# **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 O3 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 O3 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 pointto-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 (Episode1: 28 July-3 August 2014; Episode2: 8-13 July 2015; Episode3: 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<sub>3</sub> 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.** The changes in surface-layer ozone due to (a) aerosol-photolysis interaction (API), (b) aerosol-radiation feedback (ARF), and (c) the combined effects (ALL, defined as API+ARF) in the daytime (08:00-17:00 LST) during 28 July to 3 August 2014 (Episode1), 8-13 July 2015 (Episode2) and 5-11 June 2016 (Episode3). 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  $\mu$ g m<sup>-3</sup> and 80 ppb. The calculated mean changes (percentage changes) avaraged over CAPAs are also shown at the top of each panel.

Simulated air pollutants (PM<sub>2.5</sub> and O<sub>3</sub>) and meteorological variables (T<sub>2</sub>, RH<sub>2</sub>, WS<sub>10</sub>, and J[NO<sub>2</sub>]) during 28 July to 3 August 2014 (Episode1), 8-13 July 2015 (Episode2) and 5-11 June 2016 (Episode3) 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  $0.56 \sim 0.98$  and normalized mean bias (NMB) of  $-12.0\% \sim +33.4\%$ . More details about the model evaluation are listed in the section 3 in the revised manuscript (**Page 7-8, Line 181-212**). The impacts of aerosol radiation effects on meteorological variables can be found in section 4.1 and 4.2 in the revised manuscript during these three episodes (**Page 9-10, Line 223-254**).

As shown in Fig. R1(a1-a3), API alone leads to overall surface O<sub>3</sub> decreases over the entire domain with average reductions of 8.5 ppb (10.1%), 9.0 ppb (10.6%) and 8.3 ppb (10.4%) over CAPAs in the three episodes, respectively. The changes can be explained by the substantially diminished UV radiation due to aerosol loading, which significantly weakens the efficiency of photochemical reactions and restrains O<sub>3</sub> formation. However, the decreased surface O<sub>3</sub> concentrations due to ARF are only 2.9 ppb (3.1%, Fig. R1(b1)), 1.0 ppb (1.2%, Fig. R1(b2)) and 1.0 ppb (1.1%, Fig. R1(b3)) for the three episodes, which indicates that API is the dominant way for O<sub>3</sub> reduction related to aerosol-radiation interactions. Fig. R1(c1-c3) presents the combined effects of API and ARF. Generally, aerosol-radiation interactions decrease the surface O<sub>3</sub> concentrations by 11.4 ppb (13.5%), 10.0 ppb (11.9%) and 9.3 ppb (11.6%) averaged over CAPAs in the three episodes, respectively. (Page 10, Line 256-269)

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<sup>-3</sup>, and the O<sub>3</sub> 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<sub>3</sub> (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<sub>2</sub> on aerosol surfaces and resulted in the increase in O<sub>3</sub> by analyzing simulation results from the GEOS-Chem model. Meanwhile, the concentrations of O<sub>3</sub> 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<sub>3</sub> is important to well understand the co-benefits associated with reductions in both aerosols and O<sub>3</sub>.

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  $9.3 \sim 11.4$  ppb. 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 (e.g., 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<sub>3</sub> and PM<sub>2.5</sub> 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<sub>3</sub> are shown in Fig. R1, and API is the dominant factor for O<sub>3</sub> reduction related to aerosol-radiation interactions in these three episodes.

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<sub>3</sub> and PM<sub>2.5</sub> 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<sup>-3</sup> 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<sup>-3</sup>) 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 14-15, Line 391-401**)

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.** Comparison of observed and simulated AOD at 550 nm in Beijing (39.98°N, 116.38°E). The observed AOD during the three episodes are collected from AERONET.

Previous studies found that MODIS retrievals tended to overestimate AOD in the NCP during polluted events compared with AERONET AOD (Gao et al., 2015; Li et al., 2016). Therefore, we mainly focus on the comparisons between simulated AOD values and AERONET observations in this work. Figure R2 shows the correlation between observed and simulated AOD at 550 nm in Beijing. In the WRF-Chem model, the AOD at 550 nm are calculated by using the values at 400 and 600 nm according to the Angstrom exponent. Analyzing Fig. R2, the model can reproduce the observed AOD with R of 0.7 and NMB of 7.9%.

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 190-193**), 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 T2, RH2, WS10 and J[NO2] during

the three episodes. The observed  $T_2$ ,  $RH_2$ ,  $WS_{10}$  are averaged over the ten meteorological observation stations, and the J[NO<sub>2</sub>] are measured at Peking University. Most of the monitored J[NO<sub>2</sub>] in Episode3 are unavailable, so the comparison of J[NO<sub>2</sub>] in Episode3 is not shown. Generally, the model can depict the temporal variations of  $T_2$  fairly well with R of 0.98 and the mean bias (MB) of -1.9~-0.9 °C. For RH<sub>2</sub>, the R and MB are 0.91~0.97 and -4.0%~1.9%, respectively. Although WRF-Chem model overestimates WS<sub>10</sub> with the MB of 0.6~0.9 m s<sup>-1</sup>, the R for WS<sub>10</sub> is 0.70~0.89 and the root-mean-square error (RMSE) is 0.9~1.5 m s<sup>-1</sup>, which is smaller than the threshold of model performance criteria (2 m s<sup>-1</sup>) proposed by Emery et al. (2001). The positive bias in wind speed can also be reproduced in other studies (Zhang et al., 2010; Gao et al., 2015; Liao et al., 2015; Qiu et al., 2017). The predicted J[NO<sub>2</sub>] agrees well with the observations with R of 0.97~0.98 and NMB of 6.8%~6.9%.

According to the reviewer's comments, we have modified the model evaluation in the revised manuscript. (Page 8, Line 195-208)

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  $(0.5^{\circ} \times 0.5^{\circ})$  and temporal (monthly) resolution may be too coarse to validate the model performance for generating robust results.

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

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



**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<sub>2</sub>), (b) 2-m relative humidity (RH<sub>2</sub>), (c) wind speed at 10 m (WS<sub>10</sub>) averaged over ten meteorological observation stations, and (d) surface photolysis rate of NO<sub>2</sub> (J[NO<sub>2</sub>]) during 28 July to 3 August 2014 (Episode1, a1-d1), 8-13 July 2015 (Episode2, a2-d2) and 5-11 June 2016 (Episode3, a3-c3). The calculated correlation coefficient (R), mean bias (MB), normalized mean bias (NMB) and root-mean-square error (RMSE) 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 92.4~100.3 W m<sup>-2</sup> averaged over the complex air pollution areas in these three episodes. The dimming effect reduced the near-surface photolysis rates of J[NO<sub>2</sub>] and J[O<sup>1</sup>D] by  $1.8 \times 10^{-3} \sim 2.0 \times 10^{-3} \text{ s}^{-1}$  and  $5.7 \times 10^{-6} \sim 6.3 \times 10^{-6} \text{ s}^{-1}$ , respectively. However, the daytime shortwave radiation in the atmosphere was increased by 72.8~85.2 W m<sup>-2</sup>, which made the atmosphere more stable. The stabilized atmosphere decreased the planetary boundary layer height and 10 m wind speed by 129.0~249.0 m and 0.05~0.12 m s<sup>-1</sup>, respectively." (Page 2, Line 25-33)

2) Make it clear throughout the manuscript when you are referring to surface level O<sub>3</sub> and PM<sub>2.5</sub>.

#### **Response:**

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

3) Lines 179-181: The missing PM<sub>2.5</sub> 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). We have deleted this sentence in the revised manuscript.

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*<sub>3</sub> 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<sub>3</sub> concentrations by transporting the higher O<sub>3</sub> 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<sup>th</sup> and the 13<sup>th</sup> 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. S4(a3-c3)), 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<sub>x</sub> ratio is calculated to classify sensitivity regimes and to indicate the possible O<sub>3</sub> responses to changes in VOCs and/or NO<sub>x</sub> concentrations. O<sub>3</sub> production is VOC-limited if the ratio is less than 4, and it is NO<sub>x</sub>-limited if the ratio is larger than 15 (Edson et al., 2017; Li et al., 2017). The ratio of VOCs/NO<sub>x</sub> 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-f), O<sub>3</sub> are mainly formed under the VOC-limited and the transition regimes in CAPAs, which means that the increased concentrations of VOCs and NO<sub>x</sub> are favorable for ozone chemical production. As shown in Fig. R5(g-i) and (j-l), both the surface concentrations of VOCs and NO<sub>x</sub> 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<sub>x</sub> calculated from (a-c) NOALL, and (d-f) NOAPI. The changed surface-layer concentrations of (g-i) VOCs and (j-l) NO<sub>x</sub> (NO<sub>2</sub>+NO, ppb) caused by ARF during the daytime (08:00-17:00 LST) from Episode1 to Episode3. 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 49)

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 55)

*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 56-63)

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 66)

5) Lines 66-67: If this is referring to surface PM<sub>2.5</sub> concentrations, add "surface" before PM<sub>2.5</sub> concentrations.

#### **Response:**

Thanks for your suggestion. We have added the "surface" before  $PM_{2.5}$  concentrations in the revised manuscript. (Page 3, Line 66)

# 6) Line 204: should be "attention"

#### **Response:**

Thanks for your suggestion. We have changed the expression in the revised manuscript. (**Page 9**, **Line 216**)

#### 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 297**)

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 14, Line 385)

#### **Reference:**

Dai, H., Zhu, J., Liao, H., Li, J., Liang, M., Yang, Y., and Yue, X.: Co-occurrence of ozone and PM2.5 pollution in

the Yangtze River Delta over 2013-2019: Spatiotemporal distribution and meteorological conditions, Atmos. Res.,

249, 105363, 2021.

- Ding, A. J., Huang, X., Nie, W., Sun, J. N., Kerminen, V. M., Petäjä, T., Su, H., Cheng, Y. F., Yang, X. Q., Wang, M. H., Chi, X. G., Wang, J. P., Virkkula, A., Guo, W. D., Yuan, J., Wang, S. Y., Zhang, R. J., Wu, Y. F., Song, Y., Zhu, T., Zilitinkevich, S., Kulmala, M., and Fu, C. B.: Enhanced haze pollution by black carbon in megacities in China, Geophys. Res. Lett., 43, 2873–2879, https://doi.org/10.1002/2016gl067745, 2016.
- Edson, C. T., Ivan, H.-P. and Alberto, M.: Use of combined observational- and model-derived photochemical indicators to assess the O<sub>3</sub>-NO<sub>x</sub>-VOC System sensitivity in urban areas, Atmosphere., 8, 22. https://doi.org/10.3390/ atmos8020022, 2017.
- Emery, C., Tai, E., and Yarwood, G.: Enhanced meteorological modeling and performance evaluation for two Texas ozone episodes, in: Prepared for the Texas Natural Resource Conservation Commission, ENVIRON International Corporation, Novato, CA, USA, 2001.
- 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.
- Gao, J., Li, Y., Zhu, B., Hu, B., Wang, L., and Bao, F.: What have we missed when studying the impact of aerosols on surface ozone via changing photolysis rates?, Atmos. Chem. Phys., 20, 10831–10844,

https://doi.org/10.5194/acp-20-10831-2020, 2020.

- Gao, M., Carmichael, G. R., Wang, Y., Saide, P. E., Yu, M., Xin, J., Liu, Z., and Wang, Z.: Modeling study of the 2010 regional haze event in the North China Plain, Atmos. Chem. Phys., 16, 1673–1691, doi:10.5194/acp-16-1673-2016, 2016.
- Gao, M., Liu, Z., Wang, Y., Lu, X., Ji, D. and Wang, L.: Distinguishing the roles of meteorology, emission control measures, regional transport, and co-benefits of reduced aerosol feedbacks in "APEC" Blue, Atmos. Environ., 167, 476–486, 420 doi:10.1016/j.atmosenv.2017.08.054, 2017.
- Gao, Y., Zhang, M., Liu, Z., Wang, L., Wang, P., Xia, X., Tao, M., and Zhu, L.: Modeling the feedback between aerosol and meteorological variables in the atmospheric boundary layer during a severe fog-haze event over the North China Plain, Atmos. Chem. Phys., 15, 4279–4295, doi:10.5194/acp-15-4279-2015, 2015.
- Li, K., Liao, H., Zhu, J., and Moch, J. M.: Implications of RCP emissions on future PM<sub>2.5</sub> air quality and direct radiative forcing over China, J. Geophys. Res. Atmos., 121, 12,985–13,008, doi:10.1002/2016JD025623, 2016.
- 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.
- 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.
- 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<sub>2.5</sub> 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.
- Sillman, S.: The relation between ozone, NO<sub>x</sub> and hydrocarbons in urban and polluted rural environments, Atmos. Environ., 33, 1821-1845, https://doi.org/ 10.1016/S1352-2310(98)00345-8, 1999.
- Tang, G. Q., Zhu, X.W., Xin, J. Y., Hu, B., Song, T., Sun, Y., Zhang, J. Q., Wang, L. L., Cheng, M. T., Chao, N., Kong, L. B., Li, X., and Wang, Y. S.: Modelling study of boundary-layer ozone over northern China – Part I: Ozone budget in summer, Atmos. Res., 187, 128–137, 2017.
- Wang, T., Huang, X., Wang, Z., Liu, Y., Zhou, D., Ding, K., Wang, H., Qi, X. and Ding, A.: Secondary Aerosol Formation and its Linkage with Synoptic Conditions during Winter Haze Pollution over Eastern China, Sci. Total Environ., 730, 138888, 2020.
- Xing, J., Wang, J. D., Mathur, R., Wang, S. X., Sarwar, G., Pleim, J., Hogrefe, C., Zhang, Y. Q., Jiang, J. K., Wong, D. and Hao, J. M.: Impacts of aerosol direct effects on tropospheric ozone through changes in atmospheric dynamics and photolysis rates, Atmos. Chem. Phys., 17, 9869–9883, https://doi.org/10.5194/acp-17-9869-2017, 2017.
- Zhao, C., Liu, X., Ruby Leung, L., and Hagos, S.: Radiative impact of mineral dust on monsoon precipitation variability over West Africa, Atmos. Chem. Phys., 11, 1879–1893, https://doi.org/10.5194/acp-11-1879-2011, 2011.
- Zhou, M., Zhang, L., Chen, D., Gu, Y., Fu, T.-M., Gao, M., Zhao, Y., Lu, X. and Zhao, B.: The impact of aerosolradiation interactions on the effectiveness of emission control measures, Environmental Research Letters, 14(2), 024002, https://doi.org/10.1088/1748-9326/aaf27d, 2019.

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