## 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: #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 $pNO_3^-$ via $N_2O_5$ heterogeneous uptake as a function of altitude. The work shows the effects of the $pNO_3^-$ 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 ($\varepsilon$)", which also indicates the nitrate formation capacity. The equation changed as following:

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

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."

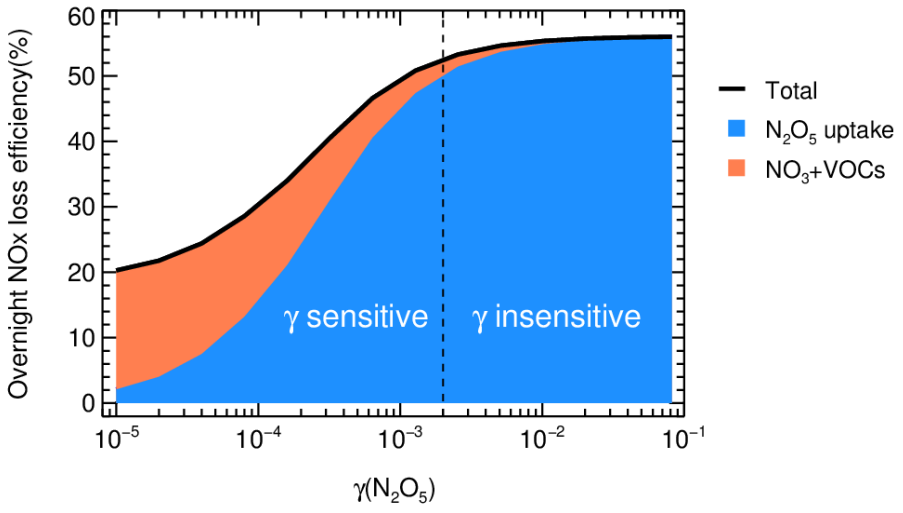


**Figure 7.** The dependence of overnight $NO_x$ loss on $N_2O_5$ uptake on $\gamma_{N2O5}$ in a typical winter pollution condition. The initial $NO_2$ and $O_3$ set to 60 ppbv and 30 ppbv, respectively, $S_a$ set to 3000 $\mu m^2$ $cm^{-3}$, the $ClNO_2$ yield is zero and $k_{NO3}$ is 0.02 $s^{-1}$. 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 ($\gamma$ = 0.002, when $N_2O_5$ uptake contribute to 90% of the maximum $NO_x$ loss) divide the loss into $\gamma$ 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.*

*1. Chang, W.; Bhave, P.; Brown, S.; Riemer, N.; Stutz, J.; Dabdub, D., Heterogeneous atmospheric chemistry, ambient measurements, and model calculations of $N_2O_5$: A review. Aerosol Sci. Technol.* ***2011,*** *45 (6), 665-695. DOI 10.1080/02786826.2010.551672.*

*2. Chang, W. L.; Brown, S. S.; Stutz, J.; Middlebrook, A. M.; Bahreini, R.; Wagner, N. L.; Dubé, W. P.; Pollack, I. B.; Ryerson, T. B.; Riemer, N., Evaluating $N_2O_5$ heterogeneous hydrolysis parameterizations for CalNex 2010. J. Geophys. Res.: Atmos.* ***2016,*** *121 (9), 5051-5070. DOI 10.1002/2015JD024737.*

*3. Lurmann, F. W.; Brown, S. G.; McCarthy, M. C.; Roberts, P. T., Processes influencing secondary aerosol formation in the San Joaquin Valley during winter. J. Air Waste Manage. Assoc.* ***2006,*** *56 (12), 1679-1693. DOI 10.1080/10473289.2006.10464573.*

*4. Brown, S. G.; Roberts, P. T.; McCarthy, M. C.; Lurmann, F. W.; Hyslop, N. P., Wintertime vertical variations in particulate matter (PM) and precursor concentrations in the San Joaquin Valley during the California Regional Coarse PM/Fine PM Air Quality Study. J. Air Waste Manage. Assoc.* ***2006,*** *56 (9), 1267-1277. DOI 10.1080/10473289.2006.10464583.*

*5. Green, M. C.; Chow, J. C.; Watson, J. G.; Dick, K.; Inouye, D., Effects of snow cover and atmospheric stability on winter $PM_{2.5}$ concentrations in Western U.S. valleys. J. Appl. Meteor. Climatol.* ***2015,*** *54 (6), 1191-1201. DOI 10.1175/JAMC-D-14-0191.1.*

*6. Wang, G.; Zhang, R.; Gomez, M. E.; Yang, L.; Levy Zamora, M.; Hu, M.; Lin, Y.; Peng, J.; Guo, S.; Meng, J.; Li, J.; Cheng, C.; Hu, T.; Ren, Y.; Wang, Y.; Gao, J.; Cao, J.; An, Z.; Zhou, W.; Li, G.; Wang, J.; Tian, P.; Marrero-Ortiz, W.; Secrest, J.; Du, Z.; Zheng, J.; Shang, D.; Zeng, L.; Shao, M.; Wang, W.; Huang, Y.; Wang, Y.; Zhu, Y.; Li, Y.; Hu, J.; Pan, B.; Cai, L.; Cheng, Y.; Ji, Y.; Zhang, F.; Rosenfeld, D.; Liss, P. S.; Duce, R. A.; Kolb, C. E.; Molina, M. J., Persistent sulfate formation from London Fog to Chinese haze. Proceedings of the National Academy of Sciences* ***2016,*** *113 (48), 13630. DOI.*

*7. Prabhakar, G., C. Parworth, X. Zhang, H. Kim, D. Young, A.J. Beyersdorf, L.D. Ziemba, J.B. Nowak, T.H. Bertram, I.C. Faloona, Q. Zhang, and C.D. Cappa, Observational assessment of the role of nocturnal residual-layer chemistry in determining daytime surface particulate nitrate concentrations. Atmos. Chem. Phys. Discuss., 2017.* ***2017****: p. 1-58*

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.