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Experimental Constraints on Barium Isotope Fractionation During Adsorption-Desorption Reactions: Implications for Critical Zone Tracer Applications

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Critical zone stores of bio-available metal nutrients are controlled by the balance between supply, via mineral dissolution, and export, by physical and chemical denudation. Barium isotopes offer a promising novel tracer of critical zone nutrient inventories, as significant proportions of the protolith-derived barium are transported as products of both physical (sediment-adsorbed barium) and chemical (dissolved barium) denudation, with a resolvable isotopic fractionation between the two phases. The desorption of sediment-adsorbed barium, upon contact with seawater, also enables these isotopic signals to be recorded in a variety of palaeo-archives (corals, foraminifera, bulk marine sediments) [1]. For this tracer to be applicable, it must be assumed that:

1) There is chemical and isotopic equilibrium between the dissolved and sediment-adsorbed barium in rivers.

2) The reactions are fully reversible.

3) There is quantitative desorption of sediment-adsorbed barium upon contact with seawater.

To test these assumptions, a series of experiments have been performed between common environmental adsorbents (clay minerals and iron oxyhydroxides), which transport the sediment-adsorbed barium, and surface waters (river waters and seawater), which host the dissolved barium, over a range of reaction timescales (seconds to months). Barium concentrations and isotope ratios of both phases have been quantified for all experiments performed.

Our experiments demonstrate that adsorption-desorption reactions between the adsorbed and dissolved phases are rapid and fully-reversible, validating assumptions 1) and 2). Measured barium isotope ratios of the clay-adsorbed and dissolved phases are offset in the same direction as field samples, but the magnitude of the offset is not sufficient to explain all the variability (Experiment: $\Delta^{138/134}$ Ba=0.14±0.05 [2SE, n=14]; Field Samples: $\Delta^{138/134}$ Ba=0.23±0.15 [2SE, n=21]). These experiments also confirm that upon contact with seawater, barium desorption from sediment is near-quantitative (96±3%) — indicating palaeo-archives of marine barium isotopes will be reflective of both denudation fluxes.

[1] Bridgestock et al. (2021), Chemical Geology 579, 120340.