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Rare earth element cycling across the chemocline of the Pettaquamscutt River estuary, Rhode Island

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Rare earth elements (REEs) concentrations were measured across the chemocline in a highly stratified estuary with permanent euxinic bottom waters (i.e., ΣS^{-II} up to ca. 4 mM). The REEs concentrations decrease with depth in the oxic waters above the chemocline, but exhibit dramatic increases in concentration through the chemocline, corresponding with substantial increases in dissolved iron [total dissolved Fe and Fe(II)] and manganese (Mn) concentrations. For example, the mean $\pm 1\sigma$ neodymium (Nd) concentration in the oxic surface water is 0.51 ± 0.18 nmol kg⁻¹, but increases by a factor of nearly 5 across the chemocline reaching values as high as 2.47 nmol kg⁻¹ at a depth of 7.4 m. Total Fe concentration increases from 0.3 ± 0.2 $\mu\text{mol kg}^{-1}$ in the oxic surface waters to 3.4 $\mu\text{mol kg}^{-1}$ within the chemocline, or by a factor of ca. 11, before decreasing to 0.4 $\mu\text{mol kg}^{-1}$ at depth in the euxinic bottom waters. The decrease in Fe concentration in the euxinic bottom waters reflects precipitation of Fe-sulfide minerals (e.g., pyrite) within the water column [1], which does not affect the REEs. Manganese concentrations increased from 1.2 ± 0.36 $\mu\text{mol kg}^{-1}$ in the oxic surface waters to 5.7 $\mu\text{mol kg}^{-1}$ within the chemocline (nearly 5-fold increase), before decreasing to 3.4 $\mu\text{mol kg}^{-1}$ in the euxinic waters. Geochemical modeling suggests REE removal from solution reflects precipitation of authigenic REE-phosphate phases. Negative Ce anomalies characterize waters of the entire water column, indicating that these anomalies can persist even in highly euxinic waters. Accordingly, processes that produce negative Ce anomalies in natural waters are more complex than simply oxidation of Ce(III) to Ce(IV) and subsequent removal from solution as, for example, cerianite (CeO₂). Our results may have consequences for the application of Ce anomalies as paleo-redox indicators.

[1] Wilkins and Barnes (1997) *Am. J. Sci.* **297**, 620.

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