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Title:	Sulfur isotope fractionation during oxidation of sulfur dioxide: gas-phase oxidation by OH radicals and aqueous oxidation by H2O2,O3 and iron catalysis.
Authors:	Sinha, B. (/jspui/browse?type=author&value=Sinha%2C+B.)
Keywords:	Chemistry Atmosphere Sulfur
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Abstract:	The oxidation of SO2 to sulfate is a key reaction in determining the role of sulfate in the environment through its effect on aerosol size distribution and composition. Sulfur isotope analysis has been used to investigate sources and chemical processes of sulfur dioxide and sulfate in the atmosphere, however interpretation of measured sulfur isotope ratios is challenging due to a lack of reliable information on the isotopic fractionation involved in major transformation pathways. This paper presents laboratory measurements of the fractionation factors for the major atmospheric oxidation reactions for SO2: Gas-phase oxidation by OH radicals, and aqueous oxidation by H2O2, O3 and a radical chain reaction initiated by iron. The measured fractionation factor for 34S/32S during the gas-phase reaction is $\alpha_{OH} = (1.0089\pm0.0007)-((4\pm5)\times10^{-5})\,T(°C)$. The measured fractionation factor for 34S/32S during aqueous oxidation by H2O2 or O3 is $\alpha_{aq} = (1.0167\pm0.0019)-((8.7\pm3.5)\times10^{-5})T(°C)$. The observed fractionation during oxidation by H2O2 and O3 appeared to be controlled primarily by protonation and acid-base equilibria of S(IV) in solution, which is the reason that there is no significant difference between the fractionation produced by the two oxidants within the experimental error. The isotopic fractionation factor from a radical chain reaction in solution catalysed by iron is $\alpha_{Fe} = (0.9894\pm0.0043)$ at 19 °C for 34S/32S. Fractionation was mass-dependent with regards to 33S/32S for all the reactions investigated. The radical chain reaction mechanism was the only measured reaction that had a faster rate for the light isotopes. The results presented in this study will be particularly useful to determine the importance of the transition metal-catalysed oxidation pathway compared to other oxidation pathways, but other main oxidation pathways can not be distinguished based on stable sulfur isotope measurements alone.
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