

## GENERAL REPLY

The authors thank the two reviewers for their constructive comments and helpful suggestions that have improved the manuscript.

As mentioned by the both reviewers, the reconstruction method had some shortcomings, but the reviewers, especially reviewer #1, gave concrete and great suggestions to resolve them. The manuscript has been largely revised and improved along the suggestions. The authors describe first the revisions in the reconstruction method as a General Reply.

The suggestions are as follows:

- (1) use of 10-year trajectory instead of 90-day, which can eliminate the remaining trajectories in the UTLS ( $k=5$ );
- (2) reconstruction of chemical passive tracers with evaluation of transport timescale (as the first step); and
- (3) reconstruction of chemical active tracers including chemical decay (as the second step).

The authors have made these procedures and some relating revisions as follows.

For point (1), as noted by the reviewers, the remaining trajectories in the UTLS ( $k=5$ ) could be eliminated, i.e., it was confirmed that the all trajectories are categorized into any origins of  $k = 1, 2, 3$ , or 4 within 10 years (Note that the criteria have also been revised to avoid some shortcomings). In addition, the inversion method to estimate tracer mixing ratio for  $k=1$  and 5 in the original manuscript could be also eliminated, i.e., the all tracers in the ExUTLS have been reconstructed only from their mixing ratios assumed in the high-latitude LT, mid-latitude LT, and tropical troposphere.

For point (2), using the 10-year trajectory, Age of Air (AoA) as well as SF<sub>6</sub> and CO<sub>2</sub> distributions have been reconstructed from the trajectories including the “Tail correction” (e.g., Diallo et al., 2012). The CH<sub>4</sub>, N<sub>2</sub>O, CO have been also reconstructed without any chemical decay in this step. AoA has been also estimated using observed SF<sub>6</sub> mixing ratios obtained by CONTRAIL, and then the two AoAs have been compared to correct transport timescale expressed in the trajectories.

For point (3), the chemical active tracers, CH<sub>4</sub>, N<sub>2</sub>O, and CO, are finally reconstructed with simulating their chemical loss along an “average path” (Schoeberl et al., 2000). The use of the concept of average path was also suggested by the reviewer #1. Based on this concept, the authors believe that the active

tracers have been successfully reconstructed together with estimation of seasonally depending their chemical loss rate.

In relation to a suite of these revisions, the latter half of section 2.1 (Estimating the origin fraction and AoA), large part of section 2.2 (Air mass original composition and reconstruction) were significantly revised, especially section 2.2 was reorganized and a new subsections 2.2.1 and 2.2.2 were created for reconstructions of chemical passive and active tracers, respectively. The analyzing results, figures, and discussions were thus also changed in association with the revision of the reconstruction method, but the main thesis was essentially not changed.

#### References:

- Diallo, M., Legras, B., and Chédin, A.: Age of stratospheric air in the ERA-Interim, *Atmos. Chem. Phys.*, 12, 12133-12154, <https://doi.org/10.5194/acp-12-12133-2012>, 2012.
- Schoeberl, M. R., Sparling, L. C., Jackman, C. H., and Fleming, E. L.: A Lagrangian view of stratospheric trace gas distributions, *Journal of Geophysical Research: Atmospheres*, 105(D1), 1537-1552, doi:10.1029/1999JD900787, 2000.

The authors believe that the revised manuscript has been improved by incorporating the more appropriate reconstruction method.

Point-by-point responses to the comments of individual reviewer are provided below.

## REPLY TO COMMENTS BY REVIEWER #1

The authors are grateful for the thorough review and constructive comments on the manuscript. All of the points raised by the reviewer have been addressed. Regarding the reviewer's major comments, please also refer to the "General Reply" section. Point-by-point responses are detailed below, in red text.

### Reviewer (Comments):

Review of "Seasonal characteristics of chemical and dynamical transports into the extratropical upper troposphere/lower stratosphere" (ExUTLS) by Yoichi Inai et al.

### Recommendation: Publication after major revision

The paper is very well organised and written. The topic discussed in this paper, transport into the ExUTLS, is in general of high relevance. Our actual limitations in simulating water vapour transport in this complex region introduce large uncertainties in the Earth radiation budget (see Riese et al., 2012). Trajectory analysis in combination with observations could be and have been used in many cases to improve our knowledge on tracer transport and distribution in the UTLS, e.g. for H<sub>2</sub>O: Fueglistaler et al. (2004, 2005a, 2005b), e.g. for CO and H<sub>2</sub>O: Hoor et al. (2010), e.g. for STE (stratosphere-troposphere exchange) and O<sub>3</sub>: Skerlag et al. (2014), e.g. for CO<sub>2</sub> and AoA: Diallo et al. (2012, 2017). This manuscript here falls a bit short of explaining what the novel aspect of the presented method really is and how the presented results augment our actual knowledge on the seasonal characteristics of transport in and into the ExUTLS, e.g. in comparison to the early studies by Appenzeller et al. (1996) and Ray et al. (1999) or to the many studies summarised in the ExUTLS review paper by Gettleman et al. (2011).

The paper should be submitted after addressing the comments below.

### General comments:

First of all, I don't fully understand the title (and/or the scope) of this paper. What is the meaning of chemical and dynamical transport? Is chemical transport a synonym for transport of chemical active tracers (N<sub>2</sub>O, CH<sub>4</sub> and CO)? Should dynamical transport describe the transport of passive tracers (SF<sub>6</sub>, CO<sub>2</sub> and AoA)? Pure Lagrangian transport (here backward

trajectories) differs from (both) tracer transports: There is no mixing and no chemistry included along the individual transport pathways.

What the authors meant to write by the phrase “chemical and dynamical transports” is 1) transport of chemical species and 2) transport of air mass which is expressed by the mixing fractions of air mass originating in the stratosphere, tropical troposphere, mid-latitude LT, and high-latitude LT. In order to make it clearer, the title has been changed to “Seasonal characteristics of trace gas transport into the ExUTLS.”

The latter is definitively a problem for CO, because the chemical decay along the 90-days backward trajectories cannot be neglected. For N<sub>2</sub>O and CH<sub>4</sub> the chemical decay is not that significant, because only a few of the initialised trajectories will travel through the sink regions of both tracers higher up in the stratosphere during the 90-days. However, the unknown (non-observed) time series of the high-latitude stratospheric background (k=1) and the tropical and extratropical UTLS background (k=5) conditions for all tracers (not only for the chemical active tracers N<sub>2</sub>O and CH<sub>4</sub>) still remain the major problem for the reconstruction of the observed tracer distributions by CONTRAIL using only 90-days backward trajectories. The reconstruction in the chosen setup could not be used for quantitative studies of neither ERA-Interim (or other reanalysis data sets) nor the transport processes in the ExUTLS, because the trajectories itself are needed to define the boundary conditions for the original time series XS\_ORG\_k in the high-latitude stratosphere (k=1) and the UTLS (k=5) which are again the prerequisite to reconstruct the observed mixing ratios in the ExUTLS derived from CONTRAIL. This is a circular reference between the trajectory analysis and the CONTRAIL observations in the ExUTLS, whereby the non-observed (inverse reconstructed) original time series for k=1 and 5 could be seen as a kind of free parameters to tune the system or in other words to close the budget for the individual tracers.

The main problem, why this paper could, to my point of view, not add to the actual state of knowledge, although it has the potential, is the limitation of the backward trajectories to 90 days. The consequence of this limitation is the circular reference explained above (the authors call this an inversion technique) that has to be introduced to reconstruct the original time series of the tracer mixing ratios in the stratospheric overworld (k=1) and in the UTLS (k=5).

The authors claim that the mixing fractions derived from the coarser resolution ERA-Interim data (1.5x1.5, 37 levels) are the same as for the finer resolution ERA-Interim data (0.75x0.75, 60 levels). If this is the case, why not using the 10-year instead of the 90-days backward

trajectories? This would at least solve the problem to reconstruct the non-observed boundary conditions for the trajectories residing in the ExUTLS ( $k=5$ ) during the 90 days and also partially for the trajectories originating from the stratospheric overworld ( $k=5$ ). The latter is unfortunately only true for the passive tracers, SF<sub>6</sub> and CO<sub>2</sub> respectively. Both tracers could be reconstructed by combining their original tropospheric time series ( $k=2,3,4$ ) and the 10-years trajectories all starting in the target region of this study – the ExUTLS – and ending in the troposphere (e.g. Diallo et al., 2017), beside a small residuum of trajectories that remains in the stratosphere and that has to be characterised (see e.g. Ploeger et al, 2016).

The strong point of this study here is to my opinion the combination of backward trajectories driven by a state-of-the-art reanalysis data set (ERA-Interim) with simultaneous measurements in the ExUTLS of five tracers with different characteristics in their lifetimes and tropospheric time series. My recommendation would be to separate the analysis on transport and chemical processes. In the first step, one could use the 10-years backward trajectories (if manageable, it would be better using the high resolution ERA-Interim data) together with the passive tracers CO<sub>2</sub> and SF<sub>6</sub> (the latter is literally the same as AoA in the UTLS) to evaluate the mixing fractions and transport timescales derived from the ERA-Interim driven backward trajectories by reconstructing the CONTRAIL observations of both passive tracers. This is already a valuable extension to the method shown by Diallo et al. (2017), because the additional simultaneous SF<sub>6</sub> observations are a second independent constraint for the evaluation due to the different (linearly independent) tropospheric time series compared to CO<sub>2</sub>.

In the next step, one could exploit the additional information from the simultaneously measured chemical active tracers. Given the transport properties (mixing fraction, air mass origin and timescale) that has been quantified and evaluated in the first step, the chemical decay along the transport pathways from the tropospheric origin into the ExUTLS could be analysed with the simultaneous measurements of the chemical active tracers CO, CH<sub>4</sub> and N<sub>2</sub>O. The difference between the reconstructed passive CO, CH<sub>4</sub> and N<sub>2</sub>O tracers without chemistry and observed values including chemical decay should allow to assign a photochemical loss for air parcels along an “average pathway” with the same AoA (see Schoeberl et al., 2000). This “average pathway” could be defined by a bulk of trajectories, e.g. by the trajectories in a given equivalent latitude-potential temperature bin. There might be many more and better approaches to derive quantitative information on the chemical decay along the transport pathways, but the huge advantage in general of using backward trajectories together with simultaneous measured passive and chemical active tracers would be that one could disentangle dynamical and chemical effects on the observed tracer

distribution in a unique way. An urgent question that has to be answered to understand the processes driving and driven by climate change in this complex and important region of the atmosphere.

If the authors decide to stay with 90-days backward trajectory setup then the limitations of the 90-days backward trajectories and the sensitivity of the results due to these limitations have to be discussed in much more detail – see also the specific comments below.

Thank you for this great constructive suggestion on how to reconstruct both the chemically passive and active compositions. Following above suggestions, the reconstruction method has been revised, and the authors believe that it has been largely improved. Please see the General Reply.

Specific comments:

P.1, L.26: “*… especially in the Arctic climate.*”

Please cite references for this statement or delete it.

The statement has been deleted.

P.1, L.27: “*… via stratospheric residual circulation (Brewer-Dobson circulation, BDC; Brewer, 1949; Dobson, 1956).*”

The stratospheric residual circulation describes the mean mass transport. The BDC includes mean mass transport and two-way mixing. The latter, by definition, does not lead to net mass exchange but may lead to net tracer exchange. Therefore, I would suggest to use stratospheric circulation instead of stratospheric residual circulation as a synonym for the BDC (see e.g. Shepard, 2002; Birner & Boenisch, 2011).

Thank you very much for informative comments. Following this comment, the statement “stratospheric circulation” has been used, instead of “stratospheric residual circulation.”

P.2, L.28: In the text is written that the trajectories have been initialised between 0° E and 140° E longitude. In Fig. 1, the initialisation is all around the globe (0° E-360° E). What is actually the correct initialisation: figure or text?

Fig. 1 in the original manuscript was not correct. It has been corrected.

P.3, L.3-5: “*The distribution of some of the particles ...*”

I can hardly see the described feature in Fig. 2. The data should be presented in a different way to illustrate this more clearly.

In association with the revision of the method using 10-year trajectory, the figure and descriptions for it have been changed.

P.3, L.6-9 & Table 1: The criteria for the classification of the air mass origins ( $k=1-5$ ) seems to me somehow uncomplete or ambiguous:

1.) How trajectories are classified, if they satisfy the criteria  $< 350$  K,  $< 4$  km and  $20^\circ$  N  $<$  lat.  $< 30^\circ$  N? Are they counted as tropical troposphere ( $k=2$ ) or as mid-latitude LT ( $k=3$ )?

The trajectories had been categorized either compartment which was satisfied first. The authors confess that it was not appropriate, so the criteria have been revised to avoid such overlap as pointed out by this comment. The revised criteria have been shown in Table 1 in the revised manuscript.

2.) How trajectories are classified, if they satisfy the criteria  $> 380$  K, lat.  $< 45^\circ$  N and pot. vorticity  $> 6$  PVU? Are the counted as UTLS ( $k=5$ )? This would mean that backward trajectories initialised e.g. at 15-16 km geopotential height and lat.  $= 45^\circ$  N which has travelled up and equatorward would be counted as UTLS (see Fig. 2 right panel all points south of  $45^\circ$  N and above 15 km). To my feeling, some trajectories that should be assigned to the shallow branch ( $k=1b$ ) of the BDC are classified here as UTLS ( $k=5$ ).

Yes, they had been counted as  $k=5$ . Following this comment, the criteria for stratosphere ( $k=1$ ) has been also revised as shown in Table 1 in the revised manuscript.

Another problem of this UTLS criteria ( $k=5$ ) in combination with the 90-days backward trajectory limitation is that the mixing ratios of the tracers assigned to this category or region spanning from  $> 350$  K in the tropics or  $> 4$  km in the extratropics up to 25 km for lat.  $> 45^\circ$  N are very different. For example CO, mixing ratios ranging from  $> 100$  ppb (extratropical UT) to  $< 20$  ppb (stratospheric values for lat.  $< 45^\circ$  N) are condensed into the original UTLS time series needed to reconstruct the observations. The consequence is that

this original time series is mainly defined by the fact where in the UTLS the trajectory stemmed from.

As described in the General Reply, 10-year trajectory has been employed in addition to the revision of the classification of original regions. The authors believe that the problem pointed out here has been fixed.

P.3, L.22: The AoA definition in this paper is different to that of Hall&Plumb (1994). Here also for purely tropospheric transport AoA values are calculated, i.e. from the UT to the lower extratropical troposphere ( $< 4$  km) or the tropical troposphere  $< 350$  K potential temperature. Hall and Plumb defined an only stratospheric AoA using the tropopause as the reference surface. However, the AoA defined here is closer to the AoA derived from tracer measurements, e.g. SF<sub>6</sub>, for which the reference surface is in most cases and for practical reasons the tropical lower troposphere. This should be mentioned and clarified somehow, because tropospheric AoA are not really common.

To mention this point, the sentence “Thus, the AoA definition used here differs from that of Hall and Plumb (1994), who defined AoA as the elapsed time an air parcel spends in the stratosphere after across the tropopause” has been added in P3, L24ff in the revised manuscript.

P.3, L.26-30: It is not evident, if the underestimation of AoA found by Inai (2018) in the midlatitude stratosphere holds for the UTLS. This could be evaluated with AoA derived from the SF<sub>6</sub> CONTRAIL observation in the ExUTLS. This issue is briefly discussed in section 4.3 and it is implicitly shown in Fig. 12f, but it would be much clearer, if the authors would show a figure with SF<sub>6</sub>-derived AoA vs. 10-years backward trajectories derived AoA. This issue is of high interest (too short transport timescales into the stratosphere for ERA-Interim driven trajectories) and it also would have implications for the interpretation of chemical active tracers, for which the exposure time to stratospheric photochemistry is of interest.

As described in the General Reply, the 10-year trajectory has been employed and the SF<sub>6</sub>-derived AoA is compared with traj-derived AoA to correct them.

P.4, L.10: No chemical decay during transport from the origin to the initial position during the 90 days of transport is included – this is definitely not true for CO (see also the general comments above).



Chemical decay of CO as well as the other species have been included as described in the General Reply.

P.4, L.19-27: Would it not be more consistent to use higher temporal resolved reference data from the NOAA/ESRL atmospheric baseline observatories for the definitions of the tropospheric time series? You already use the Barrow site (BRW) together with the Summit site (SUM, downgraded to a sampling site) to define high-latitude (lat.  $> 45^{\circ}$  N) lower troposphere ( $k=4$ ) time series. The airborne measurements at 11 km between  $10^{\circ}$  N and  $30^{\circ}$  N could be used to evaluate the differences between the remote tropical LT and the flight level.

The authors consider that there are two attitudes to incorporate such data into this analysis. The one is use of higher temporal resolved data as you pointed out, and another is use of larger special representative data. The authors have conducted such aircraft observations by ourselves and accumulate such data which have larger special representativeness. We choose own larger special representative data to use.

P.5, L.3: It is hard to believe that this equation system is not under-determined. At least there should be some auxiliary constraints, e.g. mixing ratio  $X$  for a tracer with stratospheric sink should be lower for high-latitude stratosphere ( $k=1$ ) than for the UTLS ( $k=5$ ), i.e.  $X(k=1) < X(k=5)$ .

How the minimisation of the equation 4 has been technically performed? With a simple but robust parameter sweep or with a more sophisticated (but maybe numerical more instable) algorithm? This is to my opinion quite essential for the outcome of this paper. Therefore, this (inverse) procedure and the sensitivity of the results to the choice of parameters should be explained and shown in more details (see also general comments).

The inversion method has been eliminated in association with revision of the reconstruction method.

P.5, L.13-15: This means that you exclude most or at least a large part of the upper tropospheric CONTRAIL data, because  $\text{CO} > 80$  ppb is not a spurious event in the extratropical UT of the northern hemisphere (e.g. Engel et al., 2006 and references within), especially during winter and spring. Sometimes, it would be better to use tropopause related coordinates (or filter) instead of exclusively using equivalent latitude-potential temperature

coordinates.

The number of such measurements that  $\text{CO} > 80$  ppb and  $\text{Theta} > 340$  K and  $\text{Lat}_{\text{eq}} > 60^\circ\text{N}$  is not large. Such measurements have been identified by cross-marks in Figs. 4, 5, and 8 of the revised manuscript.

P.6, L.1-2: This finding is different from the results of Hoor et al. (2005) and Boenisch et al. (2009). Are there any explanation for these differences? I would expect that there is a certain time lag between the time of maximal downwelling (winter) and the maximal stratospheric characteristic of the LMS (spring).

The result has been changed in association with the use of 10-year trajectory, and it has become consistent with the results of Hoor et al. (2005) and Boenisch et al. (2009). The statement has been changed (P7, L19-22).

P.7, L.1-2: The seasonality and the mixing ratios for CO in the high-latitude stratosphere ( $k=1$ ) and UTLS ( $k=5$ ) are unrealistic, especially for spring (see e.g. Tilmes et al., 2010 and references within). This is to my view the consequence that errors, e.g. missing chemistry, in the reconstruction of ExUTLS observations will be compensated by the reconstructed original time series of CO for the regions  $k=1$  and 5.

The estimation method of compositions in the stratospheric air mass ( $k=1$ ) has been revised as described in the first part of Sect. 3.2 (P8, L11ff), in association with revision of the reconstruction method.

P.7, L.5-10 & Figure 7: Why does the seasonality of AoA and SF6 differ, especially in the UTLS (see Fig. 7d vs. 7f)? During August, the phase of the oldest AoA in the UTLS, one would expect the lowest (detrended) SF6 mixing ratios. This seems here not to be the case, August corresponds to the season with the highest (detrended) SF6 mixing ratios, equivalent to the youngest AoA. What is the explanation for this contradiction?

The estimation method of traj-derived AoA has been revised. The revised results show in-phase seasonality of AoA and SF6, the description has been changed (P8, L25-29 in the revised manuscript).

P.7, L.19-21, & Figure 8-11: For me, it looks like April is simply the month with the most

stratospheric characteristic of the LMS – highest AoA and lowest mixing ratios of the chemical active tracers above the 4pvu-contour.

That is right. The following description has been made here (P9, L6-10): “The reconstructions and AoA for April (Fig. 16) show spatial distributions of all species that generally increase with decreasing potential temperature, equivalent latitude, or potential vorticity, as is the case for January. However, the gradients are larger, particularly for CH<sub>4</sub> and N<sub>2</sub>O mixing ratios, such that in regions where the potential vorticity is >6 PVU the mixing ratios are much smaller than those in January, but in regions where the potential vorticity is <4 PVU the mixing ratios are almost the same as in January.”

P.7, L.29 & Figure 10: Why does SF<sub>6</sub>, as a proxy for AoA, not show, in contrast to N<sub>2</sub>O and CH<sub>4</sub>, the minima at 370 K in the ExUTLS region (see Fig 10 a+b+d)?

Such “sandwich” structures have been commonly shown in Fig. 17 of the revised manuscript.

P.8, L.6-7 & Figure 10-11: “*The distribution of AoA during this season (autumn, comment by the reviewer) is similar to that during summer, with the AoA of nearly the entire region with potential vorticity of < 8 PVU being less than 1 year.*”

The seasonality of AoA here is different to that found by Boenisch et al. (2009). They found the minimum in AoA in October with AoA below 0.5 years for most of the LMS (< 8pvu). What is the explanation for the difference in the seasonality found in the study here?

The result has been changed in association with the use of 10-year trajectory, and it has become consistent with the results of Boenisch et al. (2009).

P.8, L.16-17: A direct comparison of AoA derived from SF<sub>6</sub> and backward trajectories would be better (see comment: P.3, L.26-30). How does the contradiction of different seasonality of SF<sub>6</sub> and AoA (derived from trajectories) in the UTLS fit to this result (see comment: P.7, L.5-10 & Figure 7)?

The comparison of traj-derived AoA and SF<sub>6</sub>-derived AoA has been made and the UTLS category has been removed.

P.8, L.29-30: How you confirm the impact of the Asian Monsoon (ASM) with your study? Do you use an algorithm marking and detecting ASM air, e.g. like Vogel et al. (2016)?

No, we simply confirm that the trajectories originating in Asian region increased in summer season. The statement has been corrected to “trajectories originating in the tropical troposphere over around Asia are strengthened.” (P10, L10)

P.9, L.6 & Figure 13c: “*During winter, however, tropical tropospheric air masses dominate.*” This is true not only during winter, but also during spring (until the beginning of May).

Though the figure has been changed, but it remains true, so “and spring” has been added. (P10, L18)

P.9, L.10: “*In the high-latitude lower ExUTLS, mixing fractions of the mid- and high-latitude LT are enhanced but their fractions are lower than those of the mid-latitude lower ExUTLS.*”

What is the reason? More of the trajectories started in the stratosphere in high- compared to mid-equivalent latitude lower ExUTLS (PV>2pvu at the initial starting point)?

A tropopause related analysis would help here to understand how much of the effect here is related to the starting location (UT or LMS) and how much is related to weaker uplift into the UT in mid- compared to high-equivalent latitudes.

Equivalent latitude might be not the optimal coordinate in the troposphere. In contrast to the stratosphere, where PV is dominated by the strong stratification ( $d\theta/dz$ ), in the troposphere PV is dominated by relative vorticity. A consequence is that e.g. WCBs in the UT assigned with high tropospheric PV values (e.g. Madonna et al., 2014) would be classified as high-equivalent latitude air mass.

What the authors meant to here was “In the high-equivalent latitude lower (HL; such acronym has been used in the revised manuscript) ExUTLS, origin fractions of the mid- and high-latitude LT are enhanced during summer. Origin fractions of the high-latitude LT are comparable to those in the ML ExUTLS, but smaller than those of the mid-latitude LT in the HL ExUTLS.”

Therefore, the literature has been revised (P10, L18-20), in addition, the following discussion has been added (P10, L20ff): “This can be explained by enhanced exchange at the bottom edge of the subtropical jet (i.e., along the 320–330 K surface for summer, e.g., Gettelman et al., 2011). As shown in Fig. 12d, enhanced origin fractions of the mid-latitude LT are distributed along such isentropes.”

Relating this comment, the authors find that the phase “mid- latitude lower ExUTLS” should

be changed “mid-equivalent latitude lower ExUTLS,” this revision has been also done.

P.9, L.11-13 & Fig. 13+14: The seasonal pattern in Fig 13 and 14 has not the same pattern for species with strongly varying original time series in the different compartments ( $k=1-5$ ), because the reconstructed time series shown in Fig 14 are a superposition of the mixing fractions shown in Fig. 13 with the original time series for  $k=1-5$  of the individual tracers. The difference in the seasonal pattern between Fig 13 and Fig 14 is most obvious for CO<sub>2</sub> which has a strong tropospheric seasonal cycle that is superimposed on the tropospheric mixing fractions.

The statement in the original manuscript was not correct. The statement has been revised to “Figure 20 reveals that seasonal variations in the reconstructions for each species and the trajectory-estimated AoA in each of the four locations have patterns that differ because they are based on a superposition of the origin fractions shown in Fig. 19 with the original time series for  $k=1-4$  of the individual tracers shown in Fig. 14.” (P10, L25-28)

P.9, L.33: Please add here the reference to Hoor et al. (2004)

It has been added (P11, L14).

P.10, L.20 & Fig. 15: The tracer-tracer relationships or “mixing lines” (AoA is a tracer like e.g. SF<sub>6</sub>) for sufficient long-lived tracers (chemical lifetime must be greater than at least the horizontal transport timescale) is in theory the consequence of sufficiently rapid mixing along isentropic surfaces. Please cite here the review by Plumb (2007) which includes many of the references to the pioneering works on this topic in the 80s and 90s.

Thank you very much for this informative comment. It has been cited and the following statement has been added (P12, L2-4): “Such linear “mixing lines” also suggest that the mixing took place rapidly (i.e., at a time-scale shorter than their chemical lifetimes) along an isentropic surface (Plumb, 2007 and references therein).”

P.10, L.21-23 & Fig. 15: CO<sub>2</sub> mixing ratios and AoA does not correlate below a level of about 3 years AoA, because the propagated signal of the tropospheric seasonal cycle into the stratosphere is still detectable (not smeared out over a broad enough age spectra covering several seasonal cycles). This is the simple reason why CO<sub>2</sub> mixing ratios in the LMS cannot be used to calculate AoA (see e.g. Engel et al., 2002; Boenisch et al., 2009).

Thank you for the instructive comment. CO<sub>2</sub> is deleted in the sentence and the following statements has been added: “According to Engel et al. (2002) and Bönisch et al. (2009), the mixing ratios of CO<sub>2</sub> and AoA do not correlate below a level of ~3 years AoA because the propagated signal of the tropospheric seasonal cycle into the stratosphere is still detectable. In agreement with their results, the CONTRAIL CO<sub>2</sub> measurements also converge to the sign-reversed trend with increasing AoA.” (P12, L5-8)

P.10, L.28-30 & Fig. 15f: “*Figure 15f shows seasonal variations in AoA and integrated PDF from 0 to 10 years for air masses originating in the high-latitude stratosphere*”

Fig. 15f only shows integrated PDFs for AoA from 0 to 6 years – please correct this.

The meaning of this sentence was that “The figure shows seasonal variations in AoA and the value that is calculated by integration of “age spectrum” (PDF) from 0 to  $t_f$  for air masses originating in the stratosphere” The sentence has reworded (P12, L13-15).

P.12, L.3-4: The young-bias of AoA derived from backward trajectories in the LS should be verified (see my comment P.3, L.26-30 above)

It has been verified as described in the General Reply.

P.12, L.8-9: Please show this (see comment above).

It has been shown in Fig. 4 which has been newly made.

P.12, L.15-17: “*Moreover, these estimates are indirectly validated by the CONTRAIL observations, through the reconstruction of the chemical distributions (as evident in Figs 8–11).*”

This is only partially true, because you have a kind of free parameters, these are the original mixing ratio from the deep stratosphere (k=1) and the mixing ratios resided in the ExUTLS (k=5) during the 90 days of the backward trajectory simulation. Herewith, the interaction between CONTRAIL observed and trajectory-based mixing ratios can be adjusted. This is to my opinion most obvious for CO. The estimated CO for k=1 and 5 compensates other errors, e.g. chemical decay of CO during the transport from source region to the ExUTLS (see also other specific and general comments above).

In association with the change of the reconstruction method, the free parameters have been eliminated.

P.12, L.18-24: Both problems discussed here briefly, non-linear tropospheric trend and the lack of agreement in reconstruction of CONTRAIL observations during summer (Fig. 10e), concern mainly CO<sub>2</sub>, so please clarify and mention this here.

For the non-linear trend, the statement “In this study, linear trends for CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub>, and CO<sub>2</sub> are assumed for the reconstruction. Although this is a simplified treatment, given the length of the analysis period, these trends are roughly constant over this time period with the exception of CH<sub>4</sub>, and the CH<sub>4</sub> reconstructions are more strongly affected by chemical loss, as is evident in a comparison of Figs 5a and 8a” has been added in P14, L33ff.

For the disagreement during summer, the statement “particularly for CO<sub>2</sub> (Fig. 5e)” has been added in P14, L26.

P.12, L.24-26: It is true that the equivalent latitude-potential temperature (EqLat-Theta) coordinate system accounts for dynamical features in the stratosphere, because adiabatic motion is dominant in this strongly stably stratified region of the atmosphere. This is not true for the troposphere which is much more unstable (low static stability). Potential temperature (and PV) are not conserved or only for a much shorter timescale, because diabatic motion is much more prominent there. Therefore, potential temperature and equivalent latitude are not the coordinate system of choice in the troposphere. Also the problem of tracer uplift from the PBL into the UT during summer (most prominent for CO<sub>2</sub>, see above) is not minimised in an EqLat-Theta coordinate system.

Thank you for this instructive comment. The description “which are dynamically conserved quantities in the stratosphere. In the troposphere, which is more unstable, potential temperature and potential vorticity are not conserved, or are conserved only on much short timescales, because of diabatic motion. It should be noted that tracer uplift from the LT into the UT during summer (particularly for CO<sub>2</sub>, as discussed above) cannot be reduced with the coordinate system employed here” has been added in P14, L27-30.

P.13, L.7: The mentioned role of Asian monsoon (ASM) is very likely, but it is speculation here, because it is not shown in this study, how much of the trajectories originated from the ASM (see also comment above).

The authors simply confirm that the trajectories originating in Asian region increased in summer season, so the statement “in association with the Asian summer monsoon” has been deleted.

P.13, L.15: “*The reconstructions agree well with CONTRAIL measurements in the ExUTLS.*”  
If this is one key messages of the summary then the limitations of the 90-days backward trajectories and the sensitivity of the results due to this limitation have to be discussed in much more detail (see general and specific comments above).

The 10-years trajectory has been employed as described in the General Reply.

P.13, L.23-24: “*This method provides a means to understand both dynamical transport and chemical distribution from a new perspective.*”

There has been done a lot to understand dynamical, tracer transport and chemical processes in the UTLS. Some of these studies has been mentioned in this review and should be discussed in relation to the results in this manuscript. As outlined in my general comment, I am not convinced that the actual manuscript could contribute to the actual state of knowledge, but the results should be at least discussed in this framework. The uniqueness of this approach here, combination of different tracers and backward trajectories, could to my opinion be exploited much better, if one would use 10-years instead of 90-days backward trajectories.

The 10-years trajectory has been used, and the authors believe that this study has been significantly improved.

In addition to above revision following the reviewers' comments, the authors have added new Appendix (Appendix B and relating two figures) in the revised manuscript to visualize large perspective of seasonal variation in ExUTLS.

#### References:

Appenzeller, C., J. R. Holton, and K. H. Rosenlof (1996), Seasonal variation of mass transport across the tropopause, *J. Geophys. Res.*, 101(D10), 15071-15078.  
Birner, T., and H. Bönisch (2011), Residual circulation trajectories and transit times into the extratropical lowermost stratosphere, *Atmos. Chem. Phys.*, 11(2), 817-827, doi:10.5194/acp-11-817-2011.



Bönisch, H., A. Engel, J. Curtius, T. Birner, and P. Hoor (2009), Quantifying transport into the lowermost stratosphere using simultaneous in-situ measurements of SF<sub>6</sub> and CO<sub>2</sub>, *Atmos. Chem. Phys.*, 9(16), 5905-5919, doi:10.5194/acp-9-5905-2009.

Diallo, M., Legras, B., and Chédin, A. (2012): Age of stratospheric air in the ERA-Interim, *Atmos. Chem. Phys.*, 12, 12133-12154, <https://doi.org/10.5194/acp-12-12133-2012>, 2012.

Diallo, M., B. Legras, E. Ray, A. Engel, and J. A. Añel (2017), Global distribution of CO<sub>2</sub> in the upper troposphere and stratosphere, *Atmos. Chem. Phys.*, 17(6), 3861-3878, doi:10.5194/acp-17-3861-2017.

Engel, A., M. Strunk, M. Müller, H. P. Haase, C. Poss, I. Levin, and U. Schmidt (2002), Temporal development of total chlorine in the high-latitude stratosphere based on reference distributions of mean age derived from CO<sub>2</sub> and SF<sub>6</sub>, *J. Geophys. Res.*, 107(D12), doi:Artn 4136 Doi 10.1029/2001jd000584.

Engel, A., et al. (2006), Highly resolved observations of trace gases in the lowermost stratosphere and upper troposphere from the Spurt project: an overview, *Atmos. Chem. Phys.*, 6, 283-301, doi:DOI 10.5194/acp-6-283-2006.

Fueglistaler, S., H. Wernli, and T. Peter (2004), Tropical troposphere-to-stratosphere transport inferred from trajectory calculations, *Journal of Geophysical Research: Atmospheres*, 109(D3), doi:10.1029/2003JD004069.

Fueglistaler, S., M. Bonazzola, P. H. Haynes, and T. Peter (2005), Stratospheric water vapor predicted from the Lagrangian temperature history of air entering the stratosphere in the tropics, *Journal of Geophysical Research: Atmospheres*, 110(D8), doi:10.1029/2004JD005516.

Fueglistaler, S., and P. H. Haynes (2005), Control of interannual and longer-term variability of stratospheric water vapor, *Journal of Geophysical Research: Atmospheres*, 110(D24), doi:10.1029/2005JD006019.

Gettelman, A., P. Hoor, L. L. Pan, W. J. Randel, M. I. Hegglin, and T. Birner (2011), The Extratropical Upper Troposphere and Lower Stratosphere, *Rev. Geophys.*, 49, doi:Artn Rg3003, Doi 10.1029/2011rg000355.

Hall, T. M., and R. A. Plumb (1994), Age as a Diagnostic of Stratospheric Transport, *J. Geophys. Res.*, 99(D1), 1059-1070.

Hoor, P., C. Gurk, D. Brunner, M. I. Hegglin, H. Wernli, and H. Fischer (2004), Seasonality and extent of extratropical TST derived from in-situ CO measurements during SPURT, *Atmos. Chem. Phys.*, 4, 1427-1442.

Hoor, P., H. Fischer, and J. Lelieveld (2005), Tropical and extratropical tropospheric air in the lowermost stratosphere over Europe: A CO-based budget, *Geophys. Res. Lett.*, 32(7), L07802, doi:10.1029/2004gl022018.

Hoor, P., H. Wernli, M. I. Hegglin, and H. Boenisch (2010), Transport timescales and tracer properties in the extratropical UTLS, *Atmos. Chem. Phys.*, 10(16), 7929-7944, doi:10.5194/acp-10-7929-2010.

Madonna, E., H. Wernli, H. Joos, and O. Martius (2014), Warm Conveyor Belts in the ERA-Interim Dataset (1979–2010). Part I: Climatology and Potential Vorticity Evolution, *Journal of Climate*, 27(1), 3-26, doi:10.1175/jcli-d-12-00720.1.

Ploeger, F., and T. Birner (2016), Seasonal and inter-annual variability of lower stratospheric age of air spectra, *Atmos. Chem. Phys.*, 16(15), 10195-10213, doi:10.5194/acp-16-10195-2016.

Plumb, R. A. (2007), Tracer interrelationships in the stratosphere, *Rev. Geophys.*, 45(4), Art. Rg4005, doi:10.1029/2005rg000179.

Ray, E. A., F. L. Moore, J. W. Elkins, G. S. Dutton, D. W. Fahey, H. Vomel, S. J. Oltmans, and K. H. Rosenlof (1999), Transport into the Northern Hemisphere lowermost stratosphere revealed by in situ tracer measurements, *J. Geophys. Res.*, 104(D21), 26565-26580.

Riese, M., F. Ploeger, A. Rap, B. Vogel, P. Konopka, M. Dameris, and P. Forster (2012), Impact of uncertainties in atmospheric mixing on simulated UTLS composition and related radiative effects, *J. Geophys. Res.*, 117(D16), D16305, doi:10.1029/2012jd017751.

Schoeberl, M. R., L. C. Sparling, C. H. Jackman, and E. L. Fleming (2000), A Lagrangian view of stratospheric trace gas distributions, *Journal of Geophysical Research: Atmospheres*, 105(D1), 1537-1552, doi:10.1029/1999JD900787.

Shepherd, T. G. (2002), Issues in Stratosphere-troposphere Coupling, *Journal of the Meteorological Society of Japan. Ser. II*, 80(4B), 769-792, doi:10.2151/jmsj.80.769.

Škerlak, B., M. Sprenger, and H. Wernli (2014), A global climatology of stratosphere–troposphere exchange using the ERA-Interim data set from 1979 to 2011, *Atmos. Chem. Phys.*, 14(2), 913-937, doi:10.5194/acp-14-913-2014.

Tilmes, S., et al. (2010), An aircraft-based upper troposphere lower stratosphere O<sub>3</sub>, CO, and H<sub>2</sub>O climatology for the Northern Hemisphere, *Journal of Geophysical Research: Atmospheres*, 115(D14), doi:10.1029/2009JD012731.

Vogel, B., et al. (2016), Long-range transport pathways of tropospheric source gases originating in Asia into the northern lower stratosphere during the Asian monsoon season 2012, *Atmos. Chem. Phys.*, 16(23), 15301-15325, doi:10.5194/acp-16-15301-2016.