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Re-examining deep time sulfur isotope records from a micro-scale perspective

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The early Paleoproterozoic Great Oxidation Event (GOE) [1] is a hallmark event in Earth's history. It is believed that, after the initial increase in pO_2, the continued expansion of surface oxidant reservoirs initiated a series of disturbances in biogeochemical and elemental cycles that drove secular variation in Paleoproterozoic pO_2 concentrations [2,3]. This concept is, in part, based on interpretations of bulk S isotope ($\delta^{34}S$) profiles from ancient metasedimentary rocks that presume a linkage between $\delta^{34}S$ excursions in pyrite and global perturbations to that cycle, e.g., that positive $\delta^{34}S$ excursions reflect decreasing seawater sulfate levels brought about by ocean deoxygenation events. However, a growing body of evidence indicates that pyrite genesis during early diagenesis to late metamorphism can contribute to intra-sample $\delta^{34}S$ heterogeneity and potentially bias interpretations of bulk $\delta^{34}S$ records [4–6].

Here we re-examine the environmental significance of pyrite $\delta^{34}S$ records from the metasedimentary rocks of the c. 2.1 Ga Francevillian Group (Gabon) and c. 2.0 Ga Onega Basin (Russia) that have figured prominently in concepts of Earth's early oxygenation and accompanying environmental change. We show that bulk $\delta^{34}S$ profiles are not inherently reliable records of primary global (or even local) S cycle disturbances; however, high-resolution analysis (secondary ion mass spectrometry and synchrotron-based S imaging) provide additional information that can be used to extract meaningful depositional and diagenetic information from these altered rocks. We further demonstrate that combining petrographic observations and bulk and *in situ* S isotope analysis provides a deeper understanding of the mechanisms governing S isotope fractionations in sediments, and how these signatures are preserved and altered over geologic time.

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