

## 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 N<sub>2</sub>O-O<sub>3</sub> 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 N<sub>2</sub>O-O<sub>3</sub> correlation and instead present the frequency distribution of N<sub>2</sub>O, CO, O<sub>3</sub> and SF<sub>6</sub> 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, I5. Not 'Contrary', you mean 'In contrast,...'*

Changed as suggested.

*P68, I10. I would say the LMS is vertically defined by the tropopause...*

Changes as suggested (confined → defined)

*P68, I20-23. This is a 1-sentence paragraph. It belongs at the end of the previous paragraph.*

We rephrased the section.

*P69, I1. 'gains' should be 'has gained'*

Changed as suggested (gains → has gained)

*P69, I3. Delete 'Generally'*

Changed as suggested (Generally deleted)

*P69, I 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, I9-13. 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 missions with the new German research aircraft HALO in August / September 2012 TACTS (Transport and Composition of the UTLS) and ESMVal (Earth System Model Validation). 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, I3. '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, I7. Change to 'a 3 channel...spectrometer that measures CO, CO2, and N2O...'*

Changed as suggested:

*P72, I15. 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, I17 & I19. Delete 'finally' and delete 'respectively'*

We deleted „finally” and „respectively”

*P74, I2. Delete 'Hereby' and change to 'Diabatic heating rates are used to calculate vertical velocities...'*

Changed as suggested:

*P74, I11. Change to 'On the highest flight levels of 150 and 130 hPa, air masses with...'*

Changed as suggested:

*P74, I23. Delete 'performed'.*

We deleted „performed”

*P75, I2. Change 'respectively, partly chemically processed...' to 'partially processed'*

The corresponding section was rephrased.

*P75, I4-7. 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 CH<sub>4</sub>-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, I20-22. 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, I 28. Change this sentence to 'Linearly correlated data points with enhanced CO...'*

Changed as suggested:

*P76, I3. Change this to 'During TACTS Flight 2, 5 mixing lines (ML) at the potential Temperatures...'*

Changed as suggested:

P76, I5-6. 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, I8. 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, I2-11. 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 (I.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 O<sub>3</sub> and N<sub>2</sub>O 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, I9-11. Suggested rewrite: "A tropospheric CO end member lower than typical tropopause values indicates that..."*

We changed the text as suggested.

*P78, I12. 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, I13 & I25. Delete 'determined' and 'investigated'.*

We deleted both.

*P79, I5-8. 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, I17. 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, I21. Delete 'significant'*

Deleted.

*P80, I1. Change 'suggest' to 'suggests'*

Corrected.

*P80, I6. Change to 'The calculation of 50-day back trajectories...'*

Changed as suggested throughout the manuscript.

*P80, I10, change to 'Clams calculates the vertical motion...'*

Changed as suggested.

*P80, I16, Change sentence to begin "Trace gas measurement of ..."*

Changed as suggested.

*P81, I2-6. 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, I9-15. 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 N<sub>2</sub>O, CO, and O<sub>3</sub> 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 N<sub>2</sub>O, CO, and O<sub>3</sub> in the early and later periods as well as their differences."*

Changed as suggested.

*P81, I16-17. Try this: "Figure 10 shows no significant changes for N<sub>2</sub>O and O<sub>3</sub> in the ExTL."*

Changed as suggested.

*P81, I19. 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, I20. Change to 'Both indicate...'*

Changed.

*P81, I22-24. 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, I3-7. 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..."*

I.394: changed as suggested

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

I.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<sub>2</sub>O-O<sub>3</sub> correlation. We decided to motivate the temporal change simply by the different times of the measurements rather than the temporal change of the N<sub>2</sub>O-O<sub>3</sub> 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 N<sub>2</sub>O (O<sub>3</sub>) 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.