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Mercury isotope fractionation in a meromictic paleo-ocean analogue

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Mercury (Hg) isotopes in sedimentary rocks can act as a novel proxy for reconstructing paleo-environmental changes due to their unique "three-dimensional" isotope system, which includes mass-dependent (MDF) and mass-independent isotope fractionation for both odd mass number (odd-MIF, Δ^{199}Hg and Δ^{201}Hg) and even mass number (even-MIF, Δ^{200}Hg and Δ^{204}Hg) isotopes. Yet, the application of Hg isotopes to paleoceanogrpahic problems is currently hindered by a limited understanding of isotope fractionation mechanisms in the type of anoxic settings that typified ancient-Earth oceans. Precambrian oceans were persistently redox-stratified, with oxic surface waters overlying anoxic deeper waters rich in either dissolved ferrous iron ("ferruginous") or hydrogen sulfide ("euxinic").

We analyzed Hg isotopes in sediments across a redox gradient from two US-based Precambrian ocean analogues: ferruginous Deming Lake in Minnesota and euxinic Siders Pond in Massachusetts. Sediments at both sites exhibit significant variability in Hg concentrations, ranging from 25 ng/g to 249 ng/g, and contain generally negative MDF values (δ^{202}Hg = -1.18 ± 0.07‰; 2SD to -0.57 ± 0.07‰; 2SD) and positive MIF values (Δ^{199}Hg = 0.03 ± 0.02‰; 2SD to 0.31 ± 0.02‰; 2SD). In the ferruginous lake, strong negative excursions of Δ^{199}Hg by up to -0.2‰ coincide with the oxycline where Fe(III) oxides form and with the depth where Fe(III) oxides converts to Fe(II) minerals. This juxtaposition suggests a primary control of Fe minerals on Hg isotope fractionation in ferruginous lake. In contrast, in the euxinic pond, Δ^{199}Hg shows a progressive negative shift beneath the oxycline, with a concurrent positive shift in δ^{202}Hg and increase in sulfide concentration. This fractionation pattern, replicated in previous laboratory experiments and found at multiple times in the ancient sedimentary record, is consistent with photic zone euxinia. Our results indicate that ferruginous and euxinic conditions impart unique Hg isotope fractionation patterns, paving the way for a new and novel utility for Hg isotopes in the Precambrian sedimentary record.