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Multiple isotope constraints on clay formation and alteration during early marine diagenesis

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The mineralogy and geochemistry of detrital clays is a potential archive of past climate and terrestrial weathering. Clay mineral authigenesis – 'reverse weathering' – in marine sediments can alter detrital minerals, or add a new component to the clay-sized pool, and thus may overprint any terrestrial signal. Reverse weathering also represents an important sink in ocean element and alkalinity budgets and is thus a key process in the long-term carbon cycle [1, 2]. There is growing awareness it can happen rapidly – on the timescale of years [2, 3] – and of the importance of terrigenous sediments in supplying aluminium and other reagents. A better understanding of reverse weathering is thus key to exploiting the potential of clays as archives, and to unravelling the role of reverse weathering in geochemical cycling. Despite these incentives, authigenic clays themselves are rarely directly investigated because they are typically a minor component and structurally similar to the detrital fraction, making them hard to distinguish with conventional approaches.

For many elements (e.g. Li, Be, O, Si or Sr), the difference in isotope ratios between seawater and continental waters means that the composition of the clay fraction is sensitive to the proportion of the terrestrial component relative to the authigenic component. Here, we present a geochemical comparison of the clay-size fraction of sediments from rivers and continental margins, focusing on sites along the Chilean coast. Underpinned by an elemental and mineralogical characterisation, the data includes conventional, 'novel', radiogenic, and cosmogenic isotope systems. We show that the elemental and isotopic composition of clays found in recent (coretop/Holocene) sediments is distinct from their terrestrial counterparts, in a way that reflects the systematic addition of elements during early diagenesis. We discuss how a multiple isotope approach can be used to infer the composition of the authigenic component, the manner in which it forms, and the significance of reverse weathering at a global scale.

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