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Interactive comment on "Influence of enhanced Asian NO$_X$ emissions on ozone in the Upper Troposphere and Lower Stratosphere (UTLS) in chemistry climate model simulations" *by* Chaitri Roy et al.

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Comments of Anonymous Referee #1

The study by Roy et al focuses on the radiative and dynamic impact of enhanced NOx emissions from China and India during the summer monsoon period. The increased NOx emissions are comparable to the observed increase over a 10 year period in the first decade of this millennium. This makes the study especially interesting as it describes a recent development and not a future case scenario. The results presented are interesting as NOx emissions change O3 fields in the monsoon region. The

changed O3 fields in turn affect the radiative forcing and circulation. At the end of the manuscript a connection to precipitation changes due to changed atmospheric conditions is presented. I think that the topic of the study is of interest for a large community and suitable for publication in ACP. However, there are still major and minor issues that need to be resolved before the manuscript can be considered for publication.

Reply: We thank the reviewer for their positive comments on our work. In the revised manuscript we have addressed all the major and minor issues raised by the reviewer. Changes are marked in red color.

Major Comments:

- In your study you do not consider lightning NOx. However, neglecting this NOx source in the upper troposphere could notably alter your results as the chemical background for the sensitivity studies is different. On p. 8 l. 177-181 you even mention the importance of lightning NOx during the summer monsoon season over Asia. The importance of additional NOx can be inferred from the differences in your Ind73 and Ind38 simulation. Adding lightning NOx could change the results of these simulations! Also you do not state if soil NOx is included in the simulations. This could be an additional source for NOx (see e.g. Vinken et al 2015, ACP, "Worldwide biogenic soil NOx emissions inferred from OMI NO2 observations"), if not considered already. With respect to emissions please provide a table/graphic or a section in the text summarizing the most important details of the emissions used (e.g data set, total NOx emissions per year etc.). A map plot showing the spatial distribution of NOx emissions during the monsoon season for the CTRL simulation would be useful. I personally think that giving references to previous studies only is not sufficient as the emissions are highly relevant for your study.

Ans: We are sorry for the confusion related to the inclusion of lightning NOX in our model simulations. In all simulations performed for this study, NOx emissions from lightning and soil are in fact included. We now clarify this in the text: (pg. 7, L148-155).

The difference between enhanced anthropogenic NOx emissions and CTRL simulations gives the impact of anthropogenic NOx, as NOx emissions from lightning and soil remain the same in both the simulations. In order to estimate impact of lighting NOx one needs to perform simulation with lightning-off. The difference between CTRL and lightning-off simulations shows the impact of lightning produced NOx. (pg 7, L153-155; pg. 10, L225-230)

Since focus of the current manuscript is to analyze impact of anthropogenic NOx, results from lightning-off simulations were not shown in the previous version of the manuscript. Fadnavis et al (2015) has already reported the impact of lightning NOx over the Asian region. Since lightning NOx is important during the monsoon season, we have now incorporated a figure showing impact of lightning NOx (Supplementary figure S5). As suggested by the reviewer, we have also incorporated a figure showing spatial distribution of anthropogenic emission mass flux of NOx during the monsoon season (Supplementary figure S1).

- For your CTRL and sensitivity simulations you only have 11 years (2000-2010) of data per simulation. This might be a too short time period to make robust conclusions. Please note significant differences in the plots comparing the CTRL run with the sensitivities. Showing significance will help interpreting the differences, e.g. the noisy areas such as in Fig 3 below approx. 400hPa.

Ans: We have re-plotted figure 3. Ozone production is now shown in mixing ratios instead of percentage change. Statistical significance levels of 95% is shown as contour plots.

- When showing differences compared to the CTRL simulation please use a color scale that makes it easier to see where positive or negative changes are located: this could be done using a red-white-blue color scale centered at "0" (if not centered at "0" at least include a contour indicating the "0"-line). It would be useful to show also the absolute amounts of O3/O3-production or NOx in the CTRL simulation. Otherwise percentages

can be rather meaningless.

Ans: We thank the reviewer for this suggestion and in the revised version all the figures have been changed in red-white-blue color scale centered at "0". Net ozone production (figure 3) and NOx (supplementary figure 2) are now shown in absolute amounts.

- The model set up you are using has a relatively coarse vertical vertical resolution. Please include information on the vertical resolution in Sect. 2.2 (e.g. by adding "The vertical resolution of the model set up is about x km in the UTLS")? Also, how do you expect the vertical resolution to affect your results?

Ans: We agree with the reviewer that it would have been desirable to run the model in higher horizontal and vertical resolution, but this was not possible with the available resources. We have added a supplementary figure (figure S2) depicting seasonal (June-September) mean precipitation and circulation at 850 hPa simulated in the CTRL simulation. This figure shows the general spatial pattern of precipitation and low-level circulation is well simulated (Rajeevan et al 2005). This is now mentioned in the revised manuscript (pg.7, L157-166). We now also acknowledge in the manuscript potential resolution impacts: "The accuracy of the simulation of the monsoon circulation will likely depend on the model resolution and increased vertical resolution may improve the model performance" (Druyan et al., 2008; Abhik et al., 2014) (pg.7, L160-162).

- In Sects. 3, 4 and 5 statements about cross tropopause transport by convection and consequent transport in the tropical pipe are made. In my opinion the statements are not always evident from the provided figures. Please see Specific Comments section regarding this issue.

Ans: It is now removed from the revised manuscript.

General Comments:

- I could not find any comments on how well convection is simulated by the model, please add this information e.g. by showing a map highlighting the regions of strong

convection. I know representing convection is a difficult task but at least address the possible effect of different than observed convection. Also, do the convective regions or convective activity change w.r.t the different simulations?

Ans: We have now added a figure a map of cloud droplet number concentration and ice crystal number concentration to highlight the regions of strong convection (Supplementary Figure S4(c)).

- How do VOC and CO emissions change over the period considered for the NOx emission increases? Do they remain the same? Would additional VOC and CO emissions have an impact on your results? Please add this information.

Ans: In our model simulations we have increased anthropogenic NOx emissions only. Therefore VOC and CO emissions do not change in the model perturbation experiments and therefore they do not impact our results. We have incorporated this information on pg. 7, L151-153.

- Please include some notion on the size of the Asian monsoon anticyclone. You are referring to the anticyclone several times in the manuscript and a contour showing the size would clearly help to follow your thoughts. For a 3D classificaton of the monsoon anticylcone (see e.g. Barret et al 2016, ACP, "Upper-tropospheric CO and O3 budget during the Asian summer monsoon").

Ans: Thank you for the suggestion, we now show the extent of the Asian monsoon anticyclone in Figure 1. Related discussion are incorporated in the revised manuscript (pg.8., L180-184).

- Similarly, add the contours indicating the outline/elevation of the Tibetan Plateau. This would make your argumentations easier to understand.

Ans: Thank you for this suggestion. The elevation of the Tibetan Plateau is now shown in the figures.

- Check capitalization and abbreviation: e.g. "figures" should be "Figs."

etc: See http://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html under "Manuscript composition". There are several places where this should be corrected (e.g. p 9, l 212; p 7, l 145-155,...)

Ans: The above correction regarding abbreviation has now been incorporated in the manuscript.

- Please consider to check the manuscript for grammar and spelling mistakes. I cannot vouch for the suggestions made with respect to this in the Technical Comments section. Ans: As suggested, grammar and spelling corrections have been taken care of in the revised manuscript.

Specific Comments:

Sect. 2.1.:

-You write that O3 data is convolved with the MLS AKs for comparison. However, later in the text p. 7 l. 145 you say that you use the 90hPa data from the model. Why do you not use averaging kernels also for Figure 1 a? Or are they used but it is not stated in the Figure caption!? This is confusing.

Ans: To account for the comparatively low and altitude-dependent vertical resolution, the model ozone data were convolved with the MLS ozone averaging kernel, in order to be directly compared to MLS measurements. However, Fig. 1(a) shows the spatial plot for MLS ozone at 100 hPa, which doesn't require further smoothing by averaging kernel.

Sect. 2.2:

-Please state which convection scheme is used! Although you are referring to a couple of previous studies, please mention at least the most important facts of the simulation in this section.

Ans: Thank you for the suggestion. We have now added information related to convec-

tion scheme (pg. 6-, L121-124).

Sect. 3.1:

- You show O3 profiles from 1000hPa to 10hPa. This is ok. However, I think it would be useful to make a comparison also for the relevant height section, say 500hPa to 70hPa only. This way differences are way clearer. For additional comparisons you refer to Fadnavis et al 2015, ACP. The CTRL simulation in their study includes lightning NOx! In the abstract of Fadnavis et al 2015, ACP it is stated that lightning NOx can change Ozone values by 30% and NOx by 70% in the convectively affected regions. Please comment on this issue! This also connects to the first point of the Major Comments section.

Ans: As suggested, ozone profiles are plotted on the scale 500hPa to 70hPa only and quantitative differences are now mentioned in the revised manuscript (pg,9, L195-207).

The CTRL simulation used in this study is in fact the same as in Fadnavis et al., 2015 (we mention this in the manuscript at page 6, line 141-142. As mentioned in our reply to the first major comment, lightning NOx is in fact included in all the simulations, i.e. CTRL and anthropogenic NOx emission perturbations. The impact of lightning NOx gets cancelled out when we analyze difference between CTRL and anthropogenic sensitivity simulations. In the current version we have added a figure S5 depicting lightning produced NOx. This is now made clear in the manuscript. (pg.7 L151-159). Therefore reasons for observed differences do not pertain to lightning NOx.

Sect. 3.2.:

- Please show a map plot of ICNC/CDNC to make your statement about convection over the Bay of Bengal and the southern slopes easier to follow.

Ans: Thank you for the suggestion. We have now added a map plot (supplementary figure S4(c)) of ICNC/CDNC depicting convection over the Bay of Bengal, South China Sea and the southern slopes of Himalayas.

- Please include orography in the lat/lon vs height plots. Ans: Thank you for the suggestion orography is now included in the lat/lon vs height plots.

- Due to the color scale one could think that positive anomalies in Fig. 2 (d) reach up to almost 100hPa. This is not the case. They are vertically limited to approx. 400hPa! Could it be that the color scale does not fit for Fig. 2 (d)? As I see it transport across the tropopause is only visible in Figs. 2 (c) and (f). Please comment on that or restate in the text.

Ans: The color scale has now been changed to blue-white-red (as suggested). Now it can clearly be seen that in Fig. 2 (d), positive NOx anomalies reach up to almost 100hPa ($\sim$1-4%).

- How do you attribute the signatures to convective outflow? Convective outflow could be at lower levels followed by slow upward transport to higher levels. Additionally, I think that Garny and Randel 2013 do not make a statement on transport of pollution to the deep stratosphere.

Ans: This sentence has been re-framed.

Sect. 3.3:

- The lower part of Fig. 3 looks relatively noisy please check for statistical significance or add a sentence that this should not be interpreted (see also Major Comments). You could add contours showing NOx changes (i.e. the analysis in Fig 2) to Fig. 3, so your statement in lines 212-214 is easier to follow.

Ans: We have re-plotted figure 3. Net ozone production is now expressed in ppt/day and not in percentage. We have added contour plots of 95% confidence levels to show statistical significance.

- The color scale makes it almost impossible to distinguish positive and negative differences in Figs. 4 (a) and (d) (see Major Comments section). Also I don't see real cross TP effects on O3 in Figs 4 (d)-(f). Consequent transport in the tropical pipe is only a

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hypothesis, right? Please clarify or restate.

Ans: We have re-plotted the figures in blue-white-red color scale (as suggested). This makes it easier to distinguish between positive and negative values in Figs. 4(a) and (d). Sentence related to transport in the tropical pipe is removed. We have shown arrows in Figs. 4(d) and (f), to depict cross tropopause transport of ozone (although the magnitudes are low ∼0.5-2%).

Sect. 3.4:

- The NOx maps in Fig. 5 show that at 110hPa large increases in NOx are only present in the Chin73 simulation. This fits the comment I made about cross TP transport in Sect. 3.2! Changing the color bar would be very helpful (see Major Comments section).

Ans: As suggested, the figures are re-plotted in blue-white-red color scale for better understanding. - How do you explain that in the Ind38 there is more NOx at 110hPa but less O3 than in the Ind73 simulation (Fig. 5)?

Ans: Satellite observations show a maximum in ozone precursors (CO) and low ozone in the monsoon anticyclone. The reason this is not yet known. In agreement with these observations model simulations also show more NOX and less ozone. It is now mentioned in the revised version of the manuscript. (pg. 13, L292-296).

Sect. 4:-

Why is the radiative forcing of Ind73 not stated? I think it is missing throughout the whole manuscript.

Ans: We have re-computed ozone radiative forcings using the Edwards and Slingo (1996) radiative transfer model. Figures 6 and 7 and related discussions are revised in the current version of the manuscript. The value for radiative forcing for Ind73 has now been incorporated in the revised manuscript. (pg.13,L303-304).

- Please show which regions are used to calculate the changes in radiative forcing, e.g

by including boxes in Fig. 5!

Ans: Thank you for the suggestion. The region where the changes in radiative forcing are calculated is now shown with a box in Fig. 5a. This is also mentioned in the revised manuscript (pg. 13, L303).

- Please include orography in Fig 6. This would help to follow your discussion about the TP warming in the sensitivity simulations.

Ans: As suggested orography is included in Fig 6.

- Why is there less ozone heating in Ind73 than in Ind38 although Fig. 5 (d) and (e) show that O3 anomalies are higher in the Ind73 simulation. With respect to this matter, why is the profile negative for Ind73? How does this fit with Figs 4(d)-(f)?

Ans: As mentioned above we have re-computed ozone radiative forcing using the Edwards and Slingo (1996) radiative transfer model and related discussions are rewritten.

- Why are some dots not on the lines in Fig 7(a)?

Ans: We have re-computed ozone radiative forcing using the Edwards and Slingo (1996) radiative transfer model and therefore figure 7 is re-plotted.

- To me Figs. 8 (b) and (c) look more alike than (a) and (c). In both Figs. (b) and (c) there is anomalous subsidence at approx. 20N. Also the anomalous ascent in the Ind38 is rather located at 10N, not 20N as stated in the text. Please clarify.

Ans: We have now explained figure 9 (b) and (c) as per suggestion (pg.14, L324-338).

- Clarify "Indian region" with respect to precipitation changes.

Ans: We have now defined Indian region $70°-90°$ E; $8°-35°$ N in the revised manuscript (pg.14, L330).

- Because of the open questions, I have problems to combine the results in this Sect.3 and 4 to a coherent picture.

Ans: We have re-written discussion section (section 4) and tried to produce a coherent picture. (pg. 13-15, L299-344).

Sect. 5:

- As before, you make a statement on cross tropopause transport and consequent transport via the tropical pipe that is not clear to me. See also the comments I made on this topic made before.

Ans: This statement is now removed from the revised manuscript.

Technical Comments:

p 1, l 10: change ": : : to the anticyclone." to "to the Asian summer monsoon anticyclone.". You could also add a sentence introducing the Asian monsoon anticyclone.

Ans: This sentence is re-framed pg. 1, L11-12.

p 1, l 14-16: The sentence on the emissions for the sensitivities is not precise. Maybe change to something like: "In these simulations covering the years 2000-2010 anthropogenic NOx emission have been increased by : : : with respect to the emission base year 2000. These emission increases are comparable to the observed linear trends of 3.8% and 7.3% per year during the period 2000 to 2010." Is that what you want to convey?

Ans: Above suggestion is incorporated at pg.1, L19-22.

p 1, l 19: Change ": : : are partially transported over: : :" to ": : : are partially transported to : : :", if that is what is meant by the authors Ans: This sentence is re-written.

p 1, l 22: Change ": : : produces : : :" to ": : : produce : : :"

Ans: Above suggestion is incorporated at pg.1, L23.

p 1, l 24: Change for example to ": : : India (73%) - resembling the observed increase

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over China - produces : : :". Please use present tense in the following as done in the first part of the abstract.

Ans: Above suggestion is incorporated at pg. 2, L26-27.

p 2, l 25: "It induced : : :" could be rephrased to "The higher ozone conentrations in turn induce a reversed monsoon Hadley : : :"

Ans: Above suggestion is incorporated at pg.2, L27-29.

p 2, l 26: "suppressed" should be "suppresses"

Ans: Above suggestion is incorporated at pg.2, L29.

p 2, l 27: Change ": : : the anticyclone." to ": : : the Asian summer monsoon anticyclone."

Ans: Above suggestion is incorporated at pg.2, L30

p 2, l 34: You could probably delete the etc . Ans: Above suggestion is incorporated at pg2, L37.

p 3, l 49: Change ": : :during monsoon season: : :" to ": : :during the monsoon season: : :"

Ans: Above suggestion is incorporated at pg.3, L51.

p 3, l 53: ": : :forcing increasing: : :" should be ": : :forcing due to increasing: : :"

Ans: Above suggestion is incorporated at pg.3, L55.

p 3, l 56: "and the tropopause"

Reply: Above sentence is reframed.

p 3, l 67: Just out of curiosity: Which one of the studies shows enhanced methane, ethane and N2O concentrations?

Ans: Xiong et al., (2009) have shown enhanced methane; Abad et al., (2011) has reported ethane. We have added these references in the revised manuscript. The statement related to N2O is now removed. (pg.3, L70-72).

p 4, l 74: ": : : Bay of Bengal and the Arabian Sea." Please include a reference for this statement.

Ans: This sentence was creating confusion. It is now removed in the revised manuscript.

p 4, l 74-77: "Balloonsonde observations: Thus observations show that: " Switch these sentences. Maybe cut the second "thus" in this section and change "variation" to "variations".

Ans: Above suggestion is incorporated at pg.4, L77-80.

p 4, l 90: "measurement" should be "measurements"

Ans: Above suggestion is incorporated at pg.4, L94.

p 4, l 91-92: To my knowledge MLS is an instrument onboard the Aura satellite. Please rephrase.

Ans: Above suggestion is incorporated at pg.4, L95-96.

p 5, l 119: "validation" could be changed to "evaluation", which is more accurate

Ans: Above suggestion is incorporated at pg.6, L134.

p 6, l 122: "simulation" should be "year" right? Please clarify.

Ans: Above suggestion is incorporated at pg.6, L135-137.

p 6, l 126: Add "sensitivity"

Ans: Above suggestion is incorporated at pg6, L141.

p 6, l 143 ": : :.AIRS: : :" should be ": : :MLS: : :"

Ans: Above suggestion is incorporated at pg.8, L175.

p 7, l 157: "over the various" should be "over various"

Ans: Above suggestion is incorporated at pg9, L205.

p 7, l 159: "ozone mixing ratio." should be "ozone mixing ratios."

Ans: Above suggestion is incorporated at pg.9, L207.

p 7, l 163: Add "summer" to "Asian monsoon season" to be clear

Ans: Above suggestion is incorporated at pg.9, L211.

p 8, l 169-171: "The vertical : : :" change to "Vertical distributions of NOx : : : from the CTRL simulation are : : : ". Also "figures" should be capitalized and abbreviated.

Ans: Above suggestion is incorporated at pg.9, L217; pg.10, L219.

p 8, l 172-176: In Fig. S2 I would expect number of particles per volume, i.e. môÃÃ3, as units. Could you please clarify?

Ans: Model gives cloud droplet number concentration (CDNC) and ice crystal number concentration (ICNC) in the units ' kg-1' we have converted it in mg-1.

p 8, l 174: Delete "together"

Ans: Above suggestion is incorporated at pg.10, L222.

p 8, l 184: Figs. 2 (a) and (d) only refer to the Ind38 experiment. Chin73 is shown in Figs 2 (c) and (f), right? Please change in the text.

Ans: Above suggestion is incorporated at pg.10, L233.

p 8/9, l 192/193: Change "along the descending branch of circulation" to "along the descending branch of the large scale monsoon circulation" or something like that. Please include the reference: Rodwell, M. J. and Hoskins, B. J.: "Monsoons and the dynamics of deserts", QJRMS, 1995. Here they describe the broad scale circulation with descent

in the western part of the monsoon circulation system.

Ans: Above suggestion is incorporated at pg.11, L242-243.

p 9, l 207-208: Change "estimate" to "calculate". Also add "simulation" after "CTRL".

Ans: Above suggestion is incorporated at pg.11, L250.

p 9, l 211: Add "additional" in front of "net ozone production"

Ans: Above suggestion is incorporated at pg. 11, L254.

p 9, l 214: Change to "In the Ind73: : :"

Ans: Above suggestion is incorporated.

p 10, l 222: Why should convection transport ozone upward? Usually convection is thought to bring up air with low O3 values. Are you talking about O3 from in-situ pro-duction and O3 from production in the source region which is consequently transported upward? Please clarify.

Ans: We agree with reviewer that convection is thought to bring up air with low O3 values. However we force the model with enhanced NOx emissions. Thus production of ozone increases via NOx photolysis. This increment in ozone anomalies leads to air with high 'ozone anomaly' which is transported upwards. (pg.11-12, L262-271).

p 10, l 224: Please give longitudes you associate with Arabia. Is the statement regard-ing O3 levels valid for all simulations?

Ans: As suggested we have replaced "Arabia" with broader region "west Asia" (Pg12,L269).

p 11, l 241: "estimate" should be "calculate"

Ans: This sentence is re-framed.

p 11, l 252: Change to "These figures show that the tropical easterly jet transports : :

:". Also it is clearer for O3 (Figs 5 (d)-(f)) than for NOx, right?

Ans: It is clearer for ozone (Figs 5 (d)-(f)), but is seen in case of NOx as well (Figs. 2(a)-(c)) (pg.13, L295-296).

p 11, l 264: "These simulations: : :" refers to Ind38 and Chin73 only, right? I don't see surface warming in this region for Ind73.

Ans: This sentence is re-framed.

p 12, l 276-278: In the text you write that Fig. 7(b) shows O3 heating rates over the Plateau (20N-40N), whereas in the Figure caption you state (20N-30N). Please correct.

Ans: Figure 7(b) is re plotted.

p 13, l 304: Add information that anomalies are shown!

Ans: This sentence is re-framed.

p 13, l 309: There is no "Figure 9 (e)" should be "Fig. 8(e)". Also change "(figure 8(d)) whereas they" to "(Figs. 8(d) and (f)) whereas they"

Ans: Above suggestion is incorporated at pg 14, L331-332.

p 14, l 320: "enhancement, 38% over India" could be changed to "enhancements of 38% over India "

Ans: Above suggestion is incorporated at pg. 15, L349.

p 14, l 321: Add "of" after trends

Ans: Above suggestion is incorporated at pg. 15, L350.

p 14, l 324: Change to "..that an increase: : :" or alternatively ": that increased anthropogenic: ::"

Ans: Above suggestion is incorporated at pg. 15, L355.

p 15, l 335-343: I think it would be correct to use present tense throughout this part.

Ans: Above suggestion is incorporated

p 15, l 341: "resisted" should be "impedes"

Ans: Above suggestion is incorporated at pg.16, L365.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-582/acp-2016-582-AC1-supplement.pdf

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