

Quantifying shale weathering processes by Li isotopes

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Lithium isotopes have emerged as a powerful tool to investigate abiotic weathering processes because isotope fraction is controlled by silicate weathering depending on the weathering rate. In this study, we explore Li isotopes as a proxy for shale weathering in the well-investigated Susquehanna Shale Hills Critical Zone Observatory (USA), which is a first-order catchment in a temperate climate in the Appalachian Mountain. Groundwater, soil and stream water reveal large variation in $\delta^7\text{Li}$ (14.5 to 40.0‰) controlled by variable but high degrees of Li retention by kaolinite and vermiculite formation. Parental shales, bulk soils and stream sediments decipher similar isotope signatures with little variations giving average $\delta^7\text{Li}$ values of -0.6, 0.5 and -0.3‰, respectively which is in the typical range observed for shales dominated by structural-bound Li and consistent with high Li retention. Plant tissue show a similar isotope composition as shale and soils indicating that trees rely on a water source distinct from mobile water in the soil zone and groundwater, which are both strongly effected by clay mineral formation. An isotope mass balance approach reveal that erosional weathering is the driving mechanism to remove Li from the system. This result is consisted with a high depletion of Li along with clay minerals in the soil zone whereas both are enriched in stream sediments. Overall shale weathering is dominated by clay transformation forming kaolinite through intermediate phases under highly incongruent weathering conditions followed by preferentially loss of fine-grained weathering products, a likely important mechanism in the modern global Li cycle.