Response to reviewer

We thank the reviewer for the additional comments on our manuscript. The concerns presented by Reviewer 1 in their second review are similar to those presented in their first. We provide a brief rebuttal to these points, but would also like to draw attention to our detailed responses during and at the end of the discussion phase (https://acp.copernicus.org/preprints/acp-2020-843/acp-2020-843-SC1- supplement.pdf and https://acp.copernicus.org/preprints/acp-2020-843/acp-2020-843-AC1-supplement.pdf).

In response to point 1) from the reviewer and following the suggestions of the editor, we have made several additions to the manuscript to clarify that our method only differs slightly from the method used in previous peer-reviewed publications of Allin et al. (2015) and Laube et al. (2010), which contain more details on the method. These changes are detailed in the track changes document. We feel that the method description given in the manuscript is sufficient, and in accordance with citing previously published work in scientific papers.

In response to point 2) from the reviewer we reference the comparison of our method against measurements of replicate samples made on a GC-IRMS system (Zuiderweg et al. 2011, 2012), now presented in Appendix B. This method comparison is consistent over a 50 ‰ range, which makes it highly unlikely that any scale correction is relevant for our results, given the measurement precision (as we already argued before).

Regarding the reviewer's concerns about our use of a Rayleigh fractionation model to characterise the apparent isotopic fractionation in the stratosphere, we note that that this is an empirical framework that has been successfully applied to other stratospheric gases with different lifetimes, including CH_4, N_2O and H_2, as well as chlorine isotopologues of CFCs (see references in our previous comments). Isotopic fractionations of different processes are linearly additive (Kaiser et al. 2006). The $O(^1D)$ sink is small, between 2 % and 6 % of the total sink for CFC-11, -12, and -113 (Burkholder et al. 2013). Kaiser et al. (2006) also discuss in detail the effects of mixing and transport on the apparent isotope fractionations for N_2O (which we know with much better precision). While mixing and transport are certainly relevant and reduce the observed apparent stratospheric isotope fraction compared with the intrinsic photochemical isotope effects, the *variations* in these mixing and transport effects are negligible for the precision that we report for CFCs, as can be inferred from the more precise observations for N_2O. We have added some text to the discussion about the effects of mixing and transport.

We are convinced that the data we present and their statistical interpretation are robust. While a higher measurement precision may be achievable with a larger sample size, our results are more than sufficient to draw the conclusions made in the manuscript.

Response to editor

We thank the editor for their helpful suggestions. We have implemented all of these in the revised manuscript.

Specifically, we have clarified at the end of the introduction and in the methods section that 1) our method differs only in minor ways from previous published methods (Allin et al. 2015, Laube et al. 2010); 2) the same method has been applied successfully and published previously; and 3) this method gives isotope delta values for a set of laboratory photolysis samples compatible with independent GC-IRMS analyses (as described in Appendix B).

Furthermore, we added examples of previous uses of single detector isotope mass spectrometry to Appendix B.

We have also changed the conclusions following the editor's suggestion.

The changes to our manuscript are highlighted in the track changes document submitted alongside this response. We trust to have addressed all of the editor's comments.

References

Allin, S., Laube, J., Witrant, E., Kaiser, J., McKenna, E., Dennis, P., Mulvaney, R., Capron, E., Martinerie, P., Roeckmann, T., et al.: Chlorine isotope composition in chlorofluorocarbons CFC-11, CFC-12 and CFC-113 in firn, stratospheric and tropospheric air, Atmospheric chemistry and physics, 15, 6867–6877, 2015.

Laube, J., Kaiser, J., Sturges, W., Bönisch, H., and Engel, A.: Chlorine isotope fractionation in the stratosphere, Science, 329, 1167–1167, 2010

Burkholder, J. B., Mellouki, W., Fleming, E. L., George, C., Heard, D. E., Jackman, C. H., Kurylo, M. J., Orkin, V. L., Swartz, W. H., and 25 Wallington, T. J.: Chapter 3: Evaluation of Atmospheric Loss Processes, SPARC Report on the Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species, SPARC Report No. 6, 2013.

Kaiser, J., Engel, A., Borchers, R., and Röckmann, T.: Probing stratospheric transport and chemistry with new balloon and aircraft observations of the meridional and vertical N2O isotope distribution, Atmospheric Chemistry and Physics, 6, 3535–3556, 2006

Zuiderweg, A., Holzinger, R., and Röckmann, T.: Analytical system for carbon stable isotope measurements of light non-methane hydrocarbons, Atmospheric Measurement Techniques Discussions, 4, 101–133, 2011.

Zuiderweg, A., Kaiser, J., Laube, J., Röckmann, T., and Holzinger, R.: Stable carbon isotope fractionation in the UV photolysis of CFC-11 and CFC-12, Atmospheric Chemistry and Physics, 12, 4379–4385, 2012.