Vertical distribution of stratospheric N₂O isotopocules and its temporal variation over Japan and Antarctica

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Nitrous oxide (N₂O) is a greenhouse gas in the troposphere and a major ozone-depleting gas in the stratosphere. However, it has value as a tracer of stratospheric circulation and chemical processes because known sources of N₂O are limited to natural and anthropogenic processes occurring in the lowermost troposphere and major sinks of N₂O are UV photolysis and reaction with O(¹D) (photo-oxidation) in the stratosphere. The ratio of isotopocules, isotopically substituted molecules, changes during the photolysis and photooxidation and the mixing of air masses with different origin, making it a useful indicator to estimate the relative contribution of the two decomposition processes and the mixing effect. Earlier studies on the vertical and latitudinal distribution of stratospheric N₂O isotopocule ratios showed that the magnitude of the apparent isotopocule fractionation (ϵ value) depends on altitude and latitude. To clarify the causes of variation in the apparent ϵ values, we analyzed the vertical profiles of isotopocules and their temporal variations obtained at fixed points in Japan and Antarctica over a period of 30 and 15 years, respectively.

The relationship between N₂O mixing ratio and isotopocule ratio shows little difference between sites and time of year in the lower stratosphere (< ~25 km). In the middle stratosphere (> ~25 km), however, the ε values over Japan were higher in Aug-Sep than in May and the ε values over Antarctica (Dec-Jan) were close to the former, suggesting that the effects of mixing associated with transport are relatively smaller in the summer months than in the spring. On the other hand, the relationship between isotopocule ratios suggested an increase of contribution from the photo-oxidation to N₂O decomposition in the lower stratosphere.