Response to Comments of Reviewer #2

(comments in *italics*)

Manuscript number: acp-2021-119

Title: Impacts of aerosol-photolysis interaction and aerosol-radiation feedback on surface-layer ozone in North China during a multi-pollutant air pollution episode

In this study, Yang et al. investigate the impact of aerosol-radiation interactions on O_3 formation during a multi-pollutant air pollution episode in Northern China. Additionally, the study uses process analysis to analyze how the aerosol-radiation interactions affect O_3 through various physical and chemical mechanisms. This is an interesting research topic with valid research methods and an overall well written and well-structured manuscript. However, the period of analysis is far too short (i.e., 7 days) to robustly quantify the impact of aerosol-radiation impacts in this region or to describe any variability. Additionally, the time period analyzed appears somewhat arbitrary and is nearly a decade removed from current conditions. For these reasons, the manuscript is not currently at the scientific level of the Atmospheric Chemistry and Physics Journal. However, this manuscript would be suitable for publication in ACP if either it is restructured to focus on how the methods used are unique and different from past work or if the authors investigate longer periods to generate more robust analysis and conclusions. Please find my major and minor comments below.

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 respective/combined contributions of aerosol-photolysis interaction (API) and aerosol-radiation feedback (ARF) on O_3 concentrations during multi-pollutant air pollution episodes characterized by high O_3 and $PM_{2.5}$ levels. According to the reviewer's comments, another two complex air pollution episodes are also analyzed for generating general conclusions, and we find that API is the dominant factor for O_3 reduction related to aerosol-radiation interactions during all the simulated episodes (Episode 1: 28 July-3 August 2014; Episode 2: 8-13 July 2015; Episode 3: 5-11 June 2016).

Major Comments:

1. *The novelty of this study is that it is the first time that API and ARF are investigated for synchronous occurrences of high $PM_{2.5}$ and O_3 concentrations. This is a rather broad research question to be focused on only one region and one very minor time period. Why do the authors not conduct simulations for either several of these small pollution episodes in this region or for similar episodes in other locations in China?*

Response:

The high-resolution WRF-Chem model has been widely applied to investigate the evolution mechanisms of air pollutants during short time periods (Gao et al., 2016; Qiu et al., 2017; Gao et al., 2018; Wang et al., 2020). Gao et al. (2016) summarized the general conclusion that haze events were mainly caused by high emissions of air pollutants and unfavorable weather conditions in North China Plain (NCP) by analyzing a simulated pollution episode from WRF-Chem during 14-24 January 2010. According to the results from WRF-Chem, Qiu et al. (2017) reported that the direct radiative effects of scattering aerosols were greater than that of absorbing aerosols in NCP during 21-27 February 2014. Gao et al. (2018) found that the interactions between black carbon and planetary boundary layer (PBL) could influence the surface O_3 concentration in Nanjing during 17 October 2015 by using the process analysis in WRF-Chem.

According to the reviewer's comments, another two complex air pollution episodes (8-13 July 2015 and 5-11 June 2016) in this region are also selected to conduct simulations for generating general conclusions.

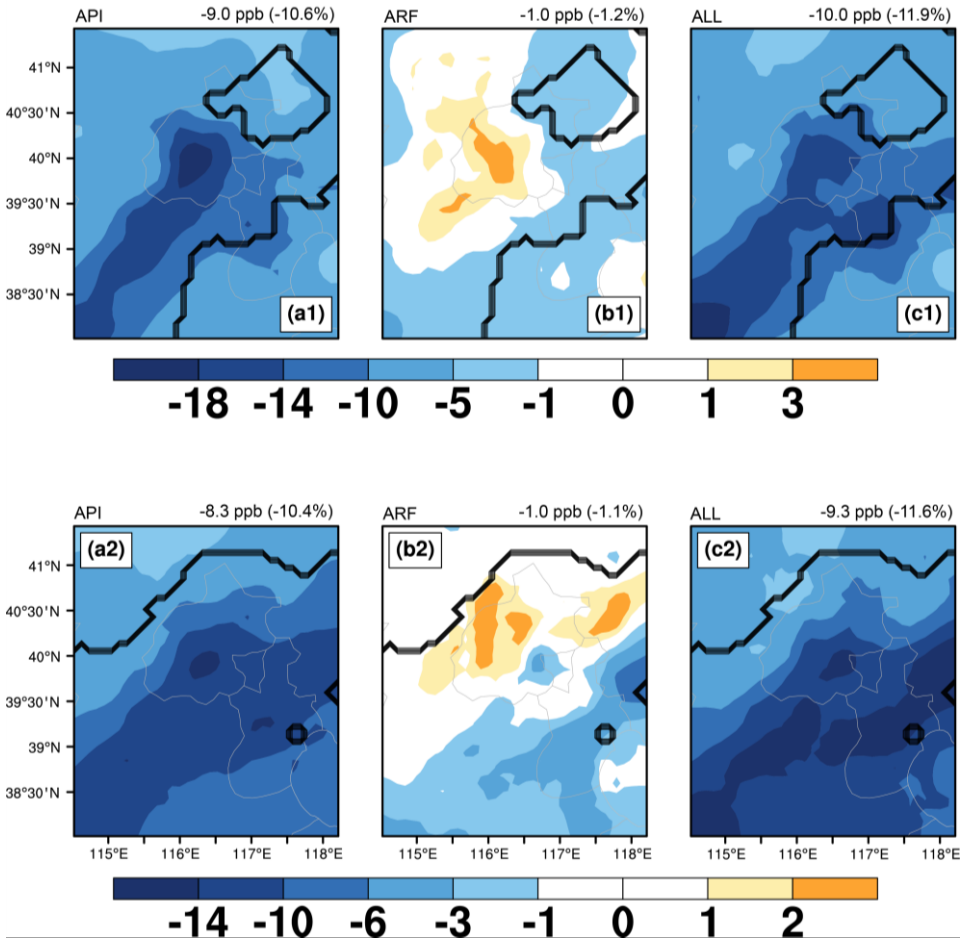

Figure R1. Changes in surface-layer ozone due to (a1-a2) aerosol-photolysis interaction (API), (b1-b2) aerosol-radiation feedback (ARF), and (c1-c2) the combined effects (ALL, defined as API+ARF) during the daytime (08:00-17:00 LST) from 8-13 July 2015 (upper) and 5-11 June 2016 (bottom). The region sandwiched between two black lines is defined as the complex air pollution areas (CAPAs) where the mean daily $PM_{2.5}$ and MDA8 O_3 concentrations in BASE case are larger than 75 μg m^{-3} and 80 ppb. The calculated changes averaged over CAPAs are also shown at the top of each panel.

Simulated air pollutants ($PM_{2.5}$ and O_3) and meteorological variables (T_2, RH_2, and WS_{10}) during 8-13 July 2015 (Episode 2) and 5-11 June 2016 (Episode 3) are compared with observations. In general, both the observed meteorological parameters and pollutant concentrations can be reasonably reproduced by the model, with correlation coefficients (R) of 0.56~0.98 and normalized mean bias (NMB) of –7.1%~+33.4%. More details about the model evaluation are listed in the supporting information (**Text S1**).

As shown in Fig. R1(a1-a2), API alone leads to the decrease in surface O_3 over the entire domain with an average reduction of 9.0 ppb (10.6%) and 8.3 ppb (10.4%) over CAPAs in Episode 2 and Episode 3, respectively. The decreased surface O_3 concentrations over CAPAs due to ARF are only 1.0 ppb (1.2%, Fig. R1(b1)) and 1.0 ppb (1.1%, Fig. R1(b2)) during Episode 2 and Episode 3, respectively. All the results indicate that API is the dominant factor for O_3 reduction related to aerosol-radiation interactions, the same as the conclusion analyzed from the case during 28 July to 3 August 2014 (Episode 1). The combined effects of API and ARF decrease surface O_3 by 10.0 ppb (11.9%) and 9.3 ppb (11.6%) over CAPAs in Episode 2 and Episode 3, respectively. (**Page 13-14, Line 343-365**)

According to the reviewer's suggestion, Figure R1 is added in the supporting information (**Figure S9**).

2. *Given that government controls have substantially changed emissions in the last decade and will continue into future, how will this research remain relevant in the future or how relevant is it to today's air pollution in China, since the period examined is 7 years ago?*

Response:

The stringent Air Pollution Action Plan has been released by the Chinese government in September 2013 to improve the $PM_{2.5}$ air quality. Although the concentrations of $PM_{2.5}$ are decreasing, the concentrations of $PM_{2.5}$ still exceed 35 μg m^{-3}, and the O_3 levels have continued to increase (Dai et al., 2021). Many studies have found that the decreased $PM_{2.5}$ can be one of the important causes leading to the increase in O_3 (Li et al. 2019; Shao et al., 2021). Li et al. (2019) pointed out that the concentrations of $PM_{2.5}$ were decreased by 40% in North China Plain from 2013 to 2017, which reduced the sink of HO_2 on aerosol surfaces and resulted in the increase in O_3 by analyzing simulation results from the GEOS-Chem model. Meanwhile, the concentrations of O_3 can also be influenced by aerosol-radiation interactions, including aerosol-photolysis interaction and aerosol-radiation feedback, which have not been systematically analyzed. The quantification of the impacts of aerosols on O_3 is important to well understand the co-benefits associated with reductions in both aerosols and O_3.

In this study, we investigate the impacts of aerosol-radiation interactions on surface O_3, and find that the combined impacts of weakened photolysis rates and changed meteorological conditions reduce surface-layer O_3 concentrations by up to 11.4 ppb (13.5%). The result can imply that the decreases in $PM_{2.5}$ can lead to the increase in O_3 due to the weakened aerosol-radiation interactions, which indicates that if the government controls the anthropogenic emissions in future by using the same strategy, higher O_3 will be observed. The result can further emphasize the importance of tighter controls in O_3 precursors (i.e., VOCs) to counteract the increased O_3 caused by weakened aerosol-radiation interactions. Therefore, the contributions of different mitigation strategies with the impacts of aerosol-radiation interactions to O_3 air quality will be discussed

detailedly in our future work.

3. *Is the focus of this research the method in which API and ARF are investigated or the impact of API and ARF in North China? If it is the former than the authors need to reword the abstract, conclusions, and objectives to make it clear that this study is a "proof-of-concept" study on how to best investigate API and ARF in high O_3 and $PM_{2.5}$ episodes. If the focus is the latter, the authors need to do additional simulations of other high multi-pollutant episodes, perhaps some closer to current conditions and others in the mid 2000s to see if there is change over time or to make the analysis and conclusions more robust.*

Response:

This study mainly focuses on the impacts of API and ARF in North China. According to the reviewer's comments, another two complex air pollution episodes (8-13 July 2015 and 5-11 June 2016) in this region are also selected to conduct simulations for generating general conclusions. The impacts of API and ARF on O_3 are shown in Fig. R1, and API is the dominant factor for O_3 reduction related to aerosol-radiation interactions. Similar results can also be concluded by analyzing the episode during 28 July to 3 August 2014.

4. *Does this version of WRF-Chem's CBM-Z and MOSAIC modules have a volatility basis set (VBS) option to simulate secondary organic aerosols and if so is it used? Given that, this is a high O_3 and $PM_{2.5}$ episode there should be a substantial amount of secondary organic aerosol from abundant oxidants and precursors that may be missed in the model without an advanced SOA scheme. How do the author's address the impact of SOA on their conclusions?*

Response:

The selected gas-phase chemical mechanism (CBM-Z) and the aerosol model (MOSAIC) in this study do not consider the impacts of secondary organic aerosols (SOA). The same schemes have been widely used in many other studies, which mainly focus on the impacts of aerosol-radiation interactions on air pollutants in North China (Ding et al., 2016; Gao et al., 2016; Qiu et al., 2017; Chen et al., 2019; Zhou et al, 2019; Gao et al., 2020).

Thanks for the reviewer's suggestion, and we will consider the impacts of SOA in our future works. A discussion about the impacts of SOA has been added in the revised manuscript as follows: "Gao et al. (2017) added some SOA formation mechanisms into the MOSAIC module by using the volatility basis set (VBS) in WRF-Chem and found that the surface $PM_{2.5}$ concentrations in urban Beijing were reduced by 1.9 μg m^{-3} due to the weakened ARF effect during Asia-Pacific Economic Cooperation (APEC). Similar magnitude can also be found in Zhou et al. (2019) (-1.8 μg m^{-3}) who did not consider the impacts of SOA in WRF-Chem when analyzing the impacts of weakened ARF on $PM_{2.5}$ during APEC. Therefore, more work should be conducted to explore the impacts of ARF on $PM_{2.5}$ and O_3 concentrations under consideration of SOA in future." (**Page 15, Line 402-412**)

5. *The authors are investigating aerosol radiation interactions, but the authors do not evaluate the model's performance against either radiation balance datasets or aerosol optical depth. Since these parameters are more important than surface*

evaluations of air pollutants to understanding API and ARF, the authors should evaluate their model configuration against satellite AOD and radiation variables such as MODIS or CERES-EBAF.

Response:

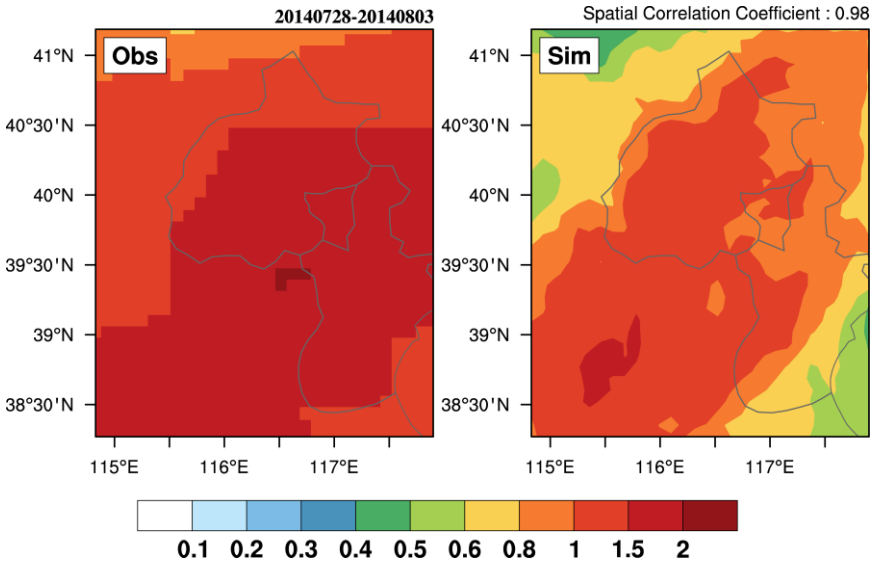

Figure R2. Spatial distributions of aerosol optical depth (AOD) at 550 nm retrieved from MODIS (left) and simulated by WRF-Chem (right) during 28 July to 3 August 2014. The MODIS retrievals are a combination of the standard (over ocean) and "Deep Blue" (over land) products.

Figure R2 shows the spatial distributions of aerosol optical depth (AOD) at 550 nm retrieved from MODIS and simulated by WRF-Chem during 28 July to 3 August 2014. In the WRF-Chem model, the AOD at 550 nm are calculated by using the values at 400 and 600 nm according to the Ångstrom exponent. Analyzing Fig. R2, the model can well reproduce the spatial distribution of observed AOD but slightly underestimate the value. The spatial correlation coefficient between the simulated and observed AOD is 0.98.

According to the reviewer's suggestion, the description of the model evaluation between observed and simulated AOD is added in the revised manuscript (**Page 8, Line 190-196**), and Figure R2 is also added in the supporting information (**Figure S1**).

6. *Are there only three meteorological observation stations in the domain against? If so, why do the authors not also validate their meteorological performance against gridded products like the Climate Research Unit (CRU) datasets to ensure their performance statistics are robust?*

Response:

Thanks to the reviewer's comments. More meteorological observations in the analyzed domain (Table R1) have been used to validate the model results, and the locations of each site are shown in Fig. R3.

Figure R4 shows the time series of observed and simulated T_2, RH_2, WS_{10} and $J[NO_2]$ during 28 July to 3 August 2014. The observed T_2, RH_2 and WS_{10} are averaged from the ten meteorological observation stations, and the photolysis rates of NO_2 are collected from Peking University. Generally, the WRF-Chem model can depict the temporal variation of T_2 fairly well

with R of 0.98 and the mean bias (MB) of -1.5 °C. For RH_2, the R and MB are 0.91 and 0.5%, respectively. Although WRF-Chem overestimates WS_{10} with the MB of 0.7 m s^{-1}, the root-mean-square error (RMSE) is 0.9 m s^{-1}, which is smaller than the threshold of model performance criteria (2 m s^{-1}) proposed by Emery et al. (2001). The predicted $J[NO_2]$ agrees well with the observations with R of 0.97 and NMB of 6.8%.

According to the reviewer's comments, we have modified the model evaluation in the revised manuscript (**Page 8, Line 198-208**).

The gridded products like the Climate Research Unit (CRU) datasets covers a large area and a longtime period, which aims to improve scientific understanding of the climate system and its interactions with society. However, the spatial (0.5° × 0.5°) and temporal (monthly) resolution may be too coarse to validate the model performance for generating robust results.

Table R1. Locations of the ten stations from NOAA's National Climatic Data Center used in this study.

Station	Latitude (°)	Longitude (°)
Yuxian	39.833	114.567
Fengning	41.2	116.633
Zhangjiakou	40.783	114.883
Huailai	40.417	115.5
Chengde	40.967	117.917
Beijing	40.08	116.585
Tianjin	39.1	117.167
Binhai	39.124	117.346
Tangshan	39.65	118.1
Baoding	38.733	115.483

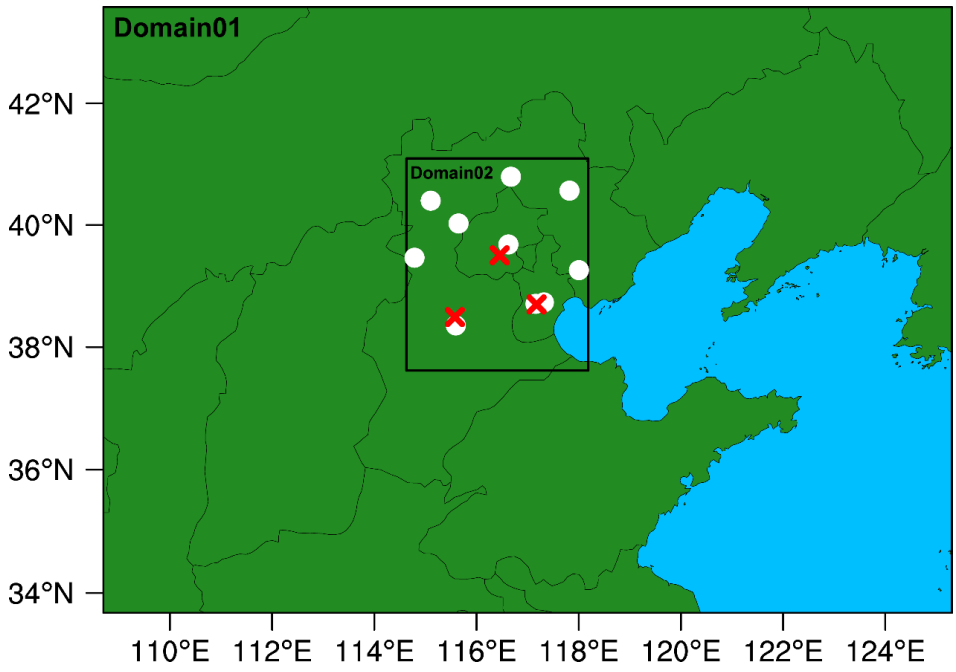

Figure R3. Map of the two WRF-Chem modeling domains with the locations of meteorological (white dots) and environmental (red crosses) observation sites used for model evaluation.

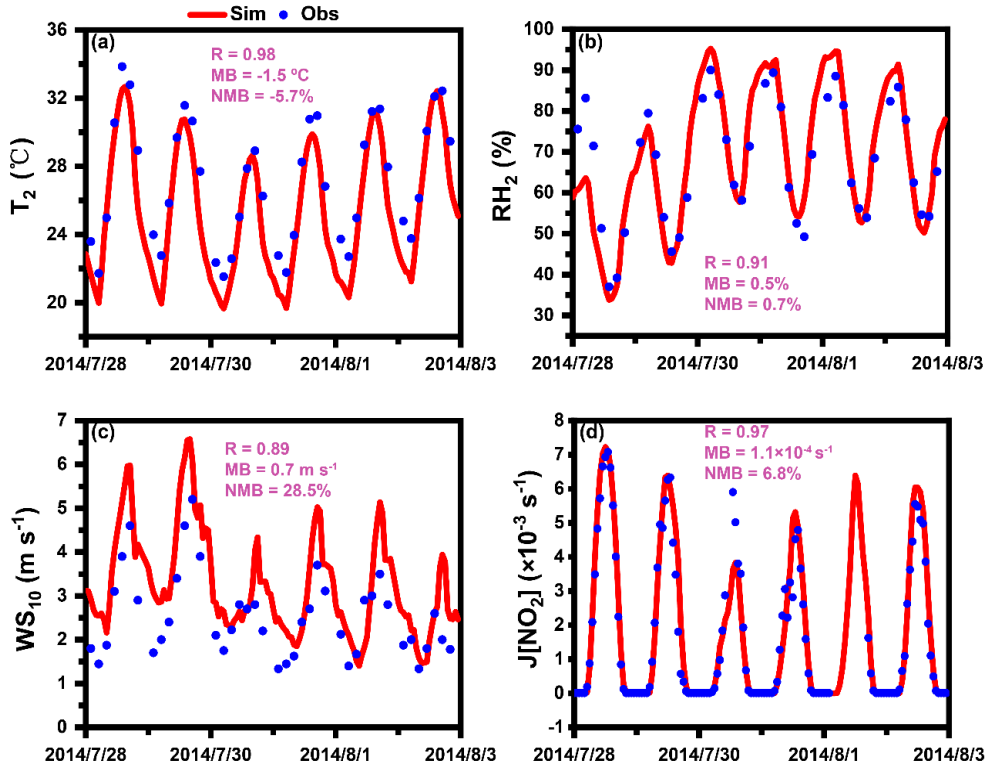

Figure R4. Time series of 3-hourly observed (blue dots) and hourly simulated (red lines) (a) 2-m temperature (T_2), (b) 2-m relative humidity (RH_2), (c) wind speed at 10 m (WS_{10}) averaged over ten meteorological observation stations, and (d) surface photolysis rate of NO_2 ($J[NO_2]$) during 28 July to 3 August 2014. The calculated correlation coefficient (R), mean bias (MB), and normalized mean bias (NMB) are also shown.

7. *Given that interactions between O_3 and $PM_{2.5}$ are non-linear, how do the authors justify using a simple ratio value (i.e., ROP) to relate these interactions? If this ratio does not account for non-linearity, how useful is this value?*

Response:

Thanks to the reviewer's suggestion. As the relationship between O_3 and $PM_{2.5}$ is non-linear, and the simple index of ROP can not fully represent the impacts of aerosols on surface O_3, so we delete the ROP in the revised manuscript.

8. *The axis labels and legends of Figure 7 are difficult to read. Either each panel should be larger overall or the font sizes of the axes and legends need increased.*

Response:

According to the reviewer's suggestion, we have modified the axis labels and legends of Figure 7 and the other figures in the revised manuscript. (**Page 32**)

Minor Comments:

1) *In the abstract, there is no context for the values listed. Further reading into the manuscript reveals that these values are the averages in the areas of the complex air pollution areas. The authors should briefly state that these values are for daytime average changes in complex air pollution areas in the*

abstract. I would also suggest adding a more processed based explanation of the changes in atmospheric state rather than simply listing a long series of values. For example, the authors could state something similar to the following: "Aerosol radiation interactions lead to shortwave dimming at the earth's surface of X, which reduce photolysis rates by X. The dimming stabilizes the atmosphere via surface cooling of X, which reduces PBL height by X. The stabilized atmosphere increases saturation in the lower atmosphere by X. etc…."

Response:

According to the reviewer's suggestion, we have revised the explanation in the abstract as follows: "Our results show that aerosol-radiation interactions decreased the daytime shortwave radiation at surface by 93.2 W m^{-2} averaged over the complex air pollution areas. The dimming effect reduced the 2 m temperature and near-surface photolysis rates of $J[NO_2]$ and $J[O^1D]$ by 0.56 °C, 1.8×10^{-3} s^{-1} and 6.1×10^{-6} s^{-1}, respectively. However, the daytime shortwave radiation in the atmosphere was increased by 72.8 W m^{-2}, which made the atmosphere more stable. The stabilized atmosphere decreased the planetary boundary layer height and 10 m wind speed by 129.0 m and 0.12 m s^{-1}, respectively, and increased the relative humidity at 2 m by 2.4%." (**Page 2, Line 24-32**)

2) *Make it clear throughout the manuscript when you are referring to surface level O_3 and $PM_{2.5}$.*

Response:

According to the reviewer's suggestion, we have revised the expressions in the whole manuscript.

3) *Lines 179-181: The missing $PM_{2.5}$ could also be from missing SOA formation pathways, as mentioned above, if no advanced SOA formulations are used.*

Response:

Thanks for your suggestion. The selected aerosol model (MOSAIC) in this study does not consider the impacts of secondary organic aerosols (SOA), and we have added the explanation in the revised manuscript as follows: "The failure to reproduce $PM_{2.5}$ peak values may be attributed to incomplete treatments of chemical reactions in WRF-Chem, e.g., missing the heterogeneous chemistry in the model (Cheng et al., 2016) and the lack of secondary organic aerosols (SOA) formation pathways in the aerosol module (Chen et al., 2019)." (**Page 7-8, Line 183-188**)

4) *Is "downward shortwave radiation in the atmosphere" the SWDNT variable from WRF-Chem? If so, the name of this variable is "downward shortwave radiation at the top of the atmosphere".*

Response:

Thanks for your comments. In the WRF-Chem model, SWDNT (SWUPT) means the download (upward) shortwave radiation at the top of atmosphere, and SWDNB (SWUPB) represents the download (upward) shortwave radiation at the surface. According to Zhao et al. (2011), the shortwave radiation in the atmosphere (ATM_SW) can be calculated as the

difference between TOP_SW (the net shortwave radiation at the top of atmosphere, i.e., SWDNT minus SWUPT) and BOT_SW (the net shortwave radiation at the surface, i.e., SWDNB minus SWUPB).

According to the reviewer's suggestion, we have changed the expressions of BOT_SW (shortwave radiation at the surface) and ATM_SW (shortwave radiation in the atmosphere) in the whole revised manuscript.

5) *Lines 217-218: If ATM_SW is the SWDNT variable, what is causing it to increase? SWDNT is usually controlled by the solar constant. Is it possible this is reflected upward shortwave (SWUPT)?*

Response:

ATM_SW represents the shortwave radiation in the atmosphere, and it can be calculated by the following equation: ATM_SW = (SWDNT - SWUPT) – (SWDNB - SWUPB).

6) *Lines 248-249: This should be revised to make it clearer that ARF primarily impacts O_3 through changing the NOx distribution.*

Response:

According to the comments of Reviewer#1, we have deleted this sentence.

7) *Lines 270-281: Is VMIX increasing surface O_3 because it is mixing down higher O_3 concentrations from aloft or because vertical mixing is suppressed due to a stable atmosphere?*

Response:

VMIX increases the surface O_3 concentrations by transporting the higher O_3 from aloft to the surface layer. Similar results can also be found in previous studies (Tang et al., 2017; Xing et al., 2017; Gao et al., 2018).

8) *Lines 282-294: Why does the VMIX contribution increase because of API?*

Response:

Analyzing the vertical profiles of the differences in contributions from each physical/chemical process to hourly O_3 variations caused by API in Fig. 8(b), we found that the contribution of VMIX_DIF is negative in the aloft (among the 9^{th} and the 13^{th} layers), while it turns to be positive at the lower seven layers, and the positive contribution increases as the height decreases. The positive variation in VMIX due to API may be associated with the different vertical gradient of O_3 between BASE and NOAPI cases.

Similar results can also be found in Gao et al. (2020), who concluded that the increased vertical gradients of O_3 due to API could enhance the vertical entrainment.

9) *Lines 295-301: Explain why VMIX_DIF and CHEM_DIF are positive during the day due to ARF.*

Response:

When the impacts of ARF are considered, PBLH is decreased over CAPAs (Fig. S3(b3)), which indicates that the suppressed PBL in NOAPI restrains the vertical turbulence and prevents O_3 being transported from aloft to surface, resulting in lower O_3 concentrations at

surface when comparing with the simulation results of NOALL. However, as the evolution in boundary layer during the daytime, more O_3 can be diffused from the upper layers to the surface in NOAPI, and the differences in hourly variation in surface O_3 due to vertical mixing between NOAPI and NOALL are positive. Similar results can also be found in Gao et al. (2018).

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 R5(a-c), O_3 are mainly formed under the VOC-limited and the transition regimes in CAPAs, which means that the increased concentrations of VOCs and NO_x are favorable for ozone chemical production. As shown in Fig. R5(e) and (h), both the surface concentrations of VOCs and NO_x are increased when the impacts of ARF are considered. Thus, the contribution of CHEM in NOAPI is larger than that in NOALL. Similar results can also be found in Gao et al. (2018).

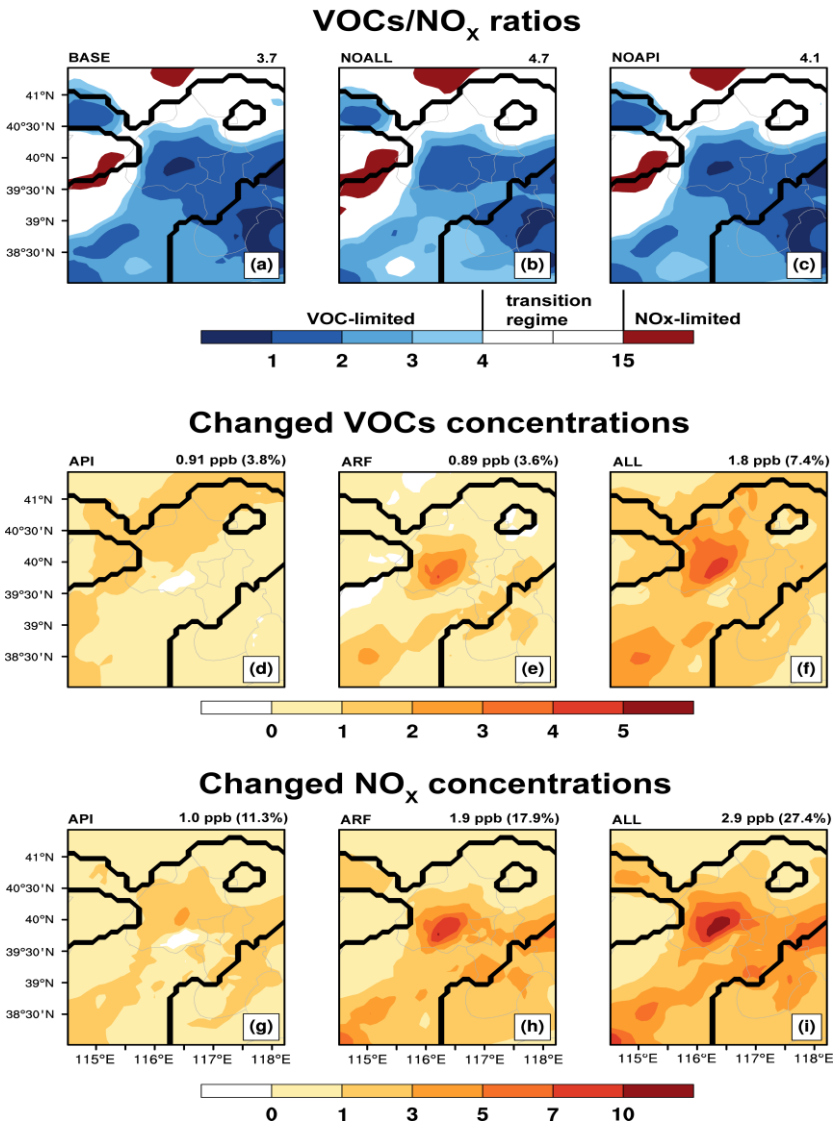

Figure R5. The ratios of VOCs/NO_x calculated from (a) BASE, (b) NOALL, and (c) NOAPI. The changed surface-layer concentrations of VOCs and NO_x (NO_2+NO, ppb) caused by (d, g) API, (e, h) ARF, and (f, i) ALL during the daytime (08:00-17:00 LST) from 28 July to 3 August 2014. The calculated values averaged over CAPAs are also shown at the top of each panel.

10) *Lines 315-316: Explain how different vertical O_3 gradients can cause this change.*

Response:

Since the VMIX is closely dependent on atmospheric turbulence and vertical gradients of O_3 concentration. The API will increase vertical gradients of O_3 to enhance the vertical entrainment (Gao et al., 2020).

Line Comments:

1) *Line 49: This should be "Earth's radiative balance" or "Earth's energy balance"*

Response:

Thanks for your suggestion. We have changed the expression in the revised manuscript. (**Page 3, Line 48**)

2) *Lines 54-56: Are these studies all focused on China? If so, state that in the sentence. Change "were" to "are".*

Response:

According to the reviewer's suggestion, we have changed the expression in the revised manuscript. (**Page 3, Line 54**)

3) *Lines 56-63: State the domain and time period of Gao et al., (2015) at the beginning of this statement rather than the end*

Response:

According to the reviewer's suggestion, we have changed the expression in the revised manuscript. (**Page 3, Line 55-62**)

4) *Line 66: Add "the" before North China Plain*

Response:

Thanks for your suggestion. We have added the "the" before North China Plain in the revised manuscript. (**Page 3, Line 64**)

5) *Lines 66-67: If this is referring to surface $PM_{2.5}$ concentrations, add "surface" before $PM_{2.5}$ concentrations.*

Response:

Thanks for your suggestion. We have added the "surface" before $PM_{2.5}$ concentrations in the revised manuscript. (**Page 3, Line 65**)

6) *Line 204: should be "attention"*

Response:

Thanks for your suggestion. We have changed the expression in the revised manuscript. (**Page 9, Line 218**)

7) *Line 256: Center align the equation.*

Response:

This equation has been deleted.

8) *Line: 259: Why are there parentheses in the units?*

Response:

This sentence has been deleted.

9) *Lines 288-289: This sentence is a little confusing. Is Net_DIF the sum of CHEM_DIF, VMIX_DIF, and ADV_DIF? If so, state that explicitly and then indicate what Net_DIF describes.*

Response:

Thanks for your suggestion. We have defined the NET_DIF in the revised manuscript. (**Page 11, Line 291-292**)

10) *Line 321: Remove "in the"*

Response:

According to the reviewer's suggestion, we have deleted it in the revised manuscript.

11) *Line 361: Remove "the contribution from VMIX and"*

Response:

According to the reviewer's suggestion, we have deleted it in the revised manuscript.

12) *Line 373: Either "A recent study" or "Recent studies have"*

Response:

According to the reviewer's suggestion, we have changed the expression in the revised manuscript. (**Page 15, Line 396**)

Reference:

Chen, L., Zhu, J., Liao, H., Gao, Y., Qiu, Y., Zhang, M., Liu, Z., Li, N., and Wang, Y.: Assessing the formation and evolution mechanisms of severe haze pollution in the Beijing–Tianjin–Hebei region using process analysis, Atmos. Chem. Phys., 19, 10845–10864, https://doi.org/10.5194/acp-19-10845-2019, 2019.

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Thank you very much for your comments and suggestions.