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Sulfur isotope effects during sulfide oxidation to sulfate: model meets reality

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The combination of laboratory experiments and numerical models in the study of stable sulfur isotope fractionation is a useful tool that contributes to a better understanding of sulfur transformations in the environment. Because sulfur isotope fractionation during oxidative sulfur cycling is expected to yield much smaller isotope fractionation than sulfate reduction, sulfur isotope effects during sulfide oxidation have not received much attention. Nevertheless, in situations where sulfide oxidation dominates over other sulfur transformations, the imprint of these small isotope effects is preserved. These isotope imprints provide crucial information about the biogeochemical conditions under which sulfide oxidation occurred. Unlike sulfate reduction, where sulfur intermediates are hardly accumulated and rarely released to the environment, sulfide oxidation to sulfate often involves accumulation of intra- and extracellular elemental sulfur as an intermediate, which may be accessed later for oxidation to sulfate by the same or different organisms. The accumulation and consumption of a pool of elemental sulfur complicates the interpretation of sulfur isotope effects during sulfide oxidation, as kinetic sulfur isotope effects from oxidation of sulfide to elemental sulfur are superimposed by sulfur isotope exchange between sulfide and elemental sulfur, a pool that is simultaneously altered by isotope effects related to the oxidation of elemental sulfur to sulfate. We derived a numerical isotope mass balance model that combines the rates for sulfur isotope exchange between sulfide and elemental sulfur, oxidation of sulfide to elemental sulfur and oxidation of elemental sulfur to sulfate with the respective isotope effects. With this model, we can predict the evolution of the isotope composition of sulfide, elemental sulfur, and sulfate during a sulfide oxidation experiment. Sensitivity tests show that these isotope trends not only strongly depend on the size of isotope fractionation, but also on the individual oxidation rates and their relative timing to each other. Our model results are compared to existing data in the literature, and with new laboratory experiments in which sulfide was oxidized to elemental sulfur and sulfate by pure cultures of *Sulfurimonas* strain CVO grown under nitrate-reducing conditions.