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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.

Anonymous Referee #1

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General Comments

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. 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. 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.

Specific Comments

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?

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 rewrit-

ACPD
14, C7372–C7378, 2014

ing 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?

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.

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

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?

P20165, L4-5: What is the reference for "the documentation"? What quality flags were actually used?

P20165, L5-6: This height range was specified by who? How was it determined? Does it only apply to PAN or all retrieved species?

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?

P20165, L17-27: This is the sort of information on PAN formation that should be in the

ACPD
14, C7372–C7378, 2014

introduction – consider moving it up there.

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. 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 200 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?

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?

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?

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.)?

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

P20168, L1-2: It's not clear here that you are talking about only three maxima, as you mention five locations. Consider revising.

P20168, L8-9: What is your evidence for your statement that the outflow of biomass

ACPD
14, C7372–C7378, 2014

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.

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?

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

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.

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

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

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.

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

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?

P20171, L5-7: This is hard to confirm as the northern and southern hemisphere plots are not all on a common color scale.

P20171, L12: From the NASA what?

ACPD

14, C7372–C7378, 2014

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?

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?

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

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.

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?

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.

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.

Figure 4: Wrong caption for the figure.

Technical Corrections

ACPD

14, C7372–C7378, 2014

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: Replace ‘;’ 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.

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