

Reviewer (Comments):

Re-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 minor revision

The revised paper has improved significantly and the authors did an impressive job and addressed the questions raised in the review thoroughly. From my side, there are only a few open questions left open that should be addressed before publications.

The paper should be submitted after addressing the comments below.

General comments:

My only general comment is that I am still convinced that the additional use of tropopause related coordinates would improve the comparison with the CONTRAIL data set. This concerns the sections 3.3 and 4.1, where the authors compare the reconstructions of the different tracers with the CONTRAIL observations (Fig. 15-18 and Fig. 21). Especially in the region close to the tropopause, i.e. the 320 K potential temperature level in high- and mid-latitudes (or more precisely equivalent latitudes), the intercomparison suffers from the small number of observations. The reason is, that the mixing ratio gradients across the tropopause are large and that the estimation of the mean values in these bins (320 +/-5 K pot. temp. and 45°/75° +/-5° eq. lat.) needs sufficient statistics therefore. This is no major issue and it is nothing wrong about the way the intercomparison is done in the paper – it is just a suggestion.

Specific comments:

P.6, L.14-17: It is the right idea to use observation, i.e. Volk et al. (1997), to determine the chemical decay along the “average path” (AP) into the ExUTLS of the northern hemisphere – the region of interest in this work here. However, there are two caveats using the data set of Volk et al. (1997) here:

a) A large part of the ASHOE/MAESA campaign took place in the southern hemisphere. Therefore, this data set might not be the best representation for the chemical decay along the AP from the troposphere into the northern hemisphere ExUTLS. This should be at least mentioned in the context here.

b) The gradients $dX/dAoA$ (X: Tracer mixing ratio and AoA: Age of Air) is not solely determined by the chemical decay along the AP in the stratosphere, but also by the tropospheric trend of the tracer that propagates into the stratosphere. That means, the observed gradients $dX/dAoA$ by Volk et al. are partially influenced by the growth rates of the tropospheric time series of N₂O and CH₄ before 1994. Luckily, the growth rates of both tracers in the time interval 5 years before the individual observations took place are rather similar. For CH₄, the growth rates are 7.9 ppb/year for ASHOA/MAESA and 5.3-7.5 ppb/year for CONTRAIL. For N₂O, the trend since 1990 is very close to linear with a growth rate of 0.81 ppb/year. Therefore, this should not be a big issue for the analysis here, but it should be mentioned.

The numbers for CH₄ and N₂O growth rates shown above are derived from the reference data sets of NOAA ESRL available on their websites:

<https://www.esrl.noaa.gov/gmd/hats/combined/N2O.html>

https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/

P.6, L.21-22: “..., so here it is assumed that the gradient of CO mixing ratio with respect to AoA is 20-times larger than that of CH₄.”

Could you please explain this in a bit more detail in the paper, why you assume that the gradient $dX_{CO}/dAoA$ is 20-time larger than $dX_{CH_4}/dAoA$. The tropospheric trends of CO and CH₄ and their chemistry in the stratosphere are slightly different, so I would expect a slightly different gradient of chemical loss along the AP. This will most probably not really be an issue for the result of this study, but the assumption should be motivated here.

Fig 6: The remaining fraction of CO in the stratosphere is not going down to zero as shown in the Figure 6, but reaches an equilibrium value due to production processes balancing the photochemical loss. The stratospheric equilibrium value is about 10-15 ppbv (e.g. Krause et al., 2018) which corresponds to about 10% of the tropospheric value.

P.8, L.15-17: “..., CH₄ and N₂O in stratospheric air masses show distinct seasonal variations but somewhat different phase,...”

Is there any explanation, why the seasonality in the stratosphere is different? Both tracers are rather long-lived in the lower stratosphere (shown in Fig. 14) and should therefore be dominated by transport processes. It is not really clear to me, how this should lead to such a difference in the seasonality. I would definitely expect for CH₄, in the same way as for N₂O and SF₆, the lowest mixing ratios in the ExUTLS in spring and not in summer.

Fig. 14 Panel (c): “Tropospheric CO”

It looks rather unlikely to me that tropospheric background values for high-latitude CO are much lower than mid-latitude CO and comparable to tropical CO. This is also in contradiction to the meridional distribution shown in GLOBALVIEW-CO provided by NOAA ESRL. I would expect at least CO mixing ratios in the high-latitudes that are as high as in the mid-latitudes. The difference here is most likely caused by the fact that NOAA ESRL background values have been used for the high-latitudes, but own aircraft based measurements have been used for mid-latitudes and tropics that are maybe not filtered in order to retrieve background values. This means that the mid-latitude and tropical tropospheric CO time series, in contrast to the high-latitudes, containing a mixture of polluted and unpolluted air masses. Please clarify this issue.

See: https://www.esrl.noaa.gov/gmd/ccgg/globalview/co/co_intro.html

Fig. 14 Panel (d): “Tropospheric SF₆”

The same seems also to be the case for tropospheric SF₆. High-latitude SF₆ should show higher values than tropical SF₆, see e.g. Rigby et al. (2010) or Waugh et al. (2013). Please clarify also.

See: <https://www.esrl.noaa.gov/gmd/hats/combined/SF6.html>

P.11, L.13-14: “This seasonal variation in the upper ExUTLS is consistent with observational estimates by Hoor et al. (2004) and Strahan et al. (2007).”

This is true, but the comparison with the results from Hoor et al. (2004) have to be taken with some caution, because this study analyse the CO₂ seasonal cycle in tropopause relative coordinates and therefore the direct comparison is not straightforward.

P.13, L.10-12: “*These overestimations of N₂O and CH₄ mixing ratios for the original stratospheric air masses might be due to overestimation of the AoA and/or overestimation of the origin fraction of air masses originating in the deep branch of the BDC.*”

Maybe I am wrong, but I think that the diagnosed overestimation of N₂O and CH₄ in May/November cannot be simply related to an overestimation of the origin fractions of deep stratospheric air. Assuming that the origin fraction from deep BDC would be smaller than the estimated N₂O values from Andrews et al. (2001) would become higher, but also the N₂O values reconstructed from the trajectories in this study would become higher.

P.13, L.33: “*..., the relationship approaches those of Andrews et al. (2001) and Röckmann et al. (2011).*”

It could be helpful to extend this sentence a bit (“*..., the N₂O-AoA and CH₄-AoA relationship approaches those of Andrews et al. (2001) and Röckmann et al. (2011) respectively.*”) for a better readability and clarification.

References:

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Rigby, M., Mühle, J., Miller, B. R., Prinn, R. G., Krummel, P. B., Steele, L. P., Fraser, P. J., Salameh, P. K., Harth, C. M., Weiss, R. F., Grealley, B. R., O'Doherty, S., Simmonds, P. G., Vollmer, M. K., Reimann, S., Kim, J., Kim, K.-R., Wang, H. J., Olivier, J. G. J., Dlugokencky, E. J., Dutton, G. S., Hall, B. D., and Elkins, J. W.: History of atmospheric SF₆ from 1973 to 2008, *Atmos. Chem. Phys.*, 10, 10305–10320, <https://doi.org/10.5194/acp-10-10305-2010>, 2010.

Waugh, D. W., et al. (2013), Tropospheric SF₆: Age of air from the Northern Hemisphere midlatitude surface, *J. Geophys. Res. Atmos.*, 118, 11,429–11,441, doi:10.1002/jgrd.50848.