Unraveling processes of atmospheric Hg sequestration by soils over annual to centennial timescales

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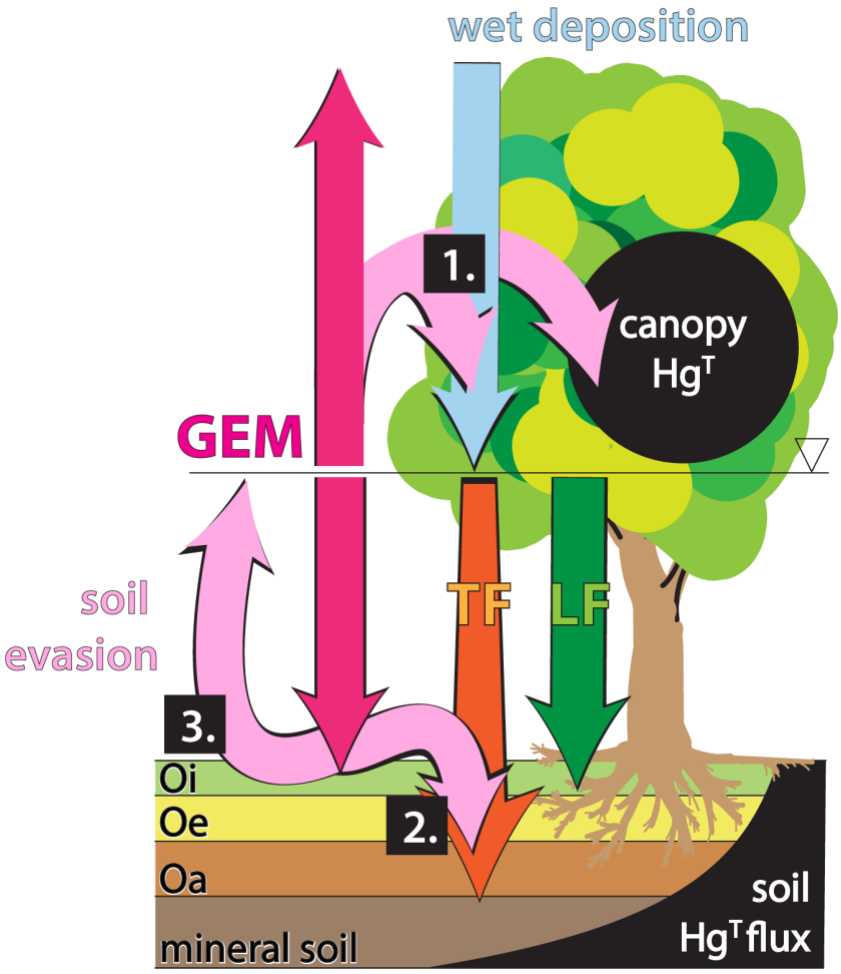
Soils are a principal reservoir of the potent neurotoxin Hg that has accumulated through the history of anthropogenic emissions to the global atmosphere, yet contemporary flux measurements between atmosphere and soil show complex, bi-directional exchange and ambiguous net flux of gaseous elemental mercury (GEM) to soil. This discrepancy reveals fundamental uncertainty regarding how Hg is incorporated into soil, while also underscoring limitations in both modeling the strength of the soil sink in global Hg cycling and understanding the sensitivity of soil Hg to future environmental change. Here we introduce a new bottom-up soil mass balance approach based on fallout radionuclide (FRN) chronometry that allows a direct and independent comparison of Hg soil accumulation rates with measured atmospheric ecosystem flux (sum of litterfall, LF, throughfall, TF, and non-foliar GEM deposition). Through this direct comparison we show that organic soil horizons are typically depleted in Hg, with accumulation rates falling 20-80% short of expected ecosystem fluxes across arctic, temperate and tropical soils. Underlying mineral soils, in contrast, accumulate Hg at rates comparable to contemporary ecosystem fluxes. Moreover, mineral soil accumulation rates increase with depth in a pattern consistent with the decadal-centennial history of atmospheric Hg emissions. We propose that while GEM is mobile in organic soils and may be re-emitted to atmosphere over diel to seasonal timescales, on balance atmospheric Hg is effectively redistributed to mineral soil where it is sequestered to break a cycle of re-emission. We propose that throughfall is likely a key process in soil mass balance, rinsing GEM deeper into soil in a manner analogous to its role in rinsing Hg from the foliar canopy, and thereby delivering GEM to soil depths beyond a surface zone of re-emission. Our ability to reconstruct soil Hg accumulation rates using radionuclide chronometers now poses opportunities to explore environmental factors that regulate both Hg emission and accumulation in soils.



Abstract Art or conceptual figure for Hg accumulation in soil. Key points include (1) rinsing of GEM from canopy by wet deposition (throughfall); (2) rinsing of GEM deeper into soil by throughfall, (3) re-emission of GEM from forest floor