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Global distribution of ozone isotopic enrichment in the middle atmosphere: Retrieval and observation

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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.

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 (TORORES) 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.

The vertical profile of the TORORES δ^{18}OOO showed an increase and a decrease (10~20%) with altitude in the stratosphere and mesosphere, respectively. The δ^{18}OOO peak, of about 20%, was located at the stratopause. The values and behaviors of the δ^{18}OOO are consistent with the past measurements and theoretical predictions in the stratosphere. The total systematic error in δ^{18}OOO 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 (γ_{air}) of the ^{18}OOO transition.

This is the first observation of the δ^{18}OOO distribution from the middle stratosphere to the lower mesosphere. 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.