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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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General Comments (1) This paper uses the chemistry-climate model ECHAM5-HAMMOZ to evaluate the transport pathways of PAN, NOX, and HNO3 from the surface of monsoon regions to the upper troposphere and lower stratosphere (UTLS). The impact of lightning on the production of these gases is also examined with the model. The model is evaluated with trace gas retrievals from MIPAS-E and a suite of aircraft data. The authors conclude that the Asian Sumer Monsoon (ASM), the North American

Monsoon (NAM) and the West African Monsoon (WAM) contribute substantial amounts of PAN to the UTLS, with the ASM convection penetrating the highest into the UTLS. They also conclude that lightning contributes substantially (25-80%) to the formation of PAN, NOx, HNO3, and O3 in the upper troposphere. The paper is clearly relevant to ACP and the title and abstract reflect the contents of the paper.

However, it was not clear to me that the research presented or the conclusions reached were novel – I think the authors need to make clearer how their work fits into the previous research on monsoon convection of pollution into the UTLS, and to the formation and budgets of PAN in the UTLS.

Reply: Thank you for this comment. We now explicitly highlight the novelty of our study in the introduction and conclusions. This is the first study, which compares the various transport pathways from the different monsoon regions to the ASM region within one consistent model framework.

(2) The model simulations and the comparison with MIPAS-E data appear competently done, but there are significant methodological issues as the data is never averaged in such a way to allow a consistent comparison with the model. For example, the model simulations appear to have been performed for the years 1995-2004 with constant anthropogenic (and fire?) emissions from the year 2000, but are then compared with MIPAS-E data from 2002-2011. In addition, the aircraft data is from multiple campaigns in the date range of 1984-2010, and the only comparison presented is a qualitative comparison of the average over the entire aircraft campaign with model maps in a figure (Figure 1) that is too small to read.

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. The main point of our study is understand transport pathways of PAN due to different monsoon systems and their recirculation in the lower stratosphere, and this can well be done with the model set-up which we have chosen. The long term seasonal means of MIPAS observations and model simulations are compared, since in these means typical patterns/structures are expected to be the same. It is now mentioned in the abstract. 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).
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.

(3) Finally, the presentation of the paper is not clear, with the color and vertical scales of different panels within a figure changing, making it difficult to confirm the discussion of the results. Thus, I feel the paper needs major revisions before it can be accepted for publication, or even reviewed adequately. My specific concerns are noted below.

Reply: Thank you for the suggestion, the presentation of figures is changed. The color and vertical scale is adjusted as per suggestion.

Specific Comments:

(4) P20161, L25-26: This conclusion doesn't necessarily follow from your evidence – the lower percentage changes with lightning over the ASM could just indicate less lightning. Can you rule that out?

Reply: It is true that there is less lightening and hence less associated NOx and PAN production. But Figure 3(b) indicates that there is more transport from the surface to the UTLS compared to the Asian summer monsoon convection than North American Monsoon (NAM) and West African monsoon (WAM). Over the WAM region there are higher values of PAN at 8 km compared to the ASM, but there is much less below 5 km. At the same time the values of PAN above 8 km over the ASM are not consistent with the much smaller amounts of lightning NOx in this region compared to the WAM. In addition, there are higher values of PAN above 12 km over the ASM, which indicates that PAN is undergoing deep convective transport in this region. The only conceivable source for the PAN maximum over the ASM is anthropogenic pollution and it is much higher compared to the WAM region.

(5) Introduction: As mentioned above, I'm not clear on what is unique or novel in this research, mainly because that context isn't clear in the introduction. Consider rewriting it to make what is unique and valuable about your study clear. Has the monsoon transport of PAN been looked at in previous modeling studies, but not with ECHAM5-HAMMOZ? Has ECHAM5-HAMMOZ not been compared with MIPAS-E PAN data before? Is it the aircraft analysis that is new? And on the lightning impact of PAN, your result isn't appreciably different from your cited estimate of Tie et al. (2001) – is there some other way your analysis improves upon theirs?

Reply: The novelty of this study is now highlighted.

(6) P20162, L25-28 and P20163, L1-2: This paragraph needs some work. First, not all NMVOCs are equally efficient at forming PAN, so you might want to mention some of the key precursors that produce acetyl peroxy radicals during their oxidation. Second, instead of simply listing source sections it would be more valuable to discuss sources of NMVOCs and NOx in the regions that can be convectively lofted by the monsoons, and what the relative magnitudes of the sources are.

Reply: As suggested this paragraph is rewritten. Information about PAN precursors, production and loss processes are now described in the introduction. The description of the relative importance of each single emission source and single NMVOC species would require a long discussion in the introduction, which we feel would not pertain the scope of this study. Nevertheless we provided in the introduction a general description of main emissions of PAN precursors, while in the model description (section 2.2) we provide a more detailed description of the emission inventories and relative importance of anthropogenic and natural emissions of PAN precursors.

(7) P20163, L22-23: There has to be a literature reference for this campaign, right? If so, add it here in addition to the website.

Reply: As suggested references for each of the campaign is added in table 1.

P20164, L24-26: You should include some details on the PAN retrievals in the text, rather than just giving a reference. What is the estimated and actual precision of the retrieval? What validation studies have been done? What are the known biases? Are there any trends in bias or sensitivity with latitude, longitude, or altitude that might affect your study?

Reply: Some details of the PAN retrievals are now included in the text, together with a short discussion of the estimated errors and their sources. There is no validation of the MIPAS PAN data set yet. Therefore the knowledge about biases, trends, sensitivities is rudimental. No major inconsistencies/problems with the data are known to date, however. Since we are mainly interested in climatological means, and especially in the transport patterns/structures, we feel that the use of the data is justified.

(8) P20165, L4-5: What is the reference for “the documentation”? What quality flags were actually used?

Reply: The text has been changed; the documentation is now referenced explicitly. A short note on the actual quality flags are is added.

(9) P20165, L5-6: This height range was specified by who? How was it determined?

Does it only apply to PAN or all retrieved species?

Reply: The height range actually used is 8-23 km. It is specified by the retrieval scientists. This restriction is based on features of the rows of the averaging kernel matrix of PAN retrievals, as it is now explained in the text and shown with two examples (given as supplementary figures). Note that here we only use PAN retrievals of MIPAS, so the height restrictions for other species are of no concern.

(10) P20165, L6-7: Why is the MIPAS-E data averaged to 4x8 degrees when the model fields are at 2.8x2.8 degrees? Why didn’t you regrid the data to match the model grid?

Reply: The model simulations were performed at a spectral resolution of T42. The true resolution is 180/42=4.3 degrees. The nonlinear spectral transform grid in this GCM and all the other spectral GCMs is designed to minimize aliasing by increasing the number of modes by 50%. Therefore now model output is re-gridded to 4x8 in order to compare with MIPAS.

We performed some test runs to decide gridding of MIPAS data. The number of contributors are less at lower resolution (visible in lat-lon cross sections of PAN). The standard error increases if we reduce the grid size less than 4x8 degrees. The longitude-altitude distributions of (1) no of contributions, (2) relative standard error and (3) PAN vmr averaged for the period 2002-2011 and latitudes 0-25N, when gridded for lon=2.8, lon=4 and lon=8 are shown in attached figure-1.pdf "Figure for response to reviewer-I".

(11) P20165, L17-27: This is the sort of information on PAN formation that should be in the introduction – consider moving it up there.

Reply: Thank you for the suggestion. It is moved in the introduction section.

(12) P20166, L10-18: This model section needs a lot more detail. It isn’t sufficient to just reference other papers detailing the model parameterizations, emissions, and validation– that should all be briefly restated here.

Reply: The model description has been improved.

(13) The use of the year 2000 emissions for the entire period from 1995-2004 needs justification as well – didn't emissions of NOx change dramatically in terms of their geographic distribution in that period? Also, I'm assuming you didn't use year 2002 fire emissions for all years, correct? Finally, why did you choose to model the period 1995-2004 when the MIPAS-E data is for 2002-2011? Why should we expect the two to be consistent with each other when the time periods don't match?

Reply: Similarly to our answer to point (2), the simulations did not aim to exactly reproduce specific meteorological years or to analyze inter-annual variability of PAN concentrations and related fields, but to look at transport pathways of PAN in different monsoon systems. For this reason we ran 10-year periods in order to obtain a reasonable statistics of transport pathways, independently from the geographical and temporal variability of anthropogenic and biomass burning emission for the considered period. The long term seasonal means of MIPAS observations and model simulations are compared, since in these means typical patterns/structures associated to monsoon systems are expected to be the same, even for different time periods. We selected the period 1995-2004 for model simulations according to the availability of the AMIP II Sea-surface temperature (SST) and Sea ice (SIC) used as lower boundary condition are available only until 2004.

(14) P20166, L22-23: "the mass of atomic nitrogen produced as NO" is a little confusing –did you use the molecular weight of N or NO? Why not just express it in moles NOx produced/flash?

Reply: The sentence was removed and included in another sentence of the model description: "Lightning NOx emissions are parameterized following Grewe et al. [2001], and are proportional to the calculated flash frequency with a production of 9 kg(N) per flash, and distributed vertically using a C-shaped profile. Lightning frequency is brought to a value that results in a global emission between 3 and 4 Tg(N) per year (Rast et al., 2014)."

(15) P20167, L3-11: This whole paragraph should be in the methods section as it describes the aircraft data used for the validation. Aren't there better references for the data than the website? Why are you using data from 1984 to 2010 to evaluate a model simulation for 1995-2004? Also, why are the aircraft data averaged to the center latitude and longitude of their flight region? Shouldn't all points within a model grid box be averaged together to allow a one-to-one comparison?

Reply: Thank you for the suggestion; paragraph is shifted in method section. The reference for each of the campaign is mentioned.

As mentioned in reply 3, the simulations did not aim to exactly reproduce specific meteorological years, and we ran 10-year periods in order to obtain a reasonable statistics. These simulations don't allow for case-by-case comparisons. However, the simulated long term seasonal mean is compared with trace gas retrievals from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS-E) and aircraft campaigns during the monsoon season (June-September), since in these means typical patterns are expected to be the same. As suggested the Aircraft observations are averaged vertically and horizontally over the coherent regions.

(16) P20167, L12-19: Figure 1 is so small it is nearly impossible to tell if the claim of good agreement between the aircraft data and the model is true. Can you give any quantitative details on the comparisons (mean bias, rms errors, etc.)?

Reply: As suggested, figure 1 is rearranged to increase clarity. Figure showing mean bias is now added as a supplementary figure 2.

(17) P20167, L24: I don't see in the text where you discuss the 16 km results.

Reply: As suggested in comment 31, figure is now replaced with figures showing PAN distribution averaged at altitudes near tropopause (14-16km). Therefore text is modified accordingly.

(18) P20168, L1-2: It's not clear here that you are talking about only three maxima, as

you mention five locations. Consider revising.

Reply: As suggested above sentence is reframed.

(19) P20168, L8-9: What is your evidence for your statement that the outflow of biomass burning smoke is underestimated in the model, and that is the cause of the error in the location of the PAN maxima? Is it the Real et al (2010) study? If so, that wasn't clear.

Reply: Thank you for the suggestion. The figure showing pan distribution over the biomass burning region of Africa (∼30S-20ON; 20W-30OE) as obtained from MIPAS and ECHAM5-HAMMOZ simulations is shown in figure S3. The difference between MIPAS and ECHAM5-HAMMOZ simulated PAN indicates that biomass burning smoke is underestimated in the model.

(20) P20168, L11-14: You mention a lot of potential explanations for the error in the location of the PAN maxima here – can you rule any of these explanations out based on your study?

Reply: All of these factors are playing a role to a greater or lesser degree so it is not possible to rule any of them out completely. The most important factors for the difference between model and observations are transport and emissions.

(21) P20168, L14-15: Anthropogenic emissions of what are underestimated?

Reply: NOx and the text is now corrected in the revised manuscript.

(22) P20168, L15-20: Where are the plots of the results from 8-23 km? We only have the maps at 14 km and 16 km. I also don't understand this discussion at all. How can the MIPAS-E PAN be 70-90 ppt higher than the model when Figure 2 shows PAN concentrations of 0-60 ppt in both the model and the measurements south of 30 S? I also don't see the evidence for the statement that in the UTLS MIPAS-E PAN is 20-60 ppt higher in the model, especially since that seems to vary with latitude and longitude.

Reply: It is not possible to show comparison of latitude-longitude variation of PAN observed by MIPAS with model simulations at every altitude hence we have given the average values over the altitude range of 8-23Km. To make it clear we have now added a supplementary figure 4 to show differences at 4 representative altitudes 13km, 15km, 17km and 20km. The latitude-altitude and longitude-altitude cross-section of differences of MIPAS-E and model are also shown in figures 3 and 5.

(23) P20168, L23-26: This speaks to the bias in the MIPAS-E retrievals, and so belongs in Section 2.1.

Reply: The work of Tereszchuk et al. is based on a data set different to the one used for the comparison with the model. To avoid confusion we removed this part of the text.

(24) P20169, L4-6: Is there a reference for this statement?

Reply: It is a conclusion based on the MIPAS and model results shown in Figure 2. There is no reference necessary and we are not aware of any papers discussing this aspect.

(25) P20169, L9-27: In general, it isn't clear in these sections when you are analyzing the model, and when you are analyzing the MIPAS-E data.

Reply: It is rewritten now.

(26) P20170, L14-15: I don't understand how low PV can confirm that there is tropospheric air in the lower stratosphere.

Reply: This statement is now removed. PV is a quasi-conservative tracer. In the troposphere, the values of PV are usually low. The potential vorticity increases rapidly from the troposphere to the stratosphere due to the significant change of the static stability. Hence low PV values at 370K level in the region 0-30ON indicate lower tropospheric air in the upper troposphere. The discussion related to the near-equatorial convective heating is now added in the revised manuscript.

(27) P20171, L2-4: What "sampling issues" could obscure the three plume structure? And isn't the coarse resolution of the MIPAS-E data due to your overly-coarse regridding?

Reply: The ASM convection at the southern flanks of the Himalayas is amongst the most intense in the world. It is persistent during the summer monsoon season as is the high altitude cloud cover. So the frequency of observations in this region is actually lower than over the WAM. This bias is reflected in the structure of the PAN which has peak values below 10 km, corresponding to periods when convection is not as deep and much less organized (particularly in the region between 90E and 100E). MIPAS has minimum PAN values above 11 km where the model has maximum PAN values. This corresponds to the location of very deep convection and it is unphysical for there to be a minimum to be there given the current knowledge of PAN chemistry and convective transport.

(28) P20174, L11-12: I don't understand – the PAN is transported from Africa to the Americas and then the Americas to Africa? Can you make this clearer?

Reply: This sentence is elaborated for clarification.

P20176, L1: You don't mean heterogeneous (e.g., gas-particle) reactions here, right? And doesn't the lightning only produce NOx, and the HNO3 and O3 comes from subsequent chemistry?

Reply: Above sentence is reframed.

(29) Table 1: You need to include references for each campaign listed, and there is a typo in the data for CAIPEEX.

Reply: Above mentioned suggestion in incorporated in the revised manuscript.

(30) Figure 1: These figures are way to small to be able to identify the data points and how they differ from the model. They should either be split into 3 figures by altitude range or four figures by species so that they are readable.

Reply: Thank you for the suggestion. Figure 1 is rearranged.

(31) Figure 2: Why are you plotting these heights? Why not do some average comparison within 2 km of the tropopause? Can you plot a map of the MIPAS/ECHAM-HAMMOZ differences to make your discussion clearer? What does the color scale value below 0 mean – missing data? If so, can you make that white or gray so it is more obvious?

Reply: Thank you for the suggestion. Figure 2 is modified. The differences are shown in a supplementary figure S2. The color scales is now changed and missing data is indicated with white color.

(32) Figure 3 and 5: These plots are very confusing, as Figures 3(c) and 3(f) have different vertical and color scales than the other plots, making it difficult to confirm that they are showing the same model output as Figures 3(b) and 3(e), respectively. I understand that you want to plot the model from the surface to 23 km, but why not just plot the MIPAS-E data the same way, using white to denote areas with no data? Then the extra panel could be used for a difference plot. Reply: As suggested by the reviewer, Figures 3 and 5 are re-plotted.

(33) Figure 3 and 5 caption: "The vertical velocity field has been scaled by 300." – This statement is meaningless as you haven't given us a scale for the wind vectors in the first place.

Reply: This was a plotting error. The scale for the wind vectors is added.

(34) Figure 4: Wrong caption for the figure.

Reply: It is corrected now.

(35) Technical Corrections P20161, L5-6: NOx and HNO3 are not really "reservoirs" of NO like PAN, HNO3 is an end product and NO is one part of the NOx family. Consider rewording this. P20162, L25: you should add a space to "peroxyacetylnitrate" so this matches P20163 L28. P20163, L17: Subscript the "y" in "NOy" P20164, L6-7: Re-

place ‘;’ with ‘,’ and add ‘its’ before ‘re-circulation’ P20164, L26: Consider cutting the phrase “measurement periods” P20165, L13: Remove the ‘s’ after ‘aerosol’ P20171, L26: Since you discuss Figure 7 before Figure 6, you should reverse the order of the figures.

Reply: All the above technical corrections are incorporated in the revised manuscript.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C9267/2014/acpd-14-C9267-2014-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 20159, 2014.

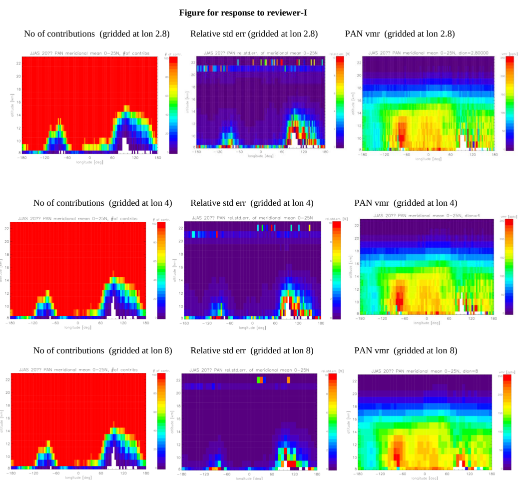

Fig. 1.