Retrieval of atmospheric trace elements concentrations using lake sediments for the past about 120 years: Impact of pollution control regulations.

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Anthropogenic emissions since the industrial revolution have resulted in enhanced accumulation of trace elements in natural ecosystems. Many of these trace elements are notable for their toxicity to humans (e.g. As, Cd, Cu, Zn, Pb), and other biological life. Anthropogenic loadings of several of these elements in the atmosphere have greatly exceeded their natural inputs. It has long been established that long-range atmospheric transport followed by wet and dry deposition is responsible for their accumulation in remote and pristine environmental sinks such as oceans, lakes and glaciers. When properly corrected for regional geochemical backgrounds and post-depositional redistribution processes, vertical variations in sedimentary trace element loads can be used to reconstruct their atmospheric deposition histories on a regional scale. Lake sediments can serve as valuable archives of historical deposition trends for a particular region and help in understanding their regional emission sources. Such historical data will be of considerable value in (1) relating historical anthropogenic emission inventories with the observed concentrations in sediments, (2) assessing the impact of pollution regulations on atmospheric deposition of toxic and non-metals in lacustrine sediments, and (3) developing future regulatory actions to minimize air pollution.

The Adirondack Park Preserve (area: 6 million ha) in upstate New York is the largest state-level protected area in the United States and contains ~3000 lakes and ponds. A majority of these lakes are high-altitude and remote, and are suitable candidates for studying atmospheric deposition histories. Previous studies from this region have focused on trace element deposition trends and geochemistry. However, these studies are mostly constrained either by the limited deposition history investigated or by the small number of analyzed trace elements.

Here we report sedimentary records of 30 major and trace elements and elemental carbon (EC) dating back to the late- to mid-19th century from two high-altitude remote lakes (West Pine Pond and Clear Pond) in the Adirondack Mountains. Since 1978, we have maintained an aerosol sampling site at Whiteface Mountain (1483 m a.s.l) in the Adirondacks. A number of our studies over a period of about four decades have shown that atmospheric SO_4^{2-}, trace elements and EC in the Adirondacks are transported from emission sources located upwind in the Northeastern and Midwestern US (e.g., Husain et al., 2004. 2008]. Therefore, in addition to reconstructing the trace element depositional history of the Adirondack region dating back to the preindustrial era, we also attempt in this paper to rationalize historical trace elements and EC trends with records of fuel use and emissions in the industrialized Midwest and the US as a whole.

Sediment cores were collected using a gravity corer with a diameter of 5cm. Cores were sliced in the field, freeze dried in our lab, age of each slice determined using ^{210}Pb dating technique, and sediment digested, and concentrations of trace elements determined using ICPMS, and EC by the thermal optical method (Sarkar et al., 2015).

Lithophilic elements showed no systematic temporal pattern, or any significant enrichment over their crustal abundances over the entire period. Anthropogenic trace elements exhibited distinct increases beginning ~1900, and peaked around 1920-1970, due apparently to energy-related emissions. Atmospheric Pb fluxes were reflective of historical smelter production and combustion of coal and leaded gasoline. Copper and zinc fluxes mimicked corresponding primary production while EC fluxes followed the long-term trend for fossil and biofuel combustion. Trace elements and EC flux trends were closely related to the growth of industrialization in the Central and Midwestern US and changing fuel consumption patterns. Peak concentrations of anthropogenic elements, except Pb and Hg, were observed at ~1921 in West Pine Pond and ~1940s in Clear Pond. Concentration of Pb peaked in 1973 in both lakes, and Hg only in Clear Pond at ~1965. Compared to the peak values, modern trace element fluxes decreased by 25-85%, whereas EC decreased by 96%. Apparently, the regulations intended to control pollutant emissions have succeeded in reducing atmospheric concentration of the species studied and have improved air quality.

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