

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.

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.
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.
3. The author themselves note that several factors are responsible for changes in UTLS PAN concentration namely changes in (i) VOC and NO_x 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 NO_x emissions and did not try to understand the role of changes in VOC emissions and lightning activity.

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.
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.
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.
4. Page 19064, L14-15: Could you please add a line showing the location of tropopause in Fig. 2b?
5. Page 19064, L16: What about the contribution of uncertainty in NO_x emissions, model transport errors and coarse resolution to these differences?
6. Page 19064, L19: I will suggest including this figure in supplementary material.
7. Page 19065, L3-4: Lightning NO_x will also contribute to elevated levels during monsoon.
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 NO_x 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.
9. 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?

10. 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?
11. Page 19068, L13-16: If NO_x is removed by wet scavenging at 12-14 km, where does NO_x at 18-19 km come from?
12. Page 19068, L21-22: Can you explain why it is so?
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.
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?
15. Why only Chn73 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 NO_x 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.
16. Page 19071: L17-18: But Indian emissions also increase PAN over northern India and Himalayas.
17. Page 19071, L26-27: Should not you compare Ind38Chn73 with Ind38 and Chn73 to arrive at this conclusion.