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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Transport pathways of peroxyacetyl nitrate in the upper troposphere and lower stratosphere from different monsoon systems during the summer monsoon season" *by* S. Fadnavis et al.

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(1) This paper uses the global chemistry-climate model ECHAM5-HAMMOZ, PAN retrievals from MIPAS-E, and aircraft observations during the monsoon season (June-September) to evaluate the transport pathways of PAN, NOx, and HNO3 from various monsoon regions to the upper troposphere lower stratosphere (UTLS). The model results suggest that three monsoon regions - the Asian summer monsoon (ASM), the North American Monsoon (NAM), and the West African monsoon (WAM) - contribute

to pollution in the South Asian UTLS. The authors also investigate the impact of lightning NOx on the distribution of these species in the UTLS, and find that the PAN in the UTLS over the ASM region is primarily anthropogenic rather than from lightning NOx. This paper is on an important topic and the authors have extensively analyzed their results. The lightning piece of the study is a fairly straight-forward model experiment to quantitatively assess the impact of lightning NOx production on O3 and NOy species in the UTLS. However, the main part of the paper is more confused, and it is not clear to me that it presents any new results or information on the transport of PAN from the boundary layer to the UTLS, which is supposed to be the main topic of the paper.

Reply: We thank reviewers for careful reading of the manuscript and valuable suggestions. We have revised the manuscript as per suggestions given by the reviewer. The revised text in the manuscript is marked in red color.

(2) P15109 L10-15 seems to be the clearest statement of the paper's conclusions, saying that pollution from North America and Europe merges with the ASM plume in the troposphere before being lofted into the UTLS. This is a true statement, but trivially so, as it is practically a restatement of the general circulation of the atmosphere. One of the advantages of a model study is that you can use the model to determine the relative importance of different factors that are difficult to separate in observations of the real atmosphere, as in the lightning NOx study in this paper. For example, if this study had used the model to estimate the relative contribution of pollution from the US, Europe, and Asia to NOy species in the UTLS over the Asian monsoon region, that would be an interesting and publishable result.

Reply: A number of studies have documented large amount Asian pollution transport cross the tropopause (Park, 2006; Fu et al., 2006; Park et al., 2007). However transport from other monsoon systems (WAM, NAM) to Asia and UTLS have gotten less attention. Until now there has been no attempt to assess the relative contributions from these source regions and to analyze the transport patterns including possible recirculation within one consistent model framework.

As suggested emission sensitivity simulations were performed and discussions related to contribution of pollution from the US, Europe, and Asia is now added in the revised version of the manuscript.

(3) As it stands, the paper goes to a lot of effort to demonstrate a qualitative result that seems obvious to me. The MIPAS-E data could also have been used to test if the model circulation is correct, and to determine how it needed to be corrected. For example, Figures 4d and 4e show major qualitative and quantitative differences in the distribution of PAN in the UTLS between the model and the MIPAS-E observations. The model could have been used to investigate if these differences are due to incorrect emissions, chemistry, or transport and thereby provide new information on the transport of PAN. Instead, while the differences between the model and observations are extensively discussed, they are explained as likely a consequence not of errors in the model, but of sampling issues with the MIPAS-E data due to clouds in convective regions, and no effort is made to screen the model results to mimic the MIPAS-E sampling and account for this effect.

Reply: Thank you for the suggestion. Screening the model results to mimic the MIPAS-E sampling would have reduced biases. We have mentioned it in section 3.2 (Page 17, lines 356-357) and conclusion section (Page 32, lines 691-692).

(4) Again, it is not clear to me what we have learned from these model-measurement comparisons that we didn't already know before. I also don't understand why the model wasn't run for the time period corresponding to the MIPAS-E dataset. The ECHAM5-HAMMOZ simulations were run using monthly mean sea surface temperatures sea ice cover data from the years 1995-2004 with anthropogenic and biomass burning emissions for the year 2000. However, the MIPAS-E data is for 2005-2012, so that there isn't a single year of overlap between the modeled period and the observations. The authors make clear that they know that changes in emission can have a significant impact on NOy species in the UTLS, but never make clear why they didn't set up their model runs to correspond to the observations they intended to use in validation, or why

they used observations that they think have substantial sampling biases in their regions of interest.

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.

However to avoid the confusion, model simulations are now performed for the period 2000-2010 (11 years to obtain reasonable statistics), Model was simulated until 2010, as amip sst and ice are available till 2010. This period of simulation overlaps the MIPAS data period 2005-2012. The features are almost unchanged in new simulations but there is marginal improvement in biases.

(5) Finally, the paper frequently gives quantitative estimates of the model bias relative to the aircraft and MIPAS-E data but does not give sufficient information on the location that the authors are referring to. For example, P15106 L8-9 says that PAN is underestimated over North and South America in Figure 6f, without noting that there is a significant model overestimate at 30 N between 8 to 10 km in altitude. There are several other statements in the paper that need to be made more specific before they can be evaluated. Although I recognize and appreciate the substantial amount of work the authors have put into their study, I do not see how it significantly adds to our knowledge of PAN transport by monsoon convection, and I have substantial concerns about the methodology as noted above. The amount of work needed to address these concerns is more significant that can be expected in a manuscript revision, so I recommend rejecting the manuscript.

Reply: As suggested this discussion is revised (page18, lines 388-389; page 19, lies

416-417). Other text is now made more specific.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 15087, 2015.