## General comments:

Firstly, the English used in the manuscript is, at times, quite poor and it is recommended that the authors make use of an English-language proofreading service to tidy up the manuscript. However, having said that, the scientific content of the manuscript is generally fine and, after following the specific recommendations below, I would fully support its publication in ACP. One other general comment relates to the consistency of units: please standardise reaction rates with the widely-used ppbv h-1 (sometimes ppbv s-1 is used instead).

Response: Thanks for the comments, which help to improve our manuscript. Please see the pointto-point response below. (Comments in Black, Response in Blue, Changes in the manuscript in Red)

We carefully read this manuscript and have improved its English. Besides, comments from the three reviewers (particularly Reviewer 1) also helped a lot to improve the English of this manuscript. Regarding the units, we unified them as suggested. Modified figures include: Figures 10, 11, 12, and 13.

## Specific comments:

Figure 2 – it would be good to see diurnal profiles of the same key species, especially considering almost all of the other results are presented as diurnal profiles rather than full time series. Response: We added diurnal variations of HONO, O<sub>3</sub>, NO<sub>2</sub>, JNO<sub>2</sub>, PM<sub>2.5</sub>, CO, NO, and SO<sub>2</sub> in the supporting information (Figure S3), which are shown below:



Figure S3: Diurnal profiles of HONO and related species measured during this campaign. Note that some of these data were also shown in the companion ACP paper for comparison between measurements at the foot and the summit stations.

Figure 10 - in A and B, the gradient-style colouring makes it difficult to distinguish the categories, please use more contrasting colours as in C and D. Also, for C and D, the caption states that unmeasured species are summarised in the "other" category. Does this refer to intermediates (e.g., OVOCs) generated in the model? i.e., model-generated species that were not measured but do contribute to the model reactivity? Please clarify.

Response: To clarify this issue, Figure 10 and its caption were improved as follows:



Figure 1: Production rates (P), loss rates (L) and reactivities of radicals. (A): L and P of OH; (B): L and P of NO<sub>3</sub>; (C): Reactivities of OH and (D): Reactivities of NO<sub>3</sub>. In (A) and (B), the top-3 sources or sinks are shown, and all the others are summarized in "Other sources" or "Other sinks". In (C), OH reactivities with different families of the measured species are shown and reactivities with all the unmeasured species are summarized in "Others". In (D), NO<sub>3</sub> reactivities from top-3 reactions are shown and all the others are summarized in "Others".

Figure 13 – in the caption, what is meant by the "integration problem"? I cannot find it mentioned in the main text. In general, the caption could do with rewording as it is not the most clear. Also, the caption states that equilibrium reactions were not considered, yet (net) PAN formation is shown in C?

Response: We didn't figure out how to integrate all the reactions that consume radicals. Therefore we summarized the top-20 radical termination reactions. The sum of the top-20 could represent the majority of total  $T(RO_x)$  as others (<0.03 ppbv h<sup>-1</sup>) were at least 2 orders of magnitude lower. Net PAN formation is a net radical loss path and was shown in Figure 13C. It could be derived from PAN production rate through reaction (1) subtracted by its loss rates through reaction (2). CH3COO2 + NO2 = PAN; (1)

 $PAN = CH3COO2 + NO2; \quad (2)$ 

The updated figure and caption are as follows:



Figure 13: Primary RO<sub>x</sub> production and net RO<sub>x</sub> loss. (A): Production from different sources and (B): their relative contributions at different hours of the day. (C): the top-20 RO<sub>x</sub> loss rates. Note that the top-20 net radical loss paths were summarized here. It could represent the majority of total T(RO<sub>x</sub>) as others (<0.03 ppbv h<sup>-1</sup>) were at least 2 orders of magnitude lower than the sum of top-20. Night-time P(RO<sub>x</sub>)<sub>HONO\_net</sub> was negative (a net sink for OH) so that its contribution was also negative at night. The same amounts of radical loss or production from equilibrium reactions (e.g., HO<sub>2</sub> + NO<sub>2</sub>  $\leftrightarrow$  HNO4; CH<sub>3</sub>COO<sub>2</sub> + NO<sub>2</sub>  $\leftrightarrow$  PAN) was excluded from radical initiation or termination. T(RO<sub>x</sub>)<sub>NO2+CH<sub>3</sub>COO<sub>2</sub> represents the net PAN formation.</sub>

Line 65 – please cite Slater, ACP, 2020 (https://doi.org/10.5194/acp-20-14847-2020) and Whalley, ACP, 2021 (https://doi.org/10.5194/acp-21-2125-2021) as examples of the importance of HONO photolysis in OH formation.

Response: Improved as suggested.

Section 2.2.2 could do with more explanation/clarity, e.g. line 160, "significantly enhanced" by how much?

Response: In Sce-3 we enlarged  $\gamma_a$  from  $2 \times 10^{-5}$  to  $1.2 \times 10^{-3}$  or EF from 7 to 400 (see Section 3.2.3) to test whether the aerosol-derived sources could well explain the observations.

Line 170 - was any back-trajectory analysis performed?

Response: We added back trajectory results in Figure S2. And the related text in Section 3.1.1: "Air masses observed at this site originated from multiple directions, including west, south, and east, which are shown in the wind rose plot in Figure S2A. Wind speed was generally low, with an average of about 2 m s<sup>-1</sup>. In addition, the wind rose results generally agree well with HYSPLIT trajectory results in Figure S2B and S2C."



Figure S2: (A): Wind rose plot for the wind measurements at the foot of Mt. Tai; (B) and (C): 1-day back trajectory from HYSPLIT (<u>https://www.ready.noaa.gov/HYSPLIT.php</u>).

Line 194 and Table 3 – please cite Crilley, AMT, 2019 (https://doi.org/10.5194/amt-12-6449-2019) as a more recent example of HONO levels in Beijing. Response: Improved as suggested.

Section 3.1.2/line 208 – misleading – the chemiluminescence method is fine (for the measurement of NO), it is the NO2 (to NO) converter chemistry that gives rise to interferences from other NOy

species.

Response: The reviewer is right. We improved the text as follows: "As the most important HONO precursor, accurate measurement of NO<sub>2</sub> plays a key role in analyzing HONO formation. The NO<sub>x</sub> monitor used in this study could specifically detect NO. To measure NO<sub>2</sub>, a Molybdenum converter is used to convert NO<sub>2</sub> to NO. However, this chemical conversion process suffers from the interference of other NO<sub>y</sub> species (Villena et al., 2012)"

Lines 211, 214, and 215 – what is organic nitrates\*? This is defined as RONO2 and ROONO2 in the Figure 3 caption, but please include the same definition in the main text. Response: In the main text, we added a sentence "In this study, we defined the sum of RONO<sub>2</sub> and ROONO<sub>2</sub> as organic nitrates<sup>\*</sup>" to make it consistent with Figure 3 caption.

Line 224 – the time is given as 11:00, should it really be 13:00? Response: Yes, it should be 13:00. We corrected it.

Line 326 – the EF value of 15.6 appears to have come from your companion paper, therefore please reference this.

Response: Improved as suggested.

Line 433 – please give the average percentage contribution of HO2+NO – same for HONO photolysis on line 434.

Response: Average contributions of  $HO_2$ +NO (70%) and HONO photolysis (11%) were added in this sentence.

Line 446 – delete "and OH" – OH reactivity is not affected by [OH]. Response: Improved as suggested.

Line 501 – give reference for the summertime O3 increase in the NCP. Response: Related references (Han et al., 2020; Li et al., 2019; Sun et al., 2016, 2019) are added.

Line 508 - ozonolysis chemistry also produces RO2

Response: Yes, RO<sub>2</sub> could also be produced during alkenes ozonolysis. This has already been considered in radical initiation (Figure 13). We improved the text: "Measurements on other radical precursors, such as H<sub>2</sub>O<sub>2</sub> (through photolysis to produce OH), HCHO (through photolysis to produce HO<sub>2</sub>), and alkenes (through ozonolysis via Criegee intermediate to produce OH, HO<sub>2</sub>, and RO<sub>2</sub>)."

## References

Han, S., Yao, Q., Tie, X., Zhang, Y., Zhang, M., Li, P. and Cai, Z.: Analysis of surface and vertical measurements of O<sub>3</sub> and its chemical production in the NCP region, China, Atmos. Environ., 241, 117759, doi:10.1016/j.atmosenv.2020.117759, 2020.

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, Proc. Natl. Acad. Sci. U. S. A., 116(2), 422–427, doi:10.1073/pnas.1812168116, 2019.

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