Title:	Sulfur isotope fractionation during heterogeneous oxidation of SO2 by mineral dust.
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Abstract:	Mineral dust is a major fraction of global atmospheric aerosol, and the oxidation of SO2 on mineral dust has implications for cloud formation, climate and the sulfur cycle. Stable sulfur isotopes can be used to understand the different oxidation processes occurring on mineral dust. This study presents measurements of the 34S/32S fractionation factor α34 for oxidation of SO2 on mineral dust surfaces and in the aqueous phase in mineral dust leachate. Sahara dust, which accounts for ~60% of global dust emissions and loading, was used for the experiments. The fractionation factor for aqueous oxidation in dust leachate is αleachate = 0.9917±0.0046, which is in agreement with previous measurements of aqueous SO2 oxidation by iron solutions. This fractionation factor is representative of a radical chain reaction oxidation pathway initiated by transition metal ions. Oxidation on the dust surface at subsaturated relative humidity (RH) had an overall fractionation factor of αhet = 1.0096±0.0036 and was found to be almost an order of magnitude faster when the dust was simultaneously exposed to ozone, light and RH of ~40 %. However, the presence of ozone, light and humidity did not influence isotope fractionation during oxidation on dust surfaces at subsaturated relative humidity. All the investigated reactions showed mass-dependent fractionation of 33S relative to 34S. A positive matrix factorization model was used to investigate surface oxidation on the different components of dust. Ilmenite, rutile and iron oxide were found to be the most reactive components, accounting for 85% of sulfate production with a fractionation factor of α34 = 1.012±0.010. This overlaps within the analytical uncertainty with the fractionation of other major atmospheric oxidation pathways such as the oxidation of SO2 by H2O2 and O3 in the aqueous phase and OH in the gas phase. Clay minerals accounted for roughly 12% of the sulfate production, and oxidation on clay minerals resulted in a very distinct fractionation factor of α34 = 1.085±0.013. The fractionation factors measured in this study will be particularly useful in combination with field and modelling studies to understand the role of surface oxidation on clay minerals and aqueous oxidation by mineral dust and its leachate in global and regional sulfur cycles.
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