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Poster Session Day 1

An Isotopic Study based on New Measured SO_2 Photoexcitation Spectrum

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Spectrums of sulfur dioxide (SO_2) isotopologues provide unique insights into SO_2 photoexcitation reaction mechanism which is thought important for contributing sulfur mass-independent fractionation (S-MIF) in the modern atmosphere during volcanic eruption processes. However, available spectra are limited, and their low resolution and precision have introduced biases in the calculation of fractionation parameters.

In this study, we present newly measured 32, 33, 34, and 36-SO_2 spectrum across the 240-320 nm wavelength range, with a resolution of 0.4 cm^{-1} and an error of 3-10%. This represents a 20-fold improvement in resolution compared to the previous study [1].

Our findings support the hypothesis that isotopic effects at the intersystem crossing is the main source of the S-MIF in SO_2 photoexcitation, as suggested by Whitehill et al. [2] Furthermore, data in this study in which $^{34}\varepsilon$, ^{33}E, and ^{36}E being small and relatively insensitive to the excitation wavelength band that potentially contributing to the S-MIF in the final organosulfur products.

The photoexcitation induced enrichment factors from reported volcano eruption data were also compared to the result in this study. The slope of $\Delta^{36}S/\Delta^{33}S$ in the self-shielding calculation does not match reported volcano data of an overall negative slope fit, and accordingly the SO_2 photoexcitation is unlikely to produce the primary trend in $\Delta^{33}S$-$\Delta^{36}S$ plot observed in stratospheric sulfate aerosol (SSA). Nevertheless, the altitude dependence and self-shielding of SO_2 photoexcitation may provide dispersion of the SSA's $\Delta^{33}S$ and $\Delta^{36}S$ plot observed in SSA.

References

[1] Danielache, S. O. et al., J. Geophys. Res. Atmos. 2012, 117(D24).
[2] Whitehill, A. R. et al., PNAS 2013, 110(44), 17697-17702.