

Response to comments by Anonymous Referee #3

We thank the anonymous reviewer #3 for their comments and for their suggestions to improve the manuscript. All comments are addressed in the following with the reviewer's comments printed in blue, and the responses in black.

General comments

This study combined upper tropospheric (UT) SF6 datasets from different measurement projects and attempted to interpret them in terms of emissions and transport. Such efforts to utilize datasets from multiple projects are of particular importance from long-term point of view. I think that a follow-up study using a 3D transport model is needed, but this manuscript has made a good job to motivate such future studies. Before agreeing to publication of this study, I would like to encourage the authors to consider my comments below to enrich scientific discussion and provide better guidance to following studies.

Orbe et al. (2021) presented detailed analyses of the SF6 age (or lag time as called in this study) including the data from the ATom campaign. This study repeats analysis of the ATom data, and extends to CARIBIC, GhOST and HIPPO, therefore number of the data in the UT is extensively increased. However, spatial distributions of the SF6 age presented in this study is Figure 4 only. With the data newly analyzed here, the authors could provide more presentations about spatial variations of the SF6 age like Figures 4 and 5 in Orbe et al. (2021). In particular, using the CARIBIC data, the authors might discuss longitudinal variation of the SF6 age over NH, which could reflect distribution of the underlying sources as well as areas of effective vertical transport. Also, the authors might confirm (or discuss difference of) the vertical gradient of the SF6 age over different latitudinal regions in comparison to Orbe et al. (2021). I think that such more in-depth analysis would highlight value of this study and further motivate follow-up studies using a 3D transport model.

In contrast to the study by Orbe et al. (2021) our study excludes stratospheric data and measurements at low altitudes. Time lags derived from a reference time series differ considerably for tropospheric and stratospheric data and to account for the different transport pathways, different reference time series would have to be chosen for tropospheric and stratospheric observations. Low altitudes were excluded because they are not covered by CARIBIC data. Therefore, a Figure similar to Fig. 5 of Orbe et al (2021) showing altitude profiles is not included.

We appreciate the suggestion of showing a spatial distribution of the derived lag times and have included a figure showing the spatial distribution of the measurement locations, colour-coded by lag-time in the revised version of the supplementary document. A corresponding figure reference has been added to the main text in section 3:

"The spatial distribution of lag-times is shown in the map in the supplementary Fig S1."

One important question (but I cannot find the answer clear) in this study is whether the SF6 age in the UT of SH has a secular decreasing trend, in contrast to Orbe et al. (2021) who

showed negative trends of the SF6 age south of 30°N at the surface. In abstract and conclusion, the authors argue that it exists, but they describe it a bit more carefully in section 3. As the authors writes, data are relatively limited in SH, so the trend line shown in Figure 5 could be unrepresentative (not free from sampling bias). It would be interesting to see the authors' argument more clearly about whether they regard the trend significant/well represented or still difficult due to limited availability of data. Regarding this, I wonder about possible use of the modeling results. The authors optimized the model (though it was tuned by more number of NH data). Figure 8 indicates almost constant value of data minus model with time in the UT exT-S. Does it mean that the model consistently shows a negative trend? What if the authors add the optimized model trend line to Figure 5? Thinking that the additional constraints from the newly analyzed data seem to be value of this study, I would like to see extended presentation of the outcome (e.g., modified Figure 5 or new figure after Figure 8) to answer the question.

As the southern hemisphere extra-tropics are clearly under-represented in the dataset we prefer to stay with the careful wording about the trends. As pointed out by another reviewer, the abstract wording and Figure 5 were not consistent regarding the southern hemisphere. The abstract wording about the trends has been changed to:

“At the most southern latitudes, a lag time of over 1 year with respect to the northern mid-latitude surface is derived, and lag times decrease over the period 2006-2020 in the extra-tropics and the southern tropics.”

In the main text, we follow the reviewer's suggestion to also discuss the trends found in the modelled lag times. Restricting the trend analysis to the years in which observational data are available in the respective latitude band, all trends are negative in the model output, but are statistically insignificant (at the 2-sigma level) except for the northern extra-tropics. There we find a negative trend of -0.07 years per decade (± 0.02) which is much smaller than what is derived from the observations (-0.14 ± 0.05 yrs/dec). The following section was added in section 4:

“Fitting a trendline in each latitude band over the time period covered by observations as visible in Fig. 5, modelled trends are negative in all four upper tropospheric boxes, but are not statistically significant (2σ). Only in the northern extra-tropics (Box 4), a statistically significant trend of -0.07 ± 0.02 years/decade is obtained, smaller than the value of -0.14 ± 0.05 years/decade in the observations.”

Figure 5 was left unchanged, because adding four more trend lines would make it more difficult to extract information from the figure. Given that of these only one line has a slope significantly different from zero, having model results in the figure would add only minor information.

Minor comments

Introduction: Interhemispheric transport with surface emissions mainly in the NH mid latitudes shapes latitudinal gradient of trace gas of interest. Citing previous studies, the authors highlight more effective interhemispheric transport in the UT than near the surface. Many

trace gases with major emissions in the NH (e.g., CO₂, CH₄, CO, SF₆ etc.) shows smaller latitudinal gradient in the UT than near the surface, but this cannot be explained by different efficiency of interhemispheric transport in different vertical layers only. I think that discussion on upward air inflow from surface to the UT over the tropics (the major pathway of surface air into the UT), is missing despite that it also mitigates latitudinal gradient in the UT. Japanese groups have made significant contributions to interpretation of latitudinal gradients of CO₂ and related gases in the UT and near the surface (Nakazawa et al. 1991; Miyazaki et al. 2008; Sawa et al. 2012; Bisht et al. 2021). The authors also mention to the Asian summer monsoon. Important in the context of this study is probably that Asian summer monsoon effectively uplift South Asian surface air (“polluted” air) to the UTLS, the gateway to interhemispheric transport as the authors cite previous studies like Yan et al. (2021) and Belikov et al. (2022). Such an express pathway from NH low latitude surface to the UT of SH also weakens the latitudinal gradient in the UT. I think that the introduction could be reformulated so as to inform readers about supply of surface air into the UT by various pathways also play an important role in determining the latitudinal gradient in the UT.

The statement on vertical transport in the introduction has been extended to better account for other transport pathways by adding the following text:

“The parametrization of convection and thus vertical transport from the surface to higher altitudes was identified as an important factor for differences of interhemispheric transport time between models (Orbe et al. 2018, Krol et al. 2018).”

While the interhemispheric gradient of SF₆ is mainly driven by the interhemispheric asymmetry of surface emissions, transport pathways from the surface to the upper troposphere also influence the latitudinal variation of mixing ratio at altitude (Miyazaki et al. 2009). In particular tropical convection can rapidly bring air masses with elevated mixing ratios of SF₆ and other tracers as for example from the Asian monsoon region or over tropical Africa to the upper troposphere (e. g. Randel and Park, 2006; Schuck et al., 2010; Vogel et al., 2016; Thorenz et al., 2017). Convection over remote marine tropical regions in contrast results in an inflow of air with low mixing ratios of anthropogenic tracers. This also implies that the interhemispheric gradient of SF₆ could vary with longitude as observed for example for CH₄ analysing measurements in the upper troposphere from IAGOS-CARIBIC (In-Service Aircraft for a Global Observing System - Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container) and CONTRAIL (Comprehensive Observation Network for TRace gases by AirLiner) flights into the southern hemisphere (Schuck et al., 2012).”

(Initial wording was *“The parametrization of convection and thus vertical transport from the surface to higher altitudes was identified as an important factor for differences of interhemispheric transport time between models (Orbe et al. 2018, Krol et al. 2018).”*)

P5 L114: I think that the original measurements were not made on the WMO X2014 scale (officially the scale is named without “NOAA”). It would be better to describe about the original measurement scale and the conversion applied retrospectively.

When revising the manuscript we changed the scale name to WMO 2014 (skipping “NOAA”) as suggested. In additional the following statement was added:

“Data measured prior to the publication of this scale were converted from the WMO 2006 scale to the WMO 2014 scale, both maintained by NOAA (https://gml.noaa.gov/ccl/sf6_scale.html). This was typically a correction of 0.01 ppt or less”

Table 1: The precision of SF6/N2O could be given in ppt/ppb, to be consistent with the following discussions. Use of ppb/ppbV is confusing. I think that the authors distinguish them on purpose, but no explanation is given. For instance, TRIHOP CO is in ppbV while UMAQS CO is in ppb despite the same measurement principle. This should be sorted out or consistent explanation should be given.

We agree that usage of ppb/ppbV was inconsistent and now use ppb. The choice of using absolute (ppt/ppb) vs relative (%) precisions depends on how these numbers are derived. For CARIBIC data, it is therefore more appropriate to report relative precisions, also for comparison with the numbers published earlier. For comparison within Table 1, absolute numbers in ppt/ppb were added.

P7 L135: “Because of their higher time resolution” high resolution does not mean observation of large variability. High-resolution measurements could capture phenomena that could not be resolved by low-frequency sampling, but underlying nature is that large variability is happening at place of interest. For instance, SF6 measurements at a very remote site (e.g., Antarctica) show small variability even if measured at high frequency.

We agree with the reviewer that the underlying variability of observations depends on the sampling location. As pointed out by the reviewer, high-resolution measurements may capture features which are unresolvable for methods with lower time resolution such as air sample collection, which in most cases averages over longer sampling times than online measurements. For observations in the same location this will lead to a higher variability in high-resolution data, therefore we think the above statement is correct and largely explains the higher variability of the research aircraft mission in comparison to the IAGOS-CARIBIC data despite their different spatial coverage.

Figure 2: The reference SF6 time series could be added to visualize the concept of the SF6 lag time.

The reference time series of SF6 has been added to Figure 2 in the revised manuscript and the figure caption was extended correspondingly.

Section 2.3: Orbe et al. (2021) pointed out that different choice of the reference time series could result in different trend of the SF6 lag time. The authors could discuss how this affects and why the simple marine boundary layer choice is made in this study.

This is now discussed in section 4, where the following paragraph was added:

„Similar results were obtained previously using the more sophisticated NASA Global Modeling Initiative chemical transport model (CTM) (Strahan et 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 south-

ern latitudes. At middle and high latitudes in the southern hemisphere, ground station observations yielded lag-times of 1.3-1.4 years, whereas the CTM results were around 1.75 years for latitudes south of 30° S. Analysing transit time distribution 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 a decreasing 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-SF₆ sites on the modelled 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.”

Figure 3: It would be interesting to see latitudinal distributions from surface sites along with those in the UT.

When preparing new figures during manuscript revision, the surface mixing ratios for the latitudes outside ±30° of each hemisphere were added for those months for which CARIBIC data is shown in Fig. 3.

Figure 4: As in my earlier comment, I hope to see more in-depth analyses of the SF₆ lag time, not only for latitude but for longitude and altitude.

The longitudinal variation is briefly touched on in the discussion of Fig 3. We’ve extended this by adding the following statement:

„Thus, the latitudinal gradient also has a longitudinal variation depending on emission and transport patterns.”

Discussion of Fig. 4 was changed and extended to:

“All four data sets agree with each other within their respective variability. In particular, this is the case for data from the HIPPO flight which cover a longitude range that is under-represented in the CARIBIC data and not covered by the included HALO missions. From this it can be concluded, that the longitudinal variability is smaller than the interhemispheric gradient. Thus, all observations are combined applying the above filter procedure to all observations simultaneously and zonal averages are discussed in the following.”

The dataset we use is restricted to measurements at pressure below 400hPa, excluding stratospheric data, covering a very small altitude range. We agree that a discussion of the vertical distribution of SF₆ mixing ratios would be interesting to look at, and we plan to do so in a future study. While the current study looks at interhemispheric transport in the troposphere, studying the vertical gradient of SF₆ should also include stratospheric data and discuss cross-tropopause transport. This is not possible without using a sophisticated model and thus beyond the scope of the current study.

P11 L195: As I commented to introduction, the smaller latitudinal gradient in the UT than

near the surface is not fully attributable to active interhemispheric transport in the UT but is also contributed by intrusion of surface air into the UT.

For the revised manuscript, the introduction was extended to account for this aspect. The here mentioned statements about consistency with the results obtained by Belikov et al. (2022) was left unchanged.

Figure 5: As in my earlier comment, this could be compared to results of the optimized model output. The trend line for ex-tropics south is hard to see in its color. Labels could be larger in size.

As the trends in the model output reflect the prescribed trend from the EDGAR emission flux time series, we deliberately do not discuss trends of the model output but use the detrended metric of the lag time for all comparisons.

When preparing new figures during manuscript revision, the label size was increased and a darker colour was chosen.

Section 4: The authors mention to possible southward shift of SF₆ emissions in the NH as discussed in previous studies (e.g. Orbe et al. 2021). I think that the authors could have made an experiment in which latitudinal emission patterns change with time in their box modeling. Yang et al. (2019) pointed out that vertical transport in the extratropics of NH (T04 in this study) is also important for north-south gradient of the SF₆ age. I wonder whether the authors have made sensitivity tests for this parameter. The authors argue that “horizontal transport seems to be more important than vertical transport” (P18 L333), at which I wondered about inconsistency with Yang et al. (2019).

In their sensitivity studies Yang et al. (2019) compared the sensitivity of tropospheric age (a_{SF_6} in their nomenclature) for which we prefer the term “lag time” and of the interhemispheric exchange time τ_{ex} . They concluded that the age a is more sensitive than the exchange time τ to perturbations of the vertical exchange between lower troposphere and upper troposphere over the northern extratropical source region (experiment S2) in their study. They obtain a similar result perturbing the transport from the northern extratropical source to the northern tropics.

Certainly the main difference between the sensitivity experiment performed by Yang et al (2019) and our current study is the multi parameter approach in which we vary all horizontal transport parameters simultaneously. We agree that vertical transport does also play a role. However, from the time lag differences between the model and the observations for neighbouring boxes presented in Fig. S4 and S5 we conclude that the vertical transport parameters have a smaller impact on modelled lag times (corresponding to tropospheric a in the Yang et al (2019) study) than the horizontal transport parameters and therefore restricted the sensitivity study to the latter.

Adding a time variation of the emission fluxes (beyond the time variation inherent to the EDGAR emission fluxes time series) would introduce an even higher degree of complexity to the sensitivity runs. A time change of the emissions would of course be reflected in modelled

mixing ratios, but as laid out in section 4, these are strongly influenced by the initialization values of the model run.

Reference

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