

WEATHERING LOSSES AND PHASE PARTITIONING OF ALUMINUM IN THE CRITICAL ZONE USING GALLIUM/ALUMINUM RATIOS

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Aluminum (Al) is a major component of primary and secondary minerals in the Critical Zone and is responsible for biogeochemical processes such as soil acidification and stabilization of soil organic matter. In addition, Al is highly toxic to plants and organisms even at relatively low concentrations in soil and water. Unlike many other chemical elements with isotopes, quantifying and tracing Al cycling has been limited because it is monoisotopic. Gallium (Ga) may be an effective geochemical tracer for Al (Shiller and Frilot, 1996, GCA) because the two elements share many physicochemical properties. To determine fluxes of Al using Ga/Al ratios, it is important to characterize fractionation of the two elements during secondary mineral precipitation and organic matter complexation.

To investigate the extent of Ga/Al fractionation during weathering and phase partitioning, we measured Ga and Al in the top 10 m of soil at three sites: Southern Sierra CZO in California, Calhoun CZO in South Carolina, and Boulder Creek CZO in Colorado. The lithology of all sites were granitoids. Contemporary climatic conditions (mean annual precipitation and temperature) strongly varied among sites. Total Ga concentrations in soils ranged from 4 – 63 $\mu\text{g g}^{-1}$ while total Al concentrations ranged from 19 – 167 mg g^{-1} . Total Ga/Al ratios in soils ranged between 0.05 – 0.12 mmol mol^{-1} . Surface horizons generally had lower Ga/Al ratios than deeper in the profile, suggesting preferential loss of Ga in the soil surface. A sequential extraction (ammonium acetate for exchangeable phase, hydrogen peroxide for organic matter bound phase, citrate-bicarbonate-dithionite for secondary oxide phase, and HF-HNO₃ for residual phase) was conducted to determine partitioning of Ga and Al across various operationally-defined phases of sorption. Additional laboratory and field studies on Ga biogeochemistry are needed to further develop the application of the Ga/Al ratio for quantifying Al fluxes in the Critical Zone.