Author Comment to Referee #3

ACP Discussions: acpd-15-9941-2015
(Editor - Federico Fierli)
'Impact of different Asian source regions on the composition of the Asian monsoon anticyclone and on the extratropical lowermost stratosphere'

We thank Referee #3 for this detailed and very helpful review. Following the reviewers advice we split the paper into three parts. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics.

General Remarks

The paper presents a model study of the Asian monsoon anticyclone; the development during the year 2012 from May to October is shown as a case study. The paper tackles a number of important topics:

1) the variation in the position and shape of the Asian monsoon anticyclone over the year from formation to break-up;

2) The chemical composition of the air within and around the anticyclone with respect to the geographical sources of the pollutants, and its temporal evolution;

3) the vertical and horizontal transport pathways out of the anticyclone, and potential transport barriers.

Over all, the paper is rather descriptive and wordy instead of analytical (in a quantitative sense) and concise. For none of the topics listed, the authors got to the bottom of the issue. For this reason, a leaner paper focussing on just one of these topics, but doing this more exhaustively, would have been probably more helpful. With such a wealth of material to analyse, the authors *should indeed think of splitting this paper into two or even three. However, this is fully at the decision of the authors*

In response to this comment, we split the paper and elaborated a revised version of the paper with the focus on 'The chemical composition of the air within and around the anticyclone with respect to the geographical sources of the pollutants, and its temporal evolution'. We removed the other parts of the paper in the revised version of the paper.

Some comparisons to satellite observations of trace gases are performed, however, these are by far not extensive. Thus, the presented results have mainly to be taken as model reality rather than real world

√ For the revised version of the paper we performed pattern correlation between MLS measurements and CLaMS simulations in the regions of the Asian monsoon anticyclone for the entire monsoon season 2012 (1 May - 31 Oct 2012), and have added a new figures (Fig.4) with comparisons between CLaMS and MLS :

We revised Section 3.1.1 as follows:

'**Comparison to MLS measurements**

'To compare our simulation with MLS O_3 and CO measurements (Version 3.3) (Livesey et al., 2008), pattern correlation between MLS measurements and CLaMS results, namely MLS(CO)/CLaMS(CO), MLS(O_3)/CLaMS(O_3) and MLS(CO)/CLaMS(India/China), were calculated (see Fig. 1). It is expected from satellite measurements that CO mixing ratios are stronger within the Asian monsoon anticyclone than outside and vice versa for O_3 indicating that air masses inside the anticyclone have a higher tropospheric characteristic than air masses in the UTLS outside of the anticyclone. At all days between 1 May 2012 and 31 October 2012, MLS measurements of O_3 and CO in a region between 15 and 50 N and 0 and 140 E (shown as black box in Fig. 2) at 380 ± 20 K potential temperature are correlated to CLaMS results as described in the following. At each day, CLaMS results are interpolated on locations of the MLS measurements transformed to synoptic 12:00 UTC positions. For each day, both MLS measurements and CLaMS results are normalised so that the maximum value of each trace gas is equal one. After-

wards the linear Pearson correlation coefficient r(t) between MLS measurement and CLaMS results is calculated for each day. This procedure allows to be compared the spatial distribution of trace gases neglecting possible differences in the absolute mixing ratios between model and measurement and to compare the spatial distribution of different quantities such as measured CO and simulated emission tracers (here India/China).
Correlation coefficients r(t) ranging between 0.72-0.86 were calculated for MLS(O_3)/CLaMS(O_3) during the monsoon season 2012 between end of June and end of September. Before the monsoon season in early May an even higher correlation coefficient up to 0.95 was found. Correlation coefficients of 0.57-0.81 were calculated between both MLS(CO)/CLaMS(CO) and MLS(CO)/ CLaMS(India/China) between end of June and end of September. These high correlation coefficients confirm that CLaMS has the capability of simulating the spatial distribution of tropospheric trace gases such as CO and stratospheric trace gases like O_3 measured by MLS. To illustrate this, the same horizontal cross-sections as in Figs. 2 and 3 at 380 K potential temperature for MLS CO and O_3 as well as for CLaMS CO and O_3 are shown in the Supplement of this paper.

Thus, high contributions of the emission tracers for India/China are simulated in regions where high values of CO are measured indicating that the emission tracer for India/China is a good proxy for the spatial distribution of tropospheric trace gases measured in the region of the Asian monsoon anticyclone. The correlation coefficient of MLS(CO)/CLaMS(India/China) increases from 0. to $\approx$ 0.8 during the formation of the Asian monsoon anticyclone, as expected because in the model the tracer has first to be transported from the ground to the UTLS. After the breakup of the monsoon anticyclone the correlation coefficient of MLS(CO)/CLaMS(India/China) decreases because further upward transport of the tracer for India/China does not occur due the the missing convection in this region and therefore the spatial CO distribution in the UTLS is dominated by other processes. In the region of the Asian monsoon anticyclone, the correlation coefficients of MLS(O3)/CLaMS(O3) are somewhat higher than those of MLS(CO)/CLaMS(CO). Reasons for that could be deficiencies in MLS CO data (v3) in the lower stratosphere as suggested by Hegglin and Tegtmeier (2015). '

Figures 4 and 5 in the ACPD paper were moved to the Supplement of the paper and were replaced by the following Figure 1:

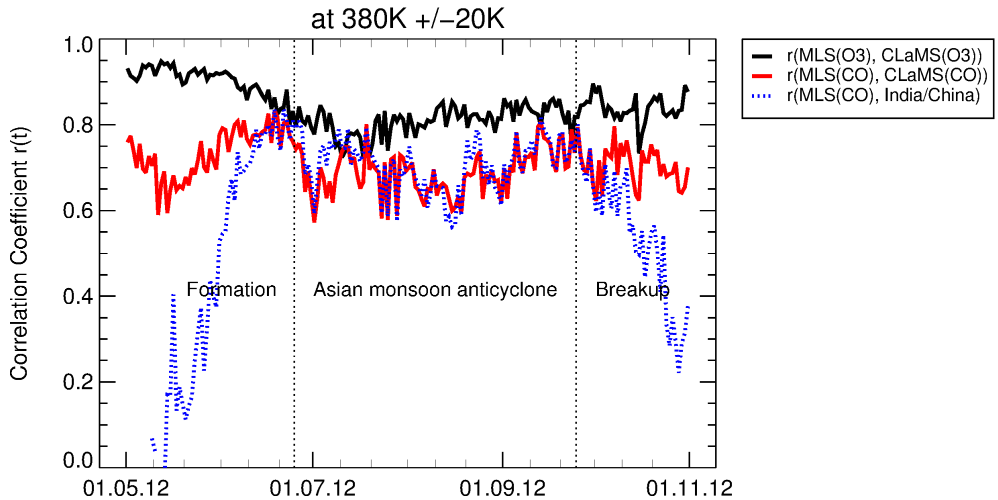

Figure 1: Correlation coefficients depending on time for tracer correlations patterns between MLS O_3 and CLaMS O_3 (black), between MLS CO and CLaMS CO (red), and between MLS CO and the CLaMS emission tracer for India/China (blue) for levels of potential temperature between 360 and 380 K (more details see text).

The section on the identification of a two-peak structure of the anticyclone (elongated or even split into two smaller ones) (discussion related to Fig. 6) is not very convincing, in my opinion. The two-peak structure searched for could easily be taken as one single broad maximum. More quantitative analysis would be needed here, should the authors decide to keep this section.

√ We removed this section.

The section on the anticyclone tropopause as vertical transport barrier (section 3.2.2) is very important and interestion, in my opinion. The authors should consider publishing this part of the paper separately, in order not to hide it at the end of a lengthy paper.

√ Yes, we agree that this point is very important. We follow the reviewers advice and will publish this part of the paper separately.

I recommend publication of the paper after consideration of my general remarks and specific comments as listed below.

Specific Comments

Abstract: General: The abstract is very detailed and a bit confusing. This is a pity because the paper might not get the attention it deserves. In line with my earlier general remarks, I find it would be easier and more interesting for the quick reader if the abstract was focused on fewer details; consider to boil down the abstract to few main messages of the paper.

$\surd$ The abstract is now focused on the remaining topic of the paper after the split and should be condensed now to the main message of the paper.We revised the abstract as follows:

'The impact of different boundary layer source regions in Asia on the chemical composition of the Asian monsoon anticyclone, considering its intraseasonal variability in 2012, is analysed by simulations of the Chemical Lagrangian Model of the Stratosphere (CLaMS) using artificial emission tracers. The horizontal distribution of simulated CO, O_3 and artificial emission tracers for India/China are in good agreement with patterns found in satellite measurements of O_3 and CO by the Aura Microwave Limb Sounder (MLS). Using in addition, correlations of artificial emission tracers with potential vorticity (PV) demonstrate that the emission tracer for India/China is a very good proxy for spatial distribution of trace gases within the Asian monsoon anticyclone. The Asian monsoon anticyclone is a transport barrier for emission tracers and is highly variable in location and shape. From end-June to early-August, a northward movement of the anticyclone and during September a strong broadening of the spatial distribution of the emission tracer for India/China towards the tropics is found. In addition to the change of the location of the anticyclone, the contribution of different boundary source regions to the Asian monsoon anticyclone strongly depends on its intraseasonal variability and is therefore more complex than hitherto believed. The largest contributions are found from North India and Southeast Asia at 380 K. In the early (mid-June to mid-July) and late (September) period of the monsoon season 2012, contributions of emissions from Southeast Asia are highest and in the intervening period (early-August) emissions from North India have the largest impact. Our findings show that the temporal variation of the contribution different convective regions is memorised in the chemical composition of the Asian monsoon anticyclone.

Air masses originating in Southeast Asia are found both within and outside of the Asian monsoon anticyclone because these air masses experience in addition to transport within the anticyclone upward transport at the southeastern flank of the anticyclone and in the tropics and can be entrained by the outer circulation of the anticyclone. Subsequently isentropic poleward transport of these air masses occurs at around 380 K with the result that the extratropical lowermost stratosphere is flooded by end of September with air masses originating in Southeast Asia. After the breakup of the anticyclonic circulation ($\approx$ end-September), significant contributions of air masses originating in India/China are still found in the upper troposphere over Asia. Our results demonstrate that emissions from India, China and Southeast Asia have a significant impact on the chemical composition of the lowermost stratosphere of the Northern Hemisphere in particular at the end of the monsoon season in September/October 2012. '

Page 9942: Lines 9-11: Isn't this obvious when the anticyclone is split into two smaller ones? I'd remove this sentence.

$\surd$ The statement is removed.

Lines 14-19: This is maybe too much detail; consider removing this sentence.

We revised the following sentence:

'The contribution of different boundary source regions to the Asian monsoon anticyclone strongly depends on its intraseasonal variability and is therefore more complex than hitherto believed, but in general the highest contributions are from North India and Southeast Asia at 380 K.'

as follows:

'The contribution of different boundary source regions to the Asian monsoon anticyclone strongly depends on its intraseasonal variability and is therefore more complex than hitherto believed. The highest contributions to the composition of the air mass in the anticyclone are found from North India and Southeast Asia at 380 K.'

Introduction: Page 9943: Lines 19-21: Really? Later you show that the tropopause above the anticyclone is an effective transport barrier; at least you should state here if this pathway is direct uplift or by isentropic poleward transport. Or maybe just change this sentence to: '. . .. The Asian monsoon circulation IS BELIEVED to provide . . .'

√ The statement is revised as follows:

'In general, the Asian monsoon circulation is believed to provide an effective pathway for water vapour and pollutants to the lower stratosphere of the Northern Hemisphere.'

Page 9944: Line 20-23: Is this in contradiction to your own findings?

√ The following sententence

'In addition to the impact on the contribution of the Asian monsoon anticyclone, deep convection at the eastern/southeastern side of the Asian monsoon anticyclone can directly transport tropospheric air into the lower stratosphere by direct convective injection (Rosenlof et al., 1997; Park et al., 2007, 2008; Chen et al., 2012).'

is revised as follows:

'In addition to the impact on the contribution of the Asian monsoon anticyclone, deep convection at the eastern/southeastern side of the Asian monsoon anticyclone is discussed as a pathway for transport of tropospheric air directly into the lower stratosphere by direct convective injection (Rosenlof et al., 1997; Park et al., 2007, 2008; Chen et al., 2012).'

Page 9951: Line 3-5: ' . . . two peaks . . .. are simultaneously found forming . . .. a double peak structure'. This is a redundant statement.

√ This section is removed in the revised version of the paper.

Line 12-18: These statements here are a somewhat speculative hypothesis, and this would be fine if further elaborated and proved in the paper. Without further proven evidence, however, theses statements remain speculative and should be removed.

√ This section is removed in the revised version of the paper.

Page 9952: Line 19: What is shown from MLS is not the same as from the model - make clear from the beginning that you show a stratospheric tracer, namely ozone, and a tropospheric tracer, namely CO.
Line 23/24: '. . . i.e. low ozone corresponds to high percentages of the emission tracers for India/China and vice versa.' This sentence is confusing. What you mean is low ozone = tropospheric air masses in contrast to high ozone = stratospheric air masses. However, polluted tropospheric air is expected to be higher in ozone than clean tro- pospheric air (still lower in ozone than stratospheric air, though). These relationships need to be clarified, otherwise it is hard to understand why polluted air loaded with emissions should come along with low ozone abundances.

√ This section is completely revised (see above, General Remarks).

page 9953: Lines 26 ff: I find the discussion in this section not very convincing; in particular, the lower panels of Fig.6 are interpreted as giving evidence to the bi-modal distribution. For me, I must admit, it looks merely like a broad maximum distributed over the entire longitude range. There is no evidence provided that the minimum between the 'two maxima' is indeed significant. For me, the only obvious and convincing feature in Fig. 6 is the shift of the tracer distributions towards the South from July/August to September/October. To prove the significance of the double peak, a statistical analysis needs to be performed. E.g. one could count the days over a larger number of periods and then assign uncertainties to the numbers; a gap between the two peaks then would be significant if the difference between the peak values and the minimum in-between is larger than 2 sigma; or any other reasonable measure.

√ We removed this Section in the revised version of the paper.

Page 9958: Lines 24 - page 9959, line 8: This justification why the 4.5 PVU *isoline can be used as boundary of the Asian monsoon anticyclone should come much earlier, e.g. page 9950, after line 9.*

√ The following sentence is added on page 9950 line 9:

' A value of 4.5 PVU is used which is in agreement with the upper limit of the PV values derived by Ploeger et al. (ACPD, 2015) to mark the transport barrier for the Asian monsoon anticyclone 2012 at 380 K.'

Pages 9959-9961, discussion of Fig. 8: How are the variations of the contributions from various source regions related to the variations of emissions from these source regions? Have the emissions assumed to be constant over time? This should then be mentioned here. In reality, emissions strengths may also have a variation over the year, thus complicating the situation.

To explain this in more detail and added the following paragraphs to the revised version of the paper:

in Sect. 2.1.1 Model Description / Emission tracers:

'The artificial emission tracers in CLaMS are designed to identify possible boundary source regions in Asia that could contribute to the composition of the Asian monsoon anticyclone in a particular monsoon season, here as a case study for the year 2012. At each time step of the model (every 24 hours) air masses in the model boundary layer are marked by the different emission tracers, i. .e. the emission tracer for North India (NIN) of an air parcel in the boundary layer over Northern India is set equal to one (NIN = 1). If an air parcel has left the model boundary layer over North India, the value of the emission tracer for NIN (=1) is transported to other regions of the free troposphere or stratosphere. Successive mixing processes between air masses from North India with air masses originating in other regions of the atmosphere (here NIN= 0) during the course of the simulation yield values of NIN differing from the initial distribution (NIN = 1 or NIN = 0). Therefore, the value of the individual emission tracer count the percentage of an air masses that originated in the specific boundary layer region since 1 May 2012 considering advection and mixing processes.'

in Sect. 3.2.1 Temporal evolution of different emission tracer:

‘The artificial emission tracers in CLaMS are designed to identify possible boundary source regions in Asia that could contribute to the composition of the Asian monsoon anticyclone during the monsoon season 2012 (as defined in Sect. 2.1) considering advection and mixing processes. E. g. the fact that the contribution of the emission tracer for Southeast Asia dominates in June demonstrates that in June upward transport or convection in the region of Southeast Asia is stronger than in other regions over Asia causing higher contributions of the emission tracer of Southeast Asia within the Asian monsoon anticyclone compared to other emission tracers in June. By this technique contributions of the boundary layer with a transport time from the boundary to the UTLS longer than one monsoon period (contributions from the boundary layer that are released before 1 May 2012) are not covered by the artificial tracers used here. Therefore, the composition of different emission tracers within the Asian monsoon anticyclone is a fingerprint of the regional and temporal variations of convective processes causing strong upward transport within the Asian monsoon anticyclone in summer 2012.

The sum of all emission tracers shown in Fig. 8 (ACPD vers.) is less than 100 % because air masses originating in the free troposphere or stratosphere also contribute to the composition of Asian monsoon anticyclone. End of June, a contribution of 35 % of the model boundary layer to the composition of the Asian monsoon anticyclone is calculated (see here Fig. 2). The remaining 65 % of the composition of the anticyclone is from the free troposphere and the stratosphere. The contribution of the model boundary layer rises to 55 % in early August and to 75 % at the end of the monsoon season in late September.’

Page 9964, section 3.2.2: I was particularly impressed by this analysis demonstrating that the Asian monsoon anticyclone tropopause acts as a vertical transport barrier, and personally I find this is a very important result that should not be hidden at the end of a lengthy paper. I’d really encourage the authors to split the paper and to make a separate short paper out of this. When discussing the results against previous literature, the paper by Randel et al., Science, 2010 must not be ignored.

We agree that this is a very important result. Many thanks for encouraging us to highlight this in an separate short paper. That is what we will do.

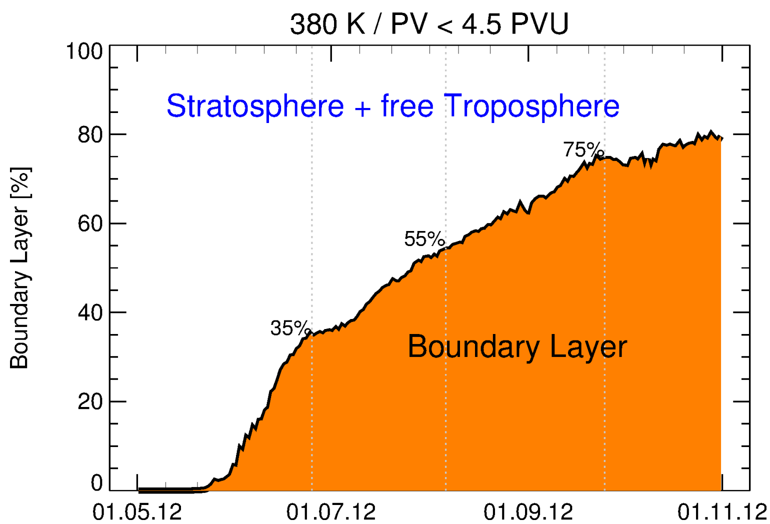

Figure 2: Temporal evolution of contributions of air masses from the boundary layer to the composition of the Asian monsoon anticyclone. The shown percentages are mean values calculated for air masses in Asia in the region between 15 and 50 N and 0 and 140 E at 380 ± 0.5 K (see black box in Fig. 1) with PV values lower than 4.5 PVU that marks the edge of the anticyclone.

Technical Comments

Page 9942, Abstract: Line 26: typo 'still'

√ done

Page 9943: Line 26: '. . . water vapour HAS a . . .'

√ done

Page 9945: Line 23 and 26: typo : 'source regions' (without s)

√ done

Page 9964: Line 6: Correct the sentence: '. . .are uplifted in the tropics are widely distributed . . .'

√ This section is removed in the revised version.

Line 13: Remove one 'the': ' . . . air masses from the the Asian monsoon . . .'

√ This section is removed in the revised version.

References

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