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On the reactivity of the fluid-solid interface: Insights from stable isotope signatures

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The chemical and isotopic composition of carbonate minerals has been extensively used by the geoscientific community as archive of the environmental conditions occurred at the time of deposition. The main assumption on the use of these signals for sedimentary carbonates of either biogenic or abiotic origin is that no exchange of elements and isotopes between solid and the fluid phase they are in contact with occurs post-depositionally, with the exception of research that is focused on alteration occurring during carbonate diagenesis. On the other hand, it has been shown that carbonate mineral surfaces are highly reactive in aqueous media. More specific, microscopic studies on carbonate mineral dissolution and precipitation revealed a large number of processes occurring at mineral surfaces. In this work, connecting the mineralogical observations with chemical and isotopic signatures, we examine the extent of mineral reactivity using stable Ba and Sr isotopes during dissolution and precipitation of witherite (BaCO₃) and strontianite (SrCO₃). The obtained results indicate that the exchange of elements between solid and fluid is not limited only to the outer surface layer of witherite and strontianite, but it affects up to 8 unit cells below the mineral surface. This observation has strong implications for the use of the elemental and isotopic composition of sedimentary carbonates as environmental archives, e.g., as isotopic and likely trace elemental exchange between solid and fluid occurs at a significant level after the achievement of chemical equilibrium in the fluid-solid system.