

二酸化硫黄光励起反応を用いて、気候変動に影響を与える火山噴火を特定する SO₂ photoexcitation mechanism links sulfur MIF in polar sulfate to climate-impacting volcanism

服部 祥平^{1*}, ヨハン シュミット², マシュー ジョンソン², ダニエラチェ セバスチアン³, 山田 明憲⁴, 上野 雄一郎⁵, 吉田 尚弘¹
Shohei Hattori^{1*}, Johan A. Schmidt², Matthew S. Johnson², Sebastian Danielache³, Akinori Yamada⁴, Yuichiro Ueno⁵, Naohiro Yoshida¹

¹ 東工大 総理工 化学環境, ² コペンハーゲン大学 化学科, ³ 上智大学, ⁴ 東京大学 地球惑星科学科専攻, ⁵ 東工大 理工 地球惑星

¹Department of Environmental Chemistry and Engineering, Tokyo Institute of Technology, ²University of Copenhagen, ³Sophia University, ⁴Department of Earth & Planetary Science, University of Tokyo, ⁵Department of Earth & Planetary Sciences, Tokyo Institute of Technology

Natural climate variation such as that due to volcanoes is the basis for identifying anthropogenic climate change. However, knowledge of the history of volcanic activity is inadequate, in particular concerning the explosivity of specific events. Stable sulfur isotope abundances contain additional information and recent studies show a correlation between volcanic plumes that reach the stratosphere and mass-independent anomalies in sulfur isotopes in glacial sulfate. We describe a new mechanism, photoexcitation of SO₂, links the two yielding a useful metric of explosivity of historic volcanic events. A plume model of SO₂ to sulfate conversion was constructed including photochemistry, entrainment of background air and sulfate deposition. Isotopologue-specific photoexcitation rates were calculated based on the UV absorption cross sections of ³²SO₂, ³²SO₂, ³²SO₂ and ³²SO₂ from 250 to 320 nm. The model demonstrates that UV photoexcitation is enhanced by altitude while mass-dependent oxidation such as SO₂ + OH is suppressed by in situ plume chemistry, allowing the production and preservation of a mass-independent sulfur isotope anomaly in the sulfate product. The model accounts for the amplitude, phases and time development of $\Delta^{33}\text{S} / \delta^{34}\text{S}$ and $\Delta^{36}\text{S} / \Delta^{33}\text{S}$ found in glacial samples. For the first time we are able to identify the process controlling mass-independent sulfur isotope anomalies in the modern atmosphere. This mechanism is the basis of identifying the magnitude of historic volcanic events.

キーワード: 成層圏硫酸エアロゾル, 地球寒冷化, 大規模火山噴火, 硫黄同位対比, 質量非依存同位体分別, 大気化学