Response to Comments of Reviewer #3

(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

The manuscript focuses on the aerosol-radiation interaction (ARI), discussing how this process has changed in the context of the abrupt aerosol decrease in East China during 2013-2017, and evaluates its contribution to the recent ozone increase in China. ARI is divided into aerosol-photolysis interaction (API) and aerosol-radiation feedback (ARF), with the WRF-Chem model used to quantify these impacts. The authors have found non-negligible ozone increase resulting from the aerosol decrease through the API and ARF processes, which has implications for the synergistic control of aerosol and ozone. This is an interesting topic and I believe it can make a novel contribution to the community. However, several important aspects need to be addressed before it can be published in ACP.

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.

General comments:

1. *The study focuses on aerosol-radiation interaction (ARI), which is split into two parts: the direct aerosol impact on radiation through scattering and absorbing (API) and the subsequent feedback on meteorology (ARF), with both influencing ozone concentrations. However, the Introduction Section could do a better job at breaking down these concepts. A detailed explanation of the distinctions between API and ARF would aid comprehension. Also, elucidating the specific ARF-related meteorological variables and their influences on ozone concentrations would be beneficial. Regarding the cited papers, such as Hong et al. (2020) and Zhu et al. (2021), the authors may consider including additional information about which ARF-related meteorological factors have been identified as important in affecting ozone concentrations.*

Response:

Thanks to the reviewer for the valuable comments and suggestions, we have added this information in the revised manuscript as follows: "API can affect O_3 directly by reducing the photochemical reactions, which weaken the chemical contribution and reduce the surface O_3 concentrations. ARF indirectly affects O_3 concentrations by altering meteorological variables, e.g. by reducing the height of the planetary boundary layer. The suppressed planetary boundary layer can weaken the vertical mixing of O_3 by turbulence and affect the concentration of O_3 precursors. Hong et al. (2020) used WRF-CMAQ in conjunction with future emission scenarios to find that weakened ARF due to reduced aerosol concentration has either negative or positive impacts on the daily maximum 1-h average O_3 concentration in eastern China from 2010 to 2050 due to the changed precursor level caused by the weakened ARF. By using WRF-CMAQ, Liu and Wang (2020b) reported that weakened API could increase the MDA8 O_3 concentrations by 0.3 ppb in urban areas from 2013 to 2017. Zhu et al. (2021) used WRF-Chem to investigate the impact of weakened ARF on air pollutants over NCP during COVID-19 lockdown and reported that the weakened ARF would increase the O_3 concentrations by 7.8% due to the increased northwesterly and planetary boundary layer height caused by the weakened ARF." (**Page 4-5, Line 95-110**)

2. *In Section 3.2, could the authors talk more about how well the model is doing in reproducing the observed decrease in $PM_{2.5}$ levels from 2013-2017. This analysis is crucial for assessing whether the model's effectively capturing the weakening of ARI.*

Response:

Thanks for your suggestion. Figure R1 demonstrates the spatial distribution of changed summer (left) and winter (right) surface (a, b) $PM_{2.5}$ and (c, d) MDA8 O_3 from 2013 to 2017. As shown in Figs. R1(a) and R1(b), the observed concentrations of $PM_{2.5}$ in eastern China are significantly reduced both in summer (-16.2 μg m^{-3}) and winter (-56.0 μg m^{-3}), and these changes can be well captured by the model (-14.3 μg m^{-3} for summer and -49.8 μg m^{-3} for winter). Therefore, the model can reproduce the observed decrease in $PM_{2.5}$ levels from 2013 to 2017. As shown in Figs. R1(c) and R1(d), the model reasonably well reproduces the seasonal patterns of changed surface MDA8 O_3 over the eastern China during summer and winter from 2013 to 2017. In summer, both the observations and simulations show the increased (decreased) MDA8 O_3 in YRD (PRD and SCB), while the model can not simulate the positive changes in MDA8 O_3 over BTH, and the potential reasons may be that this study did not consider the effect of changes in aerosol heterogeneous reactions. 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. In contrast to the changes in summer, observed MDA8 O_3 in winter generally increased over the eastern China, which can be well reproduced by the model. (**Page 12, Line 308-324**)

According to the reviewer's comments, Figure R1 is added in the model evaluation section. (**Figure 3**)

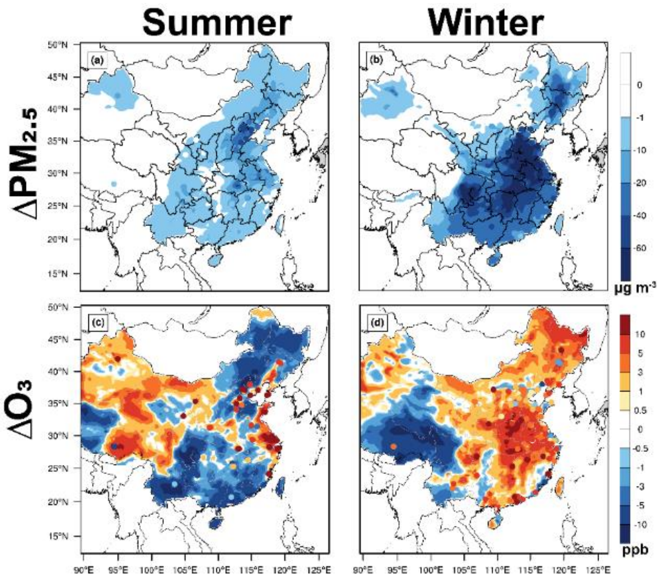

Figure R1. Spatial distribution of changed summer (left) and winter (right) surface (a, b) $PM_{2.5}$ and (c, d) MDA8 O_3 from 2013 to 2017.

3. *Section 4 needs to be better organized for clarity. I've outlined some areas for consideration:*

3.1. *The titles suggest Section 4.1 should focus on ΔO_3_MET and ΔO_3_EMI, while 4.2 should be devoted to ΔO_3_ΔARI_EMI. However, there is content overlap since 4.1 also examines ΔO_3_ΔARI_EMI, which obscures the distinctions between the two subsections.*

Response:

Thanks for your suggestion. We have changed this in revised manuscript. Section 4.1 focuses only on the ΔO_3_MET and ΔO_3_EMI, and the results of the ΔO_3_ΔARI_EMI in urban areas have been moved to Section 4.2. (**Page 12-13, Line 326-349**)

3.2. *Section 4.1 discusses ΔO_3_MET, ΔO_3_EMI, and ΔO_3_ΔARI_EMI at sparse polluted grids (so-called urban areas) while 4.2 talks about ΔO_3_ΔARI_EMI in term of regional averages. It is unclear why the discussion about ΔO_3_MET and ΔO_3_EMI focuses only on urban polluted regions. Also, the rationale for addressing urban ΔO_3_ΔARI_EMI prior to regional averages is not evident, particularly when urban results mirror the regional ones, though more pronounced. I recommend relocating the OBS-SIM ozone change comparison from Section 4.1 to Section 3.2 (to combine it with $PM_{2.5}$ change evaluation) and discussing regional ΔO_3_ΔARI_EMI before the urban analysis.*

Response:

Thanks for your suggestion. The comparison of O_3 change from 2013 to 2017 has been combined with the comparison of $PM_{2.5}$ change in Section 3. The detailed information can be found in the answer to your second question.

According to review's suggestion, in the revised manuscript we first discussed the effects of weakened ARI on O_3 at the regional level, and then in urban areas. **(Page 14-18, Line 383-495)**

3.3. Section 4.3 and Figure 7 are quite similar to Section 4.2 and Figure 5. Please consider merging Sections 4.2 and 4.3.

Response:

Thanks for your suggestion. We've combined these two sections in the revised manuscript.

4. *Could the authors explain why $\Delta O_3\_\Delta ARI\_EMI$ displays a much steeper spatial gradient in summer compared to winter (Fig. 5), whereas the $PM_{2.5}$ change suggest the opposite pattern (Fig. S8)? How does meteorology contribute to this discrepancy? Moreover, why does summertime $\Delta O_3\_\Delta ARI\_EMI$ exhibit both positive (e.g., NCP) and negative (e.g., Shandong province) values, even though the $PM_{2.5}$ decreases universally?*

Response:

The reason may be that the solar radiation flux reaches its maximum in summer seasons. The changes in meteorological variables are larger in summer than in winter due to the weakened ARI, despite the substantial decrease in aerosol concentrations during winter. Meteorology is likely to be a major contributor to this discrepancy.

Although the concentration of $PM_{2.5}$ is reduced uniformly, the changes in the components of $PM_{2.5}$ are different in different locations, resulting in different changes in single scattering albedo (SSA). As shown in Fig. R2, SSA did not change in NCP, but became smaller in Shandong Province, which may be the reason for the different changes in O_3 in these two regions. Furthermore, Fig. S7(b3) and S7(c3) show that weakened aerosol-radiation interaction leads to a decrease in T_2 but an increase in RH_2 over Shandong, which is also unfavourable for O_3 production. This could also be one of the reasons why weakened aerosol-radiation interaction leads to O_3 reduction in Shandong Province.

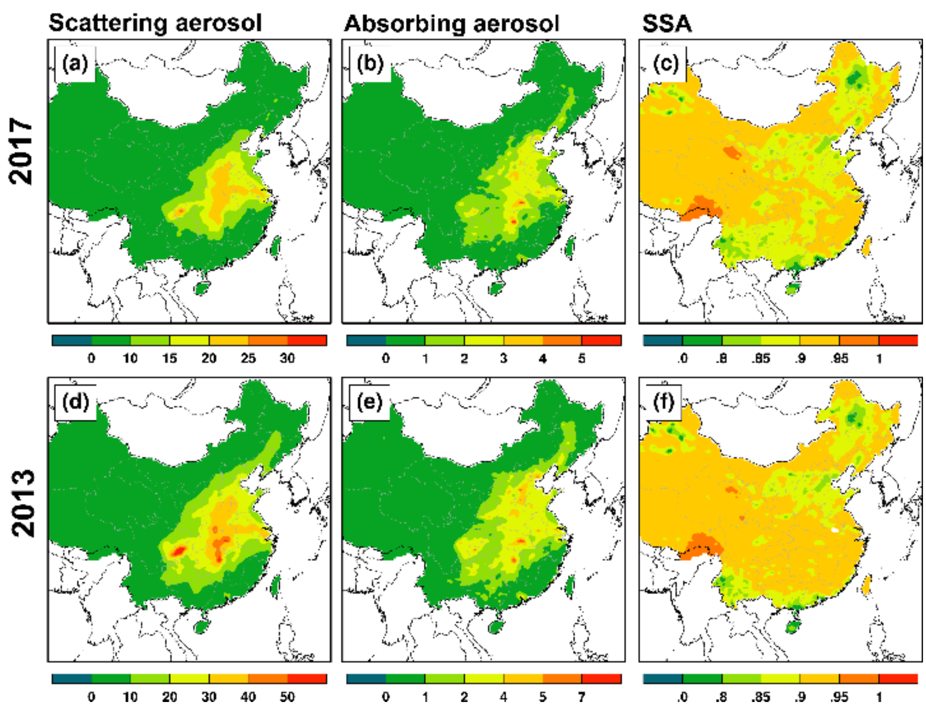

Figure R2. Spatial distribution of **(a, d)** scattering aerosol, **(b, e)** absorbing aerosol, and **(c, f)** single scattering albedo (SSA) of BASE_17E17M (upper) and BASE_13E17M (bottom) cases.

5. *From my understanding, the reduced impact of ARI on ozone is a component of the anthropogenic impact on ozone, since the reduction in ARI results from changes in anthropogenic emissions. However, the phrasing in Lines 396-398 and abstract (specifically the use of "superimposed") suggest that ΔO_3_ΔARI_EMI is and additional, separate effect rather than being nested within the broader anthropogenic impact on ozone. Please clarify.*

Response:

Thanks for your suggestion. Figure R3 shows the changed summer and winter surface-layer MDA8 O_3 concentrations caused by anthropogenic emission reduction from 2013 to 2017 with (ΔO_3_EMI) and without (ΔO_3_NOARI) ARI, including the effects of weakened ARI on the effectiveness of emission reduction for O_3 air quality (ΔO_3_ΔARI_EMI, which is also equal to ΔO_3_EMI minus ΔO_3_NOARI). As shown in Figs. R3(a1) and R3(a4), the surface-layer MDA8 O_3 concentrations increased in urban areas during summer and increased uniformly in winter due to anthropogenic emission reduction from 2013 to 2017 without the impact of ARI. The plots in the second column (Figs. R3(a2) and R3(a5)) are the same as R3(a1) and R3(a4) except that the impact of ARI is applied. When the effect of ARI is considered, the concentrations of MDA8 O_3 are increased more than that when ARI is not considered. The differences between plots in second column and first column are the consequences of weakened ARI resulted from anthropogenic emission reduction on MDA8 O_3 concentrations. As shown in Figs. R3(a3) and R3(a6), the concentrations of MDA8 O_3 are increased in both summer and winter over eastern China. Therefore, ΔO_3_ΔARI_EMI makes the superimposed impact on the effectiveness of anthropogenic emission reduction for the increased MDA8 O_3 concentrations from 2013 to 2017 over eastern China.

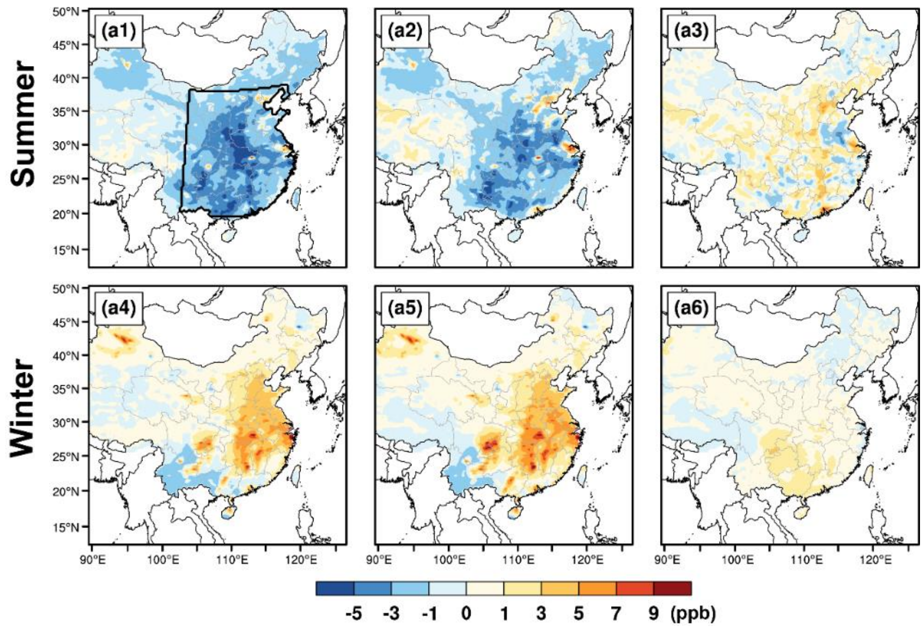

Figure R3. Spatial distribution of changed summer (upper) and winter (bottom) surface-layer MDA8 O_3 concentrations from sensitivity simulations. **(a1, a4)** Effects of anthropogenic emission reduction on MDA8 O_3 without ARI. **(a2, a5)** Effects of anthropogenic emission reduction on MDA8 O_3 with ARI. **(a3, a6)** Effects of weakened ARI on the effectiveness of emission reduction for O_3 air quality.

6. *In the Abstract, needs to explicitly clarify that the numbers presented are derived from different analysis. Lines 28-29 are for sparse polluted grids, while Lines 33-35 are for regional averages. Otherwise, readers may erroneously interpret the ratio between the numbers in Lines 33-35 and Lines 28-29 as the contribution of ARI to the total anthropogenic impacts.*

Response:

Thanks for your suggestion. We've added this information in the revised manuscript as follows:

"Sensitivity experiments show that the decreased anthropogenic emissions play a more prominent role for the increased MDA8 O_3 both in summer (+1.96 ppb vs. +0.07 ppb) and winter (+3.56 ppb vs. -1.08 ppb) than the impacts of changed meteorological conditions in urban areas. (**Page 2, Line 27-31**)

The weakened ARI due to decreased anthropogenic emission aggravates the summer (winter) O_3 pollution by +0.81 ppb (+0.63 ppb) averaged over eastern China, with weakened API and ARF contributing 55.6% (61.9%) and 44.4% (38.1%), respectively. This superimposed effect is more significant for urban areas during summer (+1.77 ppb). (**Page 2, Line 33-37**)"

Specific comments:

1. Line 61, natural emissions are also an important precursor source. Please clarify.

   Response:

   According to the reviewer's suggestion, we have changed the expression in the revised manuscript. (**Page 3, Line 65-67**)

2. Section 3.2, it should be "Fig. 2" instead of "Figs. 2". Similar typos are found in other places, e.g., Line 290, 302, 348. Please check.

   Response:

   Thanks for your suggestion. Since it's followed by a plural, we use "Figs".

3. Line 293, delete "will".

   Response:

   Deleted.

4. Lines 310-312 and figure 4, please clarify in the figure caption that ARI_EMI can be obtained by summing the bars of API_EMI and ARF_EMI.

   Response:

   Thanks for your suggestion. We have defined the ΔO3_ΔARI_EMI = ΔO3_ΔARF_EMI + ΔO3_ΔAPI_EMI in the revised manuscript. (**Page 15, Line 405-406**)

5. Lines 353-354 and figure 5, the numbers mentioned in the text are inconsistent with those presented in the figure. Please correct.

   Response:

   Correct.

6. Figure 6, the first x-axis label should be "ARI" instead of "ALL".

   Response:

   Thanks for your suggestion. We have changed the expression in the revised manuscript. (**Page 37**)

Reference:

Hong, C., Zhang, Q., Zhang, Y., Davis, S. J., Zhang, X., Tong, D., Guan, D., Liu, Z., and He, K.: Weakening aerosol direct radiative effects mitigate climate penalty on Chinese air quality, Nat. Clim. Change, 10, 845–850, https://doi.org/10.1038/s41558-020-0840-y, 2020.

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, 2020b.

Zhu, J., Chen, L., Liao, H., Yang, H., Yang, Y., and Yue, X.: Enhanced PM2.5 Decreases and O3 Increases in China During COVID-19 Lockdown by Aerosol-Radiation Feedback, Geophys. Res. Lett., 48, https://doi.org/10.1029/2020GL090260, 2021.

Thank you very much for your comments and suggestions.