Modeling insights into uranium accumulation and isotopic fractionation during early diagenesis of marine sediments

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Uranium concentrations and isotopic ratios (\(^{238}\text{U}/^{235}\text{U}\) or \(\delta^{238}\text{U}\)) can provide quantitative information about the degree of oxygenation and de-oxygenation of past oceans. The possibility of constraining changes in global redox conditions, in contrast to many other proxies that reflect local conditions, is a particular strength of the uranium isotope approach. Within organic-rich shales, these \(\delta^{238}\text{U}\) and concentration archives are also influenced by redox conditions for the sedimentary basin in which the strata were deposited. However, our current understanding of the controls on the proxy is limited, complicating interpretations of \(\delta^{238}\text{U}\) fluctuations in both the carbonate and organic-rich shale rock record. Because uranium reduction occurs in sediments rather than in the water column [1], accumulation and isotopic fractionation are modulated by processes at or below the sediment-water interface. As such, variations in productivity-driven upwelling, basin connectivity, and sedimentation rate can influence the isotopic fractionation of uranium into organic-rich shales—the largest lever on the \(\delta^{238}\text{U}\) composition of seawater. To investigate the interplay of these factors on uranium cycling, we constructed an early diagenetic biogeochemical reaction network within a reactive transport model framework to determine how the primary biological and physical controls influence microbially mediated uranium reduction. We test the sensitivity of uranium reduction and isotopic fractionation to critical factors, such as bottom water redox conditions and sedimentation rate. Our results demonstrate that these controls can result in diagnostic patterns in uranium isotopic fractionation. Moreover, local sedimentation patterns in organic-rich shales can confound our ability to use the uranium isotopic records for interpreting global redox patterns. More broadly, this work provides important new constraints on the major controls on \(\delta^{238}\text{U}\) in the past oceans, as a critical step in its development as a paleo-redox proxy.