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Isotopic investigation of atmospheric mercury sources and their influence in forest ecosystems

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Among various natural ecosystems studied, forest ecosystems act as a net sink for atmospheric mercury. While processes governing mercury integration into forests have been studied via the concentration analyses, sources and chemical species of mercury impacting forests have been difficult to constraint. We employed mercury stable isotopes to identify spatial differences in atmospheric mercury sources and their influences on five forest ecosystems across South Korea. The isotope ratios of TGM (δ^{202}Hg; -1.01 ± 0.79 ‰, Δ^{199}Hg; -0.01 ± 0.10 ‰, 1SD, n = 19) were similar to those emitted from anthropogenic activities, composed of high Hg^{2+} proportion, relative to background mercury, composed primarily of Hg^0. The TGM and foliage mercury concentrations displayed a positive relationship (r^2 = 0.67, $p < 0.05$), indicating that foliar uptake from the atmosphere is the major source to forests. Across five forests sites, the δ^{202}Hg and Δ^{199}Hg showed significant deviation between foliage and TGM (δ^{202}Hg; 1.67 ± 0.76 ‰, Δ^{199}Hg; 0.11 ± 0.23 ‰). We attribute the Δ^{199}Hg deviation to the preferential uptake of Hg^0 to foliage and the δ^{202}Hg deviation to the fractionation imparted via Hg^0 oxidation in foliar tissues. The litter samples (δ^{202}Hg; -2.32 ± 0.28 ‰, Δ^{199}Hg; -0.20 ± 0.09 ‰, 1SD, n=6) showed no significant δ^{202}Hg and Δ^{199}Hg differences compared to foliage (δ^{202}Hg; -2.50 ± 0.48 ‰, Δ^{199}Hg; -0.19 ± 0.25 ‰, 1SD, n=6) and topsoil (δ^{202}Hg; -1.82 ± 0.49 ‰, Δ^{199}Hg; -0.23 ± 0.04 ‰, 1SD, n=6), implying substantial delivery of atmospheric mercury to the forest ecosystem via foliage shedding and litter decomposition. Among two forests selected for the elevational study, we observed similar Δ^{199}Hg between the litter and topsoil with increasing elevation in Mt. Mani but not in Mt. Bihak. While mercury associated in the litter is efficiently transferred to soil via decomposition in Mt. Mani, the external influences such as anthropogenically deposited Hg_p appear to influence the topsoil of Mt. Bihak. Future studies that characterize mercury isotope ratios of various atmospheric mercury species and their cycling in the forest would improve the understanding of the fate of atmospheric mercury in diverse forest ecosystems.