

論文 / 著書情報  
Article / Book Information

題目(和文)	中層大気におけるオゾン同位体濃縮のグローバル分布：反転解析と観測
Title(English)	Global distribution of ozone isotopic enrichment in the middle atmosphere: Retrieval and observation
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種別(和文)	論文要旨
Type(English)	Summary

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## 論文要旨

THESIS SUMMARY

専攻 : Department of	化学環境学	専攻	申請学位 (専攻分野) : Academic Degree Requested	博士 (理学)	Doctor of
学生氏名 : Student's Name	佐藤 知紘		指導教員 (主) : Academic Advisor(main)	吉田 尚弘	
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

Oxygen is an important element in the planets of our solar system. Ozone, which is composed by three oxygen atoms, has a unique and key role for the earth system. In the earth's atmosphere, ozone has the largest oxygen isotopic enrichment that is a good historical tracer for chemical and physical processes. Ozone isotopic enrichments are measured to be increased (10-20%) with altitude in the lower to middle atmosphere (< 40 km) and in the northern mid-latitude. Isotopic enrichment in asymmetric heavy ozone ( $^{18}\text{O}^{16}\text{O}^{16}\text{O}$  and  $^{17}\text{O}^{16}\text{O}^{16}\text{O}$ ) showed 1.5-2 times larger values than those in symmetric ( $\text{O}^{18}\text{O}^{16}\text{O}$  and  $\text{O}^{17}\text{O}^{16}\text{O}$ ) in the lower and middle stratosphere. These measurement values are theoretically explained, but there is a lack of understanding of oxygen isotopic ratio in the upper atmosphere due to the difficulty of measurement. No global distribution of oxygen isotopic ratio in ozone was confirmed. Reveal of ozone isotopic distributions and its behavior of seasonal-diurnal variation in the middle and upper atmosphere is important to provide a fundamental knowledge of the earth atmospheric system.

The purpose of this thesis is to understand a feature and mechanism of global distribution of ozone isotopic enrichment in the stratosphere and the lower mesosphere. The limb-emission spectra acquired by the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) were used for this study. I developed the optimized retrieval algorithm for ozone isotopic ratio by SMILES (TOROROS) that includes (1) an a priori covariance matrix constrained by oxygen isotopic ratios in ozone, (2) an optimization of spectral windows for ozone isotopomers and isotopologues, and (3) an unification of retrieval altitude grid and input parameters for all windows. In the mid-latitudes (40°S-20°S, 20°N-40°N), the TOROROS  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  of about 16% agreed with past measurements between 30 and 40 km. The vertical profile of  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  showed an increase and a decrease with altitude in the stratosphere and mesosphere, respectively. The  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  peak, of about 20%, is located at the stratopause. The total systematic error in  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  was estimated to be about 5% and 7% in the stratosphere and the mesosphere, respectively. The largest error source is uncertainty in the air-broadening parameter ( $\gamma_{\text{air}}$ ) of the  $^{18}\text{O}^{16}\text{O}^{16}\text{O}$  transition.

The global distribution of  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  was estimated by isotopic fractionation of the two major reactions of ozone production and loss:  $\text{O} + \text{O}_2 + \text{M} \rightarrow \text{O}_3 + \text{M}$ ,  $\text{O}_3 + h\nu \rightarrow \text{O} + \text{O}_2$ . Temperature dependence of  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  in the three-body formation reaction was estimated from the nighttime observation at 32-52 km in the mid-latitudes. The observed  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  were fitted to a linear function of temperature with subtracting photo-induced isotopic fractionation effect. This linear approximation well explained the  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  distribution in the mid-latitudes as well as in the equator region (20°S-20°N). The  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  was underestimated in the higher latitudes (70°S-40°S, 40°N-70°N), which might indicates another mechanisms to affect ozone isotopic enrichment. Diurnal variation of  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  obtained by TOROROS showed that daytime  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  was increased with about 2-4% in the middle stratosphere, while no significant variations were observed in the upper stratosphere. The estimated  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  reproduced the diurnal variations at 37, 42 and 47 km, however, was smaller in daytime at 32 km.

These behaviors and trends in the vertical profile and diurnal variation of  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  were also observed in  $\delta^{17}\text{O}^{16}\text{O}^{16}\text{O}$ . Asymmetric heavy ozone in the stratosphere showed mass-independent fractionation (MIF). The MIF value of 6% and 2% was obtained in the daytime and nighttime, respectively, in the middle stratosphere, and was constant with 6% through the day in the upper stratosphere. No such an MIF value was observed in the lower mesosphere.

This is the first observation of the  $\delta^{18}\text{O}^{16}\text{O}^{16}\text{O}$  global distribution from the middle stratosphere to the lower mesosphere as well as  $\delta^{17}\text{O}^{16}\text{O}^{16}\text{O}$  and MIF in asymmetric heavy ozone. TOROROS also showed an opportunity of remote-sensing observation toward smaller and more precise intramolecular scale such as isotopic ratio.

備考 : 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).