Dear Editor(s),

We are submitting the point-by-point responses to the reviewer's comments. We thank the reviewer for the constructive comments and suggestions, and hope you are satisfied with our responses.

The major changes in the revised version include:

1. Correct model descriptions in the manuscript.
2. Improve Fig. 7b to ensure a better readability.
3. Add discussion about radiation uncertainties to aerosols (AOD and SSA) about Fig. 6b.

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Point-by-point responses to the Comments/Suggestions

This manuscript uses observations (including in situ and remote sensing data) and box model simulations to examine the trend of O_3 over North China Plains during 2013-2019 and its impacting factors including emissions, AOD, SSA, temperature and boundary layer height (PBLH). The contribution to O_3 formation from each impacting factor is quantified and found that reduction of emissions and aerosol radiative effects are the dominant factors that contribute to O_3 increase from 2013 to 2019. Such analysis help understand O_3 chemistry over NCP and help develop effective control strategies on reduction of ambient O_3. The manuscript is written well and clearly. This reviewer would recommend publication after addressing the following comments.

Specific comments:

1. The assumptions made in box model simulations need to be described more clearly. According to LN161, "This model computes time-dependent chemical evolution of an air parcel initialized with a known composition, assuming no additional emissions, no dilution and no heterogeneous processes", the model only takes initial concentration of chemical species, radiation and temperature and compute evolution of concentrations. It cannot directly quantify "the relative contributions of anthropogenic emissions and aerosol optical and radiative properties to the change in surface O_3" stated on LN151. A few assumptions must have been made. How does the model account for different NOx and VOCs emissions, and PBLH?

Thanks for pointing out the problem. Indeed, something was mixed up in the original description. NOx and VOCs emissions were changed to assess the impact of emissions on surface O_3 simulations (see Table 1). It is assumed that no dilution is included in the simulations given the difficulty of getting inputs to calculate the dilution rate. Dry deposition is included in all the simulations and sensitivity runs. PBLH was ingested as the model requests every 4 hours to support the calculation of entrainment term. The issue is corrected and can be found in the revised version.

LN187, using meteorological data at a 4-hour interval appears too coarse/crude to reproduce diurnal variation of O_3. Why not use hourly values?

Yes, theoretically, hourly input data could reproduce more reasonable diurnal variations in surface O_3 simulations. However, according to our tests, the computational time increased substantially when input data were ingested hourly. Meanwhile, the MM model was able to capture the diurnal variation pattern reasonably when the input data were ingested every 4 hours. Thus, we decided to ingest the input data every 4 hours for all the numerical experiments.

2. The diurnal variation of O_3 depends on the nighttime O_3 depletion due to NO titration and dry deposition and daytime O_3 formation after rush hour emissions. The box model does not account for dry deposition and additional rush hour emissions. It is a little surprising the box model can still capture the full diurnal variation of O_3 in Fig. 7.

Sorry for the confused. Please see our response to the 1st comment provided above. Dry deposition was included in our simulations. Such as information is included in the revised version.
With the emission inputs at an interval of 4 hours, the diurnal variation in surface O_3 was reproduced.

3. What is the purpose of sensitivity of jNO_2 to different solar zenith angle in Figs. 8 and 9? which should not vary from 2013 to 2019. In another word, solar zenith angle is not a factor for O_3 variation from 2013-2019.

We agree with the reviewer that the solar zenith angle is not a factor in driving the increase in surface O_3 from 2013 to 2019. The main purpose of Figures 8 and 9 is to examine the sensitivity of jNO_2, the daily 8-hr maximum O_3 and HO_2 radicals to the changes in AOD and SSA. Solar zenith angle is included in the figures for a comparison since both jNO_2 and HO_2 radicals are very sensitive to changes in solar zenith angle but show very different change trend with varying solar zenith angle. For instance, jNO_2 shows a nearly linear change with Sec (θ) wheras HO_2 doesn't. On the other hand, jNO_2 shows the largest sensitivity to AOD and SSA at noontime while HO_2 has a largest sensitivity around 3:00 pm local standard time which is consistent with the peak hour of surface O_3.

LN59, $TCNO_2$ was reported to be increased by 307% in Beijing from 1996-2011, but it decreased from 2013-2019 in Fig. 2. Are they consistent?

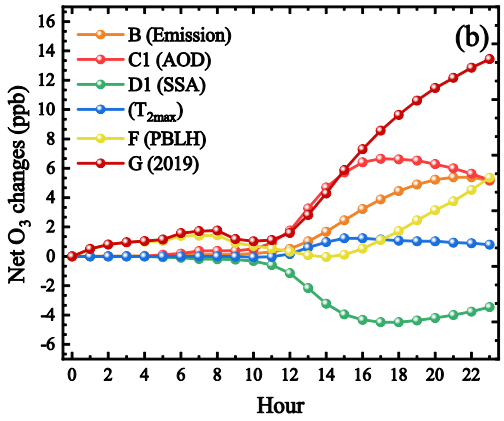
Yes, they are correct. The change trend was reversed around 2013 since a strict NOx emissions control measures were implemented in China (e.g., Huang et al., 2013; Zeng et al., 2019).

LN115 what is "ppb a-1"? ppb/year?

Yes. "ppb a-1" represent ppb per annual (year). It is corrected.

Fig. 7b needs to be improved, different lines are hard to read.

Thanks for the suggestion, the figure is replotted with better color identification to improve readability.



LN97, " reducing heterogeneous uptake of reactive gases (mainly HO_2 and O_3), of which the latter is more important". Box model appears more suitable to investigate such an impact.

Yes, the current box model needs to include a detailed aerosol chemistry and observation-based uptake coefficients to achieve the target. We added this in the revised version.

Fig. 5g may be misleading. The positive correlation between PBLH and O_3 may be simply because on those high PBLH cases, radiation and temperature are much higher, thus O_3 formation is stronger. Many papers report that shallower PBL suppresses dispersion of pollutants and leads to higher O_3, suggesting a negative correlation between PBLH and O_3.

Yes, we agree. This could represent one of the cases when radiation is strong, temperature is higher while the PBL height is higher either. Higher height of the PBL also could lead to the mixing of near surface air with the O_3 rich air aloft, resulting in the observed enhancements in surface O_3 (REDDY et al., 2012). O_3 formation could be suppressed by the a shallow PBL due to the NO titration. We have added an additional statement "whereas some other studies reported that a shallow PBL suppresses the dispersion of pollutants and leads to higher O_3, suggesting a negative correlation between PBLH and O_3 (Yan et al., 2018; Jiang et al., 2016; Wei et al., 2016; Huang et al., 2005)". Please see the revised version.

Sensitivity simulations summarized in Table 1 appears to attribute radiation uncertainties to aerosols (AOD and SSA). Cloud may also play critical roles, which might be the reason to explain the poor correlation between radiation and aerosol concentrations in Fig. 6b.

Thanks for the comment. We have added the statement with cloud may also play a critical role, which might be another reason to explain the poor correlation between radiation and aerosol concentrations in Fig. 6b" in the revised version.

References:

Jiang, Y. C., Zhao, T. L., Liu, J., Xu, X. D., Tan, C. H., Cheng, X. H., Bi, X. Y., Gan, J. B., You, J. F., and Zhao, S. Z.: Why does surface ozone peak before a typhoon landing in southeast China?, Atmos. Chem. Phys., 15, 13331–13338, https://doi.org/10.5194/acp-15-13331-2015, 2015.

Huang, J., Zhou, C., Lee, X., Bao, Y., Zhao, X., Fung, J., RICHTER, Andreas, Liu, X., and Zheng, Y.: The effects of rapid urbanization on the levels in tropospheric nitrogen dioxide and ozone over East China, Atmos. Environ., 77, 558-567, https://doi.org/10.1016/j.atmosenv.2013.05.030, 2013.

Huang, Jian-Ping & Fung, Jimmy & Lau, Alexis & Qin, Yu. (2005). Numerical simulation and process analysis of typhoon-related ozone episodes in Hong Kong. Journal of Geophysical Research. 110. 10.1029/2004JD004914.

REDDY, K.K., NAJA, M., OJHA, N. et al. Influences of the boundary layer evolution on surface ozone variations at a tropical rural site in India. J Earth Syst Sci 121, 911–922 (2012). https://doi.org/10.1007/s12040-012-0200-z.

Wei, Xiaolin & Lam, Ka-se & Cao, Chunyan & Li, Hui & He, Jiajia. (2016). Dynamics of the Typhoon Haitang Related High Ozone Episode over Hong Kong. Advances in Meteorology. 2016. 1-12. 10.1155/2016/6089154.

Yan R C，Ye H，Lin X，et al. 2018.Characteristics and influence factors of ozone pollution in Hangzhou[J].Acta Scientiae Circumstantiae，38(3) : 1128-1136

Zeng, Y., Cao, Y., Qiao, X., Seyler, B. C., and Tang, Y.: Air pollution reduction in China: Recent success but great challenge for the future, Sci. Total. Environ., 663, 329-337, https://doi.org/10.1016/j.scitotenv.2019.01.262, 2019.