

Acid Rain: What We Must Do

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Although the phenomenon of acid rain was identified in the city of Manchester, England by Angus Smith as long ago as 1852, and described more thoroughly in 1872, modern scientific research has been going on only since the mid-1950s (Cowling 1982; Gorham 1981). Public concern about the problem in North America has only arisen within the last ten years (Likens, Bormann, and Johnson 1972). In Europe public concern began in the late 1960s (Odén 1968). My purpose here is to address questions about the nature, sources, and history of acid rain. In addition I discuss the questions: Why is acid rain a problem? Is acid rain getting worse? What is the threat of further problems? Finally, I conclude that it is time to act on this problem, and I recommend an appropriate course of action.

What is the Nature of Acid Rain?

Acid precipitation, whose pH is often well below 5 and can occasionally drop below 3, is a mixture of strong mineral acids—sulfuric, nitric, and in some locations hydrochloric—in rain and snow. But we must also recognize that the acids are deposited from the air at times when rain and snow are not falling, by processes of dry deposition. Such deposition is seldom measured, and requires much more study (Engelmann and Sehmel 1976; Galloway, Eisenreich, and Scott 1980a). There may also be carbonic acid and weak organic acids present in rain, but they seldom lower annual average pH below 5.0 (Galloway, *et al.* 1982). Several toxic heavy metals including lead, cadmium, and mercury often accompany acid rain (Galloway, *et al.* 1980a), which may leach such metals from soils along with toxic aluminum (Dickson 1980). A variety of organic micropollutants is also found in acid rain, including alkanes, phthalic acid esters, fatty acid ethylesters, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons—some of which are carcinogens (Lunde, *et al.* 1976; Galloway, Eisenreich, and Scott 1980a). Some plant nutrients, for example nitrogen, sulfur, potassium, and calcium, are also deposited in acid rain, but because they are accompanied by a diverse array of toxins, the nutrients are better supplied directly as fertilizer.

What are the Sources of Acid Rain?

Several sources of acid rain are natural. For instance, Bottini (1939) recorded acid rain dominated by hydrochloric acid around the volcano Vesuvius. Hutchinson, *et al.* (1979) found acid rain falling on the Smoking Hills in arctic Canada, where natural fires in exposed beds of lignite generate large amounts of sulfur oxides that are converted to sulfuric acid. Coastal mudflats and salt marshes are a possible source as well, generating volatile sulfur compounds that may become oxidized in the atmosphere to sulfuric acid (Hitchcock,

Spiller, and Wilson 1980). It appears that ammonia is also generated from local sources and neutralizes part of the sulfuric acid generated. Convincing evidence is lacking for major acid emissions from such coastal sources, but the possibility needs more investigation. An argument against their importance can be drawn from the absence of strong acidity in rain at coastal stations in the U.S. (Likens 1976; Likens, *et al.* 1979). We can see a striking decline of chloride in rain away from the U.S. coast as the influence of sea spray declines (Junge and Werby 1958; Eriksson 1960), but we do not see similar declines of hydrogen and sulfate ions.

The major source of acid rain is not natural, but anthropogenic. The combustion of fossil fuels, chiefly coal and oil, and the smelting of sulfide minerals, generate the gaseous precursors of acid rain, sulfur and nitrogen oxides, which can contaminate not only local but also far distant regions (Drabløs and Tollan 1980; National Research Council 1981). During long-range transport the acids may be wholly or partly neutralized, for instance by alkaline fly-ash (Shannon and Fine 1974), cement dust (Barrett and Brodin 1955), blown soil particles (Gorham 1976; Munger 1982), or ammonia generated in agricultural areas (Lau and Charlson 1977). Combustion of fossil fuels appears to generate comparatively small amounts of ammonia (Junge 1958; Robinson and Robbins 1970), though urban concentrations tend to exceed those of rural areas (National Research Council 1979).

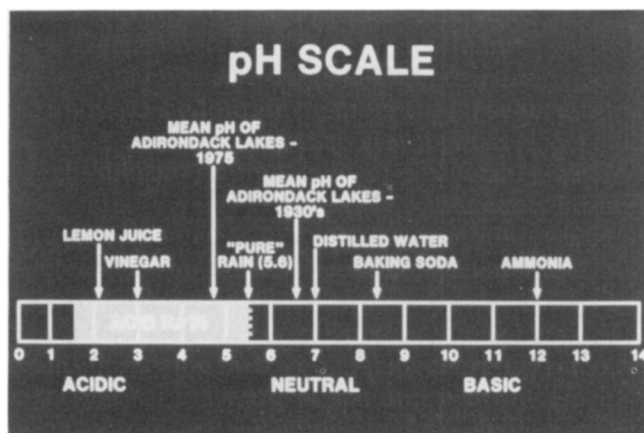
To establish the origin of acid rain most convincingly, it is best to work relatively close to a source, where the situation is less likely to be complicated by extraneous processes—mixing from other sources, neutralization by alkaline materials—than it is farther away. For example, the hydrochloric acid in rain close to Vesuvius, which brings rain pH down as low as 2.8,

must be volcanic in origin—the more so because sulfate is very low in relation to chloride. Ponds near the Smoking Hills in arctic Canada, with pH values down to 1.8 and very high sulfate concentrations, must owe their acidity to the sulfuric acid generated by burning lignite there. Similarly, Angus Smith (1852) and Crowther and Ruston (1911) found acidity to decrease sharply going away from the cities of Manchester and Leeds in England. The same decline was evident on a regional basis in Britain, as shown also by Smith (1872).

An equally convincing case for the anthropogenic sources of acid rain can be made for the area northeast of the iron-sintering plant at Wawa on the northeast shore of Lake Superior. The pH values of lakes in that region—which are normally between 6 and 7—decline to values as low as 3.2 close to the sintering plant, as sulfates rise tenfold (Gordon and Gorham 1963). There are some naturally acid bog lakes in the Wawa region, but they can be clearly distinguished from the lakes acidified by humans by their brown tea-color. For the acidity of clearwater lakes close to the sintering plant, there is no credible source other than the sulfur dioxide emitted from that plant.

Even at considerable distances it may still be possible to ascribe rain acids clearly to their anthropogenic sources. For instance, in the English Lake District the rain is acid and rich in sulfates when the wind blows from the south and east, in the direction of the major industrial districts of northern Britain. Those rains are also black and sooty. In contrast, when the wind blows from the west—from the Irish Sea—the rains are clear, lower in acidity, and rich in sea salt (Gorham 1955, 1958a). On a broader scale in Britain, bog pools fed wholly by atmospheric precipitation become increasingly acid as one travels from remote areas toward the major industrial centers of northern England. In the remote bog pools pH values are usually around 4.5 owing to natural processes of acidification, whereas close to the industrial areas the pH declines to 3.9, and even to as low as 3.2 immediately outside the industrial city of Sheffield (Gorham 1958b).

In the northeastern United States the areas of heavy sulfate deposition (Eriksson 1960) and acid rain (Likens 1976; Likens, *et al.* 1979) are in and downwind of the chief coal-using states of Ohio, Indiana, and Illinois, which produce about one-quarter of total U.S. emissions of sulfur oxides and one-sixth of nitrogen oxide emissions. Deposition of heavy metals is focused upon the same area (Bridge and Fairchild 1981). The only credible reason for this being the center of acid rain in the United States is the abundance of closely packed, urban/industrial emission sources in this region, where anthropogenic emissions strongly dominate those from natural sources (Galloway and Whelpdale 1980). Unfortunately, the large number of such sources



The degree of acidity or alkalinity of a substance is measured on the pH scale. The pH scale increases logarithmically so that each whole number is a tenfold difference.

makes it impossible to trace damage to an individual site from an individual source. There is a pool of acid air pollution over the whole northeastern part of the continent, and satellite photos show that within that area pollution can travel long distances, even into the midwest with certain weather patterns (Robinson, Husar, and Galloway 1978).

Not all situations are this clear. For example, in Nova Scotia there is a gradient of precipitation pH from 4.6 in the southwest—near urban/industrial sources of sulfur and nitrogen oxides in New England—to 5.3 in northeastern parts of the province (Underwood 1979). However, hydrogen ion is not closely related either to sulfate or to nitrate ion in these rains, presumably because of interactions among diverse emitting sources and equally diverse sources of neutralization.

One complicating factor is the mix of different acids involved. On the west coast of the United States, where vehicle emissions predominate, it appears that nitric acid is the chief acidifying agent in rain (Liljestrand and Morgan 1978; McColl and Bush 1978). In northern English cities, where coal high in chlorine is burned, hydrochloric acid appears to be the dominant acid, even though sulfate exceeds chloride in the rain (Gorham 1958c). Neutralizing agents are also of great significance. In the southeastern United States hydrogen ions correlate with sulfate minus calcium ions, and bicarbonate ions correlate with calcium minus sulfate ions, because some of the rain acids are neutralized by blown soil dust (Fisher 1968).

In Minnesota we have a mixture of different acids and different neutralizing agents as we pass across the northern part of the state from North Dakota. In the year 1978-79 precipitation pH ranged from 5.3 in agricultural southeastern North Dakota to 4.6 in forested northeastern Minnesota. Hydrogen ion was not clearly related either to sulfate or to nitrate, and the confounding factors appear to have been wind-blown calcareous dust and gaseous ammonia from the cultivated prairie (Munger 1982). In this connection it is important to remember that although ammonia may neutralize sulfuric acid in the atmosphere, producing ammonium sulfate, the product is an acidifying agent in sensitive soils (Reuss 1975). Farmers have known for a long time that if they fertilize with ammonium sulfate their soils will require liming.

When Did Acid Rain Become a Problem?

The phenomenon of acid rain may have occurred in some cities as far back as the 17th century. Sulfur pollution became serious then in London (Brimblecombe 1977, 1978). Moreover, we know that deposits of sulfate in the Greenland icecap have increased coincident with the beginning of the industrial age in Europe (Koide and Goldberg 1971). The spread of acid rain over broad rural regions, and internationally through

transboundary pollution, has probably developed chiefly in this century, and especially since World War II. In the last several decades combustion of fossil fuels has risen in western Europe and the northeastern United States, especially in power plants. Tall smoke stacks have been introduced to meet air quality standards, and this has allowed pollutants to spread more widely from their source regions. At the same time stack-scrubbing devices have come into use, removing potentially neutralizing fly-ash from the emissions.

Before passing to another question, I would like to present a brief history of acid rain studies (Gorham 1981; Cowling 1982). In cities (but not in the surrounding countryside) it was first described by Angus Smith in 1852, working in and around the city of Manchester in northern England. In 1872 Smith expanded his studies to cover the United Kingdom and published his classic book *Air and Rain*. Then in 1911 and 1912 Crowther and Ruston and Cohen and Ruston, working in and around the city of Leeds in northern England, further expanded on Smith's early work and studied the effects of acid rain upon plants and upon microbes involved in the nitrogen cycle. The acidification of rain in a number of British cities was later demonstrated by Parker (1955). Acid rain in rural areas was probably first demonstrated by Bottini (1939) near the volcano Vesuvius. Between 1939 and 1954 there were scattered pH measurements of rain in a variety of areas, but these data were not related to the acid-rain phenomenon.

A new era in acid rain studies began in the mid-1950s, when it was discovered to be widespread in rural areas far from urban, anthropogenic sources. In 1955 Barrett and Brodin discovered acid rain in Scandinavia, Houghton found fog and cloud water in New England to be acid, and I observed acid rain in the



In both Canada and the U.S., about half of the NO_x emissions come from the transportation sector, particularly from the tailpipes of moving vehicles. (Photograph courtesy of the Canadian Embassy.)

English Lake District whenever the wind blew from the urban/industrial areas. Later on I returned to Canada and worked on the acidification of lakes by acid rain from the metal-processing plants at Sudbury (Gorham and Gordon 1960) and Wawa (Gordon and Gorham 1963). Recognition that the ecological damage done by acid rain could be a major international problem came with the work of Svante Odén in Sweden (1968). Similar problems were recognized in the United States by Likens and his associates (1972).

Why is Acid Rain a Problem?

The primary reason for concern is that acid rain acidifies streams and lakes on coarse, sandy soils low in lime. The effect is seen particularly in headwater areas and in wet montane environments, wherever sulfate loading from anthropogenic sources is strong. For example, in northern England several lakes on soils poor in lime had lost their alkalinity and were very acid by the 1950s (Mackereth 1957; Gorham 1957). In southern Scandinavia thousands of lakes and streams have been severely acidified over the past few decades (Likens, *et al.* 1979). Hundreds of lakes in the Adirondacks, Maine, Ontario, and Nova Scotia have likewise become acidified in recent years, and thousands more are threatened, particularly in Canada. Acidification may be exceptionally severe during snow melt, and this may be the first sign of the phenomenon in lakes and streams that otherwise appear normal (Galloway, *et al.* 1980b).

All of these lakes are either near to or downwind from major anthropogenic sources of sulfur and nitrogen oxides. Lakes on similar substrata upwind of such sources, or far distant from them, have not become acidified, for example in northern Norway and Sweden, Finland, the Experimental Lakes Area of northwest Ontario, eastern Newfoundland, and the Boundary Waters Canoe Area of northern Minnesota. These areas are, however, at risk if acid rain continues, and especially if it worsens and spreads. Not all lakes in the sensitive regions exposed to acid rain become acidified at the same rate. In the Adirondacks, three lakes close together have exhibited very different responses that depend upon the characteristics of their watersheds, as shown in excellent research sponsored by the Electric Power Research Institute (Galloway, *et al.* 1980b).

The chemical and physical consequences of lake acidification include increased leaching of calcium from terrestrial soils (Gordon and Gorham 1963), mobilization of heavy metals such as aluminum, zinc, and manganese (Drabløs and Tollan 1980), and an increase in the transparency of the lake water (Schindler 1980). The biological consequences of such acidification include marked changes in communities of aquatic plants

and animals, with a progressive lessening of their diversity (Drabløs and Tollan 1980). In a few extreme cases, a lessening of primary productivity is observed, but in many cases there is merely a change toward lesser pelagic and greater benthic production. Whether or not primary plant production decreases, there is a progressive elimination of sensitive species of plants and animals, including fish—which are completely exterminated under severe acidification. The European roach and American smallmouth bass and walleye are extremely sensitive. Eels and perch are relatively resistant. Aluminum leached by acid rain from sensitive soils is strongly implicated in the effects on fish, chiefly through damage to gill epithelium (Muniz and Leivestad 1980). Other effects include inhibition of spawning and hatching, embryonic malformation, upset of salt balance in the blood, and growth retardation (Drabløs and Tollan 1980). Where there is a sudden acidification after snow melt, severe fish kills can result.

It should be pointed out again here that naturally acid lakes are also known, the result of drainage from adjacent, strongly acid peat bogs. Such lakes are tea-colored owing to organic matter dissolved from the peaty drainage basin. Even lakes such as these can be further acidified by acid rain, as shown by our experience at Wawa (Gordon and Gorham 1963).

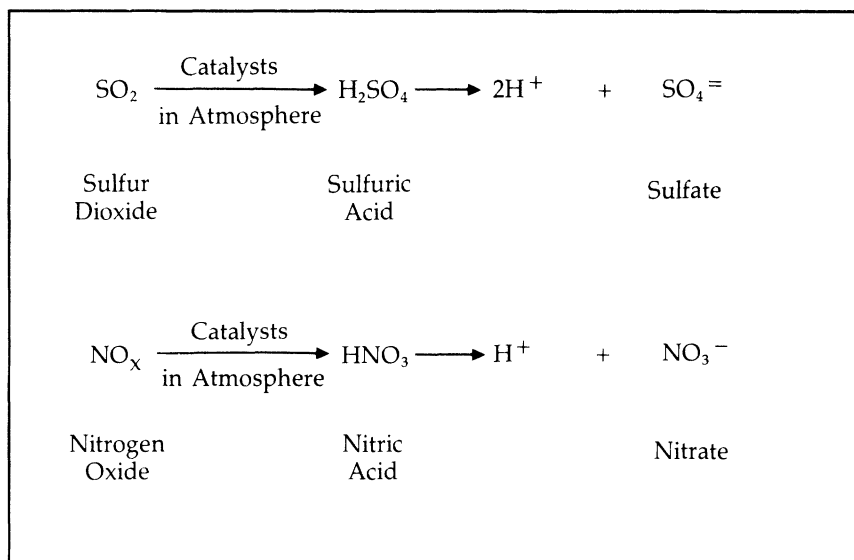
Is Acid Rain Getting Worse?

A problem of great current interest is whether acid rain is worsening and spreading. There is some evidence both from Europe and from eastern North America that suggests it is (Likens, *et al.* 1979), but the evidence is not entirely clear (Hanson and Hidy 1982). Although some criticisms have been disputed (Likens and Butler 1981), methods have changed, sites have shifted, and we simply do not have enough satisfactory trend data to resolve this problem conclusively. Nevertheless, even if acid rain is not getting worse—and we do not know at present whether this is so—it is a clear and present danger. There can be no doubt about this conclusion.

What is the Threat of Further Problems?

Many ecologists believe that over the longer term—several decades to a few centuries—acid rain may impoverish forest soils developed on sandy substrata poor in lime. Continued leaching of nutrients such as phosphorus, potassium, magnesium, and calcium from these soils may eventually result in reduced forest productivity. There is as yet no conclusive evidence for such a phenomenon. Indeed, nitrate in acid rain may actually benefit forest growth initially, because nitrogen is often the major limiting nutrient for trees on poor soil (Abrahamsen 1980). However, the

Chemical conversion of SO₂ and NO_x to acids.



deleterious effects of acid leaching may counter such a benefit, at least over the long term. Low pH may also inhibit the nitrogen cycle in forest soils, perhaps vitiating the long-term benefit of nitrogen added as nitrate in acid rain (Tamm 1976). In this context, we must remember that poor forest soils are well known to be undergoing a slow process of natural leaching and acidification that is steadily reducing their store of essential plant nutrients (Gorham, Vitousek, and Reiners 1979). Acid rain may hasten appreciably this natural process of soil impoverishment (Overrein, Seip, and Tollan 1980).

In the shorter term, recent studies of forest ecology in Germany, New England and elsewhere (Ulrich, Mayer, and Khanna 1980; Tomlinson 1981) indicate the possibility of serious forestry problems associated with the liberation of soluble aluminum by acid deposition, and its effects upon the fine tree roots in the mineral soil. Studies in New Jersey have linked a decrease in the growth rates of pines to local stream acidification; other factors such as drought, fire, pests, and atmospheric oxidants seem not to be involved (Johnson, *et al.* 1981).

Some have suggested that it is not acid rain, but changes in forestry practices and other aspects of land use, that have caused the acidification of lakes in recent decades. However, the phenomenon has been observed above treeline in Scandinavia, and in other places where forests have remained unchanged (Drabløs and Tollan 1980; Overrein, Seip, and Tollan 1980). Moreover, lakes have not acidified substantially in sensitive areas far from air pollution sources, despite major changes owing to lumbering and other human interferences.

We should remember that acid rain has effects other than the ecological ones that have been discussed. It causes severe surface damage to metals, and corrodes

limestone structures, monuments, etc. (Nriagu 1978). It can also mobilize heavy metals from pipes into supplies of potable water. Some ground waters are known to have become acidified in Sweden (Hultberg and Wenblad 1980). Moreover, the acid sulfate particles that contribute to acid rain are in the size range that penetrates deep into the lung, and they may well exacerbate lung and other diseases and increase mortality rates (Mendelsohn and Orcutt 1979; Hamilton 1980). Present epidemiological, toxicological, and clinical studies are not adequate to demonstrate conclusively that there are ill effects at ambient levels, but the problem certainly requires further attention and merits our concern (Speizer and Ferris 1978). There is also some potential for crop damage where acid rain is severe (Jacobson; Evans, *et al.* 1982).

Is it Time to Act?

There are, of course, still many things that we need to learn about the nature of acid rain, its origin, chemistry, transport, deposition, and ecological effects. In particular, we need to develop more and better dose/response data (cf. Almer, *et al.* 1978) for lake ecosystems—that is, how much damage is produced by a given degree of acid loading—of the kind developed for cultural eutrophication of lakes (Vallentyne 1974). We also need data on rates of recovery after lake acidification has been stopped. The importance of local fallout versus long-distance transport of air pollutants must also be better defined. The potential problem concerning forest productivity, about which there are so many uncertainties, could become extremely serious in the relatively near future. A major, long-term research program on this subject is urgently needed.

More immediately, the weight of all the evidence indicates that acid deposition is a serious and widespread environmental problem, caused largely by sulfur- and nitrogen-oxide pollution. Nearly all of the scientists actively studying the problem agree. The Canadian government regards acid rain as Canada's most critical environmental problem, and President Jimmy Carter's environmental message of 1979 called acid rain "one of the two most serious global environmental problems associated with fossil-fuel combustion." (The other is the "greenhouse effect" that will occur in the next 50 to 100 years as accumulating carbon dioxide, also from fossil-fuel combustion, traps heat in the atmosphere and warms the climate.) I believe—as do the governments of Canada, Norway, Sweden, and West Germany, as well as the governors of six American states, the premiers of five Canadian provinces, the Senate Committee on the Environment, and the U.S.-Canada International Joint Commission—that the evidence of damage from acid rain justifies a major attempt to reduce it. My conclusion is clear—Yes, it is time to act! Some, however, do not agree.

Industry often argues that we should wait until the evidence is much stronger, but there is ample precedent for action on environmental problems even when the chain of causation is unclear. For example, in 1952, when I was living in London, the Great Smog killed between 2,500 and 4,000 people in a week, three or more times the normal death rate. No one contests that statement, yet the chain of causation still has not been completely worked out—it was certainly far from clear at the time of the incident. Nevertheless, the authorities, acting largely upon circumstantial evidence, made great progress in cleaning up the London air. I doubt that anyone suggests they were wrong to take this very expensive action or that the regulations they issued ought to be rescinded.

Governments and industries regularly initiate far-reaching and expensive social and economic programs based on evidence not nearly as convincing as in the case of acid rain, where there is already major ecological and corrosive damage. Environmental programs should not require evidence far greater than is demanded of other initiatives, nor should the burden of proof be on the opponents of pollution. If we wait until the last scintilla of evidence has been gathered and the entire chain of causation is proved, a fragile part of life on our planet will have been damaged. Even if appropriate legislation were enacted today, it would probably take from five to ten years to affect emissions significantly.

Allowing acid rain to continue also raises questions of equity. Should one country be allowed to damage another's landscape with air pollutants, whether or not it offers to pay to clean up the damage? Acid rain violates Principle 21 of the 1972 Stockholm Declaration, endorsed by both the United States and Canada. The

declaration affirms that states have "the responsibility to ensure that activities within their jurisdiction or control do not cause damage to the environment of other states or areas beyond the limits of national jurisdiction."

What Should We Do?

Liming lakes is one temporary way to neutralize them and preserve threatened fish stocks, but it is not a long-term solution. Lime cannot readily be applied over more than a small fraction of the sensitive areas liable to undergo serious acidification. The tremendous resources necessary for a regionwide liming program would be much better directed toward controlling the problem at its source. Moreover, liming does not restore a lake to its prior state—it alters plant and animal communities. These changes would be unacceptable in preserves set aside as natural wilderness.

Instead, we should focus on the many options for reducing emissions: using coal washing, stack scrubbers, new combustion technologies, low-sulfur fuels, and alternative energy sources such as sun, wind, water, and biomass. Conservation is the most environmentally sound option, and it can be cost-effective and need not compromise human welfare.

Although acid deposition is a complicated process, I believe we can set reasonable guidelines for reducing emissions. The first question is how much we need to reduce the acidity of the rain. The National Research Council's Committee on the Atmosphere and Biosphere (1981) has suggested that the average acidity of precipitation should not exceed a pH of 4.6 to 4.7. Below that level, sensitive lakes turn acid. Because annual mean pH in the areas of the United States and Canada most affected by acid precipitation is often below 4.3—more than twice as acid as the critical level—acid deposition would have to be reduced at least 50% in those areas.

The second question is whether sulfuric or nitric acid does more harm. We know that somewhere downwind, all sulfur-dioxide emissions are eventually oxidized to sulfuric acid. Nitrogen oxides are similarly oxidized to nitric acid, but a good deal of it is consumed by plants. Thus, the amount of sulfate deposited may provide the best guide as to whether lakes will become acidified.

The third question is how much we should reduce sulfate deposition. To find the answer, we can compare the levels of deposition in areas where lakes have not been acidified with the levels that have led to various degrees of lake acidification.

Annual sulfate deposition is about 7 to 14 kilograms per hectare in northwestern Ontario and 10 to 18 kilograms per hectare in northern Minnesota. No sensitive clearwater lakes seem to have become acidified in either place. Deposition in areas harmed by acid rain

exceeds these ranges. For example, Nova Scotia receives 25 kilograms per hectare, and numerous lakes and salmon rivers have been acidified to the point of losing their fish. In the heavily polluted Adirondack Mountains, which receive 43 kilograms of sulfate per hectare, more than 200 lakes have lost their fish.

This evidence suggests that staged cutbacks to reduce annual sulfate deposition in sensitive areas to below 20 kilograms per hectare—to a little less than half the deposition in the Adirondacks—would be reasonable. Eventual further reductions to 15 kilograms per hectare would allow a margin of safety.

The fourth question is: to reduce sulfates by 50%, how much do we need to cut sulfur-dioxide pollution? I agree with the scientists at last year's Stockholm Conference on the Acidification of the Environment that on a broad geographical scale sulfate deposition is likely to be roughly proportional to sulfur dioxide emissions. Thus, an appropriate goal for the present is to reduce emissions by 50%.

Already last year the Canadian government proposed to reduce sulfur-dioxide emissions by 50% over the next decade if the United States does the same, and the West German government is already planning to reduce emissions by almost 50%. The response is up to the United States. Surely a clean environment is within our reach if we have the political will to attain it.

References

- ABRAHAMSEN, G. 1980. Acid precipitation, plant nutrients and forest growth. In Drabløs, D., and Tollan, A. (eds.) *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- ALMER, B., DICKSON, W., EKSTRÖM, C., and HÖRNSTRÖM, E. 1978. Sulfur pollution and the aquatic ecosystem. In Nriagu, J.O. (ed.) *Sulfur in the environment, Part II: Ecological impacts*. New York: John Wiley and Sons.
- BARRETT, E., and BRODIN, G. 1955. The acidity of Scandinavian precipitation. *Tellus* 7:251-257.
- BOTTINI, O. 1939. Le piogge caustiche nella regione Vesuviana. *Ann. Chim. Applic.* 29:425-433.
- BRIDGE, J.E., and FAIRCHILD, F.P. 1981. *Northeast damage report of the long range transport and deposition of air pollutants*. Northeast Regional Task Force on Atmospheric Deposition.
- BRIMBLECOMBE, P. 1977. London air pollution, 1500-1900. *Atmos. Environ.* 11:1157-1162.
- _____. 1978. Air pollution in industrializing England. *J. Air. Poll. Control Assoc.* 28:115-118.
- COHEN, J.B., and RUSTON, A.G. 1912. *Smoke*. London: Arnold.
- COWLING, E.B. 1982. An historical resume of progress in scientific and public understanding of acid precipitation and its consequences. *Environ. Sci. Technol.* 16:110A-123A.
- CROWTHER, C., and RUSTON, A.G. 1911. The nature, distribution and effects upon vegetation of atmospheric impurities in and near an industrial town. *J. Agr. Sci.* 4:25-55.
- CURTIS, C. Undated. *Before the rainbow: What we know about acid rain*. Decisionmakers Bookshelf (Edison Electric Institute) 9:1-102.
- DICKSON, W. 1980. Properties of acidified waters. In Drabløs, D. and Tollan, A. (eds.) *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- DILLON, P.J., YAN, N.D., SCHEIDER, W.A., and CONROY, N. 1979. Acidic lakes in Ontario, Canada: Characterization, extent, responses to base and nutrient additions. *Ergebn. Limnol.* 13:317-336.
- DRABLØS, D., and TOLLAN, A. (eds.) 1980. *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- ENGELMANN, R.J., and SEHMEL, G.A. 1976. Atmospheric-surface exchange of particulate and gaseous pollutants. U.S. ERDA Ser., CONF-740921 (1974). Springfield, Va: National Technical Information Service.
- ERIKSSON, E. 1960. The yearly circulation of chloride and sulfur in nature; meteorological, geochemical and pedological implications, Part II. *Tellus* 12:63-109.
- EVANS, L.S., HENDREY, G.R., LEWIN, K.F., and GMUR, N.F. 1982. *The effects of acidic precipitation on field crops*. Brookhaven National Laboratory Report 30692.
- FISHER, D.W. 1968. *Annual variations in chemical composition of atmospheric precipitation in eastern North Carolina and southeastern Virginia*. U.S. Geol. Survey Water-Supply Paper 1535-M, pp. 21.
- GALLOWAY, J.N., EISENREICH, S.J., and SCOTT, B.C. (eds.). 1980a. *Toxic substances in atmospheric deposition: A review and assessment*. National Atmospheric Deposition Program, NC-141.
- _____, SCHOFIELD, C.L., HENDREY, G.R., ALTWICKER, E.R., and TROUTMAN, D.E. 1980b. An analysis of lake acidification using annual budgets. In Drabløs, D. and Tollan, A. (eds.) *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- _____, and WHELPDALE, D.M. 1980. An atmospheric sulfur budget for eastern North America. *Atmos. Environ.* 14:409-417.
- _____, LIKENS, G.E., MILLER, J.M., and KEENE, W.C. 1982. The composition of precipitation in remote parts of the world. *J. Geophys. Res.* (in press).
- GORDON, A.G., and GORHAM, E. 1963. Ecological aspects of air pollution from an iron-sintering plant at Wawa, Ontario. *Can. J. Bot.* 41:1063-1078.
- GORHAM, E. 1955. On the acidity and salinity of rain. *Geochim. Cosmochim. Acta* 7:231-239.
- _____. 1957. The ionic composition of some lowland lake waters from Cheshire, England. *Limnol. Oceanogr.* 2:22-27.
- _____. 1958a. The influence and importance of daily weather conditions in the supply of chloride, sulfate and other ions to fresh waters from atmospheric precipitation. *Phil. Trans. Royal Soc. London, Series B*, 247:147-178.
- _____. 1958b. Free acid in British soils. *Nature* 181:152-154.
- _____. 1958c. Atmospheric pollution by hydrochloric acid. *Quart. J. Royal Meteorol. Soc.* 84:274-276.
- _____. 1976. Acid precipitation and its influence upon aquatic ecosystems—An Overview. *Water Air Soil Poll.* 6:457-481.
- _____. 1981. Scientific understanding of atmosphere-biosphere interactions: A historical overview. In National Academy of Sciences *Atmosphere-biosphere interactions: Toward a better understanding of the ecological consequences of fossil fuel combustion*. Washington, D.C.: National Academy Press.
- _____, and GORDON, A.G. 1960. The influence of smelter fumes upon the chemical composition of lake

- waters near Sudbury, Ontario, and upon the surrounding vegetation. *Can. J. Bot.* 38:477-487.
- _____, VITOUSEK, P.M., and REINERS, W.A. 1979. The regulation of chemical budgets over the course of terrestrial ecosystem succession. *Ann. Rev. Ecol. Syst.* 10:53-84.
- HAMILTON, L.D. 1980. Health effects of acid precipitation. In *Proceedings, Action Seminar on Acid Precipitation, Toronto, 1979*. A.S.A.P. Organizing Committee.
- HANSEN, D.A., and HIDY, G.M. 1982. Review of questions regarding rain acidity data. *Atmos. Envir.* 16:2107-2126.
- HITCHCOCK, D., SPILLER, L.L., and WILSON, W.E. 1980. Sulfuric acid aerosols and HCl release in coastal atmospheres: Evidence of rapid formation of sulfuric acid particulates. *Atmos. Envir.* 14:165-182.
- HOUGHTON, H. 1955. On the chemical composition of fog and cloud water. *J. Meteorol.* 12:355-357.
- HULTBERG, H., and WENBLAD, A. 1980. Acid groundwater in southwestern Sweden. In Drabløs, D. and Tollan, A. (eds.) *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- HUTCHINSON, T.C., GIZYN, W., HAVAS, M., and ZOBENS, V. 1979. Effect of long-term lignite burns on Arctic ecosystems at the Smoking Hills, N.W.T. In Hemphill, D.D. (ed.) *Trace substances in environmental health, XII*. Columbia: U. of Missouri Press.
- JACOBSON, J. 1980. Experimental studies on the phytotoxicity of acid precipitation: The United States experience. In Hutchinson, T.C. and Havas, M. (eds.) *Effects of acid precipitation on terrestrial ecosystem*. New York: Plenum.
- JOHNSON, A.H., SICCAMI, T.G., WANG, D., TURNER, R.S., and BARRINGER, T.H. 1981. Recent changes in patterns of tree growth in the New Jersey pinelands: A possible effect of acid rain. *J. Envir. Quality* 10:427-430.
- JUNGE, C.E. 1958. The distribution of ammonia and nitrate in rain water over the United States. *Trans. Amer. Geophys. Union* 39:241-248.
- _____, and WERBY, R.T. 1958. The concentration of chloride, sodium, potassium, calcium and sulfate in rain water over the United States. *J. Meteorol.* 15:417-425.
- KOIDE, M., and GOLDBERG, E.D. 1971. Atmospheric sulfur and fossil fuel combustion. *J. Geophys. Res.* 76:6589-6596.
- LAU, N.C., and CHARLSON, R.J. 1977. On the discrepancy between background atmospheric ammonia gas measurements and the existence of acid sulfates as a dominant atmospheric aerosol. *Atmos. Envir.* 11:475-478.
- LIKENS, G.E. 1976. Acid precipitation. *Chem. Eng. News* 54:29-44.
- _____, BORMANN, F.H., and JOHNSON, N.M. 1972. Acid rain. *Environment* 14:33-40.
- _____, WRIGHT, R.F., GALLOWAY, J.N., and BUTLER, T.J. 1979. Acid rain. *Scientific American* 241:43-51.
- _____, and BUTLER, T.J. 1981. Recent acidification of precipitation in North America. *Atmos. Envir.* 15:1103-1109.
- LILJESTRAND, H.M., and MORGAN, J.J. 1978. Chemical composition of acid precipitation in Pasadena, California. *Envir. Sci. Technol.* 12:1271-1273.
- LUNDE, G., GETHER, J., GJØS, N., and LANDE, M.B.S. 1976. *Organic micro-pollutants in precipitation in Norway*. Research Rept. FR9/76. Oslo, Aas, Norway: SNSF Project.
- MACKERETH, F.J.H. 1957. Chemical analysis in ecology illustrated from Lake District tarns and lakes. 1. Chemical analysis. *Proc. Linn. Soc. London* 167:159-164.
- MC COLL, J.G., and BUSH, D.S. 1978. Precipitation and throughfall chemistry in the San Francisco area. *J. Envir. Qual.* 7:352-357.
- MENDELSON, R., and ORCUTT, G. 1979. An empirical analysis of air pollution dose-response curves. *J. Envir. Econ. and Mgmt.* 6:85-106.
- MUNGER, J.W. 1982. Chemistry of atmospheric precipitation in the northcentral United States: Influence of sulfate, nitrate, ammonia and calcareous soil particulates. *Atmos. Envir.* 16:1633-1645.
- MUNIZ, I.P., and LEIVESTAD, H. 1980. Toxic effects of aluminum on the brown trout, *Salmo trutta L.* In Drabløs, D. and Tollan, A. (eds.) *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- NATIONAL RESEARCH COUNCIL. 1981. Atmospheric transport, transformation, and deposition processes. In National Academy of Sciences *Atmosphere-biosphere interactions: Toward a better understanding of the ecological consequences of fossil fuel combustion*. Washington, D.C.: National Academy Press.
- _____. 1979. *Ammonia*. Washington, D.C.: National Academy of Sciences.
- NRIAGU, J.O. 1978. Deteriorative effects of sulfur pollution on materials. In Nriagu, J.O. (ed.) *Sulfur in the environment II. Ecological impacts*. New York: John Wiley and Sons.
- ODÉN, S. 1968. The acidification of air and precipitation, and its consequences in the natural environment. *Ecol. Comm. Bull.* No. 1, Nat. Sci. Research Council, Stockholm. Arlington, VA: Translation Consultants, TR-1172.
- OVERREIN, L., SEIP, H.M., and TOLLAN, A. 1980. *Acid precipitation—Effects on forest and fish*. Oslo, Aas, Norway: SNSF Project.
- PARKER, A. 1955. *Report on the investigation of atmospheric pollution*. Dept. Sci. & Industr. Research (U.K.), Report No. 27.
- REUSS, J.O. 1975. *Chemical/biological relationships relevant to ecological effects of acid rainfall*. EPA-660/3-75-032.
- ROBINSON, E., and ROBBINS, R.C. 1970. Gaseous nitrogen compound pollutants from urban and natural sources. *Jour. Air Poll. Control Assoc.* 20:303-306.
- _____, HUSAR, R.B., and GALLOWAY, J.N. 1978. In National Academy of Sciences, *Sulfur oxides*. Washington, D.C.
- SCHINDLER, D.W. 1980. Experimental acidification of a whole lake: A text of the oligotrophication process. In Drabløs, D. and Tollan, A. (eds.) *Ecological impact of acid precipitation*. Oslo, Aas, Norway: SNSF Project.
- SHANNON, D.G., and FINE, L.O. 1974. Cation solubilities of lignite fly ashes. *Envir. Sci. Tech.* 8:1026-1028.
- SMITH, R.A. 1852. On the air and rain of Manchester. *Mem. Manchester Lit. Phil. Soc., Series 2*, 10:207-217.
- _____. 1872. *Air and Rain*. London: Longmans Green.
- SPEIZER, F.E., and FERRIS, Jr., B.G. 1978. Epidemiologic studies of health effects of sulfur oxides. In National Academy of Sciences *Sulfur oxides*. Washington, D.C.
- TAMM, C.O. 1976. Acid precipitation: Biological effects in soil and on forest vegetation. *Ambio* 5:235-238.
- TOMLINSON, G.H. 1981. *Die-back of forests—Continuing observations*. Research Memorandum, Project No. 74-7124-13, Rept. #2. Senneville, Quebec: Domtar Research Centre.
- ULRICH, B., MAYER, R., and KHANNA, P.K. 1980. Chemical changes due to acid precipitation in a loess-derived soil in central Europe. *Soil Sci.* 130:193-199.
- UNDERWOOD, J.K. 1979. pH of precipitation and annual deposition of excess sulfate, acidity and nitrate on Nova Scotia. In O'Neill, A.D.J. (ed.) *Proc. workshop on long range transport of air pollution and its impact on the Maritime Provinces*. Bedford, Nova Scotia: Atmospheric Environment Service.
- VALLENTYNE, J.R. 1974. *The algal bowl: Lakes and man*. Ottawa: Information Canada.