

Mesoarchean redox cycling from shallow to deep through coupled Fe-S stable isotope measurements

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An increasing body of work suggests the production of free oxygen in trace amounts prior to the Great Oxidation Event [1, 2], and this cryptic production may be reflected in the coupled redox cycling of iron and sulfur as early as 2.96 Ga [3]. To further explore this possibility, we performed coupled Fe-S stable isotope analyses to constrain Fe and S redox cycling across a shallow-to-deep transition captured in carbonates, mudstones, sulfidic shales, and banded iron formations of the 2.94 Ga Ball Assemblage, Red Lake Greenstone Belt, ON, Canada. Our data reveal both active iron and sulfur redox cycling, with $\delta^{56}\text{Fe}$ values ranging from -2.88 ‰ to +1.50‰, and $\delta^{34}\text{S}$ values ranging from -4.90‰ to +7.76‰. The large range in $\delta^{56}\text{Fe}$ may be explained by Fe depletion linked to precipitation of isotopically heavy Fe oxides in the offshore environment, generating low $\delta^{56}\text{Fe}$ values in the residual Fe(II) pool that upwelled onto the carbonate platform. Important variations in $\delta^{34}\text{S}$ were observed only on the carbonate platform, indicating that shallow water settings where carbonate precipitation was occurring also experienced the most important sulfur redox cycling, with negative values representing times of expansion of the SO_4 pool, and positive values suggesting near-total drawdown at times by sulfate reduction. Moreover, variations in $\delta^{34}\text{S}$ in the shallow water facies correlate broadly with $\delta^{56}\text{Fe}$ values. One possibility is that sulfate reduction modulated Fe availability on the shelf via the periodic establishment of a euxinic wedge offshore to the shelf, as suggested by the highly sulfidized shales occurring across the shallow-to-deep transition. This work provides important new data as well as a unique spatial perspective to the potential coupling of Fe and S redox cycles in Mesoarchean marine environments.

[1] Anbar *et al.* (2007), *Science* 317, 1903–1906.

[2] Planavsky *et al.* (2014) *Nature Geoscience* 7, 283–286.

[3] Eickmann *et al.* (2018), *Nature Geoscience* 11, 1–7.