Si isotope and Ge/Si ratios record successive cycles of dissolution/precipitation of pedogenic clay minerals

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Pedogenic clay minerals provide the most significant and reactive surface area in soils and as such largely govern biogeochemical processes at the interface between lithosphere and biosphere. Deciphering the mechanisms governing clay formation is therefore of the utmost importance to understand how soils will evolve and how they control the Earth's biogeochemical cycles. Pedogenetic transformations governing the dynamic of clay minerals in soils are however still not fully understood.

Compared to the original geochemical signature of clay-size minerals in the deepsoil Bw horizon of a Podzol ($\delta^{30}Si$ = -0.49±0.01‰; Ge/Si = 3.8±0.17µmol/mol), we document increasing enrichment of ^{28}Si and Ge in clay-size minerals produced during podzolization by the mobilization, transport and precipitation of carbon, metals and silicon. Partial dissolution of clay minerals previously enriched in ^{28}Si isotope and Ge in the eluvial E horizon ($\delta^{30}Si$ = -0.57±0.05‰; Ge/Si = 8.59±0.22µmol/mol) is the only process that could account for the occurrence of even lighter Si and greater enrichment of Ge in aluminosilicates in the illuvial Bh/Bs horizon ($\delta^{30}Si$ = -1.14±0.15‰; Ge/Si = 10.1±0.32µmol/mol).

This study provides consistent evidence for previously unrecognized cycles of (partial) dissolution and precipitation of pedogenic clay minerals during podzolization, leading to tertiary and quaternary silicate neoformation. This challenges the concept that pedogenic clay minerals would be the stable end soil-product in equilibrium with soil-forming factors and suggests that they are reactive over time depending on soil physico-chemical conditions. Si isotope and Ge/Si ratios record a "mineral memory" of the soil-forming processes, and as such provide a powerful tool for the understanding of biogeochemical processes governing soil formation.