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*by Ellis B. Cowling**

I. Introduction

Thirty years ago the terms "acid precipitation" and "acid rain" were bits of esoteric jargon used almost exclusively by scientists in certain specialized fields of ecology and atmospheric chemistry. Recently these terms have become worrisome household words and objects of national and international debate. While they have inspired sensational and sometimes exaggerated headlines about "death from the sky," they have also inspired a deliberate and very careful examination of the role of man in the biogeochemistry and chemical climatology of the earth.

Since the beginning of life on earth, plants have obtained an important part of their nutrients from the atmosphere. From time to time, plants and animals have also been injured by toxic substances dispersed in the atmosphere. Since the industrial revolution began, people have become a progressively more important force in the biogeochemical circulation of the earth—we have added larger and larger amounts of all sorts of substances to those that circulate naturally among the air, the water, the soil, and all the living things of the earth. Some of these man-made substances are beneficial nutrients; some are inert; others are toxic and therefore injurious to plants and animals; still others are either beneficial or injurious depending on their concentration or the nature of the organism receiving the deposition. Because man now exerts so many important influences on the chemical climate of the earth, it is essential that we understand the sources, transport, transformations, and chemistry of atmospheric deposition. We must also understand the various ecological influences that the deposited substances exert on forests, fish, crops, soils, surface waters, and man-made structures.

The story of acid rain is instructive. It shows how much we need to learn about the interacting physical, chemical, and biological worlds in which we live and work, and which we affect in many ways, both intended and unintended. It also emphasizes how badly we need more timely com-

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munication among scientists in different specialties as well as among the scientific community, leaders of government and industry, and the public at large.

This brief paper seeks to relate acid precipitation to the broad subject of atmospheric deposition and to characterize the phenomenon, its consequences, and possible approaches to its management.

A. *Atmospheric Deposition—The Fundamental Phenomenon*

Gases, liquids, vapors, and solids of many sorts from many natural and man-made sources are constantly circulated between the atmosphere and the surface waters, soils, plants, and animals of the earth. Water and air are the media through which these substances move; the atmosphere provides the linkage. Every substance emitted into the air is returned to the earth in one chemical form or another by one or more of three major mechanisms: 1) wet fallout of substances dissolved or suspended in rain, snow, hail, dew, fog, or frost; 2) gravitational settling and impaction of coarse particles and fine aerosols; and 3) adsorption or absorption of gases. Processes 2 and 3 are components of so-called dry deposition. Process 1 (precipitation) is the only part of atmospheric deposition that is now monitored routinely because we do not yet know how to construct a network to monitor dry deposition properly.

B. *Evidence of Acidity*

A special feature of the acidic substances in precipitation is their tendency to increase the solubility, and therefore the mobility, of metal cations—positively charged ions—in soil. The increase in mobility sometimes leads to leaching of toxic metal ions and their accumulation in surface and ground waters.

The acidity or alkalinity of any solution is measured in terms of pH—the negative logarithm of the concentration of hydrogen ions. The pH of a solution reflects the balance among all of the positive and negative ions it contains. At pH 7, precipitation is chemically neutral; as pH decreases, it is increasingly acidic, and as pH increases it is increasingly alkaline.

Even "clean" precipitation is slightly acidic—about pH 5.6—because it absorbs acid-forming substances naturally present in the atmosphere. The most important, carbon dioxide, reacts with water to form carbonic acid. Acid precipitation is therefore defined as precipitation having a pH of less than 5.6. Because pH is a logarithmic scale, a change of one pH unit represents a tenfold change in acidity: rain of pH 4.6, for example, is 10 times more acidic than rain of pH 5.6. For comparison, Lake Ontario has a pH of about 8.0; lakes containing fish are considered seriously acidic at pH 5.5 or 5.0.

Contemporary acid precipitation results largely from sulfur dioxide and nitrogen oxides emitted during combustion of fossil fuels and by other human activities. These gases are transformed in air, and the reaction products are absorbed by water droplets to form a dilute solution of sulfuric or nitric acids, or both.

Rainstorms with pH's as low as 3.0 to 3.5 are now fairly common in the eastern United States and Canada, where an average annual pH of rain and snow below 4.5 has been observed in recent years.

C. *Long-Distance Transport*

A distinctive feature of acid precipitation is the long-distance transport of its precursors. In part because of tall smoke-stacks, air pollutants often travel long distances on prevailing winds; the longer they remain aloft, the more likely they are to undergo physical and chemical transformations before being deposited at the surface. Sulfur dioxide and nitrogen oxides may travel hundreds or even thousands of kilometers, being converted to acidic compounds en route and deposited in acid precipitation far from their points of emission. Scientists in both Norway and Massachusetts have shown that much of the sulfur in the air over these states comes from human activities outside their borders.

D. *Ecological Consequences*

What are the biological and other consequences of atmospheric deposition in general and of acid precipitation in particular? A very important beneficial effect of atmospheric deposition is providing nutrients for the growth and development of plants. Atmospheric sources of nutrients are especially important in natural ecosystems such as lakes, estuaries, wetlands, forests, and rangelands, where nutrients from other sources are scarce and where fertilization is not a normal management procedure. Sixteen elements are essential for the growth of plants. All sixteen are dispersed in the atmosphere, and all sixteen can be taken up by plants directly through foliar organs as well as through roots.

The most significant detrimental effects of atmospheric deposition have been noted in regions where vegetation has been exposed to high concentrations of toxic gases such as sulfur dioxide, ozone, nitrogen oxides, and fluoride. The most important effects of acid precipitation have been observed in aquatic and wetland ecosystems. Rapid extinction of fish populations and changes in other forms of aquatic life have been observed in acidified waters of low ionic strength in Europe, Canada, and the United States. These changes are induced not only by direct acid stress but also by accumulation of toxic metal ions, especially aluminum. Acidification of ground water has been detected recently in western Sweden and is leading to concern about metal-ion contamination of drinking water supplies. Accelerated corrosion of metals, increased weathering of stone statuary and monuments, and damage to textiles and painted surfaces have been reported in various countries.

Much less is known about the effects of acid precipitation on crops, forests, and soils than on aquatic ecosystems. Various specific biological effects have been demonstrated in controlled laboratory and field experiments with simulated acid precipitation. These effects include: 1) erosion of protective waxes on leaf surfaces; 2) inhibition of nodulation by symbiotic nitrogen-fixing organisms associated with legumes such as soybeans; 3) leaching of calcium and other beneficial nutrients from soils; and 4) alteration of the normal growth processes and host-pathogen interactions of plants. Despite these various specific effects, however, reliable evidence of economic damage to agricultural crops, forests, and other vegetation by naturally occuring acid precipitation has rarely been reported.

E. *Early Awareness of Acid Precipitation*

Many features of the acid-rain phenomenon were discovered by an English chemist named Robert Angus Smith. In a remarkable book intitled "Air and Rain: The Beginnings of Chemical Climatology," published in 1872, Smith first used the term "acid rain." He also demonstrated that the chemistry of precipitation is influenced by such regional factors as combustion of coal, decomposition of organic matter, wind trajectories, proximity to the sea, and amount and frequency of rain or snow. Smith noted acid-rain damage to plants and materials and commented on the atmospheric deposition of heavy metals in industrial regions.

Unfortunately, however, Smith's pioneering and prophetic book seems to have been overlooked by essentially every subsequent investigator of the acid-precipitation phenomenon. Eville Gorham has developed the first detailed account of Smith's early work for a report by the National Academy of Sciences.

II. Acid Precipitation Research

Research on acid precipitation began in Europe and later spread to North America. Our present knowledge developed along parallel lines in three separate fields of science: limnology; agriculture; and atmospheric chemistry.

A. *Limnological Research*

The links between the chemistry of precipitation and the chemistry of streams and lakes were still obscure until the mid-1950's, when an aquatic ecologist, Eville Gorham, launched a series of investigations that did much to illuminate these relationships. first in England and later in Canada, Gorham and his colleagues demonstrated these critical points:

—Much of the acidity of precipitation in industrial regions is due to atmospheric emissions from combustion of fossil fuels.
—Declining alkalinity in lakes and rising acidity in bog waters are due to acid precipitation.
—Much of the free acidity in soils receiving acid precipitation is due to

sulfuric acid.
—Acid rain resulting from emissions of sulfur dioxide contributes to the deterioration of vegetation, of soils, and of the quality of lake waters around metal smelters.

By the early 1960's, Gorham had laid much of the foundation of our present understanding of the sources and the limnological and ecological consequences of acid precipitation. But his work, like that of Smith a century before, was greeted by a thundering silence from both the scientific community and the public. A plausible explanation is that Gorham's work, being highly interdisciplinary, was published in a diverse array of scientific journals. In any event, lack of recognition delayed by at least another 10 years the birth of scientific and public awareness of the acid-precipitation problem.

B. Agricultural Research

Atmospheric deposition has been recognized as a source of nutrients for plants since the 17th century. In the mid-1940's, a Swedish soil scientist named Hans Egnér began to study this process systematically. In 1948 he created the first long-term network for monitoring atmospheric deposition. Sampling buckets were placed at experimental farms all over Sweden; the collected deposition was analyzed monthly for major chemical parameters, including acidity.,

Other agricultural scientists gradually extended Egnér's network to Norway, Denmark, Finland, and, later to most of western and central Europe. In 1956, the International Meteorological Institute in Stockholm assumed responsibility for the expanded European Air Chemistry Network. In 1957, as part of the International Geophysical Year, the network was extended eastward to Poland and much of the Soviet Union. The European Air Chemistry Network has operated continuously for nearly three decades, with form 100 to 175 sampling stations. Data from the network constitute the only long-term, continent-wide record of the changing chemistry of precipitation.

C. Atmospheric Chemistry Research

The transport and deposition of water by atmospheric processes has long been recognized. But the notion that such processes play a major role in the transport, dispersal, and physical and chemical transformation of many other substances was only a working hypothesis in the early 1950's. Two Swedish atmospheric scientists, Carl Gustav Rossby and Erik Eriksson, championed these ideas and began to test them experimentally. Data from their colleague Egnér's monitoring network proved a very powerful tool. During the 1950's, Eriksson sponsored a series of European conferences of atmospheric chemistry and dispersal processes that attracted the interest of many biologist and meteorologists.

D. Integration Across Scientific Disciplines

Early in the 1960's, a Swedish soil scientist named Svante Odén began the first major integration of findings in limnology, agriculture, and atmospheric chemistry. As a young colleague of Egnér, Rossby, and Eriksson, Odén started a Scandinavian network for monitoring surface-water chemistry in 1961. When he combined data from this network with data from the European Air Chemistry Network, he began to see a general pattern of trends and relationships. Odén's analyses showed in part that:

—Acid precipitation was a large-scale regional phenomenon in Europe.
—Precipitation and surface waters were becoming more acidic.
—sulfur- and nitrogen-containing air pollutants were traveling long distances—from 100 to 2000 km—throughout Europe.

Oden also proposed a series of hypotheses about the biological and other consequences of acid precipitation:

—Decline of fish populations;
—Decrease in forest growth;
—Increase in plant diseases;
—Accelerated damage to materials.

Odén first published his ideas in Stockholm's newspaper *Dagens Nyheter* in 1967. This news story, which attracted the attention of the press all over Europe, began the process of public education about acid precipitation. A year later, Odén summarized his discoveries and postulations in an Ecology Bulletin for the Swedish National Science Research Council. Both the news story and the Ecology Bulletin stimulated a veritable storm of scientific controversy. Suddenly limnological, agricultural, forest, and atmospheric scientists began talking to each other abut acid precipitation—and designing experimental tests to prove or disprove many of Odén's unconventional ideas.

E. Governmental Activities

By the end of the 1960's, acid precipitation was stimulating a great deal of concern among both scientists and the public in Europe. Governments began to react:

—The Swedish government started an inquiry that culminated in 1972 in Sweden's Case Study for the United Nations Conference on the Human Environment—"Air Pollution Across National Boundaries: The Impact of Sulfur in Air and Precipitation."
—In 1972, three scientific organizations in Norway launched an interagency research project—" 'Acid Precipitation: Effects on Forests and Fish"—budgeted at about $2 million (U.S.) annually.
—During 1973-75, the Organization for Economic Cooperation and Development (OECD) conducted a study of long-range transport and deposition of atmospheric sulfur in eastern and western Europe.

The OECD findings, published in 1977, showed that the area of acid precipitation included almost all of northwestern Europe. The findings confirmed the long-distance transport of pollutants and showed that air quality in any European country may be measurably affected by emissions in other European countries.

In October of 1977, the Economic Commission for Europe (ECE) originated the Cooperative Programme for Monitoring and Evaluating the Long-Range Transmission of Air Pollutants in Europe, which is now in operation. Nearing completion within ECE are discussions of a multinational convention governing nations' responsibilities for combating long-range, transboundary transport of air pollutants.

III. North American Research on Acid Rain

Although concern about acid precipitation in North America arose first in Canada, no consistent and long-term measurements of the acidity of precipitation had been made in either Canada or the United States until 1972. Consequently, we still do not have a clear understanding of trends in the extent or severity of acid rain on this continent.

The first detailed studies of precipitation chemistry in the United States were done in Tennessee in the 1920's. Emphasis was given to nutrients for growth of crops. During the mid-1950's, the first regional monitoring of precipitation chemistry was undertaken by a group of State Agricultural Experiment Station scientists. The first national monitoring program was established in 1960 by the Public Health Service. As with all such studies in this country, however, these early programs were redirected or terminated before they could compile a continuing record of long-term trends.

Both Canada and the United States have recently started long-term programs for analysis of precipitation chemistry. The Canadian Network of Sampling Precipitation (CANSAP) began in 1976; the National Atmospheric Deposition Program (NADP) in the United States in 1978. By early 1980, some 40 sampling stations were operating in Canada, and 60 in this country.

The data from these programs show that the area receiving acid precipitation in the United States now embraces every state east of the Mississippi River. Although sulfuric acid has been found to be the dominant source of acidity both here and in Europe, nitric acid accounts for almost one third, and the fraction is rising.

In 1978, the governments of Canada and the United States established a Bilateral Research Consultation Group on the Long-Range Transport of Air Pollutants (LRTAP) to coordinate the exchange of scientific information on acid precipitation. LRTAP has recently documented the transboundary flow of air pollutants in eastern North America. It appears that about eleven times more oxides of nitrogen and two to four times more sulfur oxides are transported from the United

States to Canada than the reverse.

IV. Effects of Acid Precipitation

Although research on the effects of acid precipitation in North America has been limited in comparison with the European effort, some effects have been detected. Not surprisingly, they parallel the Scandinavian experience.

Acidification has caused the decline of various species of fish, especially trout and salmon, in thousands of lakes and rivers in southern Norway and Sweden during the past two decades. Similar effects have been found at many locations in the Laurentian Shield area of North America. A decline in the spawning success of Atlantic salmon has been correlated with rising acidity in streams in Quebec and the Maritime Provinces.

In the Adirondack Mountains of New York State, where rain has an average pH between 4.0 and 4.5, all the fish in about 100 lakes above 610 meters have been lost because of acidification. The decline in sport fishing is estimated to cost the region more than $1 million annually in tourism.

Although significant damage to forest soils, to agricultural crops, or to human health has not been demonstrated in North America or in Europe, experiments with simulated acid rain and other evidence indicate that the potenital is real. Sulfur and nitrogen oxides are known to damage many kinds of materials including those in historic monuments. Although acid precipitation almost certainly contributes to these effects, we cannot yet distinguish its share from that of other air pollutants.

V. A National Program

A significant effort has been under way during the past three years to respond to the problems of acid deposition: to create a permanent monitoring program that is national in scope and to encourage and coordinate research on effects. In the fall of 1977, the President's Council on Environmental Quality contracted with the Association of State Agricultural Experiment Stations of the North Central Region (organizers of the National Atmospheric Deposition Program) to draft "A National Program for Assessing the Problem of Atmospheric Deposition (Acid Rain)." This document provided the basis for a Presidential Initiative on Acid Precipitation, which President Carter announced in his Second Environmental Message.

The Presidential Initiative calls for a 10-year program of research on the causes and consequences of acid precipitation. The President directed that a standing Acid Rain Coordination Committee be established to plan and manage the program. The Committee comprises policy-level representatives from the Departments of Agriculture, Interior, Energy, Commerce, and State, the Environmantal Protection Agency, the National Science Foundantion, the Council on Environmental Quality, and the Office of Science and Technology Policy. By mid-1980, the Acid Rain Coordination Committee was well along in formulating detailed plans for the 10-year program.

VI. Acid Rain and Public Policy

Where do we stand today in the formulation of public policy on acid precipitation? Two points can be made:

—As of mid-1980, the effects of acid precipitation and associated metal-ion toxicity on aquatic ecosystems are sufficient to warrant preliminary formulation of management and ameliorative policies.
—As of mid-1980, the potenital effects of acid precipitation on terrestrial organisms are not understood well enough to justify consideration of management strategies at this time.

It is clear that the phenomenon of acid deposition demands much more scientific research and scientific and public discussion. The Presidential Initiative on Acid Precipitation and recent Congressional interest in the problem offerincreasing hope that the United States will do its part, together with Canada and other nations, in developing the new knowledge necessary for an effective program of management.

Our present energy and environmental policies are resulting in change in the chemical climate of the earth. Important changes are taking place in the lakes, streams, fields, and forests on which both the abundance and the quality of our life depends. Wide management of these resources requires communication among biologists, atmospheric chemists, meteorologists, hydrologists, regulatory strategists and legal advisors, and both industrial and political leaders in the United States and Canada. Let us continue that communication by focusing our collective knowledge and good will on the development of more adequate plans for management of air quality and its effects on plants, animals, soils, surface waters, and materials. The soon-to-be-consummated debate about possible revision of the Clean Air Act is one of the important occasions for such collective reasoning together.