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Quantifying shale weathering by Li isotopes at the Susquehanna Shale Hills Critical Zone Observatory

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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 δ7Li (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 reveal similar isotope signatures with little variations giving average δ7Li values of -0.6, 0.5 and -0.3‰ respectively which is in the typical range for shales dominated by structural-bound Li and consistent with high Li retention. An isotope mass balance approach reveal that Li is virtually quantitatively exported by erosional weathering from the system. This result is consisted with a high depletion of Li along with clay minerals in soils whereas both is 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 processes which is likely an important mechanism in the modern global Li cycle.