Editor comment and author reply

EC:

“Public justification (visible to the public if the article is accepted and published):
Given the very small uncertainties in lifetimes presented here for these gases and the very significant changes in lifetime estimates, the treatment of uncertainty stands out as a potentially significant issue in the presentation of the results. As there are many steps in the analysis, and many types of uncertainty, the presentation of how uncertainty is carried through the analysis is difficult to follow and it is unclear what has been fully accounted for. The authors articulate how this is only representative of the current state of the atmosphere, and would not account for changes in upwelling - this is a useful addition/caveat to the manuscript, but a few more details are needed:”

Author reply:

We thank the editor for raising areas of the paper where more detail and clarification are required, and have endeavoured to do so, both in our reply here, and in the text itself.

EC:
“1. These metrics have been updated based on three measurement campaigns. To what degree is the timing and spatial extent of the these campaigns representative of the current atmosphere? If this sampling was performed on a CCM, for example, does it accurately reproduce the CCMs lifetimes, FRFs etc?”

Author reply:

The manuscript text has been updated to include a discussion of the impact of the spatial coverage of the measurement campaigns on uncertainties. The use of a CCM to reproduce lifetimes is beyond the scope of this article. The following text has been added to the manuscript:

The analysis was performed on 3 different campaigns, in 4 locations (twice in Sweden, and once each in Germany, Greece, and Nepal), so only covered mid and high latitudes in the northern hemisphere. These campaigns took place during different seasons (summer, autumn, and winter). Large campaigns of this nature are rare and expensive, so this paper used the data that was available. The fact that data from all campaigns and trace gases yield a consistent picture does however give some confidence in the results.

EC:

“2. How are instrument uncertainties accounted for in this method?”

Author Reply:

Instrumental uncertainties are, similar to Laube et al. (2020), incorporated as the square root of the sum of squares of the uncertainties of the repeats of measurements of the samples as well as the calibration standard:

$$Total\ uncertainty = \sqrt{SD_{sample}{}^2 + SD_{standard}{}^2}$$

These were derived as one standard deviation and propagated for both the calculation of the mean ages as well as the FRFs. The corresponding "5n" data set is shown in Fig. 2, and includes the min/max values in the x and y directions. This then formed the basis of the bootstrapping procedure described in the manuscript. In addition, repeat draws were enabled for the bootstrapping method to reduce the dependency of the results on individual data points.

The manuscript has been revised with the following to make this clear:

Instrumental uncertainties are, similar to Laube et al. (2020), incorporated as the square root of the sum of squares of the uncertainties of the repeats of measurements of the samples and the calibration standard. These were derived as one standard deviation and propagated for both the calculation of the mean ages as well as the FRFs. The corresponding "5n" data set shown in Fig. 2 therefore includes the corresponding min/max values in both the x and y directions. So, if the measurement mixing ratio was 1 ppt, and the instrument uncertainty was 1%, then the mixing ratios of 0.99 ppt, 1.00 ppt and 1.01 ppt, were all considered.

EC:

"3. How are uncertainties in entry mixing ratio and observed mixing ratio accounted for in the FRF estimate?"

The response to the editor's previous (2) point details how the uncertainties of the observed mixing ratios are accounted for in the FRF estimate. As there are no tropospheric long-term trends near the tropical tropopause for these gases, uncertainties in entry mixing ratios are not easily accounted for. However, Leedham-Elvidge et al., (2018) showed that our southern hemispheric ground-based trends closely match observations in the upper tropical troposphere for multiple similar gases, when shifted in time by 0.5 years. We have added the following statement to Section 2.3 of the manuscript as a disclaimer:

Although the CGO data set is from the Southern Hemisphere, Leedham-Elvidge et al., (2018) showed that, for long-lived compounds that are inert in the troposphere, a CGO trend shifted by 0.5 years provides a good representation of mixing ratios in the tropical upper troposphere, where most air enters the stratosphere. Note that we do not include uncertainties in tropospheric entry mixing ratios in our FRF and mean age calculations. This represents an additional source of uncertainty to our lifetime and ODP estimates that is not accounted for in our analysis.

EC:

"4. The model uncertainty (from FRF to lifetime) is partially described with the bootstrapping method - but what about the tracer method employed? Does that account for the uncertainties in

the well known tracers? It seems unlikely given the small uncertainties presented here for the current gases."

Author Reply:

As discussed in our response to point 2, the bootstrapping uncertainties are based on instrument uncertainties. Hopefully the respective changes are now making this clearer in the manuscript.
In respect to how uncertainty is handled in the tracer method, the paper used the following procedure (method 1): The 5n data set of lifetimes and FRFs was used to create a trendline and, using the FRF of our compound of interest as 'x', this was used to estimate the lifetime of the compound of interest. The entire 5n data set trendline was bootstrapped (repeat draws again enabled), and the resulting lifetime estimates (2000 of them) were recorded (along with the frequency with which each lifetime value occurred, so a lifetime of 250 years might appear once, while a lifetime of 340 years might appear 60 times etc…). A mean, weighted by the frequency of each result, was calculated for the FRF of the compound of interest as well as its uncertainty range (so FRF, FRF+ and FRF-, to one standard deviation), resulting in a mean, high and low estimate for lifetime, thus incorporating FRF uncertainty into the estimate.

However, this method can result in an underestimation of the uncertainty range as lifetime uncertainties of the known species are not considered. In light of the editor's concerns, an additional method has been implemented (method 2): The same technique as method 1 is used, however instead of using the FRF uncertainty range to generate the lifetime uncertainty range, we take the lowest and highest estimate of lifetime using the mean FRF.
Using methods 1 & 2 we have two sets of uncertainty estimates (i.e., from the power line bootstrapping and from the FRF uncertainty), which are independent form each other, so cannot be combined as the square root of the sum of squares, and instead must be added together. So if method 1 gave 315 (287-331) years and method 2 gave 315 (245-370) years, then the full uncertainty range would be 28 (=315-287) plus 70 (=315-245)=98 on the lower side, and 16 (=331-315) plus 55(=370-315)=71 on the higher side.

The figures in the manuscript have been updated to include the combined uncertainty range from methods 1&2. During this update it was noticed that, due to a copy-paste error, Table 4 contained the lifetime estimate drawn only from the Leedham-Elvidge et al., (2018) data using SF_6 mean age of 850 years, rather than the average of all estimates. This (and any related errors) has now been corrected.

To summarise the newly derived uncertainties reflect a) the uncertainties in the FRFs of the target species, b) FRFs at different ages of air, and c) lifetimes from different sources. These uncertainties in the lifetime estimation method are now more comprehensive.
In addition, Fig.4 has been updated to show the updated uncertainty range discussed above, and an additional bar added showing the average lifetime estimate and uncertainty range, which should aid the reader.

A statement has been added to the manuscript to clarify how uncertainties were calculated and to explain the enhanced uncertainty estimates now present.

EC:

"5. How are all of these uncertainties above then combined to produce the final estimate of uncertainty?"

As described in points 1-4, uncertainty is incorporated at each stage of the method, with each stage building on the previous one.

References:

Laube, J.C., Leedham-Elvidge, E.C., Adcock, K.E., Baier, B., Brenninkmeijer, C.A.M., Chen, H., Droste, E.S., Grooß, J.U., Heikkinen, P., Hind, A.J., Kivi, R., Lojko, A., Montzka, S.A., Oram, D.E., Randall, S., Röckmann, T., Sturges, W.T., Sweeney, C., Thomas, M., Tuffnell, E. and Ploeger, F. (2020) 'Investigating stratospheric changes between 2009 and 2018 with halogenated trace gas data from aircraft, AirCores, and a global model focusing on CFC-11', *Atmospheric Chemistry and Physics*, 20(16), pp. 9771–9782. Available at: https://doi.org/10.5194/acp-20-9771-2020.

Leedham-Elvidge, E., Bönisch, H., Brenninkmeijer, C.A.M., Engel, A., Fraser, P.J., Gallacher, E., Langenfelds, R., Mühle, J., Oram, D.E., Ray, E.A., Ridley, A.R., Röckmann, T., Sturges, W.T., Weiss, R.F. and Laube, J.C. (2018) 'Evaluation of stratospheric age of air from CF_4, C_2F_6, C_3F_8, CHF_3, HFC-125, HFC-227ea and SF_6; Implications for the calculations of halocarbon lifetimes, fractional release factors and ozone depletion potentials', *Atmospheric Chemistry and Physics*, 18(5), pp. 3369–3385. Available at: https://doi.org/10.5194/acp-18-3369-2018.