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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observation of ozone enhancement in the lower troposphere over East Asia from a space-borne ultraviolet spectrometer" *by* S. Hayashida et al.

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Thank you for your positive evaluation and careful review of our study. We have revised our manuscript in accordance with your comments. The most significant revision was the merging of the Results and Discussion sections into one section, as suggested by both reviewers. Discussion on OCRB was also merged into Section 3.3 with the discussion of annual variation.

Detailed responses to the comments from the reviewers are as follows.

Major comments 1. More explanation should be given why layered information of the

tropospheric ozone can be retrieved in this study using the UV spectrum only, as other past studies failed. The principle is to include at least two spectral regions having sensitivities at different altitude levels? The authors should also address the issue of the effect of co-existing aerosol particles on the radiative transfer in the UV spectral region.

The following paragraph has been added to Section 2.1 (page 7, lines 11–19: highlighted in yellow).

"The capability to discriminate the vertical distribution of ozone profiles from backscattered UV measurements in the Hartley and Huggins bands arises primarily from wavelength-dependent photon penetration resulting from wavelength-dependent ozone absorption and its interaction with Rayleigh scattering (Bhartia et al., 1996). Vertical sensitivity to ozone in the troposphere, although much smaller compared to that in the stratosphere, arises from wavelengths longer than ∼300 nm. Temperature-dependent ozone absorption in the Huggins bands provides additional tropospheric ozone information (Chance et al., 1997). The distribution of vertical sensitivity depends on the retrieval spectral range, measurement signal to noise ratio, fitting quality, viewing geometry, cloudiness, surface albedo, and the vertical distribution of ozone."

The following paragraph has been added to Section 3.1.4 (page 14, lines 9–21: highlighted in yellow).

"The ozone profile algorithm has the capability to include monthly mean Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model fields of aerosols (profiles of six types of aerosols) in the radiative transfer simulation (Chin et al., 2002; Martin et al., 2003; Liu et al., 2005). However, the aerosol fields are not used in OMI retrievals (Liu et al., 2010a). Because the fitting of wavelength-dependent surface albedo parameters can partly account for the effects of aerosols owing to similar weighting functions, the retrievals are not very sensitive to the inclusion of aerosols (Liu et al., 2005). For example, for the OMI ozone profile retrieval in Figs. 2 and 4,

the corresponding monthly mean GOCART tropospheric aerosol optical depth is 0.65 at 347 nm with a single scattering albedo of 0.9. Using the aerosol fields only results in a small change in the retrieval: the total ozone column increases by ∼0.4 DU from 327.4 to 327.8 DU, the tropospheric ozone column increases by ∼1.0 DU from 54.9 DU to 55.9 DU, and the ozone in layer 24 decreases by ∼0.3 DU from 15.6 to 15.3 DU. Therefore, the observed ozone enhancement shown in Figs. 2 and 4 is not caused by neglecting aerosols in the retrieval."

2. Same profile as OMI is assumed for the 21th and above layers in the derivation of MOZAIC-based values convolved with the AKs. The common contribution from these high altitude layers could be the source of correlation in Figure 5 and therefore the authors need to test if such contribution is small, in order to retain the conclusion.

We have repeated the analysis using a priori data instead of the retrieved ozone profiles. The figure A corresponds to Fig. 5 except that a priori values are taken for layers 1–21. The result was a little worse but not significantly different from that in Fig. 5. In the (b) panels of the figure below, data are scatted around zero as expected. We can include the figure as another Supplementary figure if needed. However, as it is not possible to know the true ozone profile on the relevant day, it is not particularly helpful to examine the effect of upper ozone assuming a variety of ozone profiles. The effects from upper ozone layers are already shown as AKs in Figure 4.

3. The authors should not overstate the importance of OCRB in June over the Central Eastern China region. For example Yamaji et al. (2010) mentioned that even without the residues burning, the monthly average ozone level is high (73.9 ppbv), being fueled by normal anthropogenic emissions, and (only) about 7 ppbv increase from that level is attributable to the OCRB. They discussed that the OCRB is certainly important for the reproduction of the observed monthly ozone level (81.3 ppbv) in the model. Also, I feel that evidence is not enough to attribute the high ozone episode on June 22, 2005 to the influence of OCRB. In this manuscript only monthly accumulation of hotspots is discussed; the burning period should be normally much shorter than a month. I am

afraid that the rate of hotspot detection is much less in the latter half of June.

As you suggested, a relationship between OCRB and ozone enhancement in June is still only speculation. To avoid overstating the importance of OCRB in June, we have revised the later part of the Results and Discussion section. Figures 9 and 10 in the original manuscript were replaced after a discussion of monthly ozone variation. Please see Section 3.3 of the revised manuscript (page 17, line 9).

4. In sections 4.1 and 4.2, the authors frequently repeated the results already displayed in the previous section. I would suggest that Results and Discussion sections are merged to form a single section 3.

We have merged the Results and Discussion sections into one section: Section 3 in the revised manuscript (page 10).

Other technical comments 1. Page 2015, line 7. Not only infrared but also UV absorption leads to global warming. We have revised this sentence accordingly (page 2, line 10).

2. Page 2019, line 22. of which 3-7 layers are We have revised this sentence in accordingly (page 5, line 22).

3. Page 2023, section 3.1.2 Are the MOZAIC data over the airport represent the 52 x 48 km footprint size of the satellite observation?

In general, it is very difficult to evaluate spatial inhomogeneity of ozone in the lower troposphere with a reliable range using one profile obtained by airborne measurements. However, for the case discussed in this study, we have information on the ozone profile for the same day. The figure B is an ozone profile (shown in red) obtained by MOZAIC in the morning at 10:41 on 22 June 2005; these data were not used in our analysis because data taken in the afternoon (14:18) were available. Although the ozone mixing ratio at 10:41 is less than that observed ratio of 14:18, this represents a considerable enhancement of ozone, up to 120 ppbv. This means that ozone enhancement on that

day is not a sporadic phenomenon occurring in a very short time scale. In other words, it is reasonable to suppose that the ozone enhancement on 22 June 2005 is not a phenomenon confined to a limited area such as the airport. The spatial extent of ozone enhancement should have been in the range of at least several tens of kilometers or more.

4. Page 2024, line 3. How CO was measured? We have added an explanation in Section 2.5 (page 9, line 19-22: highlighted in yellow).

"MOPITT operates by sensing infrared radiation either from thermal emission/absorption at 4.7 μm for CO profiles, or from reflected sunlight at about 2.2–2.4 μm for CO column measurements in daylight. The use of solar channels enhances the instrument's sensitivity to the atmospheric boundary layer [https://www2.acd.ucar.edu/mopitt/concepts]."

5. Page 2026, Equation (1). A minus character in the term [X_t,j – X_a,i] is missing. Thank you for your careful review. This character was lost during the process of publication.

6. Page 2027, section 3.2. How much was the a priori profiles variable longitudinally for a given latitudinal band? A priori data are essentially constant longitudinally, except for some differences along with the changing surface pressure coupled with topography. Please also see page 8 line 10-13 in the revised manuscript.

7. Page 2032, line 16. What are the altitude ranges of the layers 19 and 20? The corresponding altitude layer is about 12–15 km. We have added these numbers in the text (page 17, line 3).

8. Page 2032, section 4.2. The authors need to state which relationship is expected between the position of subtropical jet and descent of stratospheric ozone. We have revised the relevant sentence as follows (page 16, line 26- page 17, lines 2: highlighted in yellow).

"In Fig. 8, we show a global map of NCEP wind at 200 hPa on the same day, with the location of the maximum wind speed indicating an STJ that is closely related to the stratospheric ozone intrusion (e.g., Holton et al., 1995)."

9. Page 2034, line 10. The authors propose using a scale factor 0.17. Considering this factor how much of ozone variation (in DU or in ppb) in the lower troposphere can be detected by OMI? We have deleted this sentence because this issue is still under investigation.

10. Page 2035, lines 25-27. Please reword "The validation has not been evaluated : : :" We have replaced the word "evaluated" with "studied" (page 19, line 16).

Other minor corrections:

Page 9, line 14: "80" has been revised to "80%". Page 14, line 7: "two layers at 23 and 24" in the original manuscript has been revised to "the summation of ΔO3 (OMI) of the three layers from 22nd layer throgh the 24th layer." Figure 3, panel (c3): Title of the y-axis was corrected to "ΔO3 22nd, 23rd and 24th(OMI) [DU]". (Figure is as before)

Inappropriate references have been removed:

Aires, F., et al., J. Geophys. Res., 117, D18304, doi:10.1029/2011JD017188, 2012.

Safieddine, S., et al., J. Geophys. Res. Atmos., 118, 10,555-10,566, doi:10.1002/jgrd.50669, 2013.

Yan, X., et al., Atmos. Environ., 40, 5262–5273, doi:10.1016/j.atmosenv.2006.04.040, 2006.

Some references have been added:

Bhartia, P. K. et al., J. Geophys. Res., 101(D13), 18,793–18,806, doi:10.1029/96jd01165, 1996.

Chin, M. et al., J. Atmos. Sci., 59, 461– 483, 2002.

Martin, R. V. et al., J. Geophys. Res., 108(D3), 4097, doi:10.1029/2002JD002622, 2003.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C2096/2015/acpd-15-C2096-2015-supplement.zip

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 2013, 2015.

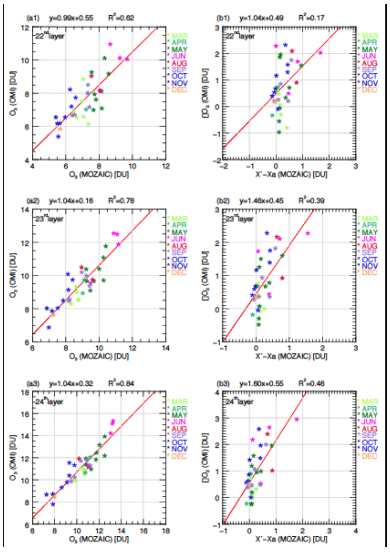

Figure A

Fig. 1.

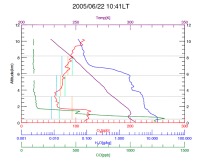

Figure B

Fig. 2.