Mass-independent fractionation of sulfur isotopes studied by 1-D photochemical model simulation

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Introduction

Mass-independent fractionation of sulfur isotopes (MIF-S) found in the geological record carries potential information on local atmospheric conditions during the Archean period. A substantial amount of research has been conducted to unravel the mechanism behind the source of the MIF-S found in the geological record. Among the latest reports Endo *et al*. [1] has demonstrated that SO_2 photolysis replicates the sulfur isotopic Archean line. Danielache *et al*. [2] has reported ultraviolet absorption spectrum of SO isotopologues and showing to have a significant the isotope effect.

Model Development

For understanding MIF-S in the Archean atmosphere, we developed a vertical one-dimentional photochemical model that takes into account stable isotopes and high-resolution absorption spectrum. Equation 1, shows that photolysis rates J_i are dependent on atmospheric opacity.

$$J_i = \int_{\lambda_1}^{\lambda_2} q_i(\lambda)\sigma_i(\lambda)I(\lambda)e^{-\tau_z(\lambda)}d\lambda \quad (1)$$

where $I(\lambda)e^{-\tau_z(\lambda)}$ is solar flux at altitude z attenuated by atmospheric opacity. The main feature of this model is that the opacity term can be calculated for each time step and the spectral resolution can be adjusted as needed. This new model has made possible simulation of photolytic processes at isotopic level in the atmosphere more accurately than conventional Archean atmosphere models.

As a verification of the model we simulated SO_2 photolysis. Our model was able to reproduce Endo *et al*. [1] the $\Delta^{33}S$ versus $\delta^{34}S$, and $\Delta^{36}S$ versus $\Delta^{33}S$ values for the unshielded solar flux at the top of the atmosphere. We also simulated SO_2 and SO photolysis considered four sulfur stable isotopes. Isotope effect of SO was observed, but was smaller than the SO_2 photodissociation induced MIF.

[1] Endo *et al*. (2015) *J. Geophys. Res-Atoms*. **120**, 2546-2557. [2] Danielache *et al*. (2014) *J. Chem. Phys*. **140**, 044319.