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 H2O: Fueglistaler et al. (2004, 2005a, 2005b), e.g. for CO and H_2O: Hoor et al. (2010), e.g. for STE (stratosphere-troposphere exchange) and O_3: Skerlag et al. (2014), e.g. for CO_2 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 Gettlemann 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_2O, CH_4 and CO)? Should dynamical transport describe the transport of passive tracers (SF_6, CO_2 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.

The latter is definitively a problem for CO, because the chemical decay along the 90-days backward trajectories cannot be neglected. For N_2O and CH_4 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_2O and CH_4) 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 $X^S_{ORG\_k}$ 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_6 and CO_2 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_2 and SF_6 (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 simoultaneous SF_6 observations are a second independent constraint for the evaluation due to the different (linearly independent) tropospheric time series compared to CO_2.
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_4 and N_2O. The difference between the reconstructed passive CO, CH_4 and N_2O 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 simoultaneous 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.

Specific comments:

P.1, L.26: "*…, especially in the Arctic climate.*"
Please cite references for this statement or delete it.

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).

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?

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.

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°N < lat. < 30°N? Are they counted as tropical troposphere (k=2) or as mid-latitude LT (k=3)?

2.) How trajectories are classified, if they satisfy the criteria > 380 K, lat. < 45° 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°N which has travelled up- and equatorward would be counted as UTLS (see Fig. 2 right panel all points south of 45°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).
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°N are very different. For example CO, mixing ratios ranging from > 100 ppb (extratropical UT) to < 20 ppb (stratospheric values for lat. < 45°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.

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_6, 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.

P.3, L.26-30: It is not evident, if the underestimation of AoA found by Inai (2018) in the mid-latitude stratosphere holds for the UTLS. This could be evaluated with AoA derived from the SF_6 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_6-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.

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).

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°N) lower troposphere (k=4) time series. The airborne measurements at 11 km between 10°N and 30°N could be used to evaluate the differences between the remote tropical LT and the flight level.

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).

P.5, L.13-15: This means that you exclude most or at least a large part of the upper tropospheric CONTRAIL data, because 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.

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).

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.

P.7, L.5-10 & Figure 7: Why does the seasonality of AoA and SF_6 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) SF_6 mixing ratios. This seems here not to be the case,

August corresponds to the season with the highest (detrended) SF_6 mixing ratios, equivalent to the youngest AoA. What is the explanation for this contradiction?

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.

P.7, L.29 & Figure 10: Why does SF_6, as a proxy for AoA, not show, in contrast to N_2O and CH_4, the minima at 370 K in the ExUTLS region (see Fig 10 a+b+d)?

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?

P.8, L.16-17: A direct comparison of AoA derived from SF_6 and backward trajectories would be better (see comment: P.3, L.26-30). How does the contradiction of different seasonality of SF_6 and AoA (derived from trajectories) in the UTLS fit to this result (see comment: P.7, L.5-10 & Figure 7)?

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)?

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).

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 (dTheta/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.

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_2 which has a strong tropospheric seasonal cycle that is superimposed on the tropospheric mixing fractions.

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

P.10, L.20 & Fig. 15: The tracer-tracer relationships or "mixing lines" (AoA is a tracer like e.g. SF_6) 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.

P.10, L.21-23 & Fig. 15: CO_2 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_2 mixing ratios in the LMS cannot be used to calculate AoA (see e.g. Engel et al., 2002; Boenisch et al., 2009).

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.

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)

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

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).

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_2, so please clarify and mention this here.

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_2, see above) is not minimised in an EqLat-Theta coordinate system.

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).

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).

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.

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