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Deconstructing the Depositional and Diagenetic Controls on Bulk Sulfur Isotope Records of Sedimentary Pyrite: Insights from SIMS-based Microanalysis

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Stable isotopic data, such as pyrite $d^{34}S$, provide a framework for reconstructing biogeochemical cycling, global redox budgets, and environmental change over Earth history. However, these reconstructions often rely on bulk sediment measurements and the assumption that the bulk values measured from a sample in a given stratigraphic section provide information about global conditions. Here we investigate a record of bulk $d^{34}S_{pyr}$ spanning a range of > 100‰ in mid-Pleistocene marine sedimentary pyrites from Valle di Manche, Italy. The first-order trend in bulk $d^{34}S_{pyr}$ is controlled primarily by changes in sedimentation rate and organic carbon loading that influence resupply of sulfate from the overlying water column during initial sulfate reduction in newly deposited sediments. Individual pyrite grains have various petrographic textures and the SIMS populations of $d^{34}S_{pyr}$ often contain several, isotopically distinct modes. The most ^{34}S-depleted mode is always characterized by nucleation textures (i.e., framboids) and it agrees well with a prediction based solely on sedimentation rate and TOC using a biological fractionation during microbial sulfate reduction that approaches equilibrium values (~72‰). Deviations between the bulk $d^{34}S_{pyr}$ and the integrated $d^{34}S_{pyr}$ of framboids are caused by petrographically distinct populations (individual equant grains and/or overgrowth/recrystallization textures) that are often anomalously ^{34}S-enriched, arising from pulses of pyrite formation during later-stage diagenesis (e.g., associated with AOM and the sulfate-methane transition zone). The stratigraphic occurrence of these deviations is associated with evidence of abrupt sedimentation (e.g., event beds) that suggest deviation from background steady-state processes. Together, these observations enable us to reconstruct the suite of biotic and abiotic processes that together give rise to bulk sedimentary $d^{34}S_{pyr}$ values and can be used to improve environmental and ecological interpretations of these records preserved in ancient strata.