Dear Editor,

We appreciate the prompt reviews and would like to thank the reviewers for insightful comments and suggestions on our manuscript entitled "What is the cause(s) of ozone trends in three megacity clusters in eastern China during 2015–2020?" (MS No.: acp-2022-781). We have carefully considered all comments and suggestions. Listed below are our point-by-point responses to all comments and suggestions of Reviewer #1 (Reviewer's points in black, our responses in blue).

Referee #1

The annual mean concentration of MDA8 increased at a high rate in BTH, YRD and PRD during the period 2015-2020. The spatial expansion of high O_3 from urban centers to surrounding regions was found during 2015-2017, accompanied by a saturation effect. The authors suggest that the occurrence of meteorological conditions of solar radiation and positive temperature under WPSH and mid-high latitude wave activities are the main reason for the increased O_3 episodes with four or more consecutive O_3-exceeding days in the three megacity clusters. The paper is logical and informative. It is a novel and interesting topic. I suggest it to be accepted after addressing the following comments.

Response: We appreciate very much your encouraging comments. In the followings, we have carefully considered and responded to your specific comments and suggestions.

1. L91-93, The increase in the contribution of O_3-exceeding days is the primary contributor to the large increase in the annual mean O_3. The contribution of O_3-exceeding days is affected by exceeding days and concentrations. Therefore, I think the mean concentration of O_3-exceeding days should be shown.

Response:

Thank you for an excellent suggestion. We have revised L91–93 in the revised manuscript to: "It is clear that the increase in O_3-exceeding days is the primary

contributor to the large increase in the annual mean O_3 in all three megacity clusters from 2015 to 2020. The contribution of O_3-exceeding days is affected mostly by the changing number of exceeding days (more than 80%), and secondly but still significantly by their changes in concentrations (less than 20%) (Tables 2–4). E.g. in BTH the exceeding days were 31, 43, 62, 74, 96 and 78 days in the individual years of 2015–2020, respectively, while their concentrations of those years were 66.42, 64.13, 69.44, 68.21, 70.19 and 69.69, respectively (Table 2 second column). Contributions from non-O_3-exceeding days are insignificant ($p > 0.1$), except that in BTH (Figure 2a) which shows a significant declining contribution ($p = 0.02$) due to the reduced number of non-O_3-exceeding days."

2. L162-163, The contribution of the increased O_3 concentration and the number of O_3-exceeding days is +20% and +34%, respectively. Please elaborate more about this method, how is the contribution calculated? L214-219, and the difference in O_3 between 2017 and 2015 can be attributed to the large number of days and higher average concentration with four or more consecutive O_3-exceeding days and those with less than four consecutive O_3-exceeding days. How are the respective contributions distinguished?

Response:

Line 162–163: Accounted for the number of O_3-exceeding days, the ratio of MDA8 O_3 in all O_3-exceeding days between 2017 and 2015 became $(64.35\times40)/(53.79\times31)=1.54$. The combined effect of ozone concentration and O_3-exceeding days was 54%, with the effect of ozone concentration is $(64.35-53.79)/53.79=0.2(20\%)$, so the effect of O_3-exceeding days is 54%-20%=34%. We acknowledge that the above calculation method shortchanges slightly the contribution of the increased O_3 concentration compared to the calculation in which the contribution of the number of O_3-exceeding days is calculated first, i.e. $(40-31)/31=29\%$ is the contribution of the number of days, and 54%-29%=25% the contribution of the O_3 concentration. This shortchanging effect is caused by the difficulty in attributing the cross term between the number of days and

the O_3 concentration.

Line 214–219: The respective contributions were distinguished by first evaluating the contribution due to Line 214–216 “One of the most remarkable differences between 2017 and 2015 in Figures 9a–9f was the large number of days with four or more consecutive O_3-exceeding days in 2017 (28 days, Figure 9d) over that of 2015 (7 days, Figure 9a)”: 28-7=21, the difference in total O_3-exceeding between 2017 and 2015 (Table 2, column 2) is 62-31=31, 21/31=68% which is fairly close to the number 62% in Line 216. Secondly, the contribution of Line 217–218 “the 10 days’ difference (2017 vs. 2015) in the number of days with less than four consecutive O_3-exceeding days” is 10/31=32%, which is fairly close to the number 30% in Line 217. The discussion above explains how the major respective contributions were distinguished. The small discrepancies of a few percent is due to the changes in O_3 concentrations.

3. L181-184, The emission of air pollutants and O_3 precursors isn’t a main cause of the expansion and saturation in BTH. Is the result the same in PRD and YRD?

Response:

Yes, the results in PRD and YRD are same as BTH, as illustrated below:

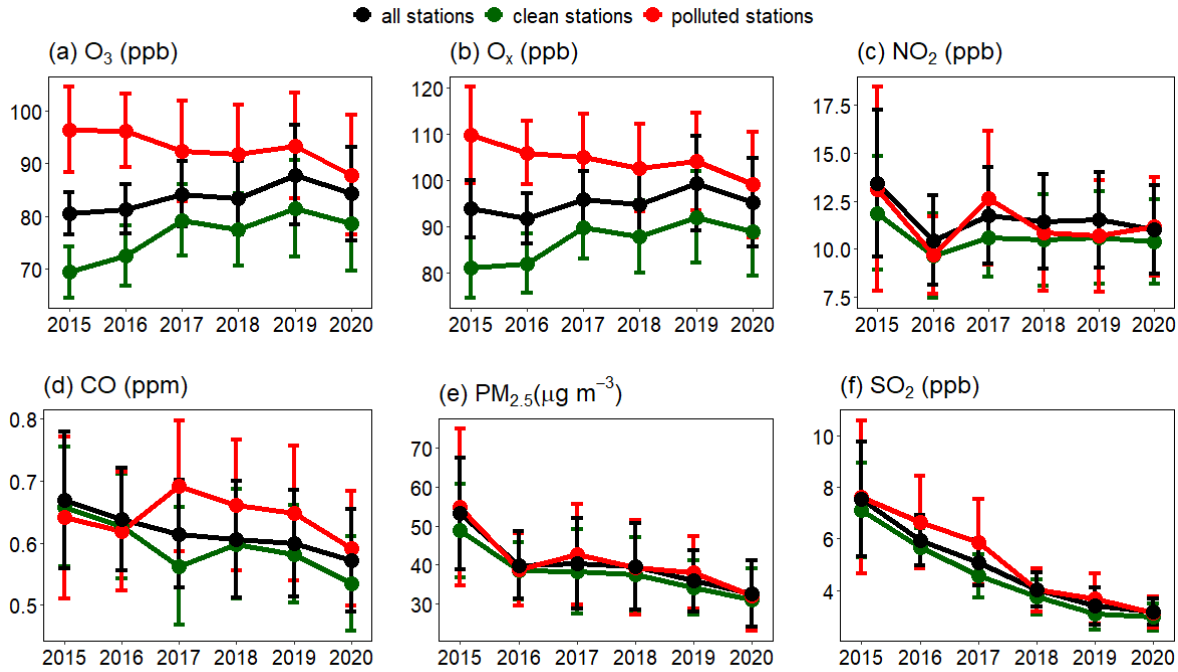

Figure R1. Annual mean concentrations of maximum daily 8-hour average O_3 in YRD during O_3-exceeding days for all stations (black), polluted stations (red) and clean stations (green) (a), same as (a) except for (b) Ox, (c) NO_2, (d) CO, (e) $PM_{2.5}$ and (f) SO_2.

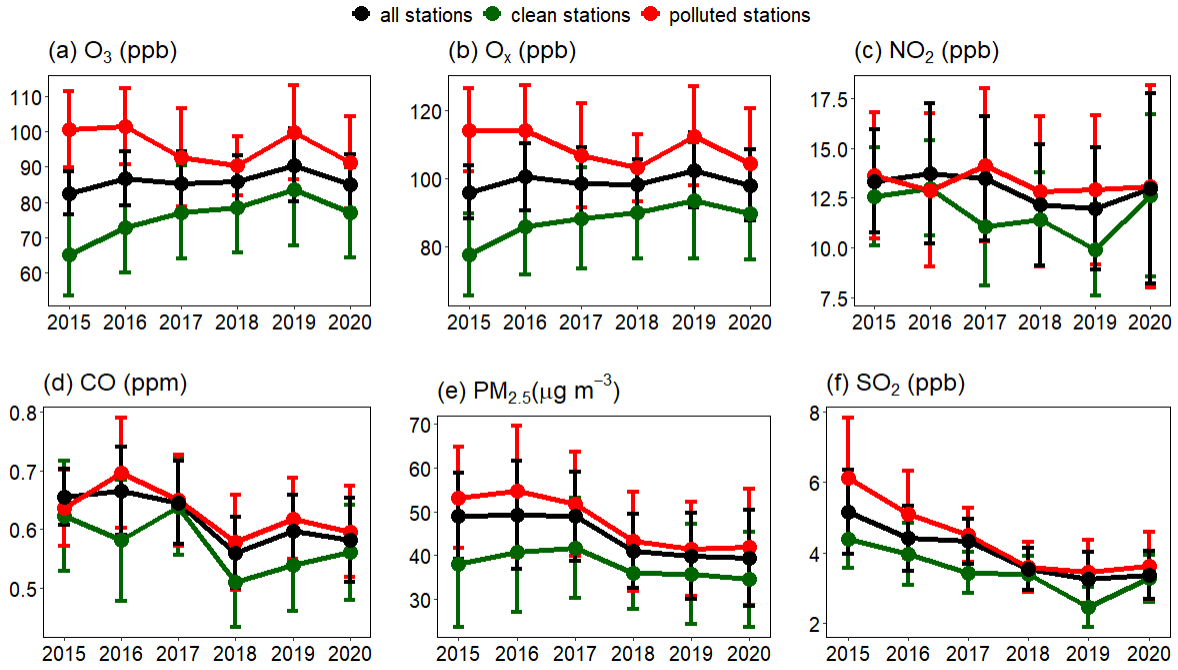

Figure R2. Same as Figure R1, except for PRD.

4. The values of SSR and T2m of the O_3 episodes with four or more consecutive O_3-exceeding days is lower than those of O_3 episodes with less than four consecutive O_3-exceeding days in PRD during 2017-2020, which are different with in BTH and YRD. In addition, O_3-exceeding days in PRD are mostly decoupled from those in BTH and YRD. Does it imply that the cause of worsening O_3 trend in PRD is different with BTH and YRD?

Response:

This is a very good and challenging question. Yes, we believe that specific causes of worsening O_3 trend are different among PRD, BTH and YRD. Nevertheless, the causes are most likely meteorological/climate oscillation in nature. For instance, we think that changes in tropical cyclones might be a factor in the worsening O_3 trend in PRD and southern YRD.

5. L280-284, The annual weighted SSR/T2m can be performed by multiplying the difference between the four or more consecutive O_3-exceeding days and clean days of SSR/T2m with the frequency of occurrence of O_3 episodes with four or more consecutive O_3-exceeding days each year. Is that right? Has this approach been used in previous studies? Do the months inside the parentheses in Figure 12 represent the months when O3-exceeding days happen in this region?

Response:

L280–284, Yes, you are right about the approach used to get the results in Figure 12. To our limited knowledge, this approach has not been used in previous studies.

Yes, the months in parentheses in Figure 12 represent the months in which the area experienced four or more consecutive O_3-exceeding days.

6. Why did the expansion and saturation occur mostly during 2015-2017? O_3 concentrations have increased in 2015-2019. The annual mean concentration of MAD8 decreased significantly in 2020. In BTH, YRD and PRD, the number of days of all O_3-

exceeding days has increased from 2015 to 2019. However, the number of O_3-exceeding days decreases in 2020. Is this change also due to the influence of WPSH and mid-high latitude wave activities?

Response:

The statement of "the expansion and saturation occur mostly during 2015–2017" was entirely based on the results shown in Figure 8: the green line and the red line converge to each other from 2015 to 2017.

Our view is that meteorology and climate played the major role in the interannual variations and trends in ozone in the three megacity clusters during 2015–2020, but it does not necessarily apply to the specific problem of "the number of O_3-exceeding days decreases in 2020".

7. Figure 7 is the same as Figure 6. Please check it.

Response:

Sorry! In the version of this article initially submitted, Figure 7 was erroneously pasted. The correct Figure 7 is shown in the following:

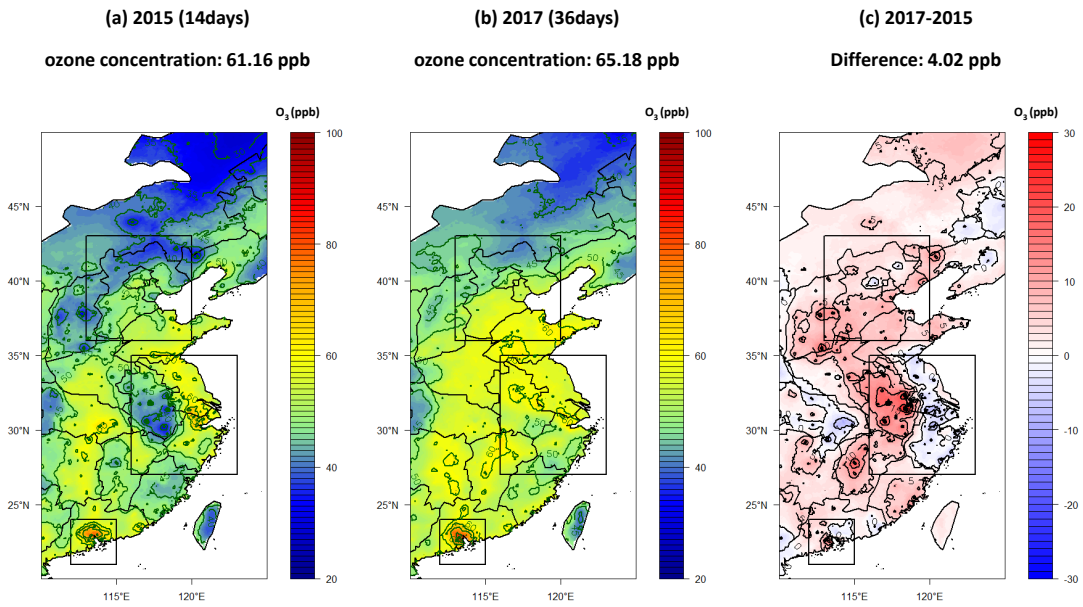

Figure R3. Spatial distribution of annual mean concentrations of maximum daily 8-hour average O_3 for O_3-exceeding days in PRD in 2015 (a), 2017 (b) and their difference (2017–2015) (c). The top, middle and bottom rectangle boxes denote BTH, YRD and PRD districts, respectively. The number inside the parenthesis behind 2015 or 2017 denotes the number of O_3-exceeding days.