

Sulfur and oxygen isotopic signatures of sulfate aerosols produced in a large atmospheric simulation chamber

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Ice-core observations have shown that sulfur isotopic composition of stratospheric sulfate produced after explosive volcanic eruptions follows a sulfur mass-independent fractionation (S-MIF) processes. Such unique isotopic imprint provides a new proxy to retrieve past climate-impacting volcanic eruptions which is essential to determine climate variability at annual to decadal timescales. However, the origin and preservation of S-MIF signals in stratospheric sulfate is still not well understood. In this study, we carry out a set of experiments simulating sulfate production in stratosphere-like environments (e.g., UV spectrum, low water vapor) utilizing a large atmospheric chamber (4 m³) with varying SO₂ and H₂O concentrations in synthetic air. The chamber is equipped with a set of analyzer, allowing to follow the evolution of precursors (e.g., SO₂, H₂O, O₃) and the production of sulfate aerosols. The large volume of the chamber also allows enough production of sulfate at relatively low SO₂ concentrations (ppb to ppm levels) and collection of time-dependent product during a single experiment. The later is useful in terms of resolving the sulfur isotope fractionation factors associated with photo- and/or OH-oxidation. Under the UV spectrum of the chamber (> 280 nm), sulfate production in the chamber with and without O₃ present are observed and the produced sulfate aerosols are collected simultaneously with residual SO₂. These samples, analyzed for O- and S-isotopic compositions, are used to constrain the mechanism responsible for the S-MIF observed in the stratosphere sulfate aerosols.