Mercury Transformations on Land and Water (METALLICUS)

David P. Krabbenhoft, USGS
John W. Goodrich-Mahoney, EPRI

And

Relating Mercury Deposition and Transformation to Methylmercury Levels in Fish

John W. Goodrich-Mahoney, EPRI

Abstract

The relationship between mercury deposition, methylmercury (MeHg) production and its presence in fish, which through consumption is the primary vector to humans, is of great interested to society. Over the past 50-60 years, we have learned that MeHg exposure to humans can result in a range of human impacts, from the lethal exposures at Minamata, Japan, to the more common condition of subtle neurological effects, especially in children exposed in utero.

In order to better understand this relationship and what, therefore, might happen to fish mercury concentrations when there is a change in atmospheric mercury deposition levels, a whole-ecosystem experiment is underway in Canada, the Mercury Experiment to Assess Atmospheric Loading In Canada and the United States (METAALICUS), as well as several regional studies. With the advent of new analytical techniques, it is now possible to measure mercury isotopes in the environment at very low levels. METAALICUS involves the application of three non-radioactive (stable) inorganic mercury isotopes to an entire watershed that contains an 8.3 ha lake in Canada at the Experimental Lakes Area (ELA). The isotopes are being added to the upland, wetland, and lake surface (^{200}Hg, ^{198}Hg, and ^{202}Hg, respectively). Isotopic mercury concentrations are being tracked in all major compartments in the lake, watershed and atmosphere. Detailed process studies are also being carried out to follow the movement and transformation of the isotopic mercury through the watershed and lake, as well as air/surface exchange of isotopic mercury. Production of MeHg is being studied in the lake sediments, upland and wetland, as is the bioaccumulation of MeHg into benthic and pelagic invertebrates and fish. Initial results indicate a rapid response in the lake, with the isotopic mercury delivered to sites of methylation, methylated and bioaccumulated within one month. The isotopic mercury is also more dynamic (i.e., available to methylating agents) then the existing mercury pools ("old" mercury). The response of the lake is exceeding faster (within a field season) than the upland response, which appears to be several years.

Similar results were found in isotopic mercury spiking mesocosm experiments in the Everglades, which are part of the Aquatic Cycling of mercury in the

Everglades (ACME) project. In that study, consecutive doses of different stable isotopes of mercury into the same mesocosms over periods of time up to six months has shown that the more recently a mercury dose enters an ecosystem, the more likely it is to be observed in food webs. In addition, the ACME project is performing multiple additions of other constituents (e.g., sulfate and dissolved organic carbon) known to affect methylation rates. In these studies, it has been observed that: (1) sulfate stimulates mercury methylation to a certain level, and beyond which it becomes inhibitory; (2) dissolved organic carbon elevated levels of mercury methylation of both “new” and “old” mercury; and (3) when mercury is added in combination with sulfate or dissolved organic carbon, a much larger response, in terms of additional MeHg production, is observed than if mercury alone is added.

Ecosystems, which have freshwater wetlands, reservoirs, and/or are affected by acid deposition, seem to be prone to higher MeHg production. Sulfate-reducing bacteria are known to be the proximal methylating agents in the environment and understanding the processes and factors that control the availability of mercury to these microbial communities (e.g., formation of neutral HgS^0 that crosses the cell) is important. The formation and cycling of MeHg is an extremely vexing question that intersects environmental chemistry, microbiology, meteorology, and biology, and as such a broad view is necessary to unravel its complexities. For example, reducing $S0_4$ and H^+, along with mercury in precipitation, maybe beneficial for those systems that received most or all of their water from precipitation. Other systems maybe buffered by the pool of “old” mercury in soils and sediments, and from global mercury sources, significantly reducing response times to changes in atmospheric mercury loading rates.