a point-by-point response to the reviews

Response to referee #1's comments

We replied to 9 main comments and 9 minor comments hereafter.

Main Comment

1-3. this paper is about the testing of the GEMS retrieval algorithm on OMI radiances, and the subsequent comparison with ozonesondes. This is interesting and worthwhile study in the run-up to the launch of GEMS. I see the paper as having two main purposes with respect to GEMS. First, it is an exercise of the retrieval algorithm on real OMI data. The fact that this is successful gives confidence that the retrieval is ready to receive the first GEMS data after launch. However, quantitative verification of the retrieval performance is harder, and the discussion of the GEMS retrieval algorithm performance on OMI radiances against sondes, compared with the OMI-algorithm retrievals against the same sondes should be expanded. The second purpose is to identify those ozonesonde measurements that might be good for GEMS validation, in as much as the work here suggests that they are useful or not for OMI validation. If there is a cross-verification here, it is really about OMI validation between OMI and the radiosondes. The fact that the GEMS algorithm is used to process the OMI radiances does not change this, especially with comparisons that should adequately account for how a priori profile and smoothing error assumptions differ between the GEMS and usual OMI algorithms. As such, the title of the manuscript does not clearly describe what is done in the paper, and I suggest that the authors modify the title to better reflect the above two goals.

- As indicated by this reviewer, the simulated GEMS retrievals are similar to OMI retrievals (PROFOZ), but with different and better implementations. Therefore the cross-verification performed in this paper is actually close to OMI validation than GEMS validation. The smoothing errors between OMI and GEMS have been addressed in Bak et al. (2013), indicating that GEMS will provide the comparable ozone profile information below ~ 22 km and the reduced information up to ~ 40 km. The tropopause-based ozone profile climatology is implemented as a priori ozone information for GEMS. However, the goal of this paper is not to detail the difference between OMI and GEMS ozone profile retrievals, but to evaluate the simulated GEMS tropospheric ozone retrievals using the limited UV information (300-330 nm) and to prepare the good reference dataset for GEMS validation. The quality comparison of simulated GEMS retrievals with OMI retrievals are additionally performed to demonstrate the confidence of the presented GEMS ozone profile algorithm. According to reviewer's suggestion, the title is changed to "Cross-evaluation of GEMS tropospheric ozone retrieval performance using OMI data and the use of ozonesonde dataset over East Asia for validation" to clearly reflect what we did through this work.

4. **Section 2.1** describes the retrieval algorithm applied to the OMI radiances. A discussion should be included as to how the retrieval algorithm characteristics are expected to change for the GEMS radiances.

- GEMS is a scanning Uv/Visible spectrometer with a single UV-enhanced CCD for the spectral measurements of 300-500 nm (FWHM: 0.6 nm, spectral sampling: 0.2 nm) with at least comparable radiometric/wavelength accuracy (4% including light source uncertainty/0.01 nm) as OMI. However, GEMS data processing is expected to be different tofrom OMI mainly in two ways: 1) OMI uses a depolarizer to scramble the polarization of light. However, GEMS has polarization sensitivity (required to be less than 2%) and performs polarization correction using an RTM-based look-up table of atmospheric polarization state and pre-flight characterization of polarization instrument polarization sensitivity in the level 0 to 1b data processing. The GEMS polarization correction is less accurate and hence additional fitting process might be required in the level 2 data processing, especially for ozone profiles that have more significant retrieval sensitivityare more sensitive to the polarization error compared to other trace-gases. 2) GEMS has a capability to perform diurnal observations and hence the diurnal meteorological input data are required to account for the temperature dependent Huggins band ozone absorption. Hence, the numerical weather prediction (NWP) model analysis data will be transferred to the GEMS Sscience Ddata Pprocessing Ccenter (SDPC). This part is clarified in the Section 2.1 of the revised manuscript.

5. **Section 3.1**. The discussion of the differences between satellite/sonde agreements at the different sites is interesting. In addition to the differences between the sonde characteristics and reliabilities, one might expect greater standard deviations at sites that are polluted and/or show greater variability in ozone loadings due to meteorology. Some further discussion would be useful about the chemical-transport environment before eliminating sites from potential GEMS validation based on instrumentation/ experimental method arguments alone. It would also help this reader if the current dense text were broken up into descriptions of the various reasons for good/bad agreement.

- We have added the figure 4 in which the seasonal mean and standard deviations of ozonesonde measurements are presented to see the stability and characteristics of ozonesonde measurements at each site. Instabilities of measurements are observed from New Delhi ozonesondes. High surface ozone concentrations at Trivandrum in summer are believed to be caused by measurement errors because low levels of pollutants have been reported at this site under the geolocation and meteorological effects (Lal et al. 2000). Besides Trivandrum, Naha could be regarded as background sites according to low surface ozone and its precursor concentrations compared to neighboring stations (Fig. 2 and Fig. 4), and previous studies (Oltmans et al., 2004; Liu et al., 2002). In the lower troposphere high ozone concentrations are captured at

Pohang, Tsukuba, and Sapporo in the summer due to enhanced photochemical production of ozone in daytime, whereas tropical sites, Naha, Hanoi, and Hong Kong show the ozone enhancement in spring mainly due to biomass burning in Southeast Asia, with low ozone concentrations in summer due to the Asian monsoon and in winter due to the tropical air intrusion (Liu et al., 2002; Ogino et al., 2013). Singapore and Kuala lump are supposed to be severely polluted area, but ozone pollution is not clearly captured over the seasons. It could be explained by the morning observation time at these two stations. In addition, instabilities of Singapore measurements are noticeable, including abnormally large variability and very low ozone concentration in the stratosphere. The effect of stratospheric intrusions on the ozone profile shape is dominant at mid-latitudes (Pohang, Tsukuba, and Sapporo) during the spring and winter when the ozone pause goes down to 300 hPa, with larger ozone variabilities in the lower stratosphere and upper troposphere, whereas ozonepause is around 100 hPa with much less variability of ozone in other seasons. This discussion has been included in Section 3.

- Lal, S., Naja, M., and Subbaraya, B: Seasonal variations in surface ozone and its precursors over an urban site in India, Atmospheric Environment, Volume 34, Issue 17, 2000, Pages 2713-2724, 2000.
- Liu, H., D. J. Jacob, L. Y. Chan, S. J. Oltmans, I. Bey, R. M. Yantosca, J. M. Harris, B. N. Duncan, and R. V. Martin, Sources of tropospheric ozone along the Asian Pacific Rim: An analysis of ozonesonde observations, J. Geophys. Res., 107(D21), 4573, doi:10.1029/2001JD002005, 2002.
- Ogino, S.-Y., M. Fujiwara, M. Shiotani, F. Hasebe, J. Matsumoto, T. H. T. Hoang, and T. T. T. Nguyen (2013), Ozone variations over the northern subtropical region revealed by ozonesonde observations in Hanoi, J. Geophys. Res. Atmos., 118, 3245–3257, doi:10.1002/jgrd.50348.

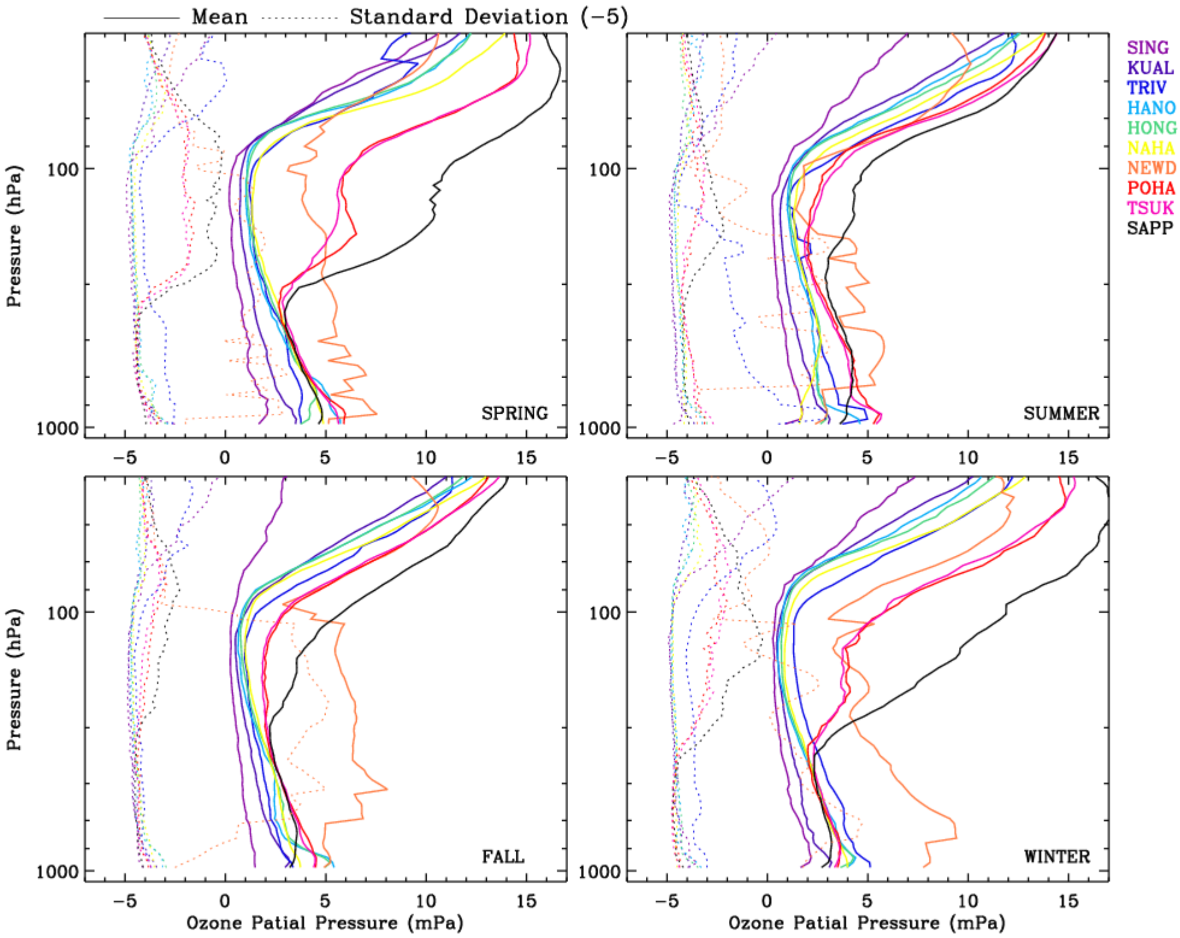


Fig. 4. Seasonal mean (solid) and standard deviation (dashed) of ozonesonde soundings from 2005 to 2015 at 10 sites. 5 mPa is subtracted to standard deviations to fit in the given x-axis.

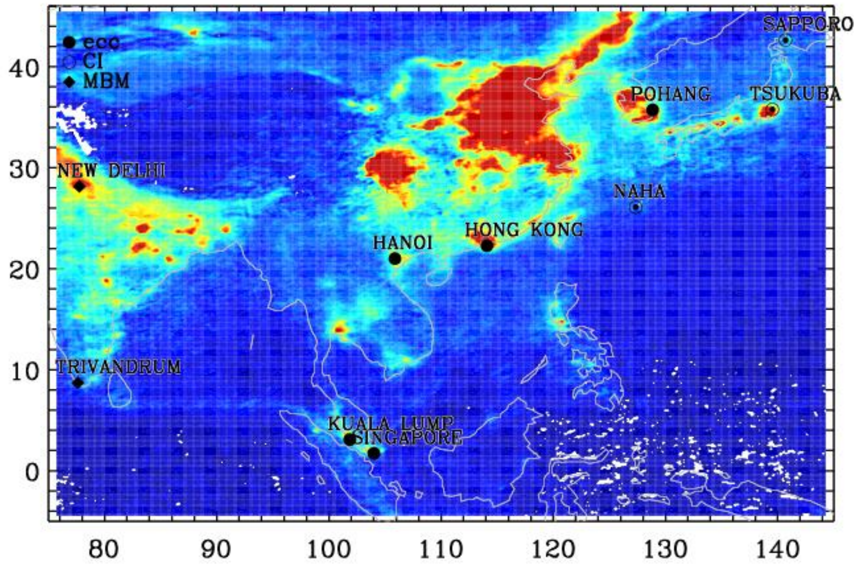
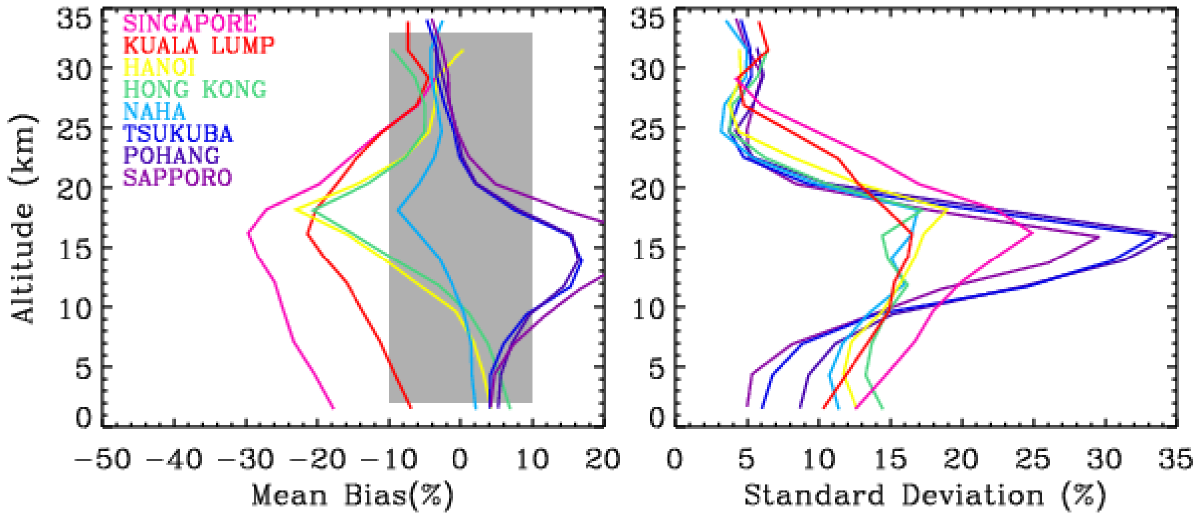


Figure 2. Geographic locations of the ozonesonde stations available since 2005 over the GEMS observation domain. The background map illustrates the OMI NO_2 monthly mean in June 2015.

6. **Section 4.2 and Fig. 7**. The impact on correlation of smoothing or not smoothing the sonde profiles might be dependent on how close the GEMS retrieval a priori profile is to the sonde "truth". How do these compare and how do they vary between locations of good and poor comparison? How far does a priori profile go toward explaining the bias?

- To explain this impact, the comparison between GEMS a priori and ozonesondes is presented below, similarly to Fig.6 (in old manuscript, it is Fig. 7 in the revised one). This a priori information is taken from the tropopause-based ozone profile climatology (TB) which adjusts a monthly and zonal mean ozone profile with a daily tropopause height. Bak et al. (2013) demonstrated that TB based a priori better represents the ozone variabilities in the extra-tropical upper troposphere and lower stratosphere, especially during the winter and spring when the atmospheric status is strongly controlled by the dynamics. A priori information is very important to the quality of UV ozone profile retrievals, but the retrieved ozone profiles show much better agreement with ozonesondes than a priori ozone profiles, implying that the independent piece of information are available from these UV measurements. We can see that the biases seen in a priori are significantly reduced for most stations except for the Singapore station. Positive biases around 15 km in the a priori at the three mid-latitude sites still remain in the retrievals but with much smaller magnitude. Negative biases around ~17 km at other lower-latitude sites (except for Singapore) are almost eliminated in the retrievals.



S1. Same as Fig 6, but for comparison of ozonesondes and a TB-based priori ozone profiles.

7-8. Section 4.2 should be split into another sub section at Line 354 that starts the discussion of the evaluation of the GEMS algorithm against the OMI algorithm. This should be presented as one of the main results sections of this paper: a quantitative evaluation of the GEMS algorithm against other widely used algorithms based on the same OMI radiances. Given the previous discussion in the paper of the various limitations of some of the sondes, it might additionally be useful to directly compare the results of the different retrieval algorithms and explain differences in results on the basis of different features of the algorithms. This would help make the case that the GEMS algorithm is performing as expected.

- In this paper, the ozonesonde measurements available in the GEMS domain are characterized and validated to better evaluate the performance of the GEMS ozone profile algorithm. The accuracy and precision of simulated GEMS ozone profiles are established against the selected true reference. Additionally, the consistent evaluation is performed for the existing OMI ozone products to check the confidence of the GEMS retrieval algorithm, demonstrating that the comparable or better performance of GEMS ozone profile retrievals in the comparison with ozonesonde measurements. The different validation results between retrieval algorithms were discussed such as "GEMS algorithm is developed based on the heritages of the SAO ozone profile algorithm with several modifications. There are two main modifications: a priori ozone climatology was replaced with a tropopause-based ozone profile climatology to better represent the ozone variability in the tropopause. Irradiance spectra used to normalize radiance spectra and characterize instrument line shapes are prepared by taking 31-day moving average instead of climatological average to take into account for time-dependent instrument degradations. These modifications reduce somewhat spreads in deviations of satellite retrievals from sondes, especially in TCO comparison. KNMI retrievals systematically overestimate the tropospheric ozone by ~ 6 DU (Fig. 9.c), which corresponds to the positive biases of 2-4 % in the integrated total columns of KNMI profiles relative to Brewer observations (Bak et al., 2015). As mentioned in Bak et al. (2015), the systematic biases in ozone retrievals are less visible in SAO-based retrievals (GEMS simulation, OMPROFOZ) as systematic components of measured spectra are taken into account for using an empirical correction called "soft calibration". The GEMS algorithm is very similar to the SAO algorithm except for the use of TB climatology and the impact of TB on the retrievals was discuss in detail in Bak et al. (2013). Also the comparison of SAO and KNMI algorithms were discussed in detail in Bak et al. (2015). So we think that it is more efficient to place the discussion related to Figures 7-9 in the same section and the direct comparison of GEMS and other OMI product is beyond the scope of this paper.

▪

9. The paper requires careful, and extensive, editing for English usage, and cut-paste typos, e.g. line 75, that should have been corrected before manuscript submission.

- This manuscript is going to be carefully revised though native English co-author before the submission of the revised manuscript.

Minor Comments

1. Several times "GEMS" measurements are described. The word "simulated" should be added each time to avoid confusion

- In this revised manuscript, "GEMS measurements" was edited to " <u>simulated GEMS measurements</u>"

2. Line 83: consistent perhaps, but not homogeneous as the authors point out in the text above.

- The indicated sentence was revised from "a homogenous, consistent ozonesonde" to "<u>a consistent ozonesonde</u>".

4. Line 105: Instrument errors certainly, but also instrument design sensitivity.

- The indicated word, "instrumental errors" was revised to "<u>Instrument errors certainly, but also instrument design sensitivity</u>"

5. Line 106: Common geophysical conditions can reduce sensitivity, not just extreme

- The associated sentence was edited from "The impact of a priori information on retrievals become important ~~ under extreme geophysical conditions to ~~ <u>under certain geophysical conditions</u>.

6. Line 123: Information may be limited but is a goal of the GEMS mission. This should be clarified.

- The GEMS mission was originally planned to develop the spectrometer for measuring the tropospheric pollutants, the spectral coverage of 300-500 nm satisfies to observe the tropospheric ozone as well as the lower/middle stratospheric ozone.

7. Line 157: Any more recent references to new measurement technique and instrumentation?

- More references (Thompson et al., 2017; Witte et al., 2017; 2018) are added in this sentence.

8. Line 200: How do these coincidence criteria for OMI and the sondes affect the results? What is the expected variation within the time and space windows? What is the representativeness uncertainty? How do these results here with the OMI comparison inform on the expected GEMS comparisons with hourly measurements at ~7km resolution?

- As mentioned in Section 2.3, the coincidence criteria between satellite and ozonesonde are: ±1.0° in both longitude and latitude and ±12 hours in time and then the closest pixel is selected. The actual spatiotemporal difference is much smaller than this criteria (57.5 km to 66.6 km, ~3 hours). The close collocation can significantly minimize the effects of spatiotemporal variability on the comparison and therefore GEMS validation accuracy could be enhanced compared to OMI, which is newly included in the summary of this paper (The impact of spatiotemporal variability on the comparison will be much reduced for GEMS due to its higher spatiotemporal resolution (7 km x 8 km @ Seoul, hourly) against OMI (48 km x 13 km @ nadir in UV1, daily).

9. Line 231: Is "troposphere" written where is should be stratosphere?

- The indicated sentence was edited to "much coarser vertical resolution of 10-14 km in the troposphere and 7-11 km in the stratosphere".

Response to referee #2's comments

We replied to 4 main comments and 25 minor comments

Main Comment

C1. GEMS ozone profile algorithm is applied to OMI BUV measurements. It should be explained why GEMS radiances has not been simulated instead and what is the impact of using LEO measurements for a GEO instrument.

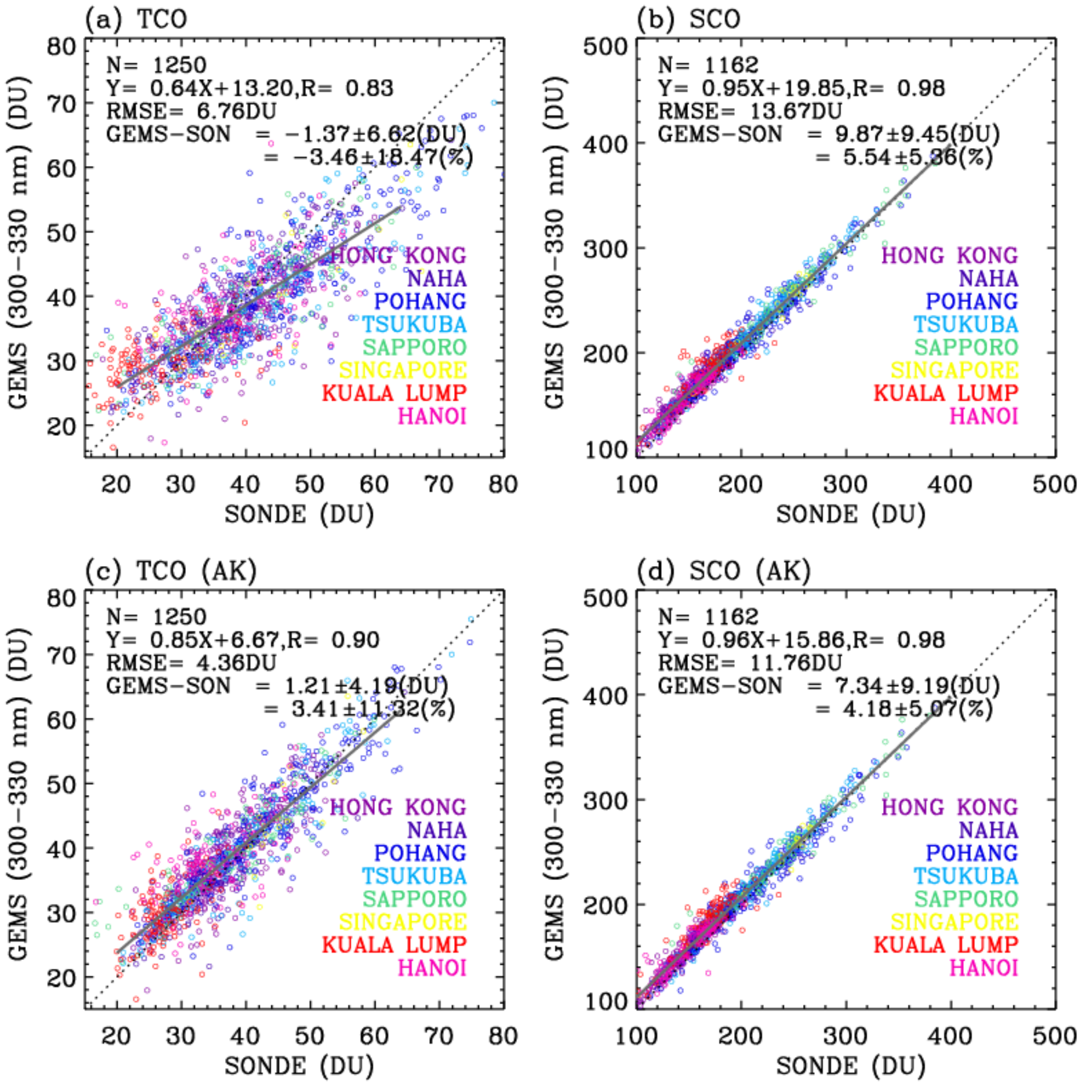
R1. The development of the GEMS L2 algorithm has been in progress with OMI measurements because the simulation of the GEMS radiances using the forward model has not been fully implemented. Two main differences in GEMS and LEO (OMI) data processing could be expected: 1) OMI use a depolarizer to scramble the polarization of light. However, GEMS has polarization sensitivity (required to be less than < 2%) and performs polarization correction using RTM-based look-up table of atmospheric polarization state and pre-flight characterization of polarization sensitivity in the level 0 to 1b data processing. The GEMS polarization correction is less accurate and hence additional fitting process might be required in the level 2 data processing, especially for ozone profiles that have more significant retrieval sensitivity to the polarization error compared to other trace-gases. 2) GEMS has a capability to perform diurnal observation and hence the diurnal meteorological input data are required to account for the temperature dependent Huggins band ozone absorption. Hence the numerical weather prediction (NWP) model analysis data will be transferred to the GEMS science data processing center (SDPC). This response has been also included in the revised manuscript, also according to the comment #4 from reviewer 1.

C2. The use of OMI measurements makes the title of the paper confusing as the validation is of OMI using GEMS algorithm, but not of GEMS. This needs to be changed.

R2. This reply is also corresponding to comment #1 from reviewer 1, the title of this paper is changed to "Cross-evaluation of GEMS tropospheric ozone retrieval performance using OMI data and the use of ozonesonde dataset over East Asia for validation".

C3. Simulated GEMS retrievals are used to verify the ozonesonde observations, i.e., to identify the good stations, and in turn, these stations are used to validate the simulated GEMS retrievals. Using this approach it is hard to expect bad results for the simulated GEMS retrievals. The ozonesonde observations should be considered as the truth, and if they need to be validated and screened, this should be done using an independent dataset, but not the same dataset that we intend to validate, in this case the simulated GEMS retrievals.

R3. Understanding the quality of the reference dataset and then selecting a good reference is a very important process in validating satellite or other in-situ measurements and then in better characterizing the retrieval accuracy and error. Satellite measurements of tropospheric ozone have previously been utilized to disclose problems in ozonesonde observations (e.g., Liu et al., 2006; Huang et al., 2018). We are also using retrievals here to identify ozonesonde measurements with significant errors. However, the station-to-station based quality control has not been typically applied in previous validation works. The figure below demonstrates how much the accuracy of the simulated GEMS retrievals from OMI measurements is underestimated if the station-to-station based quality control is not applied. We also apply the parallel validation for two independent OMI ozone profile products, OMPROFOZ and OMO3PR, respectively, demonstrating that our ozone retrievals are in comparable or better agreement with ozonesondes. As we mentioned in R1 to C1, the simulation of the GEMS radiances using the forward model has not been fully implemented.



S1. Same as Figure 8, but for including all ECC measurements.

C4. According to the results shown, the time frame established of +-12 hours seems too large for the evaluation of tropospheric ozone, especially for mid-latitudes location where a stronger daily cycle can be found.

R4. Based on the previous papers, the collocation of satellite pixel to ozonesonde stations have been performed within 6 to 24 hours. As clarified in Sect. 2.3 such as "The coincidence criteria between satellite and ozonesonde are: ±1.0° in both longitude and latitude and ±12 hours in time and then the closest pixel is selected. The Aura satellite carrying OMI crosses the equator always at ~ 1:45 pm LT and thereby OMI measurements are closely collocated within 3 hours to ozonesonde soundings measured in afternoon (1-3 pm LS)," OMI measurements are closely collocated within 3 hours to ozonesonde soundings measured in afternoon (1-3 pm) from Japanese stations, Pohang, Hong Kong, Hanoi, and Trivandrum. In this paper, the time collocation criterion is set to be 12 hours to include other stations existing over the GEMS domain.

Minor comments

C1. Line 50: Satellite name should be Sentinel-4

R1. This name has been corrected to "Sentinel-4"

C2. Line 75: ":: : have yet to be not been ..." please correct this

R2. It has been corrected to "has not been".

C3. Line 178: Among ECC stations

R3. It has been corrected to "Among ECC stations".

C4. Line 183: "Kula lump", please correct. Also all along the paper, the name of this station is written in different ways (Kuala lump, Kuala Lumpur). Please homogenize the station names in the text, figures and tables.

R4. We carefully checked what this reviewer indicated. This station name has been corrected to "Kuala Lumpur" across the manuscript.

C5. Line 221: biased -> bias

R5. It has been corrected to "bias".

C6. Line 225: Please specify the units

R6. RMS does not have the unit and thereby "RMS (i.e., root mean square of fitting residuals relative to measurement errors) less than 3" has been kept in the revised manuscript.

C7. Line 231: troposphere -> stratosphere

R7. It has been corrected to “stratosphere”.

C8. Line 234: Should be photons?

R8. It has been revised to “photons”.

C9. Line 242: xa should be placed after (1-A)

R9. Eq. 3 has been revised to “$\hat{x}_{sonde} = A \cdot x_{sonde} + (1 - A)x_a$”

C10. Line 282: Please rephrase, maybe “of” -> “with values ranging from”

R10. According to this comment, “satellite retrievals show the distinct seasonal TOC variations with the amplitude of ~ 35-40 DU” has been edited to “~ seasonal TOC variations with the values ranging from ~35 to ~ 40 DU”

C11. Line 290: “Japanese stations” or “stations from Japan”. Same in Line 296.

R11. It has been revised to “stations from Japan”

C12. Line 314: Please unify or explain the differences between LT, LS and LST across the paper

R12. There is no difference. It has been unified to “LT (Local time)”

C13. Line 322: “oznesonde” -> “ozonesonde”

R13. This word has been corrected.

C14. Line 324: Please list stations after “mid-latitude” and refer to Figure justifying this and the following statements.

R14. It has been clarified such as “mid-latitude (Pohang, Tsukuba, and Sapporo)”

C15. Line 326: “- a few %” please rephrase this

R15. It has been corrected to “a few percent”

C16. Line 338: 4.2 -> 3.2.
R16. It has been changed to “3.2”.

C17. Line 358: “... gives the good information ...” please rephrase. SOC has not been defined
R17. It has been corrected to “gives the good information on Stratospheric Ozone Column (SOC)”

“

C18. Line 367: “espeically” -> “especially”. “TCO” -> TOC.
R18. The relevant sentence has been corrected to “especially in the TOC comparison”

C19. Line 308: Shouldn’t it be “latitudinally” as it is used in other parts of the manuscript? Same in Line 398 and Line 400 (in this case, why capital L?)
R19. “latitudinally” was used at lines, 27, 308, 398, and 400, respectively. These have been revised as followings,

- At 27, “compared to latitudinally adjacent stations with Carbon Iodine (CI) and Electrochemical Condensation Cell (ECC).” to “Carbon Iodine (CI) and Electrochemical Condensation Cell (ECC) dataset measured in similar latitude regime”
- At 308, “latitudinally adjacent station, Hong Kong” to “neighboring station, Hong Kong”
- At 398, “latitudinally adjacent Japanese 398 ECC measurements at Tsukuba and Sapporo” to “Japanese ECC measurements at Tsukuba and Sapporo located in mid-latitudes (> 30 °)”
- At 400, at Naha and Hong Kong stations located in similar latitude regime.

C20. Line 399: Extra s “is similarly”
R20. This indicated one (is s similarly) has been corrected (is similarly)

C21. Figure 2: Latitudes and Longitudes are not correct.
R21. This figure has been revised.

C22. Figure 3: Please explain what is CF(O) and CF(X). Even if no CF is applied to MF sondes, it would be interesting to add them in Figure 3.
R22. To clarify, the legend in the figure has been revised to “Solid: with CF, Dash: w/o CF”.

The corresponding caption has been revised to "Effect of applying a correction factor (CF) to (a) ECC and (b) CI ozonesonde measurements, respectively on comparisons with simulated GEMS ozone profile retrievals. Solid and Dashed lines represent the comparisons with and without applying a CF, respectively, at each Japanese station."

C23. Figure 4: Please specify how you differentiate the different type of sondes. Is it using diamonds, full dots and empty dots? Which one is which? Also indicate what is the horizontal axes, eg. "time (years)"
R23. This figure has been revised to clarify the symbols and the title of x-axis.

C24. Figure 6: I would suggest rewritting the last sentence as follows "The relative difference (in %) is defined as 100 X (SONDE AK – GEMS) / (A priori)". Why is multiplied by 2?
R24. This equation has been corrected to "100 X (SONDE AK – GEMS) / (A priori)"

C25. Figure 7 and 8: Please replace TCO -> TOC and SCO -> SOC to be consistent with the text.
R25. This figure has been revised to accept this comment.

a list of all relevant changes

1. All figures have been revised for better visibility, with newly included figure 4.

a marked-up manuscript version

Cross-eEvaluation of GEMS tropospheric ozone retrieval performance using OMI data and the use

of ozonesonde dataset over East Asia for validation~~Cross-verification of simulated GEMS tropospheric ozone retrievals and ozonesonde measurements Over Northeast Asia~~

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Abstract

The Geostationary Environment Monitoring Spectrometer (GEMS) is scheduled to be launched in 2019 on board the GEO-KOMPSAT (GEOstationary KOrea Multi-Purpose SATellite)-2B, contributing as the Asian partner of the global geostationary constellation of air quality monitoring. To support this air quality satellite mission, we perform ~~the~~ a cross-verification of simulated GEMS ozone profile retrievals from OMI (Ozone Monitoring Instrument) data based on the Optimal Estimation and ozonesonde measurements within the GEMS domain, covering from 5°S (Indonesia) to 45°N (south of the Russian border) and from 75°E to 145°E. The comparison between ozonesonde and GEMS shows a significant dependence on ozonesonde types. Ozonesonde data measured by Modified Brewer-Master (MBM~~-M~~) at Trivandrum and New Delhi show inconsistent seasonal ~~-~~variabilities in ~~the~~ tropospheric ozone~~,~~ compared to ~~latitudinally adjacent stations with~~ Carbon Iodine (CI) and Electrochemical Condensation Cell (ECC) ozonesondes at other ~~equipped~~ stations in a similar latitude regime. CI ozonesonde measurements are negatively biased relative to ECC measurements by 2-4 DU~~:~~;~~;~~ ~~a bB~~better agreement ~~with GEMS simulations~~ is achieved ~~with~~ when simulated GEMS ozone retrievals are compared to ECC measurements. ECC ozone data at Hanoi, Kuala Lumpur, and Singapore show abnormally worse agreements with simulated GEMS retrievals ~~among~~ than other ECC measurements. Therefore, ECC ozonesonde measurements at Hong Kong, Pohang, Naha, Sapporo, and Tsukuba are finally identified as an optimal reference dataset. The accuracy of simulated GEMS retrievals is estimated to be ~ 5.0 % for both tropospheric and stratospheric column ozone with the precision of 15 % and 5 %, which meets the GEMS ozone requirements.

1. Introduction

The development of ~~the~~ geostationary ultraviolet (UV)/visible (VIS) spectrometers is ~~highlighted toward~~ a new paradigm in the field of the space-based air quality monitoring. It builds on the polar-orbiting instrument heritage~~s~~ for the last 40 years, which were initiated with the launch of a series of Total Ozone Mapping Spectrometer (TOMS) instruments ~~since~~ starting in 1978 (Bhartia et al., 1996) and consolidated by the Global Ozone Monitoring Experiment (GOME) (ESA, 1995), the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) (Bovensmann et al., 1999), the

Ozone Monitoring Instrument (OMI) (Levelt et al, 2006), GOME-~~/~~2 (EUMETSAT, 2006), the Ozone Mapping and Profiler Suite (OMPS) (Flynn et al., 2014), and the TROPOspheric Monitoring Instrument (TROPOMI) (Veefkind et al., 2012). Three geostationary air quality monitoring missions, including the Geostationary Environmental Monitoring Spectrometer (GEMS) (Bak et al., 2013a) over East Asia, Tropospheric Emissions: Monitoring of P~~p~~ollution (TEMPO) (Chance et al, 2013; Zoogman et al., 2017) over North America, and Sentine~~a~~l-4 (Ingmann et al., 2012) over Europe, are in progress to launch ~~their instruments~~ in the 2019-2022 time frame, ~~which will~~to provide unprecedented hourly measurements of aerosols and chemical pollutants at sub-urban scale spatial resolution (~ 10-50 km^2). These missions will constitute the global geostationary constellation of air quality monitoring.

GEMS will be launched in late 2019~~-~~ or early 2020 on board the GeoKOMPSAT-2B (Geostationary Korea Multi-Purpose Satellite) to measure O_3, NO_2, SO_2, H_2CO, CHOCHO, and aerosols in East Asia (Bak et al., 2013a). Tropospheric ozone is a key species to be monitored due to its critical role in controlling ~~the~~ air-quality as a primary component of photochemical smog, ~~the~~ its self-cleansing capacity as a precursor of the hydroxyl radical, and in controlling the Earth'’s radiative balance as a greenhouse gas.

To support the development of the GEMS ozone profile algorithm, Bak et al. (2013a) demonstrated that the GEMS spectral coverage of 300-500 nm minimizes the loss in the sensitivity to tropospheric ozone despite the lack of most Hartley ozone absorption wavelengths shorter than 300 nm. They further indicated the acceptable quality of the simulated stratospheric ozone retrievals from 212 hPa to 3 hPa (40 km) through comparisons using Microwave Limb Sounder (MLS) measurements. As a consecutive work, this study evaluates simulated GEMS tropospheric ozone retrievals against ozonesonde observations. GEMS ozone retrievals are simulated using an O~~o~~ptimal E~~e~~stimation (OE) based fitting algorithm ~~from~~ with OMI radiances ~~with using the fitting window of~~in the spectral range 300-330 nm in the same way as Bak et al. (2013a). The validation effort is essential to ensuring the quality of GEMS ozone profile retrievals and to verifying the newly implemented ozone profile retrieval scheme. In-situ ozonesonde soundings have been considered to be the best reference, but should be carefully used due to ~~its~~ the spatial and temporal irregularities in instrument types, manufacturers, operating procedures, and correction strategies (Deshler et al., 2017). Compared to TEMPO and Sentinel-4, ~~validating~~ the GEMS validation activity ~~GEMS ozone retrievals~~ is expected to be more challenging for the ozone profile product because of the much sparser distribution of stations and more irregular characteristics of ~~the~~ ozonesonde ~~dataset~~ measurements over the GEMS domain. Continuous balloon-borne observations of ozone are only available ~~from~~ at the Pohang (129.23°E, 36.02°N) site in South Korea, but this site has~~ve~~ ~~yet to be~~ not been thoroughly validated. Therefore the quality assessment of ~~the~~ its ozonesonde data is required before we use this data for GEMS validation~~ activity~~. Compared to ozonesondes, satellite ozone data are less accurate and have much coarser vertical resolution, but more homogenous due to ~~its~~ single data processing for the ~~entire~~ measurements from a single instrument. Therefore, abnormal deviations in satellite-ozonesonde differences from neighboring stations might indicate problems at individual stations (Fioletov et al. 2008). For example, Bak et al. (2015) identified 27 homogenous stations among 35 global Brewer stations available from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) network through comparisons with coincident OMI total ozone data. This study adopts this approach to select a ~~homogenous,~~ consistent ozonesonde dataset among 10 stations available over the GEMS domain based on ~~the~~ comparisons of the tropospheric ozone columns (TOC) between simulated GEMS retrievals and ozonesonde measurements, that is, simulated GEMS retrievals using OMI data ~~retrievals ones are~~ are~~is~~ used to verify the ozonesonde observations. The simulated GEMS retrievals are ultimately evaluated against the ozonesonde dataset identified as a true reference to demonstrate the reliability of our future GEMS ozone product. The simulated GEMS retrievals and ozonesonde dataset are described in Sect. 2.1 and 2.2 with the comparison methodology in Sect. 2.3. Our results are discussed in Sect. 3 and summarized in Sect. 4.

2. Data and Methodology

2.1 Ozone Profile Retrievals

The development of the GEMS ozone profile algorithm builds on the heritage~~s~~ of the Smithsonian Astrophysical Observatory (SAO) ozone profile algorithm which was originally developed for GOME (Liu et al., 2005), continuously adapted for its successors ~~such as~~including OMI (Liu et al., 2010a), GOME-~~/~~2 (Cai et al., 2012), and OMPS (Bak et al., 2017). In addition, the SAO algorithm will be implemented to retrieve TEMPO ozone profiles (Chance et al., 2013; Zoogman et al., 2017). In this algorithm, the well-known optimal estimation (OE) based iterative inversion is applied to estimate the best ozone concentrations from simultaneously minimizing between measured and simulated backscattered UV measurements constrained by the measurement covariance matrix, and between retrieved values and its climatological a priori values constrained by an a priori covariance matrix (Rodgers, 2000). The impact of a priori information on retrievals becomes important when measurement information is reduced due to instrumental errors, ~~certainly, but also~~ instrument design sensitivity (e.g. stray light, dark -current, and read-out smear), and ~~or~~ physically insufficient sensitivities under ~~extreme~~ certain geophysical conditions (e.g. the reduced penetration of incoming UV radiation into the lower troposphere at high solar zenith angles or~~,~~ blocked photon penetration below thick clouds). The described OE-fitting solution $\hat{X}_{i+1}$ can be written, together with cost function χ^2:

$$\hat{X}_{i+1} = \hat{X}_i + \left(K_i^T S_y^{-1} K_i + S_a^{-1}\right)^{-1} \{K_i^T S_y^{-1} [\mathrm{Y} - \mathrm{R}(\hat{X}_i)] - S_a^{-1}(\hat{\mathrm{X}}_i - X_a)\} \qquad (1)$$

$$\chi^2 = \left\| S_y^{-\frac{1}{2}} K_i (\hat{X}_{i+1} - \hat{X}_i) - [\mathrm{Y} - \mathrm{R}(\hat{X}_i)] \right\|_2^2 + \left\| S_a^{-\frac{1}{2}} (\hat{\mathrm{X}}_{i+1} - X_a) \right\|_2^2, \qquad (2)$$

w~~W~~here $\hat{X}_{i+1}$ and $\hat{X}_i$ are current and previous state vectors with a priori vector, X_a and its covariance error matrix, $\mathrm{S_a}$. Y and R(X) are measured and simulated radiance vectors, with measurement error covariance matrix, $\mathrm{S_y}$. K is the weighting function matrix ($\frac{\mathrm{dR(x)}}{\mathrm{dx}}$), describing the sensitivity of the forward model to small perturbations of the state vector.

The ozone fitting window was determined to~~ward~~ maximize~~zing~~ the retrieval sensitivity to ozone and minimize~~ing~~ ~~that~~ it to measurement error: 289–307 nm and 326–339 nm for GOME, 270-309 nm and 312-330 nm for OMI, 289–307 nm and 325–340 nm for GOME-~~/~~2, and 302.5-340 nm for OMPS. For OMI, GOME and GOME-~~/~~2, partial ozone columns are typically retrieved in 24 layers from the surface to ~ 60 km. However, GEMS (300-500 nm) and OMPS (300-380 nm) do not cover much of the Hartley ozone absorption wavelengths and hence the reliable profile information of ozone is limited ~~at least~~to below ~ 40 km (Bak et al., 2013a).

Fig. 1 ~~presents~~ is a schematic diagram of the ozone profile algorithm. With the input of satellite measurements, the slit function is parameterized through cross-correlation between satellite irradiance and a high-resolution solar reference spectrum to be used for wavelength calibration and for high - resolution cross section convolution (Sun et al., 2017; Bak et al., 2017); a normalized Gaussian distribution is assumed to derive analytic slit functions for OMI. To remove the systematic errors between measured and calculated radiances, "soft-calibration" is applied to measured radiances and then the logarithms of sun-normalized radiances ~~is~~ are calculated as ~~a~~ measurement vectors (Liu et al., 2010a; Cai et al., 2012; Bak et al., 2017). ~~A m~~M~~M~~easurement covariance matrices~~x~~ ~~is~~ are constructed as ~~a~~ diagonal matrices~~x~~ with ~~each~~ components taken from the square of the measurement errors as measurement errors are assumed to be uncorrelated ~~between~~ among wavelengths~~; –~~. In the OMI algorithm, ~~for OMI the~~a noise floor ~~noise~~ of 0.4 % (UV1) and 0.2 % (UV2) is used because OMI measurement errors underestimate other kinds of random noise errors caused by stray light, dark current, geophysical pseudo-random noise errors due to sub-pixel variability, ~~and~~ motion when taking a measurement, forward model parameter error (random part), and other unknown errors into account (Huang et al., 2017). GEMS is expected to ~~hvae~~have similar retrieval sensitivity to tropospheric ozone, and have at least comparable radiometric/wavelength accuracy (4% including light source uncertainty/0.01 nm) as OMI. A priori ozone information is taken from the tropopause-based (TB) ozone profile climatology~~,~~ which was developed for improving ozone profile retrievals in the upper troposphere and lower stratosphere (Bak et al., 2013b). The Vector LInearized Discrete Ordinate Radiative Transfer (VLIDORT) model (Spurr, 2006; 2008) is ~~run~~ used to calculate ~~the~~ normalized radiances and weighting function matrices~~x~~ for the atmosphere, with Rayleigh scattering and trace-gas absorption and with Lambertian reflection for both surface and cloud (Liu et al., 2010a). The ozone algorithm iteratively estimates the best ozone profiles within the retrieval converges (typically 2-3 iterations), together with other geophysical and calibration parameters (e.g., cloud fraction, albedo, BrO, wavelength shift~~s~~, R~~r~~ing parameter, mean fitting scaling parameter) for a better fitting accuracy even though some of the additional fitting parameters can reduce the degrees of freedom for signal of ozone. We should note here that GEMS data processing is expected to be different ~~to~~from OMI mainly in two ways: 1) OMI uses a depolarizer to scramble the polarization of light. However, GEMS has polarization sensitivity (required to be less than 2%) and performs polarization correction using an RTM-based look-up table of atmospheric polarization state and pre-flight characterization of ~~polarization~~ instrument polarization sensitivity in the level 0 to 1b data processing. The GEMS polarization correction is less accurate and hence additional fitting process might be required in the level 2 data processing, especially for ozone profiles that ~~have more significant retrieval sensitivity~~are more sensitive to the polarization error compared to other trace-gases. 2) GEMS has a capability to perform diurnal observations and hence ~~the~~ diurnal meteorological input data are required to account for the temperature dependent Huggins band ozone absorption. Hence, the numerical weather prediction (NWP) model analysis data will be transferred to the GEMS S~~s~~cience D~~d~~ata P~~p~~rocessing C~~c~~enter (SDPC).

2.2 Ozonesonde measurements

Ozonesondes are small, lightweight, and compact balloon-born instruments capable of measuring profiles of ozone, pressure, temperature and humidity from the surface to balloon burst, usually near 35 km (4 hPa); ozone measurements are typically reported in ~~the~~ units of partial pressure (mPa) with ~~the~~ vertical resolution of ~ 100-150 m (WMO, 2014). Ozone soundings have been taken for more than 50 years, since the 1960s. The accuracy of ozonesonde measurements has been reported as 5-10 % with ~~the~~ precision of 3-5%, depending on the sensor type, manufacturer, solution concentrations, and operational procedure (Smit et al., 2007; Thompson et al., 2007; 2017; Witte et al., 2017; 2018). T~~he t~~hree types of instruments have been carried on balloons, i.e., the modified Brewer-Master (MB-M), the carbon iodine cell (CI), and the electrochemical concentration cell (ECC).~~,~~ ~~the carbon iodine cell (CI).~~ Each sounding is disposably operated and hence weekly launched for ~~the~~ long-term operation.

Fig. 2 displays the locations of 10 ozonesonde sites focused on this study within the ~~expected~~ GEMS domain borderinge~~ed~~ from 5°S (Indonesia) to 45°N (south of the Russian border) and from 75°E to 145°E. A summary of each ozonesonde site is presented in Table 1. Most of measurements are collected from the WOUDC network, except that Pohang soundings are provided from the Korea Meteorological Administration (KMA) and Kuala Lumpur and Hanoi measurements are from the Southern Hemisphere Additional OZonesondes (SHADOZ) network. In South Korea, ECC sondes have been launched every Wednesday since 1995 ~~only~~ at Pohang, without significant time gaps. There are three Japanese stations (Naha, Tsukuba, and Sapporo) where the CI-~~-~~type~~d~~ sensor was used ~~and~~ before switchinge~~ed~~ to the ECC-type~~d~~ sensor as of early 2009, and two Indian stations at New Delhi and Trivandrum using the ~~Modified~~ modified B-M (MB-M) sensor. The rest of stations (Hanoi, Hong Kong, Kuala Lumpur and Singapore) use~~s~~ only ECC. Most stations employ ~~an~~ ECC ~~ozone~~ sensors, but inhomogeneities in ECC ozonesondes are strongly ~~addressed with respect~~correlated to ~~the~~ preparation and correction procedures. There are two ECC sensor manufactures: the ~~;~~ Science Pump Corporation (Model type: SPC-6A) and the Environmental Science Corporation (Model type: EN-SCI-Z/1Z/2Z). Since 2011 EN-SCI has been taken over by Droplet Measurement Technologies (DMT) Inc. The Standard Sensing Solution has been recommended as SST1.0 (1.0 % KI, full buffer) and SST 0.5 (2.0 % KI, no buffer) for the SPC and EN-SCI sondes, respectively by the ASOPOS (Assessment for Standards on Operation Procedures for Ozone Sondes) (Smit et al., 2012). Among ECC stations, Pohang, Hong Kong, and the Japanese stations have applied the standard sensing solution to all ECC ~~observation~~ sensors manufactured by ~~with its one manufacture~~one company. In Singapore, the ozonesonde manufacture was changed in late 2015 from EN-SCI to SPC, while SST 0.5 was switched to SST 1.0

as of 2018. Two SHADOZ stations (Kuala Lumpur~~Kuala lump~~, Hanoi) have applied the standard sensing solution just since 2015. Hanoi changed sensing solution 4 times with two different ozonesonde manufactures; Kuala Lumpur ~~Kula lump~~ operated only with SPC 6A-SST 1.0 combination until 2014, but with four different radiosonde manufactures. Therefore the~~se~~ SHADOZ datasets were ~~reprocessed~~ homogenized (~~in~~ Witte et al., ~~(~~2017) through the application of transfer functions between sensors and solution types~~s to be homogenized~~. The post-processing could be applied by data users to some WOUDC datasets given a correction factor, which is the ratio of integrated ozonesonde column (appended with an estimated residual ozone column above burst altitude) and total ozone measurements from co-located ground-based and/or overpassing satellite instruments. The above-burst column ozone is estimated with a constant ozone mixing ratio (CMR) assumption above the burst altitude (~~e.g.,~~ Japanese sites, ~~) (~~Morris et al., 2013) or satellite derived stratospheric ozone climatology (~~e.g.,~~ Indian sites, ~~) (~~Rohtash et al., 2016). No post-processing is ~~given to~~done for Pohang, Hong Kong, and Singapore. Most stations made weekly or bi-weekly regular observations, except for Indian stations with irregular periods of 0-4 per month and for Singapore with monthly observations.

In Fig. 3 the seasonal means and standard deviations of ozonesonde measurements are presented to ~~see~~show the stability and characteristics of ozonesonde measurements at each site. Instabilities of measurements are ~~apparently~~ observed from New Delhi ozonesondes. High surface ozone concentrations at Trivandrum in summer ~~is~~are believed to be caused by measurement errors because low levels of ~~the~~ pollutants ~~has~~have been reported at this site under these geolocation and meteorological effects (Lal et al. 2000). Besides Trivandrum, Naha could be regarded as a background site~~s~~ according to low surface ozone (Fig. 3) and ~~its~~ precursor concentrations (Fig. 2) compared to neighboring stations, and previous studies (Oltmans et al., 2004; Liu et al., 2002). In the lower troposphere, high ozone concentrations are captured at Pohang, Tsukuba, and Sapporo in the summer due to enhanced photochemical production of ozone in daytime, whereas tropical sites, Naha, Hanoi, and Hong Kong show ~~the~~ ozone enhancements in spring, mainly due to ~~the~~ biomass burning in Southeast Asia, with low ozone concentrations in summer due to the Asian monsoon and in winter due to ~~the~~ tropical air intrusion (Liu et al., 2002; Ogino et al., 2013). Singapore and Kuala Lumpur are supposed to be severely polluted areas, but ozone pollution is not clearly captured over the seasons. ~~It~~This ~~could~~might be explained by the morning observation time at these two stations ~~carried on in the morning~~. In addition, instabilities of Singapore measurements are noticeable, ~~such as~~including abnormally large variability and very low ozone concentration in the stratosphere.

The effect of stratospheric intrusions on the ozone profile shape is dominant at ~~the~~ mid-latitudes (Pohang, Tsukuba, and Sapporo) during the spring and winter when the ozone-pause goes down to 300 hPa, with ~~the~~ larger ozone variabilities in the lower stratosphere and upper troposphere, whereas the ozonepause is ~~placed~~ around 100 hPa with much less variability~~ies~~ of ozone ~~over the~~~~with pressure at~~in other seasons.

2.3. Comparison Methodology

The GEMS ozone profile algorithm is applied to OMI BUV measurements ~~in~~ for 300-330 nm to simulate GEMS ozone profile retrievals at coincident locations listed in Table 1. The coincidence criteria between satellite and ozonesondes are: ±1.0° in both longitude and latitude and ±12 hours in time, and then the closest pixel is selected. The Aura satellite carrying OMI crosses the equator always at ~ 1:45 pm ~~LT~~ Local T~~t~~ime (LT), ~~and thereby~~thus OMI measurements are ~~closely~~ collocated within 3 hours to ozonesonde soundings ~~measured~~ in the afternoon (1-3 pm ~~LS~~). Weekly-~~–~~based sonde measurements provide 48 ozone profiles at maximum for a year; the number of collocations is on average 40 from 2004 October to 2008, but reduced to ~ 20 recently due to the screened OMI measurements affected by the "row anomaly" which ~~is~~ was initially detected at two rows in 2007, and seriously spread to other rows with time since January 2009 (Schenkeveld et al., 2017). ~~As f~~From July 2011 the row anomaly ~~effect slowly~~ extends up to ~ 50 % of all rows. Correspondingly, the average collocation distance increases from 57.5 km to 66.6 km before and after the occurrence of the row anomaly. The impact of spatiotemporal variability on the comparison will be much reduced for GEMS due to its higher spatiotemporal resolution (7 km ×~~x~~ 8 km @ Seoul, hourly) against OMI (48 km ~~x~~× 13 km @ nadir in UV1, daily).

To increase the validation accuracy, ~~the~~ data screening is implemented ~~to~~ for both ozonesonde observations and satellite retrievals according to Huang et al (2017). For ozonesonde observations, we screen ozonesondes with balloon-bursting ~~altitudes~~ pressures exceeding 200 hPa, gaps greater than 3 km, abnormally high concentration in the troposphere (> 80 DU), and low concentration in the stratosphere (<100 DU). Among WOUDC sites, the Japanese and Indian datasets include a correction factor which is derived to make ~~a~~ better agreement between integrated ozonesonde columns and correlated reference total ozone measurements as mentioned in Section 2.2; In Fig. ~~3~~4, Japanese ozonesondes are compared against GEMS simulations when a correction factor is applied or not to each CI and ECC measurement~~s~~, respectively. Morris et al. (2013) recommended ~~to~~ restricting the application of this correction factor to the stratospheric portion of the CI ozonesonde profiles due to errors in the above-burst column ozone. Our comparison results illustrate that applying the correction factor reduces the vertical fluctuation of mean biases~~ed~~ in ozone profile differences with insignificant impact on their standard deviations. Therefore we decide to apply this correction factor to the sonde profiles if this factor ranges from 0.85 to 1.15. Because of a lack of retrieval sensitivity to ozone below clouds and lower tropospheric ozone under extreme viewing condition, ~~satellite retrievals~~GEMS simulations are limited to cloud fraction less than 0.5, SZAs less than 60°, and fitting RMS (i.e., root mean square of fitting residuals relative to measurement errors) less than 3.

Due to the different units of ozone amount between satellites and ozonesondes, we convert ozonesonde-measured partial pressure ozone values (mPa) to partial column ozone (DU) at the 24 retrieval grids heights of the satellite for the altitude range from surface to the balloon-bursting altitudes. Ozonesonde measurements are obtained at a rate of a few seconds and then typically averaged into altitude increments of 100 meters, whereas retrieved ozone profiles from nadir BUV satellite measurements have much coarser vertical resolution of 10-14 km in the troposphere and 7-11 km in the stratosphere,~~troposphere~~ based on OMI retrievals. Consequently, satellite observations capture~~s~~ only the smoothed structures of ozonesonde soundings, especially ~~in~~ near the tropopause, where a sharp vertical transition of ozone within 1 km is observed, and in the boundary layer due to the insufficient penetration of photons. Satellite retrievals unavoidably have an error compound due to its limited vertical resolution, ~~which is named~~ called "smoothing error" in ~~the~~ OE~~-~~based retrievals (Rodgers, 2000). It could be useful to eliminate the effect of smoothing errors on differences between satellites and sondes to better characterize other error sources in ~~the~~ comparisons (Liu et al., 2010a). For this reason, satellite data have been compared to ~~smoothed~~ ozonesonde measurements smoothed ~~in~~to the satellite vertical resolution~~-~~, together with original sonde soundings (Liu et al., 2010b; Bak et al., 2013b; Huang et al., 2017). The smoothing approach is:~~ following as~~

$$\hat{x}_{sonde} = A \cdot x_{sonde} + \sout{x_a}(1 - A)x_a \quad (3)$$

x_{sonde} : High-resolution ozonesonde profile

$\hat{x}_{sonde}$: Convolved ozonesonde profile into satellite vertical resolution

A : Satellite averaging kernel

x_a : A priori ozone profile

In order to define tropospheric columns, both satellite retrievals and ozonesonde measurements are vertically integrated from the surface to the tropopause taken from daily National Centers for Environmental Prediction (NCEP) final (FNL) Operational Global analysis data (http://rda.ucar.edu/datasets/ds083.2/). To account for the effect of surface height differences on comparison, ozone amounts ~~of~~ from satellite data below the surface heights of ozonesondes ~~is~~ are added to tropospheric columns of ozonesonde measurements and vice versa.

3. Results and Discussions

3.1 Comparison at individual stations

Witte et al. (2018) recently compared seven SHADOZ station ozonesonde records, including Hanoi and Kuala Lumpur in the GEMS domain, with total ozone and stratospheric ozone profiles measured by space-borne nadir and limb viewing instruments, respectively. In this comparison, the Hanoi station shows comparable or better agreement with the satellite datasets when compared to other sites. Morris et al. (2013) and Rohtash et al. (2016) thoroughly evaluated ozonesonde datasets over Japanese and Indian sites, respectively, but they did not address their measurement accuracy with respect to those at other stations. Validation of GOME TOC by Liu et al. (2006) showed relatively larger biases at Japanese CI stations and validation of OMI TOC by Huang et al. (2017) showed both larger biases and standard deviations at the India MB-M sites. In South Korea, regular ozonesonde measurements are taken only from Pohang, but these measurements have been insufficiently evaluated; only the stratospheric parts of these measurements were quantitatively assessed against satellite solar occultation measurements by Halogen Occultation Experiment (HALOE) from 1995 to 2004 in Hwang et al. (2006), but only 26 pairs were compared despite ~~its~~ the coarse coincident criteria (48 hours in time, ±4.5° in latitude, ±9 ° in longitude). Therefore, it is important to perform ~~the~~ quality assessment of ozonesonde measurements to identify ~~the~~ a reliable reference dataset for GEMS ozone profile validation

For this purpose, we illustrate tropospheric ozone columns (TOC) as a function of time ~~and~~ for individual stations listed in Table 1, measured with three different types of ozonesonde instruments and retrieved with GEMS simulations (Fig. ~~4~~5), respectively. The goal of this comparison is to identify any abnormal deviation of ozonesonde measurements relative to satellite retrievals, so we exclude the impact of the different vertical resolutions between instruments and satellite retrievals on this comparison by convolving ozonesonde data with satellite averaging kernels. At mid-latitude sites (Pohang, Sapporo, and Tsukuba) both ozonesonde and ~~satellite~~ simulated retrievals show the distinct seasonal TOC variations with values ranging from~~the amplitude of~~ ~ 35 to ~~~~~ 40 DU. Extratropical sites (Naha, Hong Kong, and Hanoi) show less seasonal variations, ~~of~~ 30 to 50 DU, whereas fairly constant concentrations are observed at Kuala Lumpur and Singapore in the tropics. Both ozonesonde observations and ~~satellite~~ simulated retrievals illustrate similar seasonal variabilities at these locations.

At New Delhi and Trivandrum, on the other hand, MB-M ozonesonde measurements abnormally deviate from 10 DU to 50 DU compared to the corresponding satellite retrievals and latitudinally neighboring ozonesonde measurements at stations in similar latitudes regimes.

In Fig. 5 6 time dependent errors in differences of TOC between ozonesonde and satellite simulated GEMS retrievals are evaluated with the corresponding comparison statistics in Table 2. Satellite retrievalsSimulated retrievals show strong correlation of ~ 0.8 or much larger with ozonesonde measurements at Pohang, Hong Kong, and three stations from JapanJapan stations, and with less correlation of ~ 0.5 at other SHADOZ stations in the tropics. However, Indian stations show poor correlation of 0.24. Mean biases and its standard deviations are much smaller at stations where a strong correlation is observed; they are ~1 DU ±~ 4DU at most ECC stations, but deviated to ~ 4 DU ±~ 10 DU at MB-M stations. In conclusion, we should exclude ozonesonde observations measured by MB-M to remove irregularities in a reference dataset for validating both GEMS simulated retrievals in this study and GEMS actual retrievals in future study. Moreover, time series of ozonesonde and satellite observationssimulated retrievals show a significant transition at three Japanese stations as of late 2008 and early 2009 when the ozonesonde instruments was were switched from CI to ECC. This transition could be affected by space-born instrument degradation, but the impact of balloon-born instrument change on them is predominant based on a less time-dependent degradation pattern at latitudinally neighboring stations during this period. CI ozonesondes noticeably underestimates atmospheric ozone by 2-3 DU compared to ECC and thereby GEMS TOC biases relative to CI measurements, are are estimated as - 2 to - 5 DU, but these biases are reduced to < 1.5 DU when compared with ECC. Therefore, we decide to exclude these CI ozonesonde observations for evaluating GEMS simulated retrievals. Compared to other ECC stations, Hanoi Sstation often changed sensing solution concentrations and pH buffers (Table 1), which might and hence might cause the irregularities due to remaining errors even though transfer functions were applied to ozonesonde measurements to account for errors due to the different sensing solution (Witte et al., 2017). This fact might affect the relatively worse performance compared to latitudually adjacenta neighboring station, Hong Kong, where the 1.0 % KI buffered sensing solution (SST 1.0) to ECC/SPC sensors have been consistently applied.

Fig. 6 7 compares differences of ozone profiles between ECC ozonesondes and GEMS simulated retrievals at each station. Among ECC ozonesondes, Singapore's ozonesondes are in the worst agreement with satellite retrievalsGEMS simulations in both terms of mean biases and standard deviations, which could be explained by the discrepancy of in collocation time. Sonde observations at Japan, Pohang, Hong Kong, and Hanoi Sstations, where balloons were launched in afternoon (~ 12-15 LSTLT), are collocated within ~ 1-2 hours to of OMI that passes the equator at 1301:45 pm LST and

~~then reaches the pole within 25 min~~, whereas the time discrepancy increases to 7 hours at Singapore, where ozonesondes are launched in the early morning. Photochemical ozone concentrations are typically denser in the afternoon than in the morning and hence ozonesonde measurements at Singapore are negatively biased relative to afternoon satellite measurements. For the reason mentioned above, the discrepancy in the observation time could also ~~impact on~~affect this comparison at Kuala Lumpur~~Kuala Lump~~, where sondes were mostly launched in the late morning, 2-3 hours prior to the OMI passing time and thereby ozonesonde measurements tend to be negatively biased. These indicate that diurnal variations of ~~the~~ tropospheric ozone are visible in ozonesonde~~oznesonde~~ measurements, emphasizing ~~on~~ the utility of hourly geostationary ozone measurements. The comparison results could be characterized with latitudes. In the mid-latitudes (Pohang, Tsukuba, and Sapporo), noticeable disagreements are commonly ~~addressed~~ seen in the tropopause region where mean biases/standard deviations are ~10 %/~15% larger than those in the lower troposphere. In the extra-tropics (Hong Kong, Naha), consistent differences of ~~–~~a few ~~%~~ percent are ~~shown~~ seen over the entire altitude range with standard deviations of 15 % or less below the tropopause (~ 15 km). Hanoi and Kuala Lumpur show significantly larger biases/standard deviations compared to other ECC stations. At Hanoi inconsistencies of solution concentrations and pH buffers might influence ~~on~~ this instability. At Kuala Lumpur the inconsistencies of observation times might be one of the reasons, considering its standard deviations of ~100 min, but mostly less than 30 min at other stations. Therefore, we ~~strictly~~ screen out Singapore, Kuala Lumpur, and Hanoi, together with all M-BM measurements at Indian stations and CI measurements at Japanese stations to improve the validation accuracy of GEMS simulated retrievals in next section. Thus~~Eventually~~, stations~~,~~ where the standard procedures for preparing and operating ECC sondes are consistently maintained, are ~~accepted~~ adopted as an optimal reference ~~in~~ for this work.

43.2 Evaluation of GEMS simulated ozone profile retrievals

The GEMS simulated retrievals are assessed against ECC ozonesonde soundings at five stations (Hong Kong, Pohang, Tsukuba, Sapporo, and Naha) identified as a good reference in the previous section. The comparison statistics include mean bias and standard deviation in the absolute/relative differences, correlation coefficients, ~~the~~ linear regression results (slope (a), intercept (b), error); the error of the linear regression is defined as $\frac{1}{n}\sqrt{\sum_i^n \left(y_{GEMS} - y_{fit}\right)^2}$, $y_{fit} = a \cdot y_{sonde} + b$. In Fig. ~~7~~8, GEMS simulated retrievals are plotted as ~~a~~ functions of ozonesondes with and without the vertical resolution smoothing, respectively, for the stratospheric and tropospheric columns. GEMS simulations underestimate the tropospheric ozone by ~ 2.27 ± 5.94 DU and overestimate the stratospheric ozone by ~ 9.35 ± 8.07 DU relative to high-resolution ozonesonde observations. This comparison demonstrates ~~a~~ good correlation coefficients of 0.84 and 0.99 for troposphere and stratosphere, respectively. This agreement is degraded if the rejected ECC sondes (Kuala Lumpur, Hanoi, and Singapore) are included; for example, the slope decreases from 0.68 to 0.64 while the RMSE increases 6.35 and 6.76 DU for TOC comparison. Smoothing ozonesonde soundings ~~into~~ to GEMS vertical resolution improves the comparison results, especially for the tropospheric ozone columns; standard deviations are reduced by ~ 5 % with mean biases of less than 1 DU. Similar assessments are performed for OMI standard ozone profiles based on the KNMI OE algorithm (Kroon et al., 2011) hereafter referred to as OMO3PR (KNMI) in Fig. ~~8~~ 9 and the research product based on the SAO algorithm (Liu et al., 2010) hereafter referred to as OMPROFOZ (SAO) in Fig. ~~9~~10, respectively. It implies that GEMS gives ~~the~~ good information on Stratospheric Ozone Columns (SOCs)~~sSOCs~~ comparable to both the OMI KNMI and SAO products in spite of ~~excluding~~ insufficient information on ~~most of~~ Hartley ozone ~~band~~ absorption in GEMS~~in GEMS retrievals~~. Furthermore, a better agreement of GEMS TOCs with ozonesonde is found than with the others due to different implementation details. As mentioned in 2.1., the GEMS algorithm is developed based on the heritages of the SAO ozone profile algorithm with several modifications. The~~re~~ ~~are~~ two main modifications are: (1) a priori ozone climatology was replaced with a tropopause-based ozone profile climatology to better represent the ozone variability in the tropopause (2)~~.~~ i~~I~~rradiance spectra used to normalize radiance spectra and characterize instrument line shapes are prepared by taking 31-day moving average instead of climatological average to take into account for time-dependent instrument degradation~~s~~. These modifications reduce somewhat the spread~~s~~ in deviations of satellite retrievals from sondes, especi~~e~~ally in ~~TCO~~ TOC comparison. KNMI retrievals systematically overestimate the tropospheric ozone by ~ 6 DU (Fig. ~~9~~10.c), which corresponds to the positive biases of 2-4 % in the integrated total columns of KNMI profiles relative to Brewer observations (Bak et al., 2015). As mentioned in Bak et al. (2015), the systematic biases in ozone retrievals are less visible in SAO-based retrievals (simulated GEMS ~~–~~data~~simulation~~, OMPROFOZ), as systematic components of measured spectra are taken into account for using an empirical correction called "soft calibration".

4. Summary

We simulate GEMS ozone profile retrievals from OMI BUV radiances in the range ~~of~~ 300-330 nm using the ~~optimal estimation~~OE-based fitting during the period ~~of~~ 2005-2015 to ensure the performance of the algorithm against coincident ozonesonde observations. There are 10 ozonesonde sites over the GEMS domain from WOUDC, SHADOZ and KMA archives. This paper gives an overview of these ozonesonde observation systems to address inhomogeneities in preparation, operation, and correction procedures which cause discontinuities in individual long-term records or ~~in~~ among ~~adjoint~~ stations. Comparisons between simulated GEMS TOCs ~~retrievals~~ and ozonesondes illustrate a noticeable dependence on the instrument type. Indian ozonesonde soundings measured by MB-M show severe deviations in seasonal time series of TOC compared to coherent GEMS simulations and ~~neighboring~~ ozonesonde observations measured in similar latitude regime~~s~~. At Japanese stations, CI ozonesondes underestimate ECC ozonesondes by 2 DU or more and a better agreement with GEMS simulations is found when ECC measurements are compared. Therefore, only ECC ozonesonde measurements are ~~first~~ selected as a reference, in order to ensure a consistent, homogeneous dataset. Furthermore, ECC measurements at Singapore, Kuala Lumpur, and Hanoi are excluded. At Singapore and Kuala Lumpur, observations were performed in the morning and thereby are inconsistent with GEMS retrievals simulated at the OMI overpass time in the afternoon. In addition, the observation time for Kuala Lumpur is inconsistent itself compared to other stations; its standard deviation is ~ 100 min, but for other ECC stations it is less than 30 min. At Hanoi the combinations of sensing solution concentrations and pH buffers changed 4 times during the period of 2005 through 2015. Therefore, GEMS and ~~ozonsonde~~ozonesonde comparisons show larger biases/standard deviations at these stations. Pohang station is unique in South Korea where ECC ozonesondes have been regularly and consistently launched without a gap since 1995; the standard 1% KI full buffered sensing solution has been consistently applied to ozone sensors manufactured by SPC (6A model). Evaluation of Pohang ozonesondes against GEMS simulations demonstrates its high level reliability, which is comparable to ~~latitudually adjacent~~neighboring Japanese ECC measurements at Tsukuba and Sapporo. Reasonable agreement with GEMS simulated retrievals is ~~s~~ similarly shown at ~~Latitudually~~ adjacent Naha and Hong Kong stations. Finally, we establish that the comparison statistics of GEMS simulated retrievals and optimal reference dataset is -2.27 (4.92) $\pm$ 5.94 (14.86) DU (%) with R = 0.84 for the tropospheric columns and 9.35 (5.09) $\pm$ 8.07 (4.60) DU (%) with R=0.99 for the stratospheric columns. This estimated accuracy and precision is comparable to OMI products for the stratospheric ozone column and even better for the tropospheric ozone column due to improved algorithm implementation~~s~~. Our future study aims to achieve this quality level from actual GEMS ozone profile product.

Author contributions. JB and KHB designed the research; JHK and JK provided oversight and guidance; JB conducted the research and wrote the paper; XL and KC contributed to the analysis and writing.

Competing interests. The authors declare that they have no conflict of interest.

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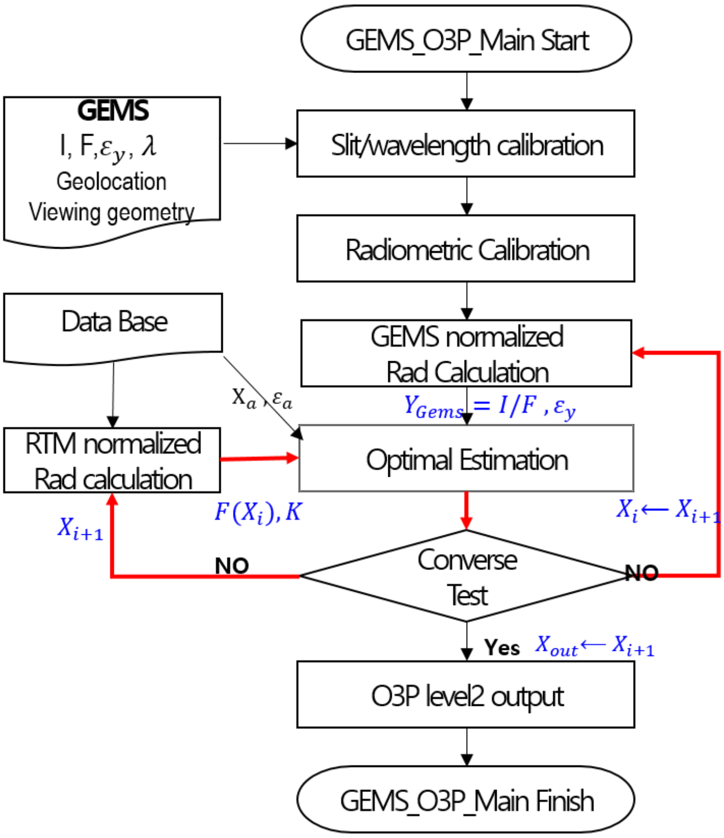

Figure 1. Flow c~~C~~hart of the GEMS ozone profile retrieval algorithm.

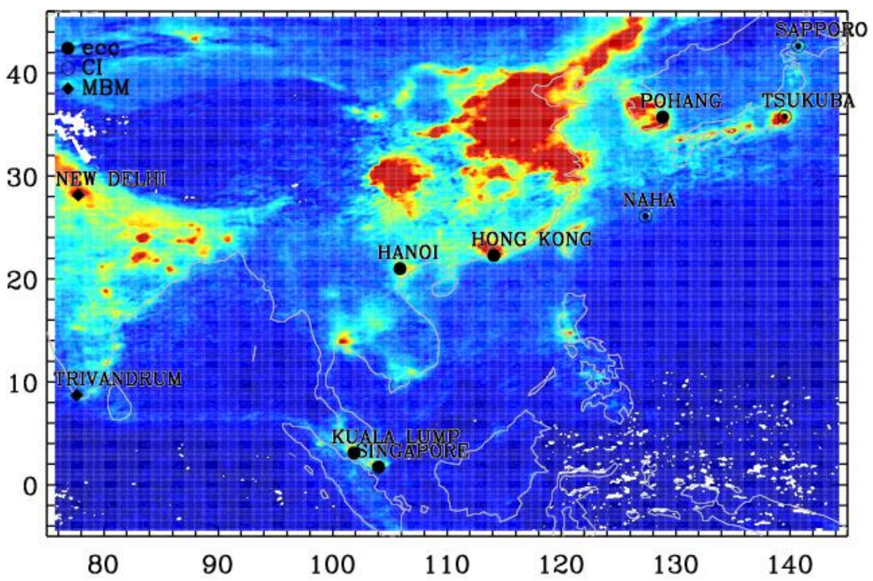

Figure 2. Geographic locations of the ozonesonde stations available since 2005 over the GEMS observation domain. Each symbpol represents a different typed sensors; the modified Brewer-Mast (MBM), the carbon iodine cell (CI), and the electrochemical concentration cell (ECC). The background map illustrates the OMI NO_2 monthly mean in June 2015.

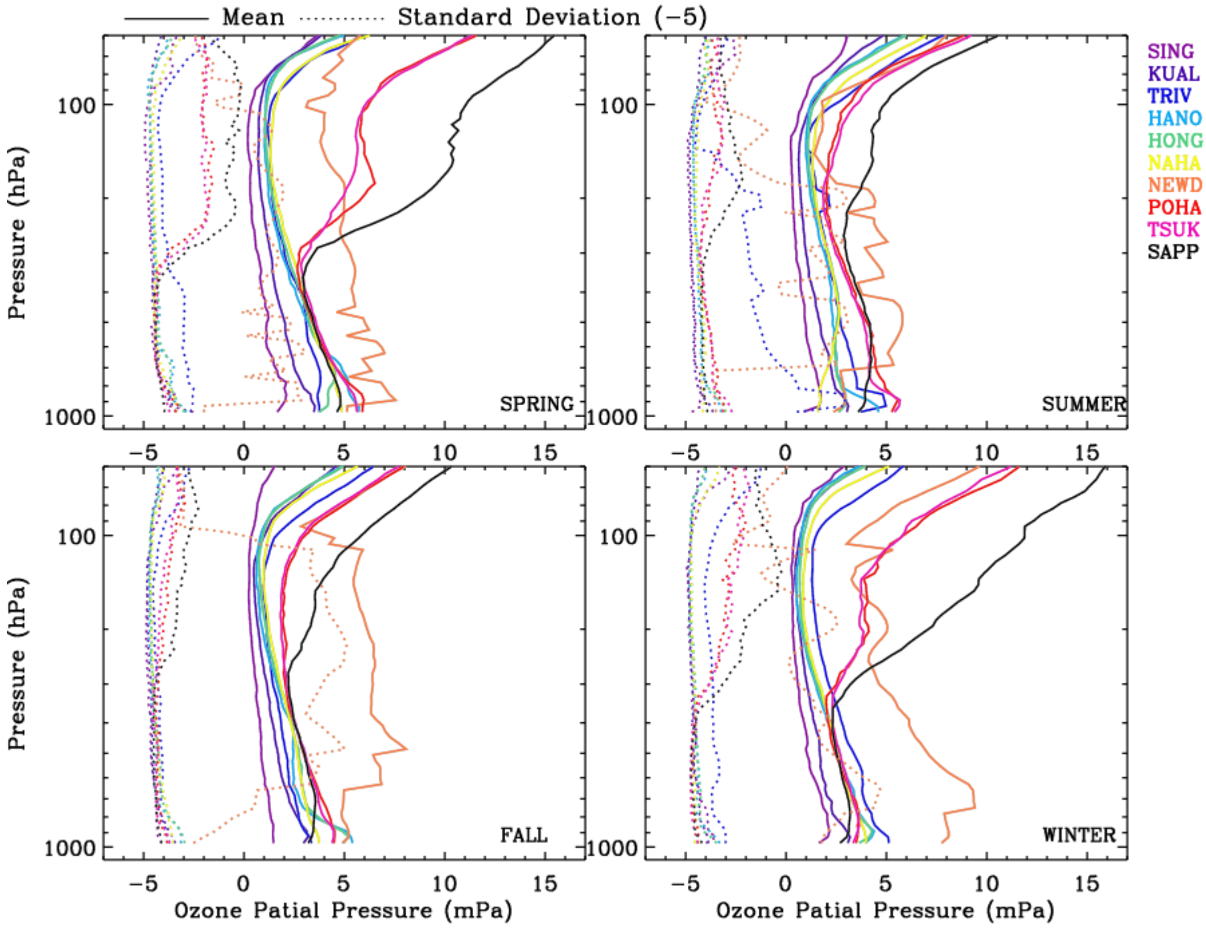

Figure 3. Seasonal mean (solid) and standard deviation (dashed) profiles of ozonesonde soundings from 2005 to 2015 at the 10 sites listed in Table 1. 5 mPa is subtracted ~~to~~from standard deviations to fit ~~in~~ the ~~given~~ x-axis.

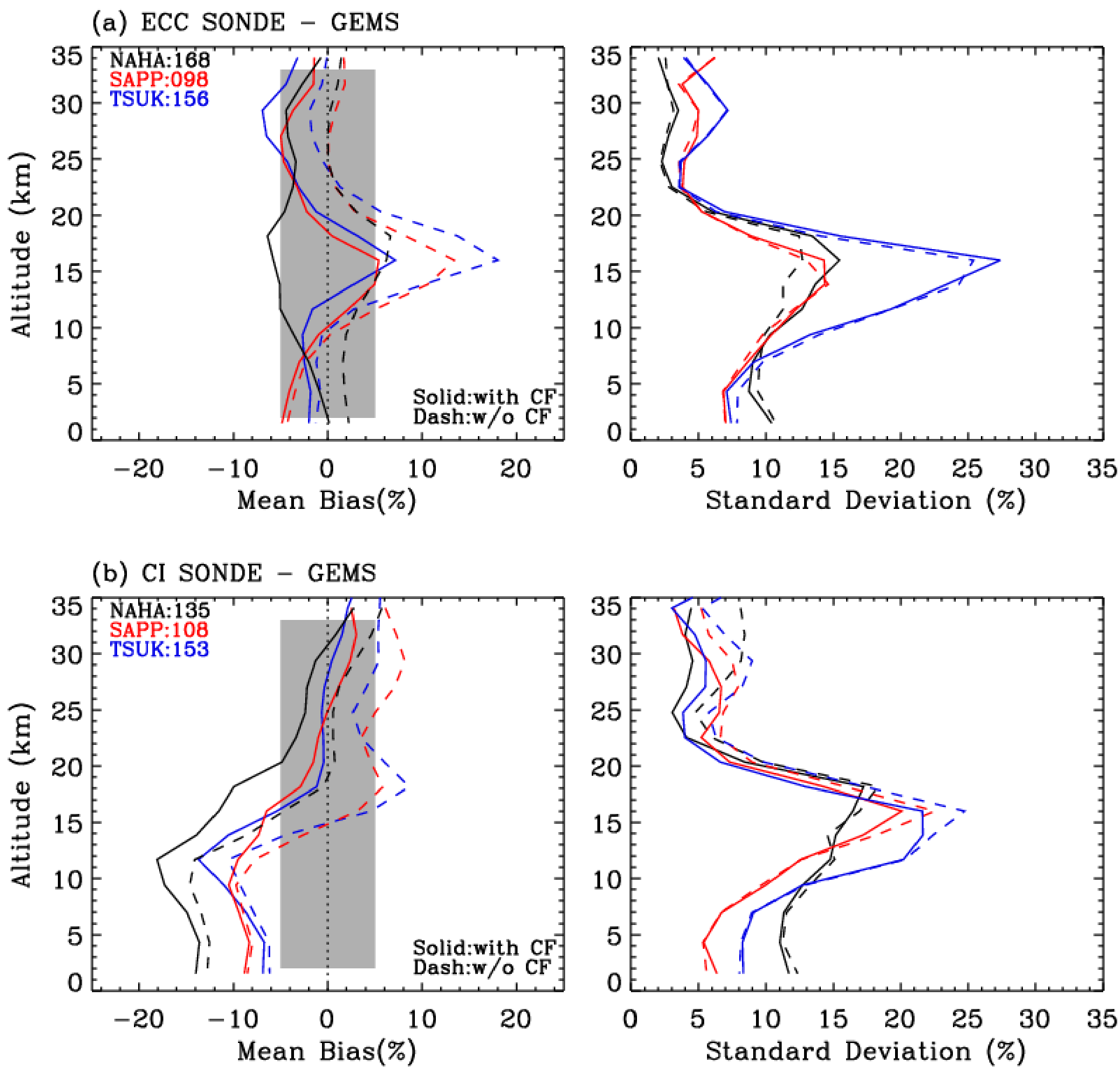

Figure 34. Effects of applying a correction factor (CF) to (a) ECC and (b) CI ozonesonde measurements, respectively, on comparisons with simulated GEMS ozone profile retrievals. Solid and dDashed lines represent the comparisons with and without applying a CF, respectively, at each Japanese station. The number of data point is included in the legends.

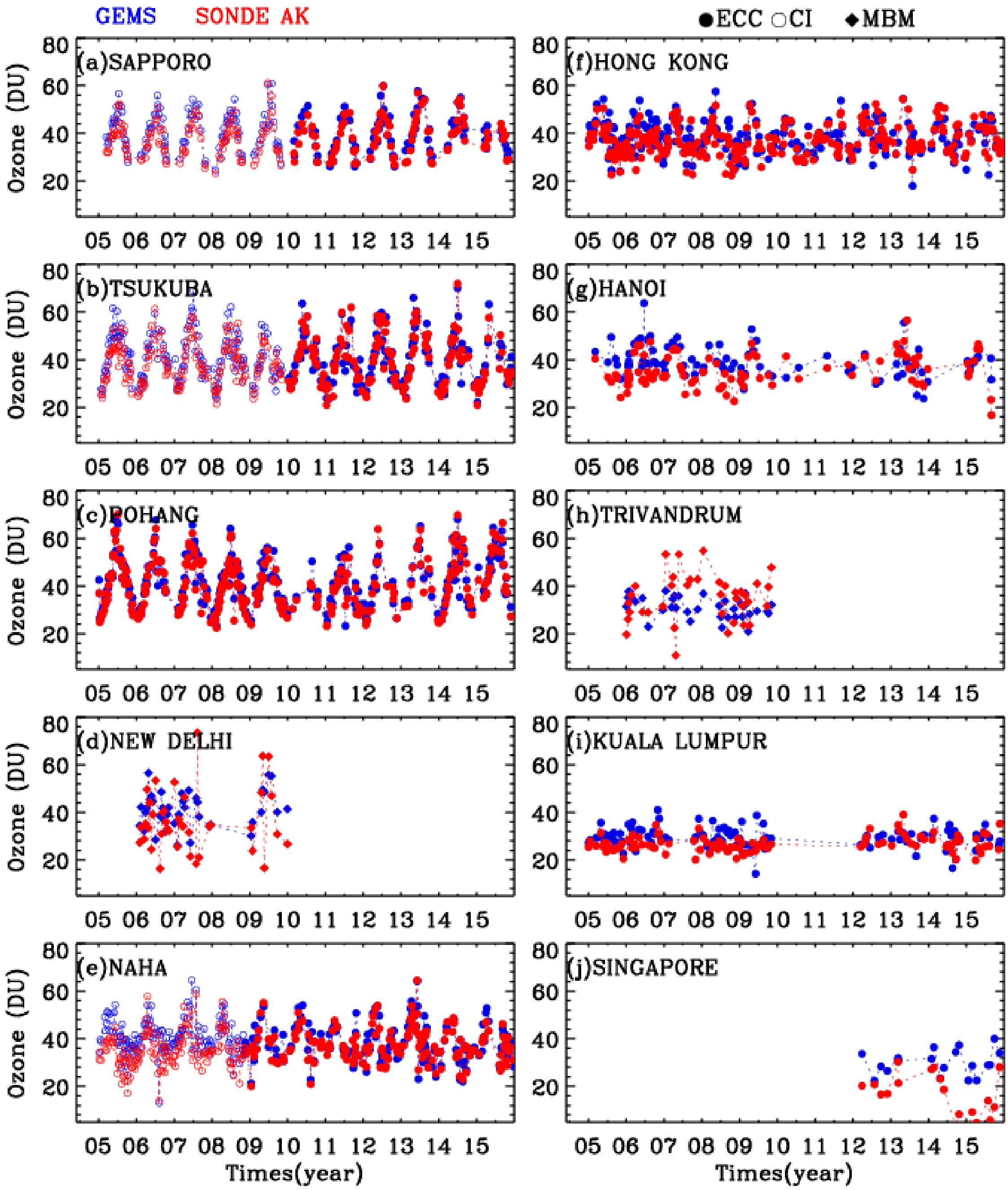

Figure 45. Time series of tropospheric ozone columns (DU) of GEMS simulated ozone profile retrievals (blue) and ozonesonde measurements convolved with GEMS averaging kernels (red) from 2005 to 2015 at 10 stations listed in Table 1.

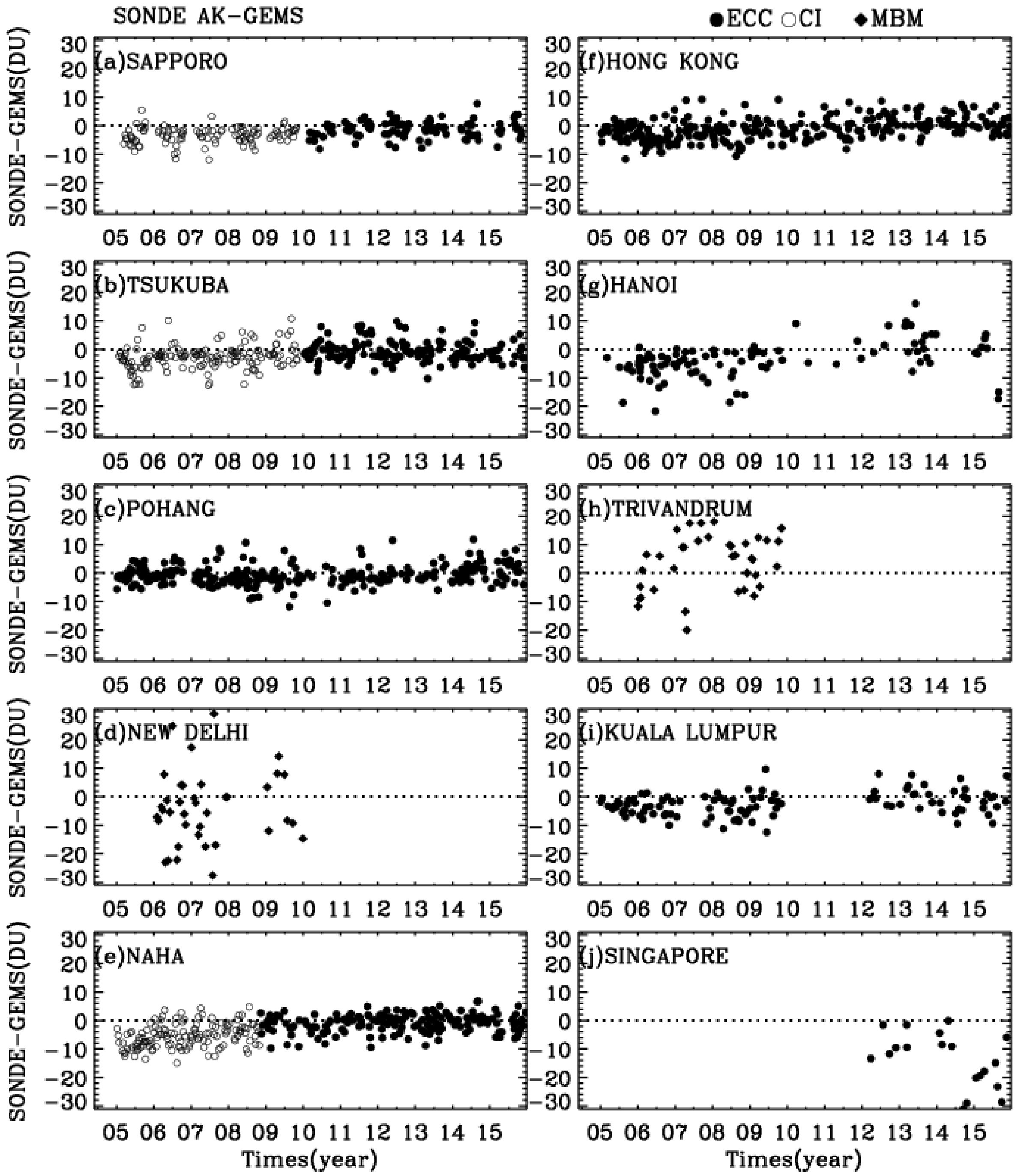

Figure ~~5~~6. Same as Figure 45, but for absolute differences of tropospheric ozone columns (DU) between ozonesonde measurements and GEMS simulated retrievals.

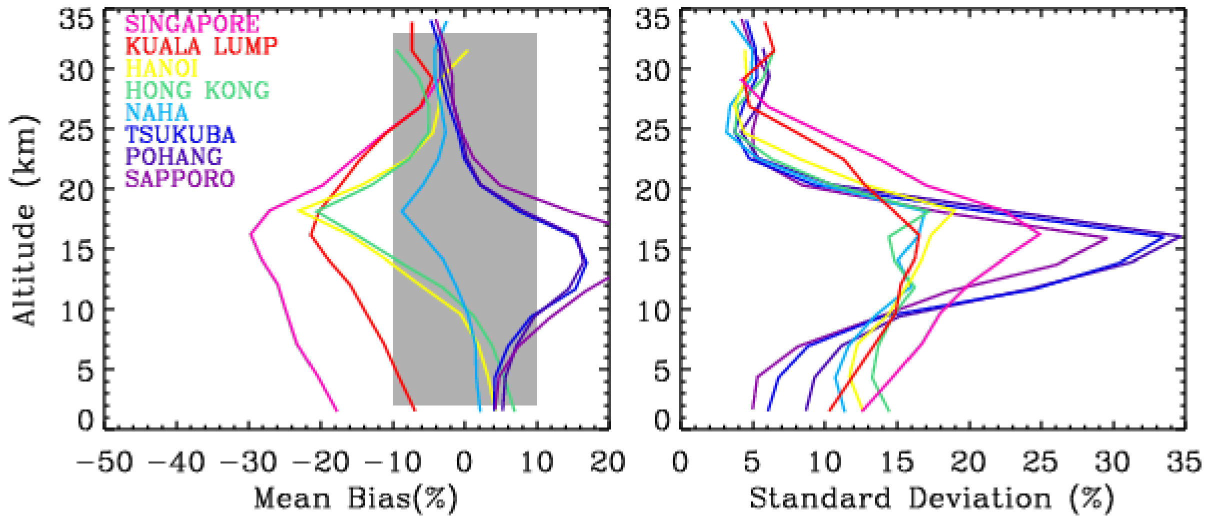

Figure ~~6~~7. Mean biases and 1σ~~σ~~ standard deviations of the differences between ozonesonde convolved with GEMS averaging kernels and GEMS simulated ozone retrievals as a function of GEMS layers, at individual ECC ozonesonde stations. The relative difference is defined as ~~2~~ (SONDE AK – GEMS) ~~X~~×100 %/ (a~~A~~ priori).

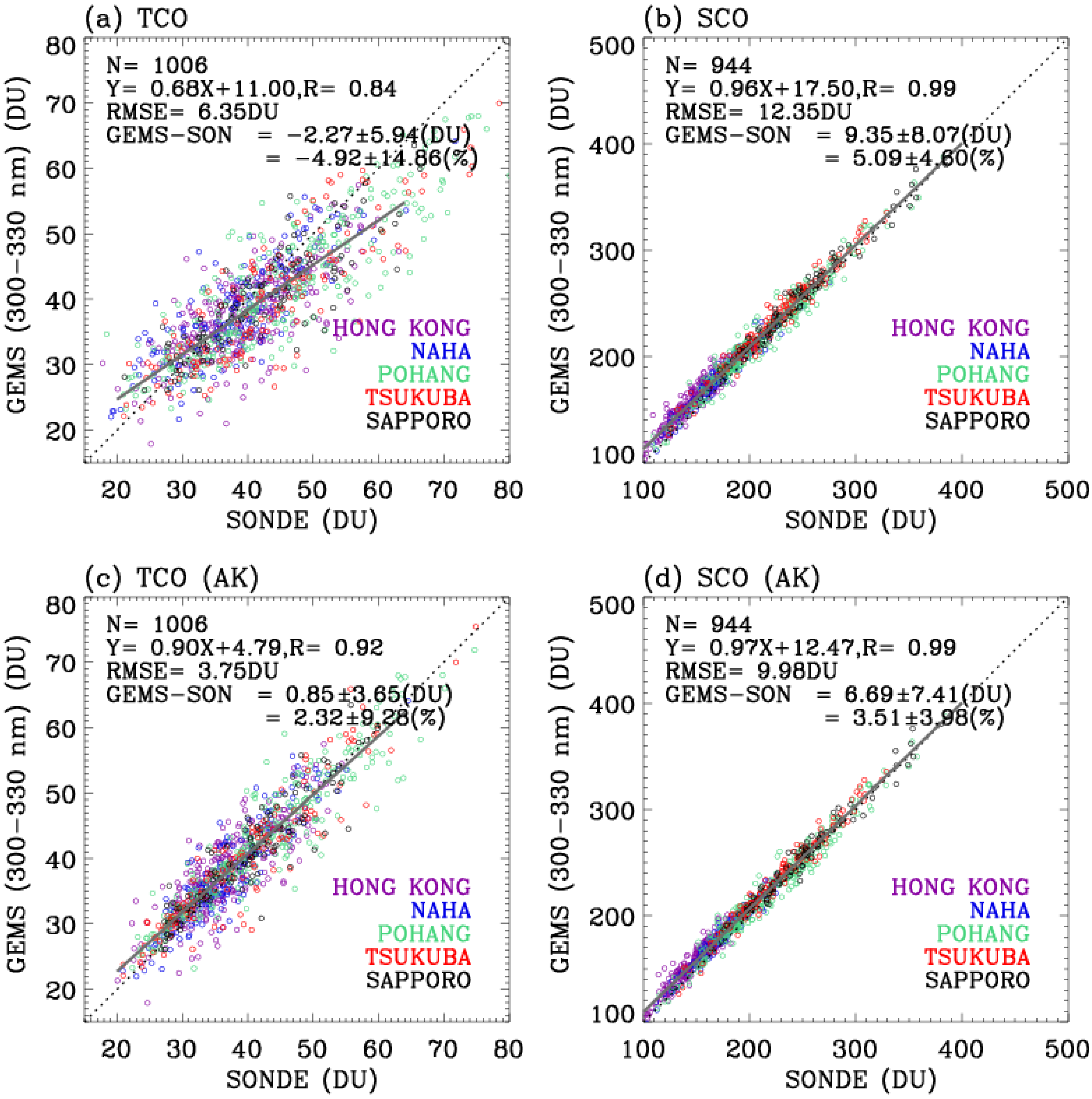

Figure 78. Upper: Scatter plots of GEMS vs. ozonesonde for tropospheric and stratospheric ozone columns, respectively. The lLower panels is are the same as the uUpper ones, except that ozonesonde measurements are convolved with GEMS averaging kernels. A linear fit between them is shown in red, with the 1:1 lines (dotted lines). The legends show the number of data points (N), the slope and intercept of a linear regression, and correlation coefficient (r), with mean biases and 1σ standard deviations for absolute (DU) and relative differences (%), respectively. Note that we use 5 stations identified as a good reference among 10 stations listed in Table 1 in this comparison.

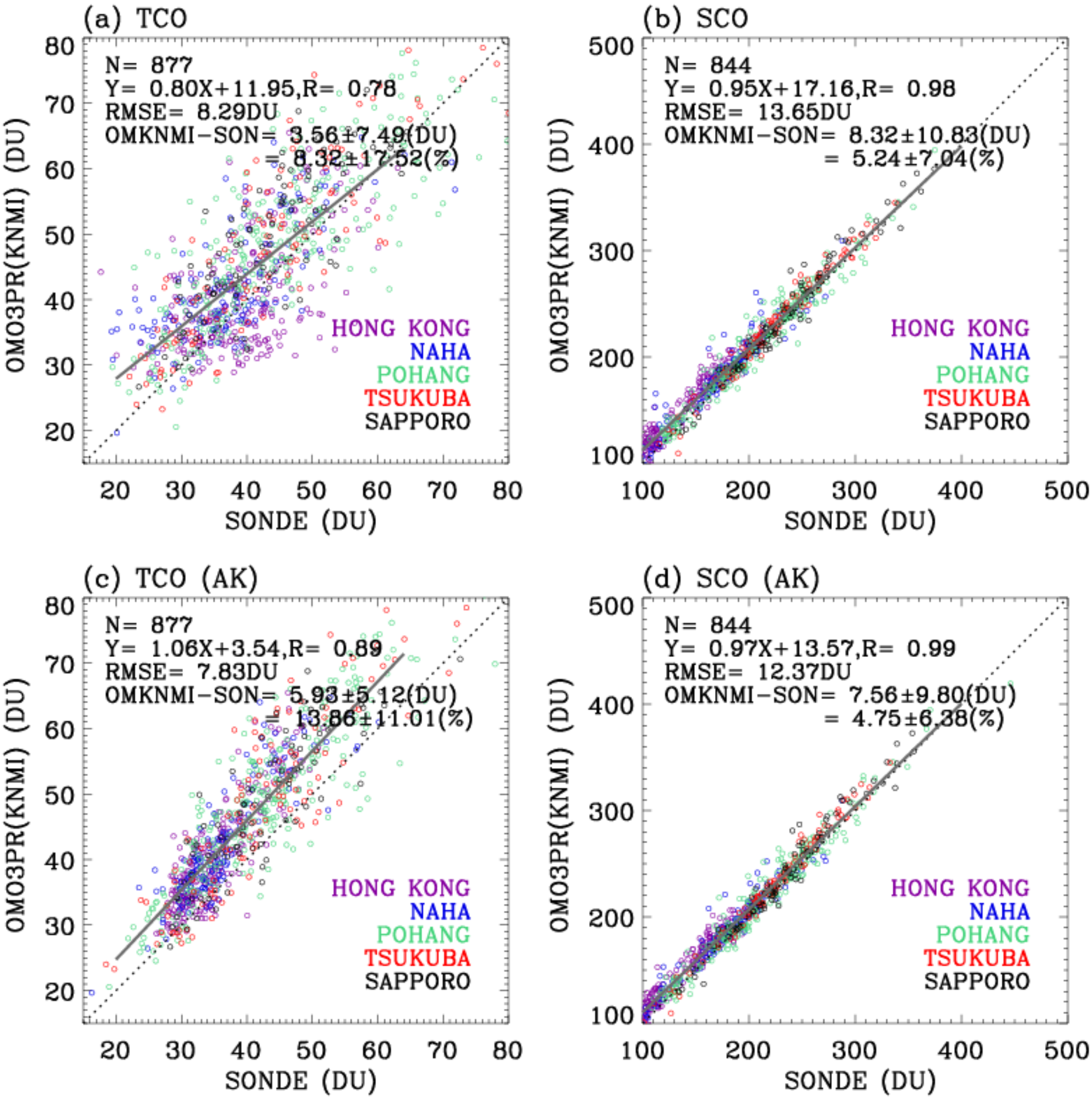

Figure 9. Same as Fig. 78, but for validating OMI standard ozone profiles (OMO3PR) produced by the KNMI optimal estimationOE--based algorithm.

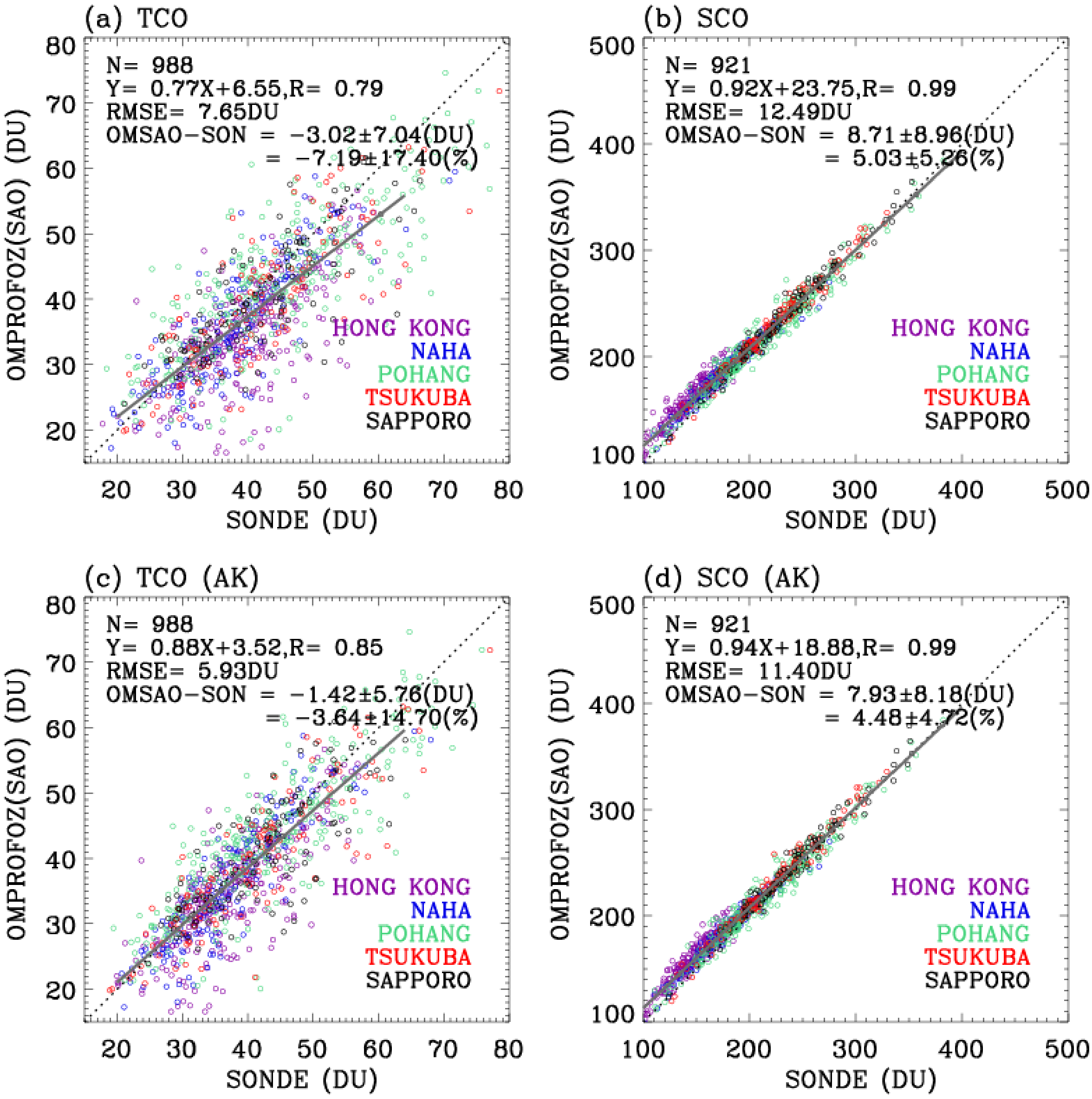

Figure 10. Same as Fig. 8, but for validating OMI research ozone profiles (OMPROFOZ) produced by the SAO ~~optimal estimation~~ OE-based algorithm.

Table1. List of ozonesonde stations.

Station[a]	Lon (°), Lat (°)	Altitude (m)	Observation Time [b]		Instrument Type[c]	ECC-SST [d]	Post Correction
Singapore	103.9, 1.3	40	07:30-08:00 (9)	Jan 12 - Sep 15	ECC/EN-SCI Z	SST0.5	No correction
				Nov15 - Dec15	ECC/SPC 6A		
Kuala Llumpur	101.7, 2.7	20	9:30-15:00 (104)	Jan 13 - Dec14	ECC/SPC 6A	SST1.0	Transfer function
				Jan 15 - Dec15	ECC/EN-SCI Z	SST0.5	
Trivandrum	77.0, 8.5	60	14:00-14:30 (34)	Jan 06 - Dec11	MBM		Correction factor
Hanoi	105.8, 21.0	10	12:00-14:00 (42)	Jan 05 - Apr 06	ECC/EN-SCI 1Z	SST2.0	Transfer function
				Apr06 - Dec 07	ECC/EN-SCI 2Z	SST2.0	
				Jan 08 - May 09	ECC/EN-SCI 2Z	SST1.0	
				Jun 09 - Dec 09	ECC/SPC 6A	SST1.0	
				Feb 10 - Dec 11	ECC/EN-SCI Z	SST1.0	
				Feb 12 - Dec 13	ECC/EN-SCI Z	SST2.0	
				Jan 15 - Dec 15	ECC/EN-SCI Z	SST0.5	
Hong Kong	114.1, 22.3	70	13:00-14:30 (11)	Jan 05 - Dec 15	ECC/SPC 6A	SST1.0	No correction
Naha	127.7, 26.2	30	14:30-15:00 (06)	Jan 05 - Oct 08	CI/ KC-96		Correction factor
				Nov 09 - Dec 15	ECC/EN-SCI 1Z	SST0.5	
New Delhi	77.1, 28.3	270	11:00-14:30 (69)	Feb 06 - Dec11	MBM		Correction factor
Pohang	129.2, 36.0	40	13:30-15:30 (24)	Jan 05 - Dec 15	ECC/SPC 6A	SST1.0	No correction
Tsukuba	140.1, 36.1	330	14:30-15:00 (08)	Jan 05 - Nov 09	CI/ KC-96		Correction factor
				Dec 09 - Dec 15	ECC/EN-SCI 1Z	SST0.5	
Sapporo	141.3, 43.1	30	14:30-15:00 (06)	Jan 05 - Nov 09	CI/ KC-96		Correction factor
				Dec 09 - Dec 15	ECC/EN-SCI 1Z	SST0.5	

[a] Data are downloaded from the WOUDC (http://woudc.org) data archive, except for Kuala Llumpur and Hanoi, which are from the SHADOZ

(https://tropo.gsfc.nasa.gov/shadoz/) network, and Pohang, which are from the Korea Meteorological Administration (KMA).
[b] The range of the observation time (LT) with 1 σ standard deviations of them (min) in ~~the~~ parentheses.
[c] Ozonesonde sensor type (ECC: Electrochemical Condensation Cell, CI: Carbon iodine cell Japanese sonde, MBM: Modified Brewer-Mast Indian sonde). ECC sensors manufactured by either ECC sensor manufactures; Science Pump Corporation (Model type: SPC-6A) and Environmental Science cooperation (Model type EN-SCI-Z/1Z/2Z).

Table 2. Comparison ~~Statistics~~ statistics (~~Mean~~ mean ~~Bias~~ bias in DU, 1σs ~~Standard~~ standard ~~Deviation~~ deviation in DU, and *R*, ~~Correlation~~ correlation ~~Coefficient~~coefficient) between GEMS simulated ~~Tropospheric~~ tropospheric o~~O~~zone c~~C~~olumn and o~~O~~zonesonde m~~M~~easurements convolved with GEMS averaging kernels.

Station	Collocation Time difference	Type	Data Period (Year)	SONDE AK – GEMS		
				#	Mean Bias + 1**σ**	*R*
Singapore	6:44	ECC	12-15	20	-13.67 ± 9.61	0.17
Kuala L~~l~~umpur	2:29	ECC	05-15	106	-2.54 ± 4.13	0.44
Trivandrum	1:46	MB-M	06-11	37	3.55 ± 9.75	0.24
Hanoi	0:32	ECC	05-15	100	-3.82 ± 6.03	0.52
Hong Kong	0:27	ECC	05-15	259	-1.19 ± 3.91	0.82
Naha	0:47	CI	05-08	135	-5.48 ± 4.07	0.85
		ECC	08-15	166	-0.94 ± 3.22	0.91
New Delhi	1:46	MB-M	06-11	39	-4.57 ± 13.36	0.24
Pohang	0:54	ECC	05-15	281	-0.75 ± 3.13	0.95
Tsukuba	1:56	CI	05-09	151	-2.98 ± 3.76	0.91
		ECC	09-15	154	-0.65 ± 3.53	0.94
Sapporo	2:18	CI	05-09	107	-3.43 ± 2.56	0.94
		ECC	09-15	95	-1.37 ± 2.79	0.93