Atmospheric pollution source attribution using coupled Hg, Pb, and Sr isotopes in preciptation

LAURA S. SHERMAN[1]*, JOEL D. BLUM[1], J. TIMOTHY DVONCH[1] AND LYNNE E. GRATZ[2]

[1]University of Michigan, Ann Arbor, MI
[2]University of Washington – Bothell, Bothell, WA
*correspondence: lsaylors@umich.edu

Atmospheric deposition of metals such as Hg and Pb is of concern to human and environmental health. These pollutants are emitted from local, regional, and global sources. Previous studies have used methods including trace element ratios, atmospheric transport modeling, and multivariate statistical receptor modeling of trace element concentrations to quantify source contributions to wet deposition. Atmospheric emissions from sources such as coal-fired power plants, metal smelters, and incinerators can also be isotopically distinct. Recently, Pb isotopes measured in daily precipitation samples were used in conjunction with trace element ratios to identify source impacts at an urban site. Similarly, Hg isotope ratios have been used to trace Hg emitted by a coal-fired power plant to local precipitation.

We measured Pb, Hg and Sr isotope ratios in 58 previously collected daily precipitation samples to develop a new method for linking emission sources with deposition sites across the Great Lakes Region. Precipitation samples were collected between 2003 and 2006 at seven sites in Michigan, Ohio, and Vermont. We observed significant variablity between sites and among individual samples in all three isotope systems. $^{208}Pb/^{206}Pb$ ratios ranged from 1.9591 to 2.0937, $^{87}Sr/^{86}Sr$ ratios ranged from 0.708591 to 0.711553, and $\delta^{202}Hg$ values ranged from -1.13 to 0.13‰. These new results corroborate previous source-receptor and atmospheric transport modeling. For example, we measured low $\delta^{202}Hg$ values (similar to those measured in soils near smelters) and low $^{208}Pb/^{206}Pb$ ratios (similar to those of North American Pb ores) in samples impacted by iron/steel production. We also found that $^{87}Sr/^{86}Sr$ ratios can be used to distinguish between sites in Michigan and southern Ohio, which we attribute to variations in locally-derived dust from different soil parent materials as well as variations in locally-emitted fly ash.

This novel multi-isotopic source attribution technique provides a new tool for assessing deposition of metal contaminants. Ultimately, the development of distinct source isotopic profiles will assist in the accurate quantification of local, regional, and global source impacts.