Response: North China Plain as a hot spot of ozone pollution exacerbated by extreme high temperatures

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Jianping Tang, L. Ruby Leung, Hong Liao

Dear Editor,

We would like to submit our revised manuscript entitled "**North China Plain as a hot spot of ozone pollution exacerbated by extreme high temperatures**" to ***Atmospheric Chemistry and Physics.***

On behalf of my co-authors, we thank you for handling the peer review of our manuscript. We appreciate your time and efforts as well as those of the two referees for the careful reviews and constructive comments that have helped improve the quality and readability of the manuscript. We have carefully revised our manuscript to address the comments accordingly. Below are the point-to-point responses to the review comments.

Kind regards,

Key:

Black: Reviewer's comments

Blue: Author's responses

Reviewer #1:

This study investigates the co-occurrences of extremes in surface O_3 and extreme heat based on observation datasets, GEOS-Chem model simulations and latest CMIP6 outputs. Detailed analysis on historical and future projections of the coupled extremes as well as the health impact is discussed. The results represent the advances in understanding the interactions between extreme weather events and air pollution. In general, I find the manuscript well written and I recommend it for publication after addressing the following comments:

Reply: We thank the reviewer for the constructive comments and suggestions, which are very helpful for improving the clarity and reliability of the manuscript. Please see our point-by-point responses to your comments below.

Major Comments:

1. The section of model evaluation: I feel the discussions can be more elaborated (Supporting information), and a bit more detailed information such as mean bias, or fractional bias, etc., is useful to indicate more confidence in interpreting the simulated results.

Reply: Thanks for your constructive and helpful comments and suggestions. To improve the model evaluation part, we've added three more statistical metrics, including mean bias (MB), mean fractional bias (MFB) and root mean square error (RMSE) to quantitively evaluate the performance of GEOS-Chem model and CMIP6 simulations, based on the equations listed in the appendix of Zhang et al. (2018). The metrics have been shown in the updated Fig.S4 and Fig.S5 (shown as below).

Accordingly, we have revised Text S1 and Text S2 by adding more interpretations：

Text S1 (Line 29-36): "The spatial correlations between the simulated and observed OPCs and CF values are all higher than 0.5 and are statistically significant at 95% confidence level, accompanied by small mean bias (MB) and root mean square error (RMSE) values. For example, the MB between the simulated and observed OPCs and CF values over China are as low as 2.34 days and -0.23%, respectively. Moreover, the mean fractional bias (MFB) for CF values is well within the limit of MFB for O_3 evaluation (15%) recommended by EPA (2007). The statistical metrics suggest that the model can reasonably reproduce the observed spatial patterns and magnitudes of OPCs and CF over NCP during 2014-2017."

Text S2 (Line 50-54): "Similarly, the MFB and RMSE for both simulated OPCs and CF values under SSP3-7.0 are the lowest among the four scenarios. The relatively higher MB and RMSE under SSP2-4.5 come from the overestimation of OPCs and CF values over the whole China, likely related to the inaccurate of SSPs emissions in China during this time period (Chen et al., 2021; Wang et al., 2021)."

Cheng, J., Tong, D., Liu, Y., Yu, S., Yan, L., Zheng, B., et al. (2021). Comparison of current and future PM2.5 air quality in China under CMIP6 and DPEC emission scenarios. Geophysical Research Letters, 48, e2021GL093197. https://doi.org/10.1029/2021GL093197

Wang, Z., Lin, L., Xu, Y., Che, H., Zhang, X., Dong, W., Wang, C., Gui, K., and Xie, B.: Incorrect Asian aerosols affecting the attribution and projection of regional climate change in CMIP6 models, npj Clim. Atmos. Sci., 4, 2, https://doi.org/10.1038/s41612-020-00159-2, 2021.

Zhang, J., Y. Gao, K. Luo, L. R. Leung, Y. Zhang, K. Wang, and J. Fan (2018), Impacts of compound extreme weather events on ozone in the present and future, Atmospheric Chemistry and Physics, 18(13), 9861-9877.

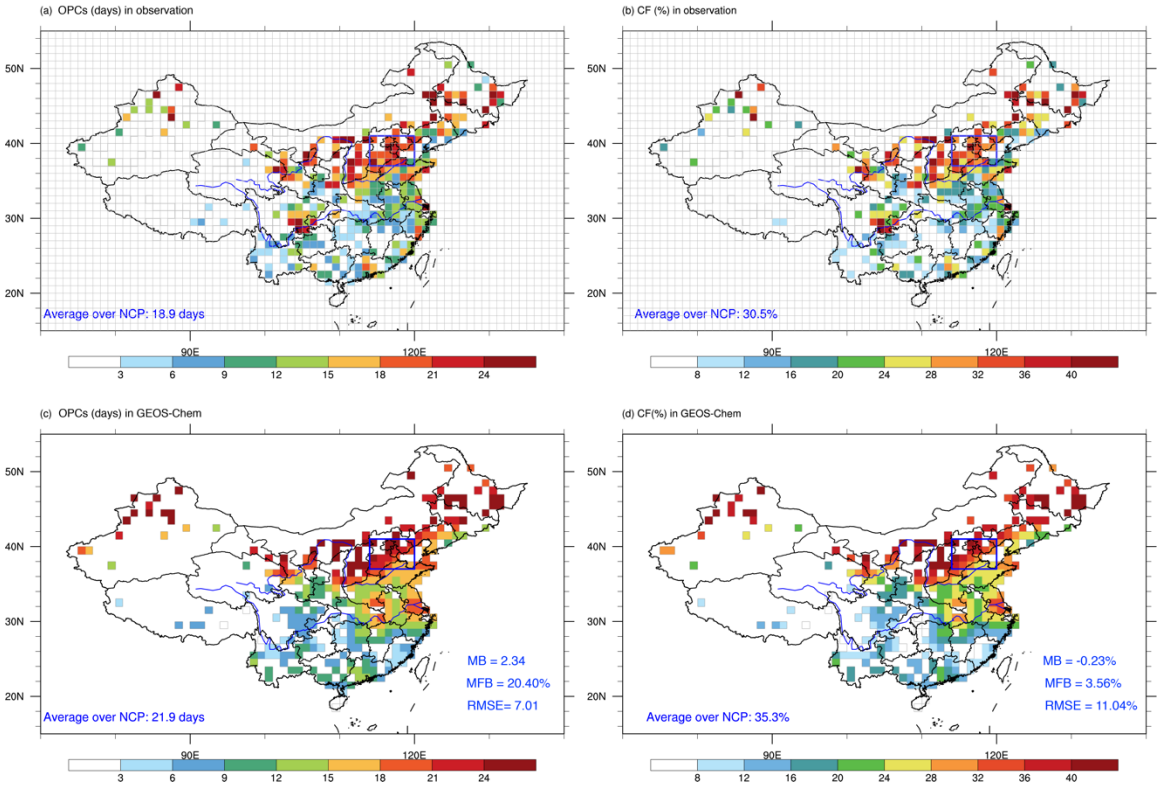

Figure S4. Spatial patterns of observed (a) OPCs (days) and (b) CF values (%) during May-September of 2014-2017. (c) and (d) are same as (a) and (b) but for the GEOS-Chem simulation. Observed and simulated values of OPCs(days) and CF averaged over NCP (37-41°N; 114-120°E) are indicated at the bottom left corner of each panel. Statistical metrics including MB, MFB, and RMSE are noted at the bottom right of panels (c) & (d). Note that the three metrics are obtained over the whole China, with equations listed in the appendix of Zhang et al. (2018).

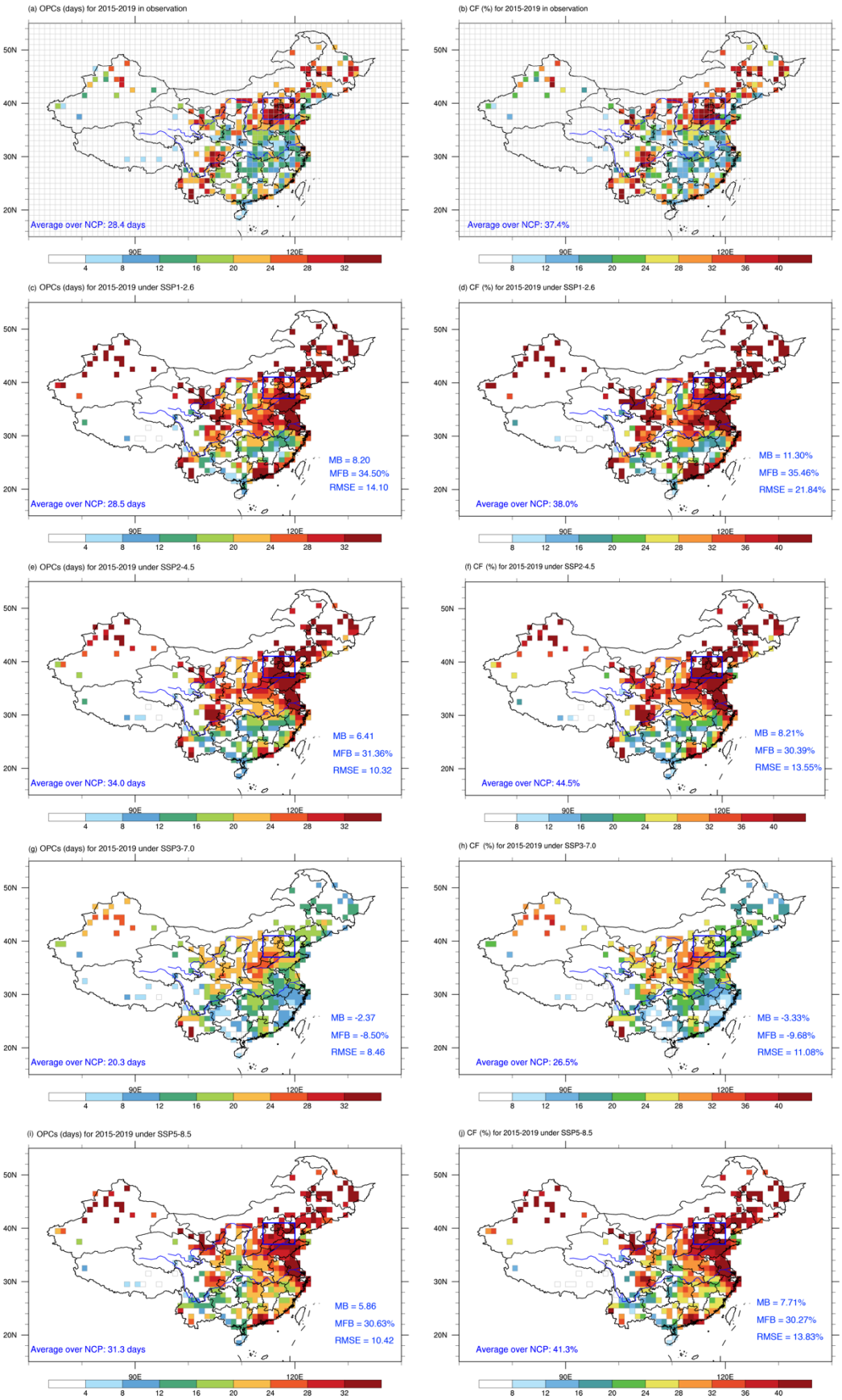
(a) OPCs (days) for 2015-2019 in observation
Average over NCP: 28.4 days
(b) CF (%) for 2015-2019 in observation
Average over NCP: 37.4%
(c) OPCs (days) for 2015-2019 under SSP1-2.6
MB = 8.20
MFB = 34.50%
RMSE = 14.10
Average over NCP: 28.5 days
(d) CF (%) for 2015-2019 under SSP1-2.6
MB = 11.30%
MFB = 35.46%
RMSE = 21.84%
Average over NCP: 38.0%
(e) OPCs (days) for 2015-2019 under SSP2-4.5
MB = 6.41
MFB = 31.36%
RMSE = 10.32
Average over NCP: 34.0 days
(f) CF (%) for 2015-2019 under SSP2-4.5
MB = 8.21%
MFB = 30.39%
RMSE = 13.55%
Average over NCP: 44.5%
(g) OPCs (days) for 2015-2019 under SSP3-7.0
MB = -2.37
MFB = -8.50%
RMSE = 8.46
Average over NCP: 20.3 days
(h) CF (%) for 2015-2019 under SSP3-7.0
MB = -3.33%
MFB = -9.68%
RMSE = 11.08%
Average over NCP: 26.5%
(i) OPCs (days) for 2015-2019 under SSP5-8.5
MB = 5.86
MFB = 30.63%
RMSE = 10.42
Average over NCP: 31.3 days
(j) CF (%) for 2015-2019 under SSP5-8.5
MB = 7.71%
MFB = 30.27%
RMSE = 13.83%
Average over NCP: 41.3%
50N
40N
30N
20N
90E
120E
4 8 12 16 20 24 28 32
8 12 16 20 24 28 32 36 40

Figure S5. Spatial patterns of (a) OPCs (days) and (b) CF values (%) during May-September of 2015-2019 in observation; (c)~(d), (e)~(f), (g)~(h), and (g)~(h) are same as (a) and (b) but for CMIP6 simulations under SSP1-2.6, SSP2-4.5, SSP3-7.0 and SSP5-8.5, respectively. OPCs (days) and CF averaged over NCP (37-41°N; 114-120°E) are indicated at the bottom left corner of each panel. Statistical metrics including MB, MFB, and RMSE are noted at the bottom right of panels (c)~(j). Note that the three metrics are obtained over the whole China.

2. In terms of the emissions: the authors only discussed anthropogenic emission inventory. How about biogenic emissions? Considering that biogenic emissions are quite important for ozone formation, particularly of the synergic effect of biogenic and anthropogenic emissions on ozone formation, it is useful to indicate how the biogenic emissions were treated in this study.

Reply: Thanks for your constructive and helpful comments and suggestions. We have added explanation on how the biogenic emissions in the updated manuscript:

Line 138-142: "Biogenic volatile organic compound (BVOC) emissions also play vital roles in modulating the formation of ozone and secondary organic aerosols (Ma et al., 2021; Y. Gao et al., 2021). For biogenic emissions in GEOS-Chem, the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 biogenic emissions are applied with updates from Guenther et al. (2012)."

Line 305-309: "In addition, Fu et al. (2015) have indicated that the enhanced biogenic emissions and the accelerated photochemical reaction rates both increased surface ozone over the US during 1988–2011. Thus, the increasing trend of biogenic emissions due to vegetation biomass variability over China (J. Gao et al., 2021) may also have potential impacts on the variations of OPCs."

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471–1492, https://doi.org/10.5194/gmd-5-1471-2012, 2012.

Ma, M., Gao, Y., Ding, A., Su, H., Liao, H., Wang, S., ... & Gao, H. (2021). Development and Assessment of a High-Resolution Biogenic Emission Inventory from Urban Green Spaces in China. Environmental science & technology.

Gao, Y., F. Yan, M. Ma, A. Ding, H. Liao, S. Wang, X. Wang, B. Zhao, W. Cai, H. Su, X. Yao and H. Gao (2021), Unveiling the dipole synergic effect of biogenic and anthropogenic emissions on ozone concentrations, Sci. Total Environ., 151722.

Cao, J., Situ, S., Hao, Y., Xie, S., & Li, L. (2021). Enhanced summertime ozone and SOA from biogenic volatile organic compound (BVOC) emissions due to vegetation biomass variability during 1981–2018 in China. Atmospheric Chemistry and Physics Discussions, 1-21.

3. About the impact of extreme events on ozone: the compound extreme events have recently been raised as a substantial concern to ozone formation. At least adding a few sentences or references to discuss the compound extremes (i.e., multiple extremes occur simultaneously) and the associated impact on ozone formation is useful.

Reply: Thanks for your constructive and helpful comments and suggestions. We have added more discussions in the Discussion and Conclusion part: "Recently, the compound extreme events (e.g., co-occurrence of two extreme weather events simultaneously) are raised as a substantial concern to O_3 formation. For example, the co-occurrences of heat wave and air stagnation promote higher O_3 concentration compared to the single extreme events of heat wave or stagnation in the U.S. in the future relative to the present (Zhang et al., 2018; Y Gao et al., 2020)."

Zhang, J., Gao, Y., Luo, K., Leung, L. R., Zhang, Y., Wang, K., & Fan, J. (2018). Impacts of compound extreme weather events on ozone in the present and future. Atmospheric Chemistry and Physics, 18(13), 9861-9877.

Gao, Y., J. Zhang, F. Yan, L. R. Leung, K. Luo, Y. Zhang and M. L. Bell, Nonlinear effect of compound extreme weather events on ozone formation over the United States (2020), Weather and Climate Extremes, 30, 100285.

Minor Comments:

1. Lines 80, 187, change "O3" to "O_3" and check throughout the entire text.
   Changed.
2. Line 208, change "MDA O3" to "MDA8 O_3".
   Changed.
3. Missing subtitle (b) in figure 2.
   Added.
4. Line 264, please be careful that the enhanced chemical production and weakened mixing and dry deposition contribute to the increase O_3 level during OPCs.
   Thanks. Modified.
5. Please use a larger font size in Figure 4 as the subtitle in each panel is hard to read. The same applies for Figure 5.
   Thanks. Both Figure 4 and Figure 5 are updated with a larger font size.
6. In terms of the health impacts of OPCs, have you considered the possible impacts of temperatures on surface ozone related health risk, i.e., higher temperatures may worsen the health impacts of surface ozone.
   As claimed in the manuscript (Line 359-362), previous studies have pointed out that O_3-related mortality may change with different air temperature levels, and yet the conclusions can be contrasting or inconsistent for different regions. Thus, this work does not consider the possible amplification/inhibition effect of combining O_3 and air temperature in affecting human health.
7. Line 212. Repeated definitions of abbreviation. An abbreviation is only needed with it

appears for the first time. Please double check the entire texts.

Thanks. Deleted.

8. As the author stated that GEOS-Chem simulations cover only the period of 2014-2017, does this mean that the definitions of OPCs and OPIs are applied to 2014-2017 for both observation and simulations? How about future?

   Thanks for your question. Yes, as addressed in the Text S1&S2, the GEOS-Chem simulations are conducted for 2014-2017. And the model simulations are evaluated based on observations during 2014-2017. Thus, both observed and simulated OPCs and OPIs are applied to 2014-2017. For the future projections, future OPCs during the mid-century (2046-2050) and end-century (2096-2100) are compared with OPCs during 2015-2019 for a consistency in time length.

9. The caption of Figure S3: downward solar radiation flux Does this mean downward surface solar radiation?

   Yes. We have made it clear in Sec. 2.1 of the updated manuscript (Line 120): "downward solar radiation flux (DSR) and sensible heat flux (SH) at surface."

10. Figure S8 includes some important information, and it is good to move it to the main manuscript.

    Thanks. Figure S8 has been put in the main manuscript and renamed as Figure 7 in the updated version.