Title:	High-Precision Measurements of 33S and 34S Fractionation during SO2 Oxidation Reveal Causes of Seasonality in SO2 and Sulfate Isotopic Composition
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Abstract:	This study presents high-precision isotope ratio-mass spectrometric measurements of isotopic fractionation during oxidation of SO2 by OH radicals in the gas phase and H2O2 and transition metal ion catalysis (TMI-catalysis) in the aqueous phase. Although temperature dependence of fractionation factors was found to be significant for H2O2 and TMI-catalyzed pathways, results from a simple 1D model revealed that changing partitioning between oxidation pathways was the dominant cause of seasonality in the isotopic composition of sulfate relative to SO2. Comparison of modeled seasonality with observations shows the TMI-catalyzed oxidation pathway is underestimated by more than an order of magnitude in all current atmospheric chemistry models. The three reactions showed an approximately mass-dependent relationship between 33S and 34S. However, the slope of the mass-dependent line was significantly different to 0.515 for the OH and TMI-catalyzed pathways, reflecting kinetic versus equilibrium control of isotopic fractionation. For the TMI-catalyzed pathway, both temperature dependence and 33S/34S relationship revealed a shift in the rate-limiting reaction step from dissolution at lower temperatures to TMI-sulfite complex formation at higher temperatures. 1D model results showed that although individual reactions could produce Δ33S values between −0.15 and +0.2‰, seasonal changes in partitioning between oxidation pathways caused average sulfate Δ33S values of 0‰ throughout the year.
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