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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trends and Emissions of Six Perfluorocarbons in the Northern and Southern Hemisphere" *by* Elise S. Droste et al.

Anonymous Referee #2

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This paper presents measurements of PFCs at Cape Grim, Australia between 1978-2017, supplemented by measurements from Tacolneston, UK 2015-2017 and campaign-based sampling in Taiwan between 2013 and 2016. Long-term trends in mixing ratio and emissions are provided by the Cape Grim data, with the Tacolneston and Taiwan data providing information about emissions in their surrounding regions and the interhemispheric gradient. Although measurements of some of these PFCs at Cape Grim were presented in Laube et al., (2012), this new study is valuable because it has improved the calibration scale, allowed separation of some isomers, and extended the trends in time. I recommend publication after consideration of the following minor comments:

General comments:

1. Near where the isomers are first mentioned in the Introduction, it might be worth providing a paragraph that has a little more background on isomers (e.g., definition, explain the nomenclature n-isomer, i-isomer), and bringing this together with explanation of the significance of measuring the different isomers. The potential for distinguishing different source types is mentioned on page 4, line 23 and the different radiative efficiency is mentioned on page 4, line 9, and there is some discussion of the implications of separation of isomers from a measurement point of view elsewhere, but these points are interspersed with lots of other details, risking losing the overall significance of the work. A paragraph that discusses the significance of isomers in general, before going on with the details of this study, would be beneficial.

2. A clearer description of uncertainties is needed in Section 2.5. E.g., page 8, line 31 - what is the "averaged model-fit uncertainty of the Cape Grim trend" and how is it calculated? page 9, line 9 - "After these uncertainties were calculated" - uncertainties in what, PFC trends? Page 9, line 9 - "the model was re-run" - the inversion for emissions? I don't really understand this paragraph (lines 9-14). I do understand why you would want trend uncertainties without calibration uncertainty, it is the details of the description that I believe should be improved.

3. I am confused about the conclusion regarding the interhemispheric gradient of n-C6F14 and i-C6F14 (last sentence of section 3.1.3). Around page 13, line 7, I thought the authors were saying that the observations were close to or slightly exceeding a ratio of N:S mixing ratio the same as c-C4F8 (1.05), but that the measurement uncertainties were too large to discern the interhemispheric gradient. The conclusion at page 13, line 20 says that the NH sources are not as substantial - is this referring to NH sources relative to SH sources, and therefore influencing the interhemispheric gradient? We know that the recent total emissions for c-C4F8 are around 10 times the emissions of n-C6F14 and i-C6F14, so it is perhaps not surprising that the interhemispheric gradient will be small in an absolute sense, but that doesn't tell us about the relative sense. But I don't think we can conclude anything about the N-S distribution of emissions due to

the N-S mixing ratio difference expected for typical N-S emissions distribution, relative to the measurement uncertainty.

Specific comments:

page 3, line 4 - "As a result" - of what? Provide a better link to the previous sentence.

page 3, line 13 - "Other sources include ..." is this still talking about the low mass PFCs? Maybe "Other sources of PFCs include..."

page 10, line 16 - perhaps replace 'sources' by 'pieces' or 'lines', and leave the word 'sources' to describe emissions to the atmosphere. Same on line 19.

page 10, line 21 - "This is slightly less..." - what is? The 2017 value is slightly less?

page 12, line 2 - specify whether this is the interhemispheric ratio of emissions or mixing ratio

page 12, line 22 - "The trends are somewhat similar" - to what? Each other? Another PFC?

page 12, line 23 - The fastest increase occurred in the mid to late 1990s.

page 13, line 4 - I would add the years, i.e. "Mixing ratios in Taiwan in 2015 and 2016 are again ..."

page 15, line 21 - change to "for n-C4F10 (Fig. 8) and n-C5F12 (emissions < 5.3x10^{-5} Gg/yr)." as only emissions for n-C4F10 are shown in Fig 8. I know this is specified in the caption, but simpler for the reader to also specify here.

page 16, line 4 - Add "Cape Grim" as follows: "However the Cape Grim data in this time period"

page 16, line 15 - "although within the uncertainties of the early data set this is not a significant difference" - isn't it the sparsity of the data (i.e. timing of the increase depends on one sample, around 1996) rather than the data uncertainties as indicated

in the figure that mean we can't be conclusive about the timing of each increase? Is there a clue to the timing (of the n-C6F14 increase, at least) from the Ivy measurements of the Cape Grim air archive, as these are more dense in time?

page 16, line 16 - What does 'It' refer to? i-C6F14 emissions, presumably, but please specify.

page 16, line 29 - 'peak emission rates have increased' - please be clear what you are comparing here (n-C6F14 alone and the sum n-C6F14 + i-C6F14?)

page 16, line 33 - what is being compared to EDGAR emissions here? Are you comparing the black dashes with the red dots? Do they agree exceedingly well?

page 18, line 1 and Table S4 - is this talking about the correlation with CO described at the end of page 9? Or just footprints? Please explain what this is.

Fig 5 - I found this figure confusing because it is combined, and I think it would be less confusing if it were split into two panels. It is the use of different axes I found most confusing - it gives the wrong impression of the relative magnitude of the two isomers. I had to keep checking the caption/legend to remember which axis to use, and which isomer was which, as I came back to the plot a number of times as I read through the paper. The gridlines (although faint) don't match the right axis. The one advantage of combining the plots is that it is easy to compare the timing of the increase around 1995 for the two isomers, but this could also be done with one plot above the other. If the plot is to remain combined, I suggest plotting the different isomers on the same y-axis. Alternatively, if the different axis ranges are to be maintained then color the axis tick labels and text blue and green to correspond to the colors of the symbols and shaded areas of each isomer, as in Fig 8. You could also put 'i-C6F14' in blue text near the i-C6F14 curve, and similar for n-C6F14 (green text). I think would still prefer it split into 2 panels.

Fig 5 - as the i-C6F14 data only begin around 1987, should the figure only show the

red line and blue band from then? Otherwise it gives the impression that mixing ratio is known before 1987, but it isn't here.

Fig 7 - could color the right y-axis text and the EDGAR data as a color other than black, to emphasise that it is on the secondary axis.

Fig 9 - it is hard to distinguish the red and magenta dotted lines on my printed copy - I suggest using a more different color or line type.

Figures - it might be good to combine the mixing ratio and emissions figures for each PFC, for example combine Figs 3 and 7 as two panels in the same figure, Figs 4 and 8 etc. I found I wanted to look at the mixing ratio plot when I was reading about the emissions and looking at the emissions plot, and combining them would remove the need for flicking back and forth. The text could still remain in the same order, just describe the top (mixing ratio) panels first and come back to the lower (emissions) panels.

The measurements should be made available in a data archive or as downloadable tables in the Supplement.

Technical corrections:

page 12, line 18 - Hemisohere

page 12, line 19 - add 'is' after 'This'

page 16, line 26 - add 's' to 'emissions'

page 18, line 1 - with with

page 20, line 13 - exist, not exists

Supplement page 2, line 3 - Section ??

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