Response to Reviewer Short Comments #SC1

We thank the reviewer for his short comments and suggestions.

Below are our replies to the reviewer's short comments.

Reviewer's short comments (*Bold Italic*) and author's response (Red font):-

General Comments:-

Paper present the comparison of the MOZART-4 model along with monthly averaged satellite distributions of ammonia emission across South Asia. The authors are trying to identify the northern region of India i.e., Indo-Gangetic Plain, IGP as a hotspot for NH_3 in Asia, both using the model and satellite observations. They highlighted a close agreement was found between yearly-averaged NH_3 total columns simulated by the model and IASI satellite measurements over the IGP, South Asia (r=0.85) and North China Plain (NCP), of East Asia (r=0.88) with a moderate correlation coefficient. Model simulated surface NH_3 concentrations and reported the under prediction with the measured surface/ground based NH_3 concentration of online pollution monitoring sites of India. The manuscript adds some new information on existing information over Indian sub-continent with model prediction which is compared with online NH_3 monitoring sits of CPCB of India. There is lot of issues/questions about the quality of the ground based data sets which is used in the comparison of model. The present study fails to establish the NH_3 emissions/scenario over Asian region due to lack of model comparison with quality controlled information of NH_3.

Reply:

We want to bring to reviewers notice that the objective of the current study is to examine the Spatio-temporal variability of atmospheric NH_3 over Asia (both South and East Asia) and focus on two hotspots regions of ammonia, the Indo-Gangetic Plain (IGP) and the North China Plain (NCP) using chemical transport modeling, satellite observations and *in-situ* ammonia measurements to understand why certain emission hotspot regions in East Asia show lower NH_3 total columns compared with similar hotspot regions in South Asia, when analyzed with both model and satellite observations. The quality control and assurance method, followed by Central Pollution Control Board (CPCB) for these air quality monitoring stations, is given in the Central Pollution Control Board (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 $\mu g/m^3$) (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 $\mu g/m^3$ 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/m3 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 time series right after the missing values. For any given day, we removed the sites from the consideration that either experience instrument malfunction and appear to be very heavily influenced by strong local sources.

Specific Comments:-

1. ***The NH3 and NOx datasets are used in the comparison of the model are taken from the online monitoring sites of the Central Pollution Control Board (CPCB), India are not quality controlled. There are a lot of issues related to calibration and validation NH3. The comparison of model should also be based on available quality controlled data sets published in a peer reviewed journals.***

   Reply
   We would like to bring to reviewers notice that we have not used nor compared the NOx data set in the present study. We have used the recent 69 stations NH_3 datasets of Central Pollution Control Board (CPCB)over South Asia and 32 stations of the Nationwide Nitrogen Deposition Monitoring Network (NNDMN) over East Asia. The quality assurance and control process followed for these air quality monitoring instruments is done as mentioned above and given in (Pollution and Board (2011), CPCB (2011),Central Pollution Control Board (2020) and Technical specifications for CAAQM station (2019)).

2. ***The instruments used in NH3 and NOx at CPCB sites are molybdenum based which converts all the gaseous nitrogen at 980oC in Nitric oxide (NO) and NOx into NO (at 350^{0} C). The difference of these two provides the NH3. Due to available moisture in the atmosphere and conversion of all gaseous nitrogen species at very high temperature it provide/estimate the high ambient NH3 concentration. Hence, for that weekly NH3 calibration is required with certified NH3 span gases.***

   Reply
   Yes, we agree with the reviewers' observations that the Conventional NH_3 measurement technique (chemiluminescence method using *molybdenum based*) overestimates ammonia measurements. This is one of the shortcomings of the chemiluminescence based measurements, which was already discussed in this manuscript. CPCB follows strict calibration of the instrument as per Pollution and Board (2011), CPCB (2011) and Technical specifications for CAAQM station (2019). Additional quality control and assurance are followed in this study is mention above.

3. ***A comparison of surface NH3 has been performed by Saraswati et al. (2019) (published in Mapan 34 (1):56-69) based on pollution monitoring sites (4 sites in Delhi) with quality controlled measurement of NH3 and reported the 2-3 times more concentration of NH3 over Delhi in compared with quality controlled data. The similar observations are reported in this manuscript in Figure 8b. In this manuscript model under predicted surface NH3 concentration. Authors are suggested to validate the model with published quality controlled datasets.***

   Reply

   Saraswati et al. (2019) carried out a comparison of NH_3 measured at NPL site in New Delhi with four other air quality measurements sites (Anand Vihar, Mandir Marg, Punjabi Bagh and R.K. Puram) in Delhi operated Pollution Control Committee (DPCC)) using a similar type of instrument and reported higher NH_3 values from other monitoring sites when compared to the reference NPL station with more than 50 % Normalised Mean Bias (NMB). However, all four sites were situated at entirely different locations from the reference site (NPL) in Saraswati et al. (2109) and may not provide the actual information on the comparison. Since NH_3 varies significantly from one location to another in Delhi, the reference site's difference partly could be due to the difference in the NH_3 measurement location. Ideally, such a comparison is made at the same location to get meaningful results.

   In order to verify the data quality, we have compared the NH_3 measurements at R.K. Puram site operated by CPCB with our own limited measurements available at IGI airport (close to R.K. Puram site) for the winter period using MARGA instrument. Our comparison shows that mean CPCB measurements are slightly on higher side than our own measurements (see below figure). The difference of 9.8 μg m^{-3}between both the measurement technique (chemiluminescence and ion chromatography (IC) based) indicates that the NH_3 measurements from the CPCB do not suffer from the calibration issue. However, rigorous validation is required in the future.

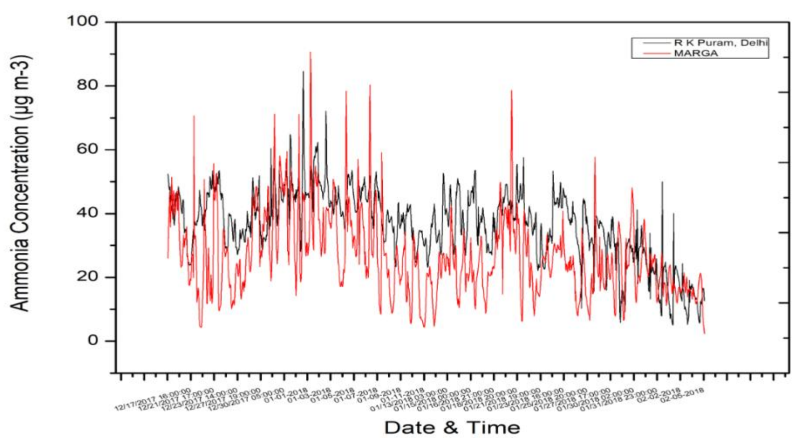

Figure: Comparison of NH_3 concentration from MARGA instrument (IGI airport) with R.K Puram (CPCB) station

4. ***Fig 4a and Fig. 6 shows the over prediction of NH3 emission by MOZART models which should be validated by quality controlled datasets. The panels are showing that sand, rocks and hillocks regions are emitting the NH3. It is sowing lack of experience/ knowledge of Indian co-authors (it seems that most of the co-authors have not hands on experience/expertise of NH3 measurements).***

   Reply

   The comparison of surface concentrations reveals that the model underestimates the ammonia observations over both South and East Asia throughout the year, which is shown by monthly mean (time series) and annual averages (scatter plot). Despite some potential calibration issues w.r.t. individual observations which are mentioned in the revised manuscript, there seems to be no obvious inconsistency with the NH_3 observations used in this study.

5. Lot of publications are available on NH3 concentration and NH3 emissions from various Indian regions. Some of them listed below which can be used for model comparison. Few papers are only sited in the manuscript.Aneja VP, Schlesinger WH, Erisman JW, Behra SN, Sharma M (2012) Reactive nitrogenemissions from crop and livestock farming in India. Atmos Environ 47:92-103 Asman WA, Sutton MA, Schjorring JK (1998) Ammonia: emission, atmospheric transport and deposition. New Phytol 139(1): 27-48 Banerjee B, Pathak H, Aggarwal PK (2002) Effects of dicyandiamide, farmyard manure and irrigation on crop yields and ammonia volatilization from an alluvial soil under a rice (Oryzasativa L.)-wheat (Triticumaestivum L.) cropping system.BiolFertil Soil 36: 207–214 Banerjee T, Singh S B,Srivastava RK (2011) Development and performance evaluation of statistical models correlating air pollutants and meteorological variables at Pantnagar, India. Atmos Res 99: 505-517 Behera SN, Betha R, Balasubramanian R (2013) Insights into chemicalcoupling among acidic gases, ammonia and secondary inorganic aerosols. Aerosol Air Qual Res 13(4): 1282-96 Behera SN, Sharma M, Aneja VP, Balasubramanian R (2013) Ammonia in the atmosphere: a review on emission sources, atmospheric chemistry and deposition on terrrestrial bodies. Environ SciPollut Res 20(11): 8092-8131 Biswas H, Chatterjee A, Mukhopadhya SK, De TK, Sen S, Jana TK (2005) Estimation of ammonia exchange at the land-ocean boundary condition of Sundarban mangrove north-est coast of Bay of Bengal, India. Atmos Environ 39: 4489-4499 Carmichael GR, Ferm M, Thongboonchoo N, Woo JH, Chan LY, Murano K, Viet PH, Mossberg C, Bala R, Boonjawat J, Upatum P, Mohan M, Adhikary SP, Shrestha AB, Pinaar JJ, Brunke EB, Chen T, Jie T, Guoan D, Peng LC, Dhiharto S, Harjanto H, Jose AM, Kimani W, Kirouane A, Lacaus J-P, Richard S, Barturen O, Cerda JC, Athayde A, Tavares T, Cotrina JS, Bilici E (2003) Measurements of sulfur dioxide, ozone and ammonia concentration in Asia, Africa and South America using passive samplers. Atmos Environ 37: 1293 – 1308 Datta A, Sharma SK, Harit RC, Kumar V, Mandal TK, Pathak H (2012) Ammonia emission from subtropical crop land area of India. Asia Pacific J AtmosSci 48 (3): 275-281 Kapoor RK, Singh G, Tiwari S (1992) Ammonia concentration vis-à-vis meteorological conditions at Delhi India. Atmos Res 28:1-9. Katyal J C, Singh B, Vlek P L G, Buresh R J (1987) Efficient nitrogen use as affected by urea application and irrigation sequence. Soil SciSoc Am J 51: 366–370. Katyal J C, Singh B, Vlek P L G, Buresh R J (1987) Efficient nitrogen use as affected by urea application and irrigation

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Reply

Thank you very much for citing some of the Indian references. We have updated some of the above relevant citations in the revised manuscript. We are aware that specific region Indian NH_3 measurements that are published in papers are available, but they have used the same chemiluminescence technique (model may differ).

In this study, we have used the global chemical transport model- MOZART4, and the domain is set to a horizontal grid resolution of 1.9° × 2.5° and driven by the same grid resolution of HTAP-V2 emission inventory. Gridded NH_3 emission data for India is not yet available, and if available, it is not available in the public domain. Specific region data cannot be used to run the model. Hence, in our study, we suggested developing India's specific NH_3 emission inventory to run the regional and global chemical transport models.

6. There are also several issues with this comparative study that needs to be taken care by Indian co-authors. They are familiar with the scenario, mainly fertilizer used and NH3 emissions from the agricultural activities. Such type of over predication/publication of NH3 emission from Indian sub-continent may create the havoc in future. We faced the problem of

CH4 emission from rice/paddy fields in India. Hence, quality controlled datasets should be used in model comparison with experimental experts.

Reply

We would like to bring to the reviewer notice that we are not addressing CH_4 emissions in this manuscript. Our intention is not to create havoc but to examine the Spatio-temporal variability of atmospheric NH_3 over Asia using chemical transport modeling, satellite observations, and *in-situ* ammonia measurements to understand why certain emission hotspot regions in East Asia show lower NH_3 total columns compared with similar hotspot regions in South Asia, when analyzed with both model and satellite observations. We have done quality control and assurance, additionally, as mention previously, before using the data. Inherent issues related to chemiluminescence are still there, which we have mentioned in the manuscript.

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