Response to Anonymous Referee #2's Comments

First of all we thank the reviewer for the positive evaluation of our study and sincerely appreciate the reviewer's insightful and helpful comments.

Below we explicitly respond to each of the items raised in the comments of anonymous referee #2. These comments are indicated in **bold**, whereas the author's response is presented in blue and revisions in red in revised manuscript.

R2C1:

Ammonia is an important short-lived pollutant with a huge global relevance for air quality, biodiversity and climate due to the wide spread food production. Improving the nitrogen use efficiency in agriculture is of key importance, which requires an understanding of the nitrogen budgets and the ability to monitor these. The atmospheric ammonia burden is difficult to model, and hence, improving our modelling capacity is an important activity. After reading the paper in detail I recommend a major revision is required to improve the paper to a level which is beyond a simple comparison between a coarse model field and observations, which is currently basically is.

A major drawback of this study is the coarse resolution the modelling is performed on. Not only in a spatial sense, also the output is available on 4 hours of the day, with IASI overpass (9:30) right in between the output times (06 and 12). The description of the comparison to the satellite data is very short. Giving the strong diurnal cycle of ammonia and the fact that the satellite data availability is affected by all kinds of factors I would like to see a much more detailed description on the method and the impacts of the choices made. - Were the monthly mean comparisons made by averaging paired observations across the month? How many valid pairs were required to allow for a valid number? If pairing was not done than a motivation/discussion why this is not important should be included. Normally the large degree of variability of ammonia column densities between days requires to pair. Satellite data availability and patterns in these within a large grid cell can also impact a non-paired comparison. How was the modelled column for 09:30 estimated? Later I read that a daily mean model value is used. . . correct? - Which quality flags of the satellite data were used? - In our experience the diurnal emission cycle largely impacts the ammonia columns at overpass. What was assumed in this study?

> Yes, we agree that we compared the monthly mean NH_3 total column from the IASI overpass in the morning (9:30) with the monthly mean model NH_3 total column averaging all 4 time-steps of the day. We also agree that the diurnal emission cycle largely impacts the ammonia columns at the overpass. To check the impacts of the diurnal cycle (driven by Boundary layer dynamics), we have again compared the monthly mean NH_3 total column from the IASI overpass in the morning (9:30) with the monthly mean model output at 11:30 LT, which near to IASI overpass (Figure 1 below). If we compare satellite and model at the nearest time-step, the Normalised Mean Bias (NMB) over IGP is reducing by 6% (with daily mean NMB=42 % and with near-time step NMB=36 %, Fig 2 (left) below), and over NCP it is increasing by

6 % (with daily mean NMB= -20 % and with near-time step NMB= -26 %) (Figure 2 (right) below). Since our model was run with the flat diurnal emissions, we have not seen any significant change compared to 4 time-step mean columns and is one of the sources of uncertainties. In the revised version, we have now compared the monthly mean NH_3 total column from the IASI overpass in the morning (9:30) with the monthly mean model output at 11:30 LT near the IASI overpass.

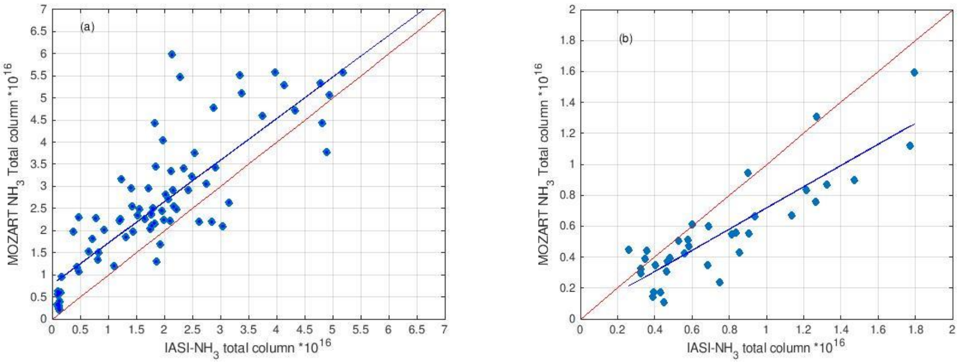

Figure 1 (a) Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH_3 ($\times 10^{16}$ molecules cm^{-2}) total columns over IGP, South Asia (rectangle: 20°N-32°N, 70°E-95°E) and (b) Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH^3 ($\times 10^{16}$ molecules cm^{-2}) total columns over NCP, East Asia (rectangle: 30°N-40°N, 110°E-120°E).

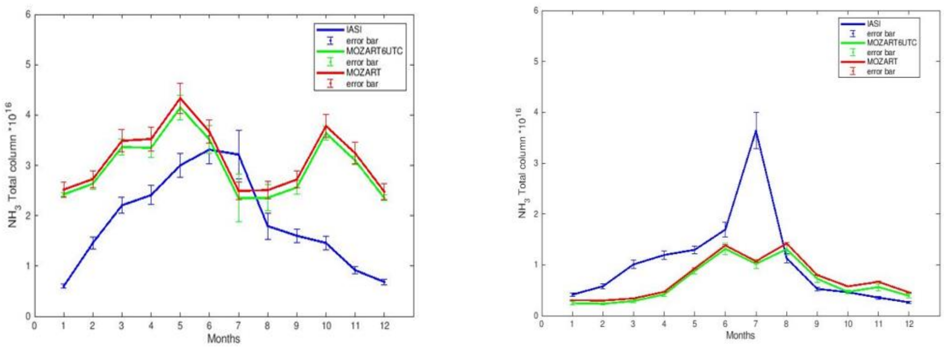

Figure 2 (left) Comparison between monthly averaged IASI and MOZART-4 simulated NH_3 ($\times 10^{16}$molecules cm^{-2}) total columns over IGP South Asia (20°N-32°N, 70°E-95°E) for daily mean (red) and near to satellite overpass (11:30, green), **(right)** Comparison between monthly averaged IASI and MOZART-4 simulated NH_3 ($\times 10^{16}$molecules cm^{-2}) total columns over NCP East Asia (30°N-40°N, 110°E-120°E) for daily mean (red) and near to satellite overpass (11:30, green)

We agree with the reviewer's comment that a large degree of variability of ammonia column densities between days requires to pair, and Satellite data availability and patterns within a large grid cell can also impact a non-paired comparison. In the present study, we are looking at monthly, seasonal and annual data. Therefore, we considered that IASI provides representative monthly, seasonal, and annual means, despite possible biases introduced by lack of days of data due to cloud cover. We compared monthly mean 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).

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. The following figures show the comparison between satellite and Model NH_3 columns on annual (Figure 3 and 4 below) and seasonal scale (Figure 5 and 6 below) calculated by averaging paired and non-paired observations. 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 pair-comparison over the NCP region than non-paired comparison (-37 %).

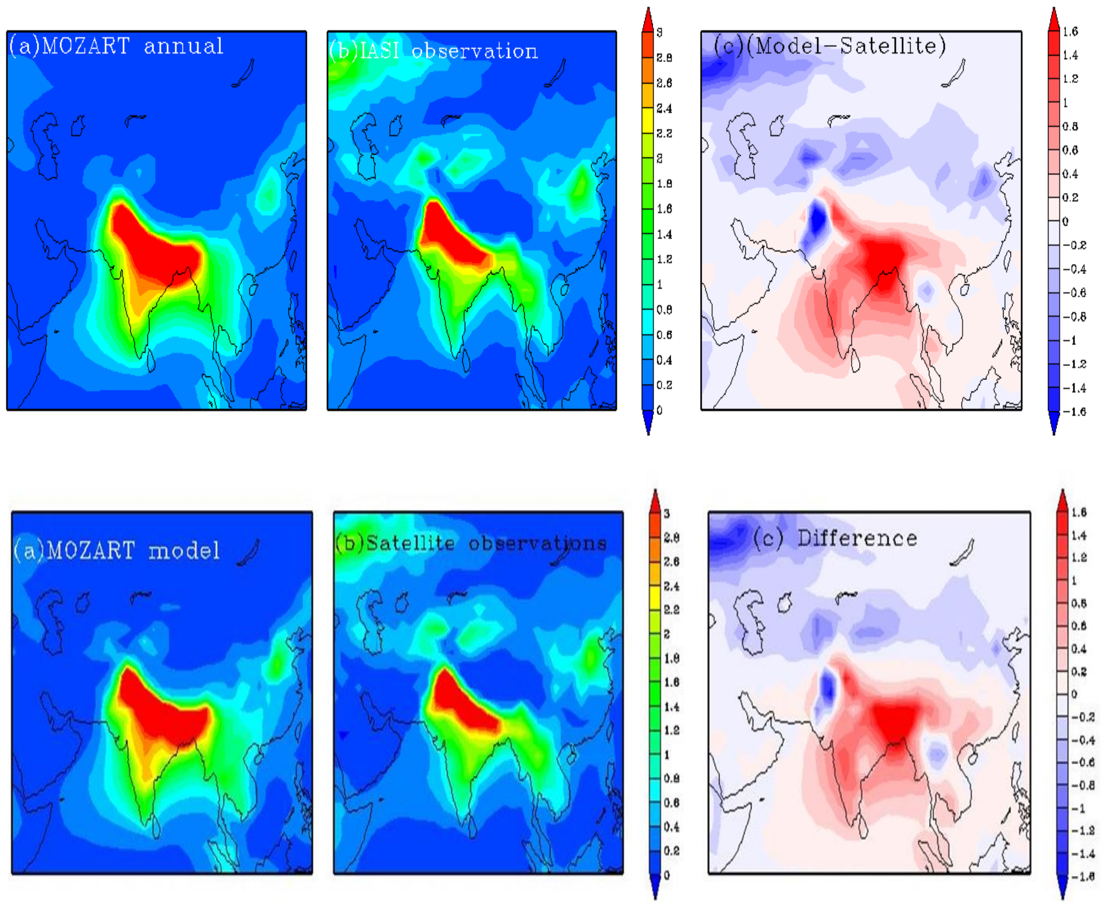

Figure 3: (top) Comparison between annual mean satellite and Model NH_3 columns calculated by averaging paired observations, **(bottom)** Comparison between annual mean satellite and Model NH_3 columns calculated by averaging non-paired observations.

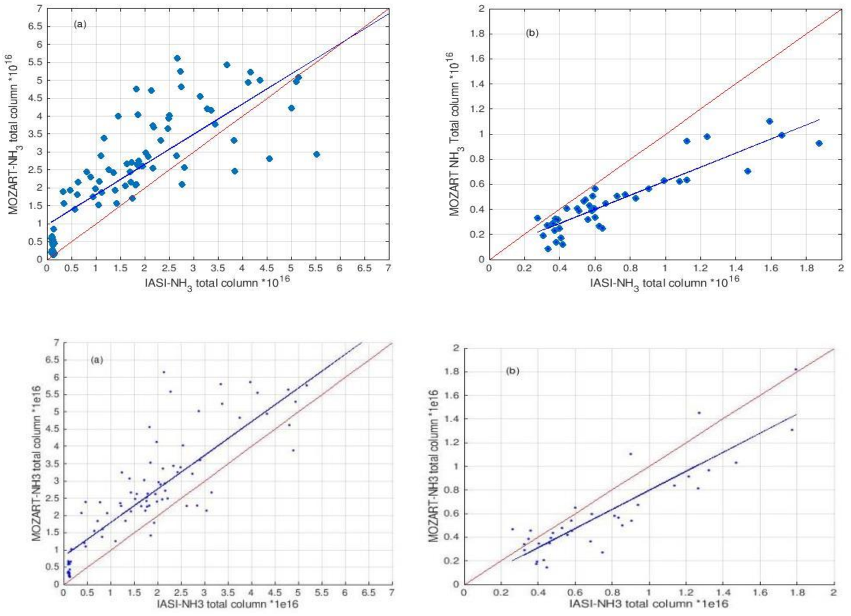

Figure 4 (top) Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH_3 total columns over IGP (left) and NCP (right) calculated by averaging **paired** observations, **(bottom)** Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH_3 total columns over IGP (left) and NCP (right) calculated by averaging **non-paired** observations

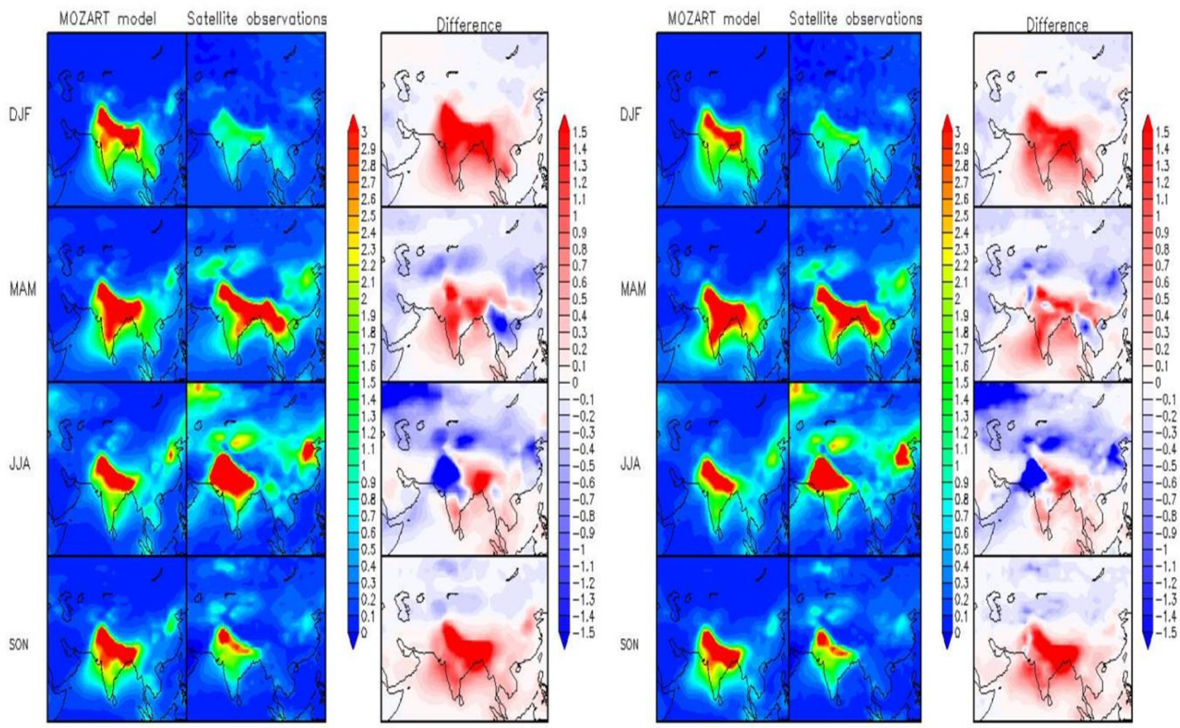

Figure 3: (left) Comparison between annual mean satellite and Model NH_3 columns calculated by averaging non-paired observations considering daily mean columns, **(right)** Comparison between annual mean satellite and Model NH_3 columns calculated by averaging paired observations close to satellite overpass time.

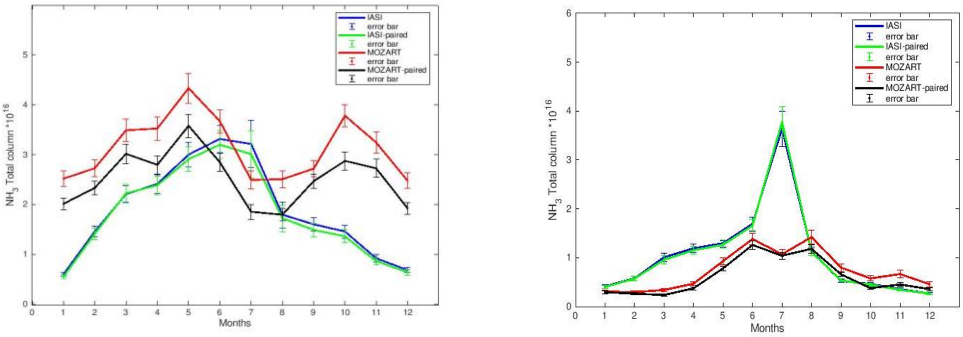

Figure 6 (left) Comparison between monthly averaged IASI (blue, non-paired) and MOZART-4 simulated NH_3 total columns for daily mean (red, non-paired) and monthly averaged IASI (paired, green) and MOZART-4 simulated NH_3 near to satellite overpass (11:30, black, paired) over IGP South Asia (20°N-32°N, 70°E-95°E), **(right)** Comparison between monthly averaged IASI (blue, non-paired) and MOZART-4 simulated NH_3 total columns for daily mean (red, non-paired) and monthly averaged IASI (paired, green) and MOZART-4 simulated NH_3 near to satellite overpass (11:30, black, paired) over NCP East Asia (30°N-40°N, 110°E-120°E).

Based on this new analysis, we have now included all the new plots (Fig. 4, Fig. 5, Fig. 6, Fig. 8) in the revised manuscript, modified Table 1, and added a detailed description of the satellite data comparison.

Further, to see the impact of finer resolution and more frequent output (1hr), we used simulated NH_3 concentration for 2011 using WRF-Chem simulation for the year 2011 from 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 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). The following figures show the comparison between satellite and Model NH_3 columns on annual (Figure 7 below) scale and its scatter (Figure 8) calculated by averaging paired and non-paired observations. It can be seen that compared to coarse simulations, the bias between the model and IASI NH_3 total columns are even larger with finer-scale simulations. We have included this for the reviewer's reference but not included in the revised manuscript. It gives a similar difference, but the magnitude of the difference is larger with WRF-Chem simulations.

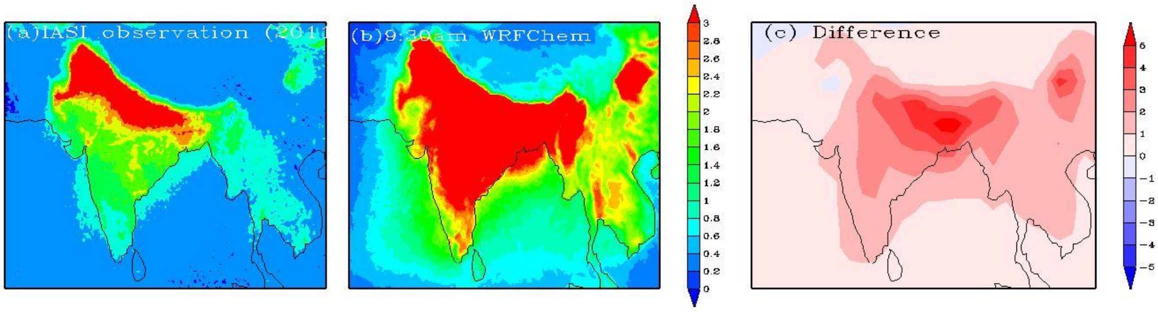

Figure 7: Comparison between annual mean IASI (left) and WRF-Chem (Middle) NH_3 columns and their difference (Right) calculated by averaging paired observations at 09:30 am on 36 km grid resolution.

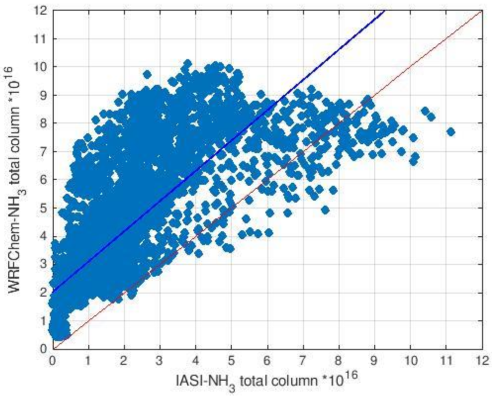

Figure 8: Scatter plot between annual averaged IASI and WRF-Chem (09:30 am) simulated NH_3 total columns over IGP.

R2C2:

Given the agricultural practices in India, is it warranted to use a flat emission cycle across the year?

We agree with the reviewer's comment. A more realistic seasonal cycle of ammonia emissions is needed for the simulations involving agriculture-based countries like India. The HTAP-V2 inventory certainly lacks this information. We aim to improve the inventory by including such a seasonal cycle for ammonia emissions in our future studies.

R2C3:

The paper is severely hampered by the coarse comparison and I am afraid that the comparison methodology may impact the systematic differences seen in this paper. The differences between overpass time and a daily mean for instance relate to the daylength (variability) and associated mixing, diurnal emission cycle, frequency and kind of precipitation events, etc. I would have like to see an analysis/consideration of such factors in this paper. Part of the observations might be useful for this purpose.

We request reviewer to refer to our reply to comment R2C1.

R2C4:

The discussion does not include a comparison to other modelling studies evaluating ammonia levels across Asia or studies on ammonia life time.

Very few studies were carried out in Asia similar to Clarisse et al. (2009), which have evaluated ammonia levels and compared model simulations with satellite retrievals. In a recent study, it is shown that higher summer-time temperature along with the higher Nitrogen (N) fertilizer application rate could cause high NH_3 emissions resulting in the high NH_3 columns over Asia, particularly during June-July-August (JJA) (Wang et al., 2020). However, satellite and model evaluation is mostly missing in this study. Studies discussing ammonia lifetime are already mentioned in the discussion part of the manuscript.

R2C5:

I could identify many grammar mistakes in the english language use. The author list includes native speakers and I would like to urge to perform a careful language check.

- A careful check for grammar has been done.

Minor comments:

R2C6:

Abstract: Please use past tense for the method description

- A careful check for grammar has been done.

R2C7:

Introduction The introduction focusses mostly on the contribution of different agricultural activities to emission estimates in south and east Asia. The challenges with respect to the emission estimation, spatial and temporal emission variability, chemistry transport modelling and model-satellite comparison are not focused on although these are relevant to the paper and partly addressed. I would like to ask the authors to address these issues in the intro.

As suggested by the reviewer, we have now added a paragraph to address the challenges for the emission estimation, spatial and temporal emission variability, and chemistry-transport modeling and model-satellite comparison in the introduction. We hope that it addresses the reviewer's concern.

R2C8:

Line 43: chemical should be synthetic

- We have replaced “chemical” with “synthetic”.

R2C9:

Line 50: 64 % of total means total global? if yes line 53 repeats this statement

No, not globally, India and China together accounted for an estimated 64 % of the total amount of NH_3 emissions in Southern Asia during 2000-2014 (Xu et al., 2018). We have now corrected this in the revised manuscript.

R2C10:

Line 60-62: could you use the recent edgar numbers or this from v4.3? Should these statements be presented with the global comparison the paragraph above?

As suggested by the reviewer, we have now provided the estimates from EDGAR v4.3.2 and included this statement where the global comparison was discussed in the introduction section in the revised manuscript. Emission estimates provided by the latest EDGAR v4.3.2 emission inventory suggests that globally about 59 Tg of NH_3 was emitted in the atmosphere in 2012, out of which agricultural soils contributed about 56 %, manure management contributed about 19 %, and agricultural burning contributed about 1.5 % (Crippa et al., 2018).

R2C11:

Line 63 and 67 are in direct contradiction to each other

Yes, we agree with the reviewer's comment that lines 63 and 67 contradict each other. In India, around 50 % of total NH_3 emissions is estimated from the fertilizer application and the remaining from livestock and other NH_3 sources. Urea is mostly used as a fertilizer and alone contributes more than 90 % of the total fertilizer used for agricultural activities (Sharma et al., 2008). We have now corrected this in the revised manuscript.

R2C12:

Data and methodology Line 85: this sentence implies only trace gases were modelled, which is not the case I guess

We have now revised the sentence in the revised manuscript.

R2C13:

Line 97: Does Mozart use a land use mosaic within a gridcell? Or dominant LUC? How do the wesely land use classes match those in the domain? Were the latter updated?

Dry deposition of gases and aerosols were calculated online according to Wesely (1989) parameterization, and wet depositions of soluble gases were calculated as described by the method of Emmons et al. (2010). Land use cover (LUC) maps used in MOZART-4 are based on the Advanced Very High-Resolution Radiometer (AVHRR) and Moderate Resolution Imaging Spectroradiometer (MODIS) data based on NCAR Community Land Model (CLM) (Oleson et al., 2010). MOZART-4 represents the land surface as a hierarchy of sub-grid types: glacier, lake, wetland, urban and vegetated land. The vegetated land is further divided into a mosaic of Plant Function Type (PFTs). These same maps are used for the dry deposition calculations (Emmons et al., 2010; Lawrence and Chase, 2007; Oleson et al., 2010). We have now included this discussion in the revised manuscript.

R2C14:

Line 122: didn't you use emissions of all sectors?

- We have used all the sectors for emissions as per the HTAP v2 emission inventory. The sectors are for all substances defined as follows:

- Air = international and domestic air,
- Shipping = international shipping,
- Energy = power industry,
- Industry = manufacturing, mining, metal, cement, chemical, solvent industry, transport = ground transport (incl. road, rail, pipeline, inland waterways),
- Residential = heating/cooling of buildings and equipment/lighting of buildings and waste treatment.
- For NH_3 there is in addition sector agriculture = agriculture (but not agricultural waste burning).
- However, for NH_3 HTAP-v2 emission inventory covers only 5 (agriculture, energy, transport, residential and industry) sectors and rest two sectors, aircraft and international shipping, is not considered for NH_3 emissions.

R2C15:
Line 133: cow dung is not fossil
Removed word “cow dung” and replaced with “biomass combustion”.

We have now corrected it in the revised manuscript.

R2C16:
Results: Line 226: the methodology describes that nitrate is present – please explain

We have corrected it in the revised manuscript.

R2C17:
250: the model has no maximum emissions in summer as antrop is flat and soil is a few percent of total, so this statement seems incorrect

We agree with the reviewer's comment that over South Asia, anthropogenic emissions are flat. Although soil emissions show some increase during summer, the percentage contribution to total emissions is small and will not affect observed NH_3 seasonal variability. We have now corrected it in the revised manuscript.

R2C18:
Figure 2: the scale on the upper left figure is misleading. It seems a seasonal cycle where it is basically flat.

As per reviewer’s suggestion, we have now revised the scale to make it consistent.

References

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