

Response to Comments of Reviewer #1

(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

Yang et al. examined the impacts of aerosols on surface ozone through the two well-known pathways, i.e., aerosol-photolysis interaction and aerosol-radiation feedback. The novelty of this study is its focus on the polluted episodes with elevated both PM_{2.5} and ozone levels over North China. They also quantified the chemical and physical processes that drive the aerosol-radiation interactions.

Overall, this is a timely study and it clearly demonstrates the impacts of aerosols on ozone pollution. The structure of this manuscript is easy to follow. Although some of the manuscript needs further clarification, the results are generally convincing. As such, I think it is publishable after the following issues are addressed.

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.

Specific Comments:

1. *In Abstract: ozone changes refer to MDA8 ozone or daytime ozone?*

Response:

The ozone changes in abstract mean daytime ozone. According to the reviewer's comments, we have added this information in the revised manuscript. (Page 2, Line 34)

2. *Line 177: a correlation coefficient of 0.66 reads like not high!*

Response:

According to the comments of Reviewer#2, another two complex air pollution episodes (8-13 July 2015 (Episode2) and 5-11 June 2016 (Episode3)) in this region are selected to conduct simulations for generating general conclusions.

Thanks for your suggestion, we have changed this sentence in the revised manuscript as follows: "As shown in Fig. 2, the temporal variations of observed PM_{2.5} can be well performed by the model with correlation coefficients (R) of 0.66, 0.56 and 0.73 and normalized mean bias (NMB) of -19.2%, -3.9% and 30.4% during Episode1, Episode2 and Episode3, respectively." (Page 7-8, Line 183-186)

3. *Lines 179-181: the oxidation of SO₂ by NO₂ in aqueous aerosols is important for summertime?*

Response:

This sentence has been deleted in the revised manuscript.

4. *Lines 248-251: this statement looks reasonable here, but in the later text the process analysis shows that chemistry will be enhanced by ARF. Instead, ARF decreases ozone through physical processes.*

Response:

Thanks for the reviewer's suggestion. We have deleted this sentence in the revised manuscript.

5. *Line 260: "is" should be "are". Please do proof-reading throughout the text.*

Response:

This sentence has been deleted in the revised manuscript. According to the reviewer's comments, proof-reading has been conducted through the whole revised manuscript.

6. *Line 310: It is Okay to use model levels (e.g., 12 levels), but it will be better to add model height in meters as well.*

Response:

Thanks for reviewer's suggestion. We have added the model height in meters in the revised manuscript. (**Page 12-13, Line 320, Line 323-325, Line 334-335**)

7. *Lines 326-327: why do you need this statement?*

Response:

Analyzing Fig. 8c we can conclude that ARF promotes the O₃ chemical production with a positive mean value of 0.72 ppb h⁻¹. The enhanced O₃ precursors due to ARF can promote the chemical production of O₃. According to the reviewer's comment, we have deleted this statement in the revised manuscript.

8. *Lines 327-328: Please provide evidence to support this conclusion.*

Response:

The typical VOCs/NO_x ratio is calculated to classify sensitivity regimes and to indicate the possible O₃ responses to changes in VOCs and/or NO_x concentrations. O₃ 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. R1(a-f), O₃ are mainly formed under VOC-limited and 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. R1(g-i) and (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. Similar results can also be found in Gao et al. (2018).

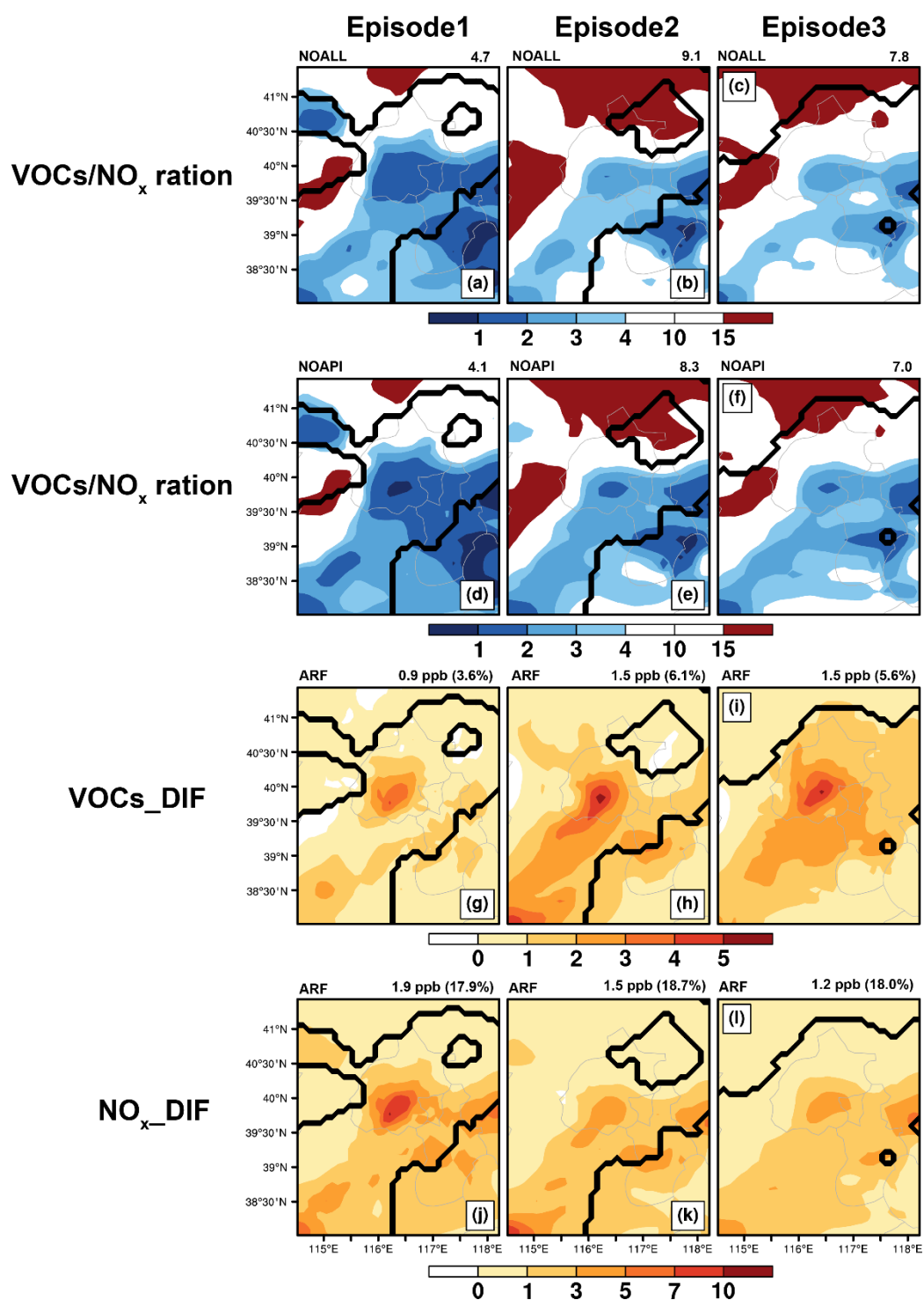


Figure R1. The ratios of VOCs/NO_x calculated from (a-c) NOALL, and (d-f) NOAPI. The changed surface-layer concentrations of (g-i) VOCs and (j-l) NO_x (NO₂+NO, ppb) caused by ARF during the daytime (08:00-17:00 LST) from Episode1 to Episode3. The calculated values averaged over CAPAs are also shown at the top of each panel.

9. *Discussion.* I think the authors should do some comparisons between your results with previous studies. This is important for readers to better understand your case study results. Moreover, how about the applicability of the calculated ROP of -0.14

ppb ($\mu\text{g m}^{-3}$)⁻¹?

Response:

According to the comments of Reviewer#2, we conduct another two complex air pollution episodes (8-13 July 2015 and 5-11 June 2016) in this region to draw the general conclusions. The three episodes feature a similar variation pattern, and the detailed information can be found in section 4. (**Page 9-13, Line 223-352**). Meanwhile, a discussion about the impacts of secondary organic aerosols (SOA) is also added in the section 5 (**Page 14-15, Line 391-401**).

Thanks to the reviewer's comments. As the relationship between O₃ and PM_{2.5} is non-linear, and the simple index of ROP can not fully represent the impacts of aerosols on surface O₃, so we delete the ROP in the revised manuscript.

10. Fig.2: It will be better to add error bars for observed PM_{2.5} and ozone.

Response:

According to the reviewer's suggestion, error bars have been added in Fig. 2 in the revised manuscript. (**Page 27**)

11. Fig.3: what are the cities these plots for?

Response:

The averaged T₂, RH₂, and WS₁₀ are collected from ten meteorological observation stations, and the detail information about the sites is listed in Table S1. The photolysis rates of NO₂ (J[NO₂]) are observed in Peking University. More details are explained in section 2.3. (**Page 6**)

12. Fig.7: what are the layers your process analysis applied for? I don't see this key information here, as well as in the text.

Response:

The surface-layer, namely, first-layer O₃ concentrations are analyzed in Fig. 7. Thanks for reviewer's suggestion, we have added this information in the revised manuscript. (**Page 10, Line 272**)

Reference:

- Edson, C. T., Ivan, H.-P. and Alberto, M.: Use of combined observational- and model-derived photochemical indicators to assess the O₃-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.
- 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.
- 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](https://doi.org/10.1016/S1352-2310(98)00345-8), 1999.

Thank you very much for your comments and suggestions.