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PERSPECTIVE

Ambio's legacy on monitoring, impact, and management of acid rain

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Abstract Early studies published in *Ambio* showed large-scale acidification of lakes in southern Sweden and Norway from acid rain. These studies were important for delimiting various scientific issues and thus for eventually contributing to legislation, which reduced emissions of sulfur dioxide and nitrogen oxides and helped to mitigate this major environmental problem. Long-term studies and monitoring in Sweden and Norway and at Hubbard Brook Experimental Forest in New Hampshire helped guide this legislation in Europe and in the USA.

Keywords Air and water pollution · Environmental legislation · Hubbard brook experimental forest · Norway · Sulfur and nitrogen oxides · Sweden

Acid rain, the common term for acidic deposition of wet (rain, snow, sleet, hail, fog water) and dry (gases and particles) deposition of acidic substances from the atmosphere to the Earth's surface, in combination with the associated air pollution from precursor emissions, has been a major and vexing environmental problem on several continents (e.g., Likens and Butler 2014). The major contributors to acid rain are emissions of sulfur and nitrogen oxides from the combustion of fossil fuels, primarily coal, and oil, which are converted into strong acids (sulfuric and nitric) in the atmosphere. Over time, national legislation has greatly reduced emissions of sulfur and nitrogen oxides in Europe and the USA resulting in a concurrent, marked decline in the acidity of deposition (e.g., Vuorenmaa et al. 2018; Likens et al. 2021). At Hubbard Brook Experimental Forest in the White Mountains of New Hampshire, the USA, where monitoring started in 1963, precipitation is about 80% less acid today than it was in the late 1960s–early 1970s (Likens et al. 2021). In spite of this remarkable success in reducing emissions and the acidity of deposition, major legacies of the long-term, environmental impact of acid rain remain in sensitive soils, biota, and water chemistry, (e.g., Likens et al. 1996; Holmes and Likens 2016; Marx et al. 2017; Johnson et al. 2018; Riise et al. 2018).

Two early papers published in *Ambio* on the acidification of Swedish and Norwegian lakes (Almer et al. 1974; Henriksen et al. 1988) were especially influential in drawing attention to this major, environmental problem in Scandinavia. These papers pointed out the large-scale impact of this air-borne pollution on lakes and streams, the rapid intensification of the problem in southern Sweden and Norway, and the probable origin of the acidifying agents, sulfur dioxide and nitrogen oxides, from anthropogenic sources located in industrial regions mostly outside Scandinavia. At that time, both the existence and the possible effects of acid rain were being hotly debated in Europe and North America (e.g., Bolin 1972; Likens et al. 1979; Likens 1989a, b, 2010).

The authors of these two papers drew on data from large-scale monitoring efforts, on detailed chemical analysis and theory and on biological impact, particularly on fish, to make the case about the emergence, origin and importance of this widespread environmental problem.

These two papers, and the studies they described, had several important features in common: large number of lakes examined ($\sim$ 400 in Sweden and $\sim$ 1000 in Norway), study areas remote from major sources of pollution, study areas located on sensitive (poor buffering capacity) soils and bedrock, lakes important for recreation and fisheries, and use of large-scale maps to show affected areas. If the lakes had been well buffered chemically, then the changes in chemistry due to acid rain inputs probably would not have been apparent or noted in a timely fashion.

At the same time in the USA and Canada, numerous studies were underway on the existence, increase in intensity of acidity and spread of the problem in eastern North America (e.g., Beamish and Harvey 1972; Likens et al. 1972; Likens and Bormann 1974; Likens 2010; Holmes and Likens 2016; Likens et al. 2021), and about the ecological impacts, particularly on fish and the role of dissolved, toxic aluminum, which had been mobilized in these acidified aquatic systems (e.g., Beamish and Harvey 1972; Schofield 1976; Likens and Butler 2018). Schofield's early research, also published in *Ambio*, on the toxic effects of dissolved aluminum on fish in lakes in northern New York State was especially important (Schofield 1976).

The visualization provided by maps, which showed the extent and temporal increases of precipitation acidity, as well as the changes in acidity of receiving systems such as lakes, was a powerful tool in communicating the message to the public and to policy makers about this controversial issue (Likens et al. 2021).

There were formidable deniers of acid rain in the 1970s and 1980s in both Europe and in North America. In fact, the 1980s was a period that I called the "Acid Rain Wars" as the Reagan Administration, in power at that time in the USA, seemed determined to find ways to deny the existence and impact of acid rain on natural ecosystems (Likens 2010). The papers by Almer et al. (1974) and Henriksen et al. (1988) were critically important in making the scientific case that acid rain was causing serious environmental damage to lakes and rivers in southern Sweden and Norway.

The Almer et al. (1974) paper reported on chemical and biological data from 383 lakes surveyed in 1970–1972 in the lake-rich, base-poor region of southwestern Sweden. They found about 50% of the lakes examined had a pH < 6 and 36% of the lakes examined were at pH < 5 during the autumn of 1970. Acidification had accelerated during the previous two or three decades, and several fish species had been extirpated from the acidified lakes. Fish loss also had been reported for acidified lakes in southern Norway during the previous 20–30 years (Jensen and Snekvik 1972). Negative effects on phytoplankton, zooplankton, and benthos were also observed in the Swedish lakes. Lakes sampled were chosen to be representative of the area to enable extrapolations. In one lake (Lake Stora Skarsjön) where data were available, pH had decreased from 6.25 to 4.5 and transparency had increased more than 7 m from 1958 to 1973.

The Henriksen et al. (1988) paper capitalized on the "Thousand Lake Survey" done in Norway in 1986 to identify changes that had occurred in the chemistry of lakes and rivers following large-scale sampling done in 1974–1975. They found that although the pH of lakes in southern Norway (40% $<$ pH 5.0 and 60% < 5.5) had changed little during the decade between surveys, sulfate concentrations in southern and eastern Norwegian lakes were significantly lower, in concert with reduced emissions of sulfur dioxide to the atmosphere affecting this area. Nitrate concentrations in lakes of the Sørlandet area doubled during this time and aluminum concentrations increased as well. At the time of the paper, sulfate emissions were predicted to decline by 30% until 1993. The authors predicted that this decline would result in a decrease in the fraction of lakes remaining acidified and void of bicarbonate buffering to 55%, from 70% in 1986. Recent studies of some of the lakes in the area through 2012 showed a continued decrease in sulfate concentrations (Riise et al. 2018), and the contribution of alkalinity to total anions increased from 10% in 1983 to 40% in 2019 (Gunnhild Riise pers. comm.). Hence, the continued mitigation of acid rain in the area has been successful for the chemical recovery of these lakes. Not only has this change been an important shift in a major state variable in these lakes, but these projections provided important management outcomes and targets.

High-quality large-scale and long-term monitoring and study are required to identify and quantify change in highly variable natural ecosystems (e.g., Lindenmayer and Likens 2018; Likens et al. 2021). Long-term ($>$ 10 years) monitoring and study can provide important, if not unique, insights about ecosystem change (Lindenmayer and Likens 2018) and have been invaluable to understanding of eutrophication (1960s), acid rain (1970–1990s), and climate change (1990s–present) in inland waters (Fölster et al. 2014). Arguably, such studies have "contributed disproportionally to ecology and policy" (Likens 2010; Hughes et al. 2017). This was the case for the Almer et al. (1974) and Henriksen et al. (1988) papers.

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