Response to Comments of Reviewer #1

(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 paper mainly investigated the impacts of aerosol-photolysis interaction (API) and aerosol-radiation feedback (ARF) on the surface ozone concentrations under the background of China's clean air action (rapid anthropogenic emission reductions from 2013 to 2017).

The effects of API on ozone concentrations are not a new finding since I have found several previous studies already addressed it (Gao et al., 2022; Liu and Wang, 2020). However, I have not found any previous studies focused on the effects of ARF on ozone concentrations. Furthermore, the authors used the IPR methodology to investigate the contribution to O_3 concentration variation from four processes (VMIX, CHEM, ADVH, ADVZ). In conclusion, I consider this paper valuable for publication, even if it has some limitations (as shown below). (1) The absence of SOA formation and heterogeneous reactions in their simulations could be a limitation of this study; even the authors have sufficiently acknowledged this. (2) Some parts/aspects are poorly elucidated, making it hard for me to understand. A major revision is needed 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.

The major innovation of this study is that **it is the first time** to quantify the response of aerosol-radiation interaction to anthropogenic emission reduction from 2013 to 2017, with the mainly focus on the contribution to changed O_3 concentrations over eastern China both in summer and winter.

According to the reviewer's comments, **another three** widely used chemical mechanisms, i.e., RADM2-MADE/SORGAM (RADM2 gas-phase chemistry coupled with MADE/SORGAM aerosol module), CBMZ-MADE/SORGAM (CBMZ gas-phase chemistry coupled with MADE/SORGAM aerosol module), and MOZART-MOSAIC (MOZART gas-phase chemistry coupled with MOSAIC aerosol module), that include SOA formation are also applied to test the impact of aerosol-radiation interaction (ARI) on O_3 with and without SOA.

Comparing the simulation results of the three additional mechanisms, the simulated $PM_{2.5}$ from MOZART-MOSAIC are closer to the actual observation. Analyzing the summer/winter MDA8 O_3 reductions due to ARI by the mechanism used in our manuscript (i.e., CBMZ-MOSAIC) and MOZART-MOSAIC, **similar results are quantified** (1.32 ppb *vs.* 1.85 ppb for summer, and 1.96 ppb *vs.* 1.60 ppb for winter). Therefore, although the CBMZ-MOSAIC used in this paper does not take into account the formation of SOA and its associated effects, the aerosol radiative effect on O_3 concentration is consistent with the results when the SOA simulation mechanism is considered.

The impacts of aerosol heterogeneous reactions on O_3 have not been considered in this

manuscript due to the **uncertainty and inconsistency** of the heterogeneous uptake shown in previous observation and simulation studies (Liu and Wang., 2020b; Tan et al., 2020; Shao et al., 2021). Shao et al. (2021) summarized that different heterogeneous uptake on the aerosol surface applied in the model simulation (e.g., 0.20 *vs.* 0.08) would cause significant deviations in simulated ozone concentrations (e.g., O_3 increased by 6% *vs.* O_3 increased by 2.5%). Therefore, the uncertainty in the heterogeneous uptake value used in the numerical simulation will finally amplify the deviation in model results.

According to the reviewer's comments about some poorly elucidated parts, such as ΔO_3_ΔARF_EMI. We have **detailedly described** in our point-to-point responses as shown below, and related descriptions have also been added in the revised manuscript.

Specific comments:

1. *In my opinion, SOAs account for a substantial portion of total aerosols. Typically, in your research, the lack of consideration of SOA can truly affect the reliability of the results (the authors also mentioned that $PM_{2.5}$ is underestimated in your model). I highly recommend the authors include SOA formation in their model.*

Response:

Thanks to the reviewer for the valuable comments and suggestions. The CBMZ gas-phase chemistry coupled with MOSAIC aerosol module (CBMZ-MOSAIC for short) used in this study does not include secondary organic aerosol (SOA), then we applied three additional chemical mechanisms that consider SOA, namely, 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), to test the impact of ARI on O_3 with and without SOA for the scenario of BASE_17E17M.

Figures R1 shows the temporal variations of observed and simulated $PM_{2.5}$ and O_3 concentrations over eastern China for the three additional chemical mechanisms. Comparing with the observed $PM_{2.5}$ (O_3) concentrations, the MOZART-MOSAIC showed the best performance in December 2017, with the R of 0.73 (0.79) and NMB of -18.7% (-20.5%). Therefore, we further used this mechanism to simulate the air pollutant concentrations during the period of June 2017. As shown in Fig. R1 (a4, b4), the temporal variations of observed $PM_{2.5}$ (O_3) can be well captured by this mechanism with R of 0.56 (0.91) and NMB of -1.7% (-20.3%).

Finally, we investigated the effect of aerosol-radiation interaction (ARI) on O_3 from the results of CBMZ-MOSAIC (this mechanism applied in this manuscript which does not include SOA) and MOZART-MOSAIC (this mechanism includes SOA and performs the best simulation results comparing with RADM2-MADE/SORGAM and CBMZ-MADE/SORGAM). As shown in Fig. R2, summer (winter) MDA8 O_3 is significantly reduced over eastern China, ARI reduces the surface MDA8 O_3 concentrations by 1.32 (1.96) ppb and 1.85 (1.60) ppb by CBMZ-MOSAIC and MOZART-MOSAIC, respectively. The O_3 reductions are of comparable magnitude in these two schemes. Therefore, we can conclude that although the CBMZ-MOSAIC applied in this manuscript does not take into account the formation of SOA and its associated effects, the aerosol radiative effects on O_3 concentrations not only in the pattern of spatial-temporal distribution but also in the order of magnitude are consistent with the results when the SOA simulation mechanism is considered.

As shown in Fig. R3, the mean SOA simulated by RADM2-MADE/SORGAM, CBMZ-

MADE/SORGAM, and MOZART-MOSAIC are 0.29, 0.45 and 0.94 μg m^{-3}, accounting for 3.4%, 3.8%, and 4.4% of $PM_{2.5}$ concentrations in winter 2017, respectively. From Fig. R4, the mean SOA simulated from MOZART-MOSAIC is 0.90 μg m^{-3}, account for 9.1% of $PM_{2.5}$ in summer 2017. Model simulated SOA concentrations are generally underestimated in most current chemical transport models (Zhang et al., 2015; Zhao et al., 2015). The low SOA concentrations simulated by the model can be explained by low emissions of biogenic and anthropogenic VOCs (key precursors of SOA), but a thorough investigation of this underestimation is outside the scope of this manuscript and it will be discussed in our future work. (**Page 18-19, Line 497-536**)

According to the reviewer's suggestion, we have added Figs. R1-R4 in the revised support information. **(Page 13-16 in supporting information)**

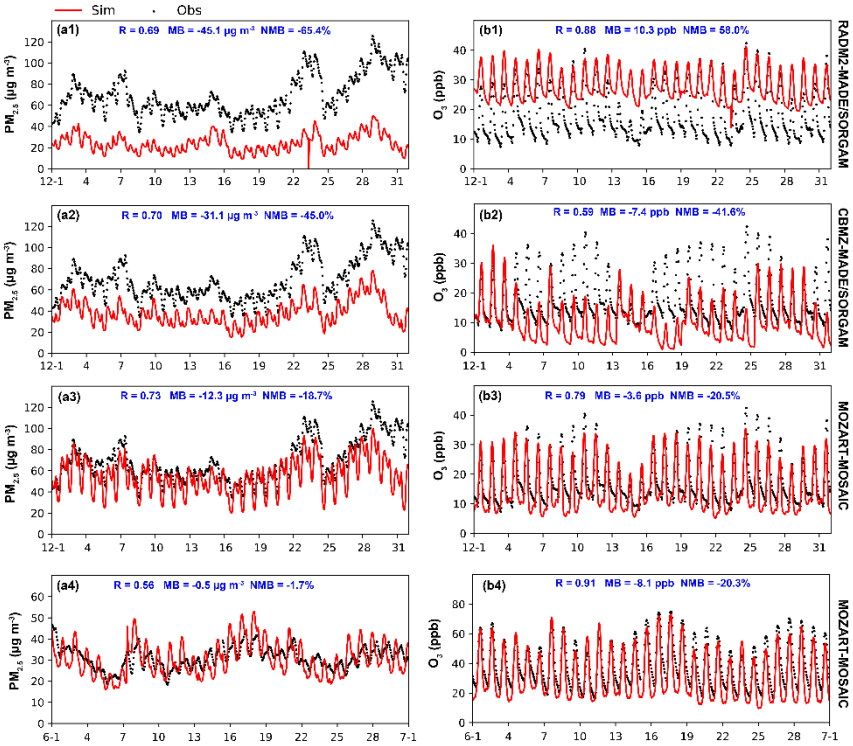

Figure R1. Time series of observed (black dots) and simulated (red lines) hourly **(a1-a4)** $PM_{2.5}$ and **(b1-b4)** O_3 concentrations averaged over the whole observation sites in eastern China during summer and winter 2017. **(a1, b1)** Simulated $PM_{2.5}$ and O_3 concentrations in winter 2017 by RADM2 gas-phase chemistry coupled with MADE/SORGAM aerosol module (RADM2-MADE/SORGAM). **(a2, b2)** Simulated $PM_{2.5}$ and O_3 concentrations in winter 2017 by CBMZ gas-phase chemistry coupled with MADE/SORGAM aerosol module (CBMZ-MADE/SORGAM). **(a3, b3)** Simulated $PM_{2.5}$ and O_3 concentrations in winter 2017 by MOZART gas-phase chemistry coupled with MOSAIC aerosol module (MOZART-MOSAIC). **(a4, b4)** is the same as **(a3, b3)**, but for summer 2017. The calculated correlation coefficient (R), mean bias (MB), and normalized mean bias (NMB) are also shown.

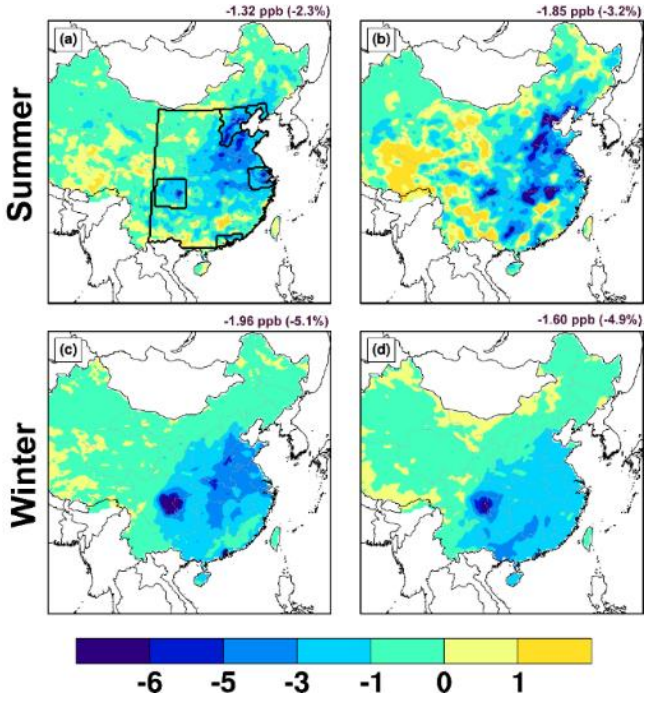

Figure R2. The effects of aerosol-radiation interaction on surface-layer MDA8 O_3 in summer (upper) and winter (bottom) 2017 calculated by **(a, c)** CBMZ-MOSAIC and **(b, d)** MOZART-MOSAIC mechanisms. The changes (percentage changes) averaged over China are also shown at the top of each panel.

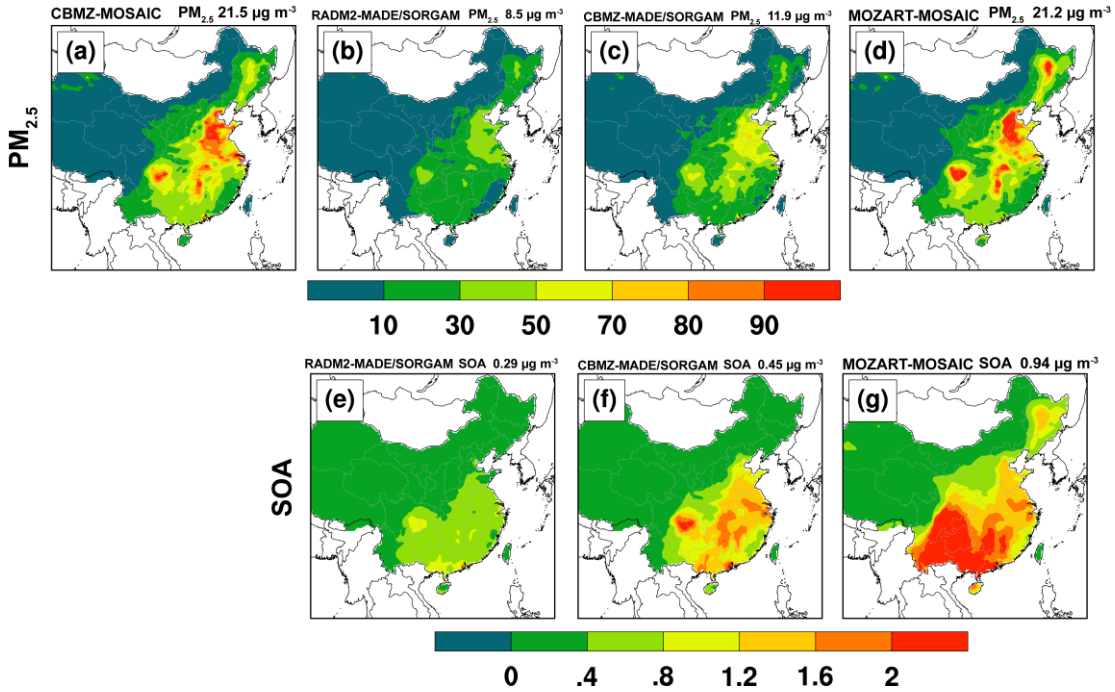

Figure R3. Spatial distributions of simulated mean $PM_{2.5}$ and SOA concentrations (μg m^{-3}) in winter 2017 by **(a)** CBMZ gas-phase chemistry coupled with MOSAIC aerosol module (CBMZ-MOSAIC), **(b, e)** RADM2 gas-phase chemistry coupled with MADE/SORGAM aerosol module (RADM2-MADE/SORGAM), **(c, f)** CBMZ gas-phase chemistry coupled with MADE/SORGAM aerosol module (CBMZ-MADE/SORGAM), and **(d, g)** MOZART gas-phase chemistry coupled with MOSAIC aerosol module (MOZART-MOSAIC). The calculated pollutant concentrations averaged over China are also shown at the top of each panel.

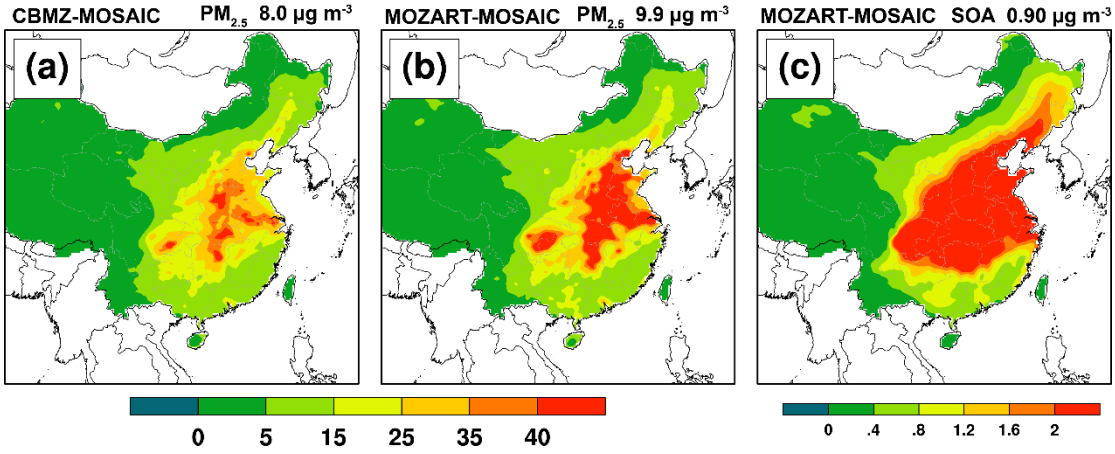

Figure R4. Spatial distributions of simulated mean $PM_{2.5}$ and SOA concentrations (μg m^{-3}) in summer 2017 by **(a)** CBMZ gas-phase chemistry coupled with MOSAIC aerosol module (CBMZ-MOSAIC), (b, c) MOZART gas-phase chemistry coupled with MOSAIC aerosol module (MOZART-MOSAIC). The calculated pollutant concentrations averaged over China are also shown at the top of each panel.

2. *Similarly, as the significant impacts of heterogeneous reactions on ozone concentrations mentioned by previous studies (Lou et al., 2014; Liu and Wang, 2020), I would expect the authors to include heterogeneous reactions in their models. If the authors have specific reasons for not including heterogeneous reactions in their models, those reasons need to be stated in the paper.*

Response:

In addition to the impacts of aerosol-radiation interaction (ARI), aerosols can also affect the concentrations of O_3 by heterogeneous chemistry (HET). Liu and Wang. (2020b) found that the rapid decrease of $PM_{2.5}$ was a major contributor for the summer O_3 increase through weakening the heterogeneous uptake of hydroperoxy radical (HO_2). However, Tan et al. (2020) launched a field campaign in North China Plain (NCP) and proposed a contradicting opinion about the importance of the impact of HET on O_3. These inconsistent conclusions generated from field observations and numerical simulations are mainly originated from the different values of heterogeneous uptake they used. Tan et al. (2020) pointed out that the heterogeneous uptake of HO_2 on aerosol surface was 0.08 ($\gamma_{HO2} = 0.08$) over NCP, which is smaller than the values ($\gamma_{HO2} = 0.2$) used in model simulations (Li et al., 2019; Liu and Wang., 2020). As shown in Fig. R5, Shao et al. (2021) found controversial results by using the different heterogeneous uptake of HO_2. When $\gamma_{HO2} = 0.2$ was used in the chemical model, the reduced heterogeneous uptake of HO_2 due to the decrease in aerosol caused the maximum O_3 increased by about 6% from 2013 to 2016, which is close to the results of Li et al. (2019) (~ 7%). When $\gamma_{HO2} = 0.08$ was used, the reduced heterogeneous uptake of HO_2 due to the decrease in aerosol led to maximum O_3 increased by only 2.5% from 2013 to 2016. Therefore, significant deviations in the model results would result from the use of different heterogeneous uptake on the aerosol surface.

Furthermore, previous laboratory studies indicate that the uptake coefficient varies widely from 0.003 to 0.5 with a strong dependence on the concentration of transition metal ions such as Cu(II) and Fe(II) in the aerosol (Zou et al., 2019). Taketani et al. (2009) reported that the uptake coefficient of HO_2 (γ_{HO2}) on seawater particles depends on relative humidity (RH), with γ_{HO2} values of 0.10 ±

0.03, 0.11 ± 0.02 and 0.10 ± 0.03 at 35%, 50% and 75% RH, respectively. Lakey et al. (2015) also found that a large humidity dependence was observed for HO_2 uptake onto humic acid aerosols. The HO_2 uptake coefficient increased from 0.007 ± 0.002 to 0.06 ± 0.01 between 32 and 76% RH for the Acros organics humic acid, and from 0.043 ± 0.009 to 0.09 ± 0.03 between 33 and 75% RH for the Leonardite humic acid. This strong dependence on aerosol composition and RH implies that a single assumed value for heterogeneous uptake used in numerical simulation may cause large uncertainty. In addition, our manuscript devoted to quantifying the effects of ARI on O_3, rather than the impacts of heterogeneous reactions on O_3. Due to the reasons listed above, we did not consider the effect of heterogeneous reactions on O_3 temporarily in the manuscript.

Thanks for the reviewer's suggestion, and we will consider the impacts of heterogeneous reaction in our future works. A discussion about the impacts of heterogeneous reaction has been added in the revised manuscript as follows:

"The impacts of aerosol heterogeneous reactions (HET) on O_3 have not been considered in this manuscript due to the uncertainty and inconsistency of the heterogeneous uptake shown in previous observation and simulation studies (Liu and Wang., 2020b; Tan et al., 2020; Shao et al., 2021). Liu and Wang. (2020b) found that the rapid decrease of $PM_{2.5}$ was the primary contributor for the summer O_3 increase through weakening the heterogeneous uptake of hydroperoxy radical (HO_2). However, Tan et al. (2020) launched a field campaign in NCP and proposed a contradicting opinion about the importance of the impact of HET on O_3. Shao et al. (2021) summarized that different heterogeneous uptake on the aerosol surface applied in the model simulation (e.g., 0.20 *vs.* 0.08) would cause significant deviations in simulated ozone concentrations (e.g., O_3 increase by 6% *vs.* O_3 increase by 2.5%). Previous laboratory studies indicate that the dependence of the uptake coefficient on aerosol composition and RH means that a single assumed value for heterogeneous uptake used in numerical simulations can lead to large uncertainties (Lakey et al., 2015; Taketani et al., 2009; Zou et al., 2019). Therefore, the uncertainty in the heterogeneous uptake value used in the numerical simulation will finally amplify the deviation in model results. Meanwhile, our manuscript devoted to quantifying the effects of ARI on O_3, rather than the impacts of heterogeneous reactions on O_3. The absence of heterogeneous chemistry on aerosol surface may result in underestimation of the effect of aerosol on O_3, which will be considered in our future work." (**Page 19-20, Line 537-556)**

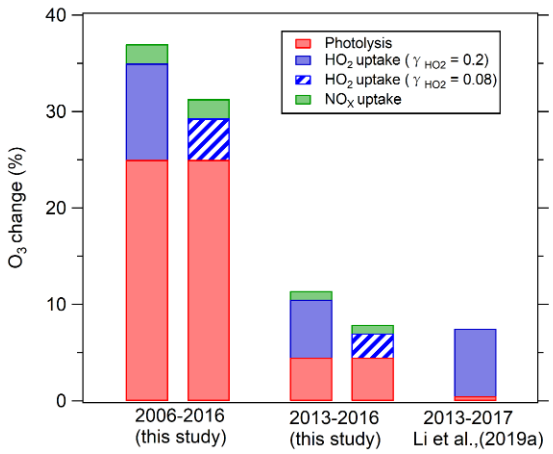

Figure R5. O_3 change due to the decrease in $PM_{2.5}$ during 2006-2016 and during 2013-2016 in the study of Shao et al., (2021) and during 2013-2017 in the study of Li et al., (2019a). This picture is from Shao et al., (2021).

3. *L160, you mentioned you fixed the meteorological field to the year 2013, can you explain how to achieve this? Can I understand that all *17M cases have exactly the same meteorological fields throughout 2017 simulation? However, I don't think all *17M cases should have the same meteorological fields, because you cannot investigate deltaO3_deltaARF_EMI if the meteorological fields are fixed in different cases. This needs to be explained more clearly in your paper.*

Response:

Thanks for your comments. ΔO_3_ΔARF_EMI represents the impacts of weakened aerosol-radiation feedback (ΔARF) due to decreased anthropogenic emission (EMI) on O_3 concentrations (ΔO_3). In order to quantify the impacts caused by the decreased EMI from 2013 to 2017, the impacts of changed meteorological variables should be removed by fixing the meteorological fields in year 2017 in sensitivity experiments, such as NOAPI_13E17M, NOALL_13E17M, NOAPI_17E17M and NOALL_17E17M (13E17M means anthropogenic emissions are from the year of 2013 and meteorological fields are from the year of 2017, more details can be found in Figure 1 in the revised manuscript).

For example, the differences between NOAPI_13E17M and NOALL_13E17M reflect the impact of ARF at the condition of 13E17M (the result is denoted as ΔO_3_ARF_{13E} for short), and the differences of NOAPI_17E17M and NOALL_17E17M show the impact of ARF at the condition of 17E17M (the result is denoted as ΔO3_ARF_{17E} for short), so the differences between ΔO3_ARF_{17E} and ΔO3_ARF_{13E} finally present the impact of weakened aerosol-radiation feedback due to decreased anthropogenic emission from 2013 to 2017 on O_3 concentrations.

For the summer simulations and the winter simulation in the year of 2013 or in the year of 2017, we use the June and December meteorological fields for the corresponding year.

The same method has been widely used in many other studies, which mainly focus on the impacts of weakened aerosol-radiation interactions on air pollutants in China (Li et al. 2019; Zhou et al., 2019; Hong et al. 2020; Liu and Wang. 2020b; Zhu et al. 2021; Shao et al. 2021).

According to the reviewer's suggestion, we have added this information in the revised manuscript. (**Page 7-8, Line 175-214**)

4. *L23-L25, you mentioned API and ARF. However, the API and ARF terminology is so abstract, making it hard for people to understand. It would help if you mentioned that API is related to the change in photolysis rates and ARF is related to the change of meteorological fields in your abstract.*

Response:

Thanks for your suggestion, we have added this information in the revised manuscript as follows: **"**Here we apply a coupled meteorology-chemistry model (WRF-Chem) to quantify the responses of aerosol-radiation interaction (ARI), including aerosol-photolysis interaction (API) related to photolysis rate change and aerosol-radiation feedback (ARF) related to meteorological fields change, to anthropogenic emission reductions from 2013 to 2017, and their contributions to O_3 increases over eastern China in summer and winter.**"** (**Page 2, Line 24-26**)

5. *L58, I think chemical species like CO and CH_4 can also lead to the formation of O_3.*

Response:

According to the reviewer's suggestion, we have changed the sentence in the revised

manuscript as follows: “As a secondary air pollutant, troposphere O_3 can be produced by nitrogen oxides (NO_x = NO + NO_2), carbon monoxide (CO), methane (CH_4) and volatile organic compounds (VOCs) in the presence of solar radiation through photochemical reactions (Atkinson, 2000; Seinfeld and Pandis, 2006).” (**Page 3, Line 60**)

6. *L57-L62 The causal relationship between the following two sentences is not clear. As a secondary air pollutant, troposphere O_3 can be produced by nitrogen oxides (NO_x = NO + NO_2) and volatile organic compounds (VOCs) in the presence of solar radiation through photochemical reactions (Atkinson, 2000; Seinfeld and Pandis, 2006). - > Consequently, the concentration of O_3 is closely related to changes in meteorological conditions and anthropogenic emissions (Wang et al., 2019; Liu and Wang, 2020a,b; Shu et al., 2020). "solar radiation" is not directly related to "meteorological conditions", try to revise those sentences to make them more logical.*

Response:

Thanks for your suggestion. we have changed the sentence in the revised manuscript as follows: “The concentration of O_3 in the troposphere is influenced by changes in meteorological conditions (e.g., high temperature and low relative humidity) and its precursors emissions (e.g., NOx and VOCs) (Wang et al., 2019; Liu and Wang, 2020a,b; Shu et al., 2020). Most precursors are from anthropogenic sources, and some precursors can come from natural sources, such as biogenic VOCs and soil and lightning NOx.” (**Page 3, Line 62-67**)

7. *2.1 Model configuration: I recommend using a chart (like Table 1 in https://www.sciencedirect.com/science/article/pii/S1352231020307378) to summarize the model configuration.*

Response:

Thanks to the reviewer’s comments, the model configuration is summarized in Table R1. We have added Table R1 in the revised supporting information. (**Table S1**)

Table R1. WRF-Chem model configurations with main physical and chemical schemes adopted in this study.

Model set-up	**Values**
Domain	East Asia
Study period	June and December 2017
Domain size	167 × 167
Domain center	34 °N, 108 °E
Horizontal resolution	27 km × 27 km
Vertical resolution	32 eta levels up to 50 hPa
Meteorological boundary and initial conditions	NCEP 1°×1° reanalysis data
Chemical initial and boundary conditions	CAM-Chem output
Physical options	**Adopted scheme**
Microphysics scheme	Lin (Purdue) scheme
Cumulus scheme	Grell 3D ensemble scheme
Boundary layer scheme	Yonsei University PBL scheme
Surface layer scheme	Monin-Obukhov surface scheme
Land-surface scheme	Unified Noah land-surface model

Longwave radiation scheme	RRTMG
Shortwave radiation scheme	RRTMG
Chemical options	**Adopted scheme**
Gas phase chemistry	CBMZ
Aerosols	MOSAIC
Photolysis	Fast-J
Biogenic emissions	MEGAN
Anthropogenic emissions	MEIC

8. *L125-L127, have you applied meteorological nudging? See above, I am not sure how you fix the meteorological fields to 2013 or 2017 when running the model.*

Response:

This work is done without nudging because only one domain is designed in our manuscript. If the nudging is turned on in only one domain simulation, the simulated meteorological field can not truly reflect the influence of the aerosol-radiation interaction feedback.

When using the 2013 (2017) FNL meteorological field data, it means that the meteorological field are from the year of 2013 (2017). For example, BASE_17E17M means that the meteorological field and anthropogenic emission are from the year of 2017. BASE_13E13M means that the meteorological field and anthropogenic emission are from the year of 2013.

9. *L151, you mentioned the biogenic emissions are calculated online by MEGAN. Have you coupled the MEGAN model with WRF-Chem dynamically? Please ascertain whether the biogenic emissions are calculated online or offline by MEGAN.*

Response:

Thanks to the reviewer's comments. In this work, we set "bio_emiss_opt = 3" in the WRF-Chem model, which represents the biogenic emissions can be calculated online by the coupled MEGAN module based upon the simulated meteorological variables (e.g., temperature, solar radiation) and underlying static data (e.g., leaf area index, plant types).

10. *L166, can you explain which aerosol optical properties are turned to zero?*

Response:

Following Qiu et al. (2017), the aerosol radiation interactions were turned off by removing the mass of aerosol species from the calculation of aerosol optical properties. Then, the aerosol optical properties such as aerosol optical depth (AOD), aerosol single scattering albedo (SSA), aerosol asymmetery factor (g) and aerosol backscatter coefficient were set to zero.

11. *L200-202, you mentioned "To avoid potential deviations caused by long-term model integration, each simulation is re-initialized every eight days". I was confused about why re-initialize the simulation every eight days can avoid potential deviations. What do you mean "potential deviations"? Can you explain this more?*

Response:

Thanks to the reviewer's comments. Lo et al. (2008) conducted three types of experiments for the entire year of 2000 to test model performance for different simulation durations: (1) continuous

integrations with a single initialization as usually done, namely, one year of uninterrupted simulation (WRFS), (2) consecutive integrations with re-initializations every 29 days (WRFM-30D), and (3) same as (2) but the model is reinitialized every 6 days (WRFM-7D). They found that the traditional continuous integration approach (WRFS) shows the worst performance. The model drifts from the forcing FNL reanalysis during the course of long integrations. It poorly simulates not only the forcing variables, (e.g., pressure, temperature, wind, and moisture), but also the model diagnostics variables (e.g., precipitation). Therefore, the simulation is re-initialized every eight days in this work, the same as the WRFM-7D, to avoid the deviation from forcing variables, (e.g., pressure, temperature, wind, and moisture) and model diagnostics variables (e.g., precipitation).

12. *L214-217 I feel confused about how many sites are operated by China National Environmental Monitoring Center (CNEMC)? You mentioned "1296 sites", does this number refer to the number of total sites of CNEMC or the number of sites chosen in your research? Moreover, are there really 1296 points (sites) on Figs. 2a and 2c?*

Response:

The CNEMC had 1484 observation sites in 2017. In this work, a single site with at least 500 actual observations during the simulated period are used for model evaluation, as we mentioned in the manuscript (**Page 9, Line 238-240**). Of course, Figs. 2a and 2c does have 1296 sites.

13. *Figure 2 shows the simulated results of which case? (BASE_17E17M?) You need to specify this point in L251 and Fig. 2.*

Response:

According to the reviewer's comment, we made it clear in Section 3 in the revised manuscript that the simulation results from the case of BASE_17E17M are used to evaluate the model performs (**Page 10, Line 264-266**).

14. *Why there are less points on Figs. 3a and 3d than Fig. 2? Please explain.*

Response:

Thanks to the reviewer's comments. The CNEMC installed only 450 sites in 2013, which grew to more than 1500 stations by 2020. In Fig. 3, only sites with continuous observations and individual site data greater than 500 were used to assess ozone trends. Thus, Fig. 3 has fewer points than Fig. 2.

15. *L221, if possible, I recommend explaining more about IPR in your paper.*

Response:

Thanks to the reviewer's suggestion, we have added this sentence in the revised manuscript as follows: "Process analysis techniques, i.e., integrated process rate (IPR) analysis, can be used in grid-based Eulerian models (e.g., WRF-Chem) to obtain contributions of each physical/chemical process to variations in pollutant concentrations. Eulerian models utilize the numerical technique of operator splitting to solve continuity equations for each species into several simple ordinary differential equations or partial differential equations that only contain the influence of one or two processes (Gipson, 1999).

In order to quantitatively elucidate individual contributions of physical and chemical processes

to O_3 concentration changes due to weakened ARI, the integrated process rate (IPR) methodology is applied in this study. IPR analysis is an advanced tool to evaluate the key process for O_3 concentration variation (Shu et al., 2016; Zhu et al., 2021; Yang et al., 2022). In this study, the IPR analysis tracks hourly (e.g., one time step) contribution to O_3 concentration variation from four main processes, including vertical mixing (VMIX), net chemical production (CHEM), horizontal advection (ADVH), and vertical advection (ADVZ). VMIX is initiated by turbulent process and closely related to PBL development, which influences O_3 vertical gradients. CHEM represents the net O_3 chemical production (chemical production minus chemical consumption). ADVH and ADVZ represent transport by winds. We define ADV as the sum of ADVH and ADVZ." (**Page 9-10, Line 245-262**)

16. *Table 2, how many sites are used for Table 2 (1296 sites?)?*

Response:

Thanks to the reviewer's comments. Table 2 contains 1296 sites, and we added this information to the revised manuscript (**Page 31, Line 811**).

17. *L284-285, you mentioned NOx-limited and VOCs-limited regions, I recommend that you could add a figure (based on your simulation results) like Fig. 5 in https://www.sciencedirect.com/science/article/pii/S1352231013000514 to your supplement, to show different O_3-sensitive regions on the map.*

Response:

The typical VOCs/NO_x ratio is calculated to classify sensitivity regimes and to indicate the possible O_3 responses to changes in VOCs and/or NO_x concentrations. O_3 production is VOC-limited if the ratio is less than 4, and it is NO_x-limited if the ratio is larger than 15 (Edson et al., 2017; Li et al., 2017). The ratio of VOCs/NO_x ranging around 4-15 indicates a transitional regime, where ozone is nearly equally sensitive to each species (Sillman, 1999). As shown in Fig R6, O_3 are mainly formed under the VOC-limited in winter and NO_x-limited and transitional regimes in eastern China, which is consistent with what our study mentioned.

According to the reviewer's suggestion, we have added Fig. R6 in the revised support information. **(Page 7 in supporting information)**

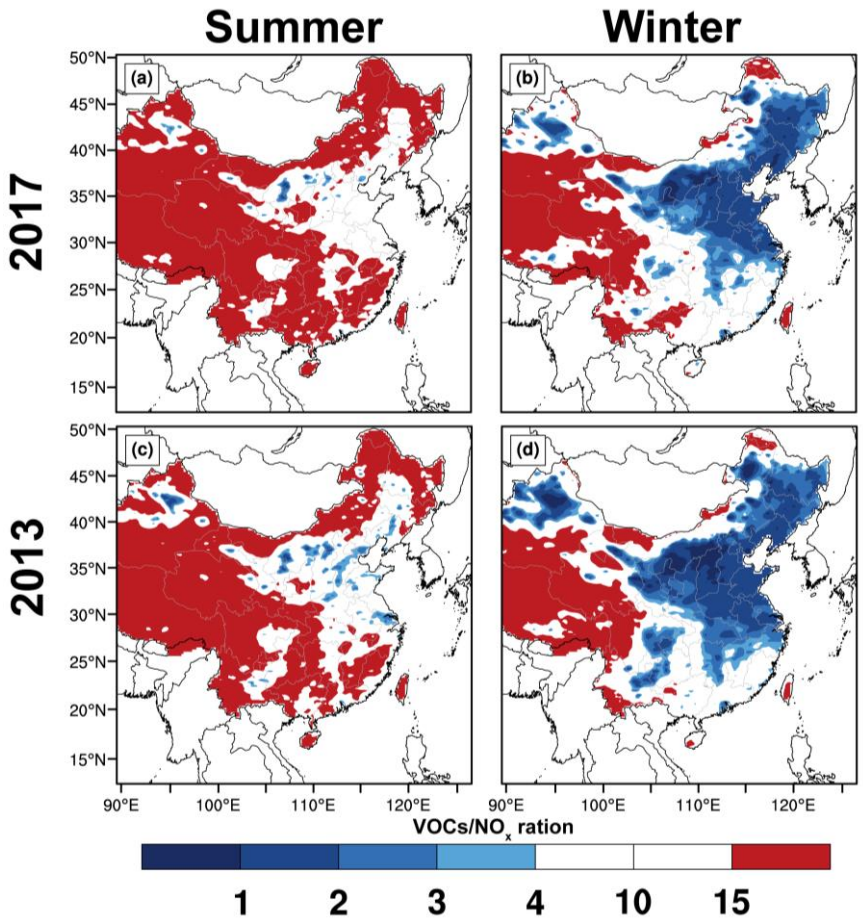

Figure R6. The ratios of VOCs/NO_x calculated from (a, b) BASE_17E17M, and (c, d) BASE_13E13M during the daytime (08:00-17:00 LST) from summer (left) and winter (right).

18. *L290-292, the meteorological effects are comparable or larger or smaller than emissions effects? This should be mentioned.*

Response:

From Figs. R7, compared with 2013, the meteorological conditions in the summer of 2017 promoted the generation of O_3 in the YRD region, but suppressed the generation of O_3 in the BTH, PRD and SCB 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 during summer.

Thanks for reviewer's suggestion, we have added this information in the revised manuscript. **(Page 13, Line 343-349)**

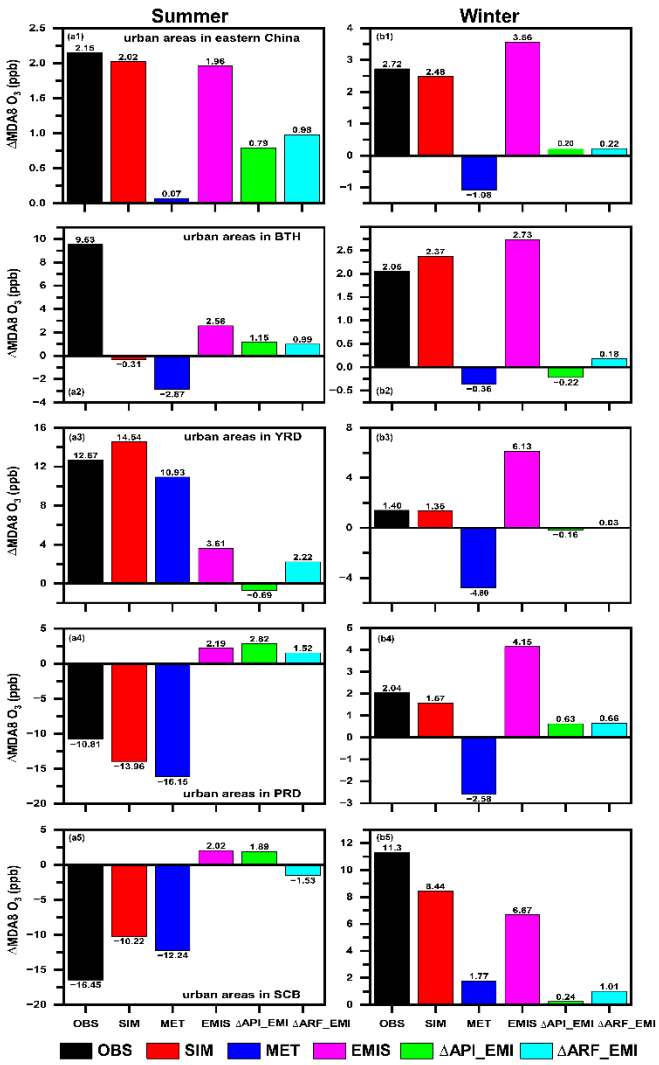

Figure R7. The observed (OBS, black bars) and simulated (SIM, red bars) changes in (left) summer and (right) winter surface-layer MDA8 O_3 from 2013 to 2017. Contributions of changed meteorological conditions alone (MET, blue bars), changed anthropogenic emissions alone (EMI, purple bars), changed aerosol-photolysis 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.

19. *L429-431, you mentioned "multi-pollutants coordinated emissions control strategies", can you specify this and give more details? Liu and Wang, 2020 suggested that "to reduce O_3 levels in major urban and industrial areas, VOC emission controls should be added to the current NOx-SO_2-PM policy". Does your research have similar insights, or can you make other recommendations that could help policymakers?*

Response:

Thanks for reviewer's suggestion. Our suggestion is consistent with Liu and Wang (2020), we hope that the government should not focus on the control of $PM_{2.5}$ pollution (NOx-SO_2-PM policy),

but should pay attention to the synergistic control of multiple pollutants such as O_3 and $PM_{2.5}$.

Technical corrections:

1. *L211, "353 stations" - > "353 meteorological stations"*

   Response:

   Thanks for your suggestion. We have added the "meteorological" in the revised manuscript. (**Page 9, Line 235**)

2. *Figure S5, "from 2013to" - > "from 2013 to"*

   Response:

   Thanks for your suggestion. We have changed the expression in the revised manuscript. (**Page 11 in supporting information**)

3. *Figure 6, "on the right side of each panel" - > "on the upper right side of each panel"*

   Response:

   According to the reviewer's suggestion, we have changed the expression in the revised manuscript. (**Page 37, Line 859**)

4. *Data and code availability should be added.*

   Response:

   According to the reviewer's suggestion, we have added the "Data availability" section in the revised manuscript. (**Page 22, Line 593-600**)

Reference:

Gao, J., Li, Y., Xie, Z., Hu, B., Wang, L., Bao, F., and Fan, S.: The impact of the aerosol reduction on the worsening ozone pollution over the Beijing-Tianjin-Hebei region via influencing photolysis rates, Sci. Total Environ., 821, 153197, https://doi.org/10.1016/j.scitotenv.2022.153197, 2022.

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, Atmospheric Chem. Phys., 20, 6323–6337, https://doi.org/10.5194/acp-20-6323-2020, 2020.

Lou, S., Liao, H., and Zhu, B.: Impacts of aerosols on surface-layer ozone concentrations in China through heterogeneous reactions and changes in photolysis rates, Atmos. Environ., 85, 123–138, https://doi.org/10.1016/j.atmosenv.2013.12.004, 2014.

Reference:

Atkinson, R.: Atmospheric chemistry of VOCs and NOx, Atmos Environ., 34, 2063–2101, https://doi.org/10.1016/S1352-2310(99)00460-4, 2000.

Edson, C. T., Ivan, H.-P. and Alberto, M.: Use of combined observational- and model-derived photochemical indicators to assess the O_3-NO_x-VOC System sensitivity in urban areas, Atmosphere., 8, 22.

https://doi.org/10.3390/ atmos8020022, 2017.

Gipson, G. L.: Science algorithms of the EPA Models-3 community multiscale air quality (CMAQ) modeling system: Chapter 16, process analysis, edited by: Byun, D. W. and Ching, J. K. S., Reported No. EPA/600/R-99/030, U.S. Environmental Protection Agency, Office of Research and Development, Washington, D.C., 1999.

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.

Lakey, P. S. J., George, I. J., Whalley, L. K., Baeza-Romero, M. T., and Heard, D. E.: Measurements of the HO_2 Uptake Coefficients onto Single Component Organic Aerosols, Environmental Science & Technology, 49, 4878-4885, 10.1021/acs.est.5b00948, 2015.

Li, K., Chen, L., Ying, F., White, S. J., Jang, C., Wu, X., Gao, X., Hong, S., Shen, J., Azzi, M. and Cen, K: Meteorological and chemical impacts on ozone formation: a case study in Hangzhou, China, Atmos. Res., 196, https://doi.org/10.1016/ j.atmosres.2017.06.003, 2017.

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 1: The complex and varying roles of meteorology, Atmos. Chem. Phys., 20, 6305–6321, https://doi.org/10.5194/acp-20-6305-2020, 2020a.

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.

Lo, J. C.-F., Yang, Z. L., and Pielke Sr, R. A.: Assessment of three dynamical climate downscaling methods using the Weather Research and Forecasting (WRF) model, J. Geophys. Res., 113, D09112, https://doi.org/10.1029/2007jd009216, 2008.

Qiu, Y., Liao, H., Zhang, R., and Hu, J.: Simulated impacts of direct radiative effects of scattering and absorbing aerosols on surface layer aerosol concentrations in China during a heavily polluted event in February 2014, J. Geophys. Res. Atmos., 122, 5955–5975, doi:10.1002/2016JD026309, 2017.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: from Air Pollution to Climate Change, second ed., John Wiley and Sons, 2006.

Shao, M., Wang, W. J., Yuan, B., Parrish, D. D., Li, X., Lu, K. D., Wu, L. L., Wang, X. M., Mo, Z. W., Yang, S. X., Peng, Y. W., Kuang, Y., Chen, W. H., Hu, M., Zeng, L. M., Su, H., Cheng, Y. F., Zheng, J. Y., Zhang, Y. H.: Quantifying the role of $PM_{2.5}$ dropping in variations of ground-level ozone: Inter-comparison between Beijing and Los Angeles, Sci. Total Environ., https://doi.org/10.1016/j.scitotenv.2021.147712, 2021.

Shu, L., Wang, T., Han, H., Xie, M., Chen, P., Li, M., and Wu, H.: Summertime ozone pollution in the Yangtze River Delta of eastern China during 2013–2017: Synoptic impacts and source apportionment, Environ. Pollut., 257, 113631, https://doi.org/10.1016/j.envpol.2019.113631, 2020.

Shu, L., Xie, M., Wang, T., Gao, D., Chen, P., Han, Y., Li, S., Zhuang, B., and Li, M.: Integrated studies of a regional ozone pollution synthetically affected by subtropical high and typhoon system in the Yangtze River Delta region, China, Atmos. Chem. Phys., 16, 15801–15819, https://doi.org/10.5194/acp-16-15801- 2016, 2016.

Sillman, S.: The relation between ozone, NO_x and hydrocarbons in urban and polluted rural environments, Atmos. Environ., 33, 1821-1845, https://doi.org/ 10.1016/S1352-2310(98)00345-8, 1999.

Taketani, F., Kanaya, Y., and Akimoto, H.: Heterogeneous loss of HO2 by KCl, synthetic sea salt, and natural seawater aerosol particles, Atmospheric Environment, 43, 1660-1665, 2009.

Tan Z, Hofzumahaus A, Lu K, Brown SS, Holland F, Huey LG, et al. No Evidence for a Significant Impact of Heterogeneous Chemistry on Radical Concentrations in the North China Plain in Summer 2014. Environ. Sci. Technolc. 54, 5973-5979, 2020.

Wang, N., Lyu, X., Deng, X., Huang, X., Jiang, F., and Ding, A.: Aggravating O3 pollution due to NOx emission control in eastern China, Sci. Total Environ., 677, 732–744, 2019.

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., Wang, S., Donahue, N. M., Chuang, W., Ruiz, L. H., Ng, N. L., Wang, Y., and Hao, J.: Evaluation of One-Dimensional and Two-Dimensional Volatility Basis Sets in Simulating the Aging of Secondary Organic Aerosol with Smog-Chamber Experiments, Environ. Sci. Technol., 49, 2245–2254, doi:10.1021/es5048914, 2015.

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.

Zhou, M., Zhang, L., Chen, D., Gu, Y., Fu, T.-M., Gao, M., Zhao, Y., Lu, X. and Zhao, B.: The impact of aerosol-radiation interactions on the effectiveness of emission control measures, Environmental Research Letters, 14(2), 024002, https://doi.org/10.1088/1748-9326/aaf27d, 2019.

Zou Q, Song H, Tang M, Lu K. Measurements of HO_2 uptake coefficient on aqueous $(NH_4)_2SO_4$ aerosol using aerosol flow tube with LIF system. Chinese Chemical Letters 2019; 30: 2236-2240.

Thank you very much for your comments and suggestions.