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.

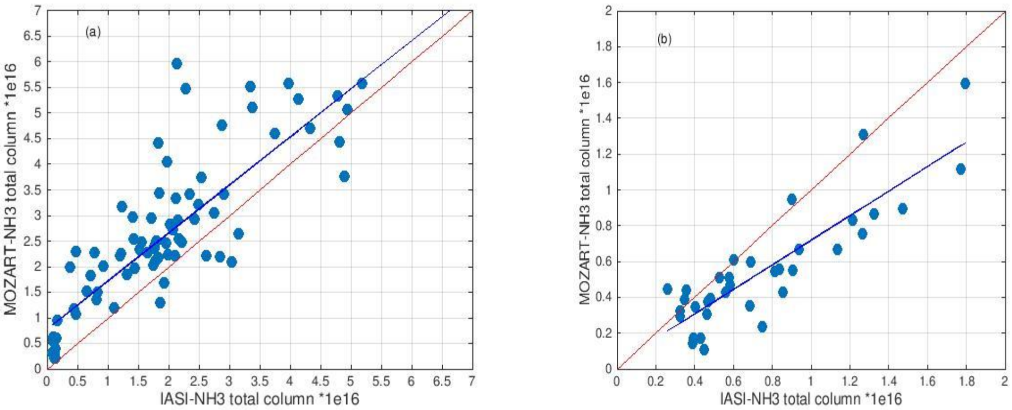
Below we explicitly respond to each of the items raised in the comments of anonymous referee #2. These comments are indicated in **bold**, whereas the author's response is presented in blue and revisions in red.

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?

- In order to add description of comparison of the model to IASI satellite, we have qualitatively compared IASI NH_3 columns with modeled NH_3 columns since the IASI-NH_3 retrieval does not produce an averaging kernel to properly weight the model values (Clarisse et al., 2009). We thus compared column to column, as IASI retrieval algorithm only provides total columns. We made un-weighted average distributions using all the morning IASI measurement available, following the recommendation for the use of the dataset provided in Van Damme et al. (2017). Furthermore, to check the impacts of diurnal cycle, we have compared the annual mean satellite overpass (9:30 am) with the nearest model timestep (11:30 am) and we have not seen any significant changes compared to 4 timestep mean columns, which we has shown in the manuscript. If we compare satellite and model at the nearest timestep, the Normalised Mean Bias (NMB) over India is reducing by 6% and over China it is increasing by 6 % (Fig 1 (a) and (b)). Our focus of the study is only on monthly, seasonal and annual data, hence we consider that IASI provides representative monthly, seasonal and annual means, despite possible biases introduced by lacking days of data due to cloud cover.



- Figure 1. (a) Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH_3 ($\times10^{16}$ 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 ($\times10^{16}$ molecules cm^{-2}) total columns over NCP, East Asia (rectangle: 30°N-40°N, 110°E-120°E).

Revision: Description added in revised version in section 2.3 and figure in the revised supplement

We compared column to column, as IASI retrieval algorithm only provides total columns. We made un-weighted average distributions using all the morning IASI measurement available, following the recommendation for the use of the dataset provided in Van Damme et al. (2017). Furthermore, considering satellite overpass (9:30 am) time with the nearest model timestep (11:30 am) does not show significant change in simulated annual mean NH_3 tropospheric column. If we compare satellite and model at the nearest timestep, the Normalised Mean Bias (NMB) over South Asia is reducing by 6 % and over East Asia it is increasing by 6 % (Fig S1 (a) and (b) in the supplement). As our aim of the study is focussed on monthly, seasonal and annual data, we consider that IASI provides representative monthly, seasonal and annual means, despite possible biases introduced by lacking days of data due to cloud cover (Van Damme et al., 2014b, 2014a, 2015). Thus, we have qualitatively compared IASI NH_3 columns with modeled NH_3 columns since the IASI-NH_3 retrieval does not produce an averaging kernel to properly weight the model values (Clarisse et al., 2009).

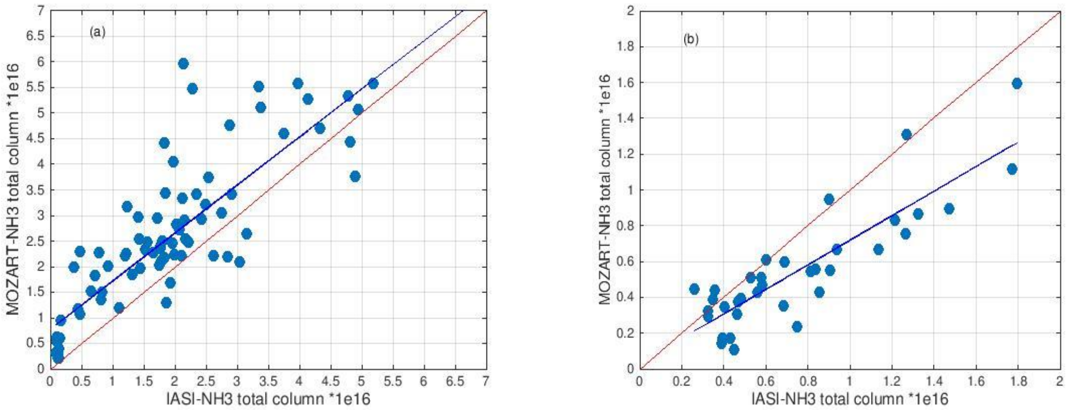


Figure S1. (a) Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH_3 ($\times 10^{16}$ molecules cm^{-2}) total columns over IGP, South Asia (rectangle: 20°N-32°N, 70°E-95°E) and (b) Scatter plot between annual averaged IASI and MOZART-4 (11:30 am) simulated NH_3 ($\times 10^{16}$ molecules cm^{-2}) total columns over NCP, East Asia (rectangle: 30°N-40°N, 110°E-120°E).

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 emissions of ammonia is needed for the simulations involving agriculture-based country like India. The EDGAR-HTAP inventory certainly lacks this information. We aim to improve the inventory by including such a seasonal cycle for emissions of ammonia 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 understand reviewer's concern with regards to the comparison methodology. We have addressed this issue in reply to the reviewer's comment R2C2.

R2C4:

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

- There are very few studies carried out in Asia similar to Clarisse et al. (2009) which have evaluated ammonia levels and compared model simulations with satellite retrievals. In 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). Studies discussing on ammonia lifetime are already mentioned in the discussion part of the manuscript.

Revision:

Higher summertime temperature along with the higher N fertilizer application rate could cause high NH_3 emissions, resulting in the high NH_3 columns over Asia (Wang et al., 2020).

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

Accepted.

Revision:

Limited availability of atmospheric ammonia (NH_3) observations, limits our understanding of control on its spatial and temporal variability and its interactions with ecosystems. Here we used the Model for Ozone and Related chemical Tracers (MOZART-4) global chemistry transport model and the Hemispheric Transport of Air Pollution version-2 (HTAP-v2) emission inventory to simulate global NH_3 distribution for the year 2010. We present a first comparison of the model with monthly averaged satellite distributions and limited ground-based observations available across South Asia. The MOZART-4 simulations over South Asia and East Asia were evaluated with the NH_3 retrievals obtained from the Infrared Atmospheric Sounding Interferometer (IASI) satellite and 69 ground-based air-quality monitoring stations across South Asia and 32 ground based monitoring stations from the Nationwide Nitrogen Deposition Monitoring Network (NNDMN) of East Asia. On the basis of model simulations and satellite observations, we identify the northern region of South Asia (Indo-Gangetic Plain, IGP) as a hotspot for NH_3 in Asia. In general, a close agreement is found between yearly-averaged NH_3 total columns simulated by the model and IASI satellite measurements over the IGP of South Asia (r=0.85) and North China Plain (NCP) of East Asia (r=0.88). However, the MOZART-4 simulated NH_3 column is seen to be substantially greater over South Asia than East Asia, as compared with the IASI retrievals, which show smaller differences. The model simulated surface NH_3 concentrations are lesser vis-a-vis the surface NH_3 measured by the ground based observation stations across South and East Asia in all the seasons, although the uncertainties prevail in the available surface NH_3 measurements. Overall, the comparison of East Asia and South Asia using both the MOZART-4 model and the satellite observations show smaller NH_3 columns in East Asia compared to South Asia for comparable emissions, indicating rapid dissipation of NH_3 due to secondary aerosol formation, which can be explained by higher emissions of acidic precursor gases in East Asia.

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.

Accepted.

Revision: Modified in the introduction section

Van Damme et al. (2015a) attempted first to validate Infrared Atmospheric Sounding Interferometer IASI-NH_3 measurements using existing independent ground-based and airborne data sets. This study doesn't include comparison of ground-based NH_3 data sets with IASI measurements particularly over South Asia (India) due to limited availability of NH_3 measurements. Liu et al. (2017a) estimated the ground-based NH_3 concentrations over East Asia, combining IASI-NH_3 columns and NH_3 profiles from MOZART-4 and validated it with forty four sites of Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN). Previous studies, based on satellite observations have suggested that the high NH_3 loading over the IGP region during summer is caused by high NH_3 emissions from intensive agricultural activities (Clarisse et al., 2009; Van Damme et al., 2015b), however validation of satellite retrievals over South Asia was largely missing. In one of the recent study over South Asia, analyses of seasonal and interannual variability of atmospheric NH_3 using IASI observations revealed large seasonal variability in atmospheric NH_3 concentrations which were equivalent with highest number of urea fertilizer plants. This study highlights the importance of role of agriculture statistics and fertilizer consumption/application in determining ammonia concentration in South Asia (Kuttippurath et al., 2020). Available global ammonia emission inventory does not include a comprehensive bottom up NH_3 emissions for South Asia compared to East Asia to be suitable for input to atmospheric models by taking into consideration actual statistical data of various NH_3 sources such as livestock excreta, fertilizer application, agricultural soil, nitrogen-fixing plants, crop residue compost, biomass burning, urine from rural populations, chemical industry, waste disposal, traffic, etc which is currently missing (Behera et al., 2013; Huang et al., 2012; Janssens-Maenhout et al., 2015; Li et al., 2017; Zhang et al., 2010). A recent study by Wang et al. (2020) examined the NH_3 column observed over the IGP during summer using regional model driven with MIX emission inventory. The study suggested that large agriculture activity and high summer temperature contributes to high NH_3 emission fluxes over IGP which leads to large total columns. However, for estimating reliable influence of NH_3 on air quality, updated emission inventory as per the source activity is essential following, which is lacking for South Asia (Han et al., 2020). Furthermore, some studies over South Asia (Datta et al., 2012; Mandal et al., 2013; Saraswati et al., 2019; Sharma et al., 2012, 2014) have reported site-specific analyses for NH_3 (ground based measurements), but are limited to a few years and scarce.

R2C8:

Line 43: chemical should be synthetic

Accepted.

Removed word "chemical" and replaced with "synthetic".

Revision:

Specifically, ammonia (NH_3) emitted from various agricultural activities, such as use of synthetic fertilizers, animal farming, etc., together with nitrogen oxides (NOx) is one of the largest sources of reactive nitrogen (Nr) emission to the atmosphere.

R2C9:

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

- No, not globally, South and East Asia together accounted for an estimated 64 % of the total amount of NH_3 emissions in the South-east part of South Asia during 2000-2014 (Xu et al., 2018).

Revision:

India and China together accounted for an estimated 64 % of the total amount of NH_3 emissions in the South-east part of Asia during 2000-2014 (Xu et al., 2018).

R2C10:

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

- Recent emission of EDGAR v4.3.2 estimated increasing emission of 59 teragram (Tg) over the period of 1970-2012 (EDGAR, 2019). While which me mentioned in the manuscript is 49.3 Tg of NH_3 estimate provided by EDGAR v4.2. Global comparison shows that EDGAR v4.3 NH_3 emission estimates are higher than v4.2 (EDGAR, 2019).

- To avoid confusion and to improve flow of the introduction part we have removed this statement which seems to be insignificant.

R2C11:

Line 63 and 67 are in direct contradiction to each other

- Line no. 63 says In India, around 50 % of total NH_3 emissions is estimated from the fertilizer application and remaining from livestock and other NH_3 sources. However, in case of fertilizer application, especially urea alone contributes more than 95 % to the fertilizer demand, consumption (Fertilizer Association of India annual report 2018-19), and more than 90 % to the NH_3 emissions (Sharma et al., 2008).
- We have removed this statement to avoid confusion.

R2C12:

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

- Yes only trace gases were modelled (NH_3, O_3, NO_x and CO), but our focus on this study is only NH_3 simulations.

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?

- 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 as used in NCAR Community Land Model (CLM). 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 dry deposition calculations. For more detail refer to Emmons et al. (2010); Oleson et al. (2010); Lawrence and Chase (2007).

R2C14:

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

- HTAP-v2 covers only 5 sectors for NH_3 emissions from agriculture, energy, transport and industry for the year 2010, rest two sectors aircraft and international shipping is not considered for NH_3 emissions.

R2C15:
Line 133: cow dung is not fossil

Accepted.

Removed word "cow dung" and replaced with "biomass combustion".

Revision:

Minor contributions from the residential sector are also observed for the Asian countries due to use of biomass combustion and coal burning which is also included in the emissions.

R2C16:

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

Yes, accepted.

Removed Line 226: In MOZART-4 chemistry, nitrate is absent and modified the statement as follows:

Revision:

In MOZART-4 chemistry, equilibrium simplified aerosol model (EQSAM)-Metzger et al. (2002) followed the assumptions which are limited to the ammonium-sulfate-nitrate-water system which is valid for only inorganic salt compounds. Since mineral cations and organic acids were neglected uncertainty can be associated in dry and wet deposition scheme which can result in overestimation (Metzger et al., 2006) (Emmons et al., 2010).

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

Accepted.

Revision:

This means that larger NH_3 emissions especially from soil may be expected in warm summer conditions than in winter, which is well represented in the emission estimate (soil) over both East and South Asia (Fig. 2). Although soil is few percent of total ammonia emissions its alkaline nature may emits higher NH_3 in the atmosphere (W. and R., 2004).

R2C18:

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

Accepted.

Revision: Figure 2 is modified

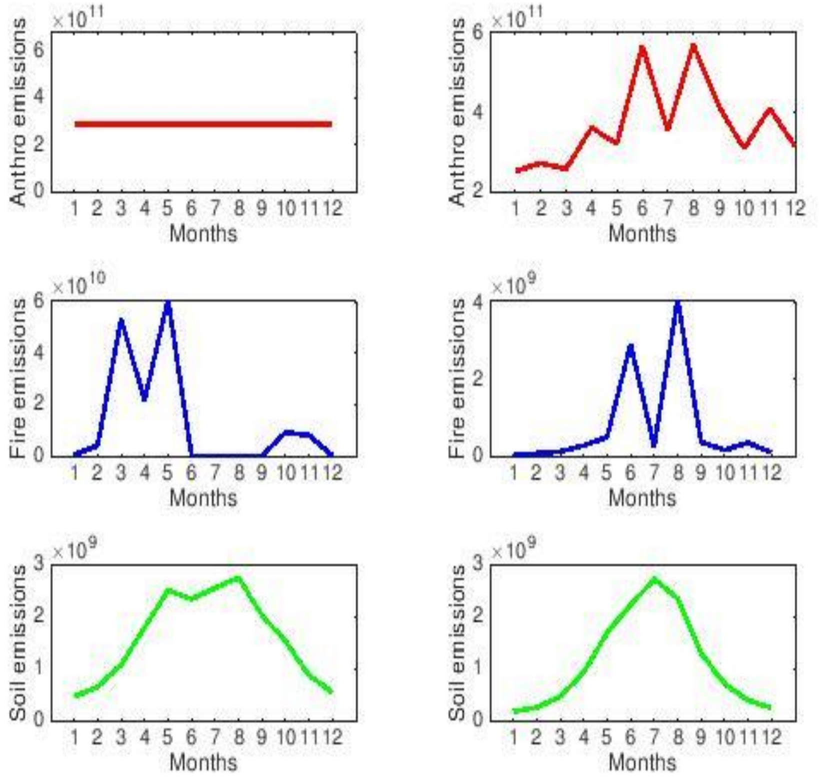


Figure 2. Monthly variation of anthropogenic (HTAP-v2) (molecules cm^{-2} s^{-1}) (top), Biomass Burning (GEFED-v3) (molecules cm^{-2} s^{-1}) (middle) and Soil (CESM) (molecules cm^{-2} s^{-1}) (bottom) NH_3 emission averaged from Indo-Gangetic plain (20°N-32°N, 70°E-95°E) and the North China Plain (30°N-40°N, 110°E-120°E).

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