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^{32}S, ^{33}S, ^{34}S, and ^{36}S Sulfur Dioxide B^1B_1-X^1A_1 Absorption Band Measurement Using LAD Regression

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Sulfur dioxide (SO_2) is a critical component in forming atmospheric aerosols, a process that significantly influences the climate system and air quality. The highly structured SO_2 UV absorption spectrum directly results from electronic transitions and a complex and nuanced set of vibrational and rotational energy levels. Isotopic substitution triggers minor differences in energy transitions and produces large isotopic effects. Modern dust samples in Greenland and other ice-core records show significant sulfur mass-independent fractionations (S-MIF). SO_2 photoexcitation in the lower stratosphere has been shown to produce signals compatible with the geological record (Hattori et al., 2013). The study of Hattori et al. (2013) was made possible by a measurement (Danielache et al., 2012) of isotopic SO_2 B^1B_1-X^1A_1 absorption spectrum with a spectral resolution of 8 cm^{-1}. A subsequent study presented by Whitehill et al. (2013) showed that the fate of the photo-exited fragment (SO_2^*) is not direct oxidation, as assumed in the Hattori et al. (2013) study, but follows a branching mechanism due to quenching. In order to revise the origin of the isotopic fractionation during the oxidation of photo-exited fragments, we started addressing the source of the larger expected isotopic effect from the absorption cross-sections of $^{3x}SO_2$ in the B^1B_1-X^1A_1 band.

Noise interference from various sources is unavoidable when conducting the spectrum measurement. And it is almost impossible to eliminate their influence on the final spectrum. Previous spectrum studies used methods such as averaging several sets of measurements and reducing the resolution of low signal-to-noise ratio (SNR) regions to obtain a relatively low-noise spectrum. This study provides a new idea on data processing by correcting the linear relationship between the absorbance and sample concentration according to the Beer-Lambert law using the least absolute deviation (LAD) method to exclude the effect of random noise on the data fit.

The spectrum resolution in this study is 0.4 cm^{-1} and is 20 times higher than the previous study (Danielache et al., 2012). Instrument optics and Fast Fourier Transform (FFT) setups were optimized for high-resolution measurements. The UV absorption cross-section of each SO_2 sulfur stable isotopologue was presented and the isotopic effect was also discussed. In addition, the raw interferograms were recorded, where the FFT can be applied to generate lower-resolution spectrums. Therefore, the relationship between spectrum resolution and the isotopic effect was also discussed.

References

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