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Sedimentary molybdenum and uranium sequestration in a non-euxinic coastal setting: role of the sulfate-methane transition zone

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Molybdenum (Mo) and uranium (U) contents in sedimentary records are commonly used to track past changes in seafloor oxygenation. However, inadequate understanding of Mo and U sequestration mechanisms in non-euxinic coastal areas limits their use as redox proxies in these settings. Because large areas of the coastal oceans are currently undergoing partial deoxygenation due to anthropogenic nutrient inputs and increased stratification, it is critical to improve our understanding of these proxies to allow robust assessment of the trajectory of environmental change. Here, we use a comprehensive set of sediment pore water and solid-phase analyses to deconvolve the mechanisms of authigenic Mo and U sequestration in a shallow non-euxinic coastal setting in the northern Baltic Sea. Despite the permanently oxic bottom waters in the area, eutrophication over the past decades has led to establishment of a shallow sulfate-methane transition zone (SMTZ) in the sediment, which is typical for human-impacted coastal settings on a trajectory towards hypoxia. Our results demonstrate remarkably synchronous patterns of Mo and U sequestration, whereby their authigenic uptake is largely predicated upon the depth and intensity of the SMTZ. Based on sequential extraction analyses, the authigenic Mo pool is dominated by refractory Fe-S phases such as pyrite and nanoscale $FeMoS_4$, signaling that authigenic Mo uptake largely proceeds through the Fe-sulfide pathway. In addition, we observe a pool of extremely labile Mo deep within the SMTZ, potentially denoting a transient phase in authigenic Mo uptake and/or partial switch in the mode of sequestration to the organic matter pathway at low levels of dissolved iron. Authigenic U is largely hosted by acid-extractable and refractory phases, reflecting sequestration into poorly crystalline monomeric U(IV) and crystalline uraninite, respectively. Analogously to Mo, authigenic sequestration of U proceeds at two distinct fronts within the SMTZ, which are characterized by shifts in dissolved sulfide concentrations, providing strong evidence for a link between sulfide-producing processes and U reduction. Our results imply that both Mo and U have the potential to capture temporal shifts in bottom water oxygenation indirectly, through the connection between oxygenation and the depth of the SMTZ. Of the two elements, Mo appears a more viable redox proxy because of the substantially higher share of the authigenic pool. However, temporal resolution of these proxies is restricted by the relatively deep authigenic uptake within the sediment column and the integrated character of the signal caused by vertical migrations of the SMTZ. These findings set a framework for interpreting sedimentary Mo- and U-based paleoredox archives in other non-euxinic coastal settings.