

Interactive comment on "Abundances, emissions, and loss processes of the long-lived and potent greenhouse gas octafluorooxolane (octafluorotetrahydrofuran, c-C₄F₈O) in the atmosphere" by Martin K. Vollmer et al.

Anonymous Referee #2

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General comments:

The article presents a large dataset and budget estimate for a newly detected compound in the atmosphere: $c-C_4F_8O$. Although its abundance is small (less than 0.1ppt) its radiative efficiency is strong and lifetime likely very high. It is still unregulated and sometimes viewed as a promising compound in terms of industrial applications (see for example Kočišek et al., 2018). I think that its scope and novelty make it adequate for a publication in ACP. I have some comments on the methodology and presentation.

C1

The Northern Hemisphere (NH) measurements are little described and commented (p 1 I5-6, p3 I14-15). The article should explain how the North Hemisphere trend (dashed line on Figure 1) was constrained and evaluate the uncertainty on emissions resulting from the lack of NH constraints.

Similarly, the mixing ratio and emission trends between 1950 and 1978 are mostly constrained by a single firn air data point undergoing a large age distribution, and having a mixing ratio (6 ppq, Table S4) very close to the detection limit (5 ppq, p4 I13). The article should explicitly discuss the constraints on the anthropogenic versus natural sources of $c-C_4F_8O$, as well as the little constrained early emissions.

However, for a well-mixed very long lived species, a reasonable estimate of global emissions can be obtained from a simple one box model calculation. Presenting this simple calculation and comparing it to the elaborate approach used would improve the description of the main uncertainties and be helpful to non-specialist readers.

A first estimate of the lifetime of $c-C_4F_8O$ is provided but some important assumptions should be better described: the basis for the estimated Lyman- α lifetime and OH reactivity (comparison with species having similar bonding structures?), the possible role of other unexplored sinks such as surface loss (to ocean and land) and heterogeneous processes should be discussed at least in terms of perspectives.

Specific comments:

p2 I10-11: The Californian regulation could be mentioned (https://ww2.arb.ca.gov/resources/documents/semiconductor-regulation)

p5 I11-14: As pumping out the interstitial air from deep firn can be difficult and induce contamination, more indications should be provided about the multi-species consistency of model results for the deep firn air sample used and the overall firn. For example, the RMSD/ σ indicator used in Buizert et al. (2012) could be provided. The reason why so few depth levels were analyzed for c-C₄F₈O should be given, sample

size issue?

p5 I14-18: The Trudinger et al. (2013) model uses both molecular and eddy diffusivity terms. As this has the same effect as modifying the diffusion coefficient, the relative roles of molecular and eddy diffusivity terms for the ABN firn should be commented.

p5 I19-26: how were the North Hemisphere concentrations evaluated?

p5 I29: Vollmer et al. (2016, 2018) used multi-depths firn air constraints from both hemispheres. The methodological adaptations to the lack of NH constraints should be described.

p6 I1: I do not understand what the Green's functions from the 12-box model are and did not see an explanation in Vollmer et al. (2016, 2018)

p6 I11-13: The emission values in Ivy et al. (2012) start in 1980 (Table 3), how was the prior estimate designed for the 1950-1980 period and what impact does it have on the final solution for this weakly constrained period?

p7 l23 and after, including section 2.2 of the Supplement: a single notation should be adopted to name reaction rates, avoid using k_R , then k_1 (implicit) and k_2 , then $k_{c-C_4F_8O}$.

p10 l9: Figure 4 is little commented, it could be shifted to the Supplement or combined with Fig. 2

p10 l26-27: circular argument, the calculated growth rate is small because the measured concentration trend is weak (in recent years), not the contrary.

p11 I6-9: the important Aspendale dataset (thousands of measurements) is briefly summarized in Table S3 and very briefly commented. A more in-depth discussion of $c-C_4F_8O$ variability at various sub-annual time scales and recent trend, as well as a plot (at least in the Supplement) would be useful.

p11 l26: the wording "a few other synthetic greenhouse gases" implicitly assumes that $c-C_4F_8O$ is purely anthropogenic but this is not discussed in the article

C3

p12 I6: due to the high cost of Antarctic field operations, research programs and logistic institutions financing them are usually explicitly named.

pS7 I16 of the Supplement: the chosen 1 ppq uncertainty seems small compared to the stdv values in Table S3, this choice should be further commented.

pS10 l6-10 of the Supplement: this discussion of background / non anthropogenic level of c-C $_4F_8O$ should be in the main article

Technical corrections:

p 5 l7 use indices in c-C4F8O

p 5 |11-14 repeats |4-8

p 5 l16 and 18 suppress))

p 6 I12 from observations of

New references:

Kočišek, J, Janečková, R. and Fedor, J., Long-lived transient anion of c-C₄F₈O, Journal of Chemical Physics, 148(7), 074303, 2018.

Buizert et al., Gas transport in firn: multiple-tracer characterisation and model intercomparison for NEEM, Northern Greenland, Atmos. Chem. Phys., 12, 4259-4277, https://doi.org/10.5194/acp-12-4259-2012, 2012.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-852, 2018.