Reviewer#1

General comments:

The article entitled “Amplified role of potential HONO sources in O3 formation in North China Plain during autumn haze aggravating processes” by Zhang et al., is in line with several studies performed by the research group on this topic. North China Plain climate and atmospheric composition received higher attention in the last decade. The authors analyzed the impact of different HONO sources on the ozone concentrations by using the WRF-Chem model and performed investigations over various scenarios to study the effect of nitrate photolysis as the main HONO source.

The article is very well written and concise.

One main aspect regarding the complete evaluation of the present work has been related to missing supplementary material and the difficulties to understand the results and discussions despite the missing figures and text.

The comments of this review have been adapted and the eventual similar comments with the other reviewers have been removed to avoid double suggestions. However, some additional comments and suggestions remain to be solved.

We sincerely thank you for the time and effort you spent in reviewing the manuscript, your kindly comments and suggestions are very precious and helpful to us. Point by point response to your comments has been made and attached beneath. The comments by the reviewer are in the plain format with black color*, and the responses are in the italic format with blue color, we hope that these improvements could make our results more reasonable.*

We are very sorry to have found that the supplementary materials are not available on the ACPD website. In our initial submission, we uploaded the manuscript and the supplementary material, while the format of Table 4 in the manuscript was in landscape format rather than in the portrait format, the landscape format might be unable to operate, and the editor reminded us to provide a new manuscript file with proper format. In the following operation of updating the manuscript with the corrected format handled by the editor, we misunderstood that the supplementary material is NOT needed, so we deleted the supplementary material. The revised supplementary material has been uploaded.

Specific comments:

1. An important finding of the present work is the effect of the photolytic and “volumic” HONO sources on the ozone high concentrations. These findings are

related to haze episodes in NCP. The NCP area is highly correlated with biomass burning events where a high amount of aromatics are released into the atmosphere. The aromatics can strongly influence tropospheric chemistry on a regional scale. Could aromatics and especially nitroaromatics, both present in gas-phase and particle-phase, through their direct photolysis explain partly the amount of HONO? Could nitroaromatics play partially the role of nitrate in this study? Including photolysis of nitroaromatics for the conditions discussed in the model would influence the HONO formation from the photolysis of nitrate. A chosen larger J(nitrate) would unbalance HONO sources from surface vs HONO sources from a higher altitude ("volumic"). Including similar sources with photolysis of nitrate could limit the J(nitrate)/J(HNO3) values.

We sincerely thank you for your support to our study. As you kindly pointed out, including similar sources with photolysis of nitrate may constrain the $J_{nitrate}/J_{HNO3}$ *value to a more reasonable range. The photolysis of nitroaromatics was supposed as a potential HONO source in the atmosphere, and their representative chemical species are nitrophenols.*

The concept of HONO formation via nitrophenol photolysis was proposed by Bejan et al. (2006) firstly, and they reported a $J_{(nitrophenol\ -\ HONO)}$ *value of (1.1–4.4)* $\times 10^{-5}\ s^{-1}$ *for four typical nitrophenols with* J_{NO2} *reaching* $10^{-2}\ s^{-1}$ *(Table 1 in Bejan et al. (2006)). They collected a ortho-nitrophenol concentration of ~60 ppt in an urban area from Harrison et al. (2005), and calculated a HONO formation rate of ~6 ppt* h^{-1} *at noontime with* J_{NO2} *reaching* $10^{-2}\ s^{-1}$ *(the photolysis of 1 ppb of ortho-nitrophenol corresponding to ~0.1 ppb* h^{-1} *HONO production around noontime) (Bejan et al., 2006). While in rural areas the concentrations of nitrophenols were very low and usually <10 ppt, the maximum HONO production rate via the photolysis of nitrophenols was < 1 ppt* h^{-1}*. Thus, the HONO production rates are quite small via the photolysis of nitrophenols, compared with other stronger potential HONO sources in urban areas (Other HONO sources usually reached several ppb* h^{-1} *in urban areas during daytime).*

Limited to the current understanding on the mechanisms of nitro-aromatic compounds and the lack of nitro-aromatic compounds in the anthropogenic inventory, the nitro-aromatic compounds are temporarily not available in the WRF-Chem model. To evaluate their potential impact on the HONO formation, we estimated by using the reported observations from the literature. Here are some recent observations of nitro-aromatic compounds in NCP. Li et al. (2020b) reported a total concentration of ~80 ng m^{-3} *for eight nitro-aromatic compounds in urban Beijing from autumn to winter of 2017–2018, being the highest concentration among the four seasons, and corresponding to ~12–15 ppt. Li et al. (2020a) reported a total concentration of 19–585 ng* m^{-3} *(mean is 173 ng* m^{-3}*) of twelve nitro-aromatic compounds in January of 2016 in urban Beijing, the maximum concentration was corresponding to ~100 ppt. Wang et al. (2018) observed the fine particulate nitrated phenols at four sites in northern China (including urban, rural and mountain areas) during 2013–2014, the* *maximum value was found in urban Jinan during winter, with a mean value of <50 ng* m^{-3}*, i.e., <10 ppt. Yang et al. (2020) reported the concentration of nitro-aromatic compounds was ~80 ng* m^{-3} *in urban Beijing and 8 ng* m^{-3} *in rural Xinglong. Recently Song et al. (2021) reported a maximum concentrations of ~ 1.2 ppb in hazy days for the gas-phase nitrated phenols in urban Beijing during winter of 2018, and the averaged concentration in clean days was ~170 ppt.*

From the above, the nitro-aromatic compounds were usually 1–100 ppt in NCP, and rarely exceeded 1ppb even in the heavy haze events in winter. The calculated maximum HONO production rate around noontime was 0.1–10 ppt h^{-1} *in most cases, quite small compared with the calculated unknown HONO sources (several ppb* h^{-1}*). Some modeling studies have evaluated the impact of this source and found that this source is minor. Lee et al. (2016) adopted an upper limit constant concentration of 1 ppb and the photolysis rate of* $\sim 3 \times 10^{-5}$ s^{-1} *in their MCM (Master Chemical Mechanism) box model but found that the contribution of this source can be neglected. Recently Chen et al. (2021) reported an average peak HONO production rate of 16.9 ppt* h^{-1} *from the photolysis of nitro-phenolic compounds in south China with the MCM model, so the HONO production rate was also small.*

Although $J_{(nitrophenol\ -\ HONO)}$ *and* $J_{nitrate}$ *values may be similar, e.g.* 10^{-5} s^{-1}*, the concentration of nitro-aromatic compounds (in the unit of "ng* m^{-3}*") in the atmosphere is quite low compared with that of nitrate (in the unit of "μg* m^{-3}*"), dwarfing their atmospheric contribution to HONO.*

2. HONO is formed from NO and OH but HONO photolysis would produce back those products. HONO formation by this source is a net production?

HONO can be formed via the NO+OH reaction, which consumes OH and mainly occurs in daytime when OH concentrations are high, and the formed HONO can in turn be photolyzed to produce NO and OH during daytime, the net OH production from HONO is its photolysis to OH minus its homogeneous formation of HONO. Near the ground, when only the NO+OH reaction was considered (Base case), the consumed OH for HONO formation was 0.11 ppb h^{-1} *at the 95 NCP sites during daytime, while the corresponding production rate of OH via HONO photolysis was 0.09 ppb* h^{-1} *during daytime, HONO had a negative contribution (-0.02 ppb* h^{-1}*) and became a small sink of OH when the NO+OH reaction was considered as the only HONO source (**section 3.3.2** in the revised version), accordingly, the daytime-averaged net HONO formation rate was 0.02 ppb* h^{-1} *at the 95 NCP sites.*

Based on the above results, the OH+NO reaction slightly increased HONO concentrations but consumed more OH as a cost, so reduced the concentrations of atmospheric oxidants essentially.

3. Table 3 has not been easy to understand since missing the definition of the metrics...

The definitions were given in the supplementary materials previously, and the supplementary materials have been uploaded.

4. In figure 2 at the end of the haze period, it seems to accumulate significant precursors of HONO, other than those included in the 6S scenario. How could be explained the missing correlations of HONO between the observed and 6S on 15.10.2018 and 22-23.10.2018?

The simulated HONO on Oct.15 and Oct.22 was indeed underestimated compared with the observations. The underestimation was mainly caused by the earlier scavenging of pollutants at the urban Beijing site (BUCT) in the model. From the comparison of $PM_{2.5}$ *in* ***Fig.2a****, we could clearly find that the observed* $PM_{2.5}$ *reached its peak on Oct.15 and Oct.22 in the first two haze episodes, especially on Oct.22; but the simulated* $PM_{2.5}$ *reached its peak on Oct.14 and Oct.21 in the first two haze episodes and decreased fast on Oct.15 and Oct.22. For a specific site like the BUCT, limited to the spatial/temporal resolution of meteorological and chemical inputs, biases may occur in the beginning or ending of a haze event, nevertheless, the model usually performs better on a regional scale because of less local effects. For example, the increasing and scavenging patterns of* $PM_{2.5}$ *were both well captured at the 95-NCP-sites in* ***Fig.4d****. Discussions on the underestimation of simulated HONO has been added in* ***Section 3.1.2*** *in the revised manuscript.*

5. In this figure, the nitrate by 6S is overestimated constantly? Why? It is correlated with the NO2 overestimation?

The detailed nitrate enhancement in case 6S at the BUCT site is given in Fig S3 and section 3.1.2 in the revised version. Among the six potential HONO sources, $Phot_{nitrate}$ *slightly consumed nitrate and lowered its concentrations, but* NO_2 *heterogeneous reactions especially* Het_{ground}*, which was overestimated due to the overestimation of* NO_2*, significantly promoted nitrate formation and increased nitrate concentrations. The photolysis of HONO elevated OH concentrations, favoring the formation of gaseous nitric acid (*HNO_3*) and nitrate, the main reactions are:*

$$OH + NO_2 \rightarrow HNO_3$$

$$HNO_3 + NH_3 \rightarrow NH_4NO_3$$

The 6S case contained the six potential HONO sources and this is why the nitrate concentration in the 6S case is higher than that in the base case. A number of studies have also proven the enhanced role of nitrate due to potential HONO sources, for example, the nitrate was significantly increased in Fig.8 of Fu et al. (2019) after considering six potential HONO sources in southern China (Fig.reply1), with a $J_{nitrate}/J_{HNO3}$ *ratio of ~120 for* $Phot_{nitrate}$. *An overestimation of ~20–40 µg* m^{-3} *for nitrate could be found on January 4 and January 6 of 2017.*

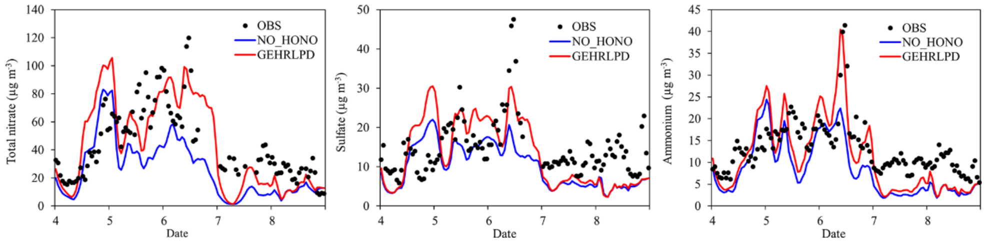

Figure 8. Temporal variation of observed and simulated total nitrate, sulfate, and ammonium concentrations in the NO_HONO and GEHRLPD cases at the Heshan site during 4–8 January 2017.

Fig.reply1 *(From Fig.8 in Fu et al. (2019)), the red line contained additional HONO sources including nitrate photolysis with a* $J_{nitrate}/J_{HNO3}$ *ratio of ~120, the blue line was the base case.*

Recently Zhang et al. (2021) compared nitrate concentrations between the base case and the case after adding six potential HONO sources including nitrate photolysis ($J_{nitrate}/J_{HNO3}=30$) *in urban Beijing, the enhanced nitrate can also be found after adding the six potential HONO sources. The overestimation of nitrate (reached 10–70 µg* m^{-3}*) could also be found in more than half of their study period. When using the hourly concentration rather than the daily mean concentration for nitrate, the overestimation would be probably even larger.*

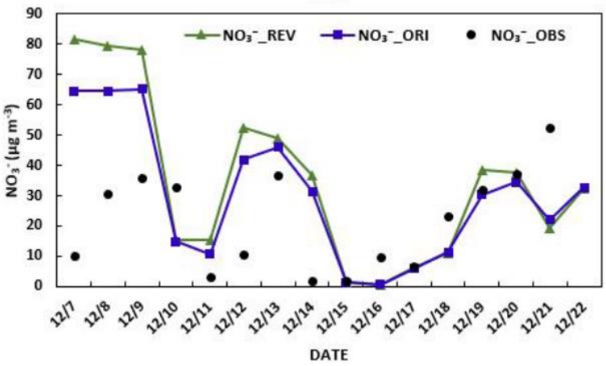

Fig.reply2 *Nitrate comparison before and after adding six potential HONO sources (From Fig.4 in Zhang et al. (2021)), the green line contained six additional HONO sources including nitrate photolysis, the blue line was the base case.*

For our nitrate results in Fig.2, the observed nitrate pattern was well reproduced. The model performed well in clean days, but overestimated by 10–50 µg m^{-3} *in some* *period of the first two haze aggravating processes. From the above, using the CMAQ model by Fu et al. (2019) and Zhang et al. (2021) or using the WRF-Chem model by us, the nitrate concentrations were usually overestimated. The overestimation could be partially caused by the uncertainties in the anthropogenic emission inventory, e.g., the overestimation of NO_x emissions, as you kindly pointed out. The inadequate understanding of the nitrate formation mechanism could also be related to nitrate simulation bias.*

6. In figure 2 the NO_2 concentrations are constantly overestimated. Could you explain why the observations are constantly lower at maxima? It may be the reason for interferences in the NO_2 measurements?

The NO_2 observations were obtained by using commercial instrument (Thermo 42i), and we deemed these observations as the truth value in the atmosphere. Generally speaking, the interferences in the NO_2 measurement like HONO or NO_y tend to slightly elevate the NO_2 observations rather than lower their values, the current bias was probably caused by the model overestimation. For the simulations, our results followed the pattern of the observations at both BUTC and 95-NCP sites but were overestimated especially during nighttime. The overestimation of NO_2 could be partially attributed to the uncertainty in the anthropogenic emission inventory due to the dramatic changes in anthropogenic emissions in China over the past few years (Zheng et al., 2018).

*In the study of Zhang et al. (2021), they used the CMAQ with the ABaCAS national emissions inventory (http://www.abacas-dss.com, last access: 14 October 2021), a large bias of NO_2 simulations compared with the observations could also be found (**Fig.reply3**).*

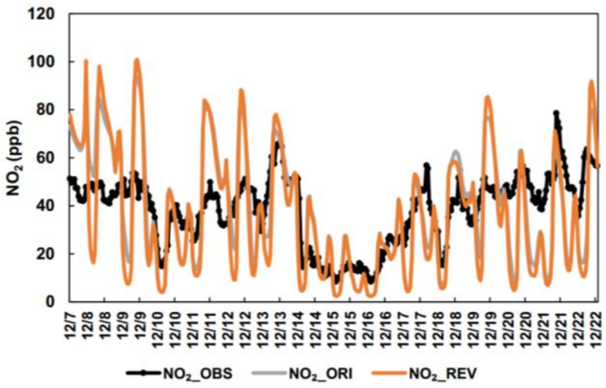

***Fig.reply3** Comparison of simulated and observed NO2 at urban Beijing (Fig.S6 in Zhang et al. (2021))*

A recent modelling study conducted by us (Lu et al., 2021) compared NO_2 simulations and observations from November of 2019 to March of 2020, covering the COVID-19-induced lockdown period in eastern China with 174 cities. The *anthropogenic inventory and the model setup were consistent with the Base case in this study. The NO_2 concentration was well simulated before the COVID-19 lockdown (November to December). In January and February an apparent bias occurred between the simulations and observations, because of the human activity came to a standstill, while the model still performed the "regular" results and approximately reflected the observations without the COVID-19 impact. When the lockdown was gradually got over the gaps were narrowed and almost appeared in late March. The overall NO_2 performance in our model is acceptable, and we speculate that on a larger spatial and temporal scale or using an updated NO_x emission inventory, the model performance on NO_2 will possibly be better and more reasonable.*

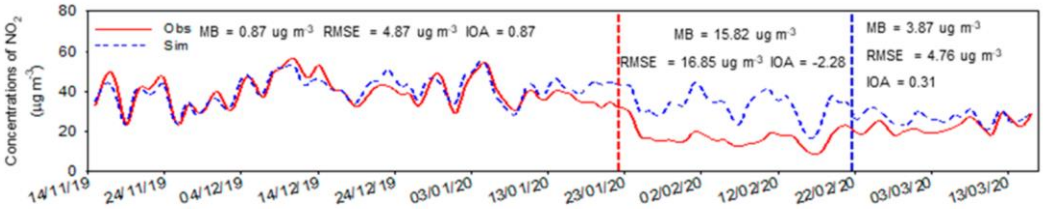

Fig.reply4 *Comparison of simulated and observed NO_2 at 174 cities in eastern China from Nov.14, 2019 to March 15, 2020 (Lu et al., 2021) by using the same model and same inventory as this manuscript.*

7. Could the authors add some information about the interferences which are related to each instrument measurements, especially for HONO, nitrate, and NO_2?

Thanks for your suggestions. HONO was measured with a homemade water-based long-path absorption photometer which was described in Chen et al. (2020). Briefly, the interferences were determined by the dual-channel absorption system because the real HONO concentration was the difference between the two channels. One of the known HONO interferences is NO_2, which had a relatively low solubility in water, and had almost the same concentration in both channels.

Other parameters were measured with respective commercial instruments. The 42i (Thermo Scientific) used molybdenum NO_2-to-NO converter, there would be a NO_2 overestimation for the conversion of HONO, HNO_3, and other NO_y. Compared with NO_2, the concentrations of other components were relatively low, so their impacts could be small. ToF-ACSM (Aerodyne) was developed via Fröhlich et al. (2013) for Non-refractory $PM_{2.5}$ measurement. The detailed usage could be found in Liu et al. (2020), where ionization efficiency calibration of nitrate was performed using 300 nm dry NH_4NO_3 every month during the observation.

These descriptions have been added into the revised manuscript, please see ***section 2.1*** *in the revised manuscript.*

8. It could be simulated a missing HONO source presence for additional contribution to ozone formation on vertical average over the J(nitrate)/J(HNO3)=30 ratio?

For the missing HONO source study, Su et al. (2008) proposed the calculation of unknown HONO source in their study, usually described as $R_{Unknown}$*. The below equation shows the processes influencing the concentration of HONO at a site:*

$$\frac{d(HONO)}{dt} = (R_{OH+NO} + R_{Unknown} + R_{Emiss} + R_V + R_H) - (R_{OH+HONO} + R_{Phot} + R_{Depo})$$

Where $\frac{d(HONO)}{dt}$ *is the observed variation of HONO concentrations,* R_{OH+NO} *is the homogeneous HONO formation rate via OH+NO ,* $R_{Unknown}$ *is the combination of HONO production from unknown sources,* R_{Emiss} *is the emission rate,* R_V *describes the vertical transport process,* R_H *describes the horizontal transport process,* $R_{OH+HONO}$ *is the HONO loss rate from reaction OH+HONO,* R_{Phot} *is the photolysis loss rate and* R_{Depo} *is the deposition loss rate.*

This method is commonly used in the $R_{Unknown}$ *calculation. By combining with the HONO observations, assuming a pseudo steady state for HONO and neglecting small terms,* $R_{Unknown}$ *was calculated via the following equation by Su et al. (2008):*

$$R_{Unknown} \approx J_{HONO}[HONO] + k_{OH+HONO}[OH][HONO] + \frac{d(HONO)}{dt} - k_{OH+NO}[OH][NO]$$

The calculated $R_{Unknown}$ *is an estimation of the whole missing HONO sources and usually used in box model for a typical observation site near the ground. The* $R_{Unknown}$*excluded horizontal and vertical transport.*

$R_{Unknown}$ *impacts on* RO_x *(=OH +*HO_2*+*RO_2*) cycles and* O_3 *formation can be found in our previous studies (Guo et al., 2020; Tang et al., 2015). In this study, each of the other five potential HONO sources except the traffic source can be grouped as* $R_{Unknown}$ *according to Su et al. (2008), and have been discussed, including your suggestion, shown in* ***sections 3.2 and 4*** *in the revised version.*

9. Line 39 and line 802: “one order of magnitude”

Thanks for your suggestions. The expression of "one order of magnitude" has been changed to "ten times" in the revised manuscript.

10. Lines 66-68: Please add more sources of HONO from homogeneous reactions ... For direct emissions and heterogeneous HONO sources are plenty of references and would be great for the consistent state of the art to include homogeneous HONO sources too, even if they have a lower contribution in comparison with NO + OH.

Thanks for your good suggestion. A number of references about homogeneous reactions formed HONO have been added to the revised manuscript, including via NO+OH reaction (Pagsberg et al., 1997; Stuhl and Niki, 1972), via nucleation of NO_2*,* H_2O*, and* NH_3 *(Zhang and Tao, 2010), via the photolysis of ortho-nitrophenols (Bejan et al., 2006), via the electronically excited* NO_2 *and* H_2O *(Crowley and Carl, 1997; Dillon and Crowley, 2018; Li et al., 2008) and via* $HO_2{\cdot}H_2O+NO_2$ *reaction (Li et al., 2015; Li et al., 2014; Ye et al., 2015). Except for the NO+OH reaction, the contribution of other homogeneous HONO sources was minor in the atmosphere. Please see the* ***Section 1*** *in the revised version.*

11. Again, I could not see the entire work and a general view of the paper since up to 9 figures in SM were missing. The work presented here adds interesting inputs for the effect of HONO sources on the concentrations of ozone, especially during haze events.

We sincerely thank you again for your precious comments and suggestions to our study.

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