

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 #2 (C11295)

Blue: Referee comment

Black: Response by author

We thank the reviewer for the careful reading of the paper and his suggestions. Below we address the criticism and the specific comments.

General Comments

This manuscript reports a data analysis work using airborne in situ measurements from the HALO research aircraft during a 2012 field campaign TACTS. The main conclusion is that strong tropospheric influence is found in the midlatitude lower stratosphere near 380K using tracer–correlations. Back trajectory analysis shows that the Asian monsoon uplifting is the main contributing process. The manuscript has some significant shortcomings and needs to be re-evaluated after major revisions.

General Comments (along the three ACP review criteria)

1. Scientific significance

The scientific significance of the manuscript is weak. The work presented has not provided new scientific insight. To improve upon this, I suggest the authors consider making the following points: What is the scientific significance if air mass in the observed mid-latitude location near 380 K is influenced by the Asian monsoon? What difference would it make if the tropospheric influenced air came through the monsoon region instead of the “regular” tropics? What’s the chemical impact of this transport pathway, qualitatively and quantitatively? In which way do you expect these observations to help improve models?

Reply:

The increasing anthropogenic sources of a variety of pollutants and aerosols in East Asia and India makes the Asian summer monsoon a key region for anthropogenic impact on the stratosphere (e.g. Richter et al.2005, Randel et al., 2010). The composition of the Asian summer monsoon as well as export of pollution from the Asian monsoon region is a major topic of current research (Glathor et al., 2009, Baker et al.2011, Schuck et al.,2010, Höpfner et al., 2015).

In our study we present strong indications that observed chemical composition changes are related to export of monsoon air. We note, that so far no other study based on in-situ measurements makes a direct link between the chemical composition of the lower stratospheric background in the extratropical mid-latitude stratosphere and a potential impact of the Asian summer monsoon (ASM).

The combination of strong pollution sources at the surface with the potential of rapid vertical transport above Theta=360 K provides a large potential to perturb the chemical composition of the lower stratosphere. This differs from the pathway via the tropical transition layer (TTL), which is associated with longer transport times to Theta = 380K. Therefore the Asian monsoon might act as a ‘short-cut’ for chemical constituent to enter the lower stratosphere.

We present quantitative estimates of transport time scales and residence times on the basis of combined tracer and trajectory studies. The observation of an enhancement of long and short-lived tracers like CO itself already indicates the potential chemical impact on the extratropical lower stratosphere. Other chemical reactive species of a similar lifetime like CO accumulate as well in the anticyclone (Park et al., 2009, Höpfner et al., 2015) and thus will be transported to the extratropics as well.

A full chemical model study is beyond the scope of this paper, our observations show, that via the monsoon pathway such pollutants raise the background mixing ratio of CO by 4 ppbv on average (which is large given a

background of less than 40 ppbv). Clearly any other pollutant with similar lifetime will enter the stratosphere via this pathway (Park et al., 2009; Randel et al., 2010; Glatthor et al., 2009; Höpfner et al., 2015). Transport to the stratosphere via the TTL is affected by other source regions and occurs throughout the year. Model studies thus have to account therefore not only for the correct chemical composition in the Asian monsoon anticyclone (AMA), where the constituents accumulate, but also on the correct transport rates to the lower stratosphere. An example is e.g. given in Vogel et al., 2013, 2015, which investigate potential processes.

Change: We added a part to the introduction, which highlights the importance of the Asian monsoon for atmospheric chemistry:

I. 58: "...The air within the Asian monsoon anticyclone is strongly affected by anthropogenic pollution, which originates from east Asian and Indian densely populated regions. During summer these pollutants accumulate in the anticyclone (Schuck et al., 2010; Baker et al., 2011; Richter et al., 2005) and strongly perturb the chemical composition of the monsoon tropopause region (Glatthor et al., 2009)".

2. Scientific quality

The scientific quality of the manuscript needs improvement, mostly because the discussion shows significant conceptual ambiguities (see Specific Comment 1). There is also a lack of quantitative results (see Specific Comments 2&3). The discussion is not focused enough on the objectives.

Reply: The focus is not on the detection of different deep stratospheric air masses, but on the effect of transport from the monsoon and mixing within the time scale of weeks.

Therefore the analysis tools valid for the deeper stratosphere are not applicable. Similar the analysis of Flocke et al., 1999 only focus on the ascending branch of the BDC, where measurement data are available.

Our study links measurements of the background far from the monsoon source to the lower stratosphere of the extratropics. Thus the budget approach by Flocke et al, is not applicable.

3. Presentation quality

The presentation quality has some shortcomings. The figures look as if they are at the exploratory stage of the data analysis. They are not refined enough to be concise and quantitative. There are too many repetitions (Figs 5&6, Figs 2&8, Fig 14). There are also a number of language and grammar issues.

Reply: We revised Figures 10,11 and 14, respectively. To reduce redundancy we removed Figs.8 and the lower part of Fig.11. We kept Figure 5 and 6 according to the comment of referee 3 regarding the location of the monsoon anticyclone and the variability of the anticyclone location. Fig. 6 shows the anticyclonic motion of the trajectories and their spatial extent, which is an important part of information for the analysis. We added, however a new Figure (new Fig.8) showing the residence times of the air parcels, which highlight the role of the anticyclonic region.

Fig.2 provides the link to the tracer time series and illustrates the method (see reply to reviewer 3). To add more quantitative information we followed the suggestion by reviewer 2 and will provide frequency distributions of the tracer data with mean and median for the ExTL as well as the lower stratosphere in the revised manuscript which complement Fig.10.

The former Figs.10 and 11 are revised with a discrete color bar to ease reading and scientific interpretation.

Specific Comments:

1. Conceptual ambiguities reflected in the choice of terms

It is difficult to get a clear physical picture of the result. The use of the term Ex-UTLS contributes significantly to this problem, especially the frequent use of "Ex-UTLS above 380 K". If the focus of the paper is on the transport of tropospheric air into lower stratosphere by the Asian monsoon, referring the region of destination as the Ex-UTLS above 380 K defeats the purpose in multiple ways: 1) Extratropical UT is almost never above 380 K, arguably with exception of the Asian summer monsoon; 2) the UT is really not of interest here, and 3) having tropospheric influence in the UT is not a meaningful statement. Because of these reasons, the statement of "Asian monsoon has impacted Ex-UTLS" is not meaningful. Furthermore, Lowermost Stratosphere (LMS) is a main component of the Ex-UTLS. It is a logical self-contradiction to conclude that there is an intensification of tropospheric influence in the Ex-UTLS and a weakening influence in the LMS the same time period (abstract).

Similarly ExTL is also a part of Ex-UTLS. To discuss them in parallel creates a lot of confusion, especially when the specific divisions are not marked in the figures.

Reply: We added a scheme (new Fig.1), which will contain the different terms and acronyms and their relation to each other. Note, that most of the terms are not clearly defined in literature. Only the terms ‘under- middle- and overworld’ (Hoskins et al., 1991) and lowermost stratosphere (LMS) as stratospheric part of the middleworld are clearly defined. Classically, the 380 K isentrope has been chosen as upper bound of the LMS and middleworld.

Since we refer to rapid transport processes affecting the region up to $\Theta=430$ K (though we measured only up to 410K) we use the term lower stratosphere for this region (which includes part of the lowermost stratosphere).

When referring to the region in the extratropics, which includes both, the lower stratosphere and the extratropical tropopause region, and the region above, we use the general term Ex-UTLS.

The ExTL has been introduced by WMO, 2006 (Fig.6) being identical with the tropopause following ‘mixing layer’ in the extratropics (Hoor et al., 2002, Pan et al., 2004), which extends around the tropopause in both directions.

It is correct that the extratropical UT is not extending above 380K, we used the term Ex-UTLS to indicate the region from the upper troposphere to the stratosphere up to 430 K, where tropospheric influence and rapid transport is visible (compare to Hegglin und Shepherd 2007).

The UT of the extratropics might play a minor role (though is not negligible, see e.g. Hegglin et al., 2004: the tropopause height of a subtropical ridge extending north to Ireland was about 380K), but the extratropical UTLS is affected by transport from the upper troposphere in the tropics (the TTL) as well as from the UT of the monsoon region. As such the UT does play a potential role for the composition of the Ex-UTLS.

Change: We added a section to clarify our definitions and terms in accordance with the new Fig.1:

I.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 $\Theta = 430$ K 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 $\Theta = 380$ K 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 $\Theta = 380$ K 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).

2. Purpose of the mixing-line discussion

The discussion of the mixing lines is a significant part of the paper but did not produce quantitative result. The analysis and discussion are somewhat narrowly conceived and the focus is on the “straight mixing lines” and whether one of the two end points represents “pure troposphere”. The early “classical” mixing line and tracer correlation papers, Waugh et al., 1997 and Plumb 2000, for example, have concise descriptions on the effects of mixing on tracer relationship. The concept of sustained mixing discussed there is very relevant to this work. Using the Waugh/Plumb framework, the slope and the shape of the mixing line would be able to help quantify the tropospheric influence through direct mixing (a single mixing with one end point consisting of the tropospheric background air) or multiple, sustained mixing. Since you are restricting the analysis to the stratospheric measurements, the Waugh and Plumb framework may be as relevant as, if not more than, the “L - shape” discussion.

Reply: Clearly the early classical mixing line discussions referred to long-lived stratospheric tracers, where large scale transport and long time scales govern transport and chemical contrasts.

Although we initially restrict the discussion of the N₂O-O₃ relationship of our analysis to the stratosphere (i.e. O₃ > 350 ppbv), the mixing processes, which are relevant for our study of the monsoon impact act on time scales of weeks and involve transport from the monsoon tropopause region.

This fact motivates the use of CO, which is ideally suited to identify the processes. Long-lived species (in the sense of Plumb and Ko, 1992) are only weakly affected by such mixing processes.

Further, slope changes of these long-lived tracers will not be affected by mixing with tropospheric air (see Boenisch et al., 2009, their Fig. 3) since mixing with a tropospheric air mass does not change the slope of these correlations. In contrast, this is the case if stratospheric air masses with different slopes (e.g. vortex / out-of-vortex air) start to mix.

Short-lived tracers like CO or tracers with a substantial variability at the tropopause lead to different slopes of the mixing lines. In our case we are close to the tropopause as indicated by minimum N₂O values of only 25 ppbv below tropospheric mixing ratios.

We study however transport from the troposphere on time scale of weeks, which motivates the use and importance of the L-shape correlation and particularly CO.

Due to the tropospheric variability the slope of such a correlation depends on the tropospheric 'endmember' and only provides qualitative information. On the other hand such a slope is a clear indication for irreversible tracer and air mass exchange within the life time of CO.

Change: We therefore totally removed the discussion of the N₂O-O₃ relationship and only focused on the CO-O₃ relationship as well as the frequency distributions of the tracers in different regions as suggested. The former section 4.1. (N₂O-O₃ correlation) was removed and replaced by the discussion of the frequency distributions).

Also note that many tracers go through sharp changes at the extra-tropical tropopause. The tracer's "tropopause value" is somewhat an ill-defined quantity.

Reply: Exactly this point is addressed with the slope analysis of CO-O₃ (former Fig.3), which tells us, that the observed slopes are below the tropopause variability of CO. Therefore we do not refer to a single CO mixing ratio as being characteristic for a source.

However, we can state, that the observed slope CO-O₃ is not associated with a direct injection or isentropic transport and mixing from the tropopause. The low CO value in combination with the slope can only be explained, if mixing between two different stratospheric air masses is considered, which both differ particularly by their CO mixing ratio (see Fischer et al., 2000, Hoor et al., 2002).

3. Seasonal evolution discussion

Overall the seasonal evolution analysis (Figs 10-15) does not bring out clear quantitative information and is also weak in physical interpretation. If the separation of the ExTL and the layer above is of strong interest, I suggest the authors to examine the (1D) distribution change in tracers with season in the two layers (histograms). Overall the 2D distribution figures are too noisy and do not serve to quantify the change well.

Reply and change: We changed the color table of the equivalent –latitude–Theta distributions (now Figs.11, 12, respectively), and added the frequency distributions in the two layers ExTL and lower stratosphere as suggested (new Figs. 13, 14).

For the change in monsoon influence, consider the seasonal change in vertical transport strength of the monsoon. For example, your earlier analyses shows that the monsoon uplifting occurred a month before the observations, which suggests that the late August observations may be associated with the late July convective activity, and the late September observations with the late August monsoon activity.

Reply: Exactly this temporal evolution and phase lag is shown in our study. Indeed our observations are affected by emissions and transport processes weeks before the measurements as shown by the trajectory analysis (according to Fig.6,7,8 and case study). Convection will surely contribute to the vertical redistribution and also the accumulation of tracers within the anticyclone.

Change: We added a supplement showing the temporal evolution of the monsoon anticyclone from July until September.

4. Relation to previous work

Although not using CO-O3 tracer correlation, a number of previous works investigated mixing in the lower stratosphere between tropics and mid-latitudes, including the potential temperature range focused in this paper. In particular the work using STRAT/POLARIS ER-2 data (Volk et al., 1996; Flocke et al., 1999) are very relevant studies. Omitting these works in the introduction and the statement P34775L20-22 give the impression that this is the first such study. Volk and Flocke also showed ways to produce more quantitative transport information in this region.

Reply: We are aware, that we are not the first ones who measured in this altitude region or applied tracer correlation analyses.

However, this work is the first study, which links the background of the ExUTLS to the monsoon and shows, that the 'flushing' is observed in many studies (satellite or airborne, Hegglin and Shepherd, 2007; Boenisch et al., 2009, Hoor et al., 2005) is at least partly linked to the effect of the Asian monsoon. This is also different from Flocke et al., 1999 and Volk et al., 1996.

Change: We added the references to the introduction (**I.32**): "...On the basis of in-situ data Volk et al., (1996) and Flocke et al.(1999) quantified entrainment rates for the tropical stratosphere highlighting the importance of mixing above Theta = 380K for the ascending part of the BDC."

Change:

Old: "In this study in-situ observed mixing lines on the CO-O₃-scatterplot are used for the first time to investigate mixing and transport processes in the Ex-UTLS above Theta = 380 K."

New:"Previous studies used the method of mixing lines to investigate exchange processes between the troposphere and stratosphere within the extratropical tropopause layer (ExTL) (Zahn et al., 2000; Hoor et al., 2004; Pan, 2004). As shown in Fig.3 (a) the observations indicate irreversible mixing above Theta= 380 K. Since this is 205 above the middleworld isentropic cross tropopause mixing is most likely not the driving mechanism for the observed mixing lines.

5. Issues with figures

There is too much repetitiveness in the figures. For example flight 2 O3-CO relationship is shown three times; Figs 5 & 6 are repetitive and so is Fig. 14. In the case of Fig 14, using a "fish born" style of plot (the mean and the error bars) instead of scatterplots would result in much better quantitative information, although I do not think the line of Figs 12, 13, and 14 is the most effective way to bring out the point the authors intended. It is probably more productive to just plot the tracer-relationship and distribution in each flight period and compare the change.

Reply: As stated above (reply to point 3) we modified or removed many Figures. We also included frequency distributions of the tracers for different regions to better quantify some aspects.

Figure color choices should be done more deliberately so the figures provide the quantitative information for the discussion. For example, the critical levels related to Fig. 5 and 6 are 370 K and 380 K. The continued rainbow color scale does not serve to indicate the trajectory point relative to these two critical levels. Similarly, consider showing clear distinction of above or below 100 ppbv in O3 scale in Fig.10 and the relevant critical values of CO.

Reply: The continuous color code of Figs. 5 and 6 are complemented by the temporal evolution of the relevant quantities over time in Figs.7 and 8. We changed to a discrete colour code in the new Figs. 11 and 12 as suggested.

6. Technical errors and language problems

- P34770L25: why "in-mixing"?

We removed the term "in-mixing" and replaced it by "mixing" throughout the manuscript.

- P34776L15: "larger 4 and 60 ppbv", check the entire sentence

We rephrased the sentence.

- P34789L26-27: The meaning of the sentence is unclear. To clarify, consider changing the location of "only" in the sentence.

Changed.

- P34781L26: "und" -> "and", also in P34806

Corrected.