

SOの紫外線スペクトルから見積もられた非質量依存同位体効果 NMD Fractionation Estimated from SO Isotopologues Photolysis UV Spectra

Danielache Sebastian^{1*}, 鈴木 智也¹, 南部 伸孝¹, 上野 雄一郎¹
Sebastian Danielache^{1*}, Tomoya Suzuki¹, Shinkoh NANBU¹, Yuichiro Ueno¹

¹ 上智大学 理工学部 物質生命理工学科, ² 東京工業大学 大学院 理工学研究科 地球惑星科学専攻

¹Department of Materials & Life Sciences, Faculty of Science & Technology, Sophia University, ²Department of Earth and Planetary Sciences, Tokyo Institute of Technology

Understanding the mechanism of sulfur isotopic fractionation phenomena has been used for some time as a tool to the understanding of reducing atmospheres. Sulfur Non-Mass Dependent (NMD) fractionation signals reported for the Archean and Early Proterozoic (>2300 Ma) atmosphere where the photodissociation of sulfur bearing species play a significant role since the concentration of oxygen is estimated to be 10-5 times present atmospheric levels and therefore ultraviolet light permeates throughout the entire atmosphere, however the underlying mechanisms are not fully understood. In order to explain the NMD signal preserved in the geological record other than SO₂ photodissociation chemistry of sulfur compound should be taken into account. In this study we consider isotopic fractionation during photodissociation of SO. Experimental studies are difficult since SO is highly reactive and unstable under most experimental and atmospheric conditions. Consequently, theoretical studies are suitable to evaluate its spectroscopic properties. We report ultraviolet absorption cross sections for ^{32,33,34,36}SO isotopologues. R-matrix theory was employed to compute absorption cross sections. Potential energy curves were calculated at multi-reference correlation interaction (MRCI) method with augmented correlation consistent polarized valence sextuple-z (aug-cc-pV6Z) basis set. The six lower-lying electronic states were explored. Because there are pseudo-crossings between electronic excited states, non-adiabatic effect has been taken into account. Isotopic fractionation constants were theoretically estimated at different temperatures by assuming that the photolysis rates are proportional to the photoabsorption cross sections for each isotopologues. We make the one box model of the atmospheric chemistry where obtained photolysis rates are utilized, and discuss the results of this model.

キーワード: 硫黄循環, 古大気, 安定同位体

Keywords: Sulfur Cycle, Archean Atmosphere, Stable Isotopes