Response to reviewer-I

Fadnavis et al. presents analysis results to examine the temporal trends of Peroxyacetyl Nitrate (PAN) during 2002-2011 over the Asian summer monsoon region using satellite PAN measurements from MIPAS and the ECHAM5-HAMMOZ model. Overall, this is a well-designed model experiment and the analysis results are presented well. I recommend the paper be published in ACP after the following minor comments are addressed, mostly editorial.

Reply: We thank reviewer for valuable suggestions and appreciating our efforts. The suggestions made by reviewer are incorporated in the revised manuscript.

1. P19057, L17: To be more accurate, may be it is better to say "NOx-limited regime for ozone photochemical production"?
2. P19060, L11: delete "the south Asian countries"
3. P19061, L21: O1D -> O(1D), 1 should be superscript, not subscript
4. Section 3.1: The model-aircraft comparisons are hard to see. May be add some quantitative comparison information, e.g. correlation, mean bias.
5. P19064, L8: variation -> distribution
6. P19065, L8: should be "distribution of PAN"
7. P19066, L16: monsoons -> monsoon
8. P19066, L19: "The boundary layer PAN transport" -> The boundary layer lofting of PAN
9. P19068, L13: "to the ASM anticyclone"?? What in the ASM anticyclone?
10. P19069, L13: increasing trend -> an increasing trend
11. Please use "the Himalayas" throughout the text. At many places, "the" is missing or Himalaya is used instead.
12. Section 5: I wonder if it is also good to add one more subsection/figure to discuss the impact on total NOy (NOx+PAN+HNO3). This will offer information on the net impact after combining changes due to transport and wet scavenging, without worrying about NOy speciation. Just a thought.

13. P19070, L27-28. This sentence is awkward. Consider rephrase.

14. P19071, L15: Nearer -> Closer

15. Please use “the Bay of Bengal” and “the western Pacific” throughout the text. “The” is missing at many places.

16. P19073, L11: The reference to the Brewer-Dobson circulation is very vague, or perhaps inaccurate. To many atmospheric scientists, the Brewer-Dobson circulation refers to the mean atmospheric circulation, including its tropical ascending branch and the extra-tropical descending branch, etc. Please be more specific.

17. P19073, L22: “latitude-longitude cross-section of ozone” -> ozone distribution

18. Section 5.3: In addition to ozone percentage changes, you may consider add ppb changes as well to be more informative.

19. P19076, L13: “vice-a versa” -> vice versa

20. P19088, figure caption: describe which season.

21. P19090, Figure 4. I think Figure 4 is a very important figure for this study. Although the current form is ok, I think it can use some improvements for easy comparison for the readers. My suggestion is to use the same color (or same color groups) for each model-MIPAS set for ASM, CHINA, and IND, respectively. I would also suggest highlight ASM with thicker lines.

Reply(1:21): All the above suggestions are incorporated in the revised manuscript.

Response to reviewer-II

Review of "Trends in Peroxyacetyl Nitrate (PAN) in the upper troposphere and lower stratosphere over Southern Asia during the summer monsoon season: regional impacts" by Fadnavis et al.

General comments:

This paper used MIPAS PAN retrievals to examine trends in PAN in the upper troposphere and lower stratosphere (UTLS) over the Asian monsoon region (ASM) during 2002-2011. The paper does a good job in this aspect and finds that PAN concentrations in the UTLS are increasing over the ASM region and trends during the monsoon season are higher compared to the annual trends. The authors then use a global chemical transport model to understand the observed trends in PAN. The results are presented in a clear way but I have several concerns with the experimental design and some of the assertions made by the authors. Although the problem addressed by the paper is suitable for publication in ACP, I can recommend its publication only after the authors address following comments.

Reply: We thank review for valuable comments and careful reading to improve the manuscript.

Major comments:

1. The first concern is related to the selection of simulation period. Since you had computational resources to conduct multiple 10 years simulation, you could have easily simulated the MIPAS period, i.e. 2002-2011. Why did you choose the period of 1996-2004? Selecting the MIPAS period would have allowed a better model evaluation as well as source attribution.

   Reply: Apparently, the reviewer misunderstood the concept of our simulations; the model was not run with "specified dynamics", but was constrained only by sea surface temperature and sea ice concentrations. Hence, the simulations did not aim to exactly reproduce specific meteorological years, and we ran 10-year periods in order to obtain a reasonable statistics. The acquisition and handling of 10 years of highly resolved meteorological data that would have been necessary to simulate the MIPAS period with specified dynamics would have been impossible. Our study does not aim to analyze inter-annual variability, nor should it be considered a detailed trend analysis. The main point of our study is the attribution of trend

signals, and this can well be done with the model set-up which we have chosen. The study is also relevant in terms of advancing the statistical methods to evaluate chemistry-climate simulations. These simulations don't allow for case-by-case comparisons. Finally, the AMIP II Sea-surface temperature (SST) and Sea ice (SIC) used as lower boundary condition are available only until 2004 (http://www-pcmdi.llnl.gov/projects/amip/amip2/AMIP2EXPDSN/BCS/amipobs_dwnld.html).
This is now mentioned in the revised manuscript.

Like in our study, recently, Fischer et al. (2014, http://www.atmos-chem-phys.net/14/2679/2014/acp-14-2679-2014.html) compared the output of a CTM model simulation with assimilated meteorology for the year 2008 with the data during the period 1987-2008.

2. Anthropogenic emissions of trace gases and aerosols with monthly variation are now available from the MACCity inventory for the period of 1960-2020. Why the authors did not use these time varying emissions for their simulations? In my opinion, it was a great opportunity to examine whether our current understanding of trends in anthropogenic emissions can explain the observed trends in UTLS PAN or not.

   Reply: In MACCity inventory anthropogenic emission data were linearly interpolated between the base years 1990, 2000, 2005 and 2010, and they are based on the RCP8.5 scenario after the year 2000. Hence, the yearly variation and a later reference year don't add any insight into real emission changes.

3. The author themselves note that several factors are responsible for changes in UTLS PAN concentration namely changes in (i) VOC and NOx emissions, (ii) increase in the frequency of deep convection, and (iii) increase in lightning activity. However, it was not clear why did the authors focus only on disentangling the role of NOx emissions and did not try to understand the role of changes in VOC emissions and lightning activity.

   Reply: VOCs are numerous with various emission sources. Therefore it is difficult to conduct numerical experiments to understand the role of regional change in VOC in distribution of UTLS-PAN. It is now mentioned in the revised manuscript. Moreover, India and China are NOx-limited regime for ozone photochemical production. Hence chemistry of the UTLS over these regions is largely affected by changes in NO_X emissions.

The effect of the lightning activity has been studied by sensitivity experiments (1) control experiment and (2) Lightning off experiment. This is described in detail in a companion paper, Atmos. Chem. Phys. Discuss., 14, 20159–20195, 2014, doi:10.5194/acpd-14-20159-2014. However, lightning induced changes in PAN over the ASM region are already mentioned in the previous version manuscript on page 19067, lines 2-4.

The increase in frequency of deep convective clouds may cause increase in frequency of vertical transport. However, a change in UTLS-PAN due to frequency of deep convection is beyond the scope of this study. This is now mentioned in the revised manuscript.

Specific comments:

1. Section 2.1: Since this is a standalone study, you should briefly discuss MIPAS sensitivity and error. A figure showing the vertical profile of averaging kernel in Asia may help the reader to better understand the information contained in MIPAS retrievals.

   Reply: As suggested brief discussion on MIPAS sensitivity and a figure showing the vertical profile of averaging kernel over Asia is incorporated as a supplementary figure S1 in the revised manuscript.

2. Page 19063, L3-8: Please explain the rationale behind Ind38Chn38 and Ind73 simulations. These simulations do not represent realistic scenarios and do not add much value to the paper given that your objective is to understand the trends in PAN from the recent past to present day. Instead, simulations with observed increase in VOC emissions similar to simulations 2-4 would have been a great addition to the paper.

   Reply: The rationale behind Ind38Chn38 and Ind73 simulations is already explained in the manuscript see page 19063, L 1-7. As pointed out by reviewer-I these experiments are well designed and are necessary to understand the regional contribution and non linear response.

   As mentioned before, at point 3, there are number of VOCs. The major VOCs are C_2H_4, C_2H_4O, C_2H_6, C_2H_6S, C_3H_6, C_3H_6S, C_3H_8, C_5H_8, CH_3OH, higher alkanes, higher alkenes, terpenes, toluene and other minor species. Therefore simulations of the observed increase in each of them over India and China are difficult. Such Investigation goes beyond the scope of this study, because one would have to define a credible VOC speciation and its changes over time. Moreover, NOx limitation in India and China, the impact of enhanced VOC emissions on the distribution of PAN is expected to be smaller.

3. Section 3.1: Model evaluation is important to establish the credibility of model. The model evaluation results presented here in terms of comparison with aircraft measurements are not relevant to the paper as all the observations except CAIPEX lie outside of the region of interest. I would rather suggest including graphs showing the vertical profiles of model results with CAIPEX measurements. In addition, the authors can try comparing model results with OMI/GOME tropospheric column NO2 and O3 retrievals. Ozonesonde observations available from WOUDC will be another potential dataset for evaluation.

   Reply: The Asian summer monsoon is not regional phenomenon and its effects are seen all over the globe (Randel 2005, 2006, 2010; Fadnavis et al., 2013). The long range transport of pollutants during monsoon (including trace gases and aerosols) is observed by number of satellites (Vinoj et al 2014; Fu et al., 2006; Scheeren et al., 2013). The present study also shows the increase in emission over India and China affects the chemistry of other regions (see section 5 and figures 5 - 8). Therefore it is necessary to evaluate model results over the globe. Hence we evaluated aircraft measurements of PAN, and related species NO_X, O_3, HNO_3 over the globe. The vertical variations of CAIPEEX measurements are depicted in figure 1. This figure is rearranged to increase clarity.

   In this study we want to understand influence of transport due to monsoon convection on the distribution of PAN in the UTLS, hence it is important to compare vertical variation of simulated trace gases with observations. If we compare the simulated column of NO_2 and O_3 with OMI/GOME measurements, we will not evaluate their variation due to transport.

   However, as suggested, a comparison of ozonesonde measurements with model simulation is now incorporated in the revised manuscript.

   Thus we have evaluated model results from surface to the UTLS. Aircraft measurements (O_3, NO_X, HNO_3 and PAN) and ozonesonde in the troposphere and MIPAS measurements in the UTLS.

4. Page 19064, L14-15: Could you please add a line showing the location of tropopause in Fig. 2b?

Reply: As suggested tropopause is shown in figure 2b.

4. Page 19064, L16: What about the contribution of uncertainty in NOx emissions, model transport errors and coarse resolution to these differences?

Reply: Above suggestion is incorporated in the revised manuscript.

6. Page 19064, L19: I will suggest including this figure in supplementary material.

Reply: Thank you for the suggestion. Suggested figure is now provided in supplementary material (figure S4).

7. Page 19065, L3-4: Lightning NOx will also contribute to elevated levels during monsoon.

Reply: Above sentence is reframed.

8. Page 19065, L19-20: I am not convinced that these elevated levels are over the Bay of Bengal. If you look at cross sections in Fig. 3d-3f, you see that PAN lifting occurs at latitudes north of 30N which is where the Himalayas are. The latitudinal extent of the Bay of Bengal is 8-21N. In addition, the winds during monsoon blow from ocean to land and thus continental emissions cannot be transported to the Bay of Bengal which in turn means that NOx and PAN levels in the lower troposphere over the Bay of Bengal will remain low as is clear from the cross sections in Fig. 3d-f.

Reply: Thank for the careful reading. The detail analysis of data indicates that PAN transport is over India and then it is transported over the Bay of Bengal in the UTLS. Therefor above sentence is reframed.

9. Page 19067, L24: Are modelled trends smaller because you did not take into account the increase in emissions of VOCs, which probably lead to an underestimation of PAN formation rate?

Reply: Thank you for the suggestion. This sentence is re-written.

10 Page 19067, L24: Highest trends near the tropopause are seen only for India and not for China and ASM. Could you explain why highest trends over the ASM and China are seen at 10-12 km?

11. Page 19068, L13-16: If NOx is removed by wet scavenging at 12-14 km, where does NOx at 18-19 km come from?

    Reply(10 and 11): This is due to a combination of choice of domains called India and China. Indian emissions are lofted higher than the Chinese emissions by the deep convective system over the region of Nepal. Chinese emissions are also lofted by shallower convective systems. The difference in the outflow over the Pacific Ocean is an indication of this. Indian emissions get trapped in the ASM anticyclone in the upper troposphere, but Chinese emissions show a significant amount of transport over the Pacific Ocean even at 8 km. The trends diagnostic is picking up on the regional convective differences. However, a large part of the Chinese emissions are also lofted into the upper troposphere near the pathway of the Indian emissions. There is a broad maximum in PAN over general region of Nepal, Bangladesh extending to the north and east. The Bay of Bengal and the South China Sea are not involved in this pathway (we looked at slices from 8 km to 20 km). The area used to designate China in the trends analysis is large and encompasses a significant fraction of deep convection that tops out below 12 km in eastern China. The area used to designate India is smaller and also has less deep convection outside the region of Nepal. Park et al. (2009) discuss the differences between China and India in regards to transport into the UTLS and the anticyclone and this reference is discussed in the paper.

12. Page 19068, L21-22: Can you explain why it is so?

    Reply: The 73% change in emissions over China must involve larger total emissions than the 38% change over India. In this step-wise "trends" analysis one would expect more over China than over India. The difference in the lower stratosphere is due to the fact that the convective transport over India is deeper than over China. The conduit over the southern slopes of the Himalayas in the region of Nepal is distinctive and the circulation extends above the tropopause (see our previous paper Fadnavis et al., 2013).

13. Page 19068, L29: I do not agree that biomass burning activity is high during monsoon season over Asia? The authors should provide MODIS fire location maps to support their statement.

    Reply: This sentence is reframed. Here we want to say that during the monsoon season the air mass in the anticyclone is from highly polluted regions of Asia and East Asia.

14. Page 19070, L12-18: It is surprising that increase in Indian emissions by 38% does not increase PAN over India but does that over other parts of the world. One would expect at least an increase over the Himalayan region (as you see in Ind73 simulation) because of the lifting of air by towering Himalayas which is noted by the authors as well at Page 19072, L1-3. Should not the spatial pattern of Ind38 and Ind73 look the same? If not, why?

    Reply: Part of the problem is the use of percentage differences instead of vmr differences for these figures. When the PAN difference at 16 km is plotted in vmr, the anomaly is much more localized with a clear maximum in the deep convective outflow region at the base of the Himalayas (see figure 6). The difference between Ind38 and Ind73 depends on more than just emissions but also on the nonlinearity of the transport. There is feedback on the radiative transfer from aerosol modification due to changes in the chemistry. This leads to non-negligible differences in the circulation between these two ensembles. (The related discussion is now added on lines 468-486).

15. Why only Chin73 simulation show increase of more than 10% at latitudes between 45-60N? If I understand correctly, the authors relate it to dynamical changes in response to changes in NOx emissions. Does the model include effects of changes in trace gases on the meteorology? If so, it should be mentioned here and/or in the model description.

    Reply: As suggested it is now mentioned in the revised manuscript.

16. Page 19071: L17-18: But Indian emissions also increase PAN over northern India and Himalayas.

    Reply: We changed discussion in revised version. Hope it is satisfactory.

17. Page 19071, L26-27: Should not you compare Ind38Chin73 with Ind38 and Chin73 to arrive at this conclusion.

    Reply: Here we want to compare equal NO_X emission over India and china (Ind73 with Chin73). The simulation Ind38Chin73 indicates combined changes over India and china. In this simulation NO_X over India is not doubled. Hence we cannot compare Ind38Chin73 with Ind38 and Chin73 to show effect of doubling of NO_X emission over India and related response.