

“Large particulate nitrate formation from N₂O₅ uptake in a chemically reactive layer aloft during wintertime in Beijing” by Wang et al.

The authors explore the mechanisms for particulate nitrate (pNO₃⁻) during wintertime haze events in Beijing, China. Comparing simultaneous ground-based and tower-based observations, the authors investigated the significance of pNO₃⁻ via N₂O₅ heterogeneous uptake as a function of altitude. The work shows the effects of the pNO₃⁻ 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.

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.
2. I am assuming eq.1 (line 164) is for the nitrate radical production rate (P_{NO₃}), not the rate of change in O₃. As the authors mentioned the availability of O₃ is driven by its reaction with NO.
3. Use subscript for O_x throughout the text
4. The authors define and discuss “particle nitrate convert efficiency” (sigma) in line 305-310. Chang et al.¹ gives an excellent review of N₂O₅ chemistry and I suggest the authors read this as they discuss and introduce parameters regarding N₂O₅ conversion. I do not believe it is necessary to introduce a new parameter “particle nitrate convert efficiency” in this case.
5. In that regard, the authors need to extend the literature search and include more references on N₂O₅ 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 N₂O₅: 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 N₂O₅ 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.; Secretst, 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