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Quantifying biotic and abiotic Si fluxes in the Critical Zone with Ge/Si ratios along a gradient of erosion rates

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Silicon (Si) is an important nutrient for many plant and algae species, and the ultimate source of Si is silicate mineral weathering reactions. These topics have inspired the application of Si isotope geochemistry to quantifying Si cycling in the Critical Zone, though the interpretations are often equivocal. Because germanium (Ge) geochemistry is similar to that of Si, the Ge/Si ratio is considered a tracer that provides additional constraints on Si cycling. Here, we provide Ge/Si ratios for three sites that span a gradient of erosion rates and thus time that material spends in the weathering zone before being removed. We present Ge/Si ratios in bulk rock, soil and saprolite, clay-size fractions, plant biomass, and river water from the Central Swiss Alps, the southern Californian Sierra Nevada, and the highlands of Sri Lanka. Our data perform two functions. First, they provide insight into the Ge/Si system. In particular, we document the presence of a substantial pool of Ge in plant biomass that is not associated with phytoliths, suggesting that overall plants do not discriminate against Ge relative to Si during uptake. We also quantify the preferential incorporation of Ge into clay minerals. We show that Ge/Si ratios in secondary clays may be a better proxy for weathering intensity (the fraction of denudation achieved chemically) than the Ge/Si ratio of river solutes. Ge/Si ratios in secondary clay minerals also perform as well as or even better than silicon isotopes as weathering intensity proxies. Second, the Ge/Si data are used in conjunction with silicon isotope data to develop a catchment Si mass-balance model. It suggests that the export of secondary, fractionated solids (largely clays and plant material) becomes increasingly important at shorter regolith residence times - about 80% of total solubilised Si in the rapidly eroding Alps sites, vs about 32% in the slowly eroding Sri Lanka site. The results also suggest that plant material is a surprisingly large contributor to Si export from these catchments, likely equivalent to 0.25 - 1.1x of dissolved Si export.