



Mercury (Hg) accumulation in terrestrial carbon (C) reservoirs: magnitude, spatial patterns, fate upon C losses, and implications of global change

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Terrestrial ecosystems are strong natural reservoirs that retain the bulk of atmospheric Hg deposition. As a result, a long-term legacy of past and present Hg pollution is sequestered in surface litter and soil pools. Hg shows a particular affinity to—and hence tends to accumulate in—terrestrial organic C. We present a summary of a comprehensive five-year investigation where we quantified: (i) relationships between Hg and C across 14 forests sites to assess the affinity of Hg to C accumulation across spatial scales; (ii) the degree to which C determines net retention and spatial accumulation of Hg; (iii) the fate of Hg upon losses of C, including losses through wildfires and mineralization; (iv) the coupling of gaseous Hg losses to CO₂ respiration; and (v) the potential sensitivity of climate-change induced changes in C on terrestrial Hg sequestration.

Results show that continental-scale spatial distribution of Hg in soils and litter is strongly related to C, and that old terrestrial C pools (as determined by C/N ratios) are particularly prone to Hg enrichment. The correlation of Hg and C is likely responsible for increasing Hg levels (concentrations and pools of total Hg, as well as methylated Hg) with higher latitude, which we attribute to a legacy of Hg sequestration in C-rich layers of northern ecosystems. Experimental studies and field observations to address fate of Hg sequestered in organic C show that: (i) fires leads to up-to-complete Hg losses in either gaseous elemental or particulate-bound form; (ii) litter decomposition also leads to evasion losses of Hg in the range of 50% of initial Hg, but little Hg is subject to runoff as dissolved Hg; (iii) soils effectively retain Hg with only about 3% of Hg subject to volatilization upon C loss during respiration; (iv) no links between CO₂ and gaseous Hg concentrations are observed in soil depth profiles in the field, indicating that fate and movement of gaseous Hg is decoupled from that of CO₂. We calculate that at a global scale as much as 4.5 x 10⁶ Mg of Hg is sequestered in soil organic C pools, and propose that climate-induced changes in terrestrial C have direct implications on terrestrial Hg reservoirs. Using organic C as a matrix to quantify surface reservoir accumulation of atmospheric pollutants may also be applicable to other compounds with strong affinities to organic C, including polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAHs), and polybrominated diphenyl ethers (PBDEs).