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

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

Referee #2

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

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

- 1. My major concern is that the authors seem to overpromise the value of OMI data for characterizing the spatial and temporal trend of ground-level ozone. The title and the abstract leave me an impression that OMI satellite data can capture the spatial distribution and the long-term trend in ground-level ozone, but the results only suggest OMI may be able to detect high ozone pollution and capture the large-scale or latitudinal variations. I suggest the authors consider revising the title, otherwise it'd be misleading to readers. The authors need to be more careful with the wording. I think this work would actually be much more valuable if the authors can clarify the limitations of OMI data, which will also be useful for preparation of next-generation satellites.
- Response. Thanks for making such a good point. Now we revised the title and also discussed the limitations in many parts of the main text.
- New title. Ability of the OMI satellite instrument to observe surface ozone pollution in China: application to 2005-2017 ozone trends
- P1 L18. OMI is much more successful at capturing the day-to-day variability of surface ozone at sites in southern China $<34^{\circ}N$ (R=0.3-0.6) than in northern China (R=0.1-0.3) because of weaker retrieval
- sensitivity and larger upper tropospheric variability in the north.

 P5 L7. This implies that OMI can only provide statistical rather than deterministic temporal information on
- ozone pollution episodes, and may be more useful in South than in North China. We return to this point in Section 4.
- 44 P5 L18. The correlation of OMI with the MEE surface ozone data likely does not reflect a direct sensitivity of

OMI to surface ozone, which is very weak, but rather a sensitivity to boundary layer ozone extending up to a certain depth and correlated with surface ozone.

P6L27. We find that the low correlation of OMI with boundary layer ozone in the northern ozonesonde data is due not only to the low DOFS but also to a large variability of ozone in the upper troposphere. Figure 4 (left panel) shows the standard deviation of daily OMI 400-200 hPa ozone during 2005-2017 summers, indicating that upper tropospheric ozone has much higher variability in the north (> 34°N) than in the south. This is related to the location of the jet stream and more active stratospheric influence (Hayashida et al., 2015). Figure 4 (right panel) displays the vertical profiles of ozone standard deviations for the five ozonesonde sites. For the two sites north of 34°N, the ozone variability becomes very large above 8 km. Since the OMI 850-400 hPa retrieval also contains information from above 400 hPa, this upper tropospheric variability causes a large amount of noise that masks the signal from boundary layer variability. For the three sites south of 34°N, the ozone variability in the boundary layer is much higher than in the free troposphere and the upper tropospheric ozone variability still remains low even above 8 km. In the rest of this paper we focus our attention on ozone episodes and the long-term trends in southern China (south of 34°N).

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

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

P7 L14. We fit the model to all daily concurrent observations of surface ozone and OMI ozone enhancements 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)

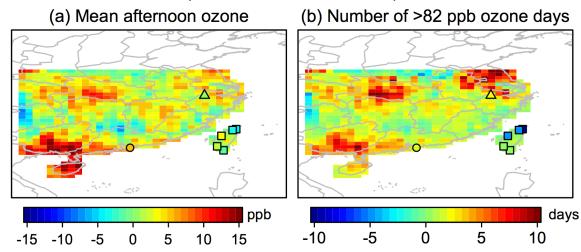


Figure 6. Changes in surface ozone pollution in China between 2005-2009 and 2013-2017 as inferred from OMI afternoon observations at around 13:30 local time. (a) Change in mean summer afternoon concentrations, obtained from the difference in the mean OMI enhancements at 850-400

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hPa and applying equation (1). Also shown with symbols are observed changes in mean MDA8 ozone from in situ observations in Lin'an, Hong Kong, and Taiwan reported by TOAR (Schultz et al., 2018). Because the TOAR observations are only reported for 2005-2014, we estimate the changes from 2005-2009 to 2013-2017 on the basis of the reported linear trends during 2005-2014 (ppb a⁻¹). The change of 12-15 LT ozone at the Hok Tsui station in Hong Kong is 5.8 ppb. (b) Change in the number of high-ozone days (> 82 ppb) per summer, calculated by applying the probability of exceeding 82 ppb (equation 8) to the daily OMI enhancements. Also shown with symbols are observed changes of the number of days with MDA8 ozone exceeding 80 ppb at the TOAR sites, similarly adjusted as the change from 2005-2009 to 2013-2017. The change in the number of days with 12-15 LT ozone exceeding 82 ppbv at the Hok Tsui station in Hong Kong is 2.1 days.

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

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

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

P9 L5. We compared the OMI trends in Figure 6 to the trends of MDA8 ozone and number of high-ozone days reported by the long-term TOAR sites (Schultz et al., 2018) and our own analysis for the Hok Tsui station in Hong Kong (Wang et al., 2009). For Lin'an, Hong Kong, and the 5 sites in Taiwan, the changes of mean ozone concentrations from 2005-2009 to 2013-2017 are 1.1, 2.3, and -0.18±2.2 ppbv (standard deviation 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 ppbv for MDA8 ozone at the TOAR sites. The changes in the number of ozone episodes per summer are 1.2, 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 TOAR sites. These OMI inferred trends are fairly consistent with the long-term records available from surface sites.