Responses to Review #1

The manuscript by Yang et al. analyzes the meteorological characteristics and processes of severe ozone pollution events in four major cities in China using the GEOS-Chem model, and examines the historical and future changes of the ozone-favorable meteorological characteristics from ERA5 re-analyses and CMIP6 simulation results. The topic is within the scope of ACP. The methods are valid and the findings are valuable for understanding the historical and future change of ozone pollution in China. In general, it is a concise and well-written manuscript. I recommend publication after some clarification.

We thank the reviewer for the helpful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

1. Introduction: I recommend further introducing what is missing in the above studies and what this study adds on then.

Response:

Thank you for the suggestion. We have now added the novelty of this study as following:

"As mentioned above, many previous studies have examined the meteorological characteristics of O₃ pollution in China. However, they focused on O₃ pollution over limited regions in China in each study (e.g., the North China Plain, southern China). These studies only examined the meteorological characteristics of O₃ pollution in a short time period due to the lack of observational data and did not consider the historical and future trends of these meteorological factors. In this study, the meteorological characteristics conducive to severe O₃ pollution in several polluted areas of China, including the North China Plain (NCP), Yangtze River Delta (YRD), Sichuan Basin (SCB), and Pearl River Delta (PRD), are respectively investigated based on the observed surface O₃ concentrations, reanalysis data, and GEOS-Chem model simulations. Besides, the contributions from various chemical and physical processes inducing regional O₃ pollution are quantified using an integrated process rate (IPR) analysis method. The historical changes in these meteorological factors favoring severe O₃ pollution over 1980-2019 are provided. Moreover, variations in future meteorological patterns during 2021–2100 leading to severe O₃ pollution in China are presented under the sustainable and high forcing scenarios according to the multi-model data from the Coupled Model Intercomparison Project Phase 6 (CMIP6)."

2. Table 1: It is quite interesting to see that ozone chemical net production is negative in Chengdu and Guangzhou. This requires some further discussions. Showing some detailed patterns of these diagnostics terms might be helpful. Is it possibly due to the relatively coarse resolution of the simulation (2x2.5)? It might be helpful to discuss the possible influence of model resolution on model diagnostics.

Response:

We agree with the reviewer that the relatively coarse model resolution could cause biases in the model results and diagnostics, since that the SCB ($102.5^{\circ}-105^{\circ}E$, $30^{\circ}-32^{\circ}N$) and PRD ($110^{\circ}-115^{\circ}E$, $22^{\circ}-26^{\circ}N$) for calculating diagnostics only cover one and four grid boxes, respectively.

We have now discussed it as "Note that, the chemical production of tropospheric O_3 decreased in SCB and PRD during the severe polluted months. It could have been biased by the relatively coarse model resolution in this study (2° latitude × 2.5° longitude), since that the SCB and PRD for calculating the chemical and physical processes only cover limited grid boxes. Further studies should be performed using a model with finer resolution or a nested simulation method."

3. Figure 8 (and future projection) uses the spatial correlation of SLP and GPH for each year with that in the targeted ozone pollution month to examine the similarity of weather patterns. While this is mostly sound and efficient, I wonder whether the magnitude of SLP and GPH should be considered. In addition, it should be clear whether the correlation is calculated for each month in 1980-2020 to compare with the targeted ozone pollution month.

Response:

In the analysis of the meteorological characteristics conducive to severe O₃ pollution, temperature at 2m (T2m) and surface relative humidity (RH), sea level pressure (SLP) and geopotential height (GPH) at 500 hPa are adopted. For T2m and RH, the changes in the magnitude of these two variables are presented because they are related to the chemical production in a specific location. For SLP and GPH, the spatial correlation over East Asia and Western Pacific (EAWP, 90°-160°E, 20°-60°N) between the variable in targeted O₃ polluted month and the month in other years is calculated, because they are more related to the large-scale circulation over a broad region. Therefore, the number of high correlation years are more suitable for SLP and GPH rather than the magnitude. This method for SLP and GPH has been used in many previous studies (Li et al., 2018; Yang et al., 2021). Also, accompanied with global warming, the magnitude of SLP and GPH could also change with time. We have added the references in the manuscript. The correlation is calculated for the same month as the targeted month in 1980-2020 to compare with the targeted ozone pollution month. We have clarified as "Similar to the analyzing method used in previous studies (Li et al., 2018; Yang et al., 2021), the SLP and 500 hPa GPH over East Asia and Western Pacific in the same month of each year similar to those during the severe polluted months in both SCB and PRD have increased (2000-2019 versus 1980-1999)".

4. Line 161: misspelling of "simulation"

Response:

Revised

References:

Li, K., Liao, H., Cai, W., and Yang, Y.: Attribution of anthropogenic influence on atmospheric patterns conducive to recent most severe haze over eastern China, Geophys. Res. Lett., 45, 2072–2081, https://doi.org/10.1002/2017gl076570, 2018.

Yang, Y., Zhou, Y., Li, K., Wang, H., Ren, L., Zeng, L., Li, H., Wang, P., Li, B., and Liao, H.: Atmospheric circulation patterns conducive to severe haze in eastern China have shifted under climate change, Geophys. Res. Lett., 48, e2021GL095011, https://doi.org/10.1029/2021GL095011, 2021.

Responses to Review #2

The authors analyzed meteorological anomalies in June 2018, July 2017, September 2019 and July 2015 corresponding to severe O_3 pollution events in the NCP, YRD, PRD and SCB, respectively. The four meteorological variables (T2M, RH, SLP and GPH) which are associated with severe O_3 pollution events in these four months are then analyzed to investigate their changes in 1980-2019 and 2021-2100, and their changes are predicted to lead to more severe O_3 pollution in the future. The topic is interesting, and the conclusion is important for better control of O_3 pollution. However, there are still issues that need to be addressed before the paper can be considered for publication.

We thank the reviewer for the helpful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

Comments:

Lines 28-31: The readers may assume the conclusion is based on the most severe O3 pollution months over these regions, but they are June 2018, July 2017, September 2019 and July 2015 for the NCP, YRD, PRD and SCB, respectively, that is, one month per region. It is unclear whether one month can represent the meteorological conditions of severe O₃ pollution events sufficiently.

Response:

Thank you for the comment. Many previous studies have analyzed the synoptic patters of regional O_3 pollution in China and found several meteorological conditions could lead to O_3 pollution. In this study, we focus on the most extreme O_3 pollution cases in many regions of China rather than the high O_3 pollution cases in a particular region. The similar scientific question and method have been applied in aerosol pollution studies (e.g., Li et al., 2018; Yang et al., 2021). We admit that other meteorological conditions besides the ones we analyzed in the manuscript can also cause regional O_3 pollution. However, these high pollution cases are much more complicated and are suit for further analysis in the individual regions.

We have now added a statement in the discussion as "In addition, this study focuses on the most extreme O_3 pollution in several polluted areas of China. However, many other meteorological conditions can also cause O_3 pollution, although they may not be as extreme as the cases analyzed in this study, which requires comprehensive analysis for individual regions in future studies." We have also revised the description in the abstract to "During the most severe O_3 polluted months, the chemical production of O_3 is enhanced under the hot and dry conditions over the North China Plain (NCP) in June 2018 and Yangtze River Delta (YRD) in July 2017, while the regional transport is the main reason causing the severe O₃ pollution over Sichuan Basin (SCB) July 2015 and Pearl River Delta (PRD) in September 2019."

Furthermore, the severe O_3 pollution events in the SCB are interpreted as regional transport from Northern China (as shown in Figure 5b). Consequently, meteorological variables SLP and GPH were selected to describe severe O_3 pollution in the SCB. However, it is hard to believe regional transport from Northern China could be the dominant factor for severe O_3 pollution events in the SCB which has strong local anthropogenic emissions and is surrounded by mountains. In lines 160-162, the authors indicated "In the top three O_3 polluted months in Chengdu, only in July 2015 the higher concentrations than the long-term averages can be captured by the simulations with fixed emissions", and it is the reason why July 2015 was selected to represent the SCB in the analysis. However, does this imply inaccuracy in the model simulations to interpret severe O_3 pollution events in the SCB? Are SLP and GPH suitable meteorological variables to describe severe O3 pollution events in the SCB in the past and the future?

Response:

In this study , we focus on the meteorological characteristics of severe ozone pollution events in different regions in China. For the GEOS-Chem simulations, the precursor emissions of O_3 are fixed to minimize the influence of interannual variations in emissions. Therefore, when comparing to the observations, the differences between the model and observations are partly attributed to the emission changes. But the possible model bias can also contribute to the difference.

We have now added a paragraph in the last section to discuss the polluted month in SCB, as "There are some limitations and uncertainties in this work that can be further addressed in future studies. For example, the model only captures the high O_3 concentrations in July 2015 in Chengdu among its top three polluted months. It is probably because the emissions are kept at 2017 levels during the simulations. The high O_3 anomalies in July 2016 and July 2018 are more likely influenced by the interannual changes in local precursor emissions in the background of country-level increases in O_3 concentration in recent years. However, we also can not rule out the possible inaccuracy in the model simulations to interpret severe O_3 pollution events in the SCB, which deserves further investigation with multi-model simulations."

To date, several studies have pointed out the importance of regional transport as the cause of O_3 pollution in SCB (Qiao et al., 2019; Shen et al., 2022; Shu et al., 2023; Yang et al., 2020). Shen et al. (2022) examined the source-receptor relationships of O_3 in China and showed that O_3 received by SCB from other areas in central and eastern China could reach 26.7%. Shu et al. (2023) investigated the mechanism of O_3 pollution at different altitudes in the western part of SCB and found that the O_3 concentration at the top of the mountain due to the regional transport. Yang et al. (2020) explored the formation and evolution mechanism of O_3 pollution in SCB by using the WRF-CMAQ model simulations and also reported that the high ozone concentration in Chengdu during one O_3 pollution case was mainly due to mixing and regional transport of O_3 precursors through vertical mixing and horizontal advection from upwind areas. Therefore, the regional transport can lead to O_3 pollution in SCB driven by anomalous atmospheric circulation pattern, which is sometimes characterized by SLP and GPH.

Lines 152-155: Why is Chengdu not mentioned in this sentence?

Response:

Added as "The top three highest frequencies of O₃ pollution days in Chengdu are in July 2016, July 2015 and July 2018 (16, 15 and 15 days per month, respectively)."

Figure 2: How are these anomalies calculated?

Response:

We have revised the figure caption to "**Figure 2.** Spatial distribution of monthly O_3 concentration anomalies (part per billion, ppb) in June 2018 (a), July 2017 (b), July 2016 (c), July 2015 (d), July 2018 (e) and September 2019 (f) relative to 40-year (1980–2019) monthly average for June (a), July (b, c, d, e) and September (f), simulated in the GEOS-Chem model. The green boxes mark NCP (a), YRD (b), SCB (c, d, e) and PRD (f)."

Figure 3a: Is it 850 hPa (figure title) or 1000 hPa (caption) wind anomalies? The same question for Figures 4-6.

Response:

We apology for the typo. It should be 850 hPa. Revised now.

Section 3.2: The discussion in this section is simple. It could be better to provide a quantitative comparison of the impacts of each meteorological variable on the high O_3 .

Response:

The impact of each meteorological variable on O_3 pollution has been fully documented in the introduction. For the quantitative analysis, we used the integrated process rate (IPR) analysis from GEOS-Chem simulations to examine the possible physical and chemical mechanisms for the O_3 pollution and discussed the meteorological characteristics related to the changes in O_3 mass. Although each meteorological variable could exert different impact on O_3 pollution, it may not be appropriate to isolate the relative role of each meteorological variable, since many meteorological variables have internal relations and change simultaneously.

Figure 8c-d: These two panels are associated with the conclusion "In SCB and PRD, the occurrence of atmospheric circulation patterns similar to those during the polluted months increased, together with the more frequent hot and dry conditions, contributing to the increases in severe O₃ pollution in SCB and PRD during 1980–2019". First, it is suggested to add uncertainty estimates particularly for July 2015 (SCB) to determine whether there are significant trends in the meteorological anomalies. Moreover, is it

possible to have quantitative estimations for the possible contributions from meteorological changes to O_3 changes in 1980-2019 to confirm the importance of meteorological changes?

Response:

For the historical changes in SLP and GPH, we did not draw a conclusion that there are clear trends of historical SLP and GPH that are similar to those in the polluted months, but only the changes in frequencies between two time period (1980–1999 vs. 2000–2019) based on meteorological reanalysis. The analysis of frequency trend requires much longer time data than 40-year during 1980–2019. Any uncertainty here can only come from the uncertainties in the reanalysis data itself, which is not the focus of this study.

The historical changes in the meteorological patterns causing severe O_3 pollution are in accordance with the elevated O_3 levels in China in the recent decade. However, the quantitative analysis of meteorological impacts needs full consideration of factors leading to O_3 pollution, including changes in anthropogenic and natural emissions of its precursors, O_3 chemical regime, other meteorological factors conducive to O_3 pollution, and stratosphere-troposphere exchange. We have added this discussion in the last paragraph of the manuscript.

Section 7: It could be helpful to provide a comparison of the impacts of meteorological changes in 1980-2019 and 2021-2100 to extend the discussion in this section.

Response:

Thank you for the suggestion. We have now added the discussion as "Unlike the historical changes in the meteorological conditions that caused the severe O_3 pollution through chemical production and regional transport, future variations in meteorological conditions conducive to the severe O_3 pollution are more related to the global warming process that enhances the O_3 production in China."

Reference

- Li, K., Liao, H., Cai, W., and Yang, Y.: Attribution of anthropogenic influence on atmospheric patterns conducive to recent most severe haze over eastern China, Geophys. Res. Lett., 45, 2072–2081, https://doi.org/10.1002/2017gl076570, 2018.
- Yang, Y., Zhou, Y., Li, K., Wang, H., Ren, L., Zeng, L., Li, H., Wang, P., Li, B., and Liao, H.: Atmospheric circulation patterns conducive to severe haze in eastern China have shifted under climate change, Geophys. Res. Lett., 48, e2021GL095011, https://doi.org/10.1029/2021GL095011, 2021.
- Qiao, X., Guo, H., Wang, P. F., Tang, Y., Ying, Q., Zhao, X., Deng, W. Y., and Zhang, H. L.: Fine Particulate Matter and Ozone Pollution in the 18 Cities of the Sichuan Basinin Southwestern China: Model Performance and Characteristics, Aerosol Air Qual. Res., 19: 2308-2319, <u>https://doi.org/10.4209/aaqr.2019.05.0235</u>, 2019.

- Shen, L. J., Liu, J. Zhao, T. L., Xu, X. D., Han, H., Wang, H. L., Shu, Z. Z.: Atmospheric transport drives regional interactions of ozone pollution in China, Sci. Total Environ., 830, 154634, <u>https://doi.org/10.1016/j.scitotenv.2022.154634</u>, 2022.
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Responses to Review #3

Ozone pollution is becoming a growing challenge in China, and the meteorology plays a significant role in how it changes. The study by Yang et al. investigated the meteorological characteristics during the high ozone months in four polluted cities in China. They also looked at how these meteorological factors have changed in the past and might change in the future, thereby providing implications for ozone control strategies. The topic is clear and interesting, and the paper is well organized and easy to follow. The results emphasize to the community that future climate warming could exacerbate ozone pollution in China.

We thank the reviewer for the helpful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

The one difficulty I have with judging the relevance of the findings is that they only analyzed one specific month with extremely high ozone in each region, and all the subsequent statements rely on the meteorological conditions prevalent during those specific months. It seems potentially non-representative. The paper would be much stronger if the authors would evaluate all the high ozone months for each region or convincingly demonstrate to readers that the representativeness of the selected months.

Response:

Thank you for the suggestion. We agree with the reviewer that more analysis would be better for the understanding the meteorological characteristics of ozone (O_3) pollution in China. Many previous studies have analyzed the synoptic patters of regional O_3 pollution in China and found several meteorological conditions could lead to O_3 pollution. In this study, we focus on the most extreme O_3 pollution cases in many regions of China rather than the high O_3 pollution cases in a particular region. The similar scientific question and method have been applied in aerosol pollution studies (e.g., Li et al., 2018; Yang et al., 2021). We admit that other meteorological conditions besides the ones we analyzed in the manuscript can also cause regional O_3 pollution. However, these high pollution cases are much more complicated and are suit for further analysis in the individual regions.

We have now added a statement in the discussion as "In addition, this study focuses on the most extreme O₃ pollution in several polluted areas of China. However, many other meteorological conditions can also cause O₃ pollution, although they may not be as extreme as the cases analyzed in this study, which requires comprehensive analysis for individual regions in future studies." Specific comments:

1. The authors looked at ozone during April-September. Please consider extend it to October since the warm season is longer in PRD.

Response:

Thank you for the suggestion. We have now included October and the results do now change.

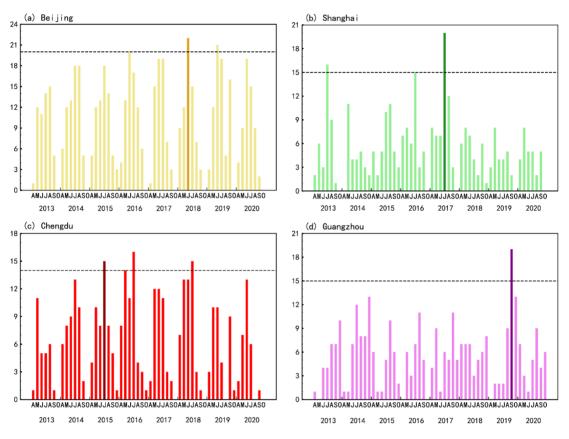


Figure 1. Time series of frequencies of severe O₃ pollution days (defined by daily maximum of 8-h average ozone (MDA8-O₃) concentration greater than 160 μ g m⁻³) in Beijing, Shanghai, Chengdu and Guangzhou (a–d) from April to October during 2013–2020. The dark-colored bars represent the most severe month (second most for Chengdu) that has the highest frequency of O₃ pollution days for the individual cities.

2. Figure 8, could the authors explain more on how they calculated the spatial correlation? Response:

The spatial correlation is calculated between SLP/GPH anomalies over East Asia and Western Pacific (EAWP, $90^{\circ}-160^{\circ}$ E, $20^{\circ}-60^{\circ}$ N) in the polluted month, June 2018 for example, and those over the same region in the target month (June as example) of each year during 1980–2020. The method has been widely used in determining the similarity of atmospheric circulation patterns. We have modified the description in the figure caption.

3. Line 161, "simulations" is misspelled.

Response:

Corrected.

4. Line 196, "northwesterly winds" is inconsistent with the following description "from the north and east".

Response:

Thank you for pointing out this typo. It has been corrected to "from the north and west".

5. Figure 2, please consider enlarging the font size.

Response:



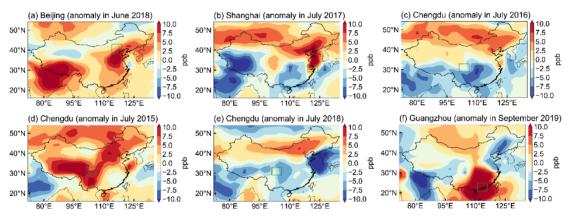


Figure 2. Spatial distribution of monthly O_3 concentration anomalies (part per billion, ppb) in June 2018 (a), July 2017 (b), July 2016 (c), July 2015 (d), July 2018 (e) and September 2019 (f) relative to 40-year (1980–2019) monthly average for June (a), July (b, c, d, e) and September (f), simulated in the GEOS-Chem model. The green boxes mark NCP (a), YRD (b), SCB (c, d, e) and PRD (f).

6. Figure 9, please consider labeling each subplot with its corresponding target region.

Response:

Added.

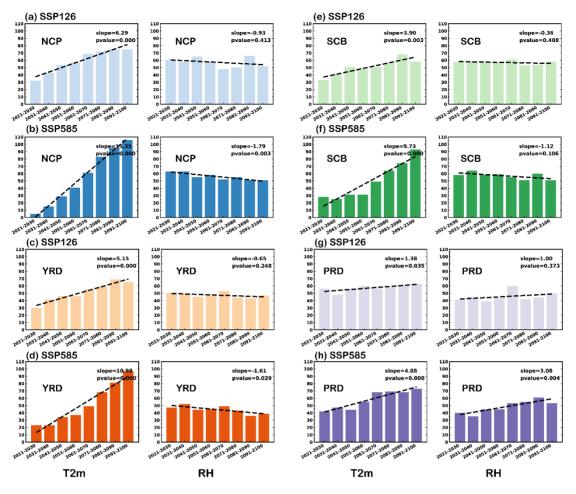


Figure 9. Frequencies of extreme months with T2m or RH anomalies exceeding the 80th percentile or below the 20th percentile of the distributions over NCP ($115^{\circ}-120^{\circ}E$, $38^{\circ}-44^{\circ}N$) (a, b), YRD ($120^{\circ}-125^{\circ}E$, $28^{\circ}-32^{\circ}N$) (c, d), SCB ($102.5^{\circ}-105^{\circ}E$, $30^{\circ}-32^{\circ}N$) (e, f) and PRD ($110^{\circ}-115^{\circ}E$, $22^{\circ}-26^{\circ}N$) (g, h) in each 10-year interval during 2021–2100 under two SSPs future scenarios of 13 CMIP6 models. The two SSPs are SSP1-2.6 and SSP5-8.5. The slope and P values of the linear regression during 2021–2100 are shown in the upper right of each panel.

References:

- Li, K., Liao, H., Cai, W., and Yang, Y.: Attribution of anthropogenic influence on atmospheric patterns conducive to recent most severe haze over eastern China, Geophys. Res. Lett., 45, 2072–2081, https://doi.org/10.1002/2017gl076570, 2018.
- Yang, Y., Zhou, Y., Li, K., Wang, H., Ren, L., Zeng, L., Li, H., Wang, P., Li, B., and Liao, H.: Atmospheric circulation patterns conducive to severe haze in eastern China have shifted under climate change, Geophys. Res. Lett., 48, e2021GL095011, https://doi.org/10.1029/2021GL095011, 2021.