Stable isotope fractionation induced from mercury biogeochemical cycling in forest ecosystems

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We determined the mass flow and isotopic compositions/fractionations associated with Hg cycling in the air-water-plant-soil compartments in an evergreen broadleaf forest site in Southwestern China to reveal mechanisms underlying the cycling of Hg. It is found that that annual Hg mass flow is -26.8±12.7 μg m^{-2} $year^{-1}$ for air-foliage Hg^0 flux, 6.7±20.5 μg m^{-2} $year^{-1}$ for air-soil Hg^0 flux, 9.1±1.2 μg m^{-2} $year^{-1}$ for bulk Hg deposition, 31.8± 10.6 μg m^{-2} $year^{-1}$ for throughfall Hg deposition, 0.3± 0.1 μg m^{-2} $year^{-1}$ for stemflow Hg deposition, and 1.8±0.4 μg m^{-2} $year^{-1}$ for Hg runoff. Mass balance analysis using the flux data shows a 50.4±44.2 μg m^{-2} $year^{-1}$ net atmospheric Hg sink. Interestingly, we observed the δ^{202}Hg shift of 0 to -3.1‰ between air and foliage as leaf age increases, and so for the Δ^{199}Hg shift of 0 to -0.15‰. The observed isotopic composition of foliage Hg and isotopic shift support re-emission of Hg^0 occurring after reductive loss from foliage. The δ^{202}Hg shift cause by Hg evasion from soil in summer ranges -0.92‰ to -0.23‰ with a Δ^{199}Hg shift from -0.25‰ to -0.10‰; while in winter only δ^{202}Hg shift is observed with range of -0.28‰ to 0.41‰. Given the isotopic compositions found in soil pore gas and ambient air, legacy Hg re-emission is largely caused by dark reduction processes in deep soil in summer, and by photo-reduction processes in surface soil in winter. Moreover, the distinctly negative Δ^{199}Hg and ~0 Δ^{200}Hg in organic soils indicate atmospheric Hg^0 deposition is the dominant source of Hg in soil. Finally, we observed positive Δ^{199}Hg and Δ^{200}Hg in bulk precipitation, however, the Δ^{199}Hg and Δ^{200}Hg in throughfall, stemflow and runoff exhibit distinctly negative values. This suggests a significant atmospheric Hg^0 source contribution in throughfall (60±13%), stemflow (96±25%) and runoff (89±23%). Results from mass balance modeling show that atmospheric Hg^0 deposition accounts for ~75% of total atmospheric Hg deposition, ~2 times greater than values by the estimate from air-foliage/soil flux. Overall, this study highlights the governing role of atmospheric Hg^0 deposition and re-emission processes in Hg cycling of forest ecosystems.