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1 Study of lower tropospheric ozone over central and eastern China:
2 Comparison of satellite observation with model simulation

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1 **Abstract**

2 The lower tropospheric ozone enhancement over Central and Eastern China (CEC) was
3 reported by Hayashida et al. (2015) using the Ozone Monitoring Instrument (OMI)
4 multiple-layer product retrieved by Liu et al. (2010), which first showed the lower
5 tropospheric ozone enhancement from ultraviolet and visible (UV-Vis) spectra
6 measurements from space. However, to clarify the enhancement in the concentration of
7 the lowermost ozone using spaceborne measurements, it is necessary to understand the
8 effect of ozone variation in the upper troposphere and lower stratosphere (UT/LS),
9 because of large smoothing errors in the retrieval scheme. In this study, a scheme was
10 developed to eliminate the artificial effect of UT/LS ozone enhancement on lower
11 tropospheric ozone retrieval using OMI. By applying the UT/LS screening scheme for
12 June 2006, we removed the artificial effect of the UT/LS ozone enhancement on the
13 lower tropospheric ozone. Even after UT/LS screening, we were able to show a clear
14 enhancement in the lower tropospheric ozone over CEC in June 2006 and confirmed the
15 conclusion derived by Hayashida et al. (2015). To clarify the reason for ozone
16 enhancement in June, the effects of emissions from open crop residue burning (OCRB)
17 in the North China Plain on lower tropospheric ozone were also examined using a
18 comparison with model simulations. On the scale of the vertical resolution of OMI
19 observations, the effect of OCRB on ozone enhancement does not seem to be significant,
20 although it may be more significant when focusing on ozone in the planetary boundary
21 layer.

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23

1 **1 Introduction**

2

3 In recent years, anthropogenic ozone (O₃) pollution has become a serious environmental
4 problem all over the world (e.g., Ordonez et al. 2005; Lefohn et al. 2010; Langner et al.
5 2012), and such hazardous air pollution events over large cities in China are now a
6 particularly great concern (e.g., Wang et al. 2009; Verstraeten et al. 2015). According to
7 the Regional Emission inventory in ASia (REAS), emissions of O₃ precursors in the
8 sectors of industry and transportation are most notable in Central and Eastern China
9 (CEC) (Ohara et al. 2007; Kurokawa et al. 2013).

10 Satellite measurements have played an increasingly important role in O₃
11 monitoring globally (e.g., Burrows et al. 2011 and references therein). However, vertical
12 discrimination of O₃ in the lower troposphere has been a big challenge for satellite-borne
13 measurements, because 90% of the total O₃ amount exists in the stratosphere. Recently,
14 Liu et al. (2010) developed an algorithm for retrieving O₃ profiles using the UV
15 radiances observed by the Ozone Monitoring Instrument (OMI). They retrieved ozone
16 profiles from the ground upward to about 60 km in 24 layers. There are 4 to 7 layers in
17 the troposphere, depending on the tropopause height. The lowermost layer, the 24th
18 layer, corresponds to about 0–3 km above the surface, although its thickness depends on
19 meteorological conditions. Hayashida et al. (2015) closely analyzed the OMI products
20 with multiple layers and revealed a significant O₃ enhancement in the lowermost layer
21 (the 24th layer) over CEC, which is most notable in June each year. That was the first
22 systematic view from satellite observation with ultraviolet spectra showing the ozone
23 enhancement in the lowermost altitude over CEC. Further comparative studies, along

1 with model simulations, are expected to clarify any unknown factors in ozone production
2 and transport mechanisms.

3 However, the effect of a large variability in O₃ amount in the upper troposphere
4 and lower stratosphere (UT/LS) on the OMI ozone retrieval must be taken into
5 consideration carefully because of the large smoothing error of the OMI retrieval scheme
6 (Liu et al. 2010). The large O₃ variability in UT/LS may exert an influence on the
7 lowermost tropospheric O₃. From this standpoint, the data selection by Hayashida et al.
8 (2015) should be reexamined, because the O₃-enhanced areas over CEC are often
9 situated near the location of the subtropical jet (STJ), where the intrusion of
10 stratospheric O₃ occurs frequently, as claimed by Nakatani et al. (2012) (see Fig. 5 in
11 Nakatani et al. 2012). For example, Dufour et al. (2015) found a good positive
12 correlation between the concentrations of O₃ and carbon monoxide (CO) in the lower
13 troposphere corresponding to the altitudes of 0–6 km over the North China Plain, both
14 of which were observed by Infrared Atmospheric Sounding Interferometer (IASI); this
15 positive correlation between O₃ and CO suggested the photochemical source of O₃. On
16 the other hand, they also pointed out signals of significant O₃ enhancement in the upper
17 troposphere (6–12 km), which correlated with the low-pressure system, suggesting an
18 effect of O₃ subsidence from the stratosphere. As these studies indicate, East Asia, and
19 CEC in particular, is one of the key regions where both stratospheric O₃ subsidence and
20 anthropogenic O₃ production are occurring actively. In this study, we present a scheme
21 to remove the effect of the O₃ variability in the UT/LS on the retrieval of the lowermost
22 O₃ layer (0–3 km). By applying this scheme, we can confirm the enhancement of the
23 lowermost O₃ every June shown by Hayashida et al. (2015).

1 To investigate the mechanism of repeatable O₃ enhancement in June over CEC
2 (see Fig. 10 of Hayashida et al. 2015), we examine an effect of open crop residue burning
3 (OCRB) in the North China Plain. Kanaya et al. (2013) and related studies of the Mount
4 Tai Experiment (MTX2006) (references in Kanaya et al. 2013) revealed that the
5 emissions from regional-scale OCRB after the harvesting of winter wheat increased the
6 concentration of O₃, together with photochemical aging. However, it is difficult to
7 estimate quantitatively the magnitude of regional-scale emissions from the burning of
8 agricultural waste. For example, the Global Fire Emissions Database (GFED) version 3
9 is a well-known comprehensive emissions inventory of biomass burning (van der Werf et
10 al. 2010) that includes the emissions of NO_x and CO originating from deforestation and
11 the burning of savanna, grassland, woodland, extratropical forest, and agricultural
12 waste and peat (see Table 5 in van der Werf et al. 2010). However, emissions from open
13 crop burning in the North China Plain have not been included in the GFED ver. 3. In
14 this study, we examine the effect of OCRB on O₃ concentration via model simulations
15 involving the OCRB emission inventory by Yamaji et al. (2010) using statistical data of
16 monthly crop residues from each province in China (Yan et al. 2006) and daily hotspot
17 data observed by the global Moderate Resolution Imaging Spectroradiometer (MODIS).
18 We focus on June 2006 in this paper because the enhancement of O₃ is most notable
19 every June, as reported by Hayashida et al. (2015), and the outstanding effect of OCRB
20 on O₃ in the North China Plain was demonstrated in June 2006 during the MTX2006, as
21 mentioned above (Kanaya et al. 2013).

22 In Section 2, we describe the data used in our analysis and the model used for
23 the simulations. In section 3.1, we present the scheme to eliminate the effect of UT/LS

1 ozone enhancement on the lower tropospheric ozone derivation. In Section 3.2, we show
2 a comparison between the satellite observations of O₃ and their precursors, such as
3 carbon monoxide (CO) and nitrogen dioxide (NO₂), and the results of the model
4 simulations. We demonstrate consistency between the observations and the model
5 results and discuss the effect of OCRB on O₃ concentration.

6

7 **2 Satellite data and model**

8

9 2.1 Satellite observation

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11 2.1.1 O₃ profile and NO₂ tropospheric column observed by OMI

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13 OMI is the UV/visible sensor on board the National Aeronautics and Space
14 Administration (NASA) EOS Aura spacecraft, which was launched in July 2004. The
15 satellite is in a Sun-synchronous polar orbit with an equatorial crossing time of 13:45
16 local time (LT). OMI measures backscattered radiances covering a wavelength range of
17 270 to 500 nm. The wavelength range is divided into three channels: UV-1 (270 to 310
18 nm), UV-2 (310 to 365 nm), and visible (350 to 500 nm). OMI has daily global coverage
19 with a spatial resolution of 13 × 24 km for the UV-2 and visible channels, and 13 × 48
20 km for the UV-1 channel.

21 In this study, we utilized the O₃ profiles retrieved by Liu et al. (2010) using the
22 OMI UV spectra from the ground to about 60 km with 24 layers. In the retrieval
23 algorithm developed by Liu et al. (2010), O₃ profiles were retrieved by applying the

1 optimal estimation technique (Rodgers 2000), with climatological mean O₃ profiles by
2 McPeters et al. (2007) as a priori profiles. Hayashida et al. (2015) analyzed the OMI
3 product of multiple layers and suggested the data reliability of O₃ at the lowermost layer,
4 the 24th layer, which corresponds to a layer from about 0 km to 3 km altitude. As in
5 Hayashida et al. (2015), the gridded O₃ data were used after screening by the criteria of
6 effective cloud fraction (ECF) < 0.2 and root mean square (RMS) defined as the root mean
7 square of the ratio of the fitting residual to the assumed measurement error of the UV-2 channel <
8 2.4.

9 We also utilized the NO₂ tropospheric column from OMI, the version 3 release
10 of the OMI NO₂ gridded level-3 (OMNO2d) product (Data DOI:
11 10.5067/Aura/OMI/DATA3007). The retrieval algorithm was described in detail by
12 Bucsela et al. (2013). Although the original OMI NO₂ data are provided with a
13 resolution of 0.25° × 0.25°, they are converted to adjust to the model resolution in the
14 later analysis.

15

16 2.1.2 CO observed by Measurements Of Pollution In The Troposphere (MOPITT)

17

18 The MOPITT instrument was launched on NASA's EOS Terra spacecraft in December
19 1999. The satellite is in a Sun-synchronous polar orbit of 705 km that crosses the
20 Equator at 10:30 LT. MOPITT covers the globe every three days with a spatial
21 resolution of 22 × 22 km. The MOPITT instrument measures at near-infrared (NIR:
22 2.3 μm) and thermal infrared (TIR: 4.7 μm) wavelengths, and CO concentration can be
23 retrieved using multispectral measurements for both the NIR and TIR wavelengths. In

1 this study, we used the CO total column product of version 6 level 3 data,
2 RetrievdCOTotalColumnDay, which are gridded at $1^\circ \times 1^\circ$ and are available at the
3 NASA website (https://eosweb.larc.nasa.gov/project/mopitt/mopitt_table).

4

5 2.2 Model simulation

6

7 2.2.1 Meteorological Research Institute—Chemistry Climate Model (MRI-CCM2)

8

9 MRI-CCM2 is the global chemistry-climate model developed by Deushi and Shibata
10 (2011). The chemistry module includes 90 chemical species with 172 gas-phase reactions,
11 59 photolysis reactions, and 16 heterogeneous reactions. The transport module includes
12 grid-scale transport using a vertically conservative semi-Lagrangian scheme, sub-grid
13 scale convective transport, and turbulent diffusion (Yukimoto et al. 2011). Emissions of
14 trace gases from various sources and dry and wet depositions are included. The
15 horizontal wind field in MRI-CCM2 is forced toward the observed field, JRA-55
16 reanalysis (Kobayashi et al. 2015) wind field, by using a nudging term. The horizontal
17 resolution is about 110 km ($1.125^\circ \times 1.125^\circ$) and the vertical range, which is divided into
18 64 layers, varies from the ground to about 80 km (0.01 hPa).

19 In this study, the MACCity database was used for the anthropogenic emissions
20 of trace gases, although they were taken from EDGAR v2.0 in the original version of
21 MRI-CCM2 (Deushi and Shibata 2011). Vegetative emission of isoprene and terpenes
22 was taken from the Global Emissions Inventory Activity (GEIA) (Guenther et al. 1995),
23 and vegetative emissions of other hydrocarbons and NO from Muller (1992). Emission of

1 NO from soils is taken from Yienger and Levy (1995); emissions of CO and N₂O from
2 soils, from Muller (1992). Emissions of CO, CH₄, and NMHCs from the ocean was based
3 on Brasseur et al. (1998) with the modifications of Horowitz et al. (2003). Emission of
4 NO by lightning was diagnosed at 6-h intervals. The global flash frequency was
5 calculated according to the parameterization of Price and Rind (1992, 1994). The details
6 of the scheme are described in Deushi and Shibata (2011). Emissions from biomass
7 burning used for this study are described in the next section.

8

9 2.2.2 Model experiment on OCRB effect

10

11 To evaluate the effect of OCRB emission on the lower tropospheric O₃ concentration
12 observed by OMI, we conducted two types of experiments: a control run (CNTL) and an
13 OCRB sensitivity study (OCRB). The emissions from biomass burning used in each
14 experiment are shown in Table 1. In the CNTL experiment, GFED ver.3 was used for
15 emissions from biomass burning. In the OCRB experiment, for the region of 7°S–50°N
16 and 70°E–142°E, the biomass burning emissions were replaced with the OCRB emission
17 inventory (Yamaji et al. 2010). Outside of this region, GFED ver. 3 was used, as in the
18 CNTL experiment. The OCRB emission inventory was developed using province-level
19 statistical data based on the bottom-up methodology of Yan et al. (2006) for the typical
20 OCRB season in CEC. To develop the daily gridded OCRB data, the annual emissions
21 from OCRB were allocated to the spatial grid of 0.5° × 0.5° and to each day according to
22 the satellite hotspots and geographical information of the land cover data. For more
23 details, readers are to refer to Yamaji et al. (2010).

1

2 Table 1. Anthropogenic and biomass burning emission inventories

	Control run (CNTL)	Sensitivity study for open crop residue burning (OCRB)
Anthropogenic	MACCity (monthly)* (Lamarque et al. 2010; Garnier et al. 2011)	
Biomass burning	GFED ver.3 (monthly)*	GFED ver.3 + OCRB emission inventory developed by K. Yamaji**

3 *Monthly values were divided by 30 to convert them to daily values for calculations.

4 **See text

5

6 3 Results and discussion

7

8 3.1 UT/LS screening for 24th-layer O₃

9

10 In this section, we describe the method used to screen out the artificial effect of the O₃
11 enhancement in the UT/LS on the O₃ concentration of the 24th layer (0–3 km). In Section
12 3.1.1, we show the variation of O₃ in the UT/LS obtained by the MRI-CCM2 simulation
13 for CNTL, which was related to the low-pressure system. In Section 3.1.2, we evaluate
14 the contribution of the O₃ enhancement in the UT/LS to the 24th-layer O₃ by applying
15 the averaging kernels (AKs) of the OMI retrieval. In Section 3.1.3, we present the
16 scheme to eliminate the cases in which the effect of the UT/LS O₃ on the 24th-layer O₃ is
17 considerably large. The results before and after the UT/LS screening are shown.

18

1 3.1.1 Enhancement of UT/LS O₃ over East Asia related to the low-pressure system

2

3 We examined the O₃ profiles and meteorological fields in East Asia in June 2006, which
4 were simulated by MRI-CCM2. The two different features in the O₃ profiles, i.e.,
5 significant UT/LS O₃ enhancement and significant lower tropospheric O₃ enhancement,
6 were found in June 2006. Here, we show two cases as representative examples. These
7 are the O₃ profiles on June 10 and June 20, 2006, which correspond to the former and
8 latter cases, respectively.

9 Fig. 1a and 1b indicate the O₃ distribution at 200 hPa simulated by MRI-CCM2
10 for CNTL. The sharp gradient in the O₃ concentration, along with the high wind speed,
11 indicate the location of the STJ. To investigate a significant subsidence of stratospheric
12 O₃ over CEC, we show a cross section of ozone along 118.125°E in Fig. 1c. It is clear that
13 O₃ is descending to an altitude of about 10 km over the region at around 30–35°N where
14 it corresponds to the center of ozone enhancement over Shandong (See Fig. 10 of
15 Hayashida et al. 2015). In contrast to the case of June 10, that the STJ shifted to the
16 north of CEC on June 20, 2006, and thus the subsidence of stratospheric O₃ was
17 significant not over CEC but in the northern part of China, as shown in Fig. 1d. The
18 lower tropospheric O₃ was significantly enhanced on June 20 over CEC at around 35°–
19 40°N, but it was not overlapped with the stratospheric O₃ subsidence, as shown in Fig.
20 1d. The longitude–altitude cross sections at 34.205°N in Fig. 1e and 1f indicate more
21 clearly the difference of O₃ distribution between June 10 and June 20 in the lower
22 troposphere. The lower tropospheric O₃ enhancement was clear at around 115–120°E,
23 while the stratospheric O₃ subsidence was not over the area.

1

2 3.1.2 Evaluation of contribution of the UT/LS O₃ enhancement to 24th-layer O₃

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4 To compare the model outputs with the OMI retrievals, the simulated O₃ amounts at the
5 model layers need to be convolved with the AKs of the OMI retrieval as in eq. 1:

$$6 \quad X'_{24} = X_{a,24} + \sum_{i=1}^{24} A(i,24)[X_{m,i} - X_{a,i}], \quad (1)$$

7 where $X_{m,i}$ is the O₃ (Dobson unit; DU) simulated from the model, $X_{a,i}$ is the a priori O₃
8 (DU) at the i^{th} layer corresponding to the i^{th} OMI layer, and $A(i,24)$ is the retrieval AKs
9 of the 24th layer at the i^{th} layer.

10 Fig. 2 shows the map of the lower tropospheric O₃ simulated by MRI-CCM2 for
11 CNTL corresponding to the OMI 24th layer (about 0–3 km altitude) after convolution
12 with eq. 1. As the OMI data were screened by the criteria of ECF and RMS, as described
13 in Section 2.1.1, the OMI data grids were sparsely selected. According to the OMI grid
14 selection, the simulated O₃ data shown in Fig. 2 are also sparse, although original model
15 data are filled in all the grids. High concentrations of O₃ are shown over CEC on both
16 June 10 (Fig. 2a) and June 20 (2b), 2006. To examine whether these high concentrations
17 were affected by UT/LS O₃ enhancement, the O₃ profiles in the area of 30–35°N, 115–
18 123°E, framed by the black rectangle, were investigated.

19 The O₃ profiles corresponding to each grid in the framed area are shown in Fig.
20 3a for June 10 and in Fig. 3b for June 20. In the figure, the a priori profiles used in the
21 OMI retrieval are indicated in gray, and differences in O₃ from the a priori profile are
22 shown in red. The center grid of the cross section shown in Fig. 1(34.205°N, 118.125°E)

1 corresponds to the profile in the second row and third column in Fig. 3. On June 10,
2 2006, differences between the MRI-CCM2 and OMI a priori profiles are prominent
3 around the 21st to 20th layer (about 10–15 km altitude), but they are not prominent on
4 June 20. Because the a priori data represent the climatological background, the O₃
5 difference shown in red can be interpreted as an enhancement from the background.

6 To elucidate the contribution of the O₃ enhancement at each layer to the 24th
7 layer, the values of the second term of eq. 1 ($\Sigma A(i,24)[X_{m,i} - X_{a,i}]$) are shown in Fig. 3c and
8 (d). On June 10 (Fig. 3c), the contribution of the O₃ enhancement in the UT/LS layers to
9 the 24th-layer O₃ is larger than that of the 24th layer itself for the most of the profiles in
10 Fig. 3c. These profiles on June 10 indicate that the high concentration of O₃ shown in
11 Fig. 2a can be attributed to the enhancement of O₃ in the UT/LS rather than to O₃
12 enhancement at the 24th layer.

13 On the other hand, on June 20, differences between the MRI-CCM2 O₃ and the
14 a priori O₃ are not significant in the UT/LS (Fig. 3b). Fig. 3d shows that the contribution
15 to the 24th layer is most notable in the lower troposphere, not in the UT/LS. Therefore,
16 the O₃ source on June 20, shown in Fig. 2b, can be attributed to O₃ production in the
17 lower troposphere, possibly by photochemical reactions. Although contributions from
18 the 23rd and the 22nd layers are not negligible, this is due to the relatively large AKs, as
19 discussed in Hayashida et al. (2015). Discrimination against the three lowermost layers
20 (22nd to 24th layers) is difficult, but this does not indicate a difficulty of elimination of the
21 artificial effect originated from the UT/LS. The two examples shown in Fig. 3 encourage
22 us to develop a screening scheme to remove the data affected by UT/LS O₃ enhancement.
23 We describe this scheme in the next section.

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3.1.3 Scheme to screen out the UT/LS effect on the 24th-layer O₃

As described above, the contribution of the O₃ enhancement at the UT/LS to the 24th-layer O₃ is not negligible, and this effect sometimes may mislead the interpretation of variation in the OMI 24th O₃. To remove the artificial effect of the UT/LS O₃ enhancement on the OMI 24th O₃, we defined criteria for screening out the data affected by UT/LS O₃ enhancement as:

$$\begin{aligned} & A(i,24)[X_{m,i} - X_{a,i}] > A(24,24)[X_{m,24} - X_{a,24}] \\ & \text{and} \quad (i = 1, \dots, 21), \quad (2) \\ & A(i,24)[X_{m,i} - X_{a,i}] > 0.5 \text{ DU} \end{aligned}$$

where $A(i,24)[X_{m,i} - X_{a,i}]$ is the second term of eq. 1 at the i^{th} layer (see Fig. 3c and 3d).

The first condition of eq. 2 removes the data when the second term of eq. 1 is greater than that of the 24th layer. The second condition that the second term of eq. 1 be greater than 0.5 DU was added because the first condition is always true when that of the 24th layer is negligibly small (almost zero), and is meaningless. Here, we introduced 0.5 DU as the threshold, although it is determined empirically based on all the data used in the analysis.

Fig. 4 shows the result of the screening based on the criteria of eq. 2. Over CEC (in the red frame), many of the grids for June 10 are screened out, while all of the grids for June 20 are accepted. In this way, we identify the grids where the contribution of O₃ variation at the UT/LS is significant, and remove these grids before the succeeding analysis.

1 Although we showed only the cases of June 10 and June 20, 2006, in this
2 section, all data were examined in the same way. Fig. 5 is the time series of the O₃
3 profiles, as shown in Fig. 3, at the grid of 34.025°N, 118.125°E, which is the same grid
4 focused upon in Fig. 1. Note that the data screening for OMI based on ECF and RMS
5 has been applied already, as mentioned in Section 2.1.1, thus only data for 14 days are
6 available. It is obvious that O₃ enhancement in the UT/LS is significant on June 1 and
7 June 9–11 (shaded in light blue), but is not significant on the other days. After the
8 UT/LS screening, 10 days remained for analysis, because four days (shaded in light
9 blue) were screened out.

10 Using the method described above, we applied the screening of eq. 2 to all grids
11 over East Asia and compared the monthly average O₃ distribution before and after the
12 screening. Fig. 6 shows the monthly mean O₃ in the 24th layer simulated by MRI-CCM2
13 before (Fig. 6a) and after (Fig. 6b) the screening for the UT/LS effect. In Fig. 6b, the
14 monthly average was obtained using only acceptable days/grids after the UT/LS
15 screening. Both Fig. 6a and 6b obviously indicate the high concentration of O₃ in the
16 lower troposphere over CEC. This result assures the validity of the 24th layer map in the
17 OMI over CEC presented in Fig. 10 of Hayashida et al. (2015). A notable difference
18 between the before and after UT/LS screening results is found in the ocean region east
19 of Japan at about 35–40°N, 160°E. In that region, the O₃ enhancement was notable
20 before the screening, as shown in Fig. 6a, in spite of no specific source over the sea.
21 Long-range transport from source regions toward the sea cannot explain the higher
22 concentration of ozone than those over the source regions such as over CEC. As in Fig.
23 6b, such abnormal high concentrations of O₃ have been screened out, which looks quite

1 reasonable. Over CEC, the picture of the monthly average after the UT/LS screening
2 clearly presents the O₃ enhancement as obtained before the screening. This is possibly
3 explained by the UT/LS effects occurring occasionally in relatively wide areas, and thus
4 being diluted on a monthly basis. A similar result was reported by Dufour et al. (2015).
5 This consistency also assures the validity of the monthly average O₃ map at the lower
6 troposphere obtained from OMI observation, and strengthens the finding of Hayashida
7 et al. (2015) for O₃ enhancement over CEC in June.

8 9 3.2 Comparison of satellite observation with model simulation

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11 In this section, we compare the satellite observation and model simulation for the two
12 scenarios (CNTL and OCRB), as shown in Table 1. Because CO and NO₂ are the
13 precursors of O₃, the outputs of these two species were examined, as well as O₃, to
14 validate the model simulations. For comparisons with satellite data, the outputs of
15 model simulations were converted to the comparable physical quantities. See Appendix
16 for details. In this section, we show comparisons of CO, NO₂, and O₃ in the map over
17 CEC and the longitude and latitude cross sections along 33°N and 117°E, where the
18 OCRB emission is at a maximum.

19 20 3.2.1 Carbon monoxide (CO)

21
22 To validate the CO concentrations simulated by MRI-CCM2, we compared them with
23 MOPITT observations in monthly basis. The MOPITT CO observation was generally

1 reproduced well by the model except for June although the figures for all months are not
2 shown here. Fig. 7a and 7b show the monthly mean total column CO in January 2006
3 from MOPITT observation and from MRI-CCM2 for CNTL, respectively. The observed
4 CO was consistent with those simulated by the model in winter as shown in Fig. 7,
5 though the results in December and February are not shown. However, in June, the
6 high concentration of CO over CEC observed by MOPITT (Fig. 7c) was not reproduced in
7 the model simulation for the CNTL scenario (Fig. 7d). The model simulation
8 considerably underestimated the CO concentration. On the other hand, the sensitivity
9 study for the OCRB scenario resulted in much higher CO, as shown in Fig. 7e, which
10 reproduced the high concentrations of CO over CEC observed by MOPITT. This result
11 indicates the validity of CO emission from OCRB estimated in the Yamaji's OCRB
12 inventory.

13 Fig. 8a and 8b are cross sections across latitudes along 33°N and longitudes
14 along 117°E, respectively. As already pointed out for Fig. 7c–e, the reproducibility of CO
15 is much better in the OCRB scenario, especially around the area where OCRB emissions
16 were added (about 30–35°N, 115–120°E) as shown in the bottom panels of Fig. 8 and 8b.
17 The results shown in Fig. 8 again demonstrate the reliability of the CO emission from
18 OCRB estimated by Yamaji et al. (2010).

19

20 3.2.2 Nitrogen dioxide (NO₂)

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22 Fig. 9 shows the monthly mean NO₂ in June 2006. The MRI-CCM2 simulations for
23 CNTL present the enhancement of NO₂ concentration over CEC corresponding to the

1 high emission in this region. However, the model simulation generally tends to
2 underestimate the NO₂ concentration, although not all the figures from throughout the
3 year are shown here; it does not reproduce the patchy hotspots of NO₂ over large cities
4 such as Beijing and Shanghai that are clearly observed by OMI (see Fig. 9a). Fig. 10a
5 and 10b are cross sections across latitudes and longitudes, respectively, as in Fig. 8. The
6 NO₂ concentration simulated by the CNTL scenario is almost consistent with OMI
7 observations in the areas of low NO₂ concentration (rural areas), as shown in Fig. 10,
8 although the discrepancy is more apparent over the large cities.

9 The NO_x emission fluxes obtained in the MACCity inventory used for the
10 model simulations indicate a smoother distribution than the NO₂ distribution observed
11 by OMI. As shown in Fig. A1(a), the geographical distribution of NO_x fluxes of MACCity
12 does not reflect hotspots over most of the large cities in CEC. This would be a major
13 reason for the discrepancy between the model result and OMI observation. Besides, it
14 may be difficult to quantitatively simulate NO₂ using the global model with a resolution
15 of ~110 km because of the short lifetime of NO₂ and the heterogeneous distribution of its
16 emission sources. To reproduce the NO₂ distribution on the scale of a city, we need to
17 use a regional model with high resolution coupled with more sophisticated emission
18 inventory reflecting a finer emission source distribution.

19 The OCRB sensitivity study (Fig. 9c) indicates the enhancement of NO₂
20 corresponding to the additional NO_x emissions from OCRB at around 30–35°N, 115–
21 120°E, where additional OCRB emission was involved. In this area, NO₂ from the OCRB
22 sensitivity study appears to be considerably higher than observed by OMI. One possible
23 reason for this difference is overestimation of NO₂ from crop burning in Yamaji's

1 emission inventory. However, as mentioned above, regional model simulations will be
2 required in the future to quantitatively determine the reason for the difference.

3 4 3.2.3 Ozone (O_3) 5

6 Fig. 11 shows the 24th-layer O_3 distribution. Fig. 11a, 11b, and 11c indicate the O_3
7 obtained from the OMI observation, MRI-CCM2 CNTL run, and MRI-CCM2 OCRB
8 sensitivity study, respectively. The observed O_3 enhancement over CEC in June 2006
9 (Fig. 11a) was reproduced very well by the model simulations. Fig. 12 shows the latitude
10 and longitude cross sections, respectively, as in Figs. 8 and 10. The peak O_3 values over
11 CEC (about 16 DU) observed by OMI are almost consistent with the values taken from
12 the CNTL scenario (solid red line). The difference between the O_3 with and without the
13 OCRB effect is not very large (about 1 DU), as shown by the red and blue solid lines; the
14 effect of OCRB emission on O_3 production looks limited.

15 To examine the smoothing effect, we also show the O_3 map without convolution
16 with AKs (Fig. 11d and e); underestimation of O_3 due to the smoothing is clear. When we
17 examine the results without AK convolution (the red dotted line and the blue dotted line
18 in Fig. 12), the effect of OCRB looks more significant. This is consistent with the report
19 from MTX2006 (Kanaya et al., 2013). However, we should note that the poor vertical
20 resolution of OMI prevents us from catching the effect of OCRB in OMI observations.
21 From the OMI retrievals, we conclude that the factors for high concentration of O_3 in
22 June are mainly anthropogenic emissions coupled with photochemical production, and
23 the OCRB effect is minor.

1 As shown in Fig. 9 and 10 of Hayashida et al. (2015), the lower tropospheric O₃
2 enhancement over CEC is notable in June every year. However, the stratospheric O₃
3 subsidence is not most active in June. We have analyzed ozone profiles in UT/LS using
4 the ozonesondes at four Japanese stations, including Sapporo, Tsukuba, Kagoshima,
5 and Naha, and the MOSAIC airborne measurement data over Beijing, Tokyo, and
6 Osaka. By analyzing all those data, we found the month of active UT/LS O₃ variability
7 depends on latitude, corresponding to the location of the STJ (Nakatani et al. 2012), and
8 June is not the most outstanding month for UT/LS O₃ variation for the latitudinal range
9 of our interest. As already mentioned in Section 3.1.3, the UT/LS effects occur
10 occasionally in relatively wide areas and should be diluted on a monthly basis. Although
11 we did not show the analysis for months other than June in this paper, the winter or
12 autumn months when the lower tropospheric O₃ enhancement is weak are out of our
13 scope. We carried out a similar analysis for May and July 2006 because O₃ enhancement
14 is not negligible in those months, though it is not as significant as in June. It was
15 confirmed that the conclusion derived from the data in June holds true for those months.
16 However, to quantitatively understand the difference between the OMI observations
17 and the model simulations, all months throughout the year should be examined in a
18 future study by utilizing a regional model with high resolution coupled with more
19 sophisticated emission inventory reflecting a finer emission source distribution.

20

21 **4 Conclusions**

22

23 In this study, we examined the effect of UT/LS O₃ enhancement on lower tropospheric

1 O₃ retrieval by OMI. We developed a scheme to eliminate cases affected by UT/LS ozone
2 enhancement. By applying the UT/LS screening scheme using model simulations of O₃
3 for June 2006, we showed clearly how the UT/LS O₃ enhancement produced an artificial
4 effect on the lower tropospheric O₃. However, even after the UT/LS screening, we were
5 able to find a clear enhancement of lower tropospheric O₃ over CEC in June 2006 and
6 confirmed the conclusion described by Hayashida et al. (2015).

7 After screening the UT/LS effect, we compared satellite measurements with
8 model simulations for CO, NO₂, and O₃, and examined the effects of OCRB emissions on
9 lower-tropospheric O₃. For the CO column, the output from the OCRB scenario was
10 fairly consistent with the MOPITT observation, although it was not consistent without
11 the OCRB emission. Therefore, we can conclude that the CO emission estimated by
12 Yamaji et al. (2010) is probable for CO. As for OMI O₃ observation, the effect of OCRB on
13 O₃ does not seem to be significant, although it may be more significant when focusing on
14 surface O₃.

15

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21

22

1 Appendix

2 A1 Emission inventories of NO_x used for simulations

3

4 Fig. A1

5 Map of NO_x emission fluxes used for simulations: (a) MACCity, (b) GFED version 3, and
6 (c) emissions used for OCRB sensitivity study.

7

8

9 A2 Physical quantity conversion

10

11 To compare the satellite observation data and the model simulation results, we
12 converted the physical quantities taken from the model simulations to the
13 corresponding quantities obtained from the satellite observation.

14

15 A2.1 Integration of CO and NO₂ to derive the tropospheric column

16

17 CO or NO₂ concentrations in the model's multiple layers were converted to total column
18 values using eq. 1:

19
$$X_{column} = \frac{1}{10^4} \left\{ X_1 (Z_1 - Z_{surf}) + \sum_{i=1}^{N-1} \frac{(X_i + X_{i+1})(Z_{i+1} - Z_i)}{2} \right\}, \quad (\text{A.1})$$

20 where X is the total column CO or NO₂ [molec/cm²] based on model simulation results,
21 and X_i and Z_i are the CO or NO₂ number density [molec/m³] and altitude [m] in the i^{th}
22 layer of the model, respectively. For MRI-CCM2, the altitude range for integration is
23 from the surface to the tropopause (about 100 hPa) ($N = 36^{\text{th}}$ layer in MRI-CCM2). We
24 integrated the number density of CO or NO₂ in each layer using the trapezoidal rule,

1 except for the region from the surface (Z_{surf}) to the center of the 1st layer (Z_1) of the model
 2 where the CO or NO₂ concentration is assumed to be constant (X_1).

3

4

5 A2.2 Integration of O₃ to derive the lowermost layer corresponding to the 24th-layer of
 6 OMI

7

8 O₃ concentrations in the model's multiple layers were converted to the lowermost
 9 tropospheric column O₃ corresponding the OMI 24th layer (about 0–3 km) using eq. (A.2):

$$10 \quad X_{column} = \frac{1}{2.69 \cdot 10^{20}} \left\{ X_1 (Z_1 - Z_{surf}) + \sum_{i=1}^{N-1} \frac{(X_i + X_{i+1})(Z_{i+1} - Z_i)}{2} + \frac{(X_N + X_{OMI24top})(Z_{OMI24top} - Z_N)}{2} \right\}, \quad (A.2)$$

11

$$12 \quad X_{OMI24top} = X_N + \frac{X_{N+1} - X_N}{Z_{N+1} - Z_N} (Z_{OMI24top} - Z_N), \quad (Z_N < Z_{OMI24top} < Z_{N+1}), \quad (A.3)$$

13

14 where X_{column} is the O₃ concentration corresponding the OMI 24th layer (DU) based on
 15 model simulation results, and X_i and Z_i are the O₃ number density [molec/m³] and
 16 altitude [m] in the i th layer of the model, respectively. $X_{OMI24top}$ and $Z_{OMI24top}$ are the O₃
 17 number density [molec/m³] and altitude [m], respectively, corresponding to the top of the
 18 OMI 24th layer (about 3 km), which can be interpolated as in eq. (A.3) with a value of N
 19 around 17 depending on meteorological conditions.

20

21

1 Figure captions

2 Fig. 1

3 (a) Map of O₃ distribution at 200 hPa simulated by MRI-CCM2 for the control run on
4 June 10, 2006. The unit of O₃ concentration is molec/cm³. Wind vectors at the same level
5 are overlain. Lines are drawn at latitude 34.205°N and longitude 118.125°E to indicate
6 the cross section in (c)–(e). (b) Same as (a) but for June 20, 2006. (c) Latitude–altitude
7 cross section at 118.125°E for June 10, 2006. Solid contour lines represent the zonal
8 wind speed (m/s), and dotted contour lines represent potential temperature (K). (d)
9 Same as (c) but for June 20, 2006. (e) Longitude–altitude cross section at 34.205°N for
10 June 10, 2006. Dotted contour lines represent potential temperature (K). (f) Same as (e)
11 but for June 20, 2006.

12

13 Fig. 2

14 Map of lower tropospheric O₃ (DU) simulated by MRI-CCM2 for the control run on June
15 10 (a) and June 20 (b), 2006. The O₃ amounts are adjusted to the OMI 24th layer and the
16 data are screened as in the OMI data (see Section 2.1.1 for the details). The black frame
17 in each panel indicates the region shown in Fig. 3.

18

19

20 Fig. 3

21 (a), (b): O₃ profiles simulated by CCM2 that are adjusted to OMI layers by convolution
22 with AKs as in eq. 1 for June 10, 2006 (a) and June 20, 2006 (b). Each profile
23 corresponds to each grid in the framed area (30–36°N, 115–124°E) shown in Fig. 2. Gray

1 bars indicate the OMI a priori O_3 [DU], and red and blue bars indicate the outputs of the
2 MRI-CCM2 control run and the MRI-CCM2 OCRB sensitivity study, respectively. The
3 scale of the x-axis of each panel is 0–50 DU.

4
5 (c), (d): Profiles of the second term in eq. 1, which indicate the contribution of the i^{th}
6 layer O_3 to the 24th-layer O_3 ($i = 1, \dots, 24$). Each profile corresponds to each profile in (a)
7 and (B). The scale of the x-axis is 0–4 DU.

8
9 Fig. 4

10 (a) Result of the grid screening to remove the UT/LS effect on the 24th-layer O_3 on June
11 10, 2006. (b) Same as (a) but for June 20, 2006. The grids in black are screened out and
12 those in gray are accepted by applying the criteria of eq. 2. The red frame in each panel
13 indicates the region shown in Fig. 3.

14
15 Fig. 5

16 Time series of O_3 profiles at 34.205°N, 118.125°E from June 1–30, 2006 as in Fig. 3. The
17 profiles shaded in light blue indicate the data affected by UT/LS O_3 enhancement on the
18 24th-layer O_3 , while those shaded in light pink do not indicate such O_3 enhancement.
19 The scale of the x-axis is 0–50 DU for the upper panel and 0–4 DU for the lower panel as
20 in Fig. 3.

21
22 Fig. 6

23 Lower tropospheric O_3 concentration (DU) simulated by MRI-CCM2 control run. The

1 data are adjusted to the OMI 24th layer, and cloud and RMS screening are applied as in
2 the OMI retrieval (see Section 2.1.1). (a) Monthly mean O₃ before UT/LS screening. (b)
3 Monthly mean O₃ after UT/LS screening.

4
5 Fig. 7

6 (a) Map of monthly mean total column CO (molec/cm²) observed by MOPITT in January
7 2006. (b) Same as (a) but for the simulation by the MRI-CCM2 control run. (c) Monthly
8 mean total column CO (molec/cm²) observed by MOPITT in June 2006. (d) Same as (c)
9 but for the simulation by the MRI-CCM2 control run. (e) Same as (c) but for the
10 simulation by the MRI-CCM2 for the OCRB scenario.

11
12 Fig. 8

13 Upper panels

14 (a) Cross section across latitude at 117.000°E. Black dotted line, red solid line, and blue
15 solid line correspond to MOPITT observation, MRI-CCM2 control run, and MRI-CCM2
16 OCRB sensitivity study, respectively. (b) Cross section across longitude at 33.084°N.
17 Lines are the same as those for (a).

18 Lower panels

19 Red bars show CO emissions of MRI-CCM2 control run, and blue bars show additional
20 CO emissions of MRI-CCM2 for the OCRB scenario.

21
22 Fig. 9

23 (a) Map of monthly mean tropospheric column NO₂ (molec/cm²) for June 2006 observed

1 by OMI. (b) Same as (a) but for MRI-CCM2 control run. (c) Same as (a) but for
2 MRI-CCM2 OCRB scenario. The grids in (a) are smoothed to $1.125^\circ \times 1.125^\circ$ (the
3 original OMI Level 3 data are provided at $0.25^\circ \times 0.25^\circ$) to adjust to the resolution of
4 MRI-CCM2.

5

6 Fig. 10

7 Upper panels

8 (a) Cross section across latitude at 117.000°E . Black dotted line, red solid line, and blue
9 solid line correspond to OMI observation, MRI-CCM2 control run, and MRI-CCM2
10 OCRB sensitivity study, respectively. (b) Cross section across longitude at 33.084°N .

11 Lines are the same as those in (a).

12 Lower panels

13 Red bars show NO_x emissions of the MRI-CCM2 control run, and blue bars show
14 additional NO_x emissions of MRI-CCM2 for the OCRB scenario.

15

16 Fig. 11

17 Monthly mean lower tropospheric O_3 (DU) in June 2006 after UT/LS screening.

18 (a) OMI observation, (b) MRI-CCM2 control run, and (c) MRI-CCM2 OCRB sensitivity
19 study. (d) Same as (b) but without convolution with AKs. (e) Same as (c) but without
20 convolution with AKs.

21

22 Fig. 12

23 (a) Cross section of O_3 (DU) across latitude at 118.125°E . Black dotted line, red solid line,

1 and blue solid line indicate OMI observation, MRI-CCM2 control run, and MRI-CCM2
2 OCRB sensitivity study, respectively. Red dotted and blue dotted lines indicate
3 MRI-CCM2 CNTL and MRI-CCM2 OCRB, as for the solid lines, but without convolution
4 with AKs. (b) Cross section across longitude at 34.205°N. Lines are the same as those in
5 (a).

6

7

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