# a point-by-point response to the reviews

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2	<b>Response to referee #1's comments</b>
3	We replied to 9 main comments and 9 minor comments hereafter.
4	Main Comment
5 6 7 9 10 11 12 13 14 15 16 17 18 19 20 21	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.
22 23 24 25 26 27 28 29 30 31 32 33 34	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"

- 35 <u>tropospheric ozone retrieval performance using OMI data and the use of ozonesonde</u>
- 36 <u>dataset over East Asia for validation</u>" to clearly reflect what we did through this work.

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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.

41 GEMS is a scanning Uv/Visible spectrometer with a single UV-enhanced CCD for the 42 spectral measurements of 300-500 nm (FWHM: 0.6 nm, spectral sampling: 0.2 nm) with 43 at least comparable radiometric/wavelength accuracy (4% including light source 44 uncertainty/0.01 nm) as OMI. However, GEMS data processing is expected to be different 45 tofrom OMI mainly in two ways: 1) OMI uses a depolarizer to scramble the polarization 46 of light. However, GEMS has polarization sensitivity (required to be less than 2%) and 47 performs polarization correction using an RTM-based look-up table of atmospheric 48 polarization state and pre-flight characterization of polarization instrument polarization 49 sensitivity in the level 0 to 1b data processing. The GEMS polarization correction is less 50 accurate and hence additional fitting process might be required in the level 2 data 51 processing, especially for ozone profiles that have more significant retrieval sensitivityare 52 more sensitive to the polarization error compared to other trace-gases. 2) GEMS has a 53 capability to perform diurnal observations and hence the diurnal meteorological input data 54 are required to account for the temperature dependent Huggins band ozone absorption. 55 Hence, the numerical weather prediction (NWP) model analysis data will be transferred to 56 the GEMS Sscience Ddata Pprocessing Ccenter (SDPC). This part is clarified in the 57 Section 2.1 of the revised manuscript.

59 5. Section 3.1. The discussion of the differences between satellite/sonde agreements at the 60 different sites is interesting. In addition to the differences between the sonde characteristics 61 and reliabilities, one might expect greater standard deviations at sites that are polluted 62 and/or show greater variability in ozone loadings due to meteorology. Some further 63 discussion would be useful about the chemical-transport environment before eliminating 64 sites from potential GEMS validation based on instrumentation/ experimental method 65 arguments alone. It would also help this reader if the current dense text were broken up 66 into descriptions of the various reasons for good/bad agreement.

67 We have added the figure 4 in which the seasonal mean and standard deviations of 68 ozonesonde measurements are presented to see the stability and characteristics of 69 ozonesonde measurements at each site. Instabilities of measurements are observed 70 from New Delhi ozonesondes. High surface ozone concentrations at Trivandrum in 71 summer are believed to be caused by measurement errors because low levels of 72 pollutants have been reported at this site under the geolocation and meteorological 73 effects (Lal et al. 2000). Besides Trivandrum, Naha could be regarded as background 74 sites according to low surface ozone and its precursor concentrations compared to 75 neighboring stations (Fig. 2 and Fig. 4), and previous studies (Oltmans et al., 2004; 76 Liu et al., 2002). In the lower troposphere high ozone concentrations are captured at

77 Pohang, Tsukuba, and Sapporo in the summer due to enhanced photochemical 78 production of ozone in daytime, whereas tropical sites, Naha, Hanoi, and Hong Kong 79 show the ozone enhancement in spring mainly due to biomass burning in Southeast 80 Asia, with low ozone concentrations in summer due to the Asian monsoon and in 81 winter due to the tropical air intrusion (Liu et al., 2002; Ogino et al., 2013). Singapore 82 and Kuala lump are supposed to be severely polluted area, but ozone pollution is not 83 clearly captured over the seasons. It could be explained by the morning observation 84 time at these two stations. In addition, instabilities of Singapore measurements are 85 noticeable, including abnormally large variability and very low ozone concentration in 86 the stratosphere. The effect of stratospheric intrusions on the ozone profile shape is 87 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 88 89 the lower stratosphere and upper troposphere, whereas ozonepause is around 100 hPa 90 with much less variability of ozone in other seasons. This discussion has been included 91 in Section 3. 92

 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.

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 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.



101 102 Fig. 4. Seasonal mean (solid) and standard deviation (dashed) of ozonesonde soundings from 2005 to

103 2015 at 10 sites. 5 mPa is subtracted to standard deviations to fit in the given x-axis.



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Figure 2. Geographic locations of the ozonesonde stations available since 2005 over the GEMS
 observation domain. The background map illustrates the OMI NO<sub>2</sub> 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?

112 To explain this impact, the comparison between GEMS a priori and ozonesondes is 113 presented below, similarly to Fig.6 (in old manuscript, it is Fig. 7 in the revised one). 114 This a priori information is taken from the tropopause-based ozone profile climatology 115 (TB) which adjusts a monthly and zonal mean ozone profile with a daily tropopause 116 height. Bak et al. (2013) demonstrated that TB based a priori better represents the 117 ozone variabilities in the extra-tropical upper troposphere and lower stratosphere, 118 especially during the winter and spring when the atmospheric status is strongly 119 controlled by the dynamics. A priori information is very important to the quality of UV 120 ozone profile retrievals, but the retrieved ozone profiles show much better agreement 121 with ozonesondes than a priori ozone profiles, implying that the independent piece of 122 information are available from these UV measurements. We can see that the biases 123 seen in a priori are significantly reduced for most stations except for the Singapore 124 station. Positive biases around 15 km in the a priori at the three mid-latitude sites still 125 remain in the retrievals but with much smaller magnitude. Negative biases around ~17 126 km at other lower-latitude sites (except for Singapore) are almost eliminated in the 127 retrievals.





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

132 7-8. Section 4.2 should be split into another sub section at Line 354 that starts the discussion 133 of the evaluation of the GEMS algorithm against the OMI algorithm. This should be presented 134 as one of the main results sections of this paper: a quantitative evaluation of the GEMS 135 algorithm against other widely used algorithms based on the same OMI radiances. Given the 136 previous discussion in the paper of the various limitations of some of the sondes, it might 137 additionally be useful to directly compare the results of the different retrieval algorithms and 138 explain differences in results on the basis of different features of the algorithms. This would 139 help make the case that the GEMS algorithm is performing as expected.

140 In this paper, the ozonesonde measurements available in the GEMS domain are 141 characterized and validated to better evaluate the performance of the GEMS ozone 142 profile algorithm. The accuracy and precision of simulated GEMS ozone profiles are established against the selected true reference. Additionally, the consistent evaluation 143 144 is performed for the existing OMI ozone products to check the confidence of the 145 GEMS retrieval algorithm, demonstrating that the comparable or better performance 146 of GEMS ozone profile retrievals in the comparison with ozonesonde measurements. 147 The different validation results between retrieval algorithms were discussed such as 148 "GEMS algorithm is developed based on the heritages of the SAO ozone profile 149 algorithm with several modifications. There are two main modifications: a priori ozone 150 climatology was replaced with a tropopause-based ozone profile climatology to better 151 represent the ozone variability in the tropopause. Irradiance spectra used to normalize 152 radiance spectra and characterize instrument line shapes are prepared by taking 31-day 153 moving average instead of climatological average to take into account for time-154 dependent instrument degradations. These modifications reduce somewhat spreads in 155 deviations of satellite retrievals from sondes, especially in TCO comparison. KNMI 156 retrievals systematically overestimate the tropospheric ozone by  $\sim 6$  DU (Fig. 9.c),

157	which corresponds to the positive biases of 2-4 % in the integrated total columns of
158	KNMI profiles relative to Brewer observations (Bak et al., 2015). As mentioned in Bak
159	et al. (2015), the systematic biases in ozone retrievals are less visible in SAO-based
160	retrievals (GEMS simulation, OMPROFOZ) as systematic components of measured
161	spectra are taken into account for using an empirical correction called "soft
162	calibration". The GEMS algorithm is very similar to the SAO algorithm except for the
163	use of TB climatology and the impact of TB on the retrievals was discuss in detail in
164	Bak et al. (2013). Also the comparison of SAO and KNMI algorithms were discussed
165	in detail in Bak et al. (2015). So we think that it is more efficient to place the discussion
166	related to Figures 7-9 in the same section and the direct comparison of GEMS and
167	other OMI product is beyond the scope of this paper.
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169	9. The paper requires careful, and extensive, editing for English usage, and cut-paste typos, e.g.
170	line 75, that should have been corrected before manuscript submission.
171	<ul> <li>This manuscript is going to be carefully revised though native English co-author</li> </ul>
172	before the submission of the revised manuscript.
173	1
174	Minor Comments
175	1. Several times "GEMS" measurements are described. The word "simulated" should
176	be added each time to avoid confusion
177	<ul> <li>In this revised manuscript, "GEMS measurements" was edited to "<u>simulated GEMS</u></li> </ul>
178	measurements"
179	2. Line 83: consistent perhaps, but not homogeneous as the authors point out in the text above.
180 181	<ul> <li>The indicated sentence was revised from "a homogenous, consistent ozonesonde" to "a consistent ozonesondo".</li> </ul>
182	" <u>a consistent ozonesonde</u> ". 4. Line 105: Instrument errors certainly, but also instrument design sensitivity.
183	<ul> <li>The indicated word, "instrumental errors" was revised to "Instrument errors certainly,</li> </ul>
	-
184	but also instrument design sensitivity"
185	5. Line 106: Common geophysical conditions can reduce sensitivity, not just extreme
186	• The associated sentence was edited from "The impact of a priori information on
187 188	retrievals become important ~~ under extreme geophysical conditions to ~~ <u>under</u> <u>certain geophysical conditions</u> .
189	<u>certain geophysical conditions</u> .
190	6. Line 123: Information may be limited but is a goal of the GEMS mission. This should be
191	clarified.
192	<ul> <li>The GEMS mission was originally planned to develop the spectrometer for measuring</li> </ul>
193	the tropospheric pollutants, the spectral coverage of 300-500 nm satisfies to observe
194 105	the tropospheric ozone as well as the lower/middle stratospheric ozone.
195 106	7 Line 157. Any more recent references to new more restricted with a line to the intervent to the
196 197	<ul> <li>7. Line 157: Any more recent references to new measurement technique and instrumentation?</li> <li>More references (Thompson et al., 2017; Witte et al., 2017; 2018) are added in this</li> </ul>
198	sentence.

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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?

- 204 As mentioned in Section 2.3, the coincidence criteria between satellite and ozonesonde 205 are:  $\pm 1.0^{\circ}$  in both longitude and latitude and  $\pm 12$  hours in time and then the closest 206 pixel is selected. The actual spatiotemporal difference is much smaller than this criteria 207 (57.5 km to 66.6 km, ~3 hours). The close collocation can significantly minimize the 208 effects of spatiotemporal variability on the comparison and therefore GEMS validation 209 accuracy could be enhanced compared to OMI, which is newly included in the 210 summary of this paper (The impact of spatiotemporal variability on the comparison 211 will be much reduced for GEMS due to its higher spatiotemporal resolution (7 km x 8 212 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**
- 219 We replied to 4 main comments and 25 minor comments

# 220 Main Comment

221 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 usingLEO measurements for a GEO instrument.

224 **R1**. The development of the GEMS L2 algorithm has been in progress with OMI measurements 225 because the simulation of the GEMS radiances using the forward model has not been fully 226 implemented. Two main differences in GEMS and LEO (OMI) data processing could be 227 expected: 1) OMI use a depolarizer to scramble the polarization of light. However, GEMS has 228 polarization sensitivity (required to be less than < 2%) and performs polarization correction 229 using RTM-based look-up table of atmospheric polarization state and pre-flight 230 characterization of polarization sensitivity in the level 0 to 1b data processing. The GEMS 231 polarization correction is less accurate and hence additional fitting process might be required 232 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.
- 239

C2. The use of OMI measurements makes the title of the paper confusing as the validation isof 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".

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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.

252 **R3**. Understanding the quality of the reference dataset and then selecting a good reference 253 is a very important process in validating satellite or other in-situ measurements and then in 254 better characterizing the retrieval accuracy and error. Satellite measurements of tropospheric 255 ozone have previously been utilized to disclose problems in ozonesonde observations (e.g., Liu 256 et al., 2006; Huang et al., 2018). We are also using retrievals here to identify ozonesonde 257 measurements with significant errors. However, the station-to-station based quality control has 258 not been typically applied in previous validation works. The figure below demonstrates how 259 much the accuracy of the simulated GEMS retrievals from OMI measurements is 260 underestimated if the station-to-station based quality control is not applied. We also apply the 261 parallel validation for two independent OMI ozone profile products, OMPROFOZ and 262 OMO3PR, respectively, demonstrating that our ozone retrievals are in comparable or better 263 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.

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

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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.

- 273 **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

275	between satellite and ozonesonde are: $\pm 1.0^{\circ}$ in both longitude and latitude and $\pm 12$ hours in
276	time and then the closest pixel is selected. The Aura satellite carrying OMI crosses the equator
277	always at $\sim$ 1:45 pm LT and thereby OMI measurements are closely collocated within 3 hours
278	to ozonesonde soundings measured in afternoon (1-3 pm LS)," OMI measurements are closely
279	collocated within 3 hours to ozonesonde soundings measured in afternoon (1-3 pm) from
280	Japanese stations, Pohang, Hong Kong, Hanoi, and Trivandrum. In this paper, the time
281	collocation criterion is set to be 12 hours to include other stations existing over the GEMS
282	domain.
283	
284	Minor comments
285	C1. Line 50: Satellite name should be Sentinel-4
286	R1. This name has been corrected to "Sentinel-4"
287	
288	C2. Line 75: "::: have yet to be not been" please correct this
289	<b>R2</b> . It has been corrected to "has not been".
290	
291	C3. Line 178: Among ECC stations
292	<b>R3</b> . It has been corrected to "Among ECC stations".
293	
294	C4. Line 183: "Kula lump", please correct. Also all along the paper, the name of this station is
295	written in different ways (Kuala lump, Kuala Lumpur). Please homogenize the station names
296	in the text, figures and tables.
297	R4. We carefully checked what this reviewer indicated. This station name has been corrected
298	to "Kuala Lumpur" across the manuscript.
299	
300	C5. Line 221: biased -> bias
301	<b>R5</b> . It has been corrected to "bias".
302	
303	C6. Line 225: Please specify the units
304	R6. RMS does not have the unit and thereby "RMS (i.e., root mean square of fitting residuals
305	relative to measurement errors) less than 3" has been kept in the revised manuscript.

306	
307	<b>C7</b> . Line 231: troposphere -> stratosphere
308	<b>R7</b> . It has been corrected to "stratosphere".
309	
310	<b>C8</b> . Line 234: Should be photons?
311	<b>R8</b> . It has been revised to "photons".
312	1
313	C9. Line 242: xa should be placed after (1-A)
<b>3</b> 14	<b>R9</b> . Eq. 3 has been revised to " $\hat{x}_{sonde} = A \cdot x_{sonde} + (1 - A)x_a$ "
1 315	
316	C10. Line 282: Please rephrase, maybe "of" -> "with values ranging from"
317	R10. According to this comment, "satellite retrievals show the distinct seasonal TOC variations
318	with the amplitude of $\sim$ 35-40 DU" has been edited to " $\sim$ seasonal TOC variations with the
319	values ranging from $\sim$ 35 to $\sim$ 40 DU"
320	
321	C11. Line 290: "Japanese stations" or "stations from Japan". Same in Line 296.
322 323	R11. It has been revised to "stations from Japan"
324 324	C12. Line 314: Please unify or explain the differences between LT, LS and LST across the
325	paper
326	R12. There is no difference. It has been unified to "LT (Local time)"
327	
328	C13. Line 322: "oznesonde" -> "ozonesonde"
329	<b>R13</b> . This word has been corrected.
330	
331	C14. Line 324: Please list stations after "mid-latitude" and refer to Figure justifying this and
332	the following statements.
333	R14. It has been clarified such as "mid-latitude (Pohang, Tsukuba, and Sapporo)"
334	
335	C15. Line 326: "- a few %" please rephrase this
336	R15. It has been corrected to "a few percent"
337	

- 338 C16. Line 338: 4.2 -> 3.2.
- **R16**. It has been changed to "3.2".
- 340
- 341 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)"
- 344 "
- 345 C18. Line 367: "especially" -> "especially". "TCO" -> TOC.
- **R18**. The relevant sentence has been corrected to "especially in the TOC comparison"
- 347
- 348 C19. Line 308: Shouldn't it be "latitudinally" as it is used in other parts of the manuscript?
  349 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.
- 359
- 360 C20. Line 399: Extra s "is similarly"
- 361 **R20.** This indicated one (is s similarly) has been corrected (is similarly)
- 362
- 363 C21. Figure 2: Latitudes and Longitudes are not correct.
- 364 **R21**. This figure has been revised.
- 365
- 366 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.
- 368 **R22**. To clarify, the legend in the figure has been revised to "Solid: with CF, Dash: w/o CF".

369	The corresponding caption has been revised to "Effect of applying a correction factor (CF) to
370	(a) ECC and (b) CI ozonesonde measurements, respectively on comparisons with simulated
371	GEMS ozone profile retrievals. Solid and Dashed lines represent the comparisons with and
372	without applying a CF, respectively, at each Japanese station."
373	
374	C23. Figure 4: Please specify how you differentiate the different type of sondes. Is it using
375	diamonds, full dots and empty dots? Which one is which? Also indicate what is the horizontal
376	axes, eg. "time (years)"
377	<b>R23</b> . This figure has been revised to clarify the symbols and the title of x-axis.
378	
379	C24. Figure 6: I would suggest rewritting the last sentence as follows "The relative difference
380	(in %) is defined as 100 X (SONDE AK – GEMS) / (A priori)". Why is multiplied by 2?
381	R24. This equation has been corrected to "100 X (SONDE AK – GEMS) / (A priori)"
382	
383	C25. Figure 7 and 8: Please replace TCO -> TOC and SCO -> SOC to be consistent with the
384	text.
385	<b>R25</b> . This figure has been revised to accept this comment.
386	
387	Response to editor #1's comments
388	C1. In the abstract: Please, to be consistent with the new title, change « cross-verification »
389	by « cross Evaluation
390 391	<b>R2</b> . The word of "verification" has been changed to "evaluation" in the abstract.
392	C2. In sect. 2.3: please add the information of spectral resolution of GEMS and OMI to show
393 394	the reader the differences between the two instruments. <b>R2</b> . To specify what this comment points out, "It is desgined to provide hyperspectral
395	radiances at a spectral resolution of 0.6 nm and spectral intervals of 0.2 nm, which are also
396	similar to OMI (spectral resolution: 0.42-0.63 nm, sampling rate: 0.14-0.33 nm/pixel)." has
397	been added in sect.2.1.
398 399	C3. Line 486: The end of the sentence is unclear to me. I would say the opposite that the
400	ozonesonde has to verify the retrievals? Please consider also to cut it into two sentences.
401	<b>R3</b> Ultimately, ozonesonde dataset are used to qualify the accuracy of simulated GEMS
402	retrievals, but as shown in this paper, ozonesonde measurements are not consistent each other
403	due to inconsistencies in the spatial and temporal irregularities in instrument types,

404 manufacturers, operating procedures, and correction strategies. Therefore we identify

- inconsistent ozonesonde data period/sites with respect to a better consistent GEMS simulated
  retrievals because single data processing is applied to the entire satellite measurements
  measured by single instrument. For clearfication, "that is, simulated GEMS retrievals using
  OMI data retrievals ones areareis used to verify the ozonesonde observations." has been
  delected because the previous sentence is enough to specify what we point out.
- 410

411 C4. Line 567-Line 745: space-born-> space borne and « balloon-born » -> balloon-borne ?

- 412 Check through the paper if necessary.
- 413 **R4**. The mis-typed word, "born" has been corrected to "borne".
- 414

415 **C5**. Line 675 : This is not clear to me what the vertical resolution represents here (10-14 km in 416 the troposphere) and (7-11 km) in the stratosphere. Is it representing the width of the broad 417 peak of the averaging kernel? (at different levels or for the columns). If yes, I would suggest 418 you to add a typical figure of averaging kernel of GEMS ozone and why not indicating the 419 degree of freedom?

- 420 **R5.** The full width at half maximum of the main peak of each averaging kernel is an estimate
- 421 of the vertical resolutiona that altitude (Rodgers, 2000). Based on this definition, the vertical
- 422 resolution of UV ozone profile retrievals is estimated as 10-14 km in the troposphere and (7-
- 423 11 km) in the stratosphere (e.g., Kroon et al., 2010; Liu et al., 2010). In associated paragraph,
- 424 I gave the general information on the vertical resolution of UV ozone profile retrievals to
- 425 emphasize on such a discrepancy with that of ozonesonde in-situ measurements and its impact
- 426 on the comparison between satellite retrievals and ozonesonde measurements. Therefore, I
- 427 added the references (Kroon et al., 2010; Liu et al., 2010) instead of adding a figure.
- 428
- 429 C6. Line 592: There is a space missing between SST and 1.0 and please define SST and KI.
- 430 **R6.** The space between SSI and version (e.g., 1.0) is removed consistenly in this manuscript.
- 431 To specify the acronyms of SSI and KI, the sentence where these acronyms are firstly used
- been modified to "The the recommend recipes of the Standard Sensing Solution (SSI) are 1.0 %
- 433 potassium iodide (KI)/full buffer (SST1.0) and 2.0 % KI/no buffer (SST0.5) for the SPC and
- 434 EN-SCI sondes, respectively by the ASOPOS (Assessment for Standards on Operation
- 435 Procedures for Ozone Sondes) (Smit et al., 2012)"
- 436
- 437 **C7.** Table 1: upper script d is missing in the caption.
- 438 **R7.** It is included in this caption as "d Potassium Iodide (KI) cathode sensing solution type
- 439 (SST) implemented in ECC ozone sensors: SST 0.5 (0.5 % KI, half buffer), SST 1.0 (1.0 % KI,
- 440 full buffer), and SST 2.0 (2.0 % KI, no buffer). Singapore station changed it to SST 1.0 % as
- 441 of 2018"
- 442
- 443

444	a list of all relevant changes
445	1. All figures have been revised for better visibility, with newly included figure 4.
446	a marked-up manuscript version
447	
448	
449	<b><u>Cross-e</u>Evaluation of GEMS tropospheric ozone</b>
450	retrieval performance using OMI data and the use
451	of ozonesonde dataset over East Asia for
452	validation Cross-verification of simulated GEMS
453	tropospheric ozone retrievals and ozonesonde
454	measurements -
455	<b>Over Northeast Asia</b>
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469 470

#### Abstract

471 The Geostationary Environment Monitoring Spectrometer (GEMS) is scheduled to be launched in 2019 472 on board the GEO-KOMPSAT (GEOstationary KOrea Multi-Purpose SATellite)-2B, contributing as 473 the Asian partner of the global geostationary constellation of air quality monitoring. To support this air 474 quality satellite mission, we perform the <u>a cross-verification evaluation</u> of simulated GEMS ozone 475 profile retrievals from OMI (Ozone Monitoring Instrument) data based on the Optimal Estimation and 476 ozonesonde measurements within the GEMS domain, covering from 5°S (Indonesia) to 45°N (south of 477 the Russian border) and from 75°E to 145°E. The comparison between ozonesonde and GEMS shows 478 a significant dependence on ozonesonde types. Ozonesonde data measured by Modified Brewer-Master 479 (MBM-M) at Trivandrum and New Delhi show inconsistent seasonal -variabilities in the tropospheric 480 ozone, compared to latitudinally adjacent stations with Carbon Iodine (CI) and Electrochemical 481 Condensation Cell (ECC) ozonesondes at other equipped-stations in a similar latitude regime. CI 482 ozonesonde measurements are negatively biased relative to ECC measurements by 2-4 DU;;; a bBbetter 483 agreement with GEMS simulations is achieved with when simulated GEMS ozone retrievals are 484 compared to ECC measurements. ECC ozone data at Hanoi, Kuala Lumpur, and Singapore show 485 abnormally worse agreements with simulated GEMS retrievals among than other ECC measurements. 486 Therefore, ECC ozonesonde measurements at Hong Kong, Pohang, Naha, Sapporo, and Tsukuba are 487 finally identified as an optimal reference dataset. The accuracy of simulated GEMS retrievals is 488 estimated to be ~ 5.0 % for both tropospheric and stratospheric column ozone with the precision of 15 % 489 and 5 %, which meets the GEMS ozone requirements. 490

491 **1. Introduction** 

492

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 polarorbiting instrument heritages for the last 40 years, which were initiated with the launch of a series of Total Ozone Mapping Spectrometer (TOMS) instruments <u>since-starting in</u> 1978 (Bhartia et al., 1996) and consolidated by <u>the\_</u>Global Ozone Monitoring Experiment (GOME) (ESA, 1995), <u>the\_</u>SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) (Bovensmann et al., 1999), <u>the\_</u>

500 Ozone Monitoring Instrument (OMI) (Levelt et al, 2006), GOME-+2 (EUMETSAT, 2006), the 501 Ozone Mapping and Profiler Suite (OMPS) (Flynn et al., 2014), and the TROPOspheric Monitoring 502 Instrument (TROPOMI) (Veefkind et al., 2012). Three geostationary air quality monitoring missions, 503 including the Geostationary Environmental Monitoring Spectrometer (GEMS) (Bak et al., 2013a) over 504 East Asia, Tropospheric Emissions: Monitoring of Ppollution (TEMPO) (Chance et al, 2013; Zoogman 505 et al., 2017) over North America, and Sentineal-4 (Ingmann et al., 2012) over Europe, are in progress 506 to launch their instruments in the 2019-2022 time frame, which will to provide unprecedented hourly 507 measurements of aerosols and chemical pollutants at sub-urban scale spatial resolution (~ 10-50 km<sup>2</sup>). 508 These missions will constitute the global geostationary constellation of air quality monitoring.

GEMS will be launched in late 2019—<u>or early 2020</u> on board the GeoKOMPSAT-<u>2B</u> (Geostationary Korea Multi-Purpose Satellite) to measure O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, H<sub>2</sub>CO, 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 <u>its</u> selfcleansing capacity as a precursor of the hydroxyl radical, and in controlling the Earth''s radiative balance as a greenhouse gas.

515 To support the development of the GEMS ozone profile algorithm, Bak et al. (2013a) demonstrated 516 that the GEMS spectral coverage of 300-500 nm minimizes the loss in the sensitivity to tropospheric 517 ozone despite the lack of most Hartley ozone absorption wavelengths shorter than 300 nm. They further 518 indicated the acceptable quality of the simulated stratospheric ozone retrievals from 212 hPa to 3 hPa 519 (40 km) through comparisons using Microwave Limb Sounder (MLS) measurements. As a consecutive 520 work, this study evaluates simulated GEMS tropospheric ozone retrievals against ozonesonde 521 observations. GEMS ozone retrievals are simulated using an Ooptimal Eestimation (OE) based fitting 522 algorithm from with OMI radiances with using the fitting window of in the spectral range 300-330 nm 523 in the same way as Bak et al. (2013a). The validation effort is essential to ensuring the quality of GEMS

524 ozone profile retrievals and to verifying the newly implemented ozone profile retrieval scheme. In-situ 525 ozonesonde soundings have been considered to be the best reference, but should be carefully used due 526 to its the spatial and temporal irregularities in instrument types, manufacturers, operating procedures, 527 and correction strategies (Deshler et al., 2017). Compared to TEMPO and Sentinel-4, validating the 528 GEMS validation activity GEMS ozone retrievals is expected to be more challenging for the ozone 529 profile product because of the much sparser distribution of stations and more irregular characteristics 530 of the ozonesonde dataset measurements over the GEMS domain. Continuous balloon-borne 531 observations of ozone are only available from at the Pohang (129.23°E, 36.02°N) site in South Korea, 532 but this site has ve yet to be not been thoroughly validated. Therefore the quality assessment of the its 533 ozonesonde data is required before we use this data for GEMS validation activity. Compared to 534 ozonesondes, satellite ozone data are less accurate and have much coarser vertical resolution, but more 535 homogenous due to its-single data processing for the entire-measurements from a single instrument. 536 Therefore, abnormal deviations in satellite-ozonesonde differences from neighboring stations might 537 indicate problems at individual stations (Fioletov et al. 2008). For example, Bak et al. (2015) identified 538 27 homogenous stations among 35 global Brewer stations available from the World Ozone and 539 Ultraviolet Radiation Data Centre (WOUDC) network through comparisons with coincident OMI total 540 ozone data. This study adopts this approach to select a homogenous, consistent ozonesonde dataset 541 among 10 stations available over the GEMS domain based on the comparisons of the tropospheric ozone 542 columns (TOC) between simulated GEMS retrievals and ozonesonde measurements, that is, simulated 543 GEMS retrievals using OMI data retrievals ones areare used to verify the ozonesonde observations. 544 The simulated GEMS retrievals are ultimately evaluated against the ozonesonde dataset identified as a 545 true reference to demonstrate the reliability of our future GEMS ozone product. The simulated GEMS 546 retrievals and ozonesonde dataset are described in Sect. 2.1 and 2.2 with the comparison methodology 547 in Sect. 2.3. Our results are discussed in Sect. 3 and summarized in Sect. 4.

548

# 549 **2. Data and Methodology**

550

# 551 2.1 Ozone Profile Retrievals

552

The development of the GEMS ozone profile algorithm builds on <u>the heritages of the Smithsonian</u>
Astrophysical Observatory (SAO) ozone profile algorithm which was originally developed for GOME
(Liu et al., 2005), continuously adapted for its successors <u>such asincluding</u> OMI (Liu et al., 2010a),
GOME<sub>2</sub>/2 (Cai et al., 2012), and OMPS (Bak et al., 2017). In addition, the SAO algorithm will be

557 implemented to retrieve TEMPO ozone profiles (Chance et al., 2013; Zoogman et al., 2017). In this 558 algorithm, the well-known optimal estimation (OE) based iterative inversion is applied to estimate the 559 best ozone concentrations from simultaneously minimizing between measured and simulated 560 backscattered UV measurements constrained by the measurement covariance matrix, and between 561 retrieved values and its climatological a priori values constrained by an a priori covariance matrix 562 (Rodgers, 2000). The impact of a priori information on retrievals becomes important when measurement 563 information is reduced due to instrumental errors, certainly, but also instrument design sensitivity (e.g. 564 stray\_light, dark\_-current, and read-out smear), and or physically insufficient sensitivities under extreme 565 certain geophysical conditions (e.g. the reduced penetration of incoming UV radiation into the lower 566 troposphere at high solar zenith angles or, blocked photon penetration below thick clouds). The 567 described OE-fitting solution  $\hat{X}_{i+1}$  can be written, together with cost function  $\chi^2$ :

568

- $\hat{X}_{i+1} = \hat{X}_i + \left(K_i^T S_y^{-1} K_i + S_a^{-1}\right)^{-1} \{K_i^T S_y^{-1} \left[Y R(\hat{X}_i)\right] S_a^{-1} (\hat{X}_i X_a)\}$ (1)
- 570

571

$$\chi^{2} = \left\| S_{y}^{-\frac{1}{2}} K_{i} (\hat{X}_{i+1} - \hat{X}_{i}) - [Y - R(\hat{X}_{i})] \right\|_{2}^{2} + \left\| S_{a}^{-\frac{1}{2}} (\hat{X}_{i+1} - X_{a}) \right\|_{2}^{2}$$
(2)

572

573 <u>w</u>Where  $\hat{X}_{i+1}$  and  $\hat{X}_i$  are current and previous state vectors with a priori vector,  $X_a$  and its 574 covariance error matrix,  $S_a$ . Y and R(X) are measured and simulated radiance vectors, with 575 measurement error covariance matrix,  $S_y$ . *K* is <u>the</u> weighting function matrix ( $\frac{dR(x)}{dx}$ ), describing the 576 sensitivity of the forward model to small perturbations of the state vector.

The ozone fitting window was determined toward maximizezing the retrieval sensitivity to ozone and minimizeing 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 leastto below ~ 40 km (Bak et al., 2013a).

Fig. 1 presents-<u>is</u> 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 <u>a</u> 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); <u>a</u> normalized Gaussian 588 distribution is assumed to derive analytic slit functions for OMI. To remove the systematic errors 589 between measured and calculated radiances, "soft-calibration" is applied to measured radiances and 590 then the logarithms of sun-normalized radiances is are calculated as a measurement vectors (Liu et al., 591 2010a; Cai et al., 2012; Bak et al., 2017). A mMMeasurement covariance matricesx is are constructed 592 as a diagonal matrices with each components taken from the square of the measurement errors as 593 measurement errors are assumed to be uncorrelated between among wavelengths; ... In the OMI 594 algorithm, for OMI thea noise floor noise of 0.4 % (UV1) and 0.2 % (UV2) is used because OMI 595 measurement errors underestimate other kinds of random noise errors caused by stray\_light, dark current, 596 geophysical pseudo-random noise errors due to sub-pixel variability, and motion when taking a 597 measurement, forward model parameter error (random part), and other unknown errors into account 598 (Huang et al., 2017). GEMS is expected to hvaehave similar retrieval sensitivity to tropospheric ozone, 599 and have at least comparable radiometric/wavelength accuracy (4% including light source 600 uncertainty/0.01 nm) as OMI. It is desgined to provide hyperspectral radiances at a spectral resolution 601 of 0.6 nm and spectral intervals of 0.2 nm, which are also similar to OMI (spectral resolution: 0.42-0.63 602 nm, sampling rate: 0.14-0.33 nm/pixel). A priori ozone information is taken from the tropopause-based 603 (TB) ozone profile climatology, which was developed for improving ozone profile retrievals in the 604 upper troposphere and lower stratosphere (Bak et al., 2013b). The Vector LInearized Discrete Ordinate 605 Radiative Transfer (VLIDORT) model (Spurr, 2006; 2008) is run-used to calculate the-normalized 606 radiances and weighting function matrices\* for the atmosphere, with Rayleigh scattering and trace-gas 607 absorption and with Lambertian reflection for both surface and cloud (Liu et al., 2010a). The ozone 608 algorithm iteratively estimates the best ozone profiles within the retrieval converges (typically 2-3 609 iterations), together with other geophysical and calibration parameters (e.g., cloud fraction, albedo, BrO, 610 wavelength shifts, Rring parameter, mean fitting scaling parameter) for a better fitting accuracy even 611 though some of the additional fitting parameters can reduce the degrees of freedom for signal of ozone. 612 We should note here that GEMS data processing is expected to be different to from OMI mainly in two 613 ways: 1) OMI uses a depolarizer to scramble the polarization of light. However, GEMS has polarization 614 sensitivity (required to be less than 2%) and performs polarization correction using an RTM-based look-615 up table of atmospheric polarization state and pre-flight characterization of polarization-instrument 616 polarization sensitivity in the level 0 to 1b data processing. The GEMS polarization correction is less 617 accurate and hence additional fitting process might be required in the level 2 data processing, especially 618 for ozone profiles that have more significant retrieval sensitivity are more sensitive to the polarization 619 error compared to other trace-gases. 2) GEMS has a capability to perform diurnal observations 620 and hence the diurnal meteorological input data are required to account for the temperature dependent 621 Huggins band ozone absorption. Hence, the numerical weather prediction (NWP) model analysis

622 data will be transferred to the GEMS Sscience Ddata Pprocessing Ceenter (SDPC).

# 624 **2.2 Ozonesonde measurements**

625

623

626 Ozonesondes are small, lightweight, and compact balloon-borne instruments capable of measuring 627 profiles of ozone, pressure, temperature and humidity from the surface to balloon burst, usually near 35 628 km (4 hPa); ozone measurements are typically reported in the units of partial pressure (mPa) with the 629 vertical resolution of  $\sim$  100-150 m (WMO, 2014). Ozone soundings have been taken for more than 50 630 years, since the 1960s. The accuracy of ozonesonde measurements has been reported as 5-10 % with 631 the precision of 3-5%, depending on the sensor type, manufacturer, solution concentrations, and 632 operational procedure (Smit et al., 2007; Thompson et al., 2007; 2017; Witte et al., 2017; 2018). The 633 three types of instruments have been carried on balloons, i.e., the modified Brewer-Master (MB-M), 634 the carbon iodine cell (CI), and the electrochemical concentration cell (ECC)., the carbon iodine cell 635 (CI). Each sounding is disposably operated and hence weekly launched for the long-term operation.

636 Fig. 2 displays the locations of 10 ozonesonde sites focused on this study within the expected 637 GEMS domain borderinged from 5°S (Indonesia) to 45°N (south of the Russian border) and from 75°E 638 to 145°E. A summary of each ozonesonde site is presented in Table 1. Most of measurements are 639 collected from the WOUDC network, except that Pohang soundings are provided from the Korea 640 Meteorological Administration (KMA) and Kuala Lumpur and Hanoi measurements are from the 641 Southern Hemisphere Additional OZonesondes (SHADOZ) network. In South Korea, ECC sondes have 642 been launched every Wednesday since 1995 only at Pohang, without significant time gaps. There are 643 three Japanese stations (Naha, Tsukuba, and Sapporo) where the CI--typed sensor was used and before 644 switchinged to the ECC-typed sensor as of early 2009, and two Indian stations at New Delhi and 645 Trivandrum using the Modified modified B-M (MB-M) sensor. The rest of stations (Hanoi, Hong Kong, 646 Kuala Lumpur and Singapore) uses only ECC. Most stations employ an-ECC ozone-sensors, but 647 inhomogeneities in ECC ozonesondes are strongly addressed with respect<u>correlated</u> to the preparation 648 and correction procedures. There are two ECC sensor manufactures: the +Science Pump Corporation 649 (Model type: SPC-6A) and the Environmental Science Corporation (Model type: EN-SCI-Z/1Z/2Z). 650 Since 2011 EN-SCI has been taken over by Droplet Measurement Technologies (DMT) Inc. The the 651 recommend recipes of the Standard Sensing Solution (SSI) are 1.0 % potassium iodide (KI)/full buffer 652 (SST1.0) and 2.0 % KI/no buffer (SST0.5) for the SPC and EN-SCI sondes, respectively by the 653 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

655 solution to all ECC observation sensors manufactured by with its one manufactureone company. In 656 Singapore, the ozonesonde manufacture was changed in late 2015 from EN-SCI to SPC, while SST 0.5 657 was switched to SST1.0 as of 2018. Two SHADOZ stations (Kuala LumpurKuala lump, Hanoi) have 658 applied the standard sensing solution just since 2015. Hanoi changed sensing solution 4 times with two 659 different ozonesonde manufactures; Kuala Lumpur Kula lump-operated only with SPC 6A-SST1.0 660 combination until 2014, but with four different radiosonde manufactures. Therefore these SHADOZ 661 datasets were reprocessed homogenized (in-Witte et al., (2017) through the application of transfer 662 functions between sensors and solution typess to be homogenized. The post-processing could be applied 663 by data users to some WOUDC datasets given a correction factor, which is the ratio of integrated 664 ozonesonde column (appended with an estimated residual ozone column above burst altitude) and total 665 ozone measurements from co-located ground-based and/or overpassing satellite instruments. The 666 above-burst column ozone is estimated with a constant ozone mixing ratio (CMR) assumption above 667 the burst altitude (e.g., Japanese sites, )-(Morris et al., 2013) or satellite derived stratospheric ozone 668 climatology (e.g., Indian sites, )-(Rohtash et al., 2016). No post-processing is given todone for Pohang, 669 Hong Kong, and Singapore. Most stations made weekly or bi-weekly regular observations, except for 670 Indian stations with irregular periods of 0-4 per month and for Singapore with monthly observations.

671 In Fig. 3 the seasonal means and standard deviations of ozonesonde measurements are 672 presented to seeshow the stability and characteristics of ozonesonde measurements at each site. Instabilities of measurements are apparently observed from New Delhi ozonesondes. High 673 674 surface ozone concentrations at Trivandrum in summer isare believed to be caused by 675 measurement errors because low levels of the pollutants hashave been reported at this site under 676 these geolocation and meteorological effects (Lal et al. 2000). Besides Trivandrum, Naha could 677 be regarded as a background sites according to low surface ozone (Fig. 3) and its precursor 678 concentrations (Fig. 2) compared to neighboring stations, and previous studies (Oltmans et al., 679 2004; Liu et al., 2002). In the lower troposphere, high ozone concentrations are captured at 680 Pohang, Tsukuba, and Sapporo in the summer due to enhanced photochemical production of 681 ozone in daytime, whereas tropical sites, Naha, Hanoi, and Hong Kong show the ozone 682 enhancements in spring, mainly due to the biomass burning in Southeast Asia, with low ozone 683 concentrations in summer due to the Asian monsoon and in winter due to the tropical air 684 intrusion (Liu et al., 2002; Ogino et al., 2013). Singapore and Kuala Lumpur are supposed to 685 be severely polluted areas, but ozone pollution is not clearly captured over the seasons. HThis could might be explained by the morning observation time at these two stations carried on in 686

the morning. In addition, instabilities of Singapore measurements are noticeable, such
 asincluding 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 variabilityies of ozone over
 thewith pressure atin other seasons.

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696

# 695 2.3. Comparison Methodology

697 The GEMS ozone profile algorithm is applied to OMI BUV measurements in for 300-330 nm to 698 simulate GEMS ozone profile retrievals at coincident locations listed in Table 1. The coincidence 699 criteria between satellite and ozonesondes are:  $\pm 1.0^{\circ}$  in both longitude and latitude and  $\pm 12$  hours in 700 time, and then the closest pixel is selected. The Aura satellite carrying OMI crosses the equator always 701 at ~ 1:45 pm LT-Local Ttime (LT), and thereby thus OMI measurements are elosely collocated within 3 702 hours to ozonesonde soundings measured-in the afternoon (1-3 pm LS). Weekly-based sonde 703 measurements provide 48 ozone profiles at maximum for a year; the number of collocations is on 704 average 40 from 2004 October to 2008, but reduced to  $\sim$  20 recently due to the screened OMI 705 measurements affected by the "row anomaly" which is was initially detected at two rows in 2007, and 706 seriously spread to other rows with time since January 2009 (Schenkeveld et al., 2017). As fFrom July 707 2011 the row anomaly effect slowly extends up to  $\sim 50$  % of all rows. Correspondingly, the average 708 collocation distance increases from 57.5 km to 66.6 km before and after the occurrence of the row 709 anomaly. The impact of spatiotemporal variability on the comparison will be much reduced for GEMS 710 due to its higher spatiotemporal resolution (7 km × 8 km @ Seoul, hourly) against OMI (48 km × 13 711 km @ nadir in UV1, daily).

712 To increase the validation accuracy, the data screening is implemented to for both ozonesonde 713 observations and satellite retrievals according to Huang et al (2017). For ozonesonde observations, we 714 screen ozonesondes with balloon-bursting altitudes-pressures exceeding 200 hPa, gaps greater than 3 715 km, abnormally high concentration in the troposphere (> 80 DU), and low concentration in the 716 stratosphere (<100 DU). Among WOUDC sites, the Japanese and Indian datasets include a correction 717 factor which is derived to make a-better agreement between integrated ozonesonde columns and 718 correlated reference total ozone measurements as mentioned in Section 2.2; In Fig. 34, Japanese 719 ozonesondes are compared against GEMS simulations when a correction factor is applied or not to each 720 CI and ECC measurements, respectively. Morris et al. (2013) recommended to-restricting the 721 application of this correction factor to the stratospheric portion of the CI ozonesonde profiles due to 722 errors in the above-burst column ozone. Our comparison results illustrate that applying the correction 723 factor reduces the vertical fluctuation of mean biasesed in ozone profile differences with insignificant 724 impact on their standard deviations. Therefore we decide to apply this correction factor to the sonde 725 profiles if this factor ranges from 0.85 to 1.15. Because of a lack of retrieval sensitivity to ozone below 726 clouds and lower tropospheric ozone under extreme viewing condition, satellite retrievalsGEMS 727 simulations are limited to cloud fraction less than 0.5, SZAs less than 60°, and fitting RMS (i.e., root 728 mean square of fitting residuals relative to measurement errors) less than 3.

729 Due to the different units of ozone amount between satellites and ozonesondes, we convert 730 ozonesonde-measured partial pressure ozone values (mPa) to partial column ozone (DU) at the 24 731 retrieval grids heights of the satellite for the altitude range from surface to the balloon-bursting altitudes. 732 Ozonesonde measurements are obtained at a rate of a few seconds and then typically averaged into 733 altitude increments of 100 meters, whereas retrieved ozone profiles from nadir BUV satellite 734 measurements have much coarser vertical resolution of 10-14 km in the troposphere and 7-11 km in the 735 stratosphere, troposphere based on OMI retrievals. Consequently, satellite observations captures only 736 the smoothed structures of ozonesonde soundings, especially in-near the tropopause, where a sharp 737 vertical transition of ozone within 1 km is observed, and in the boundary layer due to the insufficient 738 penetration of photons. Satellite retrievals unavoidably have an error compound due to its limited 739 vertical resolution, which is named called "smoothing error" in the OE-based retrievals (Rodgers, 2000). 740 It could be useful to eliminate the effect of smoothing errors on differences between satellites and sondes 741 to better characterize other error sources in the comparisons (Liu et al., 2010a). For this reason, satellite 742 data have been compared to smoothed ozonesonde measurements smoothed into the satellite vertical 743 resolution-, together with original sonde soundings (Liu et al., 2010b; Bak et al., 2013b; Huang et al., 744 2017). The smoothing approach is: following as

745

 $\hat{7}46$  $\hat{x}_{sonde} = A \cdot x_{sonde} + \frac{x_a}{a}(1-A)x_a$ (3)747 $x_{sonde}$ : High-resolution ozonesonde profile(3)748 $\hat{x}_{sonde}$ : Convolved ozonesonde profile into satellite vertical resolution749A : Satellite averaging kernel750 $x_a$  : A priori ozone profile

751 752

In order to define tropospheric columns, both satellite retrievals and ozonesonde measurements

753 are vertically integrated from the surface to the tropopause taken from daily National Centers for 754 (FNL) Operational Environmental Prediction (NCEP) final Global analysis data 755 (http://rda.ucar.edu/datasets/ds083.2/). To account for the effect of surface height differences on 756 comparison, ozone amounts of from satellite data below the surface heights of ozonesondes is are added 757 to tropospheric columns of ozonesonde measurements and vice versa.

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# 759 **3. Results and Discussions**

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- 761 762

#### 3.1 Comparison at individual stations

763 Witte et al. (2018) recently compared seven SHADOZ station ozonesonde records, including 764 Hanoi and Kuala Lumpur in the GEMS domain, with total ozone and stratospheric ozone profiles 765 measured by space-borne nadir and limb viewing instruments, respectively. In this comparison, the 766 Hanoi station shows comparable or better agreement with the satellite datasets when compared to other 767 sites. Morris et al. (2013) and Rohtash et al. (2016) thoroughly evaluated ozonesonde datasets over 768 Japanese and Indian sites, respectively, but they did not address their measurement accuracy with 769 respect to those at other stations. Validation of GOME TOC by Liu et al. (2006) showed relatively larger 770 biases at Japanese CI stations and validation of OMI TOC by Huang et al. (2017) showed both larger 771 biases and standard deviations at the India MB-M sites. In South Korea, regular ozonesonde 772 measurements are taken only from Pohang, but these measurements have been insufficiently evaluated; 773 only the stratospheric parts of these measurements were quantitatively assessed against satellite solar 774 occultation measurements by Halogen Occultation Experiment (HALOE) from 1995 to 2004 in Hwang 775 et al. (2006), but only 26 pairs were compared despite its the coarse coincident criteria (48 hours in time, 776  $\pm 4.5^{\circ}$  in latitude,  $\pm 9^{\circ}$  in longitude). Therefore, it is important to perform the quality assessment of 777 ozonesonde measurements to identify the a reliable reference dataset for GEMS ozone profile validation

778 For this purpose, we illustrate tropospheric ozone columns (TOC) as a function of time and for 779 individual stations listed in Table 1, measured with three different types of ozonesonde instruments and 780 retrieved with GEMS simulations (Fig. 45), respectively. The goal of this comparison is to identify any 781 abnormal deviation of ozonesonde measurements relative to satellite retrievals, so we exclude the 782 impact of the different vertical resolutions between instruments and satellite retrievals on this 783 comparison by convolving ozonesonde data with satellite averaging kernels. At mid-latitude sites 784 (Pohang, Sapporo, and Tsukuba) both ozonesonde and satellite simulated retrievals show the distinct 785 seasonal TOC variations with values ranging from the amplitude of ~ 35 to -~ 40 DU. Extratropical sites 786 (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 <u>the</u> tropics. Both ozonesonde
 observations and <u>satellite simulated</u> 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.

- 792 In Fig. 5-6 time dependent errors in differences of TOC between ozonesonde and satellite simulated 793 GEMS retrievals are evaluated with the corresponding comparison statistics in Table 2. Satellite 794 retrievalsSimulated retrievals show strong correlation of  $\sim 0.8$  or much larger with ozonesonde 795 measurements at Pohang, Hong Kong, and three stations from Japan Japan stations, and with less 796 correlation of  $\sim 0.5$  at other SHADOZ stations in the tropics. However, Indian stations show poor 797 correlation of 0.24. Mean biases and its standard deviations are much smaller at stations where a strong 798 correlation is observed; they are  $\sim 1$  DU  $\pm \sim 4$ DU at most ECC stations, but deviated to  $\sim 4$  DU  $\pm \sim$ 799 10 DU at MB-M stations. In conclusion, we should exclude ozonesonde observations measured by MB-800 M to remove irregularities in a reference dataset for validating both GEMS simulated retrievals in this 801 study and GEMS actual retrievals in future study. Moreover, time series of ozonesonde and satellite 802 observations simulated retrievals show a significant transition at three Japanese stations as of late 2008 803 and early 2009 when the ozonesonde instruments was were switched from CI to ECC. This transition 804 could be affected by space-borne instrument degradation, but the impact of balloon-borne instrument 805 change on them is predominant based on a less time-dependent degradation pattern at latitudinally 806 neighboring stations during this period. CI ozonesondes noticeably underestimates atmospheric ozone 807 by 2-3 DU compared to ECC and thereby GEMS TOC biases relative to CI measurements, are are 808 estimated as - 2 to - 5 DU, but these biases are reduced to < 1.5 DU when compared with ECC. Therefore, 809 we decide to exclude these CI ozonesonde observations for evaluating GEMS simulated retrievals. 810 Compared to other ECC stations, Hanoi Sstation often changed sensing solution concentrations and pH 811 buffers (Table 1), which might <u>and hence might</u> cause the irregularities due to remaining errors even 812 though transfer functions were applied to ozonesonde measurements to account for errors due to the 813 different sensing solution (Witte et al., 2017). This fact might affect the relatively worse performance 814 compared to latitudually adjacenta -neighboring station, Hong Kong, where the 1.0 % KI buffered 815 sensing solution (SST1.0) 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

820 Japan, Pohang, Hong Kong, and Hanoi Sstations, where balloons were launched in afternoon (~12-15 821 LSTLT), are collocated within ~ 1-2 hours to of OMI-that passes the equator at 1301:45 pm LST and 822 then reaches the pole within 25 min, whereas the time discrepancy increases to 7 hours at Singapore, 823 where ozonesondes are launched in the early morning. Photochemical ozone concentrations are 824 typically denser in the afternoon than in the morning and hence ozonesonde measurements at Singapore 825 are negatively biased relative to afternoon satellite measurements. For the reason mentioned above, the 826 discrepancy in the observation time could also impact on affect this comparison at Kuala LumpurKuala 827 Lump, where sondes were mostly launched in the late morning, 2-3 hours prior to the OMI passing time 828 and thereby ozonesonde measurements tend to be negatively biased. These indicate that diurnal 829 variations of the tropospheric ozone are visible in ozonesondeoznesonde measurements, emphasizing 830 on the utility of hourly geostationary ozone measurements. The comparison results could be 831 characterized with latitudes. In the mid-latitudes (Pohang, Tsukuba, and Sapporo), noticeable 832 disagreements are commonly addressed seen in the tropopause region where mean biases/standard 833 deviations are  $\sim 10$  %/ $\sim 15$ % larger than those in the lower troposphere. In the extra-tropics (Hong Kong, 834 Naha), consistent differences of -a few %-percent are shown-seen over the entire altitude range with 835 standard deviations of 15 % or less below the tropopause ( $\sim$  15 km). Hanoi and Kuala Lumpur show 836 significantly larger biases/standard deviations compared to other ECC stations. At Hanoi 837 inconsistencies of solution concentrations and pH buffers might influence on this instability. At Kuala 838 Lumpur the inconsistencies of observation times might be one of the reasons, considering its standard 839 deviations of  $\sim 100$  min, but mostly less than 30 min at other stations. Therefore, we strictly screen out 840 Singapore, Kuala Lumpur, and Hanoi, together with all M-BM measurements at Indian stations and CI 841 measurements at Japanese stations to improve the validation accuracy of GEMS simulated retrievals in 842 next section. Thus Eventually, stations, where the standard procedures for preparing and operating ECC 843 sondes are consistently maintained, are accepted adopted as an optimal reference in for this work.

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# 845 4<u>3</u>.2 Evaluation of GEMS simulated ozone profile retrievals

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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  $\frac{1}{n} \sqrt{\sum_{i}^{n} (y_{GEMS} - y_{fit})^{2}}$ ,  $y_{fit} = a \cdot y_{sonde} + b$ . In Fig. 78,

B52 GEMS simulated retrievals are plotted as a-functions of ozonesondes with and without the vertical

853 resolution smoothing, respectively, for the stratospheric and tropospheric columns. GEMS simulations 854 underestimate the tropospheric ozone by  $\sim 2.27 \pm 5.94$  DU and overestimate the stratospheric ozone 855 by  $\sim 9.35 \pm 8.07$  DU relative to high-resolution ozonesonde observations. This comparison 856 demonstrates a-good correlation coefficients of 0.84 and 0.99 for troposphere and stratosphere, 857 respectively. This agreement is degraded if the rejected ECC sondes (Kuala Lumpur, Hanoi, and 858 Singapore) are included; for example, the slope decreases from 0.68 to 0.64 while the RMSE increases 859 6.35 and 6.76 DU for TOC comparison. Smoothing ozonesonde soundings into-to GEMS vertical 860 resolution improves the comparison results, especially for the tropospheric ozone columns; standard 861 deviations are reduced by  $\sim 5$  % with mean biases of less than 1 DU. Similar assessments are performed 862 for OMI standard ozone profiles based on the KNMI OE algorithm (Kroon et al., 2011) hereafter 863 referred to as OMO3PR (KNMI) in Fig. 8-9 and the research product based on the SAO algorithm (Liu 864 et al., 2010) hereafter referred to as OMPROFOZ (SAO) in Fig. 910, respectively. It implies that GEMS 865 gives the good information on Stratospheric Ozone Columns (SOCs)sSOCs comparable to both the 866 OMI KNMI and SAO products in spite of excluding insufficient information on most of Hartley ozone 867 band absorption in GEMS in GEMS retrievals. Furthermore, a better agreement of GEMS TOCs with 868 ozonesonde is found than with the others due to different implementation details. As mentioned in 2.1., 869 the GEMS algorithm is developed based on the heritages of the SAO ozone profile algorithm with 870 several modifications. There are two main modifications are: (1) a priori ozone climatology was 871 replaced with a tropopause-based ozone profile climatology to better represent the ozone variability in 872 the tropopause (2)- ilradiance spectra used to normalize radiance spectra and characterize instrument 873 line shapes are prepared by taking 31-day moving average instead of climatological average to take into 874 account for time-dependent instrument degradations. These modifications reduce somewhat the spreads 875 in deviations of satellite retrievals from sondes, especieally in TCO-TOC comparison. KNMI retrievals 876 systematically overestimate the tropospheric ozone by ~ 6 DU (Fig. 910.c), which corresponds to the 877 positive biases of 2-4 % in the integrated total columns of KNMI profiles relative to Brewer 878 observations (Bak et al., 2015). As mentioned in Bak et al. (2015), the systematic biases in ozone 879 retrievals are less visible in SAO-based retrievals (simulated GEMS -datasimulation, OMPROFOZ), 880 as systematic components of measured spectra are taken into account for using an empirical correction 881 called "soft calibration".

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### 883 4. Summary

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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 887 of the algorithm against coincident ozonesonde observations. There are 10 ozonesonde sites over the 888 GEMS domain from WOUDC, SHADOZ and KMA archives. This paper gives an overview of these 889 ozonesonde observation systems to address inhomogeneities in preparation, operation, and correction 890 procedures which cause discontinuities in individual long-term records or in-among adjoint stations. 891 Comparisons between simulated GEMS TOCs retrievals and ozonesondes illustrate a noticeable 892 dependence on the instrument type. Indian ozonesonde soundings measured by MB-M show severe 893 deviations in seasonal time series of TOC compared to coherent GEMS simulations and neighboring 894 ozonesonde observations measured in similar latitude regimes. At Japanese stations, CI ozonesondes 895 underestimate ECC ozonesondes by 2 DU or more and a better agreement with GEMS simulations is 896 found when ECC measurements are compared. Therefore, only ECC ozonesonde measurements are 897 first selected as a reference, in order to ensure a consistent, homogeneous dataset. Furthermore, ECC 898 measurements at Singapore, Kuala Lumpur, and Hanoi are excluded. At Singapore and Kuala Lumpur, 899 observations were performed in the morning and thereby are inconsistent with GEMS retrievals 900 simulated at the OMI overpass time in the afternoon. In addition, the observation time for Kuala Lumpur 901 is inconsistent itself compared to other stations; its standard deviation is  $\sim 100$  min, but for other ECC 902 stations it is less than 30 min. At Hanoi the combinations of sensing solution concentrations and pH 903 buffers changed 4 times during the period of 2005 through 2015. Therefore, GEMS and 904 ozonsondeozonesonde comparisons show larger biases/standard deviations at these stations. Pohang 905 station is unique in South Korea where ECC ozonesondes have been regularly and consistently launched 906 without a gap since 1995; the standard 1% KI full buffered sensing solution has been consistently 907 applied to ozone sensors manufactured by SPC (6A model). Evaluation of Pohang ozonesondes against 908 GEMS simulations demonstrates its high level reliability, which is comparable to latitudually 909 adjacentneighboring Japanese ECC measurements at Tsukuba and Sapporo. Reasonable agreement with 910 GEMS simulated retrievals is similarly shown at Latitudually adjacent Naha and Hong Kong stations. 911 Finally, we establish that the comparison statistics of GEMS simulated retrievals and optimal reference 912 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 913 914 precision is comparable to OMI products for the stratospheric ozone column and even better for the 915 tropospheric ozone column due to improved algorithm implementations. Our future study aims to 916 achieve this quality level from actual GEMS ozone profile product.

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guidance; JB conducted the research and wrote the paper; XL and KC contributed to the

<sup>918 &</sup>lt;u>*Author contributions.*</u> JB and KHB designed the research; JHK and JK provided oversight and

### 920 <u>analysis and writing.</u>

935

# 921 *<u>Competing interests.</u>* The authors declare that they have no conflict of interest.

922 Data availability, The ozonesonde data used in this study were obtained though the WOUDC, 923 archives. The WOUDC SHADOZ and KMA dataset are available at 924 https://woudc.org/data/products/ozonesonde/(last access:T07 July 2019) and for the SHADOZ dataset 925 at https://tropo.gsfc.nasa.gov/shadoz/Archive.html (last access:07 July 2019). The KMA dataset is 926 available through data request at https://www.data.go.kr/. -OMI Level1b dataset are available at 927 https://aura.gesdisc.eosdis.nasa.gov/data///Aura\_OMI\_Level1/ (last access:07 July 2019). 928

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Figure 1. Flow <u>c</u>Chart 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<sub>2</sub> 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 tofrom standard deviations to fit in the given x-axis.



**Figure 34.** Effects of applying a correction factor <u>(CF)</u> to (a) ECC and (b) CI ozonesonde measurements, respectively, on comparisons with simulated GEMS ozone profile retrievals. <u>Solid and</u> 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 56. Same as Figure 45, but for absolute differences of tropospheric ozone columns (DU) between ozonesonde measurements and GEMS simulated retrievals.



**Figure 67**. Mean biases and  $1\sigma\sigma$  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)  $X \ge 100 \% / (aA priori)$ .



**Figure 78**. Upper: Scatter plots of GEMS vs. ozonesonde for tropospheric and stratospheric ozone columns, respectively. <u>The l</u>Lower 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 1 $\sigma$  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. <u>78</u>, but for validating OMI standard ozone profiles (OMO3PR) produced by the KNMI optimal estimation <u>OE</u>-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.\_

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Station <sup>a</sup>	Lon (°), Lat (°)	Altitude (m)	Observation Time <sup>b</sup>		Instrument Type <sup>c</sup>	ECC-SST <sup>d</sup>	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	
			07.30-08.00 (9)	Nov15 - Dec15	ECC/SPC 6A	5510.5		
Kuala <u>L</u> łumpur	101.7, 2.7	20	9:30-15:00 (104)	Jan 13 - Dec14	ECC/SPC 6A	SST1.0	- Transfer function	
			7.50-15.00 (104)	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	
				Jan 05 - Apr 06	ECC/EN-SCI 1Z	SST2.0		
				Apr06 - Dec 07	ECC/EN-SCI 2Z	SST2.0		
				Jan 08 - May 09	ECC/EN-SCI 2Z	SST1.0	—	
Hanoi	105.8, 21.0	10	12:00-14:00 (42)	Jun 09 - Dec 09	ECC/SPC 6A	SST1.0	Transfer function	
				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	
			14:50-15:00 (00)	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		<ul> <li>Correction factor</li> </ul>	
i sukuda				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		<ul> <li>Correction factor</li> </ul>	
				Dec 09 - Dec 15	ECC/EN-SCI 1Z	SST0.5		

Table1. List of ozonesonde stations.

<sup>a</sup> Data are downloaded from the WOUDC (http://woudc.org) data archive, except for Kuala Lumpur and Hanoi, which are from the SHADOZ

(https://tropo.gsfc.nasa.gov/shadoz/) network, and Pohang, which are from the Korea Meteorological Administration (KMA).

<sup>b</sup> The range of the observation time (LT) with 1  $\sigma$  standard deviations of them (min) in the parentheses.

<sup>c</sup> 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).

<sup>d</sup> Potassium Iodide (KI) cathode sensing solution type (SST) implemented in ECC ozone sensors: SST0.5 (0.5 % KI, half buffer), SST1.0 (1.0 % KI, full buffer), and SST 2.0 (2.0 % KI, no buffer). Singapore station changed it to SST1.0 as of 2018.

<b>Table 2</b> . Comparison Statistics statistics (Mean-mean Bias-bias in DU, $1\sigma s$ Standard standard Deviation deviation in DU, and <u>R</u> , Correlation correlation
Coefficient of the coefficient o
averaging kernels.

Station	Collocation	Туре	Data Period	SONDE AK – GEMS		
Station	Time difference		(Year)	#	Mean Bias + $1\sigma$	R
Singapore	6:44	ECC	12-15	20	$-13.67 \pm 9.61$	0.17
Kuala <u>L</u> łump <u>ur</u>	2:29	ECC	05-15	106	$-2.54 \pm 4.13$	0.44
Trivandrum	1:46	MB-M	06-11	37	$3.55\pm9.75$	0.24
Hanoi	0:32	ECC	05-15	100	$-3.82\pm6.03$	0.52
Hong Kong	0:27	ECC	05-15	259	$-1.19 \pm 3.91$	0.82
Naha	0.47	CI	05-08	135	$-5.48 \pm 4.07$	0.85
Inalia	0:47	ECC	08-15	166	$-0.94 \pm 3.22$	0.91
New Delhi	1:46	MB-M	06-11	39	$-4.57 \pm 13.36$	0.24
Pohang	0:54	ECC	05-15	281	$-0.75 \pm 3.13$	0.95
Taulauha	1.56	CI	05-09	151	$-2.98\pm3.76$	0.91
Tsukuba	1:56	ECC	09-15	154	$-0.65 \pm 3.53$	0.94
Connoro	2.19	CI	05-09	107	$-3.43 \pm 2.56$	0.94
Sapporo	2:18	ECC	09-15	95	$-1.37 \pm 2.79$	0.93