Schuck et al., The interhemispheric gradient of SF6 in the upper troposphere

Response to comments by Anonymous Referee #2

We thank the reviewer for their comments and suggestions to improve our manuscript. All comments are addressed in the following with the reviewer's comments printed in blue, and the responses in black.

This paper uses in situ trace gas measurements, primarily SF6, from the upper troposphere and surface along with a 12-box model to investigate tropospheric transport and SF6 emissions over a recent 15 year period. The combination of US and European based aircraft data sets provides a relatively complete latitudinal and temporal representation of the upper troposphere over this time period. The authors optimize both emissions and transport parameters within the 12-box model to best match the observations resulting in a number of interesting findings. This is a really nice use of a simplified model to diagnose what the observations can tell us about large scale features of atmospheric transport and trace gas emissions. The methodology and discussion of results are clearly described. I recommend publication in ACP with consideration of the minor comments below.

Specific comments:

Section 4 or 5: It would be nice to include a brief comparison of previous model estimates of the interhemispheric transport time such as from Waugh (2013) and Orbe (2016, 2021) to the original estimates from the 12 box model. The Waugh (2013) transport time was less than 2 years to the SH but still somewhat longer than the observational transport time. This comparison would help give the reader an idea of how much CTM transport needs to be adjusted to better match the observations.

We agree with the reviewer's suggestion. Following the comparison of modelled and observed lag times discussed using Figure 6, the following paragraph was added in section 4:

„Similar results were obtained previously using the more sophisticated NASA Global Modeling Initiative chemical transport model (CTM) (Strahan at al. 2007, 2016). Waugh et al. (2013) compared the CTM output to ground-based, ship-borne, and aircraft measurements from the NOAA observational network and found the model to overestimate lag-times towards southern latitudes. At middle and high latitudes in the southern hemisphere, ground station observations yielded lag-time of 1.3-1.4 years, whereas the CTM results were around 1.75 years for latitudes south of 30° S. Analysing transit time distributions derived from CTM results, Orbe et al. (2016) obtained modelled mean tropospheric age values of 1.5-2 years from the surface up to 200 hPa in the southern extra-tropics. Comparing results of a newer model run to surface observations, Orbe et al. (2021) reported good agreement between surface observations and the model results in the northern hemisphere, but an increasing overestimation by the model towards southern latitudes. In the southern extra-tropics observation-based lag-times of approximately 1.5 years were significantly below the model result of approximately 2 years. The overestimation was largely attributed to the influence of high-SF_6 sites on the reference time series used to calculate the model lag times. This supports our choice of using the marine boundary layer zonal average as the reference time series."

Line 165: Related to the above comment, it might be helpful to briefly state how the initial values of T were obtained for those not familiar with the Rigby 2013 paper. For instance,

were they based on reanalysis output or a best fit to observed mixing ratios of some trace gases?

The initial transport parameters T_{ij} were constrained by observations of anthropogenic gases such as chlorofluorocarbons. They are assumed to be applicable for gases with similar emission characteristics, a condition which is fulfilled for SF6 (Cunnold et al. 1983, 1997, 2002).

For the revised manuscript, the related paragraph was extended and now reads:

„Transport between the individual boxes is realised by parametrising bulk advection and eddy diffusion with the latter term dominating the transport (Cunnold et al. 1983, 1994). This is done with a transport matrix T, with elements T_{ij}, that quantify transport between pairs of individual boxes. These transport parameters vary seasonally but have no inter-annual variability. They were derived based on the best fit to chlorofluorocarbon measurements from the AGAGE observational network and can be assumed to yield reasonable results for gases with similar emission characteristics, which holds for gases emitted mainly from anthropogenic sources such as SF_6 (Cunnold et al. 1983, 1997, 2002).“

Lines 248-9: The difference between the PBL and UT gradient is also much smaller in the model compared to the observations. That seems worth pointing out here.

We agree and added the following statement to the revised manuscript:

„In addition, the difference between the surface and the upper troposphere is smaller in the model than observed (cf. Fig. S5).“

Line 261: ‘were performed’

Changed as suggested.

References

Cunnold, D. M., Weiss, R. F., Prinn, R. G., Hartley, D., Simmonds, P. G., Fraser, P. J., Miller, B., Alyea, F. N., and Porter, L. (1997), GAGE/AGAGE measurements indicating reductions in global emissions of CCl_3F and CCl_2F_2 in 1992–1994, *J. Geophys. Res.*, 102(D1), 1259–1269, doi:10.1029/96JD02973.

Cunnold, D. M., et al., In situ measurements of atmospheric methane at GAGE/AGAGE sites during 1985–2000 and resulting source inferences, *J. Geophys. Res.*, 107(D14), doi:10.1029/2001JD001226, 2002.

Strahan, S. E., Duncan, B. N., and Hoor, P.: Observationally derived transport diagnostics for the lowermost stratosphere and their application to the GMI chemistry and transport model, Atmos. Chem. Phys., 7, 2435–2445, https://doi.org/10.5194/acp-7-2435-2007, 2007.

Strahan, S. E., Douglass, A. R., and Steenrod, S. D. (2016), Chemical and dynamical impacts of stratospheric sudden warmings on Arctic ozone variability, *J. Geophys. Res. Atmos.*, 121, 11,836–11,851, doi:10.1002/2016JD025128.