

Response to Review#2

Title: “Bridging the spatial gaps of the Ammonia Monitoring Network using satellite ammonia measurements”

Authors: R. Wang et al.

We thank the reviewer for the helpful comments. We have revised the manuscript accordingly to help clarify and focus the manuscript. The original comments from reviewers are in *blue and italics*, our replies are in black font, and verbatim responses from the revised manuscript are in **red font**.

### Summary

*This study is well-organized and important for research community. This reviewer has two minor comments. 1) ? 2) can urban atmospheric NH<sub>3</sub> prevent the previously formed NH<sub>4</sub>NO<sub>3</sub> from evaporating? The current discussion is imbalance.*

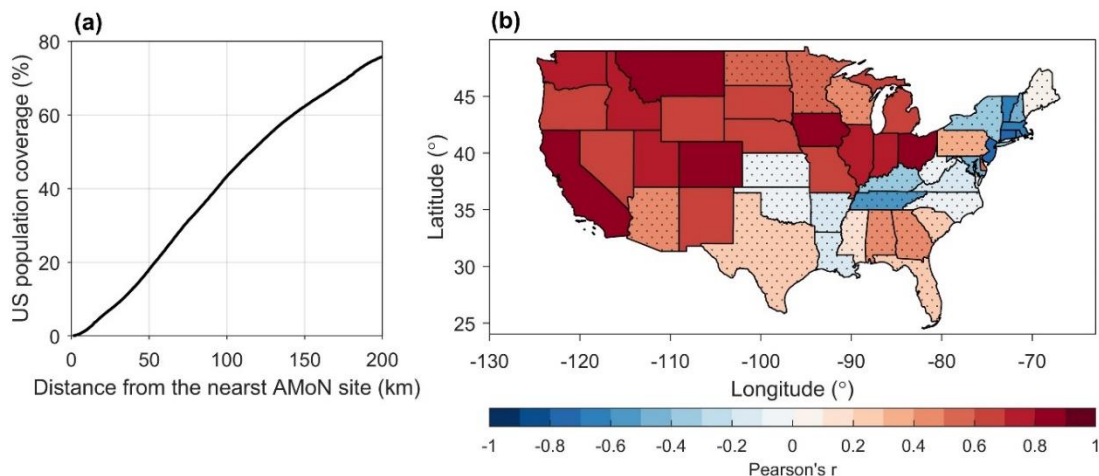
### Minor comments:

Minor comment #1: *the authors should explain why only the data ended on 2018, but not 2022*

Response: IASI observations in 2020 were excluded to rule out the possible impact of the pandemic. IASI observations in 2021 and 2022 were not included because Metop-A retired in 2021.

Minor comment #2: *can urban atmospheric NH<sub>3</sub> prevent the previously formed NH<sub>4</sub>NO<sub>3</sub> from evaporating? The current discussion is imbalance.*

Response: The equilibrium between gas phase NH<sub>3</sub> and HNO<sub>3</sub> and aerosol phase NH<sub>4</sub>NO<sub>3</sub> shifts to the aerosol phase at cold temperature and high particle pH condition (Feng et al., 2020; Guo et al., 2018; Shah et al., 2018). Gas phase NH<sub>3</sub> also plays an important role in aerosol acidity and aerosol chemistry (Lawal et al., 2018). Zhai et al. 2021 demonstrates that gas phase NH<sub>3</sub> hinders the scavenging of nitrate aerosol by slowing down the deposition of total inorganic nitrate. Wang et al. 2020 shows that gas phase NH<sub>3</sub> and HNO<sub>3</sub> can nucleate directly to form NH<sub>4</sub>NO<sub>3</sub> particles in cold atmospheric conditions and is likely to result in rapid growth of new atmospheric particles in winter, especially in urban environments with abundant HNO<sub>3</sub>. Unfortunately, reliable gas phase HNO<sub>3</sub> data in the boundary layer are not readily available to make a full evaluation of NH<sub>4</sub>NO<sub>3</sub> formation. Because trends in SO<sub>2</sub> and NO<sub>x</sub> also impact NH<sub>4</sub>NO<sub>3</sub> formation, it is difficult to evaluate how each city may respond to increases in NH<sub>3</sub> over time. We’ve added Fig. 9b and the following paragraph to discuss the NH<sub>3</sub>’s role in NH<sub>4</sub>NO<sub>3</sub> formation in urban areas in details:



**Figure 9.** (a) Cumulative distribution of CONUS population as a function of distance from the nearest AMoN sites; (b) Correlation between EPA NH<sub>3</sub> emissions and IASI observed mean NH<sub>3</sub> concentrations at state level during 2008 - 2018. The gray dots represent states without statistically significant correlations ( $\alpha = 0.05$ ).

Line 541 - 554: The urban environment with abundant HNO<sub>3</sub> and NH<sub>3</sub> emissions from vehicles nominally favors the formation of NH<sub>4</sub>NO<sub>3</sub> over rural areas. Recent studies suggest that gas phase NH<sub>3</sub> hinders the scavenging of NH<sub>4</sub>NO<sub>3</sub> by slowing down the deposition process of total inorganic nitrate (Zhai et al., 2021) and promotes new atmospheric particle formation by directly nucleate with HNO<sub>3</sub> to form NH<sub>4</sub>NO<sub>3</sub> in winter in urban areas and (Wang et al., 2020). However, ultimately the sensitivity to PM<sub>2.5</sub> from increases in NH<sub>3</sub> in any urban areas will be a complex function of trends of NO<sub>x</sub> and SO<sub>2</sub> as well (Feng et al., 2020). The NH<sub>3</sub> increase in these densely populated areas and its impact on the aerosol chemistry need needs to be further addressed. For example, Fig. 9b shows the relationship between NH<sub>3</sub> trends versus emissions trends (EPA Air Pollutant Emissions Trends Data) on the state level. For agricultural areas with high NH<sub>3</sub> (excess NH<sub>3</sub> relative to NH<sub>4</sub>NO<sub>3</sub> equilibrium), one would expect an increase in emissions to correlate very well with increasing NH<sub>3</sub> columns. In contrast, in areas with more NO<sub>x</sub>, increases in emissions may result in NH<sub>3</sub> going into NH<sub>4</sub>NO<sub>3</sub> and thereby show little or even negative correlations. To this end, Fig. 9b shows that at state level, agricultural states show strong correlations between emissions and concentrations trends, e.g., California, while the more urbanized northeast states show weak or negative correlations, e.g., New Jersey. Ultimately, co-located aerosol phase and gas phase precursor measurements are needed to fully deduce what is happening at each urban area and should be a focus of future air quality network integration.

Line 607 - 610: The comparison between NH<sub>3</sub> emission trends and IASI observed NH<sub>3</sub> concentration trends suggests that strong correlations exist in agricultural states, e.g., California, while weak or negative correlations in more urbanized northeast states, e.g., New Jersey, indicating the different contribution from emission and partitioning.

Line 43 - 45: A comparison between IASI NH<sub>3</sub> concentration trends and state-level NH<sub>3</sub> emission trends is then performed to reveal that good correlations exist in agricultural states while negative correlations in more urbanized states, suggesting the different roles of emission and partitioning in NH<sub>3</sub> increases.

## References:

- EPA, United States Environmental Protection Agency, Air Pollutant Emissions Trends Data, <https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data>, last access: August 2023.
- Feng, J., Chan, E., and Vet, R.: Air quality in the eastern United States and Eastern Canada for 1990–2015: 25 years of change in response to emission reductions of SO<sub>2</sub> and NO<sub>x</sub> in the region, *Atmos. Chem. Phys.*, 20, 3107–3134, <https://doi.org/10.5194/ACP-20-3107-2020>, 2020.
- Guo, H., Otjes, R., Schlag, P., Kiendler-Scharr, A., Nenes, A., and Weber, R. J.: Effectiveness of ammonia reduction on control of fine particle nitrate, *Atmos. Chem. Phys.*, 18, 12241–12256, <https://doi.org/10.5194/acp-18-12241-2018>, 2018.
- Lawal, A. S., Guan, X., Liu, C., Henneman, L. R. F., Vasilakos, P., Bhogineni, V., Weber, R. J., Nenes, A., and Russell, A. G.: Linked Response of Aerosol Acidity and Ammonia to SO<sub>2</sub> and NO<sub>x</sub> Emissions Reductions in the United States, *Environ. Sci. Technol.*, <https://doi.org/10.1021/acs.est.8b00711>, 2018.
- Shah, V., Jaeglé, L., Thornton, J. A., Lopez-Hilfiker, F. D., Lee, B. H., Schroder, J. C., Campuzano-Jost, P., Jimenez, J. L., Guo, H., Sullivan, A. P., Weber, R. J., Green, J. R., Fiddler, M. N., Bililign, S., Campos, T. L., Stell, M., Weinheimer, A. J., Montzka, D. D., and Brown, S. S.: Chemical feedbacks weaken the wintertime response of particulate sulfate and nitrate to emissions reductions over the eastern United States, *Proc. Natl. Acad. Sci. U.S.A.*, <https://doi.org/10.1073/pnas.1803295115>, 2018.
- Wang, M., Kong, W., Marten, R., He, X. C., Chen, D., Pfeifer, J., Heitto, A., Kontkanen, J., Dada, L., Kürten, A., Yli-Juuti, T., Manninen, H. E., Amanatidis, S., Amorim, A., Baalbaki, R., Baccarini, A., Bell, D. M., Bertozzi, B., Bräkling, S., Brilke, S., Murillo, L. C., Chiu, R., Chu, B., de Menezes, L. P., Duplissy, J., Finkenzeller, H., Carracedo, L. G., Granzin, M., Guida, R., Hansel, A., Hofbauer, V., Krechmer, J., Lehtipalo, K., Lamkaddam, H., Lampimäki, M., Lee, C. P., Makhmutov, V., Marie, G., Mathot, S., Mauldin, R. L., Mentler, B., Müller, T., Onnela, A., Partoll, E., Petäjä, T., Philippov, M., Pospisilova, V., Ranjithkumar, A., Rissanen, M., Rörup, B., Scholz, W., Shen, J., Simon, M., Sipilä, M., Steiner, G., Stolzenburg, D., Tham, Y. J., Tomé, A., Wagner, A. C., Wang, D. S., Wang, Y., Weber, S. K., Winkler, P. M., Wlasits, P. J., Wu, Y., Xiao, M., Ye, Q., Zauner-Wieczorek, M., Zhou, X., Volkamer, R., Riipinen, I., Dommen, J., Curtius, J., Baltensperger, U., Kulmala, M., Worsnop, D. R., Kirkby, J., Seinfeld, J. H., El-Haddad, I., Flagan, R. C., and Donahue, N. M.: Rapid growth of new atmospheric particles by nitric acid and ammonia condensation, *Nature*, 581, 184–189, <https://doi.org/10.1038/s41586-020-2270-4>, 2020.
- Zhai, S., Jacob, D. J., Wang, X., Liu, Z., Wen, T., Shah, V., Li, K., Moch, J. M., Bates, K. H., Song, S., Shen, L., Zhang, Y., Luo, G., Yu, F., Sun, Y., Wang, L., Qi, M., Tao, J., Gui, K., Xu, H., Zhang, Q., Zhao, T., Wang, Y., Lee, H. C., Choi, H., and Liao, H.: Control of particulate nitrate air pollution in China, *Nat. Geosci.*, 14, 389–395, <https://doi.org/10.1038/s41561-021-00726-z>, 2021.