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Assessing the deposition of atmospheric mercury in Hawaiian soils from the Big Island using Hg stable isotopes

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Mercury (Hg) is a toxic metal that undergoes reductive-oxidation cycling and exists in gaseous, aqueous, and solid forms in the environment. Mercury is emitted from both anthropogenic (e.g., mining) and natural sources (e.g., volcanism) and can be transported far from sources via atmospheric circulation due to its ~1 year residence time. One of the largest reservoirs/sinks of Hg are forest biomass and soils, which dominantly get their Hg from plant uptake of atmospheric Hg followed by deposition to soils via litterfall. Because Hg isotopes undergo mass-dependent fractionation (MDF) and several types of mass-independent fractionation (MIF), which are mostly produced during photochemical processes, stable isotopic compositions of Hg are useful in understanding the biogeochemical cycling of Hg. In particular, Hg isotopes are very useful in elucidating and quantifying pathways of deposition and transformation in soils and forests and have shown how important the plant uptake pathway is for delivering Hg to soils and effecting concentrations of atmospheric Hg.

In this study, we analyzed Hg concentrations and isotopic compositions in Hawaiian soils from the leeward side of Kohala Mountain. In addition, we also measured Hg isotopes in atmospheric gaseous elemental Hg (GEM) at several sites on the Big Island with a few samples from near the rim of Kilauea while it was actively erupting. Overall, Hg concentrations were elevated in the soil samples (560 ppb to 4300 ppb) with an average concentration of 1480 ppb, with higher concentrations at forested versus pasture sites. Soil Hg all had negative MDF consistent with other soils, but the odd isotope MIF was quite negative compared with other soils (up to -0.9 permil). The high concentrations and very negative MIF suggest that the soils may receive more atmospheric Hg deposition that undergoes significant photochemical reduction prior to incorporation into the soils. Atmospheric GEM samples at most sites on the Big Island had concentrations and isotopic compositions consistent with global background. However, near the rim of Kilauea concentrations were slightly elevated and isotopes were isotopically heavier with less MIF, which is consistent with the shift expected if there was significant influence from volcanic Hg.