

## **Reply to Editor**

Dear Editor

We greatly appreciate your efforts to help us improve our manuscript. In this version, we have worked to improve the quality of language throughout the manuscript. We would like to ask you to change the order of authors considering their contribution.

We hope that our manuscript is suitable for publication in AMT.

Sincerely yours,

Tomohiro Sato and Yasuko Kasai

National Institute of Information and Communications Technology

## **Corrections by ourselves**

Page 1 Authors

The first author: Yasuko Kasai → Tomohiro O. Sato

The last author: Sachiko Hayashida → Yasuko Kasai

Page 1 Line 1

“to” → “of”

“the amount of ozone in the troposphere” → “the tropospheric ozone”

Page 1 Line 3

“sensitivity of the tropospheric retrieval” → “retrieval sensitivity of the tropospheric ozone”

Page 1 Lines 4 – 5

“The urban and sea areas in East Asia in summer and winter seasons were selected for the feasibility study.” → “Two observation points in East Asia (one in an urban area and one in an ocean area) and two observation times (one during summer and one during winter) were assumed.”

Page 1 Line 6

“the” was added.

Page 1 Line 7

“sensitivities of retrieved ozone” → “retrieval sensitivities of the ozone profiles”

Page 1 Line 8

“values of” was removed.

“the” was added.

Page 1 Lines 8 – 9

“error reduction rate” → “the reduction rate of error”

Page 1 Line 9

“the” was added.

Page 1 Lines 9 – 10

“The measurement noises were assumed at the same level as the currently available instruments.”

→ “The measurement noise levels were assumed to be the same as those for currently available instruments.”

Page 1 Line 14

“We found that the” → “The”

Page 1 Line 16

“of” → “value for”

“It might” → “These findings”

Page 1 Line 17

“of the” → “on”

“educed” → “obtained”

Page 1 Line 18

“will be implemented in the Japanese air-quality monitoring missions, APOLLO, GMAP-Asia and uvSCOPE” → “are applicable to the upcoming air-quality monitoring missions, APOLLO, GMAP-Asia and uvSCOPE”

Page 2 Line 3

“largest single environmental health risk” → “greatest environmental health risks”

Page 2 Lines 3 – 4

“Ozone in particular causes serious damage for human health and agricultural crops.” → “Ozone in particular adversely affects human health and agricultural production.”

Page 2 Line 8

“status and to make forecasts of future ozone amount” → “situation and to forecast future ozone amounts”

Page 2 Line 9

“surface ozone” → “ozone at the surface level”

Page 2 Line 10

“impact on the health of humans” → “effect on the health of people”

“is responsible for significant reduction in crop yields” → “that can significantly reduce crop

yields”

Page 2 Line 11

“so that” → “so”

Page 2 Line 15

“in the” was added.

Page 2 Line 16

“a variety of” → “various”

Page 2 Line 33

“ozone retrieval” → “ozone profile retrieval”

Page 3 Line 1

“on” → “using”

“the retrieval sensitivity of” → “retrieval sensitivity for”

Page 3 Line 2

“the” was added.

Page 3 Line 3

“the UV and TIR spectra of the OMI and TES measurements” → “UV and TIR spectra measured with the OMI and TES instruments”

“A value of the” → “The”

Page 3 Lines 4 – 5

“among OMI, TES, and ozonesonde” → “made using the OMI, the TES, and an ozonesonde”

Page 3 Line 7

“on” → “for”

Page 3 Line 8

“, respectively” was removed.

Page 3 Line 9

“approximately 50% increased” → “increased by approximately 50%”

Page 3 Line 10

“only” → “alone”

Page 3 Line 11

“in” → “for”

Page 3 Lines 11 – 12

“They also showed a significant availability” → “The results demonstrated the effectiveness”

Page 3 Line 13

“improve” was added.

“profile” was added.

Page 3 Line 17

“can” → “should”

“missions for air-pollution” → “air-quality monitoring missions”

Page 3 Line 19 – 20

“APOLLO and GMAP-Asia mission” → “the APOLLO and GMAP-Asia missions”

Page 3 Line 21

“The missions of APOLLO and GMAP-Asia” → “These missions”

Page 3 Line 24

“the” was added.

“aid in the” was added.

Page 3 Line 25

“data-seta” → “datasets”

Page 3 Line 26

“data” → “datasets”

Page 3 Line 27

“ozone retrieval” → “ozone profile retrieval”

Page 3 Lines 28 – 29

“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” → “utilizing synergetic observation afforded by UV, TIR, and MW instruments to obtain vertically resolved information on tropospheric ozone not only at the boundary layer but also in the middle and upper troposphere”

Page 3 Line 30

“ozone retrieval adding” → “ozone profile retrieval to the addition of”

Page 3 Lines 31 – 32

“Thus, the feasibility study was performed under an ideal condition for the synergetic retrieval of the tropospheric ozone.” → “The simulation was thus performed under ideal conditions for synergetic retrieval of the tropospheric ozone profile.”

Page 4 Line 3

“used in the simulation” was added.

Page 4 Lines 3 – 4

“We assumed three spectrometers equipped in ISS that” → “Three spectrometers in the ISS were assumed to”

Page 4 Line 4

“of” → “:”

Page 4 Line 5

“use” → “used”

“measurement uses” → “one used”

Page 4 Line 7

“the” was added.

“in our simulation” was removed.

Page 4 Line 8

“set to parallel” → “set parallel”

Page 4 Lines 10 – 11

“transport when we assume a typical value of horizontal wind speed in the troposphere and the stratosphere of 1.2 km/min” → “travel if a typical value of horizontal wind speed in the troposphere and the stratosphere (1.2 km/min) is assumed”

Page 4 Line 11

“smaller than” → “less than the”

Page 4 Line 13 – 14

“Table 1 is a summary of the specification of the three assumed instruments and the radiative transfer models used in this study.” → “Table 1 summarizes the specifications of the three instruments and the three radiative transfer models.”

Page 4 Line 15

“of the simulation of” → “for”

“The UV wavelength ranges” → “UV wavelength range”

Page 4 Line 16

“ozone retrieval” → “ozone profile retrieval”

“In our simulation, we” → “We”

Page 4 Line 17

“for the” → “to”

Page 4 Lines 17 – 20

“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., 2012, 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.” → “stratospheric ozone profile retrieval using MW measurement improves the sensitivity of tropospheric ozone profile retrieval. Although there is a benefit of adding VIS wavelengths (340-505 nm) (e.g., Sellitto et al., 2012, b), we decided not to because the wavelength dependence of surface reflectance, the absorption of NO<sub>2</sub>, and the Ring effect were beyond the scope of this

study.”

Page 4 Line 28

“would be” → “was”

Page 4 Line 32

“in” → “over”

Page 5 Lines 1 – 2

“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).” →

“The assumed MW limb-sounding instrument covered two frequency bands: the 350 GHz band (345-357 GHz) and the 645 GHz band (639-651 GHz).”

Page 5 Line 8

“We also assumed that a” → “We assumed that the”

Page 5 Line 9

“that” was added.

Page 5 Line 11

“350 GHz band and 645 GHz band” → “the 350 GHz and 645 GHz bands”

Page 5 Lines 13 – 14

“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 used typical atmospheric scenarios in summer and winter for East Asia, which is one of most ozone-polluted regions and a major source of the intercontinental transport of ozone toward North America.”

Page 5 Line 16

“, centered among China, Japan, South Korea, and Taiwan” was added.

Page 5 Lines 21 – 22

“it was shown that the ozone retrieval sensitivity was most increased in small to moderate SZAs



in the simulation study performed by Landgraf and Hasekamp (2007)” → “ozone profile retrieval sensitivity is apparently higher when the solar zenith angle (SZA) is low or moderate, as shown in a simulation study by Landgraf and Hasekamp (2007)”

Page 5 Line 23

“over the two Asian areas (CEC and ECS) in June and December 2009. The characteristics of the 20 atmospheric scenarios are presented” → “, as shown”  
“, and the” → “. The”

Page 5 Lines 24 – 26

“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.” → “Of the four cases (CEC in June, ECS in June, CEC in December, and ECS in December), the ozone partial column (PC) in the LMT was the largest (approximately  $5 \times 10^{21} \text{ m}^{-2}$ ) for CEC in June and was the smallest (approximately  $2 \times 10^{21} \text{ m}^{-2}$ ) for CEC in December.”

Page 5 Line 27

“of ozone, temperature and water vapor” was added.

Page 6 Line 4

“the” was added.

Page 6 Line 7

“, and” → “;”

Page 6 Line 8

“to be equal to the value at the surface pressure,” → “equal to the value at surface pressure”

Page 6 Line 10

“in a vertical region of 985-0.01 hPa” → “for the 985-0.01 hPa vertical range”

Page 6 Lines 11 – 12

“A data product named “MERRA DAS 3d analyzed state (inst6\_3d\_ana\_Nv)” provided the three-dimensional fields of layer pressure thickness” → “The “MERRA DAS 3d analyzed state

(inst6\_3d\_ana\_Nv)" data product provided three-dimensional fields for layer pressure thickness"

Page 6 Line 14

"nearest" → "closest"

Page 6 Line 17

"model" was added.

Page 6 Line 18

"model" was added.

Page 6 Lines 21 – 22

"because there are no appropriate data to refer" → "due to the lack of appropriate reference data"

Page 6 Line 22

"in" → "for"

Page 6 Line 28

"that were described in" → "as described by"

Page 6 Line 29

". The" → " because the"

Page 6 Line 32

"selected UV ranges" → "target UV range"

Page 7 Line 2

"impact" → "effect"

Page 7 Line 3

"in" → "by"

Page 7 Line 5

"MW" → "the MW measurement"

Page 7 Lines 13 – 14

“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.”  
→ “To investigate the potential advantage of including MW observations in the retrieval of the tropospheric ozone profile, we assumed no bias between the three forward models.”

Page 7 Line 16

“Near Infrared” → “near infrared”

Page 7 Line 21

“, and it” → “and”

Page 7 Line 29

“are” → “were”

“HITRAN 2008” → “the HITRAN 2008 molecular spectroscopic database”

Page 7 Line 31

“are also included and are” → “was taken into account; it is”

Page 8 Line 2

“system and their error estimations” → “and error estimation”

Page 8 Line 12

“which” → “that”

“summation of” → “summing the”

Page 8 Line 16

“influenced” → “affected”

Page 8 Line 22

“the” was added.

Page 8 Line 29

“due to the measurement” was removed.

Page 8 Line 29 – Page 9 Line 5

“The diagonal components of  $S_\varepsilon$  were the squares of the measurement error  $\sigma_\varepsilon$ . The off-diagonal

components of  $S_\varepsilon$  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.” → “The off-diagonal components in  $S_a$  indicate the correlations between the ozone concentrations in different vertical layers. The non-zero off-diagonal components in  $S_a$  facilitate retrieval of the ozone concentration in a layer for which sufficient ozone information is not included in the 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). We set the off-diagonal components in  $S_a$  to zero to avoid the assistance of the ozone retrieval by the correlations between different vertical layers. The diagonal components of  $S_\varepsilon$  were the squares of the measurement error  $\sigma_\varepsilon$ . The off-diagonal components of  $S_\varepsilon$  were set to zero.”

Page 9 Line 6

“with” → “of”

Page 9 Line 7

“in computational calculation” was removed.

Page 9 Line 10

“was” → “is”

Page 9 Line 12

“whose” → “in which the”

Page 9 Line 16

“Using the normalized vectors and matrices, A and S are expressed as” → “A and S are expressed using the normalized vectors and matrices as”

Page 9 Line 19, 23

“ozone retrieval” → “ozone profile retrieval”

Page 10 Line 4

“the” was added.

Page 10 Lines 6 – 8

“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 4 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 sensitivities of ozone profile retrieval in terms of DFS (calculated using Eq. (13)) are plotted in Fig. 3 and summarized in Table 4. The average DFS values are plotted in red for CEC in June, in purple for ECS in June, in green for CEC in December, in blue for ECS in December, and in black for all 20 profiles. The error bars represent the standard deviation.”

Page 10 Line 9

“value” → “values”

“was” → “were”

Page 10 Line 10

“DFS average” → “average DFS”

Page 10 Line 11

“Using more than one wavelength range” → “For multiple-wavelength measurement”

“value” → “values”

Page 10 Line 12

“for the wavelength combinations of TIR+MW, UV+MW, UV+TIR, and UV+TIR+MW” → “for the TIR+MW, UV+MW, UV+TIR, and UV+TIR+MW measurement combinations”

Page 10 Line 13

“of” → “for”

Page 10 Lines 13 – 15

“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.” → “the four multiple-wavelength measurements; adding the MW measurement approximately doubled the DFS value. The addition of the MW measurement was thus the most effective way of improving the retrieval of the ozone profile in the UT region.”

Page 10 Line 16

“measurements are the main contributors” → “measurement was the main contributor”

Page 10 Line 18

“of” → “for”

Page 10 Lines 20 – 22

“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” → “, although the MW measurement provided no information on ozone in the MT region because of atmospheric opacity, it nevertheless increased the DFS in the MT calculation from 1.00 to 1.23 (approximately 23%) for the TIR+UV measurement”

Page 10 Line 22

“the” was removed.

Page 10 Line 23

“MW” → “the MW measurement”

“ozone in” → “the ozone profile for”

Page 10 Line 27

“that in” was added.

Page 10 Lines 27 – 28

“value of the UV+TIR measurements” → “value for the UV+TIR measurement”

Page 10 Line 28

“increase” was removed.

Page 10 Line 29

“in the case at” → “for”

“2009” was removed.

“the” was added.

Page 10 Line 30

“of” → “in”

Page 10 Lines 30 – 32

“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).” → “Since discontinuity generally occurs in the averaging kernel due to mathematical factors, not atmospheric physical factors, we do not discuss the DFS value for the TIR measurement in the ECS in December case.”

Page 11 Line 1X

“the” → “those of”

Page 11 Line 2

“were” → “are”

Page 11 Lines 2 – 3

“Scenarios of the simulation or the measurements are different among this work and the previous studies shown in Table 5, thus, the DFS values themselves should not be directly compared each other.” → “Since the scenarios of the simulation or measurement are different among this work and the previous studies, the DFS values cannot be directly compared.”

Page 11 Line 3

“Here we” → “We thus”

Page 11 Line 4

“difference” → “differences”

“value” → “values”

“the” was added.

Page 11 Line 5

“the” was added.

“of” → “for”

Page 11 Lines 5 – 6

“estimated to be 126%. It showed good agreement with” → “126%, which shows good agreement with”

Page 11 Lines 7 – 8

“in a range of the corresponding partial column” → “within the corresponding vertical region”

Page 11 Lines 8 – 13

“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 PMS values for the UT calculation for all cases were located within the corresponding range (215-383 hPa) when two or more wavelength ranges were combined. This was also observed for the MT calculation. For the LMT calculation, this was observed only when three wavelength ranges were combined. The PMS values for all profiles averaged for the LMT were 783 hPa and 808 hPa for the UV+TIR and UV+TIR+MW measurements, respectively. The PMS value was increased approximately 3% by adding the MW measurement to the combined UV+TIR measurement although the PMS value in the MW measurement for the LMT was lower than 300 hPa.”

Page 11 Line 14

“in the ozone partial column of ozone” → “for the partial column of ozone”  
“, is shown in the right column of” → “is shown on the right in”

Page 11 Line 15

“value of” was removed.

Page 11 Lines 15 – 19

“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 9% and 11% for UV+TIR and UV+TIR+MW measurements, respectively. Adding the MW



measurement made 2% 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.” → “It was generally higher when more wavelength ranges were combined in the ozone synergetic retrieval, as shown by the DFS values plotted in Fig. 3. The RRE value for all profiles averaged was 36% for the LMT calculation and 39% for the UV+TIR and UV+TIR+MW measurements. Adding the MW measurement increased the RRE by 3%. The DFS, PMS, and RRE values all show a certain increase in the retrieval sensitivity of the LMT ozone profile.”

Page 11 Line 20

“in this simulation” was removed.

Page 11 Lines 20 – 21

“its dependency transferred to” → “this dependency was reflected in”

Page 11 Lines 21 – 22

“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).” → “As shown in Fig. 3, the DFS values in the MW measurement for the UT in December for both areas were larger than those for June.”

Page 11 Line 23

“ozone in the UT region” → “the UT ozone”

Page 11 Line 24

“strongly depended” → “depended greatly”

Page 11 Line 25

“at CEC in June 2009” → “for CEC in June”

Page 11 Lines 25 – 26

“of UV+TIR+MW at CEC in June 2009 (red marker)” → “in the UV+TIR+MW for CEC in June”

Page 11 Line 27

“with” → “using the averaging kernel matrix”

Page 11 Line 28

“simulation using the atmospheric profile ¥#01 and ¥#12 for all of the” → “simulations using atmospheric profiles #01 and #12 for all”

Page 11 Line 29

“i.e.,” was removed.

Page 11 Line 30

“In the case of” → “For”

Page 11 Line 31

“in” → “on”

“the” was added.

Page 11 Line 33, 34

“of” → “for”

Page 11 Lines 34 – 35

“as a result of adding the two measurements” → “when the two measurements were combined”

Page 11 Line 35

“The combination of TIR and UV improved” → “Combination of these measurements improves”

Page 11 Line 35 – Page 12 Line 1

“ozone in the LMT” → “the LMT ozone amount”

Page 12 Line 2

“In” → “For”

Page 12 Line 4

“adding” → “combining”

Page 12 Line 7

“of” was removed.

Page 12 Line 10

“It was shown that the increase” → “Increase”

“amount” was added.

Page 12 Lines 12 – 14

“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.” → “Our results show that introducing MW limb measurement increases the sensitivity of tropospheric ozone profile retrieval. However, several factors may cause bias and uncertainty in the retrieval results and thus should be considered before the proposed retrieval method is implemented.”

Page 12 Line 14

“the” was added.

Page 12 Lines 14 – 15

“one of the most important error sources” → “a major source of errors”

Page 12 Lines 15 – 16

“For the ozone retrieval using the MW limb measurement, spectroscopic parameters are the largest error sources” → “For ozone profile retrieval using MW limb measurement, the spectroscopic parameters are the major error source”

Page 12 Line 16

“approximately 3-5% error” → “an error of approximately 3-5%”

“the” was added.

Page 12 Line 17

“. It” → “, which”

Page 12 Line 18

“The tangent height” → “Tangent height”

Page 12 Line 20

“thus” → “so”

“discrepancy of” → “a discrepancy between”

Page 12 Line 21

“to” was removed.

Page 12 Line 22

“amount” → “profile”

“also might” → “may”

“for correction of” → “in the correction of the”

Page 12 Line 22, 23

“the” was removed.

Page 12 Line 23

“that the” was added.

“Earth” was added.

Page 12 Line 24

“the” → “that a”

Page 12 Lines 24 – 26

“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.” → “If the time difference is actually long enough to need correction, three-dimensional atmospheric modeling including the field-of-view of the MW limb measurement should be performed.”

Page 12 Line 27

“profile” was added

“the” was removed.

Page 12 Line 28

“to increase about” → “to be increased by approximately”

Page 12 Line 29

“value was” → “values were”

Page 12 Lines 29 – 30

“future mission of IASI-NG and UVNS” → “upcoming IASI-NG and UVNS missions”

Page 12 Line 30

“the” was removed.

“this” → “the”

Page 12 Lines 31 – 32

“showed a possibility to retrieve the ozone” → “has shown that it is possible to retrieve the ozone profile”

Page 13 Line 2

“a vertically resolved ozone profile in the troposphere from” → “vertically resolved ozone profiles in the troposphere with”

Page 13 Line 3

“a combination of three separate” → “various combinations of three”

“Observation” → “The observation”

Page 13 Lines 4 – 5

“low orbit at a height of 300 km (the height of the ISS)” → “low Earth orbit (300 km, the height of the ISS)”

Page 13 Line 6

“retrieval of ozone in” → “tropospheric ozone profile retrieval for”

Page 13 Line 8

“,” → “and”

Page 13 Line 10

“ozone” → “the ozone profile”

Page 13 Line 11

“additional” → “addition of”

Page 13 Line 11

“the” was removed.

Page 13 Line 12

“of” was removed.

Page 13 Line 15

“measurements” → “measurement”

Page 13 Line 16

“ozone” → “the ozone profile”

“strongly” → “greatly”

Page 13 Lines 16 – 17

“The DFS value in the LMT became larger in the case of” → “It was larger for a”

Page 13 Lines 17 – 18

“the UV+TIR+MW measurement of  $1.03\pm 0.01$  was given by the case at” → “UV+TIR+MW measurement ( $1.03\pm 0.01$ ) was obtained for”

Page 13 Line 18

“2009” was removed.

“of” → “with”

Page 13 Line 19

“of approximately  $5\times 10^{21}\text{m}^{-2}$ ” → “(approximately  $5\times 10^{21}\text{m}^{-2}$ )”

Page 13 Line 20

“derived” → “provided”

Page 13 Line 21

“at” → “in a”

“regions” was removed.

Page 13 Lines 21 – 22

“measurements to the UV and TIR measurement combinations” → “measurement to the combined UV and TIR measurement”

Page 13 Line 23, 24

“measurements” → “measurement”

Page 13 Lines 24 – 25

“This might indicate that reducing the uncertainty of ozone abundance in the stratosphere is important for estimating an accurate tropospheric ozone profile.” → “This indicates that reducing uncertainty about ozone abundance in the stratosphere may be important for accurately estimating the tropospheric ozone profile.”

Page 18 Fig. 1

Caption, “Geometries of down-looking nadir (UV and TIR) and limb (MW) observations” → “Geometries used for observation: nadir (UV and TIR) and limb (MW).”

Page 19 Fig. 2

Caption, “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.” → “Twenty atmospheric scenarios used in this study: (a) VMR of ozone, (b) temperature, and (c) VMR of water vapor. They are divided into four groups, as denoted by color: CEC–June 2009 (red), ECS–June 2009 (purple), CEC–December 2009 (green), and ECS–December 2009 (blue). Two example profiles (#01 and #12, black and gray lines, respectively) were used to obtain results shown in Figs. 5 and 6.”

Page 20 Fig. 3

Caption, “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.” → “Values of DFS for upper troposphere (UT, 215-383 hPa), middle troposphere (MT, 383-749 hPa), and lowermost troposphere (LMT, >749 hPa): CEC–June 2009 (red), ECS–June 2009 (purple), CEC–December 2009 (green), ECS–December 2009 (blue), and 20 profiles averaged (black).”

Page 21 Fig. 4

Caption, “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.” → “Same as Fig. 3 but for PMS (left)

and RRE (right). Gray shaded area represents vertical region comprising UT, MT, and LMT.”

Page 22 Fig. 5

Caption, “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.” → “Averaging kernels for atmospheric profile 01 (more ozone enhancement in LMT, CEC–June 2009) for seven wavelength measurements: (a) UV, (b) TIR, (c) MW, (d) UV+TIR, (e) UV+MW, (f) TIR+MW, and (g) UV+TIR+MW. Measurement responses are shown by black dots and lines.”

Page 23 Fig. 6

Caption, “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.” → “Same as Fig. 5 but for profile #12 (less ozone in LMT, CEC–December 2009).”

Page 24 Fig. 7

Caption, “Values of DFS, PMS and RRE in the upper troposphere (UT, 215-383 hPa), middle troposphere (MT, 383-749 hPa), and lowermost troposphere (LMT, >749 hPa) for all of 20 profiles averaged with different  $\sigma_a$  settings. The value of  $\sigma_a$  ranges to 100% (black), 50% (red), 30% (purple), 20% (green) and 10% (blue) of the log-based a priori VMR.” → “Values of DFS, PMS and RRE for upper troposphere (UT, 215-383 hPa), middle troposphere (MT, 383-749 hPa), and lowermost troposphere (LMT, >749 hPa) for 20 profiles averaged:  $\sigma_a$  values of 100% (black), 50% (red), 30% (purple), 20% (green) and 10% (blue) of the log-based a priori VMR.”

Page 25 Table 1

Caption, “Specifications of the three assumed instruments and the radiative transfer models used in this study.” → “Specifications of three assumed instruments and radiative transfer models.”

Footnote, “Instrumental” → “Instrument”, “the” was removed, “; that is,” → “:”

Page 28 Table 4

Caption, “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.” → “Standard deviation is shown in parentheses. For example, 0.16(6) represents a mean value of 0.16 and a standard deviation of 0.06.”



Page 29 Table 5

Caption, “those of” was added, “The method and situation to derive the DFS value are different for each study, thus, the relative difference between the DFS value for the” → “Method and scenarios differed, so relative difference between DFS value for”, “this” 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

Tomohiro O. Sato<sup>1</sup>, Takao M. Sato<sup>2,1</sup>, Hideo Sagawa<sup>3</sup>, Katsuyuki Noguchi<sup>4</sup>, Naoko Saitoh<sup>5</sup>, Hitoshi Irie<sup>5</sup>, Kazuyuki Kita<sup>6</sup>, Mona E. Mahani<sup>1,7</sup>, Koji Zettsu<sup>1</sup>, Ryoichi Imasu<sup>8</sup>, Sachiko Hayashida<sup>4</sup>, and Yasuko Kasai<sup>1,9</sup>

<sup>1</sup>National Institute of Information and Communications Technology, Tokyo, Japan

<sup>2</sup>Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, Kanagawa, Japan

<sup>3</sup>Kyoto Sangyo University, Kyoto, Japan

<sup>4</sup>Nara Women's University, Nara, Japan

<sup>5</sup>Center for Environmental Remote Sensing, Chiba University, Chiba, Japan

<sup>6</sup>Ibaraki University, Ibaraki, Japan

<sup>7</sup>Tohoku University, Sendai, Japan

<sup>8</sup>Atmospheric and Ocean Research Institute, The University of Tokyo, Chiba, Japan

<sup>9</sup>Tokyo Institute of Technology, Tokyo, Japan

Correspondence to: Y. Kasai (ykasai@nict.go.jp)

**Abstract.** We performed a feasibility study of constraining the vertical profile of the tropospheric ozone 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 retrieval sensitivity of the tropospheric ozone by adding the MW measurement to the UV and TIR measurements was reported for the first time by this work. Two observation points in East Asia (one in an urban area and one in an ocean area) and two observation times (one during summer and one during winter) were assumed. Geometry of line-of-sight was nadir down-looking for the UV and TIR measurements, and limb-sounding for the MW measurement. The retrieval sensitivities of the ozone profiles in the upper troposphere (UT), middle troposphere (MT) and lowermost troposphere (LMT) were estimated using the degree of freedom for signal (DFS), the pressure of maximum sensitivity, reduction rate of error from the a priori error, and the averaging kernel matrix, derived based on the optimal estimation method. The measurement noise levels were assumed to be the same as those for 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. 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 value for the LMT ozone was also increased by adding the MW measurement. These findings indicate that better information on LMT ozone can be obtained by adding constraints on the UT and MT ozone from the MW measurement. The results of this study are applicable to the upcoming 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 **greatest environmental health risks**. **Ozone in particular adversely affects human health and agricultural production**. 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 **situation and to forecast future ozone amounts**.

Ozone plays different roles in different altitude regions in the troposphere. It is well known that **ozone at the surface level** is a harmful pollutant that has a detrimental **effect on the health of people** and plants and **that can significantly reduce crop yields**. The lifetime of ozone in the free troposphere ranges from a few days to weeks, **so** 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), **in the** middle troposphere (MT), and **in the** upper troposphere (UT) on a global scale.

Ozone has been observed from space in **various** 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 profile 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 using the OMI and TES measurements. Natraj et al. (2011) showed that retrieval sensitivity for the LMT is improved by combining the UV and TIR measurements. Fu et al. (2013) implemented a synergetic retrieval of boundary layer ozone using UV and TIR spectra measured with the OMI and TES instruments. 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 made using the OMI, the TES, and an ozonesonde 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, for 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. The DFS values were increased by approximately 50 % by combining the GOME-2 and IASI measurements, compared with the IASI measurement alone. The other approach to retrieve the tropospheric ozone profile using neural network technique was performed with the SCIAMACHY nadir measurements for the UV and VIS ranges (Sellitto et al., 2012a, b). The results demonstrated the effectiveness of combining several wavelength ranges to retrieve the tropospheric ozone profile.

Our idea is to add MW measurements to improve the synergetic retrieval of the tropospheric ozone profile. 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. 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 should be of benefit to future air-quality monitoring missions, 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 the APOLLO and GMAP-Asia missions is to measure short-lived climate pollutants for monitoring global pollution and climate change. These missions 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 the NICT air pollution prediction project is to make health index to aid in the mitigation of air pollution disasters using high-horizontal resolution (a few km scale) pollution forecasting from multiple datasets, such as satellite observation, ground-based observation, and in-situ observation datasets.

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

## 2 Observation scenario

### 2.1 Observation wavelength region and geometry

The observation scenario **used in the simulation** follows the concept of the APOLLO mission. **Three spectrometers in the ISS were assumed to** observe the three wavelength ranges: UV, TIR, and MW. Figure 1 shows the observation geometries for the 5 three spectrometers. The UV and TIR instruments **used** nadir down-looking, and the MW **one used** 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 **the ISS** was assumed to be 300 km. The azimuthal direction of the field-of-view of the MW limb-sounding was **set 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 10 of 5 minutes corresponds to approximately 6 km **travel if a typical value of horizontal wind speed in the troposphere and the stratosphere (1.2 km/min) is assumed** (Fleming et al., 1988), which is **less than the** 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 summarizes the specifications of the three instruments and the three radiative transfer models.**

15 The wavelength range **for** the UV measurement was set to 305–340 nm. **UV wavelength range** shorter than 305 nm is useful for the stratospheric **ozone profile retrieval** (e.g., Bak et al., , 2012). **We** added the MW limb measurement which is more sensitive **to** stratospheric ozone. We excluded the shorter UV wavelength ranges to clearly show whether **stratospheric ozone profile retrieval using MW measurement improves the sensitivity of tropospheric ozone profile retrieval. Although there is a benefit of adding VIS wavelengths (340–505 nm) (e.g., Sellitto et al., 2012a, b), we decided not to because the wavelength** 20 **dependence of surface reflectance, the absorption of NO<sub>2</sub>, and the Ring effect were beyond the scope of this 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). We used SNR values of the APOLLO instrument setups in the UV simulation. Three references of the SNR value were prepared for high, middle and low level of the radiance. The SNR value for the simulated radiance was 25 linearly interpolated by the two of three reference SNR values. Table 2 shows the reference SNR values at 305 nm and 340 nm, and the solar zenith angle (SZA) and surface albedo of the three conditions [Private communication with K. Gerilowski]. The values of SNR used in the UV simulation were estimated to be approximately 90 and 1400 at 305 nm and 340 nm, respectively.

We assumed that the nadir-viewing TIR instrument **was** a Fourier transform spectrometer covering the TIR spectral range (980–1080  $\text{cm}^{-1}$ ) including the ozone  $\nu_3$  absorption band, 9.6  $\mu\text{m}$  (1045  $\text{cm}^{-1}$ ) 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  $\text{cm}^{-1}$  and calculated the noise equivalent differential temperature for each wavelength, assuming that the SNR is a constant value of 300 **over** 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 assumed MW limb-sounding instrument covered two frequency bands: the 350 GHz band (345–357 GHz) and the 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 (H<sub>2</sub>O, CO, CH<sub>3</sub>CN, N<sub>2</sub>O, SO<sub>2</sub>, H<sub>2</sub>CO, and HNO<sub>3</sub>). 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 assumed that the** typical integration time was 0.5 seconds for one spectrum accumulation and **that** 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 **the 350 GHz and 645 GHz bands**, respectively).

## 2.2 Atmospheric conditions

**We used typical atmospheric scenarios in summer and winter for East Asia, which is one of most ozone-polluted regions and a major source of the intercontinental transport of ozone toward North America.** 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, **centered among China, Japan, South Korea, and Taiwan**). 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 **ozone profile retrieval sensitivity is apparently higher when the solar zenith angle (SZA) is low or moderate, as shown in a simulation study by Landgraf and Hasekamp (2007).**

We made a total of 20 atmospheric scenarios, **as shown** in Table 3. **The vertical profiles of ozone, temperature and water vapor are shown in Figure 2. Of the four cases (CEC in June, ECS in June, CEC in December, and ECS in December), the ozone partial column (PC) in the LMT was the largest (approximately  $5 \times 10^{21} \text{ m}^{-2}$ ) for CEC in June and was the smallest (approximately  $2 \times 10^{21} \text{ m}^{-2}$ ) for CEC in December.**

We interpolated the values **of ozone, temperature and water vapor** 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.

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 the WRF (Weather Research and Forecasting)/Chem model (Grell et al., 5 2005) version 3.3. The horizontal resolution of this system is approximately 40 km. The profiles over CEC and ECS were 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; 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 equal to the value at surface pressure since the effect of the temperature contrast between the atmosphere and surface is large for the TIR measurement.

10 The profiles (ozone, temperature and water vapor) for the 985–0.01 hPa vertical range were taken from the Modern Era Retrospective-Analysis for Research and Applications (MERRA) data (Rienecker et al., 2011). The “MERRA DAS 3d analyzed state (inst6\_3d\_ana\_Nv)” data product provided three-dimensional fields for 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 closest local time of 12:00 LT in CEC 15 and ECS) on the same date of the selected AQF system profiles for each region (CEC and ECS). No interpolation for local time was performed on the MERRA data.

The temperature data of the COSPAR International Reference Atmosphere (CIRA) model (Fleming et al., 1990) was used above the vertical level of 0.01 hPa. The CIRA-86 model 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 20  $40^\circ\text{N}$  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 were assumed to be equal to those at the upper boundary (0.01 hPa) of the MERRA data due to the lack of appropriate reference data. We confirmed that the effects of the assumption for 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 25 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 as described by 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 because the 30 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 target UV range (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 35 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 effect of the uncertainty of the UV surface albedo on tropospheric ozone measurements from space was discussed by 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 the MW measurement 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

10 We used the SCIATRAN radiative transfer model version 3.1 (Rozañov 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. To investigate the potential advantage of including MW observations in the retrieval of the tropospheric ozone profile, we assumed no bias between the three forward models.

15 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.

20 The LBLRTM model is an accurate and efficient line-by-line radiative transfer model and 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).

25 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 were from commonly used databases such as the HITRAN 2008 molecular spectroscopic database (Rothman et al., 2009) and the JPL spectroscopic catalog (Pickett et al., 1998). The continuum absorption due to dry and wet air was taken into account; it is 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 **and error estimation**. 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  $\mathbf{x}_a$ .

$$5 \quad \hat{\mathbf{x}} = A\mathbf{x} + (I - A)\mathbf{x}_a + G\epsilon \quad (1)$$

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

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

10 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 **that** are independently resolved is obtained by **summing the** diagonal elements of  $A$ , and is defined as DFS. The  $i$ th element of measurement response vector,  $\mathbf{m}[i]$ , is defined as

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

15 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 **affected** 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 noise  $S_m$ .

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

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

$$\epsilon_x[i] = \sqrt{\hat{S}[i, i]}. \quad (5)$$

25 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. **The off-diagonal components in  $S_a$  indicate the correlations between the ozone concentrations in different vertical layers.**

The non-zero off-diagonal components in  $S_a$  facilitate retrieval of the ozone concentration in a layer for which sufficient ozone information is not included in the 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). We set the off-diagonal components in  $S_a$  to zero to avoid the assistance of the ozone profile retrieval by the correlations between different vertical layers. The diagonal components of  $S_\epsilon$  were the squares of the measurement error  $\sigma_\epsilon$ . The off-diagonal components of  $S_\epsilon$  were set to zero.

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

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

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

The normalized weighting function is given by

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

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

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

$$S'_\epsilon = D(1/\sigma_\epsilon) S_\epsilon D(1/\sigma_\epsilon)^T \quad (10)$$

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

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

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

We evaluated the sensitivity of ozone profile 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 profile 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

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

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

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

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.  $k_B$  is the Boltzmann constant. The PCE is given by

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

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

## 5 4 Results and discussion

The sensitivities of ozone profile retrieval in terms of DFS (calculated using Eq. (13)) are plotted in Fig. 3 and summarized in Table 4. The average DFS values are plotted in red for CEC in June, in purple for ECS in June, in green for CEC in December, in blue for ECS in December, and in black for all 20 profiles. The error bars represent the standard deviation.

The DFS values in the UT region averaged for all 20 profiles were 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 average DFS values for one wavelength range was larger than unity. For multiple-wavelength measurement, the DFS values 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 TIR+MW, UV+MW, UV+TIR, and UV+TIR+MW measurement combinations, respectively. The DFS for the UV+TIR combination was the lowest among the four multiple-wavelength measurements; adding the MW measurement approximately doubled the DFS value. The addition of the MW measurement was thus the most effective way of improving the retrieval of the ozone profile in the UT region.

In the MT region, the TIR measurement was the main contributor 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 for 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, although the MW measurement provided no information on ozone in the MT region because of atmospheric opacity, it nevertheless increased the DFS in the MT region from 1.00 to 1.23 (approximately 23%) for the TIR+UV measurement. This indicates that information on ozone in the stratosphere and UT, where the sensitivity of the MW measurement is high, is also important for retrieval of the ozone profile for 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 that in the MT region, the DFS value for the UV+TIR measurement (0.46) increased to 0.60 (about 30%) as a result of adding the MW measurement. We note that the DFS value for the TIR measurement for ECS in December was larger than that for the TIR+MW measurement. The averaging kernel matrix in the LMT region for the TIR measurement in this case was discontinuously large. Since discontinuity generally occurs in the averaging kernel due to mathematical factors, not atmospheric physical factors, we do not discuss the DFS value for the TIR measurement in the ECS in December case.

We compared our results with those of previous studies of estimating the tropospheric ozone sensitivity using DFS. The DFS values for UV, TIR and UV+TIR measurements are summarized in Table 5. Since the scenarios of the simulation or measurement are different among this work and the previous studies, the DFS values cannot be directly compared. We thus calculated the relative differences between the DFS values for the UV+TIR measurements and the mean of the DFS values for the UV and TIR measurements. The relative difference for our simulation for all profiles averaged was 126 %, which shows 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 within the corresponding vertical region. The PMS values for the UT calculation for all cases were located within the corresponding range (215–383 hPa) when two or more wavelength ranges were combined. This was also observed for the MT calculation. For the LMT calculation, this was observed only when three wavelength ranges were combined. The PMS values for all profiles averaged for the LMT were 783 hPa and 808 hPa for the UV+TIR and UV+TIR+MW measurements, respectively. The PMS value was increased approximately 3 % by adding the MW measurement to the combined UV+TIR measurement although the PMS value in the MW measurement for the LMT was lower than 300 hPa.

The reduction rate of error, RRE, for the partial column of ozone calculated using Eq. (14) is shown on the right in Fig. 4. The RRE was approximately 0–20 % in the UT, MT and LMT regions. It was generally higher when more wavelength ranges were combined in the ozone synergetic retrieval, as shown by the DFS values plotted in Fig. 3. The RRE value for all profiles averaged was 36 % for the LMT calculation and 39 % for the UV+TIR and UV+TIR+MW measurements. Adding the MW measurement increased the RRE by 3 %. The DFS, PMS, and RRE values all show a certain increase in the retrieval sensitivity of the LMT ozone profile.

The sensitivity of MW measurement in the UT region largely depended on the atmospheric profile used, and this dependency was reflected in the wavelength combinations including the MW measurement. As shown in Fig. 3, the DFS values in the MW measurement for the UT in December for both areas were larger than those for June. The sensitivity of the MW measurements in the UT region increased for profiles with large amounts of the UT ozone. In the LMT, the DFS values of the UV and TIR measurements depended greatly on the atmospheric profiles. The average value of the partial column of ozone in the LMT region for CEC in June was  $5.03 \times 10^{21} \text{ m}^{-2}$ , the largest among the four cases. Only the DFS value in the UV+TIR+MW for CEC in June was larger than unity ( $1.03 \pm 0.01$ ).

More details of the vertical characteristics are discussed using the averaging kernel matrix  $A$ . Figures 5 and 6 show  $A$  and  $m$  obtained from the simulations using atmospheric profiles #01 and #12 for all 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, which were the highest and lowest values among the 20 profiles. For profile #01, the UV, TIR and MW measurements provided information on the LMT, the MT to UT, and the 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  $A$  for the UV and TIR measurements in the LMT was approximately 3 km. The peak value in the row of  $A$  increased from 0.25 for the UV and TIR measurements to 0.35 when the two measurements were combined. Combination of these measurements improves the sensitivity of retrieval of the LMT ozone

amount, 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. For 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 combining the TIR and MW measurements, the peak value in the LMT remained low.

5 Additionally, we performed sensitivity study for the DFS, PMS and RRE using several  $\sigma_a$  values in order to obtain error volume. Figure 7 shows the DFS, PMS and RRE values with  $\sigma_a$  of 100 %, 50 %, 30 %, 20 % and 10 %. Here the calculation results for all 20 profiles were averaged. The DFS values decreased as the  $\sigma_a$  value decreased for all wavelength combinations considered in this study. This behavior was also observed for the RRE results. The PMS value seemed to locate in the corresponding vertical region when the  $\sigma_a$  value increased. The behavior of the relative differences of DFS, PMS and RRE for the  
10 different wavelength combination was the same for all  $\sigma_a$  cases. Increase of retrieval sensitivity of the LMT ozone amount by adding the MW measurement was observed for all  $\sigma_a$  cases considered in this sensitivity study.

Our results show that introducing MW limb measurement increases the sensitivity of tropospheric ozone profile retrieval. However, several factors may cause bias and uncertainty in the retrieval results and thus should be considered before the proposed retrieval method is implemented. Discrepancy in the spectroscopic parameters for several wavelength ranges is a  
15 major source of errors. For ozone profile retrieval using MW limb measurement, the spectroscopic parameters are the major error source. It was reported that an error of approximately 3–5 % was caused by uncertainties in the air-broadening coefficient and line intensity in the case of the SMILES observation (Kasai et al., 2013), which 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  
20 parameter to determine the field-of-view, so uncertainty in the tangent height causes a discrepancy between the atmospheric layer assumed in the simulation and the true atmospheric layer. This discrepancy critically affects the retrieval of the ozone profile in both the stratosphere and the troposphere, and may cause bias in the correction of the time delay between MW limb measurement and other nadir measurements. In this study, we assumed that the instruments onboard the ISS (low Earth orbit) and that a time difference of approximately five minutes could be ignored. If the time difference is actually long enough to  
25 need correction, three-dimensional atmospheric modeling including the field-of-view of the MW limb measurement should be performed.

As a whole, it was shown that retrieval of the tropospheric ozone profile was improved by adding MW limb measurements to UV and TIR nadir measurements. In the LMT region, the DFS value was estimated to be increased by approximately 30 %. The DFS values were estimated to be 0.75 and 0.66 over land and ocean, respectively, for the upcoming IASI-NG and UVNS  
30 missions (Constantino et al., 2017). If MW limb measurement is implemented to the synergetic retrieval, the DFS value is estimated to increase to 0.98 and 0.86 over land and ocean, respectively. Our feasibility study has shown that it is possible to retrieve the ozone profile in the LMT with a DFS value of unity.

## 5 Conclusions

- We performed a feasibility study of obtaining **vertically resolved ozone profiles in the troposphere with** synergetic retrieval using **various combinations of three** wavelength ranges (UV, TIR, and MW). **The observation** geometries used in this study were the nadirs for the UV and TIR measurements and limb for the MW measurement from **low Earth orbit (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 **tropospheric ozone profile retrieval for** 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) **and** the reduction rate of error (RRE) for the partial column were also used as an indicator of the sensitivity evaluation.
- 10 The TIR measurement was most sensitive for retrieving **the ozone profile** in the UT when only one wavelength range was used. The **addition of** MW measurement was most effective at improving sensitivity in the UT when combining several wavelength ranges. The DFS values in the UT for all 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
- 15 more than unity by adding either the UV or MW **measurement**. The UV and TIR measurements were dominant in the retrieval of **the ozone profile** in the LMT region. The DFS value in the LMT **greatly** depended on the ozone abundance. **It was larger for a larger partial column of the LMT ozone. The largest DFS value in the LMT for UV+TIR+MW measurement ( $1.03 \pm 0.01$ ) was obtained for** CEC in June which is the case **with** the largest LMT ozone enhancement with the partial column in the LMT ozone (**approximately  $5 \times 10^{21} \text{ m}^{-2}$** ).
- 20 The MW limb measurement alone **provided** less information in the MT and LMT regions. The DFS values were less than 0.01 and the PMS was located **in a** vertical region higher than the MT and LMT. Nevertheless, adding MW **measurement to the combined UV and TIR measurement** 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 **measurement to the UV+TIR measurement. This indicates that reducing uncertainty about ozone abundance in the stratosphere may be important**
- 25 **for accurately estimating the 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

30 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.

## References

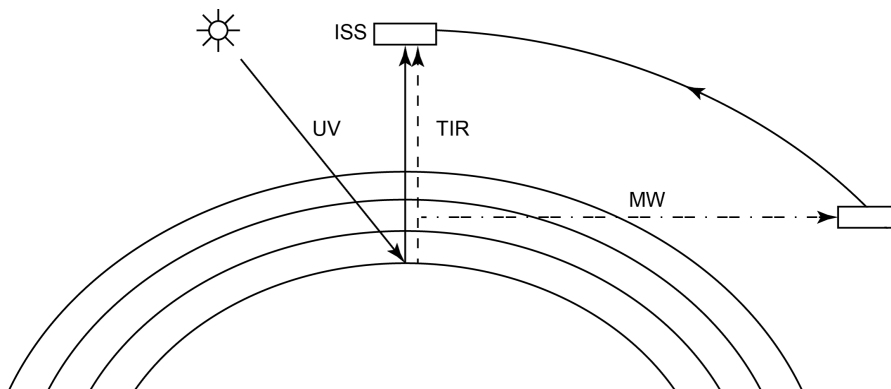
- Bak, J., Kim, J. H., Spurr, R. J. D., Liu, X. and Newchurch, M. J.: Sensitivity study of ozone retrieval from UV measurements on geostationary platforms, *Remote Sensing of Environment*, 118, 309–319, 2012.
- Baldrige, A. M., Hook, S. J., Grove, C. I., and Rivera, G.: The ASTER spectral library version 2.0, *Remote Sensing of Environment*, 113, 711–715, 2009.
- Baron, P., Mendrok, J., Kasai, Y., Ochiai, S., Seta, T., Sagi, K., Suzuki, K., Sagawa, H., and Urban, J.: AMATERAU: Model for atmospheric terahertz radiation analysis and simulation, *Journal of National Institute of Information and Communications Technology*, 55, 109–121, 2008.
- Baron, P., Urban, J., Sagawa, H., Möller, J., Murtagh, D. P., Mendrok, J., Dupuy, E., Sato, T. O., Ochiai, S., Suzuki, K., Manabe, T., Nishibori, T., Kikuchi, K., Sato, R., Takayanagi, M., Murayama, Y., Shiotani, M., and Kasai, Y.: The Level 2 research product algorithms for the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), *Atmospheric Measurement Techniques*, 4, 2105–2124, doi:10.5194/amt-4-2105-2011, 2011.
- Brinksma, E. J., Bracher, A., Lolkema, D. E., Segers, A. J., Boyd, I. S., Bramstedt, K., Claude, H., Godin-Beekmann, S., Hansen, G., Kopp, G., Leblanc, T., McDermid, I. S., Meijer, Y. J., Nakane, H., Parrish, A., von Savigny, C., Stebel, K., Swart, D. P. J., Taha, G. and PETERS, A. J. M.: Geophysical validation of SCIAMACHY limb ozone profiles, *Atmospheric Chemistry and Physics*, 6, 197–209, 2006.
- Clerbaux, C., Boynard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D., Pommier, M., Razavi, A., Turquety, S., Wespes, C. and Coheur, P. F.: Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder, *Atmospheric Chemistry and Physics*, 9, 6041–6054, 2009.
- Clough, S. A., Shephard, M. W., Mlawer, E. J., Delamere, J. S., Iacono, M. J., Cady-Pereira, K., Boukabara, S., and Brown, P. D.: Atmospheric radiative transfer modeling: a summary of the AER codes, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 91, 233–244, 2005.
- Clough, S. A., Shephard, M. W., Worden, J., Brown, P. D., Worden, H. M., Luo, M., Rodgers, C. D., Rinsland, C. P., Goldman, A., Brown, L., Kulawik, S. S., Eldering, A., Lampel, M., Osterman, G., Beer, R., Bowman, K., Cady-Pereira, K. E., and Mlawer, E. J.: Forward model and Jacobians for tropospheric emission spectrometer retrievals, *IEEE Transactions on Geoscience and Remote Sensing*, 44, 1308–1323, 2006.
- 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.
- Dentener, F., Keating, T., and Akimoto, H.: Hemispheric transport of air pollution, United Nations, ISBN, pp. 978–992, 2010.
- Fleming, E. L., Chandra, S., Schoeberl, M. R. and Barnett, J. J.: Monthly mean global climatology of temperature, wind, geopotential height, and pressure for 0–120 km, NASA Tech Memo 100687, 85, 1988.
- 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.

- 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.
- Fujinawa, T., Kasai, Y., Mahani, M., uvSCOPE team and APOLLO mission team: Japanese future mission for air quality uvSCOPE and APOLLO, 8th Atmospheric Limb workshop Abstracts, 2015.
- 5 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.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C. and Eder, B.: Fully coupled "online" chemistry within the WRF model, *Atmospheric Environment*, 39, 6957–6975, 2005.
- 10 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, doi:10.5194/acp-15-9865-2015, 2015.
- Hess, M., Koepke, P., and Schult, I.: Optical properties of aerosols and clouds: The software package OPAC, *Bulletin of the American meteorological society*, 79, 831–844, 1998.
- Kasai, Y., Kita, K., Kanaya, Y., Gmap-Asia and Apollo Mission Team: The Japanese Air Pollution Observation Missions, GMAP-Asia and  
15 APOLLO, AGU Fall Meeting Abstracts, 2011.
- Kasai, Y., Sagawa, H., Kuroda, T., Manabe, T., Ochiai, S., Kikuchi, K., Nishibori, T., Baron, P., Mendrok, J., Hartogh, P., Murtagh, D., Urban, J., von Schéele, F., and Frisk, U.: Overview of the Martian atmospheric submillimetre sounder FIRE, *Planetary and Space Science*, 63, 62–82, 2012.
- Kasai, Y., Sagawa, H., Kreyling, D., Dupuy, E., Baron, P., Mendrok, J., Suzuki, K., Sato, T. O., Nishibori, T., Mizobuchi, S., Kikuchi, K.,  
20 Manabe, T., Ozeki, H., Sugita, T., Fujiwara, M., Irimajiri, Y., Walker, K. A., Bernath, P. F., Boone, C., Stiller, G., von Clarmann, T., Orphal, J., Urban, J., Murtagh, D., Llewellyn, E. J., Degenstein, D., Bourassa, A. E., Lloyd, N. D., Froidevaux, L., Birk, M., Wagner, G., Schreier, F., Xu, J., Vogt, P., Trautmann, T., and Yasui, M.: Validation of stratospheric and mesospheric ozone observed by SMILES from International Space Station, *Atmospheric Measurement Techniques*, 6, 2311–2338, doi:10.5194/amt-6-2311-2013, 2013.
- Kikuchi, K., Nishibori, T., Ochiai, S., Ozeki, H., Irimajiri, Y., Kasai, Y., Koike, M., Manabe, T., Mizukoshi, K., Murayama, Y., Nagahama,  
25 T., Sano, T., Sato, R., Seta, M., Takahashi, C., Takayanagi, M., Masuko, H., Inatani, J., Suzuki, M., and Shiotani, M.: Overview and early results of the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), *Journal of Geophysical Research: Atmospheres*, 115, D23306, doi:10.1029/2010JD014379, 2010.
- Kleipool, Q. L., Dobber, M. R., de Haan, J. F., and Levelt, P. F.: Earth surface reflectance climatology from 3 years of OMI data, *Journal of Geophysical Research: Atmospheres*, 113, 2008.
- 30 Kyrölä, E., Tamminen, J., Leppelmeier, G. W., Sofieva, V., Hassinen, S., Bertaux, J. L., Hauchecorne, A., Dalaudier, F., Cot, C., Korablev, O., Fanton d'Andon, O., Barrot, G., Mangin, A., Théodore, B., Guirlet, M., Etanchaud, F., Snoeij, P., Koopman, R., Saavedra, L., Fraisse, R., Fussen, D. and Vanhellefont, F.: GOMOS on Envisat: An overview, *Advances in Space Research*, 33, 1020–1028, 2004.
- 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.
- 35 Levelt, P. F., van den Oord, G. H. J., Dobber, M. R., Malkki, A., Visser, H., de Vries, J., Stammes, P., Lundell, J. O. V., and Saari, H.: The ozone monitoring instrument, *IEEE Transactions on geoscience and remote sensing*, 44, 1093–1101, 2006.
- Munro, R., Eisinger, M., Anderson, C., Callies, J., Corpaccioli, E., Lang, R., Lefebvre, A., Livschitz, Y., and Albinana, A. P.: GOME-2 on MetOp, in: *Proc. of The 2006 EUMETSAT Meteorological Satellite Conference*, Helsinki, Finland, pp. 12–16, 2006.

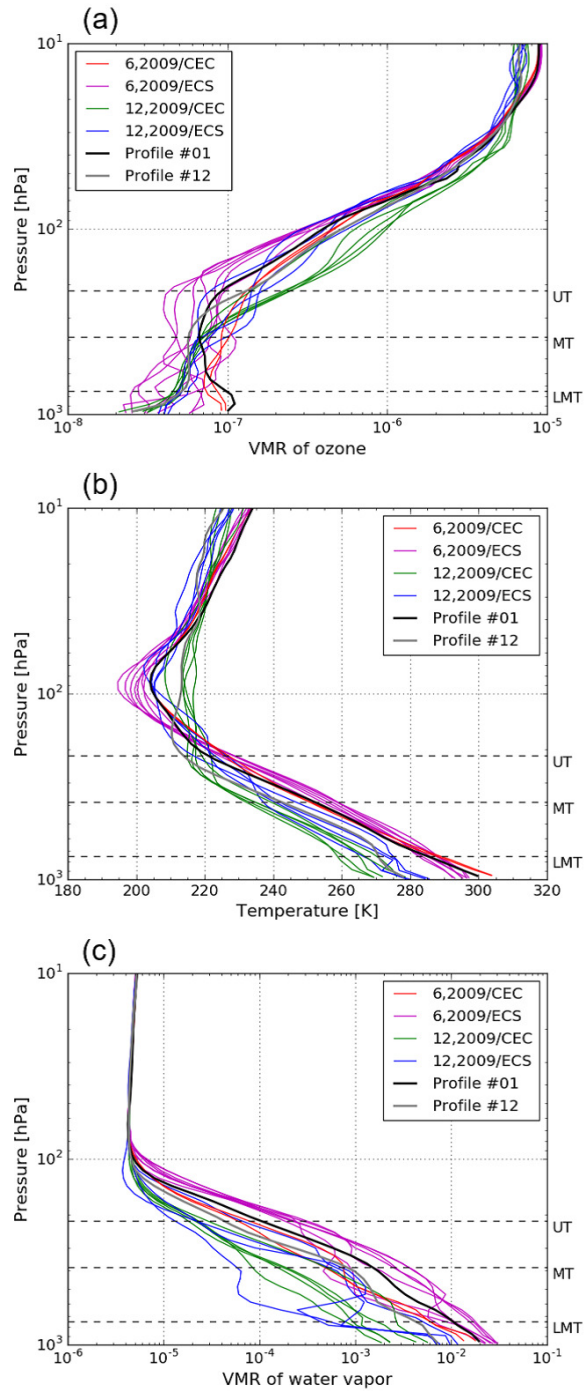


- Natraj, V., Liu, X., Kulawik, S., Chance, K., Chatfield, R., Edwards, D. P., Eldering, A., Francis, G., Kurosu, T., Pickering, K., Spurr, R. and Worden, H.: Multi-spectral sensitivity studies for the retrieval of tropospheric and lowermost tropospheric ozone from simulated clear-sky GEO-CAPE measurements, *Atmospheric Environment*, 45, 7151–7165, 2011.
- 5 Noguchi, K., Richter, A., Rozanov, V., Rozanov, A., Burrows, J. P., Irie, H., and Kita, K.: Effect of surface BRDF of various land cover types on geostationary observations of tropospheric NO<sub>2</sub>, *Atmospheric Measurement Techniques*, 7, 3497–3508, 2014.
- Osterman, G. B., Kulawik, S. S., Worden, H. M., Richards, N. A. D., Fisher, B. M., Eldering, A., Shephard, M. W., Froidevaux, L., Labow, G., Luo, M., Herman, R. L., Bowman, K. W., and Thompson, A. M.: Validation of Tropospheric Emission Spectrometer (TES) measurements of the total, stratospheric, and tropospheric column abundance of ozone, *Journal of Geophysical Research: Atmospheres*, 113, 2008.
- 10 Pardo, J. R., Serabyn, E., and Cernicharo, J.: Submillimeter atmospheric transmission measurements on Mauna Kea during extremely dry El Niño conditions: Implications for broadband opacity contributions, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 68, 419–433, 2001.
- Pickett, H. M., Poynter, R. L., Cohen, E. A., Delitsky, M. L., Pearson, J. C., and Müller, H. S. P.: Submillimeter, millimeter and microwave spectral line catalog, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 60, 883–890, doi:10.1016/S0022-4073(98)00091-0, 1998.
- 15 Rienecker, M. M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M. G., Schubert, S. D., Takacs, L., Kim, G. K., Bloom, S., Chen, J., Collins, D., Conaty, A., da Silva, A., Gu, W., Joiner, J., Koster, R. D., Lucchesi, R., Molod, A., Owens, T., Pawson, S., Pegion, P., Redder, C. R., Reichle, R., Robertson, F. R., Ruddick, A. G., Sienkiewicz, M., and Woollen, J.: MERRA: NASA’s modern-era retrospective analysis for research and applications, *Journal of Climate*, 24, 3624–3648, 2011.
- Rodgers, C. D.: *Inverse methods for atmospheric sounding: Theory and practice*, Series on Atmospheric, Oceanic and Planetary Physics, 20 World Scientific, 2, 3605–3609, 2000.
- Rothman, L. S., Gordon, I. E., Barbe, A., Benner, D. C., Bernath, P. F., Birk, M., Boudon, V., Brown, L. R., Campargue, A., Champion, J. P., Chance, K., Coudert, L. H., Dana, V., Devi, V. M., Fally, S., Flaud, J. M., Gamache, R. R., Goldman, A., Jacquemart, D., Kleiner, I., Lacombe, N., Lafferty, W. J., Mandin, J. Y., Massie, S. T., Mikhailenko, S. N., Miller, C. E., Moazzen-Ahmadi, N., Naumenko, O. V., Nikitin, A. V., Orphal, J., Perevalov, V. I., Perrin, A., Predoi-Cross, A., Rinsland, C. P., Rotger, M., Šimečková, M., Smith, M. A. H., 25 Sung, K., Tashkun, S. A., Tennyson, J., Toth, R. A., Vandaele, A. C., and Vander Auwera, J.: The HITRAN 2008 molecular spectroscopic database, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 110, 533–572, doi:10.1016/j.jqsrt.2009.02.013, 2009.
- Rozanov, A., Rozanov, V., Buchwitz, M., Kokhanovsky, A., and Burrows, J. P.: SCIATRAN 2.0—A new radiative transfer model for geophysical applications in the 175–2400 nm spectral region, *Advances in Space Research*, 36, 1015–1019, 2005.
- Saitoh, N., Imasu, R., Ota, Y., and Niwa, Y.: CO<sub>2</sub> retrieval algorithm for the thermal infrared spectra of the Greenhouse Gases Observing 30 Satellite: Potential of retrieving CO<sub>2</sub> vertical profile from high-resolution FTS sensor, *Journal of Geophysical Research: Atmospheres*, 114, D17305, doi:10.1029/2008JD011500, 2009.
- Sellitto, P., Del Frate, F., Solimini, D., and Casadio, S.: Tropospheric ozone column retrieval from ESA-Envisat SCIAMACHY nadir UV/VIS radiance measurements by means of a neural network algorithm, *IEEE Transactions on Geoscience and Remote Sensing*, 50, 998–1011, doi:10.1109/TGRS.2011.2163198, 2012a.
- 35 Sellitto, P., Di Noia, A., Del Frate, F., Burini, A., Casadio, S., and Solimini, D.: On the role of visible radiation in ozone profile retrieval from nadir UV/VIS satellite measurements: An experiment with neural network algorithms inverting SCIAMACHY data, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 113, 1429–1436, doi:10.1016/j.jqsrt.2012.04.007, 2012b.

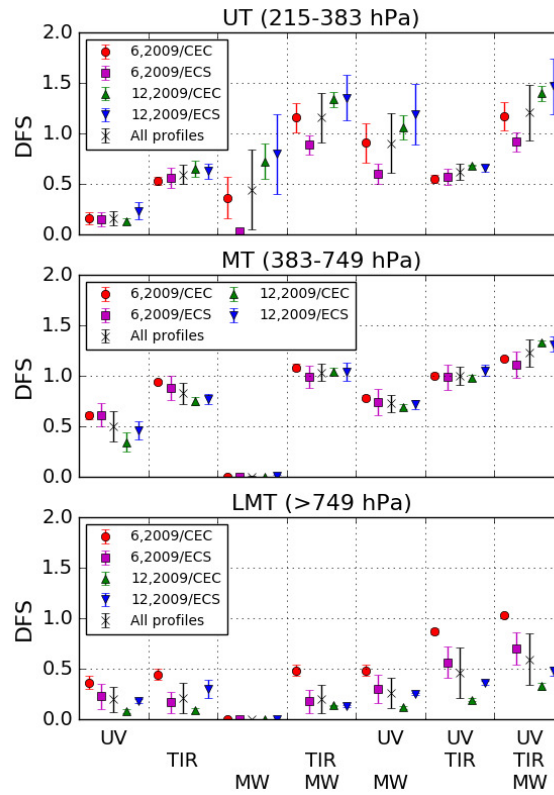
- Sudo, K., Takahashi, M., Kurokawa, J., and Akimoto, H.: CHASER: A global chemical model of the troposphere 1. Model description, *Journal of Geophysical Research: Atmospheres*, 107, 2002.
- Takigawa, M., Niwano, M., Akimoto, H., and Takahashi, M.: Development of a one-way nested global-regional air quality forecasting model, *Sola*, 3, 81–84, 2007.
- 5 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.
- Urban, J., Lautié, N., Le Flochmoën, E., Jiménez, C., Eriksson, P., de La Noë, J., Dupuy, E., Ekström, M., El Amraoui, L., Frisk, U., Murtagh, D., Olberg, M., and Ricaud, P.: Odin/SMR limb observations of stratospheric trace gases: Level 2 processing of ClO, N<sub>2</sub>O, HNO<sub>3</sub>, and O<sub>3</sub>, *Journal of Geophysical Research: Atmospheres*, 110, 2005.
- 10 Waters, J. W., Froidevaux, L., Harwood, R. S., Jarnot, R. F., Pickett, H. M., Read, W. G., Siegel, P. H., Cofield, R. E., Filipiak, M. J., Flower, D. A., Holden, J. R., Lau, G. K., Livesey, N. J., Manney, G. L., Pumphrey, H. C., Santee, M. L., Wu, D. L., Cuddy, D. T., Lay, R. R., Loo, M. S., Perun, V. S., Schwartz, M. J., Stek, P. C., Thurstans, R. P., Boyles, M. A., Chandra, K. M., Chavez, M. C., Chen, G. S., Chudasama, B. V., Dodge, R., Fuller, R. A., Girard, M. A., Jiang, J. H., Jiang, Y., Knosp, B. W., LaBelle, R. C., Lam, J. C., Lee, K. A., Miller, D., Oswald, J. E., Patel, N. C., Pukala, D. M., Quintero, O., Scaff, D. M., Snyder, W. V., Tope, M. C., Wagner, P. A., and Walch, M. J.:
- 15 The earth observing system microwave limb sounder (EOS MLS) on the Aura satellite, *IEEE Transactions on Geoscience and Remote Sensing*, 44, 1075–1092, 2006.
- WHO: Burden of disease from Ambient Air Pollution for 2012, Geneva: World Health Organization, 2014.
- Worden, J., Liu, X., Bowman, K., Chance, K., Beer, R., Eldering, A., Gunson, M., and Worden, H.: Improved tropospheric ozone profile retrievals using OMI and TES radiances, *Geophysical Research Letters*, 34, 2007.
- 20 Ziemke, J. R., Chandra, S., Duncan, B. N., Froidevaux, L., Bhartia, P. K., Levelt, P. F., and Waters, J. W.: Tropospheric ozone determined from Aura OMI and MLS: Evaluation of measurements and comparison with the Global Modeling Initiative's Chemical Transport Model, *Journal of Geophysical Research: Atmospheres*, 111, 2006.



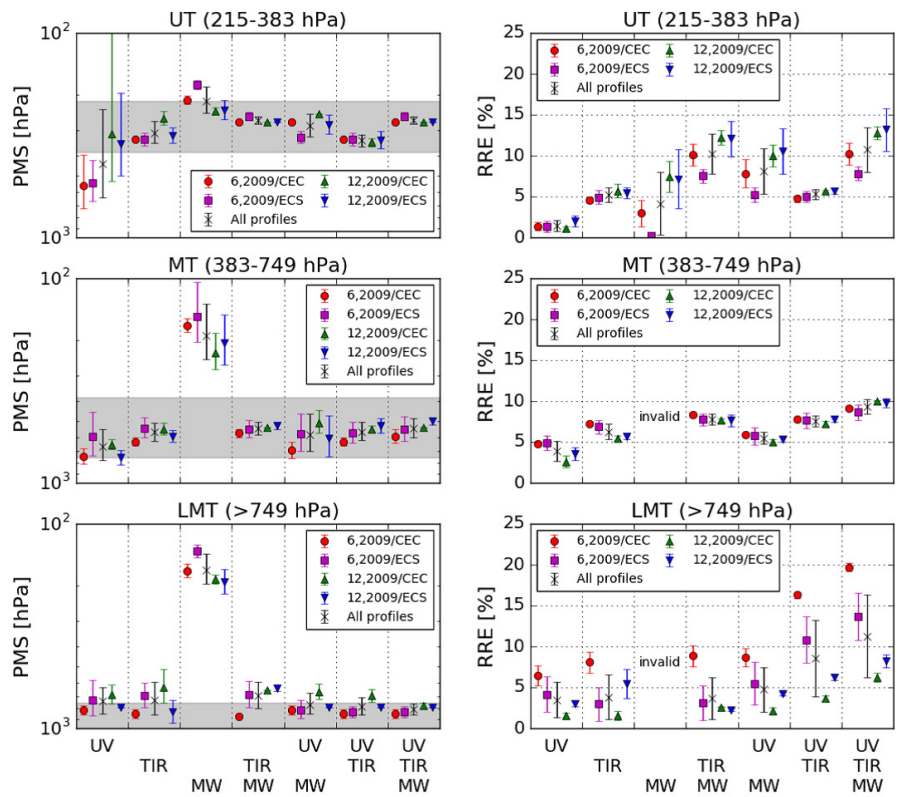
**Figure 1.** Geometries used for observation: nadir (UV and TIR) and limb (MW).



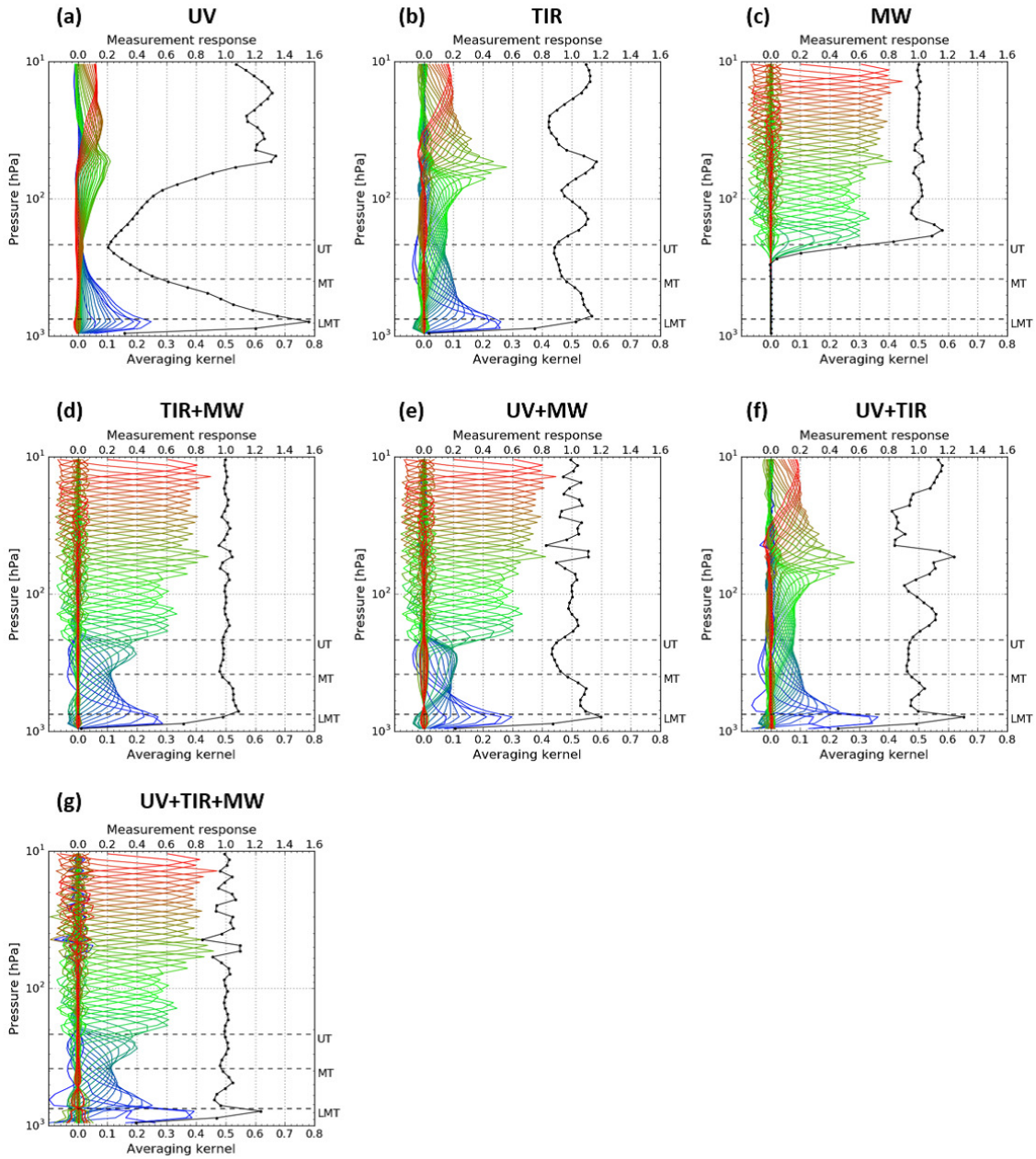
**Figure 2.** Twenty atmospheric scenarios used in this study: (a) VMR of ozone, (b) temperature, and (c) VMR of water vapor. They are divided into four groups, as denoted by color: CEC–June 2009 (red), ECS–June 2009 (purple), CEC–December 2009 (green), and ECS–December 2009 (blue). Two example profiles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



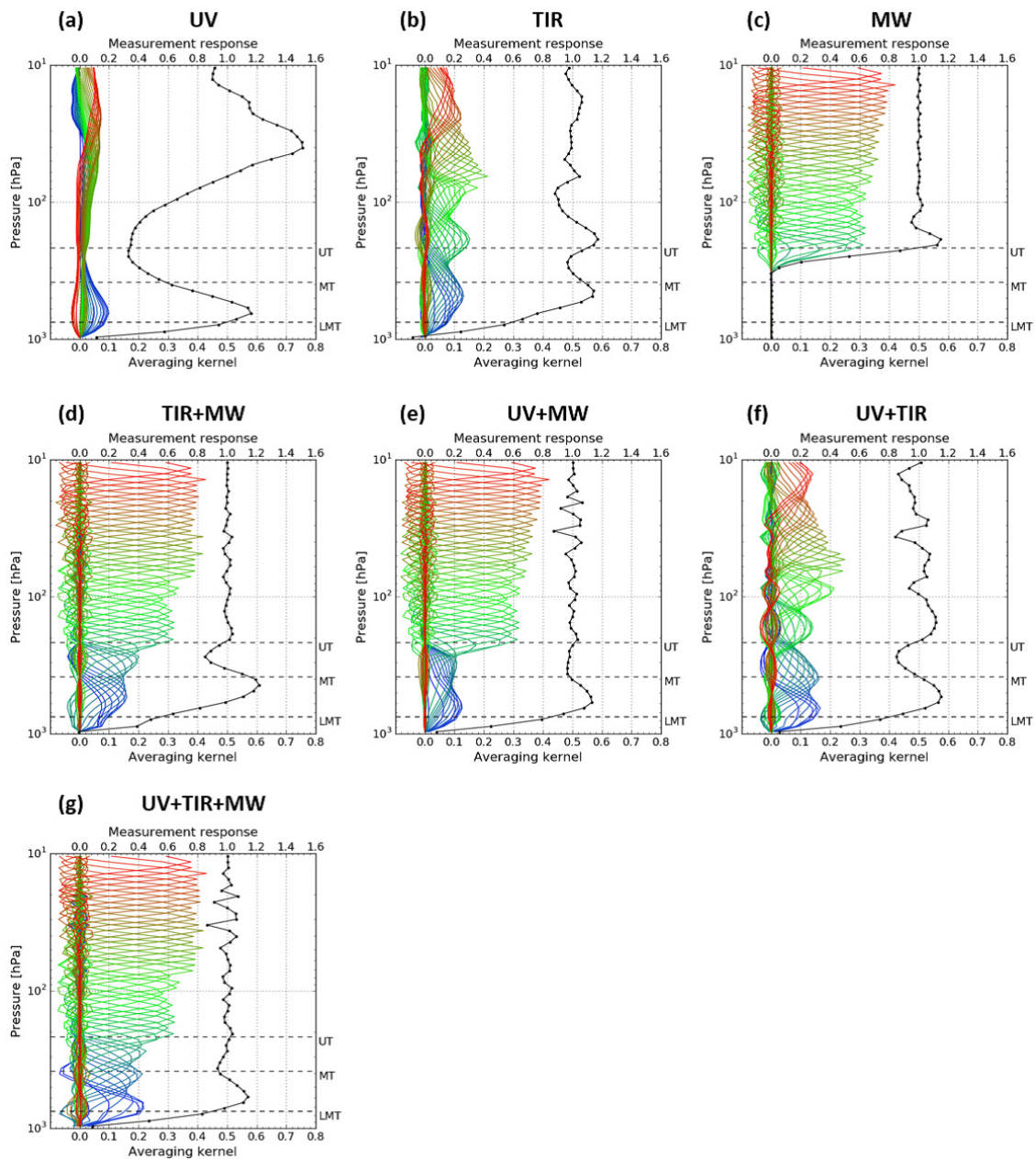
**Figure 3.** Values of DFS for upper troposphere (UT, 215–383 hPa), middle troposphere (MT, 383–749 hPa), and lowermost troposphere (LMT, >749 hPa): CEC–June 2009 (red), ECS–June 2009 (purple), CEC–December 2009 (green), ECS–December 2009 (blue), and 20 profiles averaged (black).



**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.

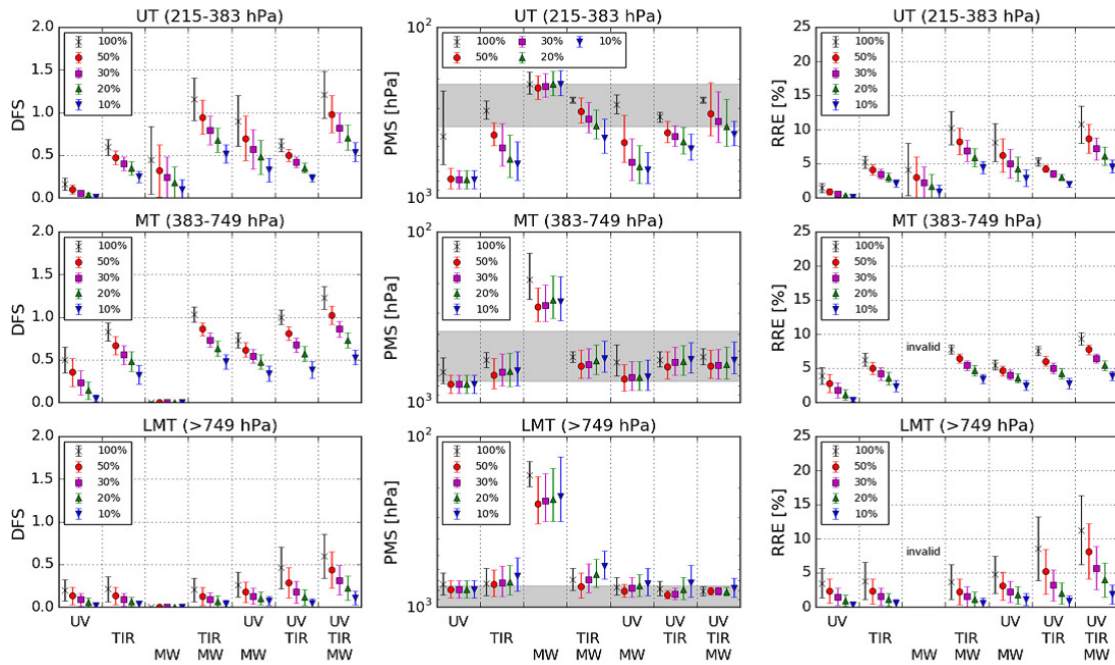


**Figure 5.** Averaging kernels for atmospheric profile #01 (more ozone enhancement in LMT, CEC–June 2009) for seven wavelength measurements: (a) UV, (b) TIR, (c) MW, (d) UV+TIR, (e) UV+MW, (f) TIR+MW, and (g) UV+TIR+MW. Measurement responses are shown by black dots and lines.



**Figure 6.** Same as Fig. 5 but for profile #12 (less ozone in LMT, CEC–December 2009).





**Figure 7.** Values of DFS, PMS and RRE for upper troposphere (UT, 215–383 hPa), middle troposphere (MT, 383–749 hPa), and lowermost troposphere (LMT, >749 hPa) for 20 profiles averaged:  $\sigma_a$  values of 100 % (black), 50 % (red), 30 % (purple), 20 % (green) and 10 % (blue) of the log-based a priori VMR.

**Table 1.** Specifications of three assumed instruments and radiative transfer models.

	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> **Instrument** sensitivity is described in commonly used way for each spectral region: SNR for UV and TIR and noise equivalent brightness temperature for MW.

**Table 2.** Estimation of three reference SNR value in the UV simulation.

	SNR at 305 nm	SNR at 340 nm
Case 1: high level radiance (albedo 90 %, SZA 0°)	~200	~2550
Case 2: middle level radiance (albedo 25 %, SZA 45°)	~60	~1200
Case 3: low level radiance (albedo 5 %, SZA 80°)	~10	~450

**Table 3.** 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 4.** 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). Standard deviation is shown in parentheses. For example, 0.16(6) represents a mean value of 0.16 and a standard deviation of 0.06.

		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 5.** Comparison of DFS in the LMT region with those of previous studies. Method and scenarios differed, so relative difference between DFS value for UV+TIR measurement ( $DFS_{UV+TIR}$ ) and mean of DFS values for UV and TIR measurements ( $DFS_{UV}$ ,  $DFS_{TIR}$ ) was used for 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.