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# Investigation of atmosphere – vegetation mercury isotopic offsets in a remote Canadian forest ecosystem

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In this study, we investigated the role of vegetation in mediating the transfer of atmospheric gaseous elemental mercury (GEM) to soils through litterfall in a Canadian boreal-deciduous mixed forest, Limberlost Forest and Wildlife Reserve (LFWR). The isotopic compositions of GEM, foliage across various plant species, mixed litterfall and soil samples from LFWR were analyzed. The Hg mass-dependent fractionation (MDF, reported as $\delta^{202}$Hg) values in foliage ($\delta^{202}$Hg = -2.16 ± 0.11 permil) were isotopically lighter relative to GEM ($\delta^{202}$Hg = -1.19 ± 0.12 permil), supporting large fractionation during foliar uptake of GEM, consistent with previous findings. Negligible even mass-independent fractionation (even-MIF, reported as $\Delta^{200}$Hg) signatures throughout all the samples ($\Delta^{200}$Hg = 0.00 ± 0.01 permil) suggest negligible contribution from atmospheric $Hg^{2+}$ species. Rather the odd mass-independent isotopic signatures (odd-MIF, reported as $\Delta^{199}$Hg) in vegetation ($\Delta^{199}$Hg = -0.31 ± 0.02 permil) compared to atmospheric GEM ($\Delta^{199}$Hg = -0.24 ± 0.06 permil)indicate GEM as the primary source of Hg to the vegetation. The small difference between GEM and foliage odd-MIF suggests some photochemical reduction and subsequent re-emission processes occurring on or within the leaves. Litterfall displayed isotopically heavier MDF values ($\delta^{202}$Hg = -1.71 ± 0.26 permil) relative to foliage, emphasizing decomposition-related processes enriching the heavier isotopes in the residual Hg pool. Surface and deeper soils also reflected odd-MIF consistent with plant derived Hg as the major source, and exhibited more positive MDF than litterfall highlighting the continued loss of lighter isotopes during decomposition within the soils. An isotopic mass balance confirmed atmospheric GEM as the predominant contributor to soil Hg. Surprisingly, the MIF and MDF offsets between GEM and foliar Hg were reasonably consistent within the LFWR ecosystem (five plant species in triplicate). Results from LFWR were also compared with GEM-plant isotopic offsets across diverse ecosystems (urban, volcanic, tropical forest - both remote, and area affected by artisanal and small-scale gold mining activities). The results revealed that significant variability is observed across different ecosystems, despite relatively consistent offsets within an ecosystem, emphasizing the need for nuanced approaches to using Hg isotopes to quantify the role of forests in Hg cycling accurately.