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 positive ozone trends in three megacity clusters in eastern China during 2015–2020?" (MS No.: egusphere-2023-1088). 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). Extensive revisions have been made in the revised manuscript to address the comments and suggestions, which we believe is significantly improved. For that we thank the reviewer.

Anonymous Referee #1

This paper analyzes the cause of 2015-2020 positive ozone trends in megacities in China. While the topic is important, the main conclusion of this study is not well supported by the analyses. In addition, the presentation, including the logic, word expression, and figure quality requires substantial improvement. I recommend at least a major revision before it can be re-considered to be published in ACP.

Response:

In the followings, we have carefully considered and responded to your comments and suggestions.

Main concern:

The authors emphasize the role of meteorology and diminish that of emission change as the key cause of the ozone rise between 2015-2020 in many places of the text, but in most places, there is no direct evidence to approve or disapprove, and the conclusions seem to be arbitrary, I list several questions related to this point below that need to be addressed.

(1) Line 166-167: Why is this simply attributed to weather system? (e.g. lines 166-168,

178-180, and many others).

Response:

Thank you for pointing out the lack of clarity on attributing the cause of interannual variability of O_3 in our paper. We clarify this problem in the revised manuscript by consolidating and elaborating statements in “lines 166–168, 178–180, and many others” into a new section near line 243 as shown below.

“3.3.1 Changing emissions as a possible cause of O_3 trends in 2015–2020

As mentioned earlier, two emission-oriented hypotheses have been proposed as a possible cause of the O_3 trends in 2015–2020. One is changing emissions of O_3 precursors NOx and VOC (Li et al., 2022). The other is the reduced removal of HO_2 radicals by diminishing $PM_{2.5}$ suggested by Li K. et al. (2021) and Shao et al. (2021). Li et al. (2022) demonstrated convincingly that the NO titration effect was the cause of the linear trend in O_3 in PRD (0.5 ppb yr^{-1}) during the period 2006–2019. But for the period 2015–2020, the NO titration effect could account for only about 10% of the linear trend in O_3 of the low O_3 stations in PRD (5.0 ppb yr^{-1}, green line, Fig. S3a).

The increase of 30 ppb in O_3 at the low O_3 stations in BTH from 2015 to 2017 (green line, Figs. 4a and 8a represents about 50% increase in O_3. The titration effect can account for only about 5% (Fig. 8f). If this increase of 30 ppb in O_3 were due to an enhancement in O_3 precursors, the enhancement would have to be substantially greater than 50% because of the well-known less-than-linear relationship between changes in O_3 and its precursors, i.e., substantially more percentage changes in precursors are needed for each percentage change in O_3 (Dodge, 1977; Shafer and Seinfeld, 1985). Figures 8d and 8f show that CO (a proxy for VOC) and NOx changed only by a few percent from 2015 to 2017, more than one order of magnitude less than the changes needed. Hence it appears that changes in meteorological conditions conducive to O_3 formation are more likely the major contributing factor to the 50% increase in O_3 at the low O_3 stations in BTH. Similar argument can be extended to YRD and PRD (Figs. S1 and S3).

The theory of reduced removal of HO_2 radicals by diminishing $PM_{2.5}$ (25%, green line of Fig. 8c) appeared to be valid qualitatively for the 50% increase in O_3 at the low O_3 stations in BTH from 2015 to 2017 (green line of Fig. 8a). But this theory was contradicted directly by the phenomenon at the high O_3 stations where a 30% reduction in $PM_{2.5}$ (red line of Fig. 8c) corresponded to a decrease rather than an increase in O_3 (red line of Fig. 8a)."

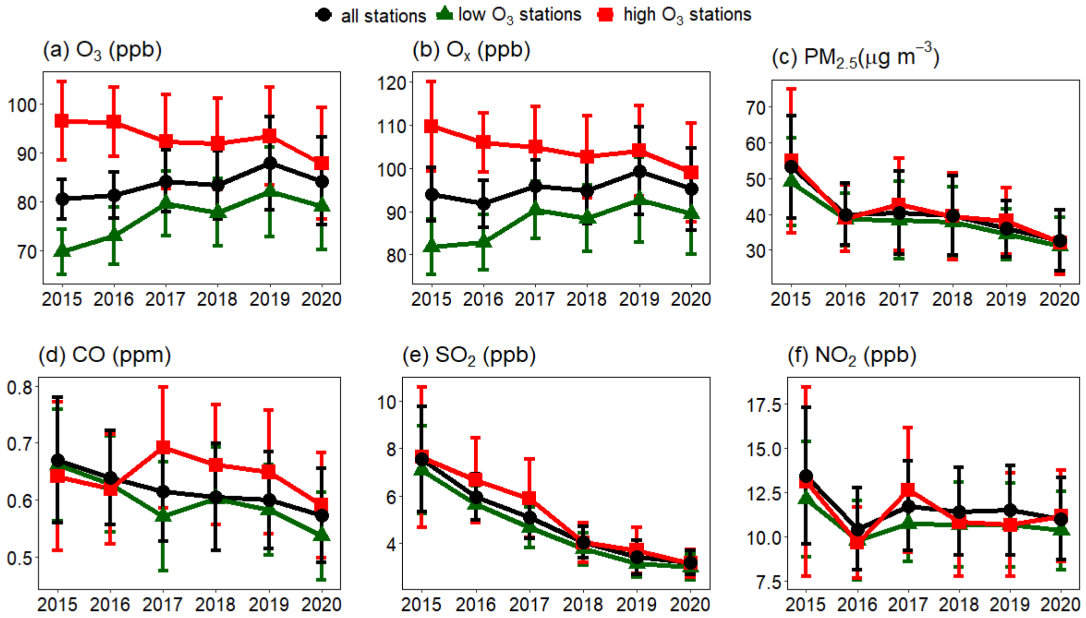

Figure S1. Annual mean concentrations of maximum daily 8-hour average O_3 in YRD during O_3-exceeding days for all stations (black), high O_3 stations (red) and low O_3 stations (green) (a), same as (a) except for Ox (b), $PM_{2.5}$ (c), CO (d), SO_2 (e), NO_2 (f).

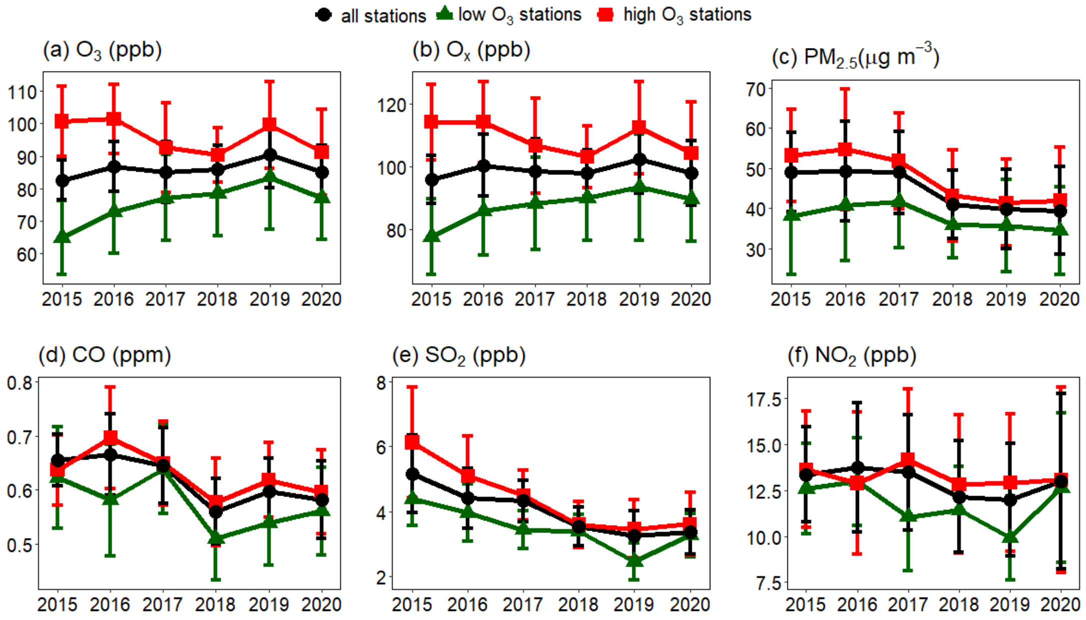

Figure S3. Annual mean concentrations of maximum daily 8-hour average O_3 in PRD during O_3-exceeding days for all stations (black), high O_3 stations (red) and low O_3 stations (green) (a), same as (a) except for Ox (b), $PM_{2.5}$ (c), CO (d), SO_2 (e), NO_2 (f).

(2) Line 177-180: For what reason it is "highly unlikely emission change"? It is not convincing to guess the cause simply based on the spatial pattern of ozone change. In addition, the ozone data used in this study is smoothed from observations in the TAP dataset, thus it may not reflect the true ozone change in the regions with no direct observation.

Response:

This part of the text (Line 177–180) has been rewritten entirely in the revised manuscript. Please see our response to your Main concern (1) about "highly unlikely emission changes".

The TAP data system integrates multiple data sources, including ground observations, satellite remote sensing data, high-resolution emission inventories, air quality model simulations, and other relevant information (Xue et al., 2020; Xiao et al., 2022). Over time, the TAP team has made consistent enhancements, resulting in a three-stage O_3

prediction model with an impressive R^2 value of 0.84 when compared to ground observations spanning the years 2013 to 2020. Consequently, we have confidence that the spatial O_3 data provided by TAP effectively captures the variations in O_3 concentrations within the megacity clusters.

(3) The function of Section 3.2 is very confusing to me. The paper is about 2015-2020 trends, but here only the difference between 2015 and 2017 is analyzed. Even though the ozone difference between 2017 and 2015 is mostly driven by weather anomalies, the authors do not explain what weather system can sustain high ozone from 2017 to 2018-2020. Analyses of yearly differences cannot be simply applied to explain the 6-year trend.

Response:

We understand your concern regarding the focus on the difference between 2015 and 2017. The time period we are examining spans only six years (2015–2020), a duration that might not typically be referred to as a trend if not for its significant environmental impact. The two years between 2015 and 2017 represent a substantial portion of the entire period, contributing one third or more to the overall trends in the three megacity clusters (as depicted in Fig. 4, black lines). In fact, if the O_3 levels in 2017 and 2019 had been the same as those in 2015, there would have been no discernible trend in any of the three megacity clusters (Fig. 4, black lines). Therefore, to understand the six-year trend, it is crucial to elucidate the factors behind the elevated O_3 levels in 2017 and 2019. Furthermore, it's worth noting that only the high values of 2019 are essential for the PRD region (Fig. 4c, black lines). We now have a good understanding that these elevated levels were influenced by the high frequency of downdrafts and stable atmospheric conditions associated with tropical cyclones in the northwest Pacific (as detailed in Section 3.3.2 of the revised manuscript).

In addition, in Section 3.3.3 we have conducted a comprehensive analysis of the role played by the Western Pacific Subtropical High (WPSH) during O_3-exceeding days in all three megacity clusters for each year from 2015 to 2020.

(4) The authors list “ozone at high ozone stations unchanged while ozone at low ozone station increase” as a major finding (Line 235). If it is driven by weather, I wonder what weather system could selectively influence sites with different ozone levels. It more likely reflects chemical factors. This is a key concern.

Response:

We appreciate this “key concern” as it points out a lack of clarity in our presentation. We actually fully agree that “It more likely reflects chemical factors.” But we believe that the “chemical factors” are driven by changes in meteorology rather than changes in emissions of air pollutants. As a photochemical product, the ozone trends are primarily controlled by the chemical production of ozone. The contribution of meteorology proposed in our manuscript is mostly through enhanced photochemical production of ozone at the low O_3 stations. This point was stated at lines 23–26 in the abstract which was made in response to a previous similar concern of the editor-in-charge. In addition, we have revised lines 235–243 as follows: “And (3), the expansions of high O_3 in the three megacity clusters were accompanied by a saturation effect that O_3 concentrations at the high O_3 stations of approximate 100 ppb in 2015 remained nearly constant or slightly declined throughout the entire period of 2015–2020 (Fig. 4).”

In regard to “what weather system could selectively influence sites with different ozone levels”? We address this point by adding the following four paragraphs and two figures (Figs. 12 and 13) near Line 300:

“Hu W. et al. (2023), in collaboration with this study, conducted a statistical analysis to assess processes that contribute to high O_3 formation in PRD when TCs were present in the northwest Pacific. They investigated the impact of the distance between TCs in the northwest Pacific and PRD on the O_3 concentration in the PRD from 2006 to 2020. They found that the large numbers of consecutive O_3-exceeding days in 2017 and 2019 relative to 2015 were primarily attributable to the greater occurrence of downdrafts and stable atmospheric conditions brought about by mid-distance category TCs. This finding clearly establishes that changing frequency of mid-distance category TCs (i.e. changing meteorological conditions) is the cause of the increases in the numbers of consecutive O_3-exceeding days as well as the higher O_3 concentrations in PRD. Ongoing study by our research group further shows that the mid-distance category TCs are predominately those TCs with tracks starting around the southern Philippines and ending near Korea and/or Japan. Since TC tracks in northwestern Pacific are strongly controlled by WPSH, we conclude that both Philippines-to-Korea/Japan track TCs and corresponding distribution and intensity of WPSH contributed to the higher consecutive O_3-exceeding days in PRD from 2015 to 2020.

Mechanically we propose that the O_3 concentrations at the high O_3 stations stayed close to a saturation level of about 100 ppb throughout 2015 to 2020, even under increased downdrafts and stable atmospheric conditions brought about by mid-distance category TCs. This saturation effect was the result of enhanced rates of atmospheric dispersion, dry deposition and photochemical loss at high O_3 concentrations, which were supported by modeling results (Li et al., 2012; Ouyang et al., 2022; Zhang et al., 2023). It is also consistent with theoretical consideration. While the low O_3 stations, where O_3 production were relatively small in 2015, experienced significant enhancements in the O_3 production (32 ppb in BTH, 12 ppb in PRD) from 2015 to 2017 because in the latter year the increased downdrafts and stable atmospheric conditions brought about by mid-distance category TCs were highly conducive to O_3 formation (Hu W. et al., 2023).

Following the analysis by Hu W. et al. (2023), the mean vertical velocity at 850 hPa during all O_3-exceeding days in PRD in 2015 (Fig. 12a) is compared to that of episodes with four or more consecutive O_3-exceeding days in 2017 (Fig. 12b). Major features in Fig. 12 compare very well with those of Fig. 7. E.g. area with positive vertical velocity (downdrafts) in 2017 (red area in Fig. 12b), which was highly conducive to O_3 formation, was by far more widespread and greater in value than that of 2015 (red area in Fig. 12a), agreeing well with the greater high O_3 area of Fig. 7b (2017) than that of Fig. 7a (2015). This agreement confirms that the increase in O_3 in PRD from 2015 to

2017 was caused by increased downdrafts and stable atmospheric conditions (meteorological conditions) brought about by TCs as suggested by Hu W. et al. (2023). The same plots for BTH are shown in Fig. 13. Features of Fig. 13 are highly consistent with those of Fig. 5. The same plot for YRD (Fig. S10) also showed more extensive and greater downdrafts in 2017 than 2015. However, the area of positive vertical velocity in YRD appeared to shift about 500 km to the east compared to the area of high of O_3 in Fig. 6b. Considering the uncertainty in evaluating the vertical velocity and that O_3 formation is also dependent on parameters other than the vertical velocity, the discrepancy is acceptable.

In summary of this section, the trends in O_3 in the three megacity clusters are critically dependent on the number of four or more consecutive O_3-exceeding days. In addition, Hu W. et al. (2023) found that the changing frequency of mid-distance category TCs (i.e. changing meteorological conditions) is the cause of the increases in the numbers of consecutive O_3-exceeding days as well as the O_3 concentrations in PRD. More importantly, our additional analyses of the mean vertical velocity at 850 hPa over the three megacity clusters (Figs. 12, 13 and S10) show that the increases in O_3 in all three megacity clusters from 2015 to 2017 were caused by enhanced downdrafts and stable atmospheric conditions (meteorological conditions) which were highly conducive to O_3 formation. The enhanced downdrafts and stable atmospheric conditions were brought about by TCs and associated WPSH. Here we bring up WPSH because it is well known that the tracks of TCs are influenced strongly by WPSH, and that WPSH affects strongly regional atmospheric dynamics and therefore O_3 formation (Chang et al., 2019; Mao et al., 2020; Ouyang et al., 2022; Zhao and Wang, 2017)."

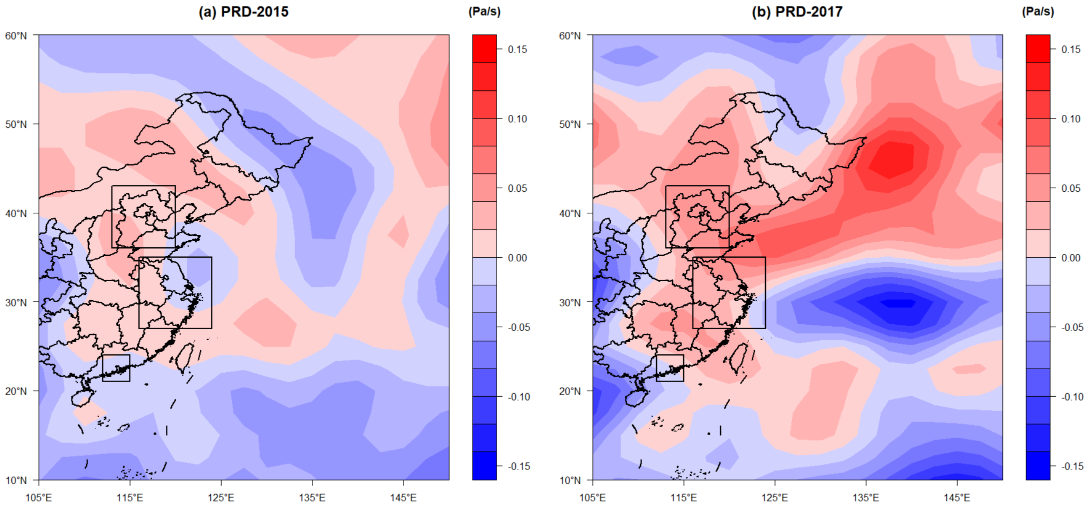

Figure 12. Mean vertical velocity at 850hPa during O$_3$-exceeding days in PRD in 2015 (a) and during episodes with four or more consecutive O$_3$-exceeding days in 2017 (b).

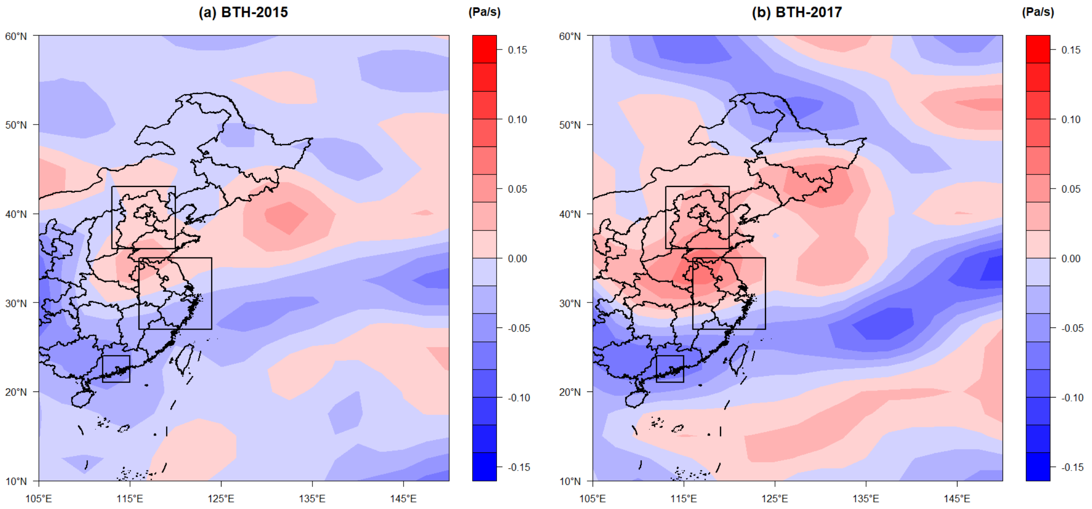

Figure 13. Mean vertical velocity at 850 hPa during O$_3$-exceeding days in BTH in 2015 (a) and during episodes with four or more consecutive O$_3$-exceeding days in 2017 (b).

(5) Line 210-223: VOCs emission change is not considered here. And again this is for difference between 2015 and 2017, it doesn’t explain the trend at all.

Response:

Please refer to our response to Main concern (3). Specifically, Lines 210–223 has been replaced in the revised manuscript by Lines 216–228 in the new section 3.3.1, which does consider VOC emissions as shown below: "The increase of 30 ppb in O_3 at the low O_3 stations in BTH from 2015 to 2017 (green line, Figs. 4a and 8a) represents about 50% increase in O_3. The titration effect can account for only about 5% (Fig. 8f). If this increase of 30 ppb in O_3 were due to an enhancement in O_3 precursors, the enhancement would have to be substantially greater than 50% because of the well-known less-than-linear relationship between changes in O_3 and its precursors, i.e., substantially more percentage changes in precursors are needed for each percentage change in O_3 (Dodge, 1977; Shafer and Seinfeld, 1985). Figs. 8d and 8f show that CO (a proxy for VOC) and NOx changed only by a few percent from 2015 to 2017, more than one order of magnitude less than the changes needed. Hence it appears that changes in meteorological conditions conducive to O_3 formation are more likely the major contributing factor to the 50% increase in O_3 at the low O_3 stations in BTH. Similar argument can be extended to YRD and PRD (Figs. S1 and S3).

The theory of reduced removal of HO_2 radicals by diminishing $PM_{2.5}$ (25%, green line of Fig. 8c) appeared to be valid qualitatively for the 50% increase in O_3 at the low O_3 stations in BTH from 2015 to 2017 (green line of Fig. 8a). But this theory was contradicted directly by the phenomenon at the high O_3 stations where a 30% reduction in $PM_{2.5}$ (red line of Fig. 8c) corresponded to a decrease rather than an increase in O_3 (red line of Fig. 8a)."

(6) Section 3.3 is not convincing as well. It should try to explain what weather system contributes to more ozone consecutive day from 2015 (as authors state that it drives the ozone increase), but figures are not helpful for this purpose. Figure 11 only shows that meteorological parameters can explain some of the ozone variability, Figures 12-13 show that WPSH can influence weather patterns, but is there any hint of an increasing influence of WPSH on consecutive ozone days? It might be useful to first clarify the weather patterns for consecutive ozone days, and explain what system can explain an increase in the frequency of such weather pattern during 2015-2020.

Response:

We take this comment to heart and have made an extensive revision of Section 3.3 as follows. First, as shown in our response to your major question (1), a new section 3.3.1 on "Changing emissions as a possible cause of O_3 trends in 2015–2020" has been added. Second, in our response to your "key concern" (4), we have added 39 lines (~750 words) of discussion and two figures (Figs. 12 and 13) near Line 300 "to first clarify the weather patterns for consecutive ozone days and explain what weather system contributes to more ozone consecutive day from 2015". Finally, we have added the following two paragraphs to Section 4 Summary and conclusions: "The trends in O_3 in the three megacity clusters are found to be critically dependent on the number of four or more consecutive O_3-exceeding days. In collaboration with this study, Hu W. et al. (2023) found that the changing frequency of mid-distance category TCs (i.e. changing meteorological conditions) is the cause of the increases in the numbers of consecutive O_3-exceeding days as well as the O_3 concentrations in PRD. Our additional analyses of the mean vertical velocity at 850 hPa in the three megacity clusters (Figs. 12, 13 and S10) show that the increases in O_3 in all three megacity clusters from 2015 to 2017 were associated with enhanced downdrafts and stable atmospheric conditions (meteorological conditions) which were highly conducive to O_3 formation. Finally, the enhanced downdrafts and stable atmospheric conditions were most likely brought about by TCs and associated WPSH.

Therefore, we propose that the O_3 concentrations at the high O_3 stations stayed close to a saturation level of about 100 ppb throughout 2015 to 2020, even under enhanced conditions conducive to O_3 formation, was the result of a relatively high rates of atmospheric dispersion, dry deposition and photochemical loss at the high O_3 concentration. While the low O_3 stations, where O_3 production were relatively small in 2015, experienced significant enhancements in the O_3 production in 2017 and 2019 because of the enhanced downdrafts and stable atmospheric conditions associated with TCs and WPSH in the northwestern Pacific, which were highly conducive to O_3 formation (Hu W. et al., 2023).".

We acknowledge that this study, like most other investigations, is an ongoing research effort. We report here some new results, but we don't have the definitive answers to all mechanisms that lead to the increasing "weather patterns for consecutive O_3 days" in the three megacity clusters. More studies are needed to address those questions.

Other comments

Line 100, Before Table 1, please consider introducing the purpose for such classification.

Response:

Thanks. we have revised the manuscript as follows to address the purpose of classification:

"Table 1 lists the criteria and corresponding numbers of low O_3 and high O_3 stations in the three megacity clusters. This classification is undertaken with the purpose of distinguishing stations with various O_3 levels within the three megacity clusters, and it is based on the number of O_3 exceeding days in 2015."

Line 116: missing ppb

Response:

Thanks. We have added "ppb".

Line 158-160. It is quite unclear what the calculation stands for, and how it leads to the conclusion that. Please clarify. Please also carefully clarify the rationale of other formulas.

Response:

In section 3.2, if we only compare the MDA8 O_3 average concentration on O_3 exceedance days between two years, the difference between 2017 and 2015 is only 3.02 ppb. However, considering that in 2015 there were only 31 days with O_3 exceedances, while in 2017 there were 62 days, characterizing the severity of O_3 exceedances between the two years solely based on the MDA8 O_3 average concentration on exceedance days is clearly insufficient. Therefore, we also take into account of the number of O_3 exceedance days and calculate a normalized MAD8 O_3 concentration by multiplying the MDA8 O_3 on exceedance days by the number of exceedance days and dividing by the total number of days in each year. When we compare the normalized MAD8 O_3 concentration for 2017 and 2015 (Fig. 2, red lines), we obtain a ratio of 2.09 between the two years.

To make it clearer, we have revised the corresponding statements as follows:

"The daily average concentration of MDA8 O_3 within the BTH box increased from 66.42 ppb in 2015 (31 days, Fig. 5a) to 69.44 ppb in 2017 (62 days, Fig. 5b), which was a difference of 3.02 ppb or a merely 4.5% increase between the two years (Fig. 5c). After accounting for the number of O_3-exceeding days, the ratio of normalized MDA8 O_3 in all O_3-exceeding days between 2017 and 2015 became 2.09. This comparison suggests together with those shown in Fig. 3a suggest that the increase in O_3 in BTH between 2015 and 2017 was driven primarily by the increase of consecutive O_3-exceeding days."

The statements pertaining to YRD and PRD have been adjusted accordingly.

Figure 3: Should "episodes" be a better word compared to "days" for the y-axis, since the variables plotted are "days"?

Response:

Thanks. We have added "Episode" to the title.

The expression needs to be improved, and the use of words needs re-consideration. For example, what does "quasi-saturation" stand for? Please revise "Same as Figure 5 except for YRD" to "Same as Figure 5 but for YRD". Please carefully check others.

Response:

Thanks. We have replaced some of the expressions, and revised “Same as Figure 5 except for YRD” to “Same as Figure 5 but for YRD” and others.

In this study, “quasi-saturation” refers to the phenomenon that O_3 concentration becomes nearly saturated and stops increasing after reaching certain level (approximate 100 ppb). Since both reviewers question the word “quasi-saturation”, we change it to simply “saturation”.

Figure quality can be improved, by increasing resolution and avoiding contours on shadings if both represent the same variable (Figs 5-6)

Response:

Thanks. We have revised all figures in the revised manuscript.

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