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 #1 (C10943)

Blue: Referee comment

Black: Response by author

We thank the reviewer for his careful and comprehensive reading and the very helpful suggestions which helped a lot to improve the paper.
Below are our detailed replies to the specific points:

General Comments

This paper uses aircraft trace gas measurements in the extratropical UT/LS from late summer 2012 to analyze atmospheric composition change. The cause of those changes is shown to be the Asian monsoon. Although this is a lengthy review, it is 98% about language and writing, not science. I think the science presented here is very good, and find only one science issue that needs to be addressed. Scientifically, I suggest revisions to Section 4 and accompanying figures. This paper would benefit substantially from language improvements and I have suggested many below; I hope they help. This paper is appropriate for ACP and will be suitable for publication after minor scientific revision and somewhat more substantial language improvement.

Reply: We tried to improve the language and some of the figures. Hopefully this makes it easier to follow the paper.

The minor science issue involves Section 4.2. I don't like the approach taken in using the N2O-O3 correlation to identify changing tropospheric influence. I think there is a better approach possible, but I am also not sure that the results of 4.2 present additional information compared to 4.1. It may make sense to combine and shorten 4.1 and 4.2 by eliminating redundant analysis. Please see the comments regarding pages 83-84.

Reply: We removed the analysis of the N2O-O3 correlation and instead present the frequency distribution of N2O, CO, O3 and SF6 in the revised version in section 4.

Specific Language and Science issues For all comments below I refer to the last 2 digits of the ACPD page number (e.g., P85), and 'l' refers to the line numbers.

Reply: We rephrased major parts of the paper and considered the suggestions of the referee. We resign to recite all formulations since most of the text has changed and a direct answer on every point is not feasible.

P67, first sentence of Intro. It is contradictory to say that the impacts are not well known but then provide 9 references on the topic – clearly something is known. I would delete this sentence and incorporate these references into the text as appropriate. You can use the 2nd sentence to start the section but delete 'In general'.

Reply: We changed the text accordingly.

P68, l2. It sounds like the tropopause is exerting a force that suppresses exchange, which is not exactly correct.

Reply: We agree and removed the statement.

P68, l3. 'In the tropics the UTLS'…I believe the tropical portion of the UTLS is referred to as the TTL, not the UTLS. Rewrite accordingly.

Reply: We agree and added a scheme (Figure 1 in the new manuscript) of the UTLS to make clearer how the various regions of the UTLS are denoted in the paper.

P68, l5. Not 'Contrary', you mean 'In contrast,…'

Changed as suggested.

P68, l10. I would say the LMS is vertically defined by the tropopause…

Changes as suggested (confined → defined)

P68, l20-23. This is a 1-sentence paragraph. It belongs at the end of the previous paragraph.

We rephrased the section.

P69, l1. 'gains' should be 'has gained'

Changed as suggested (gains → has gained**)**

P69, l3. Delete 'Generally'

Changed as suggested (Generally deleted)

P69, l 24-29. This is a 2-sentence paragraph. It makes more sense to combine it with the previous paragraph, and perhaps break that previous paragraph in two.

We rephrased the section.

P71, l9-13. This paragraph could be much clearer. There are 13 flights but you look at only 7 of them, so that's all you need to mention. Suggested rewrite: '…provide the basis for this study. A large dataset of high spatial and temperature resolution mainly in-situ trace gas data was collected in the Ex-UTLS. This paper focuses on seven research flights extending from 15 to 70N and 25W to 15E, each lasting 8-10 hours.
There are 65h of trace gas measurements in the Ex-UTLS, with 40 of those hours in the stratosphere.' Also, why 'mainly'? As far as I can tell, ALL the data used here are from the in situ instrument. If this is the case, delete 'mainly' (here and anywhere else it is used).

Reply: We rephrased the whole paragraph:
"…Our study is based on in-situ measurements performed during the first atmospheric science missionswith the new German research aircraft HALO in August / September 2012 TACTS (Transport and Composition of the UTLS) and ESMVal (Earth System Model Vaildation). This paper focuses on eight research flights covering the Ex-UTLS between 15N to 70N and 25W to 15E with typically 8-10 hours per flight (see supplement).
The measurements of TACTS can be subdivided into two phases. The first phase with approx. 35 flights hours covered the time period between the 30 August and 5 September 2012. The end phase was performed from the 23 September until 26 September 2012. 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.

P72, l3. '10s or 0.1 Hz, respectively'? 10s is 0.1 Hz and there is nothing to be 'with respect to'. Change to 'a time resolution of 10s (0.1 Hz), corresponding to…'.

Changed as suggested.

P72, l7. Change to 'a 3 channel…spectrometer that measures CO, CO2, and N2O…:'.

Changed as suggested:

P72, l15. Delete 'Therefore' and begin sentence 'The in-flight calibrations identify and correct…'

Changed to: "…The in-flight calibrations are used to identify and correct slow instrumental drifts in the post-flight data evaluation.:

P72, l17 & l19. Delete 'finally' and delete 'respectively'

We deleted ,,finally" and ,,respectively"

P74, l2. Delete 'Hereby' and change to 'Diabatic heating rates are used to calculate vertical velocities…'

Changed as suggested:

P74, l11. Change to 'On the highest flight levels of 150 and 130 hPa, air masses with…'

Changed as suggested:

P74, l23. Delete 'performed'.

We deleted ,,performed"

P75, l2. Change 'respectively, partly chemically processed…' to 'partially processed'

The corresponding section was rephrased.

P75, l4-7. What is meant by 'the stratospheric equilibrium of CO'? CO 'degradation' is 'CO loss' or 'CO oxidation'. I believe what you're saying is that CO between 20-30 ppb indicates that the air is older than tropospheric air but younger than air above the LMS. Could you instead say something about how old you think air is that has 30 ppb CO, and how old air is that has only 12 ppb?

Reply: This is correct, we see tropospheric influence with some degree of chemical ageing. The CO-loss is directly related to the OH concentration along the respective air parcel history, which introduce uncertainty. Assuming 1e6 molecules / cm-3 at 100 hPa and 220 K one can estimate a 50% CO loss over 50 days assuming no mixing. This assumption is however a very simple and unrealistic case and thus only provides an upper limit for a given OH concentration. The CO-value which establishes as a result of in-situ production from methane (slow) and CO-loss via OH is on the order of 12 ppbv and does not allow to conclude on a specific age, since it would also arise without any initial CO only from CH4-oxidation.

Regarding Fig 1a&1b, I suggest leaving off the colored overlays that indicate different mixing lines. This is confusing and it's hard to see colored lines on top of a color contour map.

Reply: We think it is important to show the locations of the flight sections, where we measured the data relevant for the individual mixing lines. We therefore kept the figure as it was, since it might help to get an idea where the mixing lines were measured with respect to the synoptic situation.

P75, l20-22. Please rewrite and clarify. Is it the analysis that is 'first time', or is it the observation of mixing lines above 380K that it new?

The corresponding section was rephrased.

P75, l 28. Change this sentence to 'Linearly correlated data points with enhanced CO…'

Changed as suggested:

P76, l3. Change this to 'During TACTS Flight 2, 5 mixing lines (ML) at the potential Temperatures…"

Changed as suggested:

P76, l5-6. If you removed the colored mixing lines from Figs. 1a and 1b (and I hope you will), this sentence will have to change.

Since we think, this information is important, we kept the figure and therefore kept the sentence.

P76, l8. Delete 'identified' ('The five identified mixing lines...') Is Table 1 necessary? What use are they to the reader?

We kept the table since it gives a quick overview of the measured mixing lines and the corresponding meteorological parameters along these mixing lines. We agree that it is not absolute essential, but we think it is worth it to show particularly potential temperature ranges, and some statistics and want to keep it in the paper.

P77, l2-11. This is a muddled and confusing introduction to this section. It needs a rewrite that is organized along these lines: 1. "This analysis seeks to determine…" 2. "The difficulties are…" 3. "We use the method of blah blah to determine the tropospheric end members…" 4. "We apply this method to all ML."

Reply: We reworded the introduction (l.237ff) : "…This analysis seeks to determine the initial mixing ratios of the air parcels, which lead to the formation of mixing lines (Hintsa et al., 1998, Hoor et al., 2002). This requires knowledge on the initial mixing ratio of one of the species involved. Since mixing ratios of trace gases like O3 and N2O at the tropopause are fairly constant compared to their stratospheric gradient, they can be used to as initial estimate to analyze mixing across the tropopause. "

After rewriting, make this a separate paragraph and start the new paragraph with line 11 ('Figure 3 displays the tropospheric end member…"). No 'exemplarily'.

Changed as suggested

P78, l9-11. Suggested rewrite: "A tropospheric CO end member lower than typical tropopause values indicates that…"

We changed the text as suggested.

P78, l12. By 'chemically unprocessed, not CO degraded' do you mean 'not photochemically aged'? If so, this is a more precise way to say this.

That's correct, we changed the text. We wanted to make clear that both mixing and photochemistry in the stratosphere lead to CO mixing ratios lower than tropospheric CO mixing ratios**.**
Changed to: " The term „pure" is used in this context to describe undiluted and photochemically unprocessed tropospheric air masses"

P78, l13 & l25. Delete 'determined' and 'investigated'.

We deleted both.

P79, l5-8. This begins awkwardly. Try: "We investigate the origin of ML 1, 2, 4, and 5, which were observed in the trough away from sharp isentropic PV gradients at the tropopause, by analyzing 50-day back trajectories calculated with the CLaMS model. Variations of potential…"

We changed the formulation as suggested.

P79, l17. Suggested rewrite: 'These trajectories appear preferably for air masses with large PV (>8 PVU) in regions with the observed mixing lines.'

We reworded the section.

P79, l21. Delete 'significant'

Deleted.

P80, l1. Change 'suggest' to 'suggests'

Corrected.

P80, l6. Change to 'The calculation of 50-day back trajectories…'

Changed as suggested throughout the manuscript.

P80, l10, change to 'Clams calculates the vertical motion…'

Changed as suggested.

P80, l16, Change sentence to begin "Trace gas measurement of …"

Changed as suggested.

P81, l2-6. Delete this entire last sentence (beginning, "Subsequently,…"). You don't need to explain what's ahead.

We deleted the following sentences:

~~***Subsequently, the overall trace gas distribution of the Ex-UTLS measured during TACTS 2012 is investigated in the following section. It will be discussed, if the boreal Ex-UTLS during August and September 2012 is affected by air masses originating from the Asian monsoon region.***~~

P81,l9-15. This introductory paragraph could be much clearer and it calls Figures 9 and 10 in the wrong order. Here is a suggestion: "We investigate the effect of the Asian monsoon on the trace gas composition of the Ex-UTLS by examining the changes in N2O, CO, and O3 from the early (28 Aug-5 Sep) to the later TACTS flights (23 Sep- 26 Sep). The distribution for each period is calculated as the average of the flight data binned by equivalent latitude and potential temperature. Figure 9 shows the data coverage for each period. Figure 10 shows the mean distributions for N2O, CO, and O3 in the early and later periods as well as their differences."

Changed as suggested.

P81, l16-17. Try this: "Figure 10 shows no significant changes for N2O and O3 in the ExTL."

Changed as suggested.

P81, l19. Delete 'during TACTS'. In fact, throughout this paper, phrases such as 'during the flight campaign' and 'during TACTS' are often unnecessary and should be deleted. It is implicit that the analyses are about the TACTs data.

P81, l20. Change to 'Both indicate…'

Changed.

P81, l22-24. These 2 sentences could be succinctly combined: "This finding is true for the relatively short-lived species CO, which decreases in the ExTL but increases slightly above it."

Changed as suggested.

P82, l3-7. The changes aren't just above 380K but are also down to 350K.

Changed to 350 K.

P82, l8-10. I'm struggling with the words here. Is this the intent: "If rapid transport of tropospheric air into the stratosphere were responsible for increased tropospheric signatures above the ExTL, CO would also have increased in the ExTL ."

This is correct, changed as suggested.

P82, l12. Instead of 'At this point…', try "It is likely that strengthening of the jet stream in September weakens transport of tropospheric air masses into the ExTL…"

l.394: changed as suggested

P82, l16. Change to "Independent of this transport…"

l.397: changed as suggested

P82, l21. Delete 'Hereby'

Deleted

P82, l20-26. Try: "The top panels of Figure 11 show the percentage of 50-day back trajectories originating in the Asian monsoon region (criteria:…) using the same coordinate system as Fig. 10. The mean residence times of the Asian monsoon trajectories between t=0 and 50 days are shown in the bottom panels." Also, move these two sentences to the paragraph below it.

Changed according to the revised Figures: ". Fig. 12 shows the percentage of 50-day backward trajectories originating in the Asian monsoon region (criteria: 25N < TRA-latitude < 40N, 40E < TRA- longitude < 110_E and Tra-Theta >360K at t = -30 days) using the same coordinate system as Fig. 11"

P82, l27. I agree that there are quite a few trajectories in the ExTL in early TACTS that originated in the monsoon, but they are not the 'predominant' origins.
Changed to: "Trajectories originating from the Asian monsoon region"

P83, l1-10. I agree in general with what you say, but I don't get the significance of the residence times – can it be better explained why these matter? If they aren't essential to your argument they could be eliminated. 'Fewer' not 'less' trajectories. Are you certain that the jet is actually creating a transport barrier, or is it that the monsoon weakens so there isn't the anticyclone 'forcing' the transport of tropospheric air? I think that the difference panel in the top row of Fig. 11 is the most convincing part of your argument, particularly because it shows differences in the same location as the N2O and CO increases in Fig. 10 while the differences along the ExTL are near zero. The spatial agreement with Fig. 10 might be emphasized there.

Indeed the spatial argument has been sharpened in the revised version. We removed the figure showing the residence times and emphasized the spatial aspect. We think, that transport across the jet has not caused the observed tracer signatures even for a potential weaker monsoon 'confinement', as indicated by the vanishing difference patterns in the ExTL.

P83, l20-22. This reasoning isn't quite right. The tropical vs. older air N2O-O3 correlation you describe is true for the stratosphere, not the troposphere. In the troposphere they are nearly uncorrelated because N2O is very high everywhere while O3 can vary. If the monsoon air is highly polluted there will be (relatively) high O3 and high N2O, but in clean air there will be low O3, high N2O. 'Relatively large O3 on a given N2O level' will be an indication of pollution, not tropical origin.

The whole section has been replaced by a detailed analysis of the frequency distributions of tracers instead of the N_2O-O_3 correlation. We decided to motivate the temporal change simply by the different times of the measurements rather than the temporal change of the N_2O-O_3 correlation.

P84, l1-25. I don't think this is a good analysis. By separating the data into the low (red) and high (blue) O3 mixing ratios you are making assumptions about the origin of these air masses. If you want to identify a change *in air mass between early and later TACTS, a more unbiased analysis would be to plot histograms for N2O (O3) for the early flights, then overlay the histograms for the later flights – similar to the plots in Fig. 13. (The histograms could be filtered by theta above the ExTL perhaps.) This should reveal the composition change during September. (The species shown in Fig. 13 should then be analyzed in the same way.) It might be useful to calculate histograms for more than one theta or height above the tropopause to make points about transport contributions at different levels.*

We thank the reviewer for this constructive suggestion and added new Figures showing overlays of the frequency distributions as suggested for the stratosphere (PV > 8 pvu) and the ExTL. The discussion has been modified accordingly.

P84. I'm not sure that this section (4.2) is actually providing any new information compared to Section 4.1 It seems like it's just another approach to getting the same information. If there is a result in 4.2 that was not shown in 4.1, please make it clear what that is. If there is no new result here, consider combining 4.1 and 4.2 and eliminating redundant analyses. Also, consider modifying the section title to alert the reader to the goal of the analysis. That is, instead of 'Trace Gas Distributions and Variability of…', Section 4 could be called 'Diagnosis of monsoon transport in the extratropical lower stratosphere'.

We followed the suggestion and merged section 4.1. and 4.2. We also changed the title: Diagnosis of monsoon transport in the extratropical lower stratosphere

Section 5 (P85 & P86). The summary should be prose, not numbered statements. I suggest that you integrate the content of the numbered statements into the discussion paragraphs on P86. By this I mean, begin Section 5 with p86, l4 ("This study shows that the transport from the Asian monsoon region…". Within this paragraph, say how the results of the analysis support the discussion. Do the same for the 3 paragraph on P86.

Regarding P86, l13, I don't think the data demonstrate this pathway, they only suggest it.

Changed as suggested.

P87, l2. Ratios are not transported, air is. Please rewrite.

Changed to 'fraction'.

P87, l14-17. Suggest combining and shortening these 2 sentences: "We conclude that the observed increase in tropospheric influence in the ExTL above 370 K originates in a region with a high tropopause. The calculation of 50-day…"

Changed as suggested.

P87, l18-22. These 2 sentences say almost the same thing. Try combining: "In agreement with the tracer observations, the CLaMS trajectories show an increasing contribution of air originating in the Asian summer monsoon to the extratropical lower stratosphere during September."

Changed as suggested.

Notes on Figures
Fig. 1 caption. 'equally to a)'??

We changed the caption at numerous places.

Figure 3. White dots with black error bars?? Can't see this.

We removed the error bars and added them separately as black crosses.

Fig. 5 caption. 'trajecetories'

Corrected

Fig. 7 caption. Time is shown in month/day, not UTC.

Corrected

Fig. 9 & 10 captions. Delete everything after '(WMO, 1957)'. That's a really ancient reference!

Corrected

Fig. 11 caption. The word 'day' is missing from '50 backward…'

Corrected.

Miscellaneous

Replace 'backward trajectory' with 'back trajectory' throughout.
In general, 1) delete words like 'Hereby' or 'Thus' or 'therefore' from the beginning of sentences, 2) change 'the presented study' to 'this study', 3) change '50 day trajectories' to '50-day trajectories', 4) don't use 'respectively'. It's either wrong or unnecessary in most cases, and 5) try to write in the 'active voice'. That is, instead of 'The effect of blah blah is investigated' try 'We investigate the effect of blah blah.'

We tried to correct the manuscript as suggested.

P67, l8. 'trajetory' (trajectory)

Corrected

Numerous places (please search), 'seperate' should be separate.

We corrected the word at various places.

P74, l9. 'Europa' (Europe)

Corrected.

P82, l17. 'Fligth' (flight)

Corrected.

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:
l. 58: "…The air within the Asian monsoon anticyclone is strongly affected by anthropogenic pollution, which originates from east Asian and Indian densly 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, refering 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 extents 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 clearify our definitions and terms in accordance with the new Fig.1:
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).

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 N_2O-O_3 relationship of our analysis to the stratosphere (i.e. O_3 > 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 N_2O 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 N_2O-O_3 relationship and only focused on the CO-O_3 relationship as well as the frequency distributions of the tracers in different regions as suggested. The former section 4.1. (N_2O-O_3 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-O_3 (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-O_3 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 ad 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 **(l.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-O_3-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.

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).

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

S. Müller[1], P. Hoor[1], H. Bozem[1], E. Gute[2,a], B. Vogel[3], A. Zahn[4], H. Bönisch[5], T. Keber[5], M. Krämer[3], C. Rolf[3], M. Riese[3], H. Schlager[6], and A. Engel[5]

[1]Institute for Atmospheric Physics, Johannes Gutenberg-University, Mainz, Germany
[2]Department of Atmospheric Chemistry, Max Planck Institute for Chemistry, Mainz, Germany
[3]Institute for Energy and Climate Research - Stratosphere (IEK-7), Forschungszentrum Jülich, Jülich, Germany
[4]Institute of Meteorology and Climate Research (IMK), Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany
[5]Institute for Atmospheric and Environmental Sciences, Goethe-University of Frankfurt, Frankfurt am Main, Germany
[6]Institute for Atmospheric Physics, German Aerospace Research Center (DLR), Oberpfaffenhofen, Germany
[a]now at: Department of Chemistry, University of Toronto, Toronto, Canada

Correspondence to: Stefan Müller
(stefan.mueller@uni-mainz.de)

Abstract. The transport of air masses originating from the Asian monsoon anticyclone into the extratropical upper troposphere and lower stratosphere (Ex-UTLS) above potential temperatures Θ = 380 K was identified during the HALO aircraft mission TACTS in August and September 2012. In-situ measurements of CO, O_3 and N_2O during TACTS Flight 2 on the 30 August 2012 show the irreversible mixing of aged with younger (originating from the troposphere) stratospheric air masses within the Ex-UTLS. Backward trajectories calculated with the ~~trajetory~~ trajectory module of the CLaMS model indicate that these tropospherically affected air masses originate from the Asian monsoon anticyclone. ~~From the monsoon circulation region these~~ These air masses are ~~quasi-isentropically transported above~~ subsequently transported above potential temperatures $\Theta = 380$ K from the monsoon circulation region into the Ex-UTLS where they subsequently mix with stratospheric air masses. The overall trace gas distribution measured during TACTS shows that this transport pathway ~~has~~ had a significant impact on the Ex-UTLS during boreal summer and autumn ~~.~~ 2012. This leads to an intensification of the tropospheric influence on the ~~Ex-UTLS with $\Delta\Theta >$ 30~~extratropical lower stratosphere with PV $>$ 8 ~~K (relative to the tropopause)~~ pvu within three weeks during the TACTS mission. ~~In~~ During the same time period a weakening of the tropospheric influence on the lowermost stratosphere (LMS) is determined. ~~Therefore, the~~ The study shows that the transport of air masses originating from the Asian summer monsoon region within the lower stratosphere ~~above Θ = 380 K~~ is of major importance for the change of the chemical composition of the

Ex-UTLS ~~from summer to autumn.~~ and significantly contributes to the flushing of the LMS during summer 2012.

1 Introduction

The impact of air masses originating from the Asian monsoon anticyclone on the trace gas composition of the upper troposphere / lower stratosphere (UTLS) ~~is not well known and therefore subject of current research (Dethof et al., 1999; Ploeger et al., 2013; Vogel et al., 2014, 2015; Park et al., 2007, 2008; Fadnavis et al., 2013, 20~~ ~~In general, the UTLS denotes the transition between the troposphere and the stratosphere (Gettelman et al., 2011). The tropopause, which suppresses exchange between the troposphere and stratosphere, is located within the UTLS region . In the tropics the UTLS is characterised by the slow diabatic ascent of air masses from the troposphere into the stratosphere within the Tropical Tropopause Layer (TTL) (Fueglistaler et al., 2009, and references therein).Contrary, diabatic cooling and descending air masses are typical for the stratospheric~~ has not yet been quantified and is only poorly known. The UTLS region (Fig.1) encompasses the global tropopause region and the lower part of the ~~Ex-UTLS (Holton et al., 1995). Within the Ex-UTLS the lowermost stratosphere (LMS) denotes the stratospheric part of the middle world (Hoskins et al., 1985), which is mainly characterised by exchange processes between the troposphere and stratosphere on time scales of days to weeks (Bönisch et al., 2009; Hoor et al., 2010). The LMS is vertically confined by the tropopause and potential temperatures~~ stratosphere up to potential temperature levels of $\Theta=$ ~~380~~430 K ~~(Holton et al., 1995) (or 400 K (Hegglin and Shepherd, 2007)). Above, the extratropical lower stratosphere (LS) is coupled with the tropical LS by the Brewer-Dobson-Circulation (BDC, Brewer, 1949; Dobson, 1956)~~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 ~~Ex-UTLS~~ extratropics via the upper stratosphere and lower mesosphere on time scales of several years (Butchart, 2014). In contrast, the shallow branch ~~links the tropical and extratropical stratosphere~~ of the BDC mainly affects the region between $\Theta = 380$ K and 430 K by quasi-isentropic transport and mixing ~~with~~ (Hegglin and Shepherd, 2007; Spackman et al., 2007; James and Legras, 2009; Birner and Bönisch, 2011). 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 Θ ~~between~~ $=380$ ~~and 450~~ K ~~(Spackman et al., 2007; James and Legras, 2009; Birner and Bönisch, 2011).~~ for the ascending part of the BDC. 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). A detailed knowledge of the relative strength of these transport pathways is of major importance to quantify the chemical composition of the Ex-UTLS, which in turn determines the radiative, dynamical and chemical impact of this atmospheric region on the climate system (Forster and Shine, 2002; Riese et al., 2012).

As shown in Bönisch et al. (2009) and Ploeger et al. (2013) the seasonality of stratospheric transport processes above $\Theta = 380\,\mathrm{K}$ leads to a seasonality of the chemical composition up to $\Theta = 430\,\mathrm{K}$ (the bottom of the tropical pipe) and a "flushing" of the lower stratosphere with young tropical air masses up to this level in summer and autumn (Hegglin and Shepherd, 2007). In this context, the Asian summer monsoon is an important pathway for the transport of tropospheric constituents into the stratosphere and ~~gains~~ has gained increasing attention in atmospheric research (Randel and Park, 2006; Randel et al., 2010; Park et al., 2007, 2008, 2009; Bergman et al., 2013; Fadnavis et al., 2013, 2014, 2015). ~~Generally, the~~ The Asian summer monsoon consists of a cyclonic flow and convergence in the lower troposphere and a large scale anticyclonic circulation in the upper troposphere between June and September centered over southern Asia (Randel et al., 2010). 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). Therefore, water vapour and tropospheric trace gases as carbon monoxide (CO) and methane (CH_4) are relatively high within the Asian monsoon anticyclone (Rosenlof et al., 1997; Park et al., 2004, 2007), whereas ozone (O_3) is relatively low (Park et al., 2008). The air within the Asian monsoon anticyclone is strongly affected by anthropogenic pollution, which originates from east Asian and Indian densly 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).

Since the monsoon tropopause is relatively high (Highwood and Hoskins, 1998), exchange processes between the troposphere and stratosphere in the region of the Asian monsoon may transport tropospheric constituents into the lower stratosphere to altitudes up to 20 km (Park et al., 2008). The pathway and strength of troposphere-to-stratosphere-transport (TST) in the tropopause region of the Asian monsoon are neither adequately understood nor quantified. In this context, the direct convective injection into the lower stratosphere (Rosenlof et al., 1997; Chen et al., 2012) and the ~~seperation of small anticyclcones~~ separation of small anticyclones from the main anticyclone, so called “eddy shedding“, is of major importance (Hsu and Plumb, 2000; Popovic and Plumb, 2001; Garny and Randel, 2013). The latter process can transport tropospheric trace gases and pollutants to mid- and high latitudes, where they mix with extratropical stratospheric air masses ~~(Roiger et al., 2011; Vogel et al., 2014). However, the~~ (Vogel et al., 2014). The impact of the Asian monsoon on the chemical composition of the northern hemispheric lower stratosphere cannot easily be ~~seperated from the general~~ separated from the upward transport of tropospheric air masses into the stratosphere in the tropics, which is regarded as the primary pathway for air masses entering the stratosphere (Holton et al., 1995). In the tropical lower stratosphere these ascending air masses are uplifted by the Brewer-Dobson circulation and zonally transported to higher latitudes by the shallow branch of the BDC.

In this paper we present in-situ trace gas measurements of CO, ~~nitrous oxide (~~N_2O (nitrous oxide), and O_3 in the northern hemisphere extratropical stratosphere up to potential temperatures Θ = 410 K obtained during the TACTS (Transport and Composition in the Upper Troposphere and Lower Stratosphere) aircraft campaign~~are presented. Analysis~~. The measurements are combined with analysis data of the ~~ECMWF (~~European Centre for Medium-Range Weather Forecast (ECMWF) and backward trajectories calculated with the trajectory module of the Chemical Lagrangian Model of the Stratosphere (CLaMS) (McKenna et al., 2002a, b; Konopka et al., 2010; Pommrich et al., 2014)~~are used for a more detailed interpretation of the measurements~~. ~~The focus is on the investigation of~~ We will focus on the ~~quasi-isentropic transport of "young stratospheric" air masses from the (sub-)tropics into the Ex-UTLS in~~ impact of tropospheric air from the monsoon region on the composition of the lower stratosphere ~~with Θ > 380 K~~on different timescales. "Young stratospheric" in this context denotes air masses, which are clearly located in the stratosphere ~~with the respective stratospheric chemical composition~~from a dynamical and chemical point of view, but which still exhibit chemical signatures of previous tropospheric influence (e.g. CO mixing ratios larger than the CO equilibrium mixing ratio of chemically processed (aged) stratospheric air masses of CO = 12.5 ± 2.5 ppbv (Herman et al., 1999; Flocke et al., 1999). Since the measurements were performed in boreal summer and autumn 2012 within a period of four weeks in a wide range of the northern hemispheric UTLS, the potential influence of the Asian monsoon on this transport pathway and therefore on the trace gas composition of the Ex-UTLS is studied. In the following we will use the term lower stratosphere (LS), when we refer to the stratospheric part of the Ex-UTLS. This refers to the lower stratosphere above the ExTL up to Θ = 430 K, which is different from the classical definition of the LMS.

The paper is organised as follows: Section 2 gives an overview of the ~~analysed measurements, model data, and backward trajectory calculations~~measurements and methods applied. Section 3 presents a case study of TACTS-Flight 2 (30 August 2012), which illustrates the identification of irreversible ~~in-mixing~~ mixing of young stratospheric air masses into the ~~Ex-UTLS~~ extratropical stratosphere above Θ = 380 K. Tracer-tracer-scatterplots of TACTS-Flight 2 are analysed in Sect. 3.2. Section 3.3 shows the results of backward trajectory calculations, which indicate that transport and mixing from the Asian monsoon anticyclone into the Ex-UTLS cause the measured mixing event. ~~Section~~In section 4 ~~subsequently analyses the influence of the identified transport path on the trace~~

~~gas distribution of the northern hemispheric Ex-UTLS during TACTS~~ we will provide strong evidence that the Asian summer monsoon strongly affected the mid-latitude lower stratosphere in summer 2012. ~~The investigation of the in-situ measured N_2O-O_3-scatterplot in Sect. ?? supports the before drawn conclusions.~~ A summary and discussion of the results (Sec. 5) and a conclusion (Sec. 6) closes the paper.

2 Project overview, measurements, and backward trajectory calculation

2.1 The TACTS campaign 2012

~~In-situ~~ Our study is based on in-situ measurements performed during the first atmospheric science ~~mission~~ missions with the new German research aircraft HALO in August / September 2012 ~~named~~ TACTS (Transport and Composition of the UTLS) ~~/~~ and ESMVal (Earth System Model Vaildation)~~provide the basis for the presented study. During 13 mission flights with a duration between 8 and 10 hours for each flight, a large dataset of high spatial and temporal resolution of mainly in-situ trace gas data was collected within the Ex-UTLS~~. This paper focuses on ~~six research flights extending from 15°N to 70°N and 25°W to 15°E. ESMVal Flight 7 (23 September 2012), which was performed in the same area, is additionally considered in the analysis.During these flights 65 hours of measurement data in the Ex-ULTS were collected, including 40 hours in the extratropical stratosphere.~~eight research flights covering the Ex-UTLS between 15N to 70N and 25W to 15E with typically 8-10 hours per flight (see supplement).

The measurements of TACTS ~~were examined during~~ can be subdivided into two phases. The ~~inital~~ first phase with approx. 35 flights hours covered the time period between the 30 August and 5 September 2012. The end phase was performed from the 23 September until 26 September 2012. The ~~time lag between both phases is used to investigate the seasonal variation of the trace gas composition of~~ 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 ~~in late summer and early autumn 2012 as shown in Sect.~~ between both phases will be compared in section ~~4.~~ 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.

2.2 In-situ trace gas measurements

The study is mainly based on in-situ data from the TRIHOP (CO and N_2O) and FAIRO (O_3) instrument aboard HALO as described in the following subsections. Basic meteorological and avionic data are taken from the Basic HALO Measurement and Sensor System (BAHAMAS). For the following analysis all data are merged to one dataset with a time resolution of 10 s ~~or~~ (0.1 Hz~~, respectively,~~), corresponding to a horizontal resolution of 2.5 km.

2.2.1 TRIHOP in-situ measurements of CO and N_2O

The TRIHOP instrument is a three channel Quantum Cascade Laser Infrared Absorption spectrometer ~~capable of the subsequent measurement of CO, CO_2,~~ that measured CO, and N_2O during TACTS and ESMVal 2012 (Schiller et al., 2008). The instrument applies Quantum Cascade Laser Absorption Spectroscopy (QCLAS) in the mid-infrared with a multipass absorption cell (type White), which is kept at a constant pressure of P = 30 hPa and has a path length of 64 m and a volume of 2.7 l. During TACTS / ESMVal the instrument was in-situ calibrated approx. every 30 minutes during the flights against a secondary standard of compressed ambient air. The mixing ratios of the secondary standard were determined before and after the campaign in the laboratory against National Oceanic and Atmospheric Administration (NOAA) standards. ~~Therefore, the~~ The in-flight calibrations ~~allow~~ are used to identify and correct slow instrumental drifts in the post-flight data evaluation. The integration time for each species was 1.5 s at a duty cycle of 8 s, which ~~finally~~ limits the temporal resolution of the measurements. During TACTS and ESMVal TRIHOP CO (N_2O) data achieved a 2σ precision of 1.0 (1.1) ppbv and a stability of the instrument of 1.5 (2.2) ppbv ~~, respectively,~~ before applying the post flight data correction. Note that the stability is based on the mean drift between two subsequent calibrations, which were performed in intervals of 30 minutes during the flights. These instrumental drifts are corrected after the flights assuming linear drift, which leads to a reduced uncertainty. Hence, the total uncertainty relative to the working standard of 1.8 (2.5) ppbv can be regarded as an upper limit.

2.2.2 FAIRO in-situ measurements of O_3

FAIRO is a new accurate ozone instrument developed for use on board the HALO aircraft. It combines two techniques, the UV photometry (light absorption of O_3 at λ = 250 - 260 nm) with high accuracy and chemiluminescence detection with high measurement frequency. A UV-LED is used as a light source for the UV photometer, which can be controlled well (in contrast to Hg lamps) for constant light emission. The 1-sigma precision is 0.08 ppbv at a measurement frequency of 4 s and a cuvette pressure of 1 bar and the total uncertainty is 2%. The chemiluminescence detector shows a measurement frequency of 12.5 Hz and a precision of 0.05 ppbv (at 10 ppbv absolute, a measurement frequency of 5 Hz, and a pressure of 1 bar) (Zahn et al., 2012).

2.3 ECMWF (meteorological data)

Global meteorological analysis data (T1279L91) of the ECMWF (European Centre for Medium-Range Weather Forecast) are used to interpret the meteorological situation during TACTS Flight

2 as shown in Sect. 3.1. The meteorological fields are available every 3 hours interpolated onto a regular grid of 0.5° horizontal spacing.

2.4 CLaMS-TRAJ (backward trajectories)

The trajectory module CLaMS-TRAJ of the Chemical Lagrangian Model for the Stratosphere (CLaMS) is used to calculate backward trajectories at the positions of the in-situ observations (McKenna et al., 2002a, b; Konopka et al., 2010; Pommrich et al., 2014). The trajectories are initialised every 10 seconds along the flight paths and calculated 50 days back in time. For this purpose ERA-Interim reanalysis data (Dee et al., 2011) with a resolution of 1° and 60 vertical levels from the surface up to 0.1 hPa are interpolated in time and space on the starting point of the backward trajectories. Meteorological data, e.g. pressure, temperature, altitude, Θ and potential vorticity (PV) are available every 1 hour along the backward trajectories. The horizontal motion of the trajectories are driven by horizontal winds from the ERA-Interim reanalysis data. ~~For vertical motion the diabatic mode of CLaMS-TRAJ is used. Hereby diabatic~~ Diabatic heating rates are ~~calculated as~~ used to calculate vertical velocities in the UTLS region for pressure levels lower than 300 hPa (Ploeger et al., 2010). Below P > 300 hPa a pressure-based hybrid vertical coordinate is applied (Pommrich et al., 2014).

3 Case study: TACTS Flight 2 on 30 August 2012

TACTS flight 2 on 30 August 2012 was performed over western Europe and the eastern Atlantic with departure and landing in Oberpfaffenhofen near Munich (see Fig. 2). An eastward propagating trough over western ~~Europa~~ Europe was crossed several times on pressure levels between 220 and 130 hPa equivalent to flight altitudes between 11.5 and 15 km. On the highest flight levels on 150 and 130 hPa, ~~respectively,~~ air masses with potential temperatures Θ between 380 and 410 K were sampled. Potential vorticity (PV) derived from ECMWF data ~~are~~ ranges between 8 and 14 pvu for these flight sections (Fig. 2(a) and (b)). Hence, these air masses are located above the dynamical tropopause, which is typically defined by ~~PV isolines between 1 and 4~~ PV-values from 1-4 pvu in the extratropics (Hoskins et al., 1985; Randel et al., 2007). In the subtropics above $\Theta = 340$ K, PV at the tropopause varies between 2 and 5 pvu (Kunz et al., 2011).

3.1 Irreversible mixing within the stratosphere

The time series of in-situ O_3, CO and pressure data for TACTS Flight 2 are shown in Fig.2(c). The CO data indicates three sections with mixing ratios lower than 40 ppbv during the flight, which is below CO mixing ratios in the northern hemispheric free troposphere ranging from 50 to 130 ppbv (Kumar et al., 2013). This indicates that the respective flight sections were ~~performed~~ within the stratosphere. Accompanying ~~O_3 >~~ ozone values exceeding 200 ppbv (Strahan, 1999) and PV values larger than 5 pvu (cf. Fig. 2) support this conclusion.

During the last two flight segments from 10:30 until 12:15 UTC and 13:00 until 14:00 UTC, ~~respectively, partly chemically processed~~ stratospheric air masses with CO ranging from 20 to 30 ppbv were sampled. These values are larger than the stratospheric equilibrium of CO = 12.5 $\pm$ 2.5 ppbv ~~(Herman et al., 1999; Flocke et al., 1999), which is reached by the CO degradation in the stratosphere on time scales~~ which establishes as a result of CO production from methane and chemical degradation of CO on the time scale of several months ~~(Xiao et al., 2007)~~(Flocke et al., 1999). Therefore, CO between 20 and 30 ppbv indicates neither pure tropospheric nor completely CO degraded stratospheric air masses. Instead, ~~mixing ratios in this range arise either due to~~ such mixing ratios are the result of either irreversible mixing of tropospheric ~~and stratospheric air masses or by~~ with stratospheric air or from the CO degradation of a former tropospheric air mass in the stratosphere (or a combination of both). In the following the correlation of CO with O_3 during TACTS Flight 2 is analysed to further examine the tropospheric influence in the Ex-UTLS for Θ between 380 and 410 K.

~~In general, without~~ Without any exchange between the troposphere and stratosphere, the CO-O_3-scatterplot would form an L-shape (Fischer et al., 2000). Irreversible mixing of tropospheric (characterised by low O_3) and stratospheric (characterised by low CO) air masses appear as „mixing lines“, which connect the mixed reservoirs on the tracer-tracer-scatterplot (Hoor et al., 2002). 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). ~~In this study in-situ observed mixing lines on the CO-O_3-scatterplot are used for the first time to investigate mixing and transport processes in the Ex-UTLS above Θ = 380 K.Figure~~ As shown in Fig.3 (a) ~~shows the stratospheric part (O_3~~the observations indicate irreversible mixing above Θ ~~>~~= ~~200 ppbv) of the CO-O_3-scatterplot of TACTS Flight 2 color coded with potential temperature Θ~~380 K. Since this is above the middleworld isentropic cross tropopause mixing is most likely not the driving mechanism for the observed mixing lines. Low CO and high O_3 are accompanied by high values of Θ. From the fact that all CO data points are well above the stratospheric equilibrium of CO = 12.5 $\pm$ 2.5 ppbv (Herman et al., 1999; Flocke et al., 1999), tropospheric influence on time scales of weeks can be deduced for all probed air masses. ~~Additional, partly linear correlated data points with enhanced CO mixing ratios on the generally curved correlation indicate recent irreversible transport of tropospheric air into the Ex-UTLS.~~

During TACTS Flight 2, five mixing lines (~~these mixing lines are in the following shortened with ML1, 2, 4, 5~~ML) at potential temperatures $\Theta \geq 380$ K ~~are~~ can be identified. These are marked by individual colors in Fig. 3 (b). The respective colors in Fig. 2 mark the location of the measured mixing lines on the flight path (Fig.2 (a) and (b)) and along the time series of CO and O_3 (Fig.2 (c)). ML 1, 2, 4, 5 ~~are measured~~ were encountered in the center of the trough (Fig. 2 (a)), ML 3 is sampled at the trough edge further south. The five ~~identified~~ mixing lines correspond to individual flight sections along the flight track with durations of typically 15 minutes for each mixing line (see

Fig. 2(c)). Table 1 lists the minimum and maximum values of CO, O_3 and Θ for each individual mixing line. Also shown are the number of data points, the flight distance, and R^2 from a linear fit regression for the respective mixing line. The applied fit is based on Press et al. (1992) and accounts for errors in both, x- and y-direction. The observed CO and O_3 ~~variability of each mixing line larger~~ range (CO: $\geq$ 4 ppbv~~and~~ , O_3: $\geq$ 60 ppbv~~, respectively,~~) of each mixing line is larger than the total measurement uncertainty of both species. Therefore, measurement artefacts ~~that could yield~~ causing these mixing lines can be excluded. $R^2 \geq 0.89$ indicates that linearity for all mixing lines is justified. In general, ML 3 ~~is measured at larger~~ constitutes of higher CO and lower O_3 mixing ratios and exhibits a larger difference between the minimum and maximum values of both species compared to ML 1, 2, 4, 5. ~~Especially significantly~~ Significantly larger CO mixing ratios up to 40 ppbv compared to CO $<$ 30 ppbv for ML 1, 2, 4, 5 in combination with a higher tropospheric endmember (see Sect. 3.2) indicate shorter time scales for the transport and mixing of CO rich air masses from the troposphere into the stratosphere for ML 3 compared to the other mixing events. The fact that ML 3 is measured at the trough edge and at lower potential temperatures compared to ML 1 ,2, 4, 5 supports the assumption that different ~~processes~~ time scales are responsible for the formation of ML 3 and ML 1, 2 4, 5. In the following section the origin of the CO enhanced air masses in the extratropical lower stratosphere is further investigated with N_2O as an additional in-situ measured trace gas.

3.2 Mixing line analysis

~~The analysis of mixing lines on tracer-tracer-scatter plots allows the determination of the~~ This analysis seeks to determine the initial mixing ratios of the air parcels, which lead to the formation of mixing lines (Hintsa et al., 1998; Hoor et al., 2002). This requires knowledge on the initial mixing ratio of one ~~trace gas, if the mixing ratio of the second trace gas before mixing is known (Hintsa et al., 1998; Hoor et al., 2002). However, in most cases the~~ of the species involved. Since mixing ratios of ~~both tracers before mixing are unknown. Therefore, a precise determination of the trace gas mixing ratio prior to the mixing event is usually not feasible~~trace gases like O_3 and N_2O at the tropopause are fairly constant compared to their stratospheric gradient, they can be used to as initial estimate to analyze mixing across the tropopause. Nevertheless, mixing ratios just below the the tropopause of trace gases like O_3 and N_2O are fairly constant compared to their stratospheric gradient. Thus, the determination of the so called „tropospheric endmember“ is possible with the ~~above assumption . This is conducted for all mixing lines (see Sect. 3.1) based on the~~ assumption of a constant tropospheric value for one of the constituents involved. This approach has been applied to ML 1-5 on the basis of the CO-O_3- and N_2~~O-CO-scatterplot.~~O-CO-scatter plot.

Figure 4 displays ~~exemplarily~~ the tropospheric endmember approximation for ML 4. The calculation of the shaded confidence region is based on the FITEXY-approach described by Press et al. (1992), which accounts for uncertainties in x- and y-direction. O_3 at the tropopause varies between

60 and 120 ppbv, with a seasonal cycle and regional variations as well. For the endmember approximation in this section a mixing ratio O_{3Trop} = 100 ppbv is applied, which is a reasonable value for the northern hemispheric summer (Zahn and Brenninkmeijer, 2003; Thouret et al., 2006). Alternatively, N_2O can be used to determine a tropospheric endmember. Superior to O_3, N_2O is inert in the troposphere and thus has an almost homogeneous distribution in the global troposphere. The sinks of N_2O are entirely in the stratosphere, which leads to a weak gradient of N_2O at the tropopause. Therefore, the N_2O mixing ratio at the tropopause is well defined, which makes N_2O an appropriate tracer for tropospheric air masses (Assonov et al., 2013; Müller et al., 2015). In August / September 2012 the free tropospheric mixing ratio of N_2O was 325 ± 0.5 ppbv, which is applied as N_2O_{Trop} (www.esrl.noaa.gov/gmd/hats/combined/N2O.html). Table 2 lists the potential range of the tropospheric endmember of CO from the calculated confidence region (shaded area in Fig. 4) for O_{3Trop} = 100 ppbv and N_2O_{Trop} = 325 ppbv, respectively. The results can be summarised as follows:

- The tropospheric endmembers of CO for ML 1, 2, 4, 5 are in a range from 35 to 55 ppbv. This is at the lower limit and even below typical tropospheric CO mixing ratios.
- Endmembers of CO based on the CO-O_3-correlation are different from the respective N_2O-CO-endmembers.
- CO endmembers between 55 and 70 ppbv for ML 3 includes typical mixing ratios at the tropopause, even though CO is at the lower limit for tropospheric CO mixing ratios.

A tropospheric ~~endmember of CO below typical CO mixing ratios at the tropopause~~ CO endmember lower than typical tropopause values indicates that the respective mixing line is not the result of mixing between pure tropospheric and stratospheric air masses. (The term „pure” is used in this context to describe undiluted and ~~chemically unprocessed , therefore not CO degraded,~~ photochemically unprocessed tropospheric air masses.) For ~~those~~ these mixing lines the ~~determined~~ tropospheric CO endmembers do not ~~describe~~ reflect the original CO mixing ratio of the unmixed air mass. ~~Lower CO endmembers than found at the tropopause~~ CO endmember values lower than the range of CO mixing ratios at the tropoapuse can only arise from the chemical degradation of CO in the stratosphere or previous mixing or a combination of both. Therefore, the observed mixing lines are the result of mixing between two stratospheric air masses with different age. Note that O_3 variability (and chemistry) in the lower stratosphere leads to differences in the tropospheric endmembers of CO based on the CO-O_3- and N_2~~O-O_3-correlation~~O-CO-correlation. Thus, the formation of ML 1, 2, 4, 5 can only be caused by the irreversible mixing of stratospheric air masses with different age. In contrast, ML 3 indicates the irreversible mixing of pure tropospheric and stratospheric air masses. These findings are supported by the meteorological situation and location where the mixing lines were measured. Air masses that form ML 1, 2, 4, 5 are probed in the centre of ~~the investigated~~ a trough far away from the jetstream (see Fig. 2). ~~From the fact that~~ Since strong convective activity ~~cannot be identified in the measurement region during the flight,~~ was absent during the flights fast transport of ~~pure~~ tropospheric air masses up to $\Theta = 400$ K can be ruled out. ML 3 arises, in contrast to the other mixing lines, presumably as a consequence of the irreversible mixing of ~~pure~~ tropospheric and stratospheric air masses at the jet stream, since this mixing event is measured at the edge of the trough in a region with high windspeed and windshear (Pan et al., 2006).

3.3 Results of backward trajectory calculations

~~In the following the~~

We investigate the origin of ML 1, 2, 4, and 5, which were observed in the trough away from sharp isentropic PV gradients at the tropopause, ~~are investigated. For this purpose 50 days backward trajectories are analysed. These were~~ by analyzing 50-day backward trajectories calculated with the ~~trajectory module of the CLaMS-model according to Sect. 2.4.~~ CLaMS model. Variations of potential temperature Θ along the backward trajectories indicate diabatic processes in the history of the respective ~~probed~~ air masses. Figure 5 shows ~~the measured~~ measured potential temperature Θ along TACTS Flight 2 (30 August 2012) color coded with PV from ECMWF analysis data. Black dots indicate the maximum potential temperature along each individual backward trajectory. These values are typically 20 K higher than the ~~measured Θ~~ potential temperature at the position of the aircraft. This is in accordance with descending air masses in the extratropical stratosphere with a rate of approx. 0.5 K/day (Butchart, 2014). Red dots indicate the minimum Θ for trajectories which show a diabatic ascent ~~from their origin at $t_{Tra} <$ -50 days~~ prior to the time of measurement. These trajectories ~~appear preferably for probed air masses with relatively large PV~~ are primarily associated with air masses showing large PV values ($>$ 8 pvu~~in regions with the observed mixing lines from the previous sections~~) in regions where the mixing lines were encountered. This finding provides an indication that diabatic upward transport of tropospherically influenced air masses ~~in the measurement region is visible in the calculated~~ is reflected by the diabatic ascent of the backward trajectories. Figure 6 displays those trajectories ~~,~~ which show a ~~significant diabatic~~ potential temperature increase exceeding 5 K. These trajectories indicate an origin in the anticyclone of the Asian summer monsoon and subsequent transport to the measurement region in the Ex-UTLS above $\Theta = 400$ K. The position of the respective trajectories for -50 days $< t_{Tra} <$ -30 days in Fig. 7 shows that ~~almost all trajectories are~~ a large fraction of the air masses is located within the Asian monsoon anticyclone for $t_{Tra} <$ -30 days. The lowest potential temperatures along the trajectories (bluish colors) appear within the Asian monsoon region. As shown in Fig. 8 the trajectories spend a significant amount of time in the region, which is affected by the Asian monsoon anticyclone. The location of the anticyclone shows a large variability within the two months prior to the measurements (see supplement) extending from 20 N to 45 N during some periods. The longest residence time of trajectories (Fig. 8) is however found in those regions which can be associated with the core region of the Asian monsoon anticyclone (Bergman et al., 2013; Garny and Randel, 2015).

The backward trajectory calculation ~~suggest that~~ suggests that the diabatic ascent within the Asian monsoon transports ~~tropospherically influenced~~ tropospheric air masses up to altitudes between 16 and 18 km. Figure 9 shows that this process is accompanied by PV values rising above 5 pvu and Θ larger 400 K. These air masses are subsequently transported at $t \approx$ -30 days to the extratropics were they mix with aged stratospheric air masses (as measured).

The calculation of ~~50 days backward~~ 50-day back trajectories cannot provide unambiguous evidence that the transport of air masses from the Asian monsoon into the Ex-UTLS causes the occurence of mixing lines on the CO-O_3-correlation. ~~However, as mentioned before, CLaMS allows to calculate~~ CLaMS calculates the vertical motion using diabatic heating rates from ERA-Interim, which tend to have a relatively small vertical dispersion in the stratosphere compared to kinematic scenarios (Ploeger et al., 2010). Notably, Vogel et al. (2014) show that ~~40 days backward~~ 40-day back trajectories, calculated with the same setup of CLaMS as used in this study, agree with trace gas measurements during TACTS Flight 6.

~~In this study, trace~~ Trace gas measurements of TACTS Flight 2 and ~~50 days backward~~ 50-day back trajectories are consistent, since ~~:~~

CO-O_3 mixing lines indicate mixing at $\Theta > 380$ K. Endmember analysis shows that recent mixing at the tropopause cannot explain these mixing lines. The mixed air masses rather have experienced a significant CO degradation in the stratosphere. CLaMS trajectory calculations confirm this conclusion, since a transport time of $t \approx 30$ days from the Asian monsoon region into the Ex-UTLS is indicated by the backward trajectories. ~~Figure ?? shows that the filtered backward trajectories belong to data points on the CO - O_3 - correlation, which are located in the region of the identified mixing lines.~~

~~The only trajectories that indicate transport from the troposphere into the Ex-UTLS above $\Theta = 380$ K originate from the Asian monsoon region.~~ A significant contribution from the TTL for this region for TACTS Flight 2 cannot be identified based on the trajectory calculations. Thus, it is concluded that the Asian monsoon affects the trace gas composition of the Ex-UTLS for $\Theta > 380$ K during TACTS Flight 2. ~~Subsequently, the overall trace gas distribution of the Ex-UTLS measured during TACTS 2012 is investigated in the following section. It will be discussed, if the boreal Ex-UTLS during August and September 2012 is affected by air masses originating from the Asian monsoon region.~~

4 ~~Seasonality~~ Diagnosis of ~~N_2O, CO and O_3 during TACTS 2012~~monsoon transport in the extratropical lower stratosphere

4.1 ~~Trace gas distribution and variability of N_2O, CO and O_3~~

The effect of the Asian monsoon on the ~~overall~~ trace gas composition ~~in~~ of the Ex-UTLS ~~during August and September 2012 is investigated . Therefore, the trace gas distribution~~ is investigated by comparing the changes of N_2O, CO and O_3 from the ~~initial~~ early measurement phase (28 ~~August to~~ Aug - 5 ~~September 2012) and final~~ Sep) to the later TACTS flights (23 ~~to~~ Sep- 26 ~~September 2012)stage of TACTS, and the mixing ratio differences of both time periods, are displayed in a potential temperature Θ - equivalent latitude ϕ_e - coordinate system in Fig.~~ Sep). The tracer distribution for each period is calculated in bins of equivalent latitude and potential temperature. Figure ~~11. The data coverage of each bin for both periods is shown in Fig.~~ 10 shows the data coverage for each period. Figure ~~10. As can be seen from Fig. 11 , no significant changes for the long-lived trace gases~~ 11 shows the mean distributions for N_2O, CO and O_3 ~~during the flight campaign within the ExTL are evident~~in the early and later period and their differences. Also given is the location of the thermal and dynamical tropopause for each period as well as isolines of PV = 6 and 8 pvu.

Comparing the tracer distribution of the two phases reveals a slight increase of tropospheric tracers in the stratosphere beyond PV > 8 pvu. This region is clearly above the ExTL which extends to $\Delta\Theta = 30$ K above the local tropopause (Hoor et al., 2004, 2010). Above the ExTL with $\Delta\Theta > 30$ K (relative to the tropopause~~(Strahan et al., 2007)~~) an increase in N_2O ~~during TACTS~~ is observed. O_3 exhibits lower values for the same region during the final phase of the measurements. Both ~~indicates~~ tracers indicate an increasing impact of tropospheric air masses in the extratropical stratosphere above the ExTL. ~~This finding is also valid for the distribution of the relatively short-lived species CO, which displays slightly larger mixing ratios above the ExTL. Within the ExTL a decrease of CO during the campaign occurs. These findings are interpreted as follows:~~

~~Lower CO in the ExTL indicates a weakening of the tropospheric influence in the ExTL during TACTS between August und September 2012. This is not detectable in the long lived tracers N_2O and O_3 due to the long chemical lifetime of bothtrace gases in the lowermost stratosphere. Above the ExTL, the transport of relatively young air masses within the stratosphere at $\Theta > 380$ K is responsible for larger mixing ratios of tropospheric (~~In the tropopause region below 8 pvu the tracer signature is less clear: The distributions of both, CO and N_2O ~~and CO) and lower mixing ratios of stratospheric tracers (O_3) in the Ex-UTLS. At mid- and high-latitudes these air masses subsequently descend to lower potential temperatures. If the transport of air masses from the troposphere below Θ = 380 K into the stratosphere on short time scales was responsible for the enhancement of tropospheric signatures above the ExTL, larger CO in the ExTL would be appearent.~~

~~Both results are in accordance with previous studies regarding the Ex-UTLS region (Hoor et al., 2002, 2004, 2010; Bönisch et al., At this point it is hypothesized that the intensification of the jet stream during autumn is responsible for a weaker transport of tropospheric air masses into the ExTL (Haynes and Shuckburgh, 2000; Berthet et al., 2007; Sawa et al., 200 which subsequently leads to lower CO mixing ratios at the final stage of the TACTS measurements. Independently from this transport pathway, the transport of Asian monsoon influenced air masses (as measured during TACTS Fligth 2) by the shallow branch of~~ indicate a slight decrease closer to the ~~Brewer-Dobson-circulation~~ tropopause, however with a large variability, which is even more pronounced in ozone. To further investigate, if transport from the Asian monsoon anticyclone is responsible for this increase of tropospheric tracers above ~~$\Theta = 400$ K leads to a stronger tropospheric influence above~~ the ExTL ~~. To further investigate this, the full 50 day backward trajectory data set is analysed in~~ we analyzed ClaMS trajectories for the whole TACTS period. Fig. 12 ~~. Hereby the relative proportion of trajectories originating from~~ shows the percentage of 50-day backward trajectories originating in the Asian monsoon region (~~criterion: 20~~criteria: 25°N < TRA-latitude < ~~50~~40°N, 40°E < TRA-longitude < ~~150~~110°E and Tra-Θ > 360 K at t = -30 days) ~~is displayed in~~ using the same coordinate system as ~~the trace gas distributions in the top array of~~ Fig. 11. These thresholds were chosen according to the mean location of the geopotential anomaly of the anticyclone which is shown in Fig. ~~12. The mean residence time of the filtered trajectories in the region of the Asian monsoon between t~~13. It shows the mean location of the geopotential anomaly relative to the mean geopotential height during JJA in the monsoon area according to Bergman et al. (2013). The dark blue line indicates the monsoon location according to the threshold from Bergman et al. (2013) at 100 ~~=~~hPa (light blue for 200 ~~0 and -50 days is shown in the bottom array of~~ hPa). The red contour marks the zero line for anomaly using the mean geopotential height from summer 2012 as threshold to calculate the anomaly. The difference of the monsoon contribution according to the trajectories in Fig. 12 ~~. Trajectories for data points within the ExTL predominantly originate~~ also shows a bimodal pattern similar to the tracer observations. Trajectories originating from the Asian monsoon region ~~during the~~ affecting the ExTL are predominantly found for the initial phase of TACTS. ~~Above the ExTL, trajectories originating from the monsoon anticyclone are preferably found for data points during the final stage~~ In the lower stratosphere above the ExTL, roughly coinciding with the 8 pvu contour, trajectories indicating air masses from the Asian monsoon region are mainly found for the second phase of TACTS. The mean residence time ~~of those~~ (not shown) of these trajectories within the Asian monsoon anticyclone shows no significant difference for both regions and time periods. ~~These results clearly indicate that the Asian monsoon has impacted the Ex-UTLS during TACTS 2012. Further, the calculated trajectories give additional~~ The trajectories provide evidence that the measured increased tropospheric influence in the extratropical lower stratosphere above the ExTL arises due to air masses from the Asian monsoon region. ~~Less trajectories~~ A decreased fraction of air masses originating from the Asian monsoon for the later measurements within ExTL indicate, in accordance to lower CO and N_2O mixing ratios, a stronger transport barrier between the tropical troposphere and extratropical stratosphere.

4.1 ~~The N_2O-O_3 correlation~~

~~The previous section shows that the composition of the extratropical lower stratosphere has significantly changed from late August to September~~ These results clearly indicate that the Asian monsoon has impacted the extratropical stratosphere above the ExTL during TACTS 2012. ~~As shown in Hegglin et al. (2006), Hegglin and Shepherd (2007) and Bönisch et al. (2011), the seasonality of transport affects the correlation between O_3 and N_2O. More O_3 relative to N_2O indicates a stronger contribution from the tropical~~

~~production region of O_3 and thus tropical influence, whereas in winter diabatic downwelling from the stratosphere leads to a decrase of O_3 relative to N_2O. Therefore, an increased tropical and Asian monsoon influence should also affect the N_2O correlation in the same way by increasing O_3 relative to N_2O. If this assumption is correct, the overall N_2O - O_3 - correlation should inherently contain the information on the composition change during TACTS 2012. Relatively large O_3 mixing ratios on a given N_2O level should be related to a more tropical or monsoonal origin, thus be encountered rather during the late TACTS phase. Lower O_3 levels relative to N_2O are indicative for a stronger contribution of descending chemically processed stratospheric air masses in the Ex-UTLS. Such air masses in the extratropical lower stratosphere were predominantly measured during the early phase of TACTS.~~

~~To test this,~~ To further analyze the tracer evolution in the ~~lower stratospheric N_2O~~LS and the ExTL we analyzed the frequency distributions of the tracers in the stratosphere for PV values exceeding PV ~~-~~= ~~O_3 correlation was subdivided in a low and high O_3 part. Fig.~~8 ~~??(a) displays the N_2O-O_3 scatter plot of all northern hemispheric data points of TACTS 2012 filtered for air masses of the lowermost stratosphere with $O_3 > 350$~~pvu (Fig. ~~ppbv. The respective parts of the correlation were analysed according to the time when measured.Figure~~14). This relatively high PV value accounts for the fact that the PV at the high tropopause in the subtropics is at 5 pvu (Kunz et al., 2011). It further marks the transition from the ExTL, which is characterized by rapid transport from the local tropopause to the stratosphere (Hoor et al., 2010). For the ExTL we analyzed the PV range from 3-8 pvu (Fig. ~~??(b)shows the histogram of the time of measurement of the blue and red points~~ 15). The frequency distribution of selected tracers was subdivided into the different phases of TACTS. As indicated in Fig. ~~??(a). Significantly more tropically influenced air masses were measured in the later phaseof TACTS. This indicates an increasing influence of air masses from the tropical (or Asian monsoon) in the extratropical lower stratosphere during TACTS 2012.Additional histograms for trace gas mixing ratios for extratropical (red) and tropical (blue) data points~~ 14 (PV > 8 pvu), during the early phase (reddish colors) N_2O, CO and SF_6 are significantly lower than in the late phase. This clearly indicates an increase of tropospheric tracers accompanied with a decreasing age of air as indicated by the increase of SF_6~~, H_2O~~. Ozone shows a slight decrease over time with a large variability. Water vapour and total water (~~2~~ CH_4 + 2 x H_2O) ~~in Fig.~~ also show an increase over time. This finding also indicates a stronger contribution from the monsoon region which tends to moisten the mid latitude lower stratosphere during this time of the year (Randel and Jensen, 2013).

Histograms for trace gas mixing ratios in the ExTL in Fig. ~~?? support this conclusion. SF_6 exhibits larger mixing ratios for blue data points. This indicates a younger age of air for the later phase of the measurements (Bönisch et al., 2009).Larger total water and water vapour mixing ratios are detected predominantly in the later TACTS phase.For the season investigated, tropical stratospheric air masses at and above $\Theta = 400$ K exhibit larger H~~15, here defined as the region between 3 and 8 pvu (Hoor et al., 2010), are less clear. The frequency distributions of CO and N_2O ~~mixing ratios~~

~~compared to mid- and high latitudes (Ploeger et al., 2013). Globally the highest water vapour mixing ratios at~~ 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 $\Theta = \sout{390}350$ K ~~occur in the region of monsoonal circulations (Randel and Jensen, 2013). Therefore , larger water vapour and total water mixing ratios indicate a stronger impact of tropical or monsoonal influenced air masses.Larger CO~~(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 $\Theta > 380$ K is responsible for larger mixing ratios ~~on~~ of tropospheric (N_2O and CO) and lower mixing ratios of stratospheric tracers (O_3 ~~isopleths for the later observations in Fig. ?? are also indicative for an increasing tropospheric influence in~~) above the ExTL. At mid- and high-latitudes these air masses subsequently descend to lower potential temperatures. These results are in accordance with previous studies regarding the Ex-UTLS ~~during TACTS. These results show that the measured N_2O-O_3 correlation also contains the finding of a strengthening influence of the Asian monsoon (or tropics) on the Ex-UTLS above $\Theta = 380$~~region (Hoor et al., 2002, 2004, 2010; Bönisch et al., 2009; Hegglin et al., 2006). It is likely that strengthening of the jet stream in September weakens transport of tropospheric air masses into the ExTL (Haynes and Shuckburgh, 2000; Berthet et which subsequently leads to lower CO mixing ratios at the final stage of the TACTS measurements. Independent of this transport pathway, the transport of Asian monsoon influenced air masses (as measured during TACTS Fligth 2) by the shallow branch of the Brewer-Dobson-circulation above Θ ~~K during TACTS 2012.~~ = 400 K leads to a stronger tropospheric influence above the ExTL.

5 Discussion and Summary

The ~~measurements and backward trajectories~~ tracer measurements over Europe during TACTS 2012 and the backward trajectory analysis provide a coherent picture, indicating a significant influence of the Asian summer monsoon on the ~~Ex-UTLS over europe during TACTS 2012. A combination of methods is applied to obtain reliability in the stated conclusion~~mid-latitude UTLS. The combination of tools provide a consistent picture, which can be summarized as follows~~:~~.

~~Section 3:~~

1. Based on in-situ trace gas measurements, irreversible mixing of different air masses in the Ex-UTLS above $\Theta = 380$ K was observed during TACTS 2012.

2. The tropospheric endmember approximation shows that the observed irreversible mixing occured ~~between air masses of different stratospheric ages rather than between~~ rather between

stratospheric air masses with different tropospheric contributions than between undiluted tropospheric and stratospheric air masses.

3. Backward trajectories indicate that the younger of the mixed stratospheric air masses were affected by the Asian monsoon circulation.

~~Section 4:~~

4. An effect of the Asian monsoon on the LS over ~~europe~~ Europe is also evident from the overall trace gas distributions measured during TACTS 2012. These show that the Ex-UTLS composition over Europe ~~significantly changes~~ has significantly changed within 20 - 30 days during the campaign. Above the ExTL a stronger impact of tropospherically influenced air masses in late September compared to late August 2012 is ~~determined~~ indicated by larger CO and N_2O, and lower O_3 mixing ratios. ~~Above the tropopause within the~~ Within the ExTL a weakening of the tropospheric influence is indicated by decreasing CO.

5. Backward trajectories ~~originating from~~ indicate that the Asian monsoon ~~region for data points measured in the lowermost stratosphere predominantly appear~~ strongly affected the LS predominantely during the late phase of TACTS 2012. Within the ExTL such trajectories are preferably found during the initial phase of the campaign. ~~The analysis of the N_2O-O_3-correlation shows an increasing impact of „younger“ stratospheric air masses in the extratropical lower stratosphere during TACTS, which originate from the tropics or monsoonal regions.~~

~~The presented study indicates~~ Our study suggests that the transport of air masses from the Asian monsoon region into the extratropical stratosphere is a major driver of the ~~modification~~ change of the Ex-UTLS chemical composition during summer and autumn of the northern hemisphere (Hoor et al., 2005; Hegglin and Shepherd, 2007; Bönisch et al., 2009). ~~Thereby,~~ In-situ data of N_2O, CO and O_3 indicate a significant increase of the tropospheric impact in the extratropical lower stratosphere above the ExTL within a few weeks~~(above the ExTL) is observed with in-situ data of N_2O, CO and O_3~~. This finding ~~shows that also other atmospheric constituents within the Ex-UTLS beside~~ indicates that water vapour (Ploeger et al., 2013; Randel and Jensen, 2013) ~~are affected by~~ as well as other atmospheric constituents (and pollutants), which were not measured, are potentially affected by transport from the the Asian monsoon circulation.

~~The presented case study indicates~~ A case study based on in-situ data shows that air masses originating from the Asian monsoon are quasi-isentropically transported above $\Theta = 380$ K into the Ex-ULTS ~~.~~ during summer and autumn 2012. This confirms the results of model ~~calculations~~ simulations and satellite data of Ploeger et al. (2013), which show an enhanced water vapour transport from the tropics (and the Asian monsoon region into the extratropics during boreal summer above $\Theta = 380$ K. A relatively large effective diffusivity between the tropics and extratropics during summer and autumn for ~~Θ between~~ potential temperatures ranging from 380 ~~and~~ to 450 K is also in accordance ~~to the~~

~~results of the presented study~~ with our results (Haynes and Shuckburgh, 2000).

Further, Haynes and Shuckburgh (2000) calculate a decreasing effective diffusivity from summer to autumn for Θ between 350 and 370 K (13 - 15 km), which indicates an increasing transport barrier from the early to the late monsoon season at the jet location. The measurement of lower CO within the ExTL during the later phase of TACTS compared to the initial phase agrees with a stronger transport barrier for mixing across the tropopause below Θ = 380 K. In accordance, ~~?~~ Ploeger et al. (2015) diagnose on the basis of the PV-gradient a strong transport barrier that ~~seperates~~ separates the anticyclone from its surrounding and inhibits isentropic transport and mixing between Θ = 360 - 380 K. Our measurements also support the hypothesis of an efficient transport of tropospheric trace gases (e.g. CO) to the upper Asian monsoon troposphere (Bergman et al., 2013). From ~~there a significant ratio~~ the upper tropospheric monsoon region a significant fraction of these air masses is efficently transported and mixed into the lower stratosphere of mid- and high-latitudes at potential temperatures $\Theta > 380$ K (Berthet et al., 2007). Thus, our data shows that the Asian summer monsoon significantly affects the overall tracer distribution over Europe with increasing efficiency from August to September 2012.

6 Conclusions

In-situ measurements of CO, O_3 and N_2O during TACTS 2012 show a significant change of the trace gas composition over the course of four weeks in the Ex-UTLS up to Θ = 410 K. From August to late September 2012 a significant increase of N_2O and CO (and ~~decrease~~ decreasing O_3) indicate a stronger tropospheric contribution above the ExTL up to Θ = 410 K. Decreasing CO mixing ratios in the ExTL below Θ = 370 K indicate a weakening of the quasi-isentropic transport across the tropopause at the jetstream into the lowermost stratosphere (Pan et al., 2006). Therefore, it is concluded that the observed increase of the tropospheric fraction above the ExTL is not caused by quasi-isentropic cross tropopause transport at the jetstream. ~~Rather,~~ We conclude that the observed increase ~~of tropospheric contribution must have originated in~~ in tropospheric influence in the ExTL above 370 K originates in a region with a high tropopause. The calculation of ~~50 days backward~~ 50-day back trajectories with CLaMS-TRAJ shows that the Asian summer monsoon significantly influences the composition of the Ex-UTLS during TACTS 2012. In agreement with the tracer observations~~these trajectories indicate an increasing fraction of air masses originating from~~ , the CLaMS trajectories show an increasing contribution of air originating in the Asian summer monsoon ~~in~~ to the extratropical lower stratosphere ~~from August to~~ during September 2012. Notably, the trajectories exhibit a mean residence time of t > 200 h in the Asian monsoon anticyclone in the last 50 days before the measurements. Within the monsoon circulation the trajectories slowly rise up to $\Theta > 400$ K. As shown in a case study in Sect. 3, the ~~monsoonally influenced~~ monsoon affected air masses are transported to the measurement region within 30 days. For the global tracer distribution increasing SF_6 values for the late phase of the measurements consistently indicate an ~~increase~~ increasing contribution of younger air in agreement with the analysis of the backward trajectories. Further, the simultaneously observed increase of (total) water vapour for the same air masses is in accordance with the horizontal tape recorder of H_2O at $\Theta = 390\,K$ caused by the Asian monsoon (Randel and Jensen, 2013). Thus, the Asian summer monsoon significantly ~~contributes to the observed „flushing "~~ affected the Ex-UTLS during TACTS 2012. This indicates, that the ASM might in general significantly contribute to the flushing of the boreal extropical lower stratosphere from summer to autumn (Hegglin and Shepherd, 2007) when the tropospheric signature is at maximum (Hoor et al., 2005; Bönisch et al., 2009).

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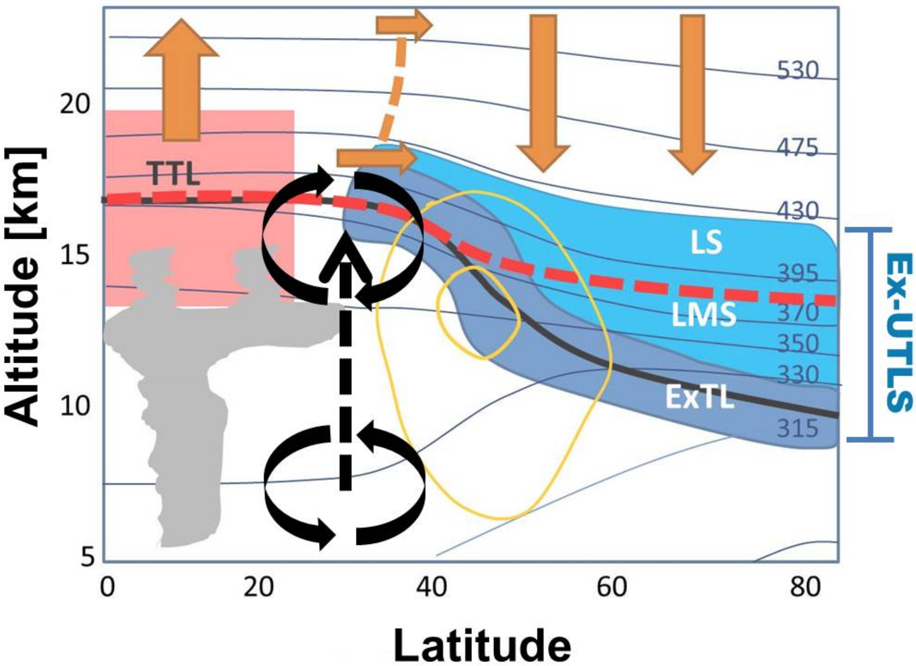

Figure 1. Illustration of the dynamical structure of the UTLS for the Northern hemispheric summer adapted from Fig. 2b of Gettelman et al. (2011) and Fig. 9 of Riese et al. (2014). The thick black line denotes the location of the thermal tropopause. Thin black lines are isentropes,the red dashed line highlights the 380 K isentrope. The location of the TTL is represented in light red. The Ex-UTLS is divided into the extratropical tropopause layer (ExTL) in dark blue and the lowermost (LMS) and lower stratosphere (LS) in light blue. The LMS and LS are vertically seperated by the 380 K isentrope. Yellow lines indicate the location of the jetstream. Orange arrows sketch the upwards transport of air masses within the TTL and descending air masses in the extratropical stratosphere. Both regions are linked by the deep (upper horizontal arrow) and shallow (lower horizontal arrow) within the stratosphere. The dashed orange line sketches the edge of the tropical pipe, which suppresses exchange between the tropical and extratropical stratosphere. The location of the Asian monsoon circulation is schematically shown in dark black.

~~Blue: CO-O_3-correlation of TACTS flight 2 (cf. Fig. 3). Black: Data points that were measured at $\Theta > 370$ K and PV > 4 pvu. Red: Data points which were measured at PV > 7.5 pvu und whose 50 day backward trajectories indicate an Θ_{min} at least 5 K below the measured Θ.~~

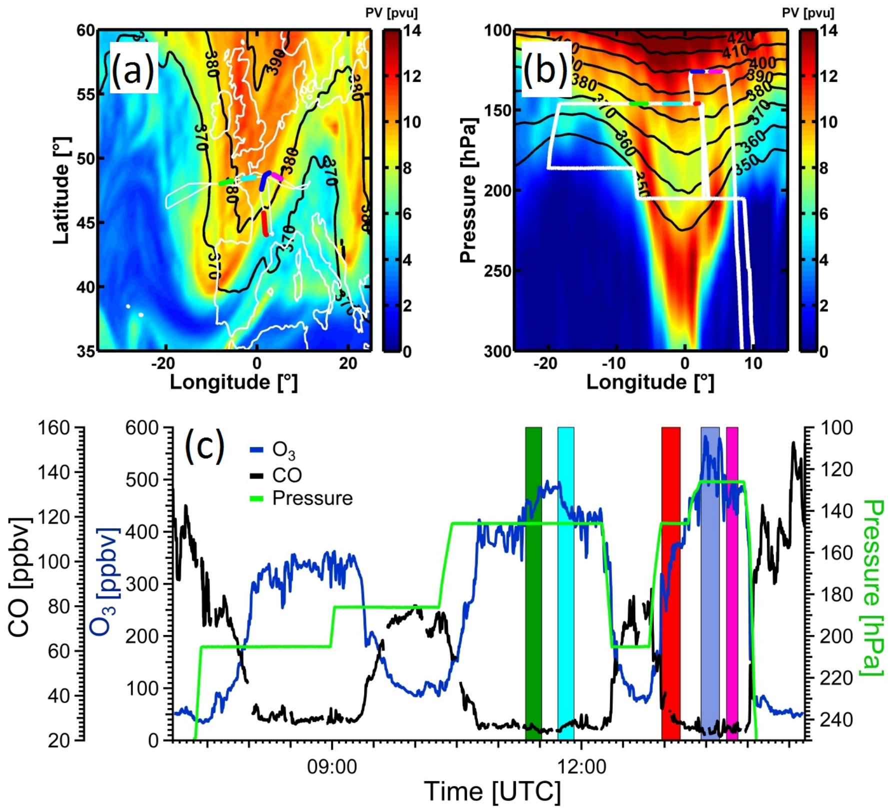

Figure 2. TACTS Flight 2 on 30 August 2012: (a) ECMWF data (15 UTC) at 150 hPa of potential vorticity (color) and potential temperature (black contour lines). The thick white line is the flight path of TACTS Flight ~~2, whereas different colors indicate flight legs with measured mixing lines~~ 2. ~~(see Sec. 3). (~~(b) Horizontal cross section of ECMWF potential vorticity data along 48.5°N~~equally to~~. Different colors in (a) and (b) indicate flight legs with measured mixing lines (see Sec. 3). (c) Time series of CO (black), O_3 (blue) and Pressure (green). Data points which form mixing lines are ~~marked in different colors~~ colored according to panel (a) and (b). The total uncertainty of CO and O_3 is mostly within the line thickness and therefore not shown (for individual error bars see Fig. 3 and Fig. 4).

~~(a): N_2O-O_3-scatterplot of all data points during TACTS 2012 with O_3 > 350 ppbv divided into higher (blue) and lower (red) O_3 mixing ratios on N_2O isopleths based on a linear fit regression. (b): Histogram of the measurement date of the red and blue data points on the N_2O-O_3-scatterplot. Purple areas display the overlap of both histograms.~~

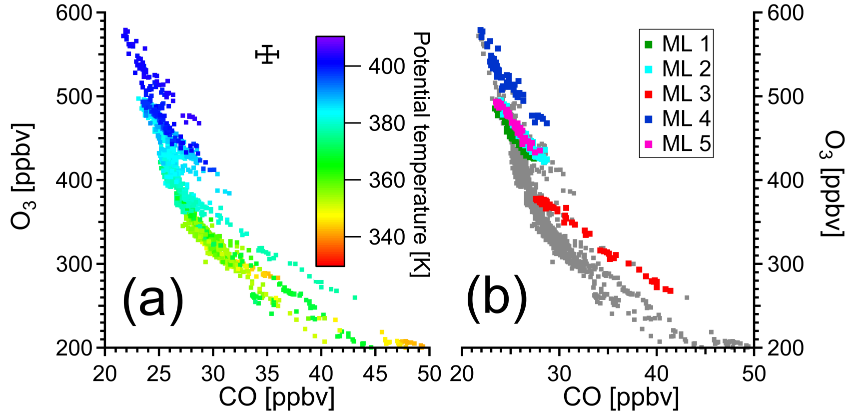

Figure 3. Stratospheric part of the CO-O_3-correlation for TACTS flight 2 on 30 August 2012: (a) Color-coded with measured potential temperature. (b) Mixing lines (ML) in different colors in accordance with Fig. 2. ~~For error bars see Fig~~Uncertainties of the CO and O3 data are separately shown as a black cross.~~4.~~

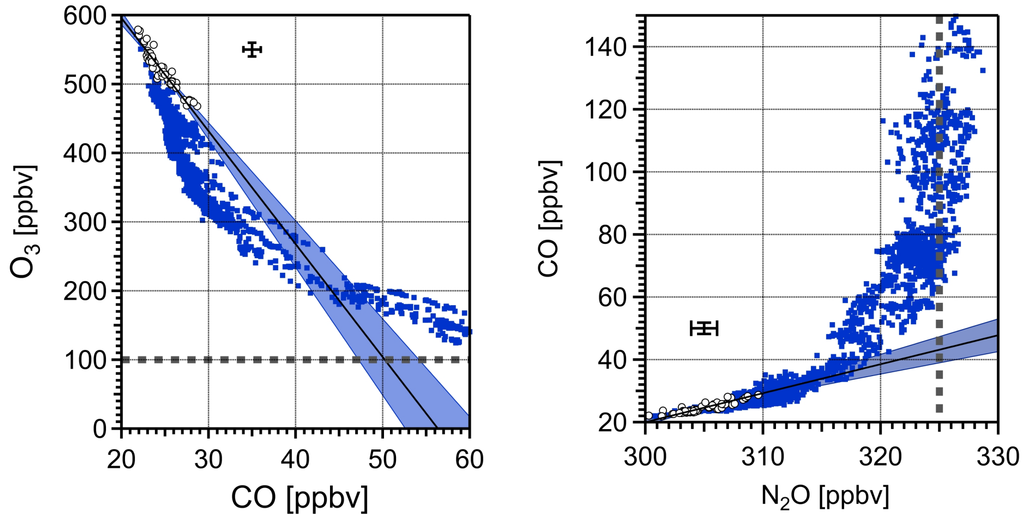

Figure 4. Linear fitting for ML 4 based on the FITEXY-routine described in Press et al. (1992). Blue dots: Scatter plot of CO and O_3 (left), and N_2O and CO (right), respectively. White dots~~with black error bars~~: Data points of ML 4 on the respective scatter plot. Solid black line: Linear fit with confidence region (shaded area). Dotted lines: Assumption of a tropospheric endmember of $R(O_3)_{Trop}$ = 100 ppbv (top) and $R(N_2O)_{Trop}$ = 325 ppbv (bottom). Uncertainties are separately shown as black crosses.

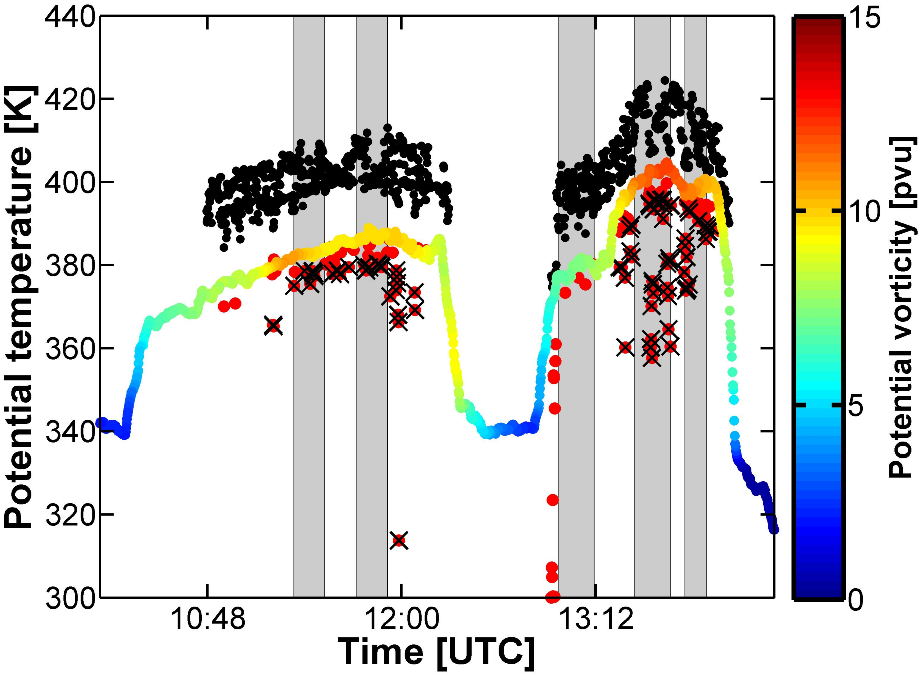

Figure 5. Solid line: Time series of the measured potential temperature Θ for the second half of TACTS Flight 2 with PV as color (from ECMWF data). Black dots: Maximum potential temperature along ~~50 days backward~~ 50-day back trajectories for all data points with $\Theta > 370$ and PV > 4 pvu at the measurement location. Red dots: Minimum potential temperature along ~~50 days backward~~ 50-day back trajectories for all data points with $\Theta > 370$ K, PV > 4 pvu, and minimum Θ along the backwards trajectories is smaller as Θ at the measurement. Red dots with PV > 7.5 pvu and minimum Θ at least 5 K smaller as Θ at the measurement are marked with a black cross (cf. Fig. 6).

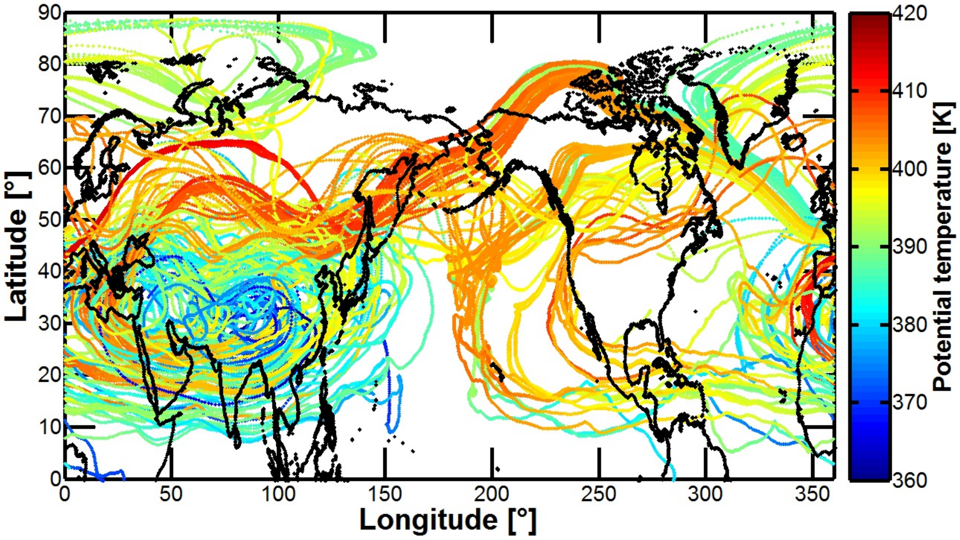

Figure 6. 50 day backward trajectories for all data points with $\Theta > 370$ K, PV > 7.5 pvu, and minimum Θ along the backwards trajectories is at least 5 K smaller as Θ at the measurement (cf. Fig. 5). Color: Potential temperature along the backward ~~trajeectories~~ trajectories based on ERA-Interim reanalysis data.

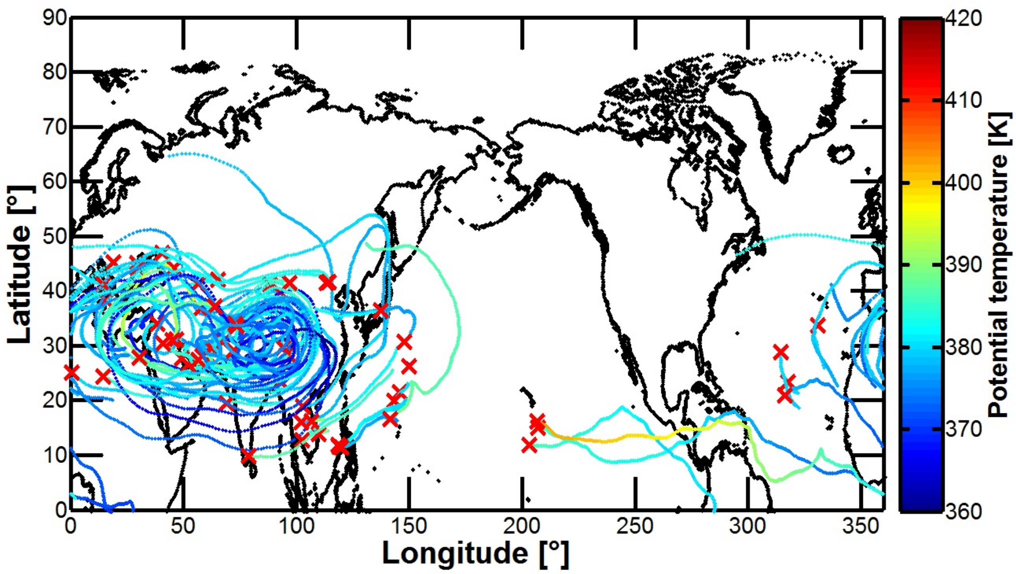

Figure 7. Pathway of ~~same~~ the trajectory ensemble as shown in Fig. 6 with $\mathrm{PV}_{Tra} < 5$ pvu. Red crosses mark the location of the trajectories where PV values exceed 5 pvu.

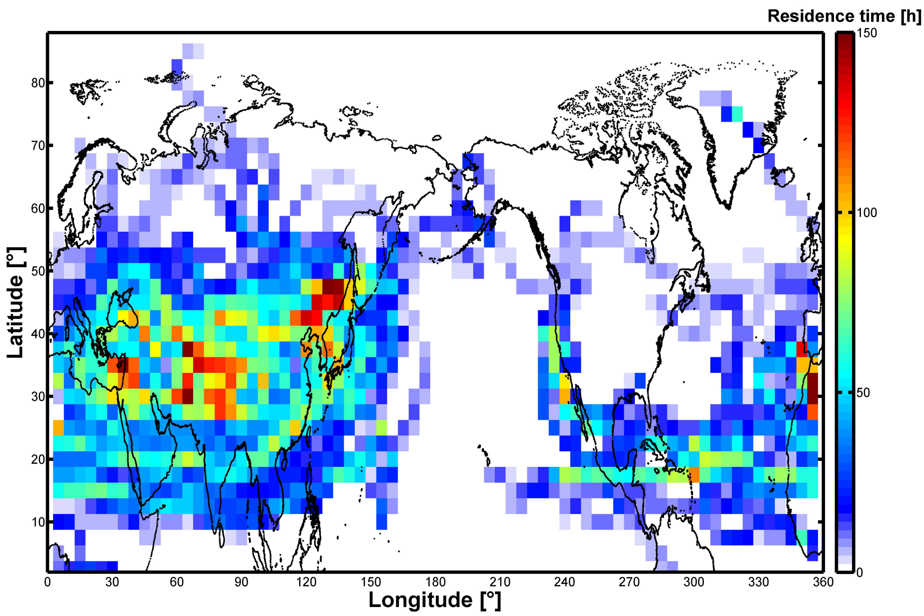

Figure 8. Residence time in hours of the trajectory ensemble as shown in Fig. 6 for -50 days < t_{Tra} < -30 days. (2.5° latitude and 5° longitude bins). Note the long residence time in the ascending region as indicated in Fig. 7.

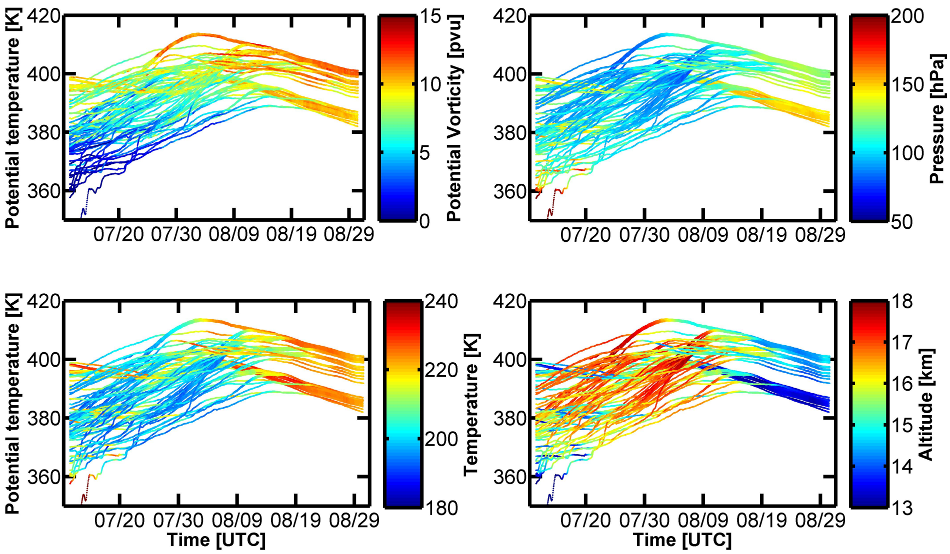

Figure 9. Potential temperature versus time ~~(in UTC)~~ along the ~~same 50 day backward trajectories~~ trajectory ensemble as shown in Fig. 6~~is shown~~. The color indicates the PV (top left), pressure (top right), temperature (bottom left), and altitude (bottom right) based on ERA-Interim reanalysis data.

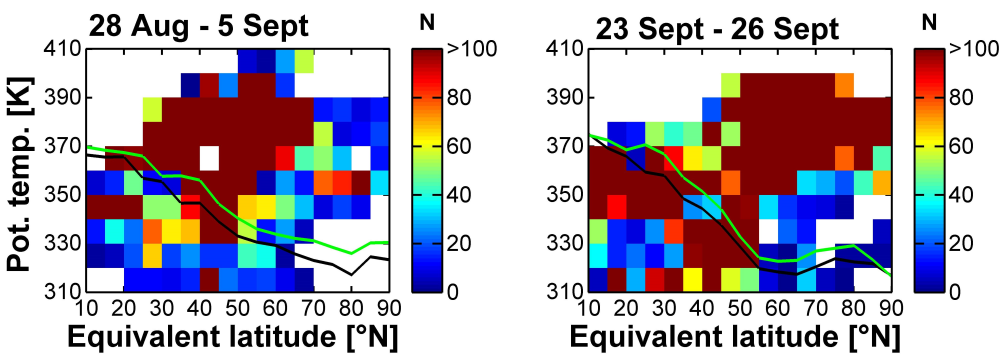

Figure 10. Data coverage (number of data points N in each bin) of TACTS 2012 in potential temperature Θ - equivalent latitude ϕ_{eq} - coordinates. Left: Data coverage of the initial phase of TACTS from the 28 August to 5 September. Right: Data coverage of the final phase of TACTS from the 23 to 26 September. The black and green line denotes the location of the dynamical (PV = 2 pvu) and thermal tropopause (WMO, 1957)~~, respectively, for the associated time period from ECMWF data~~.

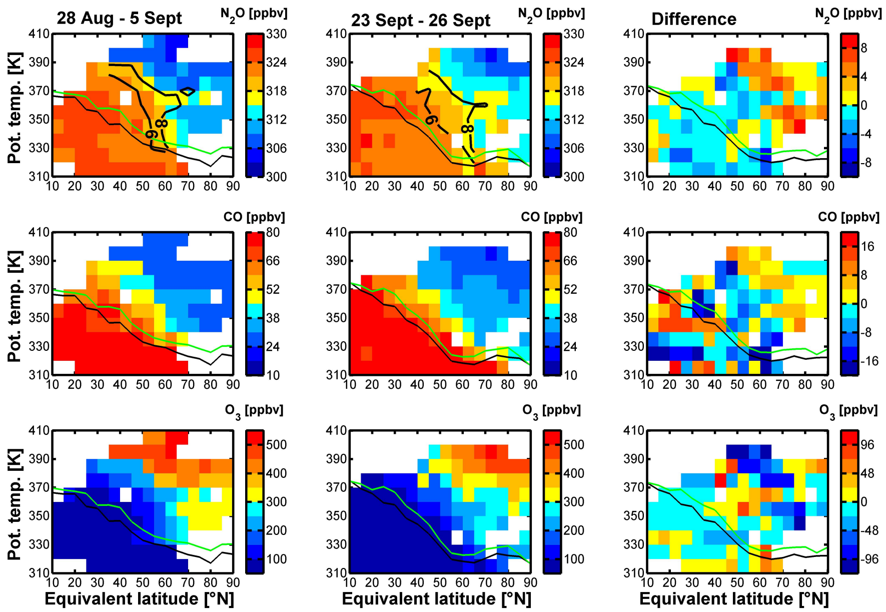

Figure 11. N_2O, CO, and O_3 distributions in potential temperature Θ - equivalent latitude ϕ_{eq} - coordinates for the TACTS 2012 campaign. Left: Distributions for the initial phase of TACTS from the 28 August to 5 September. Center: Distributions for the final phase of TACTS from the 23 to 26 September. Right: Changes in the trace gas distributions during the TACTS 2012 campaign (final minus initial phase). The black and green line denotes the location of the dynamical (PV = 2 pvu) and thermal tropopause (WMO, 1957)~~, respectively, for~~. The thick black line in the ~~associated time period~~ upper array denotes the 6 and 8 pvu isolines calculated from ECMWF data along the flight paths.

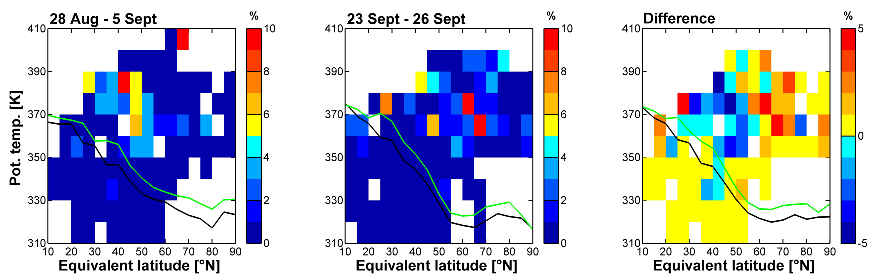

Figure 12. Same presentation as in Fig. 11 but with different color coding. ~~Top:~~ Proportion of data points for which 50 day backward trajectories calculated with CLaMS-TRAJ indicate an origin in the Asian summer monsoon anticyclone (Criterion: ~~20~~25°N < TRA-latitude < ~~50~~40°N, 40°E < TRA-longitude < ~~150~~110°E and Tra-Θ > 360 K for t = -30 days).~~Bottom: Mean residence time of trajectories in hours in the Asian monsoon anticyclone between t = -50 days and t_{meas} following the same criterion as in the middle array. Figures are filtered for bins with at least 10 data points.~~

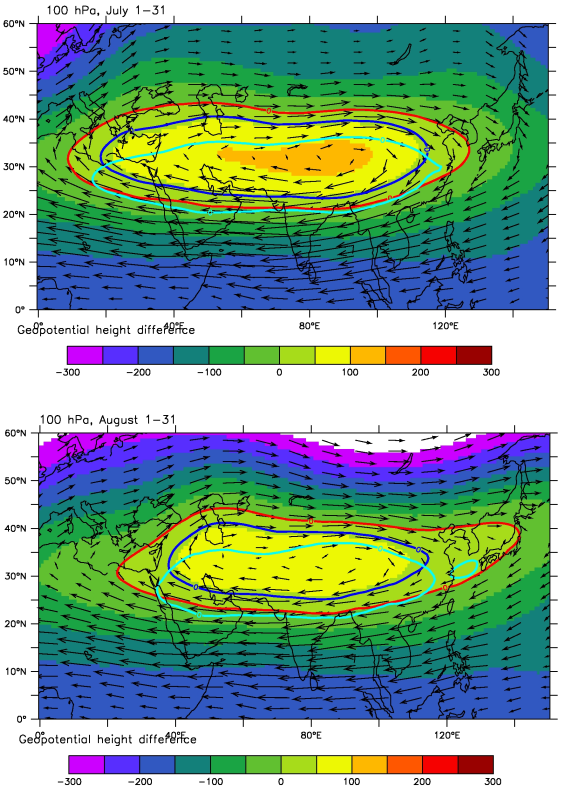

Figure 13. Geopotential height relative to the climatological mean for JJA following the method of Bergman et al. (2013). The dark blue line indicates the monsoon location according to the threshold from Bergman et al. (2013) at 100 hPa (light blue for 200 hPa). The red contour marks the zero line using the mean geopotential height from summer 2012 as threshold to calculate the anomaly.

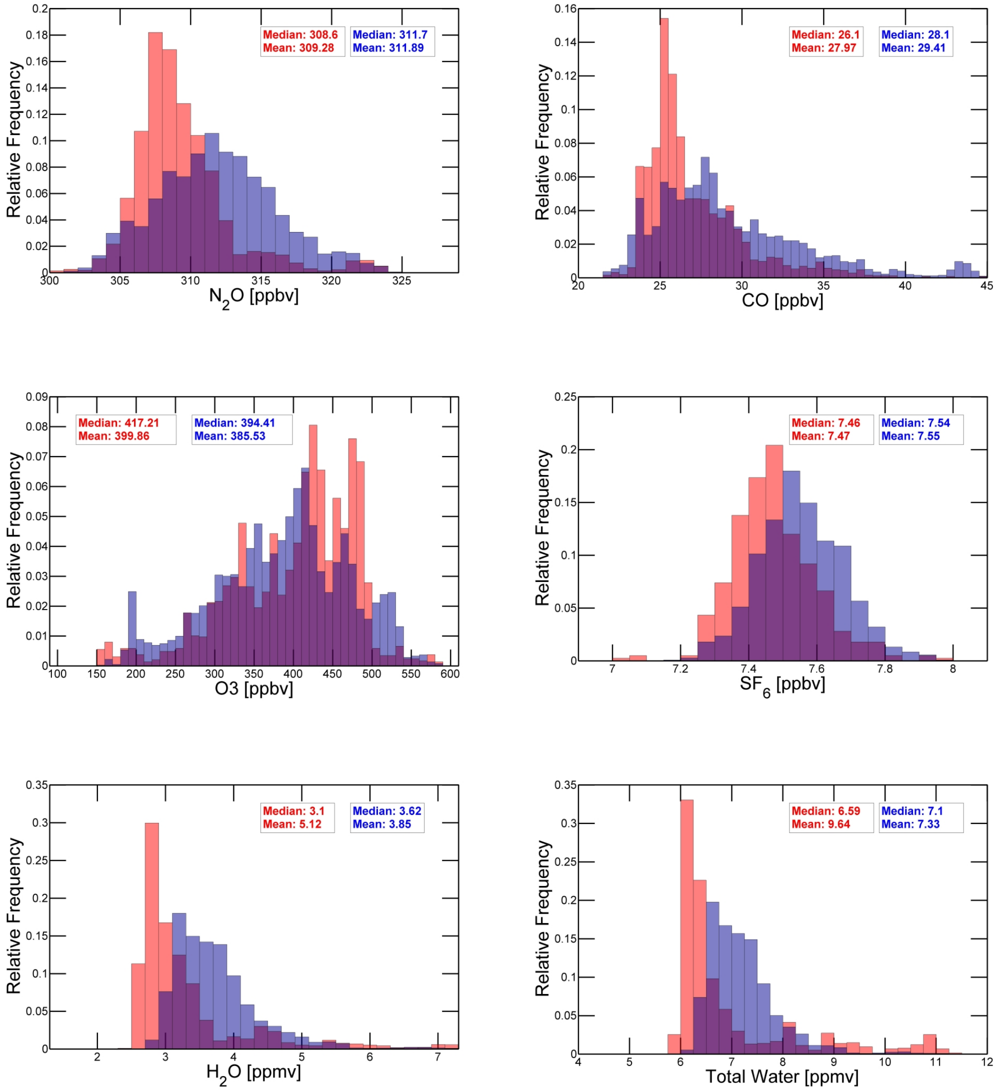

Figure 14. ~~Histogram~~ Relative frequency of ~~trace gas mixing ratios of SF$_6$~~ (~~left~~a) ~~, total water (H~~N$_2$O~~+ 2 CH$_4$~~, (b) CO, (~~center~~c) O$_3$ and (d) SF$_6$, (e) H$_2$O~~of the red and blue~~, (f) Total Water, for data points ~~on~~ with PV > 8 pvu. Redish colors denote the ~~N$_2$O-O$_3$-scatterplot in Fig~~histogram for the initial phase from the 28 August to 5 September, bluish colors for the final phase of TACTS from the 23 to 26 September. ~~?? (a).~~ Purple areas display the overlap of both histograms.

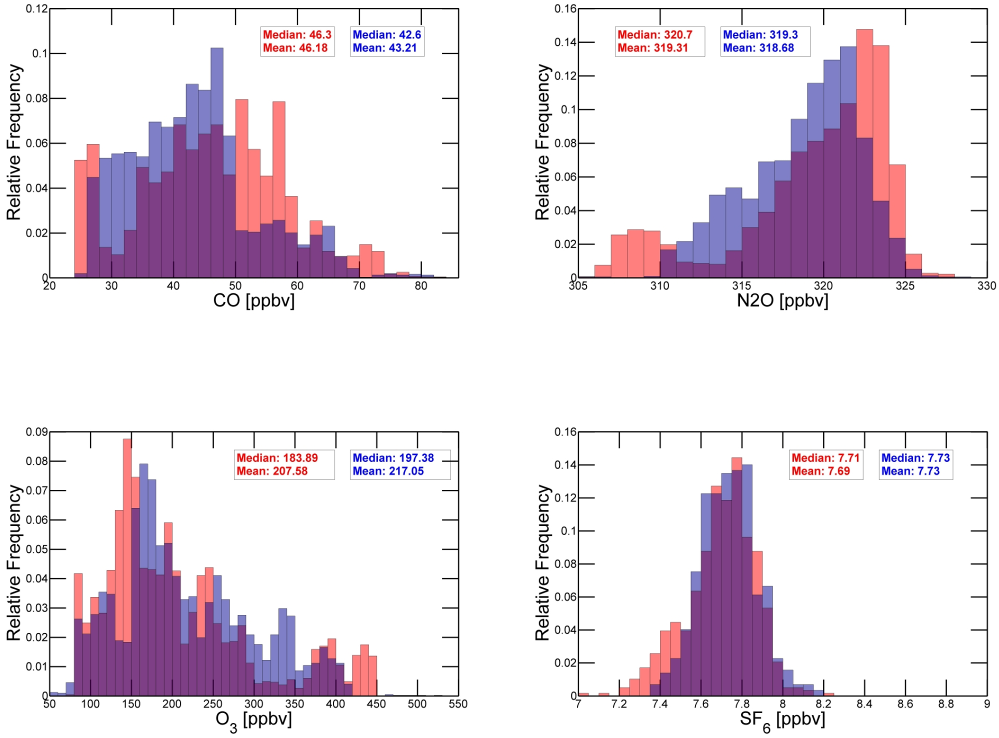

Figure 15. ~~CO-O$_3$-scatterplot for the same data points~~ Same as ~~shown in the N$_2$O-O$_3$-scatterplot in~~ Fig. ~~??~~ 14 (~~a~~without H_2O and Total Water histograms) ~~. Left: Blue~~ for data points ~~on top of red data points~~with PV ranging from 3 to 8 pvu.~~Right: Red data points on top of blue data points.~~

Table 1. Minimum and maximum values of potential temperature, and CO and O_3 mixing ratios, respectively, for every mixing line. Additonally the number of data points, the flight distance, and R^2 based on a linear regression is listed.

Mixing line	Line 1	Line 2	Line 3	Line 4	Line 5
Θ_{min} [K]	381,5	385,3	377,0	399,0	396,5
Θ_{max} [K]	384,5	389,0	382,0	405,0	401,0
CO_{min} [ppbv]	23,4	23,9	23,7	21,9	23,5
CO_{max} [ppbv]	27,4	28,7	41,4	28,7	28,0
$O_{3\ min}$ [ppbv]	425,59	421,75	248,27	467,82	433,01
$O_{3\ max}$ [ppbv]	485,56	495,11	377,92	583,16	493,54
Date points	71	71	60	65	51
Flight distance [km]	142	142	120	130	102
R^2	0,96	0,96	0,99	0,89	0,96

Table 2. Tropospheric „endmembers“ for every mixing line based on the tracer-tracer-correlations of O_3 and CO, and N_2O and CO, respectively (cf. Fig. 4). The values are based on the intercept between the calculated confidence regions with the assumed mixing ratio of O_3 and N_2O at the tropopause in Fig. 4.

Mixing line	Line 1	Line 2	Line 3	Line 4	Line 5
$R_{Trop}(O_3)$= 100 ppbv					
CO_{min} [ppbv]	42,0	45,9	59,3	47,2	44,5
CO_{max} [ppbv]	50,9	52,7	62,2	54,0	55,1
$R_{Trop}(N_2O)$= 325 ppbv					
CO_{min} [ppbv]	35,1	35,4	54,1	38,8	35,2
CO_{max} [ppbv]	44,2	39,8	68,0	46,2	47,3