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Stable Zirconium Isotopic Compositions of Riverine and Estuarine Systems

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Zirconium (Zr), an incompatible high field strength element with five stable isotopes, plays a crucial role in high-temperature geochemical studies due to its utility as tracer of rock formation processes. Yet, its behavior in low-temperature, aqueous environments remain largely unknown. Recently, significant Zr stable isotope fractionation was observed in the Pacific Ocean, indicating $\delta^{94/90}$Zr values higher than those found in the upper continental crust [1]. A quantitative understanding of Zr distribution and isotope variability across the hydrosphere is required to establish Zr isotopes as a robust (paleo-) oceanographic proxy. To this end, we developed novel methods for chemically processing and isotopically separating Zr in aqueous samples for analysis. Water samples were collected from the Colorado River, Yukon River, Gulf of Alaska, and estuaries of Amazon River tributaries. Iron co-precipitation procedure was performed to recover dissolved Zr from bulk water, followed by ion-exchange chromatography. Concentrations were determined via isotope dilution, using a calibrated double spike and multicollector ICP-MS analyses [2, 3]. Concentrations of Zr in suspended river sediment and riverine water were higher than those in marine particles and seawater. Dissolved Zr concentrations in rivers ranges from ~20 pmol/kg in the Amazon estuaries to ~595 pmol/kg in the Colorado River. Dissolved Zr isotopic composition of samples in this study ranges from $\delta^{94/90}$Zr = ~0.0 to ~ +0.6 ‰ relative to the RM 8299/SRM3169, between the average UCC and Pacific seawater values. Samples with high Zr concentrations show $\delta^{94/90}$Zr similar to the UCC average, but as Zr concentration decreases (possibly due to scavenging), $\delta^{94/90}$Zr increases. In the Amazon estuaries, a positive relationship is observed between salinity and $\delta^{94/90}$Zr, with fresher waters showing values closer to the UCC average. Further research is needed to explore how discharge rate, transport distance, bedrock geology, and climate influence Zr isotopic fractionation through chemical weathering. We hypothesize that drivers of observed fractionation patterns are a result of particle-water exchange, such as scavenging by authigenic particles.

[1] Huang et al. (2024), *Geochimica et Cosmochimica Acta* 365, 202-214.

[2] Ibañez-Mejia & Tissot (2019), *Science Advances* 5(12), eaax8648.

[3] Tompkins et al. (2020), *Journal of Analytical Atomic Spectrometry* 35, 1167-1186.