Decoupling of shallow and deep sources of nutrients at the late stages of weathering: insights from traditional and non-traditional tracers at the Luquillo CZO

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During the evolution of the Critical Zone, the predominant source of inorganic nutrients to shallow soils changes from bedrock weathering to atmospheric inputs, while vegetation is expected to cycle nutrients more tightly as soils become more leached. In parallel, the architecture of the Critical Zone also evolves, promoting a change in water flow regime as clay-rich horizons develop in the regolith. To investigate the effect of these three concurrent processes on the fluxes of inorganic nutrients in the Critical Zone, we used a suite of traditional ($^{86}Sr/^{87}Sr$ and REE) and non-traditional ($\delta^{26}Mg$ and $\delta^{7}Li$) tracers to follow the element cycle in two deep (ca. 9 m), highly-leached regolith profiles developed over two different lithologies at the Luquillo Critical Zone Observatory (Puerto Rico). Using this multi-tracer approach, we identified a shallow/deep decoupling of solutes in the regolith, at annual to kyr time scales (porewater, exchangeable fraction, bulk regolith) for both of two different lithologies (granitic and andesitic volcaniclastic). This pattern is consistent with the expected predominance of atmospheric inputs at the late stages of weathering. Our $\delta^{26}Mg$ data suggest a minor effect of vegetation on the Mg budget of soils and all tracers indicate that lithology is still the predominant control on the composition of most of the regolith profile (>1.5 m depth). At the catchment scale, the shallow/deep decoupling of solutes affects the Mg isotope signature of the streams, reflecting the input of solutes derived from deep silicate weathering and little influence from shallow porewater. Overall, our work highlights how the use of non-traditional tracers can help disentangle the overlapping physical, chemical and biological processes that determine elemental fluxes in the Critical Zone.