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Tracking oxidative U cycle with Pb isotopes in marine carbonates

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The Great Oxidation Episode (GOE) at 2.43-2.1 Ga irreversibly changed biogeochemical cycling on Earth's surface to the one controlled by presence of free atmospheric oxygen. Before that redox cycling was only locally expressed near oases with high biological productivity. Uranium is a redox-sensitive element that is insoluble under anoxic surface conditions, like Th, but decouples from Th under oxic conditions, resulting in its high concentration in the modern oxic oceans and its long seawater residence time. It is removed in anoxic setting and thus strongly enriched in organic-rich shales. Compilation of U concentration data for Precambrian and Phanerozoic black shales[1] revealed that the pre-GOE shales are not strongly enriched in U above the average upper continental crust (UCC) level, consistent with largely anoxic surface conditions during their deposition. However, concentration data cannot resolve whether U was still soluble at low concentrations in seawater at the time of deposition. A long-term record, utilizing a highly sensitive redox proxy, is therefore required to address this question.

U-Pb and Pb-Pb isotope dating of shallow-marine, sedimentary carbonates has been conducted for decades and these data might bear on U decoupling from Th in the pre-GOE surface environments. A compilation of U-Pb and Pb-Pb isotope data from literature data for sedimentary carbonates yield dates close to their depositional ages, indicating closed system behavior since their deposition. The data are used to calculate time-averaged kappa (Th/U) values to constrain U and Th decoupling in seawater from which these carbonates precipitated. While there are few pre- and syn-GOE, and ca. 1.9 Ga literature datasets with near to or even above the average UCC kappa values ([2], the majority of the datasets yielded values <1.4, well below the UCC kappa value regardless of their depositional age and displaying no secular change. These data indicate that U was decoupled from Th in the early Earth oceans since at least ca. 3.0 Ga ago and must have been soluble at low concentrations under largely anoxic terrestrial and shallow-marine conditions during the Archean.

[1] – Partin et al., 2013, EPSL 369-370, 284-293.

[2] – Rudnick and Gao, Treatise of Geochemistry, 2014.