

Decline of Acid Rain Enhances Mercury Concentrations in Fish

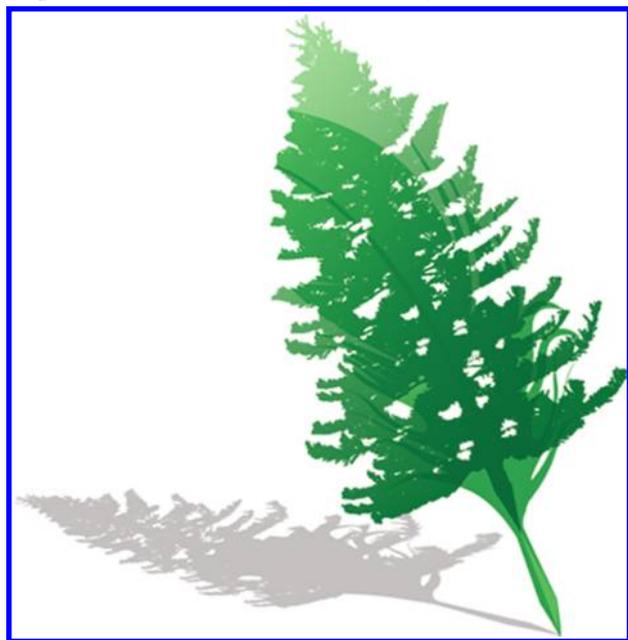
Dag Hongve,[†] Ståle Haaland,^{‡,§,*} Gunnhild Riise,[§] Inggard Blakar,[§] and Stephen Norton^{||}

[†]Norwegian Institute of Public Health

[‡]Bioforsk - Norwegian Institute for Agricultural and Environmental Research

[§]Department of Plant and Environmental Sciences, Norwegian University of Life Sciences

^{||}Department of Earth Sciences, University of Maine



Major efforts have been made to reduce sulfur emissions in Europe. Acid rain pollution in Norway has diminished substantially, thanks to the 1979 Geneva Convention on Long-range Transboundary Air Pollution, which was implemented starting in 1983.¹ Parallel with the reduction in acid rain, atmospheric concentrations and deposition of mercury (Hg) have also been reduced.² Here, we show that reduced acid rain may have increased dissolved organic carbon in runoff, and enhanced transport of mercury from catchments, and increased in-lake Hg methylation. Together, these enhanced processes result in higher concentrations of methylmercury in fish.

Mercury pollution in Norway mainly originates from distant sources in other European countries.² Similar to acid rain, the long-range atmospheric transport of Hg has declined significantly during the past decades.² It was therefore unexpected when a recent nationwide survey showed strong increased Hg concentrations in brown trout (*Salmo trutta*) and European perch (*Perca fluviatilis*) in a representative set of lakes in southern Norway.³ We selected two of these lakes for further case studies based on the availability of historic lake chemistry data. The selection includes the lake with the highest Hg-increase in perch as well as one with a slight decline.

The greatest health threat to human consumers comes from methyl-mercury (MeHg), a potent neurotoxin which is easily incorporated into living tissues. The methylation process is a key to the understanding of the potential danger posed by

environmental Hg. Environmental Hg is methylated predominantly by naturally occurring bacteria known as sulfate-reducing bacteria.⁴ The process occurs in saturated soils, anoxic lake waters, and in anoxic sediments, although the factors that control the methylation process are not fully understood.⁴ In October 2010 we sampled and analyzed modern sediments from two lakes in Norway (Figure 1).

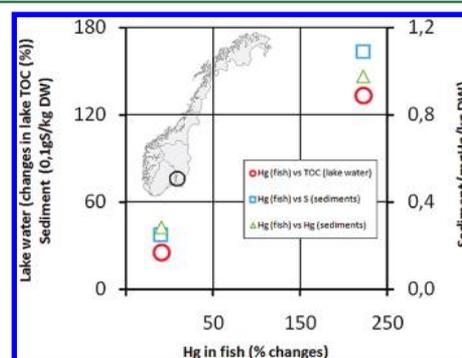


Figure 1. Change in Hg content in fish (1991 versus 2008) in two Norwegian lakes, plotted against percentage change in lake water TOC (1986–1990 versus 2003–2005), and against S and Hg in sediments (g S/kg DW and mg Hg/kg DW) (0–2 cm depth). Map shows location of the lakes.

The values of Hg in perch populations are shown in relation to recent sediment chemistry and corresponding historic lake chemistry data, obtained from the Norwegian Climate and Pollution Agency. The lake with the highest percent increase of Hg in fish also had a high percentage increase in total organic carbon, TOC (from 4 to 9 mg/L), in the lake water and high concentrations of sulfur (probably originating from acid rain) in the lake sediment (Figure 1). The lake with no percentage increase in Hg in fish had in comparison less change of TOC (from 12 to 15 mg/L).

Water color and concentrations of TOC, have increased significantly during the past decades in many lakes in Northern Europe. The TOC, originating largely from catchment soils, has increased particularly in formerly acidified lakes.⁵ The main factor governing the solubilization of soil humus is now believed to be decreased ionic strength from declining concentrations of hydrogen ions, aluminum ions, sulfate, and base cations.⁵ Lower ionic strength soil solutions increase export of TOC

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from organic soils to rivers and lakes and reducing flocculation and precipitation of organic carbon in lakes.⁵ Natural organic matter in water is a well-known complexer of metals, including Hg; solubilization of soil humus aggregates can mobilize adsorbed inorganic Hg from earlier long-term pollution. In addition sulfate, accumulated in soil and lake sediment for years during the acid rain period, can act as a terminal electron acceptor for microbial Hg methylation in the saturated soil and lake sediments. The decreased acid rain and the subsequent higher TOC concentrations in lake waters therefore seem likely to boost in-lake methylation, parallel to enhanced supply of Hg and MeHg from the catchments.

Our viewpoint is that declining sulfate in precipitation, due to the reduced sulfur emissions in Europe, may have a widespread impact on Hg concentrations in lake water ecosystems, including the Hg concentration in fish.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: staale.haaland@bioforsk.no.

Notes

The authors declare no competing financial interest.

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■ NOTE ADDED AFTER ASAP PUBLICATION

This paper published February 13, 2012 with an error in the second and third paragraphs, the caption of Figure 1, and an incorrect version of Figure 1. The correct version published February 16, 2012.