

1 **Response to referee comments on “Spatial distribution and temporal trend of ozone pollution**
2 **in China observed with the OMI satellite instrument, 2005–2017”**

3

4 We thank the referees for their careful reading of the manuscript and the valuable comments. This
5 document is organized as follows: the Referee’s comments are in *italic*, our responses are in plain
6 text, and all the revisions in the manuscript are shown in blue. The line numbers in this document
7 refer to the updated manuscript.

8

9 **Referee #1**

10
11 *This paper seeks to quantify surface ozone across China using the SAO OMI tropospheric column*
12 *ozone product. While I appreciate this effort to quantify such a relationship, the current analysis has*
13 *not demonstrated a clear and convincing link between the lower/mid-tropospheric OMI retrievals*
14 *and day-to-day ozone variability at the surface. (1) I realize that the authors are trying to find some*
15 *signal in OMI that reflects ozone at the surface, but the degrees of freedom are so small, and the*
16 *sensitivity to surface ozone is so weak, that there's no real way to distinguish between the signal that*
17 *comes from the surface and that which comes from 800 or 700 hPa. (2) As presented, the*
18 *relationship is more likely due to weather pattern variability causing ozone at the surface and in the*
19 *free troposphere to vary in tandem. (3) Far more work is required, including a thorough evaluation*
20 *of the OMI product against extensive IAGOS aircraft observations across mainland China, South*
21 *Korea, Taiwan and Hong Kong. The additional analysis required to convince me that OMI can*
22 *provide a meaningful evaluation of surface ozone across China goes beyond a standard major*
23 *revision. My recommendation to the editor is that the paper be rejected to allow the authors*
24 *adequate time to conduct additional product evaluation. If the expanded analysis can indeed*
25 *demonstrate sensitivity of OMI to surface ozone then the authors will have the basis for a new*
26 *manuscript which will make a valuable contribution to ozone monitoring across East Asia.*

27
28 **Response.** Thanks for the careful reading of our manuscript and raising so many good points. The
29 feedback has significantly improved our work. We also wish to draw the reviewer's attention to that
30 we have revised the title, discussed the limitations of this work and also validated the OMI inferred
31 ozone trends with surface observations (Figure 6).

32 [New title. Ability of the OMI satellite instrument to observe surface ozone pollution in China:](#)
33 [application to 2005-2017 ozone trends.](#)

34
35 Since this is a long comment, we decompose it into three parts and answer each part one by one.

36
37 (1) Yes, the reviewer made a very good point here that the OMI cannot distinguish the signal that
38 comes from the surface and from the lower troposphere (e.g. 800 and 700 hPa), given the relatively
39 low vertical resolution of the retrievals. We have made these changes in the text to reduce the
40 confusion.

41
42 [P5 L18. The correlation of OMI with the MEE surface ozone data likely does not reflect a direct sensitivity of](#)
43 [OMI to surface ozone, which is very weak, but rather a sensitivity to boundary layer ozone extending up to a](#)
44 [certain depth and correlated with surface ozone.](#)

45
46 The DOFS depends on what error is assumed in the prior estimate. Since the prior from McPeters et
47 al. (2007) has low boundary layer ozone with low associated error then the DOFS would
48 underestimate the ability to observe the polluted boundary layer. Now we say this.

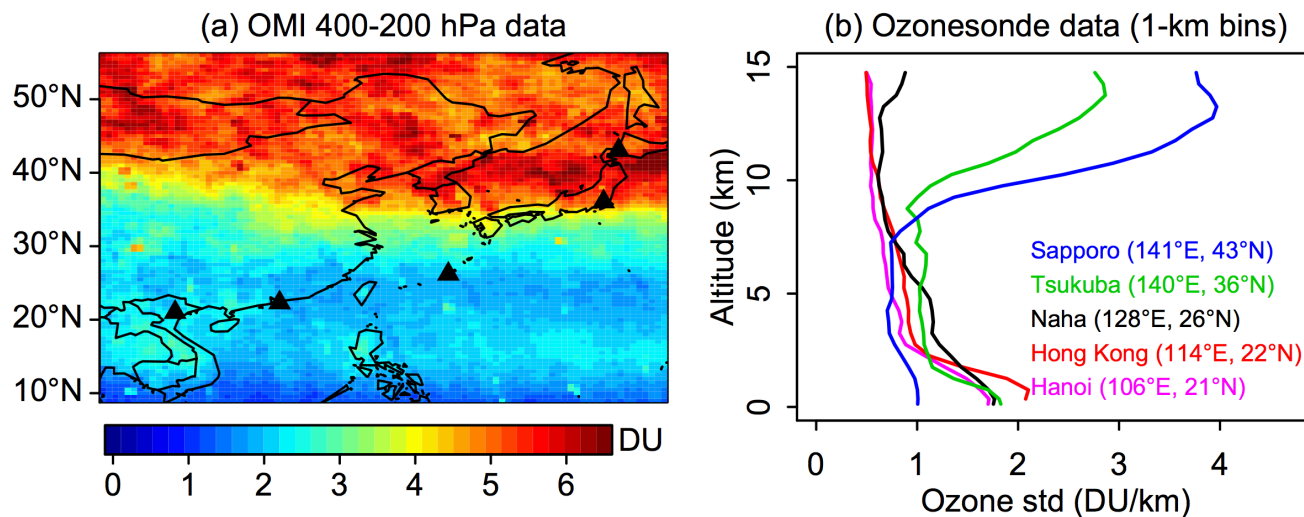
49 [P3L19. Even though a DOFS of 0.3 is still low, it is based on the prior estimate of low boundary layer ozone](#)
50 [in the McPeters et al. \(2007\) zonal mean climatology.](#)

51

52 Also the ozone sonde observations show that the ozone variability in the boundary layer is 80-100%
 53 higher than in the free troposphere. In more polluted regions like in mega city clusters, this difference
 54 should be even larger. These results indicate that the boundary layer ozone is more likely to drive the
 55 daily variability of OMI 850-400 hPa retrievals in regions like South China. We have added the
 56 following figure.

57

Standard deviation of ozone concentrations, summers 2005-2017



58 **Figure 4.** (a) Standard deviation of daily OMI 400-200 hPa ozone in East Asia during 2005-2017
 59 summers. The triangles are the locations of ozonesonde sites with observations during this period. (b)
 60 Vertical profiles of daily ozone standard deviation in 1-km bins (DU/km) in the ozonesonde data for
 61 the 2005-2017 summers.

62

63
 64 P6 L26. We find that the low correlation of OMI with boundary layer ozone in the northern
 65 ozonesonde data is due not only to the low DOFS but also to a large variability of ozone in the
 66 upper troposphere. Figure 4 (left panel) shows the standard deviation of daily OMI 400-200 hPa
 67 ozone during 2005-2017 summers, indicating that upper tropospheric ozone has much higher
 68 variability in the north ($> 34^{\circ}\text{N}$) than in the south. This is related to the location of the jet stream
 69 and more active stratospheric influence (Hayashida et al., 2015). Figure 4 (right panel) displays the
 70 vertical profiles of ozone standard deviations for the five ozonesonde sites. For the two sites north
 71 of 34°N , the ozone variability becomes very large above 8 km. Since the OMI 850-400 hPa
 72 retrieval also contains information from above 400 hPa, this upper tropospheric variability causes a
 73 large amount of noise that masks the signal from boundary layer variability. For the three sites
 74 south of 34°N , the ozone variability in the boundary layer is much higher than in the free
 75 troposphere and the upper tropospheric ozone variability still remains low even above 8 km. In the
 76 rest of this paper we focus our attention on ozone episodes and the long-term trends in southern
 77 China (south of 34°N).

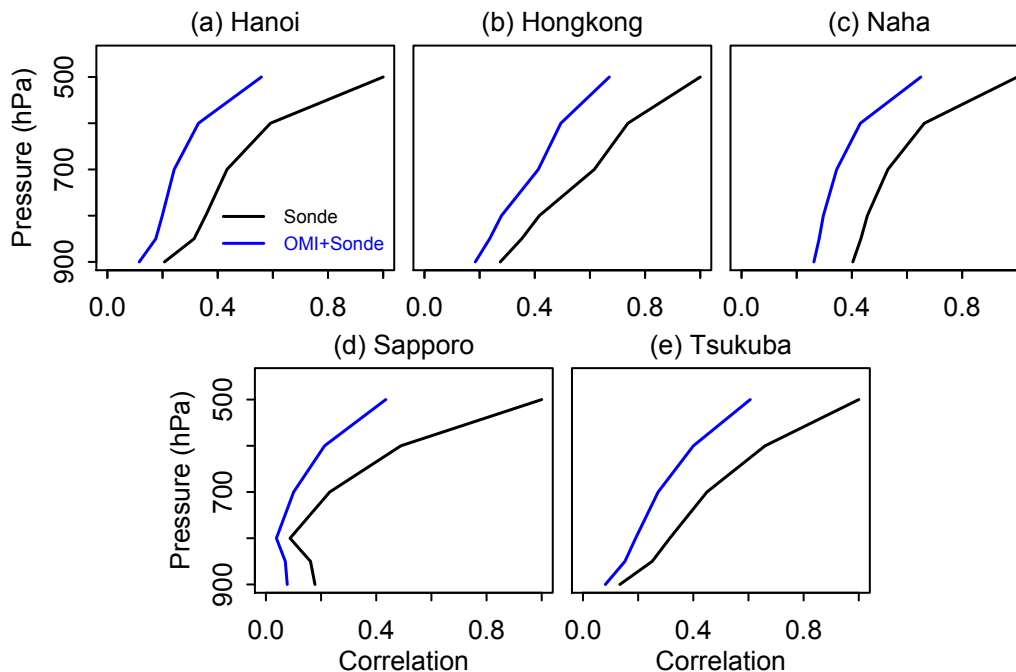
78

79 (2) We thank the reviewer for pointing out this issue. But based on our analysis, the weather
 80 patterns are insufficient to explain the observed relationship. We have conducted a sensitivity
 81 experiment and made these changes in the text.

82 P 6 L15. The correlation between boundary layer ozone pollution and the OMI ozone retrievals
 83 could be due in part to correlation between boundary layer and mid-tropospheric ozone, considering
 84 that both tend to be driven by the same weather systems. We used the ozonesonde data to examine
 85 what correlation with boundary layer (950-850 hPa) ozone would be observed if OMI were
 86 sensitive only to the free troposphere at ~500 hPa (where its sensitivity is maximum, Figure 3c) and
 87 not to the boundary layer. In that case the correlation coefficient $R_{1,3}$ of boundary layer ozone and
 88 the OMI 850-400 hPa retrievals would be given by (Vos, 2009):

$$89 \quad R_{1,3} = R_{1,2}R_{2,3} \pm \sqrt{(1 - R_{1,2}^2)(1 - R_{2,3}^2)} \quad (2)$$

90 where $R_{1,2}$ is the correlation coefficient between boundary layer and 500 hPa ozone in the
 91 ozonesonde data, and $R_{2,3}$ is that between 500 hPa ozone and the OMI 850-400 hPa retrievals. As
 92 seen from Figure S4, $R_{1,3}$ at the five sonde sites is only ~0.2, implying that direct sensitivity to the
 93 boundary layer dominates the correlation of OMI with surface ozone at least in southern China.
 94 Further evidence for this is the ability of OMI to detect the ozone enhancements in megacity
 95 clusters (Figure 1).

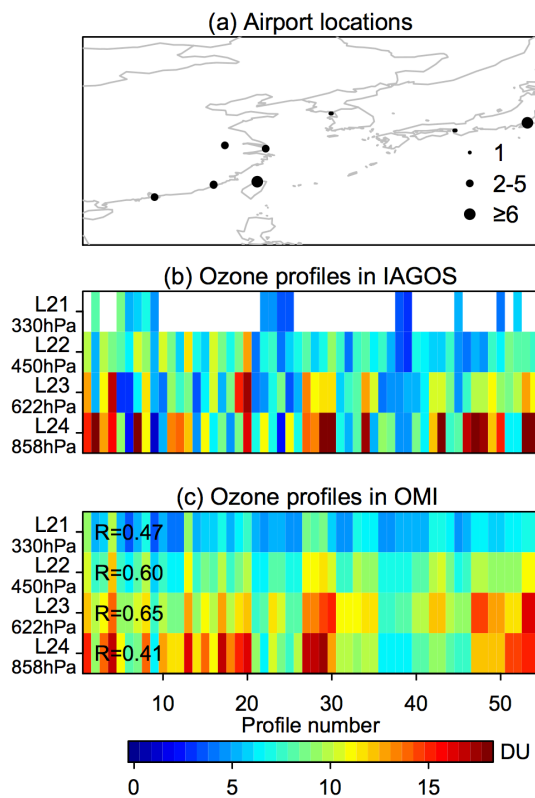


96
 97 **Figure S4.** Correlation of OMI 850-400 hPa ozone and the boundary layer ozone assuming that the
 98 OMI were sensitive only to the free troposphere at ~500 hPa (where its sensitivity is maximum,
 99 Figure 3c) and not to the boundary layer. The black line is the correlation of ozone at different
 100 pressure levels with 500 hPa ozone in the sonde observations. The blue line is the estimated
 101 correlation of OMI 850-400 hPa with ozone at different layers if the satellite can only detect the
 102 signal at 500 hPa but not from other layers, as calculated using Equation 2. See text for more details.
 103

104 (3) We have processed all IAGOS aircraft observations in East Asia during 2005-2017 summers
 105 and we only find 54 profiles in 8 airports that can be used to validate the OMI. Given so few
 106 profiles, it is unlikely to evaluate the long-term correlation of surface and mid-tropospheric ozone
 107 at each airport. But combining all profiles together, the temporal correlation coefficients of the 950
 108 hPa ozone and 850-400 hPa OMI ozone is 0.59. These results are also consistent with what we find
 109 in the Hong Kong sonde site (Figure 2). We have made these changes.

110
 111 P6 L9. We applied the same daily correlation analysis to the other ozonesonde datasets and IAGOS
 112 aircraft measurements during 2005-2017 summers. For the 54 IAGOS vertical profiles coincident
 113 with OMI observations, the correlation coefficient of the 950 hPa in situ ozone and 850-400 hPa
 114 OMI ozone is $R = 0.59$ ($p < 0.05$) (Figure S2).

Ozone profiles, IAGOS vs. OMI, JJA 2005-2017



115
 116 **Figure S2.** (a) Location and the number of tropospheric profiles at each airport in IAGOS that are
 117 coincident with OMI retrievals. We only select these profiles between 12-15 local time. (b) Ozone
 118 profiles from IAGOS, but mapped to OMI layers. The missing data at L21 is in part because we
 119 only select pixels that are within 200 km from the airport on the flight path. (c) OMI ozone profiles
 120 coincident with the ozonesondes. The correlations of unsmoothed 950 hPa ozone data in IAGOS
 121 with the OMI retrievals for different levels are shown inset. The correlation with 850-400 hPa OMI
 122 ozone is 0.59 ($p < 0.05$)

123
 124 *Further comments:*

125
 126 *When I read the title and abstract I was under the impression that the authors had made a new*

127 *breakthrough regarding the detection of surface ozone using OMI. It seemed like the instrument*
128 *could actually detect ozone at the surface and the detection was so good that daily ozone variability*
129 *at any given surface site could be determined with a precision of +/- 10.7 ppb. But when I read the*
130 *full paper I learned that this is not the case.*

131
132 *The premise that lower/mid-tropospheric OMI ozone retrievals are closely associated with surface*
133 *ozone is not shown in a convincing manner. The initial correlation of: $[O_3] = 8.9 \Delta\Omega + 15.8 \pm 10.7$*
134 *appears to be driven entirely by the latitudinal gradient of ozone at the surface and in the mid-*
135 *troposphere. Just because surface and OMI ozone have similar latitudinal gradients, when averaged*
136 *over several years, does not mean that the mid- to lower troposphere can tell us how ozone varies at*
137 *the surface from day to day. A better test of the relationship is to focus on a narrow latitude range.*
138 *This is done in Figure 3, for daily observations, where we can see that the correlation is very low*
139 *above five urban regions. For the region of Beijing the correlation is only $R=0.27$ which corresponds*
140 *to an r -squared value of 0.07, which means that the variation in OMI only explains 7% of the ozone*
141 *variability at the surface. The best case is made by the Wuhan region, but even here $R=0.53$, which*
142 *means OMI only explains 28% of the surface ozone variability. Figure 3 shows that OMI is only*
143 *weakly correlated with surface ozone and provides no convincing argument that the retrieval is*
144 *sensitive to surface ozone. The weak correlation is probably just due to weather patterns causing*
145 *surface and lower to mid- tropospheric ozone to vary in tandem.*

146
147 **Response.** Yes, the reviewer has made a very good point here that the satellite has a lot of noise. This
148 is a common problem when we use satellite data. But noisy data still contains information, and we
149 can reduce the noise by either temporally averaging the data over multiple years, or training the
150 model with a lot of data together. In our study, we tried both ways. When we average the data over a
151 five-year period, the OMI 850-400 hPa ozone displays a strong correlation ($R=0.73$) with the surface
152 observations. When we fit all the daily data in eastern China together, we find the extreme value
153 model can well simulate the distribution of high ozone concentrations. We also find the resulting
154 model can accurately estimate the probability of higher thresholds, which is a strong signal that our
155 extreme value model is well fitted.

156
157 The reviewer is correct that our old manuscript indeed overpromised the value of OMI data,
158 especially in the northern China. Due to the stronger jet wind activities and more active stratosphere-
159 troposphere exchange in the north, the upper tropospheric ozone there has much higher daily
160 variability, making OMI 850-400 hPa ozone less reliable in predicting the daily episode and inferring
161 the long-term trends. The strong jet wind also means these regions are very sensitive to global
162 background. At the same time, OMI has relatively low sensitivity in the north. So in our revised
163 manuscript, we don't fit the extreme value model or predict the trends in the north. We have added a
164 [new figure 4](#) and more discussions at [P6L26-P7L6](#). Based on reviewer's suggestion, we have
165 changed the title (see new title) and also discussed the limitations of this work (see many blue
166 highlighted text in the revised manuscript).

167
168 But the OMI data should still be useful in south China for these reasons. First, OMI has higher
169 sensitivity in the South, and the daily correlation of surface ozone and OMI 850-400 retrievals is

170 statistically significant for most sites (Figure 1). Second, the upper tropospheric ozone variability is
 171 much smaller and the boundary ozone variability can be a strong modulator of OMI 850-400 hPa
 172 ozone variability (new figure 4). Third, the inferred long-term trends of surface ozone from OMI
 173 fairly agree with these from TOAR sites (Figure 6).

174
 175 We also make it clear we have removed the latitude-dependent background.
 176 P3 L23. To remove this gradient and also any long-term uniform drift in the data, we subtract the monthly
 177 mean Pacific background (150°E-150°W) for the corresponding latitude and month

178 P4 L22. After subtracting the North Pacific background for the corresponding latitude in month, we obtain the
 179 OMI ozone enhancements shown in Figure 1d.

180 P4 L23. The spatial correlation coefficient between the OMI ozone enhancements and the MEE surface
 181 network is $R = 0.73$ over eastern China. The correlation is driven in part by the latitudinal gradient but also by
 182 the enhancements in the large megacity clusters identified as rectangles in Figure 1b. Thus the correlation
 183 coefficient is $R = 0.55$ for the 26-34°N latitude band including YRD, SCB, and Wuhan.

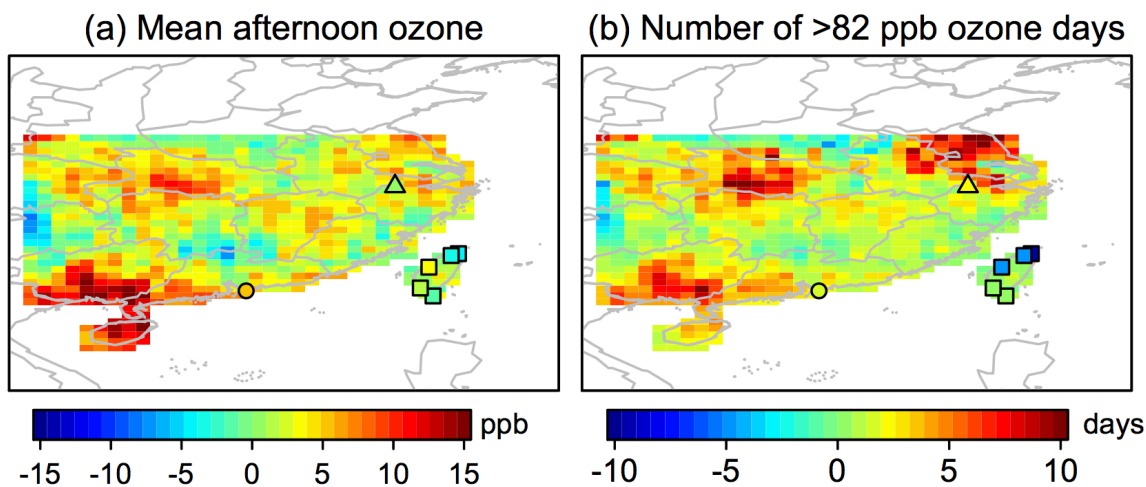
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 185
 186 *In a related comment, do the authors think that any correlation between ozone at the surface and*
 187 *ozone in the lower/mid troposphere is linked because of similar photo- chemical processes, or is the*
 188 *correlation just a coincidence due to meteorology? For example we know that in southern China the*
 189 *ozone at the surface varies strongly with the strength of the summertime Asian monsoon. When*
 190 *transport is from the south then the relatively clean air masses from the tropical Pacific bring air*
 191 *that is low in ozone, both at the surface and in the lower-mid troposphere. But when the monsoon*
 192 *winds weaken, mid-latitude air is allowed to move back into the region of southern China, bringing*
 193 *higher ozone to the lower and mid-troposphere. At the same time, the flow of clean air from the south*
 194 *also ceases at the surface, allowing ozone to build up in the polluted air masses from mainland*
 195 *China. Under this scenario ozone in the mid- troposphere is correlated with ozone at the surface*
 196 *even if the two layers are isolated from each other by strong temperature inversions.*

197
 198 **Response.** Thanks for pointing out this issue. But meteorology is insufficient to explain the observed
 199 correlation of surface ozone and OMI 850-400 hPa ozone. Please check L75-100 of this response
 200 letter for more details (or P5 L15-25 in the main text).

201
 202
 203 *OMI ozone could be compared to long-term ozone monitoring sites in rural areas which would be a*
 204 *better comparison than the urban data from the new Chinese monitoring network. It would be very*
 205 *helpful to see time series of daily OMI values (when avail- able) and corresponding surface*
 206 *observations from the following sites: Mt Tai – data can be obtained from Prof. Likun Xue, Shandong*
 207 *University [Sun et al., 2016] Hok Tsui – located on the south coast of Hong Kong, data can be*
 208 *obtained from Prof. Tao Wang, Hong Kong Polytechnic [Wang et al., 2017] Shangdianzi – see Ma et*
 209 *al., 2016 LongFengShan – located in northeastern China. Contact Dr. Xiaobin Xu at the China*
 210 *Meteorological Administration: xiaobin_xu@189.cn LinAn – Near Shanghai, Contact Dr. Xiaobin*
 211 *Xu at the China Meteorological Administration: xiaobin_xu@189.cn Xi- angGeLiLa – in south*
 212 *central China, Contact Dr. Xiaobin Xu at the China Meteorologi- cal Administration:*
 213 *xiaobin_xu@189.cn*

214
 215 **Response.** We have obtained the Hong Kong site observations from Prof. Tao Wang from the above
 216 list. And we have also used the TOAR dataset to validate our model. We find that the OMI inferred
 217 ozone trends are fairly consistent with these long term surface records.
 218 P1 L21. Comparison of 2005-2009 and 2013-2017 OMI data indicates that mean summer afternoon surface
 219 ozone in southern China (including urban and rural regions) has increased by 3.5 ppb over the 8-year period
 220 and the number of episode days per summer has increased by 2.2 (as diagnosed by an extreme value model),
 221 fairly consistent with the few long-term surface records.
 222 P9 L6. We compared the OMI trends in Figure 6 to the trends of MDA8 ozone and number of high-ozone
 223 days reported by the long-term TOAR sites (Schultz et al., 2018) and our own analysis for the Hok Tsui
 224 station in Hong Kong (Wang et al., 2009). For Lin'an, Hong Kong, and the 5 sites in Taiwan, the changes of
 225 mean ozone concentrations from 2005-2009 to 2013-2017 are 1.1, 2.3, and -0.18 ± 2.2 ppbv (standard deviation
 226 among the 5 sites) as estimated from OMI, compared to 0.7, 5.6 (or 5.8 in Hok Tsui station), and -0.75 ± 3.4
 227 ppbv for MDA8 ozone at the TOAR sites. The changes in the number of ozone episodes per summer are 1.2,
 228 1.9, and -0.17 ± 0.74 days in OMI, compared to 2.1, 1.8 (or 2.1 in Hok Tsui station), and -3.5 ± 3.9 days at the
 229 TOAR sites. These OMI inferred trends are fairly consistent with the long-term records available from surface
 230 sites.

Changes in summertime surface ozone pollution inferred from OMI
 (2005-2009 to 2013-2017)



232 **Figure 6.** Changes in surface ozone pollution in China between 2005-2009 and 2013-2017 as
 233 inferred from OMI afternoon observations at around 13:30 local time. (a) Change in mean summer
 234 afternoon concentrations, obtained from the difference in the mean OMI enhancements at 850-400
 235 hPa and applying equation (1). Also shown with symbols are observed changes in mean MDA8
 236 ozone from in situ observations in Lin'an, Hong Kong, and Taiwan reported by TOAR (Schultz et
 237 al., 2018). Because the TOAR observations are only reported for 2005-2014, we estimate the
 238 changes from 2005-2009 to 2013-2017 on the basis of the reported linear trends during 2005-2014
 239 (ppb a^{-1}). The change of 12-15 LT ozone at the Hok Tsui station in Hong Kong is 5.8 ppb. (b)
 240 Change in the number of high-ozone days (> 82 ppb) per summer, calculated by applying the
 241 probability of exceeding 82 ppb (equation 8) to the daily OMI enhancements. Also shown with
 242 symbols are observed changes of the number of days with MDA8 ozone exceeding 80 ppb at the
 243 TOAR sites, similarly adjusted as the change from 2005-2009 to 2013-2017. The change in the

244 number of days with 12-15 LT ozone exceeding 82 ppbv at the Hok Tsui station in Hong Kong is
245 2.1 days.

246
247 *Ozone at the surface and in the mid-troposphere varies greatly with transport path- way and abrupt*
248 *changes in air masses, and recent studies have shown that ozone in China varies with meteorology*
249 *[Pu et al., 2017; Zhao et al., 2018]. The authors are aware of this phenomenon as their previous*
250 *work has explored the impact of climate variability on ozone. Therefore I'm surprised that the*
251 *authors didn't first explore how surface ozone across China varies with meteorology, such as surface*
252 *temperatures (or temperature at 850 hpa) [Pusede et al., 2015], or with the height of the 500 hPa*
253 *surface [Reddy et al., 2016], both of which correlate quite well with surface ozone. The authors*
254 *should first determine the correlation between surface ozone and meteorology, and then compare*
255 *these results to what they find from OMI ozone. Does OMI give more information on surface ozone*
256 *than basic meteorology? Given that reanalysis data are available for all of China under all weather*
257 *conditions (no cloud screening) I would think that the meteorology would perform better than OMI.*
258 *If OMI performs less well than meteorology, is there any reason to use OMI to try to predict surface*
259 *ozone, when meteorological analyses are available everywhere and at all times?*

260
261 **Response.** Our focus is to evaluate the OMI observation capability, not to analyze the correlation of
262 ozone with meteorological variables which has been done before and would not capture the
263 variability in ozone driven by emissions. The reviewer makes a good point that mid-tropospheric and
264 surface ozone may respond similarly to meteorological conditions, which may in turn contribute to
265 the correlation of OMI with surface ozone. We now address this point in Section 4 by analysis of the
266 ozonesonde data (P6 L15-25 with new Figure S4). We also mention this in other parts of the
267 manuscript.

268 P5L18. The correlation of OMI with the MEE surface ozone data likely does not reflect a direct sensitivity of
269 OMI to surface ozone, which is very weak, but rather a sensitivity to boundary layer ozone extending up to a
270 certain depth and correlated with surface ozone.

271 P9L25. To better understand the correlation of OMI with surface ozone we examined vertical ozone profiles
272 from Hong Kong and other ozonesondes, and from the IAGOS commercial aircraft program. Some of the
273 correlation is driven by similar meteorology influencing ozone in the mid-troposphere (where OMI sensitivity
274 is maximum) and the boundary layer, but most of the correlation is driven by direct sensitivity to the boundary
275 layer.

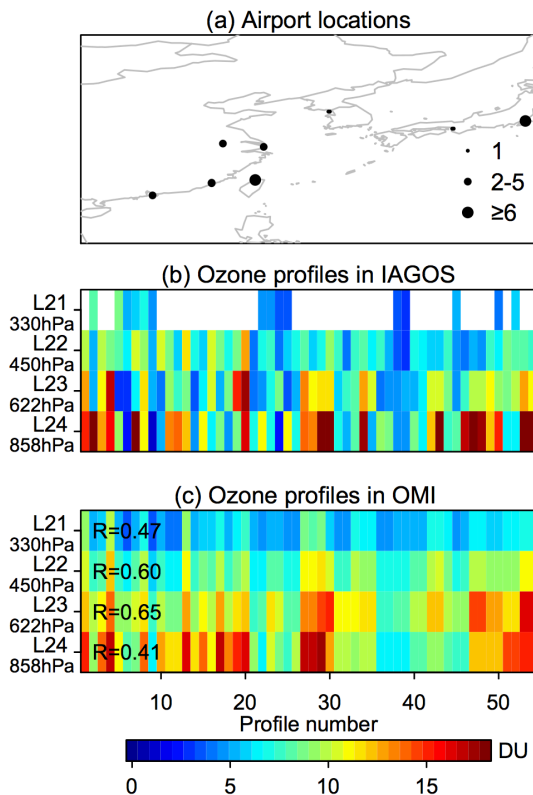
276
277 *Another necessary analysis is to see if in situ observations of ozone in the mid- troposphere are*
278 *correlated with surface ozone. I realize that the authors did look at ozonesonde profiles above Hong*
279 *Kong, but they are not very frequent and they don't tell us anything about ozone in other parts of*
280 *China, especially in the highly polluted North China Plain. The IAGOS program has hundreds of*
281 *profiles above East Asia since 1995. As shown by Ding et al. [2005] and by Gaudel et al. [2018]*
282 *ozone in sum- mertime in the boundary layer is much greater than ozone in the mid-troposphere. The*
283 *difference is due to very strong ozone production in the boundary layer, versus distant source regions*
284 *for ozone in the mid-troposphere. If the authors conducted a trans- port study for ozone in the mid-*
285 *troposphere they would find that very little of the air in this layer comes from the surface of China.*
286 *Probably 80-90% of the mid-tropospheric above China air has either been in the mid-troposphere*

287 *for days, or it comes from the boundary layer far upwind of China. The authors can freely access*
 288 *hundreds of commercial aircraft profiles of ozone and carbon monoxide above mainland China,*
 289 *Hong Kong, Taiwan and South Korea from the IAGOS database. They can then apply the OMI*
 290 *averaging kernel to the profiles and determine the relationship between IAGOS ozone in the mid- and*
 291 *lower troposphere to ozone at the surface. Does IAGOS ozone in the mid-troposphere correlate with*
 292 *ozone at the surface? Is the correlation any better than when surface ozone is correlated with*
 293 *meteorology? Then compare the IAGOS relationship to the OMI relationship. Does OMI perform*
 294 *any better than IAGOS?*

295 **Response.** We only find 54 profiles that can be used to validate the OMI data. This is because the
 296 OMI crossing time is 13:30 local time and we have to use observations close to this time window.
 297 We have added more discussion in the text.

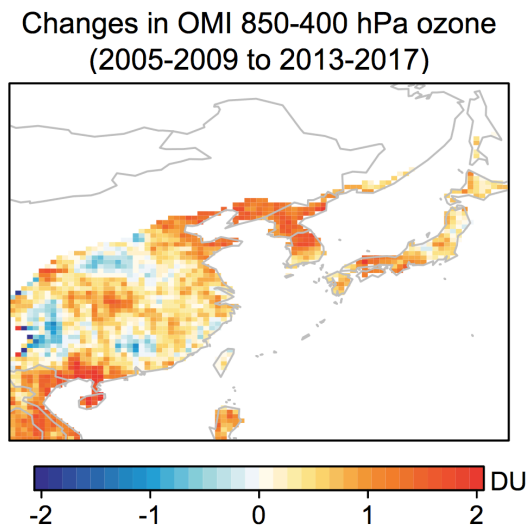
298 **P6 L9.** We applied the same daily correlation analysis to the other ozonesonde datasets and IAGOS aircraft
 299 measurements during 2005-2017 summers. For the 54 IAGOS vertical profiles coincident with OMI
 300 observations, the correlation coefficient of the 950 hPa in situ ozone and 850-400 hPa OMI ozone is $R = 0.59$
 301 ($p < 0.05$) (Figure S2).

Ozone profiles, IAGOS vs. OMI, JJA 2005-2017



302
 303 **Figure S2.** (a) Location and the number of tropospheric profiles at each airport in IAGOS that are
 304 coincident with OMI retrievals. We only select these profiles between 12-15 local time. (b) Ozone
 305 profiles from IAGOS, but mapped to OMI layers. The missing data at L21 is in part because we
 306 only select pixels that are within 200 km from the airport along the flight path. (c) OMI ozone
 307 profiles coincident with the ozonesondes. The correlations of unsmoothed 950 hPa ozone data in
 308 IAGOS with the OMI retrievals for different levels are shown inset. The correlation with 850-400
 309 hPa OMI ozone is 0.59 ($p < 0.05$)

310
311 *Figure 5 shows surface ozone trends across China which were derived from the OMI ozone product.*
312 *The strongest trends are in the far north of China and in the far south of China. Based on the summer*
313 *OMI trends (2005-2015) reported by the Tropospheric Ozone Assessment Report in supplementary*
314 *Figure S-24 of Gaudel et al. [2018], OMI has a strong trend across southern China but no trend*
315 *across northern China. Therefore I don't understand how Figure 5 can show trends across northern*
316 *China. It would be helpful to include a map that shows the OMI trends across China.*
317 **Response.** Thanks for the careful reading. The authors who plotted Figure S24 in the TOAR report
318 (Gaudel et al., 2018) are also coauthors of this work. The difference of trends in northern China
319 arises from these reasons. First, we use the 850-400 hPa ozone but Gaudel et al. (2018) uses the
320 tropospheric column ozone. Second, we use different methods to remove the background. Third, we
321 have removed the low quality L2 data but Gaudel et al. (2018) kept all of them. Now we have this
322 new figure in the supplement.



323
324 **Figure S5.** Difference of the mean OMI enhancements at 850-400 hPa from 2005-2009 to 2013-
325 2017 after correcting the Pacific background. Data are only shown for regions with DOFS below
326 400 hPa (Figure 1a) greater than 0.30.

327
328

329 **Referee #2**

330
 331 *This is a nice study that explores the potential of OMI observations of tropospheric ozone to detect*
 332 *the ozone pollution over China. While it's unrealistic to use OMI data to capture the day-to-day*
 333 *variability of ozone pollution, the authors show extreme ozone pollution may be detectable by*
 334 *aggregating long-term observations using statistical methods. Overall I think this is an important*
 335 *study to the field, which opens up the possibility to use satellite observations to detect surface ozone*
 336 *pollution, but I think the authors overpromise the value of satellite data. I have several major*
 337 *concerns:*

338
 339 **Response.** Thanks for raising these good points. This feedback has significantly improved the
 340 manuscript. Now we have a new Figure 4 showing that OMI 850-400 retrievals have limited skill in
 341 predicting the daily ozone variability in the north and we only predict the trends of ozone pollution in
 342 southern China (south of 34°N). We have new in-situ observations to validate the trends inferred
 343 from the OMI, which are shown in Figure 6.

344
 345 1. *My major concern is that the authors seem to overpromise the value of OMI data for*
 346 *characterizing the spatial and temporal trend of ground-level ozone. The title and the abstract leave*
 347 *me an impression that OMI satellite data can capture the spatial distribution and the long-term trend*
 348 *in ground-level ozone, but the results only suggest OMI may be able to detect high ozone pollution*
 349 *and capture the large-scale or latitudinal variations. I suggest the authors consider revising the title,*
 350 *otherwise it'd be misleading to readers. The authors need to be more careful with the wording. I*
 351 *think this work would actually be much more valuable if the authors can clarify the limitations of*
 352 *OMI data, which will also be useful for preparation of next-generation satellites.*

353 **Response.** Thanks for making such a good point. Now we revised the title and also discussed the
 354 limitations in many parts of the main text.

355 **New title.** Ability of the OMI satellite instrument to observe surface ozone pollution in China:
 356 application to 2005-2017 ozone trends

357 P1 L18. OMI is much more successful at capturing the day-to-day variability of surface ozone at sites in
 358 southern China (<34°N ($R = 0.3-0.6$) than in northern China ($R = 0.1-0.3$) because of weaker retrieval
 359 sensitivity and larger upper tropospheric variability in the north.

360 P5 L7. This implies that OMI can only provide statistical rather than deterministic temporal information on
 361 ozone pollution episodes, and may be more useful in South than in North China. We return to this point in
 362 Section 4.

363 P5 L18. The correlation of OMI with the MEE surface ozone data likely does not reflect a direct sensitivity of
 364 OMI to surface ozone, which is very weak, but rather a sensitivity to boundary layer ozone extending up to a
 365 certain depth and correlated with surface ozone.

366 P6L27. We find that the low correlation of OMI with boundary layer ozone in the northern ozonesonde data is
 367 due not only to the low DOFS but also to a large variability of ozone in the upper troposphere. Figure 4 (left
 368 panel) shows the standard deviation of daily OMI 400-200 hPa ozone during 2005-2017 summers, indicating
 369 that upper tropospheric ozone has much higher variability in the north (> 34°N) than in the south. This is
 370 related to the location of the jet stream and more active stratospheric influence (Hayashida et al., 2015). Figure
 371 4 (right panel) displays the vertical profiles of ozone standard deviations for the five ozonesonde sites. For the
 372 two sites north of 34°N, the ozone variability becomes very large above 8 km. Since the OMI 850-400 hPa

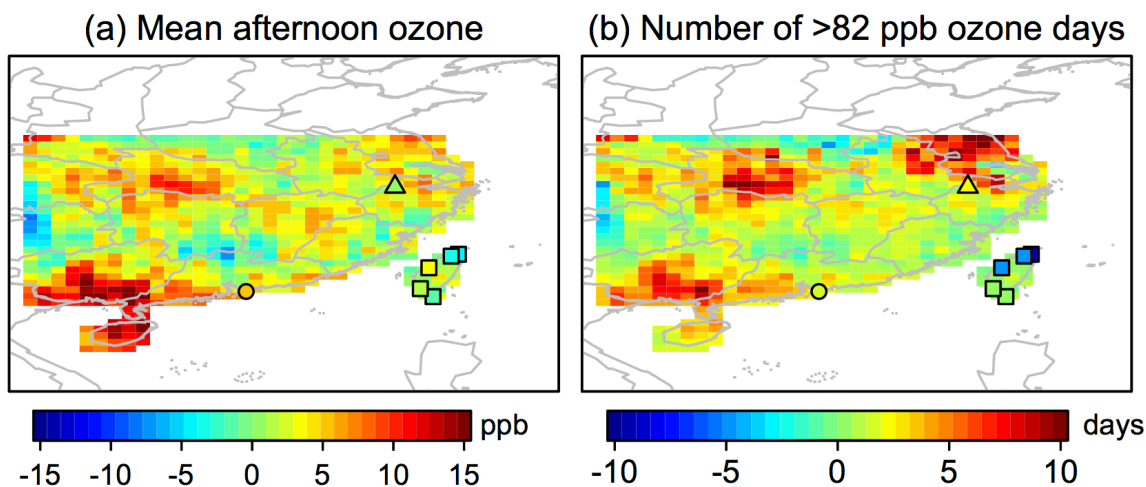
373 retrieval also contains information from above 400 hPa, this upper tropospheric variability causes a large
 374 amount of noise that masks the signal from boundary layer variability. For the three sites south of 34°N, the
 375 ozone variability in the boundary layer is much higher than in the free troposphere and the upper tropospheric
 376 ozone variability still remains low even above 8 km. In the rest of this paper we focus our attention on ozone
 377 episodes and the long-term trends in southern China (south of 34°N).

378
 379 *2. Is the point process model you used to predict ozone exceedance probability site specific? If so,
 380 how can you apply this method widely to areas without ground-based sites (as you promised in the
 381 conclusion)? The authors present the surface ozone pollution and exceedance probability only at
 382 ground-based sites, but why not show the distribution across China? For example, MEE network
 383 mainly consists of urban sites. Can you use OMI data to tell the spatial patterns of ozone pollution
 384 over rural/remote areas? If not, what's the added value of OMI data to existing ground-based
 385 network?*

386 **Response.** Thanks. The point process model makes use of all the data. Now we show the trends of
 387 ozone for all rural and remote regions in south China.

388 P7 L14. We fit the model to all daily concurrent observations of surface ozone and OMI ozone enhancements
 389 for the ensemble of eastern China sites in Figure 1 (90,601 observations for summers 2013-2017).

Changes in summertime surface ozone pollution inferred from OMI (2005-2009 to 2013-2017)



390
 391 **Figure 6.** Changes in surface ozone pollution in China between 2005-2009 and 2013-2017 as
 392 inferred from OMI afternoon observations at around 13:30 local time. (a) Change in mean summer
 393 afternoon concentrations, obtained from the difference in the mean OMI enhancements at 850-400
 394 hPa and applying equation (1). Also shown with symbols are observed changes in mean MDA8
 395 ozone from in situ observations in Lin'an, Hong Kong, and Taiwan reported by TOAR (Schultz et
 396 al., 2018). Because the TOAR observations are only reported for 2005-2014, we estimate the
 397 changes from 2005-2009 to 2013-2017 on the basis of the reported linear trends during 2005-2014
 398 (ppb a^{-1}). The change of 12-15 LT ozone at the Hok Tsui station in Hong Kong is 5.8 ppb. (b)
 399 Change in the number of high-ozone days (> 82 ppb) per summer, calculated by applying the
 400 probability of exceeding 82 ppb (equation 8) to the daily OMI enhancements. Also shown with
 401 symbols are observed changes of the number of days with MDA8 ozone exceeding 80 ppb at the
 402 TOAR sites, similarly adjusted as the change from 2005-2009 to 2013-2017. The change in the

403 number of days with 12-15 LT ozone exceeding 82 ppbv at the Hok Tsui station in Hong Kong is
404 2.1 days.

405
406 *3. Figure 5: While OMI data may be able to detect the sign of the change in ground- level ozone, the*
407 *magnitude of the change is less convincing to me. The authors suggest a 0.67 ppb /year increase in*
408 *mean ozone over China, which seems to be lower than previous studies. The point process model is*
409 *trained with ground-based observations in 2013-2017, but it's unknown how the model performs for*
410 *early years 2005 - 2009. I'd suggest the authors use available long-term ground-based ozone*
411 *observations to verify the long-term change. I understand long-term ground-based observations are*
412 *not generally available over China, but since the OMI data are global, it's possible to extend the*
413 *analysis to wider regions (e.g. Hong Kong, Japan) where long-term sites are available for*
414 *evaluation.*

415 **Response.** Thanks. We have new in-situ observations from TOAR and also from a Hong Kong site to
416 validate the trends inferred from the OMI, which are shown in Figure 6. We find the OMI inferred
417 trends are fairly consistent with the long-term records available from surface sites. We also add discussion
418 in the main text.

419 P4 L11. For evaluating the long-term surface ozone trends inferred from OMI, we use 2005-2014 trend
420 statistics for maximum daily 8-hour average (MDA8) ozone from the Tropospheric Ozone Assessment Report
421 (TOAR) (Schultz et al., 2018). We also have 2005-2017 JJA 12-15 LT mean ozone at the Hok Tsui station in
422 Hong Kong (Wang et al., 2009).

423 P9 L5. We compared the OMI trends in Figure 6 to the trends of MDA8 ozone and number of high-ozone days
424 reported by the long-term TOAR sites (Schultz et al., 2018) and our own analysis for the Hok Tsui station in
425 Hong Kong (Wang et al., 2009). For Lin'an, Hong Kong, and the 5 sites in Taiwan, the changes of mean
426 ozone concentrations from 2005-2009 to 2013-2017 are 1.1, 2.3, and -0.18 ± 2.2 ppbv (standard deviation
427 among the 5 sites) as estimated from OMI, compared to 0.7, 5.6 (or 5.8 in Hok Tsui station), and -0.75 ± 3.4
428 ppbv for MDA8 ozone at the TOAR sites. The changes in the number of ozone episodes per summer are 1.2,
429 1.9, and -0.17 ± 0.74 days in OMI, compared to 2.1, 1.8 (or 2.1 in Hok Tsui station), and -3.5 ± 3.9 days at the
430 TOAR sites. These OMI inferred trends are fairly consistent with the long-term records available from surface
431 sites.

432

433 **Referee 3**

434
435 *This paper explored the capability of OMI ozone columns to represent the surface O₃. I feel the*
436 *satellite data is over-interpreted based on the evidence provided in the paper. However, I do believe*
437 *it will be big news if substantial improvements are made to prove that the conclusion is solid.*
438

439 **Response.** We thank the reviewer for raising so many good points, which have significantly
440 improved our work. Now we have a new Figure 4 showing that OMI 850-400 retrievals have limited
441 skill in predicting the daily ozone variability in the north and we only predict the trends of ozone
442 pollution in southern China (south of 34°N). We have new in-situ observations to validate the trends
443 inferred from the OMI, which are shown in Figure 6. And we have revised the title.

444 **New title.** Ability of the OMI satellite instrument to observe surface ozone pollution in China:
445 application to 2005-2017 ozone trends
446

447
448 General comments:

449 *1. The sensitivity of OMI O₃ to the lower troposphere is very low. I feel that is the reason why no*
450 *quantitative comparison to surface observations has so far been done. I'm wondering is there any*
451 *improvements that have been made to make the quantitative comparison robust? Why does not the*
452 *quantitative comparison work for other regions, but work for China?*

453 **Response.** We now explain this better in the Introduction.

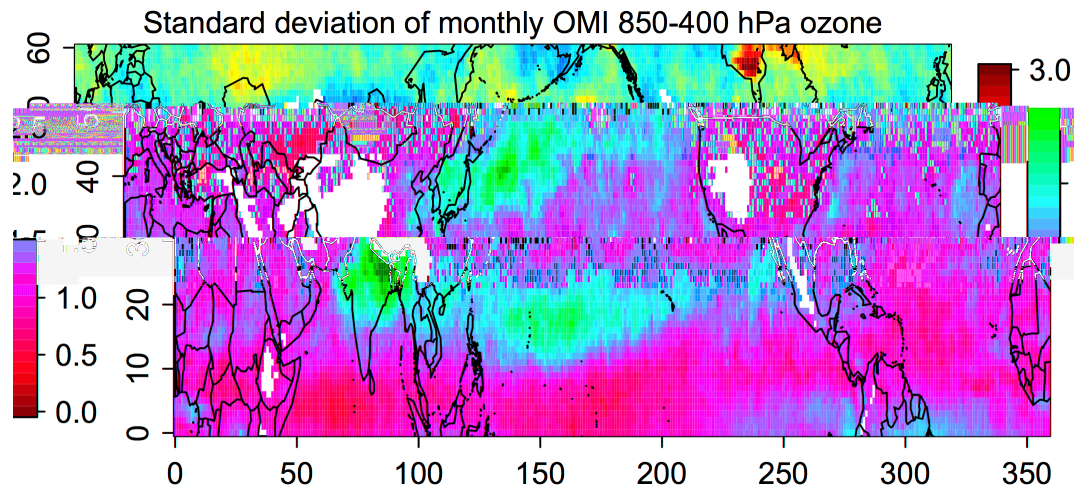
454 **P2L23.** However, no quantitative comparison of the satellite data to surface observations has so far been done.
455 Surface ozone network data are available in the US and Europe but levels are generally too low to enable
456 statistically meaningful validation. Ozone levels in China are much higher (Lu et al., 2018). The high density
457 of the MEE network, combined with vertical profile information from ozonesondes and aircraft, provides a
458 unique opportunity for evaluating quantitatively the ability of OMI to observe ozone pollution.
459

460 *2. The robustness of the residual. How large is the temporal and spatial variations of the*
461 *background? Is it likely that such variations bring significant uncertainties to the subtraction?*

462 **Response.** Thanks for making this good point. We have tested different approaches to correct the
463 background and the results are consistent with what have presented in the paper. In the text, we make
464 it simple by saying this.

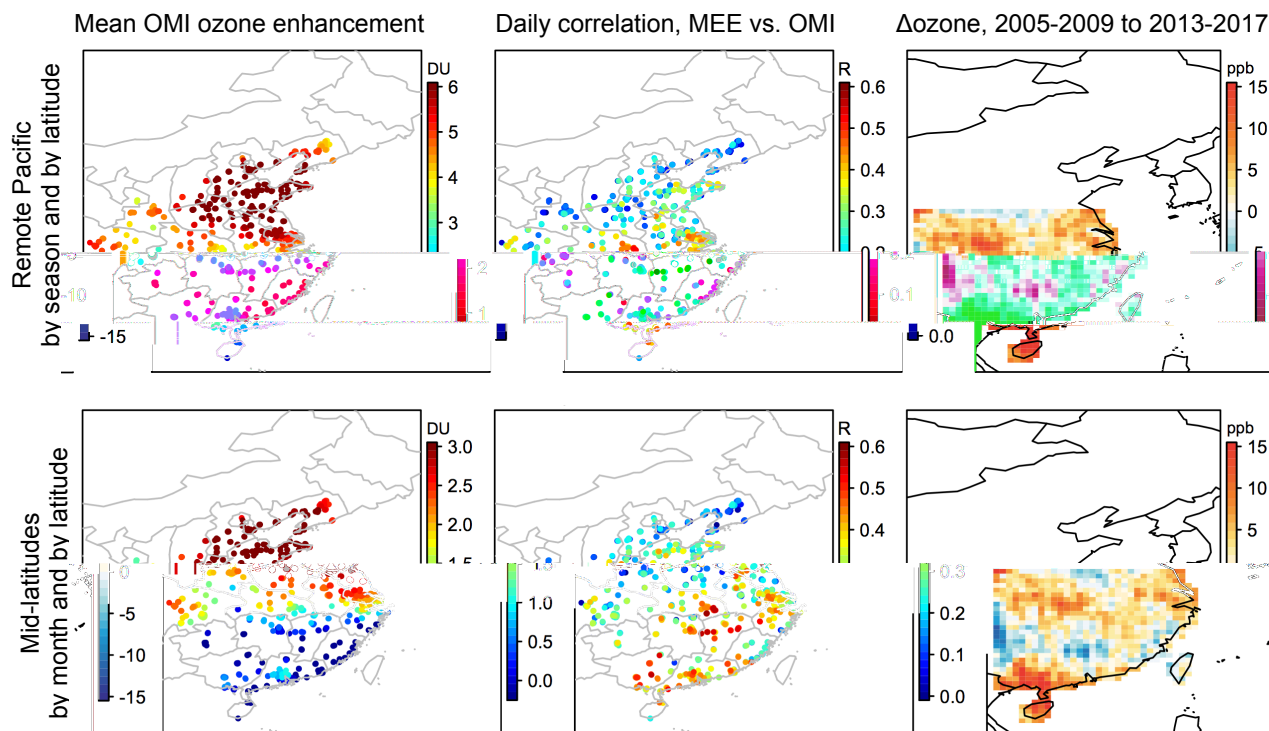
465 **P4L1.** We examined different spatial and temporal averaging domains for the North Pacific background and
466 found little effect on the residual.

467 The uncertainty related to the background correction can be found in these two figures.



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Figure SX. Standard deviation of monthly OMI 850-400 hPa ozone during 2005-2017 summers.



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Figure SX. The mean ozone enhancement (left panel), daily correlation of OMI and MEE ozone (mid panel), and the OMI inferred changes of mean ozone concentrations from 2005-2009 to 2013-2017 using different approaches correcting the OMI drift. In the top panel, we subtract the monthly mean Pacific background (150°E-150°W) for the corresponding latitude and season. In the bottom panel, we subtract the monthly mean mid-latitude ozone for the corresponding latitude and month.

479 3. The correlation between MEE and OMI. The correlation seems to be related with the dependence
480 of O₃ on latitude. I suggest additional analysis here to prove that is not the case.

481 **Response.** We are not sure if the reviewer is referring to Figure 1f here. In Figure 1f, the correlation

482 is higher in the south and lower in the north. This is because in the northern China, OMI 850-400 hPa
 483 ozone has lower sensitivity in the boundary layer, more likely to be influenced by the upper
 484 tropospheric ozone variability and stratosphere-troposphere exchange. We have added new
 485 discussion in the text.

486 P6 L27. We find that the low correlation of OMI with boundary layer ozone in the northern
 487 ozonesonde data is due not only to the low DOFS but also to a large variability of ozone in the
 488 upper troposphere. Figure 4 (left panel) shows the standard deviation of daily OMI 400-200 hPa
 489 ozone during 2005-2017 summers, indicating that upper tropospheric ozone has much higher
 490 variability in the north ($> 34^{\circ}\text{N}$) than in the south. This is related to the location of the jet stream
 491 and more active stratospheric influence (Hayashida et al., 2015). Figure 4 (right panel) displays the
 492 vertical profiles of ozone standard deviations for the five ozonesonde sites. For the two sites north
 493 of 34°N , the ozone variability becomes very large above 8 km. Since the OMI 850-400 hPa
 494 retrieval also contains information from above 400 hPa, this upper tropospheric variability causes a
 495 large amount of noise that masks the signal from boundary layer variability. For the three sites
 496 south of 34°N , the ozone variability in the boundary layer is much higher than in the free
 497 troposphere and the upper tropospheric ozone variability still remains low even above 8 km. In the
 498 rest of this paper we focus our attention on ozone episodes and the long-term trends in southern
 499 China (south of 34°N).

500
 501 If the reviewer refers to Figure 1d, we now make it clear that we have corrected the background that
 502 is dependent on latitudes.

503 P3 L23. To remove this gradient and also any long-term uniform drift in the data, we subtract the monthly
 504 mean Pacific background (150°E - 150°W) for the corresponding latitude and month

505 P4 L22. After subtracting the North Pacific background for the corresponding latitude in month, we obtain the
 506 OMI ozone enhancements shown in Figure 1d.

507 P4 L23. The spatial correlation coefficient between the OMI ozone enhancements and the MEE surface
 508 network is $R = 0.73$ over eastern China. The correlation is driven in part by the latitudinal gradient but also by
 509 the enhancements in the large megacity clusters identified as rectangles in Figure 1b. Thus the correlation
 510 coefficient is $R = 0.55$ for the 26 - 34°N latitude band including YRD, SCB, and Wuhan.

511
 512
 513 *Specific comments:*

514 1. *“We exclude outliers with over 35 Dobson Units (DU) at 850- 400 hPa (>99th percentile in*
 515 *eastern China) and exclude July 2011 when the retrievals are anomalously high.” Please give the*
 516 *reference to the exclusion. Otherwise, please quantify the influence of the exclusion.*

517 **Response.** Thanks for pointing this out. We delete this because we don’t use the July 2011 data and
 518 not excluding the extremely high data has little effect on our result.

519
 520 2. *“We see that high-ozone episodes in the 950-850 hPa sonde data are systematically associated*
 521 *with high OMI values, though the converse does not always hold.” Additional explanation for the*
 522 *reason is expected.*

523 **Response.** Thanks. Now we say

524 P6L4. We see that high-ozone episodes in the 950-850 hPa sonde data are systematically associated with high
 525 OMI values, though the converse does not always hold because free tropospheric enhancements affecting OMI

526 can also occur.

527

528

529