

Using Mercury Isotope Ratios to Understand Hg Biogeochemical Cycling in Forest Ecosystems

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Mercury (Hg) accumulation in montane forested areas plays an important role in global Hg cycling. In China, montane forests account for about 90% forested areas, 25% of which located in Southwest China. In this study, we characterized stable Hg isotopic composition at the background forest sites of Mt. Ailao, Mt. Leigong, Mt. Gongga, and 23 forests in Tibetan Plateau. We aimed to identify Hg isotope fractionation caused by the translocation of atmospheric Hg to the forest floor. We measured Hg isotope ratios of the ambient air, precipitation, throughfall, litterfall, leaf of predominant species, soil and surface water. We also investigated Hg isotope fractionation during Hg exchange between leaf and air, between soil and air, and during the litterfall decomposition process. At Mt. Ailao, the Hg isotope signature of bulk leaf sprout samples ($\delta^{202}\text{Hg} = 0.08 \pm 0.74\text{‰}$, $\Delta^{199}\text{Hg} = -0.20 \pm 0.14\text{‰}$) was similar to those of gaseous air samples ($\delta^{202}\text{Hg} = 0.37 \pm 0.44\text{‰}$, $\Delta^{199}\text{Hg} = -0.18 \pm 0.04\text{‰}$) suggesting that atmospheric Hg^0 is a major source of Hg found in foliage. Negative $\delta^{202}\text{Hg}$ -shift (3‰) and $\Delta^{199}\text{Hg}$ -shift (0.10-0.15‰) were observed during the growing season. Hg^0 effluxes from mature foliage exhibits a large, positive mass independent fractionation (MIF) signature ($\Delta^{199}\text{Hg} = 0.17 \pm 0.40\text{‰}$) in contrast to ambient air and leaf samples. During a 2-year litter decomposition period, $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$ and Hg mass varied slightly, suggesting a steady Hg accumulation in decomposing litter biomass. The negative $\Delta^{199}\text{Hg}$ in the surface soil samples were comparable to the observed $\Delta^{199}\text{Hg}$ in litter, but significantly different from the positive $\Delta^{199}\text{Hg}$ in throughfall/rainfall at 26 forest sites. This suggests that Hg input from litter is a predominant source for Hg accumulation on high montane forest floor. Statistical analysis suggests that precipitation and temperature mediated litter biomass production plays an important role in Hg accumulation in forest ecosystems. Hg isotope ratios provide new insights of Hg biogeochemical cycling in forest ecosystems.