Tracing Silicate Weathering Dynamics in a Shale-Dominated Hillslope using Lithium Isotopes in a Reactive Transport Framework

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The chemical weathering of silicates shapes the dynamics of landscapes by supplying essential nutrients to the biosphere over short timescales and regulating atmospheric CO_2 across longer periods. Despite such obvious importance, rates at which these terrestrial water-rock reactions occur remain difficult to constrain. Two factors that create this limitation are the difficulty in making direct observations across the vadose zone (VZ), where chemical weathering rates are often highest, and the complexity of biogeochemical processes and transport pathways that lead to solute generation in catchments.

Here, we take advantage of a novel Vadose Zone Monitoring System installed in an acively eroding, shale-dominated hillslope within the Eel River Critical Zone Observatory to characterize the development of solute chemistry at the inception of rock weathering. We resolve silicate weathering in the VZ and the transferral of solutes to groundwater and streams using dissolved concentrations and isotopes of lithium (Li). The evolution of these Li signatures across the hillslope resembles a boomerang-like pattern typified by depleted VZ water ([Li] = 0.3 μM & δ^7Li = -9 ‰), concentrated groundwater ([Li] = 2.8 μM & δ^7Li = 17 ‰), and isotopically heavy riverine ([Li] = 0.1 μM & δ^7Li = 29 ‰) end-members. Integration of these data into an isotope-enabled, multicomponent, 1-D reactive transport model shows: (1) the dynamic balance between dissolution of pre-existing minerals and precipitation of new secondary phases is a first-order control on solute generation in the VZ (2) secondary clay formation requires a shift from kaolinite- to illite-dominant precipitation with depth; (3) weathering processes that fractionate lithium continue beyond the VZ and into the saturated zone as groundwater migrates laterally downslope and is subject to mixing and dilution.