Mercury isotope compositions in North American forest soils and litters

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Soils represent one of the largest reservoirs of mercury on Earth and play a critical role in its natural cycle by acting as both a sink and source. However, it is not well understood how soils sequestrate and remobilize Hg. Natural variations in stable isotopes of Hg are being explored as a promising tool in studying the sources and transformations of Hg. Published Hg isotopic data in soils is scarce and significantly different from those of atmospheric Hg, which is considered as one of the major sources of Hg to soils.

We are surveying the Hg isotope compositions in mineral soils, litters and fresh vegetation from twelve forest sites across the entire contiguous United States. The current results from the first four sites (Thompson forest, WA; Truckee, CA; Niwot Ridge, CO; Howland, MA) show that all samples unanimously display negative mass dependent fractionation (MDF) with δ^{202}Hg up to -2.7‰ and negative mass independent fractionation (MIF) with Δ^{199}Hg up to -0.5‰. δ^{202}Hg depth profiles show a positive correlation with total Hg and increase from foliage to top soils. In contrast, Δ^{199}Hg shows little depth variation, suggesting this MIF signal is most likely inherited from the atmosphere and is not changed by mass dependent processes in soils. We also observe significant MIF for even isotopes (Δ^{200}Hg and Δ^{204}Hg). They display a consistent linear relationship with a Δ^{200}Hg/Δ^{204}Hg ratio of -0.5, similar to the ratio observed in atmospheric precipitation[1]. Moreover, we find a positive correlation between elevation and both types of MIF (Δ^{199}Hg and Δ^{200}Hg). We suggest that the MIF of even isotopes is linked to the production and deposition of reactive gaseous mercury (RGM), which is higher in the high elevation Western US sites than in our low elevation sites. Overall, our results indicate that atmosphere is the dominant source of Hg to soils on a continental scale, but meteorological conditions and atmospheric Hg speciation affect the relative contributions of gaseous elemental mercury (GEM) and RGM depositions and shape the eventual Hg isotope compositions in soils.

[1] Demers *et al* 2013, *Global Biogeochem. Cycles*