

Pyrite oxidation and sulfur (re)cycling in erosive landscapes as revealed by sulfate triple-oxygen isotopes

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Oxidation of the sulfur-bearing mineral pyrite (FeS_2) during weathering on land constitutes a major sink of O_2 and—indirectly via subsequent acidification of carbonate rocks—source of CO_2 to Earth's atmosphere over geologic timescales. Furthermore, by producing the terminal electron acceptor sulfate (SO_4^{2-}), pyrite oxidation may fuel microbial metabolisms and secondary sulfur recycling in sub- or anoxic groundwaters or riparian zones (collectively the 'critical zone'). A growing canon of studies has documented rapid sulfide oxidation rates in tectonically active landscapes that exhume metasedimentary silicate rock with minor pyrite. However, to understand the impact of these landscapes on the global sulfur cycle, it is necessary to quantify the role of secondary sulfur recycling. Traditional geochemical approaches—particularly sulfate stable sulfur isotopes ($\delta^{34}\text{S}$)—are not sensitive to secondary sulfur recycling since re-oxidation will dampen any expressed fractionation. To provide new insight into the role of sulfur recycling in the critical zone, here we measure the triple-oxygen isotope compositions ($\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$) of dissolved sulfate in rivers, landslide seepage, and groundwater collected across an erosion rate gradient in the Southern Alps of New Zealand. We find that catchments with high erosion rates contain dissolved sulfate with low $\delta^{18}\text{O}$ and high $\Delta^{17}\text{O}$ values, consistent with a pyrite weathering end-member observed previously in similar mountainous regions. However, for lower erosion-rate catchments, we observe a shift toward higher $\delta^{18}\text{O}$ and lower $\Delta^{17}\text{O}$ values that cannot be explained purely by admixture of additional sulfate sources (e.g., seawater, gypsum dissolution). We hypothesize that this shift implies oxygen-isotope equilibration with water during secondary sulfate reduction and re-oxidation in the critical zone. We will discuss these results within the context of the global sulfur cycle and its dependence on tectonic and geomorphic processes.