Replies to Anonymous Referee #1

(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 O_3 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.

Replies to Anonymous Referee #2

(1) It should be said that the manuscript seems to be a resubmission of a previously paper published in ACPD in 2014 http://www.atmos-chem-phys.net/14/12725/2014/acp-14-12725-2014.html that was not further published in ACP. Two versions do not show substantial differences and no indications are given in how the pertinent remarks of the reviewers in 2014 were addressed.

Reply: Point to point reply to a previously paper published in ACPD in 2014 were uploaded at the time of resubmission. We thank reviewer for careful reading and valuable suggestions. We have incorporated suggestions given by the reviewer which are marked in red color in the revised manuscript.

(2) The 2015 paper from Fadnavis et al. addresses the role of transport on the UTLS concentrations of PAN based on a 10-years CCMI simulation. The topic is certainly interesting and there is a huge amount of information on different issues that, however, is

not treated sufficiently in deep; this is a crucial weakness since the focus of the paper is smeared out and the analysis should be improved. Below a series of major and other issues that should be addressed.

Reply: The important points are discussed in depth. Considering the length of manuscript (already 63 pages) we have discussed in deep the important points and not all. We have incorporated suggestions given by the reviewer in this version of the manuscript.

(3) This paper shares a non marginal fraction of information with a previously published paper from the same authors (http://www.atmos-chem-phys.net/14/12725/2014/acp- 14-12725-2014.html). Especially concerning the comparison with satellite and aircraft observations.

Reply: The text related to comparison with satellite and aircraft observations is revised (page 15, lines 308-317). The text overlapping with companion paper is removed.

(4)The role of deep convection on PAN distribution (that is obvious) is mentioned in the companion paper and it should be essential to clearly state what is really new.

Reply: To our knowledge this is the first paper showing the pollution transport from Asia, North America and Africa into the UTLS in one consistent model framework. The numbers of papers have documented large amount pollution transport occurs cross tropopause from Asia (Park, 2006; Fu et al., 2006; Park et al., 2007). However transport from other monsoon systems (WAM, NAM) and their contribution to Asia gotten less attention (see page 3, lines 63-68).

The companion paper is different than the current paper. The companion paper shows contribution of transport of PAN from Indian and China (Asia) to the PAN trends in the UTLS. The companion paper shows Chinese emission has greater impact than Indian emission on the UTLS.

(5) The comparison with satellite data is misleading. As mentioned by the other reviewers the model n covers a different period with respect to the MIPAS database and that attribution of differences and biases is not sufficiently addressed and remains speculative. Authors claim that this simulation should be considered as a mean climatology but two points should be clarified: The simulation is driven by 1995-2004 SSTs and year 2000 emissions and hence is representative of a specific period for climatic variability and chemical regimes. The simulation is 10-years long and it should be clearly demonstrated that a relatively short period could be sufficient to include the necessary variability.

Reply: As suggested, model simulations are now performed for the period 2000-2010 overlapping period of MIPAS (2005-2011). We ran model for 11-years period in order to obtain a reasonable statistics. The simulated features are almost unchanged in new simulations but there is marginal improvement in model biases.

(6) The comparison with aircraft data has several weaknesses as well. A point-to-point correlation is to my opinion inappropriate while comparing sporadic observations (as the in-situ ones) that can be biased by specific flight strategy and a coarse simulation that should be representative of mean large scale conditions. It is necessary to define appropriate diagnostics (as done for instance in several CCMVAL analysis) to extrapolate from the aircraft database some synthetic and representative data to be compared to a CCM run (i.e. mean profiles, tracer-tracer correlations, latitudinal transects).

Reply: We want to validate performance of the model during monsoon season. Hence we computed point to point correlation as suggested by the previous reviewers of 2014 manuscript. We have checked that Aircraft observations are within area 150 km x 200 km which are comparable with the model grid (280km x 280km). Text related to comparison with aircraft data is revised (page 15, lines 308 -317).

This is common method reported by number of authors in the past. Vertical profiles of model output are compared with ozonesonde observations in our companion paper (figure S3). Also, Latitude-longitude transects of PAN, NOx, O_3 and HNO_3 are compared in our companion paper (figure 1) (http://www.atmos-chem-phys.net/14/12725/2014/acp-14-12725-2014.html). This is now mentioned in the revised manuscript (page 15, lines 308-312).

In the previous manuscript we have shown latitudinal transects which are replaced with point to point correlations plots, as it was suggested by previous reviewer of 2014 manuscript. There are number of other methods, every method has its own merit and demerit. Our aim of model validation is achieved by this method.

(7) Since the main focus of the paper is the role of transport and hence an evaluation of the model dynamics or at least of Monsoon(s) circulation should be done. This can be done evaluating the mean Monsoon annual cycles, the extent of Monsoonal circulation at the ground, the distribution of deep convection. The same applies for the Asian Anticyclone discussed in P15104 (see detailed points) Moreover, it should be discussed how vertical velocities in a large scale model can be used to infer rapid uplif in deep convective regions. - Most of the discussion refers to the role of deep convection in different regions. The fact that authors treat PAN is a secondary issue since this is considered as a proxy for NOx tropospheric emissions. So, it would be desirable to coupe that to PBL tracers (as done in many studies) to desantangle the role of emissions from different regions.

Reply: There is some misunderstanding about monsoon system and monsoon circulation. Monsoon season is for four months June to September hence it is impossible to evaluate mean Monsoon annual cycles (12 months) as suggested by the reviewer.

As suggested, figure showing distribution of deep convection at the surface, Outgoing Long-wave Radiations (OLR) and 850 hPa winds (monsoon circulation near surface) has been now added (supplementary figure S5(a)-(b)).

To show vertical uplift by monsoon deep convection (refereed to monsoon clouds) we have added a figure (supplementary figure S5(c)-(e)) showing distribution of cloud droplet number concentrations and ice crystal number concentrations over ASM, WAM and NAM. Role of vertical velocities is also mentioned (see page 17, lines 366-367). Hope, lifting up of boundary layer PAN from different emission regions by monsoon convection (clouds) is now clear.

(8) Moreover, the discussion remains often speculative and sometimes misleading , as for example in the case of overshooting convection that is often invoked in the text. I agree that this may eventually play a role in the LS chemical budget but I have some doubts that

this could be seen in satellite data (due to their coverage / vertical resolution) and it is certainly impossible to consider it with a T42 CCM.

Reply: We have replaced the word overshooting convection to deep monsoon convections.

(9) The role of horizontal vs vertical transport in UTLS is certainly fundamental and is a focus of a vast literature. Here no real information is added and, again, the simulation strategy cannot really help.

Reply: To our knowledge this is the first paper showing pollution (PAN) transport from different monsoon system into the UTLS in one consistent frame work. Large amount of literature has documented transport of pollutants Asia to UTLS; however pollution transport from WAM and NAM to Asia in the upper troposphere which is then lifted to lower stratosphere by the Asian monsoon convection is not reported yet.

The discussion on horizontal and vertical transport in UTLS highlights new findings which are listed in conclusion section. Three of them are listed below.

(1) Caribbean region is source of pollution transport into the stratosphere, convective transport from South Africa, South America and Indonesia-Australia into the UTLS.

(2) The horizontal transport of PAN analyzed from ECHAM5–HAMMOZ simulations show that the PAN from southern Africa and Brazil is transported towards America by the circulation around a large upper-level anticyclone and then lifted to the UTLS in the NAM region.

(3) The vertical distribution of simulated HNO_3 over the monsoon regimes shows low concentrations above 10 km at the foothills of the Himalayas. In contrast, the results show strong uplifting of HNO_3 into the upper troposphere with NAM convection. This may be due to the fact that NAM convection is not as intense as the ASM and there may be more wet removal of nitrogen oxides in the ASM convection. The model simulations indicate a higher efficiency of NO_x conversion to HNO_3 over the Indian region compared to NAM.

As suggested emission sensitivity simulations over Asia, North America and Africa are analyzed and results are discussed.

(10) It would have been important (as mentioned above) at least to make use of different geographical PBL tracers. - The sensitivity analysis to lightning production is certainly interesting but is somewhat disconnected to the main core of the paper and appears to be still in a rough form. As mentioned earlier it would be necessary to conduct other types of sensitivity studies.

Reply: In order to connect discussion related to transport of PBL tracers with lightning production we have revised the text (see page 28-29, lines 621-625). As suggested, emission sensitivity analysis is performed and related discussions have been added in the revised manuscript.

(11)The discussions are often lenghty with unnecessary details on well-known items and English form may be accurately revised throughout the paper - In addition to that I fully share the remarks of the 2 reviewers of the first version of the paper (that to my opinion are still valid here) and the evaluation of reviewer 1.

Reply: As suggested, we have removed the lengthy discussion text from the sections 3.1, 4.2, 5 and 6.

(12) Other issues p15098L8 The production of PAN in deep convective regions is not so striking. The high values at 16 km height may be better discussed. Model coarse resolution may play a role here. It is also not clear how to address the role of tropopause folding. The last sentence of the paragraph is not clear to me.

Reply: The PAN production in deep convective regions is ~5 ppt/day hence it is ~150 ppt/month which we think is striking. We have now mentioned maximum PAN Production in convective zones at 16 km (~12 ppt/day).

In the last sentence of the paragraph we want to say that --- In the UTLS, during monsoon season, there may be contribution of PAN due to exchange between the stratosphere and the troposphere, tropopause folding etc. Hope it is clear.

(13) P15099L4 What is a coherent location ? Are Aircraft data aggregated in some ways ?

Reply: Coherent location is a grid point. Yes, Aircraft data are aggregated in the same way.

(14) P15099 section 3.1 The database of PAN seems to be limited to few points. In addition to the main issue raised before, it seems unappropriate here to calculate any correlation or statistical significance test.

Reply: We want to validate performance of the model during monsoon season. Hence we compared the simulated NO_X, Ozone, HNO_3 and PAN with observations available during June-September. Yes, it is true that database of PAN during this season is limited but other species, ozone, NO_X and HNO_3 are comparatively better. Therefore we apply point-point correlation for all the species.

This is common method reported by number of authors in the past. Vertical profiles of model output are compared with ozonesonde observations in our companion paper (Figure S3). Also, Latitude-longitude transects of PAN, NOx, O_3 and HNO_3 are compared in our companion paper (Figure 1) (http://www.atmos-chem-phys.net/14/12725/2014/acp-14-12725-2014.html). This is now mentioned in the revised manuscript (page 15, line 308-312).

The previous reviewers of 2014 manuscript suggested computing point-to-point correlation. Our aim of model validation is achieved by this method.

(15) P15102L21 I would not use WAM (West African) to identify land convection in Africa south of the Equator.

Reply: In this paper we do not focus on land convection. In fact it discusses the global transport.

(16) P15103L3 It is not straightforward that strong vertical winds are responsible for a strong transport in the UTLS. The question of the wind intensity (not mentioned in the paper) and vertical resolution is crucial here.

Reply: I think reviewer is refereeing to discussion --- "Congo and Southern Maritime Continent (Liu and Zipser, 2005). The analyses of vertical winds show strong transport from 10–40◦ E, 100–110◦ E, 70–80◦ W (in the belt 0–10◦ 5 S). The amount of high level cloud fraction is also high over these regions". The figure related to this discussion is not included in the manuscript although we have analyzed simulated vertical winds and high level cloud fraction. Now it is clearly mentioned that figure is not included in the manuscript.

(17) P15104L3 to 16 The whole discussion is confusing and not sufficiently robust. The evaluation of the Asian Anticyclone would need a per-se evaluation. I cannot see a region of mixing and the discussion on the role of tropical heating in generating the "Gill-type" dipole is incomplete and useless here.

Reply: As suggested, above discussion is now removed from the revised manuscript.