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Stable thallium isotope fractionation in soils as affected by pedogenesis

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In this study, we investigated if variations in the stable Tl isotope ratios in soil samples from different profiles can be linked to data on the extractability and speciation of soil Tl and whether the isotopic data allow drawing conclusions on the geochemical processes linked with soil formation/rock weathering. We observed a significant accumulation of the heavy ^{205}Tl isotope in the B horizons, with ε^{205}Tl values that were up to 7 higher than in the underlying bedrock. This ^{205}Tl enrichment, however, was neither reflected in the speciation of Tl nor its chemical fractionation. Furthermore, exchangeable soil Tl in the B horizons was found to be much isotopically lighter than the bulk soil Tl. Our findings suggest that the observed isotopic shift may be linked to cyclic Tl mobilization and immobilization processes over the period of rock weathering and soil formation. Oxidative Tl uptake by Mn-oxides associated with a ^{205}Tl enrichment, continuous weathering of the Tl(III)-containing phases, followed by a Tl(I) remobilization (leading to enrichment in ^{205}Tl) are suggested to be responsible for the binding of the heavy Tl isotope fraction into other phases, mainly illite (a dominant Tl host), which is not normally expected. We therefore conclude that the use of the Tl isotopic data for phase or sorption mechanism identification in a dynamic multi-phase (soil) system can be very complicated, but, in contrast, suggesting their efficient use as a proxy for redox-controlled processes.