Impact of the Asian monsoon on the extratropical lower stratosphere: trace gas observations during TACTS over Europe 2012

Müller et al.

Point-by-point response on anonymous referee #3 (C11603)

Blue: Referee comment

Black: Response by author

We thank the reviewer for the careful reading and the suggestions, which helped to improve the paper. Below are our comments to the specific questions.

General comments:

This paper aims at better understanding the origin of the air masses in the extratropical UTLS (ExUTLS). It is based on airborne observations during the TACTS 2012 campaign and on Lagrangian trajectory modeling. The main conclusions concern the seaonal variability between late summer and fall of air masses transport between the "tropics" and the extra-tropics and the difference between transport to the region below and above 380K. Overall, the paper presents new and interesting results that are supported by evidences from the measurements. Nevertheless, the presentation and methodology are rather confusing which weakens the conclusions and I recommend some important revisions to address the comments listed below before publication.

Overall, the paper is confusing first because it deals with a number of atmospheric regions such as ExUTLS, ExTL, stratosphere above the ExTL, monsoonal region, Asian monsoon anticyclone, tropics, tropical stratosphere, extra-tropical lower stratosphere, which are not enough defined and which may or not be identicals for some of them. For instance, it could be understood that tropical and monsoonal are meaning the same (see comments below) which is not correct. A figure clearly identifying the different regions such as the Figure in in Hoor et al. (2005) would be beneficial to avoid this confusion which makes the understanding of the paper difficult. Using a single terminology for each region would also allow to avoid confusion.

Reply: We added an illustration to the paper (new. Fig.1). Additionally we changed the several passages in the text to make the paper more concise and less confusing. Particularly we rephrased the introduction according to Fig.1. and changed at several locations the text accordingly.

Change: l.21-40: The UTLS region (Fig.1) encompasses the global tropopause region and the lower part of the stratosphere up to potential temperature levels of Theta = 430K which coincides with the lower end of the tropical pipe in the stratosphere (e.g. Hegglin and Shepherd, 2009; Palazzi et al., 2009). Transport in the UTLS is thus affected by the stratospheric Brewer-Dobson circulation (BDC, Brewer, 1949; Dobson, 1956) with slow diabatic ascent in the tropics across the tropical tropopause layer (TTL) (Fueglistaler et al., 2009) and diabatic downwelling in the extratropical stratosphere." The BDC consists of two significant different transport pathways. The deep branch of the BDC transports air from the tropics to the extratropics via the upper stratosphere and lower mesosphere on time scales of several years (Butchart, 2014). In contrast, the shallow branch of the BDC mainly affects the region between Theta = 380K and 430K by quasi-isentropic transport and mixing (Hegglin and Shepherd, 2007; Spackman et al., 2007; James and Legras, 2009; Birner and Bönisch, 2011). In the extratropics below Theta= 380K the lowermost stratosphere (LMS) (Hoskins et al., 1985) as part of the extratropical UTLS (Ex-UTLS in the following) is affected by rapid isentropic transport and mixing across the subtropical jet. Transport across the extratropical tropopause layer (ExTL) further potentially contributes to the composition of the lower part of the Ex-UTLS. The ExTL and LMS are mainly characterized by exchange processes across the tropopause on time scales of days to weeks (Berthet et al., 2007; Bönisch et al., 2009; Hoor et al., 2010; Konopka and Pan, 2012; Jurkat et al., 2014).

The Asian Monsoon Anticyclone (AMA) is also a key feature of the paper but is not clearly defined and its variability is not properly accounted for. How is it possible to characterize the AMA ? Where is it located and what are its boundaries? Is it present with the same intensity during the whole period analyzed? Without answers to those questions, it appears difficult to draw conclusions about the impact of air masses fromthe AMA on the composition in the ExUTLS and ExTL during TACTS 2012. The AMA is bounded to the north by the SWJ and to the south by the TEJ. These jets and their intensities should also be documented. The dynamical situation (PV) is only given for the flight 2 of the campaign (Fig 1 a and b) which is not enough. A characterization of the dynamical situation for the different phases of TACTS and of the period up to 50 days prior to the first phase (to account for the 50 days back-trajectories calculations) should be added.

Reply and change: We added new Figures to the main manuscript to show the location of the AMA for July and August. To define the AMA we followed the method by Bergman et al., (2013) using deviations from average geopotential height as an indicator for the location of the AMA.
We also added a comprehensive supplement to the paper showing the development of the AMA in steps of five days from July to September.
We also added the information on PV for the individual flights to the supplement and changed the text of the main manuscript:
l.116-123: "…The flights, which are the basis for our study, were performed between 200 hPa and 130 hPa up to Theta = 410 K. The composition change in the Ex-UTLS between both phases will be compared in section 4. As shown in the supplement, the flights were performed mainly in regions of horizontal PV-gradients associated with Rossby wave activity. During both phases the flights covered a region from the Cape Verdes to the Arctic. PV values exceeding 10 pvu during both phases clearly indicate, that stratospheric air masses were probed during TACTS."

The conclusion about the seasonal evolution of the transport pathways from the "tropics" to the extra-tropics between the end of the summer and the beginning of fall is very general but only based on a two phases campaign during a particular year. The statements should therefore be mitigated with formulas such as "during the year 2012", "based on TACTS 2012 data". For the same reason, the last sentence of the abstract should also be mitigated "the study shows that in 2012... from summer to autumn...

Reply and change: Since we only have measurement data from 2012, we agree and changed the relevant sections accordingly.

Detailed comments:

p34769: cite Randel et al., Science, (2010) for upward transport of pollution from the AMA up to the stratosphere via the BDC based on ACE HCN observations.
We added Randel et al., (2010)
Change (l.53): "…This circulation pattern is coupled with persistent deep convection (Bourassa et al., 2012; Bergman et al., 2013) which lifts chemical constituents from the lower troposphere to the tropopause region and lower stratosphere (Randel et al., 2010)"

p34776 L3-10: there is no explanations on why 5 ML were chosen and on how the data points corresponding to each of the ML were chosen. The authors should better explain their methodology to derive the ML.

Reply: The use of mixing lines involving CO (or other short-lived species in the sense of stratospheric transport and lifetimes) is well established for transport processes involving cross tropopause transport. It makes use of the fact, that CO will equilibrate to a fixed background mixing ratio if no additional transport from the troposphere occurs. Thus any value above this background value must be the result from previous mixing. Assuming two well mixed reservoirs which mix e.g. at their boundaries, linear tracer-tracer relationships will form, as long as the mixing process is incomplete and fast enough compared to chemical CO loss. The formation of such mixing lines is not restricted to the cross tropopause mixing only and might occur between any air masses with different chemical composition.
In our case we investigate short-term processes on the order of six weeks, for which we use CO. A mixing line is identified first of all from the tracer-tracer scatter-plot as shown in Fig.5. In a second step we check, if a) the relevant points belong to a consecutive flight section and are not spurious alignments and b) if they are on isentropic surfaces since in the stratosphere mixing processes are initialized preferably by quasi horizontal isentropic processes.

P34780 L18-23: within the AMA MLS data show that CO vmr is of about 80-100 ppbv at 100 hPa (Park et al., JGR, 2007, 2009) and of 100-140 ppbv at 150 hPa (Li et al., GRL, 2005). CO lifetime in the UTLS is about 2 months. Therefore, if air masses were in the AMA 30 days before being sampled in the ExUTLS, shouldn't they be characterized by CO vmr larger than the 20-40 ppbv measured?

Reply: This is exactly true. Our argument here is, that these CO-enriched air masses comprise the 'younger' stratospheric mixing partner of the observed mixing lines and on the order of 50 ppbv. If we assume an average OH concentration of 1×10^{6} molec/cm^3 at p=100 hPa 100 ppbv of CO would degrade to 67 ppbv after 30 days of transport (51 ppbv after 50 days) including no further mixing (dilution). Thus, the mixing lines can be regarded as remnants, which indicate mixing with an air parcel, which already has a stratospheric mixing and degradation history.
Therefore we used the term 'young stratospheric air' for such an air parcel, which carries a tropospheric signature, but is chemically and dynamically a stratospheric air parcel (in contrast to a stratospheric air parcel with no recent tropospheric mixing signature).

p 34780 L1-5: it is mentioned that air masses rise to 400K with PV>5PVU within the AMA but it is difficult to see where PV values rise above 5 PVU on Fig. 7.

Reply and change: We marked the location, where PV values exceed 5 pvu in the revised Fig. 7. The revised figure shows, that there is no single location where PV exceeds 5 pvu. It shows however in combination with the newly included Fig.9 that the air parcels spent a significant time in the region of the AMA.

From Fig. 5 and 6 it seems that air masses within the AMA stay below about 390K. But the boundaries of the AMA are not defined in the paper. They could be with PV or geopotential heights threshold such as in (REFS).

Reply and change: As stated above we included a new Fig.8 showing the residence time of trajectories and also added a Figure (new Fig.13) showing the boundaries of the AMA according to Bergman et al., (2013). In addition we have included the evolution of the AMA in intervals of five days from July to September to the supplement.
The release of the air parcels from the AMA to the extra tropics as indicated by the decreasing potential temperature (Fig.9 in the revised version) occurs over a broad range of times according to the trajectories (Fig.7).

P 34781-34782: the discussion about the increase/decrease of trace gases concentrations in the ExTL and above is interesting but a bit fuzzy. First, it is not clear why CO increase above the ExTL should be coincident with N2O increase and O3 decrease and not above. Could you develop the arguments about the chemical lifetimes?

Reply: We added a more detailed paragraph to the text. The key point is, that an increase of tropospheric tracers above the ExTL should be evident in any tracer with lifetimes of months to years.
As long as TST (troposphere-to-stratosphere transport) is ongoing, both tracers should be enhanced n the same way. A decrease of the short-lived tracer in the ExTL thus indicates that the respective transport pathway is less effective. This allows the short–lived tracer to be degraded whereas a long-lived compound like N_2O is less affected.
The fact, that CO in the background LS (above the ExTL) shows a stronger enhancement than in the ExTL (with N_2O showing no significant reduction in the ExTL), thus indicates, that the isentropic pathway became less important for the observed N_2O and CO enhancements in the LS above the ExTL.
Change: We revised the whole former section 4.2. to:
l.385-395:"…Histograms for trace gas mixing ratios in the ExTL in Fig. 15, here defined as the region between 3 and 8 pvu (Hoor et al., 2010), are less clear. The frequency distributions of CO and N2O show a large variability and rather a stagnant or even decreasing tropospheric contribution. Reduced CO in the tropopause region indicates that the observed increase of tropospheric tracers in the lower stratosphere down to Theta= 350K (Fig. 11) is not due to isentropic transport across the subtropical jet.

If rapid transport of tropospheric air into the stratosphere were responsible for the increased tropospheric signatures above the ExTL, CO would also have increased in the ExTL. Therefore the transport of relatively young stratospheric air masses with a large tropospheric air mass fraction at Theta > 380K is responsible for larger mixing ratios of tropospheric (N2O and CO) and lower mixing ratios of stratospheric tracers (O3) above the ExTL. At mid- and high-latitudes these air masses subsequently descend to lower potential temperatures."

How much are they different between both stratospheric regions? The limit of the ExTL should be drawn in order to be sure about what is within the ExTL and above it.

Reply: Unfortunately it is difficult to determine an unambiguous limit for the upper bound of the ExTL. We selected a PV value of 8 PVU. As shown in Hoor et al., (2010) the 8pvu value separates short transit times of TST events from the stratospheric background. As further shown in Kunz et al., 2013, particular above Theta = 350 K 5 PVU are a good proxy for the tropopause, thus a range of 3-8 PVU covers the ExTL in the potential temperatures levels, where we focus on. We added the 6 and 8 pvu isoline to the top panel of figure 11. This figure shows that the 8 pvu isoline is a valid threshold to distinguish between the ExTL and the layer above.

Second, it is mentioned that no sign of changes are detectable for O3 and N2O in the ExTL due to their long lifetime. I do not agree, the 3 species display bimodal distribution between the ExTL and above. In the ExTL, enhanced O3 (from green to yellow) clearly coincides with decreased CO. A lower interval for the differences should be chosen for O3 to better see the increase. It is not so obvious for N2O because the color code does not really allow to see but colors from green to light blue probably correspond to a decrease.

Reply and change: The whole section 4 has been revised including the Figures. We now include tracer frequency distributions for the ExTL and the stratosphere beyond 8 pvu. We also changed the color code of the revised Fig. 11. In combination with the new Fig. 15 these plots show, that the variability in the ExTL is large and the temporal change over the measurements is weakly pronounced and less prominent.

P 34781 L21-23: in the paper the Asian monsoon region is defined as extending from 20 to 50N and 40 and 150E and Theta > 360K. It corresponds neither to the Asian monsoon identified by convective activity which is limited to South (India, Bay of Bengal) and South East (Thailand, Cambodia...) Asia. Talking about the Asian monsoon above Arabia, Kazhakstan and Mongolia is rather weird. The authors probably meant the AMA region as is mentioned in Fig. 11 caption. In that case, as already mentioned, a correct definition of the AMA should be used. The variability of the AMA is very important even on a day to day basis and no square domain can account for such a variability. This has been shown by Popovic and Plumb, JAS, (2001) (cited in the paper), and Garny and Randel, JGR, (2013) based on PV data. Even on a climatological basis, the square domain that is used is not appropriate. The AMA does never extend to 50N or 150E. Vertically, the AMA is not limited to theta > 360 K as stated in the paper but encompasses the UT down to 300 hPa. Furthermore, the AMA horizontal extension varies a lot between the UT (300 hPa) and the LS (_80 hPa). In their paper Bergman et al., JGR, (2013) try to determine the origin of the air parcels ending within the AMA. For that purpose they use geopotential heights (GH) criteria such as GH > 16.77 km at 100 hPa and GH > 12.52 km at 200 hPa. Randel and Park, JGR, (2006) use GH >14320 m at 150 hPa. For instance, with the Randel and Park (2006) criterion, for the first stage of TACTS (30 Aug to 5 Sep 2012) the AMA has a single mode and roughly extends from 35 to 115E and 18 to 38N at 150 hPa (which roughly correspond to 360K). 50 days prior to this period (10-15 July) when the back-trajectories start, the AMA is bimodal (with one mode over Tibet and one mode over Iran) and elongated from 5 to 135E. For the second phase (23 to 26 sep), the AMA has largely weakens and partly remains between 80 and 120E. A better criterion based on dynamical parameters (GH or PV) should be used in order to determine if the air masses originate from the AMA.

Reply and changes: As stated above we included maps of the geopotential to the main text and the supplement, which show the evolution of the GP at 100 hPa in intervals of 5 days from July until September. We also now included a definition of the AMA on the basis of Bergman et al., (2013). As stated by the reviewer the large day-to-day variability was exactly the reason to apply rather large thresholds in the previous version of the manuscript. We now included a new Figure (Figure 13), which shows the mean position of the AMA as defined from the geopotential and the region of a positive anomaly when applying the mean GP threshold as in Bergman et al., 2013 and a similar threshold based on the August 2012 data only.

We also added a sensitivity study to the supplement which shows the effect of varying regional thresholds. As shown in these plots, a monsoon impact on the lower stratosphere for PV > 8 pvu is still evident.

P 34783 L21-23: it is stated that "relatively large O3 mixing ratios on a given N2O level should be related to a more tropical or monsoonal origin". P 34784 L 8 "tropical" and "Asian monsoon" are further described as having a similar composition with high O3. Nevertheless, as shown in Randel and Park, JGR (2006) the AMA is an isolated region characterized by lower O3 than the surrounding tropical and extra tropical UTLS.
It is also stated that "the highest water vapour mixing ratios at theta = 390K occur in the region of monsoonal circulations". Is this not also indicative of tropospheric origin correlated with higher CO and lower O3 mixing ratios and contradictory with the above statement ? Is it not likely than transport of tropical (outside of the AMA) and "Asian monsoon" air masses are not responsible for the same O3-N2O correlation in the lower extra tropical stratosphere? This point should be clarified.

Reply: We removed the discussion of the N_2O-O_3 correlation and splitted the TACTS period in two phases simply by using time (without the N_2O-O_3 correlation) as suggested by reviewer 2 and discussion is no longer a part of the manuscript.

P34785 L22-25: this point concerning the AMA origin of air masses should be confirmed using better criteria for defining the AMA as mentioned above.

Reply: We changed this according to the comments above.

P34786 L1-3: as also mentioned above, it should be clarified weather "monsoonal" and "tropics" younger stratospheric air-masses are responsible for the same N2O-O3 correlations. If "monsoonal" and "tropical" means the same, this should be clear and "tropical" should be avoided.

Reply: As stated above we removed the discussion of the N_2O-O_3 correlation, thus this discussion is no longer a part of the manuscript.

P 34787 L14-25: the problem of a correct characterization of the AMA also weakens the conclusion of the paper concerning the larger flushing of the stratosphere above 400K in fall that in late summer. Indeed, during the same period the AMA weakens and has disappeared for the second phase of TACTS.

Reply: We improved our definition of the AMA as stated above and as documented by the revised Figures. We see over Europe the effect of export and transport from the monsoon after 20-40 days according to Fig.7. The decay of the AMA (see supplement) in September therefore has most likely not directly affected our measurements by the end of September, since we mainly observe the exported monsoon air with a phase lag from the beginning of September (see also Vogel et al., 2015).