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Investigating the role of marine authigenic clay formation in setting the δ^7Li composition of seawater

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The geochemical composition of the past oceans is frequently used to understand the interactions between climate and Earth surface processes over Earth history. One key system is lithium and the ratio of its stable isotopes (^{7}Li/^{6}Li). The large difference in the relative mass of both isotopes induces large fractionation during low-temperature geochemical processes and, therefore, distinct isotopic signatures (δ^7Li) of the different fluxes of lithium to and from seawater. At any given time, the δ^7Li composition of seawater reflects the mass balance between Li input and output fluxes and associated fractionation factors, themselves reflecting changes in Earth-surface conditions. Marine sedimentary series from active margins can help constraining some of these fluxes.

Today, many of the major fluxes and isotope fractionations in the ocean lithium cycle, such as hydrothermal fluids, river water inputs, or uptake of lithium during low-temperature alteration of basalt at the sea floor, are relatively well constrained. However, many questions remain regarding the processes taking place at the ocean floor, particularly in the presence of detrital sediments. For example, the magnitude and the isotope composition of the flux related to the formation of authigenic clays during early diagenesis are still subject to debate.

Here, we present new lithium isotope data from three marine sites along the Chilean active margin (gravity cores located at 30°S, 33°S and 36°S), supplemented by a set of core-top and riverbed sediments from the large climatic gradient between 26°S and 41°S along the Chilean margin. We observe a systematic increase in the δ^7Li values and Li/Al ratios of clay separates from continental to marine samples, consistent with the formation of authigenic aluminosilicates (i.e., reverse weathering). Interestingly, this is associated with a large fractionation between authigenic clay and seawater of ~-30‰ and the increase in δ^7Li and Li/Al is associated with clay cation incorporation. We discuss the implications of these observations for the role of marine authigenic clay formation in setting the present and past lithium isotope composition of seawater.