

Reviewer comments and author replies for 'Observationally-derived Fractional Release Factors, Ozone Depletion Potentials, and Stratospheric Lifetimes of Four Long-Lived CFCs: CFC-13 (CClF<sub>3</sub>), CFC-114 (C<sub>2</sub>Cl<sub>2</sub>F<sub>4</sub>), CFC-114a (CF<sub>3</sub>CCl<sub>2</sub>F), and CFC-115 (C<sub>2</sub>ClF<sub>5</sub>)'

RC1 (in full): 'Comment on egusphere-2025-4941', Anonymous Referee #1, 04 Nov 2025 reply  
This manuscript presents new estimates for the stratospheric lifetimes, fractional release factors (FRFs) and ozone depletion potentials (ODPs) for CFC-13, CFC-114, CFC-114a and CFC-115. The new estimates are based on stratospheric measurements rather than a combination of modelling and laboratory studies. They thus provide valuable complementary evidence regarding the current measurements and would be of direct interest for use in the Scientific Assessment of Ozone Depletion.

The analysis is good and the uncertainties are described so that it is certainly publishable material. However the structure of the manuscript makes it hard to read and understand. More thought on how the material is ordered and presented would make the paper stronger as well as more accessible. I do not think that more analysis is required.

The main problem is that there is no description on how the manuscript is structured as part of the introduction. Rather, it ends with a somewhat technical discussion of the difficulties of getting separate measurements of the two isomers CFC-114 and CFC-114a. Previous estimates of lifetimes, ODPs and FRFs are presented in Table 1. This could be presented more as a state of knowledge, a description of what new information this manuscript will provide (perhaps the current discussion section) and end with a summary of the sections that will follow. A clear discussion of the broader significance of the work is also lacking. Why should I care about FLFs, etc?

The manuscript would also benefit from similar 'heads up' material at the beginning of each main section (Methods, Results, etc.).

It is important to make clear the distinction between what information or data already exists in the literature and what new information is presented. For example, it was not clear to me on first reading that all the material in Table 1 is published and that it represents the current knowledge on which this manuscript builds. It is important that this is made clear in the accompanying text and in the caption.

The Discussion and Conclusion sections are both on the short side. Most of the material in the Discussion section could go in the Introduction as it is not really discussing the implications of the results. Can the paper do without a Discussion section by bolstering the material about implications in the Results section and ending with a stronger summary?

Minor comments

50/51 '...respectively, while the...'

Tab 1 caption Something wrong with formatting. More importantly, I think there should be a clear statement that these values are published elsewhere.

Introduction I don't think it is enough to

174/175 something gone wrong here – ‘..which is not a mathematical..’?

176 ‘..showed that the atmospheric ratio..’

208 ‘independent’

213 ‘dissociated’ more common

232 ‘..because of the non-linearities...’

243 ‘estimate’ rather than ‘prediction’

331 State what the Lickley simulation period is

364 How will the new metrics help understand the sources of these compounds?

365on This is the first time that this material has been presented. It should also be covered in the Introduction and the Discussion (if the authors decide to keep one). It is the type of broader discussion which I find lacking in the rest of the paper.

General Check correct format for references with date in brackets - ‘Laube et al, (2016)’. Is the comma needed?

Citation: <https://doi.org/10.5194/egusphere-2025-4941-RC1>

Author reply to RC1, quoting sections of RC1:

RC1 “This manuscript presents new estimates for the stratospheric lifetimes, fractional release factors (FRFs) and ozone depletion potentials (ODPs) for CFC-13, CFC-114, CFC-114a and CFC-115. The new estimates are based on stratospheric measurements rather than a combination of modelling and laboratory studies. They thus provide valuable complementary evidence regarding the current measurements and would be of direct interest for use in the Scientific Assessment of Ozone Depletion.

The analysis is good and the uncertainties are described so that it is certainly publishable material.”

Author Reply:

We thank the reviewer for this positive assessment of our work and its relevance to the Scientific Assessment of Ozone Depletion.

RC1: “However the structure of the manuscript makes it hard to read and understand. More thought on how the material is ordered and presented would make the paper stronger as well as more accessible. I do not think that more analysis is required.

The main problem is that there is no description on how the manuscript is structured as part of the introduction. Rather, it ends with a somewhat technical discussion of the difficulties of getting separate measurements of the two isomers CFC-114 and CFC-114a. Previous estimates of lifetimes, ODPs and FRFs are presented in Table 1. This could be presented more as a state of knowledge, a description of what new information this manuscript will provide (perhaps the current discussion section) and end with a summary of the sections that will follow. A clear discussion of the broader significance of the work is also lacking. Why should I care about FLFs, etc?

The manuscript would also benefit from similar 'heads up' material at the beginning of each main section (Methods, Results, etc.)."

Author reply:

We thank the reviewer for these helpful suggestions to improve the structure and readability of the manuscript. We have added the suggested 'heads-up' material to better sign-post the content for the reader.

RC1: "It is important to make clear the distinction between what information or data already exists in the literature and what new information is presented. For example, it was not clear to me on first reading that all the material in Table 1 is published and that it represents the current knowledge on which this manuscript builds. It is important that this is made clear in the accompanying text and in the caption."

Author reply:

Thank you for catching this oversight, we have now added the necessary clarification to the text and caption.

RC1: "The Discussion and Conclusion sections are both on the short side. Most of the material in the Discussion section could go in the Introduction as it is not really discussing the implications of the results. Can the paper do without a Discussion section by bolstering the material about implications in the Results section and ending with a stronger summary? "

Author reply:

There was a lively discussion amongst the co-authors of this paper regarding the discussion section of the manuscript; what should go there, what should be removed, and whether a discussion section was necessary in this case. In light of this and your comment, some restructuring along the lines you suggest, has been completed.

RC2 in full:

“This study uses stratospheric observational data and correlations between FRFs and lifetimes of well-studied ozone depleting substances to determine the steady-state stratospheric lifetimes of minor CFCs. The work in this paper provides a valuable contribution to our understanding of the lifetimes and emissions of these CFCs. Most of my comments are on clarity and framing with one major point about proper accounting of uncertainties.

The largest concern I have with this paper is with regards to the following:

uncertainties in lifetimes of well-studied compounds are largely due to uncertainty in upwelling rates. That would imply that the bias in lifetimes estimates would be correlated to bias in assumed age of air – if upwelling is faster than believed, then AoA would be younger than believed and lifetimes shorter. I may have missed this but I believe this study glosses over this relationship and assumes AoA is well constrained and that biases in AoA are independent of those in atmospheric lifetimes. How would accounting for this correlated bias change the results here? Do the uncertainties noted here account for that? Further, are the uncertainties in the atmospheric lifetimes used to fit your curve shown in Fig 3 assumed to be independent or correlated with one another? For example, how would it impact your reported uncertainties if they were perfectly correlated?

Minor comments:

Line 84 – suggestion to add uncertainties to CFC-115 lifetimes.

Line 93 – an impurity in the production of CFC-12 (Western et al, 2023). I didn't see that in the referenced paper, instead I found: “CFC-13 may be a by-product during the production of CFC-11” referencing [Vollmer, M. K. et al. Atmospheric histories and emissions of chlorofluorocarbons CFC-13 (CClF<sub>3</sub>), CFC-114 (C<sub>2</sub>Cl<sub>2</sub>F<sub>4</sub>), and CFC-115 (C<sub>2</sub>ClF<sub>5</sub>). *Atmos. Chem. Phys.* 18, 979–1002 (2018)]. Alternatively, Western references the potential for CFC-13 emissions during the plasma arc destruction of CFC-12, but references: [Murphy, A. B., Farmer, A. J. D., Horrigan, E. C. & McAllister, T. Plasma destruction of ozone depleting substances. *Plasma Chem. Plasma Process.* 22, 371–385 (2002)]. I suggest referencing the original source rather than the Western paper, as Western's paper was not directly studying the production/manufacturing processes.

I'm having a hard time interpreting the method presented to deal with the separation of CFC114 from 114a. For example, can you explain what is being correlated with what here:

A simple linear correlation (without offset) was calculated for both the Vollmer et al., (2018) and the UEA CGO data, and comparison of these linear correlations was used to derive a conversion factor “x” with the lowest residual sum of squares (RSS) (which were CFC-13=0.11, CFC-115=0.11, and  $\Sigma$ CFC-114=0.81), to attempt to make UEA measurements compatible with Vollmer et al., (2018).”

I also do not follow why the authors take the sum of CFC-114 and CFC-114a data “in order to be compare to Vollmer et al., (2018)” even though “Vollmer et al. did not distinguish between CFC-114 and CFC-114a, and so the data for these two compounds are combined to give ‘ $\Sigma$ CFC-114’ which not a mathematical sum of the two, rather a weighted sum.”

Equation 2 must assume there’s no trend in entry mixing ratios? Or is a time lag of the approximate age of air accounted for in this equation?

Line 270: Which compounds are the “well-studied” ones used here? How do you account for the uncertainties of the lifetimes of these compounds?

Line 287: I don’t think CFC-12 uncertainties are symmetric. I believe the uncertainties around the inverse of lifetimes are symmetric. Also – I would add the reference to the CFC-12 estimate provided here

Line 325 – CFC-114 is the exception because its lifetime was not updated, correct? This could be explained “, with the exception of CFC-114 (Fig. 5), whose lifetime estimate did not change significantly.”

I suggest reworking the framing of last paragraph a bit as follows: Edit the first sentence to something like:

“While this study suggests that emissions of CFC-113 and 115 are likely higher than previously estimated, it remains unclear whether additional emissions are released from long-term banks or new production.” And then the last sentence should be edited to something like: “This study does not attribute the source of emissions, but our shorter estimated lifetimes are consistent with higher inferred emissions, leaving the question of unaccounted

Citation: <https://doi.org/10.5194/egusphere-2025-4941-RC2>”

Author reply to RC2 quoting sections of RC2:

RC2: “This study uses stratospheric observational data and correlations between FRFs and lifetimes of well-studied ozone depleting substances to determine the steady-state stratospheric lifetimes of minor CFCs. The work in this paper provides a valuable contribution to our understanding of the lifetimes and emissions of these CFCs. Most of my comments are on clarity and framing with one major point about proper accounting of uncertainties. “

Author Reply:

We are pleased that our work and its potential contribution to scientific understanding of the lifetimes and emissions of these CFCs, was received so favourably by the reviewer.

RC2:“The largest concern I have with this paper is with regards to the following:

uncertainties in lifetimes of well-studied compounds are largely due to uncertainty in upwelling rates. That would imply that the bias in lifetimes estimates would be correlated to bias in assumed age of air – if upwelling is faster than believed, then AoA would be younger than believed and lifetimes shorter. I may have missed this but I believe this study glosses over this relationship and assumes AoA is well constrained and that biases in AoA are independent of those in atmospheric lifetimes. How would accounting for this correlated bias change the results here? Do the uncertainties noted here account for that? Further, are the uncertainties in the atmospheric lifetimes used to fit your curve shown in Fig 3 assumed to be independent or correlated with one another? For example, how would it impact your reported uncertainties if they were perfectly correlated? “

Author Reply:

We thank the reviewer for highlighting this oversight. In response we have added the following paragraph to the appropriate section as this provides wider context to the findings of this paper:

‘This method of calculating time-independent FRFs is able to correct for changes in the tropospheric trends of the CFCs, however it cannot account for changes in tropical upwelling. This is because the lifetimes calculated are steady-state lifetimes, and rely on the atmosphere to remain in a certain state. If the atmosphere changes, such as with a drastic change in tropical upwelling, then a new steady state would eventually be reached, with a new corresponding steady-state lifetime. There is evidence that stratospheric circulation is changing, and in turn affecting the lifetimes of long-lived tracer gases, such as Prather et al., (2023). However that paper focused on N<sub>2</sub>O, which has different sink regions to the CFCs examined here, and we do not have a global, high-quality satellite-based dataset for these CFCs. Therefore conclusions from Prather et al., (2023) cannot be transferred to this paper (as it would also be beyond the scope of our work). However it can be argued that the current observed N<sub>2</sub>O lifetime changes are relatively small, and for the four long-lived CFCs examined here, it could be expected to be well within the uncertainties that we derive. So while this method cannot completely account for the effect of upwelling, the lifetimes presented in Sect. 3.1, still represent a significant improvement to previous estimates.’

RC2: “Further, are the uncertainties in the atmospheric lifetimes used to fit your curve shown in Fig 3 assumed to be independent or correlated with one another?”

Author Reply:

The lifetimes in Leedham-Elvidge et al., (2018) were indeed calculated separately, but all rely on the same a. lifetime estimates of CFC-11, b. correlation slope with CFC-11, and c. correlation slope of CFC-11 versus mean age. They are therefore not entirely independent of each other. However, there can be some confidence to be drawn from the fact they (in particular the results from the power trendline) compare well to those in Burkholder et al., (2022) (which were derived using multiple methods, not in situ alone). Some clarifications have been added to the manuscript to reflect this.

“Minor comments:

Line 84 – suggestion to add uncertainties to CFC-115 lifetimes.”

Author reply: Good catch, added.

RC2: “Line 93 – an impurity in the production of CFC-12 (Western et al, 2023). I didn’t see that in the referenced paper, instead I found: “CFC-13 may be a by-product during the production of CFC-11” referencing [Vollmer, M. K. et al. Atmospheric histories and emissions of chlorofluorocarbons CFC-13 (CClF<sub>3</sub>),  $\Sigma$ CFC-114 (C<sub>2</sub>Cl<sub>2</sub>F<sub>4</sub>), and CFC-115 (C<sub>2</sub>ClF<sub>5</sub>). Atmos. Chem. Phys. 18, 979–1002 (2018)]. Alternatively, Western references the potential for CFC-13 emissions during the plasma arc destruction of CFC-12, but references: [Murphy, A. B., Farmer, A. J. D., Horrigan, E. C. & McAllister, T. Plasma destruction of ozone depleting substances. Plasma Chem. Plasma Process. 22, 371–385 (2002)]. I suggest referencing the original source rather than the Western paper, as Western’s paper was not directly studying the production/manufacturing processes.”

Author reply: This has now been corrected/changed in the manuscript.

RC2: “I’m having a hard time interpreting the method presented to deal with the separation of CFC114 from 114a. For example, can you explain what is being correlated with what here:

A simple linear correlation (without offset) was calculated for both the Vollmer et al., (2018) and the UEA CGO data, and comparison of these linear correlations was used to derive a conversion factor “x” with the lowest residual sum of squares (RSS) (which were CFC-13=0.11, CFC-115=0.11, and  $\Sigma$ CFC-114=0.81), to attempt to make UEA measurements compatible with Vollmer et al., (2018).”

Author reply: The correlation between mixing ratio and date was plotted for each compound (or in the case of CFC-114/a, the combined mixing ratio). This was done separately for the Vollmer et al data and the Cape Grim observatory data. These two plots were then compared to each other.

“I also do not follow why the authors take the sum of CFC-114 and CFC-114a data “in order to be compare to Vollmer et al., (2018)” even though “Vollmer et al. did not distinguish between CFC-114 and CFC-114a, and so the data for these two compounds are combined to give ‘ΣCFC-114’ which not a mathematical sum of the two, rather a weighted sum.” “

Author Reply: in Vollmer et al., (2018) CFC-114 and CFC-114a are not separated. Our analysis process is able to separate the two compounds. In order to compare our data to Vollmer et al, we needed to combine our CFC-114 and CFC-114a data. This was worded rather awkwardly in the original manuscript and has now been revised to clarify that it is OUR CFC-114/a data that is being combined, in order that it could be to the Vollmer et al data. As Vollmer et al. does not separate isomers, our data does not necessarily have to correlate to theirs. The fact that it indeed does, shows that Vollmer et al. present a reasonable approximation to the sum of CFC-114 and CFC-114a, although they are not able to observe the changing trend of CFC-114a (Western *et al.*, 2023) as the sum is dominated by CFC-114. A statement to this affect has been added to the manuscript.

RC2: “Equation 2 must assume there’s no trend in entry mixing ratios? Or is a time lag of the approximate age of air accounted for in this equation?”

Author reply: Equation 2 is a highly simplified equation to aid understanding of the process, but leaves out the precise mechanism by which entry mixing ratios are calculated (as this would simply be reproducing Ostermüller et al., (2017)’s work). Ostermüller’s ‘time-independent’ method does indeed account for changes in entry mixing ratios.

RC2: “Line 270: Which compounds are the “well-studied” ones used here? How do you account for the uncertainties of the lifetimes of these compounds?”

Author reply: These compounds are: SF<sub>6</sub>, HCFC-141b, HCFC-142b, HCFC-22, CFC-12, CFC-113, CFC-11, H1301, CCl<sub>4</sub>, CH<sub>3</sub>CCl<sub>3</sub>, H1211, and they are shown in Fig 3, though not initially listed in the text. This has now been rectified. On rereading the manuscript it was not made clear that the bootstrapping procedure was also performed on the FRF-Lifetime correlation, and that the uncertainty ranges for these compounds’ lifetimes were included in that bootstrapping procedure. This has now been clarified in text.

RC2: “Line 287: I don’t think CFC-12 uncertainties are symmetric. I believe the uncertainties around the inverse of lifetimes are symmetric. Also – I would add the reference to the CFC-12 estimate provided here

Author Reply: Added reference for the CFC-12 estimate. The lifetime uncertainties seen in Fig 3a are taken from Leedham-Elvidge et al., (2018), and those in Fig 3b are from Burkholder et al., (2022). The caption for this figure has been updated to clearly reflect this. Neither source includes asymmetric uncertainties for these compounds, and this is reflected in Fig 3.

RC2: “Line 325 – CFC-114 is the exception because its lifetime was not updated, correct? This could be explained “, with the exception of CFC-114 (Fig. 5), whose lifetime estimate did not change significantly.”

I suggest reworking the framing of last paragraph a bit as follows: Edit the first sentence to something like:

“While this study suggests that emissions of CFC-113 and 115 are likely higher than previously estimated, it remains unclear whether additional emissions are released from long-term banks or new production.” And then the last sentence should be edited to something like: “This study does not attribute the source of emissions, but our shorter estimated lifetimes are consistent with higher inferred emissions, leaving the question of unaccounted”

Author reply: Very useful suggestions – thank you! The paragraph has now been reworked along those lines for enhanced clarity.

#### References:

**RC2 Citation:** <https://doi.org/10.5194/egusphere-2025-4941-RC2>

Burkholder, J.B. and Hodnebrog, Ø. (2022) ‘ANNEX - Summary of Abundances, Lifetimes, ODPs, REs, GWPs, and GTPs’, *World Meteorological Organization (WMO). Scientific Assessment of Ozone Depletion: 2022, GAW Report No. 278*, Geneva: World Meteorological Organization (WMO), pp. 435–492. Available at: <https://ozone.unep.org/science/assessment/sap> (Accessed: 11 June 2024).

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Western, L.M., Vollmer, M.K., Krummel, P.B., Adcock, K.E., Fraser, P.J., Harth, C.M., Langenfelds, R.L., Montzka, S.A., Mühle, J., O'Doherty, S., Oram, D.E., Reimann, S., Rigby, M., Vimont, I., Weiss, R.F., Young, D. and Laube, J.C. (2023) 'Global increase of ozone-depleting chlorofluorocarbons from 2010 to 2020', *Nature Geoscience* [Preprint]. Available at: <https://doi.org/10.1038/s41561-023-01147-w>.