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

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1 Abstract

 $\mathbf{2}$ 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) multiple-layer product retrieved by Liu et al. (2010), which first showed the lower 4 $\mathbf{5}$ 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 developed to eliminate the artificial effect of UT/LS ozone enhancement on lower 10 11 tropospheric ozone retrieval using OMI. By applying the UT/LS screening scheme for 12June 2006, we removed the artificial effect of the UT/LS ozone enhancement on the 13lower tropospheric ozone. Even after UT/LS screening, we were able to show a clear 14enhancement in the lower tropospheric ozone over CEC in June 2006 and confirmed the 15conclusion derived by Hayashida et al. (2015). To clarify the reason for ozone 16enhancement in June, the effects of emissions from open crop residue burning (OCRB) 17in the North China Plain on lower tropospheric ozone were also examined using a 18comparison with model simulations. On the scale of the vertical resolution of OMI 19observations, the effect of OCRB on ozone enhancement does not seem to be significant, 20although it may be more significant when focusing on ozone in the planetary boundary 21layer.

22

1 1 Introduction

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

10 Satellite measurements have played an increasingly important role in O_3 11 monitoring globally (e.g., Burrows et al. 2011 and references therein). However, vertical 12discrimination of O₃ in the lower troposphere has been a big challenge for satellite-borne 13measurements, because 90% of the total O_3 amount exists in the stratosphere. Recently, 14Liu et al. (2010) developed an algorithm for retrieving O_3 profiles using the UV radiances observed by the Ozone Monitoring Instrument (OMI). They retrieved ozone 1516profiles from the ground upward to about 60 km in 24 layers. There are 4 to 7 layers in 17the 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 20with multiple layers and revealed a significant O_3 enhancement in the lowermost layer 21(the 24th layer) over CEC, which is most notable in June each year. That was the first 22systematic view from satellite observation with ultraviolet spectra showing the ozone 23enhancement in the lowermost altitude over CEC. Further comparative studies, along with model simulations, are expected to clarify any unknown factors in ozone production
 and transport mechanisms.

3 However, the effect of a large variability in O₃ amount in the upper troposphere and lower stratosphere (UT/LS) on the OMI ozone retrieval must be taken into 4 $\mathbf{5}$ consideration carefully because of the large smoothing error of the OMI retrieval scheme 6 (Liu et al. 2010). The large O_3 variability in UT/LS may exert an influence on the 7 lowermost tropospheric O_3 . 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 O_3 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 12correlation between the concentrations of O_3 and carbon monoxide (CO) in the lower 13troposphere corresponding to the altitudes of 0–6 km over the North China Plain, both 14of which were observed by Infrared Atmospheric Sounding Interferometer (IASI); this 15positive correlation between O_3 and CO suggested the photochemical source of O_3 . On 16the other hand, they also pointed out signals of significant O₃ enhancement in the upper 17troposphere (6–12 km), which correlated with the low-pressure system, suggesting an 18 effect of O_3 subsidence from the stratosphere. As these studies indicate, East Asia, and 19 CEC in particular, is one of the key regions where both stratospheric O_3 subsidence and 20anthropogenic O_3 production are occurring actively. In this study, we present a scheme 21to remove the effect of the O₃ variability in the UT/LS on the retrieval of the lowermost 22 O_3 layer (0–3 km). By applying this scheme, we can confirm the enhancement of the 23lowermost O_3 every June shown by Hayashida et al. (2015).

To investigate the mechanism of repeatable O₃ enhancement in June over CEC 1 $\mathbf{2}$ (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 Tai Experiment (MTX2006) (references in Kanaya et al. 2013) revealed that the 4 $\mathbf{5}$ emissions from regional-scale OCRB after the harvesting of winter wheat increased the 6 concentration of O_3 , 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 NOx and CO originating from deforestation and 11 the burning of savanna, grassland, woodland, extratropical forest, and agricultural 12waste and peat (see Table 5 in van der Werf et al. 2010). However, emissions from open 13crop burning in the North China Plain have not been included in the GFED ver. 3. In 14this study, we examine the effect of OCRB on O_3 concentration via model simulations 15involving 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 17data observed by the global Moderate Resolution Imaging Spectroradiometer (MODIS). 18 We focus on June 2006 in this paper because the enhancement of O_3 is most notable 19 every June, as reported by Hayashida et al. (2015), and the outstanding effect of OCRB 20on O₃ in the North China Plain was demonstrated in June 2006 during the MTX2006, as 21mentioned above (Kanaya et al. 2013).

In Section 2, we describe the data used in our analysis and the model used for the simulations. In section 3.1, we present the scheme to eliminate the effect of UT/LS ozone enhancement on the lower tropospheric ozone derivation. In Section 3.2, we show a comparison between the satellite observations of O_3 and their precursors, such as carbon monoxide (CO) and nitrogen dioxide (NO₂), and the results of the model simulations. We demonstrate consistency between the observations and the model results and discuss the effect of OCRB on O_3 concentration.

- 6
- 7 2 Satellite data and model
- 8
- 9 2.1 Satellite observation
- 10

11 2.1.1 O₃ profile and NO₂ tropospheric column observed by OMI

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13OMI is the UV/visible sensor on board the National Aeronautics and Space 14Administration (NASA) EOS Aura spacecraft, which was launched in July 2004. The 15satellite is in a Sun-synchronous polar orbit with an equatorial crossing time of 13:45 16local time (LT). OMI measures backscattered radiances covering a wavelength range of 17270 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 19with a spatial resolution of 13×24 km for the UV-2 and visible channels, and 13×48 20km for the UV-1 channel.

In this study, we utilized the O_3 profiles retrieved by Liu et al. (2010) using the OMI UV spectra from the ground to about 60 km with 24 layers. In the retrieval algorithm developed by Liu et al. (2010), O_3 profiles were retrieved by applying the

optimal estimation technique (Rodgers 2000), with climatological mean O_3 profiles by 1 $\mathbf{2}$ 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, the 24th layer, which corresponds to a layer from about 0 km to 3 km altitude. As in 4 $\mathbf{5}$ Hayashida et al. (2015), the gridded O_3 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 < 2.4.8

9 We also utilized the NO₂ tropospheric column from OMI, the version 3 release (OMNO2d) 10 of OMI NO_2 gridded level-3 product (Data DOI: the 11 10.5067/Aura/OMI/DATA3007). The retrieval algorithm was described in detail by 12Bucsela et al. (2013). Although the original OMI NO_2 data are provided with a resolution of $0.25^{\circ} \times 0.25^{\circ}$, they are converted to adjust to the model resolution in the 1314later analysis.

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

17

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° \times 1° 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)
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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° \times 1.125°) and the vertical range, which is divided into

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

64 layers, varies from the ground to about 80 km (0.01 hPa).

NO from soils is taken from Yienger and Levy (1995); emissions of CO and N₂O from soils, from Muller (1992). Emissions of CO, CH₄, and NMHCs from the ocean was based on Brasseur et al. (1998) with the modifications of Horowitz et al. (2003). Emission of NO by lightning was diagnosed at 6-h intervals. The global flash frequency was calculated according to the parameterization of Price and Rind (1992, 1994). The details of the scheme are described in Deushi and Shibata (2011). Emissions from biomass 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 12observed by OMI, we conducted two types of experiments: a control run (CNTL) and an 13OCRB sensitivity study (OCRB). The emissions from biomass burning used in each 14experiment are shown in Table 1. In the CNTL experiment, GFED ver.3 was used for 15emissions from biomass burning. In the OCRB experiment, for the region of 7°S-50°N 16and 70°E–142°E, the biomass burning emissions were replaced with the OCRB emission 17inventory (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 20OCRB season in CEC. To develop the daily gridded OCRB data, the annual emissions 21from OCRB were allocated to the spatial grid of $0.5^{\circ} \times 0.5^{\circ}$ and to each day according to 22the satellite hotspots and geographical information of the land cover data. For more 23details, readers are to refer to Yamaji et al. (2010).

2 Table 1. Anthropogenic and biomass burning emission inventories

	Control run (CNTL)	Sensitivity study for open crop residue
		burning (OCRB)
Anthropogenic	MACCity (monthly)* (La	amarque 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

 $\mathbf{5}$

6 3 Results and discussion

7

 $8 \quad 3.1 \text{ UT/LS screening for } 24^{\text{th}}\text{-layer } O_3$

9

10In this section, we describe the method used to screen out the artificial effect of the O_3 11 enhancement in the UT/LS on the O_3 concentration of the 24th layer (0–3 km). In Section 123.1.1, we show the variation of O_3 in the UT/LS obtained by the MRI-CCM2 simulation 13for CNTL, which was related to the low-pressure system. In Section 3.1.2, we evaluate 14the contribution of the O_3 enhancement in the UT/LS to the 24th-layer O_3 by applying the averaging kernels (AKs) of the OMI retrieval. In Section 3.1.3, we present the 1516scheme to eliminate the cases in which the effect of the UT/LS O₃ on the 24th-layer O₃ is 17considerably large. The results before and after the UT/LS screening are shown.

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

 $\mathbf{2}$

We examined the O₃ profiles and meteorological fields in East Asia in June 2006, which were simulated by MRI-CCM2. The two different features in the O₃ profiles, i.e., significant UT/LS O₃ enhancement and significant lower tropospheric O₃ enhancement, were found in June 2006. Here, we show two cases as representative examples. These are the O₃ profiles on June 10 and June 20, 2006, which correspond to the former and 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 12O₃ over CEC, we show a cross section of ozone along 118.125°E in Fig. 1c. It is clear that 13 O_3 is descending to an altitude of about 10 km over the region at around 30–35°N where 14it corresponds to the center of ozone enhancement over Shandong (See Fig. 10 of 15Hayashida et al. 2015). In contrast to the case of June 10, that the STJ shifted to the 16north of CEC on June 20, 2006, and thus the subsidence of stratospheric O_3 was 17significant 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 O_3 subsidence, as shown in Fig. 201d. The longitude-altitude cross sections at 34.205°N in Fig. 1e and 1f indicate more 21clearly the difference of O_3 distribution between June 10 and June 20 in the lower 22troposphere. The lower tropospheric O_3 enhancement was clear at around 115–120°E, 23while the stratospheric O_3 subsidence was not over the area.

- 1
- 2 3.1.2 Evaluation of contribution of the UT/LS O₃ enhancement to 24th-layer O₃
- 3

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

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

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 with eq. 1. As the OMI data were screened by the criteria of ECF and RMS, as described 1213in Section 2.1.1, the OMI data grids were sparsely selected. According to the OMI grid 14selection, the simulated O₃ data shown in Fig. 2 are also sparse, although original model 15data are filled in all the grids. High concentrations of O_3 are shown over CEC on both 16June 10 (Fig. 2a) and June 20 (2b), 2006. To examine whether these high concentrations 17were affected by UT/LS O_3 enhancement, the O_3 profiles in the area of $30-35^{\circ}N$, 115-18123°E, framed by the black rectangle, were investigated.

The O_3 profiles corresponding to each grid in the framed area are shown in Fig. 3a for June 10 and in Fig. 3b for June 20. In the figure, the a priori profiles used in the OMI retrieval are indicated in gray, and differences in O_3 from the a priori profile are 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.

To elucidate the contribution of the O₃ enhancement at each layer to the 24th 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 (d). On June 10 (Fig. 3c), the contribution of the O₃ enhancement in the UT/LS layers to the 24th-layer O₃ is larger than that of the 24th layer itself for the most of the profiles in Fig. 3c. These profiles on June 10 indicate that the high concentration of O₃ shown in Fig. 2a can be attributed to the enhancement of O₃ in the UT/LS rather than to O₃ enhancement at the 24th layer.

13On the other hand, on June 20, differences between the MRI-CCM2 O₃ and the 14a priori O₃ are not significant in the UT/LS (Fig. 3b). Fig. 3d shows that the contribution 15to the 24th layer is most notable in the lower troposphere, not in the UT/LS. Therefore, 16the O_3 source on June 20, shown in Fig. 2b, can be attributed to O_3 production in the 17lower troposphere, possibly by photochemical reactions. Although contributions from the 23rd and the 22nd layers are not negligible, this is due to the relatively large AKs, as 18 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 21artificial effect originated from the UT/LS. The two examples shown in Fig. 3 encourage 22us to develop a screening scheme to remove the data affected by UT/LS O₃ enhancement. 23We describe this scheme in the next section.

- 1
- 2 3.1.3 Scheme to screen out the UT/LS effect on the 24th-layer O_3
- 3

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:

$$A(i,24)[X_{m,i} - X_{a,i}] > A(24,24)[X_{m,24} - X_{a,24}]$$
9 and (*i* = 1,...,21), (2)
$$A(i,24)[X_{m,i} - X_{a,i}] > 0.5 DU$$

10 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).

11 The first condition of eq. 2 removes the data when the second term of eq. 1 is 12 greater than that of the 24th layer. The second condition that the second term of eq. 1 be 13 greater than 0.5 DU was added because the first condition is always true when that of 14 the 24th layer is negligibly small (almost zero), and is meaningless. Here, we introduced 15 0.5 DU as the threshold, although it is determined empirically based on all the data 16 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 O_3 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 $\mathbf{2}$ section, all data were examined in the same way. Fig. 5 is the time series of the O_3 3 profiles, as shown in Fig. 3, at the grid of 34.025°N, 118.125°E, which is the same grid focused upon in Fig. 1. Note that the data screening for OMI based on ECF and RMS 4 $\mathbf{5}$ 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 12screening. Fig. 6 shows the monthly mean O₃ in the 24th layer simulated by MRI-CCM2 13before (Fig. 6a) and after (Fig. 6b) the screening for the UT/LS effect. In Fig. 6b, the 14monthly average was obtained using only acceptable days/grids after the UT/LS 15screening. Both Fig. 6a and 6b obviously indicate the high concentration of O_3 in the lower troposphere over CEC. This result assures the validity of the 24th layer map in the 1617OMI 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 19of Japan at about $35-40^{\circ}$ N, 160° E. In that region, the O₃ enhancement was notable 20before the screening, as shown in Fig. 6a, in spite of no specific source over the sea. 21Long-range transport from source regions toward the sea cannot explain the higher 22concentration of ozone than those over the source regions such as over CEC. As in Fig. 236b, such abnormal high concentrations of O_3 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 O_3 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 O_3 map at the lower
6	troposphere obtained from OMI observation, and strengthens the finding of Hayashida
7	et al. (2015) for O_3 enhancement over CEC in June.

8

9 3.2 Comparison of satellite observation with model simulation

10

11 In this section, we compare the satellite observation and model simulation for the two 12scenarios (CNTL and OCRB), as shown in Table 1. Because CO and NO₂ are the 13precursors of O_3 , the outputs of these two species were examined, as well as O_3 , to 14validate the model simulations. For comparisons with satellite data, the outputs of 15model simulations were converted to the comparable physical quantities. See Appendix 16for details. In this section, we show comparisons of CO, NO₂, and O₃ in the map over 17CEC and the longitude and latitude cross sections along 33°N and 117°E, where the 18 OCRB emission is at a maximum.

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20 3.2.1 Carbon monoxide (CO)

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

reproduced well by the model except for June although the figures for all months are not 1 $\mathbf{2}$ 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 CO was consistent with those simulated by the model in winter as shown in Fig. 7, 4 $\mathbf{5}$ though the results in December and February are not shown. However, in June, the high concentration of CO over CEC observed by MOPITT (Fig. 7c) was not reproduced in 6 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 12inventory.

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

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20 3.2.2 Nitrogen dioxide (NO₂)

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

high emission in this region. However, the model simulation generally tends to 1 $\mathbf{2}$ underestimate the NO_2 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 such as Beijing and Shanghai that are clearly observed by OMI (see Fig. 9a). Fig. 10a 4 $\mathbf{5}$ 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 NOx 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 NOx fluxes of MACCity 12does not reflect hotspots over most of the large cities in CEC. This would be a major 13reason for the discrepancy between the model result and OMI observation. Besides, it 14may be difficult to quantitatively simulate NO₂ using the global model with a resolution 15of ~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 17use a regional model with high resolution coupled with more sophisticated emission 18 inventory reflecting a finer emission source distribution.

The OCRB sensitivity study (Fig. 9c) indicates the enhancement of NO₂ corresponding to the additional NOx emissions from OCRB at around 30–35°N, 115– 120°E, where additional OCRB emission was involved. In this area, NO₂ from the OCRB sensitivity study appears to be considerably higher than observed by OMI. One possible reason for this difference is overestimation of NO₂ from crop burning in Yamaji's emission inventory. However, as mentioned above, regional model simulations will be
 required in the future to quantitatively determine the reason for the difference.

3

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4 3.2.3 Ozone (O<sub>3</sub>)
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 $\mathbf{5}$

Fig. 11 shows the 24^{th} -layer O_3 distribution. Fig. 11a, 11b, and 11c indicate the O_3 6 7 obtained from the OMI observation, MRI-CCM2 CNTL run, and MRI-CCM2 OCRB 8 sensitivity study, respectively. The observed O₃ 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 12the CNTL scenario (solid red line). The difference between the O_3 with and without the 13OCRB effect is not very large (about 1 DU), as shown by the red and blue solid lines; the 14effect of OCRB emission on O₃ production looks limited.

15To 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 17examine 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 20resolution of OMI prevents us from catching the effect of OCRB in OMI observations. 21From the OMI retrievals, we conclude that the factors for high concentration of O_3 in 22June are mainly anthropogenic emissions coupled with photochemical production, and 23the OCRB effect is minor.

1 As shown in Fig. 9 and 10 of Hayashida et al. (2015), the lower tropospheric O_3 $\mathbf{2}$ enhancement over CEC is notable in June every year. However, the stratospheric O_3 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, and Naha, and the MOSAIC airborne measurement data over Beijing, Tokyo, and $\mathbf{5}$ 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 autumn months when the lower tropospheric O_3 enhancement is weak are out of our 1213scope. We carried out a similar analysis for May and July 2006 because O₃ enhancement 14is not negligible in those months, though it is not as significant as in June. It was 15confirmed that the conclusion derived from the data in June holds true for those months. 16However, to quantitatively understand the difference between the OMI observations 17and 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

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

7 After screening the UT/LS effect, we compared satellite measurements with model simulations for CO, NO₂, and O₃, and examined the effects of OCRB emissions on 8 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 12Yamaji et al. (2010) is probable for CO. As for OMI O₃ observation, the effect of OCRB on 13 O_3 does not seem to be significant, although it may be more significant when focusing on 14surface O₃.

15

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21

1	Appendix
2	A1 Emission inventories of NOx used for simulations
3	
4	Fig. A1
5	Map of NOx 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

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

where X is the total column CO or NO₂ [molec/cm²] based on model simulation results, and X_i and Z_i are the CO or NO₂ number density [molec/m³] and altitude [m] in the *i*th layer of the model, respectively. For MRI-CCM2, the altitude range for integration is from the surface to the tropopause (about 100 hPa) (N = 36th layer in MRI-CCM2). We integrated the number density of CO or NO₂ in each layer using the trapezoidal rule, 1 except for the region from the surface (Z_{srf}) 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 O_3 concentrations in the model's multiple layers were converted to the lowermost 9 tropospheric column O_3 corresponding the OMI 24th layer (about 0–3 km) using eq. (A.2):

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

11

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

1 Figure captions

2 Fig. 1

3 (a) Map of O₃ distribution at 200 hPa simulated by MRI-CCM2 for the control run on June 10, 2006. The unit of O₃ concentration is molec/cm³. Wind vectors at the same level 4 $\mathbf{5}$ 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

Map of lower tropospheric O_3 (DU) simulated by MRI-CCM2 for the control run on June 15 10 (a) and June 20 (b), 2006. The O_3 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

(a), (b): O_3 profiles simulated by CCM2 that are adjusted to OMI layers by convolution with AKs as in eq. 1 for June 10, 2006 (a) and June 20, 2006 (b). Each profile corresponds to each grid in the framed area (30–36°N, 115–124°E) shown in Fig. 2. Gray bars indicate the OMI a priori O₃ [DU], and red and blue bars indicate the outputs of the
MRI-CCM2 control run and the MRI-CCM2 OCRB sensitivity study, respectively. The
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 O₃ to the 24th-layer O₃ (i = 1,...,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

(a) Result of the grid screening to remove the UT/LS effect on the 24th-layer O₃ on June
10, 2006. (b) Same as (a) but for June 20, 2006. The grids in black are screened out and
those in gray are accepted by applying the criteria of eq. 2. The red frame in each panel
indicates the region shown in Fig. 3.

14

15 Fig. 5

Time series of O₃ profiles at 34.205°N, 118.125°E from June 1–30, 2006 as in Fig. 3. The profiles shaded in light blue indicate the data affected by UT/LS O₃ enhancement on the 24th-layer O₃, while those shaded in light pink do not indicate such O₃ enhancement. The scale of the x-axis is 0–50 DU for the upper panel and 0–4 DU for the lower panel as in Fig. 3.

21

22 Fig. 6

23 Lower tropospheric O₃ concentration (DU) simulated by MRI-CCM2 control run. The

1	data are adjusted to the OMI $24^{ m th}$ layer, and cloud and RMS screening are applied as in
2	the OMI retrieval (see Section 2.1.1). (a) Monthly mean O_3 before UT/LS screening. (b)
3	Monthly mean O ₃ after UT/LS screening.
4	
5	Fig. 7
6	(a) Map of monthly mean total column CO (molec/cm ²) 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/cm ²) 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 NO ₂ (molec/cm ²) for June 2006 observed

by OMI. (b) Same as (a) but for MRI-CCM2 control run. (c) Same as (a) but for
MRI-CCM2 OCRB scenario. The grids in (a) are smoothed to 1.125° × 1.125° (the
original OMI Level 3 data are provided at 0.25° × 0.25°) to adjust to the resolution of
MRI-CCM2.

- $\mathbf{5}$
- 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

Red bars show NO_x emissions of the MRI-CCM2 control run, and blue bars show
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
- study. (d) Same as (b) but without convolution with AKs. (e) Same as (c) but withoutconvolution with AKs.
- 21
- 22 Fig. 12
- (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).

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