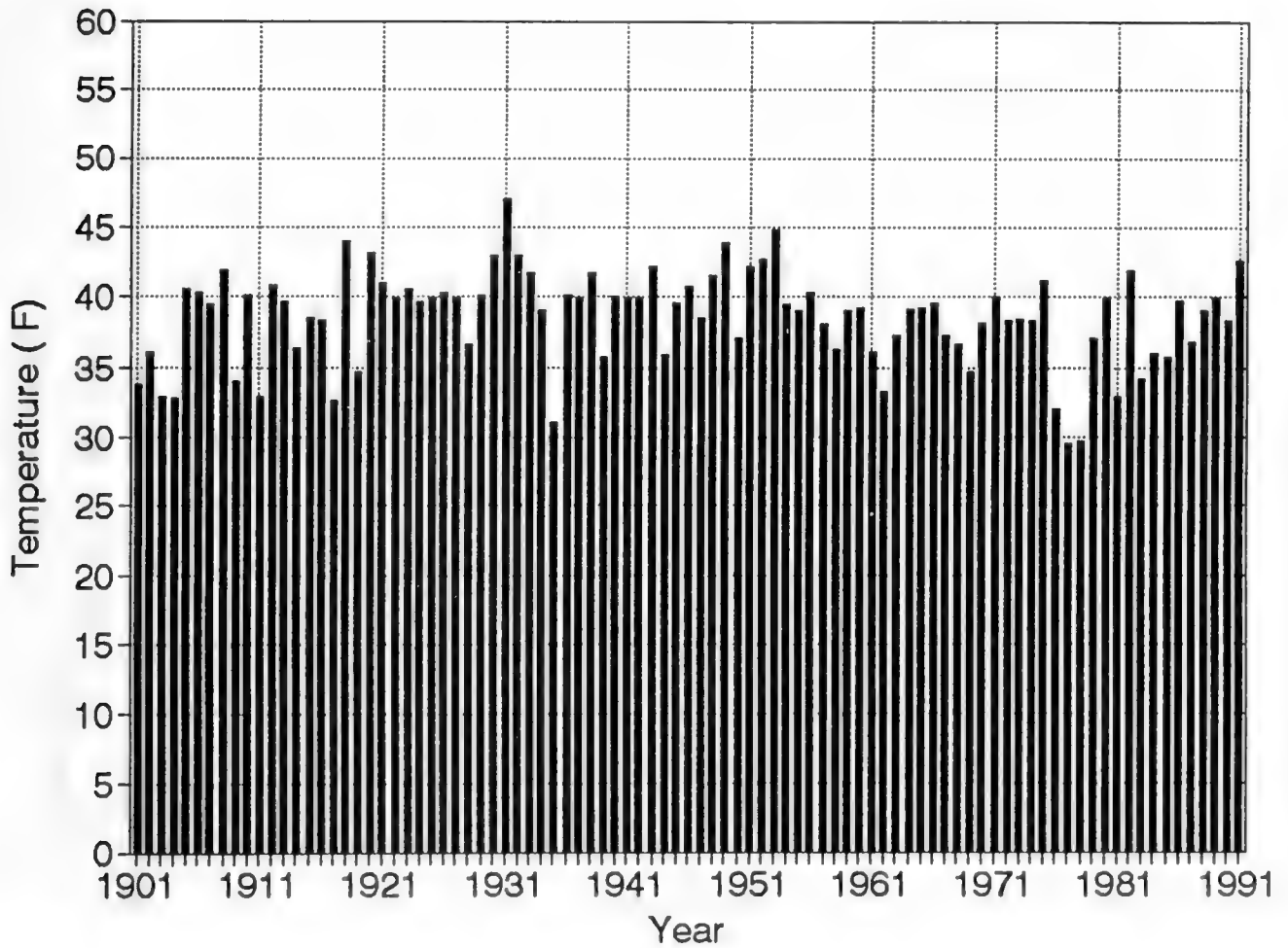
Figure 15. Statewide average maximum winter temperatures for Illinois (°F), 1901-1991.

Figure 16. Statewide average number of days per year in Illinois with maximum temperatures greater than 100°F, 1901-1991.

*Figure 17. Statewide average number of days per year in Illinois
with maximum temperature greater than 86°F, 1901-1991.*

*Figure 18. Statewide average number of days per year in Illinois
with maximum temperature less than 32°F, 1901-1991.*

*Figure 19. Statewide average number of days per year in Illinois
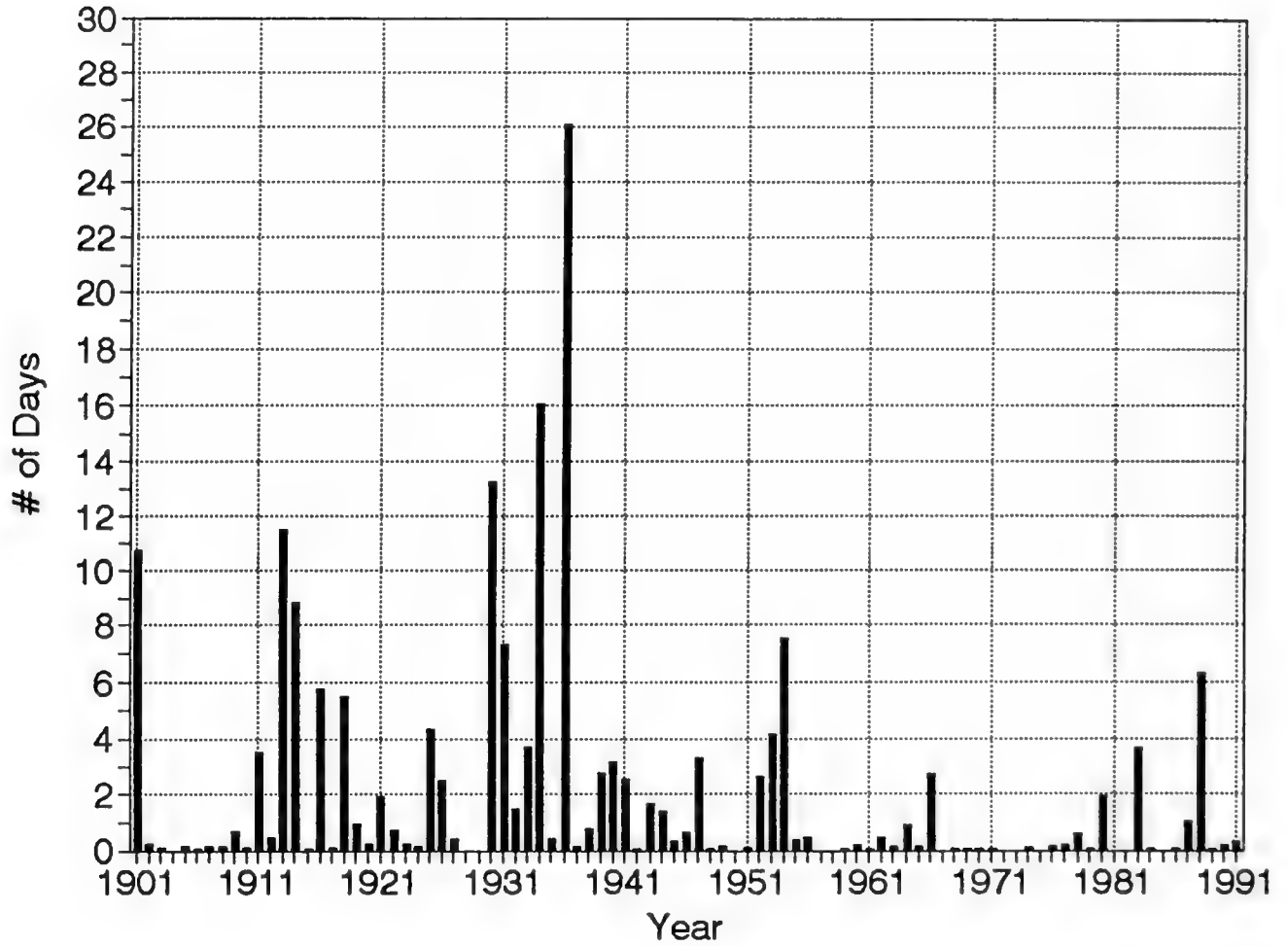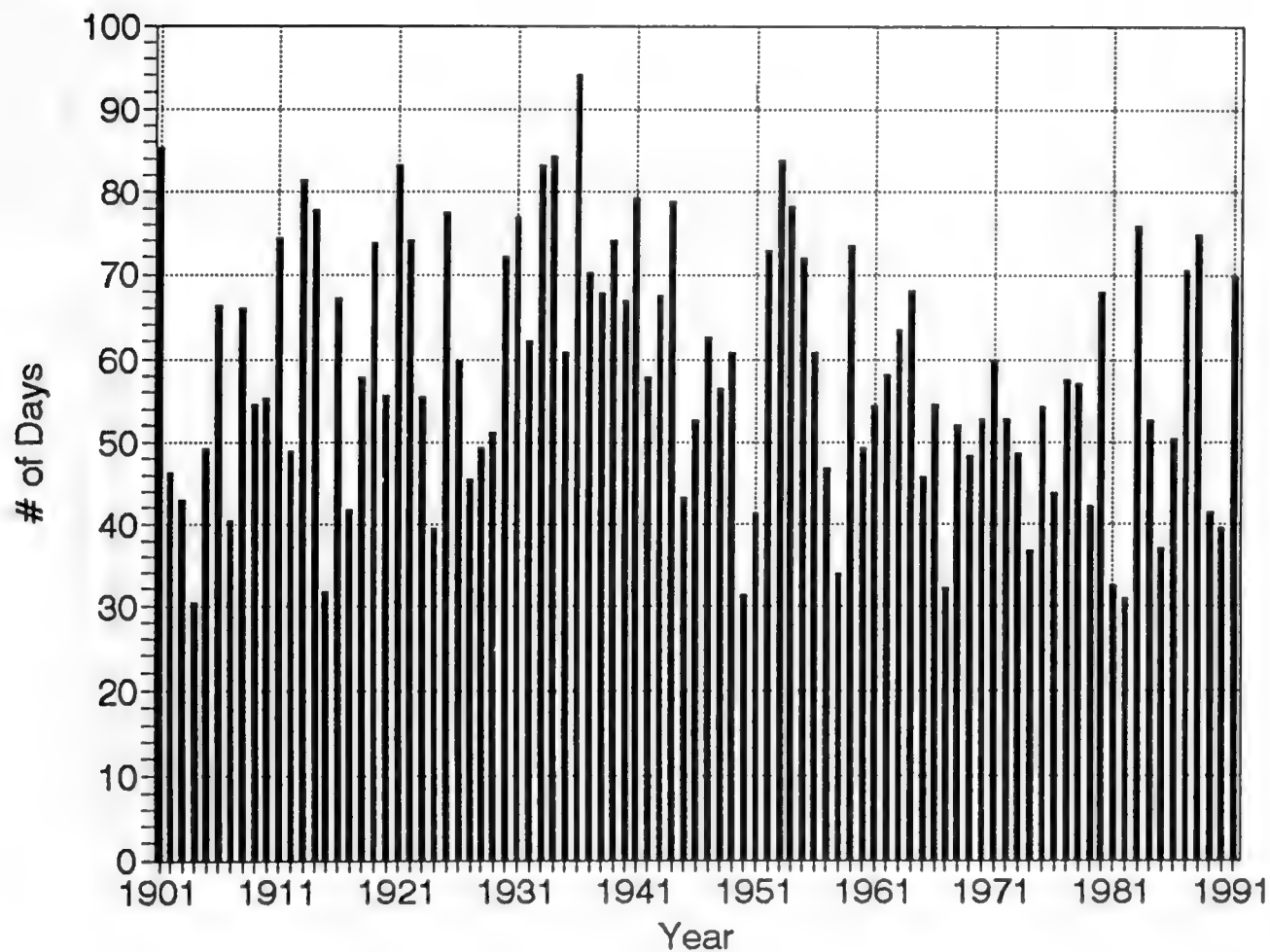with maximum temperature less than 0°F, 1901-1991.*

*Figure 20. Statewide average number of days per year in Illinois
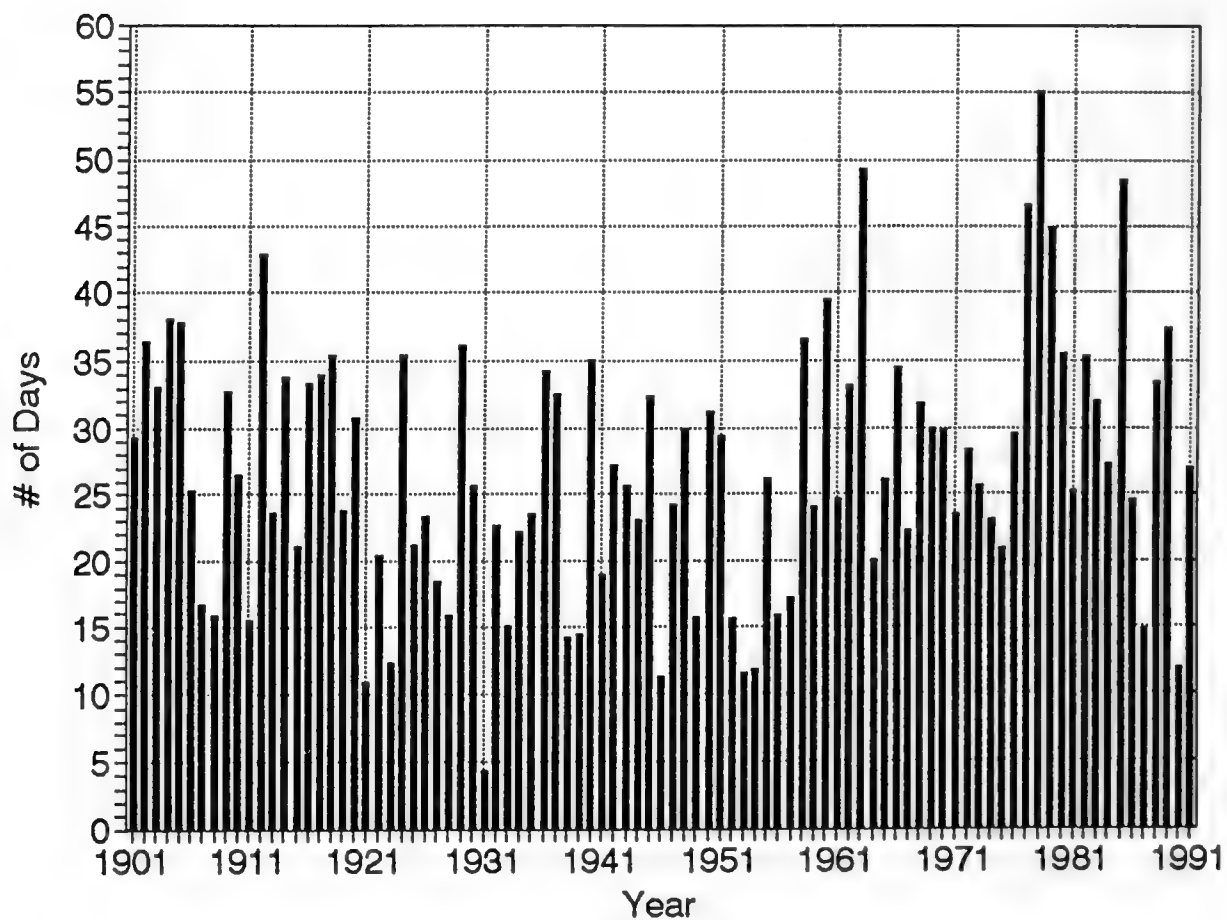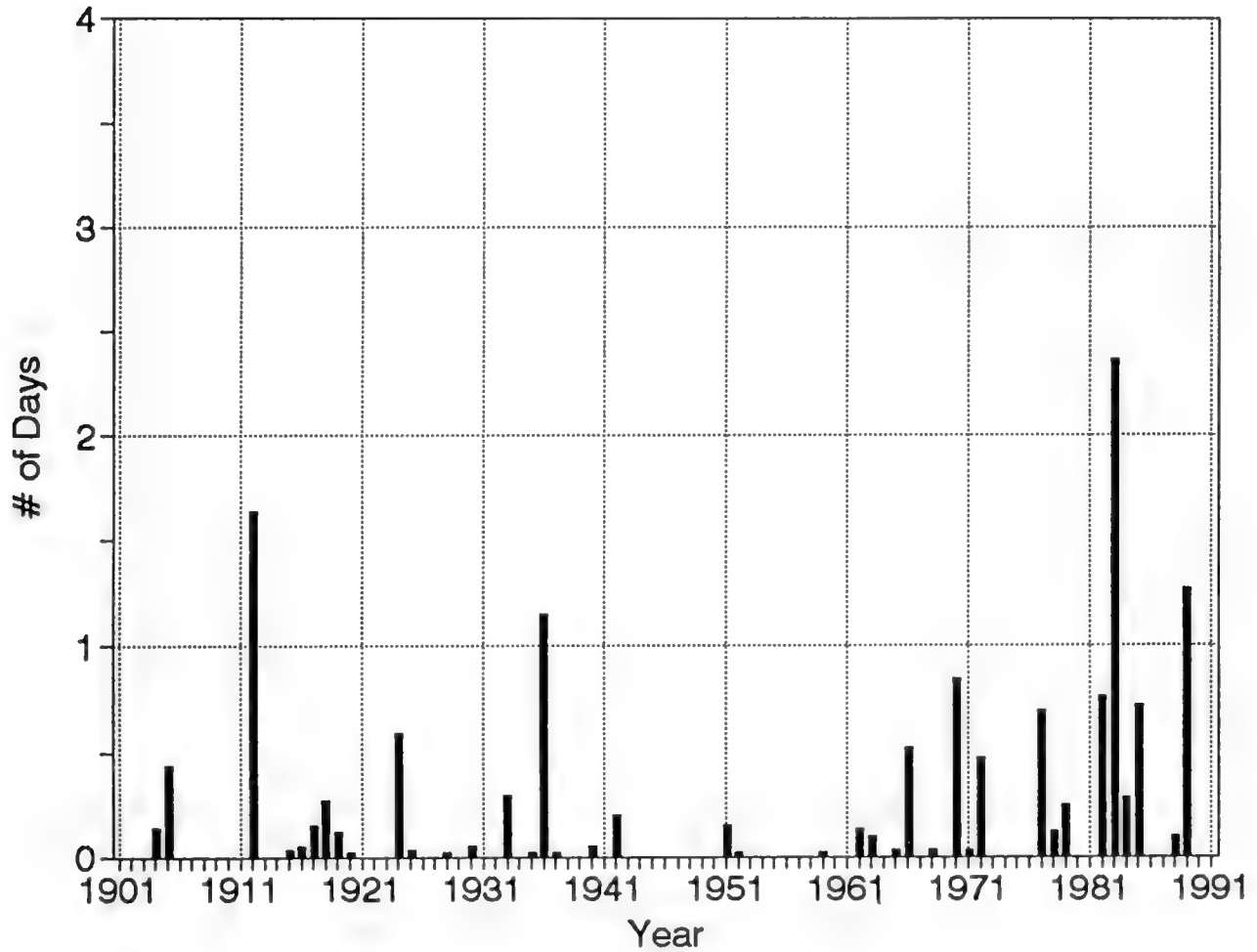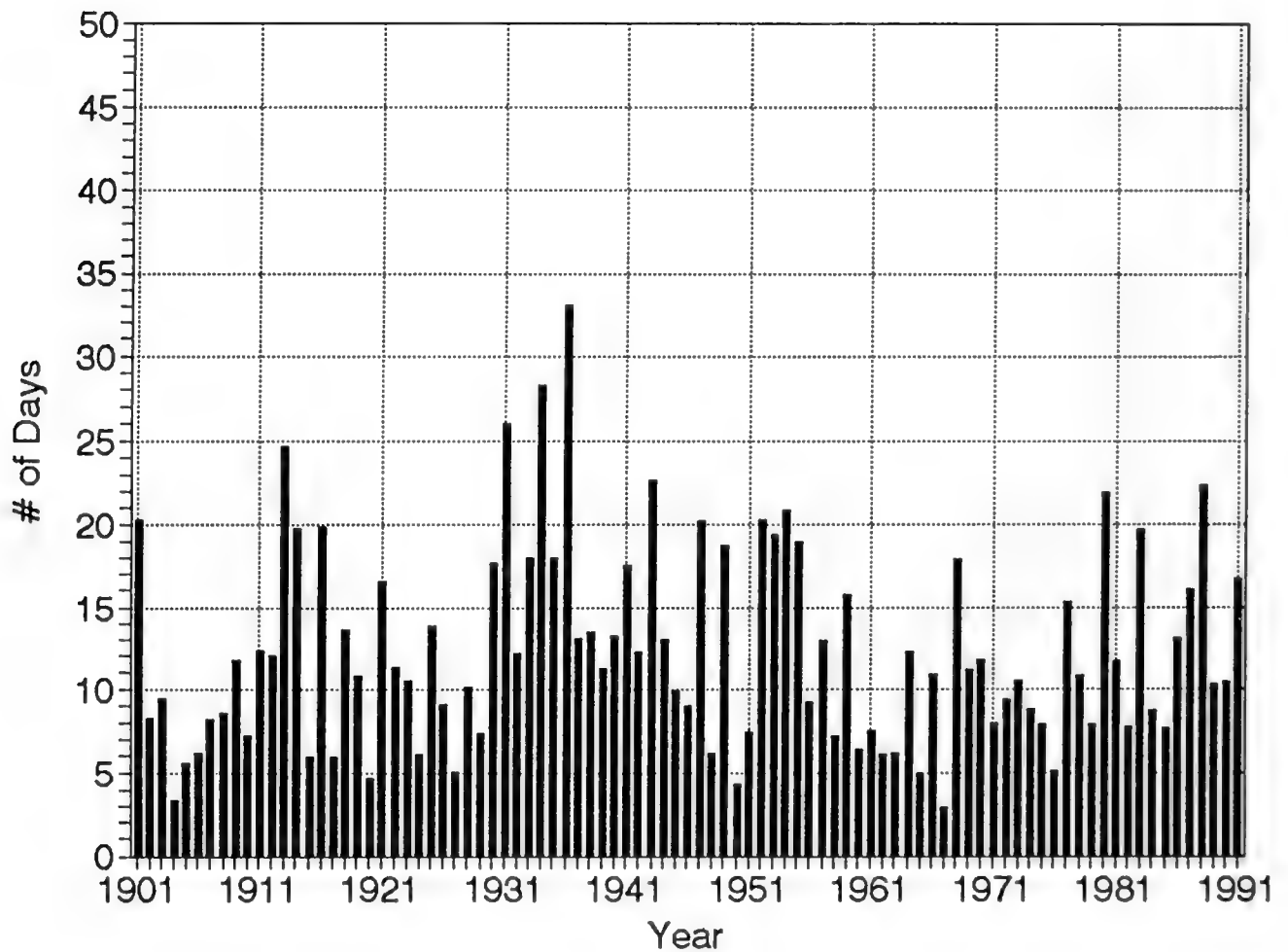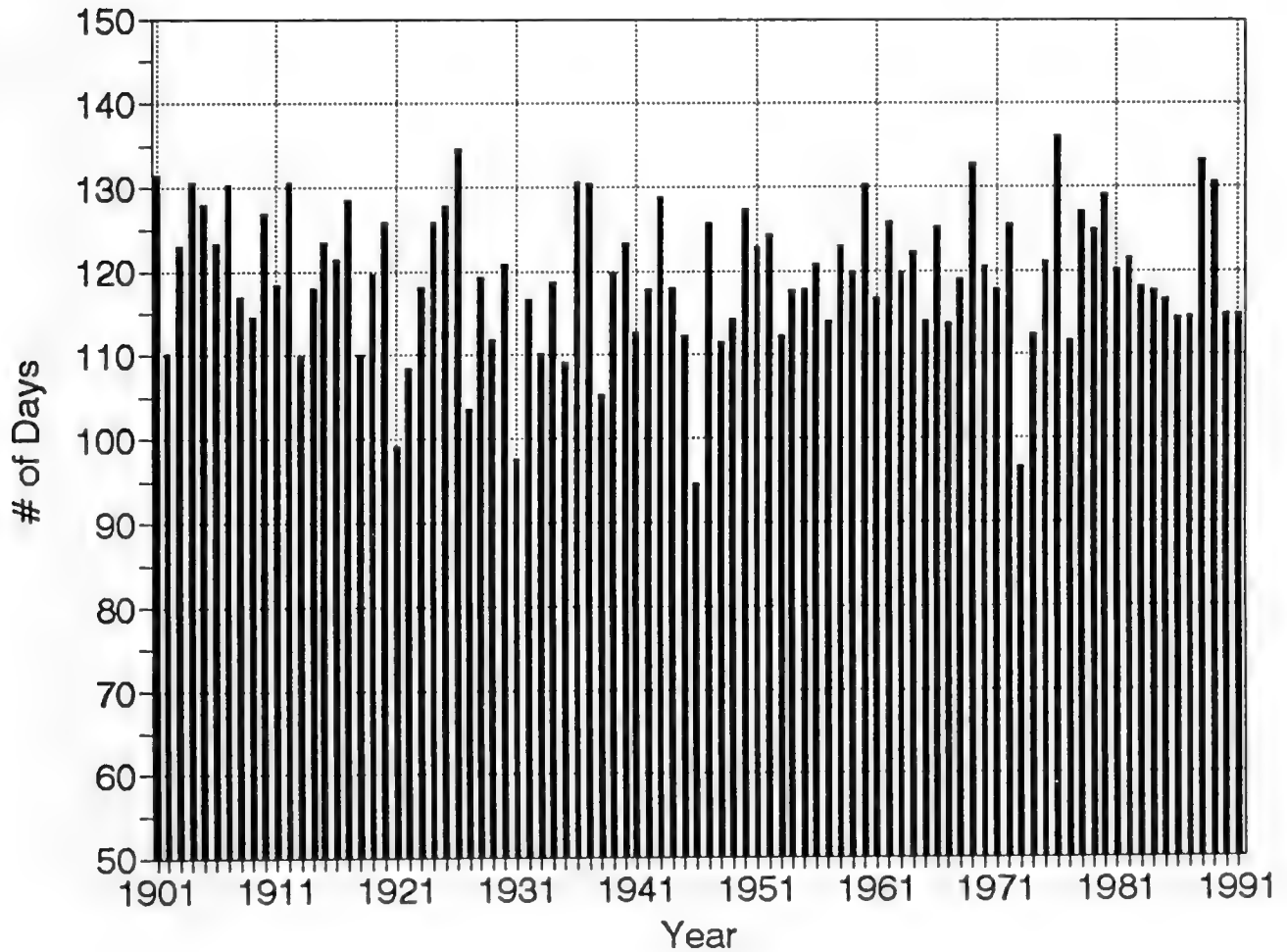with minimum temperature greater than 70°F, 1901-1991.*

*Figure 21. Statewide average number of days per year
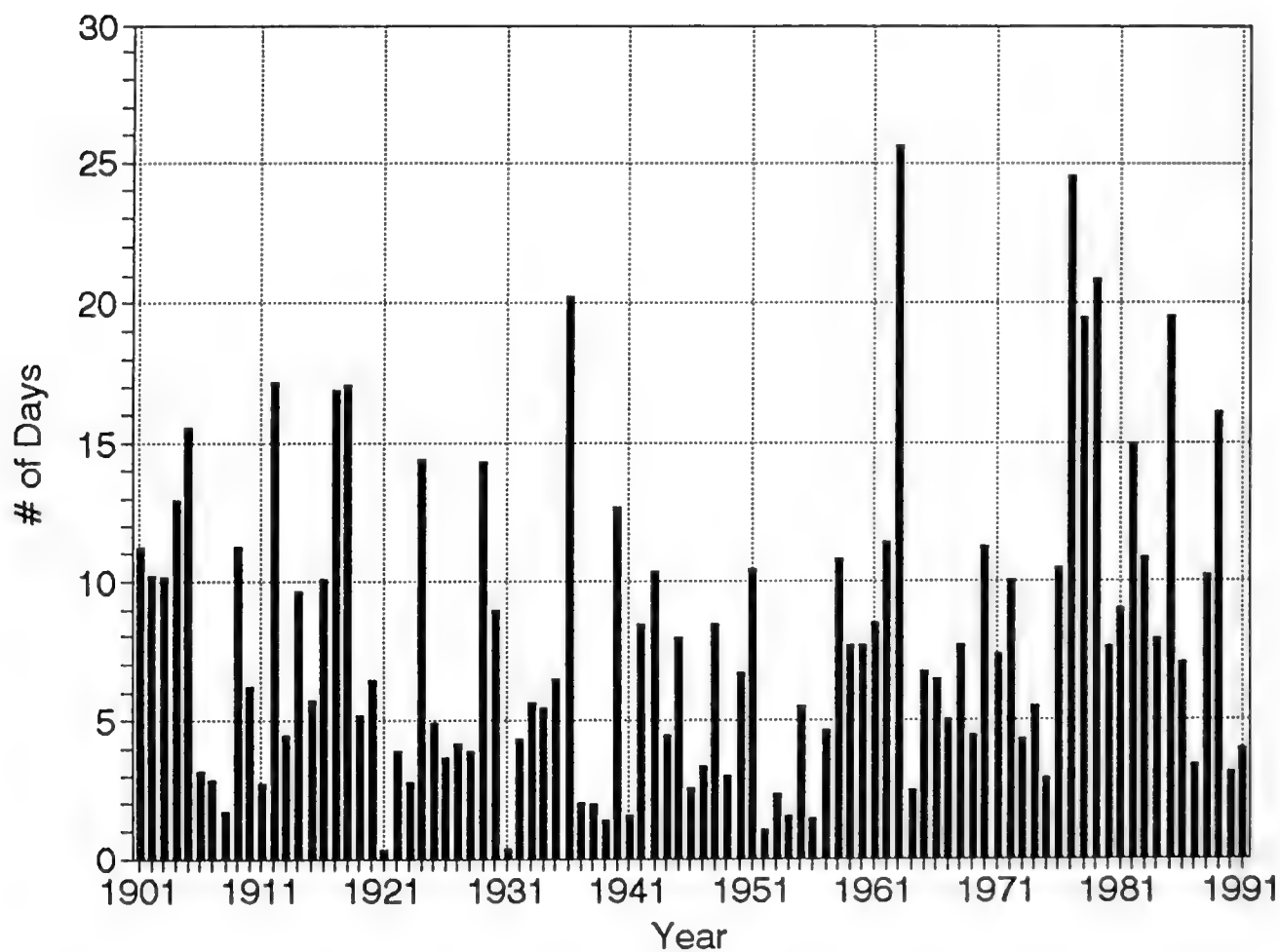with minimum temperature less than 32°F, 1901-1991.*

Figure 22. Statewide average number of days per year with minimum temperature less than 0°F, 1901-1991.

Change in Parameters Related to Temperature

Mean Statewide Annual Heating Degree-Days. Heating degree-days (HDDs) are derived from the accumulated positive differences of 65°F minus the mean daily temperature. This rather simple calculation results in a value that closely parallels heating fuel needs.

Average annual HDDs, shown in figure 23, demonstrate a crude inverse correlation to annual temperature. As one would expect, the fewest HDDs are found in the 1930s and 1940s. HDD totals during the warmest decades are about 25 percent fewer than during the colder years. Five-year running averages of this parameter (presented in deviations from the period average in figure 24) rather clearly demonstrate fewer HDDs in Illinois from the 1920s through the 1950s, the warmest decades of the century-long Illinois temperature record.

Mean Statewide Annual Cooling Degree-Days. Cooling degree-days (CDDs) represent a measure of electricity required for cooling buildings during the warm season. They represent accumulated positive differences of daily mean temperature minus 65°F.

Annual CDDs (figure 25) also show the 1930s and adjacent decades as warmer than those earlier or later, as did the HDD record. The total CDD five-year running averages (figure 26) show more CDDs during the 1930s and early 1940s, but they also show peaks during the first few years of record, in the early teens and twenties, in the 1930s and early 1940s particularly, and the mid-1950s.

The five-year running averages of heating and cooling degree days (figures 24 and 26, respectively) demonstrate a rather clear inverse relationship to each other, except for the late 1970s and 1980s.

Mean Statewide Corn Growing Degree-Days. Corn growing degree-days (GDDs) are derived from the accumulated positive differences of daily mean temperature minus 50°F (when maxima exceed 86°F, the maximum is set to 86°F; when minima are less than 50°F, the minimum is set to 50°F). Trends of temperature, mentioned earlier, are also apparent in this measure (figure 27) of growth. Annual statewide corn GDDs since 1878 have been lower than those of 1992 in only six other years, 1882, 1883, 1915, 1917, 1924, and 1967, and then only marginally! The relatively few GDDs in 1992 virtually devastated the northernmost 100 miles of the Corn Belt. As a result, corn in that area was of lower quality and did not fully mature

everywhere within that region. Interestingly, 1992 witnessed a record total corn crop in the United States by a substantial margin, largely because of reduced heat and moisture stress in the heart of the Corn Belt.

Trends in the Growing Season. Trends in the length of the growing season (figure 28), the date of the last spring freeze (figure 29), and the first fall freeze (figure 30) correspond only weakly to the *trends* indicated in average annual temperature. This is due to the fact that averages reflect the overall anomaly of the period included in the average.

Individual *events*, e.g., the date of first or last frost, are often poorly correlated with monthly or seasonal temperature averages, which are composed of many days surrounding the frost event, because the frost event is short-lived (only hours in duration). Moreover, the event is also dependent upon other specific local conditions occurring during the time of the frost, e.g., low cloud cover, wind speeds, humidity, etc.

The dominant characteristic of the growing season time series is the year-to-year variability. Changnon (1984) had noted a slight increase in the length of the growing season caused primarily by a trend towards earlier spring freezes. The results of that study, based on data from nine stations, are also seen in this analysis of 41 stations.

Table 3 (p.40) summarizes a statistical analysis of the trends from these 41 stations for two freeze thresholds (32°F and 28°F). The most obvious feature of these results is that all stations with significant trends in the date of the last spring freeze exhibit a tendency for earlier spring freezes. There is also a tendency towards later first fall freezes. The combination of these two trends leads to a tendency toward longer growing seasons.

Linear Trends in Temperature Records at Illinois Sites

Linear trends in the temperature parameters are listed for all 41 long-term stations in appendix A. Average temperature parameters, minimum temperature parameters, and maximum temperature parameters are listed in tables A.2-A.4, respectively, beginning on p.74). These trends are summarized in table 4. For most temperature parameters, statistically significant trends (at the 10 percent level) are found for less than half of the stations. However, in many cases, the direction of the trend is similar for many stations. For instance, 27 stations exhibited significant downward trends for the highest temperature of the year. This
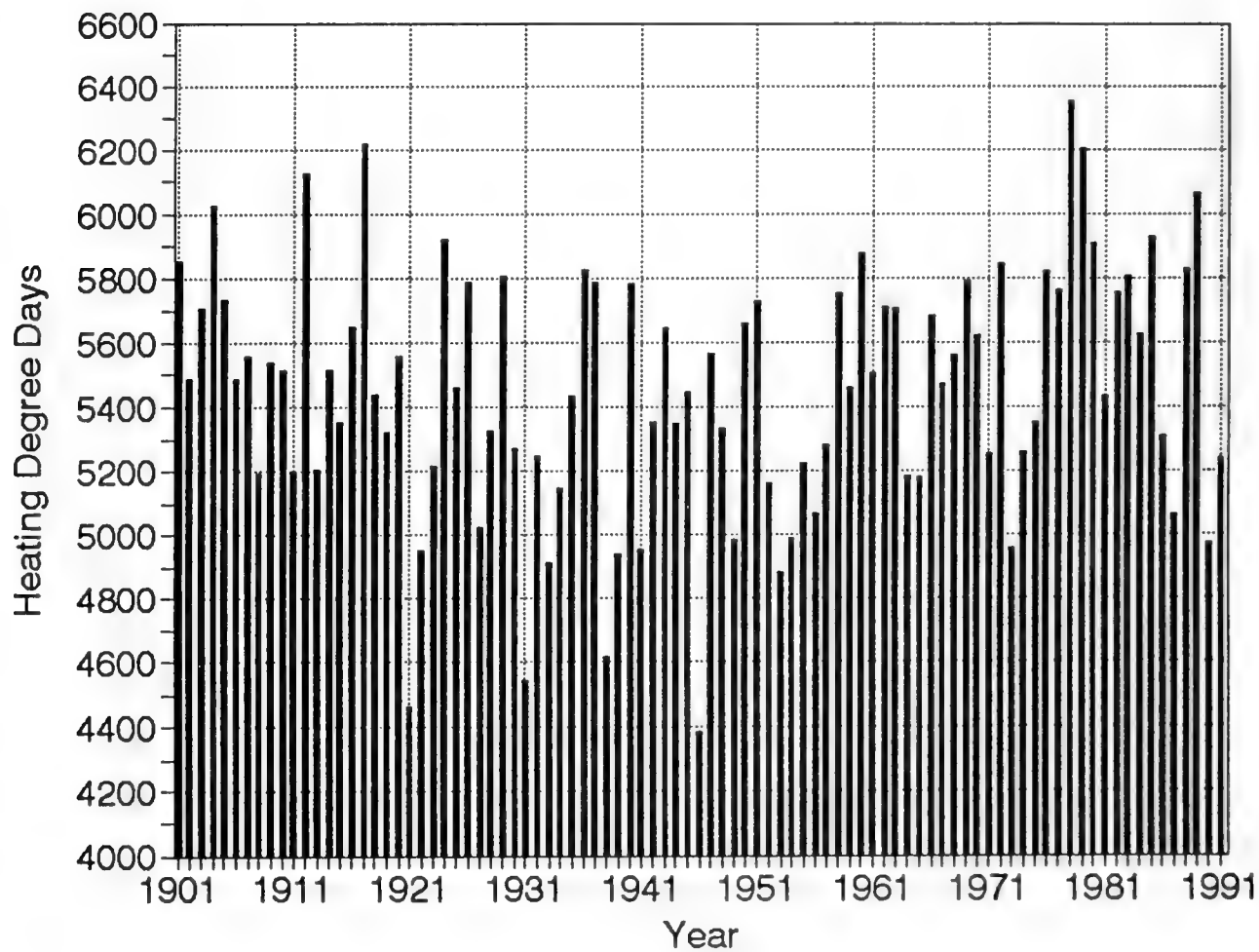
Figure 23. Statewide average annual heating degree-days for Illinois, 1901-1991.
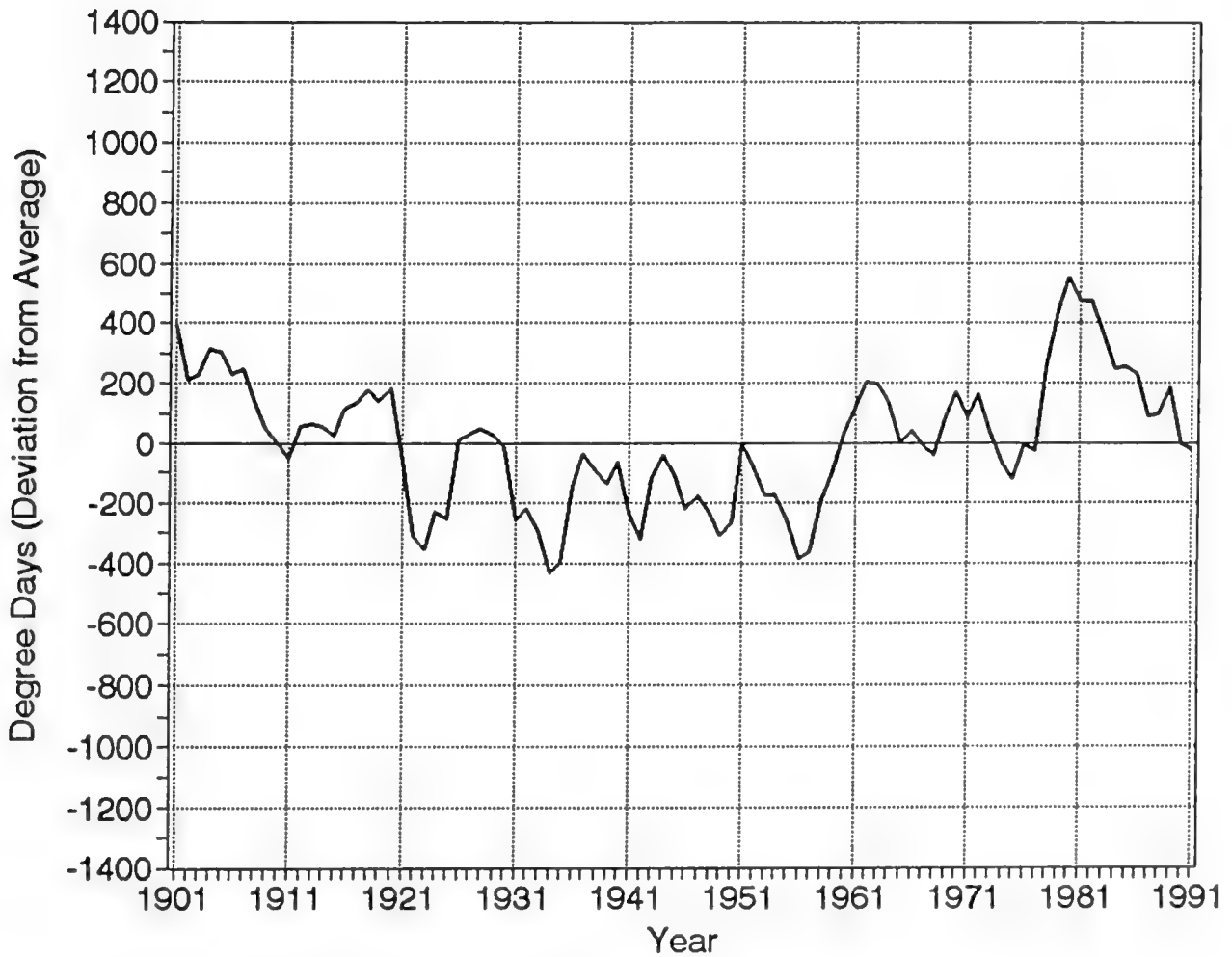
Figure 24. Five-year running averages of statewide heating degree-days,
presented as deviations from period mean, 1901-1991.
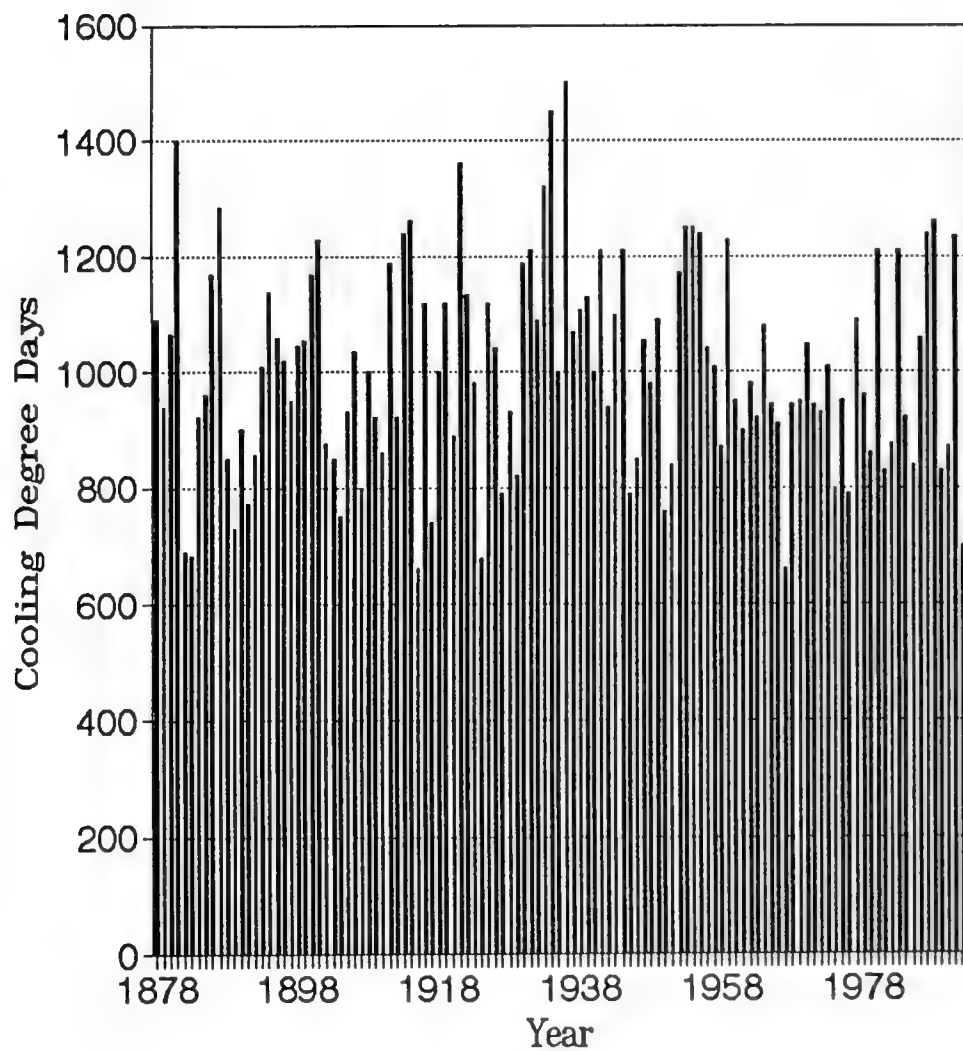
Figure 25. Statewide average annual cooling degree-days for Illinois, 1878-1992.
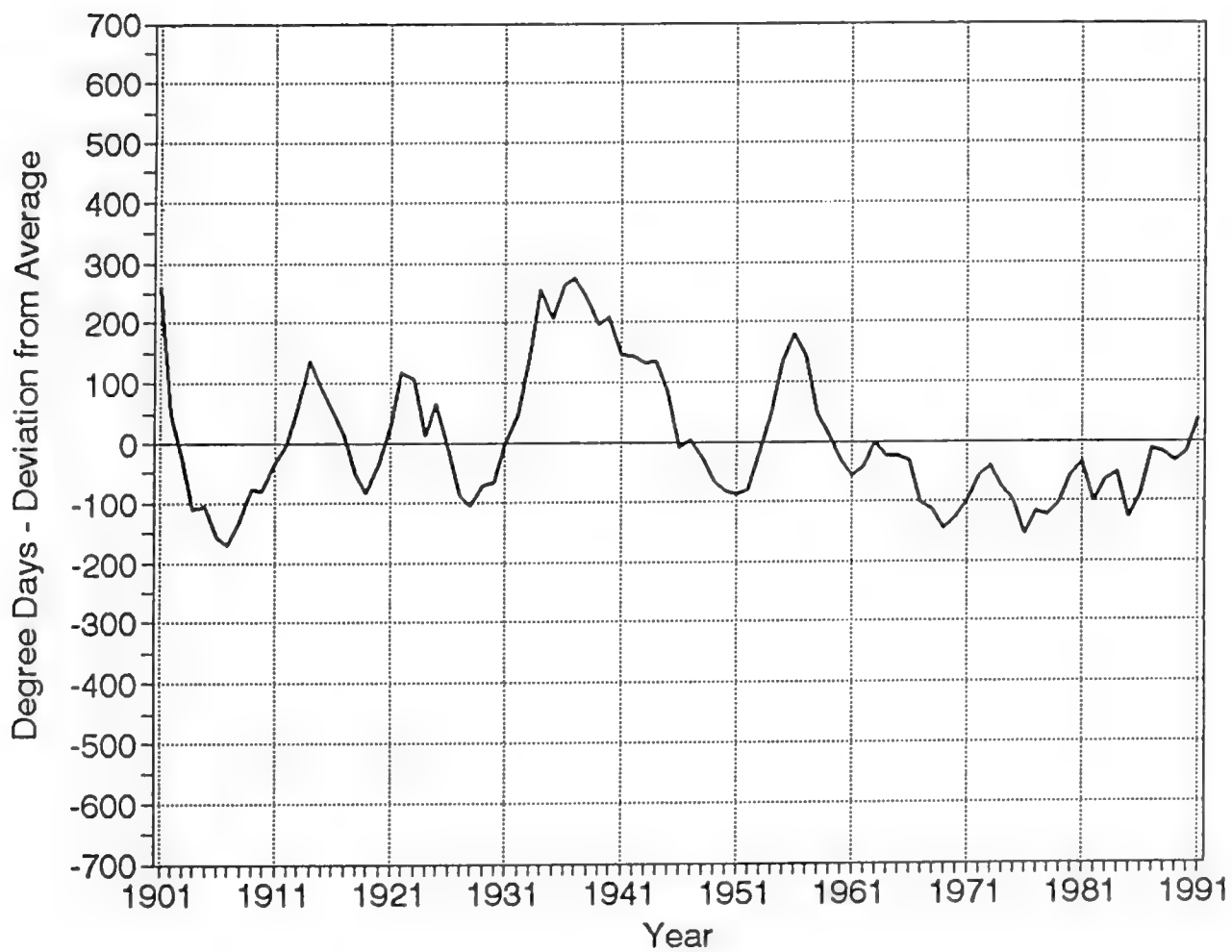
Figure 26. Five-year running averages of statewide mean cooling degree-days for Illinois, presented as deviations from period mean, 1901-1991.
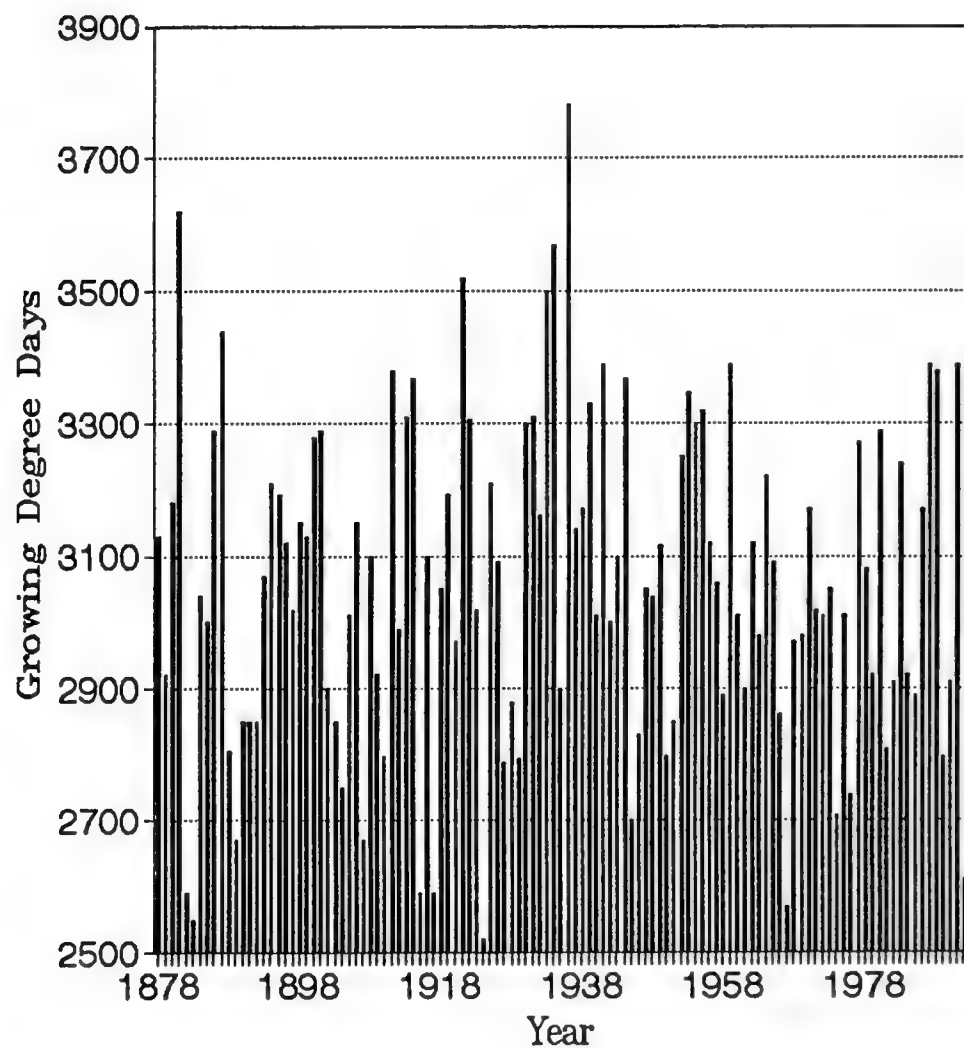
Figure 27. Average Illinois annual corn growing degree-days, 1878-1992.
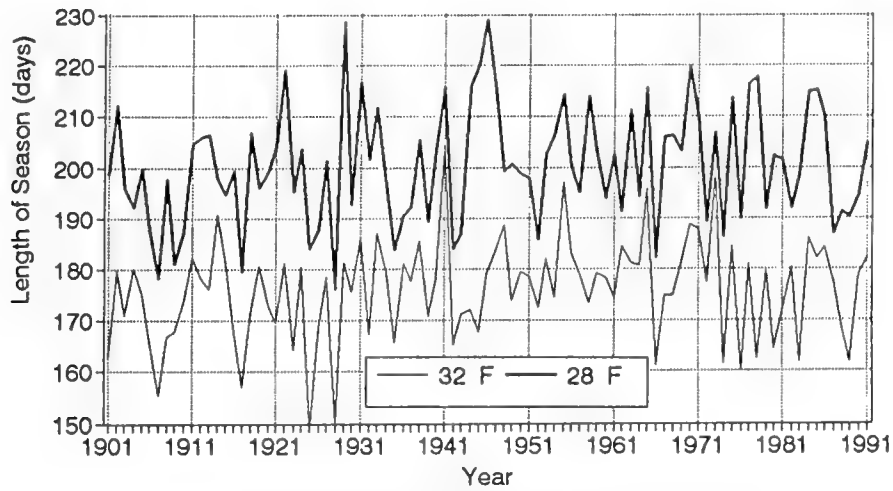
Figure 28. Statewide average length (days) of the growing season for Illinois (defined for both 28°F and 32°F), 1901-1991.
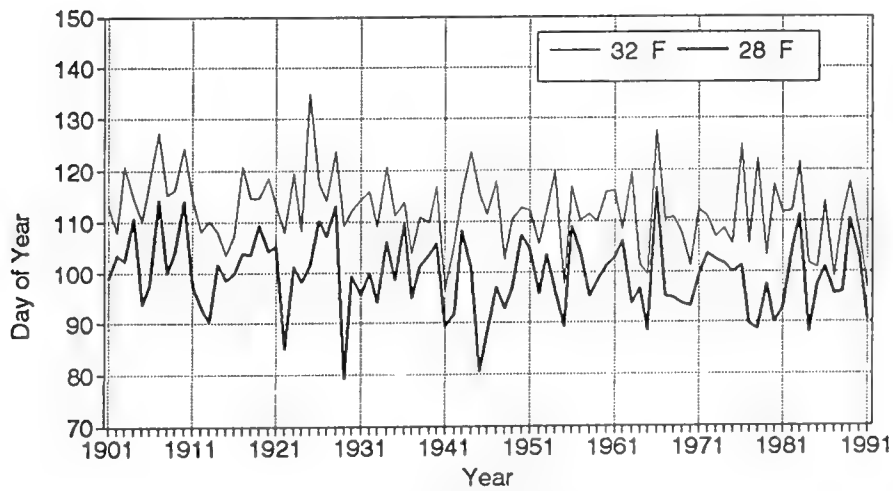
Figure 29. Statewide average day (Julian day number) of last spring frost in Illinois (defined for both 28°F and 32°F), 1901-1991.
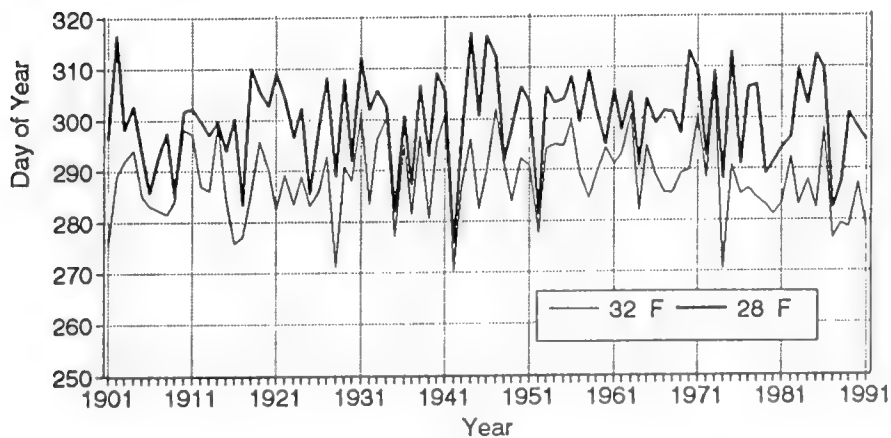
Figure 30. Statewide average day (Julian day number) of first autumn frost in Illinois (28°F and 32°F), 1901-1991.

Table 3. Summary of Growing Season Trends

Freeze thresholds	Number of stations with trends toward later (longer) freezes (growing season) (statistically significant trends given in parentheses)	Number of stations with trends toward earlier (shorter) freezes (growing season) (statistically significant trends given in parentheses)
Last spring freeze (threshold = 32°F)	5 (0)	36 (24)
Last spring freeze (threshold = 28°F)	6 (0)	35 (11)
First fall freeze (threshold = 32°F)	24 (10)	17 (5)
First fall freeze (threshold = 28°F)	24 (13)	17 (4)
Growing Season Length (threshold = 32°F)	30 (17)	11 (3)
Growing Season Length (threshold = 28°F)	26 (16)	15 (4)

Table 4. Summary of Trends in Temperature Parameters of 40 Illinois Sites

| Temperature parameter	Number of stations with upward trends (statistically significant number in parentheses)	Number of stations with downward trends (statistically significant number in parentheses)
Average monthly temperature	16 (5)	25 (9)
Average monthly minimum	18 (9)	23 (10)
Average monthly maximum	13 (3)	26 (12)
Highest temperature of year	2 (0)	39 (27)
Lowest temperature of year	23 (3)	18 (0)
Days with mean T > 50°F	22 (12)	19 (1)
Days with T-max > 86°F	7 (0)	34 (18)
Days with T-max > 90°F	2 (0)	39 (29)
Days with T-max > 100°F	0 (0)	41 (21)
Days with T-max < 0°F	40 (18)	1 (0)
Days with T-max < 32°F	36 (19)	5 (1)
Days with T-min > 70°F	18 (7)	23 (4)
Days with T-min < 0°F	38 (7)	3 (1)
Days with T-min < 32°F	21 (6)	20 (9)

reflects the high summer temperatures experienced during the 1930s and the relative absence of extreme high temperatures during the last three to four decades. This same feature is also evident in half or more of the sites in the number of days with maximum temperatures above 86°F, 90°F, and 100°F. This feature is in general agreement with the general downward trend in average temperature from the 1930s into the 1970s (figure 11). Although the average temperature record and the number of "hot" days in the last decade suggest a reversal of this trend, this change has not been of long enough duration or magnitude to overcome the large downward trend in the earlier period.

Somewhat surprisingly, the number of days of extreme cold, identified as the number of days with maximum temperatures below 0°F (figure 19), has exhibited an upward trend at many stations. This was caused by a significant number of extremely cold arctic air outbreaks during the last 15 years. These outbreaks have even occurred with some frequency during the 1980s, a period of generally very warm winters. This also reflects the strong influence of the very cold winters in Illinois during an extended period in the late 1970s and early 1980s.

Trends in selected parameters (seasonal average, minimum, and maximum temperature and seasonal precipitation) were also examined by individual seasons. The results of this analysis are given in tables A.5-A.8 (beginning on p.82) for the autumn, winter, spring, and summer seasons, respectively, and summarized in table 5. Very few stations show statistically significant seasonal trends in total precipitation. However, a number of stations show downward trends in temperature for autumn. Also, seven to eight stations show downward trends in winter temperature. Trends in spring are weakly positive.

Trends in Variability

An analysis of the trends in temperature variability are shown in figure 31 by season. For the summer season, the period 1961-1975 was characterized by the lowest variability. Figure 32 shows summer temperatures for this century. The low variability during 1961-1975 was the result of the absence of severe summer heat waves. By contrast, variability was much higher from 1976 through 1990, caused by more frequent summer heat waves. This latter 15-year period was quite similar to the first half of the century. For the fall season, low variability was experienced during 1901-1915 and 1945-1990, while higher variability characterized the years 1931-1945. For winter, 1976 through 1990 exhibited the greatest variability. This was the result of

the very cold winters during the late 1970s, contrasted with the mild winters of the middle and late 1980s. The spring season was also characterized by high variability during the period 1976-1990, as was winter.

Interestingly, trends of seasonal temperature variability in summer and fall parallel those of precipitation (figure 9), whereas those of winter and spring do not appear to be related.

TRENDS IN CLOUD COVER

Changnon (1984) presented the number of days per five years with cloudy skies (any cloud type or combination covering 70 percent or more of the sky) at Springfield, St. Louis, and Evansville, IN (figure 33) and Peoria, Moline, and Chicago (figure 34), from 1901 through 1980. All six sites showed a rather steady increase in cloudy days for the period of record, from about 90 days per year for the first few decades of the century, to about 160 days per year after about 1940 and continuing to 1980. Additional data from 1981 through 1990 maintain the higher frequencies (figure 33), except those of Evansville, IN (figure 34). Changnon (1984) questioned whether the increases might be due to greater frequencies of jet contrails, perhaps instead of or in addition to natural clouds. The question is as yet unanswered.

Although Petersen (1990) reported that cloud observing techniques were changed in June 1951 by the National Weather Service, the cloud cover record does not exhibit any discontinuity during the period shown. Prior to June 1951, fractional amounts of only two possible cloud layers were observed and therefore recorded, plus the height of the lowest scattered layer. Following 1951, sky cover and cloud height were reported. Whether this procedural change may have biased the record has not been evaluated.

Changnon et al. (1980) investigated temporal changes in cloud cover over Illinois from the turn of the century to 1977 (not shown). They demonstrated that the number of cloudy days increased by about 50 percent from the early years (about 110 days per year in southern Illinois, and 140 days per year in the north) to about 160 days per year in the south, and 180 such days in the north.

Petersen (1990) further calculated seasonal solar radiation since 1948 at several locations about the Midwest, based upon surface pressure, dew-point temperature, cloud height, and fractional sky cover. The results

Table 5. Summary of Seasonal Trends in Selected Temperature and Precipitation Parameters (Number of Stations)
(Statistically Significant Counts in Parentheses)

Season	Trend	Average temperature (°F/decade)	Average minimum temperature (°F/decade)	Average maximum temperature (°F/decade)	Total precipitation (inches/decade)
Autumn	Upward	14 (1)	17 (6)	10 (0)	35 (3)
	Downward	27 (14)	23 (11)	30 (14)	6 (0)
Winter	Upward	8 (0)	10 (0)	9 (0)	27 (0)
	Downward	33 (7)	31 (8)	32 (7)	13 (0)
Spring	Upward	26 (5)	26 (11)	24 (1)	35 (5)
	Downward	15 (2)	15 (2)	17 (3)	6 (0)
Summer	Upward	18 (5)	20 (13)	13 (0)	31 (5)
	Downward	22 (9)	21 (6)	28 (10)	10 (0)
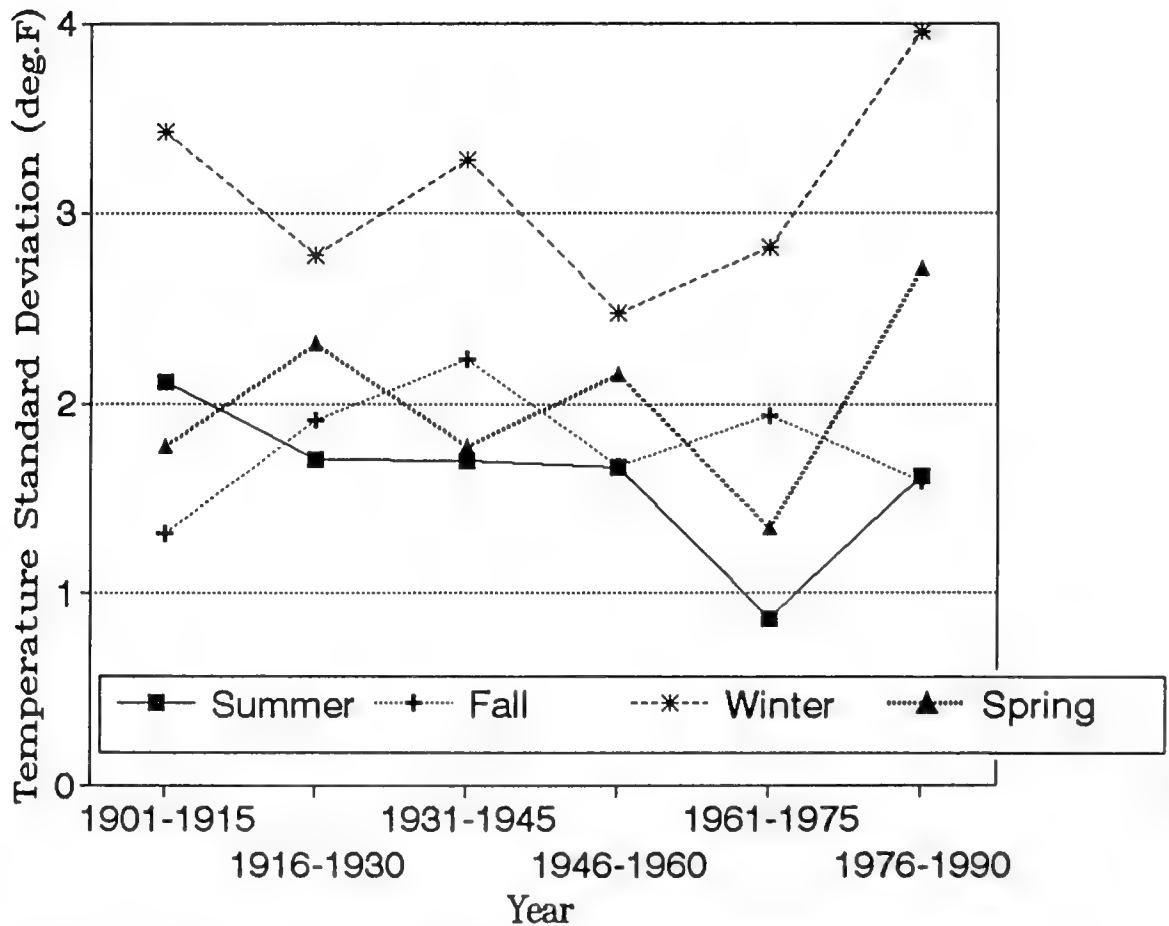
Figure 31. Standard deviations of 15-year segments of Illinois statewide average temperature,
1901-1915 to 1976-1990.
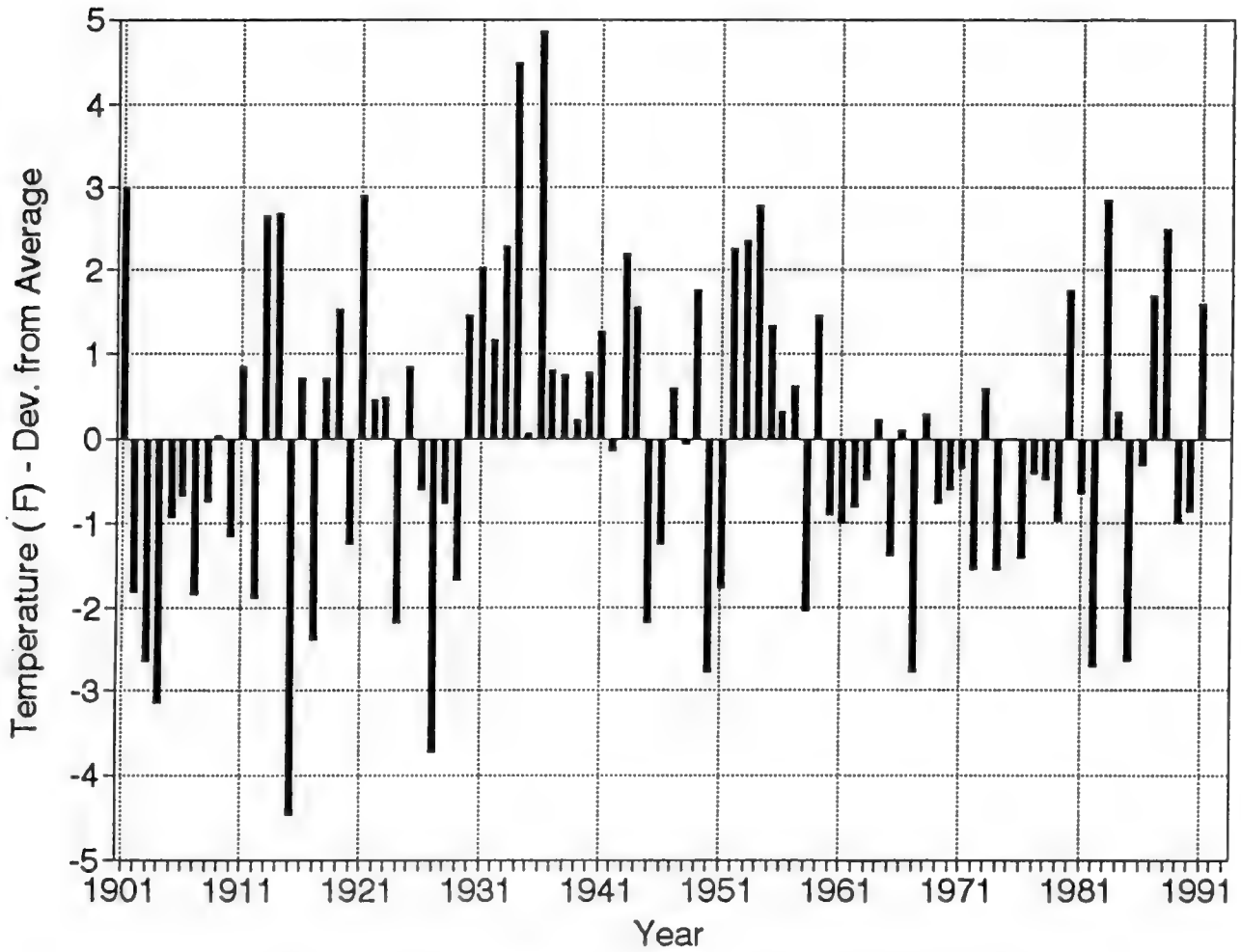
*Figure 32. Statewide summer temperature (°F) for Illinois, 1901-1991,
expressed as a deviation from the period average.*
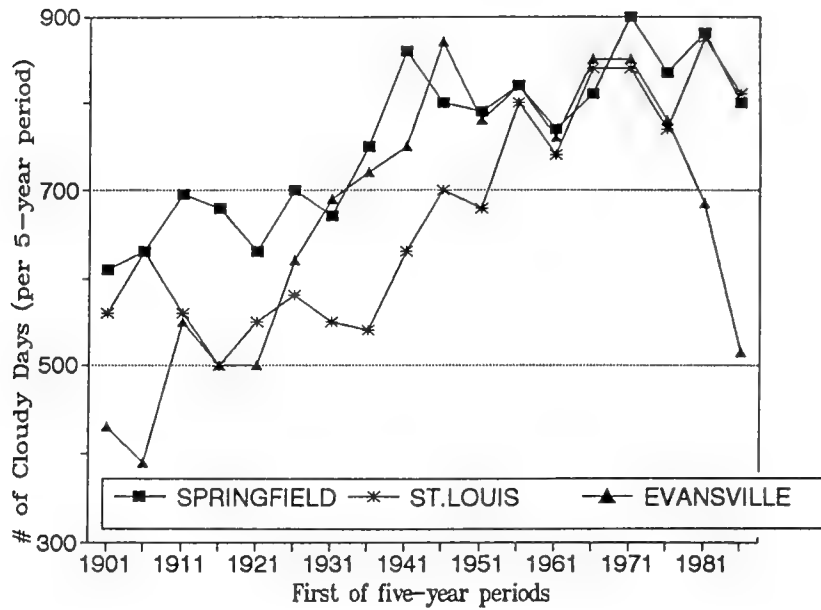
*Figure 33. Average number of days per five years with cloudy skies
at Springfield, IL; St. Louis, MO; and Evansville, IN, 1901-1990.*
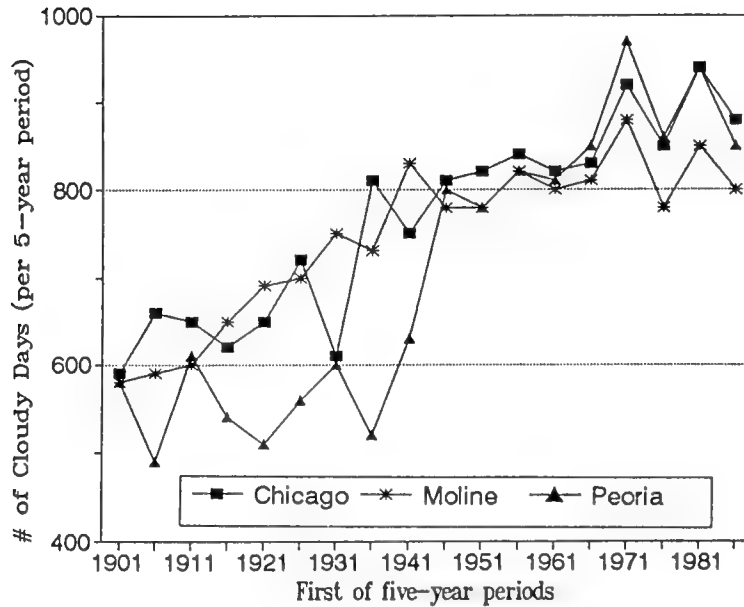
*Figure 34. Average number of days per five years with cloudy skies
at Chicago, Moline, and Peoria, IL, 1901-1990.*

relevant to Illinois are presented in table 6. Positive trends in solar radiation over much the same areas were noted at most sites and during all seasons except spring and fall. However, the increase in cloud cover occurred primarily during the first half of the century, a period not covered in the Petersen study.

As summarized by Folland et al. (1990), the upward trend in cloudiness has been observed in many other parts of the globe. They point out that some of the increase observed in the 1940s may be due to a change in observing practice; obscuring effects of smoke, haze, dust, and fog were included from the 1940s onward. However, the increases thereafter are believed to be real.

TRENDS IN SEVERE WEATHER

Trends in Freezing Precipitation since the Turn of the Century

Changnon (1984) presented the number of days per year with observations of glaze (freezing precipitation) for six NWS stations (Peoria, Moline, Chicago, Springfield, St. Louis, and Evansville, IN) from 1901 through 1980. At six sites, annual frequencies virtually doubled or more, beginning rather abruptly during the early 1940s. The higher frequencies continued until 1980, the end of the record. The absolute frequencies varied from site to site, with the greatest at Springfield, Peoria, and St. Louis.

Data were collected from 1981 through 1990 to bring Changnon's figures to the present (figures 35 and 36). Although the "extreme" frequencies reported in the 1970s at Springfield and Peoria have not continued, they remain within the range of values reported after the increase noted in the 1940s or 1950s.

Procedures for observing or reporting freezing precipitation have not changed significantly during the 90-year period. As a result, no explanation is forthcoming for the doubling (or more) of frequencies of glaze beginning in the early 1940s. A question remains whether these abrupt changes are real or an artifact of a procedural change in observations. Discussions with the NWS did not uncover any procedural changes coincident with the upward trend.

Temporal Change in Thunderstorm Frequency

Thunderstorms form in unstable air, the larger the area of instability, the greater the area of thunderstorms.

Further, greater instability leads to greater thunderstorm severity, i.e., strong, gusty winds, and possible hail and tornadoes. Changnon (1984) investigated the frequency of thunderstorms within and near Illinois from the turn of the century to 1980. We have updated those data through 1990, and present five-year totals of thunderstorm days for Chicago O'Hare and Peoria (figure 37), and Springfield, Moline, and St. Louis, MO, from 1901 through 1990 (figure 38), the pentad totals plotted at the first of the five years.

First, there are no clear temporal trends. Thunderstorm frequencies were relatively low during the late 1940s and early 1950s at both Chicago and Peoria, the very years when thunderstorms at St. Louis were most frequent! Although thunderstorms are somewhat related to temperature, i.e., warmer temperatures lead to more thunderstorms, they also depend on instability and available moisture. Also, thunderstorms are relatively small-scale phenomena, i.e., the area of any one event is usually a linear feature (associated with a cold front), confined to a one- to three-state area. Since mean frontal locations shift northward and southward with warming and cooling, respectively, the area favoring thunderstorms tends to be located north of usual during warm episodes, and south of usual during cold episodes.

Trends in Tornado Frequency

Illinois lies at the northeastern limit of "Tornado Alley," the group of states that exhibits the greatest tornado frequency on average, generally Texas through Illinois. Tornado frequencies change rather dramatically from year to year. Since 1955, the period during which tornado observations are thought to be essentially complete, Illinois has experienced 28 tornadoes per year on average. But as many as 107 were recorded in 1974, and as few as seven and eight were recorded in 1964 and 1968, respectively (Wendland and Guinan, 1988).

The annual frequency of tornadoes in Illinois is not well known prior to 1955, a period when the NWS "received" as opposed to "sought out" data on tornado occurrences within the United States. Annual Illinois tornadoes since 1881 are presented in figure 39. The data from 1845 to 1954 are known to be incomplete, having been compiled from newspapers and U.S. Weather Bureau publications, as available and summarized by Wendland and Hoffman (1993). However, it is interesting that some annual tornado frequencies prior to 1955, such as 1883, 1886, and 1890, were as great or greater than some years since 1955, the time during which tornado records in Illinois are thought to be complete.

Table 6. Period Trend (Positive or Negative) in Solar Radiation from 1948 to 1987, by Season

Location	January	April	July	October
Rockford, IL	positive	positive	positive	**negative**
Chicago, IL	**positive**	positive	positive	negative
Moline, IL	positive	positive	**positive**	**negative**
Peoria, IL	positive	positive	positive	**negative**
St. Louis, MO	positive	positive	positive	**negative**
Springfield, IL	**positive**	positive	**positive**	**negative**
Evansville, IN	positive	positive	positive	negative
Memphis, TN	positive	positive	**positive**	negative

Note: Boldface type indicates trends significant at the 10 percent level.
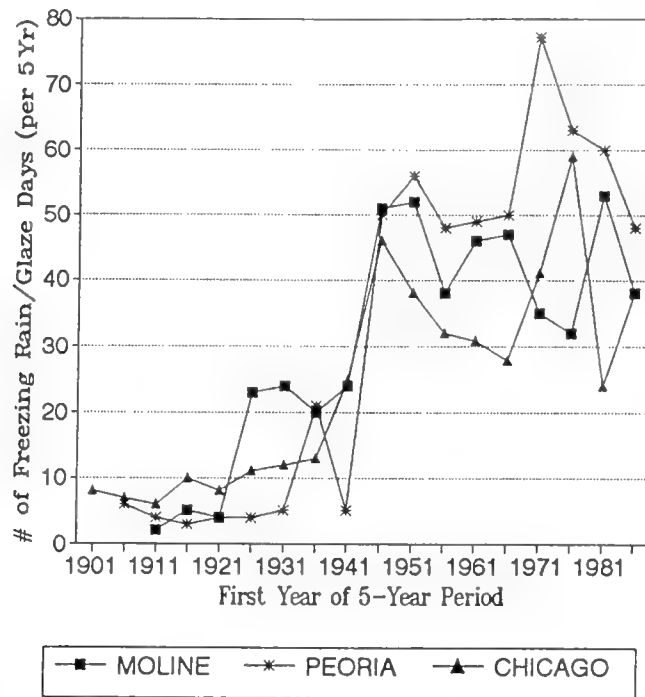
Figure 35. Average number of days per five years with glaze (freezing precipitation) at Chicago, Moline, and Peoria, IL, 1901-1990.
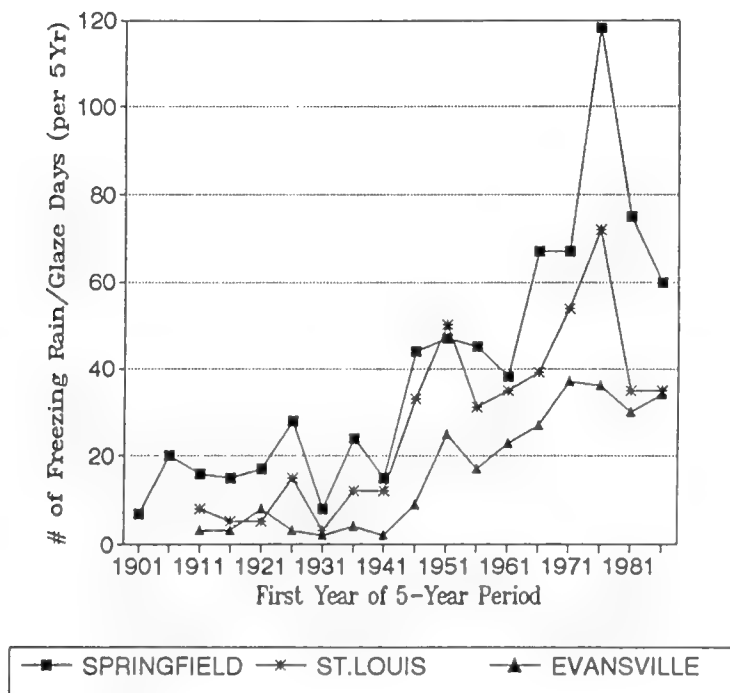
Figure 36. Average number of days per five years with glaze (freezing precipitation) at Springfield, IL; St. Louis, MO; and Evansville, IL, 1901-1990.
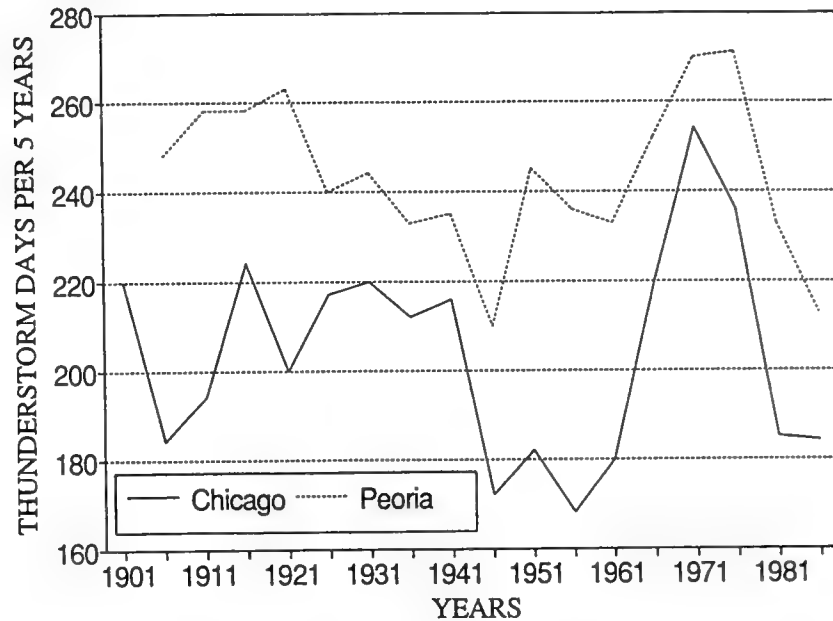
Figure 37. Average number of days with thunderstorms per five years (plotted on first of the five years) for Chicago and Peoria.
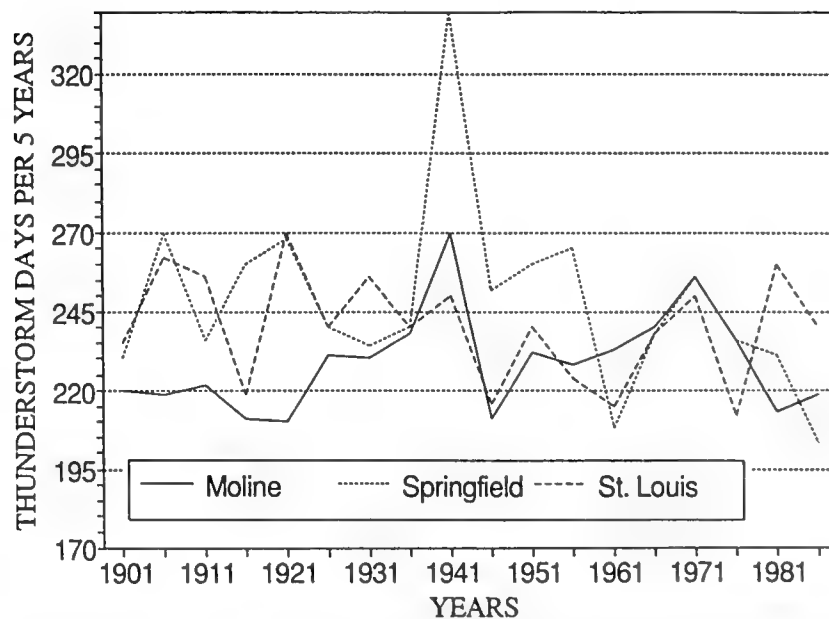
Figure 38. Average number of days with thunderstorms per five years (plotted on first of the five years) for Moline, Springfield, and St. Louis, MO.
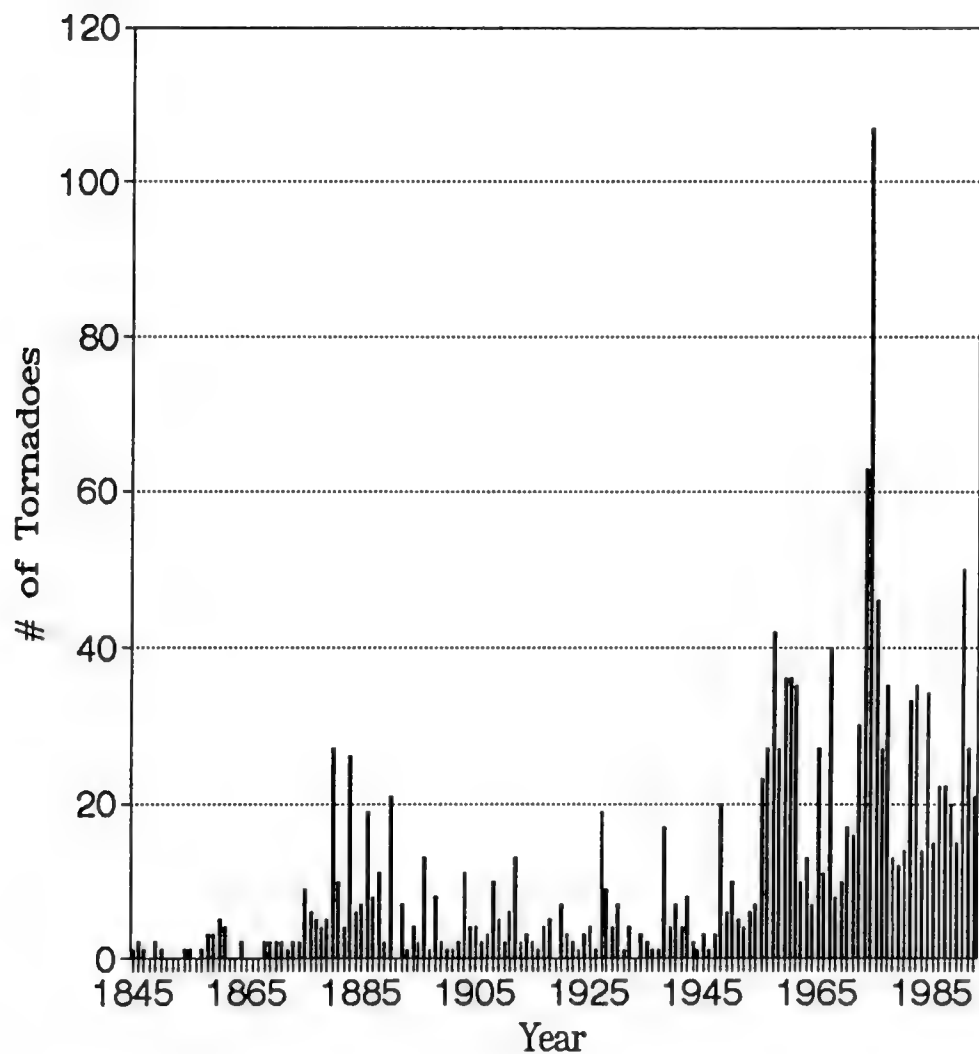
Figure 39. Number of reported tornadoes in Illinois (figure 7 from Wendland and Guinan, 1988), 1845-1990.

The annual record of the number of *days* with tornadoes in the state since 1960 (figure 40) exhibits several features:

1. Annual frequencies hover about a mean of about 11 tornado days per year from 1962 to 1971.
2. About 35 tornado days occurred in five consecutive years, 1973 through 1977.
3. About 25 tornado days occurred per year thereafter.

As to areas of the state that might seem to "favor" tornado activity, raw tornado frequencies clearly peak in those counties with greater population densities (Wendland and Guinan, 1988). The authors believe, however, that this relationship results merely from the fact that more people are likely to see and report isolated, small-scale events as tornadoes, and does not represent an actual urban enhancement of tornado frequency.

Trends in the Frequency of Severe Snowstorms

Severe snowstorms in Illinois are defined as those where 6 inches or more snow or freezing precipitation of any intensity falls over at least 10 percent of the area of the state within a 48-hour period. These storms exert tremendous hardship on those within the area of impact in terms of disruption to commerce, communication, comfort, and economics. As opposed to the historical tornado record, that for severe winter storms is believed to be rather well defined since the turn of the century because each such storm generally impacts an area of some 15,000 square miles and would therefore be recorded by observations from 50 or more NWS Coop sites, even in areas of less population density than represented today (Changnon, 1969, 1978). Newspapers have proven to be a reliable source for a relatively complete record of severe winter storms, whereas tornadoes are not necessarily so reported, due to their relatively small area of impact (on the order of a few tens of square miles).

From the turn of the century to the early 1960s, severe winter storms have occurred about five times per winter (figure 41). After the early 1960s, the frequency has declined to about three such storms per winter. Winters with greatest severe storm frequencies include 1911-1912 with 12 storms, and 10 each in 1925-1926, 1943-1944, and 1950-1951. There were no severe winter storms in ten of the last 30 winters, including 1961-1962, 1962-1963, 1965-1966, 1967-1968, and 1968-1969, 1970-1971, 1980-1981, 1981-1982, and 1982-1983, and 1991-1992.

The record of severe winter storm frequencies in Illinois somewhat parallels that of temperature, suggesting increased winter storm frequency with increased mean temperatures. This relationship may be due to a number of factors, including greater moisture supplies during warmer winters, a more northerly storm track during the middle of winter, etc. However, this study did not examine specific factors for this relationship.

Trends in Illinois Droughts

Drought can be measured by several techniques. In this instance we restrict our discussion to meteorological drought, i.e., a shortfall of precipitation. We present the percentage of the state experiencing 50 percent or less of the 30-year average precipitation during July and August from 1901 through 1991. The second uses the same threshold but for the full growing season, April through August, also from 1901 through 1991.

Five of the July-August droughts (figure 42) impacted at least 50 percent of the area of Illinois during the period 1901-1992, notably 1991 (impacting 79 percent of the state), 1930 (70 percent), 1936 (51 percent), 1971 (66 percent), and 1983 (55 percent). The recent 1988 drought impacted only about 30 percent of the state by this criterion.

According to the more stringent criterion, 50 percent or less than average precipitation from April through August (figure 43), a somewhat different distribution emerges, with extraordinary droughts in 1914 and 1936 (impacting about 60 percent of the state) and 1988 (54 percent). Neither 1914 nor 1988 were particularly noteworthy according to the different criterion, 50 percent or less precipitation during July and August. However, those years (particularly 1988) were characterized by extraordinary dryness in late spring and early summer.

Temporal Changes in Visibility

There has been and continues to be concern as to whether the atmosphere is becoming more turbid with time, perhaps due to industrialization, increased plowing and cultivation frequencies etc. Such a change could lead to reduced insolation, and related reduction in surface temperature, atmospheric clarity, and visibility. Although measurements of atmospheric turbidity are not generally available, we use several proxy records, including visibility and frequency of days with smoke or haze.

Wendland and Bryson (1970) discussed a simple relationship between mean temperature, carbon dioxide (CO_2) concentration, atmospheric turbidity and sunspots from 1880 to 1960. Using dust concentrations obtained in high snowfields of the Caucasus (Davitaia,
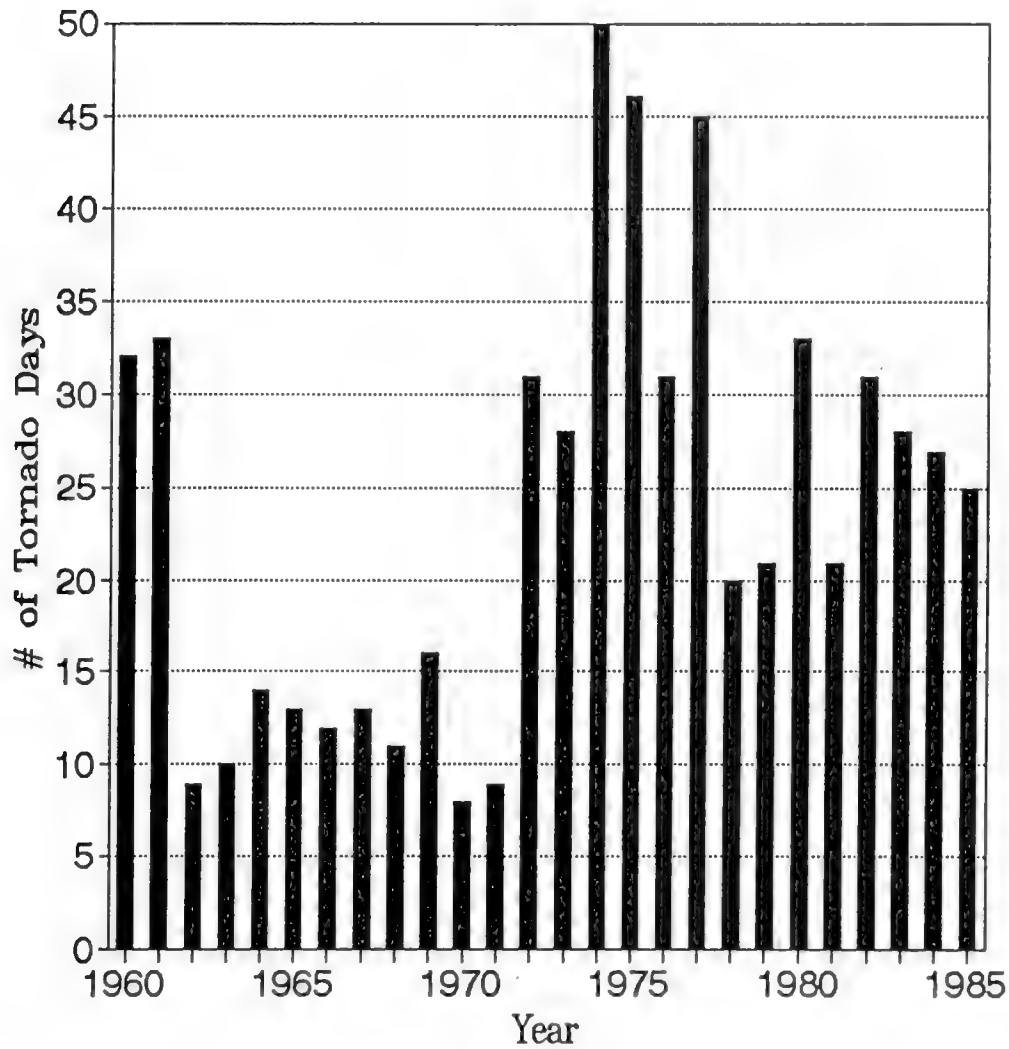
Figure 40. Number of tornado days in Illinois per year (Wendland and Guinan, 1988), 1960-1985.
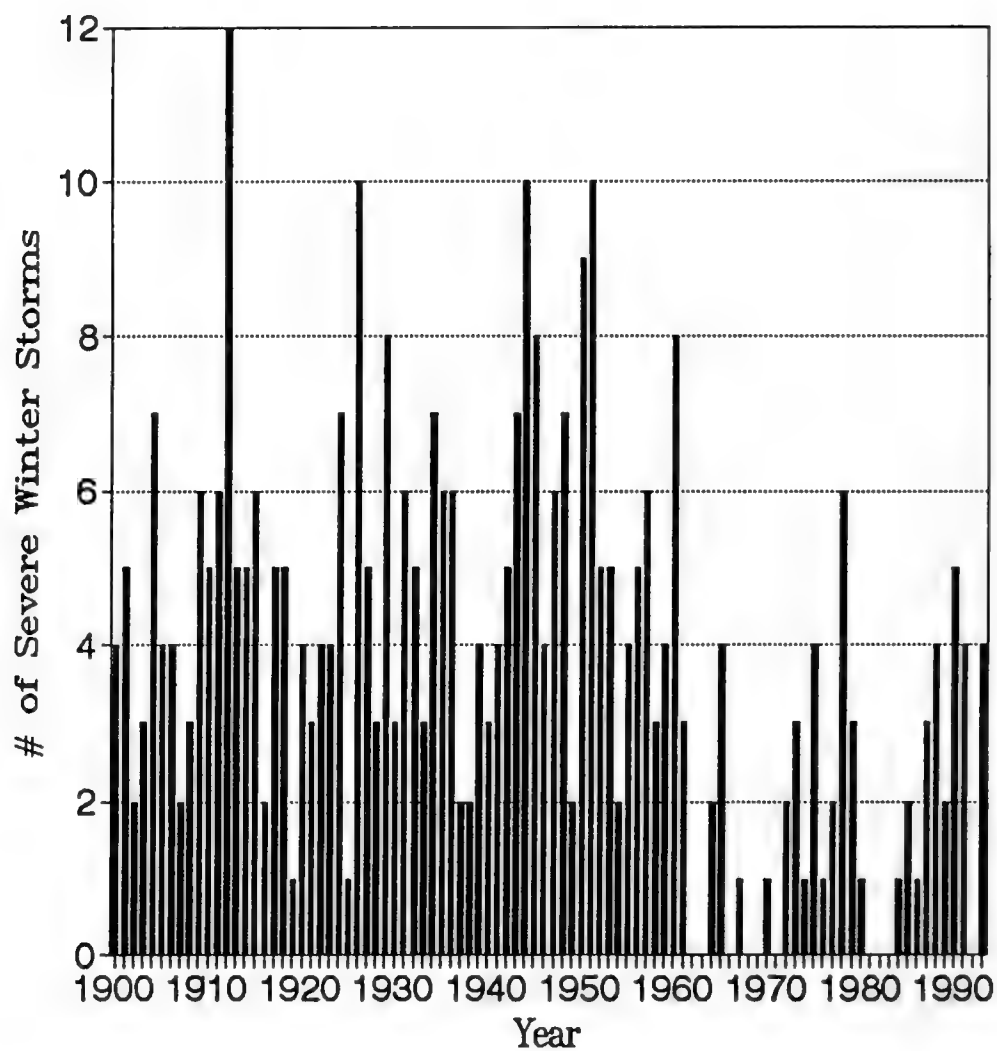
Figure 41. Number of severe winter storms per year in Illinois (defined as an area with at least 6 inches of snow or any amount of glaze within 48 hours, over 10 percent or more of the state), 1900-1992.
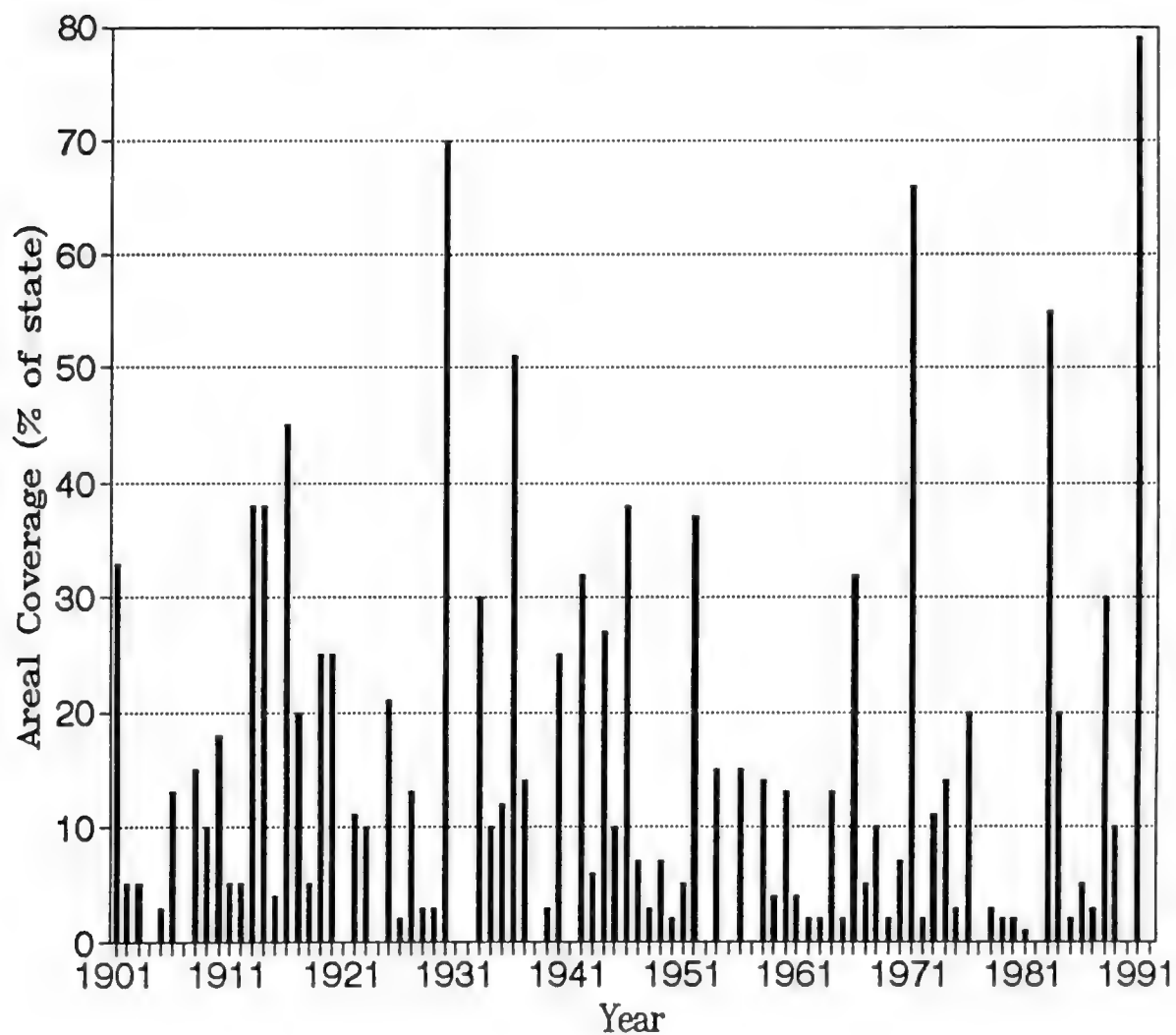
Figure 42. Percent of Illinois impacted by 50 percent or less average precipitation during July-August, 1901-1992.
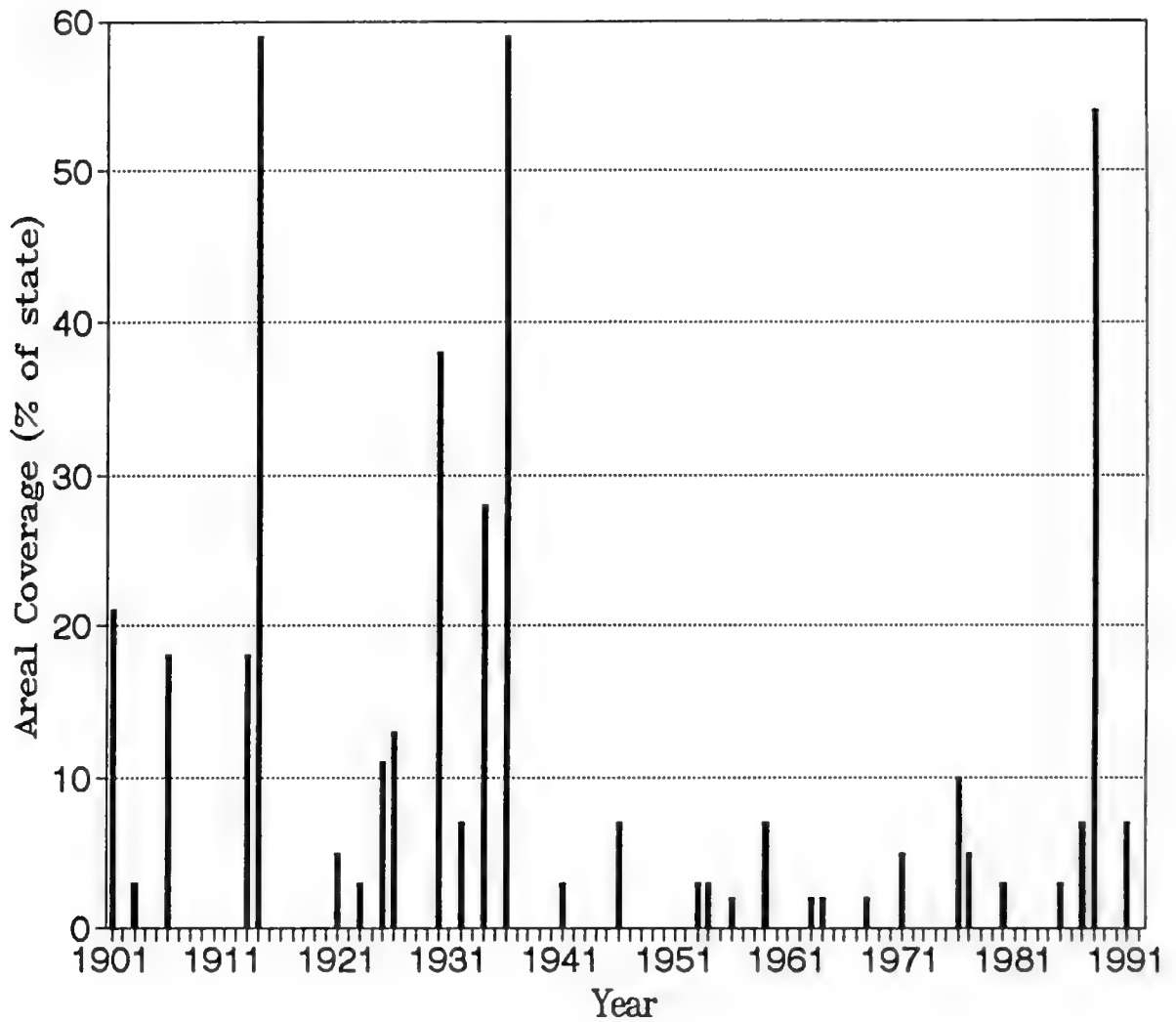
Figure 43. Percent of Illinois impacted by 50 percent or less average precipitation
during April-August, 1901-1992.

1965) which they suggest reflect integrated hemispheric loadings, they demonstrated that global mean temperature changes during those eight decades could be statistically explained by those three parameters. Specifically, the warming from 1880 through the mid 1900s was primarily related to increases in CO_2 concentration, whereas the cooling thereafter was primarily related to the dramatic increase in dust loadings, noted during the 1940s and 1950s.

Atmospheric clarity can be defined and measured several different ways. The concept of visibility, however, is straightforward, the measurement technique has changed little over time, and hence visibility measurements of decades ago are generally comparable to those of today. Vinzani & Lamb (1985) showed declining visibility at NWS sites in and around Illinois since 1950. We analyzed hourly observations of visibility since 1948 at Chicago-O'Hare, Moline, and Peoria. Frequencies of occurrences of visibilities in various categories were accumulated for each year. Following Vinzani and Lamb (1985), we estimated the values that were exceeded 60 percent and 90 percent of the hours. Trends in these values are shown in figures 44 and 45. The trends in the 60 percent and 90 percent exceedance values are upward at Chicago, indicating improving visibility. Decreasing trends in the 90 percent value are seen at Moline and Rockford. Decreasing trends in the 60 percent value are seen at Springfield. The increasing visibility at Chicago, in contrast to the negative or insignificant trends at the other sites, may be due to more stringent pollution controls on industry.

Using yet another approach, Changnon (1987) showed that the number of smoke and/or haze days at six NWS First Order Stations in and around Illinois showed a dramatic increase from a few tens of days per year prior to the 1930s, to 100 or more days per year thereafter through 1980! We have updated the frequency of smoke and/or haze data through 1990 (figures 46 and 47), and find that the increased frequencies which abruptly began in the 1930s essentially continue to the present, though the highest frequencies at all sites were noted in the 1930s and 1940s. The values of the last few pentads continue at several times greater than pre-1930 frequencies, but about half the maximum-ever values.

ANOMALOUS EPISODES FROM THE ILLINOIS CLIMATE RECORD

Global climate models (GCMs) suggest a future warming in Illinois and redistribution of precipitation during the year under greenhouse-induced effects of additional trace gases into the atmosphere. What would the impacts of such climate changes be for Illinois? The model outputs provide only average conditions, although state resources vulnerable to climate are often influenced by interannual or multiyear anomalies or by several conditions not defined by GCMs. For example, the range of certain species of insects or vegetation in Illinois depends on such conditions as the number of frost-free days, the timing of precipitation events, and changes in diurnal temperature ranges, conditions not available from GCMs.

The variability of a potential future climate may also be documented by studying the climates of the past. Identifying extreme seasons or years from the past that lie within the average conditions specified by the GCMs allows one to reconstruct variabilities and use past analogs to estimate future climates.

Over the past century, notable months, seasons, and years stand out as having been particularly warm, cold, wet, or dry. Indeed, it is often thecombination of temperature and precipitation extremes that stand out and impact Illinois resources. For example, the Dust Bowl years of the 1930s were prominent for their extreme drought *and* warm temperatures as were the 1988 drought and 1993 floods.

To understand how climate has varied over the past century, it is useful to isolate those episodes classified as 1) warm and dry, 2) cold and wet, 3) warm and wet, and 4) cold and dry. This characterization is also useful in grappling with the impacts of potential future climates in Illinois.

To identify combined temperature and precipitation extremes from the Illinois climate record, individual mean monthly temperature and precipitation values were recorded for each year between 1895 and 1991 and normalized relative to the 97-year average. The index for temperature is calculated as:

$$I_t = (T_i - T_{avg}) / T_{std}$$

where: I_t = normalized monthly temperature
T_i = average temperature for month in question
T_{avg} = 97-year monthly mean temperature
T_{std} = 97-year monthly temperature standard deviation

For precipitation, the calculation is:

$$I_p = (P_i - P_{avg}) / P_{std}$$

Figure 44. Visibility (miles) exceeded 60 percent of the time for Moline, Chicago, and Peoria, 1950-1991.

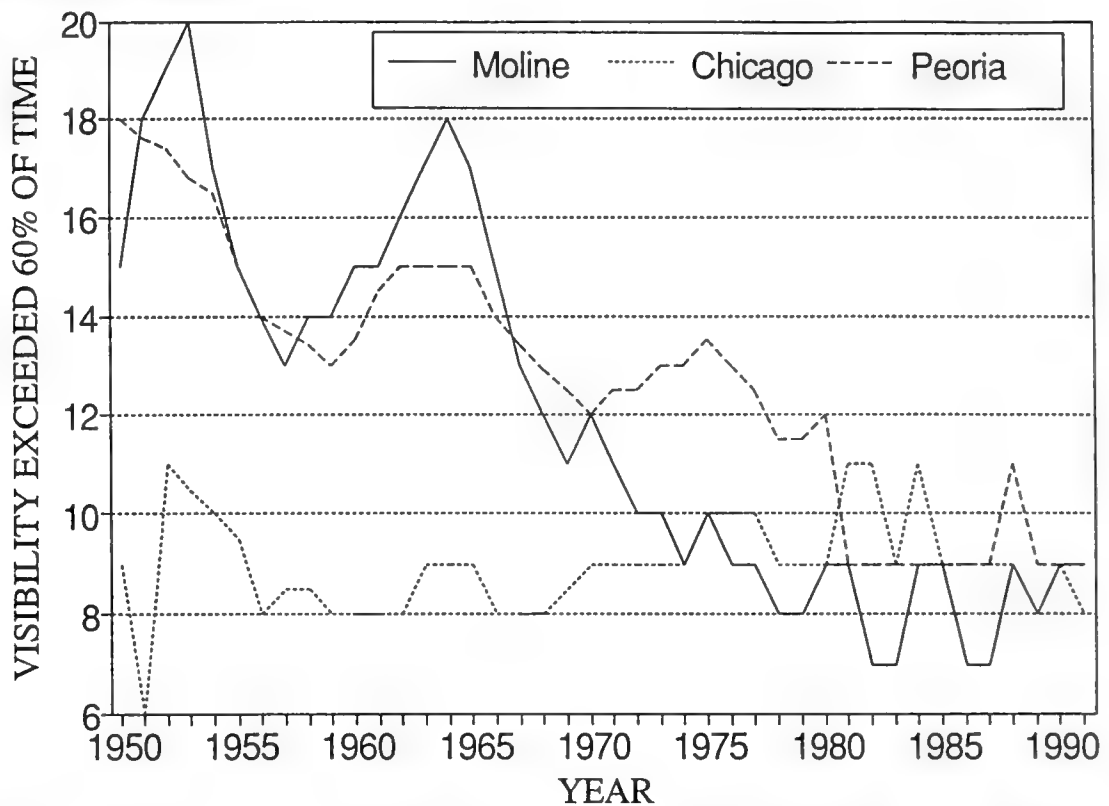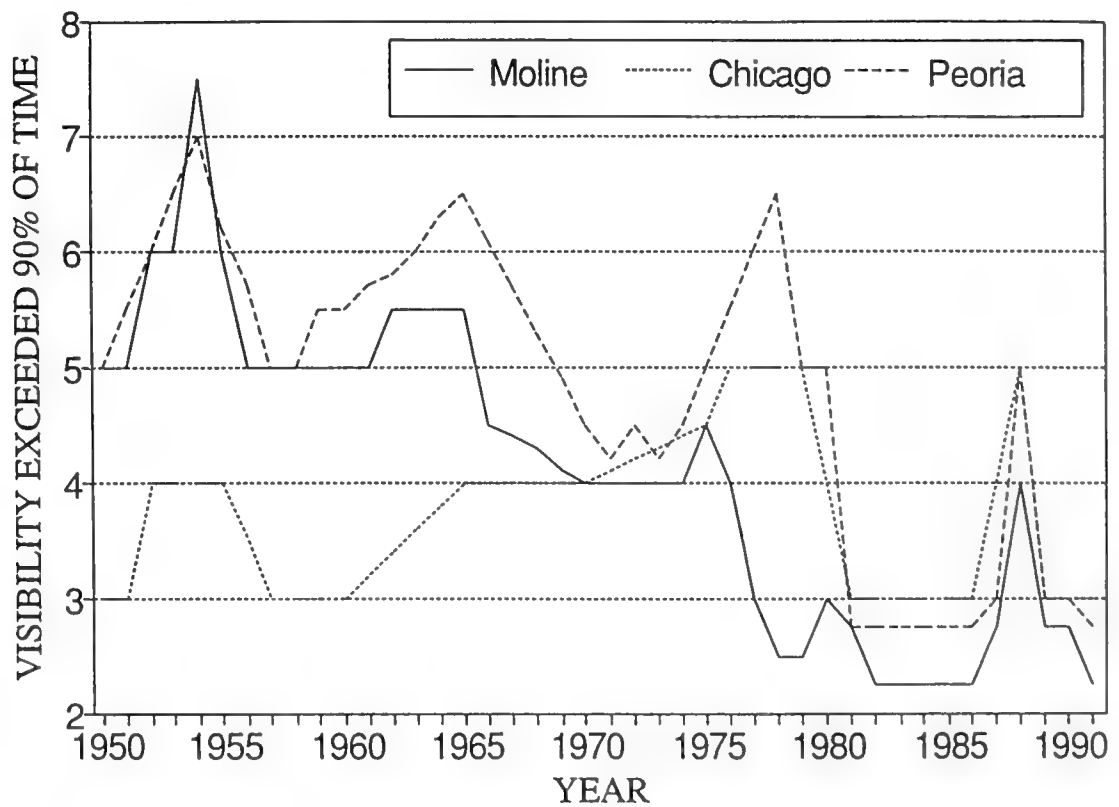Figure 45. Visibility (miles) exceeded 90 percent of the time for Moline, Chicago, and Peoria, 1950-1991.
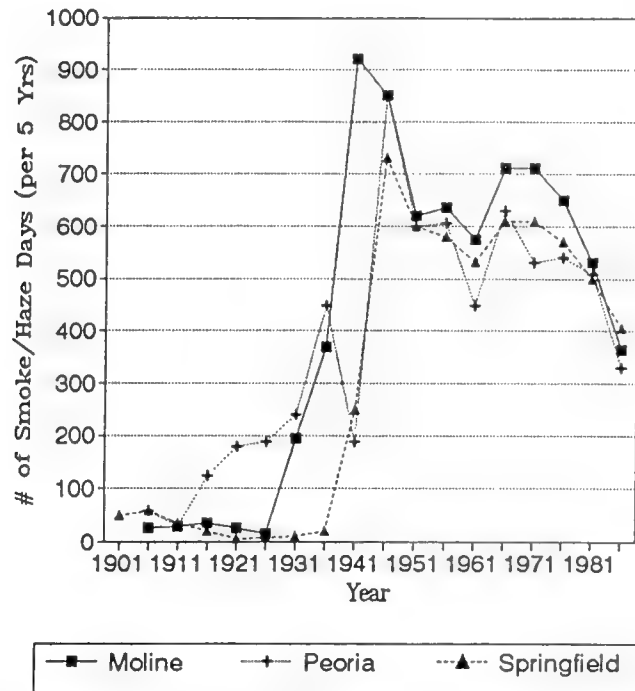
Figure 46. Number of days per five years when smoke and/or haze was reported
at Moline, Peoria, and Springfield (abscissa date in the first year of the five-year periods).
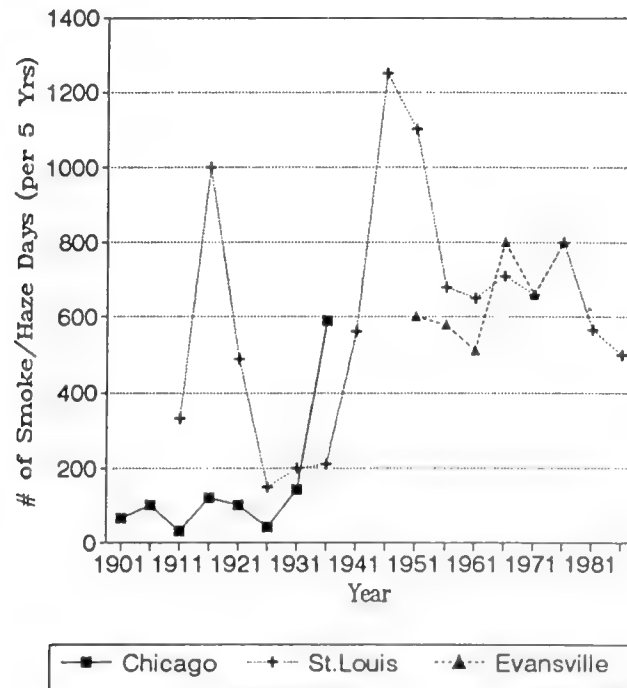

Figure 47. Number of days per five years when smoke and/or haze was reported
at Chicago, St. Louis, and Evansville.

where: I_p = normalized monthly precipitation
P_i = total precipitation for month in question
P_{avg} = 97-year monthly mean precipitation
P_{std} = 97-year monthly precipitation standard deviation

A normalized monthly value near zero indicates a temperature or precipitation value similar to that of the 97-year average. Values above zero represent positive deviations (warmer or wetter than the average). Values below zero represent a negative deviation (cooler or drier than the average). The magnitude of the deviation is expressed in standard deviation units, i.e., +1.0 indicates that the anomaly is one standard deviation greater than the period average. A seasonal normalized value was calculated for each season.

Warm and dry, and cold and wet seasons were distinguished by subtracting the normalized seasonal precipitation from normalized temperature: large positive numbers represent the former, and large negative numbers represent the latter. Warm and wet, and cold and dry seasons were distinguished by adding the normalized seasonal precipitation and temperature values: large positive results represent warm and wet seasons, whereas large negative numbers represent cold and dry seasons.

When adding the normalized values, results near zero indicate that either temperature or precipitation is greater than the mean by about the same amount that the remaining parameter is less than the mean. When subtracting normalized precipitation from normalized temperature, results near zero indicate near-equal anomalies for each parameter.

An inherent problem occurs when interpreting the result of either adding or subtracting normalized values of temperature and precipitation. One relatively large normalized value will dominate the result of either process if the other normalized value is a very small number, i.e., very close to the long-term mean. When results from either addition or subtraction were near zero, they had to be individually studied to determine the significance of the number. In this study, small normalized values, those between -0.25 and +0.25, were set to zero. (Therefore some years in the following figures exhibit no difference from the long-term mean.) Normalized precipitation and temperature values were added and subtracted to identify warm and dry, cold and wet, warm and wet, and cold and dry statewide conditions for each of four seasons between 1895 and 1991.

Statewide Summer Index

The combined indices for summer are plotted in figures 48 and 49. They show a high frequency (over three times as likely) of warm and dry, and cold and wet summers, as opposed to summers that were either warm and wet, or cold and dry. In addition, the magnitude of the latter index is much less than that for warm and dry, and cold and wet summers. As expected in a continental climate, heat waves are more likely to be associated with moisture deficiencies, and cool summers are more likely associated with excess moisture.

The combined extremes of warm and dry, and cold and wet summers are more pronounced than warm and wet, and cold and dry summers. A second feature is the persistent nature of the occurrence of warm and dry, and cold and wet summers; i.e., summers of a given character tend to occur in succession. For example, the period between 1899 and 1926 is characterized by a variety of combined temperature and precipitation extremes. The period between 1930 and 1944 is almost exclusively warm and dry, whereas the period from 1961 to 1981 is almost exclusively cold and wet. The period between 1950 and 1959 is characterized by a return to a variety of extremes, as is the period between 1981 and 1991. All but one of the cold and dry summers occurred prior to 1920.

Individual summers that stand out as warm and dry are 1901, 1913, 1914, 1930, 1933, 1936, 1983, 1988, and 1991; whereas cold and wet are 1902, 1907, 1915, 1924, 1958, and 1981; warm and wet are 1980 and 1987; and cold and dry are 1920 and 1976. For the summers with identified extremes, seasonal temperature and precipitation values and deviations from the 97-year average are presented in table 7.

Statewide Winter Index

The combined indices for the winter months are plotted in figures 50 and 51 as a function of time. In contrast to the summer months, warm and dry, and cold and wet winters are *less* frequent than warm and wet, and cold and dry conditions over the past 97 years. Cold and dry winters were three times more likely than cold and wet winters. The occurrence of cold and wet winters is restricted to the years after 1962, and three out of the four occurred after 1978. Individual winters that stand out as warm and dry are 1921, 1931, 1953, 1954, and 1987; whereas 1979 and 1985 were cold and wet; 1949, 1950, and 1983 were warm and wet; and 1963, 1977, and 1978 were cold and dry. For the winters with identified extremes, seasonal temperature and

Figure 48. Identification of statewide warm and dry, and cold and wet summers (1895-1991).
See text for definition of index.

Figure 49. Identification of statewide warm and wet, and cold and dry summers (1895-1991).
See text for definition of index.

Table 7. Statewide Summer Seasonal Combined Temperature and Precipitation Extremes

	Temperature		*Precipitation*	
Year	*Fahrenheit*	*Deviation*	*Inches*	*Percent deviation*
1901	76.7	+2.8	7.79	70
1902	71.9	-2.0	17.49	157
1907	71.9	-2.0	15.82	142
1913	76.4	+2.5	7.02	63
1914	76.4	+2.5	7.58	68
1915	69.3	-4.6	17.82	160
1920	72.5	-1.4	7.59	68
1924	71.6	-2.3	15.54	140
1933	76.0	+2.1	6.34	57
1936	78.6	+4.7	5.63	50
1958	71.7	-2.2	17.44	156
1976	72.3	-1.6	8.19	73
1980	75.5	+1.6	12.69	114
1981	73.1	-0.8	17.75	160
1983	76.6	+2.7	8.30	74
1988	76.2	+2.3	6.14	55

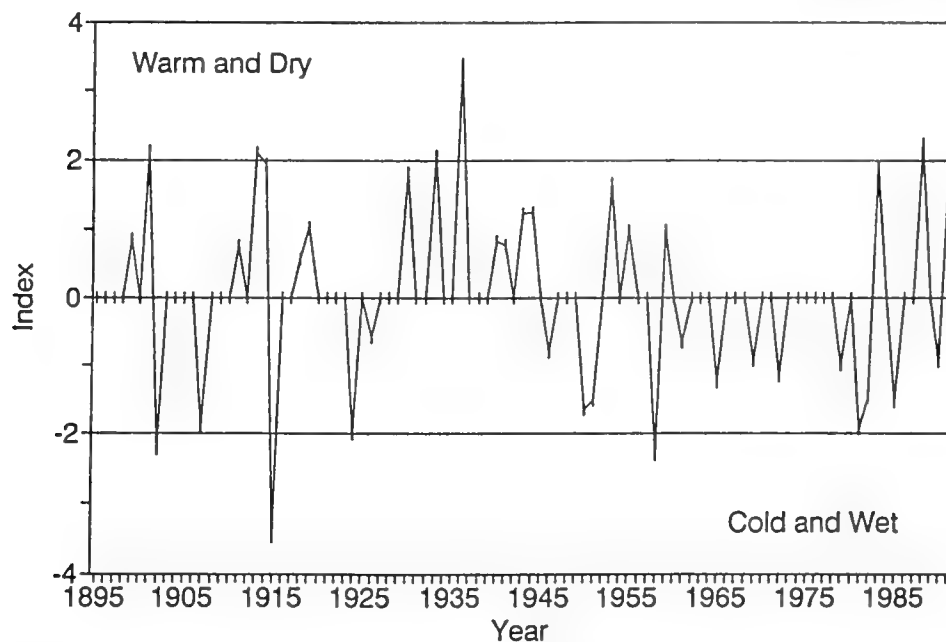Note: Deviation and percent deviation calculated from the 97-year statewide summer-season average.

Figure 50. Identification of statewide warm and dry, and cold and wet winters (1895-1991). See text for definition of index.
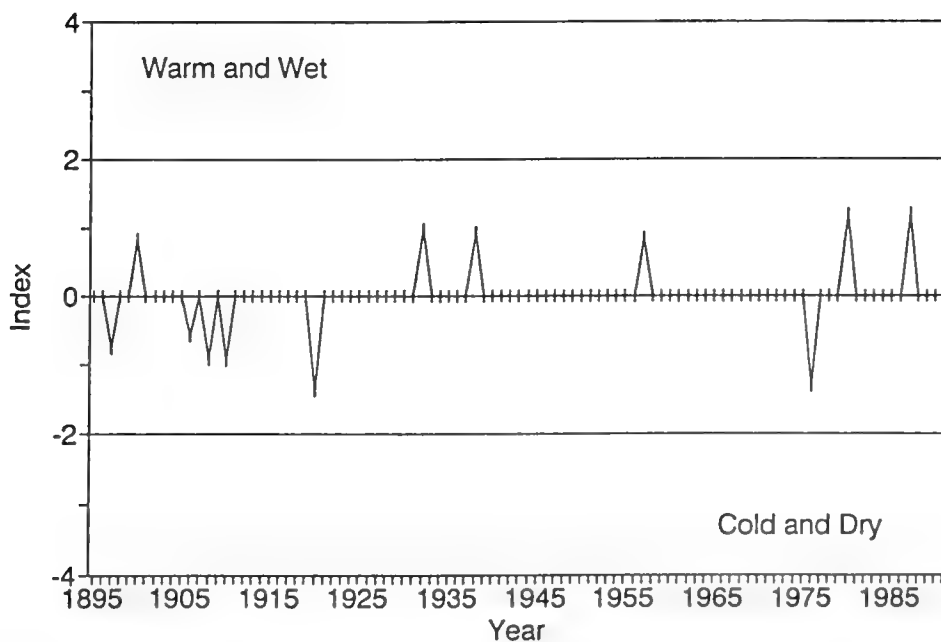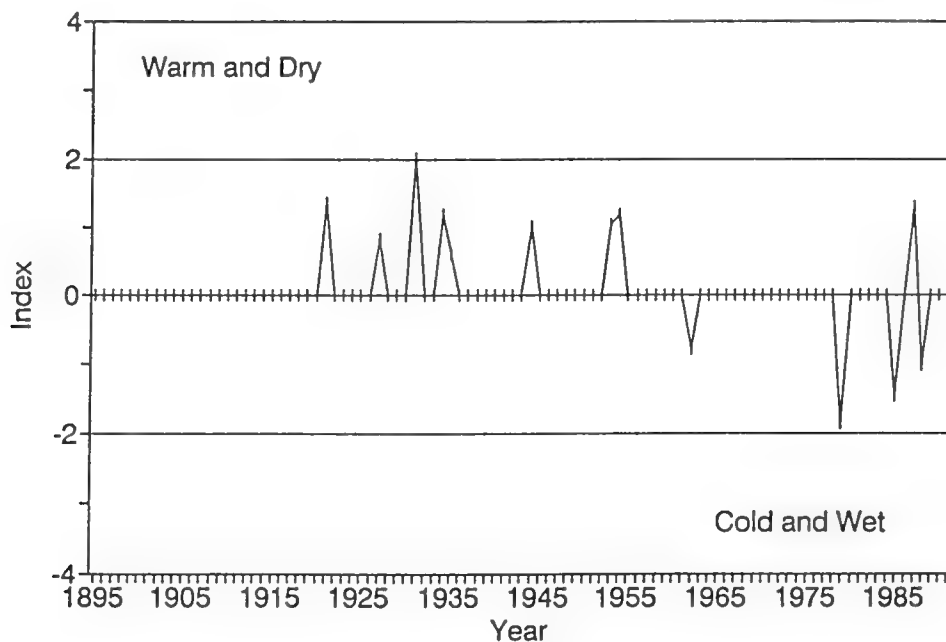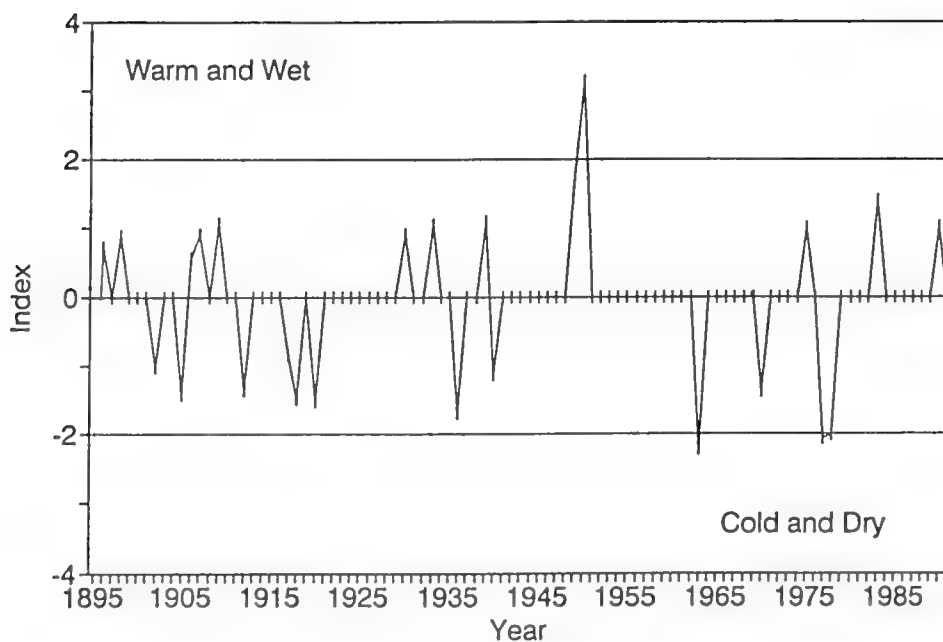

Figure 51. Identification of statewide warm and wet, and cold and dry winters (1895-1991). See text for definition of index.

precipitation values and deviations from the 97-year average are presented in table 8.

Statewide Spring Index

The combined index for the spring months are plotted in figures 52 and 53 as a function of time. In contrast to both the summer and winter seasons, springtime combined temperature and precipitation extremes are more equally distributed among the four possible combinations, reflecting the relatively large day-to-day temperature differences during transition seasons. However, the warm and dry springs were for the most part restricted to the years between the 1920s and 1930s and the late 1980s.

The spring climate record also shows active and inactive intervals. The period between the 1920s and 1930s experienced numerous deviations from average conditions (usually warm and dry, or cold and dry), whereas the late 1940s to the mid-1970s was a somewhat benign period with only minor variations from the 96-year average. Individual springs that stand out as warm and dry are 1930, 1934, 1936, 1986, 1987, and 1988; whereas cold and wet are 1983 and 1984; warm and wet are 1921, 1922, 1945, and 1991; and cold and dry are 1901, 1926, and 1971. For the springs with identified extremes, seasonal temperature and precipitation values and deviations from the 97-year average are presented in table 9.

Statewide Autumn Index

The combined indices for the autumn months are plotted in figures 54 and 55 as a function of time. Warm and dry autumns have occurred more often than the other three possible combinations, with the largest cluster of occurrences confined to the period between the late 1950s and mid-1960s. A significant warm and dry autumn has not occurred since 1971. The warm and wet autumns are clustered in the period between the early 1920s and the late 1930s. Individual autumns that stand out as warm and dry are 1938, 1939, 1953, 1963, and 1971; whereas cold and wet are 1911 and 1926; warm and wet are 1927, 1931, and 1941; and cold and dry are 1917 and 1976. For the autumns with identified extremes, seasonal temperature and precipitation values and deviations from the 97-year average are presented in table 10.

Statewide Annual Index

While warm and dry, cold and wet, warm and wet, and cold and dry seasons have been identified from Illinois'

97-year climate record, the succession of the seasons and their climate has the ultimate impact. For example, the impact of a warm and dry summer would likely be greater if the previous spring and winter were also warm and dry in relation to their respective averages. Thus, efforts to group the climates of consecutive seasons are useful. For example, an examination of calendar years (a grouping of four seasons) identified 1914, 1930, 1934, 1936, 1953, and 1987 as warm and dry; the years 1926, 1950, 1951, and 1972 as cold and wet; the years 1973 and 1983 as warm and wet; and the years 1895, 1917, 1920, and 1976 as cold and dry. The results of several GCMs suggest that the trend of the Illinois climate of the future will be toward warm and dry summers and warm and wet winters. Analog years with similar summer and winter trends are 1930, 1933, and 1983.

Regional Index

The state of Illinois measures approximately 400 miles north to south, and the climates of northern, central, and southern Illinois are not homogeneous. For example, a cold and wet summer in northern Illinois may occur in concert with a warm and dry summer in the southern third of the state. The National Weather Service divides Illinois into nine climate regions (see figure 1, p.9). For this comparison, the northern sector of the state was represented by region 1, the central district by region 5, and the south by region 9. Warm and dry, cold and wet, warm and wet, and cold and dry seasons were identified in a fashion similar to the statewide calculations. Comparison of the combined index for each of the three regions and the statewide calculation shows general agreement across the region for each of the four seasons.

A number of identified years overlap (especially those identified as extreme) and the trends over time are similar to the statewide trends. However, some year-to-year differences occur when an identified season for a particular region is not similarly identified as a statewide occurrence and vice versa. As illustration, warm and dry, cold and wet, warm and wet, and cold and dry summer seasons are identified for each of three regions (figures 56 and 57).

CONCLUSIONS

Although long-term trends can be identified in various climate parameters, the dominant characteristic of Illinois's climate is the presence of large variability on time scales of a few years or less. That is, the magnitude of long-term trends is in general considerably less

Table 8. Statewide Winter Seasonal Combined Temperature and Precipitation Extremes

	Temperature		*Precipitation*	
Year	*Fahrenheit*	*Deviation*	*Inches*	*Percent deviation*
1921	33.6	+4.9	5.10	79
1931	33.2	+4.5	2.47	38
1949	31.0	+2.3	11.41	177
1950	32.5	+3.8	15.51	240
1953	33.0	+4.3	5.48	85
1954	33.4	+4.7	5.25	81
1963	21.9	-6.8	2.37	37
1977	20.8	-7.9	2.95	46
1978	19.6	-9.1	4.56	71
1979	19.9	-8.8	7.95	123
1983	33.7	+5.0	9.01	140
1985	26.1	-2.6	8.93	138
1987	31.4	+2.7	4.03	62

Note: Deviation and percent deviation alculated from the 97-year statewide winter season average.
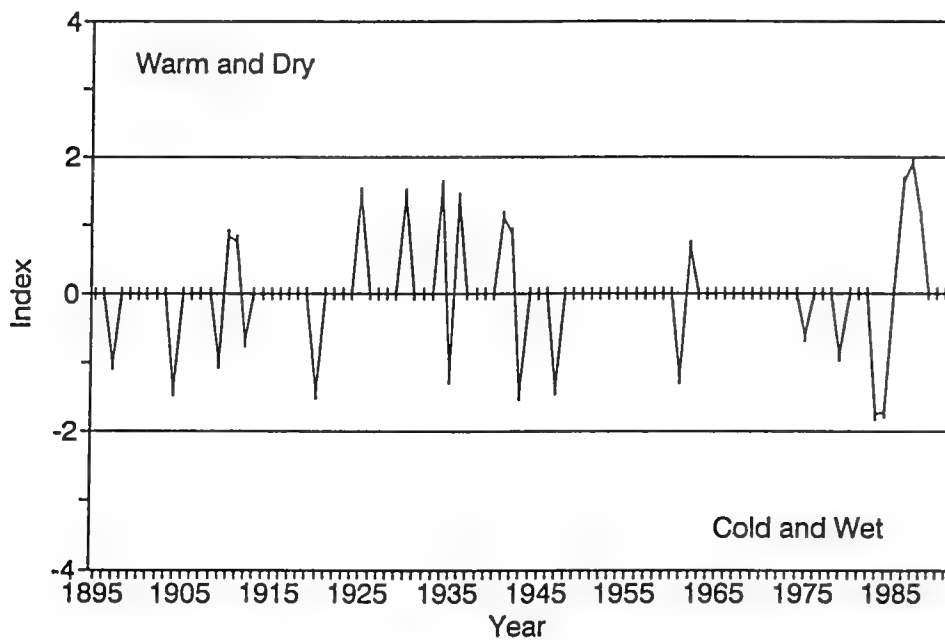
Figure 52. Identification of statewide warm and dry, and cold and wet springs (1895-1991). See text for definition of index.
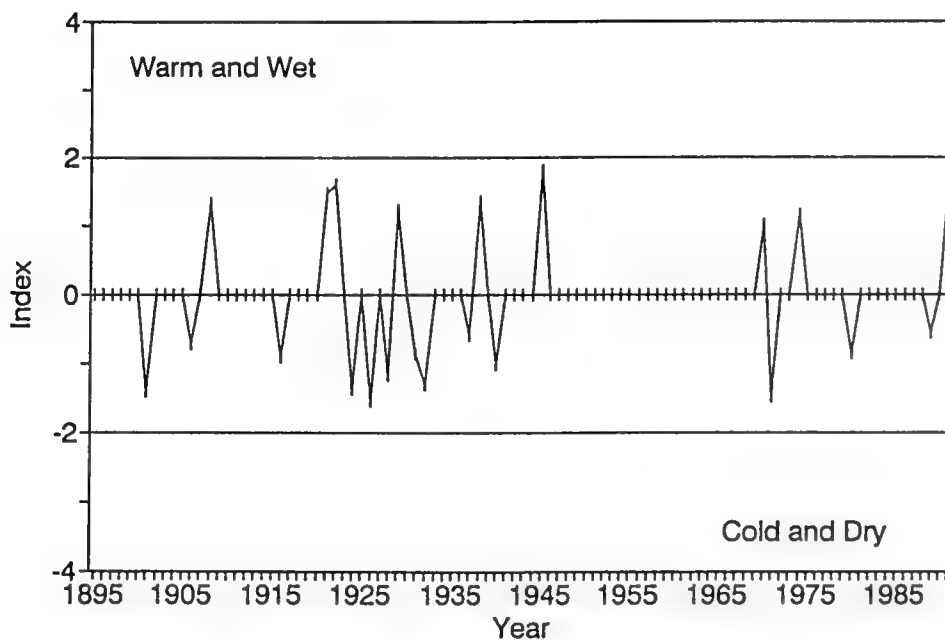
Figure 53. Identification of statewide warm and wet, and cold and dry springs (1895-1991).

Table 9. Statewide Spring Seasonal Temperature and Precipitation Extremes

| Year | Temperature | | Precipitation | |
	Fahrenheit	Deviation	Inches	Percent deviation
1901	49.4	-2.1	7.07	65
1921	56.0	+4.5	11.97	110
1922	53.6	+2.1	14.97	140
1926	47.8	-3.7	7.93	73
1930	52.4	+0.9	6.07	56
1934	52.0	+0.5	5.16	47
1936	52.7	+1.2	6.01	55
1945	53.6	+2.1	17.32	158
1971	49.6	-1.9	6.27	57
1983	48.8	-2.7	15.08	138
1984	47.3	-4.2	14.13	129
1986	54.6	+3.1	8.01	73
1987	55.3	+3.8	7.14	65
1988	52.2	+0.7	6.91	63
1991	56.0	+4.5	12.34	113

Note: Deviation and percent deviation calculated from the 97-year statewide spring season average.
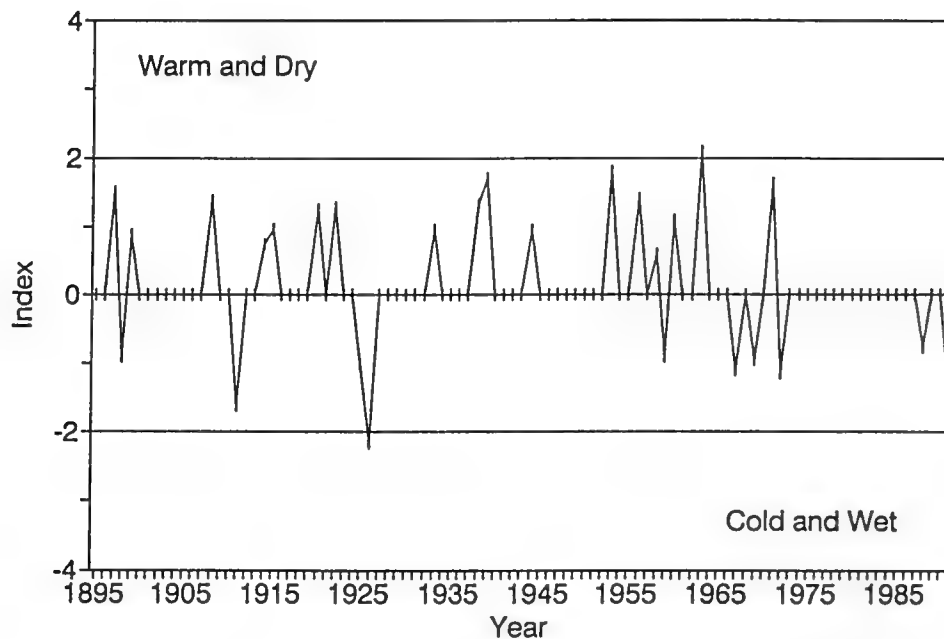
Figure 54. Identification of statewide warm and dry, and cold and wet autumns (1895-1991).
See text for definition of index.
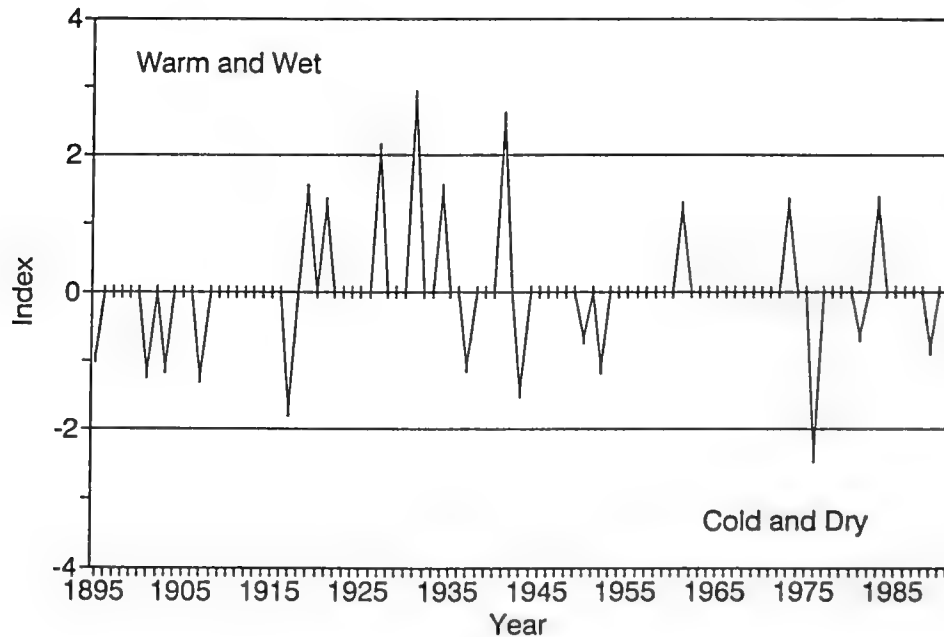
Figure 55. Identification of statewide warm and wet, and cold and dry autumns (1895-1991).
See text for definition of index.

Table 10. Statewide Autumn Seasonal Temperature and Precipitation Extremes

	Temperature		Precipitation	
Year	Fahrenheit	Deviation	Inches	Percent deviation
1911	52.5	-2.0	14.99	166
1917	50.7	-3.8	6.19	68
1926	52.6	-1.9	17.28	191
1927	57.7	+3.2	13.62	150
1931	60.3	+5.8	13.46	149
1938	57.0	+2.5	6.72	74
1939	56.4	+1.9	4.26	47
1941	56.9	+2.4	17.08	189
1953	56.7	+2.2	4.05	45
1963	58.5	+4.0	4.66	51
1971	57.9	+3.4	6.66	74
1976	48.6	-5.9	6.05	67

Note: Deviation and percent deviation calculated from the 97-year statewide autumn season average.

Figure 56. Identification of regional (north, central, and south) warm and dry, and cold and wet summers (1895-1991). See text for definition of index.

Figure 57. Identification of regional (north, central, and south) warm and wet, and cold and dry summers (1895-1991). See text for definition of index.
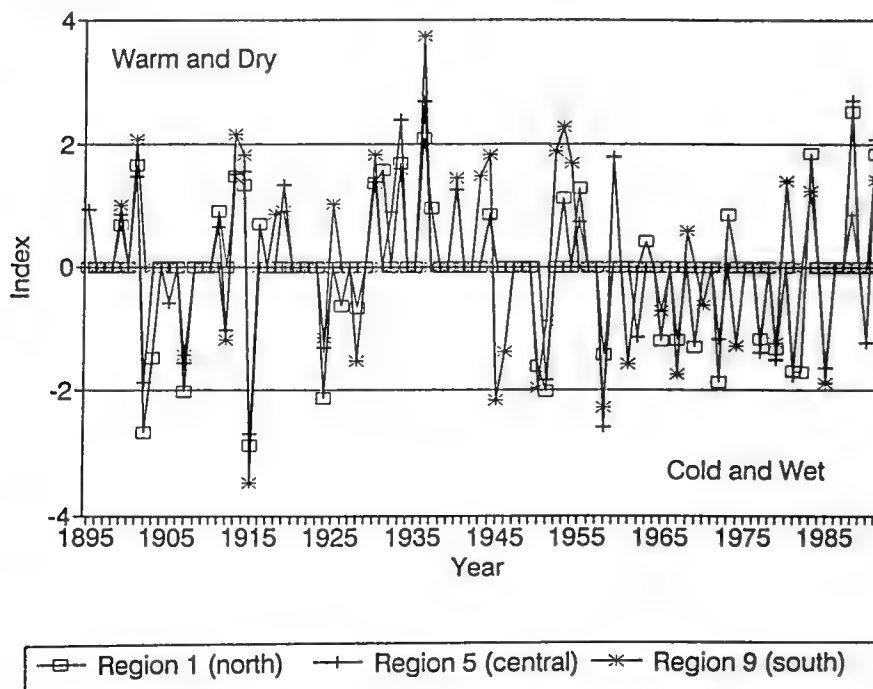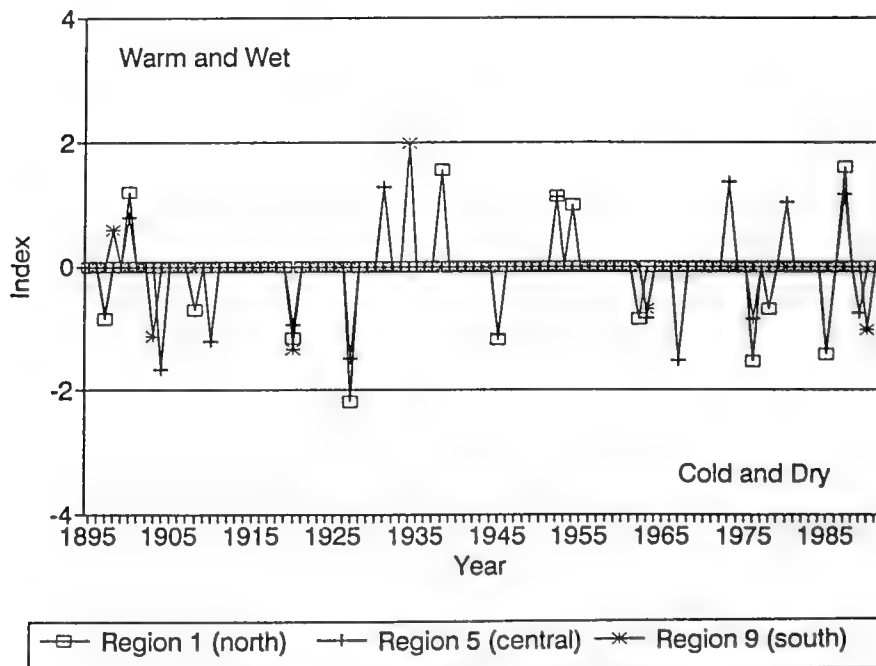
than the changes that can occur from one year or a few years to the next. With that being said, however, some identifiable longer-term features can be noted.

The most persistent and extreme summertime high temperatures occurred primarily during the 1930s, the Dust Bowl era. Indeed, average temperatures in Illinois clearly increased by 4 to 5°F from the mid- to late 1800s to the 1930s, and then cooled by about half that amount to the present. All of the temperature parameters assessed herein support those trends, including average temperatures and frequency of days with extreme temperatures, whether warm or cold. These trends are found in records of the continent, the hemisphere, and the world, although the magnitudes of change decrease with increasing areas of integration.

During the peak of Illinois temperatures during the 1930s, the frequency and intensity of hot days was unprecedented in Illinois and much of the upper Midwest. The many severe impacts of the heat and dryness of that period are well known. Since then, the frequency and intensity of summertime heat has been substantially less, particularly during the 1960s and 1970s. However, some resurgence of extreme summertime heat was noted during the 1980s, with accompanying impacts on crop yields. The extent of this trend toward higher frequencies of hot and dry summers cannot be predicted at this time.

In general, the frequency of extreme cold events increased from 1930 to the present. This upward trend reached its peak in the late 1970s and early 1980s, with an unprecedented string of extremely cold winters. They had major negative impacts on the Illinois economy and environment, affecting transportation, health, energy consumption, etc. During this cooling episode, precipitation may have increased marginally, but the major change in precipitation was the increase in annual snowfall, peaking in the late 1970s. Also notable was the abrupt decline in the annual number of severe winter storms in the state, dropping from an average of five per year prior to about 1980, to about three during recent years. The 1992-1993 winter was perceived by many to be a severe winter, but it was only near-average when compared to the situation of the last two or three decades.

Generally benign, moderate summers occurred during the 1960s and 1970s. As noted above, the 1980s were characterized by more frequent hot and dry but quite variable summers, similar to the earlier half of the century. It should be noted that midwestern agriculture experienced rapid technological advances during this relatively benign period. In addition, virtually all farmers active today have spent their entire careers during that benign period. The severity of the 1988 growing-season drought in Illinois had only been equaled in two other years since the turn of the century. The most recent Illinois drought of equal intensity occurred in 1936, long before virtually all present-day farmers were in the business.

The impact of climate variability in the state is exemplified by the cool summer of 1992. Even though overall corn yields were the highest ever in the United States, the northernmost 100 miles of the Corn Belt accumulated so few GDDs (sixth fewest since 1878) that the corn crop did not mature at its usual rate in that area. Indeed, it was not yet ripe at the time of the first frost in southern Minnesota and Wisconsin, which was within about one week of the long-term average.

The degree of cold of the 1992 summer was also demonstrated by the low total CDDs for the state. Total CDDs in 1992 were the seventh lowest since 1878. This was a boon for the consumer and a bane to the electric utility.

The tornado frequency record in Illinois exhibits no trends, rather an average of some 25 tornadoes per year, but varying from 6 to 107 per year! Of interest was the relatively high frequency recorded during the early 1880s (essentially equal to that of today), even though the tornado record for that time is known to be incomplete.

Cloud cover and the frequency of ice/glaze storms have both increased in the state. These changes do not appear to be due to changes in observing procedure, nor are they related to other parameters that might assist in their explanation.

It is clear that climate has not been stable in Illinois during the last 150 years, nor should it have been so anticipated. Many of the changes, although abrupt, were of relatively small magnitude. Yet those relatively small-scale changes levied a substantial toll on the inhabitants of the state through discomfort; lost income; increased costs; and impediments to commerce and agricultural production, in spite of major strides in hybridization and field management practices. Since climate has always changed, it will likely continue to do so. The past gives little hint as to the future direction and magnitude of these changes, but it does suggest limits within which climate may be constrained over the immediate future. The current increase to atmospheric carbon dioxide by the burning of fossil fuels, however, is suspected to change some

climate parameters at a faster rate than during the past centuries, thereby possibly rendering past climate a poor tool for evaluating future variability.

ACKNOWLEDGMENTS

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APPENDIX A

The following tables present the results of the trend analysis for individual long-term stations. The values given are the magnitude of the trend for those stations where the trend is statistically significant at the 10 percent level. If the trend is not statistically significant, an "NT" is shown.

Table A.1. Trends in Annual Precipitation Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Total precipitation (inches/decade)	Number of days with measurable precipitation (days/decade)	Number of days with precipitation > one inch (days/decade)	Total Snowfall (inches/ decade)	Number of days with measurable snowfall (days/decade)
Northwest					
Aledo	0.11	-0.59	0.01	0.97*	0.16
Dixon 1 NW	0.21	0.93	0.07	1.49*	0.73*
Galva	0.64*	0.73	0.35*	-0.29	-0.38*
Mount Carroll	0.28	1.36*	0.07	0.52	-0.06
Walnut	27	-3.05*	0.03	0.70*	-0.74*
Northeast					
Aurora	0.26	4.12*	0.07	0.38	-1.23*
Marengo	0.61*	0.25	0.22*	1.41*	-0.49*
Ottawa 4 SW	0.80*	1.87*	0.38*	0.40	0.37
West					
La Harpe	0.37	1.54*	0.14	-0.10	-0.10
Monmouth	0.27	0.54	0.10	0.36	0.00
Rushville	0.97*	0.58	0.43*	0.08	-0.04
Central					
Decatur	0.11	0.22	0.01	-0.08	-0.32
Lincoln	0.14	0.28	0.15	-0.16	-0.23
Minonk	0.55*	1.44*	0.17	0.61	0.41*
East					
Danville	1.12*	4.18*	0.21	0.62	0.26
Hoopeston	-0.01	0.63	-0.07	-1.50*	0.23
Pontiac	0.27	-0.55	0.15	-0.86*	0.04
Urbana	0.74*	2.32*	0.25*	0.72*	1.11*
West-Southwest					
Carlinville	0.02	1.03*	-0.12	0.04	0.03
Greenville 1 E	-0.16	-0.30	0.06	-0.37	-0.56*
Griggsville	0.16	2.63*	0.03	1.92*	0.83*
Hillsboro	0.27	-0.67	0.24*	-0.36	-0.48*
Jacksonville	0.62*	-0.60	0.37*	0.41	-0.73*
Pana	0.40	3.12*	0.14	1.32*	1.07*
White Hall 1 E	-0.14	1.89*	-0.30*	-0.73*	-0.50*
East-Southeast					
Charleston	0.28	-0.49	0.14	0.20	-0.43*
Effingham	0.45	1.84*	0.14	1.16*	0.47*
Flora	0.58*	1.07	0.27*	-0.90*	-0.59*
Olney	0.09	0.83	0.15	-1.06*	-0.72*
Palestine	0.15	1.85*	0.01	0.94*	0.62*
Paris Waterworks	0.72*	4.69*	0.10	2.03*	1.07*
Windsor	-0.28	0.54	-0.12	0.08	-0.32

Table A.1. Concluded

Southwest

Anna 1 E	0.51	2.23*	-0.02	0.83*	0.77*
Carbondale	-0.68	-2.15*	-0.27	-0.61	0.66*
Du Quoin 4 SE	0.32	1.18*	0.10	-0.32	-0.10
Olive Branch	0.13	2.06*	0.07	0.21	-0.01
Sparta	0.20	-0.66	0.21	-0.19	0.01
		4.12*	0.25	0.28	0.13

Southeast

Fairfield	0.91*	0.99	-0.13	-0.43	-0.26
Harrisburg	-0.06	2.37*	0.02	0.11	0.17
McLeansboro	0.46	-0.07	0.09	0.17	-0.41
Mt. Vernon	0.16				
All stations	0.35*	1.19*	0.11	0.37	0.12

Table A.2. Trends in Annual Temperature Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean temperature (°F/decade)	Number of days with mean temperature > 50°F (days/decade)	Highest daily maximum temperature (°F/decade)	Lowest daily minimum temperature (°F/decade)
Northwest				
Aledo	-0.23	-1.24	NT	0.10
Dixon	-0.01	0.52	NT	0.11
Galva	-0.02	0.37	-0.36*	0.32
Mount Carroll	-0.02	-0.06	NT	0.09
Walnut	-0.29*	09.19	-0.29*	0.08
Northeast				
Aurora	-0.22*	-0.89	NT	-0.15
Marengo	0.03	0.28	NT	-0.20
Ottawa 4 SW	0.53*	2.89*	-0.31*	-0.25
West				
La Harpe	-0.08	0.26	NT	0.17
Monmouth	0.06	0.84*	-0.34*	0.25
Rushville	0.07	0.02	NT	-0.03
Central				
Decatur	-0.29*	-0.44	-0.27*	-0.01
Lincoln	-0.21	0.03	-0.32*	0.54*
Minonk	-0.31	-1.35	-0.53*	0.22
East				
Danville	0.49*	2.68*	-0.58*	-0.04
Hoopeston	0.44*	2.58*	-0.33*	0.06
Pontiac	-0.25*	-0.59	-0.40*	-0.07
Urbana	0.06	0.70*	NT	0.40*
West-Southwest				
Carlinville	-0.09	0.41	-0.41*	0.10
Greenville 1 E	-0.11	-2.00	NT	0.18
Griggsville	-0.15	-0.68	NT	0.04
Hillsboro	-0.07	0.11	NT	0.17
Jacksonville	-0.16*	-0.54	-0.36*	-0.03
Pana	0.11	1.25*	-0.42*	0.17
White Hall 1 E	0.25*	1.95*	-0.38*	-0.01
East-Southeast				
Charleston	0.33	1.92	-0.35*	0.26
Effingham	0.17	1.70*	-0.57*	-0.04
Flora	0.34	2.76*	NT	0.07
Olney	-0.24	-0.20	-0.45*	0.08
Palestine	-0.15	0.36	NT	-0.10
Paris Waterworks	0.19*	2.00*	-0.31*	0.32*
Windsor	0.02	1.04*	-0.34*	0.31

Table A.2. Concluded

Southwest

Anna 1 E	-0.09	0.43	-0.43*	-0.14
Carbondale	-1.06*	-4.94*	-0.96*	0.07
Du Quoin 4 SE	-0.19	-0.54	-0.29*	-0.05
Olive Branch	-0.25*	-0.87	-0.46*	-0.07
Sparta	0.02	0.90*	NT	-0.04

Southeast

Fairfield	0.04	0.93	0.52*	-0.05
Harrisburg	-0.34*	-0.11	-0.37*	-0.10
McLeansboro	-0.14*	-0.23	-0.42*	-0.05
Mt. Vernon	-0.13	-0.31	-0.41*	0.20
All stations	-0.06	0.26	-0.35*	0.15

Table A.3. Trends in Annual Temperature Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean minimum temperature (°F/decade)	Number of days with minimum temperature > 70°F (days/decade)	Number of days with minimum temperature < 0°F (days/decade)	Number of days with minimum temperature <32°F (days/decade)
Northwest				
Aledo	-0.17	-0.06	0.14	-0.85*
Dixon 1 NW	0.04	0.22	0.66*	0.79
Galva	0.05	0.24	0.44	-0.54
Mount Carroll	-0.06	-0.15	0.43	0.73
Walnut	-0.32*	-0.52*	0.78*	1.23*
Northeast				
Aurora	-0.14	-0.01	0.21	-1.10*
Marengo	-0.18	0.01	0.77*	1.73*
Ottawa 4 SW	0.57*	0.78*	0.33	-0.49
West				
La Harpe	-0.13*	-0.15	0.43	0.55
Monmouth	0.11	0.20	0.12	-0.83*
Rushville	-0.04	-0.45	0.28	0.46
Central				
Decatur	-0.22*	-0.02	0.18	-1.12*
Lincoln	-0.10	-0.05	0.23	-1.31*
Minonk	-0.24	-0.21	0.45	-0.09
East				
Danville	0.45*	-0.02	0.34	1.07
Hoopeston	0.45*	0.41*	0.41	0.72
Pontiac	-0.17*	-0.32	0.39	0.40
Urbana	0.08	-0.14	-0.04	-0.75*
West-Southwest				
Carlinville	0.06	0.65*	0.03	-1.00*
Greenville 1 E	0.01	0.86	-0.74*	-2.58*
Griggsville	-0.13	-0.41	0.14*	0.60
Hillsboro	-0.06	-0.23	0.06	-0.16
Jacksonville	-0.11*	-0.28	0.30	0.57
Pana	0.16*	0.09	0.05	-0.55
White Hall 1 E	0.31*	0.53	0.38	0.57
East-Southeast				
Charleston	0.39*	0.82*	0.10	0.76
Effingham	0.23	0.27	0.57*	1.20
Flora	0.28	0.24	0.20	0.26
Palestine	-0.04	0.60	0.20	-0.50
Olney	-0.16	-0.09	0.19	-0.46
Paris Waterworks	0.31*	0.92*	0.20	-1.23*
Windsor	0.18*	0.69*	0.00	-1.90*

Table A.3. Concluded

Southwest

Anna 1 E	-0.06	-0.71*	0.08	0.61
Carbondale	-0.91*	-1.93*	0.28	1.58*
Du Quoin 4 SE	-0.10	0.24	0.25	-0.14
Olive Branch	-0.15*	-0.09	0.28*	0.35
Sparta	0.10**	1.04*	-0.10	-0.76

Southeast

Fairfield	0.13	-0.35	0.19	0.91*
Harrisburg	-0.30*	-0.45	0.06	0.41
McLeansboro	-0.15*	-0.99*	0.16	1.36*
Mt. Vernon	-0.22*	-0.17	0.84*	0.78*
All stations	-0.02	0.00	0.26	0.01

Table A.4a. Trends in Annual Temperature Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean maximum temperature (°F/year)	Number of days with maximum temperature >86°F (days/year)	Number of days with maximum temperature >90°F (days/year)	Number of days with temperatures >100°F (days/year)
Northwest				
Aledo	-0.11	0.25	-0.20	-0.16
Dixon 1 NW	-0.08	-1.07*	-0.93*	-0.12
Galva	-0.09	-1.43*	-1.43*	-0.21*
Mount Carroll	0.00	0.03	-04.1	-0.16
Walnut	-0.27*	-0.61	-1.19*	-0.11
Northeast				
Aurora	-0.40*	-0.17	-0.60	-0.10
Marengo	0.22	0.87	0.37	-0.07
Ottawa 4 SW	0.40*	-0.72	-1.19*	-0.34*
West				
La Harpe	0.00	-0.12	-0.94	-0.18
Monmouth	0.01	-0.97	-1.53*	-0.23
Rushville	0.17	0.89	0.06	-0.13
Central				
Decatur	-0.37*	-0.86	-1.41*	-0.26
Lincoln	-0.29	-1.54*	-1.99*	-0.16
Minonk	-0.38	-1.07	-1.44*	-0.27*
East				
Danville	0.52	-1.19	-2.14*	-0.34*
Hoopeston	0.44*	-0.97	-1.44*	-0.22*
Pontiac	-0.33*	-2.34*	-2.57*	-0.27*
Urbana	0.03	0.28	-0.18	-0.03
West-Southwest				
Carlinville	-0.24*	-1.24*	-1.78*	-0.52*
Greenville 1 E	-0.22	-0.84	-1.32	-0.45
Griggsville	-0.23	-1.55*	-1.89*	-0.24
Hillsboro	-1.10	-0.62	-1.29	-0.31
Jacksonville	-0.21*	-1.84*	-1.98*	-0.35*
Pana	0.01	-1.46*	-1.64*	-0.23
White Hall 1 E	0.15	-1.16	-1.85*	-0.43*

Table A.4a. Concluded

East-Southeast

Charleston	0.26	-1.93*	-2.35*	-0.40*
Effingham	0.20	-2.31*	-2.51*	-0.66*
Flora	0.46	0.81	-0.14	-0.38
Olney	-0.30	-1.07	-1.71*	-0.41*
Palestine	-0.26*	-1.08	-1.25	-0.15
Paris Waterworks	0.08	-1.48*	-1.54*	-0.24
Windsor	-0.13*	-1.44*	-2.32*	-0.37*

Southwest

Anna 1 E	-0.11	-1.63*	-1.95*	-0.42*
Carbondale	-1.20*	-4.11*	-4.68*	-1.12*
Du Quoin 4 SE	-0.29*	-2.21*	-2.32*	-0.36
Olive Branch	-0.34*	-2.45*	-3.01*	-0.61*
Sparta	-0.07	0.04	-0.73	-0.50*

Southeast

Fairfield	-0.19	-3.13*	-3.59*	-0.52*
Harrisburg	-0.39*	-0.70	-1.57*	-0.58*
McLeansboro	-0.14	-1.12	-1.51*	-0.57*
Mt. Vernon	-0.05	-1.61*	-1.42*	-0.20*
All stations	-0.10	-1.10*	-1.55*	-0.33*

Table A.4b. Trends in Annual Temperature Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Number of days with maximum temperature < 0°F	Number of days with maximum temperature < 32°F
Northwest		
Aledo	0.02	0.38
Dixon 1 NW	0.06	1.33*
Galva	0.07*	0.62
Mount Carroll	0.05	0.61
Walnut	0.04	0.62
Northeast		
Aurora	0.03	-0.40
Marengo	0.04	-0.48
Ottawa 4 SW	0.06*	0.75*
West		
La Harpe	0.01	0.39
Monmouth	0.00	-0.21
Rushville	0.01	0.47
Central		
Decatur	0.02*	-0.05
Lincoln	0.01	0.53
Minonk	0.07*	0.85
East		
Danville	0.02*	0.81*
Hoopeston	0.04*	1.30*
Pontiac	0.03	0.87*
Urbana	0.04*	0.45
West-Southwest		
Carlinville	0.01	0.67*
Greenville 1 E	-0.01	-1.93*
Griggsville	0.02	1.10*
Hillsboro	0.01*	0.12
Jacksonville	0.04*	0.93*
Pana	0.02*	0.59
White Hall 1 E	0.01	0.83*
East-Southeast		
Charleston	0.03*	0.45
Effingham	0.04*	1.81*
Flora	0.01*	0.65*
Olney	0.01	0.55
Palestine	0.01	0.69*
Paris Waterworks	0.03*	0.07
Windsor	0.03*	0.76*

Table A.4b. Concluded

Southwest

Anna 1 E	0.00	0.53*
Carbondale	0.01	0.74*
Du Quoin 4 SE	0.01	0.56*
Olive Branch	0.02*	0.64*
Sparta	0.01	0.32

Southeast

Fairfield	0.02*	1.07*
Harrisburg	0.00	0.32
McLeansboro	0.01*	0.88*
Mt. Vernon	0.07	0.51
All stations	0.03*	0.53

Table A.5. Trends in Autumn Climate Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean temperature (°F/decade)	Mean minimum temperature (°F/decade)	Mean maximum temperature (°F/decade)	Total precipitation (inches/decade)
Northwest				
Aledo	-0.31*	-0.27*	-0.22	-0.04
Dixon 1 NW	-0.22*	-0.18	-0.26*	0.02
Galva	-0.15*	-0.06	-0.23*	0.16
Mount Carroll	-0.14	-0.18**	-0.15	0.04
Walnut	-0.19*	-0.30*	-0.08	0.03
Northeast				
Aurora	-0.16	-0.10	-0.50*	0.02
Marengo	0.02	-0.27*	0.31	0.12
Ottawa 4 SW	0.38	0.43*	0.47	0.21
West				
La Harpe	-0.16*	-0.20*	-0.11	0.23
Monmouth	0.00	0.06	-0.06	0.02
Rushville	0.14	0.00	0.24	0.34*
Central				
Decatur	-0.24	-0.16	-0.32	-0.06
Lincoln	0.27	0.27*	0.35	0.00
Minonk	-0.24	-0.23	-0.25	0.27*
East				
Danville	0.11	0.20	0.03	0.13
Hoopeston	0.50*	0.50*	0.29	0.09
Pontiac	-0.17*	-0.14	-0.21*	0.07
Urbana	0.02	0.05	0.00	0.19
West-Southwest				
Carlinville	-0.11	0.05	-0.28	0.02
Greenville 1 E	0.02	0.06	-0.01	0.12
Griggsville	-0.21*	-0.19*	-0.22*	-0.06
Hillsboro	-0.14	-0.10	-0.27	0.09
Jacksonville	-0.18*	-0.16*	-0.21*	0.13
Pana	0.04	0.11	-0.20*	0.12
White Hall 1 E	0.23	0.30*	0.12	-0.18
East-Southeast				
Charleston	0.44	0.51*	0.38	0.14
Effingham	-0.17*	0.00	-0.35*	0.06
Flora	0.27	0.24	0.31	0.13
Olney	0.02	0.07	-0.03	0.11
Palestine	-0.14	-0.04	-0.25*	0.12
Paris Waterworks	0.24	0.39*	0.10	0.08
Windsor	-0.04	0.17	-0.26	0.00

Table A.5. Concluded

Southwest

Anna 1 E	-0.25*	-0.17*	-0.29*	0.02
Carbondale	-0.80*	-0.77*	-0.82*	-0.29
Du Quoin 4 SE	-0.18*	-0.06	-0.30*	0.23
Olive Branch	-0.30*	-0.15	-0.45*	0.18
Sparta	-0.03	0.10	-0.15	0.14

Southeast

Fairfield	-0.26	-0.04	-0.48*	0.25*
Harrisburg	-0.30	-0.23	-0.37	0.02
McLeansboro	-0.16	-0.18*	-0.15	0.16
Mt. Vernon	-0.32*	-0.46*	-0.08	-0.03
All stations	-0.11	-0.06	-0.15*	0.10

Table A.6. Trends in Winter Climate Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean temperature (°F/decade)	Mean minimum temperature (°F/decade)	Mean maximum temperature (°F/decade)	Total precipitation (inches/decade)
Northwest				
Aledo	-0.06	0.00	-0.11	-0.02
Dixon 1 NW	0.08	0.03	0.13	0.09
Galva	-0.20	-0.14	-0.22	0.08
Mount Carroll	-0.02	-0.09	-0.01	0.03
Walnut	-0.31*	-0.42*	-0.21	-0.06
Northeast				
Aurora	-0.16	-0.17	-0.06	0.06
Marengo	-0.04	-0.26	0.18	0.05
Ottawa 4 SW	0.10	0.09	0.11	0.05
West				
La Harpe	-0.18	-0.26*	-0.10	-0.03
Monmouth	0.06	0.08	0.05	0.07
Rushville	-0.20	-0.24*	-0.16	0.13
Central				
Decatur	-0.21	-0.18	-0.24	0.04
Lincoln	-0.76*	-0.65*	-0.87*	-0.07
Minonk	-0.18	-0.15	-0.21	0.15
East				
Danville	-0.06	-0.05	-0.06	0.11
Hoopeston	0.16	0.14	0.18	-0.16
Pontiac	-0.43*	-0.39*	-0.47*	-0.12
Urbana	-0.09	-0.04	-0.14	0.10
West-Southwest				
Carlinville	-0.16	-0.03	-0.29	0.02
Greenville 1 E	0.37	0.41	0.34	-0.06
Griggsville	-0.30*	-0.27*	-0.34*	0.10
Hillsboro	-0.06	-0.12	0.01	0.10
Jacksonville	-0.19	-0.20	-0.23	0.07
Pana	-0.04	0.01	-0.10	0.16
White Hall 1 E	-0.09	-0.11	-0.08	-0.04
East-Southeast				
Charleston	-0.07	-0.06	-0.08	0.05
Effingham	-0.09	-0.15	-0.03	0.21
Flora	0.15	0.12	0.19	0.15
Olney	-0.13	-0.10	-0.22	0.02
Palestine	0.01	0.05	-0.03	-0.05
Paris Waterworks	0.05	0.11	-0.01	0.17
Windsor	-0.15	-0.03	-0.27*	0.01

Table A.6. Concluded

Southwest

Anna 1 E	-0.18	-0.17	-0.14	0.14
Carbondale	-0.82*	-0.84*	-0.80*	0.00
Du Quoin 4 SE	-0.25*	-0.22	-0.29*	0.03
Olive Branch	-0.23	-0.23	-0.22	-0.07
Sparta	-0.21	-0.12	-0.31	-0.03

Southeast

Fairfield	-0.10	-0.03	-0.17	0.10
Harrisburg	-0.41*	-0.39*	-0.43*	-0.13
McLeansboro	-0.17	-0.22	-0.11	-0.07
Mt. Vernon	-0.08	-0.21	0.10	0.05
All stations	-0.14	-0.14	-0.14	0.04

Table A.7. Trends in Spring Climate Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean temperature ($^{\circ}F$/decade)	Mean minimum temperature ($^{\circ}F$/decade)	Mean maximum temperature ($^{\circ}F$/decade)	Total precipitation (inches/decade)
Northwest				
Aledo	0.04	0.05	0.14	0.07
Dixon 1 NW	0.06	0.15	-0.12	0.05
Galva	0.08	0.12	0.04	0.18
Mount Carroll	-0.08	-0.04	-0.18	0.12
Walnut	-0.03	-0.05	-0.01	0.24*
Northeast				
Aurora	0.03	0.08	-0.06	0.12
Marengo	0.01	-0.19*	0.21	0.10
Ottawa 4 SW	0.26*	0.37*	0.16	0.22*
West				
La Harpe	0.01	-0.05	0.07	0.13
Monmouth	0.15	0.17*	0.13	0.15
Rushville	-0.12	-0.18	-0.06	0.24
Central				
Decatur	0.14	0.11	0.16	0.03
Lincoln	0.30	0.29*	0.31	0.03
Minonk	-0.20	-0.10	-0.30	0.24
East				
Danville	0.12	0.11	0.13	0.14
Hoopeston	0.08	0.19*	-0.03	-0.03
Pontiac	-0.11	-0.07	-0.16	-0.05
Urbana	0.19*	0.19*	0.19*	0.05
West-Southwest				
Carlinville	-0.03	0.08	-0.14	0.03
Greenville 1 E	-0.13	-0.08	-0.18	-0.05
Griggsville	-0.13	-0.09	-0.17	0.08
Hillsboro	0.06	0.04	0.08	0.06
Jacksonville	-0.09	-0.04	-0.15	0.28*
Pana	0.19*	0.21*	0.18	0.11
East-Southeast				
Charleston	0.21	0.26*	0.17	-0.01
Effingham	-0.08	0.00	0.03	0.05
Flora	0.26	0.19	0.44	0.16
Olney	-0.41*	-0.33	-0.49*	0.07
Palestine	0.24	0.31*	0.07	0.13
Paris Waterworks	0.09	0.19	0.00	0.05
White Hall 1 E	0.21	0.28*	0.13	0.12
Windsor	0.21*	0.30*	0.16	-0.23

Table A.7. Concluded

Southwest

Anna 1 E	0.06	0.04	0.17	0.32
Carbondale	-0.62*	-0.53*	-0.71*	-0.03
Du Quoin 4 SE	0.12	0.15	0.09	0.24
Olive Branch	-0.18	-0.09	-0.23*	0.10
Sparta	0.23*	0.23*	0.23	0.06

Southeast

Fairfield	0.01	0.09	-0.09	0.45*
Harrisburg	0.21	0.10	0.24	0.27
McLeansboro	-0.09	-0.06	-0.12	0.27
Mt. Vernon	-0.02	-0.13	0.08	0.19*
All stations	0.04	0.06	0.02	0.13

Table A.8. Trends in Summer Climate Parameters (Asterisk Indicates Trends of Statistical Significance)

City	Mean temperature (°F/decade)	Mean minimum temperature (°F/decade)	Mean maximum temperature (°F/decade)	Total precipitation (inches/decade)
Northwest				
Aledo	-0.21	-0.17	-0.24	0.10
Dixon 1 NW	0.04	0.16*	-0.08	0.05
Galva	0.18	0.29*	0.-06	0.22
Mount Carroll	-0.22	-0.20	-0.09	0.00
Walnut	-0.36*	-0.29*	-0.42*	0.12
Northeast				
Aurora	-0.27	-0.12	-0.42*	0.11
Marengo	0.00	-0.11	0.02	0.32*
Ottawa 4 SW	0.64*	0.78*	0.20	0.22
West				
La Harpe	0.02	-0.01	0.14	0.14
Monmouth	0.01	0.12*	-0.09	0.05
Rushville	-0.27	-0.36*	-0.18	0.16
Central				
Decatur	-0.53*	-0.39*	-0.66*	0.17
Lincoln	-0.40*	-0.14	-0.65*	0.25
Minonk	0.16	0.18	0.15	0.03
East				
Danville	0.59*	0.65*	0.52	0.53*
Hoopeston	0.31*	0.42*	0.19	0.01
Pontiac	-0.29*	-0.07	-0.50*	0.37*
Urbana	0.10	0.14*	0.07	0.40*
West-Southwest				
Carlinville	-0.04	0.15	-0.22	-0.06
Greenville 1 E	-0.55	-0.27	-0.83	-0.13
Griggsville	0.03	0.04	-0.22	0.02
Hillsboro	-0.15	-0.06	-0.23	0.03
Jacksonville	-0.15*	-0.05	-0.26*	0.14
Pana	0.22	0.29	0.15	0.02
White Hall 1 E	0.63*	0.75*	0.43	-0.04
East-Southeast				
Charleston	0.16	0.40*	-0.08	0.01
Effingham	0.37	0.55*	0.39	0.01
Flora	0.49	0.39	0.67	0.09
Olney	-0.34	-0.24	-0.34	-0.06
Palestine	-0.34	-0.20	-0.42	0.02
Paris Waterworks	0.38*	0.54*	0.23	0.36*
Windsor	0.05	0.27*	-0.17	-0.06

Table A.8. Concluded

Southwest

Anna 1 E	0.03	0.06	-0.19*	0.03
Carbondale	-0.35*	-0.20*	-0.49*	-0.03
Du Quoin 4 SE	-0.18	-0.04	-0.32	-0.14
Olive Branch	-0.28*	-0.13	-0.44*	-0.08
Sparta	0.08	0.20*	-0.04	0.03

Southeast

Fairfield	-0.03	0.10	-0.26	0.09
Harrisburg	-0.43*	-0.35	-0.51*	-0.11
McLeansboro	-0.15*	-0.13*	-0.18	0.08
Mt. Vernon	-0.10	-0.80	-0.30	-0.04
All stations	-0.03	0.06	-0.13	0.08

AIR QUALITY TRENDS IN ILLINOIS

Donald F. Gatz
Illinois State Water Survey

INTRODUCTION

Purpose

There are three main purposes for including a component on air quality in a comprehensive assessment of the Illinois environment. The first purpose is simply to document the current status of Illinois air quality and any recent trends. Specifically, we will look for changes (trends) in pollutant concentrations over time within specific geographic regions, as well as state-wide, and also show spatial variations of pollutant concentrations in the Chicago area, the only area of the state where there are enough sampling sites to permit plots of spatial variability.

These analyses will reveal whether concentrations of specific pollutants in Illinois air are getting better, or getting worse, or staying the same. They may also indicate *which areas of the state* experience the highest and lowest pollutant concentrations, and whether such local or regional conditions are improving or worsening.

The second purpose is to provide the information necessary for assessments of human and ecosystem exposure to airborne pollutants. These assessments will appear in other reports in this series.

The final purpose is to identify gaps and needs in Illinois air quality monitoring and research.

Scope

This analysis, for the most part, relies on data generated by routine air quality measurements carried out by the Illinois Environmental Protection Agency (IEPA), the state agency charged with monitoring compliance with state and national air quality standards. It should

be noted that IEPA's main purpose in making these measurements is to monitor compliance, *not* to document the state's air quality. Thus, to some extent, this examination of IEPA data for descriptions of past and current air quality goes beyond the purpose for which the data were originally collected. Consequently, the goal of characterizing air quality over the whole state may not be completely attainable.

Pollutants Analyzed

The air pollutants to be examined for temporal and spatial trends include, first of all, the seven criteria pollutants, i.e., those for which national or state air quality standards have been set. These include four gaseous pollutants: ozone (O_3), sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and carbon monoxide (CO). There are also standards for three pollutants that occur as particles: These are lead (Pb); total suspended particulate matter (TSP), for which there is only a state standard; and particulate matter with aerodynamic diameters of 10 micrometers (μm) or less (PM_{10}), for which there is only a national standard.

In addition, we will examine measured concentrations of several additional pollutants for which standards have not been set. This group includes sulfate and nitrate ions and the metals arsenic (As), cadmium (Cd), chromium (Cr), iron (Fe), manganese (Mn), and nickel (Ni). These pollutants all occur as particles in the atmosphere, and they are measured by chemically analyzing the same filters used to collect the TSP samples. Several additional metals, such as beryllium, copper, selenium, and vanadium, are currently being measured or have been measured in the past by IEPA. These additional metals are not included in this assessment because their concentrations are mostly smaller than the detection limit.

Finally, we will summarize occasional (nonroutine) measurements of organic compounds. These measurements were made by IEPA or published in the scientific literature.

Period of Record

The Illinois data to be analyzed represent, at the most, the years between 1978 and 1990. Some pollutants were measured only during the later years of this time period.

DATA QUALITY

Methods of air pollutant sampling and analysis used by IEPA conform to U.S. EPA standards. However, it should be noted that artifacts can occur on certain types of filters possibly used by IEPA for high volume sampling during the data period. Positive sulfate artifacts (formation of sulfate on the filter from gaseous precursors) have been reported (Appel et al., 1984). Both positive and negative (evaporative loss) artifacts can occur for nitrate (ibid.).

METHODS

Data Source

The data used in this report were taken from annual summary reports published by IEPA for the years 1978-1990 (IEPA, 1979-1991). Questionable values were checked with IEPA personnel (Swinford, personal communications, 1992 and 1993) and corrected if necessary.

Creation of Computer Files

Data were entered by hand into computer spreadsheet and database files from IEPA reports. Sampling site locations were specified by latitude and longitude. Spreadsheet data files were also converted to Geographic Information System (GIS) files using existing data conversion software. Site locations in the Chicago area were verified by comparison of locations plotted on GIS maps against road atlas maps, and corrected when necessary.

Statistical Methods

Box Plots. Box plots were used to convey information about the distribution of a particular pollutant and its time-averaged concentrations (e.g., annual mean, highest 24-hr average, or highest 1-hr average) over sampling sites in a specific geographical region for a specific year. For a given year, a box plot shows the 10th, 25th, 50th, 75th, and 90th percentiles, respectively, as the low "whisker", the bottom of the "box", the line across the box, the top of the box, and the upper whisker. Individual values outside the 10th and 90th percentiles were also plotted as individual points. Thus, a plot of a series of boxes representing a series of

years visually shows how various percentiles of the distribution of pollutant concentration change over time. In addition to showing plots to give a visual impression of time trends, we have assessed the statistical significance, or lack thereof, of changes in pollutant concentrations over time.

Trend Analysis. Time sequences of pollutant concentrations were tested for statistical significance of time trends *over the entire period of record* using the nonparametric Spearman Rank Correlation Coefficient as described by Snedecor and Cochran (1980). This is the same trend test used by IEPA to detect the statistical significance of time trends in pollutant concentrations. Please note that the critical values of the rank correlation coefficient for significance at various confidence levels given by IEPA (e.g., IEPA, 1991) differ somewhat from critical values given by tables A11(i) and A11(ii) in the appendices of Snedecor and Cochran (1980), which were used in this study. The test was applied to the series of medians of the annual distributions over sites within each geographic area.

Where significant time trends were found, average annual percent changes were calculated by dividing the slope by the concentration computed for 1984 (the middle year of the 1978-1990 series). The 1984 concentration was computed from the linear regression line fitted to the time series of median regional concentrations.

Regional Differences. One-way analysis of variance was used to test for differences in mean pollutant concentrations between independent regions; i.e., the Chicago area, the Metro East area, and the remainder of the state. Any data sets that failed the Bartlett's test for homogeneity of variance were also analyzed by the distribution-free Kruskal-Wallace one-way analysis of variance. Differences between individual regions were evaluated from pairwise comparison probabilities computed from the Tukey HSD multiple comparisons procedure. These tests were all carried out using SYSTAT (Wilkinson, 1990).

TIME TRENDS
IN POLLUTANT CONCENTRATIONS

Trends in pollutant concentrations over time are the major emphasis of this part of the Air Resources volume. Table 1 presents computed median pollutant concentrations from the distribution of site values for each geographical region and year. Table 2 presents results of statistical tests for significance of time trend for all

Table 1. Air Pollutant Concentration Medians, from the Distribution of Individual Sampling Site Values, for Specific Geographic Regions and Years

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
1-hr maximum CO, ppm	1978	11.10	10.85	---[*]	12.25
	1979	12.10	12.10	---	16.00
	1980	12.75	12.95	---	12.75
	1981	16.10	12.65	---	18.40
	1982	13.50	10.40	---	14.50
	1983	14.15	14.40	---	12.50
	1984	14.00	13.00	---	14.85
	1985	9.95	9.65	---	10.30
	1986	8.90	8.45	---	11.30
	1987	9.75	8.80	---	11.60
	1988	9.30	9.10	---	11.35
	1989	8.60	7.30	---	12.90
	1990	9.50	8.90	---	9.95
8-hr maximum CO, ppm	1978	7.65	7.15	---	7.95
	1979	8.70	8.60	---	9.40
	1980	7.95	7.35	---	8.10
	1981	9.00	8.80	---	9.50
	1982	6.90	6.10	---	7.20
	1983	9.30	10.90	---	6.30
	1984	8.90	8.90	---	9.10
	1985	6.15	5.55	---	6.35
	1986	5.80	5.45	---	5.90
	1987	5.30	5.30	---	5.30
	1988	5.30	5.30	---	5.75
	1989	5.10	4.80	---	7.35
	1990	5.30	3.70	---	5.60
Annual mean Pb, $\mu g/M^3$	1979	0.430	0.425	0.650	0.340
	1980	0.320	0.370	0.390	0.225
	1981	0.260	0.285	0.345	0.160
	1982	0.290	0.290	0.710	0.160
	1983	0.285	0.280	0.400	0.150
	1984	0.250	0.240	0.320	0.130
	1985	0.150	0.130	0.265	0.070
	1986	0.080	0.080	0.220	0.050
	1987	0.060	0.060	0.260	0.035
	1988	0.050	0.050	0.170	0.020
	1989	0.040	0.040	0.165	0.025
	1990	0.050	0.055	0.160	0.020

Note:

[*]--- Insufficient data at enough sampling sites to justify computing a median.

Table 1. (Continued)

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
1-hr maximum NO_2, ppm	1978	0.1760	0.2250	---	---
	1979	0.1380	0.1460	---	---
	1980	0.1480	0.1480	---	---
	1981	0.2015	0.2015	---	---
	1982	0.1320	0.1320	---	---
	1983	0.1320	0.1470	---	---
	1984	0.1160	0.1290	---	---
	1985	0.1330	0.1340	---	---
	1986	0.1060	0.0990	---	---
	1987	0.1150	0.1150	---	---
	1988	0.1255	0.1290	---	---
	1989	0.1125	0.1130	---	---
	1990	0.0885	0.0890	---	---
24-hr maximum NO_2, ppm	1978	0.0970	0.0970	---	---
	1979	0.0940	0.0950	---	---
	1980	0.0965	0.0965	---	---
	1981	0.0740	0.0740	---	---
	1982	0.0850	0.0850	---	---
	1983	0.0800	0.0900	---	---
	1984	0.0570	0.0570	---	---
	1985	0.0640	0.0650	---	---
	1986	0.0550	0.0560	---	---
	1987	0.0590	0.0595	---	---
	1988	0.0625	0.0650	---	---
	1989	0.0615	0.0630	---	---
	1990	0.0480	0.0495	---	---
Annual mean NO_2, ppm	1978	0.0450	0.0450	---	---
	1979	0.0510	0.0510	---	---
	1980	0.0470	0.0470	---	---
	1981	0.0420	0.0420	---	---
	1982	0.0385	0.0385	---	---
	1983	0.0290	0.0300	---	---
	1984	0.0280	0.0290	---	---
	1985	0.0270	0.0280	---	---
	1986	0.0240	0.0250	---	---
	1987	0.0240	0.0260	---	---
	1988	0.0230	0.0220	---	---
	1989	0.0270	0.0270	---	---
	1990	0.0240	0.0250	---	---

Note:

*--- Insufficient data at enough sampling sites to justify computing a median.

Table 1. (Continued)

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
1-hr maximum O₃, ppm	1978	0.1370	0.1510	0.1710	0.1175
	1979	0.1180	0.1215	0.1265	0.1030
	1980	0.1210	0.1255	0.1420	0.1050
	1981	0.1145	0.1415	0.1230	0.0980
	1982	0.1100	0.1170	0.1180	0.0940
	1983	0.1370	0.1580	0.1540	0.1065
	1984	0.1255	0.1270	0.1380	0.1035
	1985	0.1110	0.1145	0.1270	0.1005
	1986	0.1110	0.1110	0.1310	0.0985
	1987	0.1215	0.1390	0.1260	0.1060
	1988	0.1270	0.1275	0.1465	0.1130
	1989	0.1100	0.1130	0.1165	0.1030
	1990	0.0930	0.0930	0.1180	0.0900
3-hr maximum SO₂, ppm	1978	0.1460	0.1125	---	0.1815
	1979	0.1320	0.1310	---	0.1340
	1980	0.1355	0.1450	---	0.1170
	1981	0.1350	0.1150	---	0.1505
	1982	0.1240	0.1160	---	0.1450
	1983	0.0980	0.0900	---	0.1510
	1984	0.1195	0.0955	---	0.1515
	1985	0.1195	0.1090	---	0.1365
	1986	0.1210	0.1020	---	0.1990
	1987	0.1530	0.1310	---	0.1880
	1988	0.1360	0.0880	---	0.1860
	1989	0.1245	0.0870	---	0.2410
	1990	0.1115	0.0660	---	0.1340
24-hr maximum SO₂, ppm	1978	0.0495	0.0435	---	0.0720
	1979	0.0795	0.0820	---	0.0630
	1980	0.0465	0.0400	---	0.0600
	1981	0.0640	0.0650	---	0.0550
	1982	0.0560	0.0615	---	0.0530
	1983	0.0420	0.0410	---	0.0645
	1984	0.0530	0.0460	---	0.0670
	1985	0.0505	0.0450	---	0.0690
	1986	0.0505	0.0440	---	0.0850
	1987	0.0510	0.0440	---	0.0565
	1988	0.0515	0.0420	---	0.0850
	1989	0.0455	0.0310	---	0.0640
	1990	0.0400	0.0260	---	0.0480

Note:

*--- Insufficient data at enough sampling sites to justify computing a median.

Table 1. (Continued)

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
Annual mean SO_2, ppm	1978	0.0090	0.0075	---	0.0150
	1979	0.0120	0.0120	---	0.0110
	1980	0.0080	0.0080	---	0.0095
	1981	0.0090	0.0090	---	0.0075
	1982	0.0070	0.0080	---	0.0070
	1983	0.0080	0.0090	---	0.0080
	1984	0.0080	0.0080	---	0.0080
	1985	0.0080	0.0080	---	0.0085
	1986	0.0080	0.0060	---	0.0085
	1987	0.0080	0.0080	---	0.0090
	1988	0.0080	0.0070	---	0.0080
	1989	0.0070	0.0070	---	0.0075
	1990	0.0070	0.0070		0.0070
24-hr maximum TSP, $\mu g/M^3$	1978	189.0	189.0	209.0	183.5
	1979	167.5	170.0	217.0	145.0
	1980	155.0	156.0	202.0	137.0
	1981	161.5	143.5	234.0	163.0
	1982	143.5	146.5	166.5	134.5
	1983	438.0	495.5	218.0	319.0
	1984	139.0	137.0	167.0	139.0
	1985	253.0	297.0	179.0	208.0
	1986	119.0	123.0	218.0	107.0
	1987	141.0	146.0	183.5	131.5
	1988	169.0	174.0	188.0	138.0
	1989	188.0	188.0	231.0	137.0
	1990	220.0	220.0	158.0	237.0
Annual mean TSP, $\mu g/M^3$	1978	64.0	63.0	85.5	63.0
	1979	68.0	68.0	89.5	62.0
	1980	68.0	66.0	84.0	65.0
	1981	63.0	60.0	84.0	61.0
	1982	51.0	52.5	60.0	46.5
	1983	53.0	55.0	63.0	50.0
	1984	49.0	51.0	63.0	43.0
	1985	48.0	50.0	58.5	43.0
	1986	48.0	50.0	68.0	44.0
	1987	52.0	53.0	76.0	48.0
	1988	56.0	60.0	84.0	51.0
	1989	70.5	67.0	89.5	50.0
	1990	63.0	65.0	78.0	52.0

Note:

*--- Insufficient data at enough sampling sites to justify computing a median.

Table 1. (Continued)

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
Annual mean nitrate, μg/M^3	1978	5.50	5.50	5.60	5.60
	1979	5.40	5.80	5.20	5.20
	1980	5.40	4.75	5.50	5.50
	1981	5.50	5.80	5.30	5.30
	1982	5.15	5.40	4.50	4.50
	1983	6.00	6.30	4.90	4.90
	1984	5.80	6.00	4.20	4.20
	1985	5.00	5.30	3.85	3.85
	1986	5.25	5.50	4.10	4.10
	1987	5.50	5.50	5.70	5.70
	1988	5.30	5.45	4.80	4.80
	1989	5.40	5.80	5.05	5.05
	1990	4.60	4.80	4.55	4.55
Annual mean sulfate, μg/M^3	1978	11.90	12.10	13.40	10.50
	1979	11.90	11.85	13.50	10.75
	1980	12.30	12.55	12.90	11.05
	1981	12.00	12.00	12.90	11.60
	1982	10.45	10.05	11.55	10.30
	1983	12.25	12.20	13.80	10.95
	1984	10.80	10.85	11.20	9.10
	1985	10.20	10.20	11.65	9.00
	1986	10.10	10.05	10.80	8.80
	1987	10.20	10.00	14.20	10.00
	1988	11.00	10.80	12.10	9.60
	1989	10.80	10.50	12.60	9.95
	1990	10.70	10.30	12.45	9.50
Annual mean As, μg/M^3	1978	0.0030	0.0030	0.0080	0.0020
	1979	0.0020	0.0020	0.0040	0.0020
	1980	0.0020	0.0020	0.0050	0.0020
	1981	0.0020	0.0020	0.0040	0.0020
	1982	0.0020	0.0015	0.0120	0.0015
	1983	0.0020	0.0015	0.0060	0.0015
	1984	0.0010	0.0010	0.0080	0.0010
	1985	0.0000	0.0000	0.0040	0.0000
	1986	0.0010	0.0010	0.0060	0.0000
	1987	0.0010	0.0010	0.0060	0.0010
	1988	0.0020	0.0010	0.0060	0.0010
	1989	0.0010	0.0010	0.0040	0.0010
	1990	0.0010	0.0010	0.0045	0.0010

Note:

*--- Insufficient data at enough sampling sites to justify computing a median.

Table 1. (Continued)

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
Annual mean Cd, µg/M³	1978	0.00230	0.00230	0.00760	0.00190
	1979	0.00190	0.00175	0.00480	0.00165
	1980	0.00070	0.00070	0.00450	0.00045
	1981	---	---	---	---
	1982	0.00125	0.00055	0.00900	0.00010
	1983	0.00040	0.00035	0.00540	0.00015
	1984	0.00200	0.00100	0.00500	0.00150
	1985	0.00100	0.00000	0.00450	0.00100
	1986	0.00300	0.00300	0.00900	0.00100
	1987	0.00200	0.00200	0.01000	0.00100
	1988	0.00300	0.00300	0.00500	0.00000
	1989	0.00000	0.00000	0.00450	0.00000
	1990	0.00200	0.00200	0.00400	0.00000
Annual mean Cr, µg/M³	1985	0.0010	0.0010	--	0.0010
	1986	0.0095	0.0100	--	0.0035
	1987	0.0060	0.0070	--	0.0035
	1988	0.0050	0.0050	--	0.0040
	1989	0.0040	0.0050	--	0.0030
	1990	0.0010	0.0040	--	0.0000
Annual mean Fe, µg/M³	1978	0.850	0.845	1.410	0.740
	1979	0.985	0.990	1.785	0.805
	1980	0.680	0.710	1.110	0.540
	1981	0.605	0.600	1.040	0.580
	1982	0.600	0.600	1.365	0.510
	1983	0.810	0.810	1.360	0.640
	1984	0.690	0.690	1.270	0.500
	1985	0.640	0.640	1.435	0.375
	1986	0.720	0.720	2.230	0.440
	1987	0.840	0.820	2.550	0.530
	1988	0.830	0.855	2.700	0.570
	1989	1.100	1.245	2.145	0.510
	1990	0.980	1.265	2.150	0.600

Note:

*--- Insufficient data at enough sampling sites to justify computing a median.

Table 1. (Concluded)

Variable	Year	Whole state	Chicago area	Metro East area	Remainder of state
Annual mean Mn, $\mu g/M^3$	1978	0.0435	0.0405	0.1080	0.0590
	1979	0.0560	0.0540	0.1330	0.0580
	1980	0.0380	0.0355	0.0830	0.0395
	1981	0.0380	0.0345	0.0770	0.0410
	1982	0.0345	0.0325	0.0850	0.0335
	1983	0.0450	0.0460	0.0720	0.0325
	1984	0.0430	0.0475	0.0700	0.0320
	1985	0.0450	0.0450	0.1040	0.0295
	1986	0.0415	0.0420	0.1970	0.0290
	1987	0.0430	0.0430	0.2710	0.0370
	1988	0.0440	0.0410	0.2760	0.0360
	1989	0.0480	0.0480	0.2185	0.0320
	1990	0.0470	0.0555	0.2130	0.0360
Annual mean Ni, $\mu g/M^3$	1984	0.0020	0.0020	--	0.0030
	1985	0.0010	0.0010	--	0.0020
	1986	0.0070	0.0070	--	0.0040
	1987	0.0050	0.0050	--	0.0040
	1988	0.0050	0.0070	--	0.0030
	1989	0.0030	0.0040	--	0.0025
	1990	0.0030	0.0045	--	0.0020

Table 2. Computed Mean Annual Percent Change in Regional Concentrations, Assuming a Linear Trend. Significance of Time Trend was Based on Spearman Rank Correlation Coefficients.

Species	Observation	Whole state	Chicago area	Metro East area	Remainder of state
CO	1-hr maximum	-3.5**	-3.7**	---	-2.7*
CO	8-hr maximum	-4.6***	-5.3***	---	-3.7***
Pb	Annual mean	-20.5***	-21.2***	-12.6***	-24.8***
NO_2	1-hr maximum	-4.4***	-5.5***	---	---
NO_2	24-hr maximum	5.5***	-5.2***	---	---
NO_2	Annual mean	-7.1***	-6.8***	---	---
O_3†	1-hr maximum	n.s.(D**)	n.s.(D**)	n.s	n.s.
SO_2	3-hr maximum	n.s.	-3.5***	---	n.s.
SO_2	24-hr maximum	n.s.	n.s.	---	n.s.
SO_2	Annual mean	-2.6***	-5.2*	---	n.s.
TSP	24-hr maximum	n.s.	n.s.	n.s.	n.s.
TSP	Annual Geo mean	n.s.	n.s.	n.s.	n.s.
NO_3^-	Annual mean	n.s.	n.s.	n.s.	-2.1*
SO_4^{2-}	Annual mean	-1.3*	-1.6*	n.s.	-1.4**
As	Annual mean	-8.4***	-11.1***	n.s.	-9.5***
Cd	Annual mean	n.s.	n.s.	n.s.	-12.7**
Cr	Annual mean	n.s.	n.s.	---	n.s.
Fe	Annual mean	n.s.	n.s.	+6.0**	n.s.
Mn	Annual mean	n.s.	n.s.	+10.0*	-4.4*
Ni	Annual mean	n.s.	n.s.	---	n.s.

Notes:

n.s. = not significant.

D = downward trend (not quantifiable by method used).

Significance levels: * = 5 percent, ** = 2 percent, and *** = 1 percent.

--- = not enough sites to estimate trends. (When there were not enough Metro East sites for a trend estimate, the Metro East sites are included with the remainder of the state.)

† O_3 data were tested for time trend both before and after accounting for surface temperature influences. Results in parentheses show trend and significance after accounting for surface temperature.

pollutants examined. Time trends for each pollutant are graphed and the text discusses the results of the related statistical tests in table 2. The graphs show concentrations at several percentile points on the distribution of particular pollutant concentration statistics (i.e., annual means or 24-hr maxima) over all sampling sites within a given geographic area for each of a series of years, and how these percentiles of concentration change over the period 1978-1990. The box plot is a convenient graphical device to use for this purpose, because it shows several concentration percentiles simultaneously. The concentrations plotted in a single graph are averages over a particular time period, such as one hour, 24 hours, or an entire year. For the criteria pollutants, these averaging times correspond to those *for which the standards are written for each particular pollutant*, so they vary from one pollutant to the next, and a given pollutant may have two or more graphs, corresponding to two or more averaging times.

It will become apparent later that the number of sampling sites for a given pollutant in any geographical area changed from year to year as new sites were installed or previously used sites were taken out of service. In recent years the number of sampling sites has dropped markedly for most pollutants. This suggests an alternative approach to the analysis of time trends: analysis of time trends only at sites with long-term records. This approach was rejected in favor of the analysis of time trends in regional medians because very few sites had long-term records, and such sites could not be inferred to represent broad geographic areas.

Data used in the following statistical tests are shown in table 1.

Results of the statistical tests for the significance of trends are shown in table 2, and the time sequences of box plots in figures 1-30. Each figure shows results for up to four geographical areas, depending on whether the number of sampling sites in the area was sufficient to justify a separate plot. Each figure shows results for the entire state and the Chicago area, which is the portion of the U.S. EPA's Air Quality Control Region (AQCR) 67 in Illinois. Many figures also show results for the Metro East area on the Illinois side of the Mississippi River across from St. Louis. This is the Illinois portion of AQCR 70. The fourth geographic region shown is the remainder of the state, excluding the Chicago area and the Metro East area (if shown separately). If the Metro East area is not shown separately, then the region labeled as "remainder of state" also includes any data from the Metro East area.

Criteria Pollutants

Carbon Monoxide (CO). Results for CO are shown in table 2 and figures 1 and 2. The table shows trends toward decreasing concentrations significant at the 5 percent level, or better, for both 1-hr (figure 1) and 8-hr (figure 2) maxima during the 1979-1990 period, in all three geographic areas with sufficient data to test. The overall linear trends range from -2.7 percent to -5.3 percent. Figure 1 suggests generally higher and slightly increasing 1-hr maximum concentrations at all percentile levels represented by the box plots during the 1978-1984 period, followed by lower and decreasing concentrations from 1985 onward. This pattern appears in all three geographic areas shown in the figure. The 1-hr CO standard, 35 parts per million (ppm), appears not to have been exceeded anywhere in the state during the 13 years of record.

On the other hand, the 8-hr maximum concentrations of CO (figure 2) exceeded the standard, 9 ppm, repeatedly until very late in the 1979-1990 period. Figure 2 shows a pattern of higher concentrations at all percentiles during the 1978-1984 period, followed by lower and decreasing concentrations from 1985-1990, in all geographic areas.

The statistical comparison of CO concentrations between geographical regions found a significant difference (5 percent) between the Chicago area and the remainder of the state for the 1-hr (figure 1), but not for the 8-hr (figure 2) averaging time. For both averaging times, however, it appears that early in the 1979-1990 period the Chicago area contributed the highest individual concentrations, whereas the highest values during the later years of the period came from elsewhere in the state.

For both CO averaging times, the number of sampling sites (N) remained relatively constant, with about 15 sites in the entire state, 6 to 11 sites in the Chicago area, and 6 or 7 sites elsewhere.

Lead (Pb). Airborne Pb concentrations decreased dramatically (figure 3) during the 1979-1990 period, due in part to the phase-out of leaded gasoline, beginning in about 1975 (IEPA, 1991). Table 2 shows decreasing linear trends between -12.6 percent and -24.8 percent per year in the four geographical regions. While the trends are significant at the 1 percent level in all four regions, the rate of decrease in the Metro East area is considerably smaller than the others. Figure 3 graphically shows the marked drops in all the concentrations
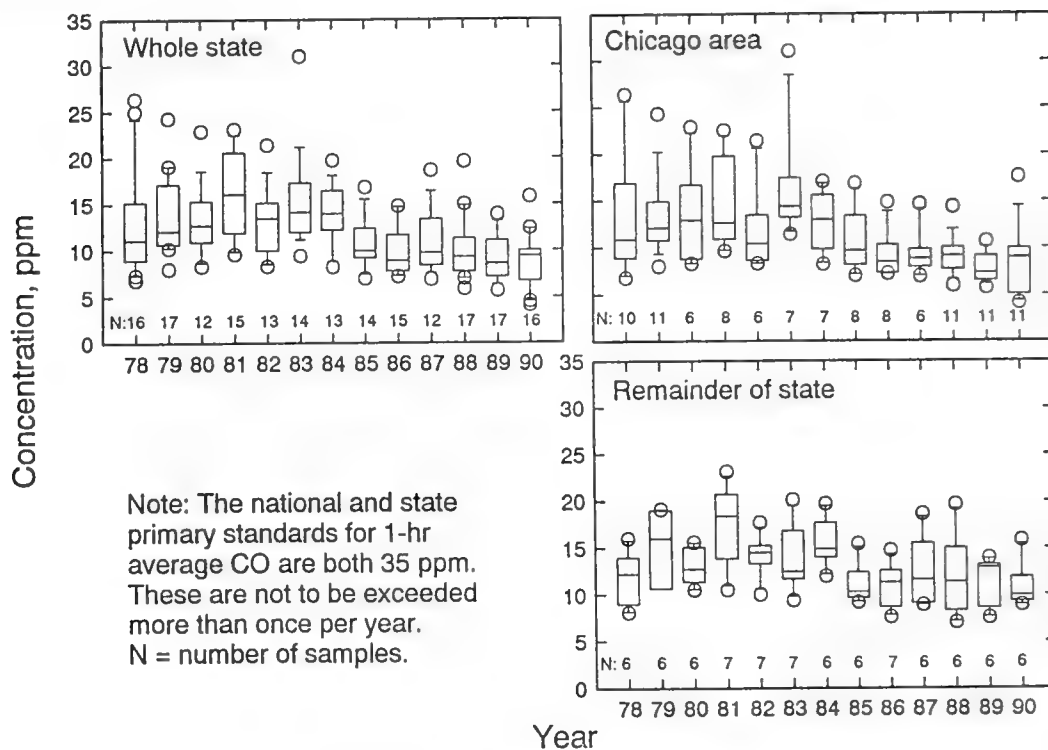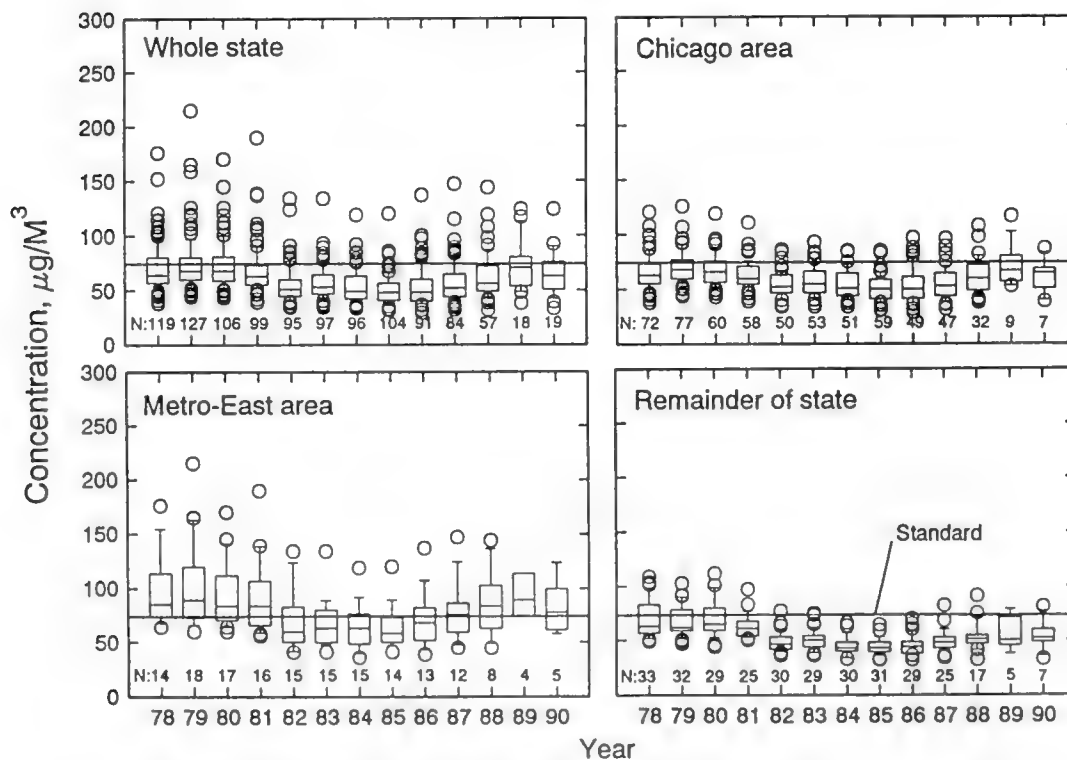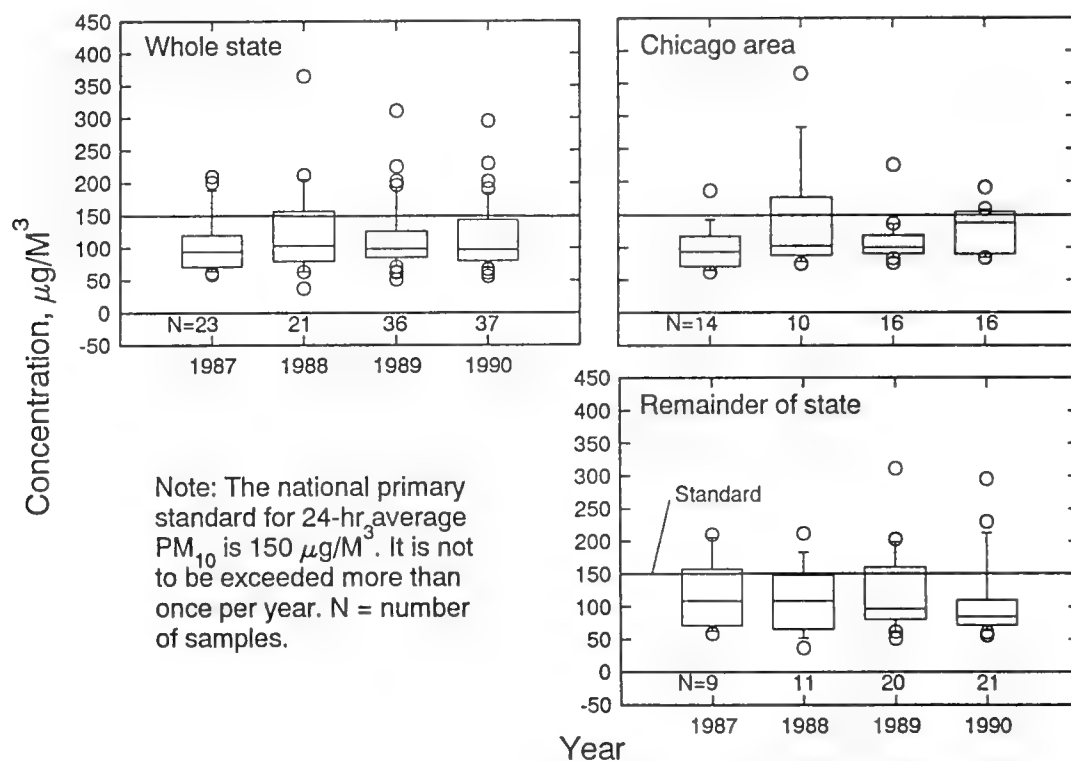
Figure 1. Trends in 1-hr maximum CO concentrations in Illinois.
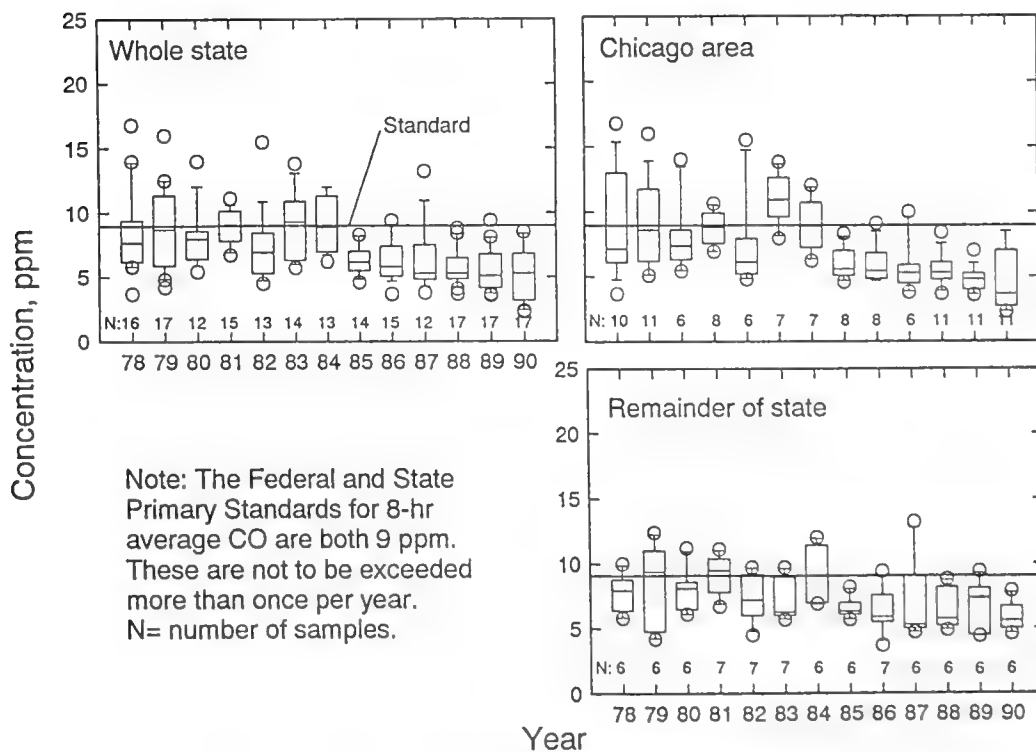
Figure 2. Trends in 8-hr maximum CO concentrations in Illinois.
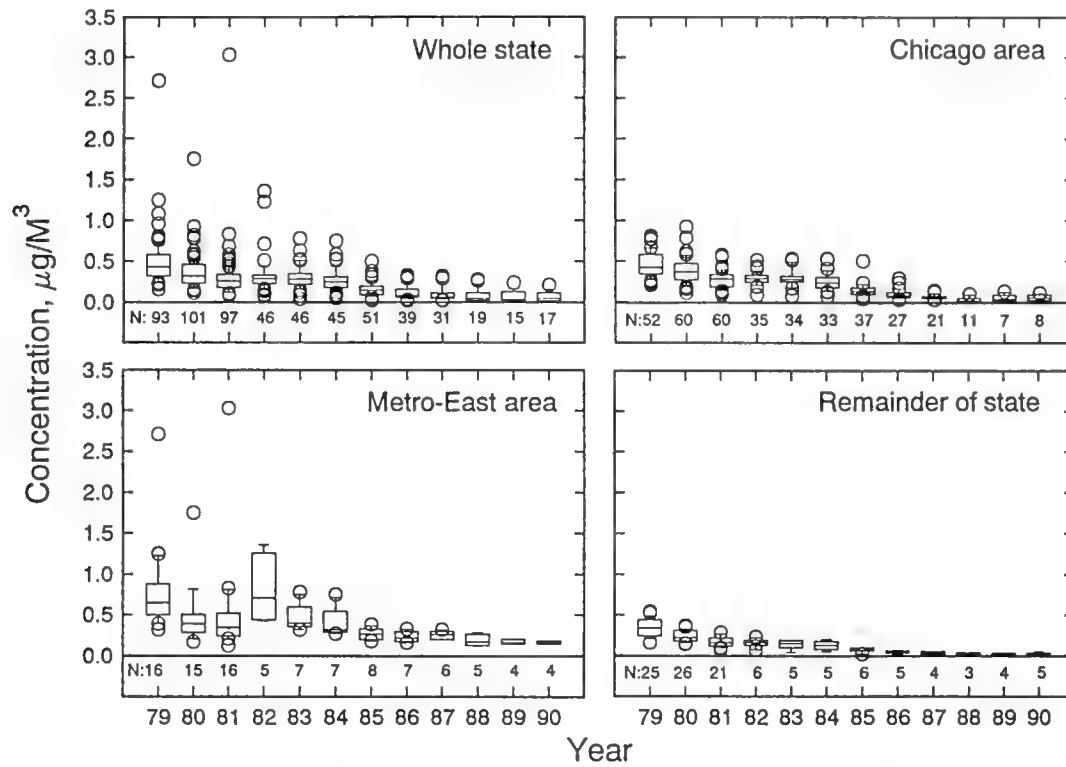
Note: Standards are based on the quarterly arithmetic mean. See text.
N = number of samples.

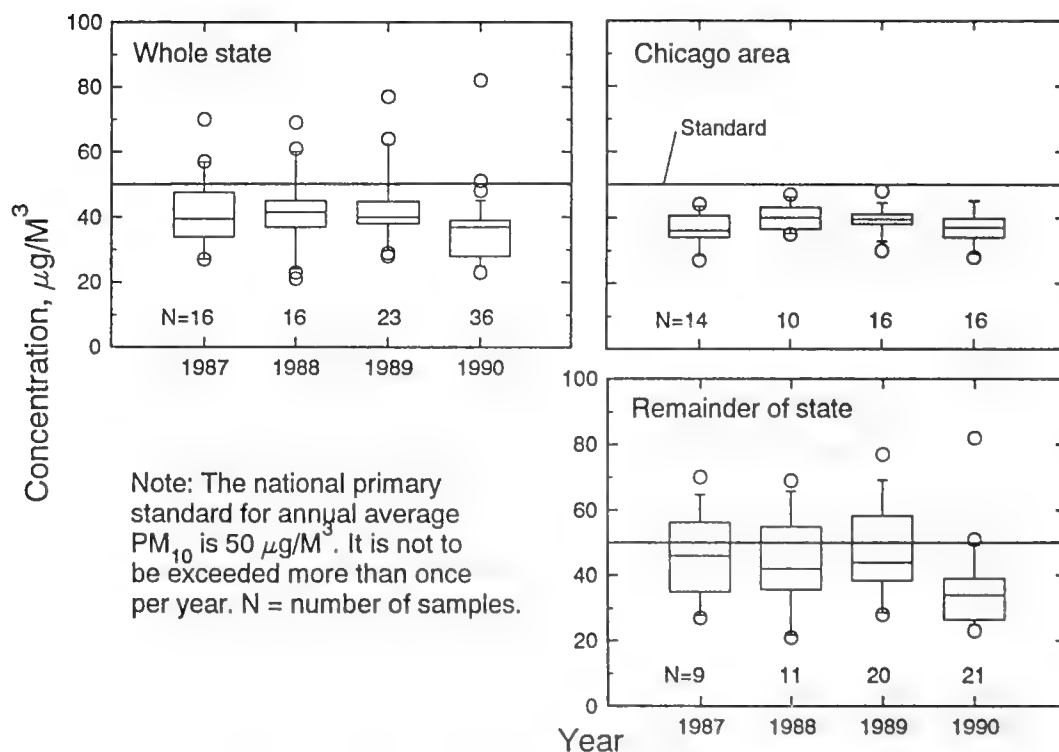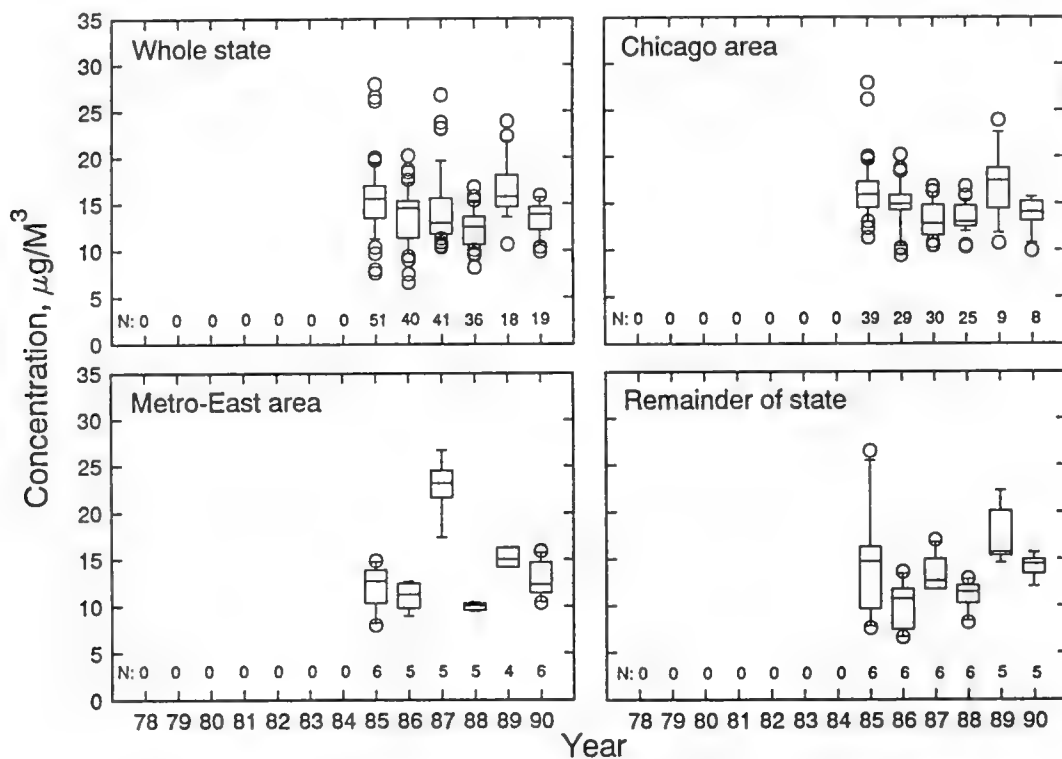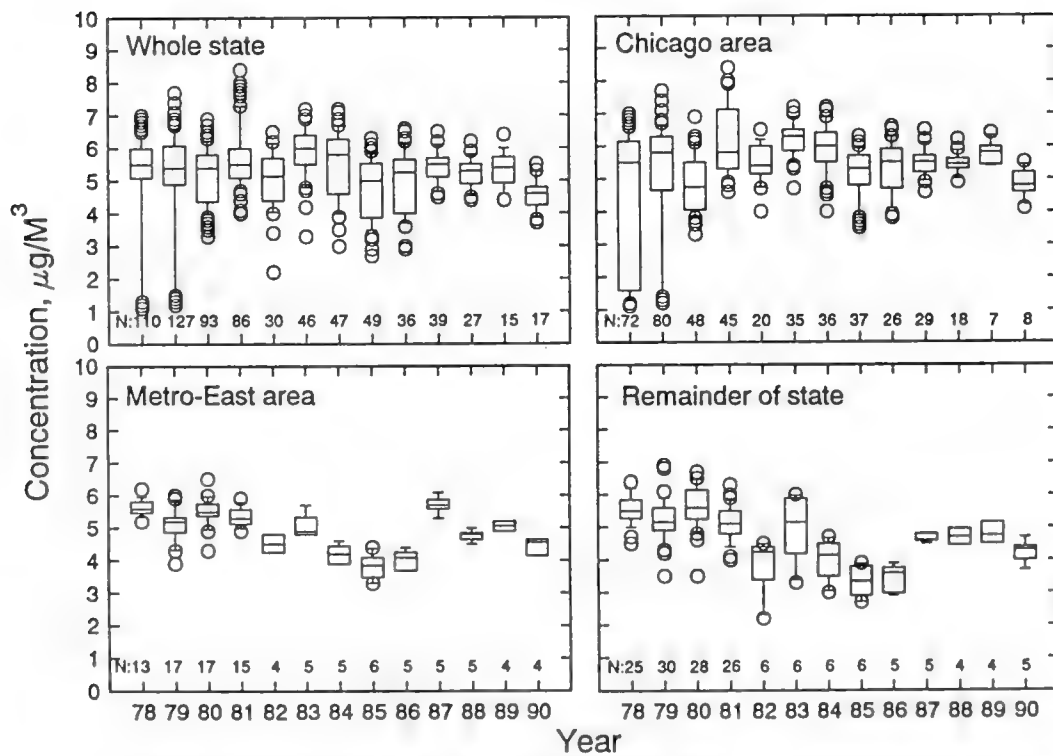*Figure 3. Trends in annual mean Pb concentrations in Illinois.*
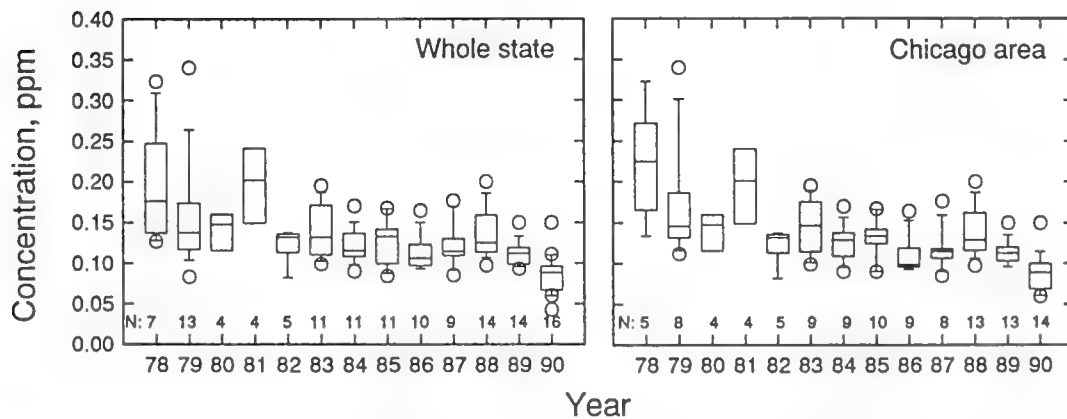
Note: There are no national or state standards for 1-hr average NO_2.

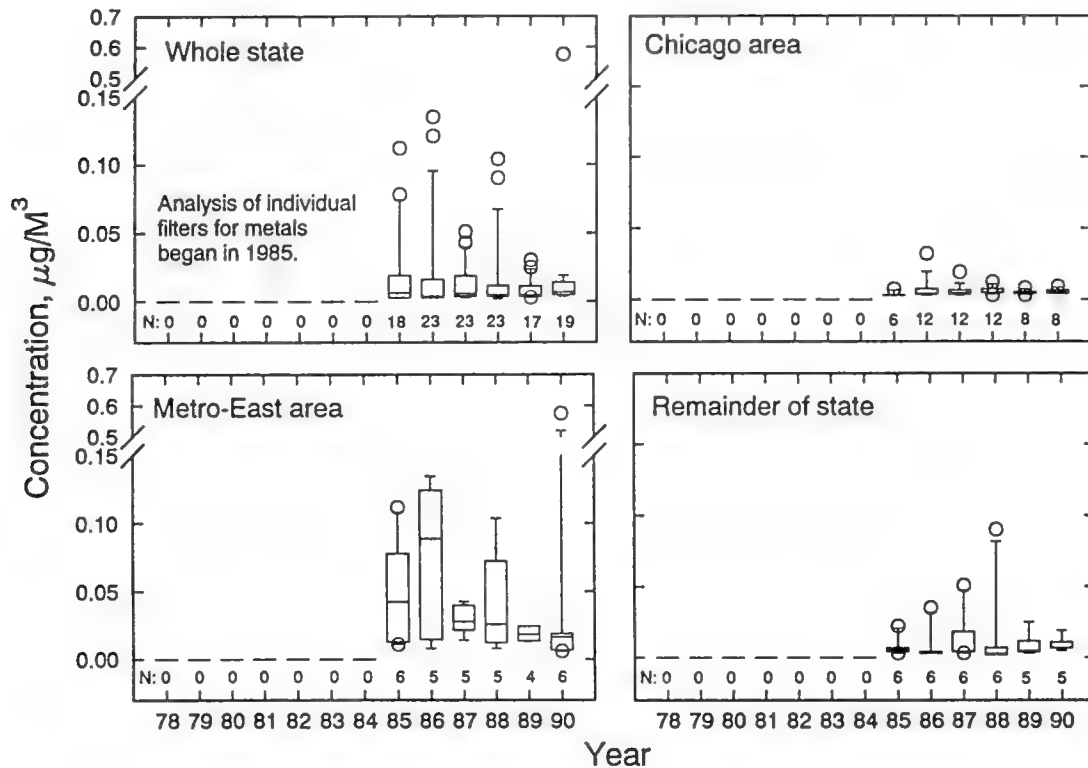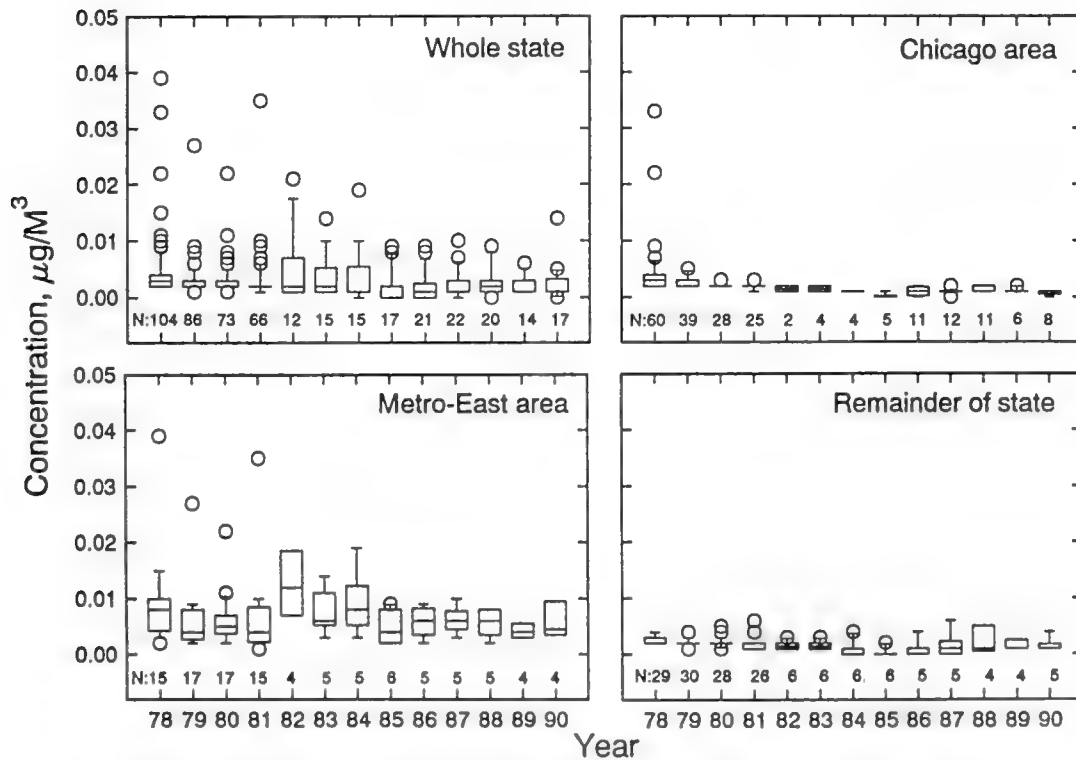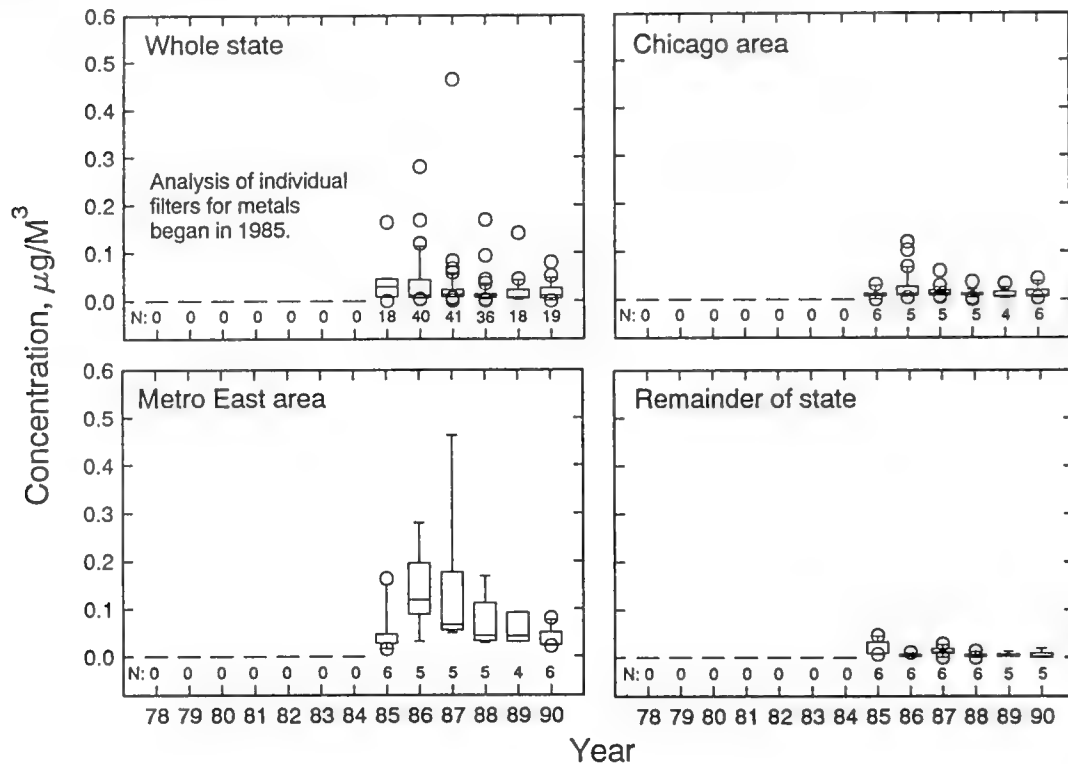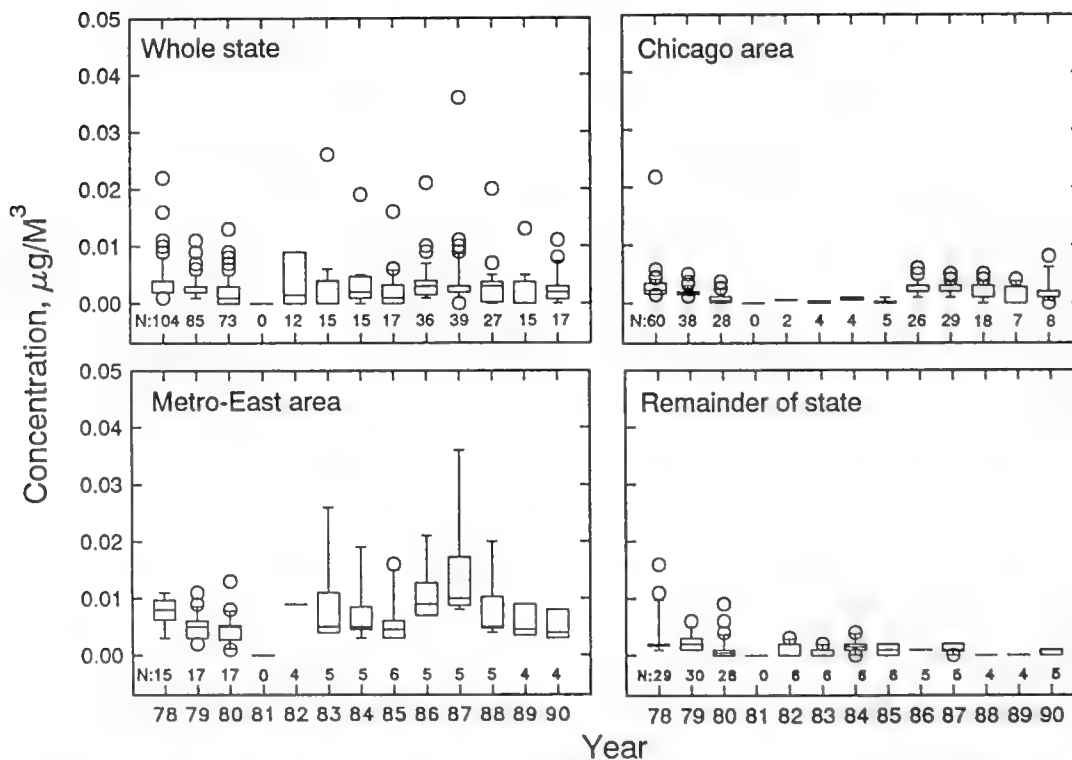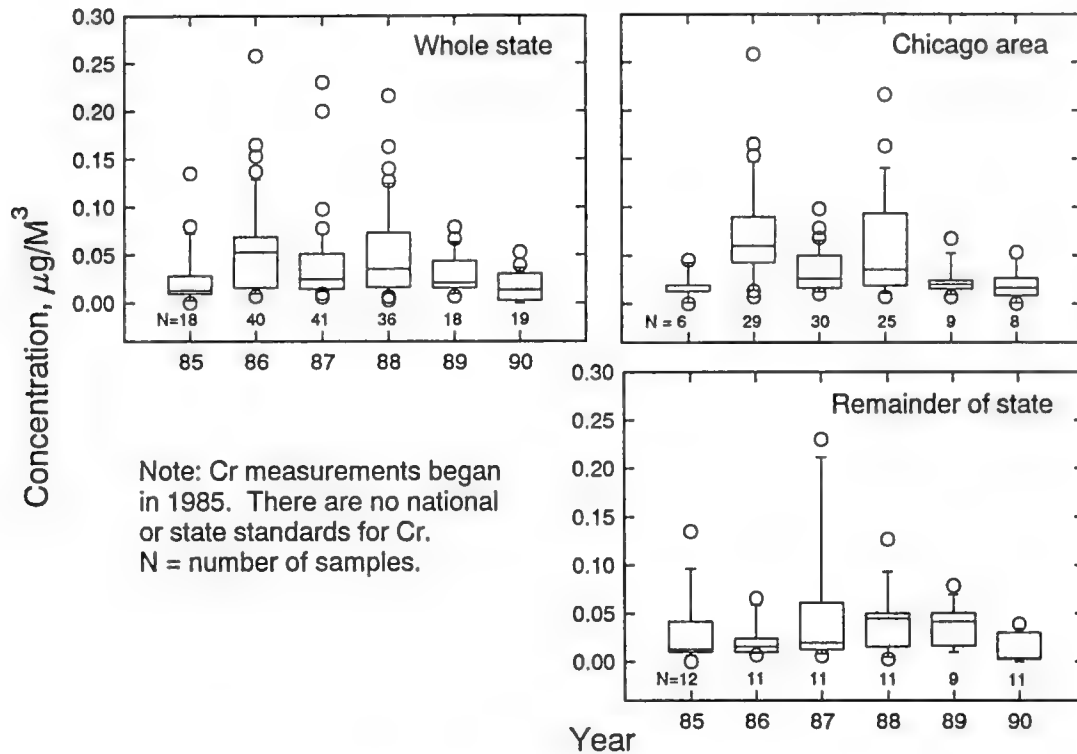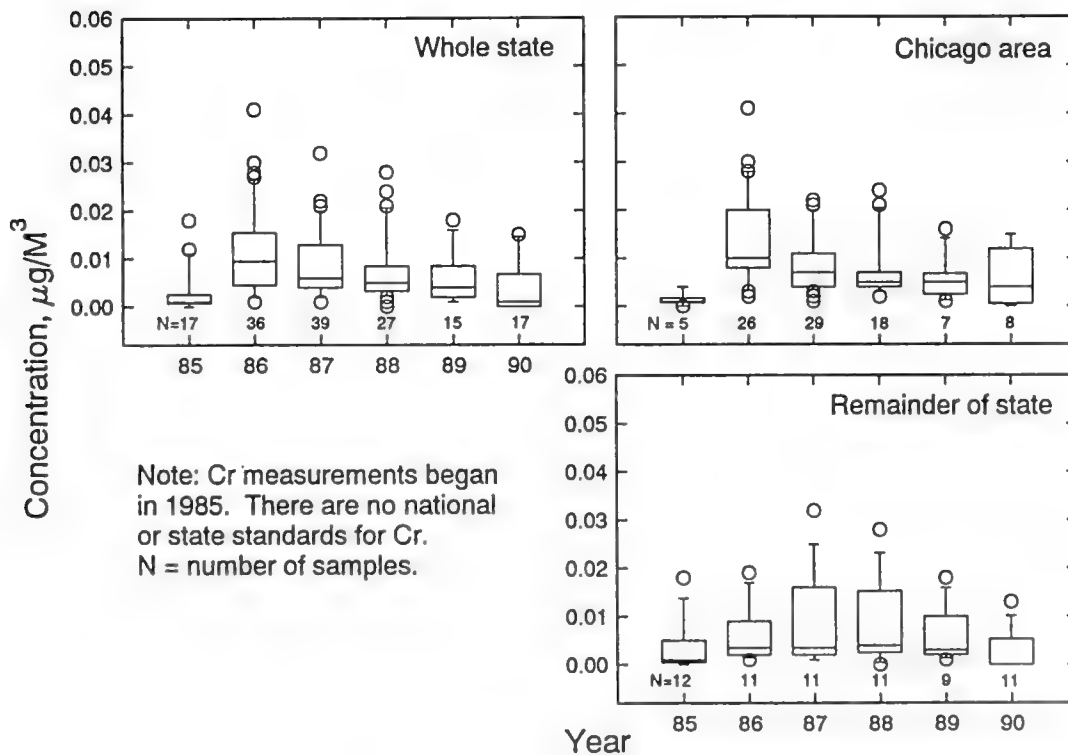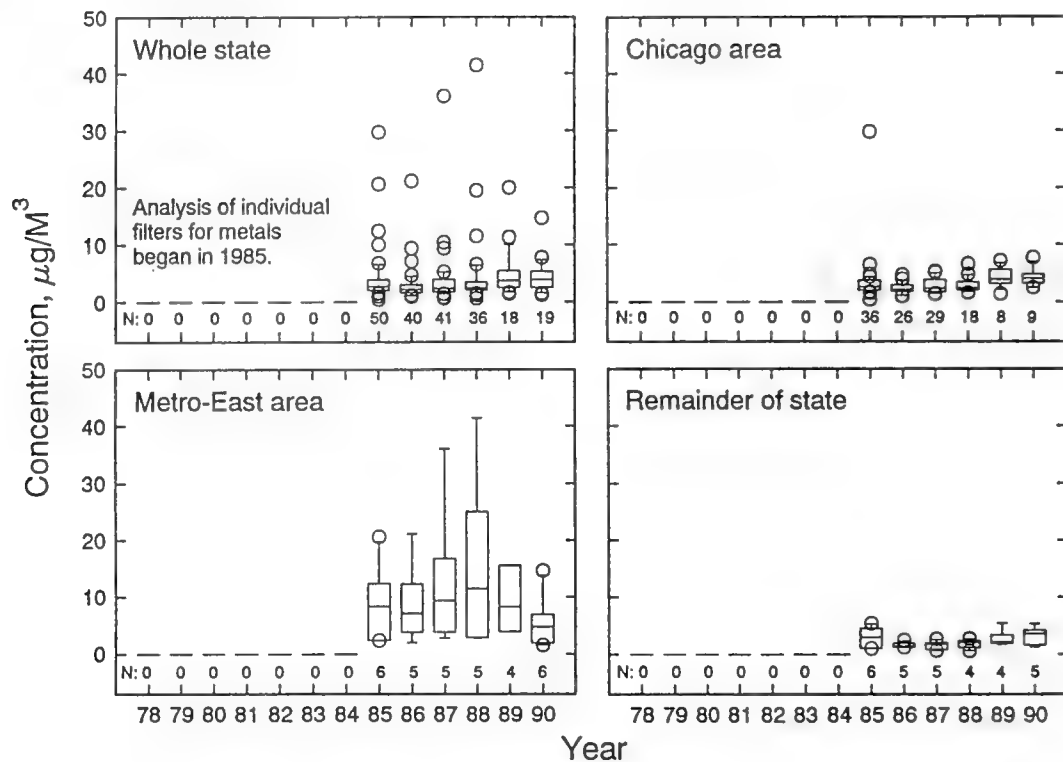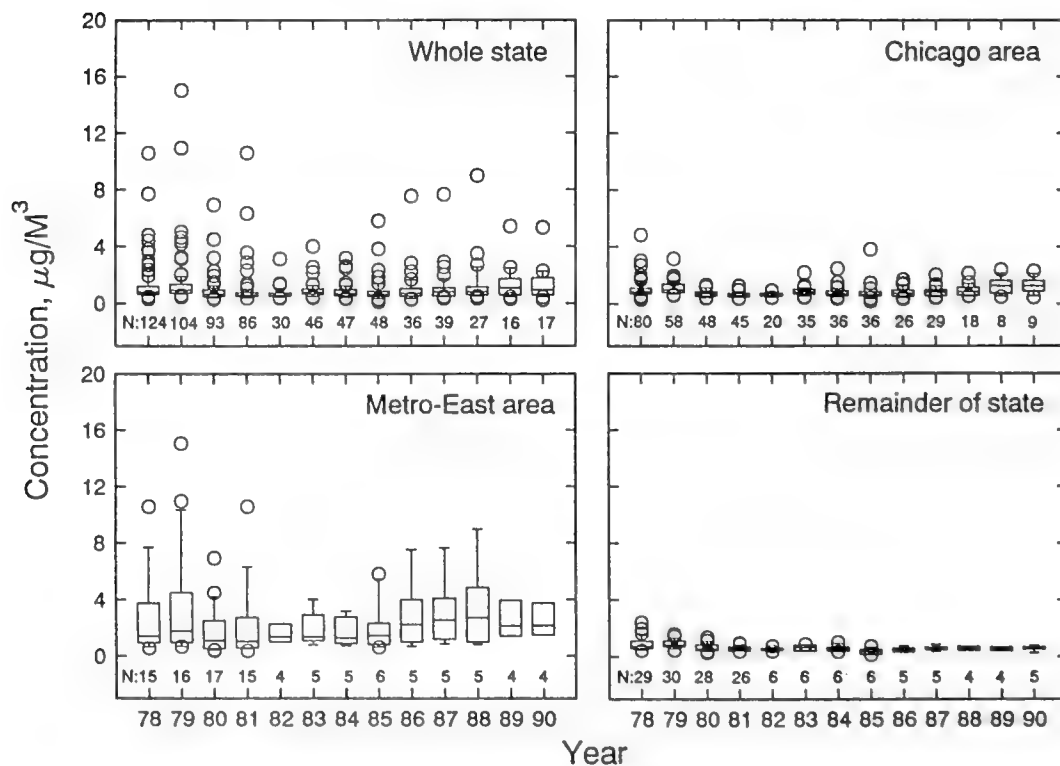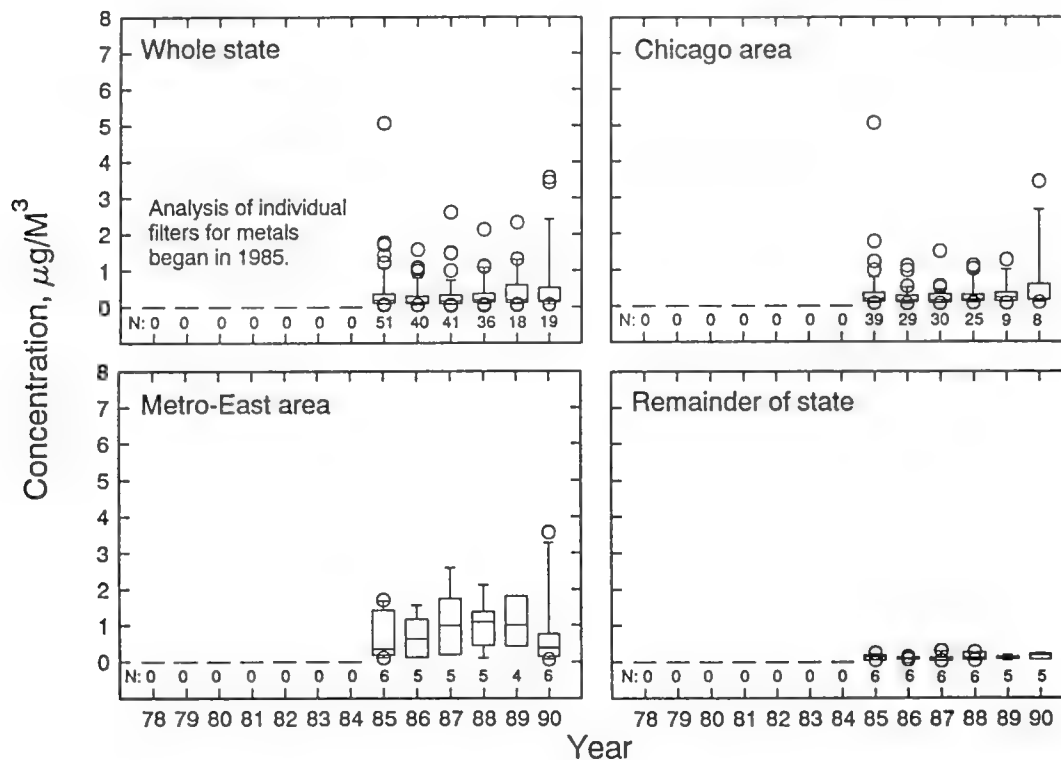*Figure 4. Trends in 1-hr maximum NO_2 concentrations in Illinois.*
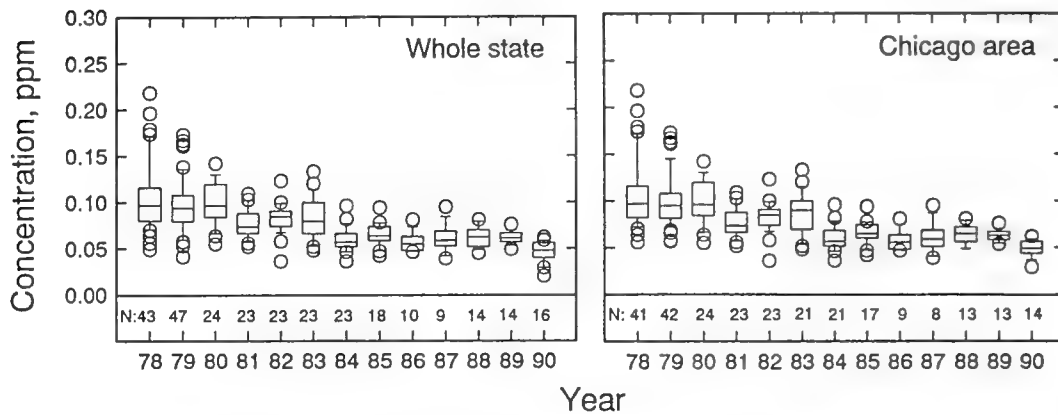
Note: There are no national or state standards for 24-hr average NO_2.
      N = number of samples.

Figure 5. Trends in 24-hr maximum NO_2 concentrations in Illinois.
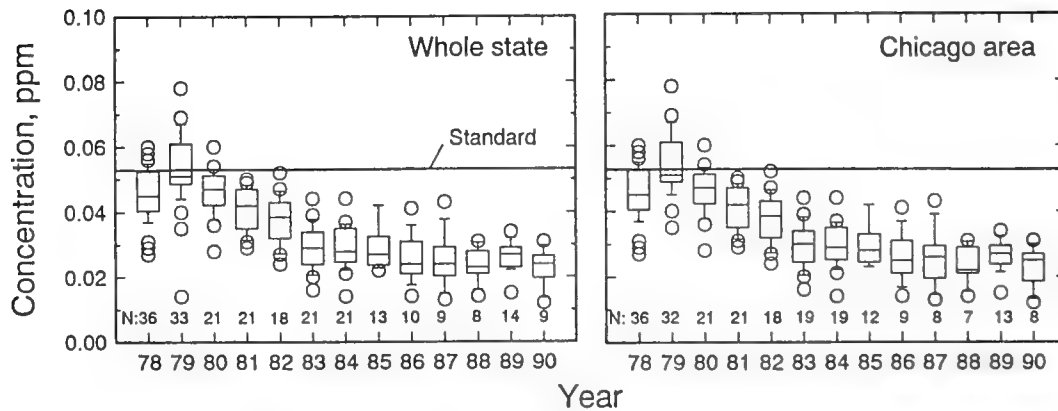
Note: The national and state standards for annual mean NO_2 are 0.053 ppm.
      N= number of samples.

Figure 6. Trends in annual mean NO_2 concentrations in Illinois.
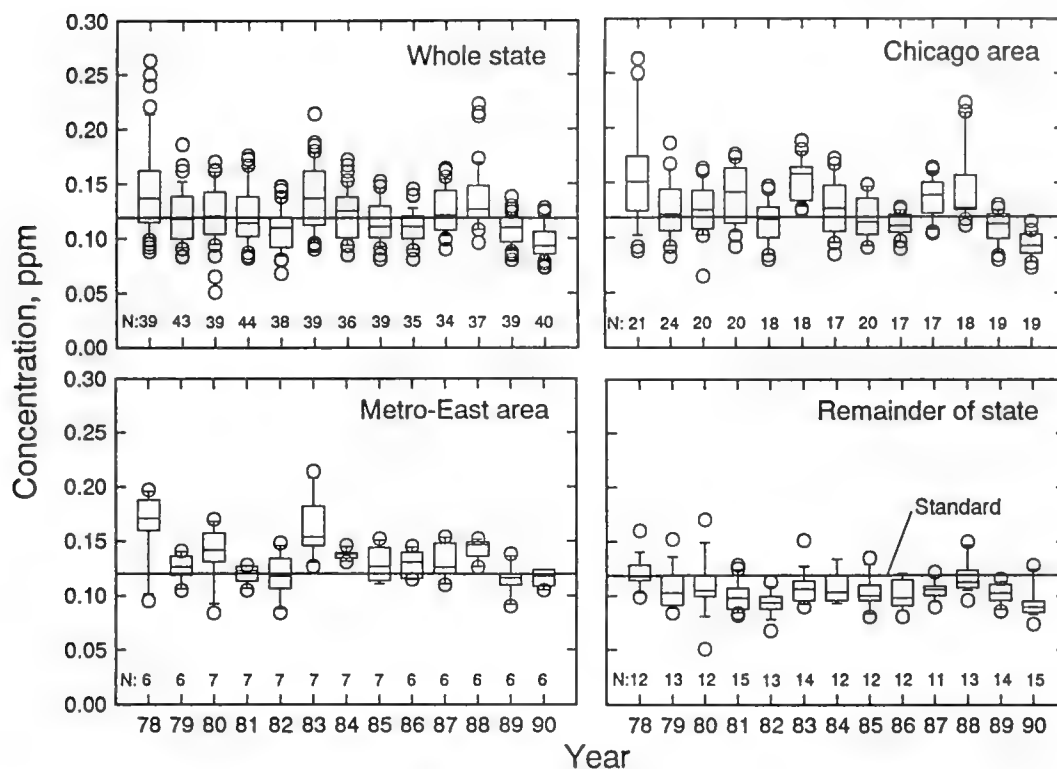
Note: The national and state primary standards for 1-hr O_3 are 0.12 ppm.
N = number of samples.

Figure 7. Trends in 1-hr maximum O_3 concentrations in Illinois.
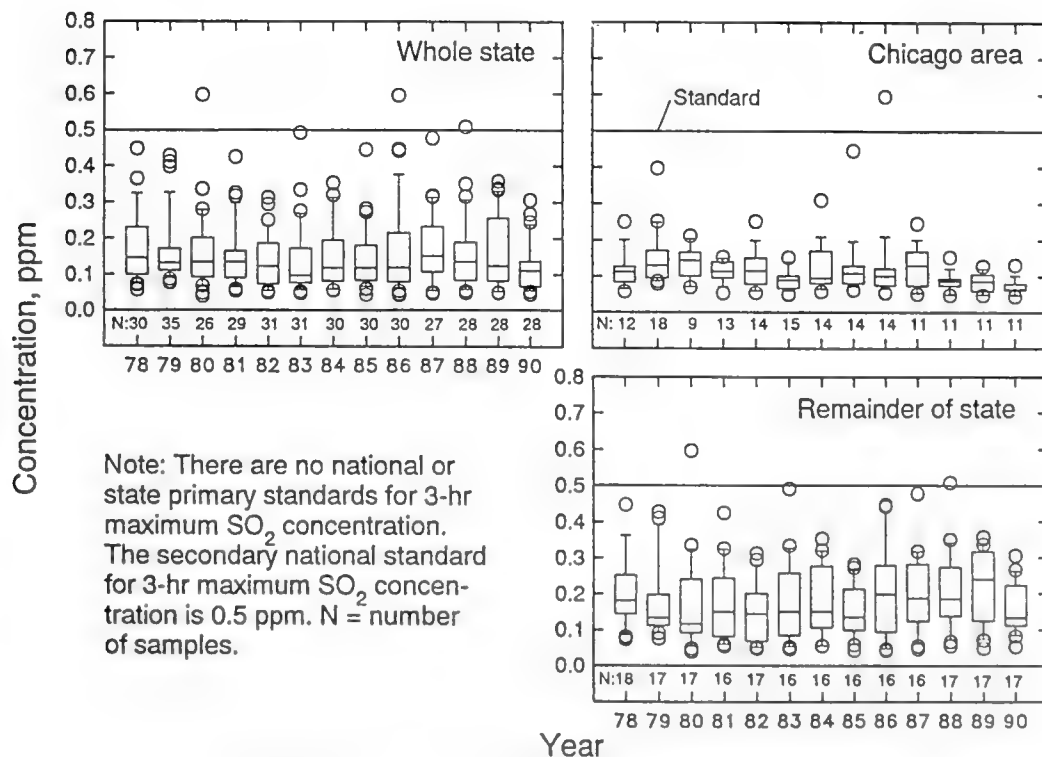
Note: There are no national or state primary standards for 3-hr maximum SO_2 concentration. The secondary national standard for 3-hr maximum SO_2 concentration is 0.5 ppm. N = number of samples.

Figure 8. Trends in 3-hr maximum SO_2 concentrations in Illinois.

Note: The national and state primary standards for 24-hr maximum SO$_2$ concentration are both 0.14 ppm. N = number of samples.
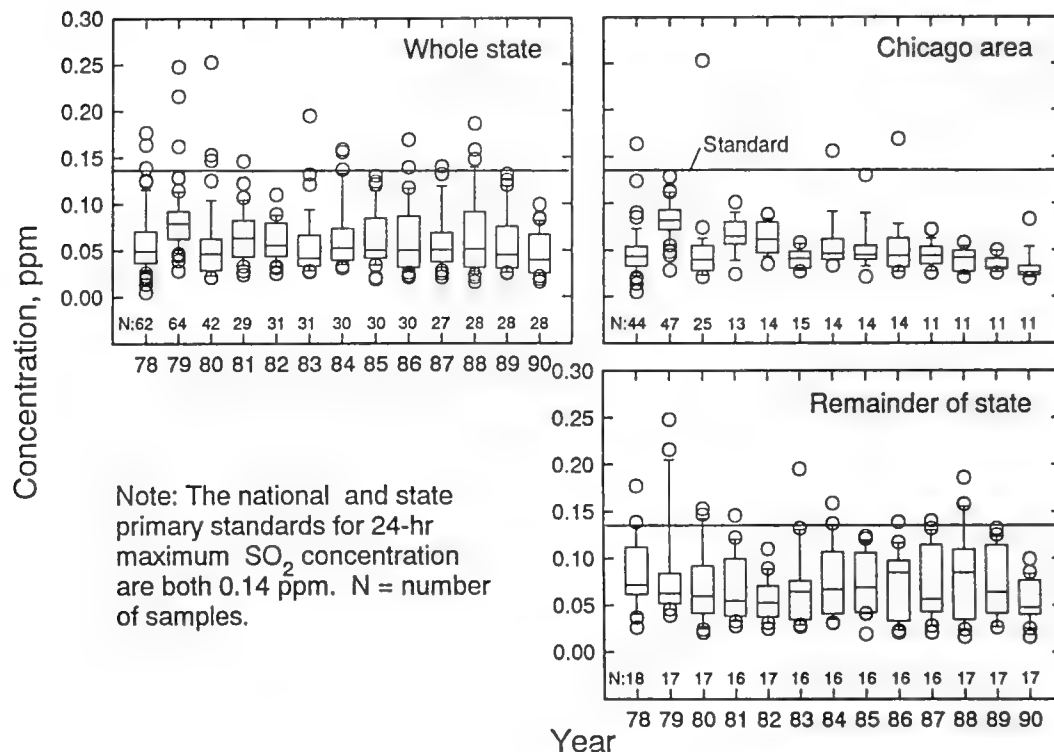
Figure 9. Trends in 24-hr maximum SO$_2$ concentrations in Illinois.

Note: The national and state primary standards for annual arithmetic mean SO$_2$ concentration are 0.03 ppm. N = number of samples.
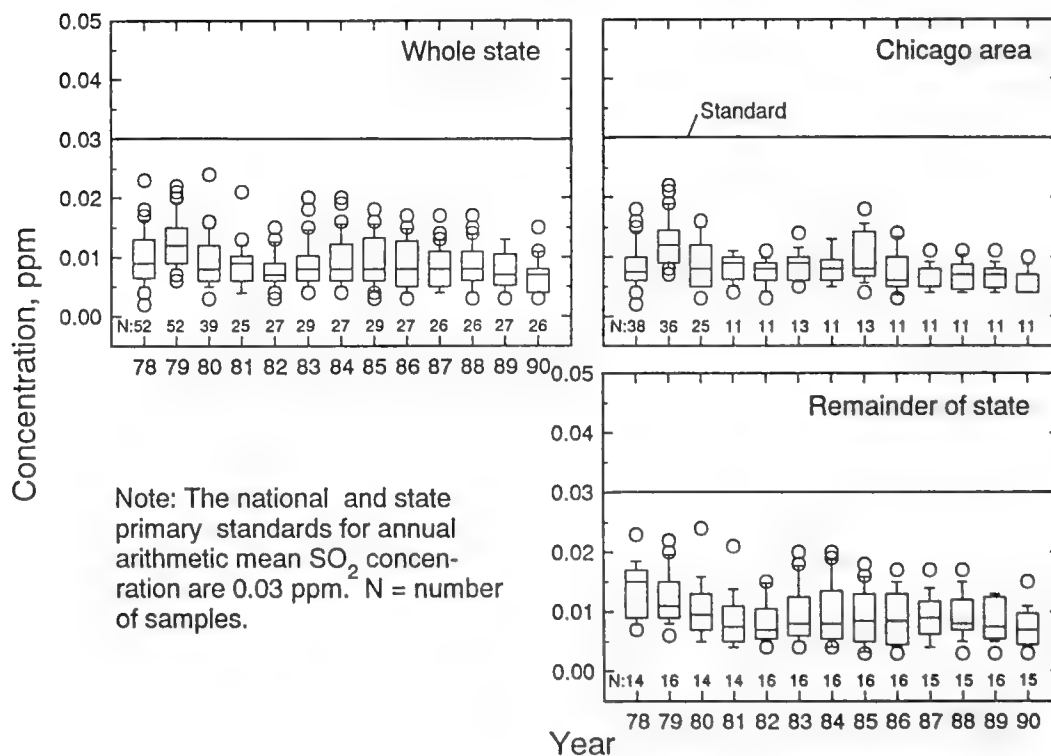
Figure 10. Trends in annual mean SO$_2$ concentrations in Illinois.
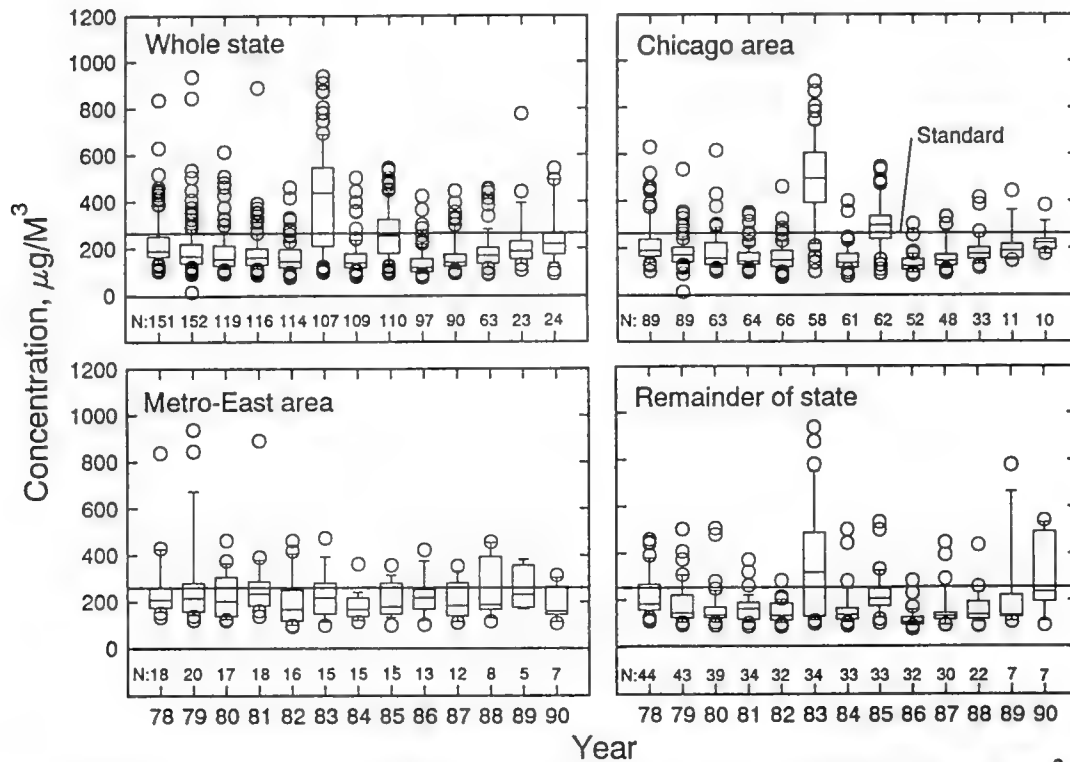
Note: The state primary standard for 24-hr maximum TSP concentration is 260 μg/M^3.
N = number of samples.

Figure 11. Trends in 24-hr maximum TSP concentrations in Illinois.

Note: The state primary standard for annual geometric mean TSP is 75 μg/M^3
N = number of samples.

Figure 12. Trends in annual geometric mean TSP concentrations in Illinois.

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Figure 13. Trends in 24-hr maximum PM$_{10}$ concentrations in Illinois.

Figure 14. Trends in annual mean PM$_{10}$ concentrations in Illinois.

Note: There are no national or state standards for NO_3^-. N = number of samples.

Figure 15. Trends in 24-hr maximum NO_3^- concentrations in Illinois.

Note: There are no national or state standards for NO_3^-. N = number of samples.

Figure 16. Trends in annual mean NO_3^- concentrations in Illinois.
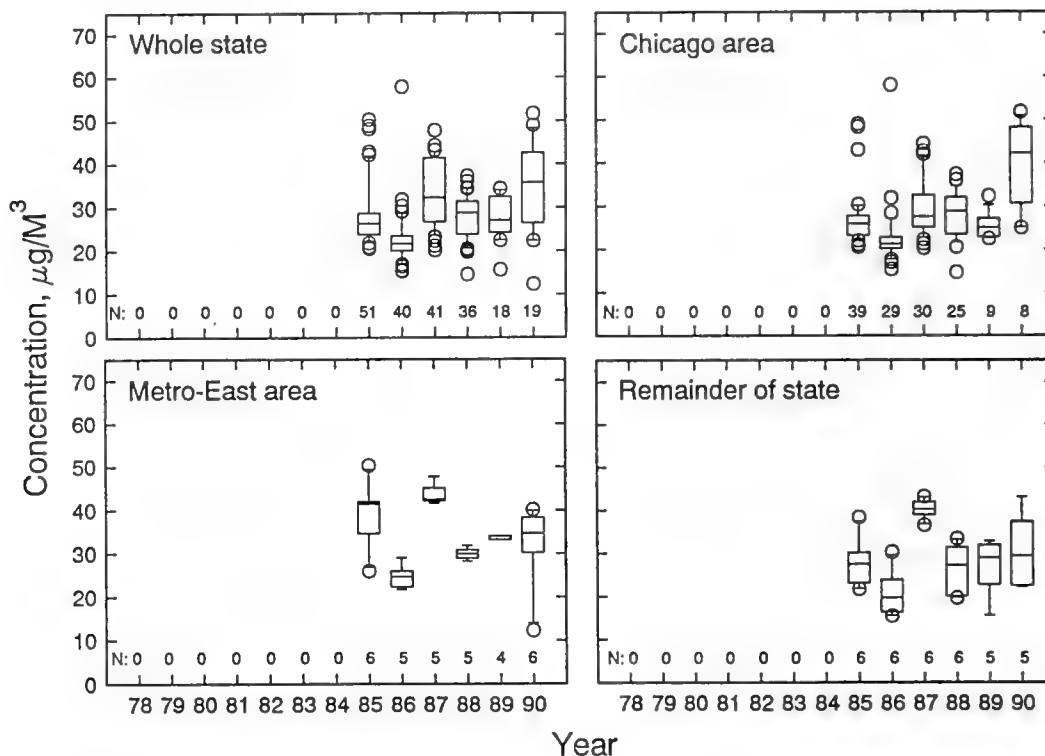
Note: There are no national or state standards for SO_4^{2-}. N = number of samples.

Figure 17. Trends in 24-hr maximum SO_4^{2-} concentrations in Illinois.
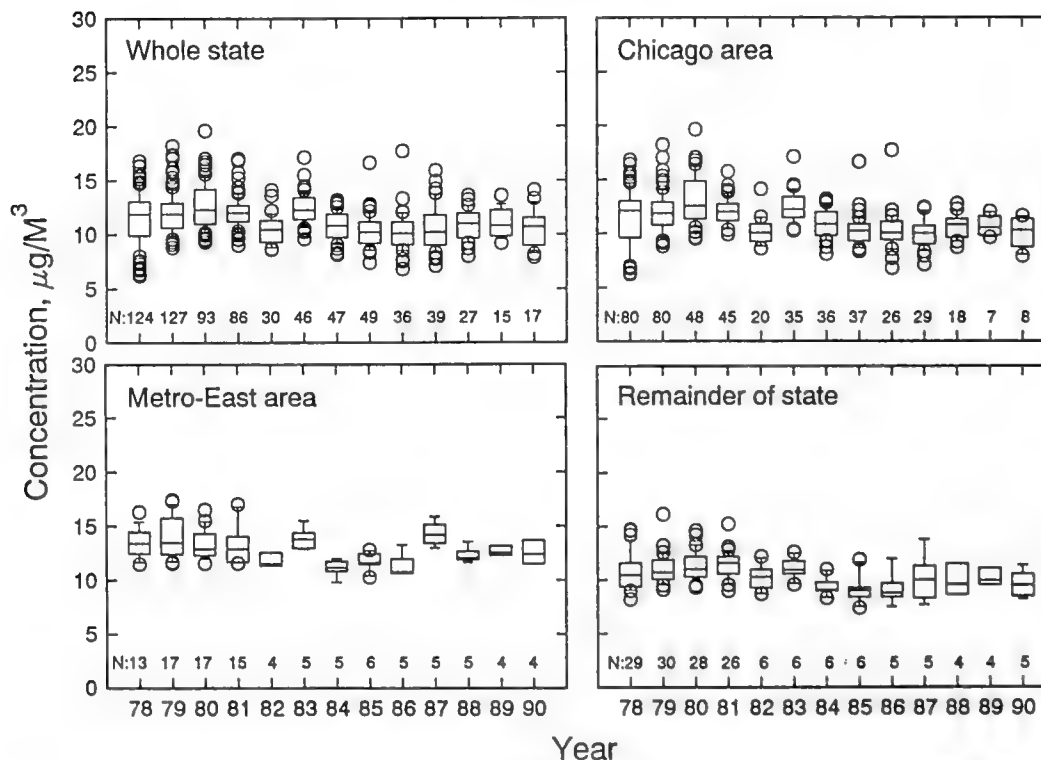
Note: There are no national or state standards for SO_4^{2-}. N = number of samples.

Figure 18. Trends in annual mean SO_4^{2-} concentrations in Illinois.

Note: There are no national or state standards for As. N = number of samples.

Figure 19. Trends in 24-hr maximum As concentrations in Illinois.

Note: There are no national or state standards for As. N = number of samples.

Figure 20. Trends in annual mean As concentrations in Illinois.

Note: There are no national or state standards for Cd. N = number of samples.

Figure 21. Trends in 24-hr maximum Cd concentrations in Illinois.

Note: There are no national or state standards for Cd. N = number of samples.

Figure 22. Trends in annual mean Cd concentrations in Illinois.

Figure 23. Trends in 24-hr maximum Cr concentrations in Illinois.

Figure 24. Trends in annual mean Cr concentrations in Illinois.

Note: There are no national or state standards for Fe. N = number of samples.

Figure 25. Trends in 24-hr maximum Fe concentrations in Illinois.

Note: There are no national or state standards for Fe. N = number of samples.

Figure 26. Trends in annual mean Fe concentrations in Illinois.

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Note: There are no national or state standards for Mn. N = number of samples.

Figure 27. Trends in 24-hr maximum Mn concentrations in Illinois.
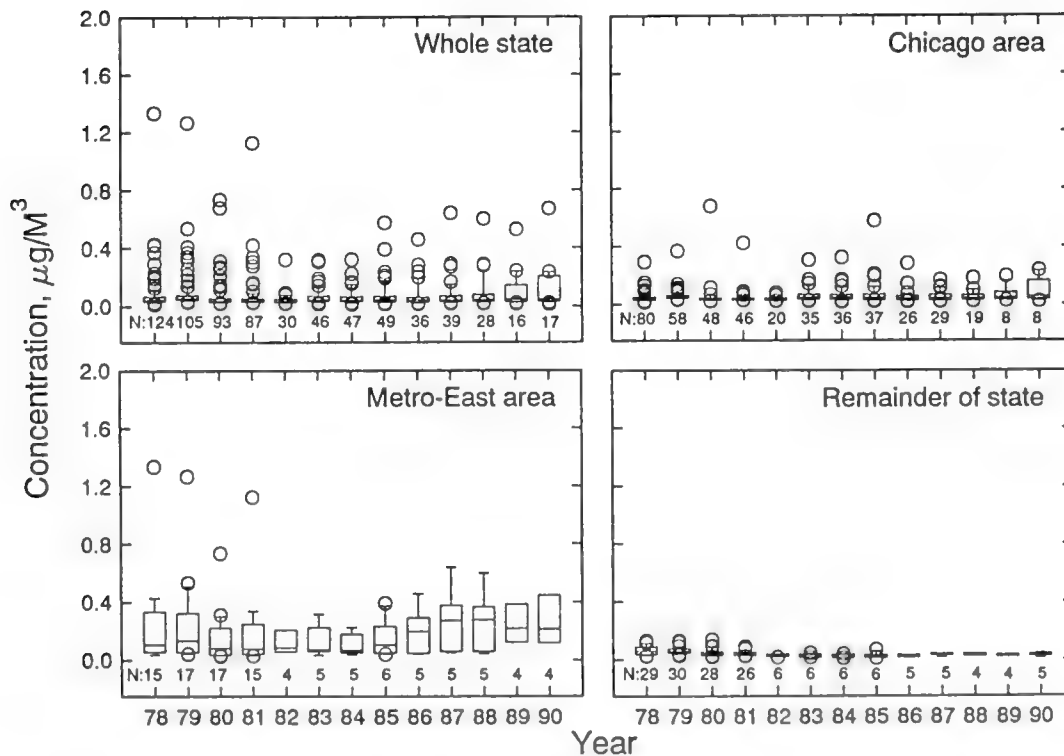
Note: There are no national or state standards for Mn. N = number of samples.

Figure 28. Trends in annual mean Mn concentrations in Illinois.
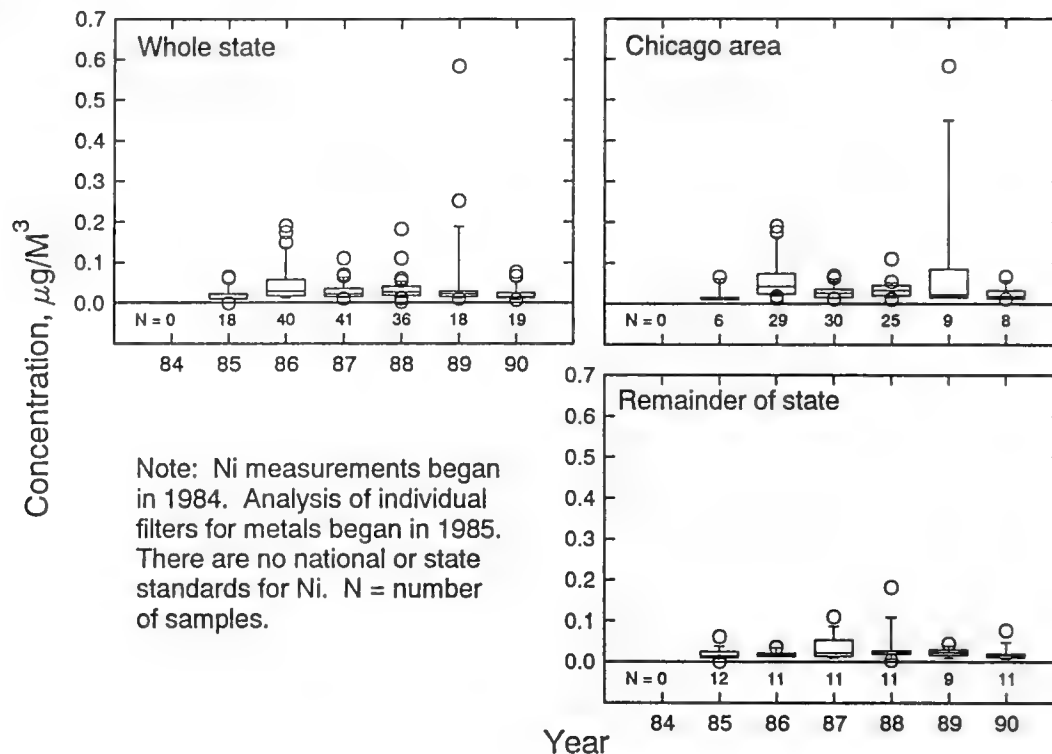
Figure 29. Trends in 24-hr maximum Ni concentrations in Illinois.
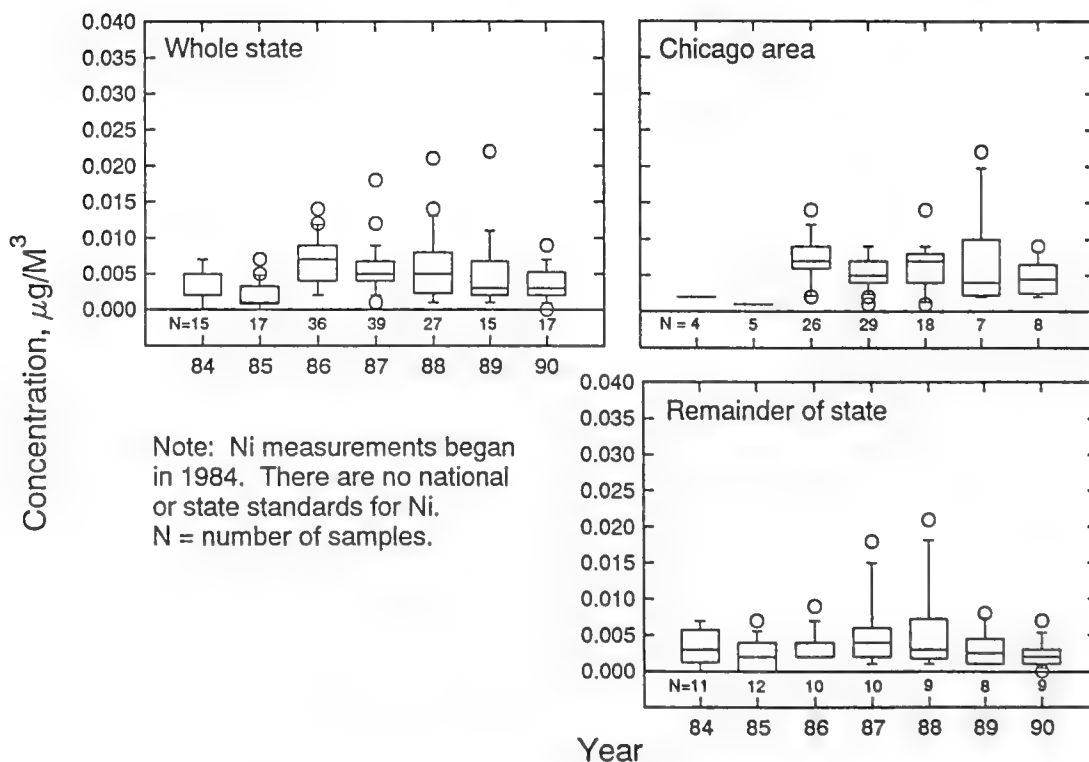
Figure 30. Trends in annual mean Ni concentrations in Illinois.

corresponding to the percentiles depicted by the box plots.

There has clearly also been a drop in the highest annual Pb concentration in each region, as depicted by the uppermost open circle. This dropoff was quite dramatic in the entire state, where numerous concentrations exceeded 1 microgram per cubic meter ($\mu g/M^3$) occurred between 1979 and 1982, but none thereafter. Comparison of the "whole state" panel with the other three panels clearly shows that high individual concentrations during the 1979-1982 period occurred in the Metro East area. The marked improvement in maximum and median Metro East airborne Pb concentrations after 1982 is likely to have been strongly influenced by the closure and subsequent cleanup of a secondary lead smelter in Granite City (Cooper and Frazier, 1983).

The national and state standard for Pb is a quarterly arithmetic mean of 1.5 $\mu g/M^3$. No violations of this standard have occurred in Illinois since 1982 (IEPA, 1991).

Inspection of figure 3 indicates systematically higher individual Pb concentrations (open circles) in the Metro East area than in the Chicago area or the remainder of the state, especially during the 1979-1984 period. The statistical test for differences between regional means found only the Metro East Pb concentrations higher than the remainder of the state at the 5 percent level. Median Pb concentrations in the Metro East area were also higher than those in the Chicago area, but the differences were not quite significant at the 5 percent level.

As Pb concentrations have decreased, and violations of the standard have dropped to zero, the number of sites measuring airborne Pb concentrations in Illinois has also dropped sharply. Statewide, the number of sampling sites with valid annual means decreased from a high of 101 in 1980 to 17 in 1990. In the Chicago area the corresponding numbers are 60 and 8; in the Metro East area, 16 and 4; and in the remainder of the state, 26 and 5.

Nitrogen Dioxide (NO_2). Concentrations of NO_2 decreased substantially in Illinois during the 1980s. Table 2 shows decreasing trends for 1-hr and 24-hr maxima, as well as annual mean NO_2 concentrations, both statewide and in the Chicago area. The trends range from 4.4 percent to -7.1 percent per year, and all are significant at the 1 percent level. There were not enough sampling sites in the other areas of the state to justify testing. Box plots for 1-hr maximum, 24-hr

maximum, and annual mean NO_2 are shown in figures 4-6, respectively. They illustrate the decreases, during the 1979-1990 period, in NO_2 concentrations both statewide, and in the Chicago area, over all three averaging times. Although only the trend of the median has been tested statistically (table 2), it is apparent that concentrations associated with all the percentiles depicted by the box plots have decreased. The same is true for the highest individual values each year (plotted as open circles).

No national or state standards exist for 1-hr or 24-hr maximum NO_2 concentrations. Numerous concentrations above the annual mean NO_2 standard (0.053 ppm) occurred in the late 1970s, but none have occurred since 1980.

Almost all of the NO_2 measurement sites are in the Chicago area, so it is impossible to look for differences in concentrations between geographic regions.

The number of sites measuring valid 1-hr NO_2 concentrations has increased recently, both statewide and in the Chicago area, from a low of four sites in the early 1980s to about 15 sites in 1990. The number of sites with valid 24-hr and annual mean NO_2 concentrations has decreased from 36-47 sites in the late 1970s to 10-15 sites in 1990.

Ozone (O_3). All pollutants are affected to some extent by weather conditions. Poor dispersion conditions caused by low wind speeds or calm conditions can allow pollutant concentrations to build up in local areas near emission sources, and long-range transport can carry pollutants from their sources for deposit at distant locations. For O_3, which is produced in the atmosphere by reactions between hydrocarbons and nitrogen oxides, however, weather conditions—especially temperature—affect how much of the pollutant is produced. Thus, it is desirable to remove the effect of weather conditions when looking for trends in O_3. We first discuss O_3 concentrations as observed, and then use a statistical regression approach to look for trends in O_3 *after the effect of temperature has been removed.*

Table 2 indicates that no significant trend could be detected in the median of 1-hr maximum O_3 concentrations in any geographic region of Illinois. The box plots of 1-hr maximum values by geographic area are shown in figure 7. Year-to-year variations are apparent, including higher concentrations statewide and in the Chicago area in 1983 and 1988, when abnormally high temperatures and little rain occurred during the summer

ozone season, but overall the visual impression of no trend toward higher or lower concentrations agrees with table 2.

The Chicago and Metro East areas clearly had higher median concentrations during the data period, compared to the remainder of the state, and these two urban centers also account for all the the highest individual site values statewide (open circles).

The national and state standard for 1-hr maximum O_3 concentration is 0.12 ppm. It is apparent that numerous violations of the standard occur most years in Illinois, although only two sites exceeded the standard (one time each) in 1990. Weather conditions over much of Illinois, and especially in the Chicago area, during the summer of 1990 were cooler, cloudier, rainier, and windier than normal (IEPA, 1991). These conditions undoubtedly contributed to the lower O_3 concentrations observed.

The test for differences between regions showed that mean concentrations in the Chicago and Metro East areas were each significantly (5 percent level) greater than that in the remainder of the state.

A number of attempts to account for effects of atmospheric conditions on O_3 concentrations have been published (e.g., Korsog and Wolff, 1991; Shively, 1991). Some of the past work has been quite sophisticated in searching for the best meteorological parameters to use in predicting O_3 concentrations, and in the use of statistical methods to characterize the relationship. In contrast, the following attempt to account for the effects of meteorological conditions on O_3 in Illinois is relatively simple. It uses only mean surface temperature during the O_3 season (April-October) in a regression equation involving a constant, a temperature term, and a time term. The model is:

$$[O_3] = \text{Constant} + \beta_1 \cdot (\text{Time}) + \beta_2 \cdot (\text{Temperature}) \quad (1)$$

where $[O_3]$ is ozone concentration, and β_1 and β_2 are coefficients of time and temperature, respectively.

The procedure is to evaluate the coefficients of the time and temperature terms using observed O_3 and temperature data, and to observe whether the coefficient of the time term is significantly different from zero. If it is, then there is a significant trend with time after the effect of temperature on O_3 concentrations has been removed. The model was evaluated for each of

the four geographical areas defined above. The O_3 data were the medians of the annual distribution of 1-hr maximum O_3 concentrations over all measurement sites in each geographical region. Corresponding April-October mean temperatures were obtained for each region from the Midwestern Climate Information System (Kunkel et al., 1990). Time was sequential year number, beginning with 1 in 1978. The regression analyses were carried out using the Multiple General Linear Hypothesis (MGLH) regression routine in the SYSTAT statistical software (Wilkinson, 1990). Results are shown in table 3.

The first thing to notice about the results is that the temperature coefficient is positive, indicating a positive correlation between O_3 and temperature, as expected, in all four geographical regions. The temperature coefficient is significantly different from zero (5 percent level) over the whole state, and also in the "remainder" region, significant at the 10 percent level in the Metro East region, and just barely misses the 10 percent significance level (P = 0.1011) in the Chicago area.

The coefficient of the time term is negative, indicating a downward trend in concentrations, in all four regions. The downward trend, after accounting for the effects of surface temperature, is significant at the 5 percent level over the whole state and in the Chicago area, and at the 10 percent level in the other two regions. (Note: although not shown in table 2, a -1.9 percent per year downward linear trend in O_3 significant at the 10 percent level was detected in the O_3 concentrations as observed.)

A further indication of the importance of the time term is the improvement in the squared multiple correlation (R^2 or the percent of the total variance explained by the regression) as the time term is added to the regression model. The model was evaluated separately without the time term to obtain R^2 values with and without the time term in each region. Results are shown in table 4. There is a substantial improvement in the percent variance, explained with the addition of the time term in each region—from 26 to 58 percent for the whole state, from 5 to 46 percent in the Chicago area; from 16 to 42 percent in the Metro East area, and from 40 percent to 55 percent in the remainder of the state.

The results regarding ozone time trends may be summarized as follows. Without accounting for temperature effects, no trends were found at a 5 percent significance level, but a 1.7 percent per year downward trend in concentrations, significant at the 10 percent level, was detected in the Chicago area (not shown in

Table 3. Results of Regression Analysis To Test Significance of O_3 Time Trend after Accounting for Effects of Average Surface Temperature

Geographic region	Variable[*]	Coefficient	P (2-tail)
Whole state	Constant	-0.30400	0.0774
	Time	-0.00177	0.0196
	Temperature	0.00565	0.0186
Chicago area	Constant	-0.36658	0.2236
	Time	-0.03108	0.0194
	Temperature	0.00707	0.1011
MetroEast area	Constant	-0.28673	0.2287
	Time	-0.00212	0.0605
	Temperature	0.00548	0.0811
Remainder of state	Constant	-0.20043	0.0651
	Time	-0.00073	0.0958
	Temperature	0.00401	0.0098

Note:
[*]Temperature = mean surface temperature, April through October, in the region.

Table 4. Comparison of R^2 Values (Explained Variance) in Regression Equations Evaluated with and without Time Terms

Geographic region	R^2 (without time)	R^2 (with time)
Whole state	0.260	0.582
Chicago area	0.048	0.464
Metro East area	0.165	0.423
Remainder of state	0.399	0.551

table 3). After accounting roughly for temperature effects, downward trends were detected at the 5 percent level statewide and in the Chicago area, and downward trends significant at the 10 percent level were detected in the other two regions. For evaluating potential effects of ozone on agricultural crops, concentrations in the non-urban "remainder" region would be the most relevant.

Sulfur Dioxide (SO_2). Table 2 shows downward trends significant at the 1 percent level for 3-hr maximum SO_2 in the Chicago area (-3.5 percent per year), and for annual mean SO_2 over the whole state (-2.6 percent per year). The table also shows a downward trend significant at the 5 percent level for annual mean SO_2 in the Chicago area (-5.2 percent per year). No trends significant at 5 percent or better were detected for any of the three types of observations in any region of the state; note, however, that the number of observing sites in the Metro East area was insufficient to estimate a trend.

Box plots for 3-hr maximum SO_2 are shown in figure 8. Any trends are difficult to perceive by the naked eye, despite the highly significant trend detected in the Chicago area (table 2). Comparison of SO_2 concentrations between regions shows that the highest 3-hr maximum SO_2 values observed statewide occurred in areas *other than* the Chicago area. There is no national or state *primary* standard for 3-hr maximum SO_2. The *secondary* national standard for 3-hr maximum SO_2 is 0.5 ppm. Figure 8 shows that this standard has been exceeded on a few occasions during the 1978-1990 period. The test for differences between regions found concentrations in the Chicago area significantly lower than those in the rest of the state. The number of sampling sites with valid data for 3-hr maximum SO_2 has remained very constant during the period.

Box plots for 24-hr maximum SO_2 are shown in figure 9. No trends are apparent to the naked eye, and no significant trends were detected in table 2. Again, the Chicago area experienced somewhat lower median concentrations than the remainder of the state, which in this case includes the Metro East area. The difference was signif-icant at better than the 1 percent level. The national and state primary standard for 24-hr maximum SO_2, 0.14 ppm, has been exceeded a few times state-wide in most years between 1978 and 1990; but only four of these occurred in the Chicago area. Numbers of sites with valid data fell by a factor of about 2 state-wide over the 1978-1990 period. Most of this reduction oc-curred in the Chicago area, which had 47 sites with valid data in 1979, but only 11 sites in 1990. Most of this reduction is the result of closing sampling sites, rather than active sites not meeting completeness criteria.

Figure 10 shows box plots for annual mean SO_2 concentrations. Decreases in median concentrations statewide are significant at the 1 percent level and in the Chicago area at the 5 percent level (table 2). Regional maximum concentrations (highest open circle) also appear to be on downward trends, at least for the "whole state" and "remainder" regions. There were no significant (5 percent) differences in concentration between geographic regions. Numbers of sites with valid data have also decreased over the 1978-1990 period, statewide by about 50 percent, and by about 70 percent in the Chicago area.

Particulate Mass (TSP, PM_{10}). The state standard for particulate mass is written for total suspended particulate matter (TSP), while the national standard is for PM_{10}, or particulate matter up to 10 μm in aerodynamic diameter.

Box plots for 24-hr maximum TSP concentrations are shown in figure 11. Median values have remained relatively constant over the 1978-1990 period with occasional anomaly years such as 1983, when spring dust storms in east central and northeastern Illinois (IEPA, 1984) caused numerous measurements > 500 μg/M^3. No significant trends in median concentrations were detected (table 2) in any of the four geographic regions for the entire 1978-1990 period, but careful examination of the individual regional plots suggests a decreasing trend for the first half of the 1980s followed by an increasing trend in the later half, at least in some regions.

It is important to know whether these recent concentration increases reflect actual regional environmental conditions, or whether they are an artifact of the year-to-year changes in the measurement network. Numbers of sites with valid samples have dropped sharply in all regions in recent years, and if the sites removed from the network were systematically in cleaner areas (which might be expected), then average concentrations at the remaining sites would be higher. This question could be investigated by comparing records of concentrations at removed sites and at remaining sites.

The test for differences between regions found none (5 percent level), but some violations of the standard have occurred each year in all regions.

Trends in annual geometric mean TSP concentrations are shown in figure 12. No overall significant trends were detected (table 2), but the pattern of decreasing concentrations up to about 1985, followed by increases

through 1989, is clear in all four regions. Again, however, the reality of the increase in the late-1980s is in doubt because the number of sites with valid data declined sharply during the 1986-1990 period. Comparison of concentrations between regions showed that concentrations in the Metro East area were significantly (1 percent level) higher than those in both the Chicago area and the remainder of the state. In addition, visual examination suggests that the highest individual site values occurred in the Metro East region.

The first published data for PM_{10} are for 1987 (IEPA, 1988), after the U.S. EPA changed its particulate matter standard from TSP to PM_{10}, although some measurements were being made in anticipation of the new national standard as early as 1984 (IEPA, 1988). Because data were available for only four years, no evaluation of PM_{10} trends was included in table 2. However, box plots for 24-hr maximum and annual mean PM_{10} are shown in figures 13 and 14, respectively.

Figure 13 shows a number of violations of the 150 µg/M^3 standard for 24-hr maximum PM_{10} in both the Chicago area and other areas of the state. The number of sampling sites increased in all three geographic areas shown in the figure during the 1987-1990 period.

Figure 14 also shows a few concentrations above the 50 µg/M^3 standard statewide, none of which occurred in the Chicago area.

This concludes the presentation of data on the criteria pollutants. There are currently no standards for the remaining pollutants, which all occur as particles in the atmosphere, and are collected on high-volume filters.

Filter Analyses: Nitrate, Sulfate, and Metals

Through 1984, annual mean concentrations of nitrate, sulfate, and metals were derived from analyses of filters composited monthly. Thus, distributions of annual means are available from 1978-1990, but distributions of 24-hr maximum concentrations are available only after 1985, when measurements of individual 24-hr filters began.

Since all of these analyses depend on the collection of filter samples, it is appropriate to discuss changes in the number of valid samples that apply to all of the following analytes. Of course there are small individual differences between pollutants in the number of valid samples, but the major trends are determined primarily by the number of active sampling sites.

Statewide, the number of sampling sites collecting filter samples dropped from >100 in 1978 to <20 in 1990; the change from 1985 to 1990 was from about 50 sites to <20. In the Chicago area the drop in sites was from >70 in 1978 to 35-40 in 1985 to <10 in 1990. In the Metro East area, there were ≥15 sites up to 1981 and ≤5 after 1985. The numbers for the remainder of the state were >25 sites up to 1981 and 4-6 thereafter.

Nitrate Ion (NO_3^-). Box plots for 24-hr maximum and annual mean NO_3^- concentrations are shown in figures 15 and 16, respectively. High year-to-year variability in the median (and other box plot percentiles) 24-hr maximum NO_3^- concentrations (figure 15) is apparent, particularly in the Metro East and "remainder" areas, where the plots are based on <10 sites.

For annual mean NO_3^-, the only significant trend (1 percent level) was a -2.1 percent per year decreasing trend in the "remainder" region (table 2). Figure 16 shows a relatively steady median concentration of about 5 µg/M^3 statewide and in the Chicago area, and somewhat lower concentrations, with more year-to-year variability in the Metro East and remaining areas of the state. The comparison between regions found the Chicago area concentrations significantly (5 percent or better) higher than those in the Metro East area and the remainder of the state.

Both statewide and in the Chicago area, the variability within individual years, as measured by the interquartile (75th-25th percentile) range, or the height of the "box" in the box plot, appears to have decreased in recent years. Again, however, this occurred along with a steep decline in the number of sites with valid data, so the smaller variability may simply reflect the fact that measurements are occurring at fewer locations.

Sulfate Ion (SO_4^{2-}). Box plots for 24-hr maximum and annual mean SO_4^{2-} are shown in figures 17 and 18, respectively. Here also relatively high year-to-year variations in 24-hr maxima occurred, particularly in years or geographic areas with <10 sites.

Table 2 shows that declines in annual mean SO_4^{2-} concentration significant at the 5 percent (whole state and Chicago area) or 2 percent (remainder of the state) levels occurred in all areas other than the Metro East area. The significant linear trends were all between -1.3 and -1.6 percent per year.

Concentrations were significantly (1 percent level) higher in the Metro East area than in the Chicago and "remainder" regions of the state.

Arsenic (As). Box plots for 24-hr maximum and annual mean As are shown in figures 19 and 20, respectively. Except for the Metro East area, where median 24-hr maximum concentrations >0.01 $\mu g/M^3$ occurred in four of the six years of record, the median 24-hr maximum was <0.01 $\mu g/M^3$ statewide. One value >0.5 $\mu g/M^3$ was observed in the Metro East area in 1990.

Differences between geographical regions are the most interesting feature of these plots. Comparison of the plots for the various regions shows clearly that the Metro East area accounts for the highest measured values statewide; this is true for both maximum 24-hr concentrations as well as mean annual concentrations. The statistical test for differences between regions was not run on the 24-hr maximum As data because of the relatively few years of data. For annual mean As, the test found that concentrations in the Metro East area were higher than those in the Chicago and "remainder" areas at the 0.1 percent significance level.

Table 2 shows declines in As concentrations of -8.4, -11.1, and -9.5 percent per year in the whole state, the Chicago area, and the "remainder" region, respectively. All are significant at the 1 percent level.

Cadmium (Cd). Box plots for 24-hr maximum and annual mean Cd are shown in figures 21 and 22, respectively. Again, differences between geographical regions are clear, and again the Metro East area accounts for most of the highest values that were observed statewide over both averaging times. The test for regional differences in annual mean concentration confirmed that the highest concentrations occurred in the Metro East area at a significance level of 0.1 percent. Table 2 shows that the decline of -12.7 percent per year in annual mean Cd in the "remainder" region was significant at the 2 percent level.

Chromium (Cr). Figures 23 and 24 show box plots of 24-hr maximum and annual mean Cr, respectively. Note that Cr was not measured on filter samples until 1985. The number of sites in the Metro East area was insufficient for a separate plot, so the Metro East sites are included in the "remainder" region. The marked differences between regions that were apparent for As and Cd are smaller or absent here. Rather than the Metro East area having the highest concentrations, Cr appears to be somewhat more prominent in the Chi-

cago area, although the differences were not statistically (5 percent) significant. No significant trends in the median annual mean Cr were detected (table 2).

Iron (Fe). Box plots for 24-hr maximum and annual mean Fe are shown in figures 25 and 26, respectively. For both averaging periods, it is again clear from the figures that the Metro East area accounts for the highest values statewide. The test for differences between regions confirmed that the highest annual mean Fe concentrations occurred in the Metro East area (0.1 percent level). Moreover, the trend test (table 2) de-tected an upward trend (6.0 percent per year) in the Metro East area, significant at the 2 percent level. While it is true that the two area sampling sites with the lowest annual mean concentrations were closed during the 1985-1990 period (one after 1985, and the other after 1988), this is not the cause of the increasing trend in concentrations. Concentrations at the remaining sites also increased during these years.

Manganese (Mn). Figures 27 and 28 show box plots for 24-hr maximum and annual mean Mn, respectively. Both the Chicago area and the Metro East area contributed to the highest values over both averaging periods observed statewide, but the Metro East area experienced noticeably higher median concentrations (figures 27 and 28). This was confirmed for annual means by the test for differences between regions, which found higher concentrations in the Metro East area compared to the Chicagto and "remainder" areas (0.1 percent level). As in the case of Fe, the trend test (table 2) detected an increasing trend (10 percent per year), significant at the 5 percent level, in annual mean Mn in the Metro East area. Also as in the case of Fe, the closing of sampling sites could have had only a minor effect on the increasing Mn concentrations over the 1985-1990 period.

Nickel (Ni). Box plots for 24-hr maximum and annual mean Ni are shown in figures 29 and 30, respectively. Ni was not measured on filter samples until 1984. Since the number of sites in the Metro East area was insufficient for a separate plot, the Metro East sites are included in the "remainder" region. For Ni, it appears that the Chicago area contributed most of the highest 24-hr maximum concentrations (figure 29). Comparing annual means between regions, it appears that the Chicago area has recently experienced somewhat higher median concentrations, but the statistical test found no significant (5 percent level) differences. Apparently, both regions contribute more or less equally to the highest annual means. The trend test (table 2)

found no significant trends in the median annual mean Ni.

Metal Enrichments, Relative to Soil. Enrichment factors are a common and convenient method of distinguishing between natural and anthropogenic sources of metals in the atmosphere. For many metals the typical natural source examined is crustal or soil aerosols. Enrichment, E, for element X is expressed as a ratio of ratios:

$$E = (X/RE)_{air}/(X/RE)_{soil} \qquad (2)$$

where RE is a reference element for which the soil or earth's crust is the dominant source in the atmosphere. Enrichments are usually evaluated for measured concentrations of X in air, using abundances (mass fractions) of X and RE in soils from compilations of global mean soil or crustal composition. Figure 31 shows element enrichments based on the median statewide annual mean concentrations of As, Cd, Cr, Pb, Mn, and Ni, using Fe as the reference element, for 1978-1990. If the ratio of the subject element to Fe in air is the same as the ratio of the same elements in soil, then the enrichment is 1.0, and the element X is assumed to come only from the soil. In practice, because of variations in soil composition, elements with enrichments between 1 and 10 are usually assumed to have primarily soil sources. Values of E >10 are assumed to indicate anthropogenic sources.

Using these criteria, figure 31 shows that Pb and Cd, with enrichments between 100 and 3000, have primarily anthropogenic sources in Illinois. Note that the enrichment of Pb has dropped from 1000-2000 in the early 1980s to 100-200 in recent years as Pb has been removed from automotive fuels. Nevertheless, the concentration of Pb currently in the atmosphere is far in excess of what would be there naturally from soil wind erosion. Sources such as Pb smelters and fly ash from coal burning may now be the major sources of Pb in the atmosphere, although Pb recycled from previously deposited auto exhaust may also be important.

Cd also has a high enrichment factor of about 1000, which has remained relatively constant during the 1980s. The absence of a Cd datapoint in 1981 is the result of a one-year hiatus in Cd measurements. Those in 1983 and 1989 were caused by median Cd concentrations of 0.000 $\mu g/M^3$.

Arsenic enrichments are mostly >10 also, suggesting a mostly anthropogenic source. They appear to be de-

creasing somewhat, in agreement with the decreasing trends in concentration seen in table 2.

Mn enrichments are firmly in the 1-10 range generally ascribed to natural sources, with very little year-to-year variation. The majority of Mn in the Illinois atmosphere may well be natural. Mn sources also tend to be Fe sources, however, so the extent to which the reference element Fe is nonnatural, may also hold true for Mn. For example, anthropogenic sources are very likely to be the cause of the significant increase (at the 5 percent level, or better) in concentrations of both Fe and Mn during the 1980s in the Metro East area.

The year-to-year variations in Cr and Ni enrichments during the 1985-1990 period are similar to each other, but unlike those of any of the other metals in figure 31. The enrichments are mostly in the 1-10 range, suggesting mainly natural sources for these metals, but the sometimes large year-to-year variations suggest that concentrations of both of these metals are enhanced by anthropogenic sources at least during some years.

SPATIAL DISTRIBUTIONS OF POLLUTANT CONCENTRATIONS

It was a very early goal of this summary of Illinois air quality to draw spatial contours of pollutant concentrations on a statewide scale. Examination of the data quickly dashed that early hope, however, as it became clear that sampling sites were concentrated in the two major industrial and population centers, the Chicago and the Metro East areas. The relatively few sampling sites not in these major centers were clustered in smaller cities and the environs of troublesome sources. Data are not available from sparsely populated regions with no major sources. For an agency such as the IEPA with the mission of documenting problems and enforcing standards, this sampling strategy is obvious. It does not provide data needed to document statewide air quality, however.

For this report, we propose that the data are adequate to show spatial variations in that portion of the state with the highest concentration of sampling sites, the Chicago area. To provide a sense of the temporal changes in Chicago-area spatial patterns, contours have been drawn for 1980, 1985, and 1990, and are presented below. The rapid drop in the number of sampling sites, even in the Chicago area, makes the 1990 contours very problematic. The 1990 contours are meant to suggest only the most rudimentary spatial
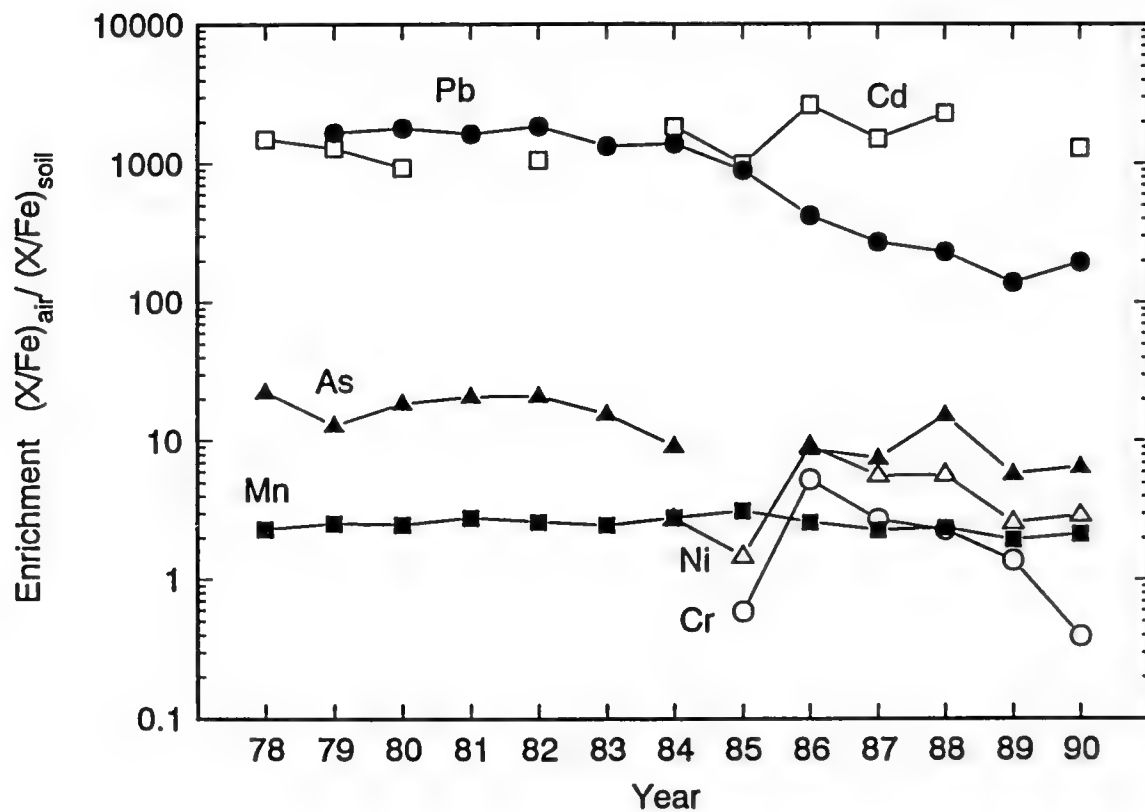
Figure 31. Trends in airborne metal enrichment, relative to soil, in Illinois.

patterns. For most pollutants the data are not adequate to do more.

The data and associated discussions for the several pollutants appear in the same order as those of the temporal trends above.

Criteria Pollutants

CO. Figures 32 and 33 show spatial distributions of 1-hr maximum and 8-hr maximum CO concentrations in the Chicago area in 1980, 1985, and 1990. The low density of sampling sites, and the changes in site locations between years preclude identification of persistent locations of high or low concentrations. In 1980 one sampling site near the lake shore had a 1-hr maximum >20 ppm. In 1985 three sites had 1-hr maxima between 10 and 20 ppm, but there were no measurements >20 ppm. In 1990, only one site recorded a 1-hr maximum >10 ppm. This general decline in concentrations agrees with the significant overall decline in Chicago-area concentrations detected by the trend test (table 2). No violations of the 35 ppm national and state standard for 1-hr maximum CO were observed.

Figure 33 shows spatial patterns of 8-hr maximum CO concentrations. Again, temporal changes in the sampling network limit discussion mostly to temporal changes in concentrations. In 1980, one lakeshore site exceeded the national and state standard for 8-hr maximum CO (9 ppm), but no violations were observed in the region in 1985 or 1990. A general decline in concentrations is apparent from 1980 to 1985 to 1990 that agrees with the significant decline detected in the Chicago area from the full 1978-1990 dataset (table 2).

Pb. Spatial distributions of annual mean Pb concentrations in the Chicago area for 1980, 1985, and 1990 are shown in figure 34. Persistent concentration maxima occurred in the central city and the southeast Chicago industrial area; however, the well-known and very significant decline (table 2) in atmospheric Pb concentrations during the 1980s is evident here as well. The value of the highest contour line declined from 0.6 µg/M^3 in 1980 to 0.2 µg/M^3 in 1985 and 0.1 µg/M^3 in 1990. Pb standards are written in terms of mean concentrations over calendar quarters, which cannot be inferred from the data shown here. See the earlier discussion related to temporal trends.

NO$_2$. Spatial patterns of 24-hr maximum and annual mean NO$_2$ in the Chicago area are shown in figures 35 and 36, respectively. In 1980 the highest 24-hr maxima occurred in suburban northwestern (DesPlaines) and southern (Flossmoor) Cook County, with single sites in each area experiencing one or more days with mean concentrations >0.14 ppm. By 1985, the two sites with the highest values in 1980 were no longer active. The data suggest an overall decline in maximum 24-hr concentrations, with the highest value (0.094 ppm) occurring in the city center. The 1990 data show a further overall decline in maximum 24-hr concentrations, with the maximum (0.062 ppm) occurring in a cluster of sampling sites near O'Hare Airport. There are no national or state standards for 24-hr maximum NO$_2$.

Figure 36 shows Chicago-area contours of annual mean NO$_2$. Shaded areas in the northwest suburbs (DesPlaines) and the central city exceeded the national and state primary standard (0.053 ppm) in 1980. Although no violations of the standard were observed in 1985 and 1990, the available data suggest that the highest annual mean concentrations still occurred in the central city area near Lake Michigan. The apparent decreasing concentrations agree with the significant trend in Chicago area annual mean NO$_2$ discussed earlier (table 2).

O$_3$. Figure 37 shows contours of maximum 1-hr O$_3$ concentrations in the Chicago area for 1980, 1985, and 1990. Most of the Chicago area was in violation of the 0.12 ppm standard in 1980, with a conspicuous area of lower values across the middle of Cook County and the city of Chicago. In 1985 the violations of the standard were confined to the northeastern and southeastern portions of Cook County. No violations were observed in 1990; however, there is evidence that this may have occurred because summer weather conditions were relatively unfavorable for O$_3$ formation, as discussed earlier. Table 2 indicâtes no significant temporal trend in O$_3$ as observed; however, as shown earlier, when surface temperature is accounted for, there is a significant trend (2 percent level) toward decreasing concentrations in the Chicago area over the 1978-1990 period.

SO$_2$. Figures 38-40 show Chicago-area spatial contours for SO$_2$: 3-hr maximum, 24-hr maximum, and annual mean concentrations, respectively. The contours drawn from the available data are very difficult to interpret. Overall, 3-hr maximum concentrations were primarily >0.10 ppm in 1980 and 1985, and primarily <0.10 in 1990. This apparent trend toward decreasing concentrations agrees with the significant trend for the Chicago area detected earlier (table 2). A single high value (0.446 ppm) was observed in the western suburbs (Bedford Park) in 1985. There are no national or state standards for 3-hr maximum SO$_2$.
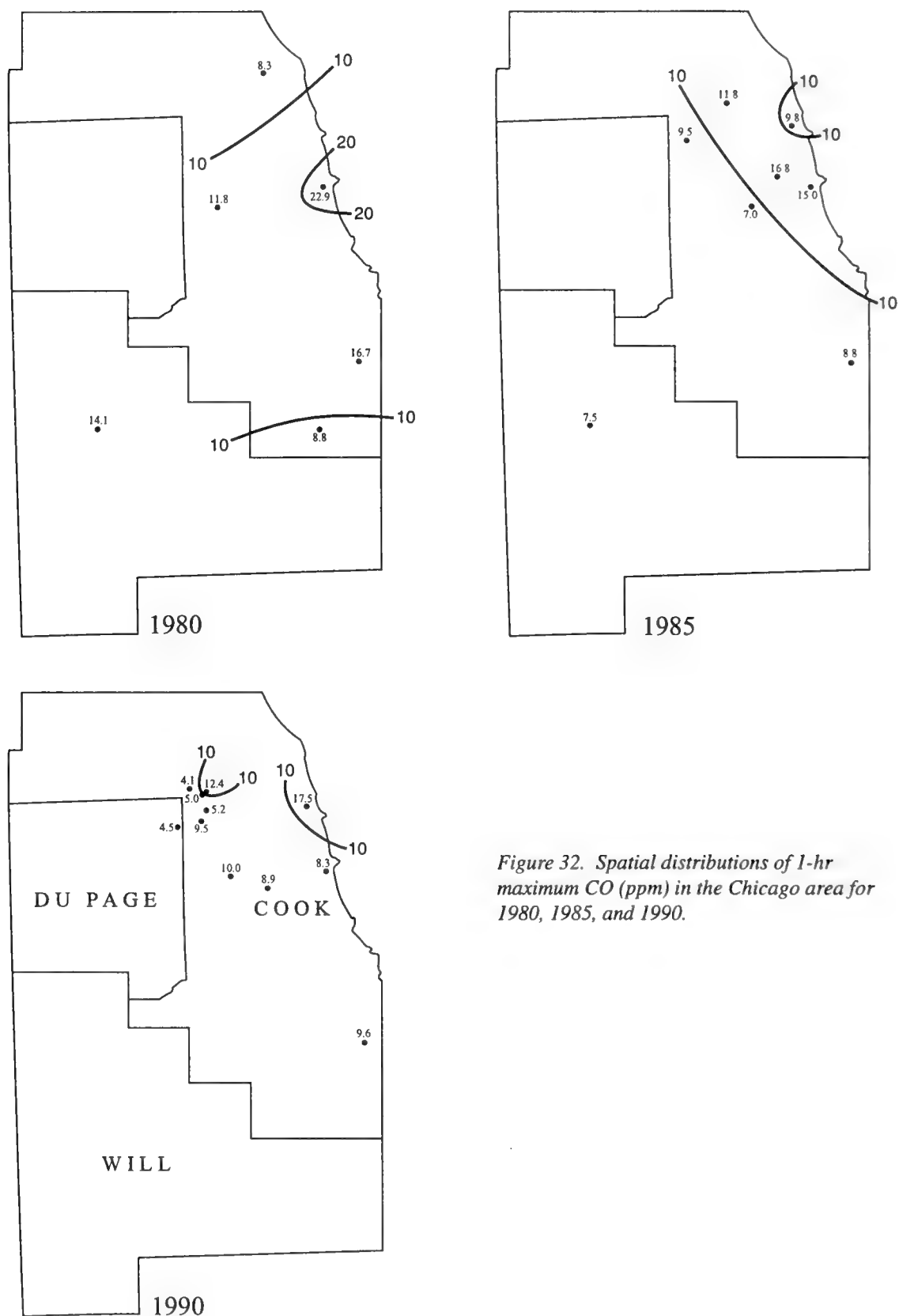
Figure 32. Spatial distributions of 1-hr maximum CO (ppm) in the Chicago area for 1980, 1985, and 1990.
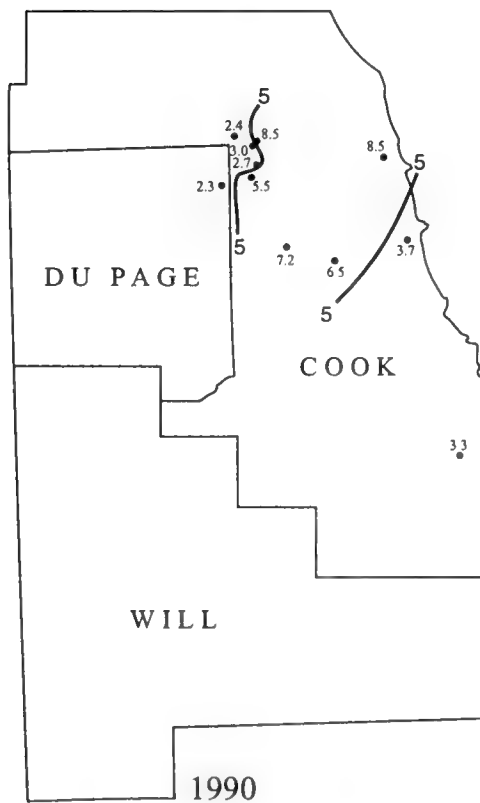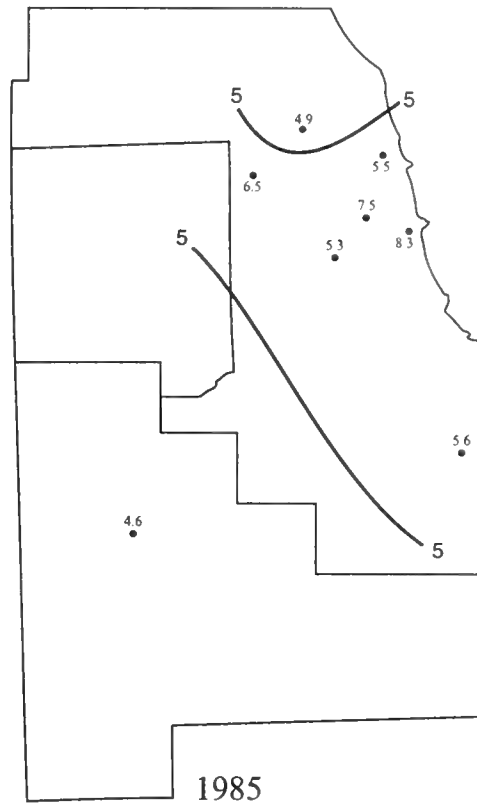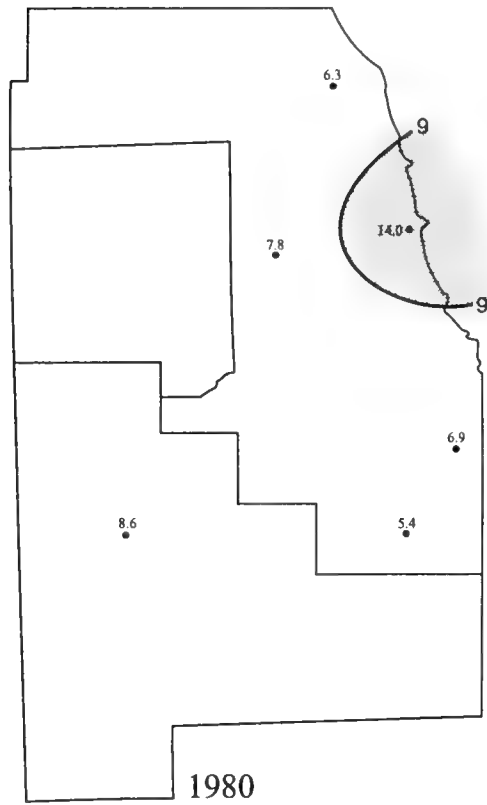
Figure 33. Spatial distributions of 8-hr maximum CO (ppm) in the Chicago area for 1980, 1985, and 1990. The shaded area denotes violation of an air quality standard.
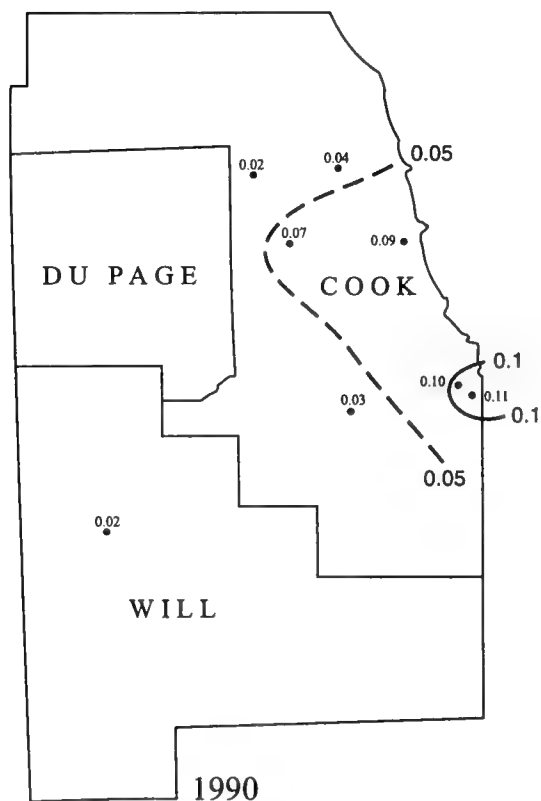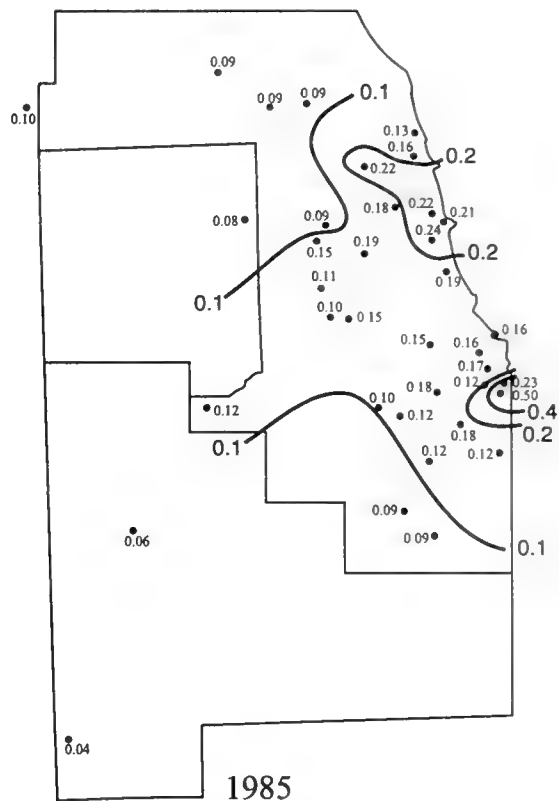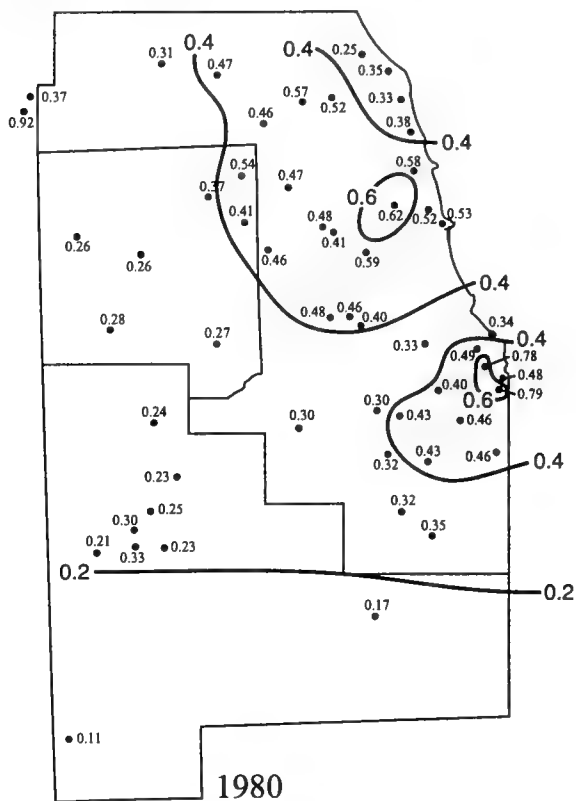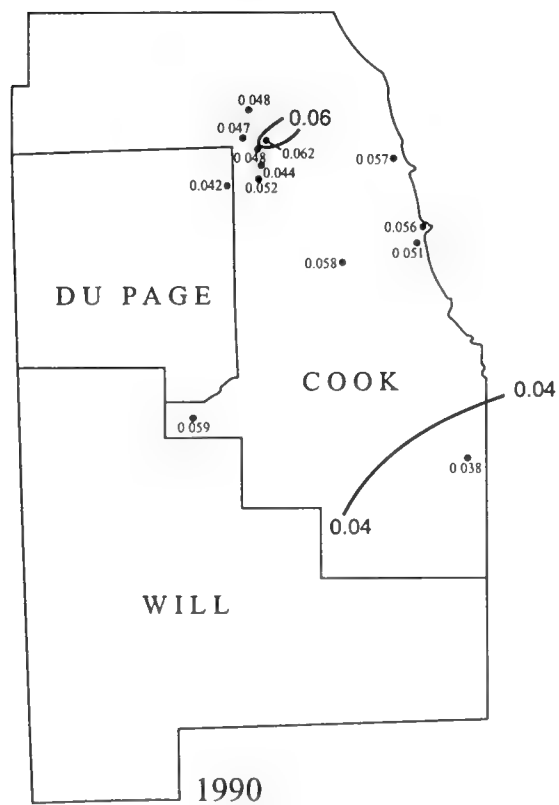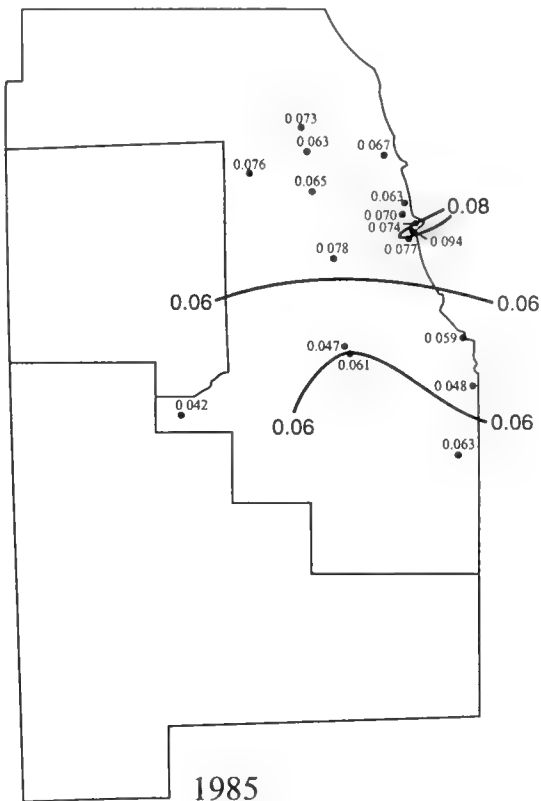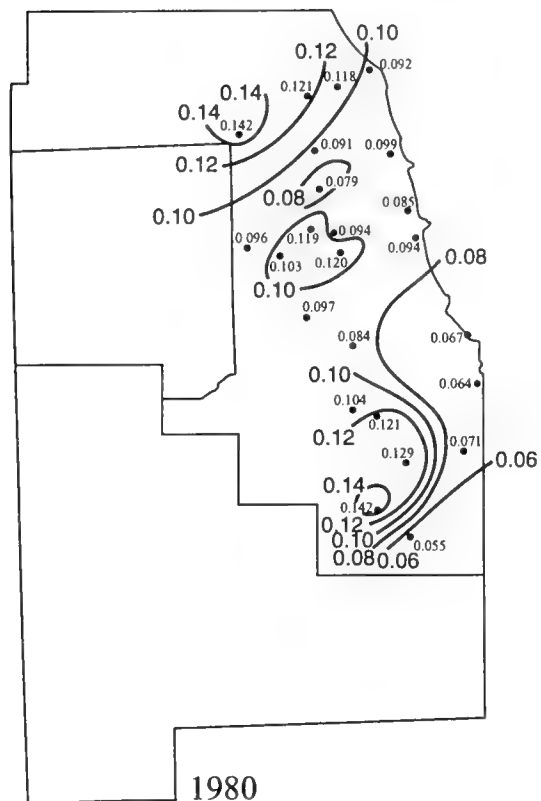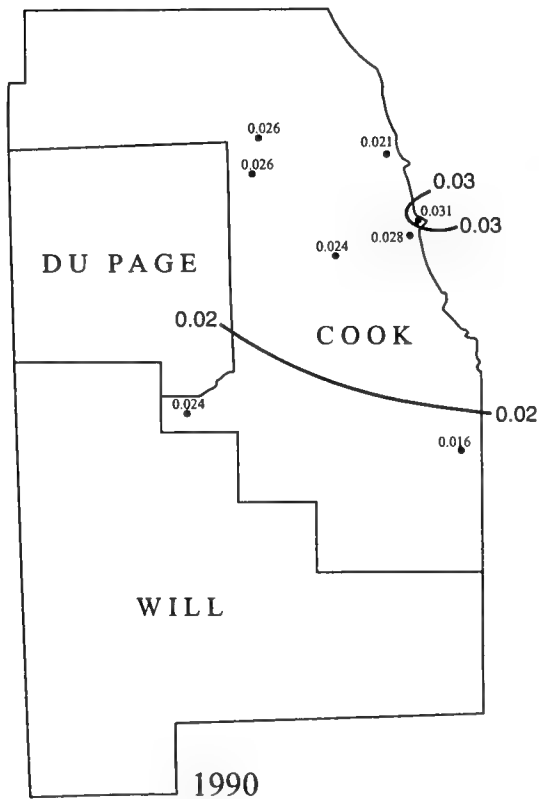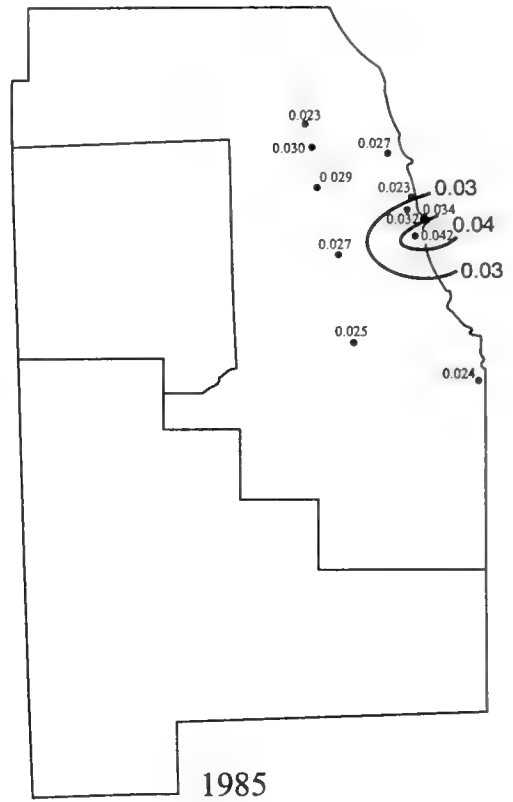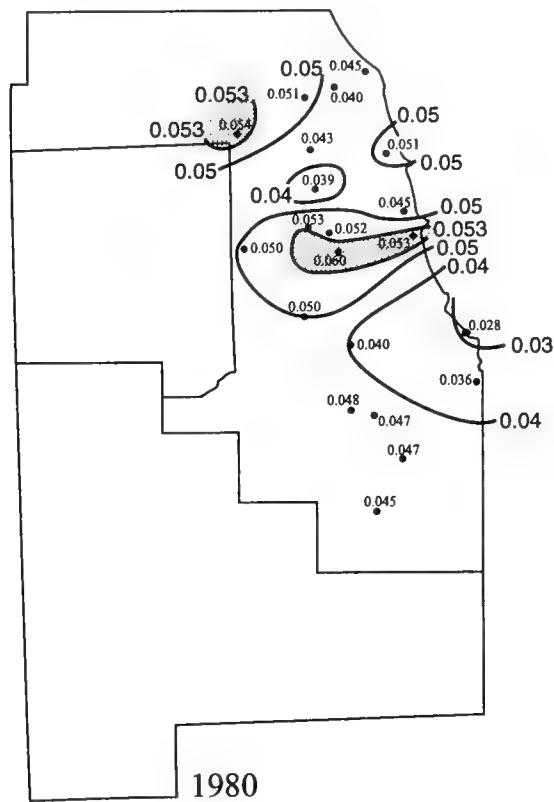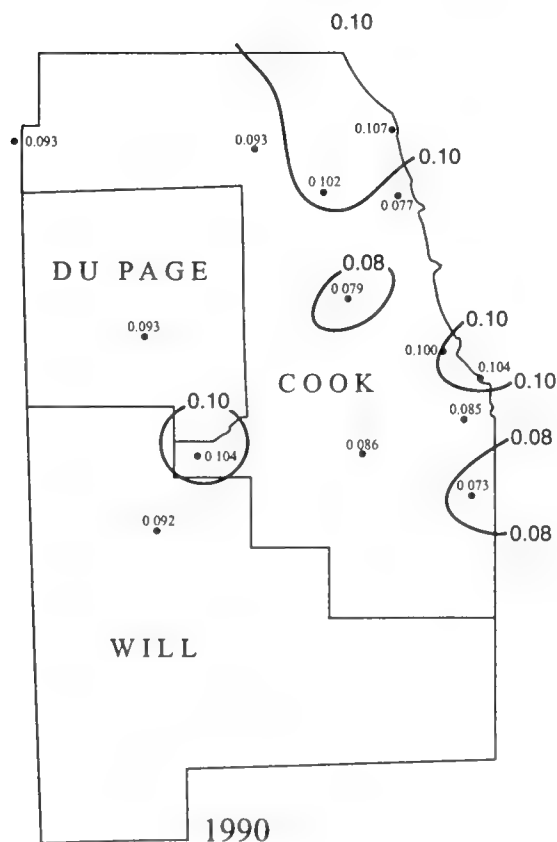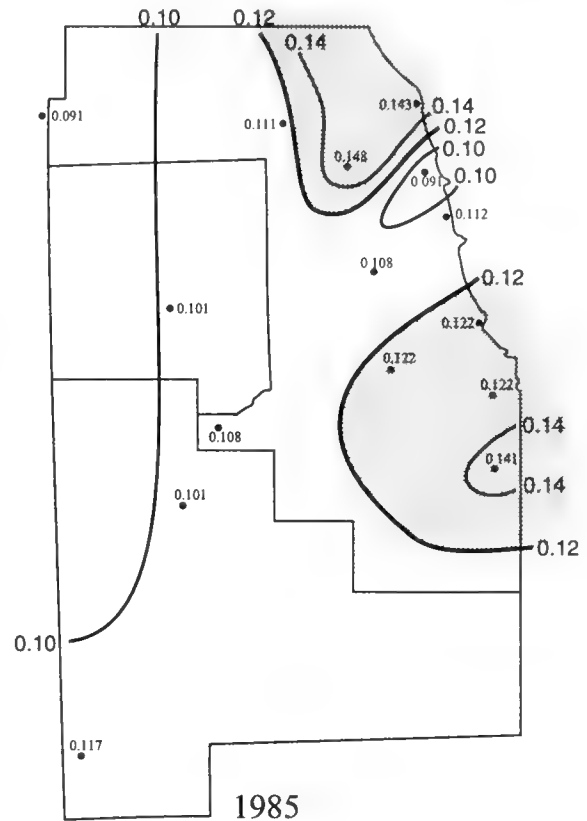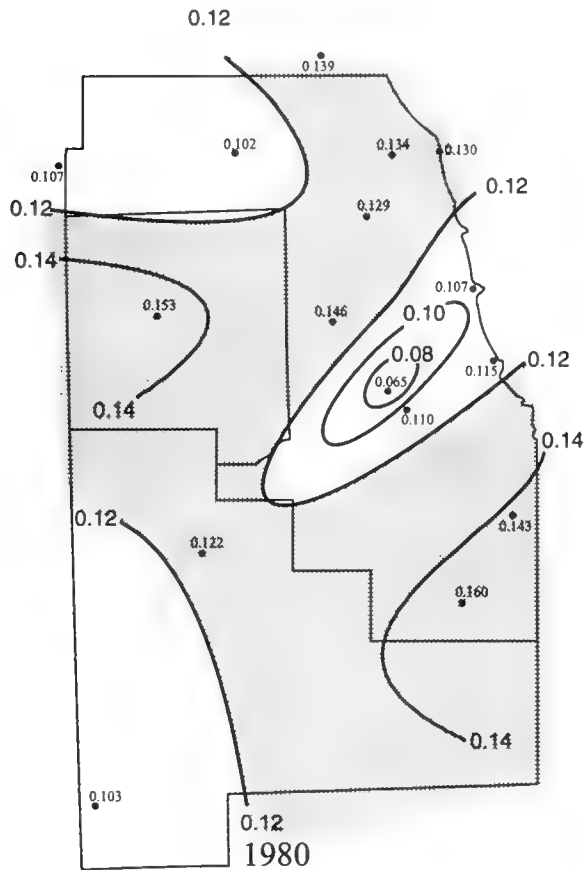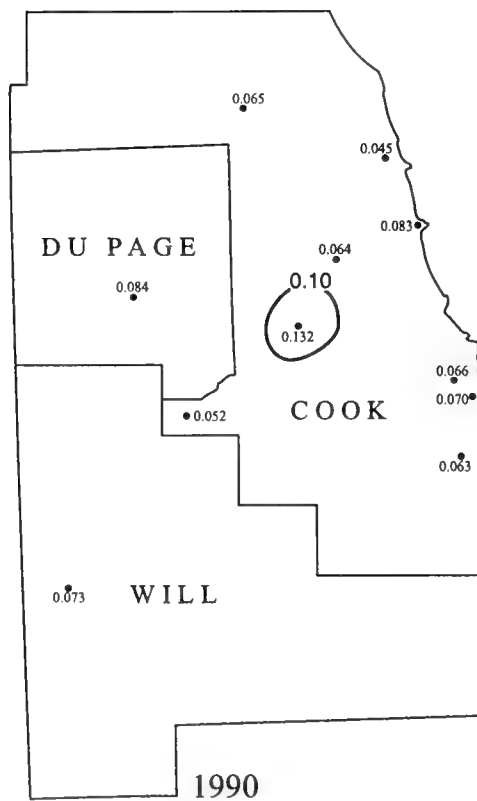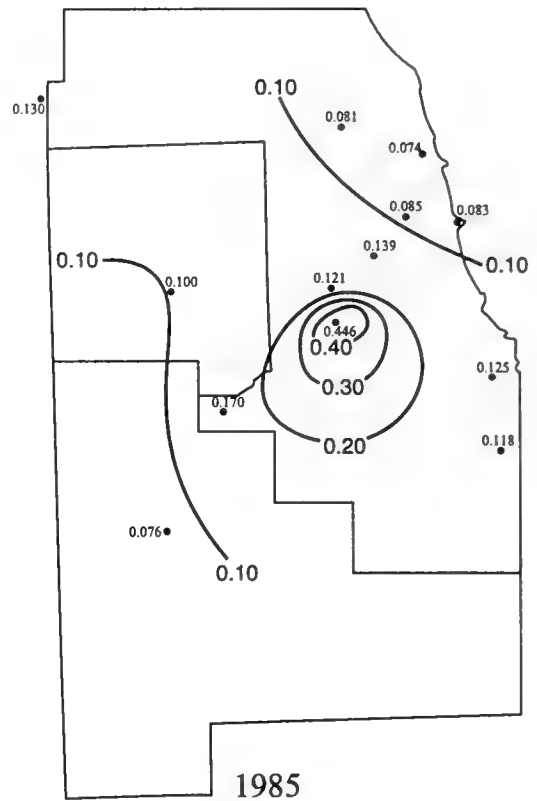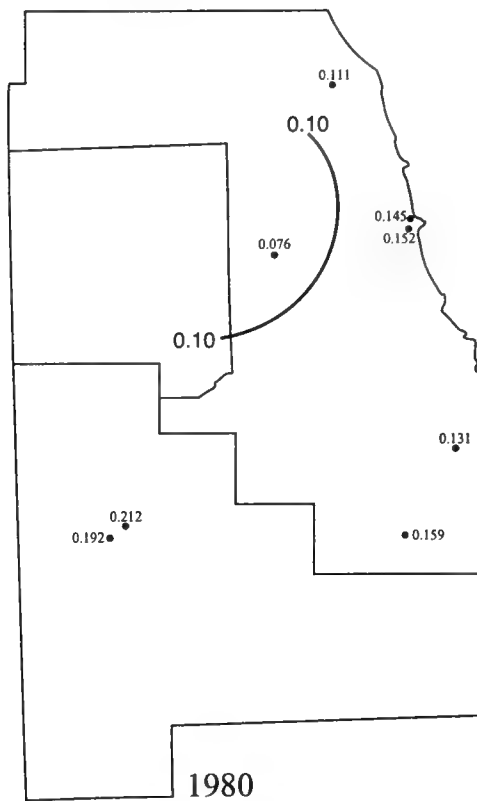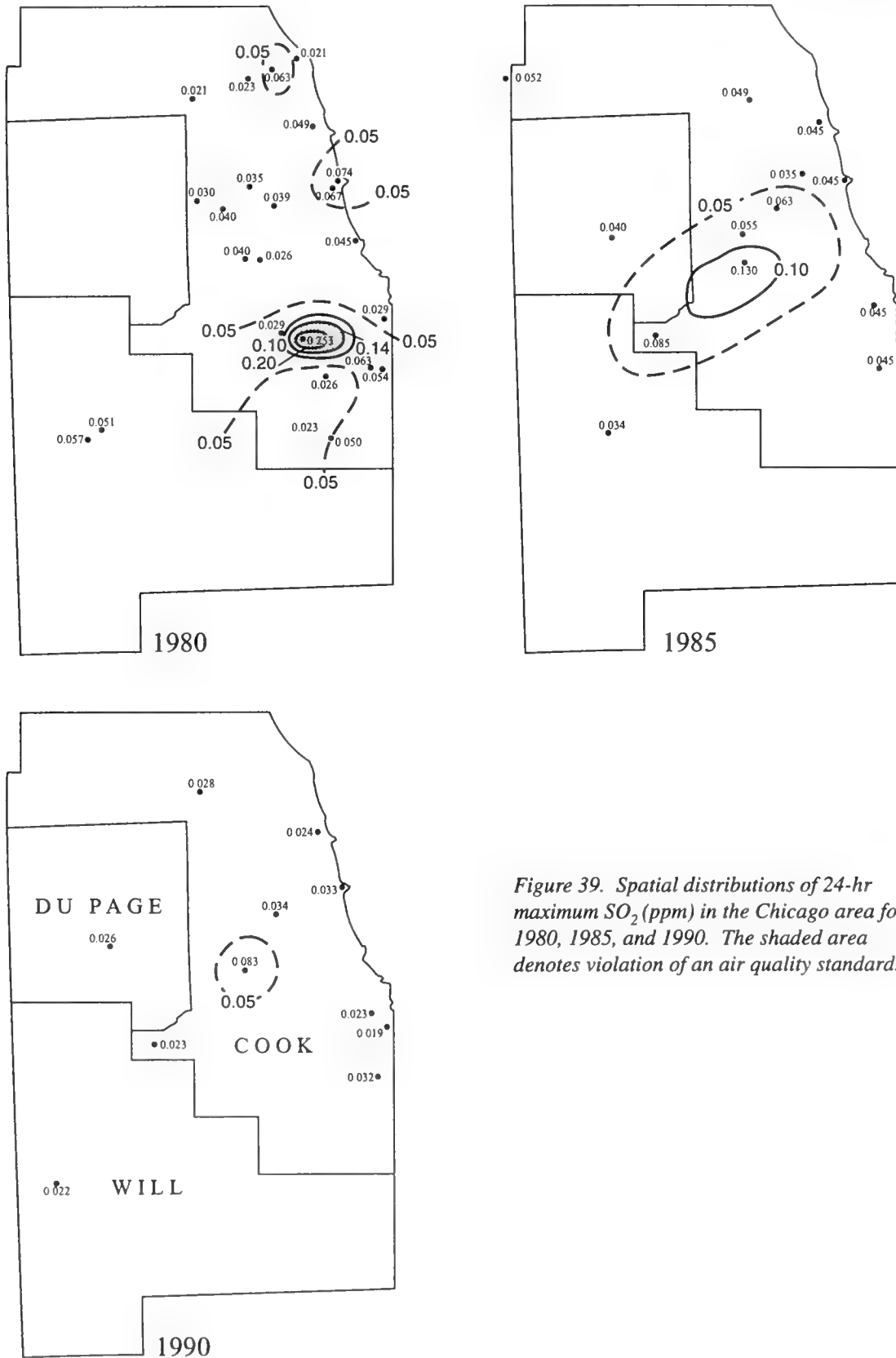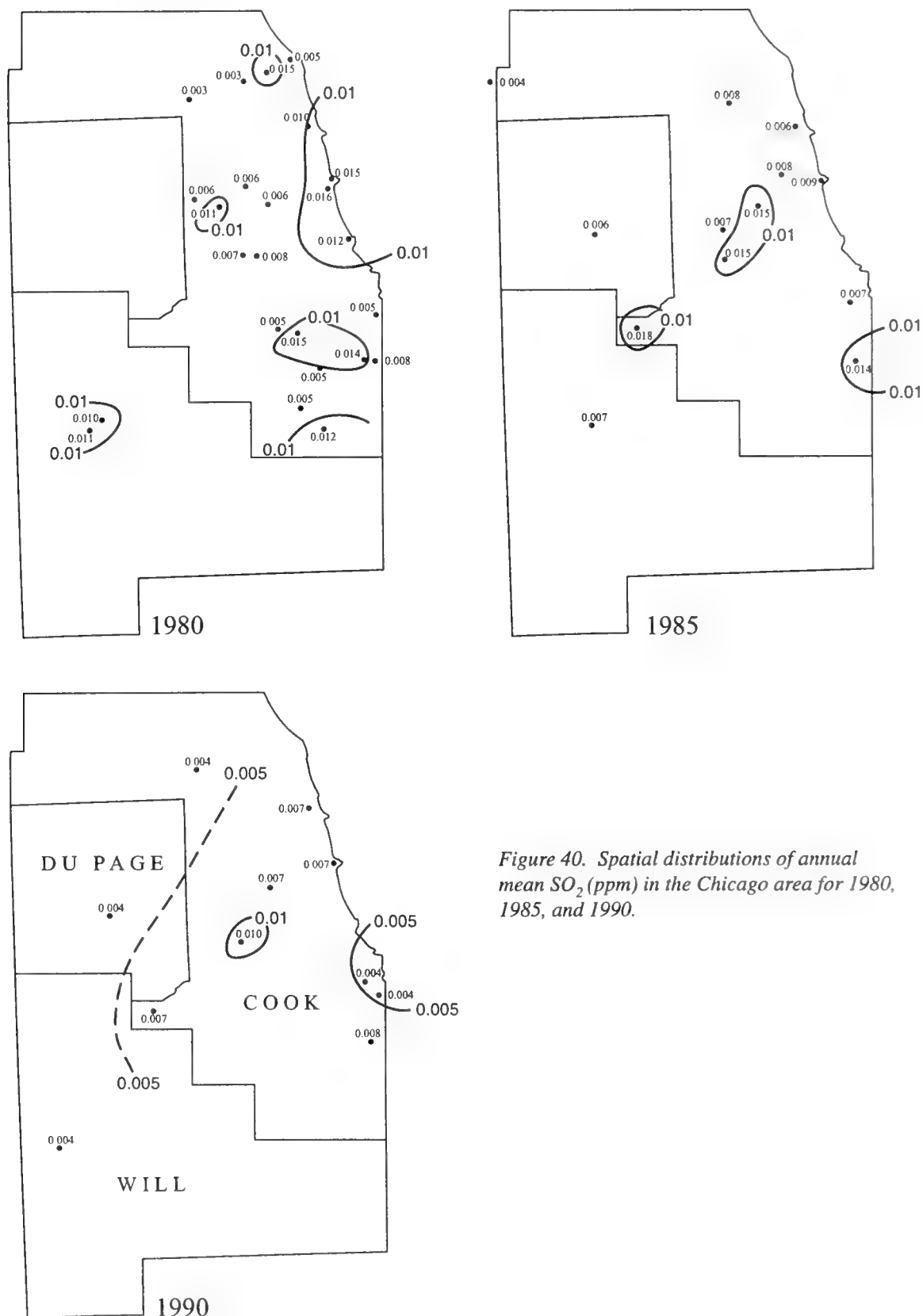
Figure 34. Spatial distributions of annual mean Pb ($\mu g/M^3$) in the Chicago area for 1980, 1985, and 1990.

Figure 35. Spatial distributions of 24-hr maximum NO$_2$ (ppm) in the Chicago area for 1980, 1985, and 1990.

Figure 36. Spatial distributions of annual mean NO$_2$ (ppm) in the Chicago area for 1980, 1985, and 1990. Shaded areas denote violations of an air quality standard.

Figure 37. Spatial distributions of 1-hr maximum O^3 (ppm) in the Chicago area for 1980, 1985, and 1990. Shaded areas denote violations of an air quality standard.

Figure 38. Spatial distributions of 3-hr maximum SO$_2$ (ppm) in the Chicago area for 1980, 1985, and 1990.

Figure 39. Spatial distributions of 24-hr maximum SO$_2$ (ppm) in the Chicago area for 1980, 1985, and 1990. The shaded area denotes violation of an air quality standard.

Figure 40. Spatial distributions of annual mean SO$_2$ (ppm) in the Chicago area for 1980, 1985, and 1990.

Chicago-area spatial contours for 24-hr SO_2 are shown in figure 39. One site in south-suburban Cook County (Blue Island) exceeded the national and state standard (0.14 ppm) in 1980. Aside from this extreme, concentrations at most locations were <0.05 ppm. The available data for 1985 and 1990 show maxima in the western suburbs (Bedford Park) in both years, but again most sites experienced 24-hr maximum concentrations <0.05 ppm. There were no violations of the standard in 1985 or 1990. Table 2 shows no significant temporal trends in 24-hr maximum SO_2 in the Chicago area.

Chicago-area spatial contours for annual mean SO_2 concentrations are shown in figure 40. The figure shows no strong spatial patterns in any of the three years, but very different patterns of minimal high and low concentrations over the three years. There were no violations of the national and state standard (0.03 ppm) for the annual mean in any of the three years.

TSP. Spatial patterns of 24-hr maximum and annual mean TSP in the Chicago area are shown in figures 41 and 42, respectively. The spatial pattern for 1980 shows areas of high concentrations, including violations of the 260 $\mu g/M^3$ standard, in (1) the industrial area of southeastern Chicago, (2) the western suburbs of Cook County (River Forest) and eastern DuPage County (Elmhurst), (3) northwestern Will County (Romeoville, Joliet, and Rockdale), and (4) southwestern Will County (Braidwood). In 1985 there were widespread violations of the standard, over almost all of Cook County, the southern half of DuPage County, and southwestern Will County On the other hand, there were no violations in the Joliet area. The widespread extent of the violations of the 24-hr standard suggests that a regional weather event might have been responsible, and indeed, a dust storm that occurred on May 31, 1985, was the cause of most of the violations (IEPA, 1986). In 1990 the much-reduced sampling network detected only one violation of the standard at Rockdale in Will County.

Figure 42 shows the spatial patterns for annual geometric mean TSP. In 1980 maximum concentrations above the national standard (75 $\mu g/M^3$) occurred in a broad band from the central city area of Chicago to suburban Summit, in the southeast Chicago industrial region, in northeast DuPage County (Bensenville), and in the Joliet area of Will County In 1985, only two sites in the Chicago area violated the standard, one in southeast Chicago and one in west suburban McCook. In 1990 only one site, in downtown Chicago, was in violation; however, the number of sampling sites was down considerably.

Filter Samples: Nitrate, Sulfate, and Metals

There are no standards for the remaining pollutants.

NO_3^-. Spatial patterns of 24-hr maximum and annual mean NO_3^- in the Chicago area are shown in figures 43 and 44, respectively. Data on 24-hr maximum NO_3^- are not available before 1985. Maximum 24-hr NO_3^- concentrations are relatively uniform over the area in both 1985 and 1990, varying by a factor of only about 2 from minimum to maximum. Thus, the importance of the locations of relative high and low concentrations is difficult to assess.

Figure 44 shows spatial patterns of mean annual NO_3^- in the Chicago area. Again, the values are spatially very uniform. It is interesting to note that in 1980 the Cook County concentrations were generally <5 $\mu g/M^3$ while those in DuPage and Will Counties were >5 $\mu g/M^3$; in 1985 the pattern was reversed, although that assessment is based on only four sites outside of Cook County There were too few sites in 1990 to justify any comment on spatial variations.

SO_4^{2-}. Spatial patterns of 24-hr maximum and annual mean SO_4^{2-} in the Chicago area are shown in figures 45 and 46, respectively. Interpretation of these patterns is limited by the same factors that affected the NO_3^- data: lack of data for 1980 and a large disparity between the number of sampling sites in 1985 and 1990. Based on only four sampling sites, the data provide a hint that maximum 24-hr concentrations >40 $\mu g/M^3$ affected more of the Cook County area in 1990 than in 1985. Further, the southeast Chicago industrial area had maximum concentrations >40 $\mu g/M^3$ in both years.

Figure 46 shows Chicago-area spatial patterns of annual mean SO_4^{2-}. The areas where mean annual SO_4^{2-} exceeded 15 $\mu g/M^3$ were scattered throughout Cook County in 1985, but were reduced to one site in 1985, and none at all in 1990.

Fe. Spatial distributions of 24-hr maximum and annual mean Fe concentrations in the Chicago area for 1985 and 1990 are shown in figures 47 and 48. In 1985, maximum 24-hr concentrations varied between 1 and 4 $\mu g/M^3$ over most of the three-county Chicago area, but there was a small area where concentrations exceeded 20 $\mu g/M^3$ in the southeast Chicago industrial area. In 1990 the typical range was still 1-4 $\mu g/M^3$, but the high concentrations in southeast Chicago were down considerably.
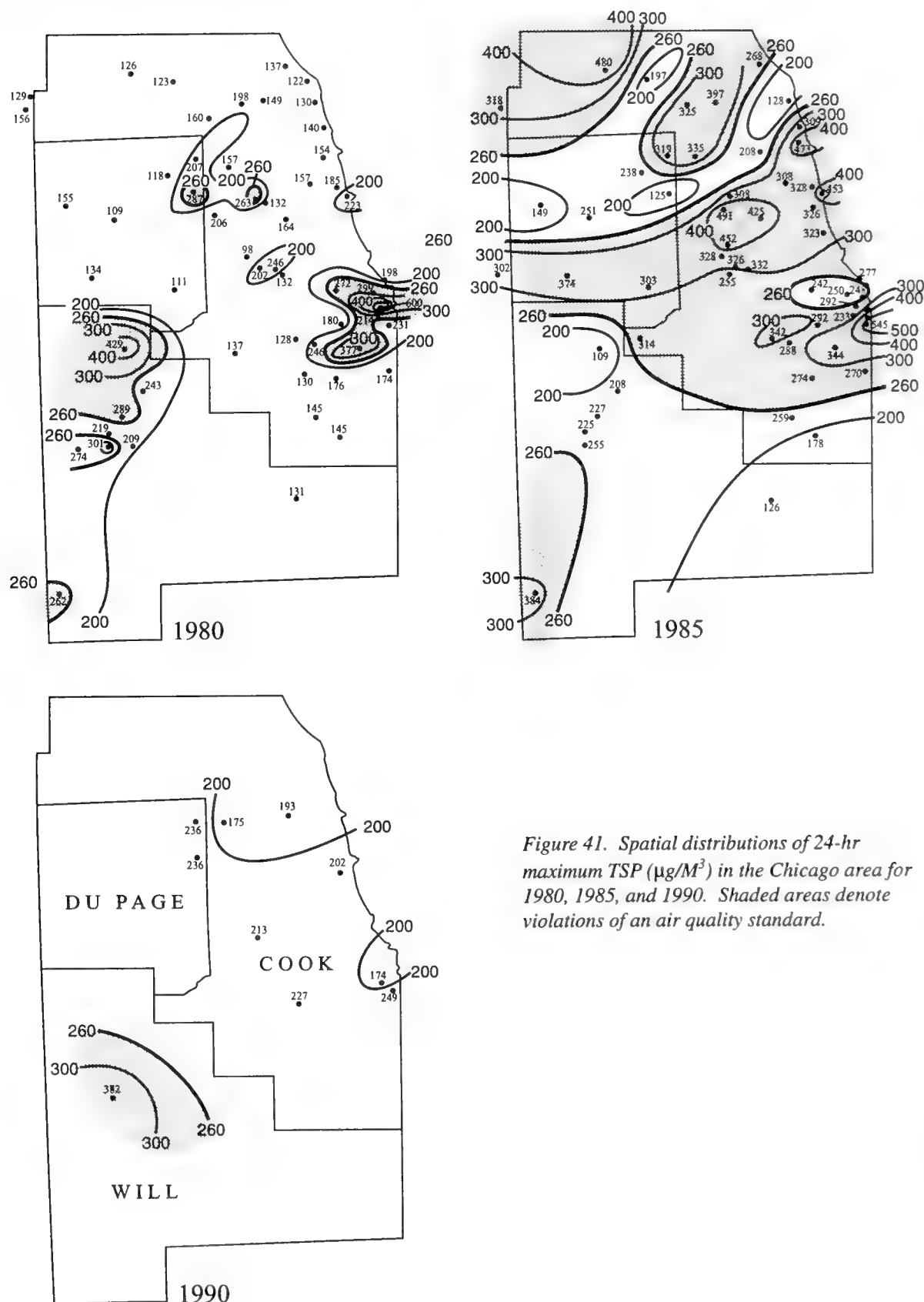
Figure 41. Spatial distributions of 24-hr maximum TSP (μg/M³) in the Chicago area for 1980, 1985, and 1990. Shaded areas denote violations of an air quality standard.
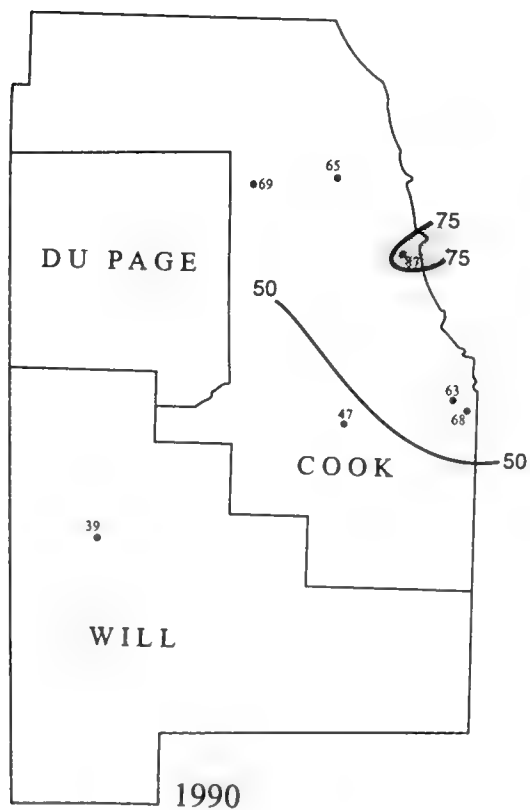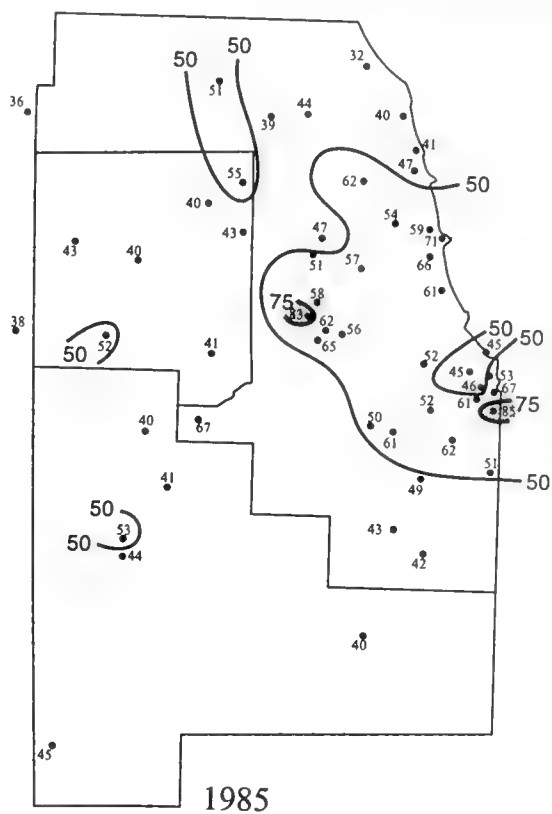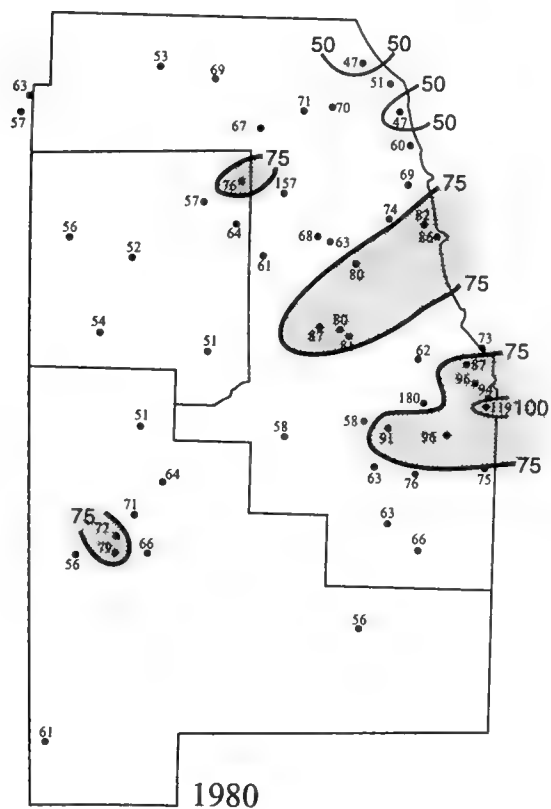
Figure 42. Spatial distributions of annual geometric mean TSP (µg/M³) in the Chicago area for 1980, 1985, and 1990. Shaded areas denote violations of an air quality standard.
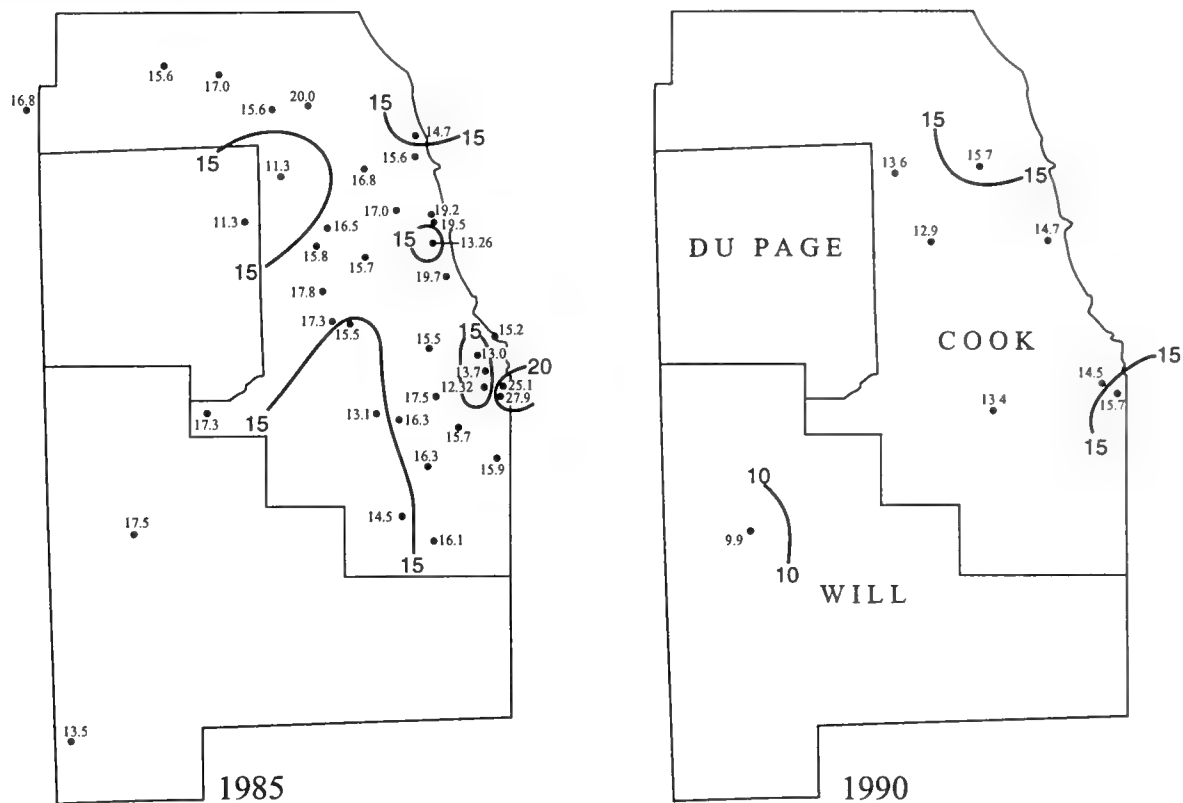
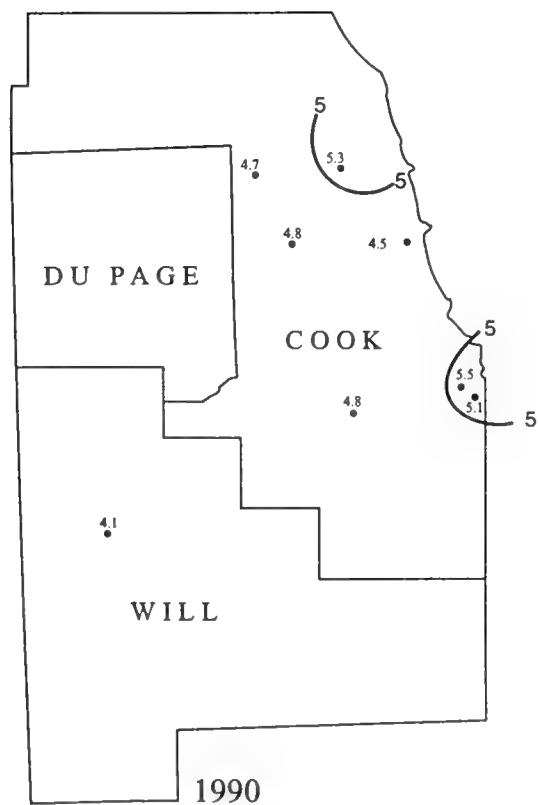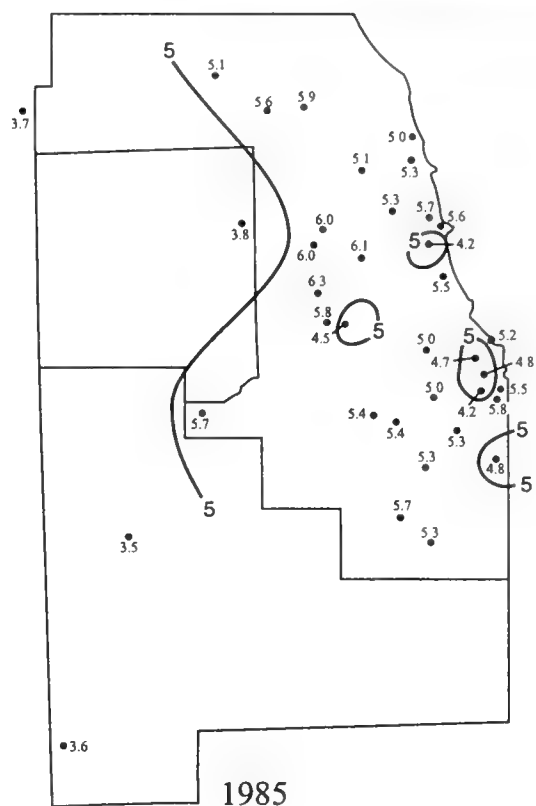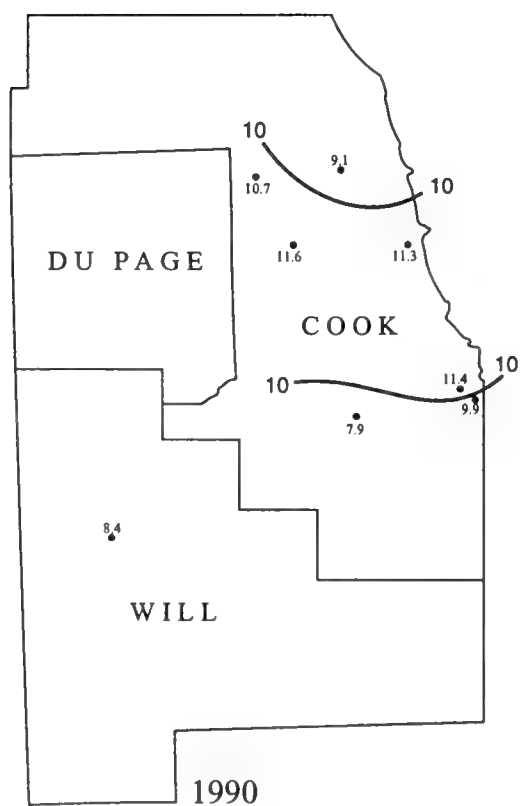*Figure 43. Spatial distributions of 24-hr maximum NO$_3^-$ (µg/M^3) in the Chicago area for 1985 and 1990.*
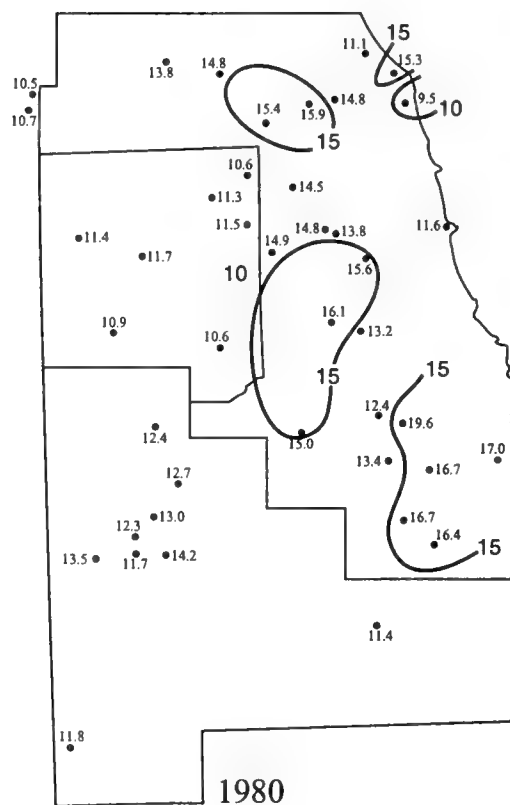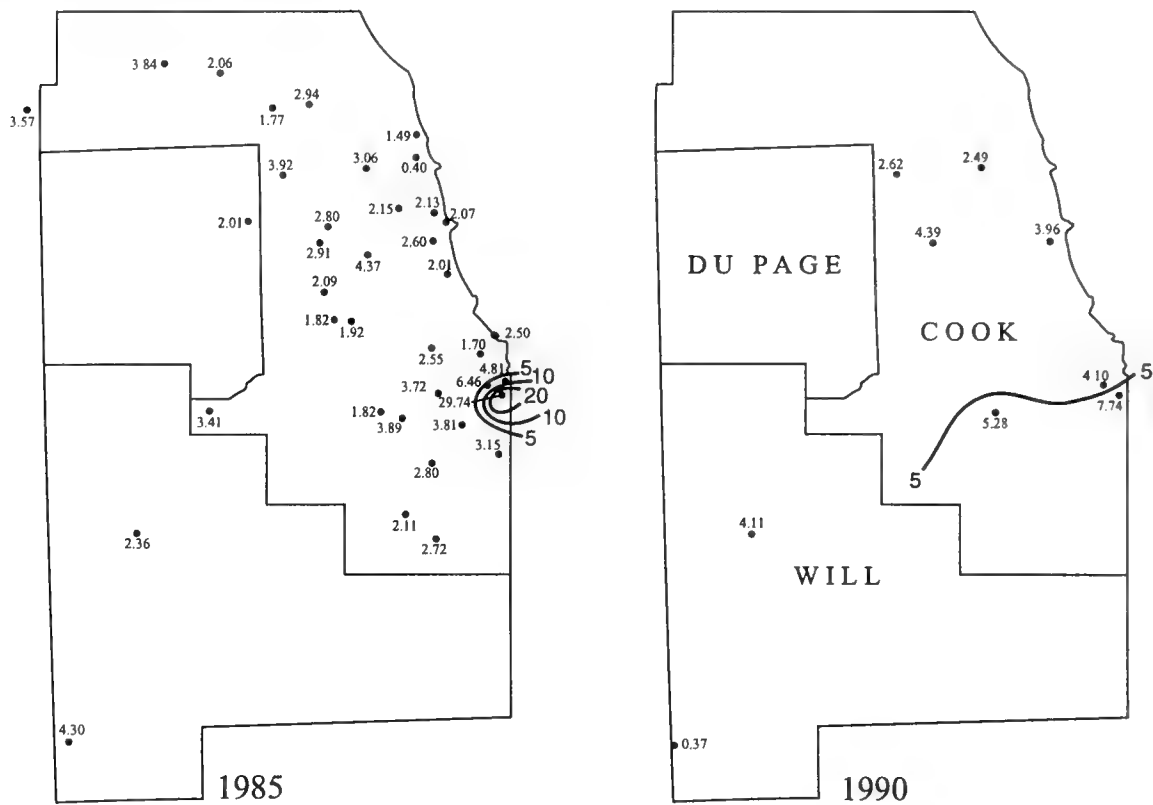
1980

1985

DU PAGE

COOK

WILL
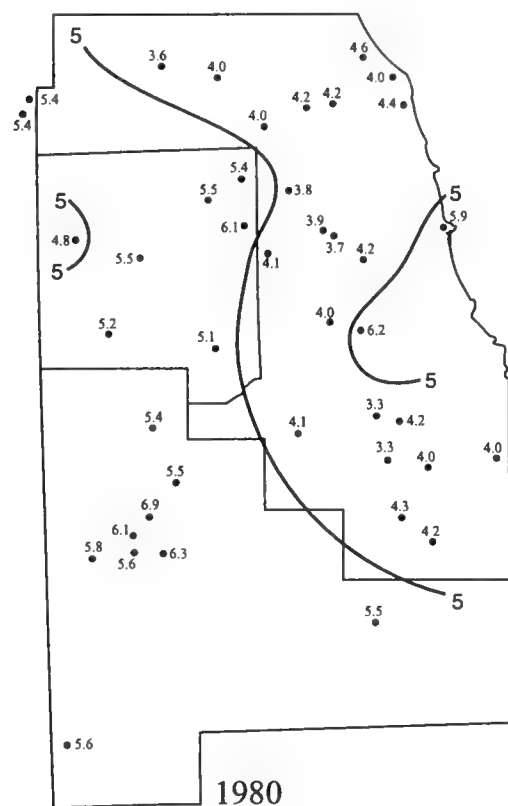
1990

Figure 44. Spatial distributions of annual mean NO_3^- ($\mu g/M^3$) in the Chicago area for 1980, 1985, and 1990.
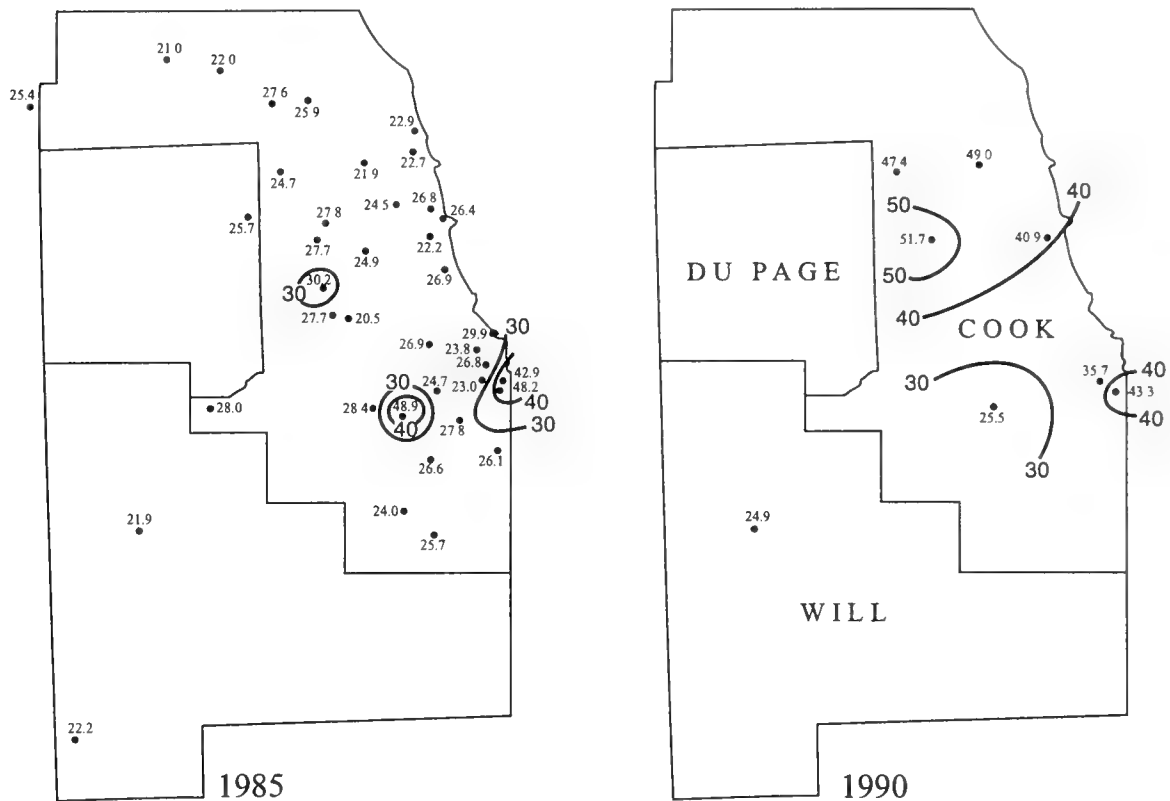
Figure 45. Spatial distributions of 24-hr maximum SO$_4^{2-}$ (µg/M^3) in the Chicago area for 1985 and 1990.

Figure 46. Spatial distributions of annual mean SO_4^{2-} ($\mu g/M^3$) in the Chicago area for 1980, 1985, and 1990.

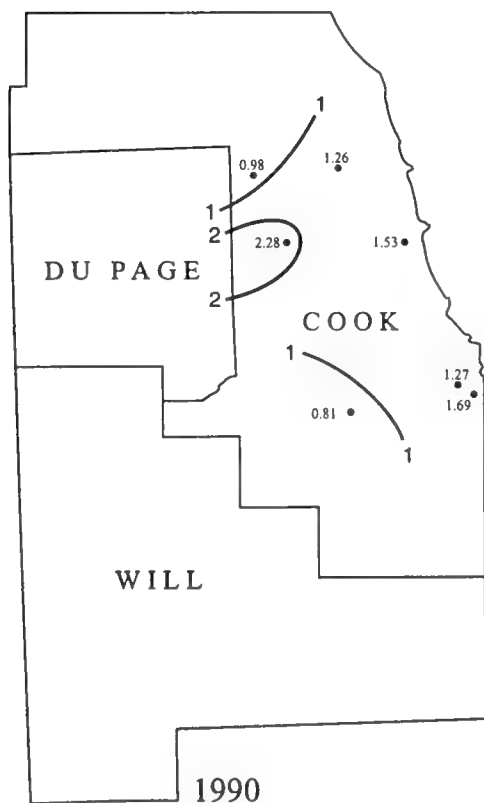*Figure 47. Spatial distributions of 24-hr maximum Fe (μg/M³) in the Chicago area for 1985 and 1990.*

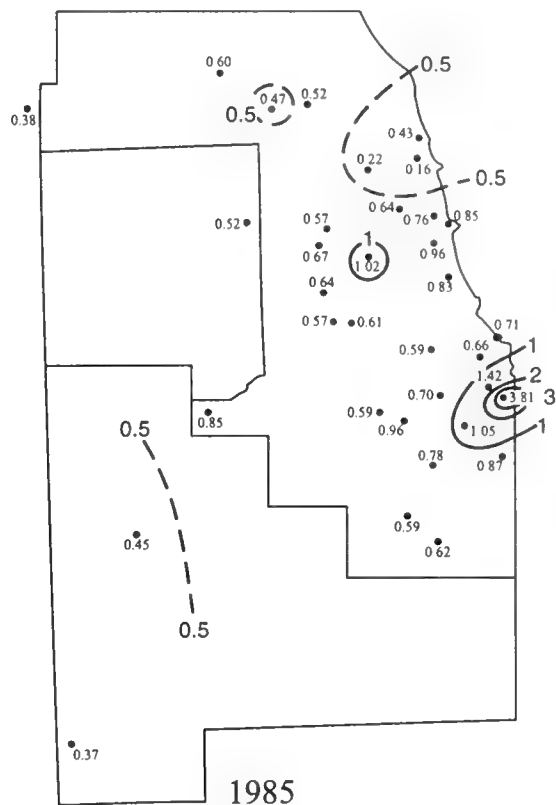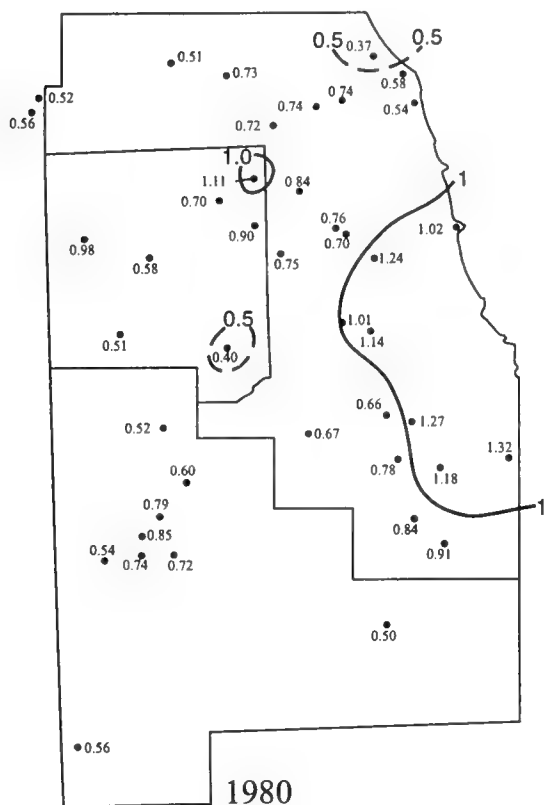*Figure 48. Spatial distributions of annual mean Fe (µg/M³) in the Chicago area for 1980, 1985, and 1990.*

Figure 48 shows spatial patterns of mean annual Fe concentrations in the Chicago area. The main feature of the 1980 pattern was a broad area of concentrations >1.0 µg/M³ in central and southeastern Cook County Note, however, the lack of 1980 measurements in the southeast Chicago industrial region, where mean values >3 µg/M³ were observed in 1985. Most of the few sampling sites remaining in 1990 reported mean Fe concentrations >1.0 µg/M³, but the southeast Chicago hot spot was gone.

Mn. Spatial patterns of 24-hr maximum and annual mean NO_3^- in the Chicago area are shown in figures 49 and 50, respectively. Notice the similarities between the spatial patterns of Mn (figures 49 and 50) and Fe (figures 47 and 48), especially in 1985. This is not surprising since baseline concentrations are probably the result of wind erosion of soil dust, while local peak concentrations are often associated with industrial activity, especially steel-making. Peak 24-hr maximum concentrations of both Mn and Fe in southeast Chicago decreased by a factor of 4 or 5 from 1985 to 1990.

Because of very limited numbers of sampling sites in the Chicago area through 1985, spatial data for the remaining four metals, As, Cd, Cr, and Ni, is limited to 1990 annual means, all presented in figure 51.

As. Figure 51 shows annual mean concentrations of As at eight sites in the Chicago area. Mean concentrations at six of the eight sites were 0.001 µg/M³, and the other two were 0.000 µg/M³. The figure presents only the data. No contours are justified.

Cd. The 1990 spatial pattern of Cd in the Chicago area is also shown in figure 51. Annual mean concentrations were 0.001 or 0.002 µg/M³ at six of the eight sites, with a maximum value (0.008 µg/M³) in southeast Chicago.

Cr. The 1990 spatial pattern of Cr in the Chicago area is also shown in figure 51. Annual means ranged from 0.000 to 0.015 µg/M³. The maximum concentrations occurred in southeast Chicago and the western suburb of Maywood.

Ni. The Chicago-area pattern of mean annual Ni concentrations for 1990 are also shown in figure 51. There was a broad concentration maximum (>0.005 µg/M³) over central and southern Cook County, with the highest value at Maywood.

TABULATIONS OF LITERATURE DATA ON OCCASIONAL MEASUREMENTS

Some pollutants, including some considered toxic, have not been measured routinely in Illinois for reasons that usually involve the cost of analysis, the ability to detect the extremely low concentrations present in the atmosphere, or both. Table 5 summarizes some recent measurements of volatile organic compounds in Illinois. Measurements in table 5 were made in Chicago, in the Metro East area, and at a rural agricultural site near Champaign between 1986 and 1990. Sweet and Vermette (1992) found that the concentrations they measured in Illinois urban areas were quite similar to those observed in other urban areas of the United States. Further, the concentrations they observed in an industrial area were quite similar to those seen in other cities without significant industry, which suggests that the major types of sources were widespread small sources, especially related to automotive emission and fuels, rather than large point sources.

The available data are not adequate to address the issue of temporal trends, except that there are a few substantial differences in mean concentrations between years. With regard to spatial variations, there are compounds for which rural and urban concentrations are similar or different, and some differences in concentration between urban areas.

As is true for many atmospheric parameters, concentrations of many volatile organic compounds are highly variable both spatially and temporally. These variations depend on sample duration, proximity of sampling to sources, variations in source strength, and especially on weather conditions.

Variability between years is seen by comparing the various measured mean concentrations in Chicago. In general, the results of McAlister et al. (1989, 1991) for 1988 and 1990 agree quite well with those of Sweet and Vermette (1992) measured over the 1986-1990 period. However, there are a few exceptions to this rule, notably carbon tetrachloride and tetrachloroethylene in 1990, and toluene and meta- plus para-xylene in 1988. The measurements of Wadden et al. (1992) were made in a different Chicago location, near the city center. There is good agreement between the Wadden et al. measurements and those already mentioned for some compounds, such as benzene, but poor agreement for others, such as chloroform and ethylbenzene. In most cases the Wadden et al. values are greater than the others, probably because of spatial variations within the city.
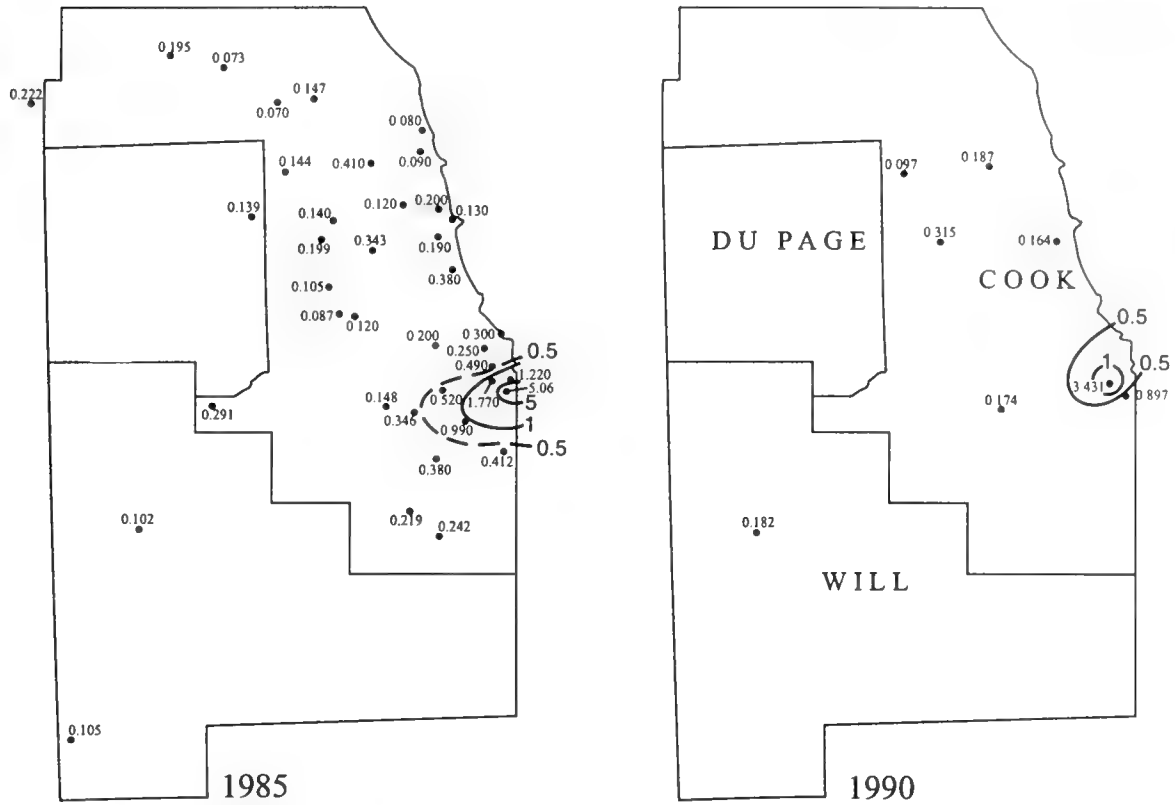
Figure 49. Spatial distributions of 24-hr maximum Mn ($\mu g/M^3$) in the Chicago area for 1985 and 1990.

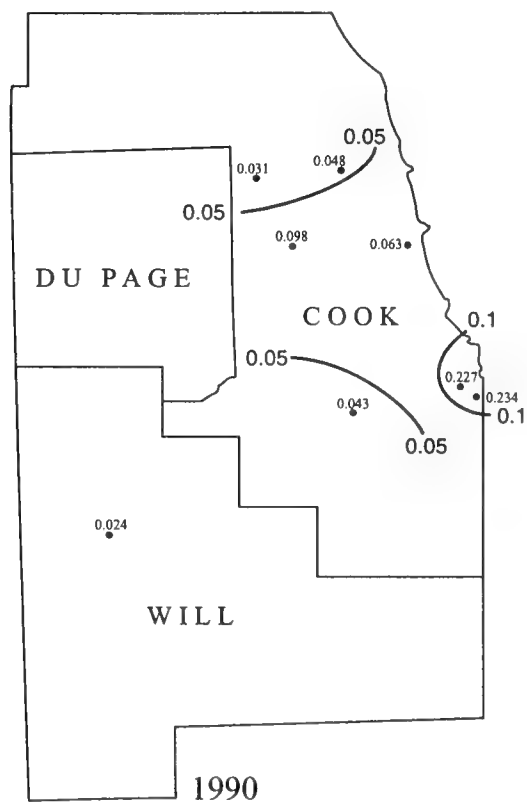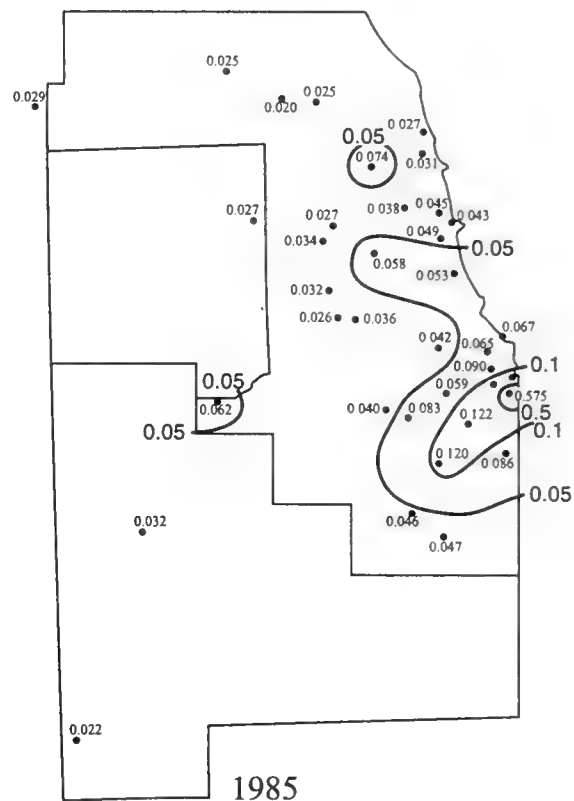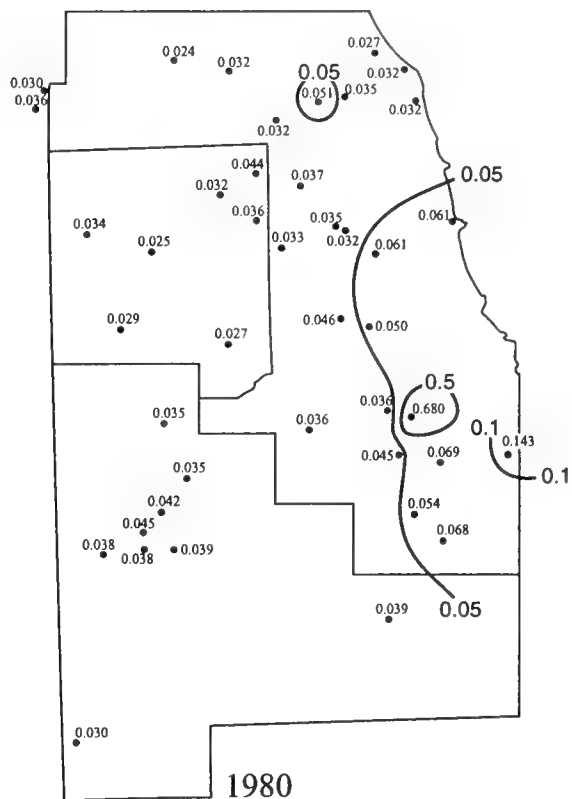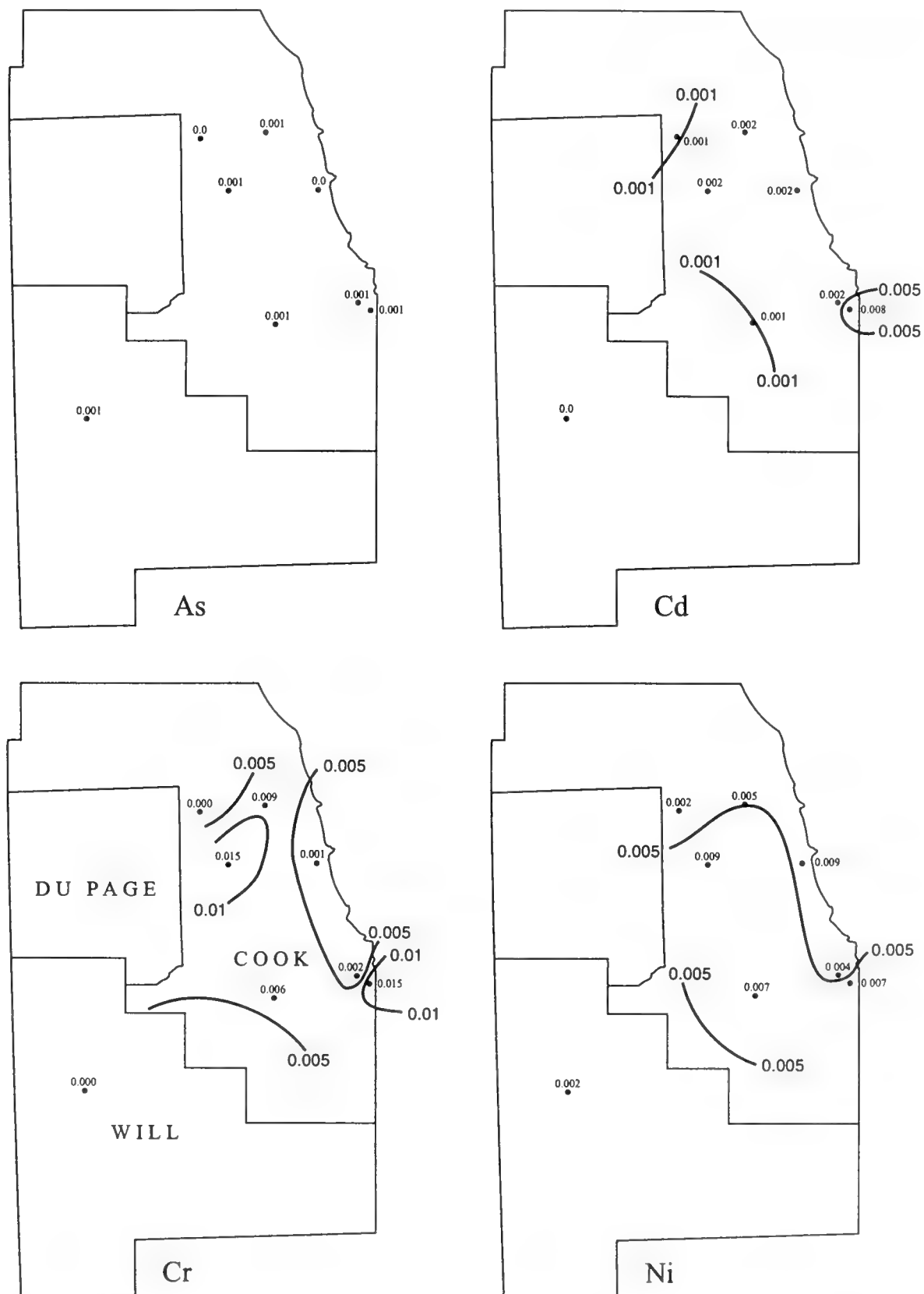*Figure 50. Spatial distributions of annual mean Mn ($\mu g/M^3$) in the Chicago area for 1980, 1985, and 1990.*

Figure 51. *Spatial distributions of annual mean As, Cd, Cr, and Ni (μg/M³) in the Chicago area for 1990.*

Table 5. Summary of Measured Concentrations of Some Volatile Organic Compounds in Illinois (N = number of samples)

Compound	Ref*	Years	Chicago**				Metro East**				Bondville (rural)**			
			N	Mean	SD	Max	N	Mean	SD	Max	N	Mean	SD	Max
acetaldehyde	3	1990	28	3.8		9								
acetylene	4	1990-91	60	5.2	2.7	13.4								
acetone	3	1990	28	7.2		19								
acetaldehyde	3	1990	28	3.8		9								
benzene	1	1986-90	103	4.6	6.6	54	83	10.6	17.2	102	23	1.3	0.5	2.4
	2	1988	34	2.8		23								
	3	1990	29	5.1		18								
	4	1990-91	78	7.6	6.8	38								
bromodichloromethane	4	1990-91	56	20.7	81.5	582								
butane	4	1990-91	71	13.0	29.2	241								
i-butane	4	1990-91	64	7.0	12.7	91								
carbon tetrachloride	1	1986-90	103	0.7	0.2	1.7	83	0.9	0.3	1.7	23	0.8	0.2	1.2
	3	1990	29	2.9		23								
	4	1990-91	63	7.4	9.5	34								
chlorobenzene	1	1986-90	103	0.3	0.2	1.6	83	3.0	6.3	36	23	0.2	0.1	0.5
chloroform	1	1986-90	103	0.3	0.2	1.6	83	0.5	0.9	6.6	23	0.3	0.1	0.4
	4	1990-91	66	4.6	7.2	40								
cumene	4	1990-91	65	6.4	34.2	277								
n-decane	4	1990-91	53	8.6	38.8	282								
dibromochloromethane	4	1990-91	63	81.8	189	1025								
o-dichlorobenzene	4	1990-91	52	20.4	33.3	150								
2-2-dimethylbutane	4	1990-91	76	4.5	7.6	41								
ethane	4	1990-91	60	9.9	5.3	22.8								
ethylbenzene	1	1986-90	103	1.4	1.2	7.6	83	6.9	17	110	23	0.4	0.3	1.6
	2	1988	34	3.0		26								
	3	1990	29	1.8		6.6								
	4	1990-91	78	8.8	22.7	195								
ethylene	4	1990-91	60	7.3	3.5	16.7								
formaldehyde	3	1990	28	6.0		17								
n-heptane	4	1990-91	56	6.4	8.3	45								
hexane	4	1990-91	78	9.2	17.9	137								
2-methylbutane	4	1990-91	77	16.7	16.8	76								
methylene chloride	4	1990-91	65	19.5	34.2	174								
2-methylpentane	4	1990-91	77	7.1	8.7	40								
3-methylpentane	4	1990-91	77	6.4	6.9	39								
n-octane	4	1990-91	39	1.5	1.1	3.7								
pentane	4	1990-91	78	8.6	12.4	72								
alpha-pinene	4	1990-91	60	0.1	0.2	1.4								
n-propylbenzene	4	1990-91	74	8.5	29.1	247								
tetrachloroethylene	1	1986-90	103	1.8	1.6	9.1	83	1.4	1.3	6.1	23	0.4	0.3	1.2
	3	1990	29	10.7		41								
	4	1990-91	60	1.8	10.7	83								

Table 5. (Concluded)

Compound	Ref*	Years	Chicago**				Metro East**				Bondville (rural)**			
			N	Mean	SD	Max	N	Mean	SD	Max	N	Mean	SD	Max
toluene	1	1986-90	103	8.9	8.9	56	83	8.5	9.3	45	23	3.0	2.3	9.5
	2	1988	34	61.1		1284								
	3	1990	29	11.8		33								
	4	1990-91	78	23.5	24.6	131								
1,1,1-trichloroethane	1	1986-90	103	3.3	3.5	25	83	3.9	6.1	31	23	1.1	0.6	1.9
	2	1988	34	4.1		23								
	3	1990	29	8.1		45								
	4	1990-91	73	17.5	25.3	125								
trichloroethylene	1	1986-90	103	1.0	1.0	5.9	83	2.1	5.8	43	23	0.6	0.5	1.5
	3	1990	29	1.2		5.0								
	4	1990-91	76	3.2	3.2	13.7								
1,3,5-trimethylbenzene	4	1990-91	77	6.8	24.4	212								
m-, p-xylene	1	1986-90	103	3.9	8.3	22	83	16	42	312	23	1.2	0.8	3.9
	2	1988	34	20.6		301								
	3	1990	29	9.8		39								
	4	1990-91	78	24.6	86.7	763								
o-xylene/styrene	1	1986-90	103	2.9	5.5	44	83	3.3	8.6	55	23	1.1	0.9	4.3
	2	1988	34	3.9		54								
	3	1990	29	3.7		13								
	4	1990-91	78	18.8	79.4	650								

Notes:

*References: 1. Sweet and Vermette (1992). 2. McAlister et al. (1989). 3. McAlister et al. (1991). 4. Wadden et al. (1992).

**For Reference 1, sampling was carried out at several sites in southeastern Chicago and several sites in the Metro East area. The compounds reported in Reference 1 were detected in 95 percent of the samples. For References 2 and 3, sampling was carried out at Carver High School in Chicago and Sauget in the Metro East area. Sampling for Reference 4 was carried out near the Chicago city center.

The compounds listed in References 2 and 3 were detected in at least 50 percent of the samples. In all cases, for purposes of computing mean values, sample concentrations reported as < the detection limit were assumed to have concentrations of one-half of the detection limit. The rural samples were collected at an agricultural site in Champaign County, 8 km west of Champaign, near Bondville, Illinois.

Variability between concentrations of organic compounds between urban areas occurs for some compounds, and not for others. For benzene, chlorobenzene, ethyl benzene, trichloroethylene, and meta- plus para-xylene, mean concentrations show differences ranging from factors of 2 to 10. Note, however, that the variability of the respective concentrations is very high, judging from the standard deviation, so that the differences may not be statistically significant. It is noteworthy that the Metro East concentration was higher for each of these compounds.

Urban/rural variability, or to be more precise, the lack of differences in concentration between urban and rural locations, is seen for a few compounds, notably carbon tetrachloride and chloroform (Sweet and Vermette, 1992). Such uniform concentrations are evidence of a well-mixed contaminant with a long atmospheric residence time and few current sources. Carbon tetrachloride is an example of a compound no longer used commercially, so its current sources are very small. However, its residence time in the atmosphere is long, so over time it has become well mixed in the atmosphere, and its concentrations are relatively uniform regardless of whether the sampling site is in a rural or urban area (Sweet and Vermette, 1992).

DISCUSSION

Trends over Time: Increasing, Decreasing, and Level Concentrations

Criteria Pollutants. Six pollutant types are included among the criteria pollutants: CO, Pb, NO_2, O_3, SO_2, and particulate matter. Some of these are measured over multiple averaging times, but there is not necessarily a standard for every averaging time. For example, NO_2 is measured over averaging times of 1 hour, 24 hours, and the calendar year, but there are state and national primary standards only for the annual mean. Particulate matter is measured as TSP, for which there are only state standards (for both annual geometric mean and 24-hr maximum concentrations), and as PM_{10}, for which there are only national standards (annual arithmetic mean and 24-hr maximum). Thus, for the six pollutant types, measurements have been made for 14 separate pollutant-averaging time combinations, 11 of which have state or national primary standards. The PM_{10} measurements have only been made for a few years, not enough to make a trend test meaningful, so of the 14 combinations, only 12 (at the

most) have been tested for time trends within individual geographic regions. In some regions there were not enough sites to carry out trend tests. The results of the trend tests within regions are summarized in table 2.

For the state as a whole, 12 pollutant/averaging time datasets were tested for time trend. Seven of these (table 2) showed trends toward decreasing concentrations significant at the 1 percent or 2 percent level, four had no significant trend (5 percent) (including O_3, before accounting for temperature effects), and none showed increasing trends. After accounting for temperature effects, O_3 showed a decreasing trend (2 percent level, table 3), rather than no trend.

The same 12 kinds of datasets were tested for trend in the Chicago area. Eight of these (table 2) showed decreasing trends at the 5 percent level (all but one of these were at the 1 or 2 percent level). Four showed no significant trend at the 5 percent level, and none showed an increasing trend. After accounting for temperature, O_3 showed a decreasing trend (2 percent level, table 3), rather than no trend.

In the Metro East area, only four pollutant/averaging time datasets were measured at enough sites to warrant testing for time trend. Only Pb showed a decreasing trend (1 percent), while three other datasets showed no significant trend (5 percent). After accounting for temperature, O_3 showed a decreasing trend at the 6 percent level, but not at the 5 percent level. No significant (5 percent) increasing trends were found for criteria pollutants.

Nine datasets were tested for time trend in the "remainder" region, four of which included a few sites from the Metro East region when there were insufficient sites for separate tests there. Three of these datasets showed decreasing trends at the 5 percent level (all but one at the 1 percent level), and five datasets showed no significant (5 percent level) trends. After accounting for temperature, the decreasing trend of O_3 with time was significant only at the 10 percent level. Again, there were no significant increasing trends.

Aside from Pb, the largest decreasing linear trends were found for annual mean NO_2, at -7.1 percent per year statewide and -6.8 percent per year in the Chicago area. The greatest magnitudes of decreasing linear trends were found for Pb: -12.6 percent per year in the Metro East area, and -20 to -25 percent per year statewide and in the other geographic areas.

Noncriteria Pollutants. Only trends in annual mean concentrations of the noncriteria pollutants measured— NO_3^-, SO_4^{2-}, and the six metals (As, Cd, Cr, Fe, Mn, and Ni)—were tested for trend over time. Currently, 24-hr average metal concentrations are also being measured, but their record is too short to permit a meaningful test of time trend. The eight noncriteria pollutants were all tested for trend in all four regions, except for the Metro East region, where the data were not adequate to test Cr and Ni. Of the 30 trend tests, 20 showed no significant (5 percent) trend. Over the state as a whole, and in the Chicago area, two species showed significant decreases: SO_4^{2-} at the 5 percent level and As at the 1 percent level. In the Metro East area, Fe and Mn showed significant *increases* (the only increasing trends found in this study): Fe at the 2 percent level, and Mn at the 5 percent level. In the "remainder" region, SO_4^{2-} (2 percent), As (1 percent), Cd (2 percent), and Mn (5 percent) showed significant decreases.

Spatial Trends: Illinois Hot Spots

Comparison of Geographic Regions. Comparison of median and maximum pollutant concentrations within geographic regions, from the yearly box plots in figures 1-30, indicates which geographic areas of the state experienced the highest concentrations of air pollutants. Chicago generally had higher median regional values of annual mean NO_3^-, and of annual mean and 24-hr Cr. It also experienced higher median annual mean Ni and the highest individual 24-hr Ni concentrations. These Cr and Ni results agree with the observations of Sweet and Gatz (1988), based on measurements of fine and coarse particles with dichotomous samplers. On the other hand, the Chicago area generally experienced lower concentrations of 3-hr and 24-hr SO_2 than the rest of the state.

The Metro East area generally experienced higher concentrations than the rest of the state for annual mean Pb, annual mean TSP, and both 24-hr and annual mean As, Cd, Fe, and Mn. Again, the As and Cd results agree with those of Sweet and Gatz (1988), but for Pb and Mn, Sweet and Gatz found higher concentrations in Chicago.

The Chicago and Metro East areas experienced higher concentrations than the rest of the state for 1-hr maximum O_3 and annual mean SO_2.

Chicago Area Concentration Contours. Based on the contour plots in figures 32-51, statements about preferred areas of relatively high pollutant concentra-tions should be regarded as best guesses, based on available data. But these statements are uncertain as to magnitude of pollutant concentrations as well as to precise location. The available data are not adequate for highly certain conclusions because: 1) sampling sites are distributed nonuniformly over the area; 2) the plotted data represent only three years (1980, 1985, and 1990); and 3) numbers of sampling sites, and their locations, have changed from one plotted year to the next, and they have recently dwindled to the extent that the meaning of contours is questionable.

Only one locale in the Chicago area stands out for being the location of high concentrations for multiple pollutants: the industrial area of southeast Chicago around Lake Calumet. This area has persistent, relatively high concentrations of some metals, in terms of both 24-hr maxima and annual means. The metals with highest 24-hr maxima in the Lake Calumet area were Fe and Mn. There was a persistent peak in 24-hr SO_4^{2-} in this area as well. Metals with local peaks in annual mean concentrations include Pb, and possibly Cd, Cr, and Mn, although the evidence for the latter three is somewhat weaker.

Other locations in the Chicago area appeared as areas of persistent high concentrations of only one or two pollutants. One-hour maximum CO was persistently high along the Lake Michigan shoreline north from downtown Chicago. Annual mean NO_2 concentrations were also persistently high near the Chicago city center (the Loop). The plotted O_3 data (figure 37) suggest a possible persistent area of high 1-hr maximum concentrations in northeast Cook County. There is also some weak evidence for relatively high 1-hr maxima of O_3 and annual mean NO_2. Southwest suburban Bedford Park had persistently high concentrations of 3-hr maximum, 24-hr maximum, and annual mean SO_2. Finally, various locations in Will County experienced persistently high 24-hr maximum TSP concentrations, perhaps related to wind erosion of agricultural soils.

CONCLUSIONS

Temporal Trends

Criteria Pollutants. Of all the criteria pollutants tested for time trend in any geographic region of the state, results indicated only decreasing trends or no significant (5 percent) trends. No increasing trends in criteria pollutants were detected.

For the state as a whole, seven of 12 pollutant/averaging time datasets tested for time trend showed significant trends (5 percent level or better) toward decreasing concentrations over the 1978-1990 data period. Pb had the largest decreasing mean linear trend statewide, -20.5 percent per year. After accounting for temperature effects, O_3 showed a significant decreasing trend (2 percent level), rather than no trend.

In the Chicago area, eight of the 12 datasets showed decreasing trends at the 5 percent level (all but one of these were at the 1 or 2 percent level). Again, Pb had the largest trend, -21.2 percent per year. After accounting for temperature, O_3 showed a decreasing trend (2 percent level, table 3), rather than no trend, in the Chicago area.

In the Metro East area, only four pollutant/averaging time datasets were measured at enough sites to warrant testing for time trend. Only Pb showed a significant (5 percent) trend, a trend toward decreasing concentrations. The Pb trend was sizable, -12.6 percent per year, but much smaller than the Pb trend observed in all other areas of the state. After accounting for temperature, O_3 showed a decreasing trend at the 6 percent level, but not the 5 percent level.

Three of nine datasets tested for time trend in the "remainder" region showed decreasing trends at the 5 percent level or better. Again, Pb had the largest trend, -24.8 percent per year.

Noncriteria Pollutants. The eight noncriteria pollutants were all tested for trend in all four regions, except for the Metro East region, where the data were not adequate to test Cr and Ni. Of the 30 trend tests, 20 showed no significant (5 percent) trend. Over the state as a whole, and in the Chicago area, two species showed significant decreases—SO_4^{2-} and As. In the Metro East area, Fe and Mn showed significant increases (the only increasing trends found in this study). In the "remainder" region, SO_4^{2-}, As, Cd, and Mn showed significant (5 percent or better) decreases.

Spatial Trends

Comparison of median and maximum pollutant concentrations within geographic regions, from the yearly box plots in figures 1-30, indicates which geographic areas of the state experienced the highest concentrations of air pollutants. Chicago generally had higher median regional values of annual mean NO_3^-, and of annual mean and 24-hr Cr. It also experienced higher

median annual mean Ni and the highest individual 24-hr Ni concentrations. On the other hand, the Chicago area generally experienced *lower* concentrations of 3-hr and 24-hr SO_2 than the rest of the state.

The Metro East area generally experienced higher concentrations than the rest of the state for annual mean Pb, annual mean TSP, and both 24-hr and annual mean As, Cd, Fe, and Mn. The Chicago and Metro East areas experienced higher concentrations than the rest of the state for 1-hr maximum O_3 and annual mean SO_2.

Within the Chicago area, only one location stands out for its high concentrations of multiple pollutants. This is the industrial area of southeast Chicago around Lake Calumet. This area has persistent, relatively high annual mean or 24-hr concentrations, or both, of SO_4^{2-}, Fe, Mn, and Pb, and possibly Cd, Cr, and Mn. The evidence for the latter three is somewhat weaker than for the others, however. Other locations in the Chicago area appeared as areas of persistent high concentrations of only one or two pollutants.

ACKNOWLEDGMENTS

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ATMOSPHERIC DEPOSITION TRENDS IN ILLINOIS

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INTRODUCTION

Purpose

In the conceptual design of Illinois' Critical Trends Assessment Project or CTAP, (IENR, 1992), atmospheric deposition is treated as an environmental process that describes how pollutants from various sources are delivered to receptors. The CTAP plan addresses six natural environmental receptors: (1) forests, (2) agroecosystems, (3) streams and rivers, (4) lakes and impoundments, (5) prairies and savannas, and (6) wetlands. Among these, atmospheric deposition is considered for forest ecosystems and lakes and impoundments, because research has shown possible damage to these receptors from certain kinds and amounts of atmospheric deposition. For example, the recently completed National Acid Precipitation Assessment Program reported evidence that acidic deposition, along with other stresses, affects some high-elevation spruce forests in the eastern United States (Shriner et al., 1990). Long-term changes in low-elevation forests may be possible as well.

This section describes the characteristics of atmospheric deposition in Illinois, which pollutants have the highest concentrations, and how the concentrations vary across the state and throughout the year. Where there are data sufficient for an analysis, changes over several years are calculated and trends are inferred, if these changes are significant. Also shown are maps of deposition fluxes, which together with the concentration data provide information necessary for assessments of the exposure of Illinois' natural environment to atmospheric deposition. While an explicit description of the source-receptor relationships for major pollutants, such as sulfur dioxide or SO_2 and nitrogen oxides or NO_x, is not considered in this section, the sources of these pollutants in Illinois and surrounding states are compared to their occurrence in atmospheric deposition. Finally, additional work is discussed that is necessary to improve our assessment of atmospheric deposition in Illinois and over Lake Michigan.

Scope

Atmospheric deposition includes gases and aerosols (solid, liquid, or mixed phase). It includes primary pollutants, which retain their chemical identity between source and receptor; and it includes secondary pollutants, which undergo physical or chemical transformation during transport in the atmosphere. Calcium is an important chemical element in Illinois atmospheric deposition. As a primary pollutant, calcium is found in dust particles that become airborne during automobile traffic on unpaved roads (Barnard et al., 1986). Dust particles are relatively large (diameter >1 micrometer or μm); and because of their size and density, the primary mechanism for their removal from the atmosphere is gravitational settling, a form of atmospheric deposition. Sulfate is a secondary pollutant in the Illinois atmosphere, because it is emitted mostly as a gas, SO_2, when fossil fuels are burned. In the air it is transformed into relatively small sulfate aerosols (diameter < 1 μm), which are removed by deposition processes other than gravitational settling.

Deposition of pollutants from the atmosphere is a continuous process, though there are large temporal variations in the deposition rate or flux. These variations relate to the kind of deposition that is occurring and to surface and atmospheric conditions. There are two kinds of atmospheric deposition, wet and dry. Wet deposition is defined as the delivery of pollutants to the surface by precipitation. Dry deposition is the delivery of gases and aerosols to the surface by mass transfer processes other than precipitation. By "surface," here, is meant soil or rock or water bodies or crops or forests or whatever man-made structures cover these natural surfaces (e.g., buildings, monuments, highways, parking lots). In principle, dry deposition occurs continuously, while wet deposition occurs episodically, e.g., when it rains.

Wet deposition includes precipitation-borne pollutants, such as dissolved gases, dissolved particles, insoluble particles, and dissolved materials that are leached from the chemical matrix of particles that do not dissolve in precipitation. An example of this latter type of wet deposition is the potassium that rainwater leaches from soil particles, even though the solid soil particles do not themselves dissolve. Precipitation-borne pollutants can be organic or inorganic. Wet deposition is measured by chemically analyzing precipitation. To measure the chemistry in samples of precipitation collected separately from dry deposition requires a device that opens only during precipitation. Such a device, a wet-only

deposition collector, involves some compromise, since dry deposition occurs continuously even during precipitation. Under most conditions and for most chemicals, however, this compromise is inconsequential, since the total, wet plus dry, deposition during precipitation is dominated by the wet part (wet > 10 x dry). An example of an exception to this statement would be rain that occurs during blowing dust, when the chemical deposition from the falling dust dominates that from the rain.

For this project, wet deposition data from the national network, the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) were used. The NADP/NTN reports the concentrations of dissolved calcium or Ca^{++}, magnesium or Mg^{++}, sodium or Na^+, potassium or K^+, ammonium or NH_4^+, sulfate or $SO_4^=$, nitrate or NO_3^-, chloride or Cl^-, orthophosphate or PO_4^{3-}, and free hydrogen ion or H^+, measured in pH units. No trace metals or organic compounds are measured by the NADP/NTN. Samples are filtered to remove insoluble materials, so the NADP/NTN provides data for the soluble major inorganic ions found in precipitation, i.e., for those chemicals that result in acidic deposition or "acid rain," which occurs over all of Illinois.

Dry deposition includes the mass transfer of pollutants to the surface by a variety of physicochemical processes: turbulent diffusion, diffusion followed by surface sorption of gases, gravitational settling of large particles, impaction, and interception of solid and liquid particles. Dew, fog, and mist are examples of liquid particles that are considered dry, not wet, deposition, because the mass transfer to the surface is by interception, not precipitation.

Dry deposition fluxes are strongly affected by atmospheric factors, which influence the rate at which pollutants are delivered to a receptor surface, and by surface factors, which influence the efficiency with which pollutants "stick" to a receptor surface. Among the atmospheric factors are wind speed and turbulence, air temperature, solar radiation, and relative humidity. Among the surface factors are roughness, wetness, surface-to-air temperature difference, and type of surface, which includes whether the surface is animate (plant) or inanimate. The relative importance of these factors in determining the dry deposition rate also depends on the physical and chemical nature of the pollutant. For example, factors that affect the mass transfer of carbonaceous soot, an unreactive, insoluble particle, are much different than the ones affecting nitric acid, a highly reactive, soluble gas.

The dry deposition of gases and submicron aerosols involves highly complex processes for which direct measurements are intractable on a spatial domain of the size and complexity of Illinois. Thus, an indirect method was applied to infer, rather than measure, dry deposition fluxes. This inferential method employs a conceptual model. This computerized model estimates an atmosphere-to-surface coupling parameter known as the deposition velocity, V_d. The dry deposition flux is the product of V_d and the measured air concentration of a particular pollutant. Model inputs include the atmospheric and surface factors discussed earlier. These are measured at a network of sites sponsored by the National Oceanic and Atmospheric Administration's Atmospheric Turbulence and Diffusion Division (NOAA/ATDD). Land use and vegetation type and status are also reported at these sites, along with the airborne concentrations of Cl^-, $SO_4^=$, particulate NO_3^-, nitric acid vapor or HNO_3, and SO_2. The NOAA/ATDD sampling system is especially designed to *exclude* large particles, since the inferential method of calculating dry deposition applies specifically to gases and submicron aerosols.

The dry deposition of large particles, which have an aerodynamic diameter > 1 μm, is typically estimated from an analysis of the mass of a pollutant accumulated on a surrogate surface (Dolske and Gatz, 1984). The term "surrogate surface" is used, because it is impractical to measure dry deposition on every animate (plant) and inanimate surface, and so a substitute surface is selected. Since the physical process that dominates large particle dry deposition is gravitational settling, the surrogate surface must receive and retain large particles in a manner equivalent to soils, rocks, streams, building surfaces, plant surfaces, etc. In principle, there is no universal surrogate for surfaces with such diverse characteristics.

To estimate the dry deposition of large particles, (i.e., sedimentation or dryfall) for the CTAP, data from the NADP/NTN were used. The NADP/NTN measures dryfall in separate samples collected by the sampler also used for precipitation. This device, a wet/dry collector, has two identical containers; it discriminates between wet and dry conditions, exposing the wet deposition container during precipitation and the dryfall container at all other times. Dryfall samples are sent for analysis of the same analytes measured in precipitation.

Evaluations of dry deposition monitoring methods (Hicks et al., 1986) have raised objections to this

method for monitoring dry deposition. One objection is that contamination by foreign matter (plant parts, insects, bird feces, etc.) compromises sample integrity. A second objection is that gaseous and fine-particle deposition are poorly characterized. For this study, however, the data were screened to minimize the effect of sample contamination from foreign matter. As to the second objection, the data were used only for dryfall or sedimentation estimates, not for the deposition of gases and submicron aerosols.

DATA SOURCES

Wet Deposition

Data for assessing the precipitation quality and wet deposition fluxes for Illinois' CTAP were taken from the NADP/NTN. This network began operations at 20 sites in 1978 and today operates at nearly 200 U.S. locations, with a site in American Samoa and one in Puerto Rico. Sites were chosen according to network design criteria (Robertson and Wilson, 1985), and equipment was installed to minimize sampling artifacts from local point and area sources (Bigelow, 1984). Based on these criteria, data from network sites are expected to be representative of the region where the sites are located. Using a wet/dry collector of a specified type, one-week, wet-only, deposition samples are collected on Tuesday mornings in the NADP/NTN (Bigelow and Dossett, 1988). All samples are sent to a single Central Analytical Laboratory (CAL), located at the Illinois State Water Survey. At the CAL, approved standard procedures are followed for sample analysis (Peden et al., 1986) and data validation (Bowersox, 1984).

Table 1 lists the locations, site types, and periods of operation of NADP/NTN sites in Illinois. Seven sites are currently active in the state; the site near Salem (IL47) was closed in 1988 and moved to Stephen Forbes State Park, near Omega (IL99), in 1989. Figure 1 shows the locations of NADP/NTN sites in and around Illinois. Data from outside the state are included with Illinois data during all spatial objective analyses of precipitation quality and deposition flux fields. This approach precludes "edge effects" from extrapolation of the data, by providing in-state and out-of-state observations that can be used to interpolate data fields out to and beyond state borders. Edge effects can produce spurious results in objective analysis schemes.

Dry Deposition

Each of the NOAA/ATDD sites, Argonne, IL19, and Bondville, IL11 (see figure 1 for locations), was deliberately co-located with an existing NADP/NTN site. The objective of the NOAA/ATDD program is to develop and verify models for estimating dry deposition from easily measured surface conditions, micro-meteorological parameters, and air quality data (Hicks et al., 1991). Weekly average air concentration data and a site-specific deposition velocity, V_d, are used to calculate dry deposition amounts for certain gases and ions associated with small particles. Surface conditions include local topography, land use, and vegetation type and status. Each of these surface factors contributes to V_d at varying length scales, and these factors vary only slowly over time. Micrometeorological parameters (wind speed and direction and their fluctuations, temperature, humidity, solar radiation, surface wetness, and precipitation rate) are monitored continuously and digitally recorded every 15 minutes.

All of these factors are input variables for a computer model that provides a set of V_d values for a predetermined time period (usually weekly). Each V_d in the set is specific to the chemical species and to the gas or particle size range in which that species is found in the atmosphere. The weekly-average airborne concentrations of particulate Cl^-, $SO_4^=$, and NO_3^-, and of gases, HNO_3 and SO_2, are reported for each site. The sampling system excludes large particles, since the inferential method of calculating dry deposition applies specifically to gases and submicron aerosol-associated materials.

The weekly accumulated flux of each chemical is then calculated as the product of the average concentration and the appropriate V_d. These calculated results are periodically compared with directly measured dry deposition data at the NOAA/ATDD sites (Meyers et al., 1991), so that the model can be calibrated and revised as needed. The current version of the NOAA/ATDD model produces dry deposition estimates that generally agree within 10 to 20 percent of the directly measured values. Further model calculations are used to "scale up" to larger areas and longer time scales.

In addition to the NOAA/ATDD data, longer term (annual) and wider area concentration data for other chemical species in Illinois' atmosphere were provided through the CTAP (see *Air Quality Trends in Illinois* chapter). For each of these chemicals (arsenic or As, cadmium or Cd, chromium or Cr, iron or Fe, manganese or Mn, nickel or Ni, nitrogen dioxide or NO_2, NO_3^-, ozone or O_3, lead or Pb, SO_2, and $SO_4^=$, as well

Table 1. National Atmospheric Deposition Program/National Trends Network Sites in Illinois

Site designation	IL 11*	IL 18	IL 19*	IL 35	IL 47	IL 63	IL 78	IL 99
Site location								
Latitude	40°3'12"	41°50'29"	41°42'4"	37°42'36"	38°38'36"	37°26'8"	40°56'0"	38°42'36"
Longitude	88°22'19"	88°51'4"	87°59'43"	89°16'8"	88°58'1"	88°40'19"	90°43'23"	88°44'57"
Elevation (m)**	212	265	229	146	173	161	229	153
Nearest town	Bondville	Shabbona	Argonne	Carbondale	Salem	Glendale	Monmouth	Omega
Site type	rural	rural	suburban	rural	suburban	rural	rural	rural
Dates of operation								
Wet deposition								
Begin	27 Feb 79	26 May 81	11 Mar 80	31 Jul 79	15 Apr 80	30 Jan 79	8 Jan 85	26 Sep 89
End	continuing	continuing	continuing	continuing	22 Nov 88	continuing	continuing	continuing
Dryfall deposition								
Begin	27 Feb 79	26 May 81	1 Apr 80	20 Jul 79	15 Apr 80	30 Jan 79	none	none
End	continuing	continuing	continuing	continuing	22 Nov 88	continuing	none	none

Notes:
*Also a NOAA/ATDD dry deposition site where airborne pollutant concentrations and atmospheric and surface factors are measured so that deposition can be inferred.
**Elevations are reported in heights above mean sea level.
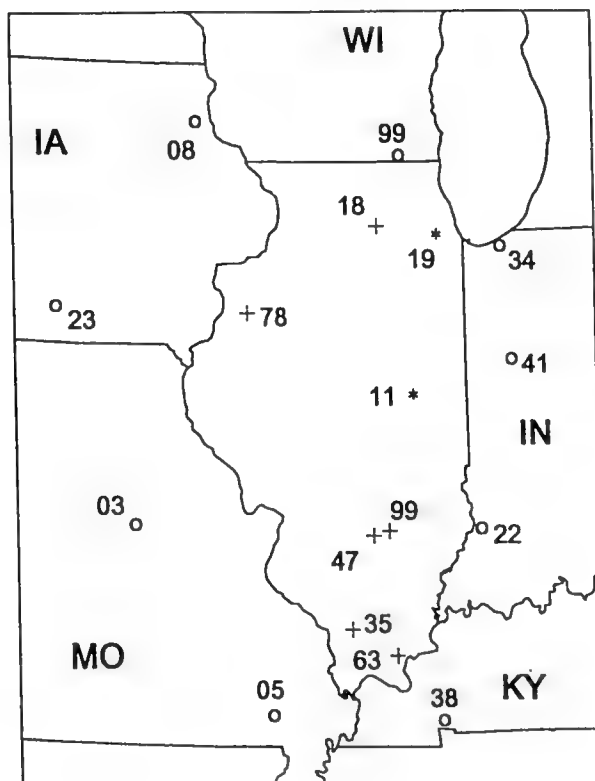
Figure 1. Map of the Illinois region, showing locations of sites providing atmospheric deposition data used in the CTAP.

as total suspended particulate matter or TSP), annual statistics from the Illinois Environmental Protection Agency (IEPA) concentration data were combined with the appropriate NOAA/ATDD V_d to estimate the median and range of dry deposition loadings.

Dryfall deposition samples are collected at a subset of NADP/NTN sites, using the "dry side" of the wet/dry collector. There are five dryfall sites in Illinois, listed in table 1. While the wet deposition samples at NADP/NTN sites are collected weekly, dryfall samples are left to accumulate for eight weeks, and then are sent to the CAL for analysis. At the CAL, 250 milliliters (mL) of deionized water is added to the samples, the mixture is swirled to remove dry deposited matter from all surfaces, and the samples are allowed to equilibrate overnight. Next, the samples are filtered (0.45 µm pore diameter), handled, and analyzed the same way as rain samples (Peden et al., 1986; Lockard, 1987). Results are reported in mass flux units.

DATA ANALYSIS METHODS

Data Screening Procedures

Data from NADP/NTN collection sites in Illinois and neighboring states were used to assess atmospheric deposition for the CTAP. These data were used only when standard network procedures for field, laboratory, and data management operations were followed. Several documents describe the NADP/NTN quality assurance program (NADP/NTN, 1990), field site sample collection and measurement methods (Bigelow and Dossett, 1988), analytical laboratory operations (Peden et al., 1986), and data verification and screening procedures (Bowersox, 1984).

Adhering to standardized procedures ensures that data comparisons among sites are not biased by *differences* in equipment or procedures and that comparisons over time at a site are not biased by *changes* in equipment or procedures. Where NADP/NTN measurements of atmospheric deposition were used for Illinois' CTAP, analyses were based on data that are comparable in space and time.

In addition to the selection of samples collected according to standard procedures, all wet deposition data must have been for wet-only precipitation samples, uncontaminated by matter foreign to the formation and occurrence of precipitation. The requirement for wet-only sample collection was described in the introduction of this chapter. An example of foreign

matter is a leaf or a bug. A report of foreign matter in a sample, while necessary, is not sufficient for exclusion of a record from the analysis. An objective system for identifying an NADP/NTN sample as "contaminated" requires the co-occurrence of foreign matter and an anomalous chemical composition (Bowersox, 1984). "Contaminated" samples are excluded from the analysis.

Finally, to create chemical concentration maps and to calculate interannual trends, the data must satisfy four completeness criteria. Expressed as percentages, these four criteria quantify the fraction of time and the fraction of the precipitation amount for which there are valid wet deposition data. In summary, these criteria require that there be:

1. Precipitation amount measurements for 90 percent or more of the time,
2. Valid chemistry measurements for 75 percent or more of the time,
3. Valid chemistry measurements for 75 percent or more of the measured precipitation, and
4. A ratio of total wet-only sample volumes to total measured precipitation of 75 percent or more, taking data records where both values are available.

These same criteria are applied in the publication of the *NADP/NTN Annual Data Summary* (NADP/NTN, 1991). Wet deposition data that pass these validation tests have an acceptable level of comparability and representativeness for making maps and performing trend analyses for the CTAP.

Figure 2 displays the record of when wet deposition measurements were made at each of the Illinois sites. Also displayed are the periods when data passed the validation tests for calculating trends. Whole years of data were used to evaluate the four completeness criteria. Consequently, a year either passed or failed the tests and is included or excluded, respectively, from the trends analyses. A meteorological year, 1 December through 30 November (Trenberth, 1983), was used to ensure proportionate representation from each season in each year (i.e., December of year 1 and January and February of year 2 comprise a winter season).

For making maps, the completeness criteria were applied to the time period on which the maps were based (multiple meteorological years). Notice in the figure that valid data from the Monmouth site (IL78) began in 1986. Given the importance of the data from this site in defining east/west gradients in several of the chemical species, it was decided that concentration
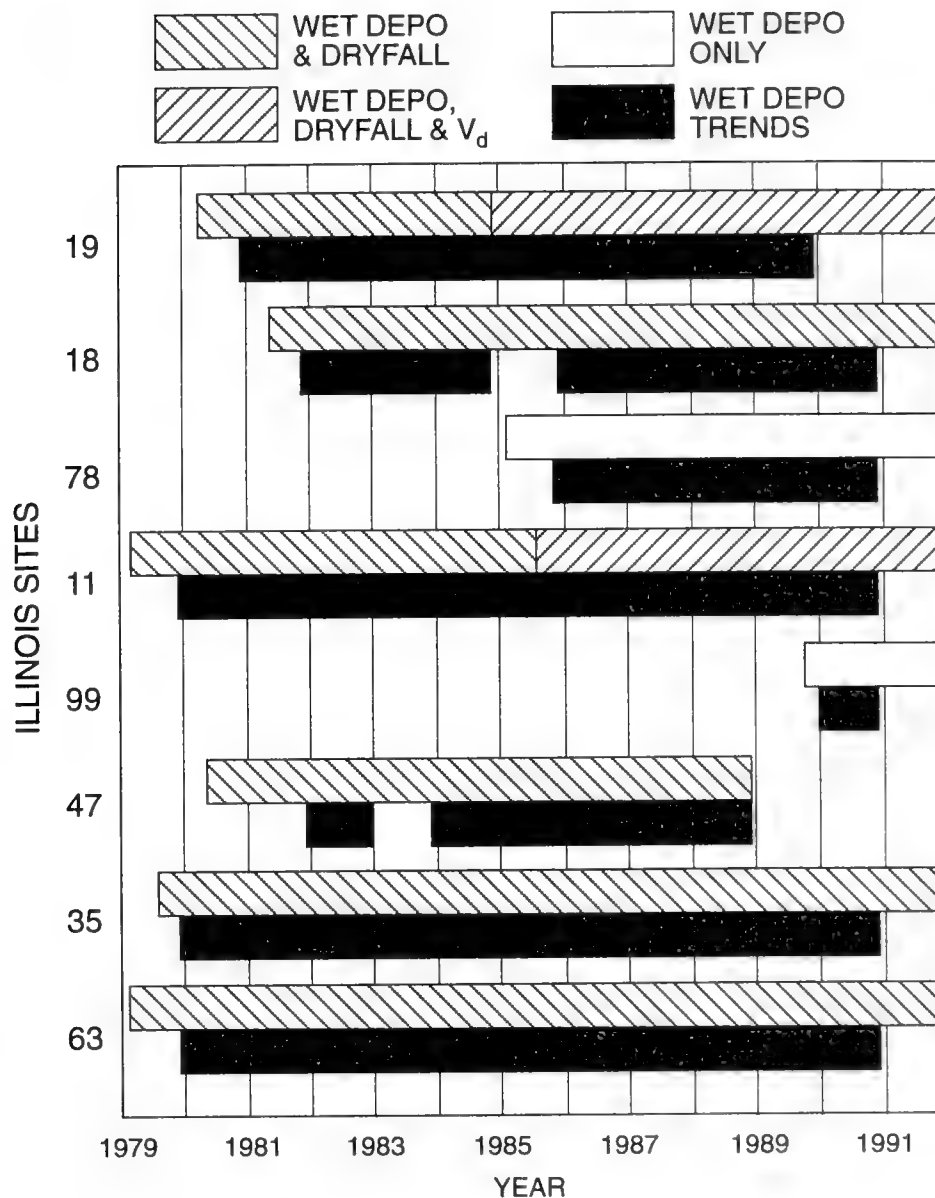
Figure 2. Record of wet and dry deposition measurements at Illinois sites (see figure 1
for site locations). Wet deposition (WET DEPO) and dryfall measurements are from NADP/NTN sites. Deposition
velocity, V_d measurements are from NOAA/ATDD sites. Valid data for long-term trends
(WET DEPO TRENDS) are also indicated.

maps would be based on valid data beginning in 1985 and ending in 1990. During this period the Salem site (IL47) terminated operations and was replaced by the Omega site (IL99), with a one-year (1989) hiatus. In the interest of having maximum spatial coverage, valid data from these two nearby sites were pooled in making concentration maps. With only a year of valid data at the Omega site, no statistical tests were done to evaluate whether these two data sets were from the same population and could be pooled without introducing a bias. This should be done when more data are available for future analyses.

Figure 2 also displays when and where dryfall (i.e., dry bucket) measurements are made at Illinois sites (see also figure 1 and table 1). Unlike NADP/NTN wet deposition data, dryfall data are not routinely screened to ensure samples that are dry-only and uncontaminated. Gatz et al. (1988) examined dry bucket data from the entire NADP/NTN to develop criteria that could be applied to remove unrepresentative contaminated samples from the data. Their approach was to examine the distribution of ionic concentrations, while applying ever more stringent screening criteria, thus eliminating more and more data. Samples were removed in groups, first due to sample collection and handling errors (i.e., nonstandard procedures), and successively due to contaminants: water, bird droppings, plant or insect debris, and other contaminants. Screening to the level where samples with visible contaminants were removed resulted in a clear and statistically significant break in the NH_4^+, K^+, and PO_4^{3-} distributions; this level sharply reduced a positive bias in these constituents. Even H^+ and SO_4^{2-} showed some effects from removal of these samples, though not as marked. For these reasons, dryfall data from Illinois sites were screened to this level for this report.

Recall from previous sections that the NADP/NTN dry bucket sampling protocol is to collect an eight-week integrated sample from the "dry side" of a wet/dry collector. This results in either six or seven samples per year. Removing samples to the screening level described above, leaves from two to six samples per year at each of the five Illinois sites. With so few samples in a year, all of the data from each site were pooled to examine spatial patterns in dryfall deposition, but the calculation of interannual trends at a single site was impractical.

The dry deposition of gases and submicron aerosols in Illinois is inferred from measurements at two NOAA/ATDD sites (Argonne and Bondville), shown in figure 1 and listed in table 1. Figure 2 shows the period of operation of these two stations. As previously de-

scribed, data from the NOAA/ATDD sites include the chemistry of weekly-integrated air samples and 15-minute-average micrometeorological measurements, digitally recorded. These are input variables to a model that infers dry deposition fluxes. These data are screened for completeness by verifying that all micrometeorological measurements are continuous during a period for which V_d is computed. Airborne concentrations also must be available for this period. This requires uninterrupted field equipment operations and complete laboratory analyses for each weekly period. Quality assurance practices (Hicks et al., 1991) help to ensure accurate and complete data from NOAA/ATDD sites. These include:

1. Weekly site operator verification of instrumental performance,
2. Monthly air quality system checks with field and procedural blanks,
3. Quarterly flow rate calibrations, and
4. Semiannual calibration of the entire system by NOAA/ATDD personnel.

Finally, the NOAA/ATDD model occasionally is tested and verified against direct measurements of dry deposition at "core" sites in the network. Argonne is one of these sites. The Bondville site is a so-called "satellite" site, where identical methods and instrumentation are used, but no on-site model verification is done. For this report, 45-51 weekly, quality-assured, data points per site per year were available to calculate dry deposition fluxes, for an overall completeness better than 90 percent.

Annual median values from the IEPA concentration data were used (see *Air Quality Trends in Illinois* chapter) with distribution statistics from the NOAA/ATDD V_d results to calculate statewide deposition estimates. Deposition amounts are reported only for those years in which sufficient data were available from both data sets to calculate annual statistics.

Finally, annual estimates of total atmospheric deposition were computed for 1985-1990 for those chemical species (calcium, sulfur, and nitrogen) for which all the various components of wet and dry deposition were available, according to each of the above criteria. The total deposition values were based on summation of annual means of NOAA/ATDD V_d values for fine particles and gases, the IEPA concentration data for sulfate, sulfur dioxide, and nitrate, the NOAA/ATDD nitric acid concentration data, and NADP/NTN dryfall and wet deposition data.

Statistical Analyses

Ion concentrations in valid wet-only deposition samples from NADP/NTN sites were used to characterize Illinois precipitation chemistry. An approach used by Semonin and Bowersox (1983) to identify the ions that dominate the stoichiometry of precipitation was applied to Illinois data. Briefly, ions were ranked in order of their average concentrations and the subset that comprised about 90 percent of the total ion strength was selected. This subset was used to calculate a charge balance for individual samples and to assess whether the distribution of charge balances was near zero. Zero is expected, if all ions are measured with zero bias and precision, which requires perfect measurements. In practice, ion balances are positive and negative due to noise, or imprecision, in every measurement; and so the distribution of ion balances clusters around zero. When the distribution of ion balances for the subset was within the distribution for all measured ions, the process of adding ions to the subset was stopped. This subset was assumed to be characteristic of Illinois precipitation.

The distributions of ions that "characterize" Illinois precipitation are summarized by site and by year in this chapter. Ion concentration distributions are presented in notched box-and-whisker plots, as in figure 3. A commercial software package on a personal computer created these plots by ranking the data in ascending order from minimum to maximum. The middle half of the data, defined as the 25th to the 75th percentile, is enclosed in a box. In figure 3 this box begins at 0.23, the 25th percentile, and ends at 0.61, the 75th percentile. The box width is proportional to the square root of the number of observations (N) in the data set it represents. Side-by-side boxes are scaled accordingly, so that the reader can distinguish how much data is represented by each box relative to its neighbors.

One generally puts more confidence in large rather than small data sets, because they are less likely to change significantly when new data are added, i.e., results are generally more robust as N grows. Along the sides of the box in figure 3 is a V-shaped notch with a horizontal line in its middle. This line marks the median, at ~ 0.36 in figure 3, which is the value in the ranked data with as many observations above as below it. Notches are drawn according to formula (1) and approximate a 95 percent confidence interval around the median (AXUM, 1992). Notches are drawn at the

$$\text{median} \pm (1.57 * \text{height of box} \sqrt{N}) \qquad (1)$$

"Informally interpreted, if the notches for two boxes don't overlap, it is evidence that the medians are different" (AXUM, 1992). To facilitate the comparison of medians, a shaded band that fills the notch for a preselected reference site was added to some figures, e.g., figure 7. Vertical lines, or whiskers, are located above and below the box and these mark the highest and lowest 25 percent of data, respectively. Short horizontal tick marks have been added to the whiskers to mark the location of the 10th and 90th percentiles. In figure 3 the 90th percentile occurs at 0.88 and the 10th percentile at 0.17. Altogether, the notched box-and-whisker plots mark the location of the minimum and the 10th, 25th, 50th (median), 75th, and 90th percentiles, and the maximum.

Statistical distributions of pollutant concentrations in Illinois precipitation are presented as notched box-and-whisker plots in figures 7-12. All valid data from each site are summarized in these figures. Sites are shown in order from north on the left to south on the right (see table 1 and figure 1 for site names and locations). The number of valid data points (N) for each site varies and depends mostly on the duration of sampling at each site (figure 2). For each site in these figures, N is 450 (IL190); 402 (IL18); 245 (IL78); 497 (IL11); 77 (IL99); 293 (IL47); 483 (IL35); and 502 (IL63).

All of the basic statistical analyses used to analyze dry deposition estimate distributions to arrive at means, medians, standard error, and ranges were computed using SYSTAT 5.0 routines (Wilkinson, 1990). The development of variables used to generate combined data, e.g.,

$$\text{dry deposition} = V_d * \text{concentration}$$

and

$$\text{total deposition} = \text{wet} + \text{dry (gas)} + \text{dry (aerosol)} + \text{dryfall}$$

was done using either SYSTAT data management utilities or BASIC programs written specifically for each task.

All graphical displays of the dry deposition data, including box plots, bar graphs, stacked bars, and scatterplots, were generated on an ARES 486-66 PC using SYGRAPH 1.0 and SIGMAPLOT 5.0 (Jandel, 1992). SYGRAPH 1.0 was used to prepare the total deposition plots, which were overlaid on a State of Illinois map.
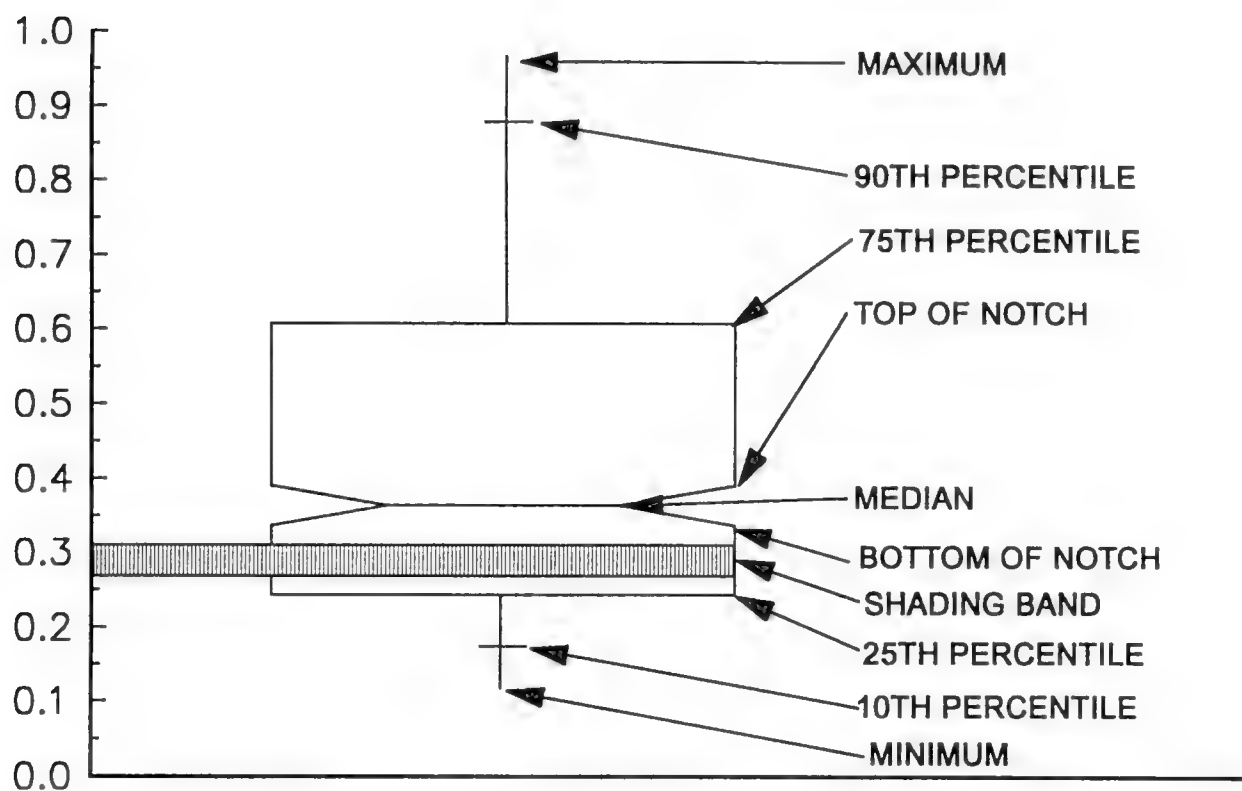
Figure 3. Example of a notched box-and-whisker plot used to summarize the distributions of ion concentrations in wet deposition.

GIS Use Disclaimer

Due to the extremely low spatial density of deposition data, no geographic information system (GIS) coverages have been developed for atmospheric deposition. The assignment of wet deposition values to nine climate zones, and dry deposition to the "Chicago area" vs. "Remainder of State" is the best spatial resolution possible with currently available data. Furthermore, for dry deposition in particular, large variations in the rate of deposition can occur for a specific receptor system (lake surface, forest, or agricultural crop) and even elements within a system (for example, various parts of a building exterior within a city).

RESULTS

Spatial Analysis

Wet Deposition. General characteristics of Illinois precipitation chemistry. Wet-only data from the NADP/NTN were used to examine the spatial variations in precipitation-borne pollutant deposition in Illinois. The NADP/NTN reports mass concentrations of ten ions, (Ca^{++}, Mg^{++}, Na^+, K^+, NH_4^+, $SO_4^=$, NO_3^-, Cl^-, PO_4^{3-}, and H^+) routinely measured in the filtrate of precipitation collected at network sites. This set of ions includes the major anionic and cationic species that cause acidic precipitation in Illinois and elsewhere. Except for orthophosphate and possibly potassium, each of these ten ions is important in determining the average ionic composition of precipitation somewhere in the United States. In many coastal areas, the proximity of a major source of airborne sea salt results in precipitation having a chemistry dominated by Cl^-, Na^+, $SO_4^=$, and Mg^{++} (Junge, 1963).

In other areas, the chemical signature of wet deposition reflects the mix of sources that dominates the air quality in the local area. Semonin and Bowersox (1983) used data from North American precipitation chemistry networks to identify these signatures, where possible. In the eastern United States and Canada, four different signatures were reported. One four-ion signature (H^+, $SO_4^=$, NO_3^-, and NH_4^+) occurred across a portion of Illinois, although it was determined from a limited set of data, one to two years of data at four sites. Data are now available from eight different locations and for up to 12 years at some of these locations (see figures 1 and 2 and table 1).

A procedure similar to the one used by Semonin and Bowersox (1983) was applied to Illinois precipitation

data. Volume-weighted average concentrations were calculated for each ion at every site. Site-specific total inorganic ion concentrations were computed by summing up these values at the respective sites. Then, ratios of the average concentrations of each anion and cation to this total inorganic ion concentration were calculated. These ratios are shown as percent of inorganic ions for all eight Illinois sites in figure 4. In this figure, anion and cation percentages are presented in a stacked bar totaling 100 percent for each site. Anion percentages are stacked on the bottom of the figure, beginning with $SO_4^=$ then NO_3^- and Cl^-, which is their order of importance. Average orthophosphate concentrations were below the analytical limit of detection and too small to show. Cation percentages are stacked on the top of the figure. Except for free hydrogen ion, H^+, the order in which cations are stacked is generally from least important (K^+) to most important (NH_4^+). The H^+ percentages are shown numerically in the clear bar that separates the anions from the other cations. This arrangement facilitates the comparison of the average precipitation chemistry among sites, and it depicts H^+ as making up the cations needed to push the total to 100 percent. Indeed, this is what happens chemically in acidic precipitation: the total cation concentration without H^+ must equal the total anion concentration, and to the extent the cations are deficient, H^+ makes up the difference.

Hydrogen ion percentages range from 17 percent at the Monmouth site in western Illinois (IL78) to 28 percent in the east-central and south-central parts of the state (Bondville-IL11, Salem-IL47, and Omega-IL99). At all sites, from IL19 in the north to IL63 in the south, there is little variation in the acidic anion contributions of sulfate, nitrate, and chloride. What changes most from site to site is the contribution of basic cations, (NH_4^+, Ca^{++}, Mg^{++}, Na^+, and K^+) that counterbalance or neutralize the acidic anions. The cations maximize at Monmouth (IL78), where H^+ is lowest, and minimize at the Bondville (IL11), Salem (IL47), and Omega (IL99) sites (see figure 4). At all sites in figure 4, $SO_4^=$ clearly is the largest contributor to the inorganic ion strength, i.e., the sum of the ion concentrations. Hydrogen ion is the next most important contributor, followed by NO_3^-, NH_4^+, and Ca^{++}. Using volume-weighted average concentrations, these five ions make up 89.4 to 93.1 percent (mean of 90.6 percent) of the inorganic ions in Illinois precipitation.

Taking concentrations in individual samples instead of averages, the sulfate concentration is highest in 65 to 82 percent of all Illinois samples. The order of importance of ions in individual samples is the same as for
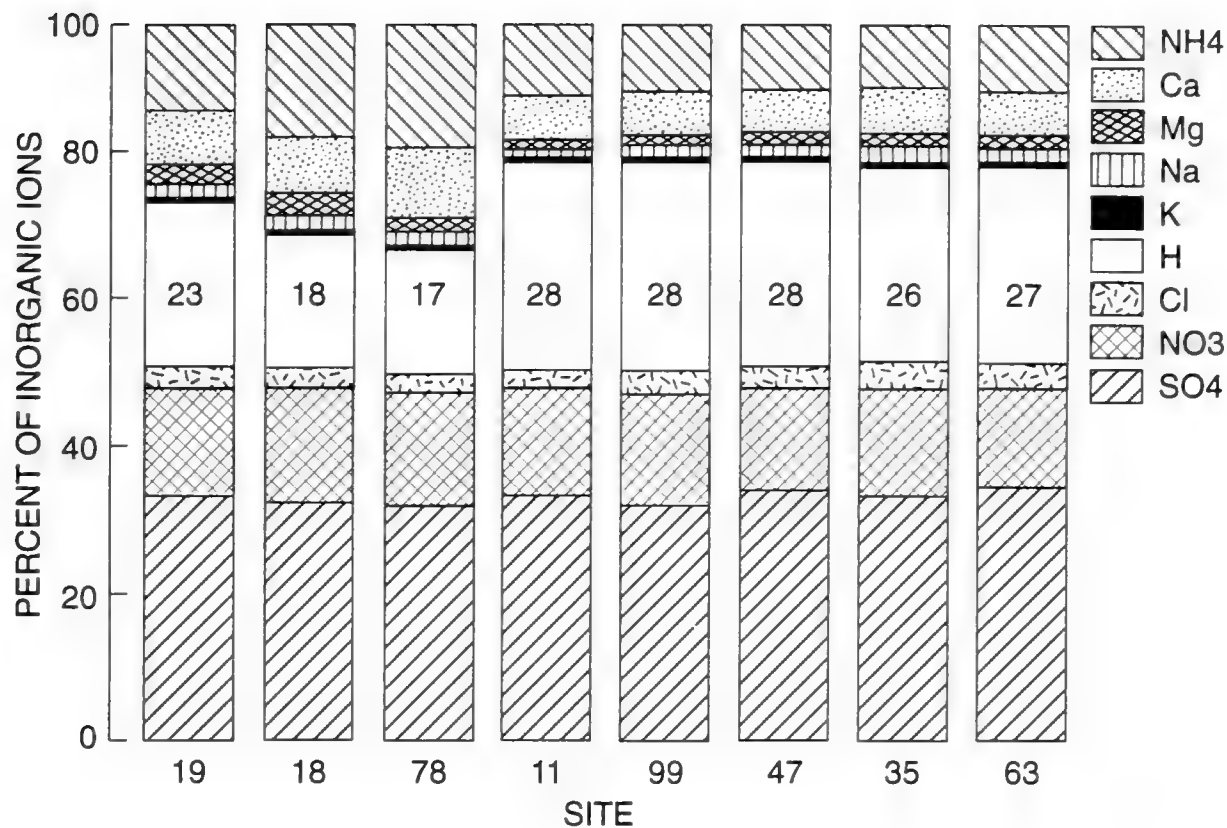
Figure 4. Ratios (in percent) of volume-weighted-average anion and cation concentrations to the total measured inorganic ion concentrations at Illinois sites (see figure 1 for site locations). Numbers in the clear stacked bar in the middle are percentages for the hydrogen ion, H⁺. Average orthophosphate concentrations were below the analytical detection limit and too small to show.

the average concentrations: $SO_4^= > H^+ > NO_3^- > NH_4^+ > Ca^{++}$. Notably, for all but 0.4 percent of the wet deposition samples in Illinois, one of these five ions has the highest concentration. Figure 5 presents the percentage ratios of the sum of these five ions to the sum of all ten ions in individual samples. These ratios are ranked from lowest to highest, and the rankings are depicted in this figure. For individual samples, 90 percent of the ratios are between 82.5 and 96.2 percent, which is consistent with the ratios when average concentrations are used.

A test of whether these five ions are sufficient to characterize the stoichiometry of Illinois precipitation is to check for a balance of positive and negative ions. In principle, liquid precipitation is an aqueous solution bearing no net positive or negative charge; thus cation (or positive ion) concentrations must equal anion (or negative ion) concentrations. To the extent that the cations and anions are out of balance, the measured set of ions is incomplete (some have been missed), the measurements are inaccurate (biased and/or imprecise), or a combination of these factors is at play. The charge balance was tested by taking the concentrations of the five largest contributors to the ion strength of individual samples and calculating a quantity labeled "ion percent difference." This quantity was defined by Stensland and Bowersox (1984) as

$$\text{Ion Percent Difference} = \frac{(\text{anion sum} - \text{cation sum})}{(\text{anion sum} + \text{cation sum})} \times 100 \text{ percent} \quad (2)$$

where (anion sum) is the chemical equivalent, or ionic charge, concentration of $SO_4^=$ plus NO_3^- and (cation sum) is the concentration of H^+ plus NH_4^+ plus Ca^{++}. Ion percent differences calculated in this way are summarized in figure 6. For 87.5 percent of the data in this figure, ion percent differences are less than 15 percent. This is consistent with the CAL experience for samples with an ion strength of about 0.2 milliequivalents per liter (meq/L) and pH values < 5.0, characteristics that are typical of Illinois precipitation. Based on the NADP/NTN Quality Assurance Plan (NADP/NTN, 1990), samples are not even targeted for reanalysis until ion percent differences are 15 percent, using the ten-ion set of measurements. A comparison of the original analysis with reanalysis results at the CAL verified that the original analyses were almost always correct, even when ion percent differences were 20 percent (Stensland and Bowersox, 1984). This study reported that 0.2 percent of the original numbers were in error. Thus, the magnitude of ion percent differences with the five-ion set is not markedly higher than for the more complete ten-ion set of measurements.

Finally, if analytical imprecision alone is to blame for the ion percent differences from the five-ion set, the number of positive and negative values taken over all samples would be virtually the same. Figure 6 shows a breakdown of the frequency of occurrence of positive and negative differences. Positive differences account for about 60 percent and negative differences for about 40 percent of the cases. This means that a cation deficit is 50 percent more likely to occur than an anion deficit (i.e., 3 cation deficits to every 2 anion deficits). This imbalance could result from an analytical bias or from ions not measured or not considered. Indeed, this imbalance is consistent with the ions not considered in the five-ion set. Looking at the averages in figure 3, Mg^{++}, Na^+, and K^+ together exceed Cl^- at every site. The skewness in the ion balance results in figure 6 reflects the charge imbalance in ions left out of the calculation.

It follows from these results that five ions dominate the ion strength of Illinois precipitation: $SO_4^= > H^+ > NO_3^- > NH_4^+ > Ca^{++}$. While these ions effectively control the solution stoichiometry, there is a small net cation deficit from the remaining ions. Illinois precipitation is best characterized as a dilute solution of mineral sulfuric (H_2SO_4) and nitric (HNO_3) acids, partially neutralized by dissolved NH_4^+ and Ca^{++}. The sulfur and nitrogen oxides result almost entirely from the combustion of fossil fuels, while the calcium and ammonium are from terrestrial sources. Since the five ions so dominate the chemistry of Illinois precipitation and since the other measured ions have no deleterious effects at the levels reported, the summary of wet deposition in this chapter focuses on the spatial and temporal distributions of these five ions.

Statistical distributions of pollutant concentrations. Precipitation in Illinois has a range of ion strengths from ~ 25 μeq/L to > 7,000 μeq/L, a factor of ~ 300 from low to high. Ion strength measures the inorganic pollutant concentration in ion equivalents, so it indicates how polluted the rain or snow is. A typical rain sample from an Illinois site might have a pH of 4.4; and if this were entirely due to dilute sulfuric acid, the ion strength would be ~ 80 μeq/L. The distributions of total ion strengths of wet deposition samples are shown for each site in figure 7. All of the median concentrations (located at the center of the notches in the box plots) are > 80 μeq/L, which is another indication that Illinois precipitation contains more than sulfuric acid (i.e., more than H^+ and $SO_4^=$). It is evident in this figure that more than half of the samples have an ion concentration between 100 and 300 μeq/L. From the results in the previous section, 90 percent of this is due to just five ions.
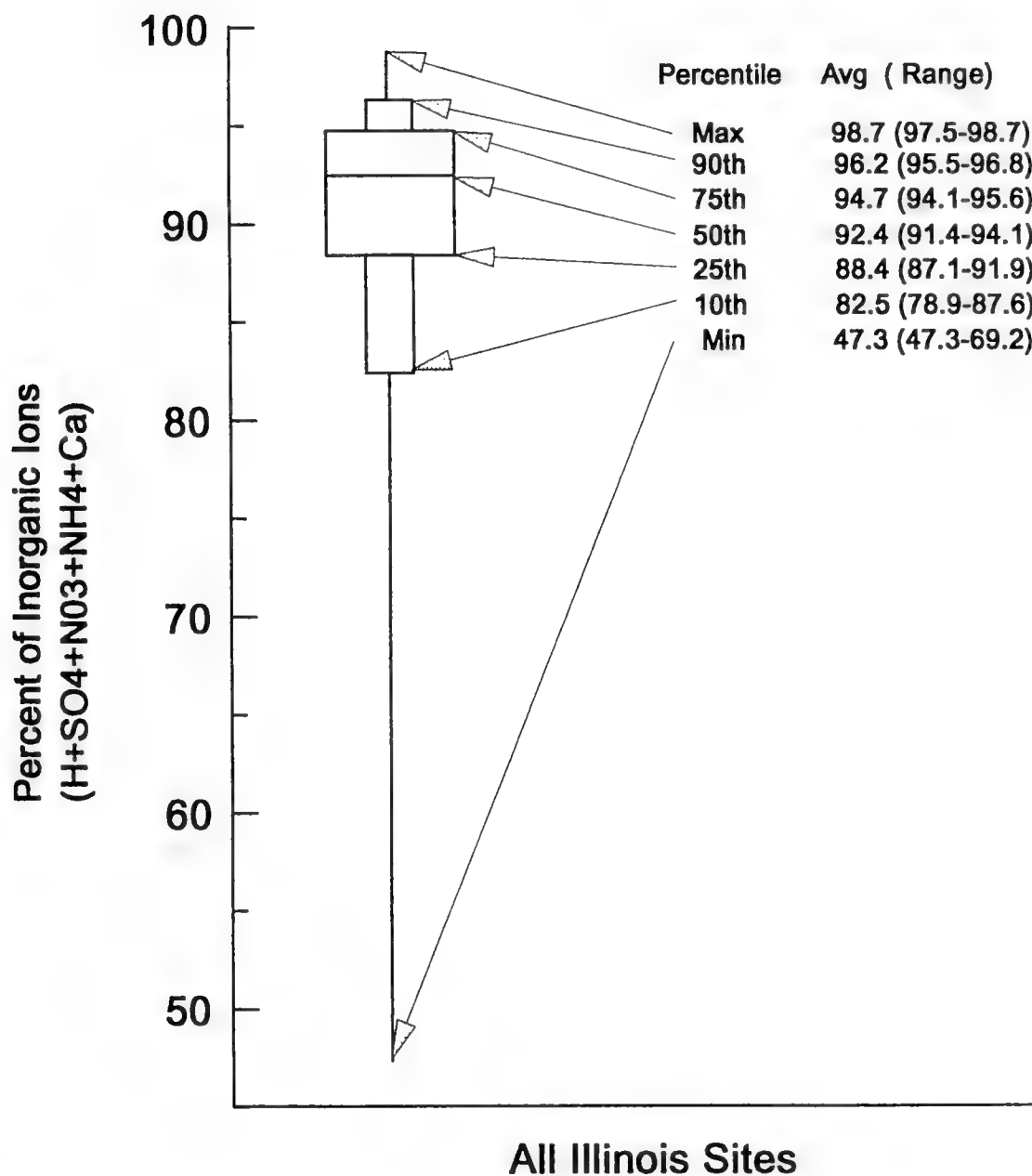
Figure 5. Distribution of the ratios (in percent) of the sum of concentrations of five ions (H⁺, SO₄⁼, NO₃⁻, NH₄⁺, and Ca⁺⁺) to the total measured inorganic ion concentrations in individual Illinois samples. The percentage values at seven reference percentiles in the distribution are listed, along with the range of percentages at the eight Illinois sites, when ranked separately (see table 1 for a list of sites).
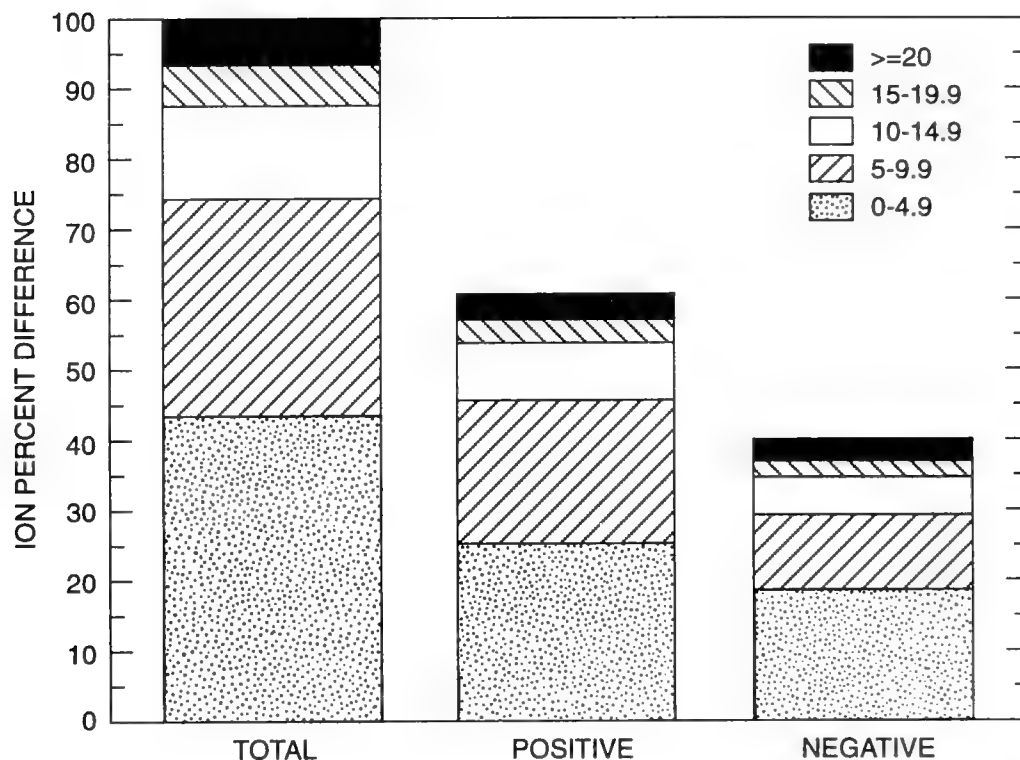
Figure 6. Ion percent differences calculated from the five-ion set ($SO_4^=$, H^+, NO_3^-, NH_4^+, and Ca^{++}). Differences appear in the five percentage frequency classes shown in the legend. Absolute values of the differences were used in compiling the frequency distributions labeled "TOTAL". Frequency distributions for net "POSITIVE" and "NEGATIVE" differences are also shown.
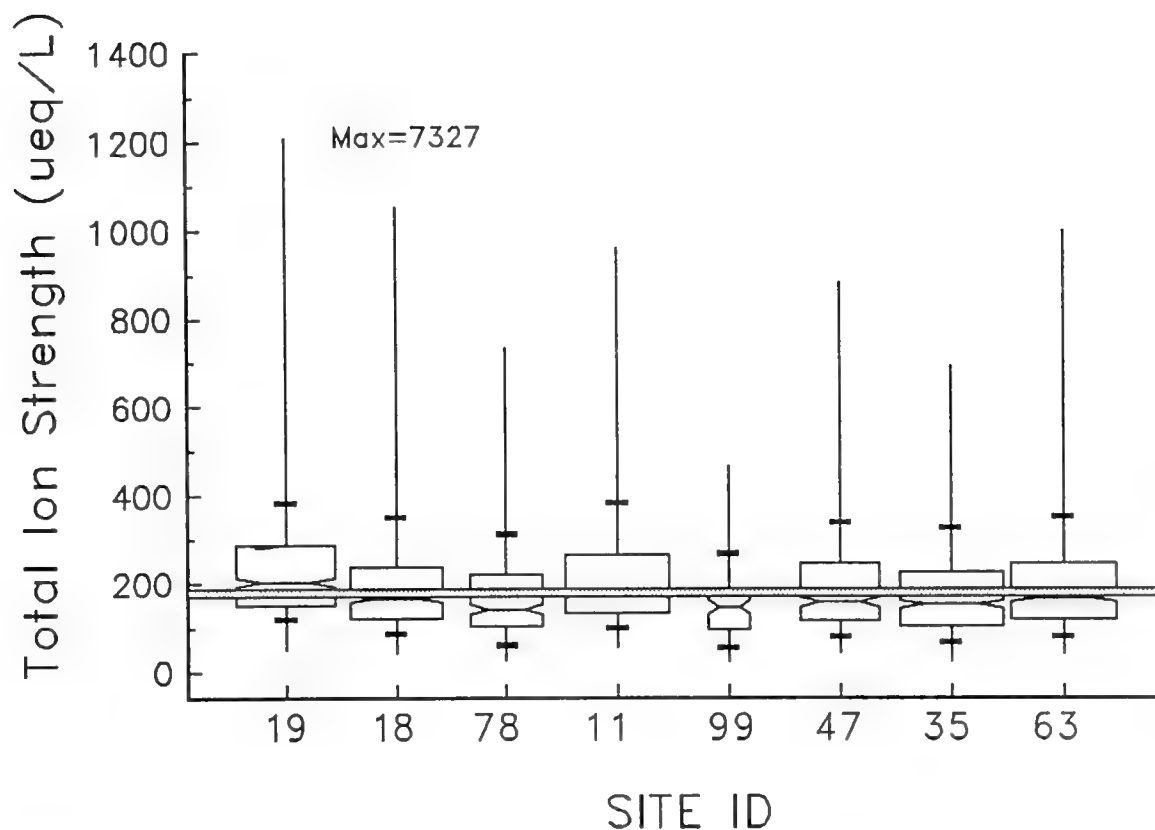
Figure 7. Distributions of total inorganic ion concentrations (Total Ion Strength) in Illinois precipitation at NADP/NTN sites (see figure 1 for site locations). Boxes enclose the 25th to the 75th percentiles and the notches mark the median and its 95 percent confidence interval. Box widths are proportional to the square root of the number of observations. Lines (whiskers) extend to the minimum and maximum except where indicated. Horizontal ticks on the whiskers indicate the 10th and 90th percentiles. For comparison, the shaded band marks the location of the notch for IL11.

A shaded band fills the notch (the median and its 95 percent confidence interval) of the box (25th to the 75th percentile) for Bondville (IL11) data in figure 7. Located in rural east-central Illinois, Bondville is roughly halfway between the three northern (IL19, IL18, and IL78) and three southern sampling locations (IL47/99, IL35, and IL63). The location of the notch for a site, either fully above or below this shaded area, indicates that the median pollutant concentration at that site is significantly above or below (respectively) the concentration at Bondville. Only at the Argonne site (IL19) in the Chicago area is the median ion strength significantly above (nearly 10 percent) that at Bondville, probably due to the influence of nearby urban sources. Median pollutant concentrations appear to be significantly below the Bondville level at the Monmouth site (IL78) in western Illinois and at the Carbondale site (IL35) in the forested southwestern part of the state. At the remaining sites, median pollutant concentrations, while below the Bondville value, are within the (shaded) confidence interval and are not significantly different than this reference. No north to south gradient is evident in these overall pollutant concentrations, outside of the possible urban influence on Argonne data from the Chicago area.

Free acidity, or the concentration of H^+ in precipitation is measured and reported in pH units; and in Illinois the observed pH range is from 3.10 to 8.45 (0.004 to 794 μeq/L). Acid rain has been defined as having a pH below 5.6, a value that is based on the free acidity of water in equilibrium with atmospheric carbon dioxide or CO_2 (Barrett and Brodin, 1955). Of Illinois wet deposition samples, 91.3 percent have pH values < 5.6. This 5.6 reference is based on the assumption that, except for CO_2, natural pollutants scavenged from the air by precipitation are neither acidic nor basic; i.e., they are neutral. Measurements from remote regions of the world, where natural pollutants dominate anthropogenic influences, belie this assumption (e.g., Galloway et al., 1982). "No single value for pH or any analyte can be regarded as a worldwide background value;" however, the average pH in humid remote areas is closer to 5.0 than 5.6 (Sisterson et al., 1990). Of Illinois wet deposition samples, 85.6 percent have pH values < 5.0. Taking either of these definitions, 8 to 9 out of every 10 weekly precipitation samples in Illinois can be termed "acidic deposition."

Figure 8 plots the distribution of pH measurements at each Illinois NADP/NTN site. The shaded band, filling the notch for the Bondville site (IL11), once again serves as a basis for comparing data among sites. Bondville pH values at the 10th, 25th, 50th, 75th, and

90th percentiles are the lowest among the sites, and the Bondville median is significantly below the medians at all but the IL63 and IL99 sites. This is consistent with the generally higher $SO_4^=$ and NO_3^- measurements at Bondville, shown in figures 9 and 10. At the IL78 site near Monmouth in western Illinois, the median pH is about 4.6 and the middle half of the distribution, the 25th through the 75th percentiles, is almost completely above the middle half of the distribution at Bondville. The free acidity at Monmouth is about half of the Bondville levels, owing to the combined effects of lower $SO_4^=$ and NO_3^- and higher Ca^{++} and NH_4^+. What's more, the spreads in the pH distributions, indicated by the differences in the 10th and 90th percentiles (horizontal reference marks on the whiskers in figure 8) are much larger at Monmouth (IL78), Shabbona (IL18), and Argonne (IL19) than they are at any of the sites in the southern half of the state. The more frequent occurrence of higher pHs at these sites causes the larger spread, which exceeds a pH unit (> 10-fold difference in H^+). Higher pHs occur when higher Ca^{++} or NH_4^+ or both are present to neutralize the $SO_4^=$ and NO_3^- acidity. These conditions occur more often at northern than southern Illinois sites.

It is the acidity of precipitation, along with the associated sulfur and nitrogen pollutants, which raises concerns about the potential for damage to agroecosystems and to unmanaged systems, such as forests. Nearly all of the reported damage to these ecosystems is the result of a combination of environmental stresses, including acidic deposition (Shriner et al., 1990; Barnard et al., 1990). It is generally not acidic precipitation alone that is the cause of reported damage. Direct damage, though rare, has been reported, however. "Phytotoxic concentrations of acidic precipitation are generally regarded to lie below pH 3.6" (Shriner et al., 1990). In Illinois, this occurs for 0.6 percent of the weekly wet deposition samples. There are no known reports of direct "acid rain" damage to Illinois crops or forests in the literature, due to these rare, very acidic events.

Figures 9 and 10 contain the notched box-and-whisker plots of $SO_4^=$ and NO_3^- concentrations at Illinois sites. Median $SO_4^=$ concentrations range from 2.14 to 3.14 mg/L, with an overall median of 2.64 mg/L; and median NO_3^- concentrations range from 1.37 to 1.84 mg/L, with an overall median of 1.55 mg/L. Sulfate to nitrate mass concentration ratios of ~ 1 1/2 to 1 are typical of Illinois precipitation. In ion equivalent (or charge) concentration units, this ratio is ~ 2 to 1, meaning precipitation would have about two units of sulfuric acid for every unit of nitric acid, if both $SO_4^=$ and NO_3^- were totally unneutralized. Using the shaded
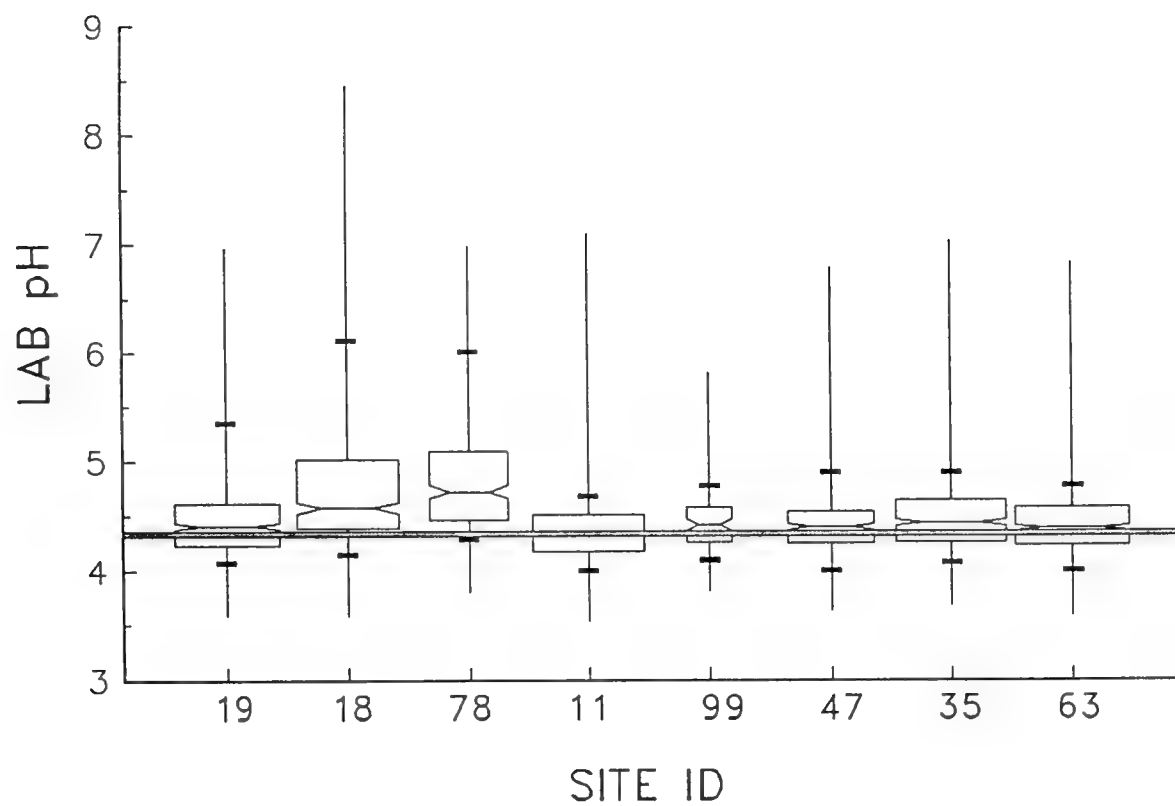
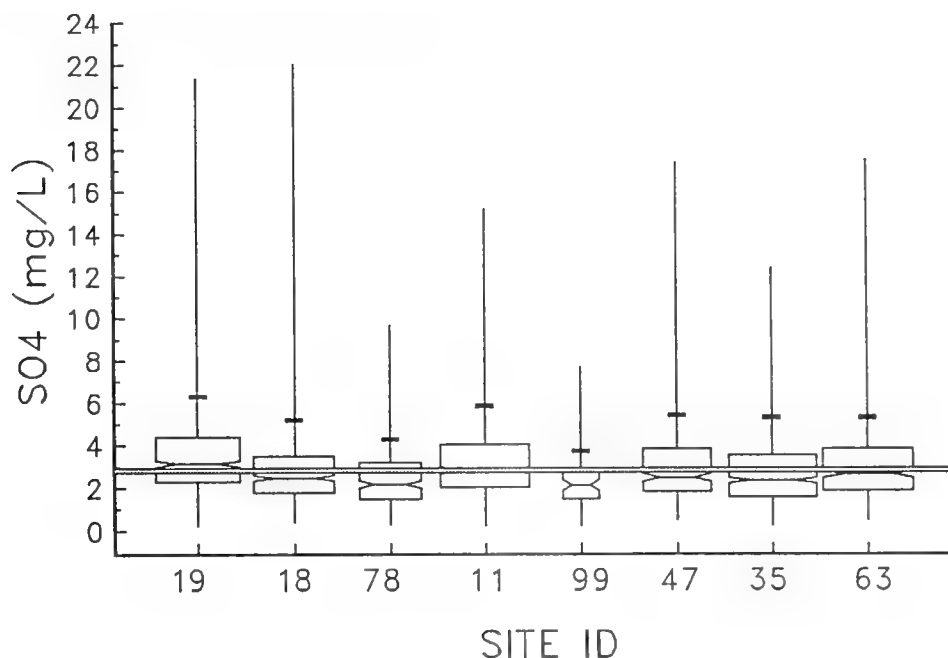Figure 8. Same as figure 7, except for laboratory (CAL) pH measurements.

Figure 9. Same as figure 7, except for sulfate (SO₄) mass concentrations in mg/L. Horizontal ticks marking the 10th percentiles on the lower whiskers were too close to the 25th percentiles to show. From left (IL19) to right (IL63) the 10th percentile values are 1.60, 1.25, 1.04, 1.51, 1.03, 1.39, 1.11, and 1.39, respectively.
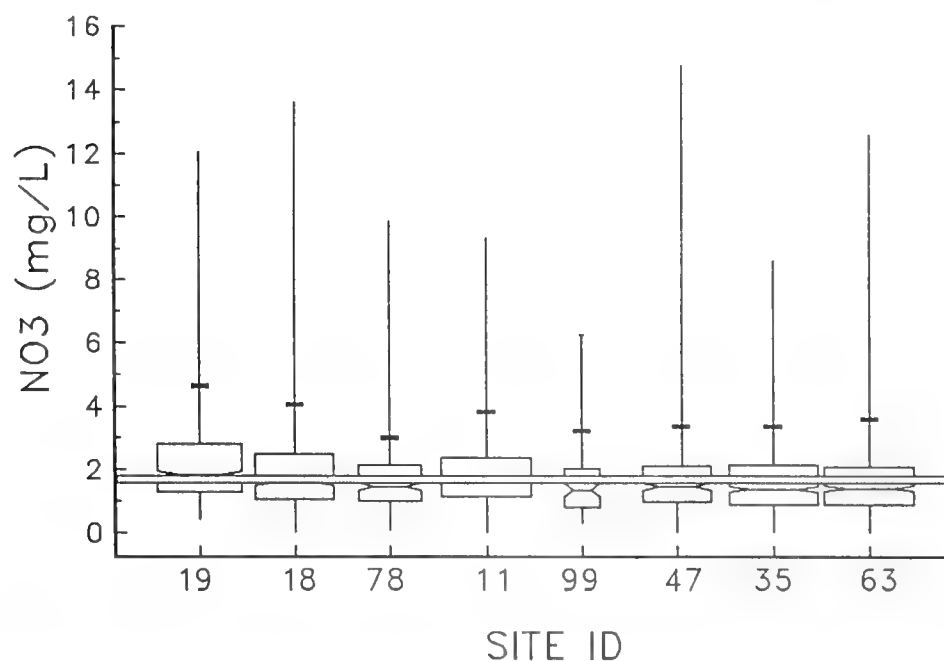

Figure 10. Same as figure 7, except for nitrate (NO₃) mass concentrations in mg/L. Horizontal ticks marking the 10th percentiles on the lower whiskers were too close to the 25th percentiles to show. From left (IL19) to right (IL63) the 10th percentile values are 0.97, 0.79, 0.64, 0.79, 0.58, 0.57, 0.60, and 0.59, respectively. Also the lowest five values at IL35 and the minimum at IL11, IL18, IL47, and IL63 were at the analytical limit of detection (0.93 mg/L).

band in the figures as a reference, the similarity in behavior of these two pollutants becomes apparent. While the magnitudes of the respective $SO_4^=$ and NO_3^- deviations from the reference (i.e., the IL11 median with its 95 percent confidence interval) are not identical, IL19 is above IL11 and has the overall highest concentrations for both ions. In general, $SO_4^=$ and NO_3^- concentrations for IL18, IL35, IL47, IL63, IL78, and IL99 are below IL11; and IL99 data are lowest. Choosing reference points other than the median with its confidence interval gives similar results when the two ions are compared; thus covariance is evident in the $SO_4^=$ and NO_3^- concentration profiles.

To quantify the covariance that appears in figures 9 and 10, correlations for $SO_4^=$ and NO_3^- concentrations were calculated. Valid samples from all eight Illinois sites were used in the calculations. As a basis for comparison, correlations among the other three major ions and $SO_4^=$ and NO_3^- were included in the analysis. Results are presented in table 2, which lists the correlation coefficients or R-values for the relationships between each ion and the other four ion concentrations in individual precipitation samples. Only linear relationships were tried, which means that the magnitude of the R-values indicates the degree to which one concentration is proportionate to the other concentration being tested. High positive R-values indicate that as one concentration goes up the other goes up in the same fractional (or proportionate) amount. A value of 1.0 indicates a perfect positive correlation. Negative R-values mean that as one ion goes up, the other goes down. It is apparent from the results in table 2 that many of the R-values are > 0.60 and are similar in value, but the $SO_4^=$ and NO_3^- correlation is best; it has the highest R-value (0.74). Squaring the R-values measures the fraction of ion concentration variance that is accounted for by covariance; and for $SO_4^=$ and NO_3^-, R^2 in percent is 55. In Illinois precipitation, over half of the variation in $SO_4^=$ concentrations can be explained by the fact that they track NO_3^- concentrations, and vice versa.

One implication of the correlative behavior of $SO_4^=$ and NO_3^- in precipitation is that ecosystem exposure to high $SO_4^=$ is likely to be accompanied by high NO_3^-. Recall from figure 4 and the discussion in the previous section that $SO_4^=$ and NO_3^- have the highest and third highest average concentrations, respectively, in Illinois precipitation. Free acidity, H^+, has the second highest average concentration. In table 2, the correlations of H^+ and $SO_4^=$ ($R^2 = 38$ percent) and H^+ and NO_3^- ($R^2 = 19$ percent) are not as high as for several other pollutants, however. Notwithstanding the lower correlations of H^+ with $SO_4^=$ or NO_3^-, comparisons of the concentration

distributions (i.e., the notched box-and-whisker plots) of these three pollutants in figures 8-10 show that where the notch falls above the shaded band in figure 8, it falls below the band in figures 9 and 10, except for the Argonne (IL19) site. In other words, generally higher pH values at a site and generally lower $SO_4^=$ and NO_3^- concentrations go together, and vice versa. Where $SO_4^=$ is high, so too are H^+ and NO_3^-; and from the correlation results, when $SO_4^=$ is high, NO_3^- is more likely than any other ion to be high too. The exceptional behavior at IL19 is another indication of urban Chicago influences on Argonne precipitation. Higher airborne dust levels with higher accompanying Ca^{++} concentrations may be neutralizing the higher $SO_4^=$ and NO_3^- in IL19 precipitation.

Other implications of the correlative behavior of $SO_4^=$ and NO_3^- in precipitation are that

1. Precipitation scavenges these two pollutants from the air similarly.
2. The mixing ratio of these pollutants in air varies over a narrow range.
3. Sulfur and nitrogen oxides are emitted from the same sources.
4. A combination of these factors is at work in the atmosphere.

Figure 11 presents the NH_4^+ concentration distributions in precipitation at Illinois sites. Median concentrations range from 0.26 to 0.45 mg/L, with an overall median of 0.35 mg/L. Unlike the $SO_4^=$ distributions, in which all values are above the analytical detection limit, and NO_3^- distributions, with only 0.3 percent detection limit values (see figure 10), the frequency of occurrence of detection limits in the NH_4^+ concentrations is 3.6 percent. Detection limit values occur at every Illinois site, from a low of 1.6 percent at Argonne (IL19) to a high of 6.0 percent at Carbondale (IL35); and so each of the lower whiskers in figure 11 terminates at 0.02 mg/L, the NH_4^+ detection limit. That 28 percent of the concentrations are within a factor of 10 of this detection limit results in greater analytical uncertainty for NH_4^+ (15-17 percent) than for the four other major ions (James, 1992). Sites north (IL19, IL18, and IL78) of Bondville have higher NH_4^+ concentrations than sites to the south (IL99, IL47, IL35, and IL63), although only IL35 is significantly different. This geographical pattern is markedly different than for pH, $SO_4^=$, or NO_3^-, but is similar to the Ca^{++} pattern in figure 12. Of note, too, is that the NH_4^+ concentrations at the Argonne site (IL19) are not the highest, as clearly was the case for $SO_4^=$ and NO_3^-.

Table 2. Linear Pearson Correlation Coefficients (R Values) and R-squared Values, for the Pairwise Correlations among the Five Major Inorganic Ion Concentrations in Illinois Precipitation (number of observations = 2711; all values significant at better than the .01 percent level except the H^+ and Ca^{++} correlation, which is significant at the 5 percent level)

	$SO_4^=$		NO_3^-		Ca^{++}		NH_4^+		H^+	
	R	R^2*	R	R^2*	R	R^2*	R	R^2*	R	R^2*
$SO_4^=$	1.0	1.0	.74	.55	.59	.35	.63	.40	.62	.38
NO_3^-	.74	.55	1.0	1.0	.66	.43	.69	.47	.44	.19
Ca^{++}	.59	.35	.66	.43	1.0	1.0	.55	.30	-.06	.00
NH_4^+	.63	.40	.69	.47	.55	.30	1.0	1.0	.11	.01
H^+	.62	.38	.44	.19	-.06	.00	.11	.01	1.0	1.0

Note:
*Bonferonni-adjusted probabilities (Wilkinson, 1990).
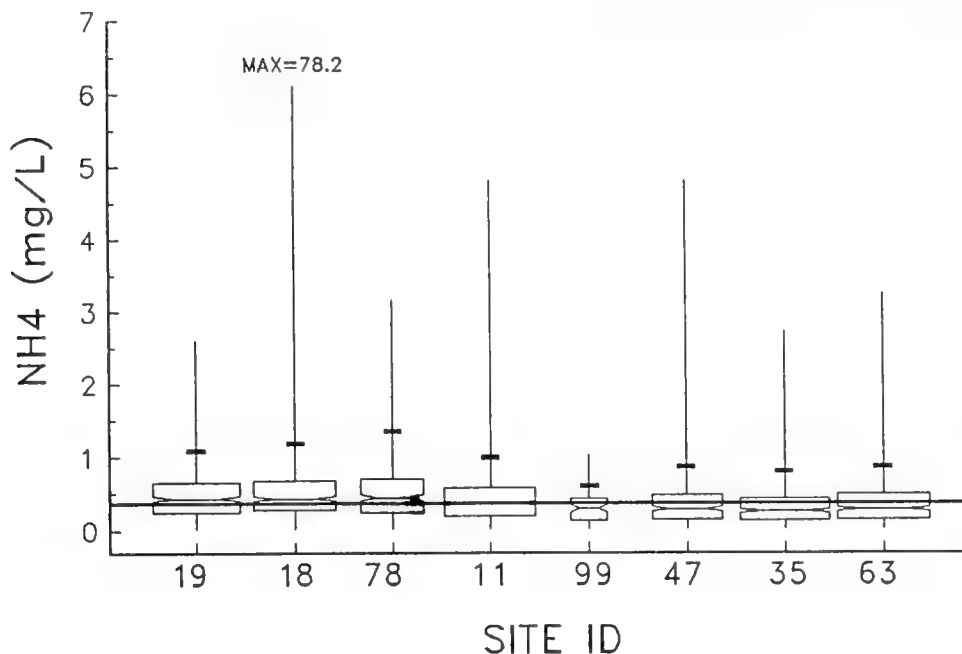
Figure 11. Same as figure 7, except for ammonium (NH$_4$) mass concentrations in mg/L. Horizontal ticks marking the 10th percentiles on the lower whiskers were too close to the 25th percentiles and the minimums to show. From left (IL19) to right (IL63) the 10th percentile values are 0.14, 0.16, 0.12, 0.10, 0.07, 0.07, 0.06, and 0.08, respectively. Also all of the minimum values and from 1.6 to 6.0 percent of the other low values in the distribution are the analytical limit of detection (0.02 mg/L).
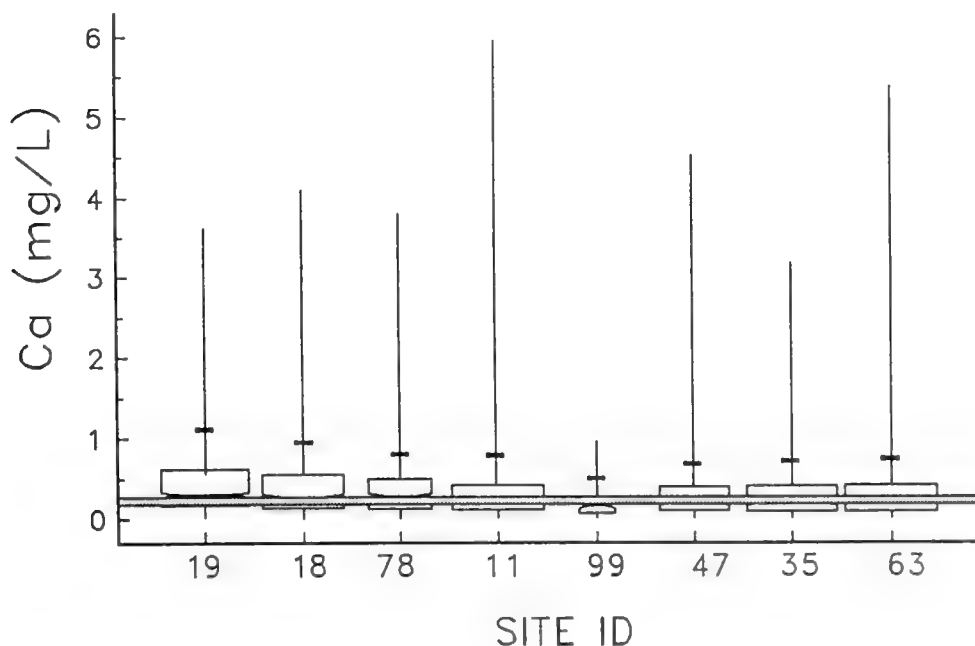
Figure 12. Same as figure 7, except for calcium (Ca) mass concentrations in mg/L. Horizontal ticks marking the 10th percentiles on the lower whiskers were too close to the 25th percentiles and the minimums to show. From left (IL19) to right (IL63) the 10th percentile values are 0.0997, 0.077, 0.060, 0.058, 0.034, 0.050, 0.050, and 0.053, respectively. Also the lowest three values at IL35 and the minimum at IL19 are the analytical limit of detection (0.009 mg/L).

Ammonium is a nutrient and is one of two nitrogen compounds deposited by precipitation, the other being NO_3^-. In mass concentrations units, NO_3^- and NH_4^+ rank third and fourth in importance in Illinois precipitation, with overall median concentrations of 1.55 and 0.35 mg/L, respectively. In these units, the NO_3^- median is over four times the NH_4^+ median. To assess the importance of these two pollutants as nutrients deposited on Illinois crops, forests, lakes, and streams, however, it is their nitrogen content that is relevant. As a fraction of the total mass, nitrogen is 22.6 percent of NO_3^- and 77.6 percent of NH_4^+. Converting the overall median concentrations, above, to nitrogen (N) mass units yields 0.35 and 0.27 mg/L for NO_3^- and NH_4^+, respectively. Expressed this way, the median N from NO_3^- is just 30 percent more than the median N from NH_4^+. What's more, NH_4^+ and NO_3^- have the second highest correlation coefficient of all the pollutants reported in table 2. R^2 is 0.47, meaning 47 percent of the total variation in these two N compounds in precipitation is accounted for by their covariance.

Exposure studies have not shown that crop or forest damage has occurred from the ambient N levels found in wet deposition in the United States (Shriner et al., 1990; Barnard et al., 1990), however, even for the most extreme events. On crops, the N from NO_3^- and NH_4^+ is likely to enhance growth and productivity. On certain unmanaged high-elevation forests, there is some evidence that the N addition from wet deposition over time may stimulate excess growth, which could predispose trees to winter damage (Shriner et al., 1990). Illinois forests are not exposed to the harsh conditions that sometimes occur at high elevations and are not likely to be affected negatively by N deposition, but this should be assessed. It is not known to what extent wet-deposited N accumulates in Illinois lakes and may contribute to water quality problems and eutrophication.

One final observation from figure 11 is the lone flier reported at the Shabbona site (IL18). The NH_4^+ in this sample was 78.2 mg/L, more than ten times the second highest value at this site or the maximum at any other site. This same sample appears as the lone flier in figure 7 and is a flier for potassium and orthophosphate, as well. Although this sample passed all of the screening criteria discussed in the earlier section on screening procedures, it is a clear anomaly. No visible contaminants were reported, but the extreme NH_4^+, PO_4^{3-}, and K^+ strongly suggest contamination by fertilizer. It has a disproportionately large influence on correlation analyses and so was excluded from the results presented in table 2.

The notched box-and-whisker plots for Ca^{++} appear at each of the eight Illinois sites in figure 12. Median Ca^{++} concentrations range from 0.159 to 0.296 mg/L, with an overall median of 0.238 mg/L. A geographic pattern of higher values in northern Illinois than in southern Illinois, described in the discussion on NH_4^+, is indicated in this figure. The highest concentrations occur at the Argonne site (IL19), where Ca^{++} values are 8 to 26 percent above the second highest site values at every reference percentile (minimum, 10th, 25th, 50th, 75th, and 90th) in the distribution except the maximum. Despite the higher concentrations at IL19, none of the notches appears either completely above or below the shaded band, which means that all of the median concentrations can be interpreted as being within the 95 percent confidence interval of the Bondville (IL11) median. This is consistent with the large relative uncertainty or variance in Ca^{++} data. Seventeen percent of all Ca^{++} concentrations are within a factor of ten of 0.009 mg/L, the analytical detection limit; and precision estimates are typically 10 to 16 percent (James, 1992).

There are no reports of real or potential ecological damage from Ca^{++} in precipitation. It is important in Illinois wet deposition because it is at sufficiently high concentrations to offset the free acidity effected by $SO_4^=$ and NO_3^-. It is the only major ion that has a negative correlation coefficient with H^+ in table 2, although almost no variance ($R^2 \sim 0$) is explained by this relationship. A study to explain the sharply lower pH (higher H^+) in eastern U.S. precipitation in the 1970s, compared with the 1950s, reported that Ca^{++} (and magnesium) decreases, not $SO_4^=$ and NO_3^- increases, were the cause (Stensland and Semonin, 1982). High airborne dust levels from widespread droughts in the central United States were cited as the major source of the higher Ca^{++} in the 1950s. Soil aerosols, road dust (Barnard et al., 1986), and construction and demolition activities are important sources of Ca^{++} today. Calcium remains one of five major ions in Illinois precipitation.

Overall, figures 9-12 exhibit some features that are common to precipitation chemistry data from all NADP/NTN sites (Knapp et al., 1988). Pollutant concentrations tend to be skewed by a few large outliers. This is evident in the asymmetry of the notched box-and-whisker plots. The upper whiskers (top 25 percent) are often several times longer than the lower whiskers (bottom 25 percent); and the distances from the tick marks at the 90th percentiles and the ends of the whiskers (the top 10 percent) are often several times longer than the distances between the 75th and 90th percentiles. This skewness is typical of many environmental variables, particularly chemical concentrations, which

are bounded by zero but have no upper limit. A log transformation of the data removes a considerable amount of this asymmetry. Indeed, log Ca^{++} concentrations have a distribution that cannot be distinguished from a normal distribution, which is the classic bell-shaped curve. A statistical test of the hypothesis: log concentration distribution equals normal distribution, was used to make this observation (STATGRAPHICS, 1992; Kolmogrox-Smirrov "goodness-of-fit" test at the one percent confidence level). Applying the same test to log NO_3^-, NH_4^+, or H^+ concentrations does not result in normal distributions, i.e., the probability is < one percent that log concentrations have a normal distribution. For H^+, this is tantamount to the observation that pH measurements do not have a normal distribution, since pH is proportional to log (H^+). Log $SO_4^=$ concentrations cannot be distinguished from a normal distribution at the one percent confidence level but can at the 10 percent level.

Observing that pollutant concentrations in precipitation are skewed (figures 9-12) and knowing how to describe the concentration distributions with mathematical expressions helps effects researched to study and understand the eccosystem response to pollutant experience. What's important is that pollutant concentrations in Illinois precipitation often range over two or three orders of magnitude, beginning near zero. This means that ecosystems are exposed to many low and moderate concentrations and to a few concentrations that may be 100 to 1000 times higher. In addition, there is a substantial amount of covariance among the ions, which means that these high concentrations tend to occur at the same time. Recall that the frequency of occurrence of pH < 3.6, at which direct damage to some plants has been reported (Shriner et al., 1990), is 0.6 percent in Illinois. For these same samples, $SO_4^=$ is in the top 1.3 percent, NO_3^- is in the top 9.6 percent, and NH_4^+ is in the top 16 percent; Ca^{++} values are low and high. This correlative behavior of precipitation concentrations is quantified in table 2: for $SO_4^=$ and NO_3^-, R^2 is 55 percent; for NH_4^+ and NO_3^-, Ca^{++} and NO_3^-, and NH_4^+ and $SO_4^=$, R^2 is 40 to 47 percent. Among the five major ions, the H^+ correlations are lowest; however, when H^+ is correlated with the other ions in a charge balance relationship, R^2 improves to 80 percent. This is yet another way to show that Illinois precipitation is a dilute solution of sulfuric and nitric acids, partially neutralized by ammonium and calcium.

Dry Deposition. The complex phenomena of pollutant mass transfer by nonprecipitation processes from the atmosphere to receptors at the earth's surface, i.e., dry deposition, are often explained and examined by way of analogy to an electrical circuit. Indeed, such a conceptual model forms the basis for coupling together the computational methods used to arrive at the NOAA/ATDD results for V_d (Hicks et al., 1991). The series resistance model shown in figure 13 (after Davidson, 1988) considers the airborne *concentration* of a particular chemical to be analogous to an *electrical potential* or voltage. The receptor surface is then taken to be a sink, at zero potential, or at the ground state. Pollutant transfer from the atmosphere to the surface actually occurs when particles or gas molecules pass through various layers in the atmosphere and there is a physical or chemical reaction at the surface. Different processes control the rate of transfer in each layer. In the series resistance analogy, this transfer is an *electrical current* that flows through a series of resistors, which control the amount of current that passes.

In their simplest form then, the three "resistors" in the series resistance model represent atmospheric processes. R_a or aerodynamic resistance represents the turbulent processes in the lower levels of the atmosphere that mix and deliver airborne materials to thenear surface atmosphere. R_b or boundary-layer resistance represents the effect of the receptor surface on small-scale air turbulence and the rate at which particles and gas molecules can be brought into close enough proximity to the receptor surface to actually react chemically or "stick" physically. R_c or surface resistance represents the ability of the surface to retain the materials brought into reactive proximity. A fourth resistor (R_g or gravitational settling resistance) represents the rate at which particles literally "fall" if they are large or massive enough that gravitational effects dominate the process.

As in the analogous electrical circuit, the higher the resistance, the lower the current. The resistance with the highest value becomes the overall rate limiter for the entire process. Mathematical tools that apply to the analyses of electrical circuits apply exactly to this conceptual framework. Additional resistances in series or parallel to the simple three-resistor model can be used to differentiate and examine the effects of additional processes. Large to moderate scale atmospheric processes, and thus daily weather patterns, affect primarily R_a. The surface type and configuration affect the way a receptor is coupled to the atmosphere, and thus R_b. Finally, the type of material at the surface, its biological state, composition, physical state, chemical reactivity, and especially wetness, impact R_c. Overlaying the resistance analogy, and adding additional complexity to the deposition rate estimation, is the fact that different pollutants have different resistances. The
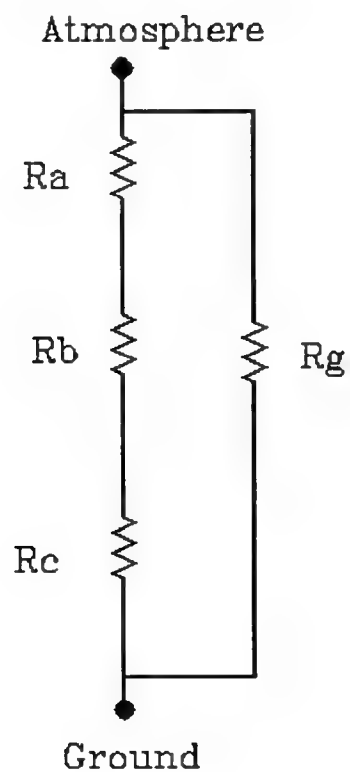
Notes:
R_a = aerodynamic resistance
R_b = boundary-layer resistance
R_c = surface resistance
R_g = gravitational settling resistance

Figure 13. Schematic representation of dry deposition processes in the so-called series resistance analogy.

aerodynamic size and solubility of aerosols, and the reactivity of gases with various types of surfaces greatly impact the R values. In a simple series resistance circuit, the overall resistance (R_{total}) is just the sum of the individual elements:

$$R_{total} = R_a + R_b + R_c$$

This is the basic form of the model for gases and fine aerosol. For these physical forms of pollutants in the atmosphere, gravity has little or no effect on the deposition rate, and R_g has a very large value relative to the other three terms. Where parallel resistances exist in an electrical circuit, R_{total} is found by inverse summation, so that including all four resistances in the model shown in figure 13 produces the equation:

$$1 / R_{total} = (1/(R_a + R_b + R_c)) + (1/R_g)$$

For very large particles, such as aerosol with aerodynamic diameters of several μm, or raindrops whose size is measured in millimeters (mm), R_g becomes a relatively very small number. In this case, the $(1/R_g)$ term becomes very large, and the effect of gravity dominates the process. V_d is a conductance, or inverse resistance term, and is calculated simply as

$$V_d = 1/R_{total}$$

Therefore, many atmospheric and surface parameters are considered to produce a single V_d value, which is specific for the pollutant type and average surface type modeled. The two NOAA/ATDD sites (Bondville and Argonne) were originally selected to represent regionally typical conditions. The Argonne site is at the southwestern fringe of the Chicago metropolitan area. and is characterized by mixed forest and suburban residential vegetation, with numerous relatively local pollution sources. In contrast, the Bondville site is farmland, rotated annually between soybean and corn production, with very few nearby pollution sources. The V_d estimation model takes into account these general site characteristics, as well as day-to-day changes in vegetation status and land use reported by the site operators. In the NOAA/ATDD model, large particles, small particles, ozone, nitric acid, and sulfur dioxide are each treated as a separate case for which V_d values are computed weekly. Finally, this conductivity term is multiplied by the potential term, concentration, and the sampling interval, in units of time (here, one week) to arrive at the dry deposition amount:

$$Deposition = V_d \cdot concentration \cdot time$$

In the data reported here, this equation was used with appropriate conversion factors to express all dry deposition amounts as kilograms of material depositing per year to one hectare of area (kg ha^{-1} yr^{-1}). To convert to pounds per year to one acre (lbs ac^{-1} yr^{-1}), multiply the deposition value shown by 1.12.

The first step in examining the spatial variation in dry deposition, involves a comparison of the V_d results for the two NOAA/ATDD sites. Figure 14 shows the distribution of weekly mean V_d values observed at the two sites during the 1985-1992 period for which data are available. For each of the three pollutants shown (HNO_3, SO_2, and aerosol), the median V_d at Argonne is somewhat higher than at Bondville due to the site characteristics and long-term climatic differences in the two sites. Most important among these factors is the greater roughness scale at Argonne, due to land use and vegetation type. These factors are expressed primarily in the R_a and R_b components of the V_d computation. Larger differences are seen in figure 14 between the three pollutant species than between sites. This is due to the R_c component, and the differences are caused largely by the varying affinities of the pollutants for surfaces. Aerosol particles may "stick" or "bounce off" surfaces, and R_c is influenced by the physical nature of the surface. SO_2 gas is very reactive with surfaces when aqueous films are present to facilitate the chemical reactions that convert SO_2 to $SO_4^=$ at the surface, so the R_c component for SO_2 is strongly controlled by surface wetness. HNO_3 is also a gas, but it has a very small R_c value for all surface types, whether wet or dry, and thus HNO_3 has the largest V_d values overall.

Figure 15 is similar to figure 14, but shows the corresponding Argonne and Bondville weekly mean air concentration distributions for HNO_3, SO_2, and $SO_4^=$ ($SO_4^=$ is strongly associated with small aerosol, i.e., diameter < 1 μm). The differences here are attributable to the suburban versus rural nature of the two sites. The SO_2 concentration at Argonne is somewhat higher, simply due to the higher density of SO_2-emitting sources in the Chicago-Joliet region, as compared to the agricultural region around Bondville. $SO_4^=$ in fine aerosol, by contrast, is largely a secondary pollutant, formed by the reaction of gas phase SO_2 over time. Thus, $SO_4^=$ concentrations tend to vary regionally, rather than locally; concentration distribution and the local sources may have somewhat less of a direct impact. HNO_3 concentrations appear to be quite similar at both sites; the high rate of dry deposition, the reactivity of the gas with aerosol constituents, and the volatile nature of some of the reaction products, preclude generalizations about HNO_3 distributions.
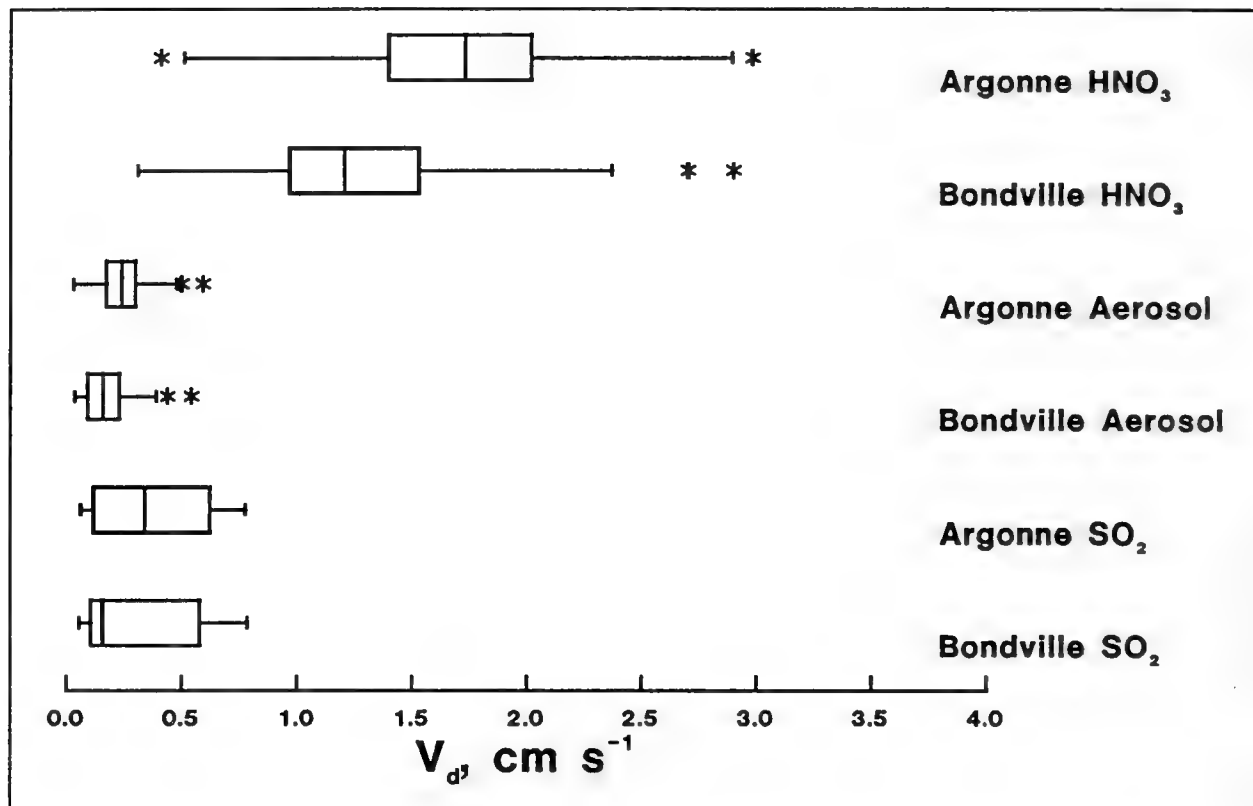
Figure 14. Comparison of the distributions of weekly mean dry deposition velocities (V_d) determined at two Illinois sites (Argonne, IL 19, and Bondville, IL11) over the period 1985-1992.
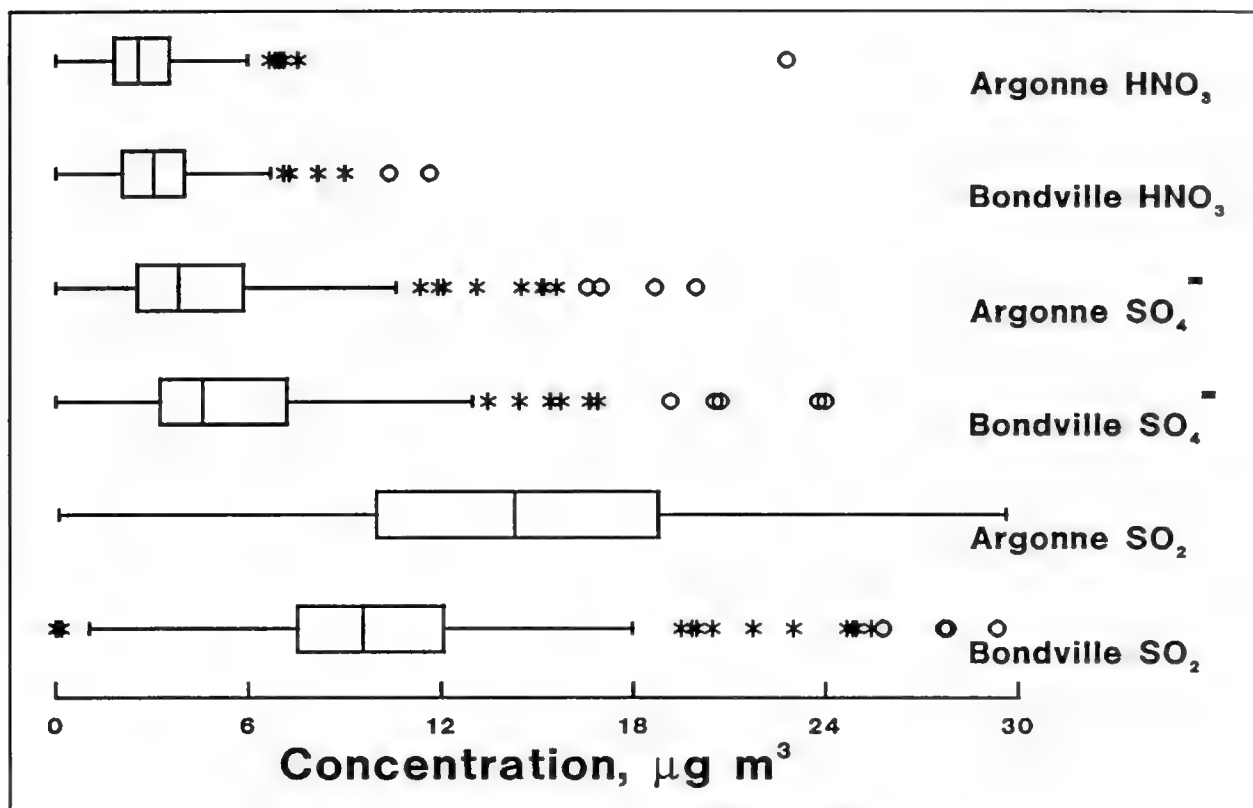
Figure 15. Comparison of the distributions of weekly mean airborne concentrations of three pollutants µg m⁻³) at two Illinois sites (Argonne, IL19, and Bondville, IL11) over the period 1985-1992.

Finally, the V_d and concentration data are combined in figure 16. This figure shows the computed dry deposition amounts for the two NOAA/ATDD sites. Dry deposition loading estimates for the period 1984-1990 were computed using annual median pollutant concentrations from IEPA monitoring data (see *Air Quality Trends in Illinois* chapter). Separate medians were derived for the Chicago and the "Remainder of State" areas. These concentration values were then combined with V_d statistics based on the model results from the NOAA/ATDD network data set, to produce the loading estimates shown in figures 17-28. Each figure presents the annual loading values in kg ha^{-1} yr^{-1} for a particular pollutant, with plot (a) showing the Chicago-area data and plot (b) showing the "Remainder of State" data. Note that while data were not available for all years of the overall period, the time scale of all plots is constant. For each year, the distribution of NOAA/ATDD weekly mean V_d data was analyzed. The product of the median pollutant concentration (from IEPA measurements) and the annual *mean* V_d value is shown as a horizontal line. The vertical line represents the loadings at ± one *standard deviation* in V_d. Finally, the triangles pointing up and down indicate the result if the *maximum and minimum* V_d, respectively, are used in the loading calculation.

The Argonne NOAA/ATDD site V_d values were used to compute the Chicago-area loadings; Bondville V_d values were used for the "Remainder of State" results. This introduces some additional imprecision in the results, since there exists much variation in land use, topography, and vegetation type in the "Remainder of State" area, while the Bondville V_d values are computed for farmland. While modeling tools (Matt and Meyers, 1993) and land-use and vegetation type data (see chapter with GIS data) exist for much finer resolution estimations of V_d, development of the calculations was beyond the scope of the current project.

From this series of plots (figures 17-28), it is obvious that dry deposition in Illinois tends to be somewhat higher in the Chicago area. This is due to both higher airborne concentrations for most pollutants and higher deposition velocities. For arsenic, manganese, nitrate, ozone, sulfur dioxide, and sulfate (figures 17, 21-24, and 26-27), the differences are on the order of 10 to 30 percent. For cadmium, chromium, iron, nickel, lead, and TSP (figures 18-20, 22, 25, and 28), dry deposition in the Chicago area exceeds the rest of the state by 200 to 400 percent. This is primarily caused by the differences in air quality (see *Air Quality Trends in Illinois* chapter). Temporal trends in dry deposition generally follow air quality trends, although additional variability is introduced into the time-series data by interannual variation in deposition velocities.

Spatial variation in dryfall was examined using the NADP/NTN "dry bucket" data from five Illinois sites for which a long-term record was available (figure 1 and table 1). Figures 29-31 show the distributions of dryfall loading amounts observed in the 8-week integrated samples at the five sites for calcium, ammonium, and sulfate. Dryfall data for other ions (the dryfall samples were analyzed for all of the same ions as the wet deposition samples) is not considered to be a reliable indicator of actual dry deposition rate. This is because the polyethylene bucket, used in the NADP/NTN wet/dry collector, cannot account for the aerodynamic and chemical reactivity characteristics of natural environmental surfaces. For calcium and the large-particle fraction of ammonium and sulfate, however, there appear to be less significant interfering chemical reactions, and there is a larger R_g component due to the larger particle sizes associated with these chemicals. Thus, gravitational processes are relatively more important for particles containing calcium and the large-particle fraction of ammonium and sulfate, and the dryfall collector is believed to provide useful data for these species. In figures 29-31, Argonne (IL19) is the leftmost box plot distribution. The other five sites are plotted in roughly north-south order from left to right. Figure 29 shows a slight tendency for calcium to be higher in the southern part of the state. Figure 30 shows that the highest and most variable values for ammonium dryfall occur at Argonne and in northern Illinois. Argonne shows a slightly higher dryfall loading (figure 31) for sulfate than the rest of the state, where a slight trend to higher values in southern Illinois is also noticeable.

Temporal Trends

Wet Deposition. Trends in the chemical composition of Illinois precipitation were assessed by first determining the annual concentration distribution of each ion at each site. Next, the year-to-year changes in concentrations were evaluated by applying a Spearman rank correlation test to percentile points in the distributions. Three points were chosen, the 25th, 50th, and 75th percentile values. A summary of the test results is shown in table 3. None of the trend estimates were identically zero. A zero trend means that there is no overall direction in the year-to-year concentrations, either up or down. As a result, the changes were labeled as either "up" or "down" in this table.
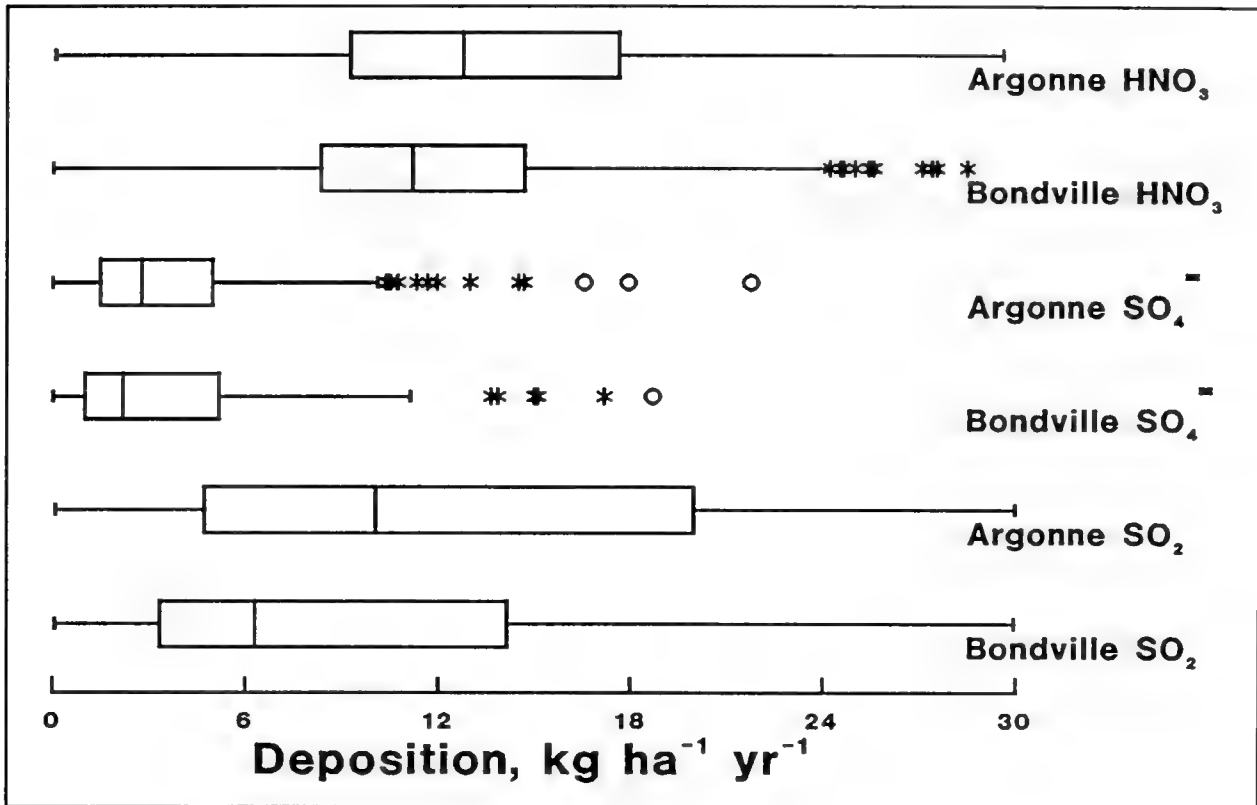
Figure 16. Comparison of the distributions of weekly estimates of annualized dry deposition (kg ha^{-1} yr^{-1}) at two Illinois sites (Argonne, IL19, and Bondville, IL11) over the period 1985-1992.

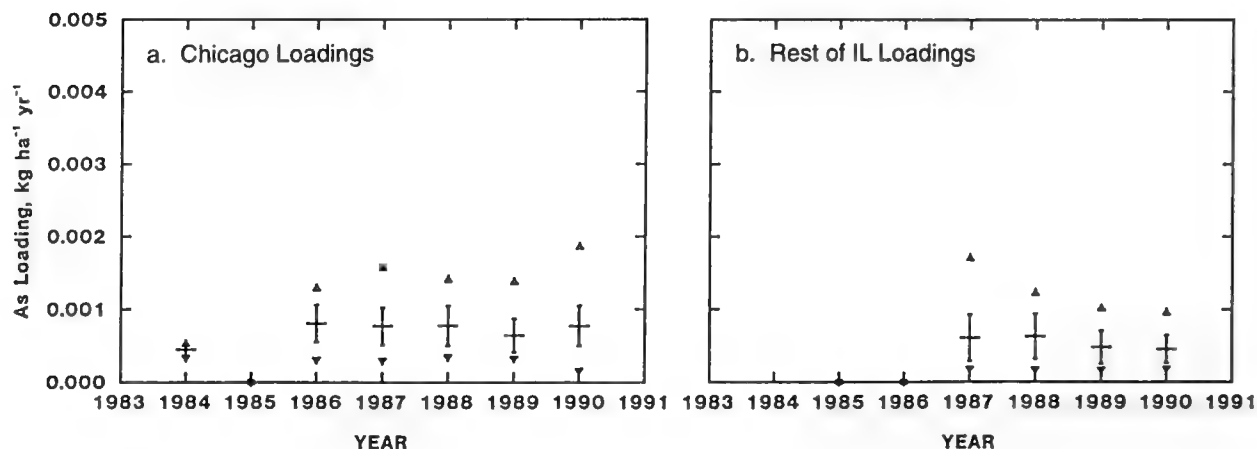Figure 17. Dry deposition loadings of arsenic (kg ha⁻¹ yr⁻¹) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.
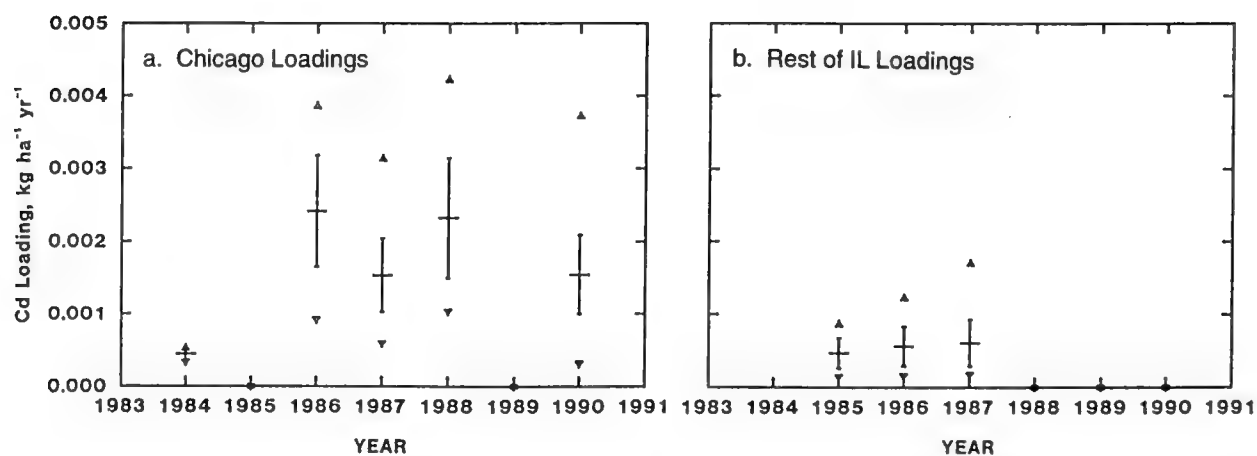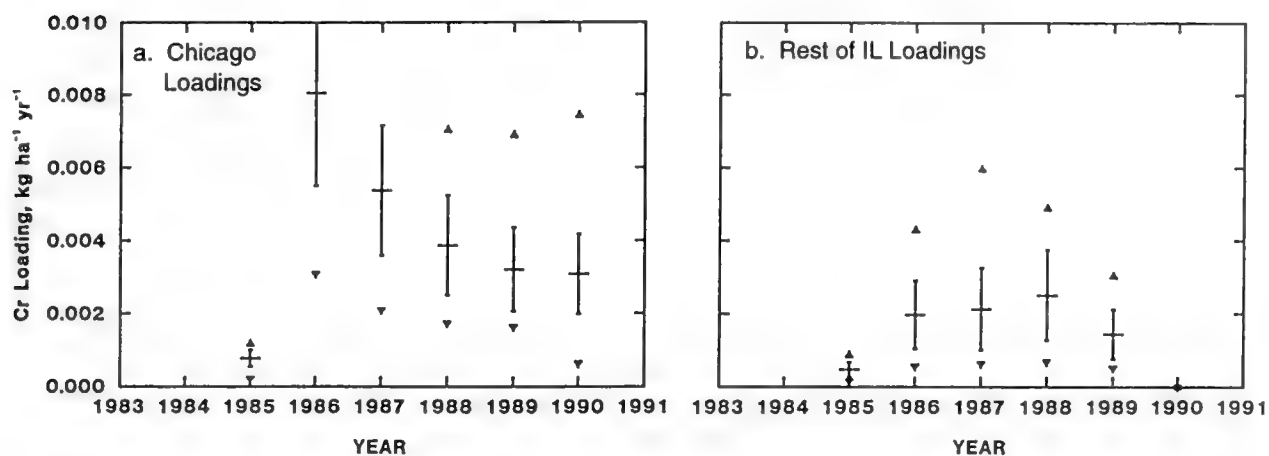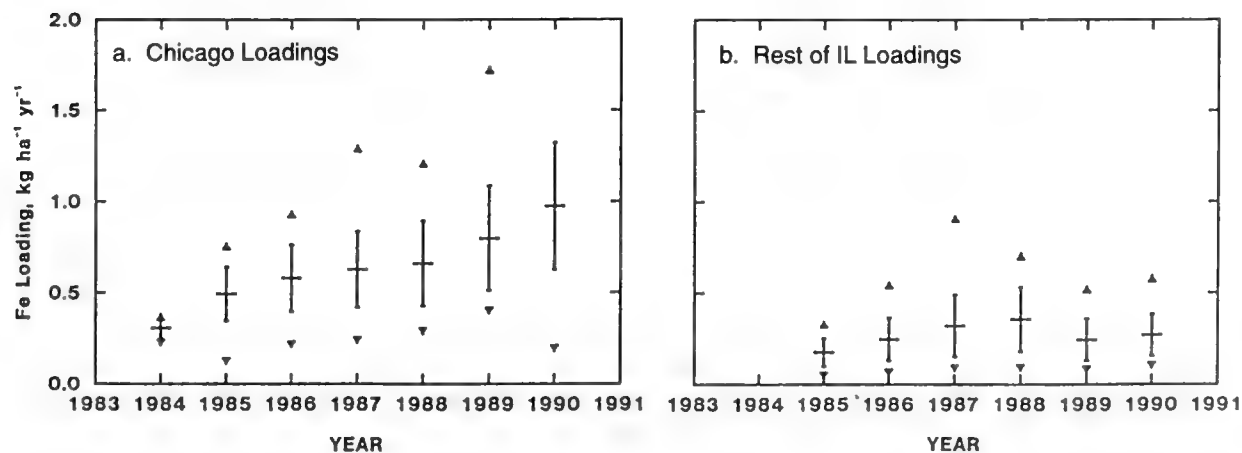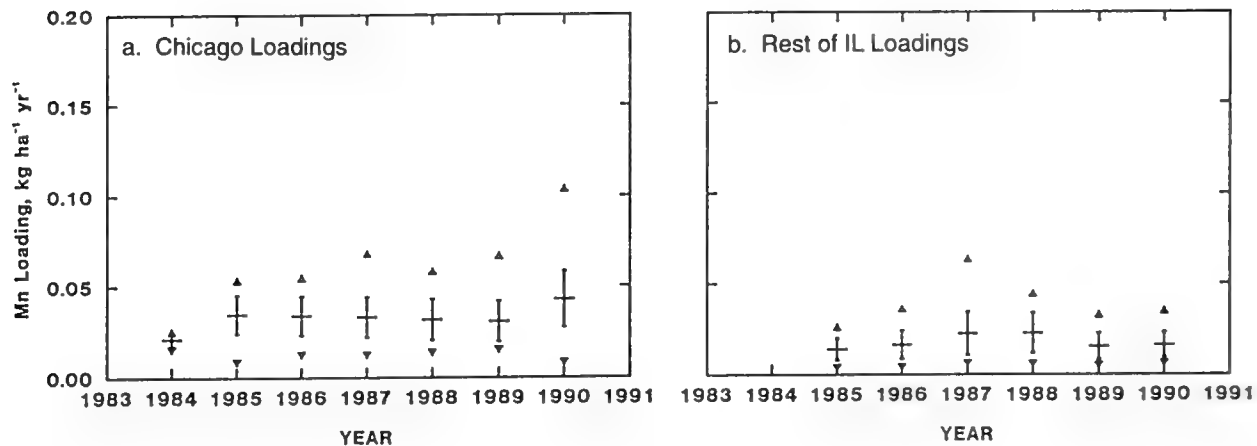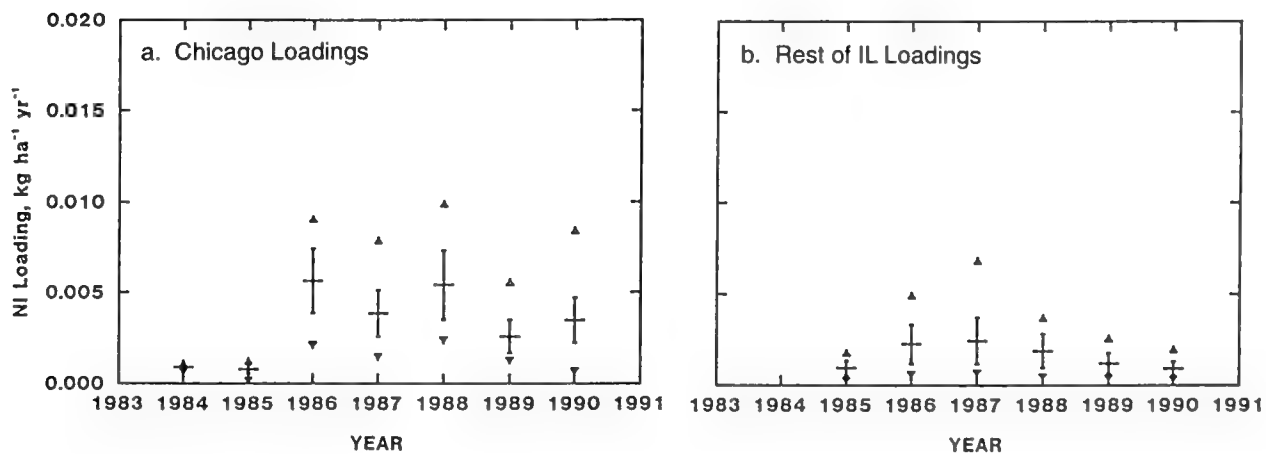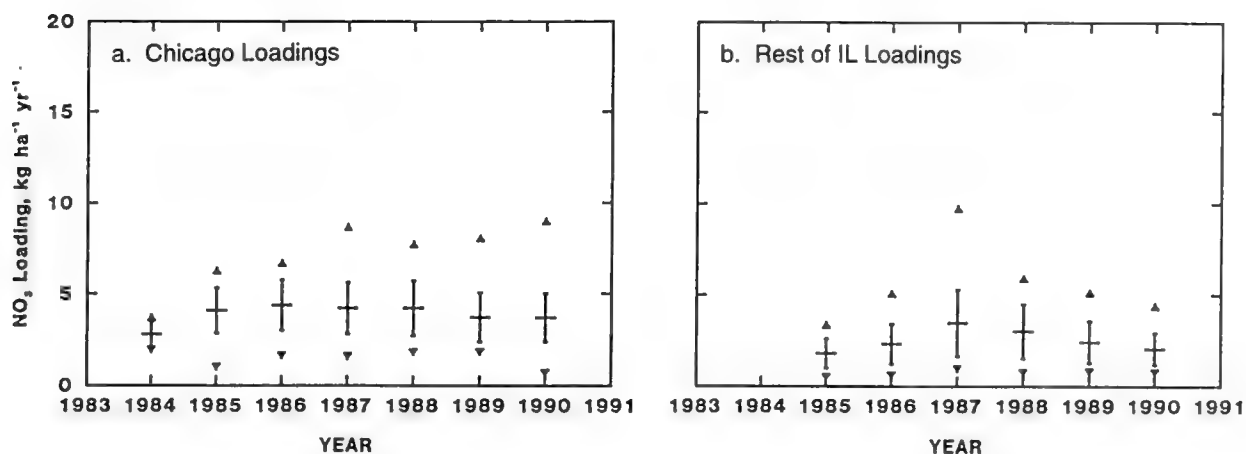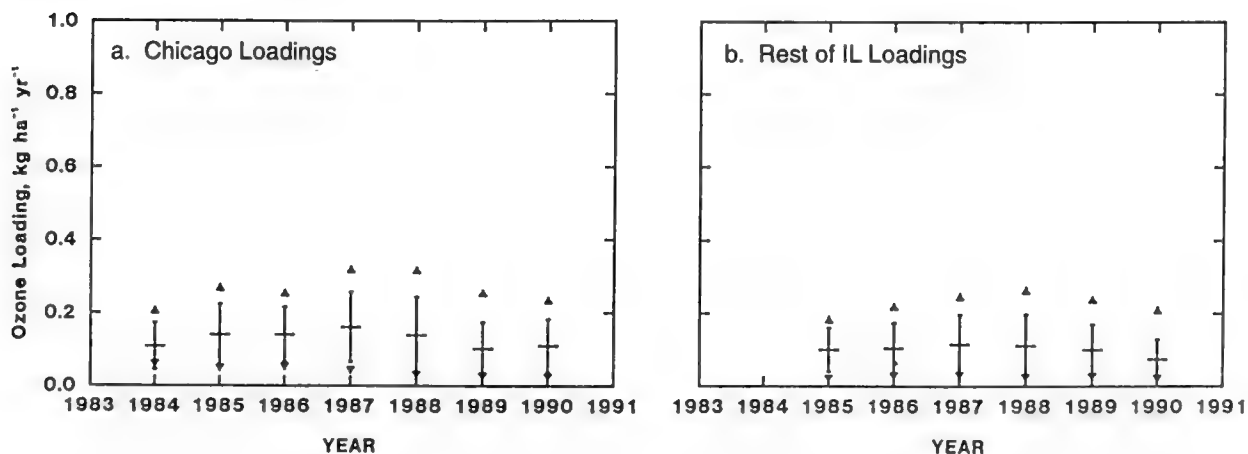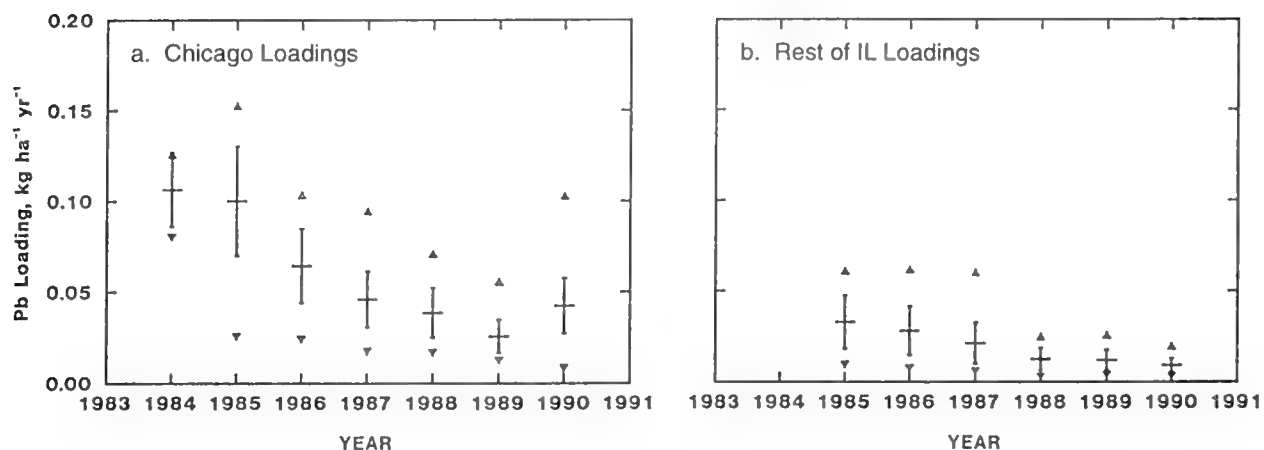

Figure 18. Dry deposition loadings of cadmium (kg ha⁻¹ yr⁻¹) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.

Figure 19. Dry deposition loadings of chromium (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.
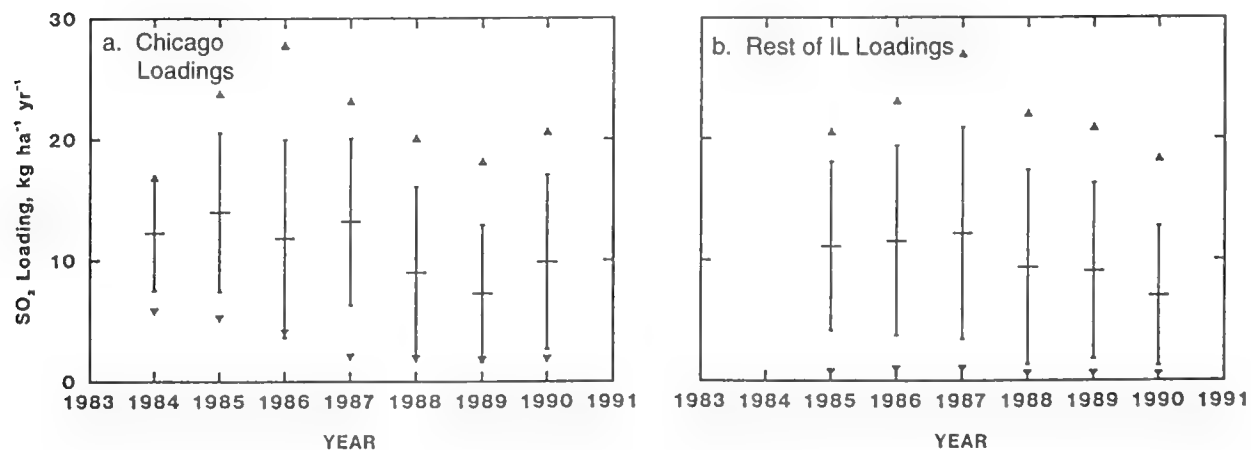

Figure 20. Dry deposition loadings of iron (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.

Figure 21. Dry deposition loadings of manganese (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V$_d$ statistics.
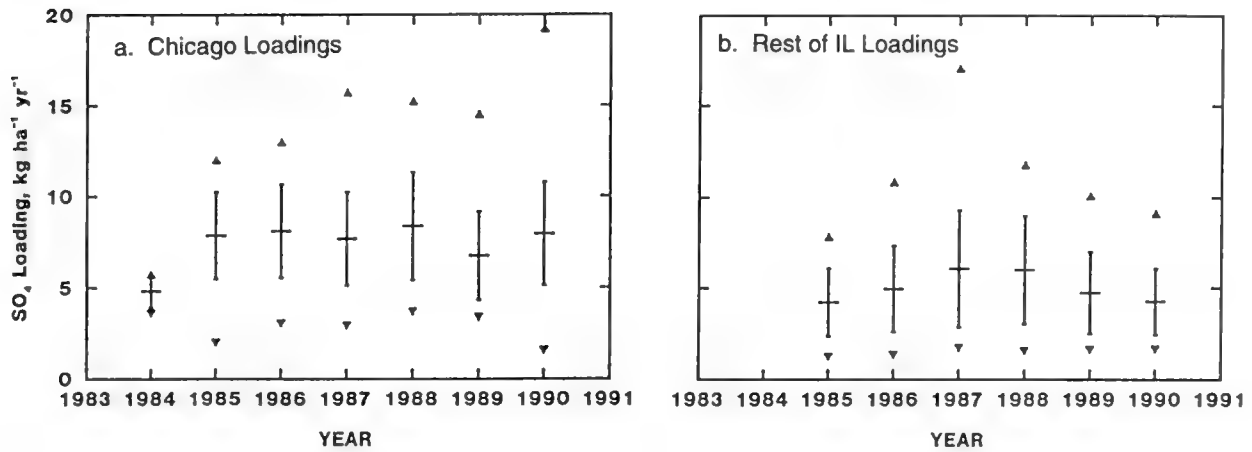

Figure 22. Dry deposition loadings of nickel (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V$_d$ statistics.
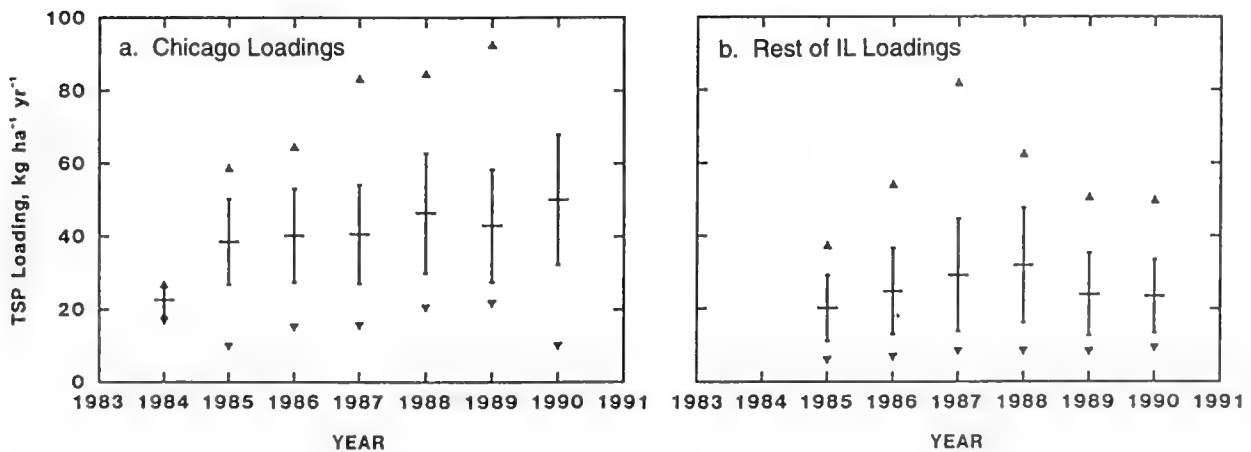
Figure 23. Dry deposition loadings of nitrate (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.

Figure 24. Dry deposition loadings of ozone (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.

Figure 25. Dry deposition loadings of lead (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V$_d$ statistics.

Figure 26. Dry deposition loadings of sulfur dioxide (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V$_d$ statistics.

Figure 27. Dry deposition loadings of sulfate (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.

Figure 28. Dry deposition loadings of TSP or mass of aerosol particles (kg ha^{-1} yr^{-1}) for the Chicago area (a) and the remainder of the State of Illinois (b), from ILEPA annual median concentration and NOAA/ATDD V_d statistics.
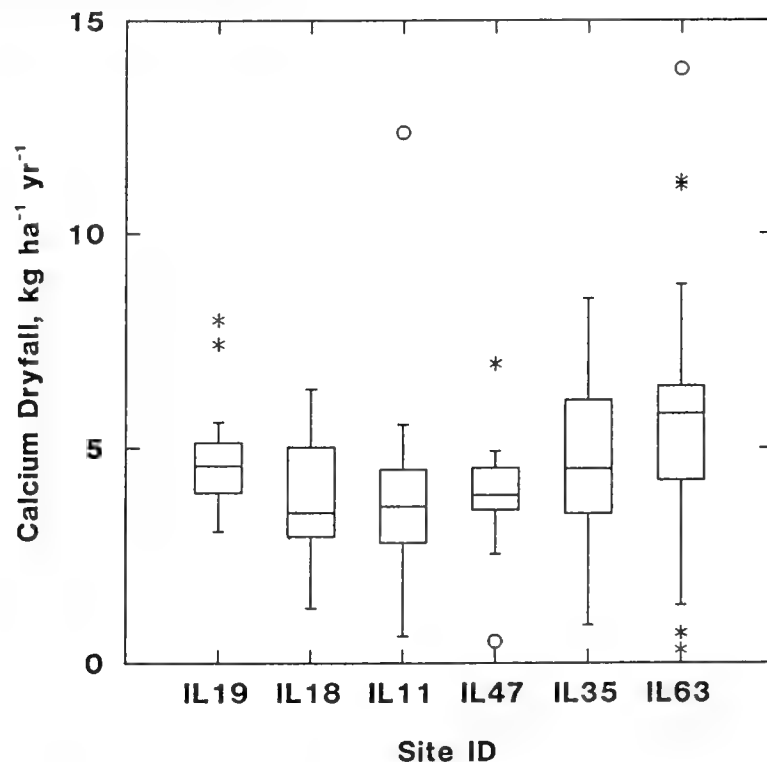
Figure 29. Box plots of the dryfall loadings for calcium (kg ha^{-1} yr^{-1}) measured at six NADP/NTN sites in Illinois (see table 1 for precise sampling intervals included, which vary slightly for each site).
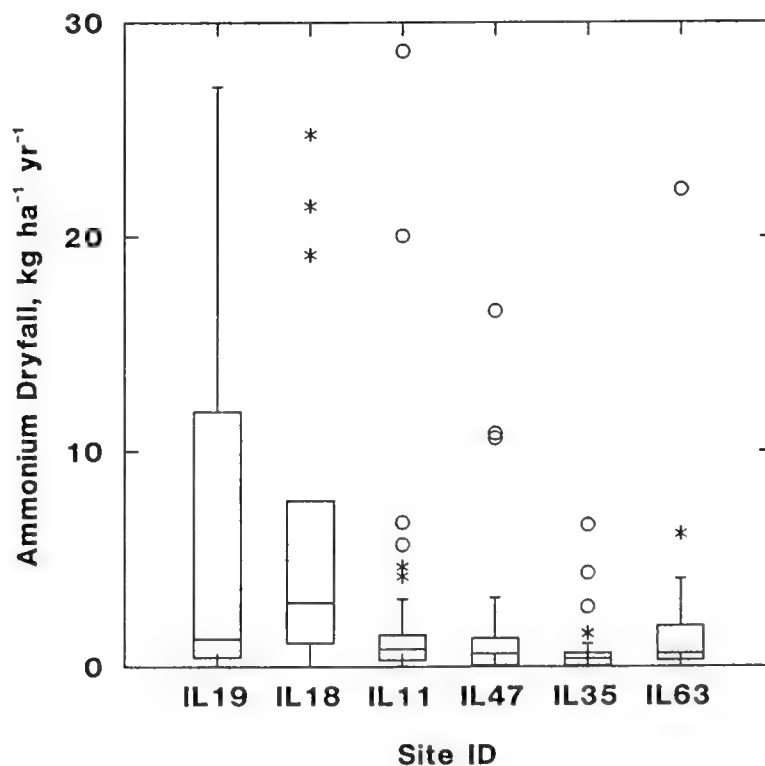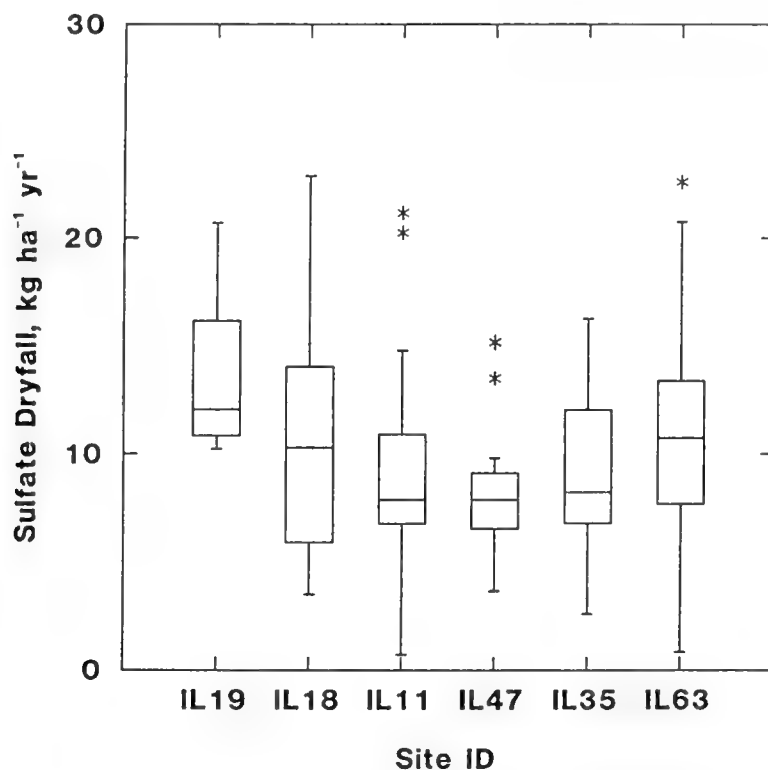

Figure 30. Box plots of the dryfall loadings for large-particle ammonium (kg ha^{-1} yr^{-1}) measured at six NADP/NTN sites in Illinois (see table 1 for precise sampling intervals included, which vary slightly for each site).

Figure 31. Box plots of the dryfall loadings for large-particle sulfate (kg ha^{-1} yr^{-1}) measured at six NADP/NTN sites in Illinois (see table 1 for precise sampling intervals included, which vary slightly for each site).

Table 3. Changes in Concentrations of Major Inorganic Ions in Illinois Precipitation

Ions	Percentile	Number of sites		Number with significant change*	
		Up	*Down*	*Up*	*Down*
	25	1	6	0	3
Ca$^+$	50	0	**7**	0	2
	75	1	6	0	2
	25	3	4	0	1
NH$_4^+$	50	3	4	0	1
	75	3	4	0	0
	25	2	5	0	0
NO$_3^-$	50	2	5	0	1
	75	1	6	0	0
	25	0	7	0	3
SO$_4^=$	50	0	7	0	0
	75	1	6	0	1
	25	7	0	0	0
pH	50	6	1	0	0
	75	5	2	0	0

Note:
*Using a Spearman rank correlation test, reject H$_o$ (5 percent level): no change in concentration.

For Ca^{++}, results show that the concentrations decreased at nearly all sites and percentiles. Of the 21 tests (3 percentiles at 7 locations), only 7 were statistically significant at the 5 percent level for NO_3^- and NH_4^+: the evidence is weak for a net change in either direction, however. These two pollutants were relatively unchanged in precipitation during the last decade. For $SO_4^=$, the weight of evidence once again shows a downward trend in concentration, although only 4 of 21 tests were significant (at the 5 percent level). And finally, for pH the test results indicate an increase over time. Although none of these tests was significant, the conclusion that pH has risen is consistent, chemically, with the drop in $SO_4^=$. Stoichiometrically, the decrease in sulfate was larger than the decrease in calcium, accounting for a net decrease in acidity or a rise in pH.

Seasonal variation sin V_d, concentration, and deposition at Bondville are shown in figures 32-34. These figures show a LOWESS-fit curve (Cleveland, 1979) through all weekly mean data from the NOAA/ATDD site collected in the period 1985-1992. The results illustrate the impact of the cyclic variation in the environment that affects the dry deposition process. In figure 32, the enhancement of the V_d of ozone and SO_2 in the summer months is obvious, and is largely due to a reduction in R_c (see figure 9) as biologically active surfaces, i.e., vegetation, and liquid water films proliferate during the growing season. R_c increases, and V_d is thus reduced for these surface-reactive pollutants during the winter season. Fine-particle V_d also shows a tendency for higher warm-season values, but there is a much smaller increase. V_d values for fine particles and HNO_3 respond to 1) a decrease in R_b, which occurs with the increase in reactive area and roughness of the surface as the vegetation canopy expands, and 2) a decrease in R_a, which occurs due to and produces increased atmospheric turbulence and instability produced by summer convection. Figure 33 shows the concentration data for fine particulate NO_3^- and $SO_4^=$, and gaseous SO_2 and HNO_3, measured at Bondville, on a corresponding annual cycle. A strong seasonal cycle appears only for $SO_4^=$. Multiplying the curves of figures 32 and 33 together produces the deposition loading curves of figure 34. When *both* concentration and V_d have seasonal cycles that are in phase, or peak simultaneously, then the deposition curve has similar but intensified cycle. This is the case for $SO_4^=$, where the fine-particle V_d and the $SO_4^=$ concentration tend to increase slightly in the warm season; the $SO_4^=$ deposition curve then peaks relatively steeply at the same time period. For SO_2, only the V_d term has a definite cyclic seasonal trend, but it is nonetheless reflected in the deposition cycle because of its magnitude. Finally,

in the case of HNO_3, the V_d and concentration curves are not distinct, but appear to be out of phase, with V_d higher in the summer, and concentration higher in the winter. The resulting deposition curve reveals much variation, but no distinct seasonality. It is important to note from figures 32-34 that simple monitoring of airborne concentration does not necessarily reveal a true picture of the delivery of pollutants to receptors at the surface.

The time series of annual distributions of dry deposition loading for several pollutant species, calculated for the Argonne and Bondville NOAA/ATDD sites, is plotted in figures 35 and 36. The box plot for each year indicated shows the distribution of weekly loading estimates, expressed as an annual total in $kg\ ha^{-1}\ yr^{-1}$. There are no apparent or statistically significant *long-term trends* for SO_2, $SO_4^=$, HNO_3, or NO_3^- loadings at either site during the 1984-1992 period. However, significant *interannual variation* exists for dry deposition of each pollutant. These variations tend to follow air quality variations, but not exactly, due to the independent variations in V_d. While slight trends toward decreases in wet deposition have been noted above, and may be attributed to lower emissions of SO_2 and $SO_4^=$ over the 1980s, such a trend is not apparent in the dry deposition data. Figures 12-23 represent additional time-series plots of dry deposition loadings, based on IEPA air quality data. Annual median concentrations for the Chicago area and the "Remainder of State" area were used in combination with the NOAA/ATDD V_d results to produce the annual deposition estimates. Trends over time in the IEPA air quality data are thus reflected in these figures and are described in detail elsewhere in this report.

Total Deposition

The wet and dry deposition data compiled throughout this section on atmospheric deposition were combined, where possible, to arrive at estimates of "total atmospheric deposition," (figures 37-39). The resultant total deposition estimates should at best be considered very tentative. The uncertainties in each of the values combined are relatively large, on the order of tens of percent. These uncertainties are compounded when summed into the total deposition estimate. However, these results are a valid statewide estimate, based on real monitoring data, of the magnitude of inputs of several chemicals to the earth's surface. The following data were used to plot the results of total atmospheric deposition for calcium (NADP/NTN wet Ca^{++} + NADP/NTN dryfall Ca^{++}), total nitrogen (NADP/NTN dryfall NH_4^+ + NOAA/ATDD HNO_3 + IEPA
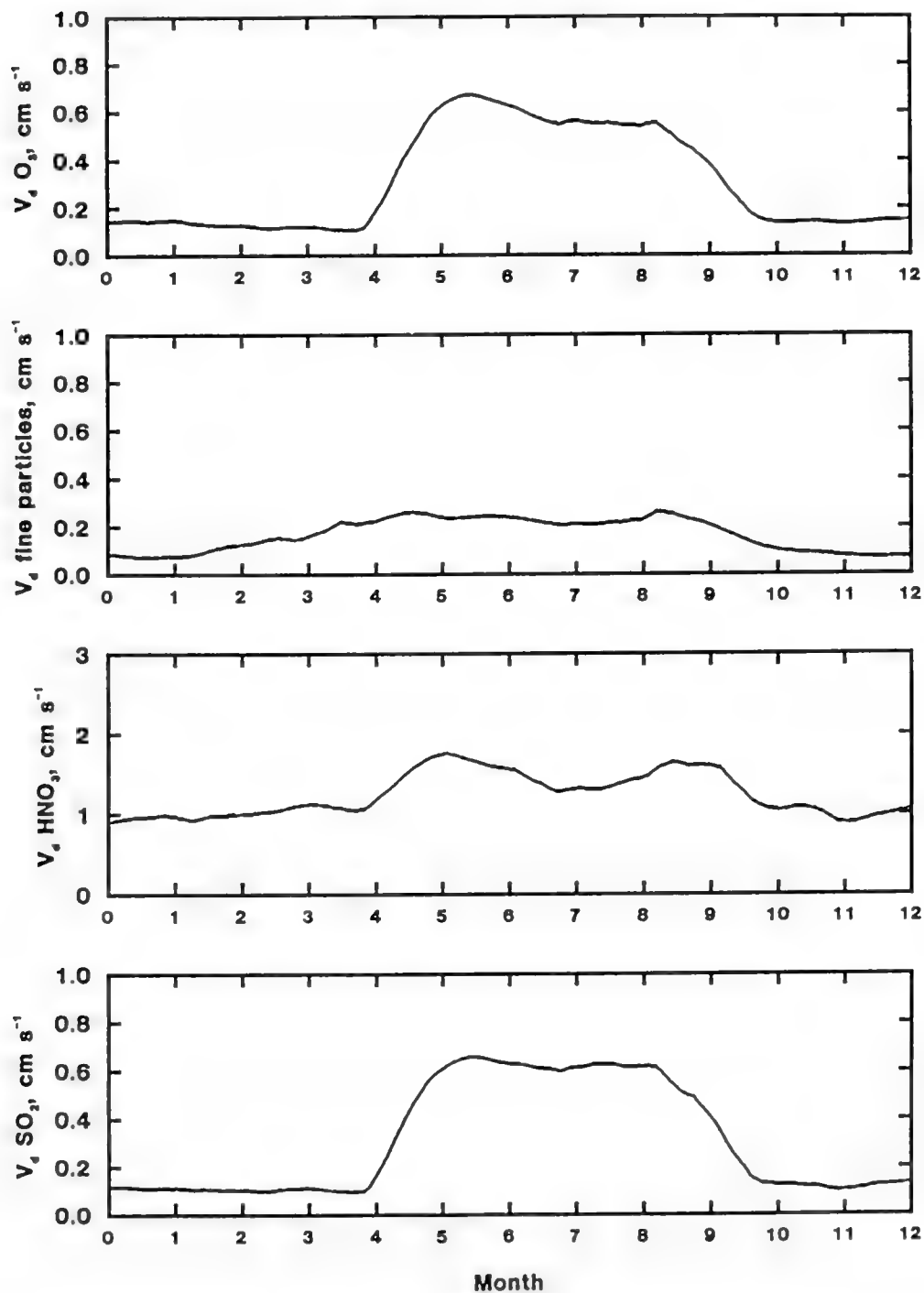
Bondville, Illinois: Dry Deposition Velocity

Figure 32. Seasonal variations of weekly mean dry deposition velocity, V_d (cm s^{-1}) for several airborne pollutants calculated from observations at Bondville, Illinois, for the period 1985-1992.
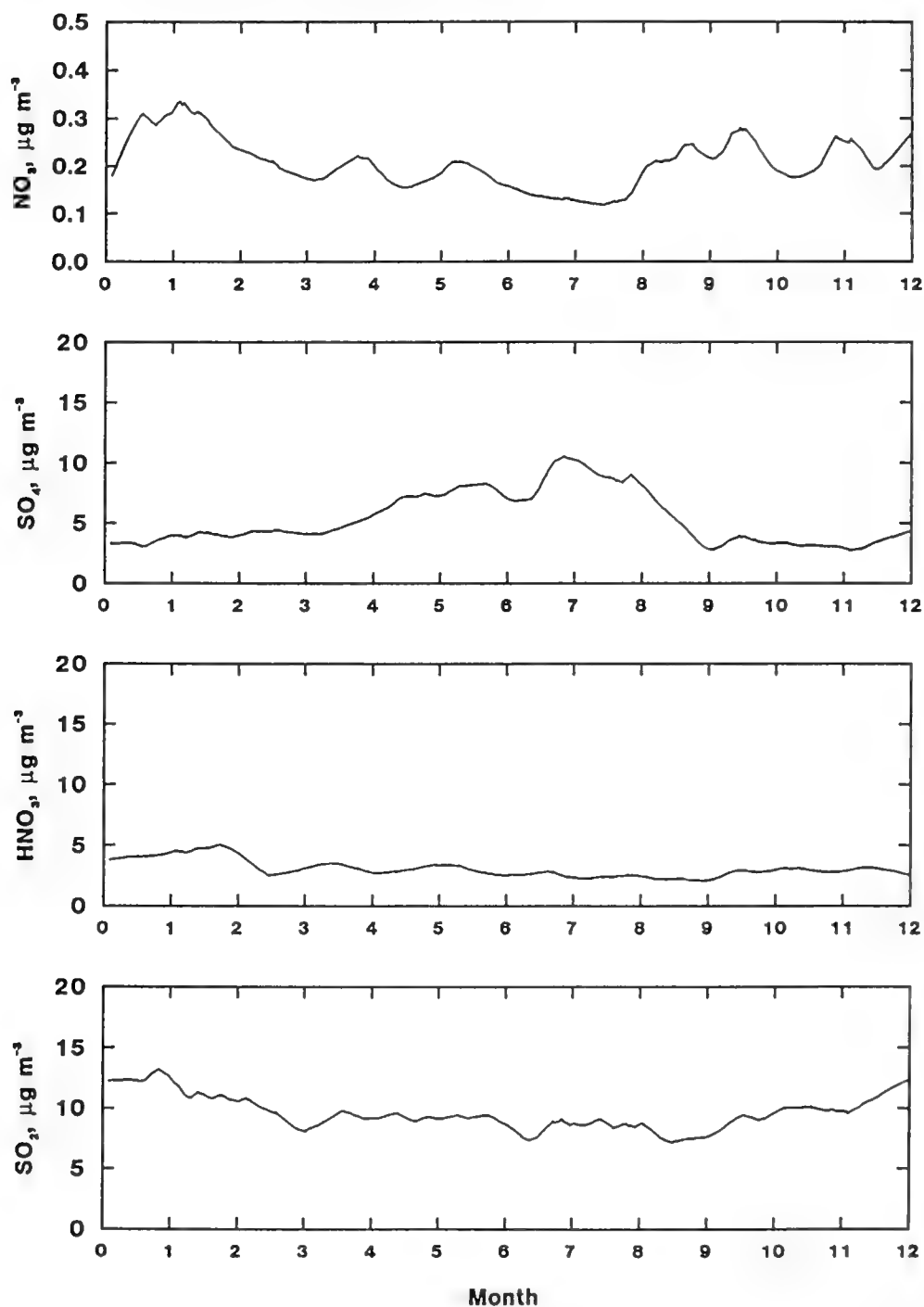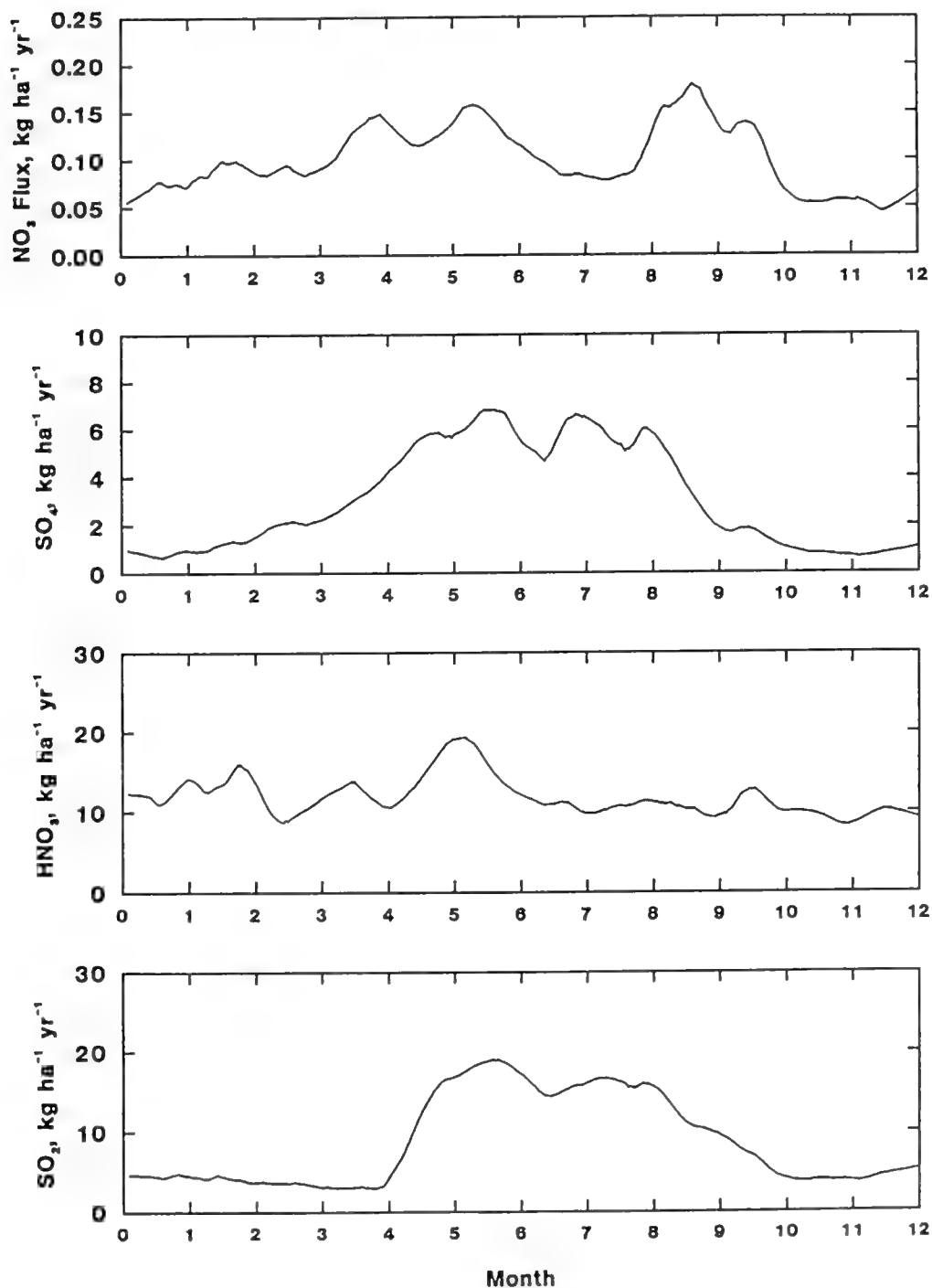
Bondville, Illinois: Concentrations

Figure 33. Seasonal variations of weekly mean airborne concentration (μg m⁻³) for several airborne pollutants calculated from observations at Bondville, Illinois, for the period 1985-1992.

Bondville, Illinois: Dry Deposition

Figure 34. Seasonal variations in annualized weekly mean dry deposition loading (kg ha⁻¹ yr⁻¹) for several airborne pollutants at Bondville, Illinois, for the period 1985-1992.

Argonne, Illinois: Dry Loadings
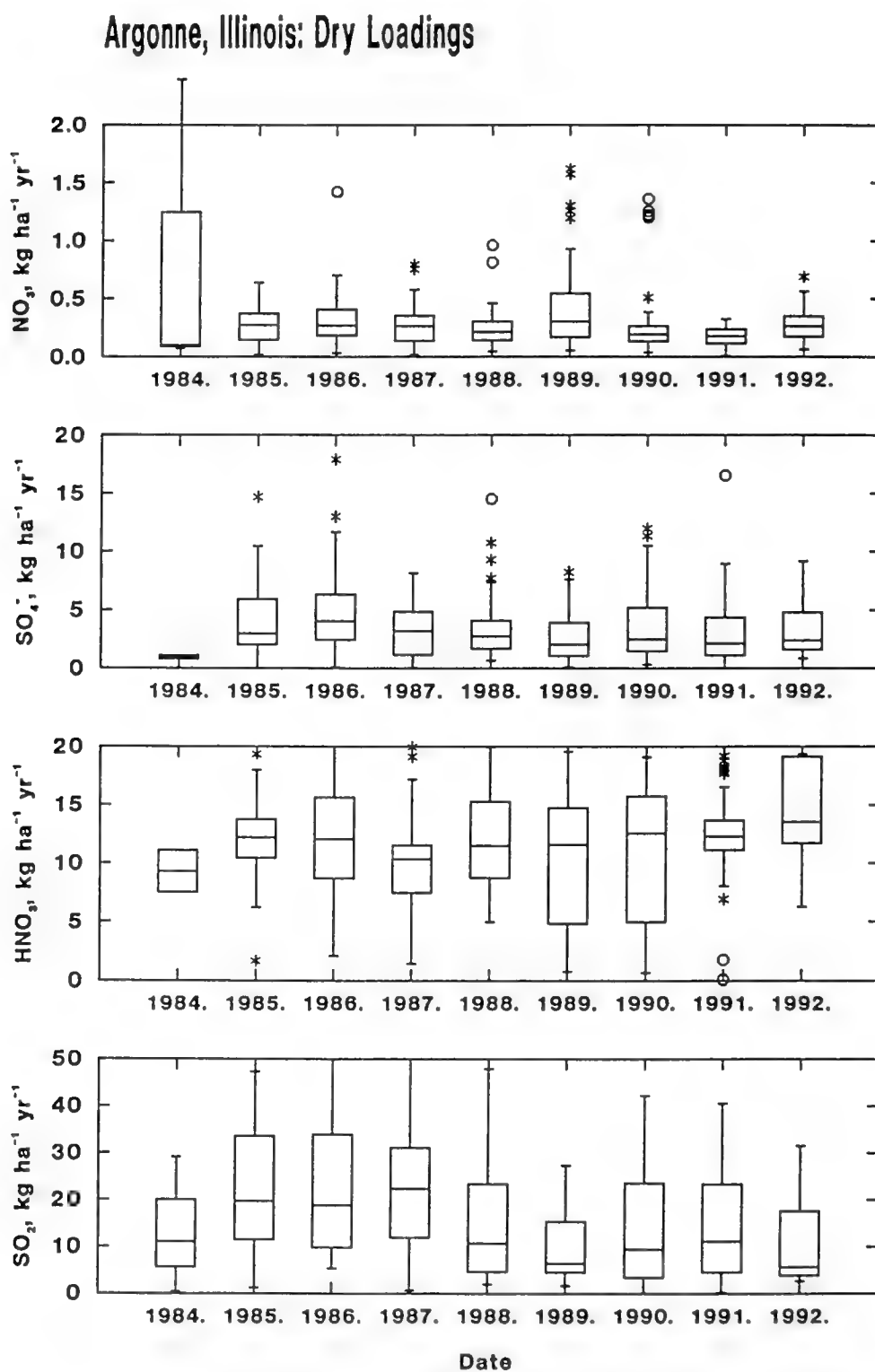
Figure 35. Annual distributions of dry deposition loading ($kg\ ha^{-1}\ yr^{-1}$) for several pollutant species, calculated for the Argonne, Illinois NOAA/ATDD site.
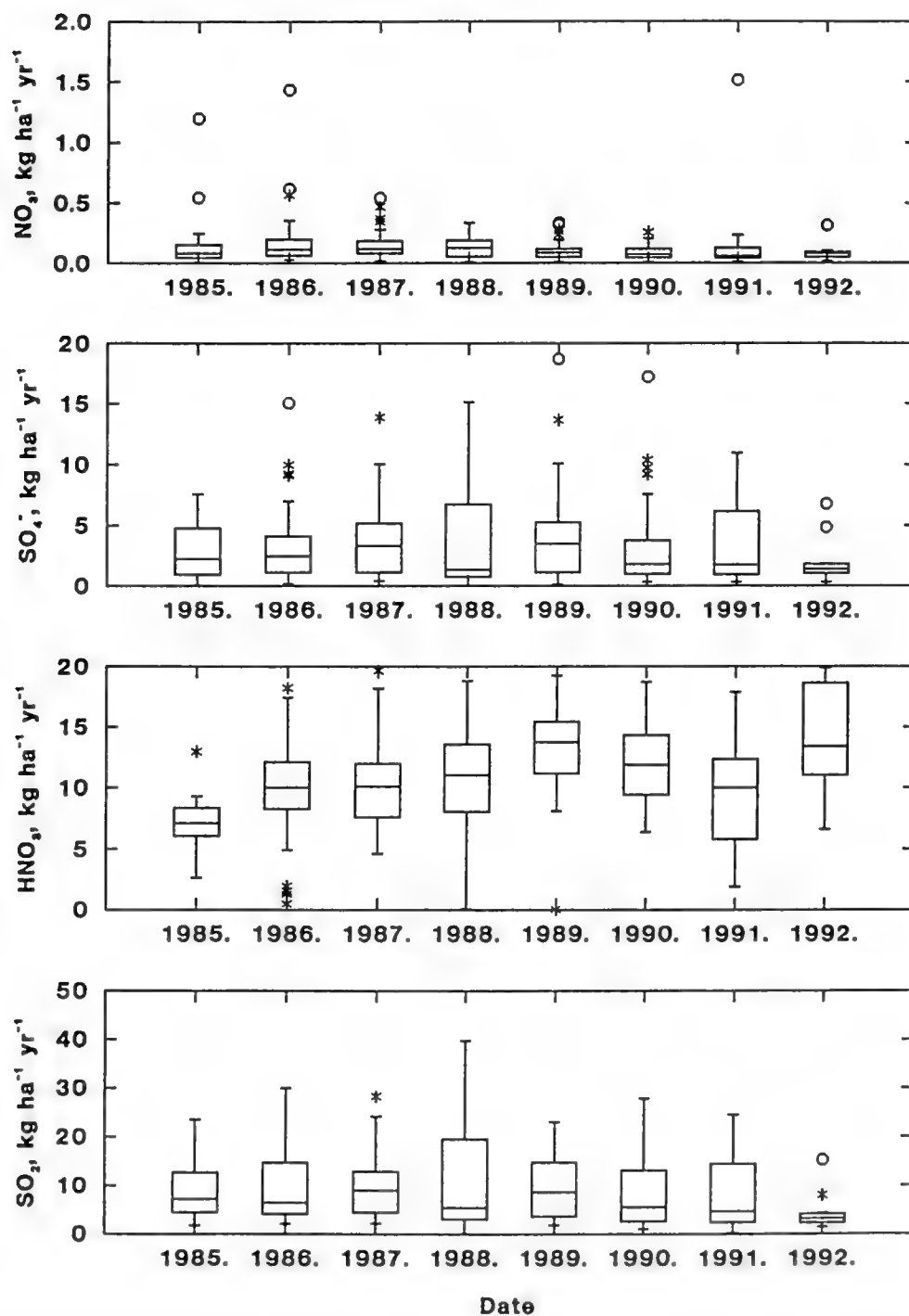
Bondville, Illinois: Dry Loadings

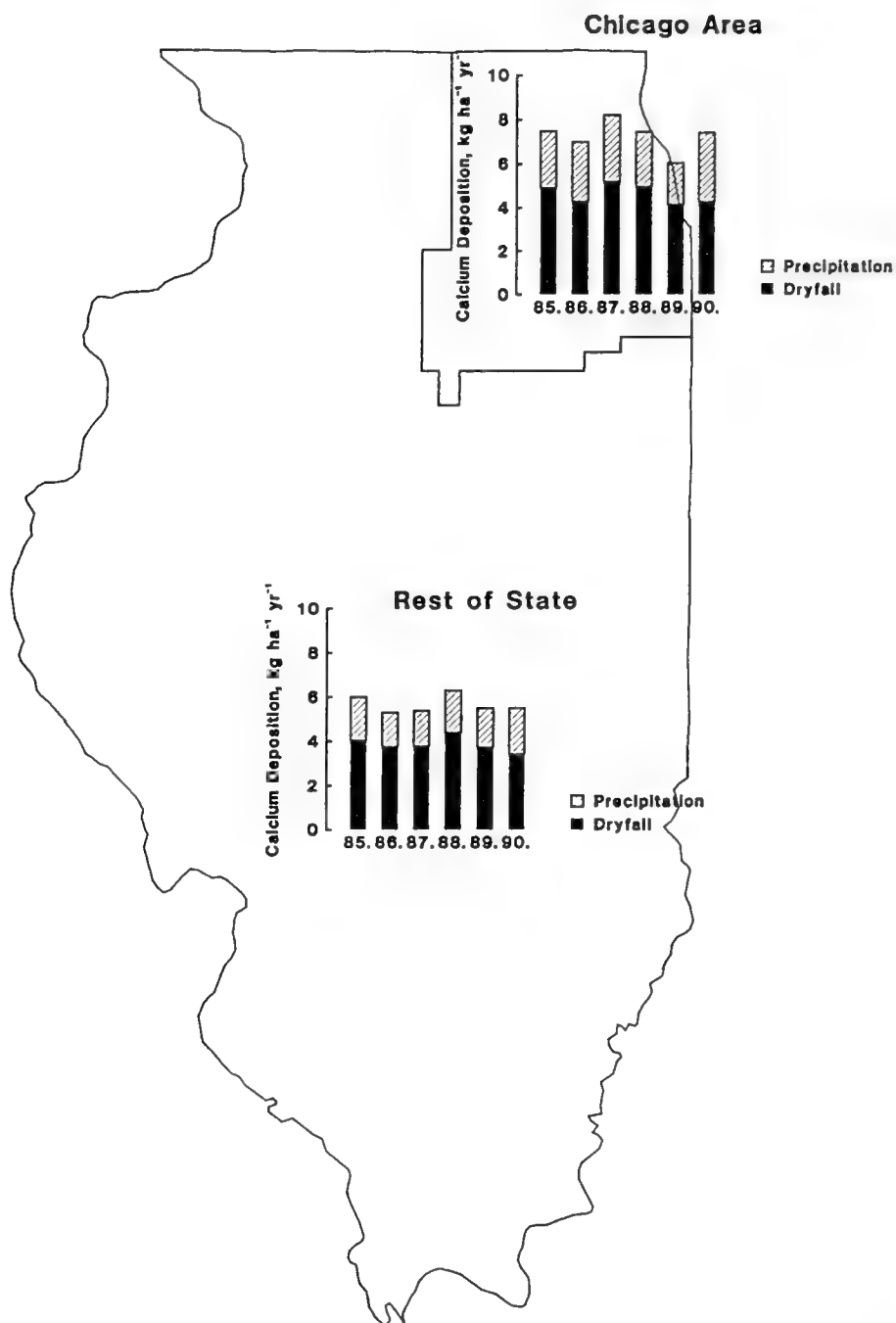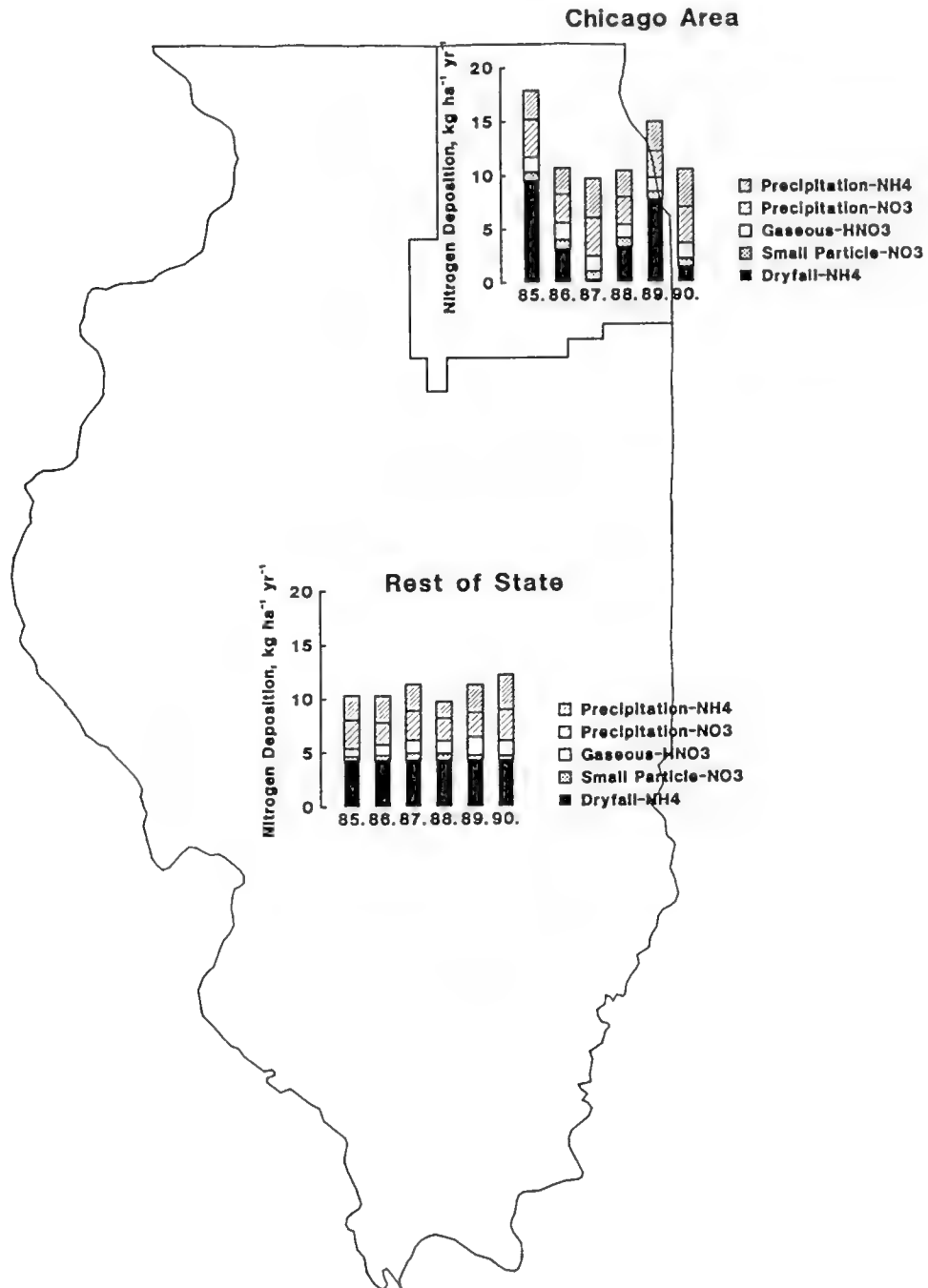Figure 36. Annual distributions of dry deposition loading (kg ha^{-1} yr^{-1}) for several pollutant species, calculated for the Bondville, Illinois NOAA/ATDD site.
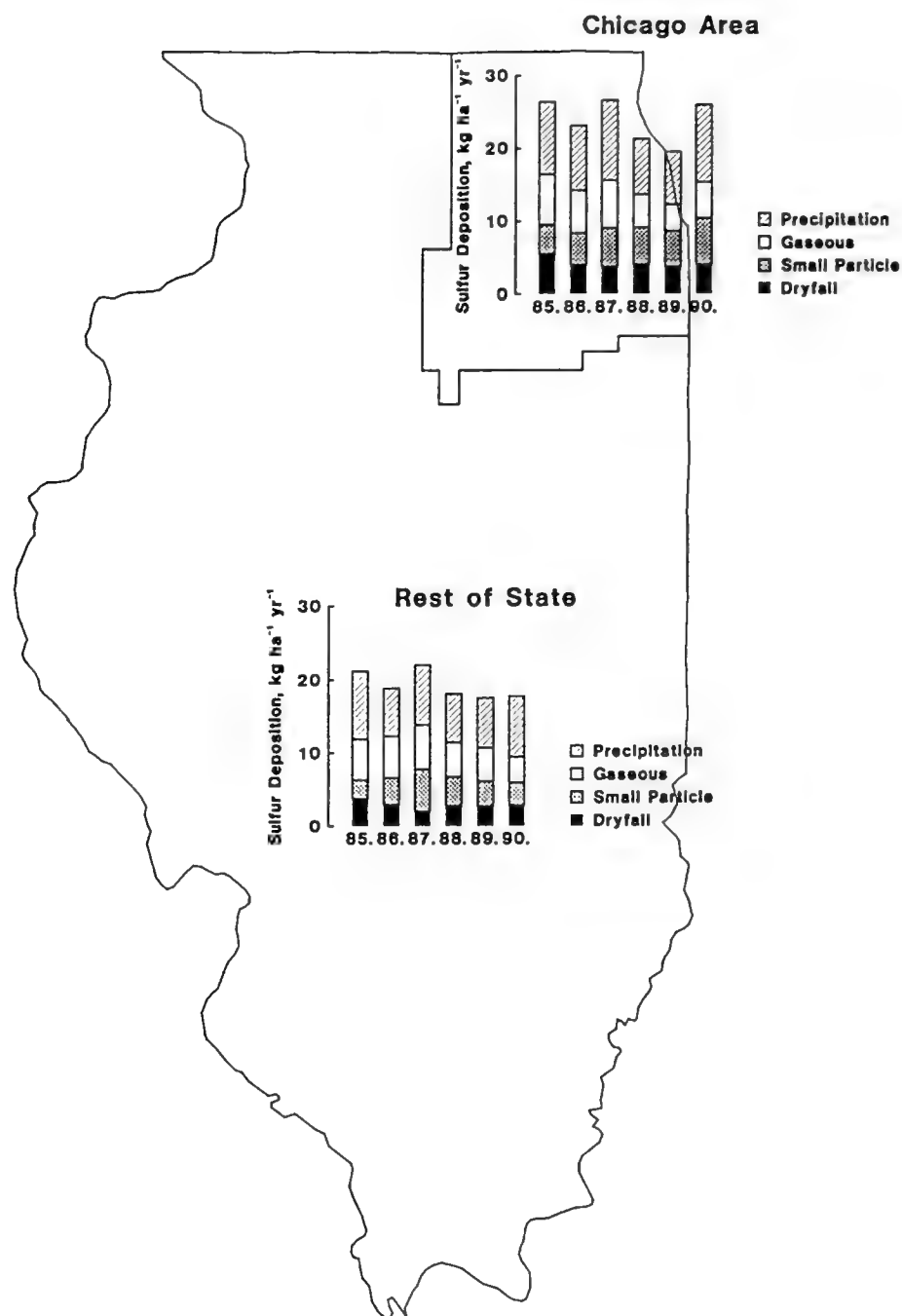
Total Atmospheric Deposition – Calcium

Figure 37. Annual estimates of total atmospheric deposition (kg ha^{-1} yr^{-1}) for calcium for the Chicago area and the remainder of the State of Illinois.

Total Atmospheric Deposition – Nitrogen

Figure 38. Annual estimates of total atmospheric deposition (kg ha⁻¹ yr⁻¹) for nitrogen for the Chicago area and the remainder of the State of Illinois.

Total Atmospheric Deposition - Sulfur

Figure 39. Annual estimates of total atmospheric deposition (kg ha⁻¹ yr⁻¹) for sulfur for the Chicago area and the remainder of the State of Illinois.

NO_3^- + NADP/NTN wet NO_3^- + NADP/NTN wet NH_4^+), and total sulfur (NADP/NTN dryfall $SO_4^=$ + IEPA $SO_4^=$ + IEPA SO_2 + NADP wet $SO_4^=$) in figures 37-39, respectively.

SUMMARY AND CONCLUSIONS

Deposition Trends and Spatial Variation

Wet deposition in Illinois has been monitored for ten years or more at eight NADP/NTN sites. The NADP/NTN reports the concentrations of ten separate chemical pollutants in precipitation, of which just five account for about 90 percent of the chemical composition that causes Illinois precipitation to be acidic. The pollutants in order of importance are: sulfate ($SO_4^=$) > hydrogen ion (H^+) > nitrate (NO_3^-) > ammonium (NH_4^+) > calcium (Ca^{++}). Illinois precipitation is most simply described as a dilute solution of mineral sulfuric and nitric acids, partly neutralized by ammonium and calcium.

Based on statistical tests of time- series data alone, there is no unambiguous trend that applies to all of the important pollutants causing acid rain in Illinois. Based on a "weight of the evidence" analysis, however, several points can be made about Illinois precipitation chemistry changes during the 1980s:

1. Sulfate decreased 2 to 4 percent per year in the southern third of the state, with smaller decreases elsewhere.
2. Calcium decreased by 3 to 7 percent per year, except at Argonne (suburban Chicago), where it remained steady.
3. Nitrogen species, ammonium and nitrate, remained unchanged.
4. pH increased slightly, but the increase is too small and too variable to be quantified.
5. Sulfur dioxide and NO_x emissions decreased slightly.

Dry deposition in Illinois tends to be somewhat higher in the Chicago area, due to both higher airborne concentrations of most pollutants and higher deposition velocities. For sulfate, nitrate, sulfur dioxide, ozone, arsenic, and manganese, the differences are on the order of 10 to 30 percent. For cadmium, chromium, iron, nickel, lead, and TSP, dry deposition in the Chicago area exceeds the remainder of the state by 200 to 400 percent; this is caused primarily by the differences in air quality (see *Air Quality Trends in Illinois* chapter). Temporal trends in dry deposition generally follow air quality trends, although additional variability

is introduced into the time-series data by interannual variation in deposition velocities. More important for ecological impacts is the seasonal nature of dry deposition loadings, with higher deposition velocities for many pollutants occurring during the warm season, when biological impacts may also be the greatest.

The total deposition of sulfur in the Chicago area is about 15 percent higher than in the rest of the state. For sulfur (sulfate *plus* sulfur dioxide), the ratio of wet to dry deposition is about 1 part wet to 1.5 parts dry. The deposition of nitrogen in the Chicago area is about 30 percent higher than in the rest of the state. For nitrogen (nitrate *plus* ammonium *plus* nitric acid vapor), the ratio of wet to dry deposition is about one part wet to three parts dry.

Deposition to Specific Receptor Systems

The spatial and temporal variation information is most useful in describing the coupling of the atmosphere to receptors that are also distributed in space and whose sensitivity varies temporally with the depositing pollutants. Acid deposition to forests for example, is most likely to have an effect during the growing season, and is much less likely to be harmful in the dormant season. Toxic deposition to forests, however, may act through a *cumulative* effect, in which the temporal variation is less important to understanding the impact on the receptor system.

Acid deposition to Lake Michigan presents a special difficulty in this analysis, since neither wet nor dry deposition is measured over the lake. The refinement of estimates based on shoreline measurements is an ongoing research topic, however.

Other Aspects of Atmospheric Deposition

Agricultural systems have been shown to be relatively insensitive to current levels of wet acid deposition, but the impact of toxic deposition and dry deposition of many pollutants is unknown. Ozone has been shown to have negative impacts on yield and quality of cash crops in several areas of the United States. The role of wet and dry toxics deposition as a contributor to nonpoint source pollution in surface and ground-water supplies for human consumption is also unknown at this time. The impact of atmospheric deposition (acid rain, toxic pollutants, and biological nutrients) to lakes and streams in Illinois (i.e., non-Lake Michigan waters) has not been documented, however, consideration of the magnitude of deposition for many chemicals

would indicate that significant impacts are possible. Finally, the impacts of $SO_4^=$, SO_2, acids, and NO_3^- deposition on exterior building materials, paints, etc., both in precipitation and dry deposition, have been demonstrated in recent federally sponsored research in many areas of the United States. Materials impacts in Illinois are as yet unquantified, but are potentially large.

Enhancement of Atmospheric Deposition Knowledge

Further analyses of the data presented here and additional existing data, which were not accessible in the context of this project, could refine the temporal and spatial resolution of a combined atmospheric deposition database. Recommendations for further work include acquisition of additional extant air quality and precipitation chemistry data, and the development of the NOAA/ATDD model to a high-resolution model specifically for Illinois.

ACKNOWLEDGMENTS

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15. Supplementary Notes

16. Abstract (Limit: 200 words)

Air quality measurements made by the Illinois Environmental Protection Agency for 1978-1990 were analyzed for time trends and spatial variations over Illinois. The Spearman Rank Correlation Coefficient was used to evaluate the statistical significance of trends in median concentrations. Spatial distributions were plotted for six criteria pollutants, and sulfate, nitrate, and several metals in the Chicago area for 1980, 1985, and 1990. Downward trends significant at 1% were noted in annual mean lead concentrations in all areas of Illinois. Particulate matter showed no significant trends statewide or in any geographical regions. Ozone concentrations were decreasing after removal of temperature effects. Annual mean sulfate had a downward trend statewide (significant at 5%), in the Chicago area, and in areas away from major urban areas (2%). No significant trends for nitrate were detected regionally or statewide. Downward trends were significant at 1% for arsenic, the only other metal with a significant trend statewide or in the Chicago area. No significant trends were detected for arsenic or cadmium in the Metro East area where both manganese and iron concentrations were increasing. Spatial concentration patterns of most pollutants were highly variable in the Chicago area for 1980, 1985, and 1990. Contouring and comparisons were hampered by recent sharp declines in sampling site number and density.

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