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Mercury isotopic fractionation in latosol profile developed from strongly weathered basalt

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Weathering is a key process transferring material from continents to the hydrosphere, and plays a critical role in the cycling of trace elements. Mercury (Hg) is a toxic heavy metal and well-known global pollutant. Soil is the largest reservoir of Hg on Earth surface, and thus the behavior of Hg during soil weathering significantly affects how Hg transports from terrestrial to aquatic environment and biota. However, the transport and fate of Hg during soil weathering are rarely studied. The fractionation of Hg stable isotopes has been widely used to trace Hg cycling. Here we measured Hg concentration and isotope ratios in a latosol profile in Zhanjiang, Guangdong Province, South China, formed through intense weathering of basalt, in order to understand how Hg isotopes fractionate during weathering. All soil samples showed significantly negative δ^{202}Hg (−2.97‰ to −2.54‰) and Δ^{199}Hg (-0.68‰ to -0.43‰), which are dramatically different from those of the bedrock (δ^{202}Hg = -7.30‰, Δ^{199}Hg = -0.05‰), suggesting that throughout the profile Hg was dominantly from atmospheric deposition rather than from bedrock. The Hg enrichment and isotope compositions both show distinct patterns below and above ~300cm. There was significant leaching loss of Hg below ~300cm, but almost no variation in δ^{202}Hg. This is likely because Hg was absorbed by Fe hydro(oxide) and clay colloid, which were completely weathered and migrated, resulting in little isotope fractionation. In contrast, there was a slight increase in Hg enrichment above 300cm, suggesting that the atmospheric input outweighs the leaching loss in the upper section. The soils above 300cm also show much more negative Δ^{199}Hg than the average Δ^{199}Hg of atmospheric Hg(0), the source of Hg in our profile, and a downward positive shift of Δ^{199}Hg. We suggest that Hg deposited from the atmosphere has undergone photoreduction on the surface, which would produce negative Δ^{199}Hg when Hg is complexed by thiols in organic matter, and was subsequently leached downward. Overall, our results suggest that weathering is unlikely to cause significant fractionation of Hg isotopes, and the Hg isotope compositions in soils are primarily controlled by atmospheric deposition and photoreduction on the surface layer.