Response to Comments of Reviewer #2

(comments in *italics*)

Manuscript number: EGUSPHERE-2023-2393

Title: Weakened aerosol-radiation interaction exacerbating ozone pollution in eastern China since China's clean air actions

This study examines the role of aerosol-radiation interaction (ARI), decomposed into aerosol-photolysis interaction (API) and aerosol-radiation feedback (ARF) on surface ozone concentration in China. Surface ozone increased remarkable in eastern China, contrasting the dramatic decline of $PM_{2.5}$ concentrations. It is therefore necessary to investigate the reasons for the ozone increase. The study found that reduced ARI due to decreased PM concentrations contributes to ozone production, with API playing a more important role than ARF. The regional differences are also briefly discussed. I think this is a nice study that is helpful in understanding the recent ozone increase in China. I only have a few minor comments.

Response:

Thanks to the reviewer for the valuable comments and suggestions which are very helpful for us to improve our manuscript. We have revised the manuscript carefully, as described in our point-to-point responses to the comments.

1. *A previous study seemed to indicate that chemical processes associated with $PM_{2.5}$ reduction, i.e., reduced removing rate of hydroperoxy radicals, is the main reason for the ozone increase in eastern China (Li et al., 2019, PNAS). I wonder how this effect compare to the ARI discussed in this study?*

Response:

As Li et al. (2019) did not directly quantify the extent of O_3 increase by weakened aerosol heterogeneous reactions, we use the results of Liu and Wang. (2020) for comparison. The increased MDA8 O_3 concentration over urban areas in summer caused by weakened aerosol-radiation interaction in this study is 1.77 ppb, which is compared to the value of 2.12 ppb increase caused by weakened aerosol heterogeneous reactions quantified by Liu and Wang (2020). According to the reviewer's comments, we have added this sentence in the revised manuscript. (**Page 18, Line 485-488**)

2. *In the WRF-Chem experiments, the authors zeroed off aerosol optical properties to exclude ARF. I wonder if aerosol microphysical properties are still included? This may affect cloud properties and still impact the radiation budget.*

Response:

The effects of aerosols on microphysical properties were not consider in this work. The most common approach to assessing the impact of aerosol-cloud interactions on air quality in model simulation is to assume a prescribed vertically uniform cloud droplet number concentration (Zhang et al., 2015; Zhao et al., 2017). In this study, we turned off aerosol optical properties in the

optical module which could not affect the cloud properties.

Figure R1 shows the spatial distributions of simulated summer and winter cloud droplet number concentration (CDNC) from BASE_17E17M and NOALL_17E17M cases in the daytime (08:00–17:00 LST). Analyzing Fig. R1, the CDNC distribution and concentration of BASE and NOALL has barely changed. Therefore, we zeroed off aerosol optical properties to exclude ARI with less impact on the cloud.

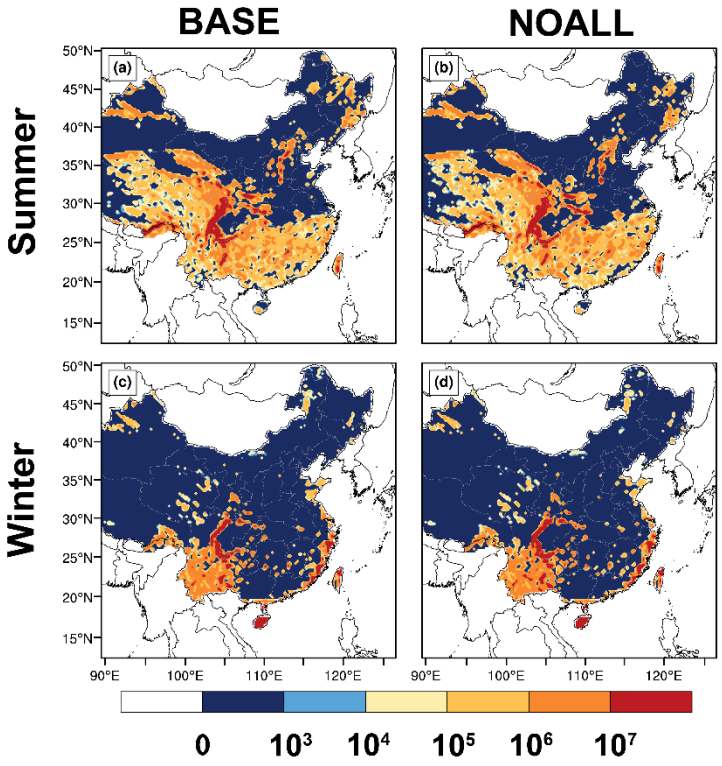

Figure R1. Spatial distributions of simulated summer (upper) and winter (bottom) cloud droplet number concentration (CDNC) from BASE_17E17M and NOALL_17E17M cases in the daytime (08:00–17:00 LST).

3. *Section 3.2, model evaluation: why not also evaluate VOCs, which is also an important precursor for ozone?*

Response:

Thanks for reviewer's suggestion. In this study, we did not evaluate VOCs due to the lack of measurements of VOCs over the China. However, the China's Ministry of Environmental Protection will include VOCs as a routine monitoring object in the future. Therefore, we will include this comparison in our future work.

4. *Line 87 and associated discussions: Does ARI always suppress O_3 formation? Could the change the meteorological variables through ARF increase O_3 concentration, say by reducing RH or increasing regional transport?*

Response:

Yang et al. (2022) reported that ARF reduced the planetary boundary layer height in North China, leading to an increase in VOCs and NO_x concentrations, which is favorable for ozone chemical production. Gao et al. (2018) also found that ARF can enhance ozone

chemical production through this pathway. Therefore, ARF can increase O_3 concentration by influencing the meteorological variables, e.g. by reducing the height of the planetary boundary layer.

5. *I suggest the authors discuss more about the summer-winter differences. Wintertime has much less radiation and lower temperature, so ARI is in general much lower. In summer, meteorology seems to make large contributions than emission changes (Figure 4, left column), what might be the reason?*

Response:

Focusing on the four developed city clusters, compared with 2013, the meteorological conditions in the summer of 2017 promoted the generation of O_3 in the YRD region (Fig. R2(a3)), but suppressed the generation of O_3 in the BTH (Fig. R2(a2)), PRD (Fig. R2(a4)) and SCB (Fig. R2(a5)) regions. In PRD and SCB, the changes in MDA8 O_3 due to meteorology even have a greater impact than that by emission changes, which highlights the significant role of meteorology on summer O_3 variations. **(Page 13, Line 343-349)**

According to the comments of Reviewer#1, another three widely used chemical mechanisms, i.e., RADM2 gas-phase chemistry coupled with MADE/SORGAM aerosol module (RADM2-MADE/SORGAM for short), CBMZ gas-phase chemistry coupled with MADE/SORGAM aerosol module (CBMZ-MADE/SORGAM for short), and MOZART gas-phase chemistry coupled with MOSAIC aerosol module (MOZART-MOSAIC for short), that include SOA formation are also applied to assess the impact of aerosol-radiation interaction (ARI) on O_3 during summer and winter is added in the discussion section. (**Page 18-19, Line 497-536**)

In summer, solar radiation flux reaches its maximum and atmospheric temperature are also higher than that in winter. The atmospheric warming can alter tropospheric O_3 concentrations by modulating the chemical kinetic, dynamic processes or biogenic emissions. Warmer temperatures often coincide with other meteorological conditions favorable to O_3 production, such as stagnation air and reduced cloud cover (Vukovich, 1995). This may be the reason why meteorological effect on O_3 is greater than that by emissions changes.

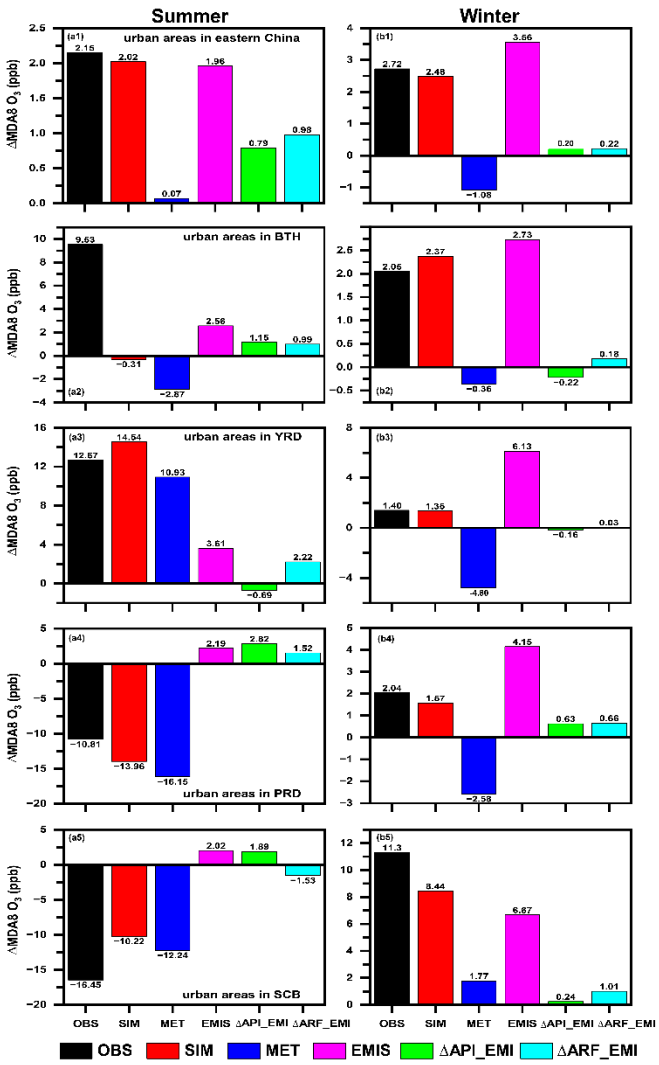

Figure R2. The observed (OBS, black bars) and simulated (SIM, red bars) changes in (left) summer and (right) winter surface-layer MDA8 O_3 from 2013 to 2017. Contributions of changed meteorological conditions alone (MET, blue bars), changed anthropogenic emissions alone (EMI, purple bars), changed aerosol-photolysis interaction alone (ΔAPI_EMI, green bars), and changed aerosol-radiation feedback alone (ΔARF_EMI, cyan bars) are also shown. Observations are calculated from the monitoring sites in the analyzed region, while the corresponding gridded simulations are averaged for SIM. **(a1-b1)**, **(a2-b2)**, **(a3-b3)**, **(a4-b4)** and **(a5-b5)** represent the urban areas in eastern China, Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (SCB), respectively.

6. *Figure 4: model seems to significantly underestimate the ozone change in BTH for summer (Figure 4a2). This area experienced the most ozone increases in the past decade. So it is important for the model to correctly represent ozone trend in this region. What might be the reason for this significant bias?*

Response:

Thanks for your suggestion. The reason for the underestimation over BTH in summer may be that this study did not consider the effect of changes in aerosol heterogeneous reactions, due to the uncertainty of the heterogeneous uptake value used in the numerical simulation. Li et al. (2019) found that the weakened uptake of HO_2 on aerosol surfaces was

the main reason for the O_3 increase over BTH. Therefore, the contributions of aerosol heterogeneous reactions to O_3 air quality will be discussed detailedly in our future work.

7. *Finally, the effects of API and ARF may not be independent, i.e., there may be nonlinear interaction between the two effects. This should be noted and discussed.*

Response:

Thanks for the reviewer's suggestion. A discussion of the separate treatment of API and ARF in this study has been added in the revised manuscript as follows: "There may be an interaction between API and ARF. However, in this study we discuss the role of API and ARF separately, which may ignore the effects of interactions between API and ARF on O_3. This may affect our results, and we will discuss their interaction in our future studies." (**Page 20, Line 557-560**)

Reference:

Gao, J. H., Zhu, B., Xiao, H., Kang, H. Q., Pan, C., Wang, D. D., and Wang, H. L.: Effects of black carbon and boundary layer interaction on surface ozone in Nanjing, China, Atmos. Chem. Phys., 18, 7081–7094, https://doi.org/10.5194/acp-18-7081-2018, 2018.

Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q., and Bates, K. H.: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China, P. Natl. Acad. Sci. USA, 116, 422–427, https://doi.org/10.1073/pnas.1812168116, 2019.

Liu, Y. and Wang, T.: Worsening urban ozone pollution in China from 2013 to 2017 – Part 2: The effects of emission changes and implications for multi-pollutant control, Atmos. Chem. Phys., 20, 6323–6337, https://doi.org/10.5194/acp-20-6323-2020, 2020.

Vukovich F. M.: Regional-scale boundary layer ozone variations in the eastern United States and their association with meteorological variations, Atmos. Environ., 29, 2259-2273, 1995.

Yang, H., Chen, L., Liao, H., Zhu, J., Wang, W., and Li, X.: Impacts of aerosol–photolysis interaction and aerosol–radiation feedback on surface-layer ozone in North China during multi-pollutant air pollution episodes, Atmos. Chem. Phys., 22, 4101–4116, https://doi.org/10.5194/acp-22-4101-2022, 2022.

Zhang, B., Wang, Y., and Hao, J.: Simulating aerosol–radiation–cloud feedbacks on meteorology and air quality over eastern China under severe haze conditionsin winter, Atmos. Chem. Phys., 15, 2387–2404, https://doi.org/10.5194/acp-15-2387-2015, 2015.

Zhao, B., Liou, K.-N., Gu, Y., Li, Q., Jiang, J. H., Su, H., He, C., Tseng, H.-L. R., Wang, S., Liu, R., Qi, L., Lee, W.-L., and Hao, J.: Enhanced $PM_{2.5}$ pollution in China due to aerosol-cloud interactions, Scient. Rep., 7, 4453, https://doi.org/10.1038/s41598-017-04096-8, 2017.

Thank you very much for your comments and suggestions.