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Restructuring of marine Fe burial pathways in the Late Archean driven by enhanced sulfur supply

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Many Archean shales are characterized by high Fe contents, but it remains unclear whether this Fe enrichment reflects Fe-rich detrital input from the provenance, or additional Fe supplied from seawater and/or pore fluids. Here we establish a detrital-controlled empirical relationship between total Fe_2O_3 and TiO_2/Al_2O_3 and use deviations from this baseline to quantify non-detrital Fe addition. Most Archean shales show systematic Fe excess relative to detrital control, indicating widespread external Fe input beyond provenance-derived supply. Combining geochemical results with XRD mineralogy reveals a temporal shift in dominant Fe burial pathways. In >2.8 Ga shales, Fe is mainly hosted by authigenic Fe-silicates (especially Fe-rich chlorite), with additional contributions from Fe-oxides and Fe-siderite. In contrast, during 2.8-2.5 Ga, pyrite becomes increasingly abundant, marking the emergence of sulfide minerals as an additional major Fe sink.

Thermodynamic calculations show that higher aqueous Fe concentrations, higher temperature (potentially >25℃), and moderately elevated pH (6.5-7) favor Fe-silicates crystallization. The widespread occurrence of authigenic Fe-rich chlorite in >2.8 Ga shales therefore may point to warm, Fe-rich, near-neutral marine conditions. In contrast, higher HS^- availability and lower temperature (potentially <30℃) conditions favor pyrite precipitation. Enhanced sulfur supply may have resulted from increased continental emergence, which would expand exposed land area for photochemical and biologically mediated pyrite oxidation and promote surface cooling.

Taken together, Archean oceans were broadly Fe-rich: prior to 2.8 Ga, Fe was mainly buried as Fe-silicates, whereas 2.8-2.5 Ga pyrite became an additional Important Fe sink, potentially reflecting enhanced sulfur input associated with continental emergence.