Response to Comments of Editor

(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

The last draft was marked "major revision" but it appears to me that no major revision was made. In fact, no changes were made at all to the Introduction and Methods sections. The major comment from the reviewer, which I reiterated, was that the paper does not sufficiently motivate the focus on 3 episodes, nor discuss the relevance of the results for other periods of time. The paper still does not state why these 3 episodes were chosen. The paper also seems to be motivated to study high pollution conditions, but that is not stated in the major objectives. And while the authors responded to my comments and the reviewer's first comment, no changes were made in the paper itself to clarify these points.

What is the goal of the paper - to consider high air pollution conditions, and for what purpose?

Why were the three episodes chosen, and what do we hope to learn more generally for other conditions?

How do the conclusions based on the three episodes relate with other conditions during the years studied (2014-16) and to what extent are they relevant for other years before or after?

Response:

Thanks for the editor's valuable comments. Following your suggestions, substantial revision has been made in the revised paper. The newly updated responses and the final revised manuscript can clearly show how the manuscript is improved.

Response to "*What is the goal of the paper - to consider high air pollution conditions, and for what purpose?*"

We have **added** the following paragraphs in the **Introduction** section to clarify the goal of this work and why we consider high pollution events:

"The characteristics of air pollution in China during recent years are changing from the single pollutant (e.g., $PM_{2.5}$, particulate matter with an aerodynamic equivalent diameter of 2.5 μm or less) to multiple pollutants (e.g., $PM_{2.5}$ and ozone (O_3)) (Zhao et al., 2018; Zhu et al., 2019), and the synchronous occurrence of high $PM_{2.5}$ and O_3 concentrations has been frequently observed, especially during the warm seasons (Dai et al., 2021; Qin et al., 2021). Qin et al. (2021) reported that the co-occurrence of $PM_{2.5}$ and O_3 pollution days (days with $PM_{2.5}$ concentration > 75 μg m^{-3} as well as maximum daily 8 h average ozone concentration > 80 ppb) exceeded 324 days in eastern China during 2015-2019. Understanding the complex air pollution is essential for making plans to improve air quality in China."

"The present study aims to quantify the respective/combined impacts of ARF and API on surface O_3 concentrations by using the WRF-Chem model, and to identify the prominent physical and/or chemical processes responsible for ARF and API effects by using an integrated process rate

(IPR) analysis embedded in the WRF-Chem model."

Response to "*Why were the three episodes chosen, and what do we hope to learn more generally for other conditions?*"

We had three episodes in the previous version of the manuscript. In order to cover different years, a new complex air pollution episode (Episode4, 28 June to 3 July 2017) is **added** in the revised manuscript. As shown in Fig. 6 (see below) of the revised manuscript, all the episodes show same conclusion that the reduction in O_3 by API is larger than that by ARF.

To clarify the reasons of choosing these four episodes, we have **added** the following paragraph in the **Introduction** section:

"We carry on simulations and analyses on four multi-pollutant air pollution episodes (Episode1: 28 July to 3 August 2014; Episode2: 8-13 July 2015; Episode3: 5-11 June 2016; Episode4, 28 June to 3 July 2017) in North China with high O_3 and $PM_{2.5}$ levels (the daily mean $PM_{2.5}$ and the maximum daily 8-h average O_3 concentration are larger than 75 μg m^{-3} and 80 ppb, respectively). These episodes are selected because (1) these events with high concentrations of both $PM_{2.5}$ and O_3 are the major subjects of air pollution control, (2) high concentrations of both $PM_{2.5}$ and O_3 allow one to obtain the strongest signals of ARF and API, (3) the measurements of J[NO_2] during 2014 and 2015 from Peking University site (Wang et al., 2019) can help to constrain the simulated photolysis rates of NO_2, and (4) selected events cover different years of 2014 to 2017 during which the governmental Air Pollution Prevention and Control Action Plan was implemented (the changes in emissions and observed $PM_{2.5}$ in the studied region during 2014-2017 are shown in Fig. S1). We expect that the conclusions obtained from multiple episodes represent the general understanding of the impacts of ARF and API."

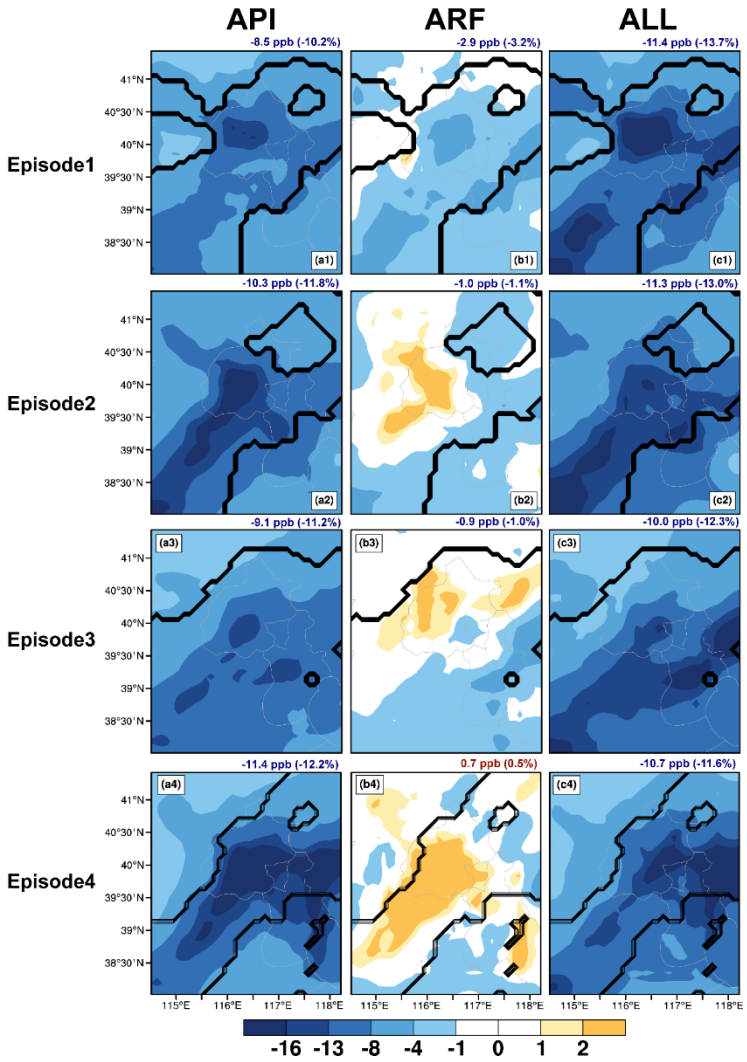

Figure 6. The changes in surface-layer O_3 due to (a) aerosol-photolysis interaction (API), (b) aerosol-radiation feedback (ARF), and (c) the combined effects (ALL, API+ARF) in the daytime (08:00-17:00 LST) during 28 July to 3 August 2014 (Episode1), 8-13 July 2015 (Episode2), 5-11 June 2016 (Episode3) and 28 June to 3 July 2017 (Episode4). The regions sandwiched between two black lines are defined as the complex air pollution areas (CAPAs). The changes (percentage changes) in O_3 concentrations caused by API, ARF, and ALL avaraged over CAPAs are shown at the top of each panel.

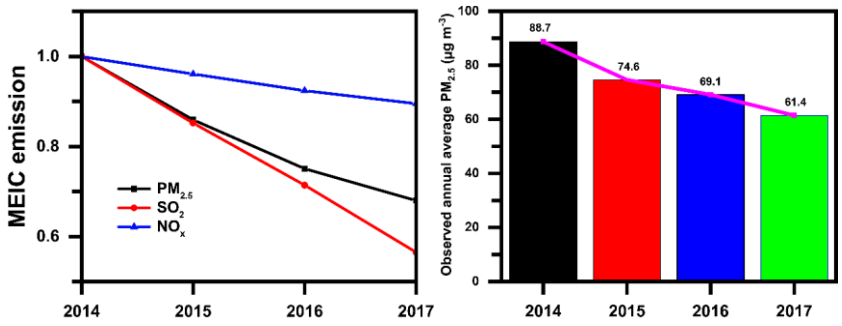

Figure S1. Trends of emissions over 2014–2017 from MEIC emission inventory and the observed annual mean $PM_{2.5}$ concentrations in the studied domain during 2014-2017.

Response to *"How do the conclusions based on the three episodes relate with other conditions during the years studied (2014-16) and to what extent are they relevant for other years before or after?"*

(1) ***How do the conclusions based on the three episodes relate with other conditions during the years studied?***

To address the editor's comments, three additional episodes with different conditions of air pollution, i.e., (1) $PM_{2.5}$ pollution alone (Episode_add1, the daily mean $PM_{2.5}$ concentration is larger than 75 μg m^{-3}), (2) neither $PM_{2.5}$ nor O_3 exceed air quality standard (Episode_add2, the daily mean $PM_{2.5}$ and maximum daily 8-h average O_3 concentration are smaller than 75 μg m^{-3} and 80 ppb, respectively), and (3) O_3 pollution alone (Episode_add3, the maximum daily 8-h average O_3 concentration is larger than 80 ppb) are simulated to examine the impacts of API and ARF on O_3. Detailed information about these three additional episodes is summarized in Table S3 (see below) of the revised manuscript.

Table S3. Three additional episodes with different levels of $PM_{2.5}$ and O_3.

Case	Time	$PM_{2.5}$ pollution (concentration)	O_3 pollution (concentration)
Episode_add1	2014.10.7-2014.10.11	√ (223.5 μg m^{-3})	× (46.9 ppb)
Episode_add2	2016.6.13-2016.6.17	× (36.5 μg m^{-3})	× (62.4 ppb)
Episode_add3	2017.6.15-2017.6.20	× (61.9 μg m^{-3})	√ (103.6 ppb)

In Episode_add1, Episode_add2 and Episode_add3, API alone is simulated to reduce surface O_3 averaged over each episode and over the entire simulated domain (38.01~41.45 °N, 114.52~118.28 °E) by 15.3 ppb (29.3%), 4.4 ppb (6.8%) and 4.5 ppb (5.3%), respectively, and ARF alone reduces surface O_3 by 3.9 ppb (6.2%), 0.6 ppb (1.0%), and 0.1 ppb (0.1%), respectively (see below, Fig. S13 of the revised manuscript). All the results confirm the same conclusion that the reduction in O_3 by API is larger than that by ARF.

We have also **added** the following discussion about these three additional episodes in the **Discussion** section:

"We presented above the results from our simulations of multi-pollutant air pollution episodes. In order to show that the conclusion of this work can be applied to other conditions of air pollution, three additional situations are carried out, i.e., (1) $PM_{2.5}$ pollution alone (Episode_add1, the daily mean $PM_{2.5}$ concentration is larger than 75 μg m^{-3}), (2) neither $PM_{2.5}$ nor O_3 exceed air quality standard (Episode_add2, the daily mean $PM_{2.5}$ and maximum daily 8-h average O_3 concentration are smaller than 75 μg m^{-3} and 80 ppb, respectively), and (3) O_3 pollution alone (Episode_add3, the maximum daily 8-h average O_3 concentration is larger than 80 ppb). Detailed information about these three additional episodes is listed in the supporting information (Text S1 and Table S3). Analyzing Episode_add1, Episode_add2 and Episode_add3 in Fig. S13, API alone is simulated to reduce surface O_3 averaged over each episode and over the entire domain by 15.3 ppb (29.3%), 4.4 ppb (6.8%) and 4.5 ppb (5.3%), respectively, and ARF alone reduces surface O_3 by 3.9 ppb (6.2%), 0.6 ppb (1.0%), and 0.1 ppb (0.1%), respectively. All the results confirm the same conclusion that the reduction in O_3 by API is larger than that by ARF."

(2) ***How do the conclusions based on the three episodes to what extent are they relevant for other***

***years before or after?*”**

Air pollution in China was characterized by high concentrations of $PM_{2.5}$ before 2014 (Li et al., 2019a; Zhang et al., 2019) and by synchronous occurrence of high $PM_{2.5}$ and O_3 or high levels of O_3 alone after 2017 (Dai et al., 2021; Li et al., 2019b; Li et al., 2020; Qin et al., 2021). Episode_add1, Episode1-Episode4, and Episode_add3 can represent these situations, respectively. Therefore, we believe that our general conclusion can also be applied to the years before 2014 and after 2017.

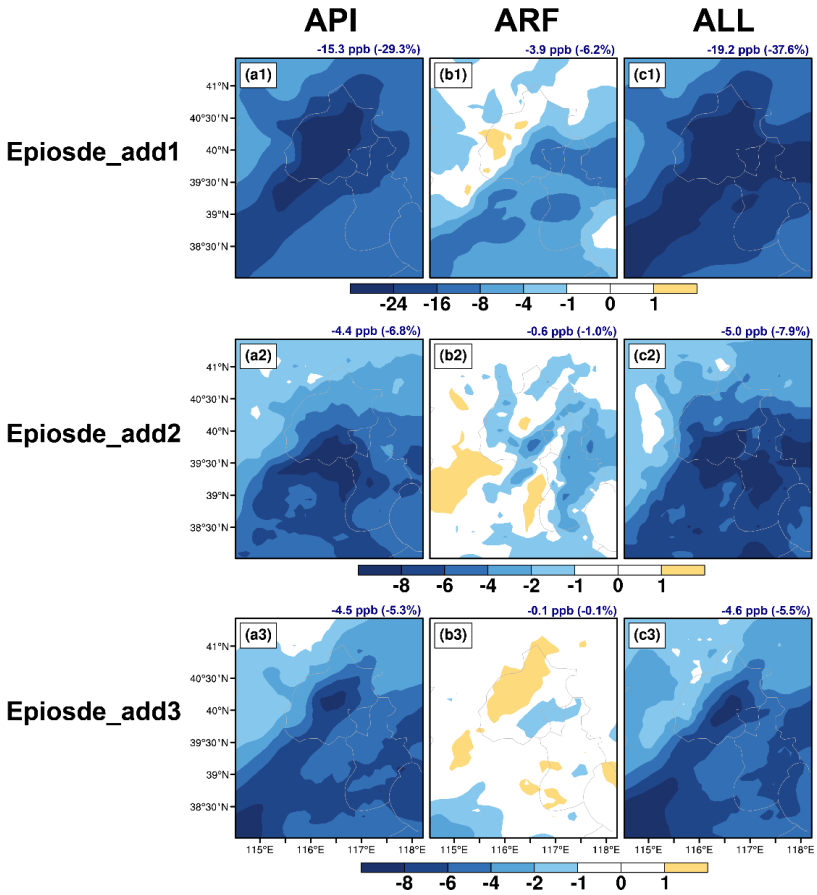

Figure S13. The changes in surface-layer O_3 due to (a) aerosol-photolysis interaction (API), (b) aerosol-radiation feedback (ARF), and (c) the combined effects (ALL, API+ARF) in the daytime (08:00-17:00 LST) of 7-11 October 2014 (Episode_add1), 13-17 June 2016 (Episode_add2) and 15-20 June 2017 (Episode_add3). The changes (percentage changes) in O_3 concentrations caused by API, ARF and ALL avaraged over the entire simulated domain are also shown at the top of each panel.

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.

Li, J. D., Liao, H., Hu, J. L., Li, N.: Severe particulate pollution days in China during 2013-2018 and the associated typical weather patterns in Beijing-Tianjin-Hebei and the Yangtze River Delta regions, Environmental Pollution,

248, 74-81, 2019a.

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, 2019b.

Li, K., Jacob, D. J., Shen, L., Lu, X., De Smedt, I., and Liao, H.: Increases in surface ozone pollution in China from 2013 to 2019: anthropogenic and meteorological influences, Atmos. Chem. Phys., 20, 11423–11433, https://doi.org/10.5194/acp-20-11423-2020, 2020.

Qin, Y., Li, J., Gong, K., Wu, Z., Chen, M., Qin, M., Huang, L., and Hu, J.: Double high pollution events in the Yangtze River Delta from 2015 to 2019: Characteristics, trends, and meteorological situations, Sci. Total Environ., 792, 148349, 2021.

Wang, W., Li, X., Shao, M., Hu, M., Zeng, L., Wu, Y., and Tan, T.: The impact of aerosols on photolysis frequencies and ozone production in Beijing during the 4-year period 2012–2015, Atmos. Chem. Phys., 19, 9413–9429, https://doi.org/10.5194/acp19-9413-2019, 2019.

Zhao, H.; Zheng, Y., and Li, C. Spatiotemporal distribution of $PM_{2.5}$ and O_3 and their interaction during the summer and winter seasons in Beijing, China. Sustainability, 10, 4519, 2018.

Zhang, Q., Zheng, Y., Tong, D., Shao, M., Wang, S., Zhang, Y., Xu, X., Wang, J., He, H., Liu, W., Ding, Y., Lei, Y., Li, J., Wang, Z., Zhang, X., Wang, Y., Cheng, J., Liu, Y., Shi, Q., Yan, L., Geng, G., Hong, C., Li, M., Liu, F., Zheng, B., Cao, J., Ding, A., Gao, J., Fu, Q., Huo, J., Liu, B., Liu, Z., Yang, F., He, K., and Hao, J.: Drivers of Improved $PM_{2.5}$ Air Quality in China from 2013 to 2017, P. Natl. Acad. Sci. USA, 116, 24463–24469, https://doi.org/10.1073/pnas.1907956116, 2019.

Zhu, J., Chen, L., Liao, H., and Dang, R.: Correlations between PM2.5 and Ozone over China and Associated Underlying Reasons, Atmosphere, 352, 1–15, https://doi.org/10.3390/atmos10070352, 2019.

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

The revised reversion of Yang et al. does address several of the manuscripts original limitations. The manuscript is still clear and well written and the scientific problem addressed is important to the scientific community. However, a few issues remain that leave questions as to whether the findings in the manuscript are robust and meaningful. My major and minor comments on the revised manuscript are as follows:

Response:

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

Major Comments:

1. *The authors have included two additional episodes to address the issue of scientific robustness. With these two additional episodes, it appears that the impact of aerosol radiation interactions, usually via API, are similar in all episodes despite variability in the magnitude and spatial extent of the CAPAs. With these finding we can reasonably conclude that these values are indeed representative of CAPAs in this region during the period of 2014-2016. However, this did not address any issues with changes in time/emissions (i.e, 2001-2005 or more currently 2018- 2020). If the authors are not going to do any additional episodes, they need to convincingly justify why the period of 2014-2016 is representative of /or important for current/future conditions.*

Response:

It's difficult to carry out simulations of complex air pollution events during 2001-2005, because the national observations of $PM_{2.5}$ and O_3 concentrations were not available until 2013. Thus, the time and the area of complex air pollution events cannot be determined for earlier years. To address your concerns, we now have four episodes (Episode1: 28 July to 3 August 2014; Episode2: 8-13 July 2015; Episode3: 5-11 June 2016; Episode4, 28 June to 3 July 2017) in the revised manuscript. These selected events cover different years of 2014 to 2017 during which the governmental Air Pollution Prevention and Control Action Plan was implemented (the changes in emissions and observed $PM_{2.5}$ in the studied region during 2014-2017 are shown in Fig. R1).

To further address yours and editor's comments, three additional situations under different conditions of air pollution, i.e., (1) $PM_{2.5}$ pollution alone (Episode_add1, the daily mean $PM_{2.5}$ concentration is larger than 75 μg m^{-3}), (2) neither $PM_{2.5}$ nor O_3 exceed air quality standard (Episode_add2, the daily mean $PM_{2.5}$ and maximum daily 8-h average O_3 concentration are smaller than 75 μg m^{-3} and 80 ppb, respectively), and (3) O_3 pollution alone (Episode_add3, the maximum daily 8-h average O_3 concentration is larger than 80 ppb) are simulated to examine the impacts of

API and ARF on O_3. All the results confirm the same conclusion that the reduction in O_3 by API is larger than that by ARF. (Please see our responses to Editor's comments).

Air pollution in China was characterized by high concentrations of $PM_{2.5}$ before 2014 (Li et al., 2019a; Zhang et al., 2019) and by synchronous occurrence of high $PM_{2.5}$ and O_3 or high levels of O_3 alone after 2017 (Dai et al., 2021; Li et al., 2019b; Li et al., 2020; Qin et al., 2021). Episode_add1, Episode1-Episode4, and Episode_add3 can represent these situations, respectively. Therefore, we believe that our general conclusion can also be applied to the years before 2014 and after 2017.

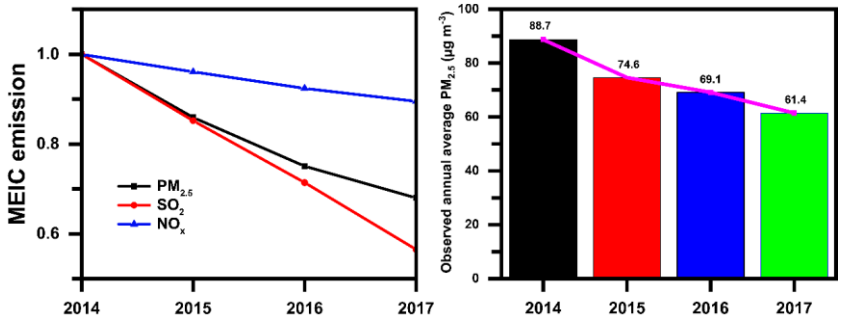

Figure R1. Trends of emissions over 2014–2017 from MEIC emission inventory and the observed annual mean $PM_{2.5}$ concentrations in the studied domain during 2014-2017.

2. *The authors have added caveats to the conclusion to address the issue of lacking SOA formation pathways in their simulations. These listed caveats are important, but the authors have overlooked the possibility that increased O_3 from $PM_{2.5}$ reductions will generate more SOA via increased oxidation. This feedback could partially compensate the increased O_3 formation the authors predict will happen.*

Response:

Thanks for pointing this out. We agree with your helpful suggestion that the lacking SOA formation pathways in these simulations may underestimate the impacts of aerosol-photolysis interaction (API) and aerosol-radiation feedback (ARF) on surface ozone.

The discussions about the impacts of SOA have been **revised** as follows: "In the current CBMZ and MOSAIC schemes, the formation of SOA (secondary organic aerosol) is not included (Gao et al., 2015; Chen et al., 2019). The absence of SOA can underestimate the impacts of API and ARF on O_3. Meanwhile, the lack of SOA may lead to weaker heterogeneous reactions to result in higher O_3 concentrations (Li et al., 2019c). The net effect of the two processes will be discussed and quantified in our future study."

Minor Comments:

1. The response to the previous Reviewer2 Minor comments 7-10 should be included in the manuscript if not already done to facilitate ease of understanding.

Response:

The previous Reviewer 2's Minor Comments 7-10 have been **added** into Section 4.4 of the revised manuscript as follows:

(1) Since VMIX increases the surface O_3 concentrations by transporting O_3 from aloft (where O_3 concentrations are high) to the surface layer (Tang et al., 2017; Xing et al., 2017; Gao et al.,

2018).

(2) The positive change in VMIX due to API may be associated with the different vertical gradient of O_3 between BASE and NOAPI cases (Gao et al., 2020), as shown in Fig. 8a.

(3) The positive VMIX_DIF is related to the evolution in boundary layer during the daytime. The 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 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 both species (Sillman, 1999). As shown in Fig. S7, (a-f), O_3 is mainly formed under the VOC-limited and the transition regimes in CAPAs. As shown in Figs. S7(g-i) and S7(j-l), 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.

(4) The positive variation in VMIX due to API may be associated with the different vertical gradient of O_3 between BASE and NOAPI again.

2. The color bars for Figures 5 and 6 needs to be the same for all episodes to facilitate easy comparison.

Response:

According to the reviewer's suggestion, we now use the same color bars in Figures 5 and 6 in the revised manuscript.

3. In Figures 2 and 3 the y-axis need to be consistent for all episodes to facilitate ease of comparison

Response:

We now use the same y-axis in Figures 2 and 3 in the revised manuscript.

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.

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.

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.

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, 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., 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, J. D., Liao, H., Hu, J. L., Li, N.: Severe particulate pollution days in China during 2013-2018 and the associated typical weather patterns in Beijing-Tianjin-Hebei and the Yangtze River Delta regions, Environmental Pollution, 248, 74-81, 2019a.

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, 2019b.

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, 2019c.

Li, K., Jacob, D. J., Shen, L., Lu, X., De Smedt, I., and Liao, H.: Increases in surface ozone pollution in China from 2013 to 2019: anthropogenic and meteorological influences, Atmos. Chem. Phys., 20, 11423–11433, https://doi.org/10.5194/acp-20-11423-2020, 2020.

Qin, Y., Li, J., Gong, K., Wu, Z., Chen, M., Qin, M., Huang, L., and Hu, J.: Double high pollution events in the Yangtze River Delta from 2015 to 2019: Characteristics, trends, and meteorological situations, Sci. Total Environ., 792, 148349, 2021.

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.

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, W., Li, X., Shao, M., Hu, M., Zeng, L., Wu, Y., and Tan, T.: The impact of aerosols on photolysis frequencies and ozone production in Beijing during the 4-year period 2012–2015, Atmos. Chem. Phys., 19, 9413–9429, https://doi.org/10.5194/acp19-9413-2019, 2019.

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.