Response to Reviewers

We thank the reviewers for their careful reading and their constructive comments on our manuscript. As detailed below, the reviewer's comments are shown as italicized font, our response to the comments are normal font. New or modified text is in blue. All of the line numbers refer to Manuscript ID: acp-2017-1217.

Reviewer: #1

The topic of the heterogeneous process of NOx is of interest to the community, and the vertical measurements of chemical composition in winter haze are particularly valuable. My main concern, however, on the present work is that the model was poorly constrained by observations and had to reply on too many assumptions. The vertical measurements only included O_3*, NO, and* NO_2 *which enabled calculation of* NO_3 *production, but several key parameters, such as* N_2O_5*, VOCs and aerosol surface area density, for loss of* NO_3*,* N_2O_5 *production and subsequent loss to nitrate were not measured, making it very difficult, if not impossible, to evaluate nighttime reactions of NOx and for nitrate formation. In addition, the analysis was only based on one profile measured in the early evening, and too many assumptions in the model calculations were not well justified. All these make it very difficult to judge the validity of the conclusions drawn from the analysis. The authors are advised to carefully consider and reduce these uncertainties which could lead to large bias and possible errors.*

1). *L16. Please elaborate what the simultaneous measurements were conducted.*

Changed as following: "Simultaneous ground-based and tower-based measurements of NO_x and O_3 were conducted…"

2). *L22. Please define the potential of* pNO_3^- *.*

This sentence change to: "The nighttime integrated production of pNO_3^- for …"

Here we deleted "potential".

3). *L57-58. It is not clear how the* N_2O_5 *uptake coefficient in winter will be different from summer.*

Since the properties of the aerosol particles (e.g., organic compounds, particle nitrate, liquid water contents, solubility, viscosity, etc.) and meteorological conditions and (e.g. temperature, relative humidity etc.) are different in summer and winter, these differences will led to changes of the N_2O_5 uptake coefficient. The explanation was added in Line 57-58: "This is because the properties of aerosol particles (e.g., organic compounds, particulate nitrate, liquid water contents, solubility, and viscosity) and meteorological conditions (e.g., temperature and relative humidity) differ between summer and winter (Chen et al., 2015; Zhang et al., 2007)."

4). *L96. The abbreviation of 'IAP' should be spelled out upon the first mention in the text.*

Changed accordingly.

5). *L110. Please specify which instruments were installed on board a movable cabin.*

Specified in the Line 110: "NO_x and O_3 instruments were installed on board a movable cabin on the tower…"

6). *L111-113. Describe in more detail about the two light-weight instruments for vertical measurements of O_3, NO and NO_2. How were they calibrated, was there any intercomparison with conventional monitors?*

The detailed description after Line 113 was added: "NO_x calibration was performed in the lab using a gas calibrator (TE-146i, Thermo Electron, USA) associated with a NO standard (9.8 ppmv). The O_3 calibration was done with an O_3 calibrator (TE 49i-PS), which was traceable to NIST (National Institute of Standards and Technology) standards annually. Before the campaign, the NO_x monitor was compared with a Cavity Attenuated Phase Shift (CAPs) Particle Light Extinction Monitor, and the O_3 monitor was compared to a commercial O_3 analyzer (TE-49i, Thermo Electron, USA). Good agreement was found between the portable instruments and the conventional monitors."

7). *L124. Were the daily cycles conducted in the same time periods for every day?*

Yes, we conducted the daily cycle measurement in the similar time periods for the three days.

8). L129-131. The assumptions of no NO influence, no physical mixing, and no transport of the air mass may not be valid here. Without continuous or intermittent measurement as constraints, it is difficult to know the evolution of the air masses, as such one cannot testify the validity of these assumptions.

Since high O_3 concentrations (20 ppbv) at the high altitude (>150 m) were observed at night, the NO concentrations have to be zero for this kind of condition. We were using a box model for the interpretation of the observed dataset. In the framework of box model analysis, the assumption is the analyzed air mass were well mixed. The assumption of well mixing is plausible since the influence of physical mixing on the reaction rate of NO + O_3 could be neglected.

9). *L143-147. The low theoretical equilibrium ratio of NO_3 to N_2O_5 at the low-temperature condition may not necessarily mean that the N_2O_5 formation dominates the NO_3 loss. More evidence is needed here. The kNO_3 value of 0.02 s^{-1} assumed in the present study was much higher than other studies, for example, Brown et al., 2016, in which the NO_3 reaction with VOC contributes more than half of the total NO_3 loss.*

Yes, high N_2O_5/NO_3 is not means N_2O_5 heterogeneous uptake dominate the NO_3 loss. While during the polluted period in winter Beijing (NO_2 = 45 ppbv, Temperature = 273 K, Sa = 3000 μm^2 cm^{-3}), the pseudo first order loss rate of N_2O_5 heterogeneous uptake will be 1×10^{-3} s^{-1} , corresponding to the N_2O_5 uptake coefficient of 5×10^{-3}, and contributed to NO_3 loss rate of 0.4 s^{-1}, which is much higher than the direct NO_3 loss by the reaction of NO_3 with VOCs, even the k_{NO3} set to a high value of 0.02 s^{-1}. Therefore we believe the N_2O_5 formation dominates the NO_3 loss in this study. With respect to k_{NO3}, Brown et al., (2016) shows the wintertime average k_{NO3} in Hong Kong was about 6×10^{-3} s^{-1}, and dominated by monoterpenes. Previous work showed the average kNO_3 is about 0.011 s^{-1} in rural Beijing in summertime, and BVOCs is the dominating part (Wang et al., 2017; Wang et al., 2018). During wintertime, the BVOCs emission would decrease due to lower temperature and weak solar radiation, the k_{NO3} set to 0.02 s^{-1} in this study represents an upper value to some extent. The differences of this study with the campaign conducted in Hong Kong (Brown et al., 2016) may cause by the higher temperature and much lower aerosol surface area in Hong Kong (Temperature = 285 K, Sa ≈ 200 μm^2 cm^{-3}).

In Line 145, we rewrite the part as following: “During the polluted period in winter Beijing (here NO_2 = 45 ppbv, Temperature = 273 K, Sa = 3000 μm^2 cm^{-3}), the ratio of N_2O_5 to NO_3 is large enough, i.e., 450, the pseudo first order loss rate of N_2O_5 heterogeneous uptake will be 1×10^{-3} s^{-1} with the N_2O_5 uptake coefficient of 5×10^{-3}. N_2O_5 uptake would contribute an NO_3 loss rate of 0.4 s^{-1}, and much higher than the direct NO_3 loss by the reaction of NO_3 with VOCs, even the k_{NO3} set to a high value of 0.02 s^{-1}. Therefore, N_2O_5 uptake was proposed to be dominantly responsible for the NO_3 loss and the initial s(t) was set to 1”

10). *L148-150. It is not clear how the author determined the initial concentration of NO_2 and O_3, which could affect the integrated concentrations. Were you using the* *iterative method suggested by Wagner et al., 2013? The retrieved results should be included in the supplementary. Any measurement constraints were used to validate this calculation?*

Yes, we used the iterative method suggested by Wagner et al., 2013. The initial NO_2 and O_3 concentration were derived according to Eq. 1 and Eq. 2, O_3 and NO_2 are integrated backward in time to sunset. The average initial NO_2 and O_3 above 150 m at sunset time is about 61 ± 3 ppbv and 27 ± 6 ppbv, respectively. The measured NO_2 concentration in PKU site at sunset time (local time, 16:55) is 61 ppbv and show good consistent with the model result.

Changed as following in Line 299: "The average initial NO_2 and O_3 above 150 m at sunset time is about 61 ± 3 ppbv and 27 ± 6 ppbv, respectively. The measured NO_2 concentration in PKU site at sunset time (local time, 16:55) is 61 ppbv and show good consistent with the model result."

11). *L146. "than" should be "that".*

Corrected accordingly.

12). *L162. Please clarify the exact time period for the model running from sunset to sunrise.*

Here we clarified as: "Sunset and sunrise time during the measurement is 16:55 and 07:30 (Chinese National Standard Time, CNST) and the length of night was about 14.5 h, the model is run from sunset to sunrise with the running time set to 14.5 h."

13). *L164-168. There are some flaws in the Eq.1 to 5 of the box model. To simplify the differential equation, the author assumes an equilibrium between NO_3 and N_2O_5 in Eq. 3 and 4, which means the loss rate of NO_2 through R2 and production rate from R3 should be equal. Therefore, the NO_2 loss rate will equal to the reaction rate of R1, which contradicts Eq. 2. As suggested by Wagner et al., 2013, "the assumption of equilibrium leads to an error which accumulates as the equations are integrated". This could affect the results when retrieving the initial NO_2 concentration and subsequent model simulation. To be more accurate, I suggest the author use the explicit equations suggested by Wagner et al., 2013.*

Thanks for the suggestion, we corrected these equations accordingly, and the fixed k_{NO3} and k_{N2O5} first, set s(t) to 1 in the first step and then iterate the s(t) till the difference between two iteration less than 0.005. The explicit equations changed as:

"(Eq. 1) $\frac{d[O_3]}{dt} = -k_{NO2+O3}[O_3][NO_2]$

(Eq. 2) $$\frac{d\,[NO_2]}{dt} = -(1 + s(t)) \times k_{NO2+O3}[O_3][NO_2]$$

(Eq. 3) $$\frac{d\,[NO_3+N_2O_5]}{dt} = k_{NO2+O3}[O_3][NO_2] - k_{N2O5}[N_2O_5] - k_{NO3}[NO_3]$$

(Eq. 4) $$\frac{[N_2O_5]}{[NO_3]} = k_{eq}[NO_2]$$

(Eq. 5) $$s(t) = \frac{\int_0^t k_{N2O5}\cdot[N_2O_5]dt + [N_2O_5]_t}{[O_3](0)-[O_3](t)}$$

(Eq. 6) $$k_{N2O5} = \frac{C \times S_a \times \gamma_{N2O5}}{4}$$ ”

14). *L169-L179. The surface area was calculated based on the measurement of particle size distribution from 0.01 to 0.6 µm and could be underestimated due to the lack of information of larger particles, resulting in large uncertainty in the calculated uptake coefficients. It is necessary to provide an uncertainty estimation of how much this will affect the results.*

During the study period, the particle number and size distribution (PNSD) larger than 0.7 µm is unavailable, it is different to quantify the contribution from lager particles. While during the following polluted episode (2017-01-01 to 2017-01-07), PNSD of $PM_{2.5}$ data are available, we found particle smaller than 0.7 µm dominated more than 95% aerosol surface area, the similar result also represented in Germany and summer Beijing (Crowley et al., 2010; Wang et al., 2018). The underestimation of S_a in this study (5%) could lead to the overestimation of N_2O_5 3.6% - 4.2%, and underestimation of pNO_3^- with 0.2% - 2.5% considering the N_2O_5 uptake coefficient varied from 1×10^{-3} to 0.05.

We added the description in Line 179: “Although the PNSD information for particles larger than 0.7 µm was not valid during the study period, the particles smaller than 0.7 µm dominated more than 95% of the aerosol surface area in a subsequent pollution episode (01/01/2017 to 01/07/2017), and similar results also were reported in other studies (e.g., Crowley et al., 2010a; Wang et al., 2018). The possible lower bias of S_a (5%) only led to a small overestimation of N_2O_5, i.e., 3.6%–4.2%, and an underestimation of pNO_3^- of 0.2%–2.5% when γ_{N2O5} varied from 1×10^{-3} to 0.05.”

15). *L240-241, L245-247, L257-258. The author attributed the lower Ox level at high altitude on the night of December 19 to missing sinks of Ox with high N_2O_5 uptake, but it could also be a result of the continuous emissions of NOx near the ground leading to accumulation of NO_2 within the nocturnal boundary layer with a height around 100m. So the Ox level in the residual layer and surface doesn't have to be conserved. Here it would be good to show vertical information on meteorological parameters.*

The emission of NO would not influence the sum of Ox (=O_3+NO_2). The emission of NO_2 would led to higher O_x at the surface layer. Nevertheless, the vertical profile measurement showed no vertical gradient of NO_2 lower than 150 m so that we do not think there could be a significant emission of NO_2. And therefore, the O_x level shall be conserved between the nocturnal boundary layer and the residual layer since no O_3 is produced at night. In addition, the nocturnal boundary layer is determined to be about 340 m through the vertical profile of temperature during the same period in Beijing (Zhong et al., 2017).

***16).** L264-265, Wrong figure number referenced here.*

Corrected accordingly.

***17).** L265. Please explicitly define the equation used for calculating the nitrate accumulation.*

We listed the calculation equation as following:

“
$$\sum \mathrm{pNO_3^-} = \int_0^t (2-f)\cdot k_{\mathrm{N2O5}}\cdot [\mathrm{N_2O_5}]dt \qquad (7)$$
”

***18).** L262-267. This model simulation assumes an ideal condition with no NO concentration above 150 m from the sunset to 21:00, which cannot be substantiated. Thus the calculated accumulation of nitrate is questionable.*

Due to the strong thermal inversion during winter haze episode (e.g. Zhong et al., 2017), the isolation is existed more easily in vertical scale in urban Beijing, the air mass in upper layer is not easily affected by surface NO emission. The theoretical framework of the box model we used is same as Wagner et al., (2013) and Yun et al., (2018). The model allow us to accumulate the pNO_3^- till sunrise, which shows an upper limit of the nitrate production via N_2O_5 uptake in the upper layer. In addition, at sunset time, we observed significant O_3 presented at the near surface layer. Before O_3 is fully titrated away, the NO concentrations shall be zero for the sunset time.

***19).** L281-282. Please elaborate how to deal with calculations of mixing.*

We rewrite the vertical mixing in L279-283 as following: “Zhong et al. (2017) showed that the NBL and PBL both were at 340 m from December 19 to 20, 2016 in Beijing. Daytime vertical downward transportation was helpful in mixing the air mass within the PBL. Assuming the newly formed pNO_3^- aloft from 150 m to 340 m is 50 μg m^{-3} during the nighttime and well mixed within the PBL by the next morning, the enhancement to the surface layer (ΔpNO_3^-) can be simplified to the calculation in

Eq. 8 as following:

$$\Delta \mathrm{pNO_3} = \frac{\int_0^{150} P(\mathrm{pNO_3})dH + \int_{150}^{340} P(\mathrm{pNO_3})dH}{340} \qquad (8)$$

Here, $P(\mathrm{pNO_3^-})$ is the integral production of $\mathrm{pNO_3^-}$ and H represents height. Owing to high NO below 150 m, the $\mathrm{pNO_3^-}$ formation via N_2O_5 uptake was zero. The enhancement of $\mathrm{pNO_3^-}$ from 150 m to 340 m was calculated as 28 µg m^{-3},"

20). *L282-285. If the author's hypothesis is true, it should be able to observe a sharp increase of particulate nitrate at the ground site in the morning of December 20. Any evidence on that?*

The particulate nitrate measurement is not available in this study, but as labelled in Figure 2(a), the red line showed PM concentration had a sharp increase of ~60 µg m^{-3}, which was purposed to be consist with the result considering a large proportion of particulate nitrate in PM mass concentration, especially during winter polluted episode in Beijing (e.g., Zheng et al., 2015).

21). *L309. Eq.6 is incorrect. The particle nitrate formation is twice of N_2O_5 loss if assuming $ClNO_2$ yield is zero.*

Thanks, we corrected accordingly.

22). *L328, "coral" should be "coal".*

Corrected accordingly.

23). *L342-343. A possible reason for the small difference on kNO_3 variation could be the NO_3 change (via VOC loss) were unaccounted for in Eq.2 that used to retrieve the initial concentration. Comparison between the full differential equations and simplified calculation is required to validate the results.*

The full differential equations was used to recalculate the $\mathrm{pNO_3^-}$ variation on k_{NO3}. The s(t) decreased from 1 to 0.99 even k_{NO3} set to 0.05, and the difference between the full differential equations and simplified calculation is negligible, suggested that the calculation result is valid.

24). *L351, delete "to be zero".*

Corrected accordingly.

25). *L357, add “that” after “found”.*

Corrected accordingly.

Reviewer: #2

The authors explore the mechanisms for particulate nitrate (pNO_3^-*) during wintertime haze events in Beijing, China. Comparing simultaneous ground-based and tower-based observations, the authors investigated the significance of p*NO_3^- *via* N_2O_5 *heterogeneous uptake as a function of altitude. The work shows the effects of the p*NO_3^- *formed aloft on the surface* $PM_{2.5}$ *the following the morning. Given the significance of this work, I recommend this manuscript for publication after significant revisions.*

We thank for the Reviewer #2’s constructive comments and suggestions to improve the quality of our manuscript.

1). *although the experiment design is well thought and the analysis appears to be solid, the technical writing needs significant improvement. I recommend the authors to use professional technical writing services in English to improve the penmanship and eliminate any grammatical errors. Example sentences to be reviewed carefully and reformulated are line 66-70, line 178-179, 180-183, 186-190, 194-195, 205-213, 242-244, 275-278, 292-295 etc.*

The resubmitted manuscript has been edited by a professional service in English.

2). *I am assuming eq.1 (line 164) is for the nitrate radical production rate (*PNO_3*), not the rate of change in* O_3*. As the authors mentioned the availability of* O_3 *is driven by its reaction with NO.*

Yes, Eq. 1 is the production of nitrate radical, but O_3 is also one reactant of this reaction. As the production of NO_3 takes place, the O_3 is consumed. This reaction is more important for O_3 losses for the conditions of the high-altitude (>150 m) air masses of which the reaction pathway of O_3 + NO is negligible due to the presence of zero NO.

3). *Use subscript for Ox throughout the text*

Corrected accordingly.

***4).** the authors define and discuss "particle nitrate convert efficiency" (sigma) in line 305- 310. Chang et al. 1 gives an excellent review of N_2O_5 chemistry and I suggest the authors read this as they discuss and introduce parameters regarding N_2O_5 conversion. I do not believe it is necessary to introduce a new parameter "particle nitrate convert efficiency" in this case.*

Thanks for your suggestion, Chang et al. (2011) reviewed the N_2O_5 chemistry systematically and comprehensively. With respect to N_2O_5 conversion, Chang et al., focused on the contribution to overnight NO_x loss. Here we revised the parameter to "Overnight NO_x loss efficiency (ε)", which also indicates the nitrate formation capacity. The equation changed as following:

(Eq. 9) $$\varepsilon = \frac{\int_0^t 2\times k_{\mathrm{N2O5}}\cdot[\mathrm{N_2O_5}]dt+\int_0^t k_{\mathrm{NO3}}\cdot[NO_3]dt}{[NO_2](0)}$$

Here the consumed NO_3 with VOCs and N_2O_5 uptake regarded as the effective NO_x loss. The Figure 7 changed the Y-axis and we did not normalize the loss efficiency, which shows the similar result with previous figure version."

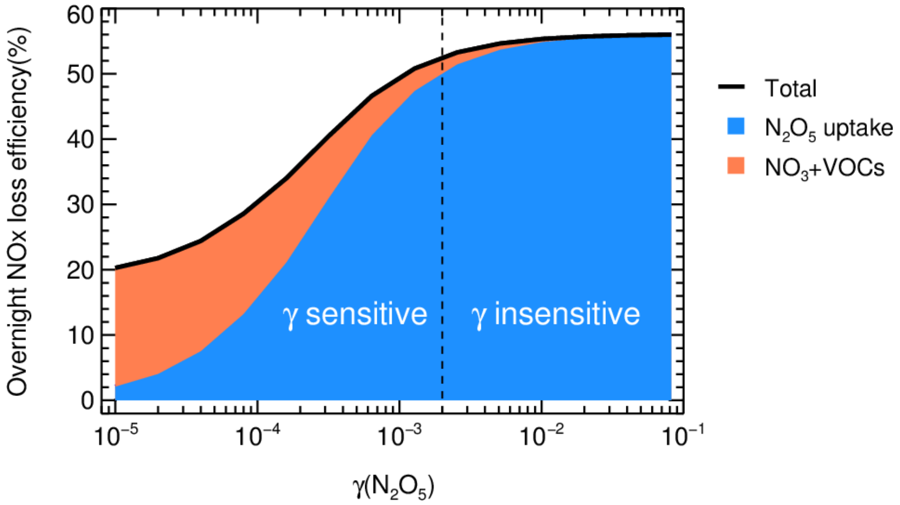

Figure 7. The dependence of overnight NO_x loss on N_2O_5 uptake on γ_{N2O5} in a typical winter pollution condition. The initial NO_2 and O_3 set to 60 ppbv and 30 ppbv, respectively, S_a set to 3000 μm^2 cm^{-3}, the $ClNO_2$ yield is zero and k_{NO3} is 0.02 s^{-1}. The reaction time set to 14.5 h. The blue and orange zone represent the contribution of NO_3+VOCs and N_2O_5 uptake, the dashed line (γ = 0.002, when N_2O_5 uptake contribute to 90% of the maximum NO_x loss) divide the loss into γ sensitive and insensitive region. The maximum nocturnal NO_x loss by NO_3-N_2O_5 chemistry is 56%.

***5).** In that regard, the authors need to extend the literature search and include more references on N_2O_5 heterogeneous uptake and wintertime haze events outside the Beijing area. For more references on relevant topic, review publications of Chang et* *al. 1-2 , Lurmann et al. 3 , Brown et al. 4 , Green et al. 5 , Wang et al. 6 , Prabhakar et al. 7 etc.*

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Thanks for the suggestion and we compared our results with these references that concern the winter haze event in other region, and cited these work in the revised manuscript.

References

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