Interactive comment on “Perfluorocyclobutane (PFC-318, c-C₄F₈) in the global atmosphere” by Jens Mühle et al.

Anonymous Referee #1

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The authors have brought together an impressive data set for atmospheric observations and modelling of PFC-318, which is global, long-term and consistent. The manuscript is well written but perhaps lacking succinctness a bit, which could be helped by moving some rather technical aspects that are not essential for the narrative to the supplement. A general concern is that the authors focus very much on their hypothesis of PTFE production as the main source of PFC-318 to the atmosphere. Other sources are barely mentioned let alone discussed, whereas the observations in my opinion point to a much more complex picture of emission sources (including unknowns). This should be given some more consideration. Other points are listed below.

L119 I don’t think a personal communication can be counted as evidence

L169-178 Have the authors ascertained that their calibration system has a linear response behaviour over a relevant mole fraction range? How was the calibration scale uncertainty estimated?

L187 “perhaps slightly better”?

L210 “perhaps”?

L214-249 It is commendable that the authors have carried out these tests. However, the high number of statistical outliers is worrying and casts some doubt on the derived long-term trends, in particular the early parts. Adding uncertainty ranges to the fits based on a) the samples that were included but showed discrepancies between Medusas and b) the sparsity of the measurements, might help here. In addition, I recommend moving this rather technical paragraph to the supplement.

L225 “eighth”

L267 “a”

L513 How high is the interhemispheric gradient and how has it evolved over time? This might e.g. reveal information on changes in emission latitudes. There is a lot of space in Figures 1 and 4 to show this.

L523 Define “good agreement”. There are no uncertainty ranges given for the two estimates in Figure S7.

L537-538 Again, are these discrepancies within uncertainties?

L555 GWP-100?

L571-574 Given that the largest emissions appear to occur near the sea, is there scope for some emissions being related to ships or submissions? What fraction of emissions did the model initially assign to have occurred over the ocean?

L590-591 This appears to be in disagreement with the statement in L576-578.

L603 FABS?
L631-638 Please add information such as measurement precisions, observed mole fraction ranges, ions used for identification and quantification, etc. on the HFP measurements to the manuscript. Please provide quantitative evidence instead of “associated with” and “virtually absent”.

L639-641 Consistent with emissions from many of these facilities, but clearly not all (as stated in L610-612). Given the problems with associating these sources can the authors confirm that the ratios between m/z 131 and 101 during pollution events were consistent with those observed in clean air? This would help to rule out interferences during pollution events.

L661 How much smaller?

L697 That is a very optimistic way of looking at that Figure.

L706-707 This is not very clear from the Figure, which is rather indicating an unknown source.

L713 What is the main purpose of this direct c-C4F8 production?

L729 Which ones did PFC-318 correlate best with (also for other pollution events in Asia etc)?

L740-741 How much larger?

L1236 Figure 1 is mostly demonstrating quality assurance purposes and one cannot see most station data anyway as it is on top of each other. As the long-term trend is shown again in Figures 3 and 4 I suggest moving it to the supplement.

L1256 Is it necessary to show years from 1900 if the first data point is after 1930? What is the uncertainty of the calculated effective ages?

Supplement

Figure S1 The caption is actually an entire section and should perhaps have its own heading.

Figure S3 There is quite some uncertainty in the 1960s and 70s. Has this been reflected in the emission uncertainties?

Figure S7 Please also show the published observational data set of Saito et al.

Table S3 RoW is not explained and web pages should be cited with the date on which they were accessed.