

Reviewer #2

Comment [2-1]: Major comments: This paper presents a large dataset of the dynamics of nocturnal O₃ in China with a special emphasis on the frequency with which nighttime concentrations are observed to increase (nocturnal ozone enhancements or NOEs). While I think there are useful results here, the paper would benefit from substantial editing to improve clarity and conciseness. I recommend publication after the following comments have been addressed.

Response [2-1]: We thank the reviewer for the positive and valuable comments. All of them have been implemented in the revised manuscript. Please see our itemized responses below.

Comment [2-2]: I recommend reframing the motivation behind this study. As written the introduction seems to argue that these NOEs are interesting because of their effects on health (of humans, plants, etc) but, at least in China, the maximum levels observed during NOEs are really very low (17 ppb in winter, 37 ppb in the summer) for health effects, especially as they occur during a time of day when most humans are asleep indoors and plants are more dormant. Do you have references that show those concentrations being associated with negative health outcomes? I don't mean to say at all that we shouldn't try to understand the full daily cycle of O₃ and the effects of vertical mixing on nighttime levels, just that it rings hollow to present it as if the NOEs themselves are a major source of concern. I also found it a bit odd that there seems to be more focus given to the frequency of occurrence of increases than to the concentrations themselves.

Response [2-2]: Thank you for the suggestion. We have reframed the second paragraph of the introduction to highlight the implication of NOE events to atmospheric dynamics and chemistry: “Analyses of the nocturnal ozone enhancement events and associated ozone peaks have important implications for understanding nocturnal atmospheric dynamics and chemistry, as well as ozone exposure to human and vegetation health. Due to the lack of nighttime chemical source, elevated nocturnal ozone levels are thought to be driven by enhanced transport or atmospheric mixing, which can be indicative of atmospheric dynamic processes such as the boundary-layer low-level jets (Klein et al., 2014). The enhanced nighttime ozone then reshapes the ozone diurnal cycle, and may increase daily integrated ozone exposure time to human and vegetation that threatens human health (Turner et al., 2016; Fleming et al., 2018) and crop yields (Yue et al., 2017; Lefohn et al., 2018; Feng et al., 2022; Li et al., 2022). Enhanced nocturnal ozone can also increase the oxidation capacity by stimulating the nitrate radical formation (Wang et al., 2021), and promote the formation of secondary pollutants such as particulate nitrate and secondary organic aerosols (Brown and Stutz, 2012; Huang et al., 2020). It can further affect atmospheric chemistry of the following day (Millet et al., 2016; Caputi et al., 2019; Zhao et al., 2020). While long-term trends of mean nighttime ozone level have been extensively studied as an important metric for assessing ozone air

quality and emission changes (Cooper et al., 2012; Lu et al., 2020), the more episodic nocturnal ozone enhancement event has been underappreciated despite its unusuality and important implication.”.

Reference

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Comment [2-3]: Similarly, I don't find the lengthy discussion of differences in frequency of occurrence of nocturnal ozone increases between the US, Europe and China compelling. In the US and Europe there are generally lower daytime O₃ peaks (which leaves a smaller enhancement in the residual layer) and, I think, fewer nocturnal NO emissions (which results in less complete titration at night) so that the difference between the surface and the residual layer is less stark and mixing has a smaller effect. What really matters is probably the 24 hour integrated O₃ exposure, which is much higher in most of China than in the EU or US, possibly exacerbated by trends of increasing NOEs over time? But not by a ton (at least not yet since the nocturnal O₃ is so low). And the nighttime exposures in the EU and US are comparably more important because the nighttime O₃ levels are higher and the daytime peaks lower (generally). You do eventually get there towards the end of the manuscript but I think the whole paper would be improved if you discuss the logical explanations for these broad differences when you are describing the observed distributions.

Response [2-3]: Thank you for pointing it out. We attempted to organize the paper into two parts: characteristics and mechanism. The higher frequency NOE event (Figure 1a) in China than in US and Europe is a new finding that motivates us to probe into the mechanism. Then we do not start the discussion with Figures 1b-d because they are closely related to the mechanisms, instead we move to Figures 2-4 for a comprehensive description of NOE statistics (spatial pattern, seasonal difference, peak value, and evolution of other pollutant), which also provides hints on the NOE mechanisms. Sections 3.2 and 3.3 focus on the mechanism and include detailed discussion on Figures 1b-d. We believe this organization is easier for readers who are not familiar with these unusual NOE

events. Showing Figures 1b-1c with Figure 1a allows a direct comparison between NOE frequency and ozone level, but indeed it confuses the logics. We further clarify this in the text “As shown in Figures 1b and 1c, the spatial pattern of NOE event frequencies is closely related to the afternoon (14-17 LT) ozone and nighttime O_x (O₃+NO₂) concentrations measured at the surface. This feature has important implications for understanding the mechanism of NOE events, which will be analyzed in Section 3.2.”

We agree with your analyses on the low NOE frequency over Europe and US. We have added them in the text “In other regions over Europe and US, we see much lower NOE event frequencies on average. This is because the daytime peak ozone is relatively lower than those over China, leading to low nighttime ozone concentrations in the residual layer (as indicated by the afternoon ozone at surface of 30 ppbv or less), in addition nighttime NO emissions there are low, which contribute to weak titration effect (as indicated by the small difference between nighttime ozone and O_x level). As such, the ozone difference between the surface and the residual layer is less stark, and residual layer ozone cannot serve as an effective source to enhance ozone at the surface even there is strong mixing or transport.”

We have also discussed the influence of NOE event on integrated ozone exposure in the introduction [Response 2-2] and in Section 3.4 “While the enhanced nocturnal mixing between the residual layer and nighttime boundary layer contributes to nocturnal ozone enhancement at the surface, the enhanced ozone is also subject to more efficient chemical destruction and dry deposition, resulting in lower ozone peak values on the next day (Hu et al., 2013; Caputi et al., 2019). As such, whether NOE events would increase or decrease the ozone level and integrated ozone exposure in the following day is yet to be determined.”

Reference

- Caputi, D. J., Faloon, I., Trousdell, J., Smoot, J., Falk, N., and Conley, S.: Residual layer ozone, mixing, and the nocturnal jet in California's San Joaquin Valley, Atmos. Chem. Phys., 19, 4721-4740, 10.5194/acp-19-4721-2019, 2019.
- Hu, X.-M., Klein, P. M., Xue, M., Zhang, F., Doughty, D. C., Forkel, R., Joseph, E., and Fuentes, J. D.: Impact of the vertical mixing induced by low-level jets on boundary layer ozone concentration, Atmospheric Environment, 70, 123-130, <https://doi.org/10.1016/j.atmosenv.2012.12.046>, 2013.

Comment [2-4]: Also related to point #2 above, I appreciate the usage of odd oxygen in your analysis but it seems like an afterthought right now and I think you should introduce it earlier (for example, it could logically be used in discussing the observed differences in nocturnal O₃ behavior between the EU, the US and China.)

Response [2-4]: We agree. We have moved Figure S4 to Figure 1 to show the O_x

pattern. Please also kindly find our revision as stated in [Response 2-3].

Comment [2-5]: NNOE (non-nocturnal ozone enhancement) is a weird acronym because it sounds like it should be an enhancement that happens during the day rather than a lack of an enhancement at night. Perhaps “non-enhanced nocturnal ozone” or “stable nocturnal ozone event” would work better?

Response [2-5]: Thank you for pointing it out. We have adopted the “non-enhanced nocturnal ozone” as the full term for NNOE in this paper.

Comment [2-6]: I would encourage the authors to think about whether certain points could be made using correlation plots rather than color-scaled maps that the reader must qualitatively compare. I had to do a lot of scrolling back and forth to see some of the trends that were being described. The top rows of Figures 1 and 2 make sense as color-scaled maps but when you are trying to compare NOE frequency to nocturnal ozone concentrations and subsequent-day afternoon O₃ I think those would be much better communicated by correlations. Actually the relationship between NOE frequency and afternoon ozone is less direct than looking at, for example, peak O₃ in a NOE compared to O₃ from the day before (or the following day). Why not plot those correlations instead? Similarly, I find that Figure 3 takes more effort than it should to look at. Would it communicate the same thing if you showed a single map that was colorscaled by the mean enhancement observed for evenings on which an NOE occurred? I believe the main point is that the sites that have the most frequent NOEs also experience the largest O₃ increases when they occur. Or perhaps that could also be a correlation plot.

Response [2-6]: Thank you for pointing it out. We have added Figure 5 (Figure R1) to quantitatively determine the relationship between NOE frequencies, afternoon ozone, and nighttime O_x. We now state in the text “Figures 1b and 1c present the mean afternoon ozone and nighttime O_x concentrations over China, Europe, and the US. Comparison of Figure 1a and 1b/1c reveals that the spatial pattern of NOE event frequencies, afternoon ozone levels and nighttime O_x level (both indicative of ozone in the nighttime residual level) are largely consistent. In particular, sites over China, Mediterranean, and mountainous western US with frequent NOE event recorded are consistently observing high afternoon ozone levels or nighttime O_x level. Figure 5 further shows that NOE frequency at Chinese sites increases with rising mean afternoon ozone level or nighttime O_x level, and is 10% (18%) higher when the afternoon ozone levels (nighttime O_x levels) exceed 50 ppb than when they are below 30 ppb.” We also derive a significant and positive linear correlation coefficient of 0.5 (*p*-value<0.01) between NOE frequencies and nighttime O_x level and 0.2 (*p*-value<0.01) between NOE frequencies and afternoon ozone level. However, we think that the value of linear correlation coefficient is somewhat misleading, because NOE is defined with a fixed threshold, so the relationship between its frequency and ozone level would not be linear. So we prefer to illustrate their relationship by comparing the mean NOE frequency at different ozone/O_x range

as shown in Figure R1. We reserve the map plot in Figure 1, because the spatial pattern itself convey additional information on where the hotspots of NOE frequency and ozone level are located.

Figure 3 aims to illustrate the frequency of different magnitudes of the NOE events at different regions in China, which may be a great concern for nighttime ozone air quality prediction. It is not for direct comparison between the NOE frequency and magnitude, so we do not prefer to replace it with a scatter plot. But we indeed find that sites that have the most frequent NOEs also experience the largest O₃ increases when they occur with a linear correlation coefficient of $r=0.4$. We have added the following text “We also find a significant positive correlation between NOE frequency and magnitude across 814 Chinese sites ($r=0.4$, p -value<0.01), indicating that sites with more frequent NOE events are more likely to experience larger nighttime ozone increase.”

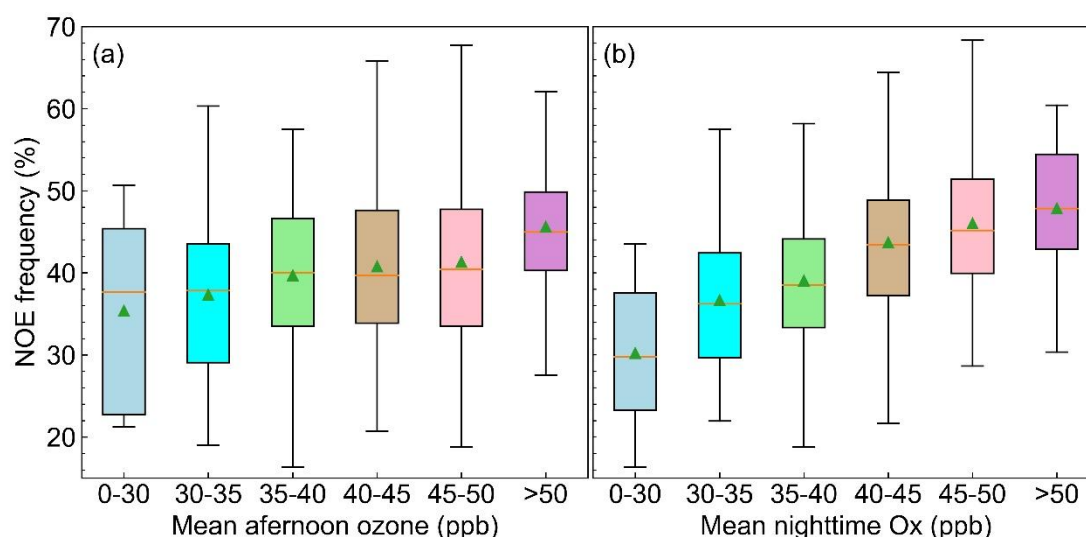


Figure R1 (Figure 5). The relationship between NOE frequencies and afternoon ozone (a) and nighttime O_x (b) at 814 Chinese sites. The colored box-and-whisker plots (5th, 25th, 50th, 75th, and 95th percentiles, and mean values denoted as triangles) show NOE frequencies at different concentrations of ozone or O_x.

Comment [2-7]: When looking at vertical profiles (eg section 3.2 and Figure 6), I think potential temperature might show your point better in terms of highlighting the altitude range that is being affected by cooling at the surface.

Response [2-7]: Thank you for the suggestion. We have used the profile of potential temperature to replace temperature, supporting the enhanced atmospheric vertical mixing in the NOE events. The new Figure R2 (Figure 7) is shown below. We have revised the text accordingly.

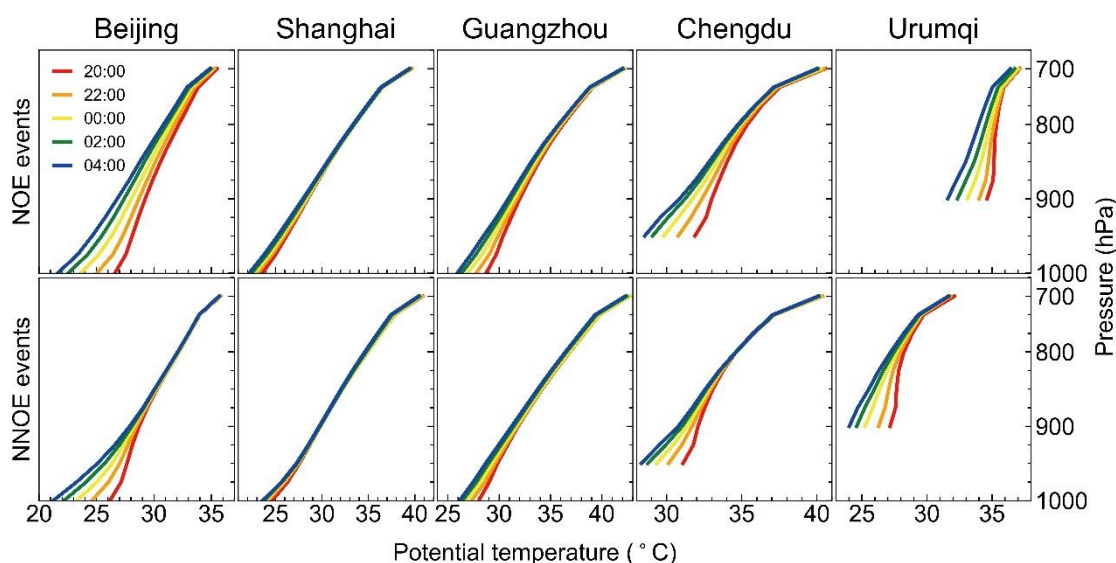


Figure R2 (Figure 7). Comparison of vertical profiles of potential temperature at five typical cities between the NOE (the top panels) and the NNOE (the bottom panels) events. The colored lines represent potential temperature profiles at different time of the night. The temperature data are from the ERA5 dataset.

Comment [2-8]: I wonder if you have considered the effects of reactions between NO_2 and O_3 to form NO_3 and N_2O_5 in the surface layer? NO_3 is quite reactive and N_2O_5 has a high deposition velocity so it could be an appreciable fraction of the observed nighttime O_3 decreases. I would consider it part of “ NO_x titration of O_3 ” but I don’t think I saw this process mentioned explicitly anywhere. If it has not been considered it certainly should be. NO_3 is highly reactive and N_2O_5 deposits very easily so they both could be substantial as a nocturnal O_x loss.

Response [2-8]: Yes, we have. Our original text described the effects of reactions between NO_2 and O_3 to form NO_3 and N_2O_5 in Line 206. We now also add it to Line 192: “This is reasonable because the decreasing rate of ozone is fast at early night, due to the rapid chemical loss through NO_x titration ($\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$, $\text{NO}_2 + \text{O}_3 \rightarrow \text{NO}_3 + \text{O}_2$, $\text{NO}_3 + \text{NO}_2 + \text{M} \rightarrow \text{N}_2\text{O}_5 + \text{M}$) when ..”.

Comment [2-9]: Does it matter that sunrise and sunset is at a different time of day across different latitudes (or between the cold and the warm season)? It seems like defining nighttime in terms of clock time rather than solar time could bias things, especially increases that are observed in the early morning in the summer when you might have sunlight for the beginning of commuting time. I’m thinking especially of the NOE events assigned to have happened between 8 and 9 pm and 4 and 5 am.

Response [2-9]: Thank you for pointing it out. We did not consider solar time in the analyses. The solar time for a given site may differ from the actual clock time by as much as plus or minus 45 minutes at a given time of year. Based on ozone time series shown in Figure 4, we find that ozone enhancement between 8 and 9 pm and 4 and 5 am is of relatively low frequency, suggesting that difference in solar time and clock time should not exert a large impact on our analyses. We

thus prefer to stick to the clock time (local time) in our analyses for easier communication to readers.

Comment [2-10]: I recommend trying to cut down on the figures that accompany the case studies. I don't think the main finding, that vertical mixing can largely explain the observed NOEs is particularly controversial so I think it should be sufficient to describe briefly the particular instances that were investigated and the consistency between them but I don't think this requires the 1-2 figures per event that are currently shown.

Response [2-10]: Thank you for the suggestion. We have moved the Figure 8 and Figure 10 to the supplementary text and revised the text accordingly.

Comment [2-11]: MINOR issues: In the abstract, I was initially confused about what an annual mean frequency of 41% meant. After reading I believe that you calculate the annual frequency of NOEs for each site and then average across all sites. While I think changing to "mean annual frequency" would be slightly clearer, I would encourage the authors to also describe this number in slightly more detail to make things easier on the reader as I started wondering early on which locations were used for each dataset. I would include a reference to S1 (the map of locations) around line 92 rather than only in the paragraph before.

Response [2-11]: Thank you for pointing it out. We have revised the text as **"We find that the mean annual frequency of NOE events is 41±10% (i.e. about 140 days would experience NOE event per year) averaged over all 814 Chinese sites in 2014-2019, which is 46% larger than those over Europe and US."** We have also added a reference to Figure S1 when introducing the ozone data from Europe and the United States in Line 94.

Comment [2-12]: Figure 1: I don't see how the inset shows mean and standard deviation.

Response [2-12]: We used observations of all sites (N is the number of sites) in three regions to calculate the mean and standard deviation (mean ± standard deviation), respectively. We put them at the top of the figure. We have clarified in the figure caption **"The values of regional mean and standard deviations among the N sites are shown inset at the top of each figure (mean ± standard deviation)."**

Comment [2-13]: Figure 2: Please label the colorscale for panels c and d. It's in ppb I think? But with the upper one % and the lower one not it is a bit confusing. Same issue with the inset as for Figure 1. In general I think insets, unless they are simply zoomed in on a particular region of the larger panel, should have their own axis labels, otherwise they are very hard to interpret.

Response [2-13]: Thank you for pointing it out. We have revised accordingly.

Comment [2-14]: Figure 4: Error bars would be good. Some of those profiles don't look super different for NOEs and NNOEs. And I would recommend that you harmonize axes for all sites in figure 4 if possible.

Response [2-14]: Thank you for the suggestion. We have added shadings to represent standard deviation of hourly ozone. We do not apply the shadings to NO₂ and CO because it would be too crowded. We prefer to have different scales for axis because it allows a clearer examination of the evolution of air pollutants, while comparison across sites is not the focus of this plot.

Comment [2-15]: Line 228, I find this sentence confusing. I can see how surface layer O_x should be comparable to residual layer O_x. And surface layer O_x would be similar to residual layer O₃ if NO₂ were a small fraction of the residual layer O_x but do we know that a priori? Also, I don't think nighttime emissions of NO need be small for this relationship to hold because it simply converts O₃ into NO₂ on a one to one basis and thus conserves O_x.

Response [2-15]: Thank you for correction. We have revised this sentence accordingly in Line 244 as “Similarly we may use the surface nighttime O_x (O₃+NO₂) concentration as an indicator of ozone in the nighttime residual layer (Kleinman et al., 2002; Wang et al., 2018; Tan et al., 2021), assuming that nighttime titration is much stronger than the effect of deposition and regional transport.”

Reference

- Kleinman, L., Daum, P., Lee, Y.-N., Nunnermacker, L., Springston, S., Weinstein-Lloyd, J., and Rudolph, J.: Ozone production efficiency in an urban area, *Journal of Geophysical Research*, 107, 10.1029/2002JD002529, 2002.
- Tan, Z., Ma, X., Lu, K., Jiang, M., Zou, Q., Wang, H., Zeng, L., and Zhang, Y.: Direct evidence of local photochemical production driven ozone episode in Beijing: A case study, *Science of The Total Environment*, 800, 148868, <https://doi.org/10.1016/j.scitotenv.2021.148868>, 2021.
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Comment [2-16]: Figure 5 is another place where I think error bars would help. These differences look pretty big, I don't really understand how it can be that there isn't a significant difference in U* or PBLH between NOE and NNOE evenings. Need to look at figures from supplement.

Response [2-16]: The difference of U* or PBLH between NOE and NNOE evenings is more obvious in their evolution with time than the absolute value itself, which indicates increasing nighttime mixing. This is the reason for showing the relative ratio to 8 p.m. value in the main text. But we also have the plot of absolute values in Figure S4 and indeed the values are also larger in NOE events in most cities in particular for the late night. Please also kindly find our revision

in [Response 1-6]. We have attempted to add standard deviation of U^* and PBLH in Figure 6. An example for Beijing and Shanghai is shown below in Figure R3. The trends of U^* and PBLH are clear even accounting for the standard deviation. We find that adding the standard deviation to all the five cities makes to the plot too crowded to read. As such we prefer to reserve the current plot.

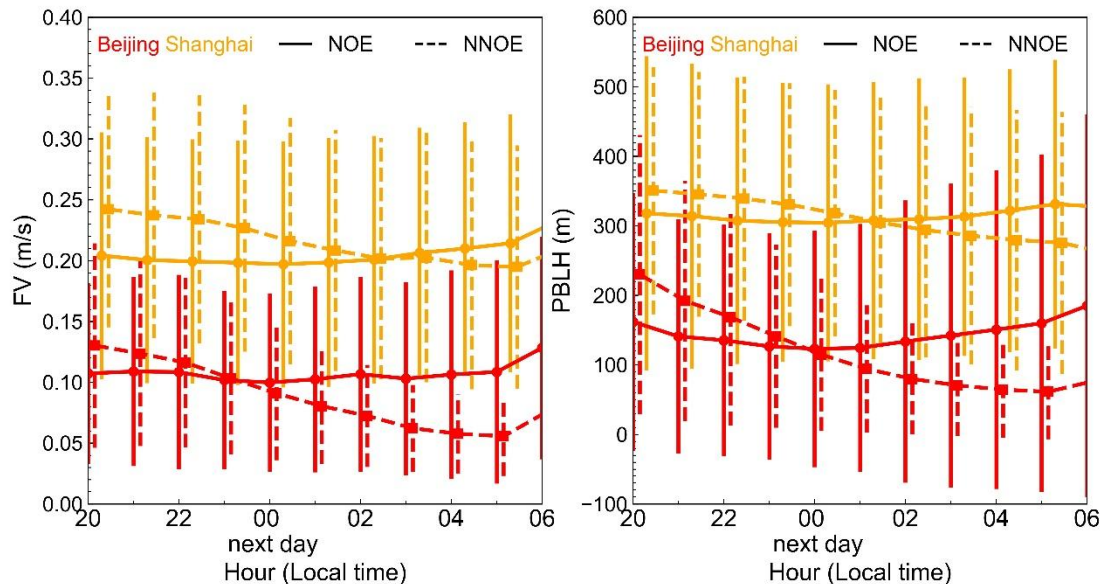


Figure R3. The absolute values of nighttime friction velocity (U^*) and planetary boundary layer height (PBLH) averaged over all NOE and NNOE events in Beijing and Shanghai. Error bars represent the standard error of U^* and PBLH at different time.

Comment [2-17]: Figure 6 – I believe this must be model data given the smoothness of the lines and your previous use of U^* and PBLH from the model. But I think it would be worth clarifying that here.

Response [2-17]: Thank you for pointing it out. The temperature, U^* and PBLH data are all from the ERA5 dataset as stated in Section 2.2. We have added this information in the figure caption for clarification.

Comment [2-18]: Your text goes right from Figure 9 to Figure 11. I recommend moving Figure 10 to wherever it is that you discuss it or removing it if it is not currently discussed in the text.

Response [2-18]: We have moved Figure 10 to the supplementary text following your suggestion.

Comment [2-19]: Figure S1: the red dots are nearly invisible. Recommend marking with stars or some other symbol that will stand out in both shape and color and making them a bit bigger.

Response [2-19]: We have enlarged the dots following your suggestion.

Comment [2-20]: Figure S2: The legend says that the inset shows the number of sites with positive trend but I don't really understand what I'm looking at. As displayed I don't think these are useful and, since I don't know what you're trying to communicate, I can't figure out how to help.

Response [2-20]: Thank you for pointing it out. We agree that this figure does not add convincing conclusion. We have removed this figure to avoid confusion.

Comment [2-21]: English language – quite a few instances including from the first few pages (but not limited to):

line 62, threat should be threaten

top of p3: only one or A few and A comprehensive view on (del “the”) general characteristics and mechanisms of (del “the”)...

Line 71: six years OF ozone...

Response [2-21]: Thank you for pointing it out. We corrected them accordingly.