Validation and Comparison of Tropospheric Column Ozone Derived from GOME Measurements with Ozonesondes over Japan

Katsuyuki Noguchi[1], Naomi Urita[1], Sachiko Hayashida[1], Xiong Liu[2], and Kelly Chance[2]
[1]*Faculty of Science*, *Nara Women's University*, *Nara*, *Japan*
[2]*Atomic and Molecular Physics Division*, *Harvard-Smithsonian Center for Astrophysics*, *Cambridge*, *Massachusetts*, *USA*

Abstract

Tropospheric column ozone derived from Global Ozone Monitoring Experiment (GOME) measurements (GOME-O_3) was validated using operational ozonesonde measurements over Japan and compared with Tropospheric Ozone Residual (TOR) derived from Total Ozone Mapping Spectrometer and Solar Backscatter Ultraviolet measurements. The GOME-O_3 validation showed positive biases with a magnitude of less than 3 DU (∼10%) and random errors of 5−9 DU (∼15−30%) at collocated sonde stations. GOME-O_3 showed better agreement with ozonesonde measurements over Japan than TOR did. The direct comparison between GOME-O_3 and TOR showed the underestimation of TOR during winter and spring in the southern part of Japan (around 30°N).

1. Introduction

Tropospheric ozone plays an important role in governing oxidation processes in the troposphere (Brasseur et al. 1999). Photodissociation of ozone and subsequent reactions with water vapor are a source of hydroxyl radicals that regulate the chemical lifetime of various chemical species in the troposphere. Moreover, tropospheric ozone is a major air pollutant and a greenhouse gas.

The increase in tropospheric ozone is of great concern in East Asia where anthropogenic emissions of ozone precursors have dramatically increased following rapid economic growth in the region (Akimoto et al. 1994). For example, observations from the Global Ozone Monitoring Experiment (GOME) show that nitrogen dioxide (NO_2), one of the most important ozone precursors, is increasing over East Asia (Richter et al. 2005). Because Japan is downwind of the Asian Continent, it is greatly affected by the emission of pollutants from the Asian Continent, and monitoring the behavior of tropospheric ozone around Japan is highly necessary.

Satellite-based observations can be used to monitor atmospheric environments. However, it is usually difficult to observe minor constituents such as ozone in the troposphere using satellite-borne sensors because of thick middle-atmospheric ozone and tropospheric clouds. Fishman and Larsen (1987) and Fishman et al. (1990) proposed the Tropospheric Ozone Residual (TOR) method to derive global distributions of tropospheric column ozone (TCO) from satellite observations. TOR is the difference between total column ozone derived from the Total Ozone Mapping Spectrometer (TOMS) and stratospheric column ozone derived from other sensors that measure vertical profiles of stratospheric ozone. Fishman et al. (1990) used Stratospheric Aerosol and Gas Experiment (SAGE) data to estimate stratospheric column ozone, and subtracted it from the total column ozone to derive TCO. They used Solar Backscatter UltraViolet (SBUV) data with empirical correction in subsequent studies (Fishman et al. 2003) due to the far greater spatial coverage of SBUV observations.

Liu et al. (2005) recently applied another approach to derive the global distribution of tropospheric ozone directly. They successfully retrieved vertical profiles of ozone directly from the spectroscopic measurements of GOME after applying extensive wavelength and radiometric calibrations and improvements in forward model inputs and radiative transfer modeling. They obtained global distributions of GOME-based TCOs (hereafter called "GOME-O_3") that were computed by integrating ozone in the vertical from the surface to the tropopause. An advantage of GOME-O_3 is that it can be compared to other species (e.g., NO_2) that are simultaneously observed by GOME. Studies on the relationships between ozone and its precursors can therefore be undertaken.

Liu et al. (2005) validated their results globally but did not focus on the region around Japan, the area of interest in the present study. An intensive validation study would be necessary to detail the behavior of tropospheric ozone around Japan with a focus on the relationship between ozone and anthropogenic emissions of precursors over East Asia. Ozonesonde measurements in Japan taken operationally at four stations (Sapporo, Tsukuba, Kagoshima, and Naha) are subject to strict quality control, and are thus highly appropriate for validating satellite data. Data from all four stations are considered to cover the region around Japan. Liu et al. (2005) did not use data from Sapporo, the northernmost station in Japan.

The present study also compared the GOME-O_3 dataset with the TOR dataset (Fishman et al. 2003), which has been widely used to investigate global tropospheric ozone behavior, to examine the similarities and differences between them.

2. Data

GOME is a nadir-viewing spectrometer with four channels in the ultraviolet and visible parts of the electromagnetic spectrum. Liu et al. (2005) retrieved ozone vertical profiles using backscattered radiance spectra measured by GOME at 289−339 nm and the optimal estimation technique that utilizes a priori information based on SBUV V8 climatology (McPeters et al. 2003). The spatial resolution of GOME-O_3 is normally 960 × 80 km^2. The O_3 is derived from measurements in Channel 1a (< 307 nm), which has pixel size 960 × 80 km^2 and Channel 2, which has pixel size 320 × 40 km^2. The GOME-O_3 dataset is provided as a swath of data for every orbit. Global mean biases and standard deviations of GOME-O_3 relative to ozonesonde observations were usually within 3 DU (15%) and 3−8 DU (13−27%), respectively (Liu et al. 2005). We used GOME-O_3 observations from 1996 to 1999 in the present study.

The TOR dataset used in the present study was a residual obtained by subtracting SBUV-based stratospheric column ozone from TOMS-based total column ozone (Fishman et al. 2003). The TOR dataset is

Corresponding author: Katsuyuki Noguchi, Faculty of Science, Nara Women's University, Kita-uoya Nishi-machi, Nara 630-8506, Japan. E-mail: nogu@ics.nara-wu.ac.jp.

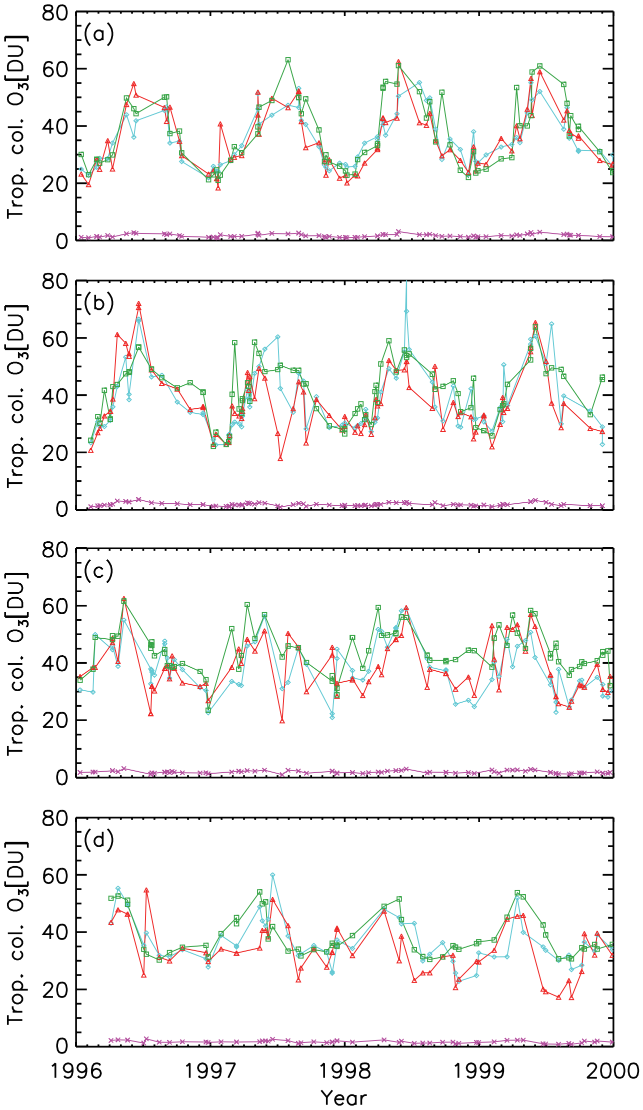

Fig. 1. Time series of a priori values of GOME-O_3 (green), retrieved values of GOME-O_3 (blue), and ozonesonde TCO (red) at (a) Sapporo, (b) Tsukuba, (c) Kagoshima, and (d) Naha. The pink crosses, which are the 5% values of the ozonesonde TCO, roughly show the measurement errors of ozonesonde.

provided as monthly averages that are gridded to 1.25° longitude by 1° latitude pixels. The SBUV-based TOR accuracy was estimated to be 4.0 DU (~13%) (Fishman et al. 2003).

The ozonesonde data used in the present study were provided by the Japan Meteorological Agency (JMA). Ozonesondes are launched from four sonde stations, Sapporo (43.1°N, 141.3°E), Tsukuba (36.1°N, 140.1°E), Kagoshima (31.6°N, 130.6°E), and Naha (26.2°N, 127.7°E), and measure profiles with vertical resolutions of less than 100 m (JMA 2002). The ozonesonde instruments were the RS2-KC79 (Kobayashi and Toyama 1966), and its successor the RS2-KC96. The relative biases and standard deviations of the concentration measurements in the troposphere were estimated to be −(2–7) % and ± (4–6) %, respectively (Smit and Kley 1998).

To derive TCO from a vertical profile of ozone, we need the information on tropopause heights. The present study used the NCEP-based tropopause heights (Kalnay et al. 1996) for GOME-O_3 and ozonesonde measurements as in Liu et al. (2005). Since TOR also utilized

Table 1. Biases (B) and random errors (σ) of a priori values of GOME-O_3 and those of retrieved values of GOME-O_3, in DU. Also shown are the correlation coefficients (R) between the a priori and the ozonesonde measurements and between the GOME-O_3 and the ozonesonde measurements.

Station	$B_{apriori}$	B_{gome}	$\sigma_{apriori}$	σ_{gome}	$R_{apriori}$	R_{gome}
Sapporo	1.9	0.2	6.9	5.3	0.81	0.86
Tsukuba	3.7	1.1	8.7	8.5	0.68	0.74
Kagoshima	6.2	0.01	8.1	8.0	0.54	0.61
Naha	5.0	2.7	7.9	6.8	0.54	0.68

the NCEP-based tropopause heights (Fishman et al. 2003), the definition of the tropopause is unified for all the TCO data throughout the present study.

Difference of tropopause height between NCEP and ozonesonde measurement will cause a different validation result. We carefully estimated the effect of different tropopause heights on the result by comparing the ozonesonde TCO values derived from the ozonesonde- and NCEP-based tropopause heights at the four stations. The estimation showed that the magnitudes of the biases and the standard deviations of the differences between the two TCOs were 0.2–2.3 DU and 3.6–6.5 DU, respectively.

3. Validation of GOME-O_3

Cases were chosen such that a sonde station was within a swath pixel from GOME-O_3, the cloud fraction of the GOME measurement was less than 0.8, and the GOME measurement and ozonesonde measurement fell on the same day. Because GOME records observations at ~10:30 local time and the ozonesonde takes measurements between 14:00 and 16:00 Japan Standard Time, a time lag of several hours exists between GOME and ozonesonde measurements. The numbers of cases (N) considered for each station were 78 (Sapporo), 90 (Tsukuba), 78 (Kagoshima), and 55 (Naha).

TCOs of the *a priori* GOME-O_3, the retrieved GOME-O_3, and the ozonesonde measurements were denoted as $I_{apriori}$, I_{gome} and I_{sonde}, respectively. Biases in I_{gome}, denoted as B_{gome}, were defined at each sonde station during the observation period (1996–1999) as $B_{gome} = \sum_{i=1}^{N} \Delta I^i_{gome}/N$, where $\Delta I^i_{gome} = I^i_{gome} - I^i_{sonde}$. Random errors in I_{gome} at each sonde station during the observation period, denoted as σ_{gome}, were defined as the standard deviations of ΔI_{gome}. As for the case of B_{gome} and σ_{gome}, biases of $I_{apriori}$ were calculated as $B_{apriori} = \sum_{i=1}^{N} \Delta I^i_{apriori}/N$, where $\Delta I^i_{apriori} = I^i_{apriori} - I^i_{sonde}$, and the standard deviations of $\Delta I_{apriori}$ were denoted as $\sigma_{apriori}$. The retrieved profile will show improvement over the a priori vertical profile if the measurement contains significant ozone information. Statistically, B_{gome} and σ_{gome} are smaller than $B_{apriori}$ and $\sigma_{apriori}$, respectively, if the measurement contains significant ozone information.

Figure 1 shows the time series of $I_{apriori}$, I_{gome}, and I_{sonde} at each sonde station. Seasonal variations in TCOs are more distinct for more northerly latitudes (i.e., Sapporo and Tsukuba) than for southerly latitudes (i.e., Kagoshima and Naha). Day-to-day fluctuations are superimposed on the seasonal variations. Offsets and such day-to-day fluctuations are more important for validating I_{gome} than patterns in the seasonal variations because a priori information already gives basic seasonal variations in TCO. The correlation coefficients between $I_{apriori}$ and I_{sonde} ($R_{apriori}$) and between I_{gome} and I_{sonde} (R_{gome}) at each sonde station are useful to check whether I_{gome} could correctly observe the day-to-day fluctuations of TCO.

The significance of the GOME retrievals can be checked by comparing B_{gome} with $B_{apriori}$, σ_{gome} with $\sigma_{apriori}$,

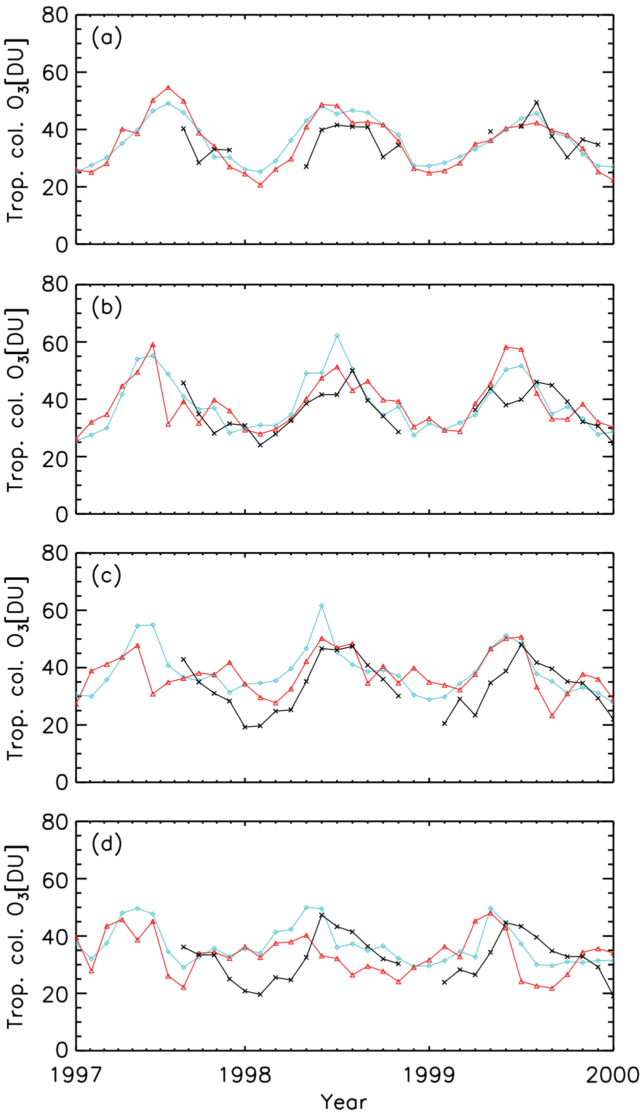

Fig. 2. Time series of monthly means of the retrieved values of GOME-O_3 (blue), TOR (black), and ozonesonde TCO (red) at (a) Sapporo, (b) Tsukuba, (c) Kagoshima, and (d) Naha.

and R_{gome} with $R_{apriori}$ at each sonde station. Table 1 summarizes $B_{apriori}$, B_{gome}, $\sigma_{apriori}$, σ_{gome}, $R_{apriori}$, and R_{gome}. Table 1 shows that B_{gome} is half or less than half of $B_{apriori}$. The σ_{gome} is also smaller than $\sigma_{apriori}$, suggesting an improvement, albeit small, over a priori values. Retrievals do add information to the a priori values. The result that R_{gome} at all sonde stations is better than $R_{apriori}$ also suggests the significance of the GOME retrievals. The retrievals of I_{gome} are deemed successful not only at Tsukuba, Kagoshima, and Naha but also at Sapporo, which was not used in Liu et al. (2005).

The B_{gome} is positive with a magnitude of 0.01–2.7 DU (~0–10%) and σ_{gome} of 5.3–8.5 DU (~15–30%) as shown in Table 1. Among the four sonde stations, Sapporo shows relatively good agreement of GOME-O_3 with ozonesonde measurements. The B_{gome} and σ_{gome} over Japan are consistent with the global means in Liu et al. (2005). The GOME-O_3 and the ozonesonde TCO data are close to the one-to-one correlation line in their scatter diagrams (not shown). The possible sources of the positive biases of GOME-O_3 include 1) an incomplete discrimination between the tropospheric ozone and the ozone just above the tropopause due to a coarse vertical resolution of the GOME measurements and the sharp increase of ozone above the tropopause, and 2) the underestimation of the JMA ozonesonde measurements (see section 2) which would cause positive biases in GOME-O_3 larger than the true biases. In addition, the spatial-temporal variability of tropospheric ozone, the coarse spatial resolution of GOME (960 × 80 km^2), and the time differences between the GOME and ozonesonde measurements can also contribute to the increases of B_{gome} and σ_{gome}.

Table 2. Biases ($\bar{B}$) and random errors ($\bar{\sigma}$) of the monthly means of retrieved values of GOME-O_3 and those of TOR, in DU. Correlation coefficients ($\bar{R}$) between the GOME-O_3 and the ozonesonde measurements and between the TOR and the ozonesonde measurements are also shown.

Station	$\bar{B}_{gome}$	$\bar{B}_{tor}$	$\bar{\sigma}_{gome}$	$\bar{\sigma}_{tor}$	$\bar{R}_{gome}$	$\bar{R}_{tor}$
Sapporo	0.7	−2.7	2.9	6.8	0.95	0.41
Tsukuba	−0.04	−3.1	5.5	7.5	0.83	0.54
Kagoshima	0.9	−4.1	6.7	7.6	0.61	0.58
Naha	3.0	−0.5	5.9	11.7	0.64	−0.27

4. Comparison between GOME-O_3 and TOR

We compared GOME-O_3 and TOR with ozonesonde measurements at the four sonde stations before the direct comparison between GOME-O_3 and TOR. To conform the spatial and temporal resolution of GOME-O_3 and ozonesonde TCO to that of TOR, denoted as $\bar{I}_{tor}$, the GOME-O_3 and ozonesonde TCO were gridded to a 1.25° longitude by 1° latitude pixel and TCOs at the pixels that included the sonde station location were averaged for each month to create $\bar{I}_{gome}$ and $\bar{I}_{sonde}$. The GOME measurements were used when the cloud fraction was less than 0.8 as in section 3. Day-to-day overlapping of observations among the datasets was required in section 3, but not in this section.

Biases in $\bar{I}_{gome}$ and $\bar{I}_{tor}$ at the gridded pixels over the sonde stations during the observation period (1997–1999) were defined as $\bar{B}_{gome} = \overline{\Delta\bar{I}_{gome}}$ and $\bar{B}_{tor} = \overline{\Delta\bar{I}_{tor}}$, respectively, where $\Delta\bar{I}_{gome} = \bar{I}_{gome} - \bar{I}_{sonde}$ and $\Delta\bar{I}_{tor} = \bar{I}_{tor} - \bar{I}_{sonde}$. A long bar over a symbol denotes uniform operation during the observation period. Random errors $\bar{\sigma}_{gome}$ and $\bar{\sigma}_{tor}$ at the gridded pixels over the sonde stations during the observation period were defined as the standard deviations of $\Delta\bar{I}_{gome}$ and $\Delta\bar{I}_{tor}$, respectively. The correlation coefficients between $\bar{I}_{gome}$ and $\bar{I}_{sonde}$ ($\bar{R}_{gome}$) and between $\bar{I}_{tor}$ and $\bar{I}_{sonde}$ ($\bar{R}_{tor}$) were also calculated as section 3.

Figure 2 shows the time series of $\bar{I}_{gome}$, $\bar{I}_{tor}$, and $\bar{I}_{sonde}$ at each sonde station. TOR shows poor agreement with ozonesonde measurements, especially at Kagoshima and Naha. The largest disagreement is that $\bar{I}_{tor}$ during winter and spring at Kagoshima and Naha is consistently 10–15 DU smaller than $\bar{I}_{sonde}$ and even seems to be out of phase with $\bar{I}_{sonde}$.

Table 2 summarizes $\bar{B}_{gome}$, $\bar{B}_{tor}$, $\bar{\sigma}_{gome}$, $\bar{\sigma}_{tor}$, $\bar{R}_{gome}$, and $\bar{R}_{tor}$. Table 2 shows that $\bar{B}_{gome}$ and $\bar{B}_{tor}$ are −0.04–(+3.0) DU and −4.1–(−0.5) DU, respectively. The magnitudes of $\bar{B}_{gome}$ are smaller than those of $\bar{B}_{tor}$ at all the stations except Naha. The smallness of $\bar{B}_{tor}$ at Naha, however, does not seem to be significant since the seasonal variation of TOR does not agree with that of ozonesonde measurements as is shown in Fig. 2d. The $\bar{\sigma}_{gome}$ and $\bar{R}_{gome}$ are better than $\bar{\sigma}_{tor}$ and $\bar{R}_{tor}$, respectively, at all the sonde stations. These results suggest that GOME-O_3 shows better agreement with ozonesonde measurements over Japan than TOR does.

Figure 3 is a direct comparison between GOME-O_3 and TOR, showing the residuals of $\bar{I}_{gome}$ minus $\bar{I}_{tor}$ around Japan (120°E–150°E, 20°N–50°N). Since the TCO in this region had a stronger dependence on latitude than longitude (not shown), we zonally averaged the residuals along latitudinal pixels between 120°E and 150°E in a given month to see the month-latitude dependence of $\bar{I}_{gome}$ minus $\bar{I}_{tor}$. Figure 3 shows that positive residuals exist during winter and spring in the southern part of Japan (around 30°N) and increase up to ~16 DU. This result suggests the overestimation of $\bar{I}_{gome}$ or the underestimation of $\bar{I}_{tor}$. By the fact that $\bar{I}_{tor}$ during winter and spring at Kagoshima and Naha was systematically smaller than ozonesonde measurements as was shown in Fig. 2c and 2d, we can infer that the residuals are attributed to the underestimation of TOR rather than the overestimation of GOME-O_3.

The underestimation of TOR may be explained by an overestimation of the stratospheric ozone which is used in the derivation process of TOR. Figure 11 of Wozniak et al. (2005) showed that the SBUV-based stratospheric ozone utilized in the TOR derivation was overestimated during winter and spring at Kagoshima and Naha compared to the stratospheric ozone derived from ozonesonde/ground-based measurements. The present study suggests that the overestimation of stratospheric ozone in the TOR derivation shown in Wozniak et al. (2005) is not limited to Kagoshima and Naha but is a general feature in the southern part of Japan (around 30°N). The TOR dataset in this region may need to be used carefully.

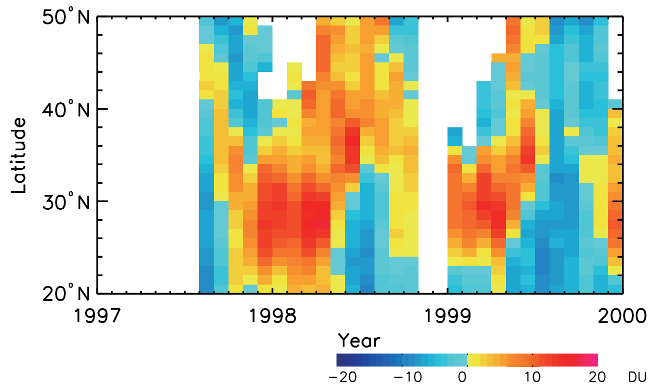

Fig. 3. Month-latitude dependence of the residuals of GOME-O_3 minus TOR. The residuals are zonally averaged between 120°E–150°E.

5. Conclusions

We used operational ozonesonde measurements, recorded at four stations in Japan, to validate a tropospheric column ozone dataset derived from GOME measurements (GOME-O_3). The results at the stations show that biases were positive with a magnitude of less than 3 DU (~10%); random errors were 5–9 DU (~15–30%). The present study also compared GOME-O_3 and Tropospheric Ozone Residual (TOR) with ozonesonde measurements. GOME-O_3 had smaller magnitudes of biases and random errors than TOR and showed a better correlation with ozonesonde measurements. Especially, TOR shows poor agreement with ozonesonde measurements at Kagoshima and Naha. The direct comparison between GOME-O_3 and TOR by using the zonal averages of the residuals of GOME-O_3 minus TOR between 120°E and 150°E showed the underestimation of TOR during winter and spring in the southern part of Japan (around 30°N). GOME-O_3 was demonstrated to be a useful dataset for studying tropospheric ozone near Japan. GOME-O_3 will contribute to better understanding the relationship between ozone and its precursors in the region.

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