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Controls on estuarine pyrite formation and implications for Paleozoic Earth system oxygenation

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Geochemical redox proxies have yielded conflicting interpretations for the rise of oxygen in the Early Paleozoic Era, a period marked by many evolutionary radiations. A primary source of oxygen to the ocean and atmosphere is the reduction of sulfate to sulfide and its subsequent burial in sedimentary pyrite; therefore, to improve our knowledge of Early Paleozoic oxygenation, it is important to understand how dynamic environmental conditions in low-oxygen basins may affect burial fluxes of pyrite in sediments. We collected sediments from the Chesapeake Bay—an estuary with multiple spatiotemporal gradients in sulfate concentration, hypoxia intensity, organic carbon flux and provenance, sedimentation rate, and bioturbation—to evaluate primary controls on pyrite accumulation in a dynamic, oxygen-stressed basin. Pyrite accumulation in the upper 0.5 m of sediment reached a maximum in sediments that occupied the mid-range of sulfate-chloride ratios, not in strongly reducing, sulfate-depleted sediments. We consider these results in terms of sulfate supply versus sulfate demand: highly reducing sediments with high microbial sulfate demand will quickly reduce the available supply of pore water sulfate, but the supply of fresh sulfate to generate more pyrite will be limited. In contrast, mildly ventilated sediment columns can match sulfate demand with sulfate supply to generate more pyrite, although excessive ventilation will introduce enough oxygen to suppress pyrite retention. Sediment cores with lower sulfate-chloride ratios also had higher FeS/pyrite ratios, suggesting that conversion of FeS to pyrite in the upper sediment pile is impeded under strong sulfate depletion. Therefore, pyrite formation may occur more readily in cores with a mildly oxidized dissolved sulfur pool, perhaps via the polysulfide reaction. Our results imply that mild ventilation of sediments, paired with the introduction of mild oxidizing power to the sediment column, may have temporarily increased pyrite burial fluxes as bioturbation rates increased in the Early Paleozoic, thus providing a positive feedback to oxygen accumulation. This contrasts with previous Earth system models that have assumed that incipient bioturbation induced a monotonic decrease in pyrite burial efficiency.