

We thank the Reviewer for taking efforts to go through our manuscript and providing his/her comments. The point by-point responses (bold text) to the comments (normal text) are given below and corresponding changes made are highlighted in red color in the revised manuscript.

Comment 1: The paper describes uncertainty of modeled ozone to emission inventories of precursors generated by three different international effort. An evaluation of two chemical mechanisms MOZART and RADM-2 are also presented for one of the inventories. Results for April 2013 are presented. As presented it is a fairly unconstrained problem in terms of evaluation of the goodness of one emission field over the other purely based on ozone alone. I have tried to learn something new from the manuscript that I could have not guessed by just looking at table 1. They all have about the same NO_x and HTAP has nearly 50% more NMVOC's than the other two emissions.

Response 1: We believe that the referee made the comparison between total emissions aggregated over all regions *in the table 2* (as Table 1 is showing abbreviations/acronyms). HTAP has about 43% and SEAC4RS has about 46% higher NO_x as compared to the INTEX-B inventory. Hence the NO_x emissions are not quite the same. Additionally SEAC4RS, the newest inventory of the three, has similar NO_x levels to HTAP whereas it has similar VOC emissions as INTEX-B (the oldest inventory of the three). Considering the non-linear dependence of O₃ formation on precursors, a set of numerical experiments is necessary to assess the influence of such large differences among the inventories. This information is added in the revised manuscript (Page:5, Lines:178-184). Finally, we explicitly emphasize the region-based evaluations of simulated ozone, and the differences in NO_x emissions over regions are as high as 200% (South – INTEX-B vs. HTAP; Central – INTEX-B vs. SEAC4RS, etc.).

Comment 2: If we are in a hydrocarbon limited regions (as it seems like most of India is) then HTAP will produce more ozone. I don't see the mystery in this conclusion. Fixing emissions to get the correct answer is patently wrong in a situation like here, where there so many physical and chemical process unknowns.

Response 2: Here, reviewer is mentioning ozone formation over the Indian region as hydrocarbon-limited, which is quite contrary to what we have reported. This highlights again the importance of studies presenting numerical experiments as compared to concluding ozone production simply by comparing emission values.

Ozone production over most of the Indian region is NO_x limited in INTEX-RADM2 simulation, as shown using the CH₂O/NO_y ratio (Figure 5). This result is in agreement with a previous study using this inventory (Kumar et al 2012b). In contrast, ozone production is relatively more sensitive to VOCs in the HTAP-RADM2 and S4RS-RADM2 simulations, with significant parts of the Indian region still being NO_x limited. We suggest that our evaluation results should therefore be considered while analysing the surface ozone pollution, budget and impacts with any of the inventories or chemical mechanisms utilised in our paper over India.

We do not agree with the reviewer that many physical and chemical processes are unconstrained/unknown here. It is to be noted that the WRF-Chem model has been extensively used to successfully reproduce the meteorology and dynamics over this region. This is discussed with numerous references in the introduction section of our paper already (Page: 2-3, Lines: 69-83). For example, Kumar et al. (2012a) explicitly conclude that the meteorology is of sufficient quality to simulate the ozone chemistry over South Asia. It is to be noted that our

configuration of the model setup is based on the findings of previous studies. In addition, nudging with ERA interim reanalysis here provides constraints to the simulated meteorology/dynamics.

The suggestion of the reviewer to evaluate additional schemes for boundary layer dynamics and convection has been incorporated in the revised manuscript (see response to your comments 4 and 6).

Comment 3: It would have been very useful if we could have some figures showing comparison between observed and measured hydrocarbon. I am sure, we will get the answer that there are not any. I would suggest that the group should collect some data on NMHC's to support this analysis if that were the case. (b) Where is the evaluation of NO_x simulated at these sites? I have never seen a ozone evaluation paper that completely ignores the precursor observations and entirely based on ozone measurement.

Response 3: We agree that there is a need to conduct the measurements for precursors over this region. However this is beyond the objectives and the possibilities of the present study as described in the manuscript (Page: 3; Lines: 93—99). The evaluation of precursors would certainly provide further information about the uncertainties in the inventories and should be a recommended next step (Page:1, Line: 33-34; Page:14, Lines: 543—545), however, our conclusions assessing the simulated ozone would not be affected, which are given as follows:

(a) noontime ozone in the model significantly differs among different inventories (and also different chemical mechanisms) in contrast with the 24-h mean values, and that the current estimates of ozone impacts on human health and crop yield over South Asia have large uncertainties.

b) Ozone simulated using the SEAC4RS inventory (latest) coupled with RADM2 chemistry is in better agreement with observations making it more suitable for simulating surface ozone relative to other inventories used in the study.

We agree that there are very limited observations of precursors, nevertheless following reviewer's suggestion, we include an evaluation of modelled NO_x, ethane and ethene against recent measurements (Table C1; Table S1 in revised Supplement). Significant differences are seen in NO_x mixing ratios at Delhi, with only INTEX-RADM2 being within 1 standard deviation of the observed value. Ozone production at Delhi is VOC limited in all simulations in the present study (seen from CH₂O/NO_y ratio in Fig. 5). This indicates the importance of conducting measurements of NMVOCs in the Delhi region. At Kanpur also NO_x from INTEX-RADM2 compares better with the observed values. At Mt. Abu in the west, NO_x from HTAP-RADM2 compares better with observed values, however it should be noted that the site is also impacted by transported ozone during spring (Naja et al., 2003). At Udaipur, all simulations tend to underpredict NO_x. At Haldia in the east, NO_x from S4RS-RADM2 compares better with observed value which is also in line with the results for ozone in the east region in this study. At Nainital, modelled NO_y is evaluated and is seen to be within 1 standard deviation variability of the observed value in all simulations.

Modelled ethane mixing ratios are quite similar in all simulations and agree well with observed values at Mt. Abu but are underpredicted at Nainital by a factor of about 2. On the other hand,

modelled ethene mixing ratios at both Mt. Abu and Nainital agree relatively well with observed values in INTEX-RADM2 and S4RS-RADM2 as compared to HTAP-RADM2. The corresponding table and a small description is now added in the revised manuscript (Page: 6-7; Lines: 235-239 in the manuscript and Table S1; Section S1 on Page: 1-2 in revised supplement).

We would again like to mention that the observations of precursors are very sparse in the south Asian region and it is important to have an evaluation over a network of observations, as we present for ozone in this study, to understand their contribution into ozone formation and also the budget of NMVOCs over the region. However this does not affect the conclusions of the present study.

Table C1. Comparison of modeled monthly average (for April) precursor mixing ratios (in ppbv) with observations at several stations

Specie	Site	Reference	Observations $\pm 1 \sigma$ std	HTAP- RADM2	INTEX- RADM2	S4RS- RADM2	HTAP- MOZ
NO _x	Delhi	SAFAR data	59.8 \pm 27.5	208.7	64.4	187.2	188.9
	Kanpur	Gaur et al. (2014)	5.0	10.2	6.5	30.5	9.1
	Mt. Abu	Naja et al. (2003)/ Kumar et al (2012b)	2.1	1.7	1.1	1.1	1.4
	Udaipur	Yadav et al. (2014)	8.7 \pm 4.2	2.1	1.6	1.5	2.0
	Haldia	Purkait et al. (2008)	12.6	4.4	3.5	8.2	4.6
NO _y	Nainital	Sarangi et al. (2014)	1.8 \pm 1.6	3.2	2.7	2.9	2.6
NMVOC (ethane)	Nainital	Sarangi et al. (2016)	2.3	1.2	1.2	1.1	1.0
	Mt. Abu	Sahu and Lal (2006)	1.3	1.1	1.1	1.1	1.0
NMVOC (ethene)	Nainital	Sarangi et al. (2016)	0.9	1.2	0.9	0.8	0.9
	Mt. Abu	Sahu and Lal (2006)	0.3	0.7	0.5	0.5	0.6

Comment 4: The comparison between MOZART and RADM-2 also hinges on an unknown in the model performance over India. I have seen a few papers on WRF from India that shows huge (+/- 1000 mts or more) differences in PBL heights by just using two different PBL schemes in the model. If MOZART is producing more ozone in the upper troposphere and is getting entrained into the PBL, where is the evaluation of PBL heights or entrainment rates in the study.

Response 4: We agree that choice of PBL scheme could affect local pollutant concentration especially over complex terrains, however Singh et al. (2016) observed little impact on surface ozone and larger impact on aerosols in this season during the Ganges Valley field campaign. The usage of the MYJ PBL scheme in this study is motivated from previous studies (Kumar et al., 2012a; Ojha et al., 2016). Nevertheless, following the reviewer's suggestion we conduct a simulation using another parametrization (Yonsei University Scheme) and analyse its effect on our conclusions.

Comparison of monthly average (in April) planetary boundary layer heights between the two PBL schemes (Fig. C1; Fig. S8 in revised supplement) revealed that the differences are mostly within ± 150 m with Yonsei scheme generally resulting in higher PBL heights over India. Nevertheless, the chemical tendencies combined with vertical mixing tendencies of surface O_3 are found to be nearly similar with Yonsei scheme (Fig. C2; Fig. S9 in revised supplement) as in the base runs using the MYJ scheme (Fig. 9b in manuscript) with MOZART still producing higher ozone aloft (not shown) as in the original runs. Thus changing the PBL scheme still results in production of more ozone aloft in MOZART which is getting mixed with near surface air showing that our conclusions are not affected. This information is provided in the revised version of manuscript (Page: 10, Lines: 374-382).

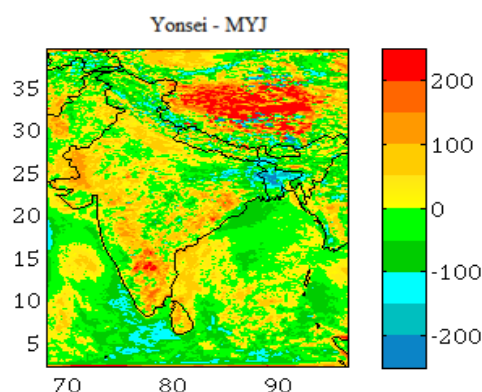


Figure C1. Difference in monthly average (in April) PBL height in meters between simulations with Yonsei and MYJ parameterization (i.e. base run) with HTAP-RADM2 setup.

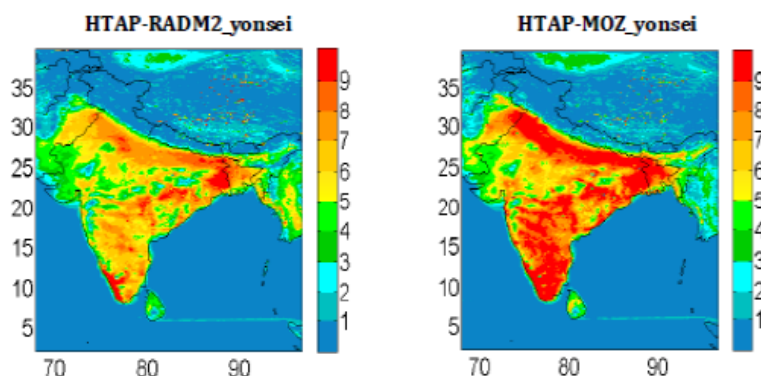


Figure C2. Average net daytime surface ozone chemical + vertical mixing tendency (in ppbv h^{-1}) for April during 0630-1230 IST for HTAP-RADM2 and HTAP-MOZ setup but with the Yonsei PBL scheme.

Comment 5: Why is MOZART producing more ozone in the upper troposphere than RADM? Is it because the photolysis rates used in RADM different than the ones used in MOZART? I am guessing the photolysis code used for both RADM and MOZART are the same – but please check.

Response 5: Because of the way the two mechanisms RADM2 and MOZART are implemented into WRF-Chem, they use different photolysis schemes: RADM2 uses the Madronich TUV or Fast-J scheme, and MOZART uses the “Fast” TUV (Madronich F-TUV) scheme, which is based

on the same physics as the Madronich TUV scheme, but designed to run faster. The differences between the two Madronich photolysis schemes is further described in the supplementary material to Mar et al. 2016.

In the present study although RADM2 uses the Fast-J photolysis scheme, a sensitivity simulation with Madronich TUV revealed similar surface ozone mixing ratios and chemical tendencies at various model levels with small differences (<5%) over most of Indian region (not shown). So our results would be similar if we use Madronich TUV scheme instead of Fast-J scheme with RADM2. Further, Mar et al. (2016) used Madronich TUV scheme with RADM2 and Madronich F-TUV scheme with MOZART chemical mechanism and reported that the two different Madronich photolysis schemes had only a small contribution to the differences in the predicted ozone by two chemical mechanisms.. The major difference between two chemical mechanisms was due to differences in inorganic reaction rates (Mar et al, 2016). Hence we conclude that in our study too, the differences over Indian region are primarily due to choice of the chemical mechanisms irrespective of photolysis scheme used. Moreover, as the aerosol radiation feedback is turned off hence the observed differences are mainly result of differing gas phase chemistry. This is discussed and clarified in the revised version (Pages: 10-11; Lines: 394-405).

Furthermore, as also discussed in Section 4.1 in the manuscript (Page: 10, Lines: 383-386), RADM2 exhibits greater VOC sensitivity than MOZART, and the higher VOC concentrations at the surface relative to aloft favour ozone production at the surface relative to aloft for RADM2. The increasing NO_x-sensitivity with increasing height results in MOZART producing more ozone in the upper troposphere in comparison to RADM2.

Comment 6: It seems like the ensemble based cloud scheme (GD) doesn't perform well over India. It has too much downward flux of air from the upper troposphere to surface. I recommend you try with a different scheme or carefully evaluate the UT/PBL fluxes in the model with observations.

Response 6: The GD scheme has been used successfully to reproduce the spatio-temporal distribution of black carbon during this season (pre-monsoon) (Kumar et al., 2015), as well as aircraft-based measurements of water vapor profiles during summer-monsoon (Ojha et al., 2016). Following the reviewer's suggestion to further strengthen our results, we now compare radiosonde observations of water vapor profiles over several stations which shows good agreement between model and observations (also see response to comment 8).

Additionally, following the reviewer's suggestion we evaluate modelled ozone using a different convection parameterization (Kain-Fritsch scheme). The differences in the modelled surface ozone mixing ratios over most of the Indian domain are found to be within $\pm 5\%$ (Figure C3; Fig. S5 in revised supplement). Relatively large differences, seen over some of the Indian region, show that Kain-Fritsch scheme tends to predict higher surface ozone mixing ratios relative to the base run (incorporating Grell 3D Ensemble Scheme) which would only add up to biases in the original runs. Therefore our conclusions remain unchanged. This is now discussed in the manuscript (Page: 7, Lines: 262-267).

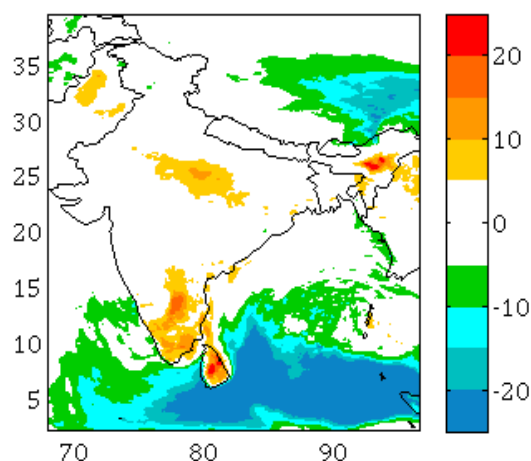


Figure C3. Percentage difference in monthly average surface ozone (ppbv) during April between S4RS-RADM2_kf run (using Kain-Fritsch convection scheme) and S4RS-RADM2 base run (using Grell 3D scheme).

Comment 7: I have also noticed that lines 130/131 probably refer to spectral nudging and not really a FDDA. Do you have or assimilated any observational meteorological data from the Indian Meteorological Department (sondes, surface weather stations etc) to perform the FDDA? Performing spectral nudging to ERA probably is not a good idea, unless you can establish that it is a good representation of synoptic scale conditions over India during this period. Many instances (specially at 12 km resolution) it is better to run the model in data poor areas with model physics than nudging the entire wind profile to ERA or any other reanalysis

Response 7: No we did not use spectral nudging. Grid analysis nudging (`grid_fdda=1`) has been used to nudge the model towards the Era interim reanalysis fields. Such nudging is shown to well represent the synoptic scale conditions over India (Kumar et al., 2012a; Ojha et al., 2016; Girach et al., 2017).

Comment 8: Have you evaluated the model synoptic scale meteorology for the simulation period with any observations?

Response 8: Numerous studies have shown that WRF-Chem reproduces the synoptic scale meteorology over the Indian region with sufficient quality for its use to drive chemical simulations (e. g. Kumar et al., 2012a). Further nudging towards the reanalysis fields limits the errors in simulated meteorology (e. g. Kumar et al., 2012a; Ojha et al., 2016; Girach et al., 2017). Nevertheless, we now include evaluation of model simulated water vapour, temperature and wind speed against radiosonde observations (Fig. C4; Supplementary material, Fig. S3). We also find that model simulated meteorology is in good agreement (within 1-standard deviation variability) with the observations. This is discussed in the revised version of the manuscript (Page: 6, Lines: 208-217).

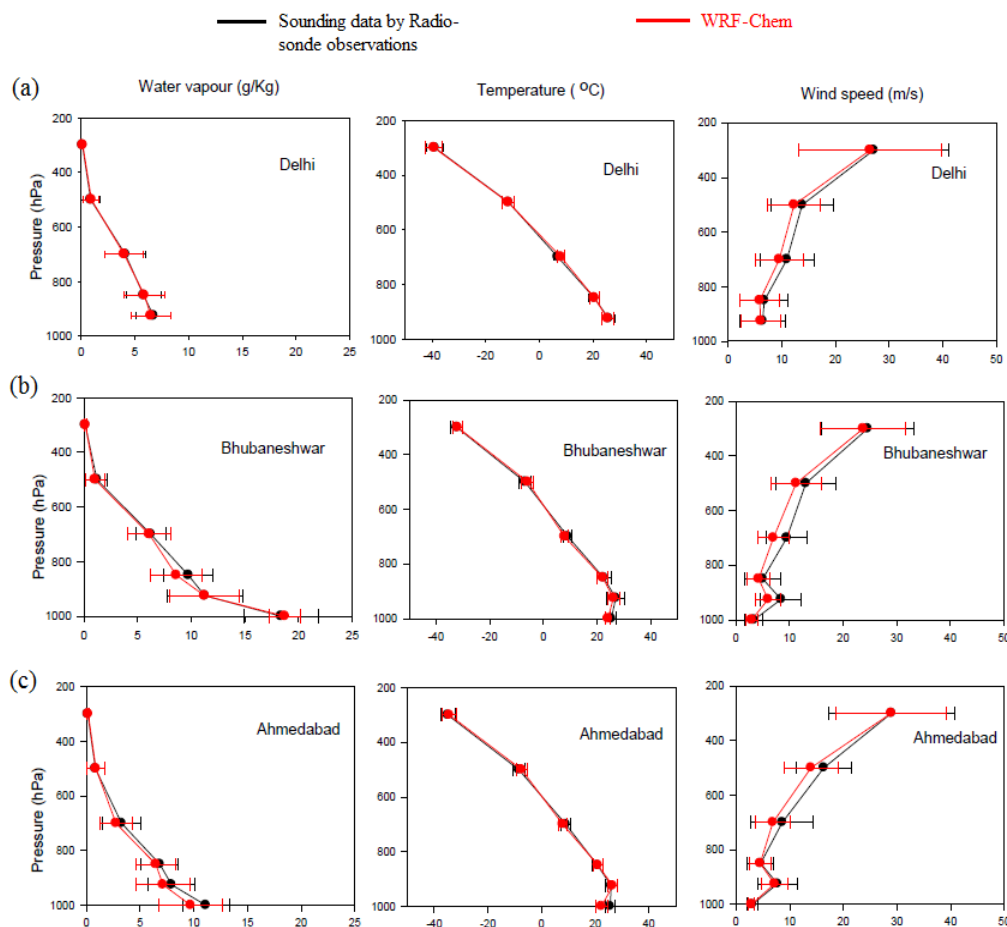


Figure C4. Vertical profiles of monthly average (April 2013) water vapour mixing ratio (g/Kg), temperature (°C) and wind speed (m/s) from WRF-Chem (in red) and sounding data (in black) at (a) Delhi (in north India); (b) Bhubaneswar (in east India); and, (c) Ahmedabad (in west India). Horizontal bars represent temporal standard deviation of monthly averages.

Comment 9: Line 85/86 cites a paper that shows the differences between simulated ozone is 4.5% with different emissions. Is the goal to improve upon that. I personally will be quite happy if you can predict ozone at less than 5% accuracy using a model.

Response 9: The cited paper is referring to “Southeast Asia”, which is the region covering the Indo-China peninsula and the Indonesian archipelago. Our objective is to investigate if over “South Asia /India” the modelled ozone is similar among different inventories or not. Interestingly we found significant differences in modelled ozone over India especially around noontime when photochemistry is most intense. Our study highlights stronger uncertainties in emissions over India causing considerable spatial-heterogeneity in the model performance in simulating ozone pollution across different south Asian regions.

Comment 10: A Taylor diagram makes lots of sense when you are trying to find out which model (or model physics) is getting close to a reference point. Emissions by themselves have no real value and improving them is not really a model issue, more of an inventory developers problem. I don’t see the point of this as the errors could be due to any number of physics or chemistry issues and not related to emissions at all. I can simply scale the HTAP emissions to a lower value and get closer to the other two emissions, that doesn’t lead to a model improvement.

Response 10: We have used the Taylor diagram to present evaluation statistics for a general overview and inter-comparison i.e. how the model reproduces the “diurnal variation” at different stations, irrespective of the emission inventory (Page:9; Lines: 327-334).

The ability to simulate diurnal variation is dependent on model performance and hence we use it to extract features of model performance instead of reporting the statistics in the form of large tables. For example, the model does not capture the diurnal variation at unresolved complex terrains, irrespective of the emission inventory used, and no scaling in inventories could improve model performance at these stations. This is clarified in the revised manuscript (Page: 9, Lines:334-336). For further details regarding the use of Taylor diagram please refer to Taylor (2001).

Comment 11: The metric CH₂O/NO_y was presented in several figures. What am I supposed to learn from this? I am guessing the RADM scheme has no methane and MOZART has methane in its chemical trace list. How is NO_y defined, does it include HNO₃? The variability you see is most likely because of different loading of NMHC from each emission. Doesn't tell much about anything in my opinion.

Response 11: An explanation for why the metric CH₂O/NO_y is a more useful diagnostic to determine ozone production regime than by simply analysing the NO_x and NMHC loadings is found in the reference of Sillman (1995). A value of 0.28 for CH₂O/NO_y ratio is suggested to be the transitional value from VOC limited regime to NO_x limited regime. This is now discussed in revised manuscript (Page: 8, Lines: 281-284). The metric CH₂O/NO_y has been successfully used as a diagnostic of chemical regime in other regional modelling studies, e.g., Kumar (2012b), Mar et al. (2016).

In the present study also the metric CH₂O/NO_y has been utilized to investigate the ozone production regime (NO_x limited, VOC limited) that could vary with changing emissions or chemical mechanism. IGP is one example where there are clear differences (Fig. 5 in the manuscript). Further, the regime also shows variability with altitude (Fig. S10 in supplement). All this information cannot be comprehended just by analyzing the NO_x/NMHCs loadings.

Regarding methane: yes, in contrast with the RADM2, MOZART has methane in the tracer list. NO_y is the summation of NO_x, HNO₃, PAN, NO₃ and N₂O₅. So yes, NO_y includes HNO₃.

Comment 12: During this time of the year the atmosphere over the central plains in India is loaded with dust. What role does dust play in the ozone production / removal?

Response 12: Dust could reduce ozone mixing ratios by influencing photolysis rates and through the heterogeneous chemistry, especially over the northern Indian stations (Kumar et al. 2014 a,b).

In the present study aerosol radiation feedback is kept switched off to investigate the effects of precursors on modelled ozone. Similar procedure had been utilised previously to compare emissions inventories for modelled ozone over the Southeast Asia (Amnuaylojaroen et al., 2014).

Further, large variabilities (500 to 6,000 Tg/yr globally) have been reported in dust emissions depending on dust parameterization in the model (Ginoux et al., 2001; Huneus et al., 2011;

Prospero et al., 2010; Textor et al., 2006; Wu and Lin, 2013; Li et al., 2017) and uptake coefficients due to its complex composition (Bauer et al., 2004; Zhang and Carmichael, 1999; Li et al., 2017)). Kumar et al (2014 a) tuned a dust parameterization in the model to match the modeled AOD with Aeronet observations for a dust storm event in the year 2010. In view of these issues, it is important to conduct extensive research to deal with uncertainties in heterogeneous chemistry related to dust loadings using multi-year observations or by strategic field experiment to provide more confidence into the dust schemes, however, this is beyond the objectives of this study.

Comment 13: The biomass burning identified has a major source of precursors also produces copious amounts of aerosols and in particular brown carbon. Brown carbon can change photolysis rates quite significantly and reduce ozone formation. How much of the disagreement is due to not accounting for these types of effects that are unique to India? We may have to fix these issues before trying to fix emissions. This only adds one more bad scientific processes to an already poor decision making in India for pollution control.

Response 13: We agree that there are factors unique to India inducing additional uncertainties in simulating the ozone production. Here we have focussed on analysing the effects of differences in anthropogenic emissions, which certainly play a major role in the ozone formation. The spatial heterogeneities in emissions are apparent in the study which makes a strong case to examine its effect on ozone estimation. Nevertheless, Jo et al. (2016) have reported that on an annual average basis, changes in surface ozone mixing ratios because of brown carbon aerosols over this part of the world (South Asia) are <5%. Again, we wish to thank the reviewer for bringing this out and further studies should be taken up to investigate the impact of brown carbon on surface ozone. This is also mentioned in the revised manuscript (Page:14; Lines: 536-541).

Comment 14: Have you evaluated the water vapor in the model during these months. Does the error in water vapor in the model explain some of the differences?

Response 14: Simulated water vapour has now been evaluated with radiosonde data (Supplementary material, Fig. S3). Model simulated water vapor is in very good agreement with the observations (within 1-standard deviation variability). As meteorology is kept unchanged in all simulations, it doesn't explain the differences.

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