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Tracing the source and transport of Hg during pedogenesis in strongly weathered tropical soil using Hg isotopes

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Soil is the largest reservoirs and re-emission sources of mercury (Hg) on Earth's surface. The accumulation and remobilization of Hg during pedogenesis significantly affect how Hg transports from terrestrial to aquatic environment and biota, but these processes are poorly understood. We present Hg concentration and isotope ratios of soil and bedrock samples from latosol profile formed through intense weathering of Cenozoic basalts in Zhanjiang, Guangdong Province, China, to trace the sources, transport and transformation processes of Hg during pedogenesis, and evaluate the potential impact of tropical soils on global Hg cycling. The profile shows significantly negative mass dependent (MDF, δ^{202}Hg = −2.97‰ to −2.54‰) and mass independent fractionation (MIF, Δ^{199}Hg = −0.68‰ to −0.43‰), with a distinct positive shift of δ^{202}Hg value from the bedrock to the partially weathered horizon (Layer C) and a general downward positive shift of Δ^{199}Hg value. These isotopic signatures suggest a dominant Hg input from atmospheric Hg(0) in the form of litterfall (Hg_{Litter}) and limited contribution (<15%) of geogenic Hg (Hg_{Geo}) from the bedrock. The Hg_{Litter} is subject to significant photoreduction on soil surfaces and the subsequent downward migration in association with soil organic matter, which are likely responsible for the strongly negative Δ^{199}Hg value throughout the latosol profile. Moreover, the positive shift of δ^{202}Hg value from bedrock to the weakly weathered rock indicates a substantial loss of Hg_{Geo} during bedrock weathering, likely due to the dissolution of primary minerals in basalt. In addition, the δ^{202}Hg value shows a strong correlation with soil pH at some horizons, attributable to the isotopic fractionation during the change of Hg speciation likely triggered the adsorption on soil surfaces, which are affected by surface charge properties and soil pH values. Overall, our results provide new insights into the source and mechanisms/processes driving Hg transport, transformation and remobilization during pedogenesis of a tropical latosol profile, and offer direct evidence for the loss of Hg_{Geo} during the weathering of bedrock, which may serve as important sources of Hg in tropical regions and have a significant impact on the Hg isotope signatures of water and biota in river/lake and marine environments.