

## **Reply to Referee #2**

Dear Referee #2

We would like to thank the Referee #2 for the valuable comments and suggestions. Please find the manuscript with major revision. We answered to your comments and suggestions in point by point. We also improved the abstract and conclusion to make the purpose of this paper clear. We hope that current manuscript is significant for the publication in AMT. We still wonder our English is not perfect as a manuscript of AMT even after the Native English checking. We will ask to our colleague, who is native English speaker, only once before the final version, since we do not want to disturb him frequently.

Sincerely yours,

Tomohiro Sato and Yasuko Kasai

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Note

**GC:** General comments

**MC:** Minor corrections or Additional corrections

## **Answers to Referee #2**

### **Overview comment**

The paper by Kasai et al. presents an interesting simulation study of the potential of a synergetic retrieval using UV, IR and microwave (UV+IR+MW) spectra for retrieving tropospheric ozone vertical profiles. This multispectral approach could be implemented in future Japanese air-quality monitoring missions. Whereas the topic and relevance of the paper are suitable for a possible publication at AMT, the current manuscript needs substantial revision in order to add and complete with important information and correct some inconsistencies of the results. As described below, part of the calculations are needed to be done again (UV radiative transfer and error/averaging kernels estimations) and many aspects should be more detailed (comparisons with previous multispectral methods, analysis of sensitivity as function of height, error estimations, etc.).

The major revisions needed for the manuscript are the following:

### **Answer to Overview comment**

We would like to thank you for the helpful comments and suggestions. We carefully checked our calculation script according to your suggestion and found a mistake to estimate the increase ratio of DFS values. The increase ratio was modified to 30% from 40% in the LMT region. Thank you so much for this suggestion. We answered the comments point-by-point and revised according to your suggestions. We also improved the abstract and conclusion in accordance with your suggestions.

### **Revisions to Overview comment**

Page 1 Abstract

“We performed a quantitative feasibility study of constraining the vertical profile of the amount of ozone in the troposphere by using a synergetic retrieval method on multiple spectra, i.e., ultraviolet (UV), thermal infrared (TIR) and microwave (MW) ranges, measured from space. Twenty atmospheric scenarios for East Asia in summer and winter seasons were assumed in this study. Geometry of line-of-sight was nadir down-looking for UV and TIR measurements, and limb-sounding for MW measurement. The sensitivities of retrieved ozone in the upper troposphere (UT), middle troposphere (MT) and lowermost troposphere (LMT) were estimated using values of the degree of freedom for signal (DFS), partial column error, and averaging kernel matrix, derived based on the optimal estimation method. The measurement noises were assumed at the same level as the currently available instruments. The weighting functions for the UV, TIR and MW ranges were calculated using the SCIATRAN radiative transfer model, the Line-By-Line

Radiative Transfer Model, and the Advanced Model for Atmospheric Terahertz Radiation Analysis and Simulation, respectively. In the UT region, the DFS value was enhanced by about 200% by adding the MW measurements to the combination of UV and TIR measurements. We found that the DFS value of the LMT ozone was increased by approximately 40% by adding the MW measurements to the combination of UV and TIR measurements; nevertheless, the MW measurement alone has no sensitivity for the LMT ozone. Better information of the LMT ozone can be deduced by adding constraints on the UT and MT ozone from the MW measurement. The results of this study will be implemented in the Japanese air-quality monitoring missions, APOLLO, GMAP-Asia and uvSCOPE.”

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“We performed a feasibility study to constraining the vertical profile of the amount of ozone in the troposphere by using a synergetic retrieval method on multiple spectra, i.e., ultraviolet (UV), thermal infrared (TIR) and microwave (MW) ranges, measured from space. A quantitative evaluation of the sensitivity of the tropospheric retrieval by adding the MW measurement to the UV and TIR measurements was reported for the first time by this work. The urban and sea areas in East Asia in summer and winter seasons were selected for the feasibility study. Geometry of line-of-sight was nadir down-looking for UV and TIR measurements, and limb-sounding for MW measurement. The sensitivities of retrieved ozone in the upper troposphere (UT), middle troposphere (MT) and lowermost troposphere (LMT) were estimated using values of the degree of freedom for signal (DFS), pressure of maximum sensitivity, error reduction rate from the a priori error, and averaging kernel matrix, derived based on the optimal estimation method. The measurement noises were assumed at the same level as the currently available instruments. The weighting functions for the UV, TIR and MW ranges were calculated using the SCIATRAN radiative transfer model, the Line-By-Line Radiative Transfer Model, and the Advanced Model for Atmospheric Terahertz Radiation Analysis and Simulation, respectively. The DFS value was increased by approximately 96%, 23% and 30% by adding the MW measurements to the combination of UV and TIR measurements in the UT, MT and LMT regions, respectively. We found that the MW measurement increased the DFS value of the LMT ozone; nevertheless, the MW measurement alone has no sensitivity for the LMT ozone. The pressure of maximum sensitivity of the LMT ozone was also increased by adding the MW measurement. It might indicate that better information of the LMT ozone can be deduced by adding constraints on the UT and MT ozone from the MW measurement. The results of this study will be implemented in the Japanese air-quality monitoring missions, APOLLO, GMAP-Asia and uvSCOPE.”

Page 13 Conclusions

“ We performed a feasibility study of obtaining a vertically resolved ozone profile in the

troposphere from synergetic retrieval using a combination of three separate wavelength ranges (UV, TIR, and MW). Observation geometries used in this study were the nadirs for the UV and TIR measurements and limb for the MW measurement from low orbit at a height of 300 km (the height of the ISS). Twenty atmospheric profiles at CEC and ECS in June and December 2009 were used for the simulation. We evaluated the sensitivities of retrieval of ozone in the three vertical regions (UT (215-383 hPa), MT (383-749 hPa) and LMT (>749 hPa)) in terms of the DFS, PCE and A based on the OEM calculation.

The TIR measurement was most sensitive for retrieving ozone in the UT when only one wavelength range was used. The additional MW measurement was most effective at improving the sensitivity in the UT when combining several wavelength ranges. Adding the MW measurement to the UV+TIR measurements increased the DFS value in the UT by a factor of two. In the MT region, the contribution of the TIR measurement was dominant in the DFS calculation. The average DFS value of the TIR measurement for all of the 20 profiles used in this study was  $0.83 \pm 0.11$ . It increased to more than unity by adding either the UV or MW measurements. The UV and TIR measurements were dominant in the retrieval of ozone in the LMT region. The DFS value in the LMT strongly depended on the ozone abundance. Our calculation shows that the combination of UV, TIR and MW measurements is able to retrieve the ozone abundance in the LMT with a DFS value larger than unity, when the ozone abundance is as high as approximately  $5 \times 10^{21} \text{ m}^{-2}$  in the LMT and the peak of A is in the LMT for both the UV and TIR measurements.

It is worth noting that adding MW measurements to the UV and TIR measurement combinations improved sensitivity not only in the UT but also in the MT and LMT; nevertheless, less information on ozone in the MT and LMT was derived from the MW measurement alone. The DFS values increased by 95%, 23% and 40% in the UT, MT and LMT by adding the MW measurements to the UV+TIR measurements. This indicates that reducing the uncertainty of ozone abundance in the stratosphere is important for estimating an accurate tropospheric ozone profile.”

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“ We performed a feasibility study of obtaining a vertically resolved ozone profile in the troposphere from synergetic retrieval using a combination of three separate wavelength ranges (UV, TIR, and MW). Observation geometries used in this study were the nadirs for the UV and TIR measurements and limb for the MW measurement from low orbit at a height of 300 km (the height of the ISS). The urban (CEC) and ocean (ECS) area in June and December 2009 were assumed in this study. We evaluated the sensitivities of retrieval of ozone in the three vertical regions (UT (215-383 hPa), MT (383-749 hPa) and LMT (>749 hPa)) in terms of the degree of freedom for signal (DFS) based on the OEM calculation. The pressure of maximum sensitivity (PMS), the reduction rate of error (RRE) for the partial column were also used as an indicator of

the sensitivity evaluation.

The TIR measurement was most sensitive for retrieving ozone in the UT when only one wavelength range was used. The additional MW measurement was most effective at improving the sensitivity in the UT when combining several wavelength ranges. The DFS values in the UT for all of 20 profiles averaged were  $0.62 \pm 0.08$  and  $1.21 \pm 0.28$  for the UV+TIR and UV+TIR+MW measurements, respectively. In the MT region, the contribution of the TIR measurement was dominant in the DFS calculation. The average DFS value of the TIR measurement for all profiles averaged was  $0.83 \pm 0.11$ . It was increased to more than unity by adding either the UV or MW measurements. The UV and TIR measurements were dominant in the retrieval of ozone in the LMT region. The DFS value in the LMT strongly depended on the ozone abundance. The DFS value in the LMT became larger in the case of larger partial column of the LMT ozone. The largest DFS value in the LMT for the UV+TIR+MW measurement of  $1.03 \pm 0.01$  was given by the case at CEC in June 2009 which is the case of the largest LMT ozone enhancement with the partial column in the LMT ozone of approximately  $5 \times 10^{21} \text{ m}^{-2}$ .

The MW limb measurement alone derived less information in the MT and LMT regions. The DFS values were less than 0.01 and the PMS was located at vertical region higher than the MT and LMT regions. Nevertheless, adding MW measurements to the UV and TIR measurement combinations improved sensitivity not only in the UT but also in the MT and LMT. The DFS values were increased by 96%, 23% and 30% in the UT, MT and LMT, respectively, by adding the MW measurements to the UV+TIR measurements. This might indicate that reducing the uncertainty of ozone abundance in the stratosphere is important for estimating an accurate tropospheric ozone profile.”

### **GC 2-1**

The choice of some of the parameters used in the simulation are neither clearly justified nor realistic: in page 3 lines 41-43, it is mentioned that a surface albedo of 90% and solar zenith angle of  $0^\circ$  are considered. These parameters are of course not realistic. Indeed, surface albedo at the UV is often from 5% to 15% and only greater than 30% for exceptionally bright surfaces. Solar zenith angles are also key parameters affecting UV spectra that greatly vary depending on overpass time and location of the measurement. Simulations of UV spectra and jacobians should be done again with realistic values of surface albedo and diverse solar zenith angles. The use of appropriate values is key for a proper estimation of the performance of the retrieval approach and they should be realistic for each scene observed.

### **Answer to GC 2-1**

We thank the referee for giving us this valuable comment. The statements in our manuscript were misleading, as you mentioned. In our simulation, the surface albedo was assumed to be 0.056 for CEC June, 0.063 for CEC December, 0.065 for ECS June, and 0.084 for ECS December, respectively, as described in Page 7 Lines 5 – 6. These values were obtained from the database that contains the monthly global map of the Earth's surface Lambertian equivalent reflectance (LER) deduced from the Aura/OMI measurements [Kleipool et al., 2008].

The surface albedo value of 90% that you pointed out was used only for estimating the noise equivalent spectral radiance (NESR). The NESR was calculated by dividing the simulated backscattered radiance by the signal-to-noise ratio (SNR). The SNR was estimated by a linear interpolation with the three radiances (the brightest, middle and the smallest) [Private communication with K. Gerilowski]. The smallest radiance was calculated with the condition of 90% albedo and 0° SZA (cloudy condition). The middle radiance was done in 25% albedo and 45° SZA. The largest radiance was done in 5% albedo and 80° SZA. Figure GC2.1.1 shows the results of the SNR simulations. The SNR values were approximately 90 and 1400 (mean of the three spectra) at 305 nm and 340 nm, respectively.

The solar zenith angle (SZA), which corresponds to local time, is also a key parameter for simulation of the UV radiance, as you mentioned. The aim of this article is to investigate the sensitivity of the tropospheric ozone retrieval by adding the MW limb measurement to the synergistic retrieval approach. To purely bring out the improvement of the ozone retrieval sensitivity by combining several wavelength ranges, we performed the simulation in an ideal condition. Landgraf and Hasekamp (2007) showed that the increase of the ozone retrieval sensitivity was largest in the condition of small to moderate SZA (around noon). Therefore, we set the local time to be 11:46am and 00:29pm (around noon) for CEC and ECS, respectively. These local times correspond to the time when the surface ozone is likely increased because the photo-chemistry becomes active and the sensitivity of the ozone retrieval probably becomes high. We thought the local time condition in our simulation was sufficient for investigating the ozone retrieval sensitivity with combination of UV, TIR and MW in an ideal condition. To make it clear, we improved the statements in our manuscript as follows.

#### Reference

- Landgraf, J., and Hasekamp, O. P.: Retrieval of tropospheric ozone: The synergistic use of thermal infrared emission and ultraviolet reflectivity measurements from space, *Journal of Geophysical Research: Atmospheres*, 112, 2007.

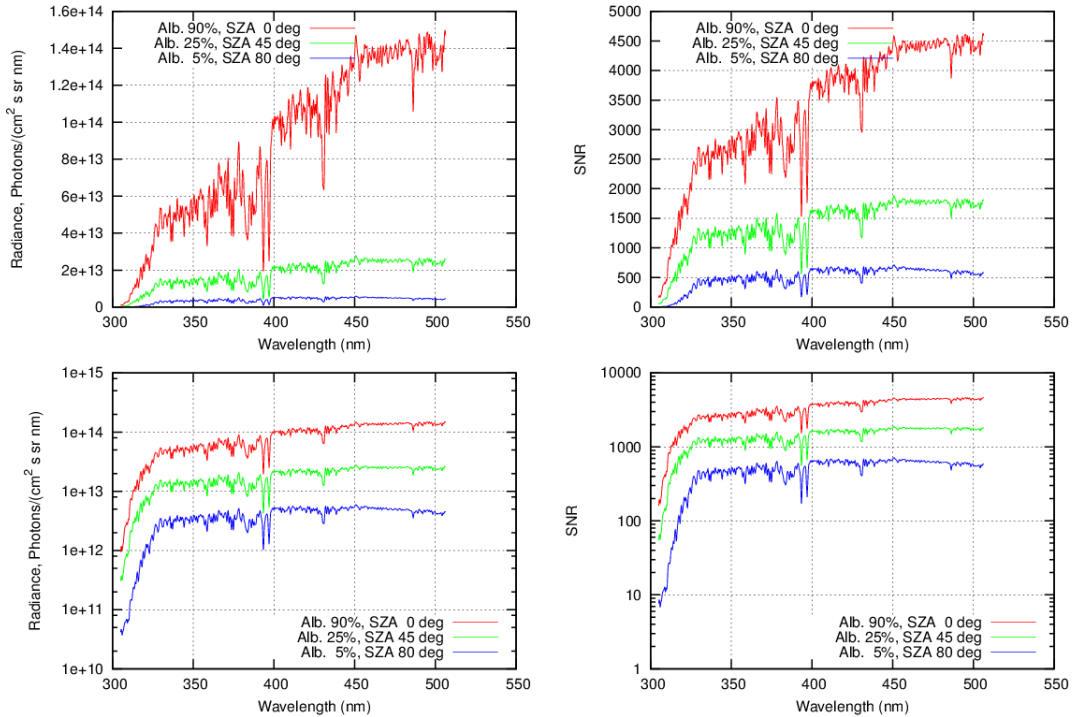


Figure GC2.1.1: Estimation of SNR in the UV wavelength range. The left column figures show the radiance spectra, and the right ones show the SNR. The top and bottom panels show the results with a linear and log scales, respectively.

**Revisions to GC 2-1**

Page 4 Lines 23 – 27

“The noise equivalent spectral radiance (NESR) for a surface albedo of 90% and the solar zenith angle of 0° were approximately 90 and 2600 at 300 and 340 nm, respectively (private communication with K. Gerilowski). The NESR value was obtained by dividing the simulated backscattered radiance by the signal-to-noise ratio (SNR).”

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“The noise equivalent spectral radiance (NESR) was obtained by dividing the simulated backscattered radiance by the signal-to-noise ratio (SNR). The SNR values were estimated by an interpolation with the small, middle and large values of the radiance calculated in the conditions that the surface albedo and solar zenith angle (SZA) of 90% and 0°, 25% and 45°, and 5% and 80°, respectively [Private communication with K. Gerilowski]. The mean values of SNR were estimated to be approximately 90 and 1400 at 305 nm and 340 nm, respectively.”

Page 5 Lines 22 – 24

“The local times around noon were set because it was shown that the ozone retrieval sensitivity was most increased in small to moderate solar zenith angles (SZAs) in the simulation study performed by Landgraf and Hasekamp, (2007).” was added.

P 24 Table 1

“~90 (300 nm), ~2600 (340 nm)”

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“90 (300 nm) – 1400 (340 nm)”

P 24 Table 1

“<sup>b</sup> In the case that surface albedo is set to 90% and solar zenith angle is set to 0°.” was deleted.

### **GC 2-2**

Retrieval error estimations show extremely high levels (i.e. PCE from Figure 3). These values (around 65%) are too high for a proper retrieval. This is the case for all retrievals using diverse spectral domains (even for single band retrievals). We expect typical retrieval errors for tropospheric ozone around 20 to 30% (IR, UV, etc) and NOT 65%. Retrieval errors of diverse tropospheric ozone columns for most single band retrievals and even UV+IR approaches implemented to real data are indeed of 20-30%. This aspect should be revised in the simulations presented by Kasai et al.. For example, retrievals can be more constrained for obtaining lower errors and therefore will provide lower sensitivity. Sensitivity is then overestimated when accepting too high errors. Therefore, the retrieval approach used in the paper to estimate averaging kernels and errors in the results presented in the paper (Figures 3, 4 and 5) should be performed again.

### **Answer to GC 2-2**

We thank the referee for giving us this valuable comment and drawing our attention to this point. We assumed 100% a priori error in our simulation to clearly see the rate of the error reduction by the measurement. This is the reason of high partial column error (PCE) such as 65%. No feasibility studies of adding MW limb measurement to the ozone synergetic retrieval approach have been reported as far as we know. Our study is the first attempt. We believe that a feasibility study in an ideal condition is quite important as the first step to introduce the MW limb measurement into the synergy retrieval of the tropospheric ozone. The values of PCE were approximately 55-65%, so that 35-45% error was reduced by the measurement spectra.



But as you pointed it out, 65% error is higher than typical retrieval error of 20-30% and showing the 65% error might be misleading for readers. Therefore, we employed the reduction rate of error (RRE) for the partial column as an alternative of PCE. The RRE value was given by

$$\text{RRE} = \frac{\text{PCE}_{\text{apriori}} - \text{PCE}_{\text{retrieved}}}{\text{PC}},$$

where  $\text{PCE}_{\text{apriori}}$  is PCE for the a priori state,  $\text{PCE}_{\text{retrieved}}$  is PCE for the retrieved state, and PC is the partial column of ozone. The manuscript was improved as follows.

### **Revisions to GC 2-2**

Page 8 Line 28 – Page 9 Line 3

“The diagonal components of  $S_a$  were the squares of the a priori error  $\sigma_a$  (100% of the log-based VMR) at each vertical pressure grid).”

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“The diagonal components of  $S_a$  were the squares of the a priori error  $\sigma_a$  at each vertical pressure grid). The value of  $\sigma_a$  was set to 100% of the log-based a priori VMR to simply quantify the error reduction from the error in the a priori error to the error in the retrieved state due to the measurement.”

Page 9 Line 24 – Page 10 Line 9

“The ozone partial column (PC) for the same vertical layers is defined as

$$\text{PC} = \sum_{i=i_{\min}}^{i_{\max}} \frac{p[i] \cdot \text{VMR}[i]}{k_B \cdot T[i]} \Delta z[i]. \quad (14)$$

Here,  $p[i]$ ,  $\text{VMR}[i]$ ,  $T[i]$  and  $\Delta z[i]$  are pressure, VMR of ozone, temperature, and the vertical length of the  $i$ th layer, respectively.  $k_B$  is the Boltzmann constant. We defined partial column error, PCE, as the relative error in PC by

$$\text{PCE} = \frac{1}{\text{PC}} \sum_{i=i_{\min}}^{i_{\max}} \frac{p[i] \cdot \varepsilon_{\text{VMR}}[i]}{k_B \cdot T[i]} \Delta z[i]. \quad (15)$$

$\varepsilon_{\text{VMR}}[i]$  is the total retrieval error in ozone VMR at the  $i$ th layer.”

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“The RRE is given by

$$\text{RRE} = \frac{\text{PCE}_{\text{apriori}} - \text{PCE}_{\text{retrieved}}}{\text{PC}}, \quad (14)$$

where  $\text{PCE}_{\text{apriori}}$  and  $\text{PCE}_{\text{retrieved}}$  are the partial column error, PCE, for the a priori state and the retrieved state, respectively. PC represents the partial column of ozone and the value of PC from the  $i_{\min}$ th vertical layer to the  $i_{\max}$ th layer is given by

$$PC = \sum_{i=i_{\min}}^{i_{\max}} \frac{p[i] \cdot VMR[i]}{k_B \cdot T[i]} \Delta z[i]. \quad (15)$$

Here,  $p[i]$ ,  $VMR[i]$ ,  $T[i]$  and  $\Delta z[i]$  are pressure, VMR of ozone, temperature, and the vertical length of the  $i$ th layer, respectively.  $k_B$  is the Boltzmann constant. The value of PCE is given by

$$PCE = \sum_{i=i_{\min}}^{i_{\max}} \frac{p[i] \cdot \varepsilon_{VMR}[i]}{k_B \cdot T[i]} \Delta z[i]. \quad (16)$$

$\varepsilon_{VMR}[i]$  is the total retrieval error in ozone VMR at the  $i$ th layer ( $\sigma_a$  for  $PCE_{\text{apriori}}$  and  $\varepsilon_x$  for  $PCE_{\text{retrieved}}$ ).

Page 11 Lines 24 – 29

“The relative error in the partial column of ozone, PCE, calculated using Eq. (15), is shown in the right column of Fig. 3. PCE generally decreased when more wavelength regions were used. PCE was approximately 55-70% in the LMT, MT and UT regions. PCE for the MW measurements alone could not be estimated in the MT and LMT because the DFS values were almost zero and there was no sensitivity with which to retrieve the ozone amount from the MW measurements.”

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“The reduction rate of error, RRE, in the ozone partial column of ozone calculated using Eq. (14), is shown in the right column of Fig. 4. The value of RRE was approximately 25-45% in the UT, MT and LMT regions. The RRE generally increased by combining more wavelength ranges in the ozone synergetic retrieval as DFS shown in Fig. 3. The RRE value for all profiles averaged in the LMT region was 36% and 39% for UV+TIR and UV+TIR+MW measurements, respectively. Adding the MW measurement made 3% increase of RRE value. A certain increase of the retrieval sensitivity of the LMT ozone was shown by DFS as well as PMS and RRE.”

### **GC 2-3**

Explicit comparison with other multispectral synergisms: The added value of the method proposed in the paper should gain great clarity with an explicit comparison with previous work on multispectral retrieval of ozone. The performance of the UV+IR+MW synergism (sensitivity and probed height) and characteristics of the instruments (spectral resolution, signal-to-noise ratio) considered in the paper should be compared to previous multispectral approaches to retrieve ozone from space. This comparison should be done with the multispectral methods implemented with real measurements (OMI and TES, Fu et al., 2013 and GOME-2 and IASI, Cuesta et al. 2013) and also future approaches (e.g. UVNS and IASI-NG, Costantino et al., 2017).

### **Answer to GC 2-3**

We deeply appreciate your valuable comment. As you mentioned, our calculation results should be validated with a comparison with previous studies of the synergetic retrieval of the tropospheric ozone. The DFS values themselves vary in the simulation situations and instrumental characteristics, so we compared a relative difference between the mean of the DFS values of UV only and TIR only and the DFS value of the UV+TIR measurements. The DFS relative difference was compared with those reported from Fu et al., (2013) and Cuesta et al., (2013). We focused on the LMT region because of our most interest.

In our simulation, the DFS values for UV only and TIR only were 0.195 and 0.210, respectively, for all profiles averaged. Their mean value is 0.203. The DFS value for the UV+TIR measurement was 0.458. The relative difference was estimated to be 126%  $(=(0.458-0.203)/0.203)$ . The same calculation was applied to the DFS values of Fu et al., (2013) and Cuesta et al., (2013) as follows. Fu et al., (2013) showed that the DFS values for the UV (OMI) and TIR (TES) were 0.10 and 0.21, respectively. Their mean value was 0.155. The DFS for UV+TIR (OMI+TES) was 0.37. The relative difference was estimated to be 139%  $(=(0.37-0.155)/0.155)$ . In the case of Cuesta et al., (2013), the DFS values over land and ocean were averaged. The DFS value of UV (GOME-2) was 0.08 for both land and ocean, and their mean value was also 0.08. In the same way, the DFS values of TIR (IASI) were 0.24 and 0.16 for land and ocean, respectively, and their mean was 0.20. The mean of the DFS values for UV and TIR was 0.14. The DFS values of UV+TIR (GOME-2+IASI) were 0.34 and 0.23 for land and ocean, respectively, and their mean was 0.285. Therefore, the relative difference between the mean of the DFS values of UV only and TIR only and the DFS value of the UV+TIR measurements was calculated to be 104%  $(=(0.285-0.14)/0.14)$ . The DFS relative difference in our simulation (126%) showed good agreement with those in the previous studies and was between the two (139% and 104%). The simulation results performed by Natraj et al., (2011) was also compared. We summarized these comparisons in one Table, and added the description in our manuscript as follows.

Costantino et al., (2017) performed multispectral synergy retrieval for the future missions of IASI-NG and UVNS. They discussed increase of the sensitivity of the tropospheric ozone retrieval by improvement of the instrument rather than combining the UV and TIR measurements, and showed that the DFS values of combination of IASI-NG and UVNS were 0.75 and 0.66 over land and ocean, respectively. Our simulation showed approximately 30% increase of DFS value in the LMT. We estimated the DFS values in the LMT ozone by adding the MW limb measurement to the IASI-NG+UVNS synergetic retrieval to be 0.98  $(=0.75 \times 1.3)$  and 0.86  $(=0.66 \times 1.3)$  for land and ocean, respectively. We added this estimation as the perspective of the future achievement of the ozone synergetic approach.

## References

- Costantino, L., Cuesta, J., Emili, E., Coman, A., Foret, G., Dufour, G., Eremenko, M., Chailleux, Y., Beekmann, M. and Flaud, J.-M.: Potential of multispectral synergism for observing ozone pollution by combining IASI-NG and UVNS measurements from the EPS-SG satellite, *Atmospheric Measurement Techniques*, 10, 1281-1298, doi:10.5194/amt-10-1281-2017, 2017.
- Cuesta, J., Eremenko, M., Liu, X., Dufour, G., Cai, Z., Höpfner, M., von Clarmann, T., Sellitto, P., Foret, G., Gaubert, B., Beekmann, M., Orphal, J., Chance, K., Spurr, R., and Flaud, J.-M.: Satellite observation of lowermost tropospheric ozone by multispectral synergism of IASI thermal infrared and GOME-2 ultraviolet measurements over Europe, *Atmospheric Chemistry and Physics*, 13, 9675-9693, 2013.
- Fu, D., Worden, J. R., Liu, X., Kulawik, S. S., Bowman, K. W., and Natraj, V.: Characterization of ozone profiles derived from Aura TES and OMI radiances, *Atmospheric Chemistry and Physics*, 13, 3445-3462, 2013.

### **Revisions to GC 2-3**

Page 11 Lines 10 – 16

“We compared our results with the previous studies of estimating the tropospheric ozone sensitivity using DFS. The DFS values for UV, TIR and UV+TIR measurements were summarized in Table 4. Scenarios of the simulation or the measurements are different among this work and the previous studies shown in Table 4, thus, the DFS values themselves should not be directly compared each other. Here we calculated the relative difference between the DFS value for the UV+TIR measurements and mean of the DFS values for UV and TIR measurements. The relative difference of our simulation for all profiles averaged was estimated to be 126%. It showed good agreement with those of Fu et al., (2013) (139%), Cuesta et al., (104%) and Natraj et al., (2011) (115%).” was added.

Page 12 Line 31 – Page 13 Line 2

“As a whole, it was shown that retrieval of ozone in the LMT was improved by adding the MW limb measurements to the UV and TIR nadir measurements.

When ozone is enhanced in the LMT and also the averaging kernel matrices of UV and TIR measurements have their peaks in this region, it may be possible to retrieve the ozone in the LMT with a DFS value of more than unity.”

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“As a whole, it was shown that retrieval of the tropospheric ozone was improved by adding the MW limb measurements to the UV and TIR nadir measurements. In the LMT region, the DFS

value was estimated to increase about 30%. The DFS value was estimated to be 0.75 and 0.66 over land and ocean, respectively, for the future mission of IASI-NG and UVNS (Constantino et al., 2017). If the MW limb measurement is implemented to this synergetic retrieval, the DFS value is estimated to increase to 0.98 and 0.86 over land and ocean, respectively. Our feasibility study showed a possibility to retrieve the ozone in the LMT with a DFS value of unity.”

#### Reference

“Costantino, L., Cuesta, J., Emili, E., Coman, A., Foret, G., Dufour, G., Eremenko, M., Chailleux, Y., Beekmann, M. and Flaud, J. M.: Potential of multispectral synergism for observing ozone pollution by combining IASI-NG and UVNS measurements from the EPS-SG satellite, Atmospheric Measurement Techniques, 10, 1281-1298, doi:10.5194/amt-10-1281-2017, 2017.” was added.

P 28 Table 4 was added.

Table 4. Comparison of DFS in the LMT region with previous studies. The method and situation to derive the DFS value are different for each study, thus, the relative difference between the DFS value for the UV+TIR measurement ( $DFS_{UV+TIR}$ ) and mean of DFS values for UV and TIR measurements ( $DFS_{UV}$ ,  $DFS_{TIR}$ ) was used for this comparison. The relative difference was calculated by  $(DFS_{UV+TIR} - \text{Mean}(DFS_{UV}, DFS_{TIR})) / \text{Mean}(DFS_{UV}, DFS_{TIR})$ .

	$DFS_{UV}$	$DFS_{TIR}$	$\text{Mean}(DFS_{UV}, DFS_{TIR})$	$DFS_{UV+TIR}$	Rel. dif.	Definition of LMT
This work	0.20	0.21	0.203	0.458	126 %	>749 hPa
Fu et al. (2013)	0.10	0.21	0.15	0.37	139 %	>700 hPa
Cuesta et al. (2013) <sup>a</sup>	0.08	0.20	0.14	0.285	104 %	<3 km
Natraj et al. (2011)	0.26	0.27	0.265	0.57	115 %	>800 hPa

<sup>a</sup> DFS values over land and ocean are averaged.

#### **GC 2-4**

Heights of maximum sensitivity for each partial column: in the paper, only the degrees of freedom for the retrieval of ozone partial column are used as diagnostic for sensitivity. However, averaging kernels explicitly provide information on the height of maximum sensitivity that varies for each retrieval method and measuring conditions. Therefore, I strongly recommend to provide the heights of maximum sensitivity for each ozone partial column retrieved from satellite spectra, and compare them explicitly for each spectral combination (UV, IR, MW, UV+IR+MW, etc)..

### Answer to GC 2-4

We thank the referee for giving us this valuable comment and drawing our attention to this point. We employed pressure grid in our simulation, and calculated the pressure of maximum sensitivity (PMS) for each partial column. The PMS was defined as the pressure of the maximum of the corresponding the sum of rows of the averaging kernel for the partial column [Cuesta et al., (2013)]. Figure GC2.4.1 shows the result of PMS calculation. Some PMSs for the single wavelength range were not located in the corresponding vertical pressure region, especially for the LMT region. It was clearly shown that PMSs were located in the corresponding upper/middle/lowermost tropospheric regions by combination of several wavelength ranges. The UV+TIR+MW combination was the only case that all PMSs for the UT, MT and LMT regions were in ranges of the corresponding vertical region. We improved our manuscript by adding information of PMS as follows.

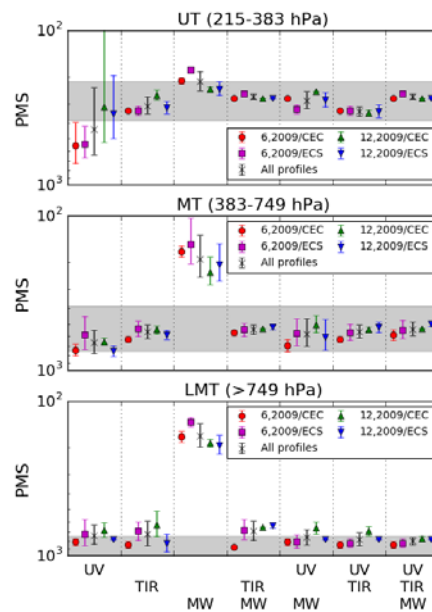


Figure GC2.4.1: Values of PMS in the upper troposphere (UT, 215-383 hPa), middle troposphere (MT, 383-749 hPa), and lowermost troposphere (LMT, >749 hPa): (red) June 2009 in CEC, (purple) June 2009 in ECS, (green) December 2009 in CEC, (blue) December 2009 in ECS and (black) all of 20 profiles. The gray shaded area represents the vertical region that corresponds to UT, MT and LMT.

### Reference

- Cuesta, J., Eremenko, M., Liu, X., Dufour, G., Cai, Z., Höpfner, M., von Clarmann, T., Sellitto, P., Foret, G., Gaubert, B., Beekmann, M., Orphal, J., Chance, K., Spurr, R., and Flaud, J. M.: Satellite observation of lowermost tropospheric ozone by multispectral

synergism of IASI thermal infrared and GOME-2 ultraviolet measurements over Europe, Atmospheric Chemistry and Physics, 13, 9675-9693, 2013.

#### **Revisions to GC 2-4**

Page 9 Line 24 – Page 10 Line 1

“We evaluated the sensitivity of ozone retrieval for seven wavelength combinations in terms of the DFS and partial column error (PCE). We calculated DFS and PCE for the UT, MT and LMT regions as follows. The value of DFS from the  $i_{\min}$ th vertical layer to the  $i_{\max}$ th layer is given by

$$\text{DFS} = \sum_{i=i_{\min}}^{i_{\max}} A[i, i] \quad (13)''$$

→

“We evaluated the sensitivity of ozone retrieval for seven wavelength combinations in terms of DFS. We calculated the DFS values for the partial column in the UT, MT and LMT regions. The value of DFS from the  $i_{\min}$ th vertical layer to the  $i_{\max}$ th layer is given by

$$\text{DFS} = \sum_{i=i_{\min}}^{i_{\max}} A[i, i]. \quad (13)''$$

We also evaluated the sensitivity of ozone retrieval using the pressure of maximum sensitivity (PMS) and the reduction rate of error (RRE) for the partial column. The PMS was defined as the pressure of the maximum of the sum of rows of the corresponding A for the ozone partial column.”

Page 10 Line 12

“The left column of Fig. 3 shows” → “Figure 3 and Table 3 show”

Page 11 Lines 17 – 23

“The pressure of maximum sensitivity, PMS, for the ozone partial column should be located in a range of the corresponding partial column. In the UT region, the PMS values for all cases were located in a range of the corresponding region (215-383 hPa) by combining more than two wavelength ranges. It was also observed in the PMS values in the MT region. But in the LMT region, only the PMS values of combination of the three wavelength ranges were located in the vertical region of LMT. The PMS value for all profiles averaged in the LMT region was 783 hPa and 808 hPa for the UV+TIR and UV+TIR+MW measurements, respectively. The PMS value was increased by approximately 3% by adding the MW measurement to the UV+TIR measurements, although the PMS value of the MW measurement itself was lower than 300 hPa in the LMT region.” was added.

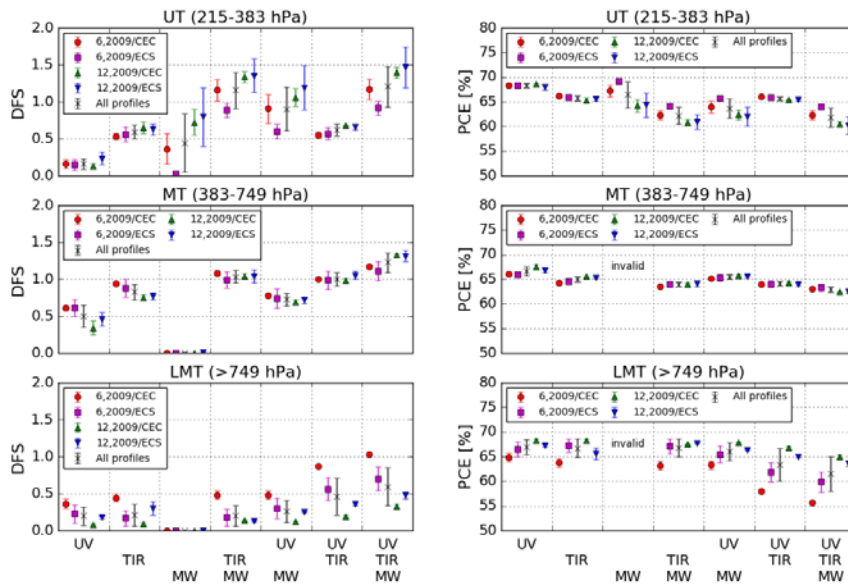


Figure 3. Values of DFS and PCE in the upper troposphere (UT, 215-383 hPa), middle troposphere (MT, 383-749 hPa), and lowermost troposphere (LMT, >749 hPa): (red) June 2009 in CEC, (purple) June 2009 in ECS, (green) December 2009 in CEC, (blue) December 2009 in ECS and (black) all of 20 profiles.

→

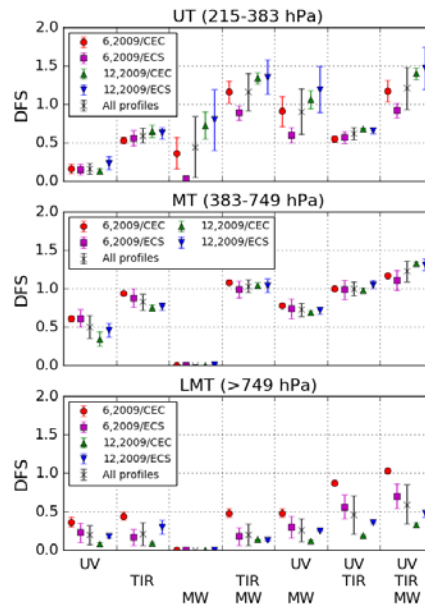


Figure 3. Values of DFS in the upper troposphere (UT, 215-383 hPa), middle troposphere (MT, 383-749 hPa), and lowermost troposphere (LMT, >749 hPa): (red) June 2009 in CEC, (purple) June 2009 in ECS, (green) December 2009 in CEC, (blue) December 2009 in ECS and (black) all of 20 profiles.



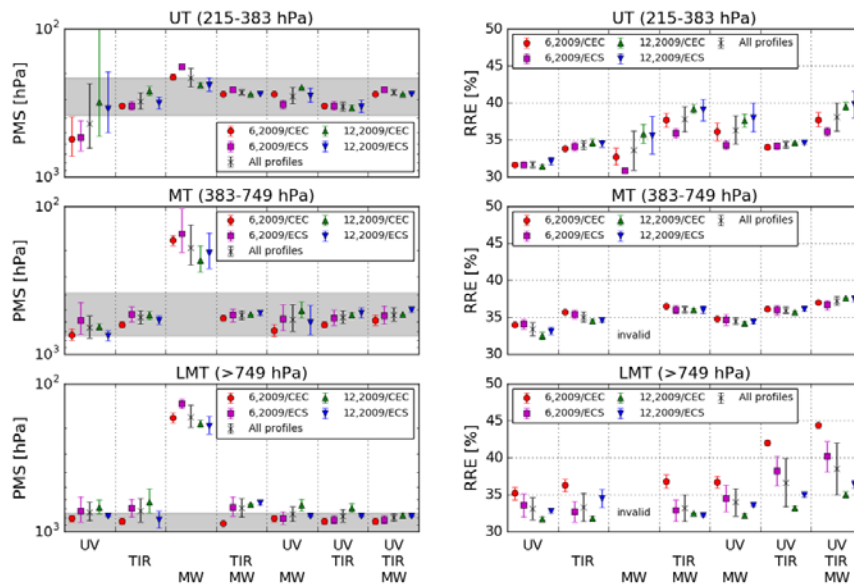


Figure 4. Same as Fig. 3 but for the PMS (left) and RRE (right). The gray shaded area represents the vertical region that corresponds to UT, MT and LMT.

Pages 23 – 24 Figures 5 and 6

The figure numbers of the averaging kernel plots were renumbered to Figures 5 and 6.

### GC 2-5

Additional terms for error estimations: for such a new multispectral retrieval, I strongly recommend to estimate the contribution of additional terms of errors such as cross errors from joint fit of surface albedo and emissivity and other systematic errors (atmospheric and surface temperature, water vapor, etc).

### Answer to GC 2-5

We deeply appreciate the referee for pointing it out. The main purpose of this feasibility study is evaluation of adding the MW limb measurement to the synergetic retrieval of the tropospheric ozone. We added the description of large error sources in the MW limb measurement of spectroscopic parameters. Time difference between the MW limb measurement and UV and TIR nadir measurements was also discussed as follows.

### Revisions to GC 2-5

Page 12 Lines 16 – 30

“In this study, we showed that an introduction of the MW limb measurement had a certain effect

to increase the sensitivity of the tropospheric ozone retrieval. However, following issues might cause bias and uncertainties in the retrieval results and should be considered to implement this retrieval method to real measurements. Discrepancy in spectroscopic parameters for several wavelength ranges is one of the most important error sources. For the ozone retrieval using the MW limb measurement, spectroscopic parameters are the largest error sources. It was reported that approximately 3-5% error was caused by uncertainties in air-broadening coefficient and line intensity in the case of the SMILES observation (Kasai et al., 2013). It is comparable to the approximately 4% uncertainty in the spectroscopic parameters in the UV and TIR wavelength ranges (Gratien et al., 2010). The tangent height correction can also be a large error source for the MW limb measurement (Kasai et al., 2013). The tangent height is a key parameter to determine the field-of-view, thus uncertainty in the tangent height causes discrepancy of the atmospheric layer assumed in the simulation and the true atmospheric layer. This discrepancy critically affects to the retrieval of the ozone amount in both the stratosphere and the troposphere, and also might cause bias for correction of time delay between the MW limb measurement and the other nadir measurements. In this study, we assumed instruments onboard the ISS (low orbit) and the time difference of approximately five minutes could be ignored. If the time difference was long and its correction was required, three-dimensional atmospheric modeling should be performed including the field-of-view of the MW limb measurement.” was added.

#### **GC 2-6**

A better description of the atmospheric scenario is needed. How model and airborne data are combined? How the 20 cases are chosen? What is the main characteristic (time of the day, region, ozone load, surface and atmospheric properties) of each scenario? These aspects should be explicitly commented and described in the text of the paper.

#### **Answer to GC 2-6**

We deeply appreciate this suggestion. We synthesized the vertical profiles of ozone, water vapor and temperature from three atmospheric profiles; a one-way nested global-regional air quality forecasting (AQF) system [Takigawa et al., 2007, 2009], the Modern Era Retrospective-Analysis for Research and Applications (MERRA) data [Rienecker et al., 2011] and the COSPAR International Reference Atmosphere (CIRA) [Fleming et al., 1990]. The aircraft observation data was used in the MERRA data, but we did not use the aircraft observation data directly. The statement “We made a total of 20 atmospheric scenarios from the model calculation and air-born observations made over two Asian areas (CEC and ECS) in June and December 2009. (Page 4 Lines 32 – 33)” was quite misleading, and we improved this statement as follows.

The reason why we chose the 20 scenarios over Asian area, CEC and ECS, was that China is one of the largest air-polluted countries in the world and the prediction of transboundary pollution is one of the main purposes of the Japanese future missions. The CEC, located between Beijing and Shanghai, is the area where largest amount of ozone in the lowermost troposphere was observed over East Asia from the Aura/OMI measurement [Hayashida et al., 2015]. This work also showed the amount of ozone in the lowermost troposphere in the urban area was largest in June (summer season) and smallest in December (winter season). The ECS was selected for a comparison between the urban area (CEC) and ocean (ECS). The main characteristics of the 20 scenarios are shown in Table 2, and we added the explanation of the 20 scenarios in our manuscript as follows.

#### References

- Fleming, E. L., Chandra, S., Barnett, J. J., and Corney, M.: Zonal mean temperature, pressure, zonal wind and geopotential height as functions of latitude, *Advances in Space Research*, 10, 11-59, 1990.
- Hayashida, S., Liu, X., Ono, A., Yang, K., and Chance, K.: Observation of ozone enhancement in the lower troposphere over East Asia from a space-borne ultraviolet spectrometer, *Atmospheric Chemistry and Physics*, 15(17), 9865--9881, 2015.
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- Takigawa, M., Niwano, M., Akimoto, H., and Takahashi, M.: Development of a one-way nested global-regional air quality forecasting model, *Sola*, 3, 81-84, 2007.
- Takigawa, M., Niwano, M., Akimoto, H., Takahashi, M., and Kobayashi, K.: Projection of surface ozone over East Asia in 2020, *Journal of Agriculture Meteorology*, 65, 161-166, 2009.

#### **Revisions to GC 2-6**

Page 5 Lines 14 – 30

“We performed a feasibility study of tropospheric ozone retrieval for typical atmospheric scenarios in summer and winter. We made a total of 20 atmospheric scenarios from the model calculation and air-born observations made over two Asian areas (CEC and ECS) in June and December 2009. The characteristics of the 20 atmospheric scenarios are presented in Table 2, and

vertical profiles of ozone, temperature and water vapor are shown in Figure 2. We assumed the following quasi-clear sky cases for all scenarios. A no-cloud condition was considered for all wavelength ranges. Basic background aerosol was taken into account only in the UV calculation. We used the vertical profiles of urban and maritime aerosols of a standard mixing state that were described in Hess et al. (1998). The scale height of the vertical profiles that we used was 3 km. These profiles were adjusted to be 0.2 of the total optical thicknesses of the aerosols (moderate pollution).”

→

“ We performed a feasibility study of tropospheric ozone retrieval for typical atmospheric scenarios in summer and winter. The target area of this study is East Asia, one of most serious ozone polluted areas and the source of ozone intercontinental transport toward North America from Asia. We chose two observation points in East Asia, 35°N, 116.5°E (Central-East China, CEC, located between Beijing and Shanghai) and 31°N, 127.25°E (East China Sea, ECS). The CEC is the area where largest amount of the boundary layer ozone was observed from the Aura/OMI measurement (Hayashida et al., 2015). The ECS was chosen for a comparison of urban area and ocean. We selected June and December for representatives of summer and winter seasons, respectively. Hayashida et al., (2015) reported the amount of the boundary layer ozone in the area near CEC was maximized in June and minimized in December. The observation time was set to 04:00am GMT, which corresponds to 11:46am and 00:29pm for CEC and ECS local times, respectively. The local times around noon were set because it was shown that the ozone retrieval sensitivity was most increased in small to moderate solar zenith angles (SZAs) in the simulation study performed by Landgraf and Hasekamp (2007).

We made a total of 20 atmospheric scenarios over the two Asian areas (CEC and ECS) in June and December 2009. The characteristics of the 20 atmospheric scenarios are presented in Table 2, and the vertical profiles of ozone, temperature and water vapor are shown in Figure 2. We discuss the simulation results, dividing into four cases (CEC in June, ECS in June, CEC in December and ECS in December). The ozone partial column (PC) in LMT at CEC in June was largest (approximately  $5 \times 10^{21} \text{ m}^{-2}$ ) among those of four cases, and the smallest value (approximately  $2 \times 10^{21} \text{ m}^{-2}$ ) was taken from the case at CEC in December.”

Page 6 Line 27 – Page 7 Line 5

“The information on surface albedo for simulating UV radiance spectra was taken from the database described by Kleipool et al. (2008). This database contains the monthly global maps of the Earth’s surface Lambertian equivalent reflectance (LER) deduced from the Aura/OMI measurements. We assumed that surface albedo was constant in the selected UV ranges (305-340 nm).”

→

“We assumed the following quasi-clear sky cases for all scenarios. A no-cloud condition was considered for all wavelength ranges. Basic background aerosol was taken into account only in the UV calculation. The aerosol profile was included as a known parameter because it was reported that the inclusion of aerosol profile changed within 2% of the LMT ozone amount in case of the Aura/OMI measurement (Hayashida et al., 2015). We used the vertical profiles of urban and maritime aerosols of a standard mixing state that were described in Hess et al., (1998). These profiles were adjusted to be 0.2 of the total optical thicknesses of the aerosols (moderate pollution). The aerosol profile was not included in the TIR calculation. The extinction of radiation due to aerosol particles with a scale of approximately 9.6  $\mu\text{m}$ , which corresponds to the wavelength of the TIR range, is negligibly small for the synergetic retrieval of the LMT ozone with the TIR measurement (e.g., Natraj et al., 2011). We assumed that surface albedo was constant in the selected UV ranges (305-340 nm). The information on surface albedo for simulating UV radiance spectra was taken from the database described by Kleipool et al. (2008). This database contains the monthly global maps of the Earth's surface Lambertian equivalent reflectance (LER) deduced from the Aura/OMI measurements.”

### **GC 2-7**

Spectroscopy coherence should be mentioned as an important issue for real retrievals. There exist previous multispectral retrievals combining microwave spectra with other domains? Spectroscopic consistency has been analyzed as for UV and IR (e.g. Gratien et al., 2010)?

### **Answer to GC 2-7**

We thank the referee for drawing our attention to this point. As far as we know, our study is the first attempt to introduce the MW measurement to the synergetic retrieval of the tropospheric ozone. We referred Gratien et al., (2010) and discussed the tropospheric ozone retrieval error due to uncertainties in the spectroscopic parameters in the MW ranges as follows.

### **References**

- Gratien, A., Picquet-Varrault, B., Orphal, J., Doussin, J.-F., and Flaud, J. M.: New Laboratory Intercomparison of the Ozone Absorption Coefficients in the Mid-infrared (10  $\mu\text{m}$ ) and Ultraviolet (300-350 nm) Spectral Regions, *The Journal of Physical Chemistry A*, 114(37), 10045-10048, doi:10.1021/jp103992f, 2010.

### **Revisions to GC 2-7**

Page 12 Lines 18 – 23

“Discrepancy in spectroscopic parameters for several wavelength ranges is one of the most important error sources. For the ozone retrieval using the MW limb measurement, spectroscopic parameters are the largest error sources. It was reported that approximately 3-5% error was caused by uncertainties in air-broadening coefficient and line intensity in the case of the SMILES observation (Kasai et al., 2013). It is comparable to the approximately 4% uncertainty in the spectroscopic parameters in the UV and TIR wavelength ranges (Gratien et al., 2010).”

#### Reference

“Gratien, A., Picquet-Varrault, B., Orphal, J., Doussin, J.-F., and Flaud, J. M.: New Laboratory Intercomparison of the Ozone Absorption Coefficients in the Mid-infrared (10  $\mu\text{m}$ ) and Ultraviolet (300-350 nm) Spectral Regions, *The Journal of Physical Chemistry A*, 114(37), 10045-10048, doi:10.1021/jp103992f, 2010.” was added.

### **GC 2-8**

Aerosol in the UV: how are they accounted in simulations? Are aerosols jointly fitted or considered as perfectly known? The error of this assumption should be explicitly analysed and quantified.

### **Answer to GC 2-8**

Thank you so much for pointing it out. We used the typical aerosol profiles in the urban and ocean area for the calculation at CEC and ECS, respectively. The aerosol profile was taken into account as known parameter because the inclusion of aerosol profile changed within 2% of the LMT ozone amount in case of the Aura/OMI measurement (Hayashida et al., 2015). Our main purpose of this feasibility study is evaluation of the tropospheric ozone retrieval sensitivity by adding the MW limb measurement, and the 2% change in the LMT ozone retrieval of the aerosol profile could be ignored.

#### References

- Hayashida, S., Liu, X., Ono, A., Yang, K., and Chance, K.: Observation of ozone enhancement in the lower troposphere over East Asia from a space-borne ultraviolet spectrometer, *Atmospheric Chemistry and Physics*, 15(17), 9865--9881, 2015.

### Revisions to GC 2-8

Page 6 Lines 28 – 30

“The aerosol profile was included as a known parameter because it was reported that the inclusion of aerosol profile changed within 2% of the LMT ozone amount in case of the Aura/OMI measurement (Hayashida et al., 2015).” was added.

### GC 2-9

In Fig. 3, why DFS for IR+MW are lower than for IR only at the LMT???? Please justify and verify calculations.

### Answer to GC 2-9

We thank the referee for drawing our attention to this point. We guess that you pointed the DFS results in the LMT for the ECS December case. We carefully checked our simulation process again, but could not find any mistakes. Figure GC2.9.1 shows the averaging kernel of the TIR simulation for the profile #19 (ECS December). We found a discontinuous in the averaging kernel matrix in the LMT region. In general, the discontinuity in the averaging kernel occurs because of mathematical issues not atmospheric physical issues. We added the description of this point in our manuscript as follows.

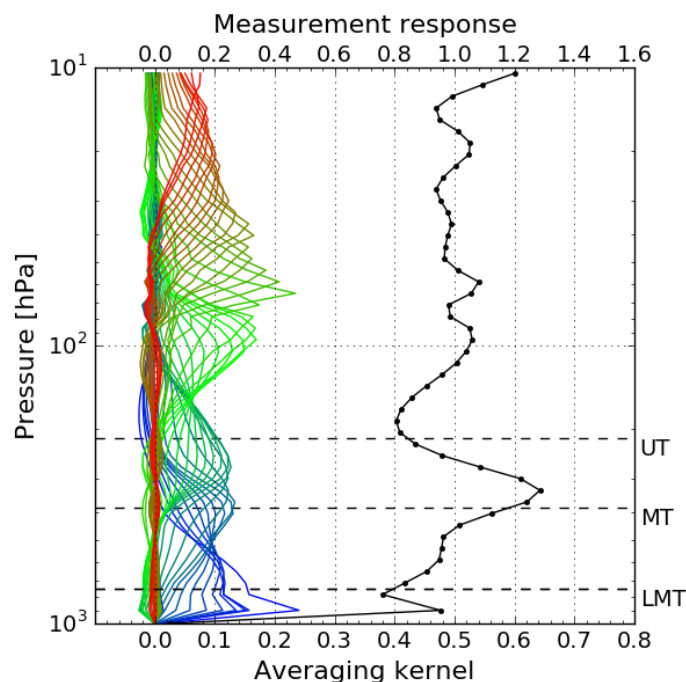


Figure GC2.9.1: Averaging kernel of the TIR simulation for the profile #19.

### **Revisions to GC 2-9**

Page 11 Lines 5 – 9

“We note that the DFS value for the TIR measurement in the case at ECS in December 2009 was larger than that for TIR+MW measurement. The averaging kernel matrix in the LMT region for the TIR measurement of this case was discontinuously large. In general, the discontinuity in the averaging kernel occurs because of mathematical issues not atmospheric physical issues, thus we avoid any scientific discussion with the DFS value for the TIR measurement in this case (ECS in December 2009).” was added.

### **MC 2-1**

In page 2, lines 61-65: Here the performance of the combination of is done for that of OMI and TES. Similar characteristic should be provided for the synergism of IASI and GOME2 measurements, in terms of DFS, errors and altitude of maximum sensitivity.

### **Answer to MC 2-1**

We thank the referee for pointing it out. We improved our manuscript as follows.

### **Revisions to MC 2-1**

Page 3 Lines 6 – 10

“Although the DFS value of boundary layer ozone is much improved by combining UV and TIR measurements, it is less than unity.”

→

“The DFS values for ozone up to 3 km were estimated to be  $0.34\pm 0.04$  and  $0.23\pm 0.04$  over land and ocean, respectively, on 19-20 August 2009 over Europe (see Table 1 in Cuesta et al., (2013)). The corresponding heights of maximum sensitivity were  $2.20\pm 0.50$  km and  $3.42\pm 0.59$  km, respectively. The DFS values were approximately 50% increased by combining the GOME-2 and IASI measurements, compared with the IASI measurement only.”

### **MC 2-2**

Page 3: please provide the units for NESR.

### **Answer to MC 2-2**

The noise equivalent spectral radiance (NESR) was obtained by dividing the simulated backscattered radiance by the signal-to-noise ratio (SNR). Therefore, the unit of NESR is the



same as the radiance. In our UV simulation using the SCIATRAN model, the unit of radiance is [photons / (cm<sup>2</sup> s sr nm)].

### **MC 2-3**

Page 3: The UV domain <305 nm does provide useful information on stratospheric ozone. Please clarify why it is not used in the retrieval.

### **Answer to MC 2-3**

We would like to thank the Referee for pointing it out. As you mentioned, the UV wavelength range (< 305 nm) might be useful for the retrieval of the stratospheric ozone. But the MW limb measurement is much more useful for it. We decided not to use the UV wavelength ranges shorter than 305 nm to clearly show effects of adding the MW measurement to the synergetic ozone retrieval.

### **Revisions to MC 2-3**

Page 4 Lines 15 – 18

“The UV wavelength ranges shorter than 305 nm is useful for the stratospheric ozone retrieval (e.g., Bak et al., 2012). But the MW limb measurement is more sensitive for the stratospheric ozone. We excluded the shorter UV wavelength ranges to clearly show whether the stratospheric ozone retrieval by the MW measurement improves the tropospheric ozone retrieval sensitivity.” was added.

### **MC 2-4**

Page 3 : reference Boynard et al 2009 for IASI is not appropriate. Please use Clerbaux et al 2009 instead.

### **Answer to MC 2-4**

Thank you so much for pointing it out. We changed the reference to Clerbaux et al., 2009.

### **Revisions to MC 2-4**

Page 4 Line 30

“... (Boynard et al., 2009).” → “... (Clerbaux et al., 2009)”

#### Reference

“Boynard, A., Clerbaux, C., Coheur, P.-F., Hurtmans, D., Turquety, S., George, M., Hadji-Lazaro, J., Keim, C., and Meyer-Arnek, J.: Measurements of total and tropospheric ozone from IASI: comparison with correlative satellite, ground-based and ozonesonde observations, Atmospheric chemistry and physics, 9, 6255--6271, 2009.” was removed.

#### **MC 2-5**

Page 7, lines 1-3 : Also Fu et al 2013 and Cuesta et al 2013, Landgraf et al 2007 showed the advantages of a combined UV+IR retrieval of ozone. Please add these references.

#### **Answer to MC 2-5**

Thank you so much for pointing it out. We added the references as follows.

#### **Revisions to MC 2-5**

Page 12 Lines 11 – 12

“as shown in Worden et al., (2007) and Natraj et al., (2011)”

→

“as shown in the previous studies (Landgraf and Hasekamp, 2007; Worden et al., 2007; Natraj et al., 2011; Cuesta et al., 2013; Fu et al., 2013)”

**Other corrections by ourselves**

Page 10 Lines 28 – 19

“from 1.00 in the TIR+UV measurements to 1.23 (about a 20% increase)”

→

“from 1.00 to 1.23 (about 23% increase) for the TIR+UV measurements”

Page 11 Line 1

“in the MT and LT” → “in the UT and MT”

Page 11 Line 5

“a 40%” → “30%”

Page 12 Line 2

“This shows that retrieval combining UV, TIR and MW measurements can possibly retrieve the ozone amount in the LMT region with a DFS value of unity when the LMT ozone is enhanced as large as approximately  $5 \times 10^{21} \text{ m}^{-2}$ .” was removed.

# Vertical profile of tropospheric ozone derived from synergetic retrieval using three different wavelength ranges, UV, IR, and Microwave: sensitivity study for satellite observation

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**Abstract.** We performed a feasibility study to constraining the vertical profile of the amount of ozone in the troposphere by using a synergetic retrieval method on multiple spectra, i.e., ultraviolet (UV), thermal infrared (TIR) and microwave (MW) ranges, measured from space. A quantitative evaluation of the sensitivity of the tropospheric retrieval by adding the MW measurement to the UV and TIR measurements was reported for the first time by this work. The urban and sea areas in East Asia in summer and winter seasons were selected for the feasibility study. Geometry of line-of-sight was nadir down-looking for UV and TIR measurements, and limb-sounding for MW measurement. The sensitivities of retrieved ozone in the upper troposphere (UT), middle troposphere (MT) and lowermost troposphere (LMT) were estimated using values of the degree of freedom for signal (DFS), pressure of maximum sensitivity, error reduction rate from the a priori error, and averaging kernel matrix, derived based on the optimal estimation method. The measurement noises were assumed at the same level as the currently available instruments. The weighting functions for the UV, TIR and MW ranges were calculated using the SCIATRAN radiative transfer model, the Line-By-Line Radiative Transfer Model, and the Advanced Model for Atmospheric Terahertz Radiation Analysis and Simulation, respectively. The DFS value was increased by approximately 96 %, 23 % and 30 % by adding the MW measurements to the combination of UV and TIR measurements in the UT, MT and LMT regions, respectively. We found that the MW measurement increased the DFS value of the LMT ozone; nevertheless, the MW measurement alone has no sensitivity for the LMT ozone. The pressure of maximum sensitivity of the LMT ozone was also increased by adding the MW measurement. It might indicate that better information of the LMT ozone can be deduced by adding constraints on the UT and MT ozone from the MW measurement. The results of this study will be implemented in the Japanese air-quality monitoring missions, APOLLO, GMAP-Asia and uvSCOPE.

## 1 Introduction

The World Health Organization (WHO) estimates that around seven million people died as a result of the effects of air pollution in 2012 (WHO, 2014), and it cites air pollution as being one of the world's largest single environmental health risk. Ozone in particular causes serious damage for human health and agricultural crops. Tropospheric ozone has been increasing globally at rates of 0.3–1.0 ppb yr<sup>-1</sup> over past few decades in the northern hemisphere (Dentener et al., 2010, and references therein). Ozone is formed by sunlight-driven oxidation from ozone precursors such as methane (CH<sub>4</sub>), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), and nitrogen oxides (NO<sub>x</sub>) in the troposphere. Monitoring of the amount of the tropospheric ozone is required to understand the current status and to make forecasts of future ozone amount.

Ozone plays different roles in different altitude regions in the troposphere. It is well known that surface ozone is a harmful pollutant that has a detrimental impact on the health of humans and plants and is responsible for significant reduction in crop yields. The lifetime of ozone in the free troposphere ranges from a few days to weeks, so that the transport scale of ozone is potentially intercontinental and hemispheric. Upper tropospheric ozone is the third most important warming gas and is responsible for a large part of the human enhancement of the global greenhouse effect. For further understanding these different characteristics of tropospheric ozone, it is important to obtain information on the vertical distribution of ozone separately in the lowermost troposphere (LMT), middle troposphere (MT), and upper troposphere (UT) on a global scale.

Ozone has been observed from space in a variety of spectral ranges, including the ultraviolet (UV), visible (VIS), thermal infrared (TIR), and microwave (MW) with different observation geometries (nadir-looking and limb-sounding). Observations at different wavelengths have sensitivity to ozone at different altitudes. Generally, nadir-looking observations in the UV/VIS range are sensitive to ozone in the LMT (e.g., the Ozone Monitoring Instrument, OMI, onboard the Aura satellite (Levelt et al., 2006) and the second Global Ozone Monitoring Experiment, GOME-2, onboard the MetOp satellite (Munro et al., 2006)), while nadir-looking in the TIR range is sensitive to ozone in the MT (e.g., the Thermal Emission Spectrometer, TES, onboard the Aura satellite (Osterman et al., 2008) and the Infrared Atmospheric Sounding Interferometer, IASI, onboard the MetOp satellites (Clerbaux et al., 2009)). Limb-sounding and stellar/solar-occultation is used to sound ozone in the stratosphere and above. Limb-sounding in the UV/VIS region sounds ozone in the stratosphere, and stellar occultation instruments observe ozone above the stratosphere (e.g., the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography, SCIAMACHY, (Brinksma et al., 2006) and the Global Ozone Monitoring by Occultation of Stars, GOMOS, (Kyrölä et al., 2004) both onboard the Envisat satellite). Limb-sounding in the MW spectral range is sensitive at altitudes above the UT (e.g., the Microwave Limb Sounder, MLS, onboard the Aura satellite (Waters et al., 2006) and the Superconducting Submillimeter-Wave Limb-Emission Sounder, SMILES, onboard the International Space Station (Kikuchi et al., 2010)).

Measurement using several wavelength ranges is an advanced method of deriving a vertically resolved ozone profile. Ziemke et al. (2006) derived the global distribution of the tropospheric ozone column by subtracting the stratospheric ozone column measured using the MLS MW spectra from the total ozone column measured using the OMI UV spectra. A feasibility study of the tropospheric ozone retrieval using the optimal estimation method (OEM) (Rodgers, 2000) combining UV and TIR measurements was performed by Landgraf and Hasekamp (2007). Worden et al. (2007) implemented the concept of synergetic

retrieval on the OMI and TES measurements. Natraj et al. (2011) showed that the retrieval sensitivity of the LMT is improved by combining UV and TIR measurements. Fu et al. (2013) implemented a synergetic retrieval of boundary layer ozone using the UV and TIR spectra of the OMI and TES measurements. A value of the degree of freedom for signal (DFS) for ozone from the surface to 700 hPa was estimated to be  $0.37 \pm 0.09$  for 22 coincident measurements among OMI, TES, and ozonesonde  
5 from 2004 to 2008 (see Table 2 in Fu et al. (2013)). Cuesta et al. (2013) also performed a synergetic retrieval of boundary layer ozone, using the GOME-2 (for UV) and IASI (for TIR) measurements. The DFS values for ozone up to 3 km were estimated to be  $0.34 \pm 0.04$  and  $0.23 \pm 0.04$  over land and ocean, respectively, on 19–20 August 2009 over Europe (see Table 1 in Cuesta et al. (2013)). The corresponding heights of maximum sensitivity were  $2.20 \pm 0.50$  km and  $3.42 \pm 0.59$  km, respectively. The DFS values were approximately 50 % increased by combining the GOME-2 and IASI measurements, compared with the  
10 IASI measurement only. The other approach to retrieve the tropospheric ozone profile using neural network technique was performed with the SCIAMACHY nadir measurements in the UV and VIS ranges (Sellitto et al., 2012a, b). They also showed a significant availability of combining several wavelength ranges to retrieve the tropospheric ozone profile.

Our idea is to add MW measurements to the synergetic retrieval of the tropospheric ozone. To the best of our knowledge, no study has attempted to show how MW measurements improve the retrieval of the vertical profile of tropospheric ozone.  
15 In this study, we performed a feasibility study of obtaining a vertically resolved ozone amount in the troposphere by using synergetic retrieval from a combination of UV, TIR and MW measurements covering wide wavelength ranges. This work can be of benefit to future missions for air-pollution, such as Air POLLution Observation (APOLLO), Geostationary mission for Meteorology and Air Pollution (GMAP-Asia) (Kasai et al., 2011), uvSCOPE (Fujinawa et al., 2015), and air pollution prediction project in National Institute of Information and Communication Technology (NICT). The objective of APOLLO  
20 and GMAP-Asia mission is to measure short-lived climate pollutants for monitoring global pollution and climate change. The missions of APOLLO and GMAP-Asia assume atmospheric monitoring from the International Space Station (ISS) and from geostationary orbit, respectively. The uvSCOPE mission, a candidate for the earth observation section of ISS, aims to detect hot-spots of air pollutant with a high horizontal resolution (such as  $1 \times 1$  km<sup>2</sup>) for better understanding of the inventory of air pollution. The target of NICT air pollution prediction project is to make health index to mitigation of air pollution disasters  
25 using high-horizontal resolution (a few km scale) pollution forecasting from multiple data-sets, such as satellite observation, ground-based observation, and in-situ observation data.

In this paper, we report a feasibility study of the tropospheric ozone retrieval based on the concept of APOLLO, i.e., to obtain vertically resolved information of ozone within the troposphere not only at the boundary layer but also in the middle and upper troposphere by utilizing synergetic observation afforded by UV, TIR and MW instruments. Our major aim of this  
30 feasibility study is to evaluate the sensitivity of the tropospheric ozone retrieval adding the MW measurement to the multi-spectral synergetic retrieval. Thus, the feasibility study was performed under an ideal condition for the synergetic retrieval of the tropospheric ozone.

## 2 Observation scenario

### 2.1 Observation wavelength region and geometry

The observation scenario follows the concept of the APOLLO mission. We assumed three spectrometers equipped in ISS that observe the three wavelength ranges of UV, TIR, and MW. Figure 1 shows the observation geometries for the three spectrometers. The UV and TIR instruments use nadir down-looking, and the MW measurement uses limb-sounding at tangent heights from 10 to 80 km. In this feasibility study, we assumed spherically homogeneous atmosphere along the line-of-sight of the MW measurement. The height of ISS was assumed to be 300 km in our simulation. The azimuthal direction of the field-of-view of the MW limb-sounding was set to parallel to the ISS's orbital motion. The tangent point of the MW limb-sounding passes the UV and TIR nadir down-looking point approximately five minutes before the UV and TIR nadir down-looking. **The time delay of 5 minutes corresponds to approximately 6 km transport when we assume a typical value of horizontal wind speed in the troposphere and the stratosphere of 1.2 km/min (Fleming et al., 1988), which is smaller than typical horizontal resolution of the MW limb measurement (8 km for Aura/MLS (Waters et al., 2006)). Therefore, we ignored this time difference between the UV and TIR measurements and MW measurement.** Table 1 is a summary of the specification of the three assumed instruments and the radiative transfer models used in this study.

The wavelength range of the simulation of the UV measurement was set to 305–340 nm. **The UV wavelength ranges shorter than 305 nm is useful for the stratospheric ozone retrieval (e.g., Bak et al., , 2012). In our simulation, we added the MW limb measurement which is more sensitive for the stratospheric ozone. We excluded the shorter UV wavelength ranges to clearly show whether the stratospheric ozone retrieval by the MW measurement improves the tropospheric ozone retrieval sensitivity.** We also decided not to include the VIS (340–505 nm) range in this study, although the benefit of adding VIS wavelengths has been reported (e.g., Sellitto et al., 2012a, b). The reason why we excluded these ranges is because the wavelength dependence of the surface reflectance, absorption of NO<sub>2</sub> and the Ring effect were out of the scope of the study. The spectral resolution, defined as the full width at half maximum (FWHM), and the sampling step were assumed to be 0.6 nm and 0.2 nm, respectively. **The noise equivalent spectral radiance (NESR) was obtained by dividing the simulated backscattered radiance by the signal-to-noise ratio (SNR). The SNR values were estimated by an interpolation with the small, middle and large values of the radiance calculated in the conditions that the surface albedo and solar zenith angle (SZA) of 90 % and 0°, 25 % and 45°, and 5 % and 80°, respectively [Private communication with K. Gerilowski]. The mean values of SNR were estimated to be approximately 90 and 1400 at 305 nm and 340 nm, respectively.**

We assumed that the nadir-viewing TIR instrument would be a Fourier transform spectrometer covering the TIR spectral range (980–1080 cm<sup>-1</sup>) including the ozone ν<sub>3</sub> absorption band, 9.6 μm (1045 cm<sup>-1</sup>) as TES (Osterman et al., 2008) and IASI (Clerbaux et al., 2009). We set the maximum optical path difference to 8.33 cm, which corresponds to a spectral resolution of 0.12 cm<sup>-1</sup> and calculated the noise equivalent differential temperature for each wavelength, assuming that the SNR is a constant value of 300 in the entire spectral range.

Several ozone transitions in the microwave/submillimeter range have been employed by recent space-borne instruments, e.g., 206.1 and 235.7 GHz for Aura/MLS (Waters et al., 2006), 501.8 and 544.6 GHz for Odin/SMR (Urban et al., 2005),

and 625.4 GHz for Aura/MLS and JEM/SMILES (Kikuchi et al., 2010). The MW limb-sounding instrument considered in this study was designed for covering two frequency bands, i.e., the 350 GHz band (345–357 GHz) and 645 GHz band (639–651 GHz). There are ozone lines at 352.3, 352.8, and 355.0 GHz in the former and at 640.1, 642.3, 644.8, 645.6, 647.8, and 650.7 GHz in the latter. These frequency bands were selected for detection of not only ozone but also other molecules related to global warming and air-pollution ( $\text{H}_2\text{O}$ ,  $\text{CO}$ ,  $\text{CH}_3\text{CN}$ ,  $\text{N}_2\text{O}$ ,  $\text{SO}_2$ ,  $\text{H}_2\text{CO}$ , and  $\text{HNO}_3$ ). The channel separation width of the spectrometer was assumed to be 25 MHz. The frequency resolution, defined by FWHM was set to be identical to the channel separation width. The antenna diameter was assumed to be 40 cm. The Earth's limb was assumed to be scanned vertically from 10 to 80 km with an interval of 2 km and total 35 spectra were acquired in one vertical limb scan. We also assumed that a typical integration time was 0.5 seconds for one spectrum accumulation and it took 17.5 seconds for one vertical limb scan. The brightness temperature noise was estimated to be 0.7 K and 1.7 K for the 350 GHz and 645 GHz bands, respectively, assuming the system noise temperature of a typical Schottky-barrier mixer (2500 K and 6000 K for 350 GHz band and 645 GHz band, respectively).

## 2.2 Atmospheric conditions

We performed a feasibility study of tropospheric ozone retrieval for typical atmospheric scenarios in summer and winter. The target area of this study is East Asia, one of most serious ozone polluted areas and the source of ozone intercontinental transport toward North America from Asia. We chose two observation points in East Asia,  $35^\circ\text{N}$ ,  $116.5^\circ\text{E}$  (Central-East China, CEC, located between Beijing and Shanghai) and  $31^\circ\text{N}$ ,  $127.25^\circ\text{E}$  (East China Sea, ECS). The CEC is the area where largest amount of the boundary layer ozone was observed from the Aura/OMI measurement (Hayashida et al., 2015). The ECS was chosen for a comparison of urban area and ocean. We selected June and December for representatives of summer and winter seasons, respectively. Hayashida et al. (2015) reported the amount of the boundary layer ozone in the area near CEC was maximized in June and minimized in December. The observation time was set to 04:00am GMT, which corresponds to 11:46am and 00:29pm for CEC and ECS local times, respectively. The local times around noon were set because it was shown that the ozone retrieval sensitivity was most increased in small to moderate solar zenith angles (SZAs) in the simulation study performed by Landgraf and Hasekamp (2007).

We made a total of 20 atmospheric scenarios over the two Asian areas (CEC and ECS) in June and December 2009. The characteristics of the 20 atmospheric scenarios are presented in Table 2, and the vertical profiles of ozone, temperature and water vapor are shown in Figure 2. We discuss the simulation results, dividing into four cases (CEC in June, ECS in June, CEC in December and ECS in December). The ozone partial column (PC) in LMT at CEC in June was largest (approximately  $5 \times 10^{21} \text{ m}^{-2}$ ) among those of four cases, and the smallest value (approximately  $2 \times 10^{21} \text{ m}^{-2}$ ) was taken from the case at CEC in December.



We interpolated the values from the following three original atmospheric profiles by using cubic splines to make the atmospheric profile smooth in the overlapping regions for a vertical pressure ( $p$ ) grid defined as follows.

$$p[i] = \begin{cases} 10^{3-(i+1)/24} \text{ [hPa]} & i = 1, 2, \dots, 71 (\geq 1 \text{ hPa}) \\ 10^{3-(i-35)/12} \text{ [hPa]} & i = 72, 73, \dots, 108 (< 1 \text{ hPa}) \end{cases}$$

The scale height of the vertical profiles that we used was 3 km.

5 The profiles of ozone, temperature, and water vapor in the vertical region from the surface to 65 hPa (approximately 20 km) were simulated by a one-way nested global-regional air quality forecasting (AQF) system (Takigawa et al., 2007, 2009). This system is based on the CHASER (Chemical Atmospheric General Circulation model for the Study of Atmospheric Environment and Radiative Forcing) model (Sudo et al., 2002) and WRF (Weather Research and Forecasting)/Chem model (Grell et al., 2005) version 3.3. The horizontal resolution of this system is approximately 40 km. The profiles over CEC and ECS were  
10 spatially averaged for the periods of June 1 to June 30 and December 1 to December 31, 2009. The surface temperature was simulated with the AQF system, and the temperature difference between the surface and the lower boundary of the lowest atmospheric layer was less than 1 K. We set the surface temperature to be equal to the value at the surface pressure, since the effect of the temperature contrast between the atmosphere and surface is large for the TIR measurement.

The profiles (ozone, temperature and water vapor) in a vertical region of 985–0.01 hPa were taken from the Modern  
15 Era Retrospective-Analysis for Research and Applications (MERRA) data (Rienecker et al., 2011). A data product named “MERRA DAS 3d analyzed state (inst6\_3d\_ana\_Nv)” provided the three-dimensional fields of layer pressure thickness, air temperature, specific humidity, and ozone mixing ratio at six-hour intervals (00:00, 06:00, 12:00, 18:00 GMT). The MERRA data covered a  $0.66^\circ \times 0.5^\circ$  latitude-longitude grid. We averaged the MERRA data at 06:00 GMT (the nearest local time of 12:00 LT in CEC and ECS) on the same date of the selected AQF system profiles for each region (CEC and ECS). No  
20 interpolation for local time was performed on the MERRA data.

The temperature data of the COSPAR International Reference Atmosphere (CIRA) (Fleming et al., 1990) was used above the vertical level of 0.01 hPa. The CIRA-86 includes monthly and zonally mean temperatures and pressures (0–120 km) with almost global coverage ( $80^\circ\text{N}$ – $80^\circ\text{S}$ ) at an interval of  $10^\circ$ . We averaged the two temperature data at  $30^\circ\text{N}$  and  $40^\circ\text{N}$  for CEC, and used the temperature data at  $30^\circ\text{N}$  for ECS. The mixing ratios of ozone and water vapor at pressures less than 0.01 hPa  
25 were assumed to be equal to those at the upper boundary (0.01 hPa) of the MERRA data because there are no appropriate data to refer. We confirmed that the effects of the assumption in the upper vertical range were negligibly small for our calculation.

We assumed the following quasi-clear sky cases for all scenarios. A no-cloud condition was considered for all wavelength ranges. Basic background aerosol was taken into account only in the UV calculation. The aerosol profile was included as a known parameter because it was reported that the inclusion of aerosol profile changed within 2 % of the LMT ozone amount  
30 in case of the Aura/OMI measurement (Hayashida et al., 2015). We used the vertical profiles of urban and maritime aerosols of a standard mixing state that were described in Hess et al. (1998). These profiles were adjusted to be 0.2 of the total optical thicknesses of the aerosols (moderate pollution). The aerosol profile was not included in the TIR calculation. The extinction of radiation due to aerosol particles with a scale of approximately  $9.6 \mu\text{m}$ , which corresponds to the wavelength of the TIR

range, is negligibly small for the synergetic retrieval of the LMT ozone with the TIR measurement (e.g., Natraj et al., 2011). We assumed that surface albedo was constant in the selected UV ranges (305–340 nm). The information on surface albedo for simulating UV radiance spectra was taken from the database described by Kleipool et al. (2008). This database contains the monthly global maps of the Earth’s surface Lambertian equivalent reflectance (LER) deduced from the Aura/OMI measurements. We obtained monthly and spatially averaged albedo values of 0.056 (June) and 0.063 (December) for CEC and 0.065 (June) and 0.084 (December) for ECS, respectively, from the LER data at the wavelength of 328.1 nm, which is the shortest wavelength in the database. The impact of the uncertainty of the UV surface albedo on tropospheric ozone measurements from space was discussed in Noguchi et al. (2014). The surface emissivity for modeling the TIR radiance spectra was estimated by linear regression analysis based on the Advanced Space-borne Thermal Emission Reflection Radiometer (ASTER) Spectral Library (Baldrige et al., 2009). The surface emissivity for MW was set to 1.0 for the entire range. MW limb measurements are generally insensitive to the surface emissivity since the atmosphere is strongly opaque in this wavelength range.

### 3 Synergetic retrieval simulation

#### 3.1 Forward models of UV, TIR, and MW regions

We used the SCIATRAN radiative transfer model version 3.1 (Rozanov et al., 2005), the Line-By-Line Radiative Transfer Model (LBLRTM) version 12.1 (Clough et al., 2005), and the Advanced Model for Atmospheric Terahertz Radiation Analysis and Simulation (AMATERASU) (Baron et al., 2008) for the calculation of spectra in the UV, TIR, and MW wavelength ranges, respectively. In the presented study, no bias is assumed between the three forward models in order to investigate potential advantage of including MW observation to retrieval of tropospheric ozone.

The SCIATRAN model was developed by the Institute of Remote Sensing/Institute of Environmental Physics (IFE/IUP) of the University of Bremen, Germany, for fast and precise simulation of radiance spectra in the UV, VIS and Near Infrared ranges as measured by spaceborne instruments, e.g., GOME (240–790 nm) and SCIAMACHY (240–2400 nm). SCIATRAN is applicable to spectral regions ranging from 175.44 nm to 2400 nm, and is basically compatible with arbitrary observation geometries and sensor positions in space, in the atmosphere, and on the ground. The spherical shape of the Earth’s atmosphere, including the refraction effect, is properly taken into account when simulating the radiance spectra.

The LBLRTM model is an accurate and efficient line-by-line radiative transfer model, and it has been extensively validated for atmospheric radiance spectra from UV to submillimeter-wave ranges. The line-by-line calculation of the optical thickness of the atmospheric layers is conducted on the basis of the spectroscopic line parameter database (HITRAN 2008) with its updates (Rothman et al., 2009). This model is used as the forward model in retrieval algorithms for analyzing spaceborne measurements such as EOS-Aura/TES (Clough et al., 2006), and GOSAT/TANSO-FTS (Saitoh et al., 2009).

The AMATERASU model consists of a line-by-line radiative transfer calculation allowing for a multi-layered horizontally homogeneous shell atmosphere. This model has been implemented in the retrieval analysis of the SMILES measurements (e.g., Baron et al., 2011) and in the feasibility study of a submillimeter instrument for planetary science (Kasai et al., 2012). The spectroscopic parameters are from commonly used databases such as HITRAN 2008 (Rothman et al., 2009) and the JPL

spectroscopic catalog (Pickett et al., 1998). The continuum absorption due to dry and wet air are also included and are based on the formulation in Pardo et al. (2001).

### 3.2 Theoretical retrieval basis and error estimation

The optimal estimation method (OEM) (Rodgers, 2000) was used for the synergetic retrieval system and their error estimations.

5 The retrieved state vector  $\hat{\mathbf{x}}$  was estimated by minimizing the differences between the observed radiance spectra  $\mathbf{y}_{\text{obs}}$  and the modeled radiance spectra  $\mathbf{y}_{\text{mod}}$ , using a constraint from an a priori state vector  $\mathbf{x}_a$ .

$$\hat{\mathbf{x}} = A\mathbf{x} + (I - A)\mathbf{x}_a + G\boldsymbol{\epsilon} \quad (1)$$

In this equation,  $\mathbf{x}$  is the true state vector,  $A$  is the averaging kernel matrix,  $G$  is the gain (contribution function) matrix, and  $\boldsymbol{\epsilon}$  is the measurement noise vector. The averaging kernel matrix characterizing the sensitivity of the retrieved state vector  $\hat{\mathbf{x}}$  to

10 the true state vector  $\mathbf{x}$  is given by

$$A = \frac{\partial \hat{\mathbf{x}}}{\partial \mathbf{x}} = GK = (K^T S_\epsilon^{-1} K + S_a^{-1})^{-1} K^T S_\epsilon^{-1} K, \quad (2)$$

where  $S_a$  and  $S_\epsilon$  are the a priori covariance matrix and the measurement error covariance matrix, respectively.  $K$  is a weighting function matrix ( $K = \partial \mathbf{y}_{\text{mod}} / \partial \mathbf{x}$ ).  $A$  corresponds to the identity matrix when the retrieved profile is equal to the true atmospheric profile. The number of state vector elements which are independently resolved is obtained by summation of diagonal

15 elements of  $A$ , and is defined as DFS. The  $i$ th element of measurement response vector,  $\mathbf{m}[i]$ , is defined as

$$\mathbf{m}[i] = \sum_j A[i, j]. \quad (3)$$

A value of the measurement response element near unity indicates that almost all information in the retrieval result comes from the observation spectra, while a small value indicates that the retrieval result is largely influenced by the a priori.

The total retrieval error covariance  $\hat{S}$  is calculated using the covariance matrices of the smoothing error  $S_s$  and measurement  
20 noise  $S_m$ .

$$\begin{aligned} \hat{S} &= S_s + S_m \\ &= (I - A) S_a (I - A)^T + G S_\epsilon G^T \\ &= (K^T S_\epsilon^{-1} K + S_a^{-1})^{-1} \end{aligned} \quad (4)$$

The square root of the  $\hat{S}$  diagonals is the total retrieval error in  $\hat{\mathbf{x}}$  ( $\boldsymbol{\epsilon}_x$ ). The value of  $\boldsymbol{\epsilon}_x$  at  $i$ th layer is given by

$$25 \quad \boldsymbol{\epsilon}_x[i] = \sqrt{\hat{S}[i, i]}. \quad (5)$$

We evaluated the sensitivity of the vertical profile of ozone from the synergetic retrieval for seven different combinations of the wavelength ranges, i.e., UV, TIR, MW, TIR+MW, UV+MW, UV+TIR and UV+TIR+MW, in the 20 atmospheric scenarios. The state vectors  $\mathbf{x}$ ,  $\hat{\mathbf{x}}$  and  $\mathbf{x}_a$  were calculated using logarithm units of the volume mixing ratio (VMR). **The diagonal**

components of  $S_a$  were the squares of the a priori error  $\sigma_a$  at each vertical pressure grid). The value of  $\sigma_a$  was set to 100% of the log-based a priori VMR to simply quantify the error reduction from the error in the a priori error to the error in the retrieved state due to the measurement. The diagonal components of  $S_e$  were the squares of the measurement error  $\sigma_e$ . The off-diagonal components of  $S_e$  were set to zero. The off-diagonal components in  $S_a$  indicate the correlations between the ozone concentrations in different vertical layers. Non-zero off-diagonal components in  $S_a$  assist the retrieval of ozone concentration in a layer in which sufficient ozone information is not included in a measurement spectrum with the correlations with other layers in which sufficient ozone information is included in a measurement spectrum (e.g., Saitoh et al., 2009). One of the aims of our feasibility study is to investigate the ozone retrieval sensitivity for each vertical region in an ideal condition. We set off-diagonal components in  $S_a$  to be zero to avoid the assistance of the ozone retrieval by the correlations between different vertical layers.

We normalized the state vector  $\mathbf{x}$  and measurement vector  $\mathbf{y}$  with  $\sigma_a$  and  $\sigma_e$  because values with different order in a vector and a matrix often cause undesirable mathematical errors in computational calculation.

$$\mathbf{u} = \frac{\mathbf{x} - \mathbf{x}_a}{\sigma_a} \quad (6)$$

$$\mathbf{v} = \frac{\mathbf{y}_{\text{obs}} - \mathbf{y}_{\text{mod}}}{\sigma_e} \quad (7)$$

The normalized weighting function was given by

$$K' = KD(\sigma_a/\sigma_e). \quad (8)$$

Here,  $D(\mathbf{a})$  is a diagonal matrix whose diagonal elements are equal to the components of the vector  $\mathbf{a}$ .  $S_a$  and  $S_e$  were normalized in the same way.

$$S'_a = D(1/\sigma_a)S_aD(1/\sigma_a)^T \quad (9)$$

$$S'_e = D(1/\sigma_e)S_eD(1/\sigma_e)^T \quad (10)$$

Using the normalized vectors and matrices,  $A$  and  $\hat{S}$  are expressed as

$$A = \left( K'^T S'^{-1}_e K' + S'^{-1}_a \right)^{-1} K'^T S'^{-1}_e K', \quad (11)$$

$$\hat{S} = \left( K'^T S'^{-1}_e K' + S'^{-1}_a \right)^{-1}. \quad (12)$$

We evaluated the sensitivity of ozone retrieval for seven wavelength combinations in terms of DFS. We calculated the DFS values for the partial column in the UT, MT and LMT regions. The value of DFS from the  $i_{\text{min}}$ th vertical layer to the  $i_{\text{max}}$ th layer is given by

$$\text{DFS} = \sum_{i=i_{\text{min}}}^{i_{\text{max}}} A[i, i]. \quad (13)$$

We also evaluated the sensitivity of ozone retrieval using the pressure of maximum sensitivity (PMS) and the reduction rate of error (RRE) for the partial column. The PMS was defined as the pressure of the maximum of the sum of rows of the

corresponding  $A$  for the ozone partial column. The RRE is given by

$$\text{RRE} = \frac{\text{PCE}_{\text{apriori}} - \text{PCE}_{\text{retrieved}}}{\text{PC}} \quad [\%] \quad (14)$$

where  $\text{PCE}_{\text{apriori}}$  and  $\text{PCE}_{\text{retrieved}}$  are the partial column error, PCE, for the a priori state and the retrieved state, respectively. PC represents the partial column of ozone, and the value of PC from the  $i_{\text{min}}$ th vertical layer to the  $i_{\text{max}}$ th layer is given by

$$\text{PC} = \sum_{i=i_{\text{min}}}^{i_{\text{max}}} \frac{p[i] \cdot \text{VMR}[i]}{k_{\text{B}} \cdot T[i]} \Delta z[i]. \quad (15)$$

Here,  $p[i]$ ,  $\text{VMR}[i]$ ,  $T[i]$  and  $\Delta z[i]$  are pressure, VMR of ozone, temperature, and the vertical length of the  $i$ th layer, respectively.  $k_{\text{B}}$  is the Boltzmann constant. The PCE is given by

$$\text{PCE} = \sum_{i=i_{\text{min}}}^{i_{\text{max}}} \frac{p[i] \cdot \epsilon_{\text{VMR}}[i]}{k_{\text{B}} \cdot T[i]} \Delta z[i]. \quad (16)$$

$\epsilon_{\text{VMR}}[i]$  is the total retrieval error in ozone VMR at the  $i$ th layer ( $\sigma_{\text{a}}$  for  $\text{PCE}_{\text{apriori}}$  and  $\epsilon_{\text{x}}$  for  $\text{PCE}_{\text{retrieved}}$ ).

## 10 4 Results and discussion

The sensitivity of ozone retrieval for the UT (215–383 hPa), MT (383–749 hPa), and LMT (>749 hPa) regions was investigated in terms of DFS. **Figure 3 and Table 3 show** the DFS values calculated with Eq. (13) for the seven wavelength combinations: UV alone, TIR alone, MW alone, TIR+MW, UV+MW, UV+TIR and UV+TIR+MW. The DFS values were averaged in June in CEC (shown by red markers in Fig. 3), June in ECS (purple), December in CEC (green), December in ECS (blue) and all 20 profiles (black). The error bar represents the standard deviation.

The DFS value in the UT region averaged for all 20 profiles was calculated to be  $0.16 \pm 0.08$ ,  $0.59 \pm 0.10$  and  $0.44 \pm 0.41$  for the UV, TIR, and MW wavelength range, respectively. None of the DFS average values for one wavelength range was larger than unity. Using more than one wavelength range, the DFS value increased to  $1.15 \pm 0.25$ ,  $0.90 \pm 0.30$ ,  $0.62 \pm 0.08$ , and  $1.21 \pm 0.28$ , for the wavelength combinations of TIR+MW, UV+MW, UV+TIR, and UV+TIR+MW, respectively. The DFS of the UV+TIR combination was the lowest among those of more than one wavelength range, and adding the MW region increased the value by about two times. The additional MW region was hence most effective at improving the retrieval of ozone in the UT region.

In the MT region, the TIR measurements are the main contributors of DFS information. The DFS values were  $0.50 \pm 0.16$ ,  $0.83 \pm 0.11$ , and less than 0.01 for the UV, TIR and MW wavelength ranges, respectively. The DFS values increased in the same way as in the UT calculation by adding measurements in different wavelength ranges. The average DFS values of the 20 profiles were  $1.03 \pm 0.09$ ,  $0.73 \pm 0.09$ ,  $1.00 \pm 0.09$  and  $1.23 \pm 0.13$  for TIR+MW, UV+MW, UV+TIR and UV+TIR+MW, respectively. It should be noted that the MW measurements, which have no information on ozone in the MT region because of atmospheric opacity, certainly increased the DFS value in the MT region **from 1.00 to 1.23 (about 23 % increase) for the TIR+UV measurements**. This indicates that the information on ozone in the stratosphere and UT, where the sensitivity of MW is high, is also important for retrieval of ozone in the MT region.

The DFS values in the LMT region were generally smaller than those in the UT and MT regions. They were calculated to be  $0.20\pm 0.13$ ,  $0.21\pm 0.15$ , less than 0.01,  $0.20\pm 0.14$ ,  $0.26\pm 0.15$ ,  $0.46\pm 0.25$  and  $0.60\pm 0.27$  for UV, TIR, MW, TIR+MW, UV+MW, UV+TIR and UV+TIR+MW, respectively. The DFS values of the UV and TIR wavelength ranges were almost the same, while the MW measurements had no sensitivity in the LMT region. Similar to the MT region, the DFS values of the UV+TIR measurements (0.46) increased to 0.60 (about 30 % increase) as a result of adding the MW measurement. We note that the DFS value for the TIR measurement in the case at ECS in December 2009 was larger than that for TIR+MW measurement. The averaging kernel matrix in the LMT region for the TIR measurement of this case was discontinuously large. In general, the discontinuity in the averaging kernel occurs because of mathematical issues not atmospheric physical issues, thus we avoid any scientific discussion with the DFS value for the TIR measurement in this case (ECS in December 2009).

We compared our results with the previous studies of estimating the tropospheric ozone sensitivity using DFS. The DFS values for UV, TIR and UV+TIR measurements were summarized in Table 4. Scenarios of the simulation or the measurements are different among this work and the previous studies shown in Table 4, thus, the DFS values themselves should not be directly compared each other. Here we calculated the relative difference between the DFS value for the UV+TIR measurements and mean of the DFS values for UV and TIR measurements. The relative difference of our simulation for all profiles averaged was estimated to be 126 %. It showed good agreement with those of Fu et al. (2013) (139 %), Cuesta et al. (2013) (104 %) and Natraj et al. (2011) (115 %).

The pressure of maximum sensitivity, PMS, for the ozone partial column should be located in a range of the corresponding partial column. In the UT region, the PMS values for all cases were located in a range of the corresponding region (215–383 hPa) by combining more than two wavelength ranges. It was also observed in the PMS values in the MT region. But in the LMT region, only the PMS values of combination of the three wavelength ranges were located in the vertical region of LMT. The PMS value for all profiles averaged in the LMT region was 783 hPa and 808 hPa for the UV+TIR and UV+TIR+MW measurements, respectively. The PMS value was increased by approximately 3 % by adding the MW measurement to the UV+TIR measurements, although the PMS value of the MW measurement itself was lower than 300 hPa in the LMT region.

The reduction rate of error, RRE, in the ozone partial column of ozone calculated using Eq. (14), is shown in the right column of Fig. 4. The value of RRE was approximately 25–45 % in the UT, MT and LMT regions. The RRE generally increased by combining more wavelength ranges in the ozone synergetic retrieval as DFS shown in Fig. 3. The RRE value for all profiles averaged in the LMT region was 36 % and 39 % for UV+TIR and UV+TIR+MW measurements, respectively. Adding the MW measurement made 3 % increase of RRE value. A certain increase of the retrieval sensitivity of the LMT ozone was shown by DFS as well as PMS and RRE.

The sensitivity of MW measurement in the UT region largely depended on the atmospheric profile used in this simulation, and its dependency transferred to the wavelength combinations including the MW measurement. The DFS values of the MW measurement in the UT region for profiles in December 2009 (green and blue markers in Fig. 3) were larger than those in June 2009 (red and purple markers). The sensitivity of the MW measurements in the UT region increased for profiles with large amounts of ozone in the UT region. In the LMT, the DFS values of the UV and TIR measurements strongly depended on the atmospheric profiles. The average value of the partial column of ozone in the LMT region at CEC in June 2009 was

$5.03 \times 10^{21} \text{ m}^{-2}$ , the largest among the four cases. Only the DFS value of UV+TIR+MW at CEC in June 2009 (red marker) was larger than unity ( $1.03 \pm 0.01$ ).

More details of the vertical characteristics are discussed with  $A$ . Figures 5 and 6 show  $A$  and  $m$  obtained from the simulation using the atmospheric profile #01 and #12 for all of the wavelength combinations. The DFS values in the LMT for the UV+TIR+MW measurements of profiles #01 and #12 were respectively estimated to be 1.04 and 0.27, i.e., which were the highest and lowest values among the 20 profiles. In the case of profile #01, the UV, TIR and MW measurements provided information in the LMT, MT to UT, and UT to the stratosphere, respectively. The UV and TIR measurements were important to retrieve the ozone amount in the LMT when only one wavelength range was used because their peaks in the row of  $A$  were located in the LMT region. The FWHM of the row of  $A$  of the UV and TIR measurements in the LMT was approximately 3 km. The peak value in the row of  $A$  increased from 0.25 of the UV and TIR measurements to 0.35 as a result of adding the two measurements. The combination of TIR and UV improved the sensitivity of retrieval of ozone in the LMT, as shown in the previous studies (Landgraf and Hasekamp, 2007; Worden et al., 2007; Natraj et al., 2011; Cuesta et al., 2013; Fu et al., 2013). Adding the MW measurement further increased it to 0.4. In profile #12 (less ozone in the LMT), the peak of the row of  $A$  for the UV measurement was located in the MT (maximum value of 0.1). Although the peak value in the MT increased to 0.23 as a result of adding the TIR and MW measurements, the peak value in the LMT remained low.

In this study, we showed that an introduction of the MW limb measurement had a certain effect to increase the sensitivity of the tropospheric ozone retrieval. However, following issues might cause bias and uncertainties in the retrieval results and should be considered to implement this retrieval method to real measurements. Discrepancy in spectroscopic parameters for several wavelength ranges is one of the most important error sources. For the ozone retrieval using the MW limb measurement, spectroscopic parameters are the largest error sources. It was reported that approximately 3–5 % error was caused by uncertainties in air-broadening coefficient and line intensity in the case of the SMILES observation (Kasai et al., 2013). It is comparable to the approximately 4 % uncertainty in the spectroscopic parameters in the UV and TIR wavelength ranges (Gratien et al., 2010). The tangent height correction can also be a large error source for the MW limb measurement (Kasai et al., 2013). The tangent height is a key parameter to determine the field-of-view, thus uncertainty in the tangent height causes discrepancy of the atmospheric layer assumed in the simulation and the true atmospheric layer. This discrepancy critically affects to the retrieval of the ozone amount in both the stratosphere and the troposphere, and also might cause bias for correction of time delay between the MW limb measurement and the other nadir measurements. In this study, we assumed instruments onboard the ISS (low orbit) and the time difference of approximately five minutes could be ignored. If the time difference was long and its correction was required, three-dimensional atmospheric modeling should be performed including the field-of-view of the MW limb measurement.

As a whole, it was shown that retrieval of the tropospheric ozone was improved by adding the MW limb measurements to the UV and TIR nadir measurements. In the LMT region, the DFS value was estimated to increase about 30 %. The DFS value was estimated to be 0.75 and 0.66 over land and ocean, respectively, for the future mission of IASI-NG and UVNS (Constantino et al., 2017). If the MW limb measurement is implemented to this synergetic retrieval, the DFS value is estimated to increase

to 0.98 and 0.86 over land and ocean, respectively. Our feasibility study showed a possibility to retrieve the ozone in the LMT with a DFS value of unity.

## 5 Conclusions

We performed a feasibility study of obtaining a vertically resolved ozone profile in the troposphere from synergetic retrieval using a combination of three separate wavelength ranges (UV, TIR, and MW). Observation geometries used in this study were the nadirs for the UV and TIR measurements and limb for the MW measurement from low orbit at a height of 300 km (the height of the ISS). The urban (CEC) and ocean (ECS) area in June and December 2009 were assumed in this study. We evaluated the sensitivities of retrieval of ozone in the three vertical regions (UT (215-383 hPa), MT (383-749 hPa) and LMT (>749 hPa)) in terms of the degree of freedom for signal (DFS) based on the OEM calculation. The pressure of maximum sensitivity (PMS), the reduction rate of error (RRE) for the partial column were also used as an indicator of the sensitivity evaluation.

The TIR measurement was most sensitive for retrieving ozone in the UT when only one wavelength range was used. The additional MW measurement was most effective at improving the sensitivity in the UT when combining several wavelength ranges. The DFS values in the UT for all of 20 profiles averaged were  $0.62 \pm 0.08$  and  $1.21 \pm 0.28$  for the UV+TIR and UV+TIR+MW measurements, respectively. In the MT region, the contribution of the TIR measurement was dominant in the DFS calculation. The average DFS value of the TIR measurement for all profiles averaged was  $0.83 \pm 0.11$ . It was increased to more than unity by adding either the UV or MW measurements. The UV and TIR measurements were dominant in the retrieval of ozone in the LMT region. The DFS value in the LMT strongly depended on the ozone abundance. The DFS value in the LMT became larger in the case of larger partial column of the LMT ozone. The largest DFS value in the LMT for the UV+TIR+MW measurement of  $1.03 \pm 0.01$  was given by the case at CEC in June 2009 which is the case of the largest LMT ozone enhancement with the partial column in the LMT ozone of approximately  $5 \times 10^{21} \text{ m}^{-2}$ .

The MW limb measurement alone derived less information in the MT and LMT regions. The DFS values were less than 0.01 and the PMS was located at vertical region higher than the MT and LMT regions. Nevertheless, adding MW measurements to the UV and TIR measurement combinations improved sensitivity not only in the UT but also in the MT and LMT. The DFS values were increased by 96 %, 23 % and 30 % in the UT, MT and LMT, respectively, by adding the MW measurements to the UV+TIR measurements. This might indicate that reducing the uncertainty of ozone abundance in the stratosphere is important for estimating an accurate tropospheric ozone profile.

*Acknowledgements.* This work was supported by the Funding Program for Next Generation World-Leading Researchers (NEXT Program) (No. GR101). This research was partly supported by Coordination Funds for Promoting Space Utilization by the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan. We are grateful to Dr. K. Takigawa for providing us with his numerical simulation data for this study. The MERRA data used in this study was provided by the Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight Center through the NASA GES DISC online archive. We made use of CIRA-86. We thank Dr. K. Kikuchi for discussing



with us the specifications of the MW instrument proposed for the Air Pollution Observations from the ISS. We thank our colleagues in the Aura MLS team (JPL) for valuable discussion on the frequency selection of the APOLLO MW instrument. We are grateful to Dr. A. Rozanov for giving a lot of useful comments of SCIATRAN.

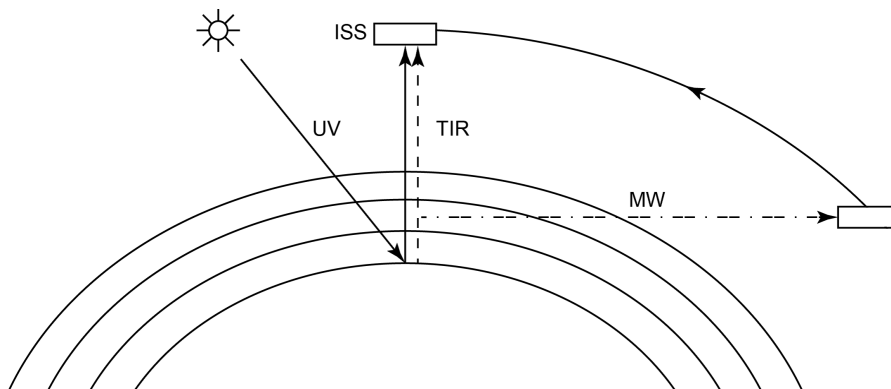
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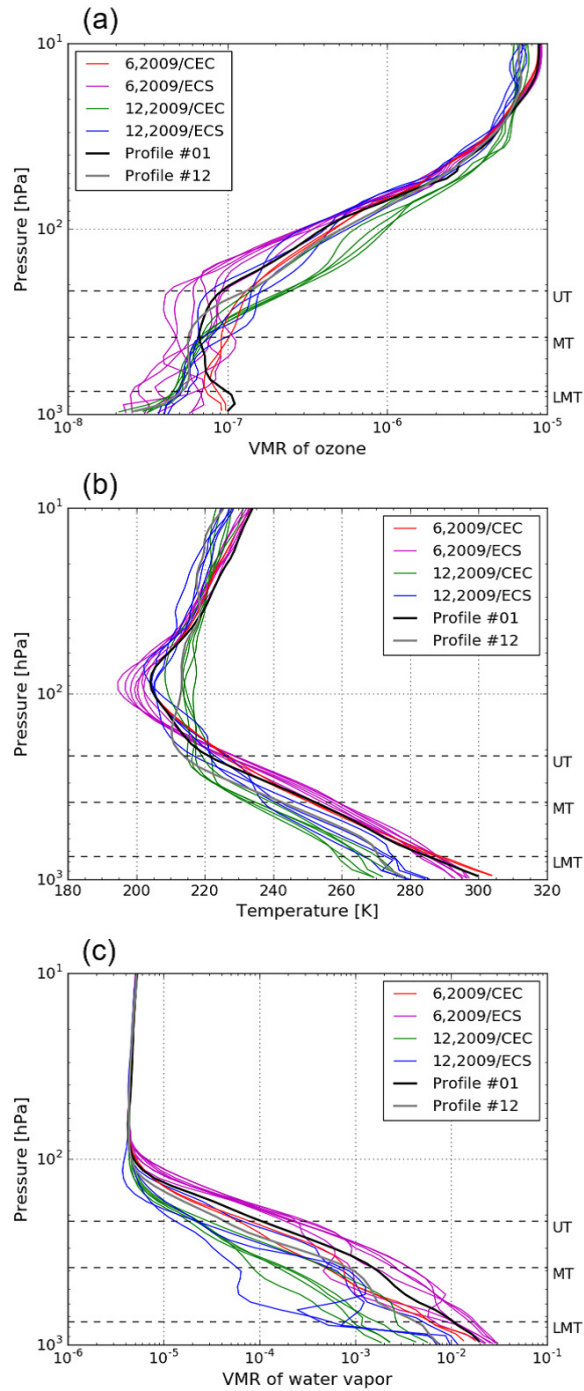
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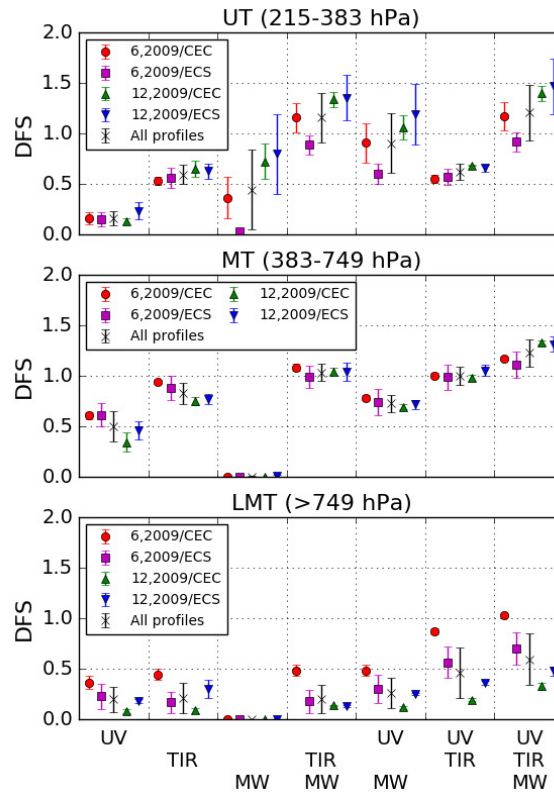
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**Figure 1.** Geometries of down-looking nadir (UV and TIR) and limb (MW) observations.

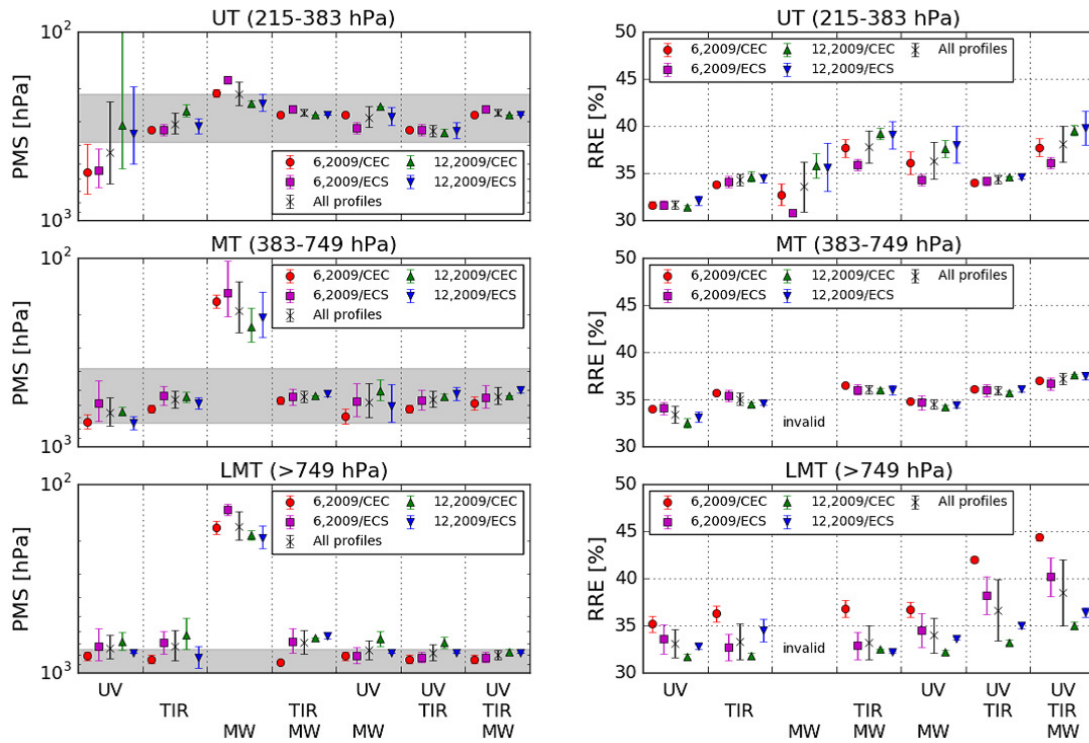


**Figure 2.** The 20 atmospheric scenarios used in this study: (a) VMR of ozone, (b) temperature, and (c) VMR of water vapor. These atmospheric scenarios can be divided into four groups, as denoted by four different color curves: (red) June 2009 in CEC, (purple) June 2009 in ECS, (green) December 2009 in CEC, and (blue) December 2009 in ECS. Two example profiles #01 and #12, represented by the black and gray lines, respectively, are the ones used to obtain the results shown in Figs. 5 and 6.

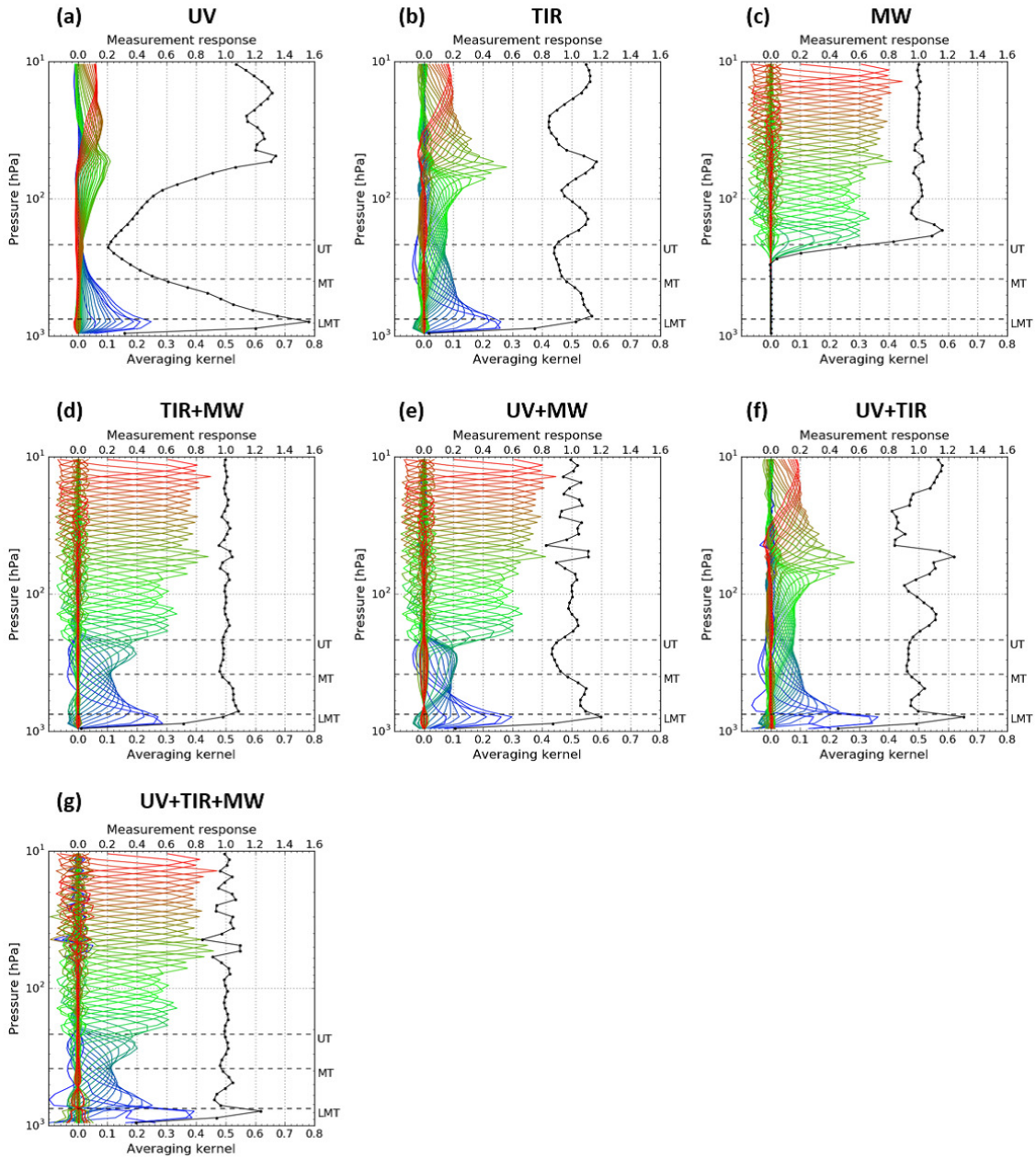


**Figure 3.** Values of DFS in the upper troposphere (UT, 215–383 hPa), middle troposphere (MT, 383–749 hPa), and lowermost troposphere (LMT, >749 hPa): (red) June 2009 in CEC, (purple) June 2009 in ECS, (green) December 2009 in CEC, (blue) December 2009 in ECS and (black) all of 20 profiles.

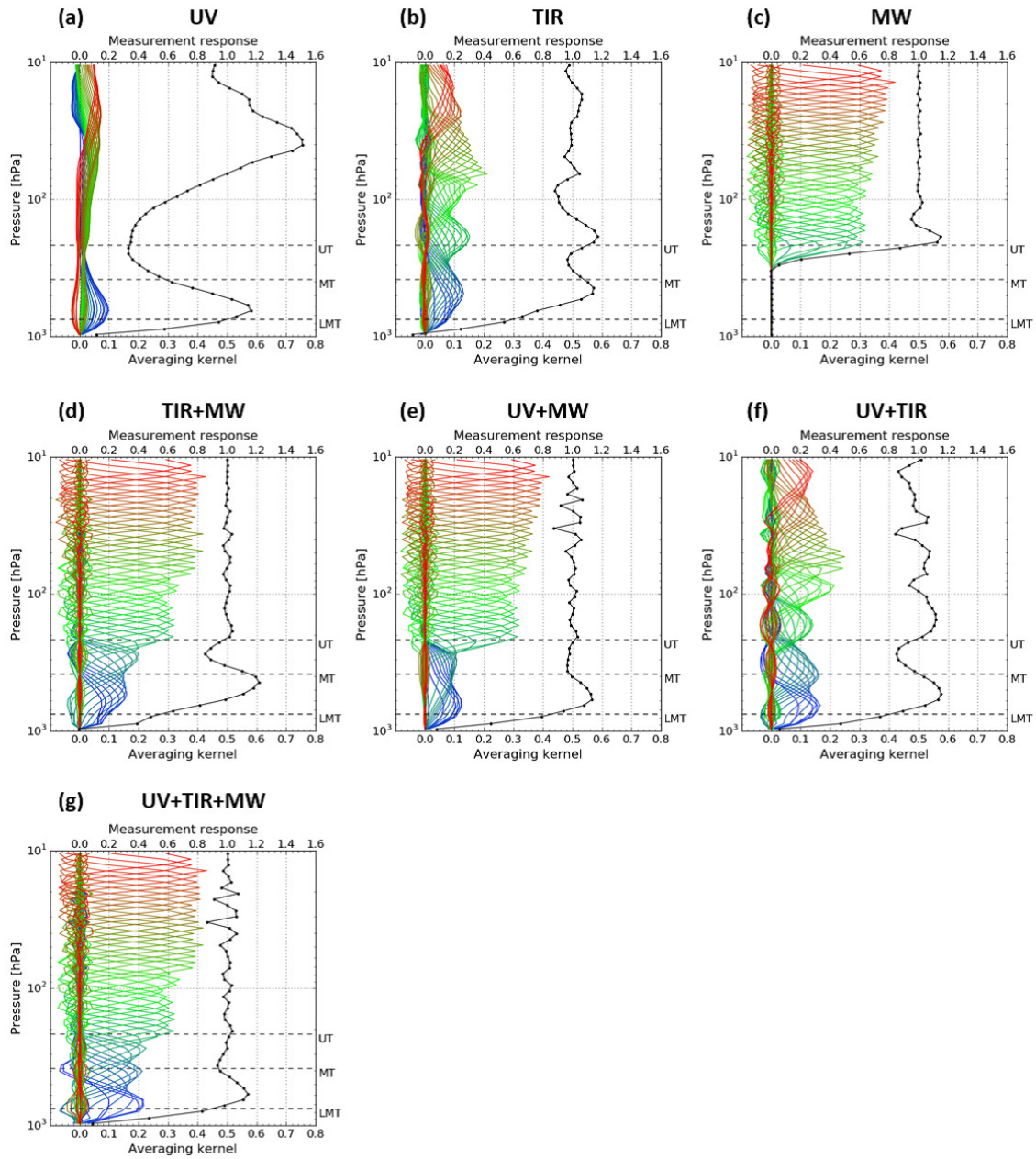




**Figure 4.** Same as Fig. 3 but for the PMS (left) and RRE (right). The gray shaded area represents the vertical region that corresponds to UT, MT and LMT.



**Figure 5.** Averaging kernels of the atmospheric profile #01 (June 2009 in CEC) for seven wavelength selection cases: (a) UV, (b) TIR, (c) MW, (d) UV+TIR, (e) UV+MW, (f) TIR+MW, and (g) UV+TIR+MW. The measurement responses are also plotted by black dots and lines. Atmospheric profile #01 is a scenario which has an ozone enhancement in the LMT.



**Figure 6.** Same as Fig. 5 but for atmospheric profile #12 (December 2009 in CEC). This profile is a scenario that there is less ozone in the LMT.

**Table 1.** Specifications of the three assumed instruments and the radiative transfer models used in this study.

	UV	TIR	MW
Observation geometry	Nadir-viewing	Nadir-viewing	Limb-viewing
Wavelength	305–340 nm	980–1080 $\text{cm}^{-1}$	345–357 GHz, 639–651 GHz
Spectral resolution	0.6 nm	$\sim 0.12 \text{ cm}^{-1}$	25 MHz
Sampling step	0.2 nm	$\sim 0.12 \text{ cm}^{-1}$	25 MHz
Sensitivity <sup>a</sup>	90 (305 nm)–1400 (340 nm)	300	0.7 K (350 GHz band), 1.7 K (645 GHz band)
Scattering	Yes	No	No
Emission	No	Yes	Yes
Forward model	SCIATRAN	LBLRTM	AMATERASU

<sup>a</sup> Instrumental sensitivity is described in the commonly used way for each spectral region; that is, SNR for UV and TIR and noise equivalent brightness temperature for MW.

**Table 2.** Summary of 20 atmospheric scenarios used in the simulation.

#	Date <sup>a</sup>	Area <sup>b</sup>	$T_s^c$ [K]	$P_s^c$ [hPa]	PC (UT) <sup>d</sup> [ $m^{-2}$ ]	PC (MT) <sup>d</sup> [ $m^{-2}$ ]	PC (LMT) <sup>d</sup> [ $m^{-2}$ ]	H <sub>2</sub> O <sup>e</sup> [g/cm <sup>2</sup> ]
01	6/16	CEC	301.4	976.0	$2.57 \times 10^{21}$	$5.87 \times 10^{21}$	$5.66 \times 10^{21}$	3.4
02	6/24	CEC	304.5	970.2	$4.13 \times 10^{21}$	$6.14 \times 10^{21}$	$4.51 \times 10^{21}$	2.1
03	6/25	CEC	305.0	970.2	$3.65 \times 10^{21}$	$6.52 \times 10^{21}$	$4.93 \times 10^{21}$	2.3
04	6/3	ECS	293.9	999.6	$1.49 \times 10^{21}$	$4.39 \times 10^{21}$	$3.72 \times 10^{21}$	4.5
05	6/9	ECS	294.9	1010.9	$1.68 \times 10^{21}$	$4.17 \times 10^{21}$	$3.96 \times 10^{21}$	4.2
06	6/20	ECS	296.0	1004.2	$2.92 \times 10^{21}$	$6.34 \times 10^{21}$	$1.86 \times 10^{21}$	3.7
07	6/21	ECS	296.5	1002.9	$3.00 \times 10^{21}$	$4.26 \times 10^{21}$	$2.39 \times 10^{21}$	6.0
08	6/26	ECS	296.8	1010.0	$3.27 \times 10^{21}$	$6.20 \times 10^{21}$	$2.25 \times 10^{21}$	4.1
09	6/27	ECS	297.6	1006.6	$2.13 \times 10^{21}$	$3.13 \times 10^{21}$	$1.49 \times 10^{21}$	6.0
10	6/30	ECS	298.1	1004.5	$2.71 \times 10^{21}$	$3.05 \times 10^{21}$	$1.80 \times 10^{21}$	5.9
11	12/2	CEC	280.7	993.1	$3.20 \times 10^{21}$	$4.47 \times 10^{21}$	$1.83 \times 10^{21}$	1.1
12	12/11	CEC	280.0	988.8	$2.66 \times 10^{21}$	$4.38 \times 10^{21}$	$2.10 \times 10^{21}$	1.4
13	12/20	CEC	271.6	997.3	$4.58 \times 10^{21}$	$4.14 \times 10^{21}$	$2.22 \times 10^{21}$	0.3
14	12/22	CEC	278.1	985.9	$4.25 \times 10^{21}$	$4.27 \times 10^{21}$	$2.21 \times 10^{21}$	0.6
15	12/27	CEC	271.3	992.6	$4.51 \times 10^{21}$	$4.41 \times 10^{21}$	$2.10 \times 10^{21}$	0.4
16	12/28	CEC	274.0	985.8	$4.12 \times 10^{21}$	$4.33 \times 10^{21}$	$2.15 \times 10^{21}$	0.4
17	12/4	ECS	286.7	1019.2	$4.04 \times 10^{21}$	$4.48 \times 10^{21}$	$2.54 \times 10^{21}$	1.1
18	12/12	ECS	287.9	1019.9	$2.35 \times 10^{21}$	$4.81 \times 10^{21}$	$2.92 \times 10^{21}$	2.0
19	12/21	ECS	282.4	1025.7	$4.91 \times 10^{21}$	$4.74 \times 10^{21}$	$2.68 \times 10^{21}$	0.8
20	12/26	ECS	281.3	1019.3	$3.72 \times 10^{21}$	$4.38 \times 10^{21}$	$2.68 \times 10^{21}$	0.9

<sup>a</sup> All simulation data are from 2009.<sup>b</sup> CEC and ECS stand for Central East China (30°N–40°N, 110°E–123°E) and the East China Sea (29°N–33°N, 125°E–129.5°E), respectively.<sup>c</sup> Temperature and pressure at the surface.<sup>d</sup> PC means ozone partial column. PC is presented for each altitude region: upper troposphere (UT, 215–383 hPa), middle troposphere (MT, 383–749 hPa), and lowermost troposphere (LMT, > 749 hPa).<sup>e</sup> H<sub>2</sub>O column amount in the troposphere.

**Table 3.** Mean and standard deviation ( $1-\sigma$ ) of DFS in the upper troposphere (UT, 215–383 hPa), middle troposphere (MT, 383–749 hPa), and lowermost troposphere (LMT, >749 hPa). The standard deviation is listed in the parenthesis. For example, 0.16(6) means 0.16 of the mean value and 0.06 of the standard deviation.

		UV	TIR	MW	TIR+MW	UV+MW	UV+TIR	UV+TIR+MW
6, 2009/CEC	UT	0.16(6)	0.53(4)	0.36(20)	1.16(14)	0.91(20)	0.55(4)	1.17(14)
	MT	0.61(3)	0.94(2)	<0.01	1.08(4)	0.78(3)	1.00(2)	1.17(1)
	LMT	0.37(6)	0.44(6)	<0.01	0.48(6)	0.48(5)	0.87(3)	1.03(1)
6, 2009/ECS	UT	0.15(7)	0.56(10)	0.03(3)	0.89(9)	0.60(10)	0.57(8)	0.92(10)
	MT	0.61(11)	0.88(12)	<0.01	0.99(11)	0.74(13)	0.99(13)	1.11(13)
	LMT	0.23(13)	0.17(11)	<0.01	0.18(11)	0.30(14)	0.56(15)	0.70(16)
12, 2009/CEC	UT	0.12(3)	0.65(8)	0.72(17)	1.34(8)	1.06(12)	0.68(1)	1.40(7)
	MT	0.34(9)	0.75(4)	<0.01	1.04(3)	0.69(2)	0.98(3)	1.33(2)
	LMT	0.08(2)	0.09(3)	<0.01	0.14(1)	0.12(2)	0.19(1)	0.33(3)
12, 2009/ECS	UT	0.23(8)	0.63(8)	0.80(40)	1.35(23)	1.19(30)	0.66(4)	1.47(27)
	MT	0.46(9)	0.77(5)	<0.01	1.04(9)	0.72(5)	1.05(5)	1.31(7)
	LMT	0.18(2)	0.30(9)	<0.01	0.13(1)	0.25(1)	0.36(2)	0.48(5)
All profiles	UT	0.16(7)	0.59(9)	0.44(40)	1.16(25)	0.90(30)	0.62(8)	1.21(28)
	MT	0.50(15)	0.83(11)	<0.01	1.03(8)	0.73(9)	1.00(9)	1.23(13)
	LMT	0.20(12)	0.21(15)	<0.01	0.20(14)	0.26(15)	0.46(25)	0.59(26)

**Table 4.** Comparison of DFS in the LMT region with previous studies. The method and situation to derive the DFS value are different for each study, thus, the relative difference between the DFS value for the UV+TIR measurement ( $DFS_{UV+TIR}$ ) and mean of DFS values for UV and TIR measurements ( $DFS_{UV}$ ,  $DFS_{TIR}$ ) was used for this comparison. The relative difference was calculated by  $(DFS_{UV+TIR} - \text{Mean}(DFS_{UV}, DFS_{TIR}))/\text{Mean}(DFS_{UV}, DFS_{TIR})$ .

	$DFS_{UV}$	$DFS_{TIR}$	$\text{Mean}(DFS_{UV}, DFS_{TIR})$	$DFS_{UV+TIR}$	Rel. dif.	Definition of LMT
This work	0.20	0.21	0.203	0.458	126 %	>749 hPa
Fu et al. (2013)	0.10	0.21	0.15	0.37	139 %	>700 hPa
Cuesta et al. (2013) <sup>a</sup>	0.08	0.20	0.14	0.285	104 %	<3 km
Natraj et al. (2011)	0.26	0.27	0.265	0.57	115 %	>800 hPa

<sup>a</sup> DFS values over land and ocean are averaged.