Are Clay Minerals the Primary Control on the Oceanic Rare Earth Element Budget?

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The rare earth elements (REEs) are an important tool for understanding biogeochemical cycling and sedimentary processes in the global ocean. However, their application is hindered by ambiguities in the marine REE budget surrounding a likely significant missing source. Recent work identifies a benthic flux via the pore water as the potentially dominant source of REEs to the ocean. Here, we use new pore water REE, microbeam imaging and mineralogical data to identify the sedimentary phases that interact with the pore waters and the nature of this interaction to mechanistically understand the sedimentary flux. Fe-Mn oxihydroxides are considered a ubiquitous, reactive sedimentary REE host phase, and are commonly targeted for paleocirculation reconstruction using Nd isotopes. However, the distribution, preservation, and formation of Fe-Mn phases remains poorly understood with no direct, imaging-based identification of their systematic presence to date. Our new mineralogical and direct imaging observations suggest that authigenic Fe-Mn oxihydroxides are not sufficiently abundant to account for the large proportion of REEs recoverable in leaching procedures targeting these phases. We propose that pore water REEs are instead largely sourced from early diagenetic dissolution of ubiquitous, Fe-bearing clay minerals. We demonstrate that pore water REE signatures are similar to those of river sourced clays, consistent with a detrital clay dissolution source. This finding agrees with recent suggestions that, more broadly, the dissolution of detrital siliciclastic materials in the ocean is a potentially important source of solutes to the ocean. We argue that the spread in REE patterns in pore waters relative to this clay source can be explained by fractionation during authigenic clay uptake of REEs. We conclude that clay mineral dissolution and authigenesis are likely the primary influences on REE cycling near the seafloor, with the balance between clay dissolution and authigenesis controlling the concentration, ratio of heavy and light REE abundances, and the isotopic composition of pore waters. We explore the implications of this hypothesis for the oceanic REE budget and the geographic distribution of oceanic REE inputs.