Acidic Deposition
in New Jersey

A report to the Governor and Legislature of New Jersey by the Panel on Acidic Deposition in New Jersey Under the auspices of the Governor's Science Advisory Committee.

September 1985

Governor's Science Advisory Committee

ACIDIC DEPOSITION IN NEW JERSEY

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by

The Panel on Acidic Deposition in New Jersey

under the auspices of

The Governor's Science Advisory Committee

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PREFACE

Assembly Concurrent Resolution No. 134, passed by the New Jersey Assembly on December 16, 1982 and by the New Jersey Senate on June 20, 1983, petitioned "the Governor's Science Advisory Committee to assess and evaluate known environmental and economic effects of acid deposition upon water, wildlife, marine-life, vegetation, property, recreation, and agriculture in New Jersey and to consult with appropriate persons and groups in other affected states in order to ascertain the effects of acid deposition on a regional basis." It is also requested that the committee "within 18 months of the effective date of this concurrent resolution, report its findings and recommendations to the Governor and the Legislature, accompanying the same with whatever proposal for State, regional, or federal action it may deem appropriate."

While the Governor's Science Advisory Committee (GSAC) was formulating its mode of response to the Concurrent Resolution, the Committee learned that Governor Kean desired an interim report on the subject before the December 4-5, 1983 meeting of the Coalition of Northeastern Governors (CONEG) that Governor Kean, as 1983 Chairman, was to host at the Meadowlands Hilton Hotel. Governor Kean had placed "acid rain", as acidic deposition is popularly called, on the meeting agenda and was to deliver a speech on the effects of acid rain and on the need to adopt a regional abatement strategy.

GSAC thus joined with the Clean Air Council (CAC) to hold a public hearing on "The Effects of Acid Rain in New Jersey" on October 17, 1983 in the State Museum, Trenton. Dr. Donald F. Nelson of GSAC and Dr. Paul J. Lioy of CAC acted as cochairmen of the hearing. Experts in various aspects of the acidic deposition problem from New Jersey and surrounding states were invited to testify at the hearing. Testimony from the public was also received. A report by the same name as the hearing was written and forwarded to Governor Kean in mid-November, 1983 and made public at the CONEG meeting.

At the beginning of 1984 GSAC formed a panel of experts from within New Jersey to make an in-depth study of acidic deposition in New Jersey in response to the Concurrent Resolution. The panel began its work in February. The New Jersey Department of Environmental Protection offered staff support services to the panel through John C. Elston, Chief, Bureau of Air Quality Management & Surveillance. During 1984 the panel members have given much time in the spirit of public service in order to prepare this report. It has been peer-reviewed by nine experts in the diverse fields encompassed by acidic deposition, revised by the panel, and finally approved by GSAC.

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INTRODUCTION

Acidic deposition is the combination of acidic precipitation in the form of rain, snow, and fog and the dry deposition of acidic aerosols and gases from the atmosphere. The term "acid rain", used in the popular press, actually refers to only one component of the acidic deposition phenomenon. A number of harmful effects have been purported to result from acidic deposition, including the acidification of lakes and streams with a consequent alteration of their biota; decreased growth rates in some agricultural crops and trees; corrosion damage to buildings, bridges, monuments, and other exposed metallic and stone structures; increased dissolution of toxic metals in surface waters and, in a few cases, ground waters; and increased breathing difficulties caused by acidic aerosols for people suffering from lung disorders such as asthma and emphysema. However, the causal relationship between these problems and acidic deposition is still a subject of intense debate and research.

Acidic deposition is both an old and a new problem. Statements that "the air is full of acid and sulphureous particles" can be found in the literature as early as 1738. In the mid-nineteenth century, Smith in England gave a remarkably accurate and encompassing characterization of the effects of acidic deposition (Smith, 1852, 1872). In fact, it was Smith who first used the phrase "acid rain". In the century following Smith's seminal studies concern about acidic deposition grew slowly as did the number of studies devoted to it. The work of Oden (1968) in Sweden and the subsequent report to the United Nations Conference on the Human Environment (Bolin, 1971) did much to focus world-wide concern on the problem. Shortly after that Likens and his associates drew attention to the problem occurring in the eastern United States (Likens et al., 1972). Thus, as a problem having wide recognition, it is only somewhat over a decade old.

The past decade has seen an increasing number of studies of the acidic deposition phenomenon and, more recently, a number of important reports summarizing the status of present knowledge have appeared. Prominent among these are two National Research Council reports, "Atmosphere-Biosphere Interactions: Toward a Better Understanding of the Ecological Consequences of Fossil Fuel Combustion" (NRC, 1981), "Acid Deposition: Atmospheric Processes in Eastern North America" (NRC, 1983), the Environmental Protection Agency report "The Acidic Deposition Phenomenon and Its Effects: Critical Assessment Review Papers" (Altshuller and Linthurst, 1982), the U.S./Canada Work Group #2 report "Atmospheric Science and Analysis" (US/CWG #2, 1982), and the yearly National Acid Precipitation Assessment Program reports (NAPAP, 1982, 1983). This committee has made considerable use of these reports as well as the original literature.

In spite of the rapid growth of knowledge concerning acidic deposition the complexity of the problem leaves much left to be learned. Nevertheless, the overall outlines of the problem are known. Figure 1a presents the geographic distribution in North America of the mean value of pH of precipitation, while Figures 1b and 1c present the geographic distribution in North America of the annual wet deposition of sulfate and nitrate (anions of sulfuric and nitric acids, respectively). As stated in the second National Research Council report (NRC, 1983), "There are no known natural causes that can account for either the distribution or the value of acidity in eastern North America". However,

anthropogenic sources of the precursor compounds, sulfur dioxide (SO_2) and various oxides of nitrogen (NO_x), that result from fossil fuel combustion in electric power generation, manufacturing, and transportation are well known. Figures Id and Ie map the state-by-state and province-by-province anthropogenic emissions of SO_2 and NO_x in North America. The geographical pattern of emissions in Figures Id and Ie, in conjunction with the prevailing westerly winds, offers a hard-to-refute mesoscale explanation of the origin of acidic deposition. The first National Research Council report (NRC, 1981) found "the circumstantial evidence for their [anthropogenic emissions] role overwhelming". This Panel concurs in these judgements.

An examination of these Figures reveals that, of the thirty one states contiguous to or east of the Mississippi River, New Jersey is the twenty third from the top of the list of emitters of SO_2 and seventeenth from the top of the list of emitters of NO_x. When balanced against New Jersey's ranking as the seventh most populous state within these thirty one states and against New Jersey's substantial industrialization, these emission figures testify to the successful emissions abatement instituted in New Jersey over the last two decades. However, part of this abatement has resulted from the New Jersey utilities' choice of importing a substantial portion (37%) of this state's consumed electrical power from out-of-state. A truer ranking should include the emissions from this generated power in New Jersey's total. In spite of the emission abatement, Figure Ia shows that New Jersey's average pH value of precipitation, about 4.3, is only somewhat better than the worst state-wide average, about 4.1, in Ohio. This attests to the regionalization of the problem and the consequent need in any abatement strategy of considering the entire eastern part of the United States as well as eastern Canada.

The first National Research Council report (NRC, 1981) pointed out the agreement between studies in both the United States and Scandinavia that there is a critical pH value of precipitation below which acidification of sensitive waters (those having a small buffering capacity) occurs within a decade or two. This critical value is around 4.6. Based on this value and pH values occurring in the eastern United States, the report concluded that a 50% reduction in acidic deposition (a rise of 0.3 pH units) was needed to protect the numerous sensitive waters in this region.

The second National Research Council report (NRC, 1983) addressed the question whether a 50% reduction in deposition could be accomplished by a 50% reduction in emissions. Such a conclusion cannot be taken for granted because of the many nonlinear processes that occur in the atmosphere. A nonlinear process is one in which the output or result of the process is not linear in, that is, is not proportional to, the amount of an input substance. Though admitting that the data base was sketchy, the report (NRC, 1983) concluded that deposition was proportional to emission, or more precisely, "that there is no evidence for a strong nonlinearity in the relationships between long-term average emissions and deposition." Thus, on the basis of this report, a 50% reduction in deposition would be expected to result from a 50% reduction in emission when considered on a regional basis over a period of a year or so. A 50% reduction in deposition in New Jersey would cause the pH value of wet deposition to rise from about 4.3 to 4.6.

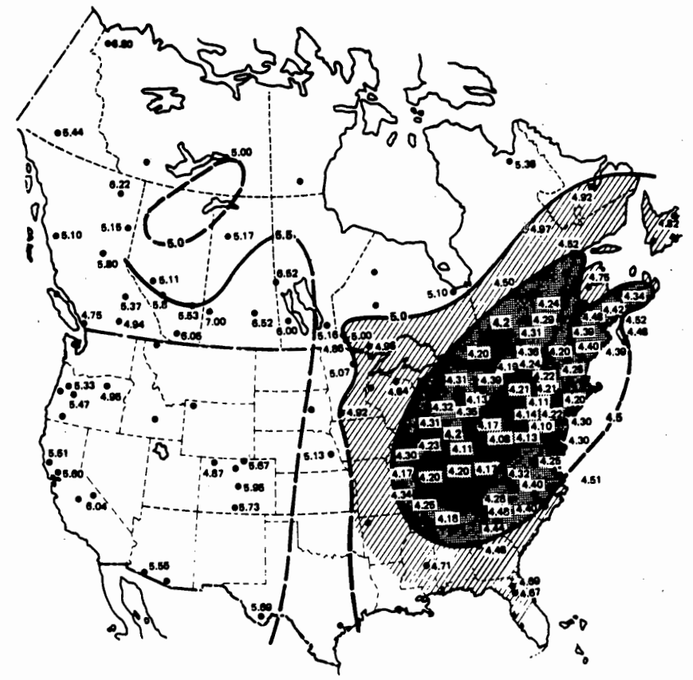

FIGURE 1a Annual mean value of pH in precipitation weighted by the amount of precipitation in the United States and Canada for 1980. SOURCE: U.S./Canada Work Group #2 (1982).

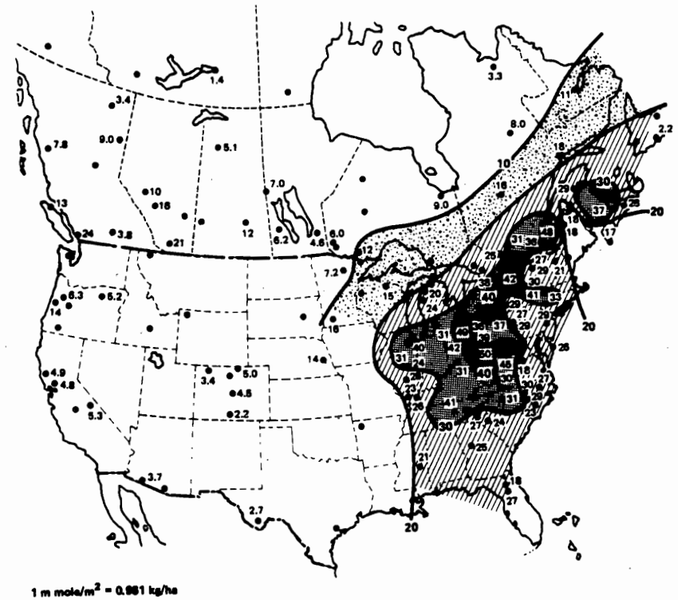

FIGURE 1b Spatial distribution of mean annual wet deposition of sulfate weighted by the amount of precipitation in North America in 1980 (mmoles/m^2). SOURCE: U.S./Canada Work Group #2 (1982).

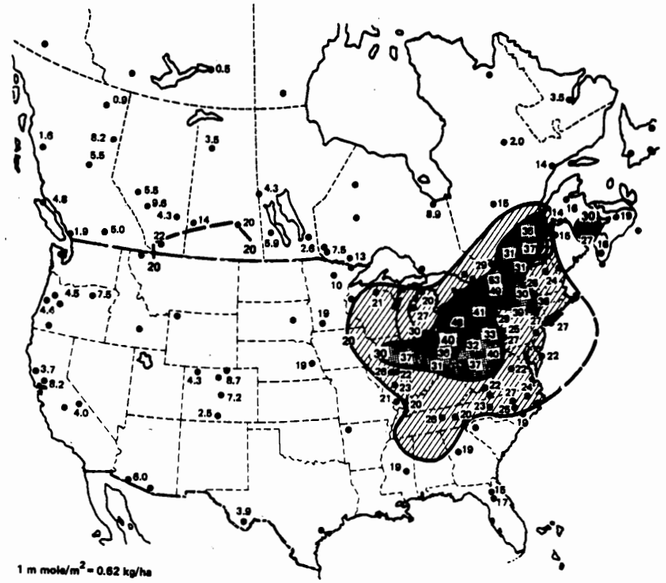

FIGURE **1c** Spatial distribution of mean annual wet deposition of nitrate weighted by the amount of precipitation in North America in 1980 (mmoles/m²). SOURCE: U.S./Canada Work Group #2 (1982).

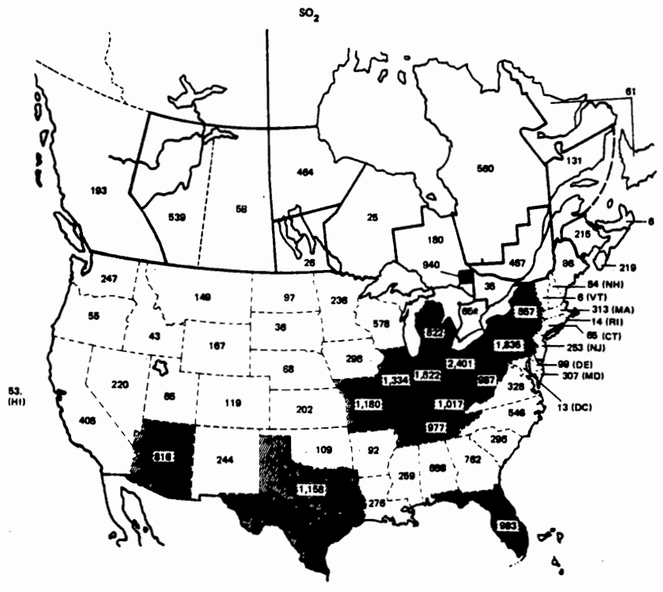

FIGURE **1d** Representative values of SO_2 emissions in the United States and Canada in 1980 (thousands of metric tonnes). SOURCE: U.S./Canada Work Group #3B (1982).

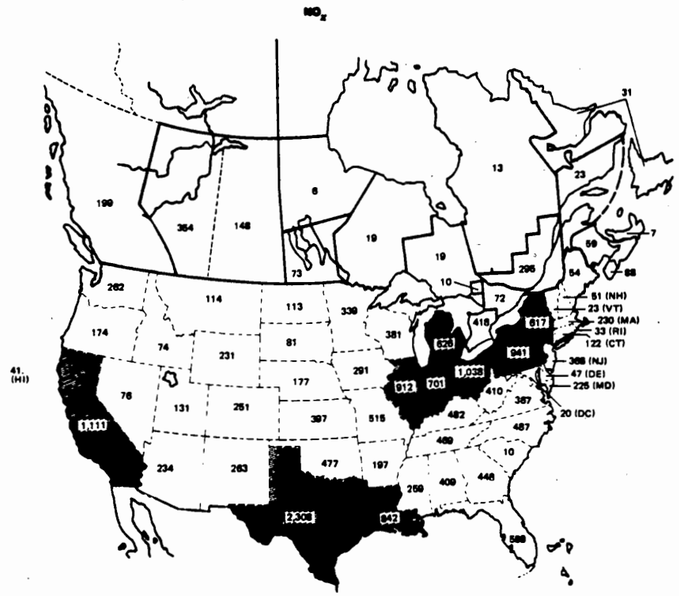

FIGURE **1e** Representative values of NO_x emissions in the United States and Canada in 1980 (thousands of metric tonnes). SOURCE: U.S./Canada Work Group #3B (1982).

However, studies published since issuance of the second National Research Council report (NRC, 1983) have raised questions about the linearity conclusion. Though there is no doubt that "what goes up comes down" when the entire global surface is considered, the percentage reduction in deposition may differ from the percentage reduction in emissions depending on the distance of the receptor from the source as determined by the chemical processes in the atmosphere during transport. Though the newer studies have not quantified the nonlinearity, they do suggest that the dominant nonlinearity will act to lower the percentage reduction of deposition below the percentage reduction in emissions for the eastern United States.

Rain naturally has some acidity as a result of the carbon dioxide component of the atmosphere and natural emissions of sulfur compounds. However, at pH values of rainfall of 4 to 4.5, typical of much of the northeastern United States, the acidity of rain can be easily 10 times higher than acidity of rain in remote areas of the world. Thus, anthropogenic emissions of SO_2 and NO_x have caused a very large change in an important property of our atmosphere. Since our atmosphere is such a pervasive influence on our environment, it seems eminently reasonable that the increased acidity will lead to some changes in our environment in time. This very general consideration along with the goal of preserving our environment leads naturally to a desire to reduce acidic deposition and hence its SO_2 and NO_x emission precursors.

In many quarters, however, it is felt that an emissions reduction strategy cannot be based solely on such general reasoning and that the specific adverse effects of acidic deposition must be determined in order to evaluate properly a cost/benefit tradeoff. It is toward this end that many present studies of acidic deposition are aimed. The proof of cause and effect in a scientifically

convincing manner, however, is a difficult task for two major reasons: (1) an adequately complete characterization of any ecosystem in a state undisturbed by acidic deposition (the "baseline") is not known and (2) any ecosystem contains a myriad of constituents and interactions and so attributing a specific effect to a specific cause is very difficult.

Nevertheless, it is the purpose of this report to assemble all the available information concerning the effects of acidic deposition in New Jersey, to draw those conclusions that, based on the experience and judgement of the panel, appear warranted scientifically, and to make recommendations of actions that are deemed needed.

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SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

Acidic deposition is primarily generated from emissions of sulfur dioxide, nitrogen oxide, and organic species which, in turn, largely result from the combustion of fossil fuels. A fraction of these emissions is converted in the atmosphere to sulfuric, nitric, and organic acids and various salts of these acids. These materials form aerosols which are deposited by wet and dry mechanisms on forests, lakes and cities both locally and many miles downwind. New Jersey is both an emitter of these precursors and a recipient of acidic deposition generated by the emissions of neighboring states. This report attempts to assess the damage that acidic deposition may be doing to New Jersey's ecosystems, its water supply, its manmade structures and the health of its citizens.

Emissions

The history of sulfur oxide (SO_x) emissions for New Jersey and for several neighboring states is summarized below. Sulfur dioxide (SO_2) accounts for most of the SO_x emissions.

	SO_x Emissions, Thousands of Metric Tons/yr				
	1950	1960	1970	1978	1980
N.J.	1188	452	536	294	253
N.Y.	772	1296	1021	954	857
Penns.	881	2145	2103	1725	1835
Ohio	813	2664	2837	2829	2401
33 Eastern States	10,784	18,853	23,492	21,742	21,436

New Jersey's reduction in SO_2 emissions began in the 1950's with the switch from coal to natural gas and oil, and continues to the present as a result of strict environmental regulations. It should be noted, however, that New Jersey imports a substantial amount of its power, partially accounting for the current low level of emissions. Our nearest neighbors with major emissions have shown decreases in SO_2 emissions from the 1970 levels, largely a result of the 1970 Clean Air Act. New York State has recently passed legislation to reduce its SO_2 emissions by an additional 245,000 metric tons per year by the 1990's. This legislation is specifically intended to help counter the damages from acidic deposition. While some of the southeastern states, due to their rapid growth and industrialization, continued to increase their emissions through 1980, the combined emissions of 33 eastern states have shown a reduction of about 10% since 1970, and somewhat more than that since the peak year of 1973.

Nitrogen oxide (NO_x) emissions increased quite rapidly up to 1978 (see Table below).

NO_x Emission, Thousands of Metric Tons/yr

	1950	1960	1970	1978	1980
N.J.	257	329	489	449	368
N.Y.	448	697	908	826	617
Penns.	435	927	990	1017	941
Ohio	453	872	1058	1160	1038
33 Eastern States	6,386	10,817	15,300	17,609	15,060

Automotive and other transportation emissions account for about 40% of the NO_x emissions. These emissions were reduced between 1978 and 1980 by about 15%, as a result of control on power generation and industrial emissions and auto emissions. As might be anticipated from its small size, high industrialization, and heavy automotive usage, New Jersey has the highest level of NO_x emissions per year per square mile of any of the states in the U.S.

The generation of organic acids in the atmosphere is poorly understood. It is known that they are related to hydrocarbon emissions (which have decreased over the past decade in New Jersey), but the relationship is obscure. Since organic acids appear to be a significant but not dominant influence on precipitation acidity and since their sources and effects are not well known, they have not yet been studied to the same extent as have sulfuric and nitric acids.

Deposition

Acidic deposition occurs in dry form and in wet form as in rain or snowfall. Wet deposition is readily measured, whereas dry deposition (the impaction and interception of aerosols and gases from the atmosphere) is significant but there is a lack of agreement on whether the measurements are representative of forested areas, open fields, etc. Wet deposition monitoring is, therefore, the most widely used technique in measuring acidic deposition.

There are two wet deposition sites operated by the New Jersey Department of Environmental Protection (DEP) that have sufficiently accurate records to permit estimates of the annual wet deposition rate. One site is at Washington Crossing State Park in Mercer County and the other is at Millville in Cumberland County. At these sites wet deposition has been estimated at 24-27 kg/hectare/yr of SO_4^{2-} ion with a corresponding average precipitation pH of 4.3. A similar level is found across our northern border in New York State. A recent study of the data from monitors in the eastern United States operated by the National Atmospheric Deposition Program (NADP) indicated that the average deposition rate was 23.1 kg/ha/yr in 1980-1981, with a corresponding pH of 4.35. Hence New Jersey, based on the limited monitoring within the State, is experiencing wet deposition with acidity similar to other parts of the northeastern United States. For comparison, the most polluted regions in the eastern United States received

40 kg/ha/yr, with an average rainfall pH of about 4.0, and the least polluted areas in the northeast received 15 kg/ha/yr with a pH of 4.7. The industrialized area of New Jersey close to New York City is indicated to receive 30 kg/ha/yr and above.

There is no long term record of wet deposition in New Jersey, but it is apparent from the history of sulfur dioxide and nitrogen oxide emissions that acidic deposition has been occurring for decades. Indeed, with the reduction in emissions both in New Jersey and in our neighboring states, acidic deposition would be expected to have been declining slightly over the past decade, but there is no critical evidence of such a reduction in New Jersey.

The damage to New Jersey from acidic deposition was assessed in the following ways:

1. The literature was reviewed for the types of damage that acidic deposition has done or has the potential to do within New Jersey and in adjacent areas.

2. The pertinent data within the Department of Environmental Protection on air and water quality were studied and evaluated.

3. Continuing acidic deposition research which is being carried out in New Jersey has been incorporated in the report. In several cases, the research is being directed by panel members.

As was observed in an NRC report (NRC, 1981), "in many respects man is now operating on nature's own scale." This poses the danger that our activities can cause a long term alteration in our environment. The acidity of rainfall (and other acidic deposition) is a case in point. From 90 to 95% of that acidity in the eastern United States arises from anthropogenic sources (NRC, 1983). A man-made change of that magnitude in such an important and pervasive environmental influence is bound to cause significant alterations in some portions of our environment in time. Thus, based on this very general consideration alone, reduction of emissions that cause the acidity is a sound goal.

It was found that the damage resulting from acidic deposition is extremely difficult to determine. Acidic deposition does not occur alone; it is always in conjunction with other factors. For example, the health effects can be the result of a combination of the acidic aerosols and the other pollutants. Lakes and streams can be acidified by natural causes, as well as by acidic deposition. Forests suffer from diseases, pest infestations and air pollution. The effects of acidic deposition must be separated from the variety of simultaneously occurring influences. In short, there are many confounding variables, the historical natural baseline is often poorly known, and thus proof of cause and effect is very difficult. The lack of data specific to New Jersey also makes it difficult to estimate damage in the state. The conclusions, then, contain some uncertainties, but they do reflect the experience and judgement of the panel.

Summary of Known and Suspected Effects of Acidic Deposition in New Jersey

- Sulfate aerosol concentrations average 10-11 ug/m^3 in the atmosphere and reach levels of 30-35 ug/m^3 during stagnation episodes in the summer. The present levels of acidic aerosols pose no threat to healthy adults. There is a potential threat of temporary pulmonary impairment to adolescent asthmatics during summer aerosol peaks.

- Ground and surface waters with pH much below 5.5 can, under some circumstances, dissolve toxic metals from soils and surface litter. In a study by the New Jersey Geological Survey, about 14 per cent of the domestic wells in one community in Ocean County have shown lead concentrations above the drinking water standard of 50 ppb. Recent information from the U.S. Geological Survey indicates that lead from solder and copper from pipes used in domestic plumbing systems may be leaching into the tap water in homes. This problem is currently under study. In northern New Jersey, Clyde Potts, Split Rock and Clinton reservoirs are near neutral but have low alkalinity. Limited analyses show that the metal content of these waters is quite low at the present time.

- Pinelands' ground waters are acidic and have low alkalinity with sulfate often the dominant anion. Preliminary data indicate that the sulfate adsorption of the soils is low, and the soils are probably saturated with sulfate. Whereas hydrogen ion is generally the major cation in surface water, the major cation in ground water is often calcium.

- Surface waters in the Pinelands have long been acidic (pH 3.5 - 4.5) as data taken over nearly 30 years confirm. The acidity is largely due to the combination of the low buffering capacity of the soil and to organic acids (fulvic, humic, and tannic acids) from decaying vegetation. This results in the typical brown-colored waters of the Pinelands. The organic acids are effective in complexing toxic aluminum ions. As a consequence, Pinelands ecosystems appear to be adopted to the acidity in as much as the waters support many kinds of acid-tolerant organisms including fish. Acidic deposition has been falling on the Pinelands in gradually increasing amounts over the last several decades. This acidic deposition has been estimated to have lowered the pH in the Pineland's waters by an additional 0.1 to 0.2 pH units. An investigation of the acidity of McDonald's Branch and Oyster Creek waters has indicated a pH decline of perhaps 0.2 to 0.26 pH units over the 24 years and 13 years respectively during which data have been gathered. The pH data are scattered such that the data base could also be interpreted as implying no decline has occurred over the years. Assigning causes to small overall declines in pH in a natural system subject to floods, drought, fires, and human activity becomes highly problematical, but the large fire in 1963 and the increasing acidic deposition which occurred from the 1950's to the mid-1970's have probably played significant roles in the pH trends. Any subtle effects of continued acidic deposition at the present levels on aquatic biota and on the terrestrial ecosystems in the Pinelands will probably be difficult to discern, but continued monitoring appears to be a prudent course.

- There are several lakes on the Kittatinny Ridge in northwestern New Jersey with pH ranging from 3.5 to 4.5. Two of these lakes, Long Pine Pond and Crater Lake, are severely stressed, containing only yellow perch, one of the most acid-tolerant fish. All of the lakes have high clarity and contain Sphagnum, an acid-tolerant moss, at the lake edges. These lakes share characteristics with systems elsewhere which have been acidified by acidic deposition. Continuing research in lakes on the Kittatinny Ridge indicates that the most acidic lakes are characterized by increased levels of such trace elements as lead in the water column and in various organisms.

- There are lakes in the Highlands of north-central New Jersey which are nearly neutral in acidity but which have low buffering capacity. These lakes could, in the future, become acidified. The streams and rivers in this area generally seem to have sufficient buffering to prevent a serious decline in pH.

- One study has noted a reduced growth rate in about two-thirds of the pitch pine and shortleaf pine in the Pinelands. However, no such effect was observed in white pine in this study. Given the relatively small number of trees sampled and the possibility that drought and other stresses may have contributed to the growth reduction, this study should be considered suggestive rather than conclusive. Other conifers have suffered similar reductions in growth rate in New England and as far south as Georgia. The cause of the reduction in growth rate is unknown, but acidic deposition is suspected of being one of a number of interacting factors. A survey on oaks, one of the predominant species in the State, is underway.

- Experimental evidence obtained in various parts of the U.S. indicates that rainfall of pH 4.0 does not generally have an adverse effect on agricultural crops. In particular no adverse effect has been found on New Jersey crops from presently occurring acidic rainfall, but, of course, not all crops or varieties have been studied. The traditional use of lime to produce a maximum crop yield provides additional protection against any acidifying effects on the soil.

- The available data give no indication of consistent changes in acidity levels nor elemental composition of soils from sections of northern New Jersey over the past several decades ascribable to acidic deposition. No generalization can be made of other sections of New Jersey.

- Environmental exposure probably has a major impact on exposed metals, alloys and stone of building structures and cultural or historical artifacts. However, weathering of these materials is not well understood and the added component due to acidic deposition is not well delineated. Documented materials damage in New Jersey due to acidic deposition has not been established nor has any economic cost of the materials damage been attempted.

- Sulfate aerosols are primarily responsible for limiting the visibility in New Jersey to 10-15 miles. The major visibility impairment occurs at high relative humidity during peak concentrations of sulfate aerosols in the summer months.

The data base relating acidic deposition to recreational activities within the state is limited or non-existent. The panel did not feel the estimation of this potential secondary type of damage was possible.

Likewise the panel felt there was insufficient economic data on the different types of damages to warrant attempting estimation of the value to the state of a reduction in acidic deposition.

The panel thought it interesting that the many months of review and study of the information on New Jersey uncovered no new or more serious set of damages or adverse effects than revealed in the one-day hearing held jointly by the Clean Air Council and the Governor's Science Advisory Committee in November, 1983.

Recommendations

Although known adverse effects of acidic deposition in New Jersey are, to date, not extensive, acidic deposition must nevertheless be regarded as a threat to our ecosystems, to our historical and cultural artifacts, and possibly to the health of some of our citizens. It is, therefore, in our best interest to monitor these potential threats for the purpose of developing a firm baseline of information and an estimation of future trends. A small amount of research is underway in New Jersey surveying damage to lakes, streams, groundwater, forests, crops and materials. A study of the health effects of summer air pollution peaks on adolescents is also underway in New Jersey. This research is obviously important to our knowledge of the acidic deposition phenomenon within the state.

Our more specific recommendation are as follows.

1. Continue monitoring precipitation pH and acidic deposition. Reinstitute the measurement of ambient concentrations of sulfate and nitrate aerosols as a measure of the future health threats and other adverse effects from acidic deposition. Measurements should include hydrogen ion concentrations on fine particulates as the hydrogen ion has been shown to be the essential "health stress indicator".

2. Study possible strategies for reducing emissions of acidic deposition precursors. Small scale dispersion models could be used to identify the strategies which would result in the largest reductions in wet and dry deposition in New Jersey. Since effects found by the panel are not overwhelming, no recommendation for a further general reduction in sulfur dioxide emissions for New Jersey in the absence of a regional or national program is felt to be justified at this time.

3. Expand the study of forests and urban trees to learn whether the reduced growth rate observed in some conifer species in the Pinelands occurs elsewhere in New Jersey. Determine whether the reduced growth is caused by acidic deposition, other pollutants or natural causes. Studies should include other important species within the state, such as oaks. Evaluate forest soils throughout the state to determine the potential for the loss of nutrients through leaching and for the mobilization of toxic metals due to acidic deposition.

4. Evaluate surface and ground waters throughout the state in areas identified as potentially susceptible to acidic deposition, principally in the Pinelands, the Highlands and on Kittatinny Ridge. This program should include water chemistry measurements and estimates of the status of aquatic communities, including their pH and trace element tolerance. A study of major ion chemistry including hydrogen ion flux through susceptible watersheds should also be undertaken to clarify the relationship of acidic deposition to soil, groundwater and surface water acidity. Consider various approaches for treatment and/or management of severely acidified lakes on Kittatinny Ridge.

5. Require routine testing of potable water supplies to include such parameters as pH, toxic metals and sulfate concentrations. The New Jersey Safe Drinking Water Act of 1984 establishes a mechanism for monitoring the water quality of all reservoirs and commercial wells. The act establishes a Drinking Water Quality Institute which shall, among other duties, make recommendations concerning which contaminants should be tested. The panel strongly advises that the Institute include pH and toxic metals which are soluble in acidified water among the parameters for which testing is required in both treated and untreated waters. The panel also recommends that sulfate concentration be similarly monitored in the untreated water to aid in establishing a possible influence of acidic deposition.

6. Maintain an inventory of cultural and historic artifacts subject to damage by acidic deposition. Photographic, chemical and dimensional studies should be used to establish the present conditions of these artifacts in order to document future change or damage.

7. Assist the health studies being carried out in New Jersey on adolescents during air pollution peaks by monitoring sulfate and ozone levels especially. Institute studies of the effect of acidic aerosols in the range of actual peak concentrations occurring in New Jersey on elderly people and on those with chronic lung disease.

8. Allocate funds to carry out the expanded research program which has been recommended herein. Areas specifically needing emphasis are (1) a further assessment of the health effects on adolescents and the elderly with chronic lung disease which can arise from acidic aerosols, (2) the delineation of the role of acidic deposition on forest decline, and (3) determination of the effect of acidic deposition on lakes, streams, and ground water in susceptible areas. Although no detailed cost estimate has been made, the recommended research could cost in the order of 2.0 million dollars/-year for 3 to 5 years. This would include funding for DEP to coordinate the significantly expanded acidic deposition research in New Jersey.

CHAPTER I: ATMOSPHERIC PROCESSES

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PHYSICAL, CHEMICAL AND METEOROLOGICAL ASPECTS OF ACIDIC DEPOSITION

A. MAJOR PHYSICAL AND CHEMICAL PROCESSES INVOLVED IN ACIDIC DEPOSITION

1. Sources of Acidic Gases in the Atmosphere

In any assessment of the acidity of precipitation, it is important to recognize that rainwater would be acidic even if the atmosphere were unpolluted. Carbon dioxide, a natural constituent of the atmosphere, dissolves in water to form carbonic acid. The equilibrium concentration of carbonic acid in rainwater would be such as to make its pH value equal to approximately 5.6, as opposed to a value of 7.0 that would be classified as completely neutral. When one adds in the effect of natural sources of sulfur, which vary widely in space and time, unpolluted pH values in the range 4.5 - 5.6 are obtained (Charlson and Rodhe, 1982). However, there are also natural substances, such as ammonia, that tend to increase the pH. It is probably impossible to find a location on earth where the atmosphere can be thought of as completely unpolluted, but several recent investigations of pH values in remote areas (e.g., Galloway et al., 1982) show values of 4.8 - 5.0.

The primary anthropogenic sources of acidic gases in the atmosphere are the large quantities of sulfur and nitrogen oxides that are discharged from the combustion of fossil fuels, such as coal, oil, and gas. The sulfur oxide (SO_x) emissions come about because sulfur is usually found as an ingredient in oil and coal, although in widely variable amounts. Nitrogen oxide (NO_x) emissions result from any high-temperature combustion processes involving air. Although nitrogen and oxygen are the principal natural constituents of air, it is only at the high temperatures associated with combustion that they combine into nitrogen oxides.

Table 1 gives estimates of annual emissions of SO_x for states in the eastern half of the country. Husar (1985) believes that emissions for 1950 were probably much higher than those shown in the table because of the existence of categories of sources, such as railroads, that were not considered. He additionally believes that SO_2 source strengths in New Jersey and elsewhere in the Northeast were probably fairly high even during the 19th century. When viewed from the standpoint of emissions density (emission per unit area) as shown in Table 2, the northeastern states and those in the Ohio Valley rank highest. In 1978, New Jersey had the seventh highest SO_x emissions density in the United States. Table 3 gives a county by county breakdown of SO_2 emissions in New Jersey. It should be noted that the figures given in this table do not agree precisely with analogous figures for 1980 given by Gschwandtner (1981), in Table 1. The New Jersey figures compiled by the DEP are believed to be the most reliable. Although the annual emissions have increased with time in most cases, they have not increased nearly as much as has the use of electric power. The reason for this is primarily the gradual transition to fuels that are lower in sulfur content (Gschwandtner et al., 1981), primarily natural gas and low-sulfur oil. The figures for New Jersey, Connecticut, and Delaware are particularly noteworthy examples of this trend. Nevertheless, fossil-fueled electric utility plants are now and are projected to remain the largest sources of sulfur dioxide (SO_2), emitting more than four times as much as industrial, commercial, automotive, and residential sources combined.

Table 4 gives estimates of annual emissions of NO_x for states in the eastern half of the country. When viewed from the standpoint of emissions density (emission per unit area) as shown in Table 5, the northeastern Atlantic coastal

TABLE 1

ESTIMATES OF ANNUAL EMISSIONS OF SULFUR OXIDES
(10^6 kg yr^{-1}) (Gschwandtner, 1981)

	1950	1960	1970	1978	1980
Alabama	126.2	557.3	888.6	728.2	821.2
Arkansas	38.0	23.8	36.0	114.1	92.1
Connecticut	127.6	219.4	288.1	101.7	65.2
Delaware	95.7	178.1	202.8	170.9	99.2
District of Columbia	29.4	35.0	70.9	15.9	13.4
Florida	203.9	309.9	873.4	622.6	993.3
Georgia	108.9	180.0	372.6	641.2	761.7
Illinois	789.5	2227.5	2290.0	1586.8	1334.1
Indiana	484.0	1673.2	1762.8	1678.3	1821.5
Iowa	157.6	331.0	336.3	349.6	298.2
Kentucky	102.7	573.0	1163.1	1207.8	1016.7
Louisiana	211.2	199.8	288.8	326.5	276.0
Maine	34.3	49.7	74.4	59.9	86.0
Maryland	362.3	370.0	424.7	324.4	306.6
Massachusetts	822.2	340.9	522.5	365.1	312.5
Michigan	469.7	986.1	1380.5	1014.7	822.7
Minnesota	367.9	355.9	409.5	344.1	236.2
Mississippi	42.5	37.2	72.4	248.1	250.5
Missouri	649.2	529.7	1004.9	1186.8	1180.4
New Hampshire	63.0	26.5	87.1	61.5	84.3
New Jersey	1188.3	452.1	535.8	294.0	253.3
New York	772.0	1295.7	1021.3	953.9	856.7
North Carolina	278.3	211.6	483.6	510.9	546.4
Ohio	812.6	2663.7	2837.3	2828.5	2401.1
Pennsylvania	880.8	2145.4	2102.8	1725.2	1834.5
Rhode Island	61.3	79.5	54.6	19.4	13.8
South Carolina	40.4	105.3	168.0	265.3	295.8
Tennessee	88.3	663.8	897.4	1056.4	976.6
Texas	830.4	817.3	1050.0	1157.3	1158.2
Vermont	3.7	7.8	11.7	7.9	6.2
Virginia	143.1	155.6	431.0	326.4	327.5
West Virginia	321.3	481.2	889.1	952.8	986.8
Wisconsin	196.8	548.2	293.3	602.3	1031.9
	10784.1	18852.6	23492.1	21741.6	21435.6

TABLE 2

ANNUAL EMISSIONS DENSITIES OF SULFUR OXIDES
(kg km^{-2} yr^{-1}) (Gschwandtner, 1981)

	1950	1960	1970	1978	1980
Alabama	944	4168	6646	5446	6142
Arkansas	276	173	262	829	4858
Connecticut	9834 (6)	16907 (7)	22201 (5)	7836	5024
Delaware	17960 (4)	33414 (1)	38063 (1)	32061 (1)	18616
District of Columbia	169070	200904	407038	91344	77059
Florida	1344	2043	5757	4104	6547
Georgia	714	1180	2443	4204	4994
Illinois	5403 (10)	15245 (9)	15672 (9)	10860 (10)	9130
Indiana	5148	17797 (6)	18750 (6)	17851 (3)	19374
Iowa	1081	2270	2306	2397	2045
Kentucky	981	5475	11114	11541 (9)	9711
Louisiana	1680	1589	2297	2597	2195
Maine	398	577	865	696	997
Maryland	13220 (5)	13502 (10)	15500 (10)	11840 (8)	11187
Massachusetts	38436 (2)	15935 (8)	24425 (4)	17070 (4)	14608
Michigan	3114	6538	9153	6728	5454
Minnesota	1689	1634	1880	1580	1084
Mississippi	344	301	586	2007	2027
Missouri	3596	2934	5566	6574	6538
New Hampshire	2615	1099	3614	2551	3499
New Jersey	58539 (1)	22273 (4)	26396 (3)	14483 (7)	12478
New York	6022 (9)	10088	10288	7427	6682
North Carolina	2043	1553	3550	3750	4011
Ohio	7609 (7)	24943 (3)	26568 (2)	26486 (2)	22483
Pennsylvania	7500 (8)	18269 (5)	17906 (7)	14691 (6)	15620
Rhode Island	19486 (3)	25288 (2)	17352 (8)	6174	4386
South Carolina	502	1308	2088	3305	3675
Tennessee	807	6065	8199	9652	8925
Texas	1199	1180	1516	1671	1672
Vermont	149	313	470	317	249
Virginia	1353	1471	4076	3087	3096
West Virginia	3532	7682	14192	15209 (5)	10847
Wisconsin	1353	3768	2016	4140	7094

Note: Numbers in parentheses indicate numerical ranking of 10 highest emissions densities (D.C. excluded).

TABLE 3

COUNTY BY COUNTY INVENTORY OF SO_2 EMISSIONS IN NEW JERSEY DURING 1980
AS DETERMINED FROM EMISSIONS INVENTORIES COMPILED
BY THE NEW JERSEY DEPARTMENT OF ENVIRONMENTAL PROTECTION

COUNTY	INDUSTRIAL EMISSIONS (tons per year)	HIGHWAY EMISSIONS (tons per year)	OTHER EMISSIONS (tons per year)	TOTAL EMISSIONS (tons per year)
Atlantic	2479	306	3519	6304
Bergen	12008	1751	5487	19246
Burlington	2048	599	2001	4648
Camden	5849	850	3329	10028
Cape May	57677	135	837	58649
Cumberland	12374	237	3114	15725
Essex	17747	1389	6479	25615
Gloucester	17578	312	1357	19247
Hudson	7973	759	4190	12922
Hunterdon	6769	158	1289	8216
Mercer	18904	591	2895	22390
Middlesex	23928	1104	3784	28816
Monmouth	1567	938	2459	4964
Morris	1632	817	2466	4915
Ocean	1312	575	3462	5349
Passaic	1199	866	2712	4777
Salem	22166	115	1167	23448
Somerset	3157	333	1254	4744
Sussex	36	194	1188	1418
Union	16049	1180	4191	21420
Warren	4780	161	1254	6195

TOTAL 309,036

TOTAL EQUAL TO: 280×10^6 Kg/yr

Note: 280×10^6 Kg/yr compares with the 1980 emissions for New Jersey of 253×10^6 Kg/yr listed in Table 1.

TABLE 4

ESTIMATES OF ANNUAL EMISSIONS OF NITROGEN OXIDES
(10^6 kg yr^{-1}) (Gschwandtner, 1981)

	1950	1960	1970	1978	1980
Alabama	156.6	279.2	377.6	429.7	480.5
Arkansas	104.0	105.0	174.9	197.4	197.2
Connecticut	91.9	127.8	183.4	166.1	121.6
Delaware	18.0	46.5	65.3	64.1	47.1
District of Columbia	28.9	31.8	52.9	30.4	19.9
Florida	187.4	292.0	501.4	705.3	588.0
Georgia	155.1	206.4	361.5	498.6	448.3
Illinois	544.0	813.3	1025.6	1036.1	912.0
Indiana	268.9	531.0	523.3	545.5	701.3
Iowa	132.3	196.0	280.7	291.4	290.9
Kentucky	132.1	253.7	451.4	511.2	482.0
Louisiana	292.1	486.2	923.4	1447.3	842.2
Maine	38.6	44.6	68.8	69.5	53.9
Maryland	98.8	202.3	271.2	284.9	225.1
Massachusetts	149.2	231.5	326.7	331.4	230.0
Michigan	289.0	532.7	768.3	765.6	625.9
Minnesota	150.3	217.6	302.5	362.0	338.8
Mississippi	83.1	137.0	164.9	247.0	258.8
Missouri	180.4	267.2	385.3	311.4	314.9
New Hampshire	16.6	28.2	57.8	60.6	75.5
New Jersey	256.6	329.4	488.8	448.8	368.3
New York	447.9	697.4	908.4	825.7	616.5
North Carolina	174.4	263.5	496.0	536.9	586.5
Ohio	452.8	871.8	1057.9	1159.8	1038.4
Pennsylvania	435.1	926.6	989.5	1017.2	941.2
Rhode Island	30.4	41.0	43.8	39.5	33.1
South Carolina	79.7	136.6	215.6	272.5	236.1
Tennessee	150.1	305.1	424.3	533.6	469.2
Texas	786.1	1302.9	2313.9	3012.1	2307.7
Vermont	10.2	12.4	29.9	23.5	22.4
Virginia	167.1	268.8	398.7	395.6	367.1
West Virginia	108.1	204.2	315.1	419.8	410.3
Wisconsin	178.4	269.4	413.5	429.3	381.4
	6386.0	10817.2	15299.6	17609.4	15059.7

TABLE 5

ANNUAL EMISSIONS DENSITIES OF NITROGEN OXIDES
($kg\ km^{-2}\ yr^{-1}$) (Gschwandtner, 1981)

	1950	1960	1970	1978	1980
Alabama	1171	2088	2824	3214	3593
Arkansas	756	763	1271	1435	1433
Connecticut	6311 (4)	9851 (4)	14137 (3)	12803 (3)	8350
Delaware	3378 (10)	8726 (5)	12249 (5)	12031 (5)	8839
District of Columbia	165891	182598	304180	174790	114229
Florida	1235	1925	3305	4649	3657
Georgia	1017	1353	2370	3269	2939
Illinois	3723 (6)	5566 (10)	7019	7019 (10)	6241
Indiana	2860	5648 (9)	5566	5802	7459
Iowa	1044	1344	1925	1998	2295
Kentucky	1262	2424	4313	4885	4604
Louisiana	2324	3868	7346 (9)	11513 (6)	6700
Maine	449	518	799	808	627
Maryland	3604 (8)	7382 (8)	9897 (7)	10397 (8)	8211
Massachusetts	6973 (3)	10823 (3)	15272 (2)	15490 (2)	10749
Michigan	1916	3532	5094	5076	4149
Minnesota	690	999	1389	1662	1555
Mississippi	672	1108	1334	1998	2092
Missouri	999	1480	2134	2833	1743
New Hampshire	690	1171	1397	2515	3138
New Jersey	12639 (1)	16226 (1)	24080 (1)	22110 (1)	18140
New York	3487 (9)	5430	7073 (10)	6429	4799
North Carolina	1280	1934	3641	3941	4304
Ohio	4240 (5)	8163 (6)	9906 (6)	10860 (7)	9723
Pennsylvania	3705 (7)	7890 (7)	8426 (8)	8662 (9)	8014
Rhode Island	9670 (2)	13048 (2)	13910 (4)	12240 (4)	10528
South Carolina	990	1698	2679	3387	2932
Tennessee	1371	2788	3877	4921	4285
Texas	1135	2170	3341	4349	3331
Vermont	409	499	1162	944	898
Virginia	1580	2542	3723	3741	3471
West Virginia	1725	3260	5030	6701	6547
Wisconsin	1226	1852	2842	2951	2621

Note: Numbers in parentheses indicate numerical ranking of 10 highest emission densities (D.C. excluded).

states, Ohio, and Pennsylvania rank highest. In 1980, New Jersey had the highest NO_x emissions density of any state in the United States. Table 6 gives a county by county breakdown of NO_x emissions in New Jersey. It should be noted that the figures given in this table do not agree precisely with analogous figures for 1980 given by Gschwandtner (1981) in Table 4. On a nationwide basis, emission rates increased by more than a factor of two between 1950 and 1970, but have leveled off since then, and even decreased slightly. New Jersey and surrounding states reflect this trend. Emissions by mobile sources appear to be responsible for about 40 percent of the NO_x in most of the northeastern states, but represent only about 25 percent of the total in Ohio and Illinois. Industrial sources and electric utilities are the major remaining contributors.

2. Chemical Reactions

The principal acid components of precipitation are generally nitric and sulfuric acids. Lesser amounts of hydrochloric acid and low molecular weight organic acids (i.e., formic and acetic) are present as well. The acidic compounds may arise from one or a combination of the following:

- Compounds formed in the gas phase and incorporated as gases into cloud droplets or precipitation (Schwartz, 1984).
- Compounds present on particles around which cloud droplets nucleate or which are scavenged by precipitation during its fall (Gill et al., 1983).
- Compounds formed in the liquid phase (in clouds or precipitation) from incorporated gases (Chameides and Davis, 1982; Graedel and Goldberg, 1983). These formation reactions may involve dissolved or undissolved transition metal compounds or soot (Martin, 1983) functioning as catalysts.

In order to assess any acidic deposition reduction strategy, it is necessary to know which of the processes above are significant and which species control those processes.

a. Nitric acid

Nitric acid (HNO_3) is generated in the atmosphere by a gas phase reaction involving nitrogen dioxide (NO_2) emitted from combustion processes,

$$NO_2 + OH \rightarrow HNO_3$$

HNO_3 is very soluble in water, and is readily incorporated into droplets. Unlike the SO_2-H_2SO_4 pair, oxidation of NO_2 to nitric acid in solution appears unlikely because of the very low solubility of NO_2 (and also of NO). If this surmise is correct, one would anticipate a higher nitrate concentration in rain

TABLE 6

COUNTY BY COUNTY INVENTORY OF NO_x EMISSIONS IN NEW JERSEY DURING 1980
AS DETERMINED FROM EMISSIONS INVENTORIES COMPILED
BY THE NEW JERSEY DEPARTMENT OF ENVIRONMENTAL PROTECTION

COUNTY	INDUSTRIAL EMISSIONS (tons per year)	HIGHWAY EMISSIONS (tons per year)	OTHER EMISSIONS (tons per year)	TOTAL EMISSIONS (tons per year)
Atlantic	1460	8785	2185	12430
Bergen	19039	30707	10123	59869
Burlington	7924	17449	3630	29003
Camden	7521	17478	4805	29804
Cape May	23252	3079	988	27319
Cumberland	4185	4040	2307	10532
Essex	29904	19940	11107	60951
Gloucester	12832	9198	2378	24408
Hudson	13523	9479	7027	30029
Hunterdon	6684	4891	1456	13031
Mercer	14402	11738	3818	29958
Middlesex	15067	29063	7567	51697
Monmouth	1658	19818	4426	25902
Morris	2141	16195	4537	22873
Ocean	1214	10229	2474	13917
Passaic	2012	12896	5215	20123
Salem	10036	3873	1412	15321
Somerset	3021	9397	2729	15147
Sussex	33	2871	1198	4102
Union	14963	17509	7147	39619
Warren	6320	5349	1578	13247
TOTAL				549,283
TOTAL EQUAL TO:				498 x 10^6 Kg/yr

Note: This compares with 368 x 10^6 Kg/yr listed for 1980 New Jersey emissions on Table 4.

in the summer than in the winter, since OH is much more abundant in the summer. Such a relationship is, in fact, seen in the rather sparse data which are available (Galloway and Likens, 1981).

b. Sulfuric acid

Sulfuric acid (H_2SO_4) can be formed in the atmosphere by gas phase chemistry. Although several sulfur-containing compounds may act as sulfuric acid precursors, sulfur dioxide (SO_2) appears to be the species of primary interest in the northeastern United States. Its gaseous reaction sequence is:

$$SO_2 + OH \rightarrow HSO_3$$

$$HSO_3 + O_2 \rightarrow SO_3 + HO_2$$

$$SO_3 + H_2O \rightarrow H_2SO_4$$

The sulfuric acid formed has a very low vapor pressure and will promptly condense to form sulfuric acid droplets, which in turn may be scavenged by water droplets.

The limiting step in the above reaction sequence is the initial reaction, since SO_2 must compete with other atmospheric trace gases for the scarce OH radicals, and since OH concentrations at night, in winter, or on heavily overcast days are very low.

Sulfur dioxide is very soluble in water and can undergo equilibrium transformation within the droplets as well as in the gas phase.

$$SO_{2(aq)} + H_2O \rightarrow H^+ + HSO_3^-$$

$$HSO_3^-(aq) \rightarrow H^+ + SO_3^{2-}$$

At acidities typical of New Jersey raindrops, most of the dissolved SO_2 will be in the bisulfite (HSO_3^-) form.

Several routes for bisulfite oxidation in atmospheric droplets are known. Two involve the oxidizers hydrogen peroxide (H_2O_2) and ozone (O_3):

$$HSO_3^- + H_2O_2 \rightarrow HSO_4^- + H_2O$$

$$HSO_3^- + O_3 \rightarrow HSO_4^- + O_2$$

Depending on the acidity of the solution, the sulfate may be present as bisulfate (HSO_4^-) ion, or may rapidly be transformed to the sulfate (SO_4^{2-}) ion.

Three catalytic processes, involving dissolved iron [Fe(III)], dissolved manganese [Mn(II)], and carbon [C] (soot), are also known to oxidize bisulfite ions in solution. The catalysts are present on aerosol particles absorbed into the water droplets. The details of the catalytic reactions are not understood, but they may be expressed schematically as:

$$HSO_3^- + Fe(III) \rightarrow HSO_4^- + Fe(II)$$

$$HSO_3^- + Mn(II) \rightarrow HSO_4^- + Mn(I)$$

$$HSO_3^- + C \rightarrow HSO_4^- + C$$

Other possibilities for bisulfite oxidation include reactions with organic compounds and with solid catalytic metal particles.

The importance of the possible solution processes for bisulfite oxidation depend on the concentrations of the reactants (many of which are unmeasured in cloud or rain droplets) and on the droplet pH. One comparison of these processes was made by Martin (1983), who constructed the graph shown in Figure 2a. The curves in Figure 2a were calculated by choosing typical concentrations for the reactants, and thus apply only to droplets of that chosen composition. H_2O_2 is seen to dominate the bisulfite oxidation, except at pH levels of 5 and above, where ozone oxidation of bisulfite is controlling. Given the enormous range of concentrations that have been measured in clouds and rain, other equally plausible scenarios can be put forward. In Figure 2b developed by Dr. Graedel, the competing bisulfite oxidation rates are shown for a situation where H_2O_2 is reduced by a factor of five below that used for Figure 2a (this is typical of winter conditions) and iron and manganese concentrations are increased by a factor of one hundred (typical of rain near industrial areas). In this case, the metal ion interactions play significant roles in the oxidation.

The rate of oxidation of bisulfite to sulfate is thus expected to vary with season of the year, with the presence and chemical reactions of at least five different oxidizers from different sources, with the atmospheric moisture content, and with the intensity of solar radiation. The variation in rate could easily be a factor of ten or more, a variation that translates directly into variations in distance from the source of SO_2 before oxidation occurs. Winter studies of rain chemistry in the northeastern United States (Camerota et al., 1983) show only about twenty percent as much sulfate as during the summer. Since SO_2 emissions are thought to be relatively constant throughout the year, this implies a substantial decrease in the oxidizing ability of the atmosphere during the winter months.

c. Hydrochloric acid

The evidence suggests that hydrochloric acid (HCl) is incorporated into droplets from the gas phase rather than being formed in the liquid phase, since HCl is very soluble in water, is present only at very low concentrations in the gas phase, and is not known to be generated efficiently by any solution chemistry process applicable to the atmosphere.

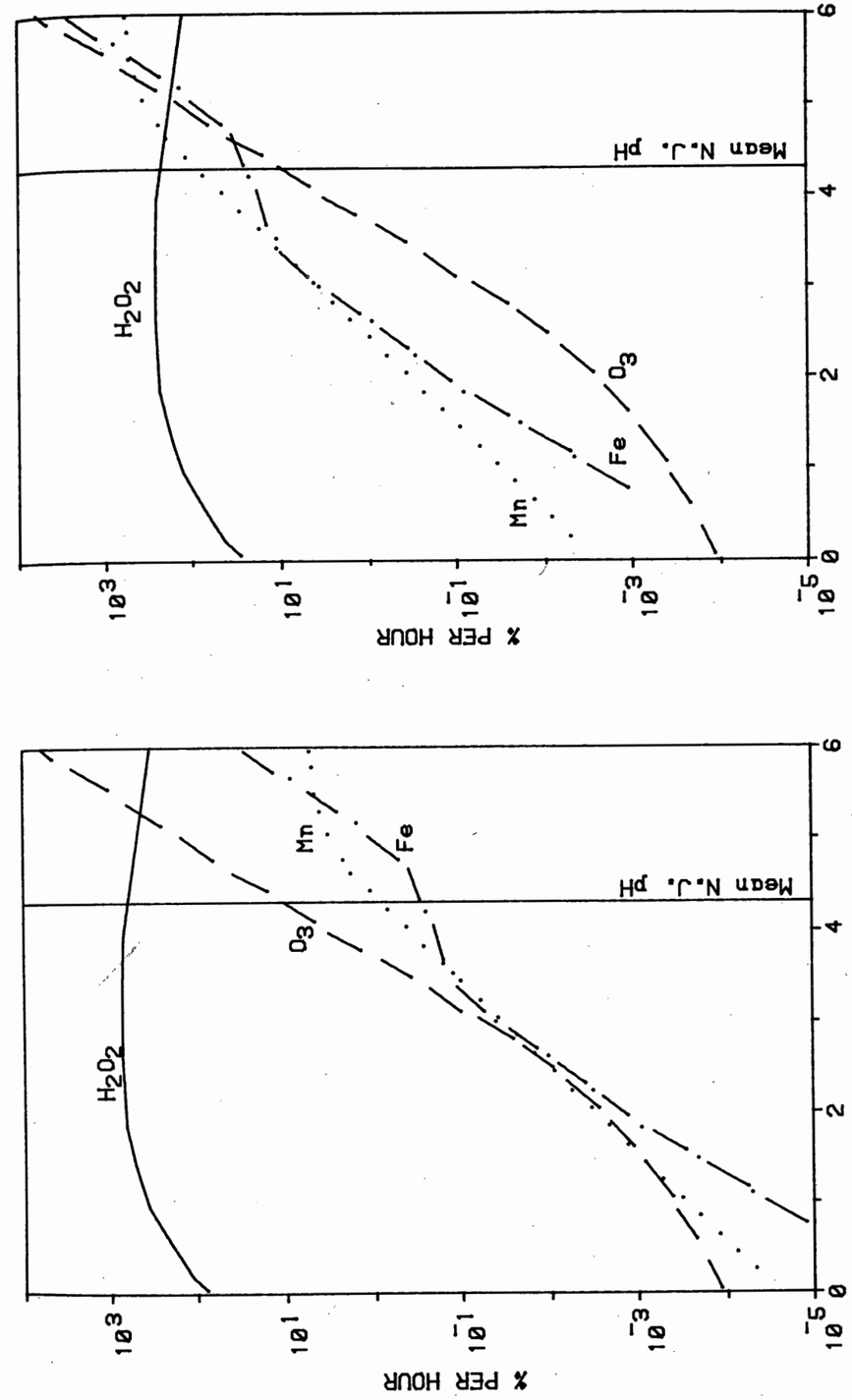


Figure 2a,b; Rates of Bisulfite Oxidation in Water Droplets New Jersey Annual Mean pH Shown for Comparison

d. Organic acids

In certain areas of the world well removed from sources of gaseous sulfur and nitrogen compounds, the pH of rain is controlled by the presence of organic acids (Keene et al., 1983). The principal sources of these acids are not known. In the liquid phase, organic acids can be readily formed by a variety of oxidation processes. The most likely acid precursors are the aldehydes, which are generated in the atmosphere by gas phase chemical reactions. It appears that organic acids will be relatively unimportant in northern United States precipitation (Cuiang et al., 1984), but until additional information is available, it is impossible to rule out organic chemical influences on acidic precipitation.

3. Dispersion of Air Pollutants, Particularly as Related to Questions Surrounding Acidic Deposition

The meteorological effects that influence the transport and dispersion of plumes over large distances are well-known, but not readily quantified. Plumes are carried horizontally by the wind flow at the altitude where they are located. Wind patterns often vary greatly with height, hence plumes released at different altitudes may travel at different speeds and in widely different directions. Systematic vertical air motions that are associated with large-scale weather patterns (e.g., low and high pressure systems) can cause plumes to ascend and descend as they travel. Turbulent (non-systematic) air motions, whose magnitudes are affected by wind, sunshine, the presence of physical obstructions, and other factors, cause plumes to gradually spread as they travel. Temperature inversions (regions of the atmosphere in which temperature increases with height) above a plume limit its vertical expansion. Plumes lying above inversions tend to be limited in their interaction with the ground. The height and intensity of inversions often vary greatly with time of day, thereby imposing a 24-hour periodicity on the behavior of plumes.

A number of experimental studies have been done to investigate plume behavior on scales of several hundreds of miles, and mathematical models have been developed (see, for example, Heffter, 1980; Draxler and Taylor, 1982). Such models, in conjunction with models for the chemical and physical processes that take place within the plume, have been used in studies of both wet and dry deposition.

4. Mechanisms by Which Acidic Materials Are Deposited onto the Ground

Sulfur and nitrogen oxides and their related compounds may exist in plumes in gaseous, liquid, and/or solid form. In none of these three forms do these materials readily "fall out" of the atmosphere. In fact, in the absence of deposition mechanisms, these compounds have been shown to remain airborne for thousands of miles.

In the case of wet deposition (that is, deposition in the form of rain, snow, or fog), the pollutants become incorporated into ice crystals or droplets of water, and fall out of the atmosphere together with the larger, heavier water droplets or snowflakes. In order for this to occur, the pollutant materials must be transported to some region of the atmosphere in which condensed water

exists, and they must become incorporated into the condensed water. Often, once association occurs, chemical reactions in the aqueous phase may occur. This entire process is known as precipitation scavenging, and may occur in one or a combination of the following ways (Pruppacher and Klett, 1978). These include:

a. Nucleation - The reaction of liquid droplets or ice crystals forming around pollutant particles because of the particles' thermodynamic affinity for water.

b. Diffusion - The migration of a pollutant toward a liquid droplet or ice crystal in response to the fact that the pollutant concentration in the immediate vicinity of the droplet or crystal is lower than in the surrounding area. This occurs for pollutant gases or for particles less than a few tenths of a micrometer in diameter. For pollutant gases or for particles less than a few tenths of a micrometer in size, the pollutant may migrate toward a liquid droplet or ice crystal in response to the fact that the pollutant concentration in the immediate vicinity of the droplet or crystal is lower than in the surrounding area.

c. Impaction - This occurs for pollutant droplets or particles larger than about one micrometer. Incorporation into raindrops or ice crystals may occur as the result of collisions.

These processes may occur within the clouds themselves or in the region beneath the clouds where precipitation is falling. Lazrus et al. (1983) indicate that most attachment occurs near the bases of clouds.

Deposition of pollutant material in particulate or gaseous form (dry deposition) usually occurs through direct attachment of the pollutant to exposed surfaces at or near the ground. The mechanisms by which this occurs are similar to processes (2.) and (3.) above, except that the capturing occurs on leaves, soil, water, snow, etc. The rates at which deposition onto various materials occurs have been studied by a number of investigators. Summaries of this work are given by Sehmel (1980), Garland (1979), and Chamberlain (1980).

5. Visibility Reduction as a Manifestation of Acidic Materials in the Atmosphere

The chemical and physical processes described in this report lead to the formation of aerosols whose sizes often lie in the 0.1 - 1.0 micrometer range. In fact, studies have shown that a large portion of aerosols found in the northeastern U.S. whose sizes lie in this range are composed of sulfates and nitrates. Because these aerosols are so numerous in the atmosphere, and because their sizes lie in the same range as the wavelengths of visible light, they are particularly effective in reducing visibility. We observe this visibility reduction as a milky haziness that is pervasive in the northeastern United States. Trijonis and Yuan (1978) and others have shown that sulfate concentrations and visibility are closely related in the northeast. However, at least part of this relationship can be attributed to the fact that the same meteorolo-

gical conditions that produce high sulfate concentrations also tend to increase humidity and concentrations of other pollutants that affect visibility.

B. CLIMATOLOGY OF NEW JERSEY AS IT RELATES TO ACIDIC DEPOSITION

1. Wind Patterns

Surface winds in New Jersey most often blow from the northwest in winter and from the southwest in summer (U.S. Dept. of Commerce, 1981). However, because there are certain preferred wind directions associated with precipitation, average surface wind conditions during precipitation events tend to be more southerly and easterly than the overall average values. It has been shown, however, that the winds that are most relevant to acidic precipitation are those at cloud level (see for example Lazrus et al. [1983]). Wind directions tend to become more westerly with increasing elevation. At heights of 5000-10000 feet, typical of rain-producing clouds, wind directions usually range from southwest to northwest. However they may be from the northeast during intense coastal storms. Wind speeds at those elevations usually lie in the range 5-50 miles per hour, with an average of about 25 miles per hour.

2. Precipitation Type, Intensity and Duration

New Jersey receives abundant precipitation. Also, when averaged over many years, uniform amounts occur throughout the year. This precipitation can be considered to have three main origins: coastal storms, fronts, and air mass instability. Precipitation from coastal storms occurs primarily in the fall and winter, is generally widespread, and may last for one or two days. The air that reaches New Jersey during these storms usually has traveled for a considerable distance over water, and is not heavily laden with pollutants. Cold fronts and warm fronts usually arrive from the north, south or west. Air associated with frontal precipitation often originates from the area of the Gulf of Mexico, and has taken a path over the southern and/or central U.S. before it reaches New Jersey. This air may be heavily polluted. It should be noted here that the origin of the air associated with a given storm or front is not necessarily the same as the origin of the storm or front itself. As these weather systems move from place to place, air continuously moves through them. Frontal precipitation occurs around the year, but occurs most often in winter and spring. Air mass instability occurs when the air is heated from below or cooled from above. It is manifested in the form of showers or thundershowers. Showers that are produced by air mass instability occur most frequently in spring and summer. The air in these showers has similar origins to that found in frontal precipitation.

C. SUMMARY OF EXISTING OBSERVATIONS IN NEW JERSEY

1. Observations of Wet and Dry Deposition

Measurements of acidic deposition are usually made by collecting in a nonreactive container the wet or dry material that falls out of the air. The composition of this material is then analyzed in a laboratory. The device that is used for obtaining the samples is an automatic wet/dry collector. It consists

of two plastic containers with a movable cover whose position is controlled by a moisture sensor. When no liquid moisture is present, the container for wet deposition is covered, and dry deposition is collected. When rain begins, the moisture sensor causes the cover to move, thereby exposing the container for collecting wet deposition and closing the other container.

When measurements of acidic deposition are compared to each other, it is necessary first to ascertain the sampling procedure. In New Jersey, samples are taken on a daily, weekly, and event basis. In the case of a daily (weekly) sample, the collected material is removed for analysis at a fixed time each day (week). During the course of a day or week, significant wind shifts and other meteorological events may occur, and the collected sample represents a composite of all of that period's conditions. In event sampling, the material is removed for analysis at the conclusion of a particular event (usually an individual precipitation occurrence). Although event sampling is the greatly preferred method from the standpoint of interpretation of results, it is difficult to implement in that it is labor intensive. As a result, event sampling is currently accomplished only at the Washington Crossing State Park site. No continuous sampling of storms is conducted.

Figure 3 illustrates the current acidic precipitation monitoring sites in New Jersey and vicinity. Informal measurements have been made in New Jersey for about ten years by Dr. David Letcher at Trenton State College and by Dr. Eileen Brennan at Cook College. In 1978, the first station affiliated with the National Atmospheric Deposition Program (NADP) went into operation in New Jersey. Since that time, other sites have been established. At present, there are five monitoring stations in New Jersey at which the NADP protocol is used. Site locations in adjoining states are also shown in Figure 3. Of the New Jersey sites, three are operated by the New Jersey Department of Environmental Protection (NJDEP) and two are operated by the U.S. Department of Energy. Of the three NJDEP sites, only observations from Washington Crossing State Park are reported to NADP.

Figure 4a shows event precipitation pH recorded at Washington Crossing State Park during 1983. Some correlation exists with precipitation volume with lowest pH values generally occurring during summertime convective storms. However, within this seasonal pattern, large short-term variability exists, reflecting the differing origins of the air during precipitation events. Reiss (1982) studied the relationship between pH and the trajectory of the air from which the precipitation fell. He found that lowest pH values in New Jersey were associated with trajectories that had passed over the Ohio Valley. The same trajectories were also associated with the highest sulfate and nitrate concentrations. Figure 4b shows seasonally-averaged pH values, weighted by precipitation amount, measured at Washington Crossing State Park for 1982 and 1983. Summer rain events, in addition to being fewer in number, are also more independent of each other in the sense that they are not produced by a single long-persisting weather situation. We should, therefore, not be surprised to see variability, even in seasonally averaged pH values. This variability makes the identification of trends and cycles difficult. Similar problems occur in the analysis of ion concentrations derived from rainfall measurements.

1. WASHINGTON CROSSING STATE PARK (NJDEP)-WEEKLY & EVENT
2. MILLVILLE (NJDEP)-WEEKLY
3. ANCORA (NJDEP)-WEEKLY
4. CHESTER (USDOE)-MONTHLY
5. NEW BRUNSWICK (USDOE)-EVENT
6. MILFORD, PA. (NADP)-WEEKLY
7. WEST POINT, N.Y. (NADP)-WEEKLY
8. MANHATTAN (USDOE)
9. QUEENS (USDOE)

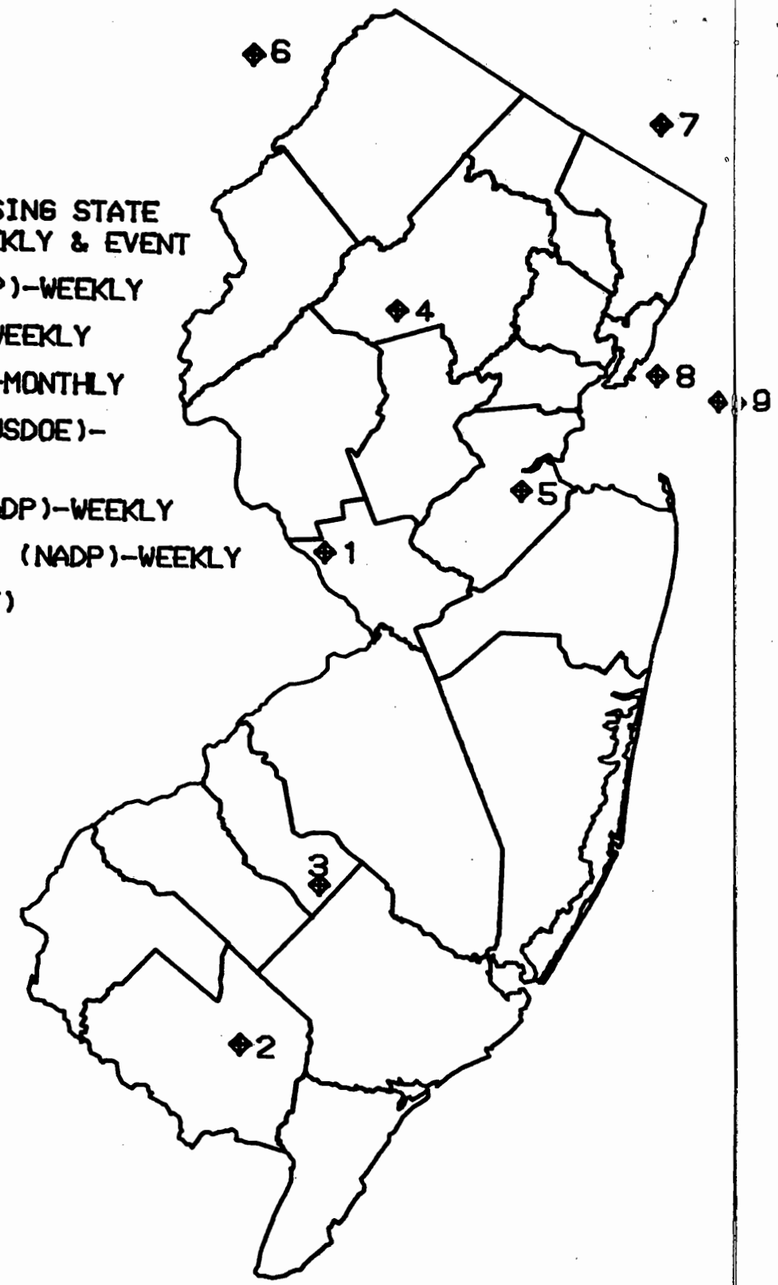


Figure 3. Acid Precipitation Monitoring Sites in New Jersey and Vicinity

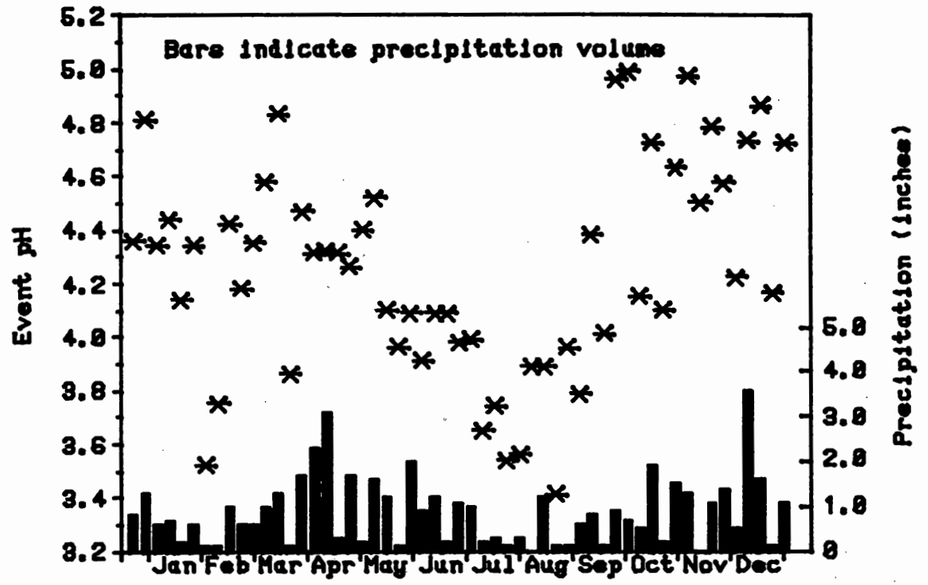


Figure 4a. Event pH and precipitation volume observed at Washington Crossing State Park during 1983.

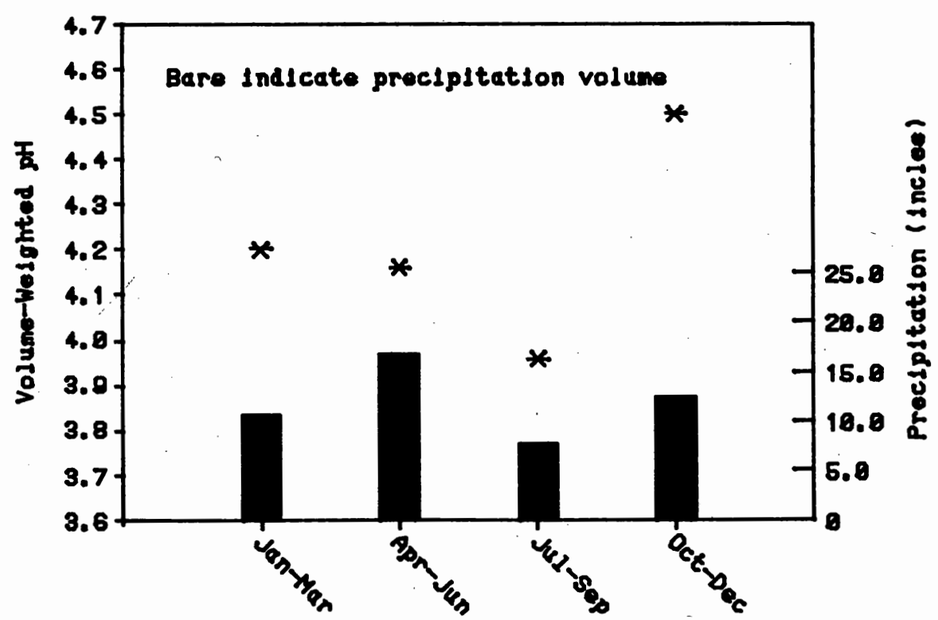


Figure 4b. Seasonally-averaged volume weighted pH and precipitation volume measured at Washington Crossing State Park. 1982 &1983

In addition to pH, an important parameter to gauge acidic deposition is to define the rate, or mass loading of acidic contamination per given area over time. Units for loading are usually expressed as kilograms/hectare/year (kg/ha/yr). In 1982, the rate at which sulfate in rainfall was deposited at Washington Crossing State Park was about 24 kg/ha/yr. This can be compared to wet sulfate loadings of 11-20 kg/ha/yr that were suggested by the U.S. - Canadian Work Group on Impact Assessment (1981) as being "protective for the majority of surface waters". In a more recent study of acidic deposition in the northeastern United States, Gorham et al. (1984) estimated that wet sulfate loadings of 14-16 kg/ha/yr would be needed to raise pH levels to the damage threshold range of 4.6-4.7. Available wet sulfate loading in the New Jersey area is summarized in Table 7.

TABLE 7

WET DEPOSITION LOADING IN NEW JERSEY AND VICINITY

	Wet Loading Kg of SO_4 ha/yr	Time Period (yr)
Manhattan	45	1980
Chester	32	1980
	37	1981
	30	1982
Millville	27	1983
West Point (New York)	25	1983
Washington Crossing	24	1983

For the time periods shown, the Chester and Manhattan sites received the highest annual deposition. This may be due to their proximity to nearby industrial areas of New Jersey and New York City or to elevated values during the 1980-82 period. It would seem fair to say that most of New Jersey receives 24-27 kg/ha/yr but that near the industrial area close to New York City, the deposition is probably 30 kg/ha/yr or more.

Gorham et al. (1984) compiled data showing the concentration of various components of wet deposition compared with sulfate deposition. Hydrogen, nitrate ammonia, and calcium ions were plotted for all NADP sites in the eastern United States. These data show that the hydrogen ion concentration is a better indicator of sulfate deposition than either the nitrate concentration or any of the other ion or combination of ion concentrations. Weekly hydrogen and nitrate ion concentration data from the Washington Crossing State Park site have been plotted in a similar way and compared with Gorham's correlations in Figures 5a and 5b. The data indicate that the composition of the wet deposition at this site is quite similar to the average composition for the eastern United States. The pH to sulfate loading relationship for 1983 is also illustrated in Figure 5a.

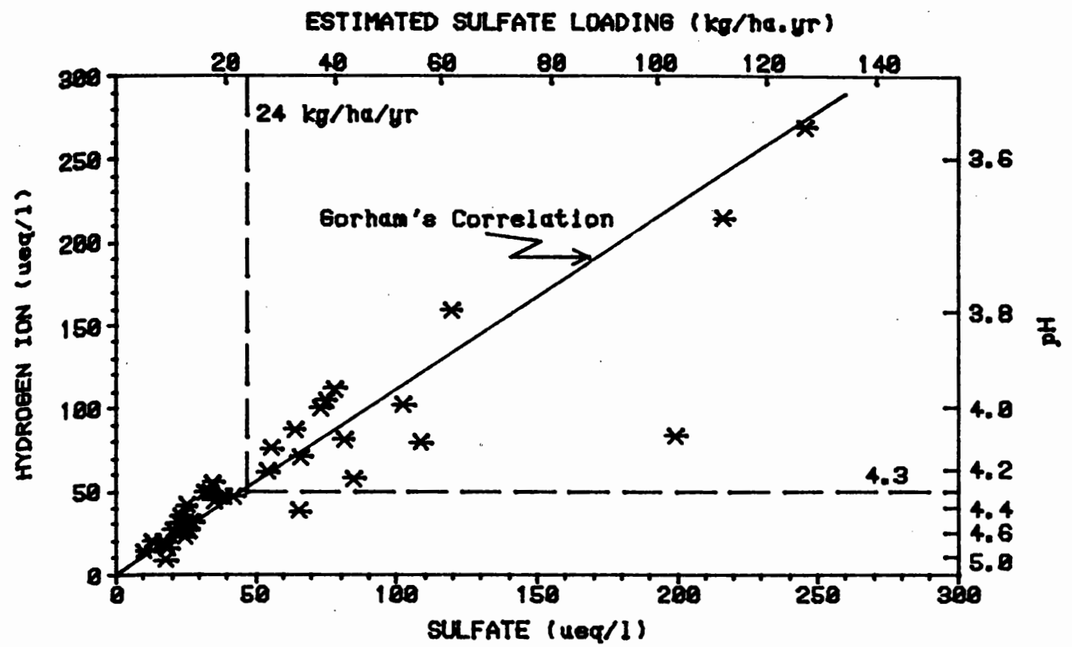


Figure 5a: Weekly hydrogen ion deposition vs sulfate deposition at Washington Crossing monitoring site. Data compared with Gorham's correlation for Eastern United States NADP sites (see Reference) and mean pH (4.3) corresponding to an estimated sulfate loading at 105 cm annual precipitation (24 kg/ha.yr)

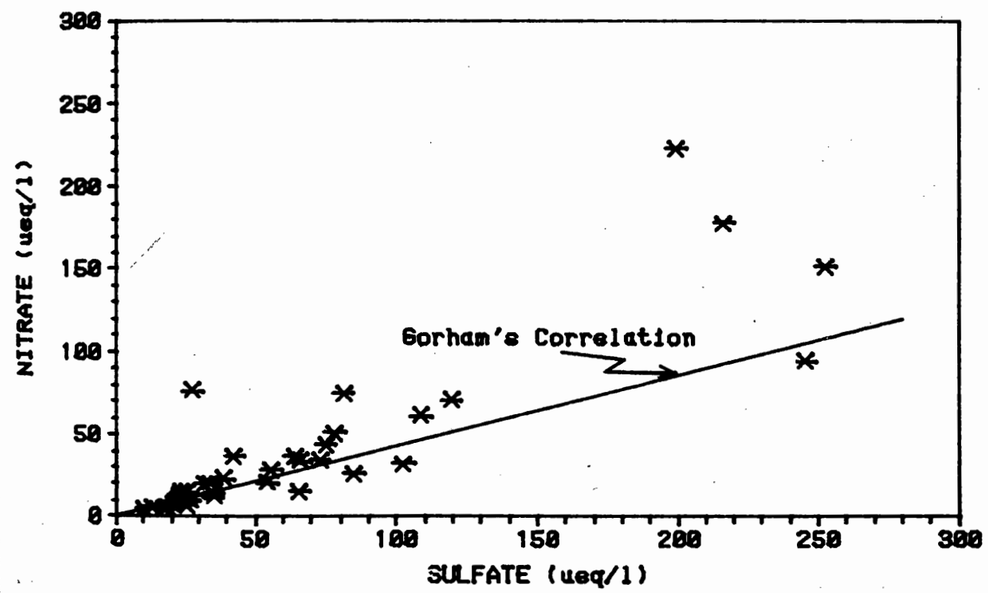


Figure 5b: Weekly nitrate ion deposition vs sulfate deposition at Washington Crossing monitoring site. Data compared with Gorham's correlation for Eastern United States NADP sites (see Reference)

Table 8 gives concentrations of other ions found in precipitation, as measured in New York City and New Jersey. In all cases, average ion concentrations at the urban New York City site are higher than those in the surrounding areas.

TABLE 8

OTHER IONS FOUND IN PRECIPITATION IN NEW YORK CITY AND NEW JERSEY (ueq/m_2)

	Manhattan (8/81-8/82)	Cook College New Brunswick, N.J. (11/81-8/82)	Chester, N.J. (9/78-9/79)
Hydrogen $(H)^+$	107	76	94
Sodium $(Na)^+$	41	28	21
Potassium $(K)^+$	2.9	2.2	2.1
Calcium $(Ca)^{++}$	42	14	12
Magnesium $(Mg)^{++}$	17	6.2	9.1
Chloride $(Cl)^-$	42	23	18

Sulfate is deposited not only in rainfall but also in dry form. Measurements of dry deposition of sulfate have been difficult because of the long exposure times needed to obtain a proper sample. Such measurements taken as part of NADP are therefore of questionable accuracy. NADP recently reduced the status of its measurements of dry deposition to a voluntary activity for its participants. It is generally believed, however, that the amount of sulfate deposited via dry deposition is of the same order of magnitude as that deposited through precipitation.

2. Other Air Quality Observations that Relate to the Acidic Deposition Question

In addition to the observations of deposition cited above, routine observations are made in New Jersey of the major precursors of acidic deposition, including sulfur dioxide (SO_2) and nitrogen oxides (NO_x), sulfate and ozone (see Table 9) . As shown in Figure 6a, SO_2 levels in New Jersey's industrial and heavily urbanized areas decreased greatly from 1969 to the mid-1970's, a period during which air quality regulations were made more stringent. Since that time, SO_2 levels have remained constant or drifted slightly higher. Outside those areas, increased stringency has largely been balanced by growth in the number of sources. Table 9 shows that SO_2 concentrations in the state are well below the National Ambient Air Quality Standards (NAAQS). It should be noted, however, that much of the SO_2 found in New Jersey's air does not originate from nearby sources. The SO_2 concentrations shown for Atlantic City in Figure 6a, for example, probably reflect the influence of more distant sources, while the excess of the Newark concentrations over those for Atlantic City represents the contribution of sources that are local to the Newark area. Seasonal variation of SO_2 is given in Figure 6b. SO_2 levels are highest during the winter months and lowest in summer. The low summer values are the result of enhanced photochemical conversion to sulfate that takes place at higher temperatures and sunshine levels.

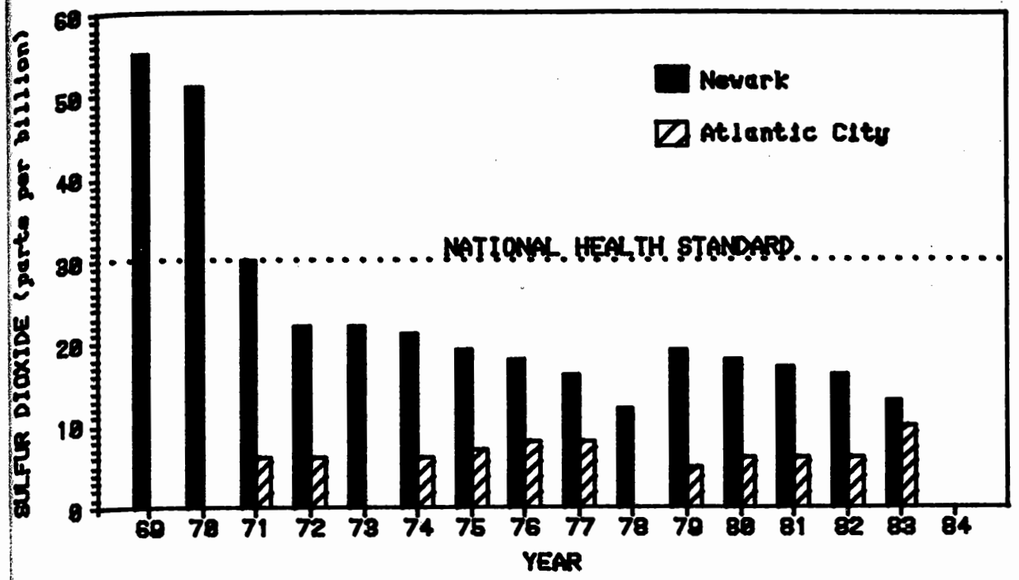


Figure 6a. Mean annual sulfur dioxide concentrations measured at Newark and Atlantic City, N.J.

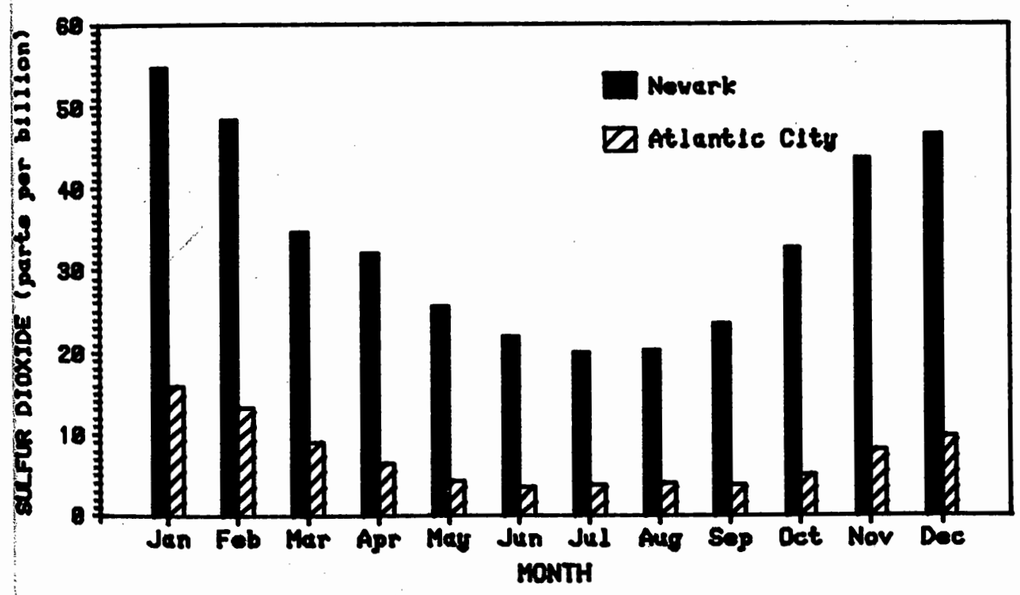


Figure 6b. Mean monthly concentration of sulfur dioxide measured in Newark (1968-1984) and in Atlantic City (1978-1984).

TABLE 9

New Jersey Air Quality Summary
1983

	Observed Concentration	NAAQS*
Sulfur Dioxide		
3-Hr., Max.	0.17 ppm	0.5 ppm
24-Hr., Max.	0.085 ppm	0.14 ppm
Annual Avg., Max.	0.011 ppm	0.03 ppm
3-Hr. Peaks, averaged over all state monitors	0.085 ppm	
24-Hr. Peaks, averaged over all state monitors	0.05 ppm	
Annual Avg., averaged over all state monitors	0.009 ppm	
Nitrogen Dioxide		
Annual Avg., Max.	0.037 ppm	0.05 ppm
Annual Avg., averaged over all state monitors	0.03 ppm	
Ozone		
1-Hr., Max.	0.28 ppm	0.12 ppm
1-Hr. Peaks, averaged over all state monitors	0.20 ppm	
Average Number of Days with Hours Above 0.12 ppm = 13		
Sulfate**		
24-Hr. Peaks	30-35 ug/m^3	
24-Hr. Means, averaged over all state monitors	10 ug/m^3	

* NAAQS is the National Ambient Air Quality Standard. The listed standard is the primary standard to protect health with an adequate margin of safety.

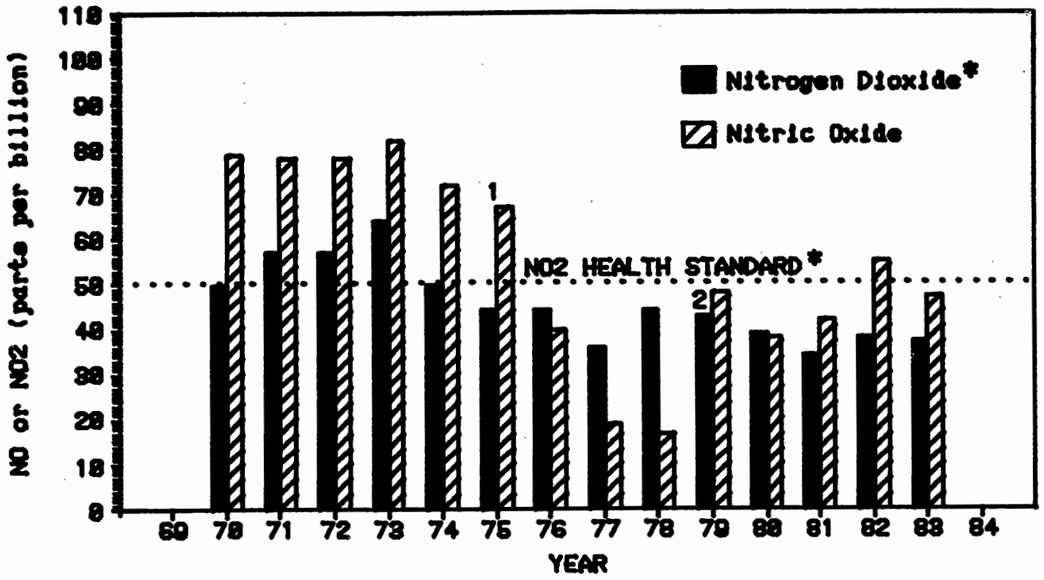
** Data is for Summer, 1981. Note, there is no NAAQS for sulfate.

Figure 7a shows how concentrations of nitric oxide (NO) and nitrogen dioxide (NO_2), the two predominant oxides of nitrogen, have varied since 1970. Again, increased stringency of air quality regulations during that period resulted in a gradual decline in concentrations, but a leveling off has occurred during the past few years. Still, NO_2 concentrations are below the NAAQS even at the most polluted sites. Figure 7b shows the seasonal behavior of NO and NO_2 concentrations. The two gases vary inversely with respect to one another, reflecting the fact that a series of photochemical processes in the atmosphere produce NO_2 at the expense of NO. These processes operate most efficiently at the higher temperatures and sunshine levels that exist during the summer months.

As mentioned earlier, no reliable measurements of dry sulfate deposition are available. However, some indication of trends in dry sulfate deposition can be obtained by examining the amount of sulfate present in suspended particulate matter. Since sulfate particles are most commonly found in the 0.1 - 1.0 micrometer size range, it would be of greatest value to examine the composition of particles whose sizes lie specifically within that range. Only recently, during the ATEOS project (Lioy et al., 1983), have such measurements been made. However, during the nine-year period, 1970-1978, analysis of sulfate and nitrate collected by high-volume samplers was conducted (National Aerometric Sampling Network). High-volume samplers collect a very wide range of particle sizes, and the mass of collected material is dominated by particles that are several micrometers or larger in size. A sample of the results of the high-volume sampler measurements which were analyzed for sulfates and nitrates are shown in Figure 8a and 8b. With the limited data available, a small but perceptible downward trend is discernable in the early 1970's followed by increases in 1977 and 1978. These data seem to parallel New Jersey's effort in reducing SO_2 emissions in the early 1970's but are over-shadowed by increasing upwind emissions thereafter. The nitrate data show a general increase throughout the period, reflecting growth in the motor vehicle population and industry. It is interesting that trends in both data sets better reflect the regional SO_2 and NO_x inventories (Table 1 and Table 4) than the New Jersey ones. This is because sulfates and nitrates in particulate form are rarely emitted directly into the atmosphere; they are generally formed as a result of chemical conversion of SO_2 and NO_x. The amount of time needed for this conversion to take place varies widely, depending on the particular source, temperature, sunshine, and the presence of other atmospheric pollutants that participate in the chemical reactions. Thus, the time lag between SO_2 and NO_x emission into the atmosphere and subsequent sulfate and nitrate formation indicates a regional contribution to New Jersey's air.

Finally, ozone is of interest because it is photochemically reactive. In the summer months high levels of ozone are sometimes found in New Jersey. As Table 9 shows, the NAAQS was exceeded on 13 days during the summer of 1983 and the average of the peak values was 0.20 ppm.

Visibility, described earlier in this report as having a strong inverse relationship to sulfate concentrations, has been routinely measured for many years by the National Weather Service. Although the measurements are semi-quantitative in nature (Reiss and Eversole, 1978), they provide perhaps the only indication as to the magnitude of sulfate concentrations in earlier decades. Figure 9 shows the nature of long-term visibility trends at Newark. Visibilities have increased as SO_2 and NO_x concentrations have decreased, indicating that



1 Station relocated-1975 2. Measurement method changed-1979

Figure 7a. Mean annual nitric oxide (NO) and nitrogen dioxide (NO2) concentrations measured at Newark, N.J.

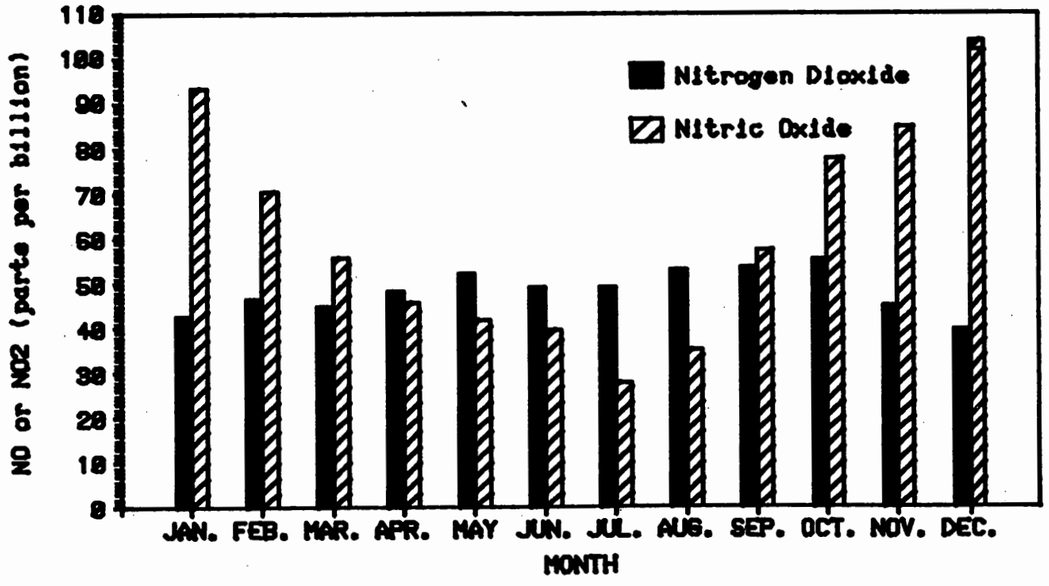


Figure 7b. Mean monthly concentrations of nitric oxide (NO) and nitrogen dioxide (NO2) measured at Newark from 1968-1984.

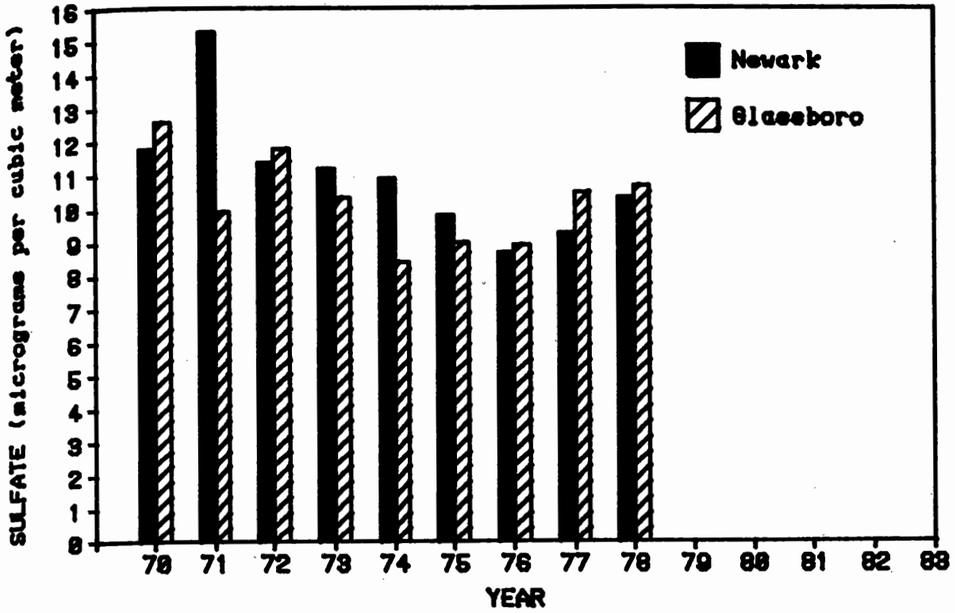


Figure 8a. Mean annual concentrations of particulate sulfate, as derived from high volume sampler observations.

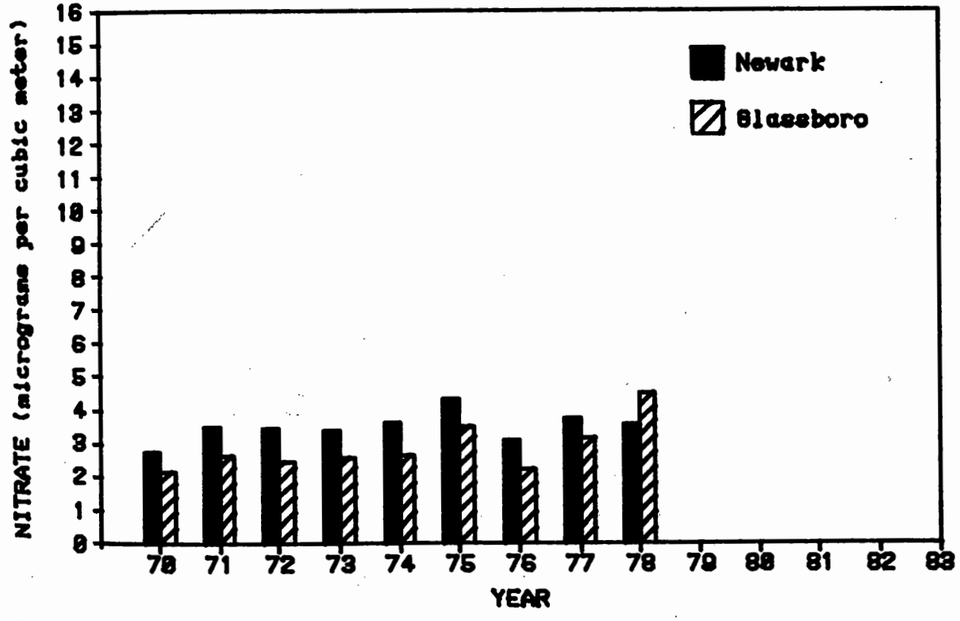


Figure 8b. Mean annual concentrations of particulate nitrate, as derived from high volume sampler observations.

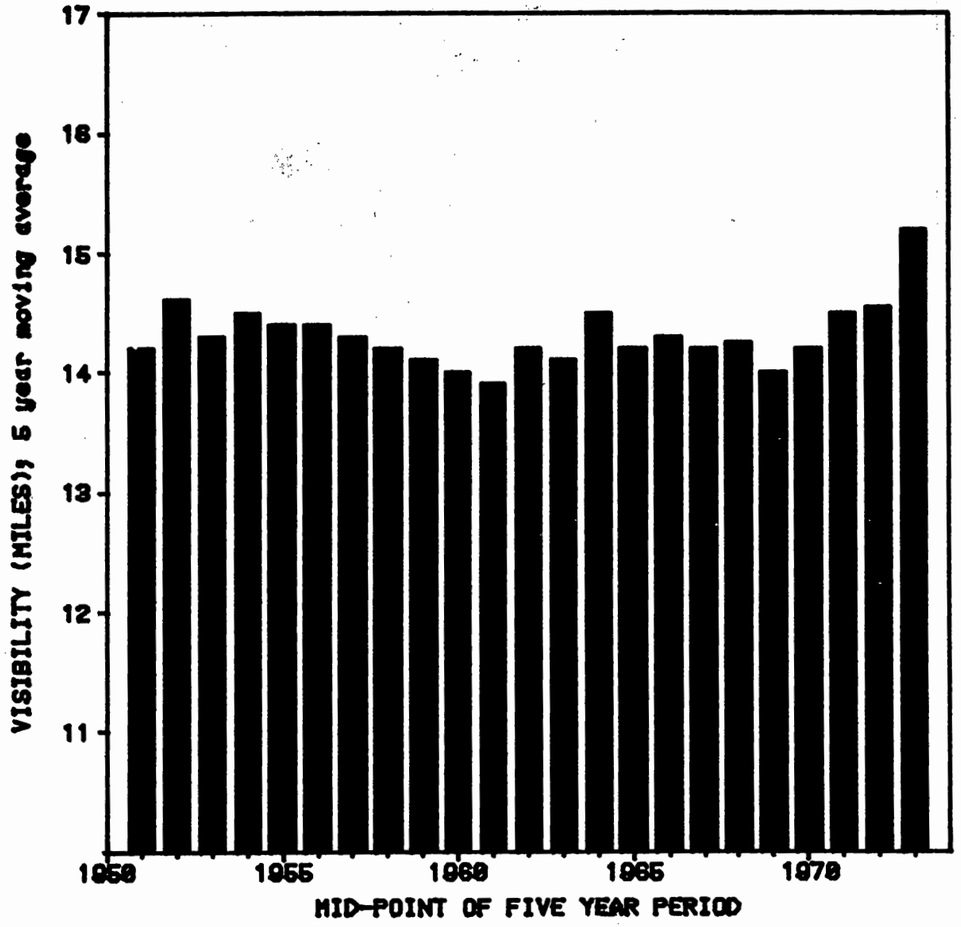


Figure 9. Visibility observations, Newark Airport. Data is normalized to 40% relative humidity.

decreases in concentrations of those precursor gases have probably resulted in decreases in sulfate concentrations. Whereas the mean annual midday visibility in New Jersey is about 15 miles, mean visibilities in some more pristine areas of the United States (e.g. Utah) are in the neighborhood of 80 miles.

D. GAPS IN OUR KNOWLEDGE OF METEOROLOGICAL ASPECTS OF ACIDIC DEPOSITION

The largest gap in our knowledge of the meteorological aspects of acid deposition seems to be in accurately specifying the three-dimensional motion of the plume. A basic problem in this regard is that routine observations of the upper atmosphere are taken only once every twelve hours at locations that are often up to several hundred miles apart. This greatly limits the accuracy of any analysis that can be done and the spatial and temporal scale of motions that can be depicted. There are also gaps in our ability to couple these models, crude as they are, to the models that depict the chemical and physical processes that take place within the plume.

Another problem in our understanding is that it has been only relatively recently that a widespread network of uniformly-calibrated observations of atmospheric deposition has come into existence. Thus, the observational data set is still very small, considering the complexity of the phenomenon that is being observed.

E. SOURCE FLUX/DEPOSITION RELATIONSHIPS

If a reduction in precipitation acidity is sought, it is of interest to determine what reduction in precursor emissions will achieve the desired precipitation acidity reduction. This question was addressed in a National Research Council report (1983), which chose to address reductions only in oxides of sulfur, rather than in those of both sulfur and nitrogen. The NRC committee was unable to find evidence contradicting a hypothesis that reductions in SO_2 emissions would lead to proportionate reductions in sulfate deposition over spatial scales of the order of a thousand miles and with time averages of a year or more. This result has been termed the "linearity hypothesis," and has been the basis for a number of calls for the reduction of sulfur oxide emissions. Because of the central role of this hypothesis in potential legislation, it is worth examining it in some detail, particularly as it pertains to New Jersey.

"Everything that goes up must come down." The key question, as pointed out by Schwartz (1983), is not whether the acidic material comes down, but where it does and in what form. The issue intrinsically involves discussions of spatial scale, since in order for an emitted molecule to contribute to rainfall acidity it must be oxidized to its acidic form, incorporated into atmospheric water droplets, and fall to the ground before being carried by the wind past the region of interest. For New Jersey, therefore, it is of interest to know whether SO_2 and NO_2 can be oxidized and incorporated in the time available between when they are emitted in the midwest and in neighboring states and when they are carried out over the Atlantic Ocean.

The data available from New Jersey measurements are not detailed enough nor long enough to use for this examination, but suitable data exist from New York, Pennsylvania, and other sites in the northeastern United States. In the summer months, chemical studies of rain show little dissolved SO_2 compared with sulfate, suggesting that the supply of oxidizers (H_2O_2, O_3, etc.) is not a limitation (Pena et al., 1982; Dana, 1984), and sulfate levels in precipitation are relatively high. In contrast, sulfate deposition at Whiteface Mountain, NY (about as far as New Jersey from the major midwest sulfur sources) in the winter is only about twenty-five percent of the summer deposition (Camerota et al., 1983), a pattern consistent with measurements throughout the northeastern United States (Dana, 1984). Some of this difference may be due to seasonal differences in meteorology, but most of it must be a result of much longer time scales (and thus spatial scales) for SO_2 oxidation during the winter months. In addition, ten to twenty percent of the dissolved sulfur in winter rain is unoxidized. Both of these statistics indicate a shortage of oxidizers in the winter.

The NRC committee based its long term "linearity" hypothesis on a correlation between SO_2 emission throughout the region east of the Mississippi River with sulfate deposition at a site in New Hampshire. This correlation could not be performed in a scientifically rigorous manner and, as a result, its validity has been contested. Lipfert (1983) states "The range of elasticities ... indicate that no conclusions on linearity or proportionality can be made." Williams (1983) says "It is clear that the decision of how to weight regional emissions in the index determines the sign and the statistical significance of linear dependence of SO_4^{2-} deposition on SO_2 emissions," and "further analyses of SO_4^{2-} data should be done separately for the yearly records of depositions during warm and during cool seasons."

The discussion above demonstrates that the basic existence of "linearity" in the northeastern United States remains in dispute and that, in any case, non-linearities are well established for time and space scales shorter than the one year, one thousand kilometer scales considered by the NAS committee. In interpreting this hypothesis from an action standpoint, several questions can be presented and discussed:

1. Does more recent evidence substantiate this hypothesis?

   There is no significant new evidence on distance scales as large as a thousand miles or time scales as long as a year. On shorter distance and time scales, however, a number of new results are of interest.

2. What are the effects when shorter time scales are considered?

   Evidence from several sources indicates that strong seasonal dependences exist in the S(IV) (i.e., sulfur dioxide) to S(VI) (i.e., sulfate) transition. In summer the transformation seems to occur within a few hundred kilometers if clouds are present (Pena et al. 1982; Dana, 1984). In winter, 10-20% of the S(IV) in rain is unconverted over distance scales of several hundred kilometers (Pena et al. 1982; Guiang et al. 1984; Dana, 1984), probably because of a shortage of oxidizers (Chameides, 1984; Seigneur et al., 1984).

Seasonal patterns in the precipitation chemistry are important because many of the potential effects of acidic deposition are seasonally dependent. For example, some lakes receive much of their input as a result of spring snowmelt. The chemical characteristics of the snow, which are established by winter deposition, are thus very important. Where growth of vegetation is involved (trees, crops, etc.), the precipitation chemistry during the growing season may be important. Thus, the winter and spring periods which appear potentially crucial are also the periods when atmospheric oxidation over distances of several hundred kilometers is only partly effective.

3. What are the effects when shorter space scales are considered?

   Even under summer daylight conditions, the conversion of S(IV) to S(VI) takes a number of hours, depending partly on cloud conditions, and is a strong function of the interplay of concentrations of different emittants (Pena et al., 1982; NRC, 1983, p. 141). In an admittedly preliminary model study, Seigneur et al. (1984) find that "for the conditions considered, reductions in SO_2 and NO_x ...lead to significantly less reductions in acid levels in the presence of clouds." In a typical result, on time scales of ten to fifteen hours (distance scales of 400-600 km), a reduction of 50% in SO_2 emissions produced only about a 35% reduction in droplet sulfate.

4. Is sulfate what one wishes to reduce or is acidity what one wishes to reduce?

   Although sulfuric acid is, on average, the primary source of precipitation acidity, other acids play non-negligible roles. In summer, sulfuric acid contributes about 70% of the acidity at Hubbard Brook, NH, for example (Galloway and Likens, 1981). Thus, a 50% reduction in SO_2 emissions, even if reflected proportionally in precipitation sulfate, would reduce acidity by about 35%. In the winter, nitric acid is proportionally more important: in this season a 50% SO_2 reduction, if reflected linearly in the droplets, would reduce the precipitation acidity by about 20%. It is therefore very important to determine whether the deleterious effects of acidic precipitation are due primarily to hydrogen ions or sulfate ions. As seen in Figure 1, precipitation in New Jersey is generally more acidic in the summer months.

5. What are the effects of other atmospheric contaminants?

   Many of the impacts proposed for atmospheric precipitation involve, as well, pollutants other than sulfate. For example, ozone is thought to be involved in forest damage (Cowling, 1984) and in materials degradation (Graedel et al., 1984). Cowling cautions that it would be foolhardy to believe that the acid rain/air pollution problem will be solved once lower sulfur emissions are mandated. What is needed, he says, is "an integrated policy involving controls on emissions of sulfur dioxide, oxides of nitrogen, volatile organic compounds, and heavy metals."

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CHAPTER II: TERRESTRIAL PROCESSES

Soils -- Dr. J. C. F. Tedrow

Forest Vegetation -- Dr. Joan Ehrenfeld

Production Agriculture -- Dr. Eileen Brennan

EFFECTS OF ACIDIC DEPOSITION ON SOILS

A. INTRODUCTION

Acidic deposition upon reaching the land surface, has the potential for inducing chemical, physical and biological changes within the soil body which, in turn, may affect the entire ecosystem including changes in plant and animal populations, plant growth, and groundwater composition, among others. Most of the state has always been under an intense, natural leaching environment - one which has induced impoverished chemical levels within a majority of soils. This impoverishment is well exemplified by the gray-colored soils of the Pinelands as well as in isolated locations in Kittatinny Mountain. There are, however, other extremes as exemplified in the limestone valleys of Warren and Sussex Counties in which the soils have a much greater potential for withstanding possible negative effects of acidic deposition.

Before addressing the problem of acidic deposition and soils, it is first necessary to define the term soil. Engineers, agronomists, geologists and others tend to have their own concepts as to the term soil. For the purposes of this discussion, soil is defined as the natural material at the earth's surface (which is generally 2 to 5 feet deep but may range between extremes of a few inches to 8 or more feet) which has been altered and transformed from parent material to a naturally generated body. Figure 10 shows a vertical section of an idealized soil with an organic surface layer underlain by various layers (horizons) which grade downward to the underlying bedrock.

In appraising the long-term effects brought about by acidic deposition, soil investigators have relied heavily on the following criteria: a) cation exchange of the soil, including cation exchange capacity (CEC) and saturating cations of the exchange complex; b) degree of acidity (pH level) of the soil which, indirectly, is a reflection of the ratio of certain saturating cations; c) nature of the organic litter where the forest cover remains intact; d) anion exchange; and e) other factors, including the trace element status, general chemical composition, mineral composition, particle-size distribution, soil structure, soil drainage, and the microbiological activity of the soil.

The problem of possible negative effects of acid precipitation on the soil system has been discussed peripherally for some years but there have been few in-depth studies. Likens et al. (1979) and others reviewed the overall question of acid precipitation and showed that in recent decades there has been increasing acidity over wide areas. While there have been some factual statements concerning acid precipitation on plants, fish population, etc., the overall effects of acid precipitation on soil remain more uncertain. Overrein, Seip and Tollan (1980) state "a number of studies have yielded evidence in support of the theoretical assumption that acid precipitation like any other change in climate may result in changes in the properties of soil e.g. decreases in pH and base saturation. At present it is difficult, however, to draw any definite conclusions on the time required for the reactions and intensities." The Soil Science Society of American under the aegis of CAST (1984) published a position paper on acid precipitation. From an agricultural standpoint, the question of acid

precipitation inducing soil changes was not alarming. This report stated "short-term consequences of such losses [cations from acid precipitation] are insignificant because of the reserve supplies of cations. Chronic losses over several decades could lead to nutrient deficiencies but this has not been demonstrated."

Krug and Frink (1983b) point out that the effects of acid precipitation on soils cannot be considered in generalities alone. Instead they state that it is necessary to consider individual soil properties on a site by site basis. They also bring out the importance of regional climate on evaluating possible negative effects of acid precipitation on soils.

1. Cation Exchange (Base exchange)

Cation exchange capacity (CEC) is a chemical term used to characterize the acid neutralizing capacity of the soil. First observed in England in the 1850s, cation exchange has come to be recognized as one of the most important reactions in soils. Certain materials such as clays and organic matter (humus forms) have high cation exchange capacities but sands tend to have extremely low values (Table 10).

Cation exchange capacity is loosely defined as the sum of the cations that the exchange mechanism of the soil can adsorb. Quantitatively it is generally expressed as milligram equivalents per 100 grams (meq/100g) of soil. In order to illustrate this point Figure 11 shows, in idealized form, potassium (K) existing under three different conditions. The crystalline form of potassium in silicate rock is virtually insoluble. The exchange mechanism as shown serves as the "storehouse" for potentially available potassium (as well as other ions). Potassium in solution is in a soluble and highly mobile state and can be taken up by plants or leached into the groundwater. In addition to potassium, the elements sodium (Na), calcium (Ca), magnesium (Mg), hydrogen (H), aluminum (Al) and others such as iron, manganese and the ammonium ion may also occupy the cation exchange complex. Under acid leaching Na, K, Ca and Mg tend to be replaced by H and some Al - a reaction which signifies soil impoverishment. The reaction is shown graphically in Figure 12. As leaching proceeds (going from right to left in the diagram), the Na, K, Ca and Mg on the exchange complex decrease in quantity while H and Al show a corresponding increase. If, for example, the Na, K, Ca and Mg make up 60 percent of the exchangeable cations, the soil would be 60 percent base saturated or, putting it another way, the soil would be 40 percent hydrogen (and aluminum) saturated. In depicting the leaching potential of a soil it is necessary to consider both the cation exchange capacity together with the quantity and ratio of the saturating cations.

2. Soil Acidity

The pH measurement is of critical importance in characterizing the degree of acidity of soil. The pH level will indirectly give some indication as to the degree of leaching that has occurred. It is universally expressed on a pH scale as shown in Figure 12 where, as the percentage of Na^+, K^+, Ca^{2+} and Mg^{2+} on the exchange complex decreases, there is a corresponding general decrease in pH value and vice versa.

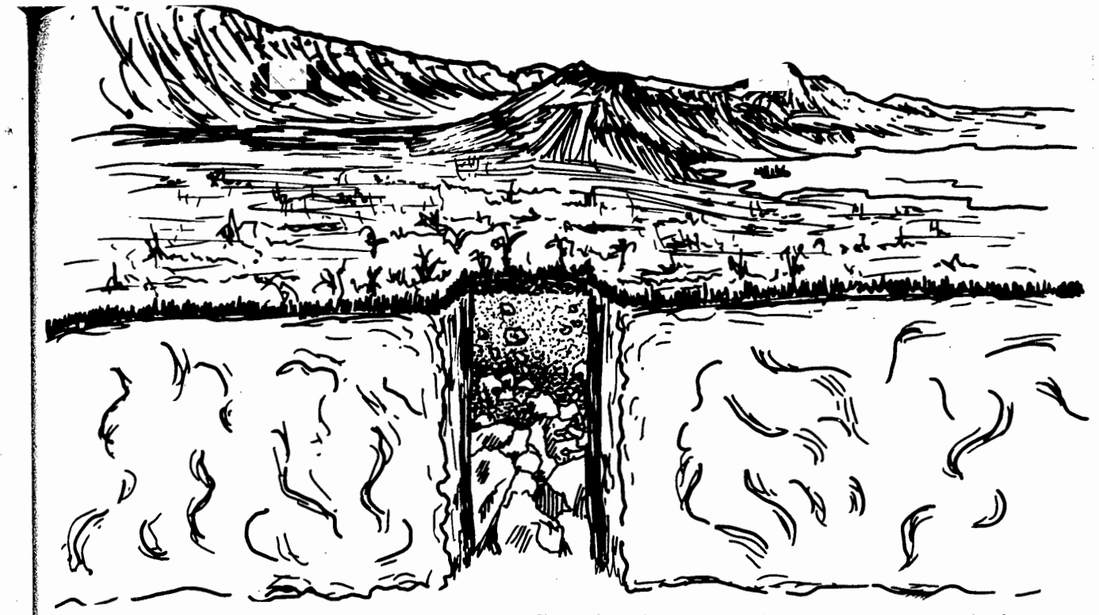


Fig. 10 Idealized diagram of a soil profile showing organic matter accumulation at the soil surface below which are various mineral soil layers and finally broken bedrock.

TABLE 10

CATION EXCHANGE CAPACITIES OF VARIOUS SOIL COMPONENTS

MATERIAL	CATION EXCHANGE CAPACITY (meq/100g)
Organic matter	100-200
Sand	1*
Silt	1-3*
Clay	
kaolinite	3-15
illite	10-30
glauconite	20-30
chlorite	10-40
vermiculite	100-150
smectite (montmorillonite)	80-150
gibbsite	nil

* Values for the sand - and silt - size particles may be much higher with greensand and shale material (Tedrow, 1966).

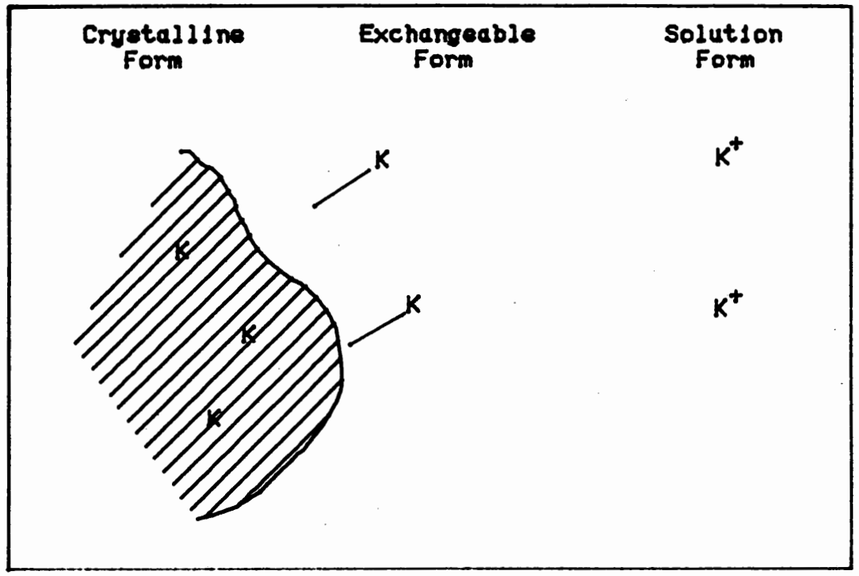


Figure 11. Three states of solubility of potassium. The left shows potassium in a near insoluble state (crystalline silicate form), the center represents exchangeable potassium on the exchange complex, and the right side indicates potassium in solution.

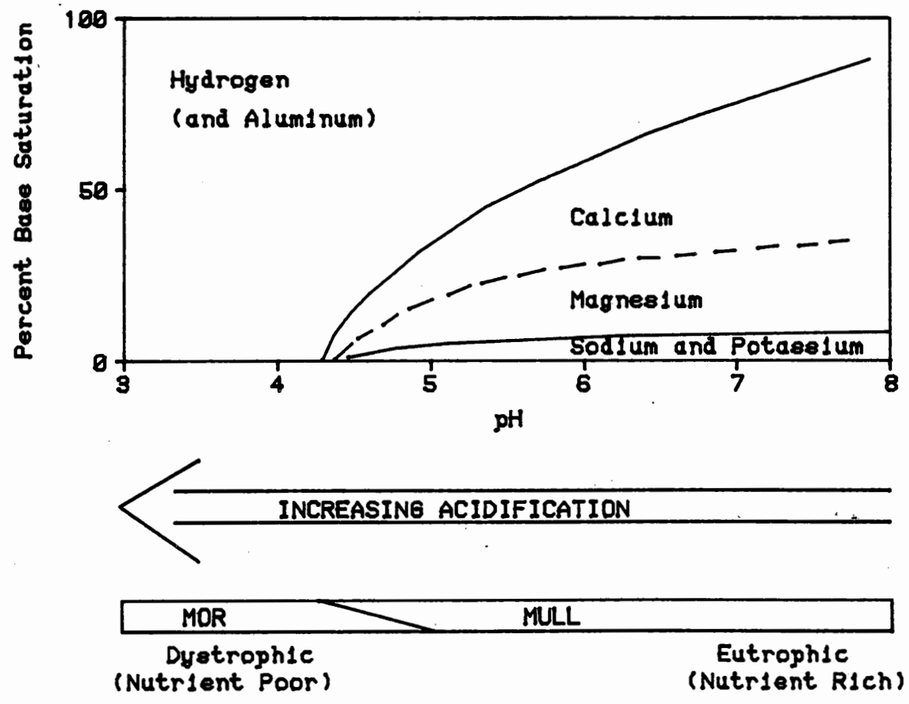


Figure 12. Diagram showing the relation of percent base saturation to pH. Also shown are the humus types in relation to acidity level.

3. Organic Litter in Forest Soils

The type of organic matter on the forest floor reflects, in part, the degree of soil acidity. A century ago in Denmark P. E. Muller found that the organic matter on the forest floor could be separated into two distinct biological types, depending upon the degree of plant nutrients in the soil matrix and the degree of humification of the forest litter (Handley, 1954). The mull type was considered to be a layer of well-decomposed forest detritus lying loosely on top of the mineral soil horizons. Such a humus type forms at the higher (although still generally acid) pH levels under high levels of trophism. When the mull type of humus is present it generally signals a high level of vital activity within the soil with a consequent high growth rate of plants. Mor, on the other hand, was characterized by Muller to be a "firm organic matter that does not give under foot." The mor type of humus is strongly acid and depauperate of plant nutrients. Figure 12 shows the type of humus form present according to soil acidity. Where there is a transition between mull and mor it is generally designated as a moder. If the pH values of the soil drop measurably from acidic deposition or other causes, it would be expected that the humus forms would change accordingly. Thus the character of humus forms can be used as indexes for possible degrading changes in soil systems. No organized studies have been made along these lines in New Jersey but casual observation over the past 35 years indicate no measurable changes in the humus forms in forested soils.

4. Anion Exchange

Soils have a capacity for adsorbing both cations (Na^+, K^+, Ca^{2+}, Mg^{2+} etc.) and anions (Cl^-, SO_4^{2-}, PO_4^{3-} etc.). Cations bear a positive charge whereas the anions bear a negative one. Anion exchange has been studied far less than has cation exchange and our knowledge of the former is more limited. Mattson (1929, 1931) and Toth (1939) demonstrated that Cl^-, SO_4^{2-} and PO_4^{3-} adsorption shows an approximate linear increase as pH values of the soils decrease. The pH effect on anion absorption has been ascribed to the neutralization of the hydroxyl groups by added cations. It should be stated, however, that these authors were dealing with low levels of saturation and, therefore, their data are of limited value in dealing with modern problems of possible excessive quantities of sulfur in the ecosystem. Mattson did speculate, however, that at low pH levels there is probably some destruction of certain mineral lattices. Wiklander (1964) pointed out that anion adsorption is also closely related to the nature of the soil colloids such as the clay mineral species, content of hydrous oxides, content of organic matter and other factors. The capacity of most soils for adsorbing anions such as SO_4^{2-}, Cl^- and NO_3^- is quite low and these ions are, for the most part, easily lost by leaching.

Kretchmer (1952) found that increasing the sulfate concentration in New Jersey soils had little, if any, effect on changing the rate of plant growth or the composition of the plants. Kretchmer too was working primarily with low levels of sulfur.

B. ACIDIC DEPOSITION ON NEW JERSEY SOILS

We now take up the problem of some probable effects of acidic deposition on New Jersey soils. With few exceptions natural pH levels in New Jersey range from about 4.0 to 6.0. From Figure 13, it is apparent that most of New Jersey virgin soils have less than 50 percent base saturation under natural conditions and many have only about 10 to 15 percent. Thus acidic precipitation with pH values of 4.0 to 4.5 alone does not always signal alarming changes within the soil. It should be emphasized, however, that a soil can still be partially depleted of bases even though the pH values of precipitation are higher than those of the soil. But the more acidic the precipitation (assuming soaking rains), the greater will be the rate of impoverishment of the soil.

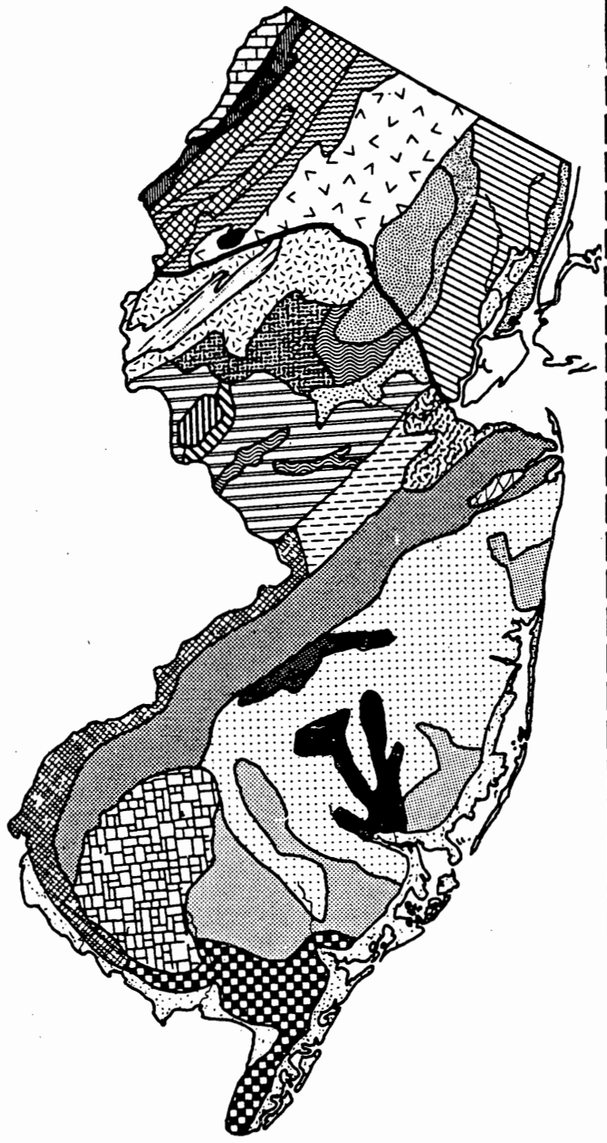
We now consider potential changes in New Jersey soil properties resulting from acidic deposition. Since only a few preliminary studies have been made on this complex subject, most of the following information is professional judgment. Figure 13 shows a map of New Jersey with 28 different kinds of soil.

Very Acid Soils:

This group includes soils with a natural pH range of about 4.0 to 4.2 - the most acidic soils of New Jersey. The major soils are Cattaraugus-Swartswood of the Kittatinny Mountain, Rockaway of Morris and Passaic Counties, Lakewood of the Pinelands uplands, St. Johns (a.k.a. Berryland) of the swamps within the Pinelands, Aura of the Salem-Gloucester Counties area and Colts Neck of the red hills of Monmouth County. This group of soils constitutes the most impoverished sites in New Jersey. The soils, already in a depauperate state, will probably undergo few additional changes under conditions as they are now known. These soils have low acid neutralizing capacities (low CECs [discounting the swampy St. Johns soil]) but acidic deposition may induce still greater acidity in the nearby lakes and streams as well as ground water. Krug and Frink (1983) have pointed out that podzolic soils (ashy gray-colored soils), such as the Cattaraugus-Swartswood and the Lakewood, have long been subjected to a strongly acid environment. They state "acid rain is widely believed responsible for acidifying soil and waters of North America and Northern Europe. However, factors commonly considered to make landscapes susceptible to acidification by acid rain, are the same factors long known to strongly acidify soils through the natural processes of soil formation." Joffe (1931) outlined the natural depletion of nutrients in such soils as listed above. Jordan (1966) found that the Lakewood soil had a low pH and, through the use of lysimeters, established that the bases on the exchange complex were low in quantity and were rapidly removed from the soil by natural leaching processes.

Moderately acid soils formed on carbonate rock:

This group of soils should be the least susceptible to acidic deposition. The group is made up of the Wallpack, Palmyra - Squires and Washington soils, all located in northwestern New Jersey. Even though these soils do not always have the highest pH values at the surface, they contain carbonate rock and are alkaline at depth. Therefore, as drainage waters pass downward through the soil,



SOIL NAME	Parent material	Dominant texture	Approx. pH level of original soil	Cation Exchange capacity	Percolation rate	Special soil properties
Wallpack	shale & limestone★	silt loam	5.5	medium	moderate	Limestone at 24 in.
Cattaraugus–Swartswood undifferentiated	acid sandstone★	stony loam	4.0	medium-low	moderate	Mor type humus
Nassau–Dutchess–Cossayuna undifferentiated	shale & limestone★	shale loam	4.5	medium	moderate	Limestone at some sites
Palmyra–Squires undifferentiated	limestone★	silt loam	4.5	medium	moderate	Limestone at 24 in.
Rockaway	gneiss★	stony loam	4.2	medium	moderate	–
Wethersfield	shale★	gravelly loam	4.5	medium	moderate	–
Holyoke	basalt & diabase★	stony silt loam	5.0	medium	moderate	–
Muck	– ★	–	6.0	very high	low	Organic
Whippany	lake deposits★	silty clay loam	6.8	high	very low	Poorly drained silts
Dunellen	shale★	sandy loam	4.5	medium-low	rapid	–
Annandale	gneiss★	gravelly loam	4.5	high	moderate	–
Washington	limestone★	loam	5.2	medium-high	moderate	Limestone at depth
Norton	shale	loam	4.8	high	moderate	–
Penn	shale	silt loam	4.6	high	moderate	Shale at 24 in.
Montalto	basalt & diabase	stony silt loam	5.5	medium-high	moderate	–
Lansdale	sandstone	silty clay loam	4.8	medium-high	moderate	–
Croton	argillite	silt loam	4.5	medium-high	low	Poorly drained silts
Sassafras	sands+	sandy loam	4.8	medium	moderate	–
Sassafras–Hammonton Phase	sands+	loamy sand	4.8	medium-low	rapid	–
Sassafras–Cape May Phase	sands+	loamy sand	4.5	medium-low	rapid	Water table at 2-3 ft.
Sassafras–Keyport undifferentiated	sand & silt+	loam	5.0	medium	moderate	Some poor drainage
Aura	sand & gravel+	gravelly sandy loam	4.2	medium-low	rapid	–
Greenwich	silt & sand+	silt loam		medium	moderate	–
Freehold–Collington undifferentiated	green sands+	sandy loam		medium-high	moderate	Glauconite soil
Lakewood	gray sands+	sand	4.2	very low	very high	Very dry sands
St. Johns	sands+	sand	4.0	variable	moderate	Water table at surface
Colts Neck	red sands+	sandy loam	4.2	low	rapid	–
Tidal Marshes	–	–	6.0/7.5	very high	–	Tidal marsh & sandbars

★glacial material +coastal plain material

Fig. 13 The major soils of New Jersey. The map is from Tedrow (1962) as redrawn by B. Robichaud and M. F. Buell in Vegetation of New Jersey. 1973. Rutgers University Press.

the base formers (Ca and Mg particularly) will be solubilized and the acids will be largely neutralized. For practical purposes these soils have a nearly inexhaustable supply of calcium and magnesium for neutralizing the groundwater. Toth and Ott (1969) found that quantities of calcium and magnesium in the surface waters from the limestone areas were much higher than in the case of acid materials. They were even able to differentiate limestone (calcitic) from dolomite areas by recording the calcium and magnesium contents of the drainage waters.

Moderately to strongly acid soils:

This is the largest area of soils within the state. Rather than a group of soils having similar properties in common, this group constitutes a residue of soils after the very acid soils and the moderately acid soils have been segregated. This group, therefore, includes many soil varieties ranging from shallow, rocky soils of the northern mountains to the deep, sandy soils of southern New Jersey. It is believed that this group has greatest potential for some negative changes if subjected to acidic deposition for extended periods. But even within this group there are variations and special situations relative to possible soil degradation. For example, the glauconite-bearing soils have higher cation exchange capacities than the nonglauconite sands. Also included with this group are clay deposits and peats with relatively high exchange capacities.

In order to show the wide range of soil properties in New Jersey, Table 11 has been compiled. Hazen soil has a relatively high base saturation and pH values. On the other hand the Lakewood, Rockaway and Swartswood soils are among the most acid and depauperate in the state.

1. Acidic Deposition in Agricultural Areas

Of the 4,813,440 acres of land in New Jersey, about 676,000 are harvested cropland, 59,000 are in pastured cropland and 127,000 are in permanent pasture. Nearly all of the 862,000 acres of cropland plus pasture in the state receive applications of lime and fertilizer. Recommendations from the New Jersey Agricultural Experiment Station generally focus on maintaining agricultural soils at a pH of 6.0 to 6.5 in New Jersey. During the 1980-1982 period, yearly purchases of agricultural liming materials averaged 168,000 tons (NJ DOA, 1983). This value also includes lawn and garden sales. With this quantity of lime being applied to cultivated and pastured areas, it is assumed that acidic deposition would have few, if any, negative effects on such lands. But liming the soil will not remove the possibility of acidic deposition causing damage to aboveground plant parts.

In addition to liming materials there were about 198,000 tons (NJ DOA, 1983) of fertilizer and fertilizer materials sold within the state in 1983. It is calculated that from that amount there was an equivalent of 250 tons of elemental sulfur in the fertilizer. Further, there are about 1000 tons of gypsum ($CaSO_4 2H_2O$) applied to New Jersey land each year, mainly for improving soil drainage. While gypsum and conventional fertilizers are in a near neutral state, this does not address the problem of the ultimate fate of the applied sulfur. Sulfur materials themselves are generally slightly acid in reaction but the potential for increased acidity is compensated for by additions of limestone.

TABLE 11

Selected chemical properties of some New Jersey soils. Hazen typifies the moderately acid soils formed on carbonate rock. Lakewood, Rockaway and Swartswood exemplify the very acid soils.

	Hazen gravelly loam			Lakewood sand			Rockaway stony loam	Swartswood stony loam
	Well-drained limestone drift in Sussex County			Dry, Gray sands of the Pine Barrens			Drift of the northern Highlands	Drift of the Kittatinny Mt.
	pH	CEC.[1] (me/100g)	Base Sat.[2] (%)	pH	CEC.[1] (me/100g)	Base Sat.[2] (%)	pH	pH
Topsoil[3]	5.5	10	50	4.2	3	10	4.2	4.0
Subsoil[3]	6.0	6	60	4.4	2	8	4.2	4.8
Parent Material	7.5	5	100+	4.6	2	8	4.2	4.8

[1]/Cation Exchange Capacity.

[2]/Percent base saturation.

[3]/The topsoil is generally up to 1 foot in thickness. The subsoil is generally 1 to 1½ feet thick.

This panel, focusing on acidic deposition, has been concerned about the effects of increased quantities of sulfur on the ecosystem. This view is proper because with "high" concentrations of sulfur there may be negative reactions in the ecosystem but, on the other hand, small quantities of sulfur are needed for plant and animal growth. Sulfur plays an important function in plant and animal metabolism. Many soil areas of the world are actually listed as being sulfur deficient. Cultivated soils in humid areas such as New Jersey generally have 100 to 1500 ppm of sulfate present. Kretchmer (1952) found that sulfur applications of up to 30 meq/100g of soil did not have a depressing effect on crop yields.

2. Soils in Forested Areas

Approximately 40 percent of New Jersey land is in forest, excluding urban areas with tree cover. Since forested areas rarely receive applications of lime, some may be subjected to a degree of potential damage from acidic deposition.

3. Wind-Blown Cement Dust

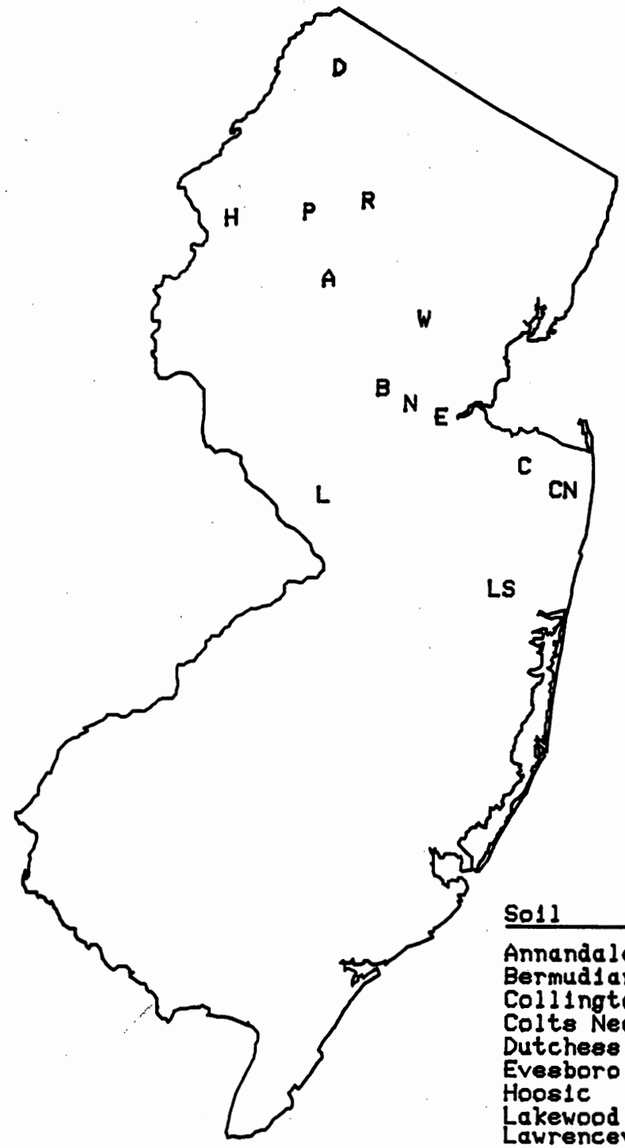
In southwestern Warren County and nearby areas there have been considerable quantities of dust blown over the landscape from former local cement plants. This ground cement and limestone dust has had a neutralizing effect on the soil acids and acidic deposition should impart no adverse effects to the soils in these areas for many years (Clucas and Tedrow, 1957).

4. Soil pH Measurements Over Time

Only in the last few decades has there been a "public-awareness" of acid rain. Changing populations of animals (including aquatics) and plants, changes in water chemistry and other possibilities have been discussed. The problem of possible changes in soil pH has not, however, been so highly evident except in extreme cases. Profs. S. D. Faust, A. McIntosh, S. Toth and T. McNevin of Rutgers University (unpublished manuscript, 1984) have investigated possible pH changes in New Jersey soils over the past 35-year period.

Soil samples were collected from 13 soils throughout northern New Jersey (Figure 14) and were analyzed by major horizon (A & B) for pH, cation exchange capacity (CEC), exchangeable bases (Ca, Mg, K, Na), and heavy metals (Fe, Cu, Ni, Zn, Mn, Cr, Pb, & Cd). Soil excavations were conducted as close as possible to the original sampling sites of 1945-1947. Comparisons were made between the analytical values of the new (1982-1983) and the originals.

Based on the old records, the new samples were obtained at locations as close as possible to the original sampled sites. PH, exchangeable bases and heavy metals were examined to determine if the New Jersey soils had been amended in these important characteristics. No significant difference was noted in the pH values of the new samples and the value of the stored soils excavated in the 1945-1947 period. Some soils appeared to have lost small amounts Ca, K and Mg but other soils appeared to have gained slightly in these elements. No over all trend existed suggesting that at least for these soils no net leaching of Ca, K,



Soil	Code	Characteristic
Annandale	A	Road cut
Bermudian	B	Field
Collington	C	Wooded roadside
Colts Neck	CN	Field
Dutchess	D	Cultivated field
Evesboro	E	Road cut
Hoosic	H	Road cut
Lakewood	LS	Wooded roadside
Lawrenceville	L	Wooded area
Norton	N	Wooded roadside
Palmyra	P	Cultivated field
Rockaway	R	Road cut
Whippany	W	Field

Figure 14. Location of soil sample sites within New Jersey for pH study. (Faust, et al, 1983)

and Mg occurred. Likewise, no trend was evident in the heavy metals. Because of the limited number of samples, and the uncertainty of the exact soil excavation sites, the significance of the results is not as high as would be desireable.

C. SOME NATURAL PROBLEMS IN MEASURING pH CHANGES IN SOILS

Soil chemistry is a very complex field which involves a multiplicity of reactions. Unlike grinding up a mineral sample and determining its pH value, the soil has natural seasonal ranges of pH values. Therefore, making projections by comparing pH changes in soils over a short or long time period produces many uncertainties. This was first brought out by Hermerling [a.k.a. Gemmerling] (1927), who found wide variations in pH values of soils, depending upon the time of the year the sampling was done. In order to expand on this problem of comparative soil pH studies, Ugolini (1964) followed pH changes on a forested site of the red shale soils near East Millstone, New Jersey. Table 12 shows the variations recorded over a 1-year period. The uppermost horizon at this one site was found to vary as much as 0.9 pH unit during the year.

TABLE 12

SOIL PH VALUES AT VARIOUS DEPTHS AND TIME INTERVALS 1957-1958*.
DATA FROM A RED SHALE SOIL AT EAST MILLSTONE, NEW JERSEY.

Depth	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	Aug	Aug.
A	5.1	4.8	4.9	4.8	5.1	4.8	4.4	4.5	4.5	5.3	4.8	4.7	4.7
A	5.6	5.2	5.2	5.0	5.9	5.6	5.3	5.2	5.3	5.0	5.4	5.2	5.2
0-3"	4.8	4.6	4.8	5.0	5.6	5.3	4.6	5.0	5.0	5.0	5.2	5.0	5.0
3-6"	5.0	4.7	4.7	5.0	5.6	5.2	4.9	4.8	5.0	5.0	5.3	4.8	4.8
6-12"	5.0	4.7	5.1	5.0	5.4	5.0	4.9	4.9	4.9	4.8	5.2	4.8	4.8
15"	4.9	4.5	5.1	5.0	5.4	4.9	4.7	4.7	4.9	4.7	5.2	4.9	4.9
20"	5.0	4.5	5.0	4.9	5.4	5.8	4.7	4.7	4.8	4.6	5.2	4.8	4.8

*Data of Ugolini (1964).

D. HEAVY METALS

The nature of the parent material is a major factor in determining the quantities of heavy metals present in soils. The earlier soils literature focused mainly on deficiencies from a standpoint of crop growth and, accordingly, there was little information regarding excessive and/or toxic levels in soils except near smelters, ore dumps and coal-burning furnaces.

Table 13 gives some approximations of heavy metal contents in selected New Jersey soils. There is a tendency for a slight concentration of heavy metals in the clay fraction as compared to the total soil. The lead concentrations in the Squires and Annandale samples for total soil are higher than in the clay fraction.

While there have been dramatic increases of heavy metals in the soils from certain locations within the state, it appears that most of these increases have been from industrial sources including sewage systems. For example, soils along the lower Hackensack and Passaic Rivers have extremely high concentrations of heavy metals.

Overall if the objective is at least to partially stabilize certain heavy metals in the soil, those with a high cation exchange capacity would have more possibilities (Griffin et al., 1977). Further, it has been shown that there is an increase in solubility of heavy metals at lower pH levels in the soils. Therefore, soils with higher pH levels and greater acid neutralization capacities should contribute lower amounts of heavy metals to streams and aquifers. But this is a short-term expediency rather than a permanent solution to the problem. It should be emphasized, however, that characterizing the complex heavy metal reactions within the soil is still quite speculative.

TABLE 13

HEAVY METAL CONTENT OF SOME SOILS (TOPSOILS) FROM NEW JERSEY
(CONNOR ET AL., 1957)

	Cu*	Mn	Zn	Ni	Co	Sn	Pb	Cr	V	Ga
					ppm					
LIMESTONE SOIL (SQUIRES)										
Total Soil	13	121	169	18	3	2	138	22	56	8
Clay	56	156	318	45	9	1	30	83	119	28
GNEISSIC SOIL (ANNANDALE)										
Total Soil	17	106	320	30	4	14	155	28	70	16
Clay	79	125	249	129	14	1	26	34	122	37
RED SHALE SOIL (NORTON)										
Total Soil	30	520	86	38	5	12	30	20	55	8
Clay	69	500	440	121	10	80	44	167	193	32

* Cu - copper
Mn - manganese
Zn - zinc
Ni - nickel
Co - cobalt
Sn - tin
Pb - lead
Cr - chromium
V - vanadium
Ga - gallium

EFFECTS OF ACIDIC DEPOSITION ON FOREST VEGETATION

A. INTRODUCTION

Forest vegetation is potentially affected by acidic deposition through a large number of pathways, including direct and indirect effects of wet and dry atmospheric deposition on the leaves and stems, direct effects on root processes, and indirect effects on soil chemistry and microbiology. Furthermore, acidic deposition may affect ecosystem processes within forests separately from the trees; changes in ecosystem processes not only will impinge on tree growth, but will also affect other aspects of ecosystem function. Because of this complexity of interactions of forest vegetation with acidic deposition, it is extremely difficult to identify the mechanism(s) by which acidic deposition affects forests, and to make predictions about the potential for damage in New Jersey.

Concern about the potential impact of acidic deposition on forest vegetation has been generated because extrapolation from known and theoretically expected effects on soil chemistry and soil nutrient status suggests the likelihood of damage, and because there have been increasingly frequent observations of extensive, unexplained decline and death of trees in both North America and Europe. In the attempt to unravel the myriad possible effects of acidic deposition, numerous studies have been undertaken involving laboratory applications of simulated acid rain, field experiments with simulated rain or acidified irrigation water, and correlation of field-derived data with environmental parameters. The most comprehensive, long-term studies of the interaction of acid deposition with forests have been conducted in Norway and Sweden; these studies have involved both analysis of field conditions and experimental manipulations (Overrein et al., 1980; SMA, 1982). Despite the wealth of data from these various sources, it still remains difficult to assess the current impact of acid deposition on forests, and even more difficult to predict future trends. These difficulties derive not only from the complexity of the interactions involved, but also from the considerable uncertainty of extrapolating from experimental conditions of very low pH to ambient pH levels, from laboratory simulations to field conditions, from studies of single response variables to ecosystems of great complexity, from experimental effects on seedlings to effects on mature trees, and from models based on limited quantitative data for a small number of parameters.

It is particularly difficult to use the existing data to assess current and potential damage to New Jersey forests. There are inadequate baseline data on forest tree growth and mortality in the state to establish trends, or to disentangle the separate effects of air pollution, gypsy moth damage, drought, and other stresses. One recent study (Johnson et al. 1981) presented data suggesting a recent sharp decrease in the growth of pitch pines, shortleaf and loblolly in the Pinelands; however, the generality of the phenomenon and its relationship to acidic deposition stress are, at present, unknown. Furthermore, most of the published data on tree response to acidic deposition concern the growth of conifers but most of New Jersey's trees are hardwoods (primarily oaks). Therefore, an assessment of the threat of acidic deposition to New Jersey's forests must rest on extrapolation from data obtained in other areas, (particularly the

extensive work in Scandinavia and Germany), and on other types of forest communities.

In order to assess the potential of acidic deposition to damage the health of New Jersey's forests, it is first necessary to outline the pathways by which effects may occur, and the evidence that particular effects are important. This information can then be used to assess possible risks to New Jersey's forests. The following survey of potential impacts of acidic deposition on forest ecosystems simply outlines the multiple pathways without attempting to review evidence for detailed mechanisms.

B. REVIEW OF THE EFFECTS OF ACIDIC DEPOSITION ON FOREST VEGETATION

1. Above-ground Effects

Acidic deposition interacts with leaves and stems of trees as both dry deposition of gases and particles, and wet deposition of acidic substances dissolved in rain, snow and fog. The following list outlines the direct effects and indirect effects of such deposition. (Information is from Linthurst, 1983, 1984; Hutchinson and Havas, 1980; D'Itri, 1982; and Drablos and Tollan, 1980; except as noted.)

a. Damage to leaves

There is evidence from laboratory studies that treatment with acidic solutions causes the cuticle of many, but not all species, to suffer degradation and increased rates of weathering. The degree of susceptibility to damage varies considerably among species, and is, in part, related to leaf morphology and surface structure. Virtually all studies of cuticular damage have reported that no effect is observed until the pH approaches or falls below 3.0. Foliar damage comparable to that observed in laboratory studies has been observed on trees growing near point sources of air pollution, (primarily SO_2) but there is, as yet, no good evidence of damage to field-grown plants from regional acidic deposition. There have been limited reports, from laboratory studies, of changes in chlorophyll content and in photosynthetic rate; however, such studies have been thus far confined to herbaceous plants.

b. Change in susceptibility to insects and disease

There exists an extensive literature on the effects of air pollution on plant pathogens and phytophagous insect populations; however, little of this work can be specifically related to the acidic component. The available information suggests that the effects of air pollution on the infectivity and virulence of pathogens, and the population dynamics of insects, vary greatly with the particular pest; plant susceptibility to insects and pathogens shows no general response to air pollution or increased acidity. Damage to the leaf cuticle is believed to be a critical variable, because of the importance of the cuticle's integrity and wettability in disease processes. The available literature suggests that, whereas there is not likely to be any generality in the response of insect and microbial pathogens to acidic deposition, it is nevertheless likely that many disease and insect pathogens will be affected by acidic deposition in some ways.

c. Reproductive biology: seed production, seed germination, and seedling establishment

There has been little research on flowering and seed set processes in trees with respect to acidic deposition. However, research on the effects of general air pollution has shown deleterious effects on cone development in conifers. Clearly, the confounding effects of oxidant gases cannot yet be separated from acidic components in evaluating the potential effects on seed set.

Seed germination has been studied in experimental situations with simulated acid rain. Different species show different responses: seeds of some species (e.g., yellow birch, red maple) are inhibited at low pH (3.0); others experience enhanced germination (e.g., white pine, white spruce); and others show no effect of rainfall pH (e.g., eastern hemlock). In sugar maple, simulated acidic rain had a stimulatory effect on seed germination only in nutrient-poor soils.

Seedling establishment is expected to be more sensitive to acidic conditions. Some workers have found that species (e.g., sugar maple) whose seeds are unaffected by simulated rain of pH 3.0 suffer reduced growth of the seedlings at this pH. However, the extensive studies of Wood and Borman (1974, 1977) on yellow birch and sugar maple showed that, despite extensive loss of nutrient cations through soil and leaf leaching, seedling growth showed no effect, or was enhanced even at very low pH (2.3). However, Raynal et al. (1982) found that injury of sugar maple seedlings could occur at a pH of 3.0 or less, depending on soil nutrient status; susceptibility to bacterial infection was an important variable in determining the experimental outcome. Stimulation of seedling growth was attributed to N-fertilization from the NO_3^- in the simulated rain. These results illustrate the fact that the complexity of the interactions between tree growth and the soil and atmospheric environments is sufficiently great that prediction from a simple set of assumptions is risky at best.

d. Foliar leaching

The loss of nutrient cations from leaves because of leaching by acidic precipitation is generally considered one of the most important effects on trees, and one of the most important causes of potential deleterious effects on growth and survival. It has been extensively studied in laboratory and field situations. Throughfall in general shows significant enrichment of nutrient cations (Ca, Mg, K, Na), sulfates, organic acids and other organic molecules; there is also a loss or gain of H^+, depending on species. Conifer and hardwood canopies have been found to behave quite differently; in general, throughfall from hardwood canopies shows an increase in pH over gross precipitation, while conifer canopies cause a decrease in pH. Conifers have been found to intercept more precipitation and accumulate more dry deposition from impaction of aerosols than do hardwoods; they also show, in some studies, higher cation concentrations in throughfall than deciduous species receiving the same incident precipitation. There is some evidence that foliar uptake of NH_4^+-N may enhance throughfall acidity (because the uptake process exchanges H^+ for NH_4^+). Representative changes in throughfall pH are those reported by Alcock and Morton (1981), who

found that gross precipitation of mean pH 5.8 declined to pH 4.1 under a pine canopy. Several studies have shown that throughfall that is significantly higher in pH during the growing season in a chestnut oak forest is equal to or lower than open rainfall pH during the winter (Hoffman et al., 1980). There is some evidence that, at least in hardwood canopies, there is an increase in the proportion of weak (probably organic) acids in throughfall, although total acidity is conserved (Hoffman et al., 1980).

The interaction of precipitation with leaf surfaces is particularly important in the dynamics of sulfur accumulation and deposition. Mayer and Ulrich (in Hutchinson and Havas, 1980) have stressed the importance of leaves as sulfate filters in determining both the acidification effects of precipitation and the ecosystem budgets of sulfur; they estimate that dry deposition is equivalent to wet deposition in amount. Other workers have reported similar findings, with a range of 30-40 kg/ha/yr of total sulfur deposition (90-120 kg/ha/yr as $SO_4^=$) in throughfall. The dominance of sulfate in the anion budget of throughfall precipitation may strongly affect the impact of the precipitation on soil nutrients.

In summary, there is abundant evidence that rainfall is enriched in cations as it passes through tree canopies, and that this enrichment is from a combination of cation loss from leaf tissues and washoff of dry deposition on the leaf surfaces. If cation uptake from the soil cannot balance this loss of nutrients from the leaves, there could be deleterious effects on the plant.

e. Other effects

Although effects on drought stress have frequently been postulated, there has been little work done on this subject. One study of cuticular damage in Scots pine reported a slight increase in the conductance of water through the cuticle; however, the trees in this study were subject to a point source of SO_2, rather than regional acidic deposition, and in any case, the effects observed were small.

Numerous studies have demonstrated that the heavy metal content of bolewood increments increases in trees exposed to point sources of these pollutants; recently, some studies have suggested that similar phenomena may occur in trees exposed to regional acidic precipitation. Baes and McLaughlin (1984) have presented evidence of increased Al, Fe, and other metals in shortleaf pine trees exposed only to regional air pollution, and Scherbatskoy (pers. comm.) has presented similar data for spruce and sugar maple in Vermont.

Analyses of foliar nutrient contents and root nutrient contents have suggested that loss of Ca and Mg may occur to a significant extent in trees showing stress; Al concentrations, however, are very variable, and no general patterns of increased levels of Al in roots can be found that correlate with the occurrence of tree decline or dieback. However, it is evident that analysis of nutrient and heavy metal balance in tree tissues is in its infancy, and no generalizations are yet justified.

2. Below-ground Effects

Below-ground effects include a complex array of soil and plant responses. McFee (1983) has summarized many of these (Table 14).

a. Cation leaching

The loss of nutrient cations, particularly Ca, Mg, K and Na, results from the movement of mobile sulfate and other anions in percolating soil water. The net loss of cations reflects a complex balance between soil structure and chemistry and the chemical and biological processes that determine cation exchange capacity and mineral weathering rates. Experimental studies have suggested that as the pH of the leaching solution falls, leaching increases at a faster rate than weathering, resulting in a decrease in base saturation; in some studies, experimental acidic inputs have also increased nitrification, resulting in greater internal generation of H^+. Calcium and magnesium appear most sensitive to loss. A number of authors (Abrahamsen, in Drablos and Tollan, 1980; Johnson et al., 1982) have suggested that anion mobility and sulfate fixation capacity are the critical variables in determining the sensitivity of soil to cation loss. Tomlinson (1983) pointed out that in low-pH soils, (usually considered resistant to acidic deposition) if H^+ and Al saturate the small number of cation exchange sites, cations liberated from mineralized organic matter are subject to rapid loss before they can be reabsorbed by vegetation; studies of soil percolates in northern New England and in Europe support this contention. It has also been pointed out that if sulfate is not immobilized, it can cause cation loss without a significant change in soil pH.

Although there is no data from New Jersey specifically addressing this issue, there is data from the New Jersey Pinelands on nutrient inputs and outputs from the soil. Wang (1984) estimated ecosystem nutrient budgets for a typical pine-oak upland site; his data are summarized in Table 15.

Wang's studies demonstrated that in the Pinelands, the mineral subsoil has an extremely low capacity to supply nutrient cations through weathering, and that the nutrient cation stocks in the soil are also very low, compared to ecosystems on soils with a richer complement of minerals. Nutrient cation input is derived chiefly from precipitation. The nutrient cation capital of the Pinelands ecosystems is stored primarily in the organic fractions - the living biomass and the organic matter of the forest floor. The ecosystem is highly efficient at retaining these nutrients, since water leaching through the soil contains very low quantities of cations, even after low to moderate-intensity fires (a common feature of this ecosystem). This ecosystem is thus an example of the kind of system in which cation leaching induced by anion mobility could pose a major problem: although the sulfate levels currently found in the drainage water do not seem to be causing excessive leaching at present, a saturation in the soil of anion adsorption sites could cause a rapid loss of the small cation pool available to the vegetation

Seip and Dillon (1984) have referenced a number of studies in which increased cation loss due to increased sulfate flux has been demonstrated.

TABLE 14

THE POTENTIAL IMPACT OF ATMOSPHERIC DEPOSITION ON LITTER AND SOILS

Process or property	Hypothetical impact of acidic deposition	Evidence (+) Confirmation (-) Rejection
I. Decomposition-mineralization		
Organic matter turnover	Decreased rate of C mineralization as a result of soil acidification and/or associated trace metal toxicity	+ Francis et al., 1980 + Lohm, 1980
Microbial community dynamics	Shift from bacteria toward more acid-tolerant fungi	+ Lohm, 1980
Nitrogen mineralization	Decreased ammonification	+ Francis et al., 1980
	Decreased nitrification	+ Francis et al., 1980 + Alexander, 1980
Root uptake	Trace metal toxicity associated with soil acidification	+ Mayer and Ulrich, 1977
II. Soil exchange complex		
Clay mineral morphology	Increased formation of hydroxy-Al interlayers under acid weathering	+ Jackson, 1963
Exchange capacity	Decrease in CEC as a result of clay alumination	+ Sawhney, 1968
	Increase in the CEC of Ultisols as a result of sulfate adsorption	+ Johnson, 1980
Base saturation and exchangeable acidity	Decrease in base saturation and increase in soil acidity	+ Abrahamsen, 1980 + Farrel et al., 1980 + Stuanes, 1980 + Bjor and Teigen, 1980 - Linzon and Temple, 1980
III. Element leaching		
Aluminum	Increased mobilization and leaching	+ Cronan, 1980 + Abrahamsen et al., 1976 + Mayer and Ulrich, 1977 + Baker et al., 1977
Manganese	Increased leaching	+ Mayer and Ulrich, 1977
Heavy metals	Increased leaching	- Tyler, 1978
Nutrient cations	Increased leaching loss	+ Abrahamsen, 1980 + Overrein, 1972 + Mayer and Ulrich, 1977 + Stuanes, 1980 + Cronan, 1980
Sulfur	Retained thus reducing S loss and cation loss	+ Farrell et al., 1980 + Johnson, 1979
IV. Primary minerial weathering	Increased weathering loss	+ Gjessing et al., 1976 + Johnson, 1979

SOURCE: McFee (1983). See original for references.

IV. Primary minerial weathering — Increased weathering loss — + Gjessing et al., 1976; + Johnson, 1979

SOURCE: McFee (1983). See original for references.

TABLE 15

SOIL NUTRIENTS, INPUTS, WEATHERING RELEASES, OUTPUTS AND NUTRIENT LOSSES DUE TO FIRE IN A PINELANDS UPLAND ECOSYSTEM, (WANG, 1984)

	N	K	Mg	Ca
	UNDISTURBED SYSTEM			
Soil exchangeable (g/sq.m)				
0 - 10 cm	24	10.8	3.9	19.9
0 - 30 cm	43	14.7	5.3	30.6
0 - 100 cm	93	33	8.2	56
Input (bulk precip.) (g/sq.m/yr)	0.42	0.12	0.12	0.27
Weathering release (g/sq.m/yr)	0.00	0.15	0.00	0.14
Output (leaching) (g/sq.m/yr)	0.002	0.15	0.08	0.14
Net accumulation (g/sq.m/yr)	0.42	0.12	0.04	0.27
	BURNED ECOSYSTEM			
Fire loss (g/sq.m/yr)	13.0	2.2	0.30	0.35
Return-time (years)	31	18	8	2

b. Metal toxicity

The increased solubility of metals at low pH has been frequently discussed as a major effect of soil acidification. In Germany, considerable evidence has been obtained by Ulrich to support the contention that aluminum has become solubilized in toxic amounts. Increases in soluble aluminum have been reported from northern New England (Cronan and Schofield, 1979), but have not been observed in Norway. Johnson et al (1984) have pointed out that in streams in which pH is controlled by strong mineral acid anions (i.e. anthropogenically derived sulfate), solubilized aluminum is in inorganic form, whereas in streams dominated by weak organic and carbonic acid acidity aluminum is organically bound and thus not in a form toxic to aquatic biota. Indirect evidence of increased aluminum availability is found in the tree-ring studies of Baes and Scherbatskoy cited above. The biological significance of increased soluble aluminum is harder to evaluate, however, since trees vary widely in their tolerance to aluminum, and some studies (e.g., for the declining red spruce), show no differences in aluminum content of healthy and declining trees. However, the interactions of acidic deposition with heavy metals are increasingly being stressed as the likely locus for forest impacts (Aber, 1984).

Increased mobility of other heavy metal toxicants has been discussed. There is some experimental evidence that increased percolate acidity mobilizes heavy metals; this topic is the subject of current research and little is yet established.

c. Impacts on soil microbial processes

The effects of acidic deposition on organic matter decomposition and on the microbial components of the nitrogen cycle are of critical importance, in part because they are the controlling variables for many aspects of ecosystem function, and in part because (as reviewed above) the apparent nitrogen limitation of many forest types appears to be a critical variable in the assessment of acidic deposition effects. Many studies have shown that while nitrogen mineralization (release of NH_4 from organic combination) is not pH-sensitive, autotrophic nitrification (oxidation of NH_4^+ to NO_3^-) is highly sensitive. There is also considerable evidence that decomposition is inhibited under acidic conditions. Thus, both field and laboratory studies suggest an increase in litter and organic matter content and a decrease in nitrate availability in forest soils. There is also some evidence that blue-green algal populations in soil and leguminous nodulation may decrease in acidified soils, thus resulting in a decrease in nitrogen fixation.

There is virtually no information concerning the potential effects of acidic deposition on mycorrhizae, despite the critical importance of these fungi in tree nutrition. However, an absence of mycorrhizae has been observed in declining German forests.

d. Direct effects on roots

Research in Germany has shown that in declining forest stands, fine root growth is seriously inhibited. Ulrich has related the loss of fine feeder roots to aluminum toxicity. Although there have not yet been published detailed studies of root growth in affected American trees, Siccama et al. (1982) reported that declining red spruce had roots that looked "unhealthy." Johnson et al.

(in Linthurst 1984) found that roots of declining red spruce did not differ from those of healthy trees in element composition. Studies in Europe have suggested that tree roots in acid-sensitive areas have Ca and Mg imbalances.

3. General factors

The interaction of vegetation with atmospheric conditions is subject to several general influences which will affect the potential for acidic effects on the various factors described above.

a. Elevation and aspect

The extensive studies undertaken in northern New England have shown that the amount, form, and chemistry of precipitation entering a forest is strongly influenced by the elevation and aspect of the stand. In particular, the exposure to fog and mist appears to be important, because these forms of atmospheric moisture usually have lower pH than rainfall, and are in contact with the vegetation for longer periods of time. Some scientists (e.g., Aber, 1984) now believe that only high-elevation stands are liable to damage from acidic deposition (primarily from fog-drip). This factor should be considered for the ridges of northern New Jersey, but the elevations are much lower than those in northern New England, and therefore not likely to be strictly comparable.

b. Rainfall timing

Lindberg et al. (in D'Itri, 1982) have shown that the length and intensity of particular precipitation episodes combine with the length of the preceeding dry period to strongly affect the actual concentrations of chemicals on a leaf surface. They show that sulfate, for example, can reach 15 ppm (mg/1) on the leaf surface, and 1.5 grams/1 as a potential concentration as the water film on the leaf evaporates. Thus, leaves may be exposed, albeit briefly, to much higher levels of toxics and total acidity than is evident from bulk precipitation measurements. Their results support both the previously described need for better understanding of dry deposition processes, and a need for better quantification of rainfall events and their interactions with tree canopies.

4. Tree growth and Survival

The effects of acidic deposition on both above-ground and below-ground factors are ultimately summed in the extent to which tree growth and survival is altered. Conversely, because tree growth is the end product of the numerous factors discussed here, it is very difficult to unequivocally ascribe observed changes in growth in forest trees to acidic deposition. In Germany, extensive die-back (death of part or all of the tree canopy) and decline (a complex of symptoms variably including early loss of foliage, poor color, drooping of branches, abnormally slow growth) of a variety of both coniferous and hardwood trees is occurring on diverse sites throughout the country; it is estimated that up to 25% of the forest area of West Germany may be affected (Cowling, 1983). In part because no specific stress can be found that is common to all areas, the problem has been ascribed to air pollution, possibly acting through some combination of oxidant gas effects, nutrient imbalance effects, physiological effects on photosynthesis and metabolism, and soil aluminum toxicity effects. In America,

extensive decline and die-back has been so far described only for red spruce (Johnson and Siccama, 1983) growing north of New Jersey; high-elevation populations (2,000 m) in the southern Appalachians are also experiencing severe decline, but lower elevation stands are still vigorous. The symptoms of the condition in spruce are not similar to those of the German trees, and Al toxicity in the soil has been eliminated as a hypothesis. The only other case that has been described is the abrupt decrease in growth of short leaf and pitch pines in southern New Jersey (Johnson, in Linthurst, 1984). In this study, 60% of the sampled trees showed evidence of abrupt and prolonged decrease in increment growth since the 1950's. However, no other symptoms of decline have been observed in these trees. For both the New England spruce and the New Jersey pitch pines, it is now thought that drought stress may be interacting with acidic deposition effects, and possibly other stress factors, in causing the decline; but no certain, or simple, hypothesis has seemed reasonable (Johnson, pers. comm.). Puckett (1982) has found that pitch and white pines, and chestnut oak in southern New York have experienced a change in the relationship of growth rate to climate, but the role of acidic deposition in this phenomenon is at present entirely speculative.

The most extensive studies of tree growth have been conducted in Scandinavia, where comprehensive studies of the effects of acidic deposition on forests have been continuing for a decade. Despite extensive sampling of forest trees (Norway spruce and Scots pine) throughout Norway and Sweden (6,150 increment cores examined in Norway alone), there is no evidence of growth reductions attributable to acidic precipitation. Although in Sweden there was some evidence in early studies of poorer growth in areas classed as having more susceptible soil, subsequent analyses suggested a more complex relationship between soil quality and soil processes, climate and tree growth, which could account for the observed growth patterns independently of the rainfall acidity. Experimental studies within the Scandinavian research program have shown that even with prolonged (4-7 yr) application of simulated rain and irrigation water, detrimental effects on growth are small or non-existent. In fact, many of the experimental studies have suggested that nitrogen is a limiting resource in the Norway spruce-Scots pine forests, and the nitrogen content of the simulated rain is a potent fertilizer. Modeling studies have also suggested that nitrogen mineralization from leaf litter by acidic deposition has a greater effect on tree growth than any direct effect of acidity on the trees.

Some of the American work has presented similar findings. Cogbill (1977) sampled four species of trees in New England and Tennessee, and reported that although some species showed reduced growth in recent decades, no regional or synchronized decreases in growth could be discerned. The work of Wood and Borman (1974, 1977) and Aber et al. (in D'Itri, 1980) have also suggested that nitrogen limitation, and the rate of nitrification in litter, are the most important controls on the hardwood trees and forests in their studies. Nevertheless, there are numerous studies of tree growth with respect to point sources of SO_2 and NO_x which show clear decreases in growth, in many cases without visible evidence of tree injury (Tomlinson, 1983). Thus, the studies of eastern American forests suggest complex and variable effects of regional air pollution on forest health.

It is clear that the effects of acidic deposition on tree growth remain an open question. The Norwegian scientists (Overrein et al., 1980) concluded that

"it is difficult to forecast the ultimate results of the atmospheric acidification and related air pollutants on terrestrial systems and to judge the rate and even the direction of changes. In the more susceptible areas it seems, however, to be a question of proportion and time required rather than whether any ecological effects appear or not." Linthurst (1983) concludes "At present there is no proof that acidic deposition is currently limiting growth of forests in either Europe or the United States... It is too early to conclude that acidic deposition has not nor will not affect forest productivity." The recent dieback and decline of conifers in the northeast (including New Jersey's Pinelands) and various species in Germany, although as yet not definitively linked to air pollution, pose a serious question for research and management decisions.

5. INTERPRETING FOREST DATA

The above review of factors mediating the impact of acidic deposition on forest vegetation has given a very brief and incomplete summary of the complexity of the problem. Complicating an application of this existing data-base to a particular region (i.e., New Jersey) is the fact that most of the research available to date concentrates on a very small number of species - Norway spruce and Scots pine in Europe, red spruce, pitch pine, shortleaf pine, and northern hardwoods in North America, and a few other species in a small fraction of the literature. A slightly larger number of species have been studied with respect to point sources of air pollutants. Nevertheless, the fact that comparative studies have invariably documented significant differences in behavior among tree species, and that examination of several response factors in a single forest stand has been accomplished for only a few species in a few locations, makes generalization unreliable.

The consensus of opinion concerning the effects of regional acidic deposition on forest vegetation appears to be that (1) no clearcut effects have been documented in field situations, but (2) there are data suggesting that at least some (primarily conifer) species in the northeastern and high-elevation southeastern United States are suffering decreases in growth, and (3) in the recent die-back and decline of conifers in Germany and New England acidic deposition has been identified as one likely candidate among other factors in a complex of stresses, and finally (4) there is sufficient evidence that acidic deposition can affect processes important to forest growth (such as microbial cycling of nitrogen, cation leaching from both soils and leaves, and direct effects on leaf and root morphology) to support the conclusion that negative impacts of acidic deposition are likely to occur in the future to some species in some areas.

C. FORESTS IN NEW JERSEY

1. Commercial Forest Land

The most recent data on the forest resources in New Jersey is the Timber Resources of New Jersey survey (Ferguson and Mayer, 1974). This summary is based on data collected in 1971-1972. Although these data are over a decade old, they at least provide an estimate of the nature of the forest resources in the state.

Forty percent (1.93×10^6 acres) of the state's land area was classified as forested; another 14% (0.69×10^6 acres) was classified as urban forest land (including urban parks, watershed property, etc). As of 1971, forest land (not including urban land) had decreased 12% since the previous survey in 1956; it is thought that a somewhat smaller decrease of 7-9% will be observed in the timber resource survey scheduled to be undertaken next year. Of the total commercial forest land, 62.6% (1.16×10^6 acres) is found in the eight southern counties that have sandy, poorly buffered, sensitive soils; these counties grow 54.2% of the total growing stock of $1,470.3 \times 10^6$ cu. ft. of commercial wood produced in the state. The other likely sensitive areas are smaller: Sussex, Warren, Passaic, and Morris counties, which include the sensitive areas of the Kittatinny Ridge and the Highlands, together account for 24.9% of the forest land and 31.3% of the growing stock. The forests can be classified into several major types by their dominant species; although seven major types can be identified in the state, two account for most of the commercial forest land. The yellow pine type (pitch pine and shortleaf pine; i.e., Pinelands forest communities) account for about 25% of the forest land, and oak-hickory stands account for 40%. Other minor types include white pine-hemlock stands, oak-gum stands (which includes Atlantic white-cedar and swamp hardwoods) and northern hardwoods.

Table 16 shows the distribution of major tree species by county, in terms of the commercial measure of wood volume. Conifers make up 18% of the total growing-stock volume. Table 16 shows that oaks are important in all counties of the state, whereas yellow pines (primarily shortleaf and pitch pine) are restricted to counties within the Pinelands area. As of 1972, oaks were increasing in volume, whereas all conifers except pitch pine were decreasing. It is apparent from Table 16 that the species most extensively studied in published acidic deposition research - red spruce, sugar maple, yellow birch - are poorly represented or absent from the state. Chestnut oak, as a member of the white oak group, was among the species found by Puckett (1982) to have experienced changes in growth rate unrelated to climate. However, there have been no published regional studies of oak growth patterns, nor have any of the experimental studies utilized oak species. Unpublished, preliminary data from the FORAST program have suggested that the oaks are not showing any change in growth unrelated to climate (Johnson, pers. comm.). Thus, without data on either the sensitivity of oaks to the various factors outlined above, or on growth rate changes during the past several decades, it is very difficult to judge whether this large component of New Jersey's forests is faced with an immediate threat from acidic deposition.

The data of Johnson and his colleagues on pitch pine and shortleaf pine have suggested that growth reductions in this species may be occurring. In the original study, approximately one-third of the 176 trees sampled in the central Pinelands showed no growth change. The data for the other trees were reported in terms of diameter increment, rather than basal area increment; since diameter increment normally decreases with tree age, it is unclear how much of the observed growth decrease was actually a normal growth trend. Unpublished, preliminary results of the FORAST research program have suggested that pitch pine does show regional unexplained change in the relationship of growth to climate (Johnson, pers. comm.). Johnson and his co-workers have recently attributed the growth problems to an initiating drought stress, compounded by other continuing environmental stresses, possibly including acidic deposition. In the original study, the growth reductions were correlated with stream pH, and the assumption

TABLE 16

NET VOLUME OF GROWING STOCK ON COMMERCIAL FOREST LAND,
BY SPECIES AND COUNTIES, NEW JERSEY, 1972
(In millions of cubic feet)

COUNTY	YELLOW PINES	OTHER SOFT-WOODS	TOTAL SOFT-WOODS	WHITE OAKS	RED OAKS	HICKORY	SOFT MAPLES	SWEET-GUM	OTHER HARD-WOODS	TOTAL HARD-WOODS	ALL SPECIES
Atlantic	35.0	5.3	40.3	26.6	37.6	1.7	14.1	13.1	6.7	99.8	140.
Burlington	48.8	5.7	54.5	27.2	37.0	2.0	15.4	9.7	10.4	101.7	156.
Camden	11.5	2.0	13.5	5.4	8.0	.5	2.8	2.4	2.3	21.4	34.
Cape May	11.7	1.9	13.6	10.1	14.4	.6	5.3	5.6	2.6	38.6	52.
Cumberland	26.2	6.0	32.2	19.5	24.4	1.4	9.3	6.5	8.0	69.1	101.
Gloucester	9.4	2.0	11.4	10.2	14.0	.2	6.8	5.4	4.8	41.4	52.
Hunterdon	--	2.8	2.8	24.3	27.5	6.6	3.2	1.3	25.9	88.8	91.
Mercer	--	.7	.7	7.6	9.6	2.6	1.2	.2	8.9	30.1	30.
Middlesex	--	1.5	1.5	10.9	13.7	3.2	1.7	.6	12.5	42.6	44.
Monmouth	12.0	1.5	13.5	15.7	22.8	.4	11.1	10.2	6.2	66.4	79.
Morris	--	4.0	4.0	41.0	45.8	12.2	5.3	1.7	41.6	147.6	151.
Ocean	42.2	4.6	46.8	18.3	27.8	1.4	10.4	10.8	7.5	76.2	123.
Passaic	--	1.4	1.4	11.1	12.9	4.0	1.4	.4	12.4	42.2	43.
Salem	9.6	2.0	11.6	11.5	14.9	.5	8.0	4.3	5.1	44.3	55.
Somerset	--	1.4	1.4	11.4	14.2	3.9	1.6	.1	14.9	46.1	47.
Sussex	--	5.5	5.5	43.1	50.2	15.6	5.9	1.6	48.7	165.1	170.
Warren	--	3.3	3.3	24.0	27.9	7.0	3.3	1.3	27.4	90.9	94.
TOTAL	206.4	51.6	258.0	317.9	402.7	63.8	106.8	75.2	245.9	1,212.3	1,470.

Source: Ferguson and Mayer, 1974

was made that the sandy, inert soils had little effect on the pH of percolating rainwater; however, the assumption that streamwater pH showed the effects of acidic precipitation has been questioned (see section on Effects of Acidic Deposition on Aquatic Biota). If this phenomenon described by Johnson is verified in more extensive studies of the species throughout southern New Jersey, and can be more definitively associated with acidic deposition, it will suggest a major threat to the commercial forests and the forestry industry of the state.

2. Potential Economic Effects

The economic stakes involved are substantial. In 1972, growth of economically-sized trees accounted for 56% of the total increase in tree volume; the rest came from growth of saplings into economic tree classes. Of the 11.4 x 10^6 cu ft harvested at that time, 29% was from softwoods. Table 17 gives more recent data for the dollar value of forest products in New Jersey. Commercial forest land produced over $5 million dollars of raw wood value in 1977, nearly $89 million of manufactured product value, and employed 4,500 people with an annual payroll of $42 million. Although the number of employees in the primary industry (cutters, mill operators, etc.) is small, it is estimated that about 30,000 employees are involved in all timber-based industries (Pierson, pers. comm.).

The forested areas of the state also have recreational value, whose economic importance is hard to quantify. In 1983, the 30 state parks received 6.6 x 10^6 day visitors and 0.22 x 10^6 overnight visitors; the 12 state forests received 1.3 x 10^6 day visitors and 0.28 x 10^6 overnight users. The fees received for the overnight use represent a small fraction of the total economic value generated by this use of state forested lands (Guidotti, pers. comm.), and there are not data available to adequately characterize this contribution to the state's economic activity. The esthetic value of the forests to the visitors is undoubtedly even more important - and less quantifiable - than the economic value.

Trees in urban and suburban areas of the state are subjected to numerous stresses, of which acidic deposition may be a relatively minor component. The sensitivity of this tree population deserves more attention, however, because the low diversity of tree species, and the trend toward an even lower diversity in the future, creates conditions for a potentially disastrous loss of urban trees.

D. CONCLUSIONS

1. The data base on forest responses to acidic deposition consists of laboratory and field experimental studies of a large variety of processes in small number of species, and some field analyses of tree growth in an equally small number of populations. Conclusions at present must rest on extrapolation from studies of individual factors to complex ecosystems and modelling of system properties. The consensus is that this data base is not yet sufficient to warrant strong conclusions about the role of acidic deposition - as opposed to other components of air pollution - in forest growth.

TABLE 17

FOREST PRODUCTS HARVESTED ANNUALLY IN NEW JERSEY

PRODUCT	ANNUAL VOLUME HARVESTED	STUMPAGE VALUE	[1]VALUE AT POINT OF DELIVERY	VALUE ADDED BY MANUFACTURER
Sawtimber	22,910 MBF	$1,489,150	$2,978,300	$44,674,500
Veneer	1,500 MBF	360,000	960,000	14,400,000
Pulpwood	27,000 Cds.	162,000	945,000	9,450,000
Furnace Poles	8,000 Tons	16,000	256,000	2,560,000
Posts & Fencing	11,200 Cds.	224,000	2,448,000	4,480,000
Piling	900 MBF	90,000	360,000	3,600,000
Firewood	85,000 Cds.	595,000	5,100,000	5,100,000
Landscape Wood Chips	2,144,600 Cu. Ft.	317,700	317,700	476,500
Christmas Trees	143,100 Trees	1,576,000	1,576,000	1,576,000
Miscellaneous	500,000 Cu. Ft.	250,000	500,000	2,500,000
TOTAL		$5,079,850	$15,441,000	$88,817,000

Statistics for the Lumber and Wood Products Group for the Annual Survey of Manufacturers - 1975 shows the following:

Number of employees	4,500
Payroll	$ 42,000,000
Value added by manufacturer	$ 85,900,000
value of industry shipments	$186,500,000

[1]Sawmill, Pulpmill, Copper Smelter, Retailer, etc.

[2]A total harvest in excess of 73,700,000 Bd. Ft. plus 85,000 cds. of firewood is indicated.

Data Source: Forest Survey - New Jersey 1972 updated by Timber Drain Survey 1977 (unpublished).
Annual Survey of Manufacturers, 1975.
Average Prices Paid for Products, 1977.

Date compiled by the Bureau of Forestry, NJDEP.

2. Extrapolation to New Jersey is rendered particularly difficult by the paucity of data on the dominant species (oaks).

3. Results from a study by Johnson of pitch pine growth suggests that a portion of the tree populations may be experiencing up to 40% decreased stemwood growth; circumstantial evidence associates precipitation acidity with the decline. Given the relatively small number of trees sampled, and the possibility (suggested by Johnson) that drought and other stresses have contributed to the growth reduction, this original report must be considered as a warning and a stimulus for further research on this species.

4. Extensive, long-term studies in Scandinavia have resulted in the conclusion that although no effects on tree growth are yet apparent, there is good reason to believe that in susceptible areas, effects will eventually occur. The rapid onset of forest stress in Germany supports this conclusion, and reinforces the idea that acidic deposition-related stress may cause sudden environmental changes in systems that showed no previous impact.

5. The following research is recommended to better evaluate the threat to New Jersey's forests:

   a. Research is needed to assess the sensitivity of New Jersey's forest soils to acidification, to cation loss (i.e., anion mobility) and to mobilization of soluble aluminum.

   b. More extensive research is needed on growth patterns of the dominant species in New Jersey, to determine if growth reduction trends are found throughout the state, and to better determine the relationships of growth trends to climate, fire history, and other environmental factors. It is strongly suggested that such growth trend research be incorporated in the timber resources survey now being planned by the Bureau of Forestry.

   c. Research is needed to better describe the natural and anthropogenic sources of hydrogen ion in soil systems. It is difficult to assess the importance of atmospheric deposition on forest soils without some information on the extent of internal generation of H^+ (through organic acid formation, nitrification, etc.).

6. A monitoring program should be undertaken to regularly assess foliar and root tissue nutrient contents of major species in the major forested areas. Such a monitoring program could give evidence of impending problems before they become manifest as extensive decline and death of trees. Given the possibility that the problems in New England and in Germany are indeed air pollution-related, a monitoring system to provide an early warning could be invaluable in New Jersey.

EFFECTS OF ACIDIC DEPOSITION ON PRODUCTION AGRICULTURE

A. THE VALUE OF PRODUCTION AGRICULTURE OF NEW JERSEY

New Jersey in years past earned its nickname of "the Garden State" and, although it has undergone extensive urbanization and industrialization, the State still retains vegetation resources of significant importance.

Production agriculture persists despite the pessimism reflected in "New Jersey Trends" published in 1974. A most important characteristic of New Jersey agriculture is the diversity of crops grown in the State; some 9 different field crops, 12 vegetable crops, and 5 fruit crops are raised for the commercial market. In the rural counties in the southern part of the State are grown most of the vegetables including asparagus, cabbage, lettuce, onion, peppers, sweet corn, sweet potato, and tomato. These same counties contain apple and peach orchards, blueberry bogs, strawberry fields, and potato fields. A field crop, either soybean, corn, wheat, alfalfa, rye, barley, or white potato, is grown in every county with the exception of Passaic, Bergen, Essex, and Union.

The estimated value of all crops produced in New Jersey in 1982 (latest data available) was $288.7 million (Crop Reporting Service, 1983). Field crops were valued at $113.9 million (41.0% of total), vegetables at $95.7 million (32.9%) and fruits and berries at $74.2 million (26.1%). Soybean and corn were ranked as the most valuable field crops at $22.5 million and $36.8 million, respectively. In 1983 the average value of an acre of farmland in New Jersey was the highest in the nation, $3,148. By comparison, the value was $793 in New York, $1,381 in Pennsylvania, $1,692 in Delaware, and $1,925 in California.

In addition to agronomic crops, a significant component of production agriculture in New Jersey is represented by shrubs, trees, flowers, and grasses that are purposely planted and cultivated outdoors for amenity (non-product) purposes. These horticultural species enhance the urban environment from greenbelts to gardens, from cemeteries to city streets, from rights-of-way to rooftops, from parks to planters (Santamour, 1978). The magnitude of this component is reflected in statistics relating to the nursery and turfgrass industries in New Jersey. According to the Crop Reporting Service, 1,031 certified nurseries operated in 1982 on 11,822 acres of land and had a total sales volume of $110 million, placing New Jersey in the number six position for the entire country. Two of the state's nurseries, Princeton and Sunnybrook, ranked in the top 100 United States' nurseries. The distribution of nurseries in New Jersey is as follows: 399 in the northern part of the state, 338 in the central part, and 294 in the southern part, with an acreage of 1,889 acres, 6,366, and 3,569, respectively. A survey is currently underway in New Jersey to identify not only the certified nurseries but all nurseries. The importance of horticultural plantings is dramatically illustrated by the finding that hospital patients recover from surgery more rapidly if they view trees rather than brick walls (Ulrich, 1984).

Tate (pers. comm.) has recently estimated that there is an average of 80 trees per street mile in New Jersey cities and suburbs. The most common are Norway maple, flowering pear, red and pin oaks, silver maple followed by

little-leaf linden, sugar and red maple, and honeylocust. Although the magnitude of the turfgrass industry is only now being documented, it is currently estimated that some 2 million home lawns and 200 golf courses have a high investment in turfgrass. Just as with agricultural crops and woody plants, there is a great diversity among the kinds of turfgrass planted in the state, and breeding programs continue to introduce many genetic variants into use to meet the particular needs of the state.

B. A HISTORY OF THE IMPACT OF AIR POLLUTION ON THE NEW JERSEY CROPS AND ORNAMENTALS

To place the phenomenon of acidic precipitation in proper perspective, it is appropriate to review very briefly the major air pollutants that have affected vegetation of the state. In 1946 peach and corn crops in certain areas along the Delaware River sustained such acute damage from a suspected air pollutant that the Legislature charged the Agricultural Experiment Station to make a study of the effect of industrial fumes on agricultural lands (Assembly Bill 376). HF emissions from industrial processes involved with the fabrication of the atomic bomb were identified as the cause of the crop damage (Daines et al., 1952). In succeeding years fluoride emissions from other sources, including the manufacturers of bricks and glass, an oil refinery, and a molybdendum reduction plant, were also found to injure not only agriculture crops but various types of evergreen trees and flowers (Brennan and Rhoads, 1976).

In the 1950's episodes of sulfur dioxide injury to vegetation were commonplace, especially in the northern counties in the spring of the year. Agricultural extension agents from the various counties often submitted plant samples with suspected SO_2 damage to the Air Pollution Staff at the Experiment Station for diagnosis. Spring flowers, scarlet hawthorne and crabapple trees, and newly emerged conifer needles exhibited foliar intercostal lesions symptomatic of SO_2 pollution (Leone et al., 1965). Since 1970 SO_2 symptoms have not been observed on vegetation in New Jersey (Brennan and Rhoads, 1976), and their absence coincides with the decrease in SO_2 emissions brought about by stricter regulations imposed on burning high sulfur coal in New Jersey.

In the 1960's photochemically-generated air pollution emerged as a formidable threat to agriculture. Ozone toxicity symptoms were common in rural and urban areas on many crops including grape, onion, potato, oats, squash, alfalfa, tomato, and scallions (Daines et al., 1960). Where the marketable crop was the leaf, as in spinach, there was obvious reduction in value. However, where the product was the seed, tuber, root or stem, the effect on yield could not be predicted from the amount of foliar injury. Field tests with various potato cultivars commonly grown in New Jersey revealed that foliage could develop a significant amount of foliar ozone injury (12-22%) without any resulting effect on yield; but when 75% of the foliage was affected, then yield was reduced 25 to 31% (Clarke et al., 1983). Currently field tests are being conducted with soybean cultivars to determine the effect of ambient O_3 on foliar injury and seed yield and quality. Contrary to a predicted loss of 20-24% by the National Crop Loss Assessment Network (NCLAN), the Williams cultivar has apparently suffered no yield loss during the past 3 years although 5-10% of the foliage has exhibited O_3 symptoms.

When photochemical pollution was first studied in California, PAN (peroxyacetylnitrate) was considered to be an important phytotoxicant. In the early 1960's what appeared to be PAN symptoms was observed on a few leafy vegetables around the Carlstadt area, but they have not been noticed in New Jersey in the last 15 years. Recent PAN measurements made in New Brunswick support the view that current concentrations are not high enough to cause plant injury (Lewis et al., 1983). It is interesting that in California where PAN levels are consistently higher than in New Jersey, injury has been recorded on herbaceous plants but never on trees in the field (Taylor, pers. comm.).

Other ingredients of photochemical smog, e.g. NO_x, have not been found to cause plant injury at concentrations that occur in ambient air in New Jersey. However, some data accumulated in New Brunswick (Brennan et al., 1964) indicated that high aldehyde levels (0.20 ppm) might correlate with a particular type of injury to petunia plants.

Following Likens' pronouncement that rainfall in the Northeast had become acidic, samples from rain events were collected at several sites in New Jersey during 1973-1974, and pH was determined. About 75% of the measurements were between pH 3.0-4.5, although some values were as high as 7.5. Agricultural agents and crop specialists were contacted to determine if any problem of unknown etiology was occurring in the State that might have been related to acid rain. No such problem could be identified.

In 1980 experiments were initiated in the greenhouse to investigate the effect of acid rain on crop plants. One study evaluated the role of epicuticular wax in determining the sensitivity of leaves to acid rain. A wax-reducing pesticide was found to enhance the wettability of cabbage foliage and its sensitivity to acid rain (Blum and Brennan, 1982). A second study supported by N.J. DEP in 1981 evaluated the effect of a combination of ozone exposures and acid rain on the yield of alfalfa (cv Saranac). The yield was found to be similar in plants exposed to simulated rain of pH 3.0 and 5.6. There was no evidence for a synergistic interaction between acid rain and ozone treatments (Rebbeck & Brennan, 1984).

C. AN ECONOMIC ASSESSMENT OF AIR POLLUTION DAMAGE TO VEGETATION

The impact of air pollution on agriculture in terms of economics is difficult to assess for two reasons (1) a simple relationship does not exist between foliar injury (which is readily observed and measured) and marketable yield except in leafy crops and (2) yield loss may or may not equate to economic loss, depending upon events that occur in the marketplace. Nevertheless estimates have been made. In 1970 scientists at Stanford Research Institute (Benedict et al., 1971) estimated that crops and ornamentals nationwide sustained a loss of $130 million from air pollution and, in the Middle Atlantic States, a loss of $17.6 million (2.4% of the value of crops and ornamentals). Over 90% of the losses were attributed to ozone. These predictions were based on fuel consumption, point source emission data, crop sensitivity and market value. Actual on-site

surveys of vegetation injury were conducted in New Jersey in 1971 and 1972. Feliciano (1971) reported a loss of $1.2 million and Pell (1972), a loss of $128,000. At that time, the relationship between foliar injury and yield was unknown and, therefore, assumed to follow a "rule-of-thumb": if 1-5% leaf injury occurred, then a 1% loss was applied for that crop; 6-10% foliar injury, a 2% loss, etc. In the last fifteen years estimates of nationwide losses have increased from $130 million to somewhere between $1 and $2 billion. By these calculations soybean production in New Jersey is reportedly decreased 13.8% ($5 million) and corn, 3% ($1 million) because of 0_3 pollution (Heck et al., 1982). These predictions have not been confirmed. A[3] soybean field test in New Brunswick from 1981 through 1983 revealed no significant yield change in the Williams cultivar due to ozone pollution.

D. A LITERATURE REVIEW OF ACIDIC PRECIPITATION AND ITS EFFECT ON CROPS AND ORNAMENTALS

The realization that rainfall in the Northeast has a pH of about 4.0 provided the stimulus for investigations of the effect of acidic precipitation on vegetation. This sequence of events contrasts sharply with previous air pollution research efforts where the actual observation of plant damage preceded a study of its causes. The former is the more difficult approach because theoretically thousands of plant species (3,000 in New Jersey) (to say nothing of cultivars/varieties) must be examined to detect the small number that are likely to be susceptible to acidic deposition. The investigations are also complicated by the fact that acidic precipitation, unlike the gaseous pollutants previously discussed, is not a pure substance nor is it chemically defined. Acidic rain is a complex mixture in which not only is pH a variable, but also its nitrogen, sulfur, and heavy metal content, and that of any number of other soluble airborne chemicals.

In the earliest acidic precipitation experiments with agronomic crops and trees, researchers applied simulated rain to various plant species in search of some characteristic visible symptom, such as found with other pollutants. No direct effect was observed until pH was decreased to pH 3.5 or lower, considerably below ambient levels (Linzon, 1981). Later reports indicated that a few foliar lesions could be observed on some species (beet, swiss chard, soybean, green pepper and bluegrass) following treatment with simulated rain at pH 4.0 (Lee et al., 1981). Only garden beet has been known to develop foliar symptoms as a result of ambient rain. Evans (1984) made this observation on the cultivar Perfected Detroit V-904 following a rainfall of pH 3.9 in Long Island. However the phenomenon has not been confirmed by any other researchers.

To date National efforts to assess the impact of acidic rain on vegetation have focused on the effect of simulated acidic precipitation (pH of about 4.2 compared to pH 5.6, the assumed value for uncontaminated rain) on the yield of agricultural crops, usually in controlled greenhouse experiments and less frequently in the field. Irving (1983) has provided an excellent review and analysis of research up to 1983. In controlled environment studies of some 34 crop varieties (28 species), 17 crops exhibited no effect from acidic rain, 8 had increased yield, 6 had decreased yield, and 3 showed a variable response. Those

adversely affected were pinto bean, mustard greens, broccoli, radish, beet, and carrot. In field studies of some 14 cultivars (9 species) where simulated acidic rain treatments were added to ambient rain, the majority of species again exhibited no effect, 6 had increased yields and 1 decreased yield (beet), and 3 showed a variable effect.

Unfortunately, the dose-response data generated from these experiments cannot be applied to the real world. This can be illustrated by examining data from EPA-sponsored soybean tests in which 4 different cultivars were exposed to simulated rain at pH's ranging from 5.3 (or 4.0) to 2.3 at four sites in the East and Midwest. The effect of increasing acidity on soybean yield was found to be variable: the Davis cultivar in Raleigh, North Carolina and the Wells cultivar in Argonne, Illinois showed no change; the Beeson cultivar grown in Yonkers, New York showed an increase; and the Amsoy 71 cultivar grown in Long Island, New York, a decrease in yield. One might infer from these results that the cultivars are differentially sensitive to simulated acidic precipitation, but as a review by Jacobson (1982) points out, many differences in methodology prevent a comparison of results between locations. In fact, 11 points related to the general protocol limit the "relevance of experimental results to ambient field conditions in agricultural regions with acidic rain." In her review, Irving (1983) reiterated many of these limitations, including the use of rain simulant with a higher SO_2/NO_3 ratio than that found in ambient air and also the failure to characterize the conditions under which plants were grown. Actually, if the experiments were repeated with a standardized protocol, the result would be only of academic interest to New Jersey, since none of the 4 cultivars tested is important to the New Jersey soybean industry.

Evans of the Brookhaven National Laboratory who contributed the data on the Amsoy cultivar to the above study has continued with research on soybean. In 1984 he reported the results of an experiment in which he compared the response of Amsoy and Williams to simulated rain treatments. Seed yields of Amsoy exposed twice weekly to simulated rain of pH 4.1, 3.3, 2.7 were respectively 3.0, 9.0, and 12.8% below yields of plants exposed to simulated rain of pH 5.6. A treatment-response function of seed yield versus rainfall pH for Amsoy was $y = 10.20 + 0.587 X$ and had a correlation coefficient of 0.96 (y = seed mass/plant and X is pH of simulated rain). The seed yield of Williams, on the other hand, was not influenced by pH of the rain; the correlation coefficient between seed mass and rain pH being only 0.038. These results are favorable for New Jersey agriculture since Williams is a commonly-grown cultivar and Amsoy has not been planted in many years.

In general, the results obtained by treating agricultural crops with acidic precipitation reaffirmed many of the principles demonstrated earlier with gaseous pollutants.

- Crop tolerance is the rule; and susceptibility, the exception.
- Interspecific and intraspecific variations occur in respect to foliar and yield response.

- There is little evidence for a linear response function.
- Year to year variation in response is observed with a given species, indicating the importance of growth environment.
- Field-grown plants tend to be less sensitive than chamber-grown plants.
- A significant amount of foliar injury does not always translate into a yield reduction.

In addition to agricultural crops, the forest ecosystem has been the focus of acidic precipitation studies (reported separately in this document) but the man-selected and man-managed trees, shrubs, and grasses that comprise an important component of urban vegetation have been, for the most part, neglected. Since this "artificial" ecosystem has characteristics all its own, inferences cannot be made from studies with the natural forest ecosystem. Very often different species are involved, with genetic qualities unlike those of plants that have undergone natural selection (Steiner and Townsend, 1978), and with growing conditions grossly unlike the natural forests (Neely, 1978). In the artificial system trees or shrubs may be released from the competition experienced in forests and may have the advantage of soil management, but the stresses of constrained root and aerial space, physical damage, chemical injury, and drought come into play. How these affect plant response to acidic precipitation is unknown.

In past research with gaseous pollutants, synergistic or antagonistic effects have been observed on vegetation resulting from the interaction of two or more pollutants (Unsworth and Ormrod, 1982). Acidic precipitation could also be involved in such interactions. In soybean chamber tests conducted at Yonkers, New York, the experimental design permitted researchers to compare the effect of simulated rain treatments in the presence and absence of ambient ozone. Although there was an increase in yield with increasing acidity in the "clean" air chamber, there was no such increase in the ambient air chamber; hence, the authors concluded that the presence of ozone negated potential gains in yield due to acid rain. Troiano et al. (1983) suggested that the reduction in soybean yield reported by Evans et al. (1981) might also have resulted from the interaction of simulated acid rain treatments with elevated concentrations of O_3 or even SO_2, as suggested by the experiments of Irving and Miller (1981). Inasmuch as New Jersey has ozone levels comparable to those occurring in lower New York State, plant response in this state could represent an interaction between acidic rain and ozone pollution. Saranac alfalfa, a cultivar that is commonly grown in New Jersey, was alternately exposed to ambient levels of O_3 and simulated rain of pH 5.6 and 3.0 in greenhouse experiments in 1981-82. Increased rain acidity did not reduce the yield of this particular cultivar nor did O_3 and acid rain have a synergistic effect on yield (Rebbeck and Brennan, 1984).

Experience with air pollutants in general has made researchers aware that not only direct effects, but also indirect effects, can stem from plant contamination. If acidic precipitation conforms to that model, then it may well affect plant-pathogen and plant-symbiont relationships, and perhaps plant-pesticide and

plant-insect interactions. At this writing the few experiments that have been done with diseases of crop plants have required an acid rain simulant of pH 3.2 to influence the disease reaction (Bruck, 1984). A consensus has not been reached on the effect of acidic precipitation, even at pH 3.2, on a symbiont of leguminous crops. Shriner (1978) reported a decrease, and Heagle (1983), no effect in nodulation. Regarding pesticides, acidic precipitation could have an adverse effect either by rendering a chemical less effective against its target or more toxic to the plant. Little information is available except for a paper indicating that increased acidity causes increased loss of triphenyltin hydroxide from leaf surfaces (Troiano and Butterfield, 1984). Lastly, insect populations on crop plants might be affected, but no information on this subject is at hand.

In summary, it can be stated that experimental evidence obtained in various laboratories in the United States indicates that rainfall of pH 4.0 does not generally have an adverse effect on agriculture crops. The standard practices of incorporating over 150,000 tons of lime/year into New Jersey soils tends to mitigate any direct effect of rainfall on soil pH. Although experimental evidence indicates that sulfur and nitrogen contained in rainfall can have a beneficial effect on agricultural crops that are nutrient poor, this condition is not likely to occur in New Jersey agricultural soils. There is insufficient evidence at this time to evaluate the impact on agricultural crops of acidic precipitation in combination with gaseous and particulate pollutants that occur in urban states of the Northeast. There is also insufficient evidence to evaluate the impact of acidic precipitation on important ornamental plants of New Jersey.

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CHAPTER III: GROUND-WATER PROCESSES

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EFFECTS OF ACIDIC DEPOSITION ON GROUND WATER

INTRODUCTION

The potential effects of acidic deposition on ground water are influenced by the chemistry of the atmospheric deposition, the nature of the soil and geologic material through which precipitation infiltrates, and the dynamics of the subsurface flow system.

The concentration of atmospheric contaminants and the amount of precipitation falling on the land surface define the input of acidic deposition to the hydrogeologic environment. The chemistry of atmospheric deposition was discussed in Chapter I. The susceptibility of soil or geologic material to acidic deposition is related to the reactivity of the material and to changes in the pH and alkalinity of the water moving through the material (McFee, 1980). Cation leaching from these materials may cause changes in solution composition or pH. The susceptibility of soil or geologic material to acidic deposition may be measured by cation exchange capacity, base saturation, and sulfate adsorption capacity; the susceptibility of surface or ground waters to acidic deposition may be measured by alkalinity.

Cation exchange capacity is the sum of exchangeable cations in a given quantity of soil or geologic material. In most soils, significant cation exchange capacity is derived from organic matter and clay, while the cation exchange capacity of subsurface geologic material is derived primarily from clays. The greater the exchangeable basic cations, the greater is the ability of a medium to neutralize acidic deposition.

Base saturation is the fraction of the cation exchange capacity derived from the basic cations. Of the exchangeable cations within a soil or geologic material, calcium, magnesium, sodium, and potassium are basic cations, while hydrogen ion and aluminum are acidic cations. The larger the base saturation, the greater is the ability of the medium to neutralize acidic deposition.

The capacity of a medium to adsorb sulfate is an important measurement in determining the sensitivity of a material to acidic deposition. Cation leaching can occur only in the presence of mobile anions. Because sulfate is the dominant anion in acidic deposition, the mobility or retention of sulfate may strongly influence leaching of cations from soil and geologic material. Materials which exhibit an ability to adsorb sulfate are less susceptible to cation leaching (McFee et al., 1983).

The dynamics of the ground-water flow system (direction and velocity of ground-water flow as it moves through the saturated and unsaturated zones) influences the destination of acidified water. The most direct route of water movement is the infiltration of precipitation directly to the water table. Precipitation interacts with soil and geologic material as it moves toward the water table. Depending upon the chemistry of the solid material contacting precipitation, mineral weathering, ion exchange, leaching, and complexation reactions may occur. Flow paths may permit the recharge of surface-water bodies by ground-water flow. Trace metals dissolved in acidified ground water may be

transported to lakes and streams. Surface-water bodies may act as reservoirs to the ground water, permitting acidified surface water to move into the ground water. Interaction between acidified water and geologic materials may dissolve trace metals such as aluminum and manganese, causing a net export from a basin. The flow path through which precipitation moves and the reaction time with soil and geologic material affect the chemical reactions that will occur.

Because of the complexities of subsurface flow systems, knowledge of surface water-ground water relationships is important in understanding how acidic deposition may affect the quality of ground water.

SUSCEPTIBILITY OF THE HYDROGEOLOGIC ENVIRONMENT TO ACIDIC DEPOSITION

The ability of soil and geologic material to assimilate or neutralize acidic deposition is controlled by the chemical composition, the reactive surface area and the hydraulic properties of the medium.

Media containing carbonate minerals (such as calcite and dolomite) or organic matter exhibit a very high buffering capacity (McFee et al., 1983). Sandstones and shales exhibit medium to low buffering capacity, and acidification of water bodies within these areas would be restricted to headwater streams and small lakes (Hendrey et al., 1980). Granitic gneisses (because of extremely slow reaction rates) and quartz sandstones exhibit little or no measureable buffering capacity (Hendrey et al., 1980).

Soil and geologic material which contain neither carbonate minerals nor organic matter may leach major cations, iron and aluminum, when exposed to acidic pore water. Cations may accumulate in the ground water or they may be exported from the watershed (McFee et al., 1983).

The reactive surface area of a material exerts a control on the assimilation of acidic deposition. Impermeable bedrock with few fractures or joints would have minimal contact with acidic deposition. Little change in the geochemistry of the water passing through the bedrock might be observed. Unconsolidated sand and gravel-sized aluminosilicate minerals would provide a significant reactive surface area for acidic deposition. The geochemistry of the water passing through these materials may be significantly altered.

The hydraulic properties exert additional controls over the ability of a medium to assimilate acidic deposition. The flow characteristics of the medium influence the geochemical reactions which occur. The permeability of a medium and the hydraulic gradient determine the contact time between the medium and the water percolating through it. For the same gradient, the lower the permeability, the greater the residence time of water within the medium. Slowly occurring chemical reactions such as those associated with chemical weathering are favored by long residence times (N. M. Johnson et al., 1981). The geochemistry of the pore water is more likely to be altered if such reactions occur.

The thickness of the media through which acidified water passes also influences assimilation. Hydrogeologic units that are very thin provide less

volume of media for interaction with acidified water. Consequently, the assimilative capacity of a small volume of media is less than that of a thick water-bearing zone or aquifer.

Several investigators have attempted to map regions of soil and bedrock that may be susceptible to acidic deposition. Maps of soil regions susceptible to acidic deposition have been determined primarily by measuring cation exchange capacity (McFee, 1980) and alkalinity (Omernik and Powers, 1982). Hendrey et al. (1980) identified regions predicted to be susceptible to acidic deposition, based on bedrock geology maps.

In certain areas of northwestern and north central New Jersey, a major component of the subsurface geology consists of certain types of crystalline bedrock, which exhibit little buffering capacity. Ground water flow in crystalline bedrock is primarily along joints and fractures and is fairly rapid. Acidified ground water may pass through crystalline bedrock relatively unaltered chemically, and the bedrock may act as a conduit for the movement of acidified ground water.

Unconsolidated sediments, such as silts and sands, are a major component of the geology in the New Jersey Coastal Plain. These geologic materials are very susceptible to acidic deposition, because they are generally low in weatherable minerals and have low base saturation and cation exchange capacity.

CHEMICAL MECHANISMS INVOLVED IN THE ACIDIFICATION OF GROUND WATER

The primary chemical process by which acidic deposition may alter ground water involves the leaching of cations. The source of the cations may be atmospherically-deposited trace metals or components of soil and mineral surfaces.

Atmospheric deposition may provide a significant influx of lead, cadmium, zinc and other trace metals to the land surface. In areas sensitive to acidic deposition, the soil water and ground water pH may be low enough to favor the dissolution of trace metals (Hultberg and Wenblad, 1980). Previously deposited trace metals may dissolve in the precipitation as it infiltrates toward the ground water. Significant concentrations of trace metals may be transported in this manner from the land surface into the ground water system.

Cations associated with mineral surfaces may be involved in ion exchange and adsorption reactions. Base cations such as calcium and magnesium may be exchanged for hydrogen where the influx of acidic deposition is large. Base cations as well as aluminum and other trace metals may be exported from a particular soil horizon or geologic strata under the influence of acidic deposition. Cronan and Schofield (1979) demonstrated the effect of acidic deposition on increasing the export of aluminum from a subalpine environment. N. M. Johnson et al. (1981) also reported that acidic waters release dissolved aluminum from the soil zone in the Falls Brook watershed (in the Hubbard Brook Experimental Forest, New Hampshire).

The influx of mobile anions and hydrogen ions is necessary for cation leaching to occur (Ulrich, 1980). In atmospheric deposition in the northeastern United States, hydrogen ion is the dominant cation and sulfate is the dominant anion. Cronan (1980) pointed out that sulfate was the dominant anion for solutions infiltrating through strata affected by acidic deposition. He hypothesized the importance of sulfate in leaching aluminum from soil water to ground water.

For cation leaching by sulfate to occur, the sulfate adsorption capacity of the soil or geologic material should be low. Soils and geologic materials which exhibit an ability to adsorb sulfate may receive greater inputs of sulfuric acid from atmospheric sources before significant pH or percent base saturation changes occur. The capacity for different soils and geologic materials to adsorb sulfate is largely unknown, although some soils (Ultisols, Oxisols, Alfisols) may exhibit some sulfate adsorption capacity (McFee et al., 1983).

Sulfate adsorption capacity increases when the amount of amorphous iron and aluminum oxides or hydroxides in a medium is large (McFee et al., 1983). As pH decreases, sulfate adsorption capacity tends to increase. However, the presence of organic matter decreases the sulfate adsorption capacity, even when the iron and aluminum oxide content is high (McFee et al. 1983; Johnson et al. 1979, 1980). The concentration of sulfate in solution also affects the sulfate adsorption capacity of a medium, that is, sulfate adsorption is concentration-dependent (McFee et al., 1983).

If the ability of a soil or geologic material to adsorb sulfate is surpassed, there should be free movement of sulfate (accompanied by cations) to lower soil horizons and/or geologic strata.

The transport of deposited trace metals to the ground water may be aided by the presence of soluble organic matter. In forested areas, inland bogs or swamps, there is generally an abundance of soluble organic matter, primarily fulvic acids and humic acids. The organic matter may act as organic anions, providing chemical bonding sites for complexation with trace metal cations.

In areas where atmospheric deposition of trace metals occurs, the formation of organo-metallic complexes may accelerate the transport of lead and other trace metals to ground water.

OBSERVED EFFECTS OF ACIDIC DEPOSITION ON GROUND WATER

Little information is available to document the effects of acidic deposition on ground water in the United States primarily because data collection programs have not been established for this purpose. Documented effects of acidic deposition on potable ground-water supplies have come primarily from Swedish research. In Sweden, long term data collection programs have demonstrated regional changes in ground-water chemistry in response to acidic deposition. Hultberg and Wenblad (1980) examined data for a period of 10 to 20 years from nine shallow wells (10 to 20 m) in sandy soils and bedrock areas. They observed a decrease in bicarbonate concentrations over time as well as an increase in

calcium and magnesium concentrations. The observed increase in calcium and magnesium may indicate that acidic deposition is affecting soils above the ground-water zone by leaching out base cations and replacing them with hydrogen ions.

In a study of ground-water quality near a sulfur point source, Hultberg and Wenblad (1980) indicated that acidification of ground water occurred as the result of high sulfur input (as sulfate and sulfur dioxide) to the soil and changes in the ground-water hydrology. In soils containing abundant iron and organic matter, sulfur deposition accumulated as pyrite in the lower soil horizons. After two consecutive dry years, the ground-water levels dropped considerably. As a result, sulfide was oxidized to form sulfuric acid, and heavy leaching of aluminum, manganese, calcium, magnesium, silica, and sulfate occurred.

Hultberg and Johansson (1981) conducted a regional study on the occurrence of acidified ground water in an area of southwestern Sweden where acidified lakes have been documented. In the province of Bohuslan, well depth was correlated with observed acidity in ground water; as depth decreased, acidity increased. Forty-nine percent of the shallow wells had a pH less than 5.5, and aluminum concentrations increased sharply as pH decreased. Copper from water pipes was found in samples of tap water from 6 of 10 wells at greater than 1.0 mg/l where the pH range was 5.50-5.95. Taste problems and diarrhea in young children have been attributed to the high concentrations of copper. Thirteen percent of the home owners complained of corrosion of copper water pipes with several homes experiencing corrosion through the walls of the copper piping.

Hultberg and Johansson (1981) studied the effects of lime treatment on two acidified lakes in southwestern Sweden. A lime treatment applied to the lakes was calculated to last approximately 10 years, based on acidic deposition rates and the lake's hydrology. After four years of treatment, however, there was a rapid decrease in pH and buffering capacity of the two lakes. A subsequent geochemical study showed that an inflow of acidified ground water accounted for the re-acidification.

EFFECTS OF ACIDIC DEPOSITION ON GROUND WATER IN SOUTHERN NEW JERSEY (PINELANDS)

A. Hydrogeochemistry of the Pinelands

1. Geology

The New Jersey Pinelands lie within the Atlantic Coastal Plain physiographic province, which is composed primarily of marine and marginal marine, unconsolidated quartzose gravel, sand, silt, and clay strata (Rhodehamel, 1979a). Figures 15a and 15b illustrate the location of the Pinelands' boundries, the two most studied watersheds (McDonalds Branch and Oyster Creek), and a profile of the areas' hydrology. Most of the region is underlain by the Cohansey Sand (up to 76 m of quartz sand with local clay and gravel beds) of Tertiary age. The Cohansey Sand consists predominantly of limonitic quartz sand with small amounts of weathered feldspars and local lenses of kaolinitic clay (Rhodehamel, 1979a). In minor parts of the uplands, the Cohansey Sand is unconformably overlain by thin (approximately 6.1 m) deposits of the Beacon Hill Gravel (Tertiary age).

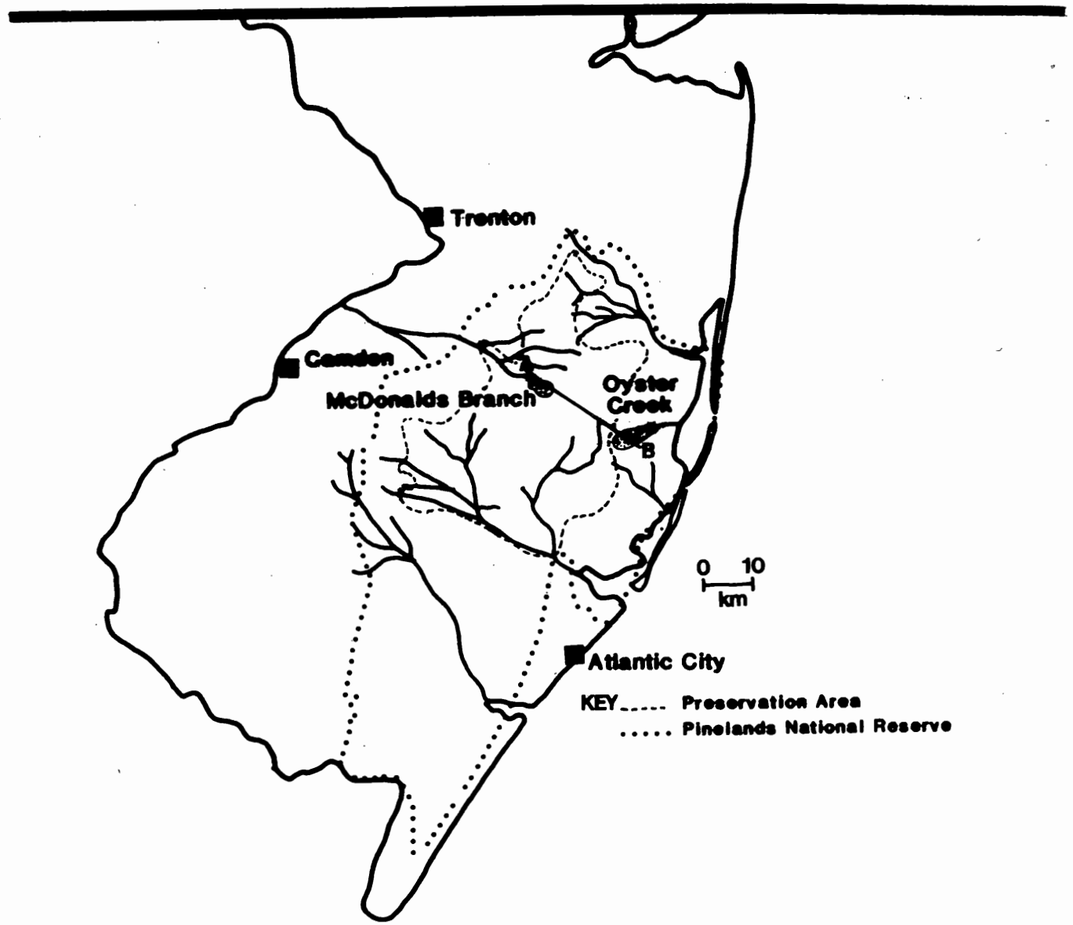
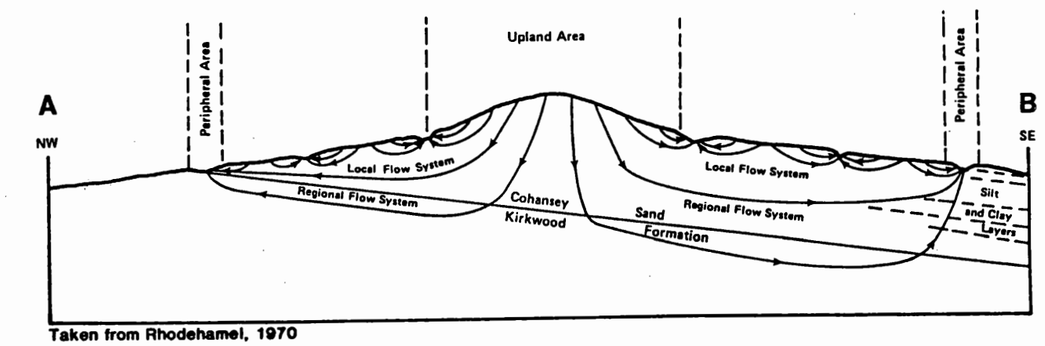


Fig. **15a** Location of longterm pH monitoring at McDonalds Branch and Oyster Creek in the Pinelands. Shading indicates watersheds. For cross section see below.



Taken from Rhodehamel, 1970

Fig. **15b** Idealized cross section showing ground-water flow patterns in Pinelands region.

2. Soils

The soils which have developed on the Cohansey Sand are highly leached, extremely acid to very strongly acid (pH ranging from 3.6 to 5.0) soils (Markley, 1979). Upland soils are generally well drained to excessively well drained; soils occupying intermediate positions in the landscape and lowland soils are moderately well drained to very poorly drained.

The sand fractions of Pinelands' soils are composed of greater than 90 percent quartz, contain minor feldspar and less than one percent muscovite (Douglas and Trela, 1979). Silt fractions of the soils are also dominated by quartz and contain limonite, some feldspar and mica, or kaolinite and vermiculite. Predominant clay minerals are kaolinite, gibbsite, and small amounts of muscovite which alters through dioctahedral vermiculite to aluminum-interlayered vermiculite or aluminum-interlayered smectite. The dominance of quartz and the small amount of clay in the Pinelands' soils result in a low cation exchange capacity, ranging from 1 to 14 meq/100 g, but generally well under 10 meq/100 g (Markley, 1979; Douglas and Trela, 1979).

The sulfate adsorption capacity of the Pinelands' soils is not known. Some sulfate adsorption is common in Ultisols and Entisols (McFee et al., 1983; Singh et al., 1980), but sulfate adsorption is reportedly low in Spodosols (Johnson and Todd, 1983). Pinelands' soils are roughly 35 percent Ultisols, 30 percent Spodosols, 20 percent Entisols, and 10 percent Histosols (highly organic soils) (Markley, 1979; Markley, 1971; Douglas and Trela, 1979). It is likely that the sulfate adsorption capacity of Pinelands' soils is low to moderate.

The effect that acid deposition has on soils is determined not only by soil chemical properties (such as buffering capacity, pH, sulfate adsorption capacity, and presence and composition of weatherable minerals), but is also strongly influenced by soil physical properties (McFee et al, 1983). Water flow is the most important physical property that influences the effect of acid deposition on soils. Water passing through sandy, well-drained soils such as those found in the Pinelands may be changed only slightly; thus these soils may be described as "sensitive" in terms of soil buffering capacity (McFee et al, 1983).

3. Surface and Ground Waters

Surface and shallow ground waters in the Pinelands are acidic (pH ranging from 3.5-5.5) and have low alkalinity (Means et al., 1981). They are low in total dissolved solids (TDS), with streams ranging from 20-35 mg/l TDS and ground waters ranging from 25-100 mg/l TDS. Surface waters are often brownish in color and contain considerable concentrations of dissolved organic carbon (up to 30 mg/l soluble organic carbon). Ground-water quality data collected by the U.S. Geological Survey (USGS) in Wharton State Forest in 1978 showed that shallow ground water (less than 50 feet deep) was acidic (pH 3.8 - 5.6), low in total dissolved solids (6.0 - 81.0 mg/l), and low in trace metals (see Table 18). Arsenic, cadmium, cobalt, lead, and nickel are the trace metals of most concern because of their toxicity. The maximum concentrations are about 15 ug/l (15 ppb); median concentrations are 5 ug/l or less.

TABLE 18

STATISTICAL SUMMARY OF GROUND-WATER QUALITY IN THE COHANSEY-KIRKWOOD FORMATION WITHIN WHARTON STATE FOREST, 1978

[Chemical constituents are reported as dissolved in mg/l unless otherwise noted]

	Number of Samples	Median	Minimum Value	Maximum Value
Alkalinity, Total (as $CaCO_3$)	35	1.0	1.0	10
Solids, Residue at 180°C	38	23	6.0	81
pH	38	4.6	3.8	5.6
Color (Platinum cobalt units)	38	10	5.0	400
Nitrogen, Nitrate	38	0.02	0	0.43
Nitrogen, Ammonia	38	0.01	0.01	0.23
Carbon, Organic	37	1.2	0	18
Calcium	38	1.2	0.2	7.8
Magnesium	38	0.60	0.2	1.9
Sodium	38	1.7	1.2	7.5
Potassium	38	0.35	0.2	1.8
Chloride	38	3.3	0.2	11
Sulfate	38	9.2	0.4	45
Silica	38	3.45	1.1	9.3
Arsenic (ug/l)	38	1.0	1.0	2.0
Cadmium (ug/l)	30	2.0	2.0	5.0
Cobalt (ug/l)	38	2.0	2.0	10
Iron (ug/l)	38	20	10	7000
Lead (ug/l)	30	2.0	2.0	13
Manganese (ug/l)	38	30	10	230.
Nickel (ug/l)	38	2.0	2.0	15
Zinc (ug/l)	38	20	20	70

(USGS, unpublished data)

4. Hydrology

The hydrology of the Pinelands region is strongly influenced by ground-water flow patterns. Rhodehamel (1970) estimated that approximately 89 percent of the total annual stream discharge within the Pinelands is derived from ground water, i.e. water reaching streams first flows through soils and shallow ground water. The principal ground-water reservoir in the Pinelands is the Kirkwood-Cohansey aquifer which is primarily a water-table, or unconfined, aquifer. Variation in precipitation causes fluctuations of the water levels within the aquifer system; the amount of water stored in the aquifer at any given time is a function of precipitation frequency. The average water levels, however, provide a subdued image of the topography sloping from elevation highs toward low-lying areas and major streams. Depth to the water table varies from about 18.3 m (60 ft) in the uplands to within centimeters of the surface in the lowlands.

An average annual precipitation of 114.3 cm falls in the Pinelands, of which approximately 50 percent is lost through evapotranspiration (Rhodehamel, 1979b). The remaining water percolates rapidly through the sandy soils, recharging the ground water. Rhodehamel (1979b) calculated a long-term hydrologic budget for the region: precipitation (114.3 cm) = interception (15.0 cm) + evapotranspiration from undrained depressions (2.3 cm) + evapotranspiration from soil and ground water (39.9 cm) + direct runoff (6.3 cm) + ground-water runoff (50.8 cm).

B. Potential Effects of Acidic Deposition on Ground Water in the Pinelands

1. Consequences of sulfate mobility on cation leaching from soil, ground, and surface waters.

The following discussion has three purposes. Initially, we review the effects of acidic deposition on soil, ground, and surface waters in parts of the northeastern United States (remembering that, in many ecosystems, reactions occurring in the soil zone may strongly influence ground and surface waters). Secondly, we briefly review the importance of the ground-water contribution to surface water in the Pinelands. Finally, we compare the scant data available on soil, ground, and surface waters in the central Pinelands with published literature on acidic deposition effects and, based on these data, postulate effects that may be occurring in the Pinelands.

Cronan and Schofield (1979) compared an acid-deposition-affected forest soil in the Northeast (Mount Moosilauke, New Hampshire) to an unaffected northwestern (Findley Lake, Washington) forest soil (see Figure 16). They found that the anionic composition of the soil solution in northeastern forest soil was dominated by sulfate, whereas the anionic composition of the northwestern soil was organic-anion dominated. In the Findley Lake soil, organic anions complexed aluminum in upper soil horizons. These complexes were transported down into lower soil horizons where organic anions were used up and aluminum was precipitated. By contrast, in the Mount Moosilauke soil, where the solution was sulfate-dominated, the low pH of the solution apparently caused aluminum to remain dissolved. In a sulfate-dominated solution, because the charge balance must be maintained, when sulfate moves through the soil solution, it must be accompanied by cations (in this case, hydrogen ion and ionic aluminum). As a

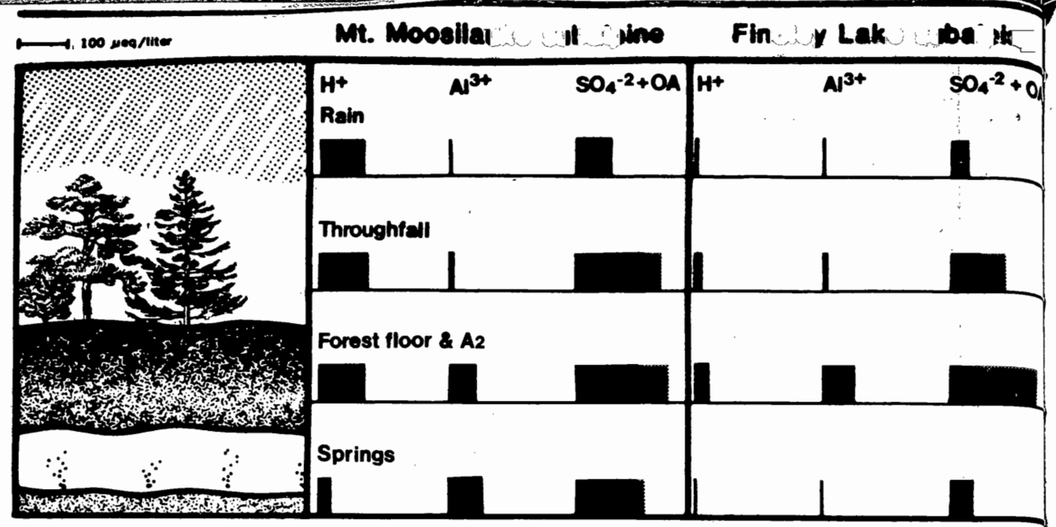


Figure 16 Average trends of change in the ionic concentrations of free hydrogen, aluminum, fate, and organic anions (OA) as rainfall penetrates two contrasting ecosystems, both of wh exhibit podzolized soils: the H_2SO_4-dominated soils of the Mount Moosilauke subalpine z and the organic acid-dominated soils of the Findley Lake watershed in Washington. The aluminum concentrations in each case actually represent the summed equivalents of three ionic species: Al^{3+}, $AlOH^{2+}$, and $Al(OH)_2^+$. The concentration of OA is indicated by shaded portion of the bars in the SO_4^{2-} + OA columns. A certain percentage of the alumin and OA equivalents may exist together in solution as complexes. Modified from Cronan & Schofield,

result, aluminum was not precipitated in lower soils horizons but was transported to ground water.

N.M. Johnson et al. (1984) also discuss the effects that acidic deposition can have on the chemistry of acid soils. They stress that the impact acidic deposition can have on a soil depends on both the amount of acid present and the type of acid anions present. They further state that "the kind of acid present has a profound effect on certain chemical reactions taking place in the soil zone and in the waters draining the soil." They compare the chemistry of two streams draining watersheds with similar soil types, developed on glaciated granitic terrain. Jamieson Creek, near Vancouver, British Columbia drains a natural, unpolluted area. Falls Brook, New Hampshire, by comparison, is located in an area which has been affected by acidic deposition.

The chemical analyses of these two streams are given in Table 19; Johnson et al. (1984) indicate that the headwaters of Jamieson Creek are acidified by organic acids and that farther downstream, these acids are replaced by carbonate alkalinity. In Falls Brook, however, the pH of the stream is controlled by sulfuric and nitric acids. They conclude that the source of hydrogen ion in Jamieson Creek is from organic and carbonic acids, but that the source of at least half of the hydrogen ion in Falls Brook is from acidic deposition. Johnson et al. further state that the difference in the kind of acid dominating each system results in a change in dissolved aluminum speciation. In Jamieson Creek (dominated by weak acids), aluminum is present largely as organic complexes. In Falls Brook, however, because the pH of the stream is controlled by strong acids, dissolved aluminum is present predominantly as free aluminum, aluminum hydroxide and fluoride complexes (Johnson et al., 1981).

Data on the effects of acidic deposition on ground-water quality in the Pinelands are scarce. Rhodehamel (1970), however, estimated that 89 percent of the surface water in the Pinelands consists of ground-water discharge. An idealized cross section of the ground-water flow patterns in the Pinelands (see Figure 15b) provides an indication of the overwhelming contribution of ground water to the surface water.

Water enters the Pinelands ecosystem as precipitation. It passes through the forest canopy, into soils and ground water, and ultimately discharges to surface-water bodies. The water table in the Pinelands is the surface of the unconfined aquifer under atmospheric pressure (Rhodehamel, 1979b). At various locations in the Pinelands, the water table is exposed at the surface in the form of swamps, streams, and rivers.

Because of the hydrology in the Pinelands and, specifically, the large ground-water component of surface water, it is important when considering acidic-deposition effects on Pinelands' ground-water chemistry to also examine soil- and surface-water chemistry. Data on both soil and surface waters in the central Pinelands are now reviewed and compared to those data previously discussed from the northeastern studies (Cronan and Schofield, 1979 and Johnson et al., 1984). Several Pinelands' ground-water analyses are then presented and related to surface-water chemistry.

The concept that mobile sulfate, when accompanied by hydrogen ion, can cause cation leaching from soils has appeared frequently in the literature (e.g., McFee et al., 1980; Cronan and Schofield, 1979; Christophersen and Wright, 1981). The importance of the sulfate adsorption capacity of a soil with respect to the mobility of sulfate is clear. Laboratory experiments were conducted for the U S G S on four major soil series in the Pinelands (Lakewood, Evesboro, Lakehurst and Berryland). The preliminary data indicate that these soils have a relatively low sulfate adsorption capacity and that they are saturated with sulfate, i.e., further adsorption was not observed with additional sulfate inputs (Schuster and Uchrin, 1985). It appears that in the Pinelands hydrologic cycle, atmospherically deposited sulfate may pass through the sandy soils relatively unaltered, liberating both acid and base cations to the soil solution, shallow ground and surface waters, particularly during wet periods.

Turner et al. (1983) present data from soils and the soil solution in McDonalds Branch basin (see Figure 15a). The acid soils (predominantly Spodosols and Entisols) in the watershed have developed on quartz sand with minor gravel and clay lenses. The data indicate that aluminum concentration in the soil solution increases with depth in the soil profile and reaches a maximum in the C horizon (Turner et al., 1983). They found that aluminum was transported from the forest floor into the A and B horizons and was associated with organic acids and probably sulfate. In the B horizon, organic matter was either deposited or metabolized. Aluminum concentrations, however, increased through the B and C horizons. Turner et al. (1983) also noted that aluminum concentrations in the soil and surface waters were highest in the winter and spring (wet periods). Aluminum transport through the lower soil horizons was apparently dependent on moisture fluxes and the concentration of mineral acids in the soil. The movement of aluminum through the B and C horizons in these Pinelands' soils may be similar to that described by Cronan and Schofield (1979) in the northeastern (acid-deposition affected) soil.

A chemical analysis of stream water from McDonalds Branch is shown in Table 19. The stream has a low pH (mean value of 4.2 from 1973 through 1981) and low alkalinity (mean value of 0.01 meq/l from 1966 through 1981) (Smith and Alexander, 1983). The chemical analysis shown in Table 19 illustrates that the anionic composition of the solution is dominated by sulfate (131 ueq/l) with organic anions contributing only 27 ueq/l to the total anionic charge. The dominant cation in the solution is hydrogen ion (99 ueq/l). It appears from this analysis, then, that this solution is dominated by sulfuric acid, not organic acids. It should be noted that there are natural, as well as anthropogenic, sources of mineral acidity in this system. Processes such as iron oxidation, pyrite oxidation, (Means et al, 1981) and biological exchange contribute both hydrogen and sulfate ions to the system. Acidic deposition, however, contributes an additional, and probably a major, amount of hydrogen and sulfate ions to this system. A further discussion of McDonalds Branch surface water appears in Chapter IV: Aquatic Processes.

A comparison of trends in stream chemistry at USGS hydrologic Bench-mark stations to trends in SO_2 emissions for a 10- to 15-year period was performed by Smith and Alexander (1983). Their study indicates that between 1965 and 1980, SO_2 emissions declined substantially in the northeastern United States (Gschwandtner and Gschwandtner, 1983). Smith and Alexander reported that between 1966 and 1981, sulfate concentrations in McDonalds Branch also declined (slope = -2.9 ueq/l/yr, significance level = 0.034), and pH showed no significant trend from 1972 through 1981. They concluded that these analyses support the hypothesis that stream sulfate trends reflect regional trends in sulfur deposition rates.

The lack of long-term ground-water quality records for the New Jersey Pinelands prevents a similar trend analysis of ground water. However, comparison of ground-water quality to surface-water quality analyses may be useful. Because McDonalds Branch surface water appears to be dominated by sulfuric acid and the soils are probably saturated with sulfate, it is probable that sulfate is being transported to ground water. Although there is a lack of ground-water data to confirm or dispute this hypothesis, typical shallow ground-water analyses (Table 20) indicate that the ground water, like surface water, is characterized by low pH and relatively high sulfate. DOC in the ground water is low and contributes very little to the charge balance (less than 20 ueq/l); sulfate is the major anion, contributing 162 ueq/l (Mullica 27S) and 179 ueq/l (Mullica 52S). For the cations, hydrogen ion contributes much less to the charge balance in ground water than in the surface water. In the ground water, calcium is the major cation (80 and 95 ueq/l for the two wells) instead of hydrogen ion.

Although ground-water data do not exist at present for long-term trend analysis, the chemical composition of surface and ground waters is such that changes in chemistry from acidic deposition appear to be possible. It is important to remember that ground water cannot be isolated from soil and surface waters. These components of the system are interrelated; acidic-deposition-induced changes in soil waters may ultimately affect ground and surface waters.

TABLE 19

Analyses of stream water from Jamieson Creek, British Columbia, Falls Creek, New Hampshire (source: Johnson et al., 1984), and McDonalds Branch, Lebanon State Forest, New Jersey (source: USGS). Analytical results are given in milligrams per liter (mg/l) and microequivalents per liter (ueq/l)[1]. DOC means dissolved organic carbon.

CHEMICAL CONSTITUENT	JAMIESON CREEK JULY 23, 1982		FALLS BROOK MARCH 6, 1979		MC DONALDS BRANCH JANUARY 24, 1980	
	pH = 4.71		pH = 4.75		pH = 4.0	
	mg/l	ueq/l	mg/l	ueq/l	mg/l	ueq/l
H^+	.02	19	.02	18	.10	99
Ca^{2+}	.29	14	.72	36	.6	30
Na^+	.40	17	.43	19	1.8	78
K^+	.13	3	.11	2.8	.6	15
Mg^{2+}	.05	4	.19	16	.4	33
NH_4^+	.05	2.8	.01	.6	n.d.	n.d.
SO_4^{2-}	1.0	21	5.3	110	6.3	131
NO_3^-	.03	.5	1.56	25	.09	1.5
Cl^-	.42	12	.62	17	2.8	79
DOC (OA^-)[5]	4.8	34	2.5	18	5.0[4]	27
Al (total)	.30	n.d.[2]	.61	26-65[3]	n.d.	
SiO_2	3.9		2.2		3.2	

1 Microequivalents per liter were calculated from analyses given in mg/l.

2 n.d. means "no data."

3 A range was calculated from data given by Johnson et al. (1981) for Al^{3+} and soluble aluminum-hydroxide, fluoride, and sulfate complexes.

4 No value for DOC was available; a value of 5.0 mg/l total organic carbon is reported instead.

5 The equivalent value for DOC was calculated as organic anion (OA^-) contribution using DOC and pH, a method described by Oliver et al. (1983).

TABLE 20

Analyses of ground water from shallow (less than 50 ft) wells from the Mullica River basin in Wharton State Forest (source: USGS). Analytical results are given in milligrams per liter (mg/l) and microequivalents per liter (ueq/l)[1]. DOC means dissolved organic carbon.

CHEMICAL CONSTITUENT	MULLICA 27S APRIL 25, 1978 pH = 4.5		MULLICA 52S FEBRUARY 16, 1978 pH = 4.9	
	mg/l	ueq/l	mg/l	ueq/l
H^+	.03	31	.01	12
Ca^{2+}	1.6	80	1.9	95
Na^+	1.5	65	1.2	52
K^+	.3	7.7	.6	15
Mg^{2+}	.4	33	.8	66
NH_4^+	0.0	0.0	0.0	0.0
SO_4^{2-}	7.8	162	8.6	179
NO_3^-	.09	1.5	.09	1.5
Cl^-	3.0	85	2.6	73
HCO_3^-	0	0	1	20
DOC $(OA^-)^3$	1.8	18	1.2	11
Al	n.d.[2]		n.d.	
SiO_2	3.2		5.4	

[1] Microequivalents per liter were calculated from analyses given in mg/l.

[2] n.d. - means "no data".

[3] The equivalent value for DOC was calculated as organic anion (OA^-) contribution using DOC and pH, a method described by Oliver et al. (1983).

2. Trace metal contamination

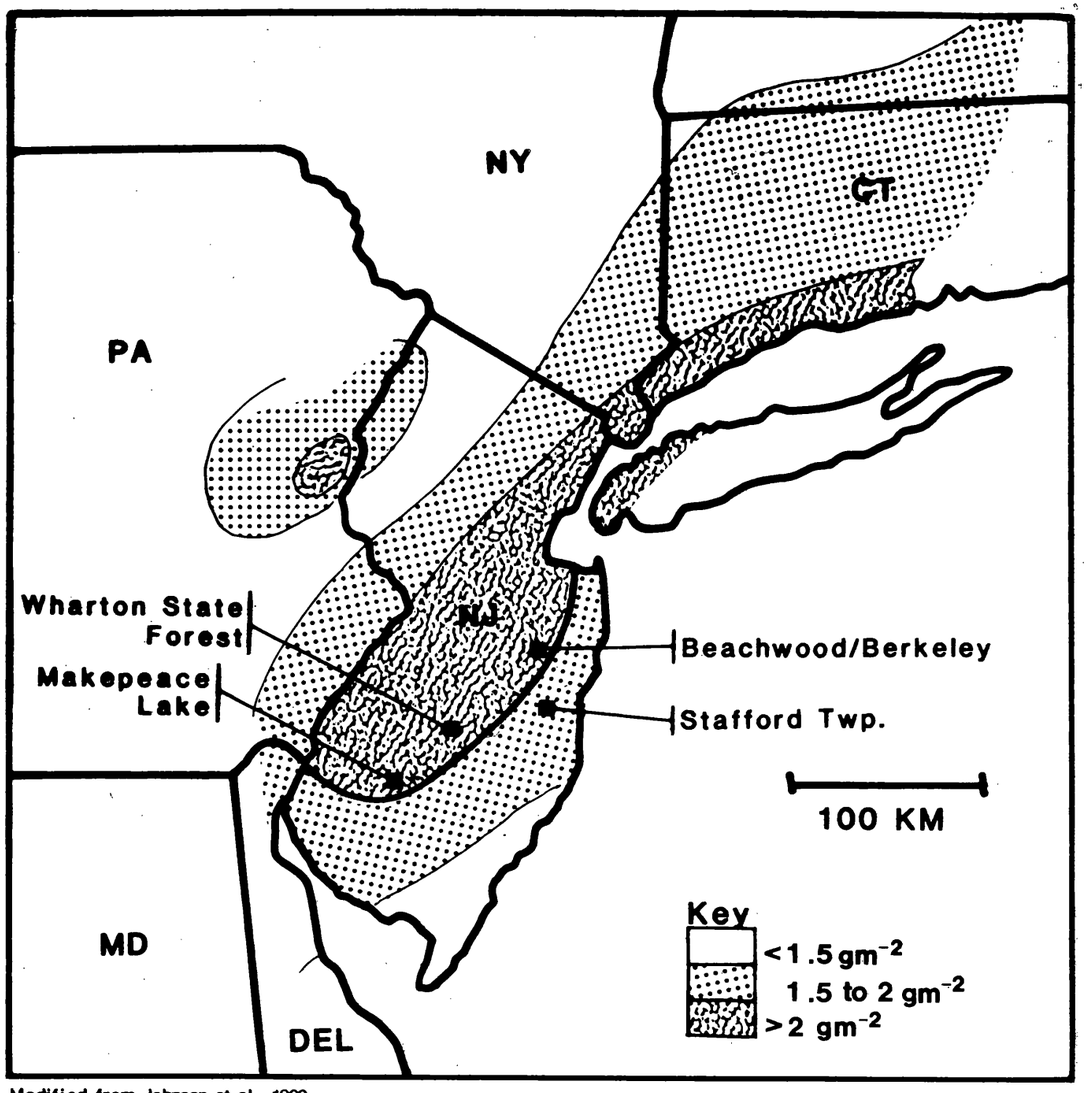
As industrial activity in the state has increased over the years, the atmospheric deposition of some trace metals of anthropogenic origin has occurred. Because the solubility of some trace metals increases with decreasing pH, the potential exists for contamination of acidic ground waters. The presence of trace metals such as lead, cadmium, copper and aluminum in ground water is of particular interest because of possible toxic effects on living organisms.

Trace metal mobilization in the soil solution or in ground water may be increased by several mechanisms. If the alkalinity of the ground water is decreased by acidic deposition, as has been observed in Sweden (that is, sulfate replaces bicarbonate as the dominant anion), pH may decline, resulting in increased solubility of certain trace metals (ie., aluminum, copper, and cadmium) and elevated concentrations of these metals in ground water. A second mechanism by which trace metal concentrations may be increased in ground water and/or the soil solution is through increased deposition of these metals to the land or vegetative surfaces. If the atmospheric deposition of trace metals is increased, these metals may be complexed by natural organic compounds or solubilized by mineral acidity which may increase the concentrations of these metals in solution.

Lead deposition (from atmospheric sources) to the forest floor has increased substantially during the twentieth century and may be 5 to 10 times greater than levels of 100 years ago (Johnson et al., 1982). However, lead from automotive emissions has been decreasing since the late 1970's as the lead in gasoline was controlled. Lead is strongly retained by the forest floor, especially in some areas of the New Jersey Coastal Plain (see Figure 17). Disturbance of the soil, within forested areas where soil lead content is elevated, can cause elevated lead levels in soil moisture and shallow ground water (Johnson, pers. comm., and U. S. Geological Survey, unpublished data).

Certain areas in the Coastal Plain have experienced lead contamination of domestic water supplies, probably by leaching of lead from plumbing systems by acidic ground water. Approximately 14 percent of the samples taken from taps of domestic wells (in a portion of the New Jersey Coastal Plain) in Ocean County showed lead concentrations above the drinking water standard of 0.05 mg/l (see Table 21). The average pH of ground water in the area is approximately 4.8. These wells have been shut down and the homes have been connected to the public water-supply system. Elevated lead concentrations have also been observed in localized areas in Stafford Township, Ocean County, and Makepeace Lake, Atlantic County; however, the cause of the contamination is unknown. These areas are within or near the area of greatest lead contamination in forest litter as described by Johnson et al. (1982).

Recent unpublished data from the U S G S seems to indicate that lead from solder used in the domestic plumbing systems is leaching into the water. Table 22 lists trace metal concentrations found in samples taken from household taps in the Beachwood Borough area. The data from Beachwood Borough indicates that a substantial reduction in trace metal concentration occurs when a household tap is permitted to run until water in the pipes is flushed out. Although the standing water concentrations of lead, copper and zinc are within drinking water



Modified from Johnson et al., 1982,

Fig. 17 Estimated lead content of the forest floor in the northeast United States (Modified from Johnson et al., 1982.). Shows locations within the New Jersey Coastal Plain where studies have been conducted to determine the lead content of ground water.

TABLE 21

STATISTICAL SUMMARY OF LEAD IN DOMESTIC WELLS FOR BEACHWOOD BOROUGH AND BERKELEY TOWNSHIP, NEW JERSEY

	TOTAL NUMBER OF WELLS SAMPLED	NUMBER OF WELLS BELOW 0.01 mg/L	NUMBER OF WELLS BETWEEN 0.01-0.05 mg/L	NUMBER OF WELLS ABOVE 0.05 mg/L STANDARD	PERCENTAGE OF WELLS ABOVE STANDARD
Beachwood	590	370	138	82	14
Berkeley					
Area 1	464	316	121	27	6
Area 2	133	111	21	1	1
Area 3	338	295	40	3	1

From Preczewski and Hayes, 1983 (unpublished data)

TABLE 22

CONSTITUENT CONCENTRATIONS FOUND IN SAMPLES FROM HOUSEHOLDS IN BEACHWOOD BOROUGH

Constituent	Number of Samples	Concentration (ug/L) Standing Mean	Standing Maximum	Running Mean	Running Maximum
Lead	7	27	89	8.7	17
Copper	7	146	650	63	270
Zinc	7	163	350	22	55
pH	7	5.3*	--	4.8*	--

* median

standards, there are significant reductions in the concentrations of lead, 27 ug/l to 8.7 ug/l; copper 146 ug/l to 63 ug/l and zinc 163 ug/l to 22 ug/l when the water is permitted to run. The data compares favorably with other areas where metal leaching from plumbing systems have been observed (Neff, 1984 and Lassovszky, 1984). Note that the pH of the water sampled during the running periods was below 5.

Previous investigators in other soft-water areas have shown a relationship between lead used in plumbing systems and lead dissolved in tap water. Beattie et al. (1972) in Glasgow, Scotland showed that lead concentrations in tap water averaged 358 ug/L in households where lead was used in the plumbing system. In a survey of tap water in Great Britain, Matthew (1981) found that 7.8%, 34.4% and 8.8% of the households in England, Scotland and Wales, respectively had lead concentrations greater than 50 ug/L. Karalekas et al. (1976) in a study of drinking water in the Boston area found that of 936 samples collected, 15.4% exceeded the federal drinking water standard of 50 ug/L.

In several states, elevated lead levels in tap water have been observed where copper pipes connected with lead-tin solder have been used. In addition to Massachusetts, the states of Oregon, Washington, Wisconsin, Minnesota, Delaware, and New York have reported problems of lead leaching from household plumbing. Three states, Oregon, Wisconsin, and Delaware have banned the use of lead-tin (50%/50%) solder in home construction. The recommended alternative in these states is a tin-antimony (95%/5%) solder normally containing less than 0.2 percent lead (N.E. Murrell, 1985, personal comm.).

Although conclusive evidence is not yet available in New Jersey, it appears that lead used in home plumbing systems may be a significant contributor to the elevated lead levels found in homes in the New Jersey Coastal Plain. Because the ground water is soft and acidic, the dissolution of trace metals from plumbing materials is favored. Further evidence to support the theory that plumbing materials are a source of lead in tap water is very low lead concentration in shallow ground water in Wharton State Forest (U S G S, unpublished data). Thirty-eight wells less than 50 feet deep were sampled in 1978 and only 15 samples showed lead concentrations above the analytical detection limit. The average concentration of lead in the 15 samples was 5.5 ug/l and the maximum value was 13.0 ug/l (see Table 18).

The U S G S is conducting a systematic ground-water sampling program to determine the distribution of lead in ground water in the New Jersey Coastal Plain. The role of acidic deposition as it relates to the transport of lead to ground water is being investigated as part of this research.

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CHAPTER IV: AQUATIC PROCESSES

Surface Water Quality -- Dr. Samuel D. Faust
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EFFECTS OF ACIDIC DEPOSITION ON SURFACE WATER QUALITY

A. INTRODUCTION

Whereas the impact of acidic deposition has been documented in various areas of eastern Canada and the northeastern United States, little is known about the effects of this environmental phenomenon on the quality and chemistry of New Jersey's surface waters (Faust and McIntosh, 1982). In 1980, a series of water quality studies were initiated in an effort to assess those lakes, rivers, streams, and reservoirs that may be succeptible to acidic deposition in New Jersey. A summary of these studies is given below.

In any surface water, several factors, including the nature of the bedrock geology, physical and chemical properties of soils, and other watershed characteristics will influence the sensitivity of water quality to acidic deposition. Sampling sites for New Jersey's waters were selected on the basis of its geology. Initially, pH and total alkalinity were determined on 31 rivers and streams and 32 lakes and reservoirs in northern New Jersey. Additional water quality analyses were performed on those waters that had alkalinities less than 30 mg/l as $CaCO_3$.

B. CHEMICAL ASPECTS OF SURFACE WATERS RECEIVING ACIDIC DEPOSITION

Before presenting the general effects of acidic deposition on surface water chemistry in New Jersey, a brief description is given of the sensitivity indicators: pH, alkalinity, acidity, and buffer capacity. These chemical parameters are employed by most researchers and others to evaluate the effects of acidic deposition on surface water chemistry and quality.

1. pH:

Mathematically, this parameter is equal to $-\log[H^+]$, where [ ] represents the molar concentration. The pH value (a logarithmic number) of pure water in equilibrium with gaseous CO_2 is 5.65 at 25°C. Sulfuric and nitric acids from the atmosphere lower the pH value of rain to approximately 4.0. Analytically, pH is a relatively quick and simple measurement (provided proper precautions are observed with calibration of the instruments).

2. Alkalinity:

Frequent reference is made in the acidic rain literature that total alkalinity is a measure of the buffer capacity of a natural water. This, simply, is not the case. Total alkalinity (TAlk) is a measure of the acid neutralizing capacity, in equivalents per liter (eq/l), of a water. In most natural waters, the carbonate system provides this neutralizing capacity:

$$C_t = [H_2CO_3] + [HCO_3^-] + [CO_3^{2-}] \qquad (1)$$

where C_t = mass balance or sum of molar concentrations of the three carbonate species from which the total alkalinity is expressed:

$$TAlk\ (eq/l) = [HCO_3^-] + 2[CO_3^{2-}] + [OH^-] - [H^+] \qquad (2)$$

where TAlk = the sum of major acid neutralizing chemical species in a natural water. Inasmuch as the pH values of most surface waters in New Jersey lie below 8.3, the total alkalinity is, for all intents and purposes, equal to the $[HCO_3^-]$. Any contributions of the $[CO_3^{2-}]$ and $[OH^-]$ are nil. Consequently, the bicarbonate alkalinity, reported frequently as mg/l (or ppm) as $CaCO_3$, is the capacity of a natural water to neutralize acidic deposition.

Another difficulty with reported alkalinities of natural waters lies with the incorrect analytical methodology often used for this parameter. Most researchers employ a fixed pH endpoint of 4.5 when waters are titrated for alkalinity. This is incorrect and results in an overtitration with an error of considerable magnitude (Faust, 1983).

3. Buffer Capacity:

Total alkalinity and pH values, considered individually, cannot give an accurate assessment of the impact of acidic deposition on a natural water. A trend of pH values with time towards more acidic values does not necessarily indicate that acidification is occurring. The same statement is valid for total alkalinity in natural waters. Rather, it is necessary to combine pH and total alkalinity into the buffer capacity concept in order to assess accurately and to calculate the capacity of a natural water's carbonate system to resist the effect of acidic deposition (Faust and McIntosh, 1983).

The buffer capacity (B) as originally defined by Van Slyke (1922) is the relation between the increment of a strong base or strong acid that causes an incremental change in the pH value of the water. If one solution has two times the buffer value of another, then twice as much base or acid is needed to change the pH value by a given quantity. Consequently, the buffer capacity of a natural water is a direct measurement of how much acidic deposition is required to lower the pH value by one unit. In the carbonate system cited above, the critical pH value is 6.3. Once a natural water reaches this pH value, its buffer capacity decreases rapidly to a minimum value below pH 5.0. Note that in this discussion, natural water assumes that other ions beside carbonates are low.

4. Acidity:

The total acidity (TAcy) of a natural water affected by acidic deposition is:

$$TAcy\ (eq/l) = 2[H_2SO_4] + [HNO_3] + 2[H_2CO_3] \qquad (3)$$

This equation shows the presence of the two "strong" acids from acidic deposition, H_2SO_4 and HNO_3, with natural acidity from the "weak" acid, H_2CO_3 (CO_2 acidity). The significance of the latter in natural waters can be estimated by comparison of the CO_2 acidity to the TAcy. Whenever the total and CO_2 acidities have the same value (usually expressed as mg/l or ppm as $CaCO_3$), the TAcy is the naturally-occurring, or carbon dioxide (CO_2, H_2CO_3), acidity. When the TAcy is greater that the CO_2 acidity, the difference represents acidic compounds other than H_2CO_3. Such compounds could be naturally-occurring organic acids (such as those found in the Pinelands of southern New Jersey), H_2SO_4 and HNO_3 from acidic deposition, and iron and other metallic ions.

C. General Effects of Acidic Deposition on Surface Water Chemistry

1. pH of Fresh Water Systems

Continued inputs of acidic deposition into a fresh water system will, of course, lower the pH value, with a concomitant increase of $[H^+]$, assuming that there is a limited or non-renewable source of bicarbonate and carbonate alkalinity for neutralization. One of the deficient areas in current research is in the attainment of a quantitative method for calculation of the rate of acidification of a fresh water system. One possibility is to use the buffer capacity concept cited above. The accuracy and precision of the concept in the prediction of acidification are seen from the calculation of the time needed to exhaust a lake's neutralizing capability. For example, it has been calculated that a mean $[H^+]$ of 70-100 ueq/l in one meter (m) of rain results in a loading of 70-100 meq H^+ per square meter per year (Dillon et at., 1978). For a lake with an alkalinity of 500 meq per square meter, a depth of 10 m, and specifying a pH of 7.0, this calculation yields a value of -0.211 eq $pH^{-1}\ m^{-2}$ (equivalents of acidity per pH unit per square meter). At a loading of 0.1 eq H^+ per square meter per year, 2.11 years are needed to lower the pH value to 6.0. Furthermore, three additional years are needed to lower the pH from 6.0 to 5.0. Thus, 5.11 years would be needed to lower the pH of this water from 7.0 to 5.0, provided, of course, that there is no additional input of alkalinity. That is, the value gives the precise equivalents of acid rain needed to lower the pH value by one unit in a carbonate system. The B-values are given in Table 26. From the information in this table and an estimation of the H^+ loading to the watershed, a rough approximation of the time required for the given pH reduction can be made assuming no neutralization is obtained as the rain water flows through the watershed to the lake. Actually calculating the rate of acidification of a lake due to acidic deposition is an exceedingly complex problem requiring detailed data on the lake, the acidic deposition, and the neutralizing factors which are present in the watershed.

2. Major Ionic Constituents

All natural waters (pollution excepted) contain dissolved constituents from attendant geologic formations and sediments. In terms of concentration, the "major" cations are Ca^{2+}, Mg^{2+}, Na^+, and K^+, whereas the major anions are HCO_3^-, SO_4^{2-}, NO_3^-, and Cl^-. Inasmuch as SO_4^{2-} and NO_3^- are also constituents of acidic deposition, it is tempting to use these anions as indicators of historic trends in the acidification of fresh waters. This is extremely difficult for several

reasons: (a) there are many anthropogenic sources of SO_4^{2-} and NO_3^-; (b) these two anions are the end products of decomposition of organic compounds containing sulfur and nitrogen; and (c) these two anions do not react conservatively in natural waters subject to biological activity of various descriptions. That is, SO_4^{2-} and NO_3^- may be used as electron acceptors in various biological processes and become reduced. Consequently, any historic trends towards an increase in their water concentration would be affected. However, if SO_4^{2-} and NO_3^- contents predominate and the pH value of the water is "low" (less than 6.3) or remains "low", then a qualitative statement may be made about the effect of acidic deposition on the system. These two ions indicate the presence of acidic deposition. Table 25 gives the sulfate contents of several surface waters in New Jersey for the year 1981.

3. Trace Elements

Much concern has been expressed about the release of such metallic ions as Al^{3+}, Pb^{2+}, Hg^{2+}, Zn^{2+}, Cd^{2+}, etc., due to the acidification of waters. This is a complex question and is incompletely understood. Sources (pollution excepted) are many in the geologies and sediments of lakes, streams, and reservoirs. These elements may occur as definite compounds, i.e., relatively insoluble carbonates and oxides, may be absorbed onto the surface of various sediments, iron oxides, organics, etc., may be chemically bound within the internal structure of clay mineral fraction of soils and sediments, or may be chelated by organics in soils and sediment. The question of whether or not these elements are leached, exchanged, dissolved, etc., from lake sediments by the H^+ ions from acidic deposition is obviously complex and poorly understood in the fresh surface waters systems of New Jersey.

D. EFFECTS OF ACIDIC DEPOSITION ON NEW JERSEY'S SURFACE WATERS

Before discussing the effects of acidic deposition on surface waters in specific parts of New Jersey, the roles that several critical factors play within New Jersey are outlined below.

1. Geology:

The role of geology in determining a region's sensitivity to acidic deposition is largely dependent upon the chemical characteristics of the weathered bedrock and the soils lying above the bedrock. For instance, limestone terrains yield infinite acid-neutralizing capacity to acidic precipitation, while granites and related igneous rocks, their metamorphic equivalents (gneisses and schists), and non-calcareous sandstones and shales yield minimal neutralizing capacity.

Efforts have been made to identify those areas within New Jersey where geological formations are likely to provide little or no neutralizing materials to surface waters. Four such provinces have been identified. These include, from north to south, (1) portions of the Valley and Ridge Province, namely Kittatinny Mountain; (2) the metamorphic rock uplands of the New Jersey Highlands Province; (3) most of the Piedmont Lowlands Province; and (4) the unconsolidated silt, sand, and gravel areas of the Coastal Plain which underlie all of the Pinelands in the southern part of the state.

2. Soils:

Certain researchers have advocated the use of soils data for determination of "acid sensitive" areas (Kaplan et al., 1981). However, as Evans et al. (1981) indicate, when considering acid transport from terrestrial to aquatic systems, several factors, including percent of land area covered with soil vs. barren rock, the amount of surface runoff vs. soil percolation, and the initial acidity of soils, must be evaluated. In addition, the thickness of soils within a given watershed must be considered. While some of this information, such as acidity, is available for New Jersey's soils, most of it is not; hence, assessment of sensitivity based on soils data is difficult.

3. Naturally-Occurring Organic Acids

Most of the surface waters in southern New Jersey, especially in the Pine Barrens, are acidic with pH values in the range of 4.0 - 0.5. These waters have a yellow-brown color due to the leaching of iron and humus (and other organic matter) from soils and decaying vegetation. The organic material, when suspended in water, releases H^+ ions from a strong acidic group (carboxylic, pKa=4.2) and a weak acid group (phenolic, pKa=8.7; Wilson and Kinney, 1977). What, then, might be the effect of acidic deposition on the pH value of these already acidic humic waters? Preliminary evidence suggests that at current levels of acidic deposition, very little effect would be observed. Several laboratory experiments were conducted by the Environmental Science Department, Rutgers University, whereby aqueous solutions of humic acid (25 ppm) were titrated with dilute sulfuric acid (pH=3.24). The pH value of the aqueous mixture stabilizied at pH 3.8 despite the addition of a considerable quantity of acid. The naturally occurring humic waters of the Pine Barrens might respond similarly to acidic deposition. That is, the addition of H^+ from the mineral acids act to repress the ionization of the organic acids, thus minimizing the drop in pH expected for the addition of the mineral acid. If so, the pH values of these waters may have been lowered by at most 0.1-0.2 pH units from the continuous input of acidic deposition. It is interesting to note that soil scientists have long reported on the "enormous acidifying and buffering capacity" of humus materials (Krug and Frink, 1983), and this concept seems to be confirmed by these analyses.

Another "effect" of this naturally-occurring organic matter is its ability to chelate metals. It is well known that iron is bound to such organic matter. Should other metals, Pb^{2+}, Zn^{2+}, Cd^{2+}, etc., become solubilized by deposition in the presence of this organic matter, their chelation and transport is a distinct possibility.

4. Clay Materials in Lake and Stream Sediments:

Aluminosilicate compounds comprise a major portion of the soils and lake and stream sediments in New Jersey. These compounds have originated from eons of physical and chemical weathering reactions. Their significance in a fresh water system lies in their ability to provide neutralization of acidity through ion exchange reactions. For example, current research on the water chemistry of White Meadow Lake in Morris County, New Jersey (in a non-limestone area) strongly suggests that these reactions are occurring.

a. Northern New Jersey (Geologic regions of Valley Ridge, Highland and Piedmont)

An initial study (Phase 1) of the sensitivity of northern New Jersey's major lakes, streams, ponds, reservoirs, etc., to acidic deposition was undertaken during 1980 - 1981 (Faust and McIntosh, 1982). Determinations of pH and total alkalinity were conducted on waters from 31 streams and rivers and from 32 lakes and reservoirs. In general, the data indicated that, while most of the region's streams and rivers have substantial neutralizing capacities (alkalinity) (Table 23), several lakes and reservoirs have limited alkalinities (Table 24). Of the 32 lakes and reservoirs sampled, 16 had alkalinities less than 10 ppm as $CaCO_3$ and 7 had pH values less than 6.3. These data were based upon a one-time only sample collection. Thus, the water quality is indicative only of the date of collection.

An additional study (Phase 2) was conducted (July and August 1981) on those lakes and reservoirs in northern New Jersey whose alkalinities were less than 10 ppm as $CaCO_3$ and on streams and rivers whose alkalinities were less than 20 ppm (Figure 18). In most cases, the alkalinities were similar to those in the initial study (Table 25). Exceptions were Sawmill Lake, whose alkalinity was 4.7 and 12 ppm (two samples) and Mt. Hope Lake, with 5.9 and 15.5 ppm. pH values ranged from 4.7 for Long Pine Pond to 7.3 for Green Pond. Note the high sulfate level in essentially all the lakes and streams listed in Table 25.

Calculation of buffering capacity indicated that two lakes, Catfish Pond and Long Pine Pond, (Nos. 13 and 14 in Figure 18) were severely affected chemically. That is, these two waters have no buffer capacity, since they have no alkalinity and pH values of 4.8 and 4.7, respectively (Table 26). These pH values and elevated sulfate levels may have been the result of acidic deposition over many years. However, the lack of historical data on these two lakes prohibits a firm conclusion on the cause of the acidic pH values. In addition, 7 lakes considered to be in the "threatened" category due to low buffer capacity and alkalinities are Green Pond, Clyde Potts Reservoir, Split Rock Reservoir, Clinton Reservoir, Hank's Pond, Butler Reservoir and Lake Denmark. The four reservoirs supply drinking water to Newark (Clinton and Butler), Jersey City (Split Rock) and Morristown (Clyde Potts). No brooks, streams or rivers were found to be dangerously low in alkalinity and buffer capacity. However, five brooks in the second phase study had pH values of 6.0 and below.

Two intensive studies were conducted on seven lakes and ponds in northern New Jersey during 1982-84. In the first study, Clyde Potts Reservoir and White Meadow Lake in Morris County were sampled on a weekly basis from September 1982 to March 1983. There was a wide range of weather conditions and a considerable amount of rainfall. For Clyde Potts Reservoir, the pH values were remarkably stable, 6.91 to 7.79, and alkalinity ranged from 8.0 to 13 ppm as $CaCO_3$. An unknown factor held these two chemical parameters relatively constant, despite a

TABLE 23

pH AND TOTAL ALKALINITY OF STREAMS AND RIVERS SAMPLED DURING INITIAL PHASE

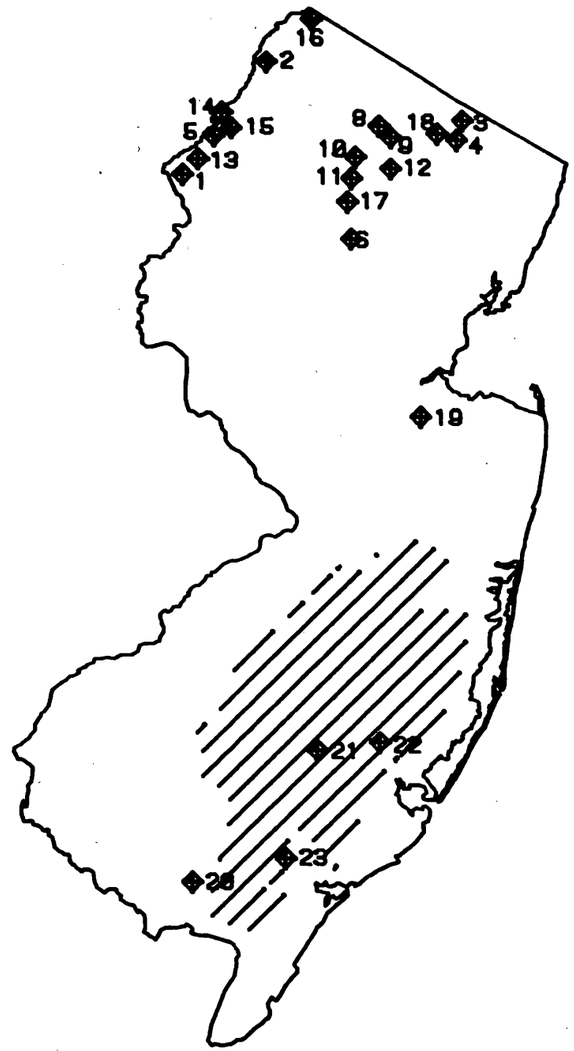
SYSTEM	DATE SAMPLED	FIELD pH	TOTAL ALKALINITY ppm as $CaCO_3$
Whippany River	6/17/81	7.3	61.1
Harmony Brook	6/17/81	7.2	54.3
Mine Brook	6/17/81	6.7	40.4
Jackson Brook	6/17/81	6.9	23.2
Burnett's Brook	6/17/81	6.9	55.9
Dawson's Brook	6/17/81	7.2	62.3
India Brook	6/17/81	7.1	36.4
North Branch Raritan River	6/17/81	6.9	73.2
Gladstone Brook	6/17/81	7.6	105.9
Peapack Brook	6/17/81	7.4	50.6
South Branch Raritan River	6/17/81	7.3	42.2
Turkey Brook	6/17/81	6.6	22.6
Stony Brook (I)	6/17/81	7.1	39.7
Trout Brook	6/17/81	6.9	56.4
Rhinehart Brook	6/17/81	6.7	41.1
Black River	6/17/81	7.4	67.7
Big Flatbrook	6/19/81	7.1	31.7
Stony Brook (II)	6/19/81	6.6	18.4
Yard's Creek	6/19/81	7.1	17.4
Tillman's Brook	6/19/81	6.2	6.9
Van Campen's Brook	6/19/81	6.6	6.1
Kanouse Brook	6/26/81	6.3	32.3
Saddle River	6/26/81	8.1	171.6
Hewitt Brook	6/26/81	7.1	67.9
Cooley's Brook	6/26/81	6.5	38.6
Green Brook	6/26/81	6.6	35.9
West Brook	6/26/81	6.9	44.4
Clinton Brook	6/26/81	6.3	25.4
Tributary to Pequanock River	6/26/81	7.5	120.7
Bear Swamp Brook	6/26/81	5.7	6.3
Stag Hill Brook	6/26/81	6.3	7.4

TABLE 24

pH AND TOTAL ALKALINITY OF RESERVOIRS AND LAKES
SAMPLED DURING INITIAL PHASE

SYSTEM	DATE SAMPLED	FIELD pH	TOTAL ALKALINITY ppm as $CaCO_3$
Catfish Pond	3/18/81	5.3	1.1
Long Pine Pond	3/18/81	4.7	0.3
Fairview Lake	3/18/81	7.8	8.3
Sawmill Lake	3/18/81	8.1	4.7
Lake Marcia	3/18/81	8.1	20.0
Steeny Kill Lake	3/18/81	7.9	*
Clinton Reservoir	5/14/81	5.4	1.2
Clinton Reservoir	6/03/81	5.7	3.0
Green Pond	5/14/81	6.5	6.7
Green Pond	6/03/81	6.2	5.8
Green Pond	6/15/81	6.9	7.1
Splitrock Reservoir	5/14/81	5.8	5.4
Splitrock Reservoir	6/03/81	6.2	3.4
Splitrock Reservoir	6/15/81	6.4	6.8
Oak Ridge Reservoir	5/14/81	6.4	19.3
Oak Ridge Reservoir	6/03/81	6.9	25.6
Echo Lake	5/14/81	6.6	24.6
Echo Lake	6/03/81	6.9	23.1
Canistear Reservoir	5/14/81	6.5	18.2
Charlottesburg Reservoir	5/14/81	6.7	16.6
Lake Wawayanda	5/14/81	7.0	35.3
Girard Lake	5/14/81	6.3	18.9
Glen Lake	5/14/81	7.6	53.0
Butler Reservoir	6/03/81	6.6	7.7
Hank's Pond	6/03/81	5.7	1.9
Lake Denmark	6/03/81	6.2	6.1
Mount Hope Lake	6/03/81	6.5	5.9
Wanaque Reservoir	6/03/81	7.6	19.9
Shepard Lake	6/03/81	7.1	17.1
Cupsaw Lake	6/03/81	7.0	30.8
Greenwood Lake	6/03/81	6.8	18.3
Upper Greenwood Lake	6/03/81	6.5	20.0
Macopin Reservoir	6/03/81	6.6	19.8
Spruce Run Reservoir	6/15/81	7.1	46.6
Round Valley Reservoir	6/15/81	7.6	63.3
Mountain Lake	6/15/81	8.8	137.4
Lake Mohawk	6/15/81	8.5	164.4
Lake Hopatcong	6/15/81	7.5	40.8
Clyde Potts Reservoir	6/17/81	7.1	12.6

* Sample lost



STREAMS AND RIVERS

1. Yard's Creek
2. Tillman's Brook
3. Bear Swamp Brook
4. Stag Hill Brook
5. Van Campen's Brook

LAKES AND RESERVOIRS

6. Clyde Potts Reservoir
7. Butler Reservoir
8. Clinton Reservoir
9. Hank's Pond
10. Green Pond
11. Lake Denmark
12. Splitrock Reservoir
13. Catfish Pond
14. Long Pine Pond
15. Fairview Lake
16. Sawmill Lake
17. Mount Hope Lake
18. Ramapo Lake
19. Weston's Mill Pond
20. Union Lake
21. Lake Ateion
22. Lake Oswego
23. Lake Lenape

Shaded area indicates pine barrens

Figure 18. Location of Sites Sampled During Phase 2

TABLE 25

WATER CHEMISTRY DATA FROM SYSTEMS SAMPLED DURING PHASE 2

LOCATION	DATE	FIELD pH	TOTAL ALKA-LINITY	TOTAL ACIDITY	CO_2 ACIDITY	SULFATE	NH_4^+-N	Mg^{2+}	Ca^{2+}	Mn	SPECIFIC CONDUCTANCE
			ppm as $CaCO_3$				ppm				umhos/cm
Lakes and Reservoirs											
Clyde Potts Reservoir	7/11/81	7.3	8.1	1.06	1.06	6.90[1]	0.89	1.78	5.2	N.D.[2]	76[1]
Butler Reservoir	"	7.0	10.0	1.72	1.72	17.40[1]	0.64	2.91	7.7	N.D.	111[1]
Clinton Reservoir	"	6.7	3.0	2.70	2.70	13.50[1]	0.64	1.51	4.3	N.D.	47[1]
Hank's Pond	"	6.4	3.6	1.18	1.18	11.20[1]	0.67	1.52	3.4	0.03	40[1]
Green Pond	"	7.3	5.1	1.32	1.32	8.90[1]	1.20	1.29	4.1	N.D.	48[1]
Lake Denmark	"	6.8	8.1	2.29	2.29	8.90[1]	1.50	2.38	4.9	N.D.	66[1]
Splitrock Reservoir	"	7.1	5.7	1.23	1.23	12.50[1]	1.00	1.86	4.8	N.D.	50
Weston's Mill Pond	7/21/81	6.2	24.1	5.63	5.63	---	2.17	3.65	9.7	0.06	155
Catfish Pond	7/22/81	4.8	0.0	2.96	0.53	9.57	1.10	1.54	2.73	0.13	33
Long Pine Pond	"	4.7	0.0	6.56	0.06	9.24	2.90	0.76	1.5	0.16	39
Fairview Lake	"	5.5	10.2	1.90	1.90	12.50	2.50	2.00	5.0	0.06	77
Sawmill Lake	"	6.7	12.0	2.38	2.38	9.89	3.10	1.77	5.7	0.08	54
Mount Hope Lake	7/29/81	6.4	15.5	4.64	4.64	12.60	2.17	2.02	6.4	0.04	98
Ramapo Lake	"	5.7	7.4	5.80	2.51	9.30	1.78	1.25	5.6	0.03	50
Union Lake	8/19/81	5.8	27.7	5.80	5.80	8.90	5.00	2.58	4.24	0.05	88
Lake Atsion	"	3.7	0.0	5.10	1.26	7.60	4.50	0.86	1.75	N.D.	50
Lake Oswego	"	4.2	0.0	2.70	0.10	4.50	4.20	0.66	1.55	N.D.	38
Lake Lenape	"	5.2	8.3	3.20	0.50	9.90	4.80	4.80	1.52	3.01	58
Streams											
Yard's Creek	7/22/81	5.0	15.0	3.54	3.54	28.60	2.70	2.78	12.9	0.04	89
Van Campen's Brook	"	6.0	9.1	1.90	1.90	8.91	2.30	1.96	4.4	N.D.	49
Tillman's Brook	"	5.7	5.2	2.32	2.32	2.16	4.20	1.88	2.7	0.06	38
Bear Swamp Brook	7/29/81	5.6	4.2	2.70	2.70	9.50	2.39	1.03	4.1	N.D.	38
Stag Hill Brook	"	6.0	6.3	2.78	2.78	16.50	0.83	2.71	9.0	N.D.	160

[1] Sample collected on 8/17/81

[2] Not detectable.

TABLE 26

BUFFER VALUES OF SOME FRESH WATERS IN NEW JERSEY

SYSTEM	BUFFER VALUE	pH	TOTAL ALKALINITY	$\frac{C_t}{M}$ [c]
Lakes and Reservoirs	$X10^{-5}$[a]		ppm[b]	$X10^{-4}$
Catfish Pond	0	4.8	0	-
Long Pine Pond	0	4.7	0	-
Atsion Lake	0	3.7	0	-
Lake Oswego	0	4.2	0	-
Green Pond	-2.37	7.3	5.1	1.14
Clyde Potts Reservoir	-3.77	7.3	8.1	1.8
Splitrock Reservoir	-3.96	7.1	5.7	1.34
Clinton Reservoir	-4.26	6.7	3.0	.87
Hank's Pond	-7.8	6.4	3.6	1.36
Butler Reservoir	-8.42	7.0	10.	2.45
Lake Denmark	-9.76	6.8	8.1	2.2
Sawmill Lake	-17.08	6.7	12.	3.48
Ramapo Lake	-27.8	5.7	7.4	8.11
Mt. Hope Lake	-33.7	6.4	15.5	5.87
Lake Lenape	-35.7	5.2	8.3	25.2
Fairview Lake	-41.2	5.5	10.2	16.5
Weston's Mill Pond	-65.	6.2	24.1	11.6
Union Lake	-99.6	5.8	27.7	25.2
Streams				
Bear Swamp Brook	-16.4	5.6	4.2	5.57
Tillman's Brook	-19.6	5.7	5.2	5.7
Stag Hill Brook	-20.05	6.0	6.3	4.09
Van Campen's Brook	-29.	6.0	9.1	5.90
Yard's Creek	-51.	5.9	15.	11.5

a. Equivalents//pH unit, 25°C.

b. As $CaCO_3$.

c. $C_t = [H_2CO_3^*] + [HCO_3^-] + [CO_3^{=2}]$.

continuous exposure to acidic deposition. Conversely, White Meadow Lake showed a trend towards lower pH values and alkalinities during the study. The pH of this lake was 9.44 in October 1982. This high pH level was due to eutrophication of the lake. By February 1983, the pH level was 6.87, and the pH level stayed relatively constant at that level thereafter.

A second short-term intensive study (August 1983 to July 1984) was begun on seven lakes in northern New Jersey. Three lakes, Fairview, Sawmill and Marcia, are underlain with or lie within limestone geology. Catfish Pond was intensively sampled also, but it is isolated from limestone. The three other lakes, Split Rock Reservoir, Clyde Potts Reservoir and White Meadow Lake, lie outside of limestone geology. The intent of the seven lake study was to determine the chemical factors controlling the pH values and to monitor the concentrations of four trace elements: Al, Cd, Pb and Zn. The pH values for the three limestone lakes did not show any trend toward permanent acidification, with one exception. On March 31, 1984, a field pH value of 4.55 measured in Sawmill Lake may have been due to melting of "acid ice." Alkalinity values remained relatively constant in the three lakes. There were no unusually "high" trace element concentrations in the three lakes. Occasionally, the dissolved Al content exceeded .100 ppm, and .681 ppm Al was measured in Fairview Lake at pH 6.8.

Results from the three non-limestone lakes were similar to the above. No trends toward permanent acidification were detected in 1983-1984. There was an occasional "low" pH value; for example, a field value of 4.2 under ice was measured in Clyde Potts Reservoir on February 21, 1984. Lead and Cd contents were low. However, there were several occasions when Zn concentrations ranged from 20. to 45. ppb. Al concentrations ranged from 129 (Split Rock) to 533 (White Meadow Lake) ppb. Catfish Pond was included in this study primarily to determine the trace element concentrations in a lake tnat has become acidified (pH range of 4.58-5.64). For this water, dissolved Al contents ranged from 78. to 557. ppb and Zn ranged from 14. to 129. ppb. Both ranges were consistently higher than for the other six lakes. Table 27 is a summary of the heavy metal content of two drinking water reservoirs in Morris County. Presumably, these metals were leached from sediments in the reservoirs. None of the contents, however, exceeded drinking water standards of the NJDEP.

TABLE 27

SUMMARY OF HEAVY METALS LEACHED FROM SEDIMENTS
IN TWO DRINKING WATER RESERVOIRS IN NEW JERSEY 1983-1984

RESERVOIR	# SAMPLES	pH	Al ppb	Pb ppb	Cd ppb	Zn ppb
Split Rock	16	6.5-7.23	4-129	2-10	0.3-2	0.5-2.7
Clyde Potts	17	5.1-7.42	24-400	0.2-5.7	0.1-1.8	1.5-43

b. Southern New Jersey (Outer Coastal Plain)

An approach commonly used to assess acidic deposition effects on surface water pH is analysis of long-term chemical trends in otherwise undisturbed surface waters. If a surface water pH trend is found, and it can be correlated with precipitation pH, then a strong, albeit circumstantial, case can be put forth that atmospheric inputs were responsible for the observed stream response. Although such continuous long-term surface water pH data sets are rare, New Jersey is fortunate that such records exist for two headwater streams in southern New Jersey (the Pinelands). The United States Geological Survey (USGS) has collected reasonably regular water quality data (including pH) at McDonalds Branch since 1958, and at Oyster Creek since 1965 (beginning in 1979, the Oyster Creek data have been collected by the Ocean County Health Department). These data offer an unique opportunity to assess the direct effect of acidic deposition on Pinelands surface waters.

The first such assessment of the Pinelands data was provided by Johnson (1979a,b) who used regression analysis to document trends in stream pH prior to 1979. He reported that stream pH declined significantly over the 20 yr interval for which data were available. The decline amounted to about 0.7 pH units. Johnson discussed and dismissed the role of several possible natural causes of the observed trend. Instead, he noted that measured precipitation pH in the early 1970s and in 1978-1979 declined significantly from about 4.4 to 3.9 and concluded "the data collected to date are consistent with the postulation of an atmospheric source for the increased (stream) H^+."

As previously noted, Pinelands surface waters are naturally acidic due to dissolved CO_2(g), humic and fulvic acids (components of highly colored waters), tannic acids, iron compounds, and very little buffering capacity. In addition, there are many natural watershed processes which can both raise and lower stream pH. Thus, the role of acidic deposition in controlling surface water pH in the Pinelands might not be as clear cut as suggested by Johnson (cf. empirical estimates of the buffering ability of these humic waters to strong acid addition). These considerations led Morgan (1984) to re-analyze the McDonalds Branch and Oyster Creek data, including data through 1982.

All of the data for both streams are depicted in Figure 19, and the relevant regression equations for Johnson's and Morgan's analyses are presented in Table 28. For McDonalds Branch, Morgan found that when 1979-1982 pH data were added to those analyzed by Johnson, the significant downward trend no longer existed. Even if the 1958 data were omitted, the trend was still not significant ($r=-0.12$). In addition, when the data were analyzed only for the interval between 1971 and 1978, when there was documented evidence for a dramatic decline in precipitation pH from 4.4 to 3.9 (Yuretich et al., 1981; Johnson, 1979a) stream pH did not decline significantly. In fact, the trend was toward increasing pH (Table 28). A recent analysis of these same data using non-parametric trend analysis yielded similar results (J. Slack, USGS, pers. comm.).

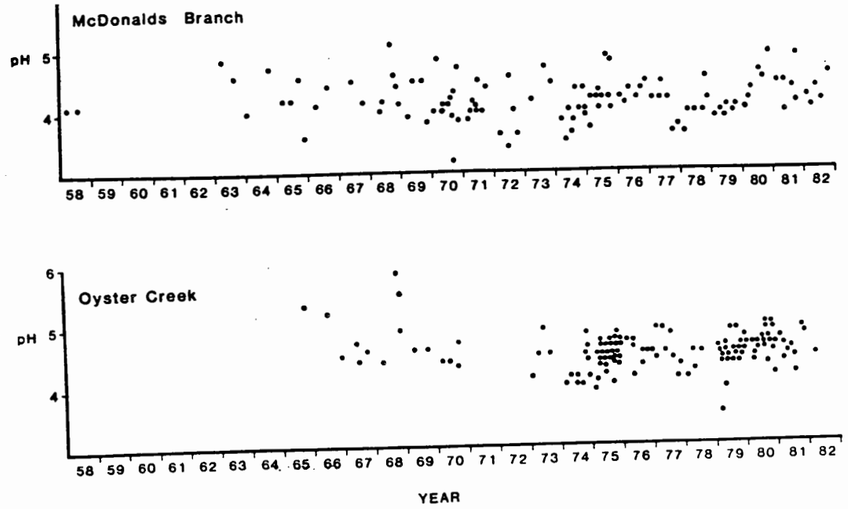


Fig. 19 pH of McDonald's Branch and Oyster Creek from 1958 to 1982.

In Morgan's analysis of the Oyster Creek data, there was a slight discrepancy between his and Johnson's results for the same time period, but the conclusion remained unchanged: stream pH declined significantly from 1965-1978. Even when 1979-1982 data were added, the relationship, although considerably weaker, held. Several lines of evidence, however, led Morgan to conclude that the decline may have had little to do with acidic deposition. For instance, if only data from 1969-1982 were examined (representing 92% of the entire data set and including the interval from 1971-1978 when precipitation pH was known to have declined), there was no significant relationship between pH and time, and the trend was toward increasing, not decreasing, pH. Non-parametric analyses of these data also failed to show a consistent downward trend (J. Slack, USGS, pers. comm.). If the total pH decline based on the regression analyses are examined over the indicated time intervals up to 1978, the McDonalds Branch decline is estimated at 0.4 pH unit and the Oyster Creek decline is estimated at 0.5 pH unit. If all the data are examined up to 1982, the most probable respective drops for the McDonalds' Branch and Oyster Creek waters is 0.2 and 0.3 pH unit. Declines of 0.2 - 0.3 pH units over many years in natural systems are obviously difficult to establish accurately and more difficult to assign causes. This is why these declines are not statistically significant, or just barely so. Assuming no other factors affect acidity, declines of 0.2 pH unit are in line with what might be expected from adding acidic deposition to waters already acidified by organic acids but otherwise free of mineral acids.

TABLE 28

Regression of pH on time according to the relation: pH=b+mx, where m and b are the regression coefficients, x is time in months, n is the number of samples, r is the correlation coefficient, and P is the level of significance. NS means not significant. The warm season is defined as May-October; the cold season, as November-April. The U.S. Geological Survey (USGS) data were provided by the Water Resources Division, and the Ocean County Health Department data were provided by the New Jersey Department of Environmental Protection, both in Trenton, NJ. (After Morgan 1984)

	n	b	m		r	P	pH**
McDonalds Branch							
USGS 1963-1978 Data (Johnson)	90	4.42	-0.0022	*	-0.22	0.05	-0.4
USGS 1963-1978 Data (Morgan)	90	4.40	$-0.0021^{\pm}.0019$		-0.23	0.05	-0.4
All USGS Data, 1958-1982 (Morgan)	115	4.27	$-0.0007^{\pm}.0012$		-0.10	NS	-0.2
USGS Data, 1971-1978 (Morgan)	55	4.06	$0.0002^{\pm}.0032$		0.03	NS	
Oyster Creek							
USGS 1965-1978 Data (Johnson)	78	5.10	-0.0047		-0.56	0.01	-0.7
USGS 1965-1978 Data (Morgan)	78	4.80	$-0.0034^{\pm}.0017$		-0.42	0.01	-0.5
USGS + Ocean County Data, 1965-1982 (Morgan)	128	4.64	$-0.0013^{\pm}.0009$		-0.24	0.01	-0.3
USGS + Ocean County Data, 1969-1982 (Morgan)	118	4.45	$0.0005^{\pm}.0009$		0.09	NS	

*95% Confidence Intervals of the Slopes

** pH decline calculated from the regression equations over the entire time period of the data.

In considering the data from both Oyster Creek and McDonalds Branch for the periods 1963-78 and 1965-78 respectively, Morgan suggested that the trend in pH reported by Johnson may not have resulted from a steady decline in pH due to increased acidic deposition. Instead, the apparent trend may have resulted from unusually high pH levels in the mid 1960s followed by a return to naturally low and stable pH levels starting in the late 1960s. Morgan noted that elevated pH in the Pinelands is clear evidence of watershed disturbance. Although both watersheds are considered undisturbed by human activity, he suggested that a major fire, such as the one in 1963, could have accounted for the elevated pH values in the mid 1960s. The 1963 fire burned over 65000 ha, or about 10% of the entire Pinelands region, including all of the McDonalds Branch watershed and about 20% of the Oyster Creek watershed. Significant leaching of base cations (Ca^{2+}, Mg^{2+}, K^{+}) occurs for a least 3 years following wildfire in the Pinelands (Boerner and Forman 1982). Leaching of these cations tends to raise the pH of infiltrating water and should eventually result in increased surface water pH. Significantly elevated NO_3^- levels coincident with the elevated pH in the mid 1960s provided additional support for this "fire" hypothesis.

In conclusion, Morgan stressed that his analysis did not show that "acid deposition has had no past or has no present effect on surface water pH in the Pinelands, just that such a conclusion is unwarranted on the basis of historical data." He emphasized that acidic deposition was just one of many factors (some completely natural) affecting surface water pH and until "a quantitative understanding of hydrogen ion flux through Pinelands soils, groundwater, bogs, and swamps is obtained, current and future effects of acid deposition, even with complete precipitation pH records, will be hard to verify."

EFFECTS OF ACID DEPOSITION ON AQUATIC BIOTA

A. REVIEW OF THE KNOWN EFFECTS OF ACIDIFICATION ON AQUATIC BIOTA

The manner in which acidification exerts its influence on aquatic organisms is complex, including both direct and indirect effects. Increased hydrogen ion concentration itself can be directly toxic to organisms by disruption of various physiological processes such as ion transport and enzyme function (Fromm, 1980). Acidification may also increase the solubility and, therefore, the concentration of various trace elements such as Al, Fe, Pb, and Hg which can be directly toxic to many organisms. However, the most disruptive effects of acidification, and the most difficult to predict, may be indirect effects brought about by changes in the overall structure of the aquatic system. Because of the high degree of interaction among the biological and nonbiological components of the aquatic environment, a direct effect on one dominant species can affect virtually every other member of the community (Eriksson, et al., 1980). Thus, an understanding of the true effects of acidification requires more than knowledge of which organisms are sensitive to low pH or elevated trace element concentrations. It requires a sophisticated understanding of how the entire ecosystem functions. While the disappearance of fish may appear to be the most significant effect of acidification to the public at large, it may be merely a symptom of more fundamental changes within other components of the aquatic system. Therefore, an appraisal of the effects of acidification on aquatic biota must necessarily consider all aspects of the aquatic environment.

1. Microorganisms (bacteria and fungi)

Microorganisms perform two critical functions in the aquatic system: they are primarily responsible for decomposition of organic matter, and they play a major role in several important nutrient cycles. Studies on the impact of acidic deposition on microorganisms have, therefore, concentrated on these two functions.

Some investigators have reported that decompositional processes slow down as waters become acidified from pH 6.5 to 6.0 (Hendrey, et al., 1976), although this phenomenon has not been universally demonstrated (Schindler, 1980; Kelley et al., 1984). There is some evidence that the microbial community shifts from one dominated by bacteria to one dominated by fungi (Grahn, 1977). If decompositional rates are reduced by acidification, the resulting accumulation of organic matter would have significant effects on a variety of essential chemical and biological processes within the aquatic system.

Little is known about the effect of acidification on microbial nutrient cycling. There is some evidence that sulfur reducing bacteria, such as Desulfovibrio and Desulfomonas, may act to buffer against acidification when the primary source of acidity is H_2SO_4 (Schindler, et al., 1980). On the other hand, at least in areas affected by acid mine drainage, sulfur oxidizing bacteria increase acidity by the production of H_2SO_4. The role these organisms may play in lakes affected by acidic deposition, however, is presently difficult to evaluate because acid mine waters are chemically quite different from the poorly buffered waters affected by acidic deposition. The impact of acidification on nitrogen cycling (in surface waters) is also poorly understood. Several studies

have indicated that soil acidification may reduce nitrogen mineralization (Francis, 1982; Novick et al., 1984). A similar reduction in surface waters could severely limit the availability of this essential nutrient to primary producers.

2. Periphyton

The periphyton community consists of algae and associated organisms which live attached to plants and sediment. In most bodies of water, particularly streams, these algae are major primary producers and serve as important food sources for many macroinvertebrates. Several responses by the periphyton to acidification have been noted. Both comparative and experimental studies have reported a dramatic decline in periphyton species richness when the pH falls below 5.0 (Almer et al., 1974; Muller, 1980). This decline is accompanied by a trend toward increased biomass and reduced growth (Hall et al., 1980). Perhaps the most dramatic response of periphyton to acidification is the formation of solid mats of algae covering the substrate in some systems (Stokes, 1981). These mats may be several meters thick and may essentially cut off the bottom from the rest of the lake.

3. Phytoplankton

Numerous studies have shown that there is a dramatic shift in phytoplankton species composition and a reduction in species richness as acidification proceeds. These trends are particularly evident when pH drops below 5.0 (Kwiatkowski and Roff, 1976). In general, species in acidified lakes are lost from all phytoplankton groups, although some groups are affected proportionately greater than others. Members of the phylum Pyrrophyta (dinoflagellates) appear least affected by acidification and often come to dominate acidic lakes. The Chlorophyta (green algae) and Crysophyta (crysophytes) have also been noted to increase in importance (Conroy et al. 1976). The group which appears most severely and negatively affected is the Bacillariophyta (diatoms). Many studies have shown a reduction in importance of diatoms in acidified lakes relative to similar circumneutral lakes (Haines, 1981).

Despite changes in species composition and richness, the impact of acidification on phytoplankton biomass and productivity is not clear. Reports of both reduced and increased biomass and productivity associated with acidification have appeared (Kwiatkowski and Roff, 1976; Haines, 1981). Much of the ambiguity with respect to this point may occur because most of the studied acidified lakes were also severely nutrient poor. It is, therefore, difficult to separate the effects of acidification from low nutrient levels. An increase in water transparency also associated with acidification allows phytoplankton growth to occur at greater than normal depths. Acidification, then, may simply lead to a spatial redistribution of phytoplankton growth and productivity with little net change in amount.

4. Aquatic Plants

Most effects of acidic deposition on aquatic plants are indirect, including decreased carbon supply for photosynthesis, nutrient depletion, increased trace element concentrations and decreased rates of nutrient recycling. Peverly (1983) speculates that these factors may become limiting as the pH decreases during acidification, and species more tolerant of low nutrient supplies and higher trace element concentrations may become dominant. Lakes susceptible to acidic deposition, soft water, low alkalinity, oligotrophic lakes, generally are characterized by isoetids (quillworts), lilies, Nitella, Utricularia (bladderwort) and mosses (Peverly, 1983).

The relationship between increased trace element activity in acidic lakes and aquatic plants has been assessed by several investigators. Miller et al. (1983) found greatly elevated levels of lead, copper, and nickel in the isoetid vegetation of soft-water lakes in central Ontario. While previous research (Stanley, 1974) has shown that elevated aluminum, zinc and copper in lake sediments may result in a 50 percent reduction in root weight in Myriophyllum spicatum, whether or not elevated trace element levels noted in aquatic macrophytes in acidic lakes are resulting in toxic effects is not known.

One significant occurrence in some acidifying lakes is the invasion of the bryophyte Sphagnum. Grahn (1977) reported that in five of six Swedish lakes with pH values between 4.4 and 5.4, Sphagnum replaced communities of Lobelia and Isoetes, typical species in soft-water lakes. The Sphagnum invasion of acidic lakes may have several deleterious consequences. Possessing a significant ion exchange capacity, Sphagnum may remove such cations as calcium, magnesium, and iron in exchange for hydrogen (Clymo, 1963). In addition to adversely affecting primary production by limiting these essential nutrients, this phenomenon may increase acidity in waters surrounding Sphagnum. Also, by forming dense mats, Sphagnum may physically eliminate other species and, acting as a barrier, may retard the exchange of materials between the sediment and water column. Finally, the presence of Sphagnum in littoral areas represents an unfavorable habitat for certain benthic macroinvertebrates and fish species.

5. Zooplankton

Numerous field surveys have found that zooplankton species numbers decrease in acidified lakes when compared with similar circumneutral lakes. For example, Sprules (1975) found 9 to 16 zooplankton species in Ontario lakes with pH levels above 5.0, but only 1 to 7 species in lakes with pH values below 5.0. Contrary to observations of other components of the aquatic system, few new species appear to invade acidified lakes. The species found in acidified lakes are also found in circumneutral lakes. As acidification proceeds, shifts in species composition occur as non-acid tolerant fauna drop out.

Cladocera tend to increase in importance in acidified lakes. Acidification generally results in a shift in dominance among the cladocera from Daphnia to Bosmina (Almer et al., 1974; Hobaek and Raddum, 1980). Daphnids often disappear completely from lakes with pH levels below 7.0-5.0. Among the two other dominant zooplankton groups in acidified lakes, copepods are primarily represented by members of the genera Diaptomus, Cyclops and Mesocyclops, and rotifers, by the genera Kereatella and Polyarthra (Roff and Kwiatkowski, 1977; Malley et al., 1982).

6. Benthic Invertebrates

Most groups of benthic invertebrates are sensitive to acidification but perhaps the most dramatic effects are found among the mollusks. The calcareous shell of these animals is highly sensitive to dissolution when the pH drops below 7.0. Because it becomes increasingly difficult for mollusks to replace the lost $CaCO_3$, few clams and snails are found in water below pH 5.0, and most disappear when the pH drops below 6.0 (Okland and Kuiper, 1980). Mollusks are valuable food sources for fish and other vertebrates (e.g. waterfowl) and their disappearance from acidified waters may adversely affect vertebrate populations.

Studies have consistently shown that among the crustacea several species of amphipods (scuds), isopods (sow bugs), decapods (crayfish) and various other groups are negatively affected when pH is artificially reduced from above 6.0 to below 4.0 (Costa, 1967; Malley, 1980; Havas and Hutchinson, 1982; Schindler and Turner, 1982). The primary negative effects of acidification have been reported to be either direct mortality from higher acidity or indirect mortality, resulting from alternatives in associated chemical parameters (especially reduced $CaCO_3$ availability) or changes in food quality or quantity.

In many aquatic systems, the benthic insects are the single most important invertebrate group, and studies of the effect of acidification on insects have generally revealed reductions in species richness, diversity, and biomass (Conroy et al., 1976; Haines, 1981); although there are particular insect species in each order that can tolerate very low pH (below pH 3.0). In general, Ephemeroptera (mayflies) appear to be most sensitive to low pH values and are rarely found in waters below pH 5.0. Diptera (true flies), Coleoptera (beetles), Hemiptera (true bugs), and Megaloptera (alderflies) appear to be least sensitive and Plecoptera (stoneflies), Trichoptera (caddisflies) and Odonata (dragonflies) exhibit intermediate sensitivities (Bell and Nebeker, 1969; Bell, 1971). Studies have shown that members of the least sensitive orders may dominate communities in which they were fairly insignificant prior to acidification (Weiderholm and Erikson, 1977).

The underlying structure of benthic insect communities also may be altered by acidification. Intense fish predation is usually a major factor structuring insect communities. Since fish populations are often decimated as a result of acidification, fish predation on the insects is lessened. Very often certain insect predators such as odonates and coleopterans increase and replace fish as the dominant predators in these waters (Stenson et al., 1978; Henrikson et al., 1980). Thus, the organization of the insect community is drastically altered, and much secondary production is shifted from harvestable (fish) to non-harvestable (insect) biomass.

Annelids, the last major group of benthic invertebrates, have not been studied as extensively as the other groups. The limited data available suggest that oligochaetes decrease dramatically in acidified lakes (Raddum, 1980). Hirudinea (leeches) are also rare below pH 6 (Haines, 1981).

7. Fish

a. Direct Effects

Evidence of direct effects on fish includes both results of laboratory experiments and field observations. Haines (1981) speculated that at very low pH levels (less than 3.5), respiratory failure may be the most critical response. At higher pH's (4 to 5), failure of ion regulation due to increased gill membrane permeability may be the primary effect. Leivestad and Muniz (1976) noted that brown trout (Salmo trutta) collected in highly acidic portions of the Tovdal River in Norway exhibited lower plasma sodium and chloride concentrations than fish from more alkaline sections of the river. Apparently, exposure to increased hydrogen ion concentrations increased gill membrane permeability, with more hydrogen ions entering and sodium and other ions leaving.

A major physiological impact of increased hydrogen ion concentrations on fish is reproductive failure. A variety of effects on reproductive processes have been suggested: abnormal development of the ovaries due to disrupted calcium metabolism (Lockhart and Lutz, 1977); delayed or impaired hatching of eggs due to disruption of critical enzymes (Peterson, et al., 1980); failure of adults to release eggs (Beamish, 1976) and reduced spermatogenesis. Reproductive behavior may also be adversely affected, as low pH may prevent spawning, with exposed species actively avoiding spawning in low pH waters.

Field observations in acidic lakes and rivers indicate that fish populations may be eliminated, with losses reported in Scandinavia (Wright et al., 1976), Canada (Harvey, 1980) and the United States (Schofield, 1976). While direct acute mortality has been observed in streams in association with rapid changes in pH because of snowmelt, loss of fish from affected lakes is likely to be gradual, resulting from effects described earlier.

Differences in pH tolerance among species have been suggested by studies in Canadian lakes (National Research Council of Canada, 1981), with yellow perch (Perca flavescens) and pumpkinseeds (Lepomis gibbosus) most tolerant (surviving at pH levels as low as 4.2) and bluntnose minnows (Pimephales notatus), least tolerant (eliminated at pH levels of 5.7). Fish populations close to their tolerance limits are typically small, probably representing a final stage prior to extinction in acidifying lakes (Beamish, 1974; Harvey, 1975).

The structure of fish populations in acidic lakes typically may reflect the failure of recruitment, with significant decreases in young age classes. Shifts in age-class structure may result from mortality of larval forms, spawning failures or decreased egg maturation.

Data of the effects of acidification on growth of fish are mixed, with both increased (due to reduced competition for food) and decreased (due to metabolic stress) growth of exposed populations (National Research Council of Canada, 1981).

b. Indirect Effects

Acidification of surface waters may result in increased trace element activity; both enhanced accumulation of these elements and toxicity may occur. Perhaps the greatest concern has been the increased activity of mercury in acidifying lakes, since methylmercury is rapidly accumulated by aquatic biota and may reach high levels in tissues of large fish. Elevated concentrations of mercury have been noted in fish from acidic waters (Dillon et al., 1984: Haines, 1981; Kelso et al., 1982).

Other trace elements may be found in increased levels in fish in acidifying waters. Fraser and Harvey (1982) found substantially increased levels of manganese in bones of white suckers (Catostomus commersoni) from acidified lakes in Ontario. Kelso et al. (1980) found increased lead concentrations in white suckers from acidic Ontario lakes and suggested that both mercury and lead might be mobilized from lake sediments under acidic conditions. Although zinc and other elements may be mobilized from sediments under acidic conditions (Schindler et al., 1981), significant accumulation may not occur because these elements are essential.

Increased toxicity of trace elements due to acidification has been hypothesized primarily for aluminum (Cronan and Schofield, 1979; Muniz and Leivestad, 1980). While the critical level of exposure to aluminum will vary between systems, levels between 100 and 200 ppb aluminum are generally thought to be chronically toxic to fish.

Data on enhanced toxicity of other trace elements are limited. However, since such elements as cadmium and mercury are highly toxic to fish, any increased activity of these substances in acidifying waters may be expected to have deleterious effects on exposed biota.

B. EFFECTS OF ACIDIFICATION ON AQUATIC COMMUNITIES

1. Northern New Jersey

Faust and McIntosh (1982) reported that the areas of northern New Jersey potentially susceptible to acidic deposition included the Highlands area in north-central New Jersey and Kittatinny Ridge in northwestern New Jersey (See Figure 1 for location of susceptible surface waters). Each area will be discussed separately.

a. Highlands

Surface waters of greatest concern in the Highlands include a number of lakes and reservoirs, with some of the impoundments serving as water supplies for New Jersey municipalities. Apparently no systematic survey of the biota in the area's lakes and reservoirs has been undertaken, although various relevant reports do exist. For example, fisheries surveys completed during the early 1950's by the New Jersey Department of Fish and Game identified various game species in some of the lakes in the Highlands region. In addition, some systems, such as Green Pond, are routinely surveyed by consultants for lake associations.

Several of the reservoirs in the Highlands provide water for the city of Newark; four, Echo Lake, Canistear, Oak Ridge and Clinton Reservoirs, were recently reported by an outdoor sports writer to be excellent sites for largemouth bass and pickerel fishing. Since largemouth bass (Micropterus salmoides) populations may be stressed by pH values between 5.0 and 5.5 (National Research Council of Canada, 1981), populations in low-alkalinity systems like Clinton Reservoir may be at risk, even though current pH values are typically above 6.0 (see Table 3).

b. Kittatinny Ridge

Surface waters of concern atop the ridge include a number of small lakes and at least one stream originating from these lakes. As is the case with the Highlands, little background biological data have been collected from affected systems. Continuing study by Rutgers University personnel is focusing on six lakes atop the ridge: Blue Mountain Lake, Catfish Pond, Crater Lake, Long Pine Pond, Lake Success and Sunfish Pond (Figure 20). Five of these systems lie on National Park Service land and one (Sunfish Pond) lies in Worthington State Forest. All the lakes are clear-water and moderately to highly acidic (Table 29). Only Blue Mountain Lake has demonstrated any measurable alkalinity. Values for calcium and magnesium in the lakes are generally lower, and, for sulfates, higher than data generated earlier for other surface waters in northern New Jersey (Faust and McIntosh, 1982).

The six lakes are characterized by widely varying biological communities. Although thorough biological surveys are not an objective of the current study, some preliminary conclusions can be reached. Two of the lakes, Crater Lake and Long Pine Pond, appear particularly stressed, with only yellow perch, among the fish, known to survive. Reductions in the diversity of benthic invertebrates and aquatic plants are also apparent in the most acidic lakes. Sphagnum is present in varying quantities at all sites.

Current research is focusing on the activity of the trace elements aluminum, cadmium, lead, mercury and zinc in the six lakes. Initial results (Table 30) indicate that elevated concentrations of aluminum, lead and zinc occur in the water column of all lakes except Blue Mountain, with the highest values occurring in the most acidic systems.

Initial analyses of aquatic plants from three of the lakes (Table 31) show highly variable results, with levels of zinc consistently exceeding those of lead in plants from Catfish Pond, while in more acidic Crater Lake, the reverse occurs, with lead greatly exceeding zinc. Data may indicate that changes in the biological availability of lead in the sediments of Crater Lake may have occurred. Further analyses will indicate whether or not increased lead activity is noted in other compartments of the acidic lakes.

While historical data on the water chemistries of the lakes are largely lacking, it is likely that acidic deposition has contributed to the high acidities noted.

Van Campen's Brook, a trout production stream originating in Blue Mountain Lake and Long Pine Pond, is characterized by alkalinities of less than 10 mg/l as $CaCO_3$ (See Table 29). Following a heavy March snow, decreases in pH of nearly 2

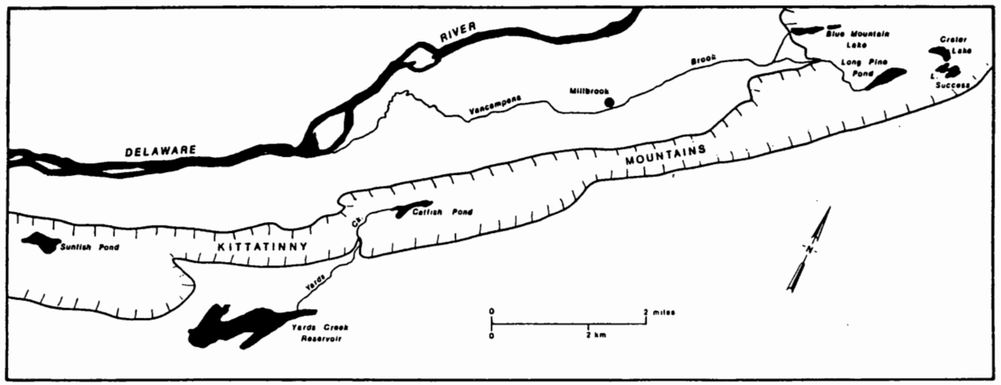


Fig. 20 Map of upper Delaware showing location of six study lakes.

TABLE 29

RANGE OF WATER CHEMISTRY PARAMETERS DETERMINED IN SIX STUDY LAKES

SYSTEM	pH	TOTAL ALKALINITY (mg/l AS $CaCO_3$)	SULFATES (mg/l)	CALCIUM (mg/l)	MAGNESIUM (mg/l)
Blue Mountain Lake[a]	5.3-6.2	2.5-4.2	12.5-22.2	3.6-4.2	1.1-1.4
Lake Success[a]	4.0-5.15	ND	7.9-16.0	3.0-3.8	0.6-0.8
Catfish Pond[b]	4.1-4.9	ND	9.3-13.6	3.2-4.6	0.7-1.0
Sunfish Pond[b]	3.8-6.6	ND	9.3-13.0	3.4-4.3	0.6-0.8
Crater Lake[a]	3.8-4.9	ND	12.5-21.9	4.2-5.1	0.8-1.0
Long Pine Pond[b]	3.6-4.7	ND	9.3-14.5	1.7-2.0	0.4-0.5

[a] based on three sampling dates from 5/84 through 10/84

[b] based on five sampling dates from 9/83 through 10/84

ND = None Detectable

TABLE 30

TOTAL TRACE ELEMENTS (ug/1) IN WATER SAMPLES COLLECTED FROM SIX STUDY LAKES IN MAY AND AUGUST, 1984. VALUES REPRESENT THE RANGE OF SIX DETERMINATIONS.

SYSTEM	pH RANGE	ALUMINUM	CADMIUM	LEAD	ZINC
Blue Mountain	5.3-6.2	26-85	0.05-0.4	0.1-0.9	8-31
Lake Success	4.0-5.15	50-100	0.05-0.5	0.9-7.1	23-55
Catfish Pond	4.1-4.9	79-139	0.1-0.7	0.7-2.6	36-95
Sunfish Pond	3.8-6.6	72-195	0.05-0.2	0.5-2.3	29-36
Crater Lake	3.8-4.9	106-306	0.5-0.7	1.3-4.3	49-68
Long Pine Pond	3.6-4.7	287-508	0.05-0.2	1.6-3.6	46-68

TABLE 31

TRACE ELEMENT CONCENTRATIONS (ug/g DRY WEIGHT) IN SELECTED AQUATIC MACROPHYTES FROM STUDY LAKES. NUMBER OF SAMPLES GIVEN IN PARENTHESIS.

SYSTEM	SPECIES	LEAD	ZINC
Catfish Pond (pH=4.1-4.9)	Utricularia sp. (Bladderwort)	39.5-110.7 (n=7)	81.5-270.0 (n=7)
	Myriophyllum sp. (Water Milfoil)	7.3-13.2 (n=2)	55.1-82.2 (n=2)
	Brasenia sp. (Water Shield)	20.0-52.7 (n=3)	283.3-369.7 (n=3)
	Nymphaea sp. (White Water Lily)	3.6-21.6 (n=6)	33.8-49.1 (n=6)
Long Pine Pond (pH=3.6-4.7)	Utricularia sp. (Bladderwort)	249.7-278.2 (n=2)	97.6-104.0 (n=2)
	Isoetes sp. (Quillwort)	171.7	533.9
	Eriocaulon sp. (Pipewort)	13.9-236.0 (n=4)	33.1-96.1 (n=4)
Crater Lake (pH=3.8-4.9)	Potamogeton sp. (Pondweed)	401.2-487.9 (n=3)	48.8-98.8 (n=3)
	Eriocaulon sp. (Pipewort)	457.8	158.1

units, ranging between 4.3 and 4.5, were noted at several sampling sites along the brook. Although the impact of such short-term influxes of acidity on the native trout are unknown, it seems probable that the stress adversely affects either adult trout or their eggs.

More study of the nature of biological communities present in affected or potentially affected surface waters in the Highlands and on Kittatinny Ridge is needed, particularly in those watersheds where delayed responses to acidic deposition may occur.

2. Southern New Jersey

Assessment of direct effects of acidic deposition on Pinelands aquatic organisms is presently difficult for several reasons. First, there are few historical records of Pinelands aquatic biota, and those that do exist (e.g., Fowler, 1906; Stone, 1911; Boyer, 1916) are primarily general compilations of species found in the entire region. Since documentation of long-term changes requires comprehensive, site-specific historical data, confirmation of any trends, despite much recent work on Pinelands aquatic communities (e.g., Pinelands Commission, 1980; Ehrenfeld, 1983; Morgan et al., 1983; Morgan, 1983; Hastings, 1984), is not possible. Continued study of Pinelands aquatic biota will allow for such documentation in the future.

A second factor is that Pinelands surface waters are naturally quite acidic. pH levels normally observed in the Pinelands are at or below those found in regions with documented acidification effects on the aquatic biota (e.g., The Adirondacks, Ontario, Scandinavia). Thus, the documented biotic changes in other impacted regions cannot be used to infer potential acidic deposition related changes in the Pinelands. Aquatic communities in the Pinelands naturally consist of many of the acid tolerant species whose presence is evidence of acidic deposition effects in other regions (cf. Morgan et al., 1983; Morgan, 1983).

Lastly, assessment of acidic deposition impacts in the Pinelands is complicated by other sources of disturbance that affect pH. Agricultural and residential development of Pinelands watersheds typically results in significantly elevated surface water pH (Pinelands Commission, 1980; Morgan, 1984). In other regions affected by acidic deposition, comparative studies of surface water of varying pH have been effective tools in documenting the effects of acidification (e.g., Wright and Henriksen, 1978; Watt et al., 1979). Because both elevated pH from development and reduced pH from acidic deposition may act simultaneously on Pinelands surface waters, there is considerable uncertainty as to the true pH of an undisturbed water body. Thus, comparative studies within the Pinelands are of limited value in documenting the effects of acidification.

Despite these difficulties in demonstrating direct effects of acidification on Pinelands aquatic organisms, the potential threat to these organisms is very real. Although Pinelands species, by their very nature, are adapted to low pH, this does not mean they prefer high acidity. Their ability to exist in the Pinelands may simply indicate that these species are not as severely affected by low pH as other species, and so they predominate. Tests of pH tolerance on acid loving organisms similar to those found in the Pinelands consistently show a lower tolerance limit of about pH 3 (Bell and Nebeker, 1969; Bell, 1971).

Although the average pH of precipitation in the Pinelands is about 4.0, the pH of specific events may sometimes approach 3.0. In addition, concentration of acidity by evapotranspiration could further increase H^+ concentration. Thus, even in these naturally acidic waters, precipitation events could significantly reduce the pH organisms encounter, at least over the short term. Such pulses of high acidity may be particularly significant for the biota, especially if they occur during critical life history periods (e.g., during breeding). Thus, continued acid inputs into the environment, even for these naturally acid tolerant species, could lead to massive disruption of aquatic communities. Determination of which particular species would be most severely affected by further acidification must await detailed acid tolerance tests and better information on how Pinelands aquatic communities are structured.

Wholesale disruption of these aquatic communities would have ramifications well beyond this particular ecosystem. The Pinelands harbor many species found nowhere else in the State and possibly the nation. Some examples of these rare and endangered aquatic plants, fish, amphibians, and reptiles are listed in Table 32. Pinelands algal and macroinvertebrate communities are too poorly known to be classified as rare and endangered, but many members of these communities are probably also largely restricted to the Pinelands. Loss of even a few of these species through atmospheric induced acidification would greatly diminish the value of the Pinelands as a natural resource for the State and the country. Such an event would be particularly tragic in view of the extensive efforts recently undertaken to preserve this unique ecosystem.

TABLE 32

THREATENED AND ENDANGERED ORGANISMS IN THE NEW JERSEY PINELANDS
AS DETERMINED BY THE NEW JERSEY DIVISION OF FISH, GAME AND WILDLIFE
(AFTER PINELANDS COMMISSION 1980)

AQUATIC MACROPHYTES

Sensitive-joint-vetch
Aeschynomene virginica

Red milkweed
Asclepias rubra

Pine Barrens reedgrass
Calamovilfa brevipilis

Barratt's sedge
Carex barrattii

Rose-colored tickseed
Coreopsis rosea

Knotted spike rush
Eleocharis equisetoides

Resinous boneset
Eupatorium resinosum

Crested yellow orchid
Habenaria cristata

Southern yellow orchid
Habenaria integra

New Jersey rush
Juncus caesariensis

Loesel's twayblade
Liparis loeselii

Boykin's lobelia
Lobelia boykinii

Canby's lobelia
Lobelia canbyi

Hairy ludwigia
Ludwigia hirtella

Yellow asphodel
Narthecium americanum

Floating heart
Nymphoides cordata

Hirst's panic grass
Panicum hirstii

Narrow panic grass
Panicum hemitomon

Slender rattlesnake root
Prenanthes autumnalis

Awned meadow beauty
Rhexia aristosa

Capitate beakrush
Rhynchospora cephalantha

Slender beaked rush
Rhynchospora inundata

Knieskern's beaked rush
Rhynchospora knieskernii

Long's bulrush
Scirpus longii

Slender nut rush
Scleria minor

Sclerolepis
Sclerolepis uniflora

Humped bladderwort
Utricularia gibba

Purple bladderwort
Utricularia purpurea

White-flowered bladderwort
Utricularia olivacea

Reclined bladderwort
Ultricularia resupinata

Yellow-eyed grass
Xyris flexuosa

TABLE 32 (continued)

THREATENED AND ENDANGERED AQUATIC ORGANISMS IN THE NEW JERSEY PINELANDS
AS DETERMINED BY THE NEW JERSEY DIVISION OF FISH, GAME AND WILDLIFE
(AFTER PINELANDS COMMISSION 1980)

FISH*

Ironcolor shiner
Notropis chalybaeus

Yellow bullhead
Ictalurus natalis

Pirate perch
Aphredoderos sayanus

Mud sunfish
Acantharchus pomotis

Blackbanded sunfish
Enneacanthus chaetedon

Banded sunfish
Enneacanthus obesus

Swamp darter
Etheostome fusiforme

AMPHIBIANS

Eastern tiger salamander
Ambystoma t. tigrinum

Eastern mud salamander
Pseudotriton m. montanus

Pine Barrens tree frog
Hyla andersoni

Southern gray tree frog
Hyla chrysoscelis

REPTILES

Wood turtle
Clemmys insculpta

Bog turtle
Clemmys muhlenbergi

* Although these species are not officially designated as threatened or endangered, their distribution in New Jersey is severely limited and mostly restricted to the Pine Barrens. Any alteration in the Pinelands environment would therefore threaten these species within the state.

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PART V: DAMAGE TO MAN-MADE MATERIALS

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EFFECTS OF ACIDIC DEPOSITION ON MAN-MADE MATERIALS

A. INTRODUCTION

The degradation of materials when exposed to the air is dependent on the presence of moisture, reactive gaseous constituents, and airborne particulate matter. The gaseous molecules include nitric acid and its precursors nitric oxide (NO) and nitrogen dioxide (NO_2), hydrochloric acid, and sulfuric acid and its precursor sulfur dioxide (SO_2).[2] The particulate matter may include solids such as soot or liquid droplets of heterogeneous composition. It appears likely that the soot particles will catalyze the degradation reactions. The moisture and reactive ingredients can be provided by condensation of water vapor on a humid day, followed by adsorption of reactive molecules from the gas phase. Alternatively, rain which contains reactive molecules may provide large quantities of reactive species. Where rain is trapped and held, however, such as in bridge supports or statuary with horizontal surfaces containing depressions, the potential for damage is severe.

No rigorous studies of materials damage in New Jersey resulting from acidic deposition have been performed. As described in other sections of this report, however, New Jersey is situated downwind of major industrial areas and is thus expected to be subject to effects similar to those occurring in nearby states. Present information is insufficient to assess the impact of long-range transport in causing materials damage in New Jersey. Very few field studies have been conducted within the state, and only one active corrosion monitoring site (Chester, Morris County) has been established.

B. POTENTIALLY SUSCEPTIBLE MATERIALS

1. Metals and Alloys

The atmospheric corrosion of metals and alloys is an electrochemical process governed by the properties of the materials involved and by the transport of moisture, oxygen, and reactive trace gases to the surfaces. The presence of moisture is crucial to the corrosion process (Phipps and Rice, 1979), and thus rain is potentially very significant, particularly if it contains acidic components and if the surface topography of the materials is such that the rain is retained rather than allowed to run off. The materials of concern are those used in construction, architecture, or electronics. These include iron, steel, aluminum, zinc, copper, bronze, silver, nickel, and tin.

Most metals oxidize upon exposure to the air, and many form mixed surface films that contain chloride, nitrate, carbonate, and sulfate salts. Sulfide and hydroxide salts are also seen on some materials. These findings indicate that degradation of metals involves the complex interactions of a variety of contaminants. Field studies of matched samples in which shielding from rain is assessed suggest that exposure of materials to atmospheric trace gases is responsible for a greater portion of materials degradation than exposure to precipitation (Yocom and Baer, 1984).

2. Masonry, Stone and Marble

Stone composed primarily of calcium carbonate (limestone, marble, etc.) or stones whose mortar is calcium carbonate are vulnerable to damage from atmospheric acidic components. Although the chemical transformation processes are not understood, the principal product is calcium sulfate, which is highly susceptible to erosion from surfaces (Gauri and Holdren, 1981).

Concrete is not attacked to any significant degree by sulfur-containing gases, but is susceptible to cracking induced by the chloride ion. Chloride may be delivered to the concrete surface as a result of rainfall, particle deposition, sea salt deposition, or road salting. The last process appears likely to be the most important in the majority of New Jersey locations.

3. Organic Materials (Rubber, Paints, Polymers, Fabrics)

Organic materials are generally resistant to chemical reactions with the inorganic species responsible for airborne acidity (Yocom and Baer, 1984). Interactions sometimes occur if inorganic pigments and extenders are contained in paint formulations - the reaction of gaseous hydrogen sulfide with lead oxide pigment is an example. Some rubber formulations, polymers, and fabrics are materials that are degraded by exposure to oxidants and sunlight, but there is not definitive evidence of degradation resulting from exposure to acidic precipitation (Shaver et al., 1983).

C. ACIDIC PRECIPITATION EFFECTS

The assessment of the impact of acidic deposition on materials is complicated by the difficulty of separating several potential processes: deposition of acidic gases on dry surfaces, deposition of acidic gases on wet surfaces, deposition of acidic particles on dry or wet surfaces, and deposition of acidic materials in rain or snow. Further, for any field measurements program it is difficult to isolate specific effects attributable to acid deposition when the effects of other atmospheric processes are operating at the same time. The manner in which these effects interact, and the responses of the materials to these interactions are not well understood. New Jersey's heavily industrialized environment, as well as the surrounding area's similar industrial makeup, very likely add many air contaminants that complicate efforts to isolate acidic deposition effects. Few definitive experiments have been performed to examine the different processes; one is thus forced to assess damage due to exposure to the atmosphere rather than damage due to acidic deposition.

1. Metals and Alloys

The chemical composition of the surface films formed on metals and alloys indicates that a variety of atmospheric gases must interact with the metallic surfaces, as do the results of several laboratory experiments (Rice et al., 1981; Graedel et al., 1984). Among the materials to which this conclusion applies are copper, iron, silver, palladium, aluminum, and zinc. Limited statistical studies tend not to reveal correlations of damage with trace gas concentrations, but rather with humidity. There is no satisfactory evidence that major changes in

the concentrations of acidic gases or of ions in precipitation will have any effect on rates of metal degradation. The rates of degradation are known to be quite sensitive to the humidity, however, suggesting that the presence or absence of a water film on the surfaces provides the limitation to the degradation process. In a limited series of laboratory experiments designed to simulate some of the complexity of urban atmospheric environments, a statistically significant relationship between steel corrosion and sulfur dioxide concentration was found (Haynie et al., 1976).

2. Masonry, Stone and Marble

Although the deterioration of marble and concrete over a number of decades is well established, cause and effect relationships for each of these materials are not. The degradation of calcium carbonate-type stone may occur by a number of atmospheric processes, and the predominant mechanism may be site-specific (Cheng and Castillo, 1984). As with metals, it appears that degradation of these materials is generally better correlated to factors other than ambient trace gas concentrations.

3. Organic Materials

Organic materials appear to suffer no significant effects attributable to acidic deposition.

D. ECONOMIC IMPLICATIONS

A draft EPA report on the same topic as this report (Yocom and Baer, 1984) includes the following comments: "Without exception, all of the generalized estimates of material damage costs related to all types of air pollution existing at the time of this report are of questionable value. The reasons for this include the following:

- It is usually not possible to isolate the specific portion of damage and therefore the associated costs created by a given air pollution effect.
- Unrealistic or improper scenarios of use, repair, and replacement of materials susceptible to air pollution damage.
- Incomplete knowledge of substitution scenarios where more expensive material systems may replace more susceptible materials.
- Inadequate knowledge of the exposure conditions of susceptible materials, for example, coexistence of pollutants with other environmental effects such as moisture and temperature, and the physical aspects of exposure such as orientation and degree of sheltering."

If the economic impact of acidic deposition on materials cannot be quantified, however, it is surely of concern in such applications as exposed steel (galvanized or not), other metallic roofing materials (such as copper), and

cultural and historic artifacts (such as bronze statues, historic buildings constructed with susceptible materials, etc.). Even if the exact relationship of materials damage to acidic deposition were known, it would be difficult to assign a value to the damage caused to cultural and historic artifacts. A beginning, however, would be to assemble a list of such artifacts in New Jersey, together with an assessment of their current condition. A preliminary list appears at the end of this section.

A full understanding of the magnitude and causes of acidic deposition effects will require separating the effects of pollutants from the effects produced by unpolluted rain and of separating the effects of rain-supplied reactants from those of gas-phase reactants. These are difficult problems, and it is reasonable to anticipate that several years will be needed to solve them.

SELECTED NEW JERSEY ARTIFACTS POTENTIALLY SUBJECT TO ACIDIC DEPOSITION DEGRADATION (PRELIMINARY)

Masonry Structures

1. Essex County Courthouse (c. 1900), Newark
   National Register file

2. Hunterdon County Courthouse, Flemington
   Part of the Flemington Historic District, HABS-NJ-771

3. Trenton Battle Monument (1891) - includes bronze reliefs
   National Register file

4. New Jersey State House, Trenton
   Part of State House Historic District

5. World War I Memorial, Atlantic City
   National Register file

Metal Structures

1. Finns Point Rear Range Light (1876), Pennsville Township
   National Register file, HAER

2. Lucy, The Margate Elephant, Margate City
   National Register file, HABS-NJ-816

3. New Hampton Pony Pratt Truss Bridge (c. 1868), Lebanon and Washington Townships

4. Statue of Liberty (1886), Jersey City
   National Register file

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PART VI: DAMAGE TO HUMAN HEALTH

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EFFECTS OF ACIDIC DEPOSITION ON HUMAN HEALTH

A. INTRODUCTION

This report summarizes the evidence of the health effects of inhalation of acid sulfates. The conclusions of this report are based on animal experiments and epidemiological and laboratory studies of acid sulfates in humans. The primary focus of this report will be on the effects on the respiratory system. Another potential health consequence of acidic deposition is toxicity of trace metals, since acidic deposition may increase the solubility of these metals in water sources and this may lead to higher levels of consumption of trace metals.

Acid rain, as rain, is unlikely to be associated with adverse health effects, and no direct injury to skin or mucous membranes, such as conjunctiva, has been reported. However, the constituents of acid rain, in aerosol form, combined perhaps with ozone, might cause injury to the lung when inhaled.

A distinction should be made between two types of sulfur oxides in the atmosphere -- particulate oxides and gas phase oxides. Particulate oxides are oxidized forms of sulfates associated with particulate matter. These particles can be carried great distances in the atmosphere and are the oxides usually associated with acidic deposition. Gas phase sulfur oxides, such as sulfur dioxide (SO_2), are generally formed locally and have a more limited range of travel.

A large fraction of SO_2 gas that is emitted into the air is ultimately transformed into particulate sulfate compounds. One of these particulate compounds is sulfuric acid (H_2SO_4). The primary pollutant in acid rain is thought to be particulates containing compounds such as sulfuric acid and its salts. This review will focus mainly on the health effects of H_2SO_4, although the effects of SO_2 will also be described. Very little work on the health effects of nitric acid and its salts have been done, but fortunately the nitrates occur in smaller concentrations in the atmosphere than do the sulfates.

B. RESPIRATORY SYSTEM

The major target of atmospheric pollutants is the lung which is susceptible to injury by air pollutants because of the large surface area exposed. The effects of air pollutants depend on their physical form (e.g. gas versus particles), interaction with other pollutants, and the physiological characteristics of the lung.

The pattern of deposition of particles in the lung depends on the size, shape, and physical properties of particles inhaled. Particles greater than 1 um in diameter tend to be deposited in the upper airways and never reach the alveoli. An important function of the nose is to humidify the inhaled gas. Particles which absorb water can grow from less than 1 um in diameter to up to 4 um in diameter and thus enhance upper airway deposition. About 25% of particles less than 1 um in size are deposited in the upper and middle airways (tracheobroncheal region) and these are carried upward by the mucocilliary blanket and

eventually expectorated. Particles less than 0.2 um in diameter may reach the alveolar spaces. The primary mechanism of removal of these particles is ingestion by resident alveolar macrophages, phagocytic cells normally present in the alveoli.

1. Health Effects of Particulate Sulfur Oxides

a. Methods of study

The health effects of particulate sulfur oxides have been studied in several ways including short-term toxicologic studies in animals, exposure of healthy subjects and patients with lung diseases to the compounds, and epidemiologic studies in populations. The effects on the respiratory system are usually assessed by measurements of pulmonary function tests. It should be noted that when abnormalities of pulmonary function are noted following exposure, substantial injury to the respiratory tract is usually present. It is likely that less severe injury, such as edema and inflammation of the exposed mucous membranes, are not detected using this approach. Therefore, low-level injury of the respiratory system may not be detected using present approaches.

Most studies of H_2SO_4 have shown effects on the lung at concentrations higher that those encountered in the atmosphere in New Jersey. For example, the 24-hour peak sulfate level in the atmosphere in New Jersey in the summer of 1981 was 30-35 ug/m^3 and the 24-hour mean average for the state was 10 ug/m^3 (see Atmospheric Processes Section). These levels of H_2SO_4 cause no detectable disease in healthy adult animals or humans as a result of short-term exposure. Thus, the ambient levels of H_2SO_4 in New Jersey would not be expected to produce pulmonary dysfunction based on the findings of published studies. It is unknown, however, whether low-level exposure may cause disorders not measurable by pulmonary function testing which may constitute a health problem.

b. Animal studies

The effects on the lung of exposure to sulfates has been shown in animal and human studies. These effects have been demonstrated at levels as little as 2-5 times those observed in peak atmospheric concentrations (NIEHS conference, 1984). This leaves little margin of safety and suggests there are potential effects of exposure to sulfates.

Acute toxicity of sulfur oxide has been estimated by mortality studies in experimental animals. The results suggest that fairly high levels of aerosols of sulfuric acid (about 20,000 ug/m^3) are needed to cause mortality in previously healthy animals (Amdur et al., 1952). It appears that concentrations of acid sulfate aerosols up to 1,000 ug/m^3 are largely unsuccessful in altering respiratory function in previously healthy animals with acid sulfate aerosols (Greenberg et al., 1978; Greenberg, 1982). There is some variation in sensitivity among animals; in particular, guinea pigs are more sensitive to sulfur oxides than other species (Amdur et al., 1978, 1978a). There also appear to be differences in relative irritancy of sulfur oxide aerosols in terms of altering pulmonary mechanics in animals. Sulfuric acid generally appears to be more irritating than any of the sulfate salts. For example, guinea pigs show increased airway

resistance in response to low levels of sulfuric acid; those levels of sulfur oxides do not increase airway resistance (Amdur, 1977; Corn et al., 1972).

Chronic exposure to sulfuric acid and sulfates has resulted in changes in respiratory mechanics in animals (Alarie et al., 1973, 1975). However, the effects are less certain than those following acute exposure, and the observed results depend on the duration of exposure, particle size, and chemical composition of the aerosols inhaled at low concentrations.

c. Human studies

There are several factors which may affect the response of humans to inhaled H_2SO_4. The dose and duration of exposure are obviously important. In addition, age, presence or absence of lung disease such as asthma, nose vs. mouth breathing, and exercise will influence the response to inhaled H_2SO_4. It is apparent that H_2SO_4 has toxic effects in the human respiratory system. Controlled studies in humans of the pulmonary effects of H_2SO_4 have shown no effects in normal or asthmatic adults at concentrations of 75 ug/m^3 (Avol et al., 1979) or 100 ug/m^3 (Utell et al., 1981; Sackner and Ford, 1980). However, adult asthmatic subjects have shown significant effects after acute exposures at concentrations of 500 and 1,000 ug/m^3 (Utell et al., 1981). In adolescent asthmatics, exposure to H_2SO_4 in concentrations of 100 ug/m^3 during exercise caused bronchoconstriction (Koenig et al., 1983). Although these studies differ in clinical characteristics of the subjects, particle size, and concentrations of H_2SO_4 used, it is apparent that asthmatics are much more sensitive to the effects of inhaled H_2SO_4 than non-asthmatics. None of these short term studies have shown, however, that bronchoprovocation can be induced at levels of H_2SO_4 present in "worst case" atmospheric conditions, e.g. 75 ug/m^3.

Studies in humans show that exposure to SO_2 may also cause respiratory dysfunction, although SO_2 appears to be less potent on a per weight basis than H_2SO_4. In healthy subjects, studies using SO_2 have shown a consistent effect on respiratory and cardiovascular function at exposure levels of 5 ppm (1,300 ug/m^3). This effect can be potentiated by mouth breathing and increased levels of exercise. For example, breathing SO_2 causes bronchoconstriction in healthy adults at concentrations of 1.0-2.5 ppm (Lawther et al., 1975; Snell and Luchsinger, 1969). In addition, new studies have indicated that H^+ is the most important factor in achieving any physiologic response and the total dose is more important than the sample peak level (NIEHS conference, 1984).

Concentrations of SO_2 of less than 1 ppm have caused bronchoconstriction in some asthmatics (Sheppard et al., 1980, 1981). Cold dry air may potentiate the response to SO_2 since levels of 0.5 ppm have caused bronchoconstriction in the presence of a cold dry environment (Bethel et al., 1983). These observations suggest a heightened responsiveness of asthmatics to SO_2. These levels of exposure in the laboratory are considerably higher than levels of SO_2 in the ambient atmosphere in New Jersey. The observed concentrations of SO_2 in ambient air in New Jersey have been measured as 0.17 ppm (3-hr. maximum), 0.085 ppm (24-hr. maximum), and 0.009 ppm (annual average for 1983 for monitors around the state) (see Atmospheric Processes Section). It thus appears that the highest levels of SO_2 in ambient air in New Jersey are at levels below those causing pulmonary function changes following short-term exposures in asthmatics.

Although the evidence indicates that asthmatics are more likely to be sensitive to H_2SO_4 and SO_2 than non-asthmatics, there may be a subset of non-asthmatics, who are at risk. Recent data shows that the distribution of measured airway reactivity in the general population is unimodal (Cockcroft et al., 1983). Presumably, anyone with increased airway reactivity could be regarded as possibly at increased risk -- this could arbitrarily be set as the upper 10% of the normal population in terms of airway reactivity.

d. Epidemiological studies

The epidemiological evidence implicating particulate sulfate compounds in the aggravation of respiratory symptoms is ambiguous (Holland et al., 1979; Shy, 1979). Some studies have found a positive association between sulfates and mortality and morbidity (Thibodeau et al., 1980; Detels et al., 1981; Knelson et al., 1978) while others have shown no significant correlations between the two (Schwing and MacDonald, 1976). These studies have been criticized for problems with experimental design making it difficult to ascertain with certainty any associations between mortality and morbidity and individual pollutants (Ware et al., 1981).

Although these negative studies have been criticized for problems with experimental design (Ware et al., 1981), a recently published study (Chappie and Lave, 1982) has taken into account all the original objections of these studies, and reanalysis of the data has shown a relationship between ambient sulfate levels and respiratory mortality. Recent data from a carefully conducted French study show a direct relationship between impaired pulmonary function in children and adults and SO_2 levels (Groupe Cooperatif PAARC, 1982a, 1982b). A recent study conducted in Canada showed a significant relationship between acute hospital admissions for respiratory diseases and O_3 and SO_2 levels in the atmosphere (Bates and Sizto, 1983). More importantly, this relationship involved not only admissions for asthma but other acute respiratory disease categories.

It must be noted that these recent epidemologic studies involve morbidity due to exposure to a mixture of pollutants, e.g. sulfates and ozone. It is difficult to separate the effects of sulfates alone since the levels of sulfates and ozone are temporally related. Sulfate levels of 30-35 ug/m^3 are observed in New Jersey during the summer months during periods of stagnant high pressure systems. Peak ozone levels occur during the same meteorological conditions and can reach levels of 18-20 ppm or 1.5 times the ambient standard for ozone.

C. TRACE METAL INTOXICATION

A potential indirect health effect of acidic deposition is trace metal intoxication. This problem has been studied less thoroughly than the effects of inhaled products of sulfates, and researchers are attempting to determine the extent to which acidic deposition predisposes to heavy metal intoxication. Acidified water is known to be capable of dissolving toxic metals to a greater extent than non-acidified water. Toxic metals such as lead, mercury, cadmium and aluminum can be leached out of soils and rocks by acidified water. In addition, acidified water can enhance leaching of trace metals from pipes and conduits in the water supply. No direct relationship has yet been established between acidic

deposition and the degradation of drinking water. Nevertheless, trace metal intoxication is a potential problem resulting from acidification of ground and surface water used for drinking. The effects of acidic deposition on ground water in New Jersey is discussed elsewhere in this report. This section will review the problem of trace metal intoxication and acidic deposition.

1. Overview of Trace Metal Toxicity

Man has always been exposed to trace metals through natural concentrations in soil and water. Large scale mining and certain occupations cause a variety of toxic metal poisonings. Metallic constituents of pesticides and therapeutic agents are an additional source of hazardous exposure. The burning of fossil fuels containing trace metals and the addition of tetraethyl lead in gasoline have now made environmental pollution the major source of trace metal poisoning. Trace metals cannot be easily metabolized and may persist in the body and exert their toxic effects on a variety of physiologic functions. The trace metals of leading concern are lead, mercury, cadmium and aluminum. These metals cause a variety of toxic effects in humans, and most of the illnesses are insidious and are difficult to diagnose because of the subtleties of their effects. This is a problem in epidemiological studies since there is no easily recognizable disease produced by trace metal intoxication.

2. Lead

Lead enters the aquatic system from atmospheric precipitation, street run-off, industrial discharge, and corrosion of lead in pipes and plumbing systems. Corrosion of lead is facilitated by water of low pH (less than 6.5). The overall incidence of lead concentrations above the drinking water standard of 50 ppm throughout New Jersey is not known. However, approximately 14% of domestic wells in parts of Ocean County showed concentrations of lead above this standard (Lord and Kish, Section on Ground Water Processes). Elevated levels have also been found in localized areas of Stafford Township and in Atlantic and Sussex counties (Lord and Kish, pers. comm.). The source of lead levels is not fully established, but the low pH in the ground water makes it easier for the water to dissolve the various possible lead sources. The known wells above the drinking water standard for lead have been shut down. A recent survey of seven lakes and reservoirs in northern New Jersey did not show elevated lead levels (Faust, pers. comm.). Among the water supplies surveyed were Split Rock Reservoir which serves Jersey City and Clyde Potts Reservoir which serves Morristown. There are no data on lead levels in blood or urine in exposed populations to determine the incidence of lead contamination.

3. Mercury

Mercury poisoning from environmental pollution has become a problem as a result of increased concentration of mercury in soil, air, and water due to greater use of mercury in industry and agriculture. However, the effect of acidic deposition on the increased concentrations of mercury is conjectural. Studies in several countries including the United States have shown a correlation between elevated levels of mercury in fish and the acidity of lake and stream habitats (OTA, 1984). No cases of mercury intoxication from consumption of contaminated freshwater fish has been reported in the United States. An epidemic

of mercury poisoning following consumption of contaminated fish in Japan (Minamata disease) was due to industrial contamination (McAlpine and Shukuro, 1958). Several lakes in acid-sensitive regions in the United States have yielded fish with mercury concentrations above the public health standards (OTA, 1984). There are no available data on mercury contamination in freshwater fish in New Jersey.

4. Aluminum

Aluminum is leached from watersheds as acidity of water increases and pH drops below 6.0. Samples from some acidified wells in this country have been found to contain levels of aluminum as high as 1,700 ug/l a content which represents a substantial portion of an individual's daily intake (OTA, 1984). No restrictions have been placed on aluminum concentrations in drinking water or foods. Little is known about the toxicity of aluminum from drinking water in this country. Patients undergoing renal dialysis may develop an encephalopathy due to aluminum intoxication (Alfrey et al., 1976), and water containing more than 50 ug/l of aluminum is thought to be unsafe for dialysis treatment. Some samples of surface water in New England contained greater than 0.10 ppm (OTA, 1984). A comprehensive survey of aluminum concentrations in New Jersey drinking water is not available, but aluminum concentrations in New Jersey surface waters have been quite variable with some samples above 0.10 ppm (100 ug/l) (Section D).

5. Other Metals

Cadmium occurs in nature in association with lead and zinc. Extraction of these ores pollutes the environment. Coal and fossil fuels contain cadmium, and their combustion releases the element into the environment. Drinking water normally does not significantly contribute to cadmium intake. The New Jersey standard for cadmium in drinking water is 0.01 ppm. Shellfish, animal liver and kidney may have concentrations greater than 0.05 ppm, even under normal circumstances. Another potential source of increased cadmium intake is wheat and rice contaminated with cadmium. There is little evidence that cadmium corrosion from pipes or conduits exists (OTA, 1984). Samples from Split Rock Reservoir and Clyde Potts Reservoir in northern New Jersey showed cadmium concentrations of 0.001 ppm or less in a survey conducted in 1983-84 (Faust, pers. comm.). These low levels represent a minimal fraction of the amounts known to cause chronic health effects under industrial exposure conditions. However, cigarette smoke is also a major source, and even small amounts in drinking water may raise a smoker's daily intake above recommended levels. Cadmium in drinking water, however, is not thought to represent a threat to human health.

Copper deficiency is a more widespread problem than copper toxicity. Acidic waters have been shown to be capable of corroding copper from pipes in household distribution systems, and elevated copper levels have been reported in several drinking water samples in New York State and in New England (OTA, 1984). No comparable data from water supplies in New Jersey are available. The hazard to the general population from currently observed copper levels appears quite small.

It is apparent that we must continue to be vigilant about lead levels in drinking water and that we need to understand the variability of aluminum levels in the water from reservoirs. The levels of other toxic metals seem to be low, but, of course, bear watching.

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GLOSSARY OF TERMS

GLOSSARY

A horizon - The surface horizon of a mineral soil having maximum organic matter accumulation, maximum biological activity and/or eluviation of materials such as iron and aluminum oxides and silicate clays.

Acid - A compound, which when dissolved in water, usually gives a pH value less than 7.0

Acid rain - Acid rain refers only to the wet portion of acidic deposition.

Acid soil - A soil with a preponderance of hydrogen and aluminum ions in proportion to hydroxyl ions. Specifically, soils with a pH value less than 7.0. For most practical purposes a soil with a pH value less that 6.6. (The term is usually applied to the surface layer or to the root zone unless specified otherwise.)

Acidic deposition - The transfer of acidic compounds from the atmosphere to the earth's surface. These compounds may be wet or dry. Wet deposition consists of dissolved sulfuric and nitric acids in rain, cloud droplets, and snow. Dry deposition consists of acidic compounds in gaseous or particle form.

Adsorption - The attraction of ions or compounds to the surface of a solid. Soil colloids adsorb large amounts of ions and water.

Aerobic - (i) Having molecular oxygen as a part of the environment. (ii) Growing only in the presence of molecular oxygen, as aerobic organisms. (iii) Occurring only in the presence of molecular oxygen (said of certain chemical or biochemical processes such as aerobic decomposition).

Aerosol - Any suspended solid or liquid particle in the atmosphere.

Alkaline soil - Precisely any soil that has a pH value greater than 7. Practically, a soil with a pH of greater than 7.3. The term is usually applied to surface layer or root zone but may be used to characterize a horizon or a sample thereof.

anaerobic - (i) The absence of molecular oxygen. (ii) Living or functioning in the absence of air or free oxygen.

Anion - An ion with a negative charge.

Anion-exchange capacity - The sum total of exchangeable anions that a soil can adsorb. Expressed as milliequivalents per 100 grams of soil (or of other adsorbing material such as clay).

Aquifer - A formation that contains sufficient saturated permeable material to yield significant quantities of water to wells and springs.

B horizon - A soil horizon usually beneath the A which is characterized by one or both of the following: (1) an accumulation of silicate clays, iron and aluminum oxides, and humus, alone or in combination; (2) a blocky or prismatic structure.

Base saturation - A measure of the degree to which the soil is saturated with exchangeable cations; Ca^{2+}, Mg^{2+}, K^{+}, and Na^{+}.

Buffer capacity - A measure of the resistance of a water to alter its pH value when a strong acid (eg. sulfuric or nitric acids from acidic deposition) or a strong base is added. (See Chapter IV).

Buffer compounds (soils) - The clay, organic matter, and compounds such as carbonates and phosphates which enable the soil to resist appreciable change in pH.

Cation - An ion with a positive charge.

Cation exchange capacity - The sum total of exchangeable cations that a soil can adsorb. Sometimes called "total-exchange capacity", "base-exchange capacity", or "cation-exchange capacity". Expressed in milliequivalents per 100 grams of soil (or of other adsorbing material such as clay).

Combining weight - The formula weight of an ion divided by the charge.

Dry deposition - The transfer of acidic compounds in gaseous or particle form from the atmosphere to the earth's surface.

Eutrophic - Having concentrations of nutrients optimal (or nearly so) for plant or animal growth. (Said of nutrient solutions or of soil solutions.)

Gram equivalent - Otherwise known as equivalent weight, which is defined as the mass in grams of one equivalent. One equivalent of an acid, for instance, is the quantity of the acid which will supply or donate one mole of hydrogen ions. (Ex. 1 mole of $H_2\ SO_4$ furnishes 2 moles of H^{+}. This means that 1 mole of $H_2\ SO_4$ is really the same quantity as 2 equivalents of $H_2\ SO_4$.)

Humus - That more or less stable fraction of the soil organic matter remaining after the major portion of added plant and animal residues have decomposed. Usually it is dark colored.

Hydraulic equivalent - Change in static head per unit of distance in a given direction.

Leaf cuticle - The waxey or fatty layer on the outer wall of the leaf surface cells; it forms an impermeable layer preventing water loss through the leaf surface.

Leguminous nodulation - Nitrogen-fixing nodules on pea or bean plants. Nitrogen fixation is the transformation of nitrogen gas (N_2) in the atmosphere to chemically-bound forms available to plants.

Mor - Raw humus; a type of forest humus layer of unincorporated organic material, usually matted or conpacted or both; distinct from the minerial soil, unless the latter has been blackened by washing in organic matter.

Mull - A humus-rich layer of forested soils consisting of mixed organic and mineral matter. A mull blends into the upper mineral-layers without an abrupt change in soil characteristics.

Mycorrhizae - Certain groups of fungi which invade root tissue and form a mutually beneficial association; these fungi are important in assisting the plant to obtain mineral nutrients from the soil.

Organic acid - Any acid that contains a carbon atom bonded to at least one carbon or hydrogen atom (e.g., acetic acid, $H_3CC(O)OH$).

Oxidant gas - Any gas which is able to give up an electron in an oxidation - reduction reaction (a reaction in which an element that loses electrons is oxidized, and an element that gains electrons is reduced).

Particle - Any solid or liquid substance suspended in the air.

Permeability - A measure of the ability of a porous medium to transmit water.

pH - A logarithmic measure of the hydrogen ion concentration ($-\log [H^{+}]$). A pH value of 7 represents neutrility; values above 7 represent basicity; values below 7 represent acidity. A pH value change of one unit represents a change of acidity by a factor of 10 and a pH value change of 13 represents a change of acidity by a factor of 2.

Soil morphology - The physical constitution, particularly the structural properties, of a soil profile as exhibited by the kinds, thickness, and arrangement of the horizons in the profile, and by the texture, structure, consistency, and porosity of each horizon.

Soil series - The basic unit of soil classification being a subdivision of a family and consisting of soils which are essentially alike in all major profile characteristics except the texture of the A horizon.

Sulfate adsorption - The ability of the soil to adsorb sulfate. The capacity of soil to absorb sulfate increases with high iron and aluminum oxide content, and decreases with high organic matter content.

Throughfall - Precipitation which passes through the canopy of a plant community and falls to the ground. It is usually altered in both amount and composition from precipitation falling directly on the ground.

Total acidity - A measure of the base neutralizing capacity of a water. It is the sum of all compounds that can be neutralized by a base such as NaOH (see Chapter IV).

Total alkalinity - A measure of the acid neutralizing capacity of a water (see Chapter IV).

Total dissolved solids - Total quantity of materials dissolved in a given quantity of water, such as sodium chloride (NaCl).

Virgin soil - Soil that has never been plowed or dug up or otherwise disturbed.

Volume weighted pH - First calculate the volume weighted $[H^+]$ as follows:

Volume weighted $[H^+]$ = the sum of the volume of rain fall for each event in a given time period multiplied by the respective $[H^+]$ for each event divided by the total volume of rainfall.

$$\text{Vol. weighted } [H^+] = \frac{\sum_i [H+]_i \times V_i}{\sum_i V_i}$$

The negative logarithm of this vol. weighted $[H^+]$ is the vol. weighted pH. Thus, if for a month the volume weighted $[H^+]$ at a New Jersey monitor was 100 mmols/liter, the volume weighted pH would be 4 for that month.

Units

ppm	parts per million -- a unit of concentration.
mg/l	milligrams per liter -- a unit of concentration, based to measure dissolved substances in water. 1 ppm = 1 mg/l since 1 liter of water weights 1,000,000 mg.
ug/m^3	micrograms per cubic meter -- unit of concentration.
meq/100g	milliequivalents per hundred grams -- a unit of concentration used to measure cation exchange capacity in the soil.
um	a unit of length equal to one millionth of a meter.
eq	equivalent
meq	one thousandth of an equivalent.
hectare	2.5 acres.
meq/l	milliequivalent per liter, the concentration value in milligrams per liter divided by the combining weight. For a magnesium concentration of 10 mg/l, the combining weight would be the formula weight (24.3) divided by the charge (2), or 12.15. Milliequivalents per liter is expressed by dividing the concentration (10 mg/l) by the combining weight (12.15) resulting in a value of 0.82 milliequivalents per liter.