

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

### **Anonymous Referee #3**

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This manuscript presents measurements of the atmospheric mixing ratio of six perfluorocarbons (PFCs) obtained from a background site (Cape Grim) in SH for 1978-2018, and from two regional sites – in Tacolneston, UK for 2015-2017 and in Taiwan for four campaign periods between 2013 and 2016. It includes updates of previous measurements shown in Laube et al., (2012) by separating the isomers of n-C6F14 and i-C6F14 and improving the corresponding calibration scale. Authors also provide global emission estimates for these six PFCs in comparison with inventory-based and previous observation-based emissions estimates. This paper contains significant addition to current observation database and thus is worthy of publication in Atmospheric Chemistry and Physics after the following discussions are considered.

1. P6 LN 14-16: Since the update of the calibration scale was suggested as one of

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the key purposes for this article, the absolute calibration procedure needs to be over-viewed more in detail than in the current version, and thus readers don't have to search for and read through previous studies cited here.

2. P 7, LN 8: Clarify that the combined influence is <2.8%. Is it a relative difference between the n-C4F10 background concentrations determined based on the two different scales?

3. P 7, LN 14: What does the ion ratio means? Is it a ratio of peak heights for two chromatograms of  $m/z=219$  and 169? Or a ratio of peak areas?

4. P 7, LN 14: Fig. S2 was shown earlier than Fig. S1 in the text.

5. P 7, LN 25: the accuracy derived from the CFC-11 reference compound seems to imply only an accuracy of the dilution process, not "the accuracy of the calibrations" that stated in the text. Otherwise more clarification is needed.

6. P 7, LN 30: How was the uncertainty of 0.3% determined? Was it 1-sigma? Then how many data were analyzed ( $n=?$ )?

7. P 7, LN 31: How could the ranges (4.0-7.8%, and -5.5-2.8%) of calibration accuracy be determined? More detailed explanation should be given.

8. P 7, LN 34: Again, please describe how the overestimation by 20% and 11% could be determined?

9. P 8, LN 15-19: The emission distribution for all PFCs seems to be based on the global distribution of C6F14 emissions recorded in the EDGAR database. The figures 7-10 showed the individual EDGAR emission estimate for each PFCs. Then authors need to discuss about how different (or consistent) the emission modeling results would be if the EDGAR emission estimate for each compound were used, instead of the C6F14 emissions record.

10. P 9, LN 6-8: This statement conflicts with a previous comment in page 7 (lines 8-

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10): “Due to the leak-tightness of the system and a lack of observed isomers, a revision of the calibration scales for c-C4F8 and n-C5F12 was deemed unnecessary. If within the calibration uncertainty, their re-calibration was not necessary, why an additional uncertainty besides the calibration uncertainty should be added for these compounds?”

11. P 10, LN 6-11: Explain more in detail how a certain correlation between modeled CO and the PFC mixing ratio can represent which emission source is more associated with PFCs and what extent as well?

12. P 10, LN 28: Clarify where the number of 1.05 came from.

13. P 10, LN 29-32: Please compare the current c-C4F8 measurements in Tacolneston with those from Mace Head in Muhle et al. (2019) and discuss and justify the statement of “the lack of variability in the data suggests that Tacolneston is not in close proximity to any major sources of c-C4F8. When considering the simulated atmospheric trend from the emission model for Tacolneston, it can be noted that it overestimates the observed concentrations, despite the well-fitted Southern Hemispheric trend.”

14. P 11, LN 6-8: Are they based on the current simulation? Otherwise, add the correspond references.

15. P 11, LN 30-31: Please discuss how authors could exclude a possibility that the found similarity in the mixing ratio trends between n-C4F10 and n-C5F12 might be due to the fact that they were determined using a same m/z (119).

16. P 11, LN 18: Much lower mixing ratio elevations were observed where?

17. P 12, LN 33 – P 13, LN 2: Please explain how author can confirm the ratio shift from 2003-2008 to 2013-2018 periods was statistically significant. How was the uncertainty of +/-0.01 determined? Are there any possibility that the ratio shift could be related with the re-calibration after 2010?

18. P 13, LN 10-11: For the observed high mixing ratio, authors need to examine their trajectories to argue an influence of a pollution plume.

19. P 18, LN 1: remove the first “with”.

20. P 18, LN 10-17: Authors should describe much more in detail about the correlation analysis between PFCs mixing ratio versus CO mixing ratio derived CO source type, which was not given even in the Supplementary information. How can the CO mixing ratio be distinguished by the sources? The description and resulting discussion should be provided in the main text.

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