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Mercury stable isotope biogeochemistry

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Methods for high precision measurement of natural Hg isotope ratios by multiple collector inductively coupled plasma mass spectrometry (MC-ICP-MS) have been developed recently by our group and allow the use of Hg isotopes to trace the biogeochemistry of Hg. Mercury has seven stable isotopes ranging from 196 to 204 amu. We have found that the isotopic composition of Hg varies by both mass dependent fractionation (MDF) and mass independent fractionation (MIF). Even and odd isotopes of Hg are fractionated by mass-dependent processes, whereas odd isotopes are also fractionated in a mass-independent way by photochemical reactions. Isotope ratios are measured relative to the NIST 3133 Hg standard reference material. MDF is reported as δ^{202}Hg ($\pm$0.08‰, 2SD) which is the difference in ^{202}Hg/^{198}Hg between a sample and NIST 3133 in permil (‰). MIF is reported as Δ^{201}Hg ($\pm$0.05‰, 2SD), which is the difference in ^{201}Hg/^{198}Hg ratio in permil from what the ^{201}Hg/^{198}Hg ratio would be if the fractionation were entirely mass dependent.

In this presentation we summarize the range of Hg isotopic compositions of a variety of environmental and geological materials. In previous work we described biotic and abiotic laboratory fractionation experiments demonstrating the range of mass dependent and mass independent fractionation caused by mercury redox transformations in the surface environment. Thus far we have found that MIF occurs during photochemical reduction of methyl-Hg and Hg^{2+} following a Rayleigh-like fractionation. Bacterial reduction causes Rayleigh-like MDF but no MIF. Coastal-marine and freshwater fish from North America have positive Δ^{201}Hg values (0.2 to $>$3‰) reflecting transfer of methyl-Hg into the food web after partial reduction by photochemical re-

actions. Most coals and the organic horizons of soils from North America have negative Δ^{201}Hg values (-0.1 to -0.4‰), possibly reflecting the influence of Hg that was photochemically reduced in aquatic systems, transported in the atmosphere and redeposited to organic matter. Igneous and sedimentary rocks, marine sediments, and Hg ore deposits generally do not display large MIF (Δ^{201}Hg $\leq \pm$0.05‰).

We have measured a range in δ^{202}Hg of over 3‰ in natural samples. Pre-anthropogenic Mediterranean marine sediments have δ^{202}Hg of -0.8‰, and a sample of contaminated sediment from a New Jersey coastal waterway has δ^{202}Hg of -0.2‰. Samples of Hg ore from New Idria, California and Almadén, Spain have δ^{202}Hg of -0.3‰ and -0.5‰, and overlying hot spring deposits contain Hg with δ^{202}Hg of -3.5 to 0.4‰. Sedimentary and igneous rocks from the Franciscan Complex, Great Valley Sequence and Clear Lake Volcanics in California have δ^{202}Hg of -0.3 to -1.0‰. Samples of coals and the organic horizons of soils from North America display a wide range of δ^{202}Hg values from -0.8 to -2.4‰. Finally, coastal-marine and freshwater fish from North America range in δ^{202}Hg values from 1.2 to -1.6‰, reflecting a variety of Hg sources to aquatic systems and possibly some MDF within food webs. Preliminary studies indicate that Hg stable isotopes show great promise in elucidating several aspects of the global Hg biogeochemical cycle and in differentiating sources of Hg pollution to the environment.