

## 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 #2

The author is grateful for the thorough review and constructive comments on the manuscript. All of the points raised by the reviewer have been addressed. The major revision was made following the #1 reviewer's comments, please see first the "General Reply" section. Point-by-point responses are provided below, in red text.

Interactive comment on "Seasonal characteristics of chemical and dynamical transports into the extratropical upper troposphere/lower stratosphere" by Yoichi Inai et al.

Anonymous Referee #2

Received and published: 2 January 2019

The paper by Inai et al. investigates the air mass composition of the extratropical upper troposphere and lower stratosphere (exUTLS), and relates to CONTRAIL in-situ observations of several trace gas species (e.g., CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub>, CO, CO<sub>2</sub>). The focus of the study lies on seasonal variations in air mass fractions and mixing ratios. In particular, it is found that seasonality in CH<sub>4</sub>, N<sub>2</sub>O and SF<sub>6</sub> mixing ratios is controlled by transport from the deep stratosphere, due to the locations of the main chemical sink regions, whereas CO and CO<sub>2</sub> are mainly controlled by transport from the tropical troposphere.

The air mass and tracer composition of the exUTLS is of particular relevance for global climate due to the radiative characteristics of this region. Hence, the present study fits well into the scope of ACP. The paper is well written and presented, and the current literature is appropriately discussed. I recommend publication after taking into account the several comments below, which I regard somewhere between major and minor.

Detailed comments:

1. Initialization: The trajectory initialization is somewhat unclear to me. In the respective text part it is said, that back trajectories are initialized between 0-140 deg E, but the corresponding Fig. 1 shows initialization locations for 0-360 deg E (P2/L27). How is the initialization done exactly?

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

2. Model-measurement comparison: The CONTRAIL measurements are mainly from Siberia. How is the model-measurement comparison done, exactly at the measurement locations/times, or just averaged over specific regions? I would suggest to explain this clearly directly after the description of the trajectory initialization (P2).

Indeed, it could be designed to release trajectories exactly at the measurement location/time and it may make directly comparison with the CONTRAIL measurements; this study, however, attempts to reconstruct spatial-extending and uniform spatiotemporal tracer distributions as well as their transport, therefore, we choose to employ the grating initialization. Following the suggestion, the statement "Although trajectories could be released at the exact CONTRAIL measurement locations and times, the grating initialization is employed because this study attempt to obtain uniform spatiotemporal tracer distributions as well as their transports by capitalizing on the CONTRIAL measurements" has been added in P2, L31ff in the revised manuscript.

3. Reconstruction method: It would be good to mention (around P4/L10) that Eq. (2) holds only for species which are chemically inert along the trajectories. Can you give some quantitative information how well this assumption holds for the species and regions considered here? Perhaps some of the difference between reconstruction and measurements (e.g., Figs. 7-10) is related to neglecting chemistry effects?

As described in the General Reply, the reconstruction method has been revised. The chemically active species are reconstructed taking the chemical decay into account.

4. Origin mixing ratios (P4/L28): Why not using higher altitude in-situ measurements (e.g., from balloons, Geophysica/Halo/ER2/... aircrafts) or global satellite observations for the reference mixing ratios? At least the "inversion method" outlined below could be validated with such data.

As described in the General Reply, the reconstruction method has been revised. In association with the revision, the "inversion method" has been eliminated.

5. Minima in tracer distributions around 370K in spring/summer (P7/L21ff): I do not think these minima are just artifacts of the reconstruction. The fact that spring/summer transport of young tropical air strengthens first around 380-400K, leading to a "sandwich" structure with older air masses below is consistent with recent findings by Krause et al. (2018) (see e.g.

their Fig. 14) and Ploeger and Birner (2016) (e.g., their Fig. 7).

In agreement with these papers, Fig. 9/10 show evidence for strongest poleward transport above about 380K, causing the mixing ratio minima below. I would suggest to discuss these distributions more appropriately.

Thank you for this informative comment and suggestion. Though the sandwich structures have changed their aspects due to the revision of reconstruction method, they have appeared at around 350 K as shown in Fig. 17. Following above suggestion, the following statements have been added (P9, L13-17): “In particular, all five chemical species show minima at ~350 K north of 60° N equivalent latitude. These minima might be formed by remainder of the deep stratospheric air masses which were transported during spring. The tracer minima near ~350 K at high equivalent latitudes begin forming in June. This “sandwich” structure in the ExUTLS has been reported by Ploeger and Birner (2016) for summer and by Krause et al. (2018) for spring. In agreement with their studies, the sandwich structures can show evidence for strong poleward transport above ~400 K, leading to mixing ratio minima at lower altitudes.”

6. Trajectory method: Kinematic trajectories show stronger dispersion compared to diabatic trajectories (e.g., Schoeberl et al., 2003). Are the results presented here robust also for diabatic transport? At least include appropriate discussion in Sect. 4.3 (“Limitations of the current study”).

The authors have not used diabatic trajectory, so we do not explicitly know how much the results change. Instead, statements “Trajectory results also generally depend on the vertical condition, i.e., kinematic (employed by the current study) or diabatic (employed by, for example, Diallo et al., 2017). Previous studies suggest that using kinematic trajectories leads to a stronger dispersion and somewhat young bias in AoA estimates compared with using diabatic trajectories (e.g., Schoeberl et al., 2003; Diallo et al., 2012). Therefore, using diabatic trajectories in this analysis might result in a correction factor ( $\gamma_{TT}$ ) of <1.5.” have been added in Sect. 4.3 (P14, L1-4).

Specific and technical comments:

P1/L29: maybe better "at/along the subtropical jet"?

Change made as suggested.

P3/L23: "...where IT satisfies..."?

Corrected.

P3/L28: What is the "actual value" what is referred to here? Observations? Which?

It is referred to observation. Statement "actual value" has been changed to "observed value."

P7/L29: ware -> were

Corrected.

P9/L10ff: The sentence "In addition ..." sounds unclear to me - I suggest rewording.

The sentence has been reworded to "In addition to seasonal variations in origin fractions, seasonal variations in the tracer mixing ratios in origin regions (Fig. 14) also affect chemical distributions in the ExUTLS." (P10, L24-25)

P9/L19: shown -> show

Corrected.

P10/L28ff: I don't understand the description of Fig. 15f. What PDF is integrated here (transit time pdf?). What is the unit of the y-axis? Please clarify and improve the description.

The integration is done for age spectrum, so statement "the value that is calculated by integration of "age spectrum" (PDF)" was added in P12, L14.

P12/L7: The Ploeger and Birner reference cited here is not in the reference list.

The reference has been added.

P12/L20: non-linear

Corrected.

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:

Krause et al. (2018), Atmos. Chem. Phys., 18, 6057-6073.

Schoeberl et al. (2003), J. Geophys. Res, 118, D3.