Response to Editor Comments

I have taken note of the rebuttal to the reviewer's comments. Several (main) concerns have voiced with regard to the use and quality of surface observations for comparison, the use of coarse resolution model and low temporal resolution for comparison with IASI satellite data, and issues with the gas-particle partitioning. Although the responses were to some extent addressing the reviewers concerns, I encourage the authors to avoid relaying issues for 'future work', and where appropriate extend the analysis with some sensitivity studies. Further discussion is warranted wrg to quality issues of surface observations: information on calibration procedures, and in particular for what it means for this study should be described carefully.

Reply: We thank Editor for his comment and suggestions. We agree with both the reviewers' comments and suggestions and address their concerns that have greatly improved the manuscript's quality. We have used an additional set of simulations using the WRF-Chem model over South Asia with a consistent emission inventory and chemical scheme to see the impact of finer model resolution and high temporal resolution compared to the IASI satellite data.

We have also added the following discussion on the data quality and the quality control procedure adopted in this study.

The quality control and assurance method, followed by Central Pollution Control Board (CPCB) for these air quality monitoring stations, is given in the CPCB (2011 and 2020). Furthermore, we take the following steps to reassure the quality of NH_3 observations from the CPCB network stations. For data quality, we rejected all the observations values below the lowest detection limit of the instrument (1 µg m^{-3}) (Technical specifications for CAAQM station, 2019) because most of the sites are situated in the urban environment. For cities where more than one monitoring station is available, we rejected all the observations above 250 µg m^{-3} at a given site if other sites in the network do not show values outside this range. This step aims to eliminate any short-term local influence that cannot be captured in the models and retain the regional-scale variability. Second, we removed single peaks characterized by a change of more than 100 µg m^{-3} in just one hour for all the data in CPCB monitoring stations. This step filters random fluctuations in the observations. Third, we removed some very high NH_3 values that appeared in the timeseries right after the missing values. For any given day, we removed the sites from the consideration that either experience instrument malfunction, or appear to be very heavily influenced by strong local sources. In order to verify the data quality of CBCB monitoring site, we have inter compared the NH_3 measurement at CPCB monitoring station (R.K. Puram) in Delhi with the NH_3 measurements at Indira Gandhi International (IGI) Airport taken during Winter Fog Experiment (WiFEX) (Ghude et al., 2017) using Measurement of Aerosols and Gases (MARGA) instrument during winter season of 2017-2018. More details on the NH_3 measurements using MARGA is available with Acharja et al. (2020). Both sites were situated in the same area of Delhi (less than 1km). Our inter-comparison show that NH_3 measured at CPCB monitoring station by chemiluminescence method are slightly (on an average 9.8 µg m^{-3}) on higher side than NH_3 measured by ion chromatography (IC) using MARGA (Fig. S1 in the revised Supplement). The differences that were observed could partly be related to the different NH_3 measurement techniques and partly to the locations of the two monitoring sites which were not place exactly at same location. Apparently, the difference of 9.8 µg m^{-3} indicates that the NH_3 measurements from the CPCB do not suffer from the calibration issue.

In particular, I would like to see a somewhat more in depth discussion on the potential biases derived from mismatch of temporal matching and boundary layer dynamics in the MOZART model, in particular in winter when high atmospheric stability prevents mixing, and IASI may not observe all NH_3 close to the surface. A case study with higher temporal (and spatial) resolution for a limited and more frequent output and realistic assumptions on IASI effective kernels may be helpful to illustrate the sensitivity of results.

Reply: We have also discussed the potential biases derived from the mismatch of temporal matching and boundary dynamics in the MOZART mode. However, as suggested by the reviewer, in the revised manuscript, we have now compared the monthly mean columns by averaging paired observations across the months. We have considered the daily NH_3 cloud-free satellite total column data and compared it with the modeled daily NH_3 total column averaging paired observations across the months, seasons, and year. For consistency with satellite retrievals, first, the model output (11:30 LT) at each day close to satellite overpass time (09:30 LT) is interpolated in space to the location of valid satellite retrievals. Since the IASI retrieval algorithm only provides total columns, in the second step, we made the unweighted average distribution of the daily paired data to obtain a monthly mean value of satellite and model total NH_3 columns at each model grid location. We find that the normalized mean bias (NMB) over IGP decreased to 38% with pair-comparison than non-paired comparison (58%) considering the model columns close to satellite overpass time. However, normalized mean bias (NMB) increased to -41% with paired-comparison over the NCP region than non-paired comparison (-37%).

IASI retrieval method used for NH_3 does not produce averaging kernels as it is not based on optimal estimation. Therefore, IASI retrievals' limitation is that it does not allow the calculation of an averaging kernel to account for the vertical sensitivity of the instrument sounding to different layers in the atmosphere. We refer to Van Damme et al. (2017); Whitburn et al. (2016) for a comprehensive discussion on the advantages and disadvantage of constrained versus unconstrained retrieval approaches for NH_3. In brief, the current approach's main advantage is that a priori information does not influence the retrieval. Therefore, the NH_3 column value is derived from the measurement only. We compared column to column, as the IASI retrieval algorithm only provides total columns. We made unweighted average distributions using all the morning IASI measurements available, following the recommendation for using the dataset provided in Van Damme et al., (2017). In this paper, we have use NH_3 total columns retrieved from the IASI instrument morning overpass (AM) observations (i.e., 09:30 local time).

Further, in order to see the impact of finer resolution and more frequent output (1hr), we used simulated NH_3 concentration for the year 2011 using WRF-Chem simulation for the year 2011 from work reported in Ghude et al. (2016) over south Asia at 36 km grid spacing. The model uses MOZART-4 gas-phase chemistry linked to the GOCART aerosol scheme, similar to the one which is used in MOZART-4 simulation in the present work. Again, we have considered the daily NH_3 cloud-free satellite total column data for 2011 and compared it with the modeled daily NH_3 total column averaging paired observations across the year. For consistency with satellite retrievals, first, the model output (9:30 LT) at each day is interpolated in space to the location of valid satellite retrievals at an overpass time of 09:30 LT. Since the IASI retrieval algorithm only provides total columns, in the second step, we made the unweighted average distribution of the daily paired data to obtain a yearly mean value of satellite and model total NH_3 columns at each model grid location (36 km). We found that the bias between the model and IASI NH_3 total columns is even larger with finer-scale simulations compared to coarse simulations. We have included this for the reviewer's reference but not included it in the revised manuscript as it gives a similar difference, but the magnitude of the difference is larger with WRF-Chem simulations.

We requested Editor to refer to our responses and figures provided in the 'Response to Anonymous Referee #1's and Anonymous Referee #2's Comments' document enclosed with the revised manuscript.

Likewise some first order estimate of the impact of applying a temporal profile on agricultural NH_3 emission would be preferable.

Reply: Unfortunately, the application of agriculture has significant spatial and temporal variability over South Asia, which depends on the cropping season and cropping pattern, is not well documented. However, we agree that it will contribute to the mismatch observed between observed and modeled NH_3 columns to some extent. Under the on-going South Asia Nitrogen Hub (SANH) project (The Global Challenges Research Fund (GCRF) South Asia Nitrogen hub), it is planned to develop a high-resolution NH_3 emission inventory over South Asia that will account for the temporal profile of agricultural NH_3 emission based on agricultural statistics.

I encourage the author to resubmit, taken the review comments and my instructions as much as possible into account.

Reply: We requested Editor to refer to our responses in the 'Response to Anonymous Referee #1's and Anonymous Referee #2's Comments' document enclosed with the revised manuscript.

References

Acharja, P., Ali, K., Trivedi, D. K., Safai, P. D., Ghude, S., Prabhakaran, T. and Rajeevan, M.: Characterization of atmospheric trace gases and water soluble inorganic chemical ions of PM1 and PM2.5 at Indira Gandhi International Airport, New Delhi during 2017–18 winter, Sci. Total Environ., 729, 138800, doi:10.1016/j.scitotenv.2020.138800, 2020.
CPCB: Guidelines for Real Time Sampling & Analyses., 2011.
CPCB: Central Pollution Control Board (2020), [online] Available from: https://cpcb.nic.in/quality-assurance-quality-control/.
Van Damme, M., Whitburn, S., Clarisse, L., Clerbaux, C., Hurtmans, D. and Coheur, P.-F.: Version 2 of the IASI NH_3 neural network retrieval algorithm; near-real time and reanalysed datasets, Atmos. Meas. Tech. Discuss., 1–14, doi:10.5194/amt-2017-239, 2017.
Ghude, S. D., Chate, D. M., Jena, C., Beig, G., Kumar, R., Barth, M. C., Pfister, G. G., Fadnavis, S. and Pithani, P.: Premature mortality in India due to $PM_{2.5}$ and ozone exposure, Geophys. Res. Lett., 43(9), 4650–4658, doi:10.1002/2016GL068949, 2016.
Ghude, S. D, D. M. Chate, C. Jena, G. Beig, R. Kumar, M. C. Barth,G.G. Pfister, S.

Fadnavis, and P. P.: Premature mortality in India due to PM2.5 and ozone exposure, Geophys. Res. Lett., 1–8, doi:10.1002/2013GL058740.Received, 2016.
Ghude, S. D., Bhat, G. S., Prabhakaran, T., Jenamani, R. K., Chate, D. M., Safai, P. D., Karipot, A. K., Konwar, M., Pithani, P., Sinha, V., Rao, P. S. P., Dixit, S. A., Tiwari, S., Todekar, K., Varpe, S., Srivastava, A. K., Bisht, D. S., Murugavel, P., Ali, K., Mina, U., Dharua, M., Rao, Y. J., Padmakumari, B., Hazra, A., Nigam, N., Shende, U., Lal, D. M., Chandra, B. P., Mishra, A. K., Kumar, A., Hakkim, H., Pawar, H., Acharja, P., Kulkarni, R., Subharthi, C., Balaji, B., Varghese, M., Bera, S. and Rajeevan, M.: Winter fog experiment over the Indo-Gangetic plains of India, Curr. Sci., 112(4), doi:10.18520/cs/v112/i04/767-784, 2017.
Technical specifications for CAAQM station: Technical specifications for continuous ambient air quality monitoring (CAAQM) STATION (real time) Central Pollution Control Board East Arjun Nagar , Shahdara., 2019.
The Global Challenges Research Fund (GCRF) South Asia Nitrogen hub: The Global Challenges Research Fund (GCRF) project, [online] Available from: https://gtr.ukri.org/projects?ref=NE/S009019/.
Whitburn, S., Damme, M. Van, Clarisse, L., Bauduin, S., Heald, C. L., Hurtmans, D., Zondlo, M. A., Clerbaux, C. and Coheur, P.: A flexible and robust neural network IASI-NH_3, , 6581–6599, doi:10.1002/2016JD024828.Received, 2016.