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Multi-isotopic constraints on pyrite oxidation and the sulfur cycle

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Pyrite oxidation plays a key role in chemical weathering and the sulfur and oxygen cycles by generating dissolved sulfate and acidity, consuming O_2, and releasing CO_2 and metals. Despite significant insights gained from isotopic approaches, the multiple isotope compositions of sulfur ($\delta^{34}S$, $\Delta^{33}S$, and $\Delta^{36}S$) and oxygen ($\delta^{18}O$ and $\Delta^{17}O$) from pyrite-derived sulfate remain poorly constrained. We propose that sequential experiments can separate pyrite oxidation pathways over time, enabling isotopic fingerprinting of pyrite-derived sulfate end members. To fingerprint oxidation pathways, we conduct abiotic pyrite oxidation experiments lasting minutes to years, time scales not captured under typical laboratory conditions, and measure $\delta^{34}S$, $\Delta^{33}S$, $\Delta^{36}S$, $\delta^{18}O$, and $\Delta^{17}O$ of the resulting sulfate. Oxygen isotope results show that during the initial (<2h) oxidation of fresh pyrite surfaces over half of the sulfate's oxygen is derived from atmospheric O_2. Afterwards, the O_2-derived-oxygen content in sulfate diminishes, following a power law, and becomes negligible after 2 years. The balance of sulfate oxygen is derived from water through pyrite oxidation by Fe^{3+}. Sulfur isotope compositions of the same sulfates further support the presence of two dominant pyrite oxidation pathways with distinct oxygen sources, direct oxidation by atmospheric O_2 and oxidation by Fe^{3+}. Following the results of our experiments, we use our updated $\delta^{18}O$ and $\Delta^{17}O$ constraints to reevaluate published continental-scale sulfate input to the ocean, estimating sulfate from O_2 and Fe^{3+} pyrite oxidation pathways and the proportion reset by biological sulfur cycling.