Tracing controls on lithium isotope behavior in Hawaiian regoliths: Soil-atmosphere-biosphere exchanges

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Lithium (Li) isotopes may be an important proxy for tracing silicate weathering. However, the impacts of soil-atmosphere-biosphere exchanges on Li isotope behaviors in surficial environments require careful examination. In this study, we analyzed Li geochemistry of soils/saprolites/rocks, sequential extractions and vegetation in two regolith profiles developed in the humid and arid sites along the Hawaiian climosequence.

Here we present Li isotope results in regolith from the humid and arid regions, respectively. Because of opposite water discharge balance, vertical substance migration is dominant in the humid site experiencing advanced weathering, whereas advective migration prevails in the arid site undergoing incipient weathering. Decoupled shallow (<1m) and deep regolith (>1m) processes exerted distinct regulations. Shallow regolith exhibits absolute Li accumulations, peak enrichments in rhizospheric soils (humid, $\tau_{Li,Nb}$=10.93; arid, $\tau_{Li,Nb}$=2.77) and heavy Li isotopic compositions (humid, 10.3-17.2‰; arid, 12.4-16.5‰), contradict to upward enhanced weathering. In contrast, deep regolith shows Li abundance close to bedrocks, and is featured by heavy Li at the regolith discontinuities, i.e. the corestone zone (humid, 7.6-9.1‰) and carbonated-enriched layer (arid, 11.8‰). Shallow regolith was primarily influenced by atmospheric addition, vegetation cycle and pedogenesis. The results demostarte dust accretions mainly affected the δ^7Li in the humid site, and rainfall (sea spray) controlled the δ^7Li in the arid site. Both exchangeable and reducible pools are rich in heavy Li isotope (humid, 10.3-17.2‰; arid, 12.4-16.5‰), indicating isotope exchanges with soil solutions. In spite of Li as a non-essential element of low abundance in plants, the fidelity of vegetation cycle accounted for Li enrichments and isotope behaviors in soils over a long-term stability of ecosystem. The bio-interference is supported by light Li in the oxidisable (organic) soil pools, in accordance with our experimental humic acid-Li interactions. Pedogenesis is critical for the retention, mobilization and isotope fractionation of Li. Deep regolith was dominantly affected by percolation/accumulation, inheriting signals of downward transported pore fluids. Our results point out the climate-regulated biotic and abiotic contributions to Li geochemical cycle in natural systems.