Mg isotope fractionation during shale weathering in the Shale Hills Critical Zone Observatory: Accumulation of light Mg isotopes in soils by clay mineral transformation

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Mg stable isotopic ratios have great potential as a natural tracer to elucidate weathering processes and biogeochemical pathways in surficial environments. Here, we investigated the Mg isotope fractionation during shale weathering under a temperate climate in northeastern USA. Mg isotope ratios were measured systematically in soil profiles and pore waters, groundwaters, and stream waters along a planar hillslope from a shale bedrock catchment (Shale Hills) in central Pennsylvania. Significant fractionation of Mg isotopes during shale weathering is clearly observed: δ^{26}Mg values of stream and soil pore waters are about ~0.5 ‰ to 1 ‰ lighter than the bedrock. Soil samples are up to ~0.14‰ lighter than the bedrock, suggesting a heavy Mg isotope reservoir is "missing" from our measurements. Based on a mass balance model, we infer that this missing heavy Mg reservoir (δ^{26}Mg ~1.6‰) is particulate Mg lost from the hillslope transect during shale weathering. Furthermore, soil samples show a clear decreasing trend of δ^{26}Mg values with increasing weathering duration toward the surface. We suggest that the accumulation of light Mg isotopes in surface soils at Shale Hills is due to a combined effect of i) mobilization of isotopically light Mg solute during clay dissolution and re-precipitation in vermiculite; and ii) loss of isotopically heavy particulate Mg as micro-sized particles. Our study provides the first field evidence that changes in clay mineralogy affect the Mg isotope compositions of residual bulk soils. This example also demonstrates that loss of isotopically distinct fine particles from clay-rich systems could be a new and important mechanism to drive the Mg isotope compositions of silicate weathering residuals in an opposite direction as normally expected, i.e. to lighter Mg values instead of heavier values.