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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Distributions, long term trends and emissions of four perfluorocarbons in remote parts of the atmosphere and firn air" *by* J. C. Laube et al.

Anonymous Referee #2

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This is a nice study reporting the measurements of four longer chain (C4-C7) perfluorinated n-alkanes from a number of different sampling programs. The data tell a consistent story about increasing abundances of these chemicals over the past few decades related most likely to human-derived emissions, and I believe it will be of interest to readers of ACP. Comparisons are made to inventory-based emissions estimates and large differences are noted. I only have minor comments.

One concern relates to the section on deriving past atmospheric trends from stratospheric measurements of the chemicals (p. 4081-4082). The authors mention some limitations to this derivation (measurement precision is not as good), but they should

also indicate the limitations of the technique they have chosen. One concern is the suggestion that mean age is a concrete quantity with only small errors (implied from their use of a linear regression to derive rates), when in fact a stratospheric air parcel represents of air with a distribution of ages that are not well defined. The authors have rightly not presented their firn data as discrete points with dates for this reason. Because similar processes are at play in the stratosphere, I would think it also quite suspect to plot the stratospheric data as discrete points with distinct ages. I would think it important to mention these caveats and the simplifications incorporated in the analysis so as to not mislead the reader into thinking the results of the analysis of this stratospheric air are particularly robust. Given these issues, it seems inappropriate to use any consistency in rates to suggest that the stratospheric lifetimes of these chemicals are long; Instead, this seems reasonable and appropriate to be taken as an assumption in the analysis. It also seems incorrect to presume that similar growth rates will be derived in the SH and globe if averaged over an 8-year period (line 8, p 4082).

Figure 6, this latitudinal gradient for C5F12 during 27/10/2009 seems quite improbable. Did other predominantly NH-derived chemicals show this distribution, or could it represent a sampling/analysis issue?

p. 4082, lines 19-21, were other trace gases measured that could provide information about stratospheric influences on particular samples?

p. 4083, it could be mentioned that errors in lifetime cannot reconcile the differences between the inventory emissions and those derived from atmospheric data.

Figure 8. I would suggest that this figure be made into 2 panels showing the different compounds. I find it quite difficult to interpret as is.

It would add useful context to provide, for example, the current annual fossil-fuel derived CO2 emission for comparison to annual or total cumulative PFC emissions (last paragraph). It also seems a bit surprising that no mention of the radiative forcing contributed by these chemicals is made. This would be a useful and fairly easy addition.

Abstract: Line 12, the presence of a chemical in the stratosphere does not necessarily confirm that it has a “very long atmospheric lifetime”

Line 14-15, could be clearer, differ by five orders of magnitude among the measured compounds or between atmosphere- and EDGAR-derived values? It is the latter, of course, but this is clear only upon reader the paper.

Line 17, Be clear that you are using 100-yr Global Warming Potentials here, in the text, and in Figure 10 caption.

Line 19, Emissions and CO2-eq emissions are quoted with no indication of certainty even though the 100-yr GWP of one chemical isn’t known.

p. 4079, lines 16-20. how is it determined that the firn reconstruction through 2007 is OK but into 2008 is more uncertain? Is the point referring to seasonal influences on near-surface firn results?

p. 4080-81, lines 25-4. Are the authors suggesting that the errors in the reconstruction are actually larger than expressed by the dashed line in the figure? Some clarification is needed. It is also suggested that an inverse modeling approach gave similar results though they aren’t discussed at all; were they within the uncertainties presented in the figure?

I find that the supplemental information contains useful and worthwhile information that adds to the paper. It might be worth mentioning the degree to which an isotopic impurities would affect the inventory vs atmosphere-based emissions figures (very negligibly, I presume). Is there a reason point instead of lines are used to express growth rates from fits in Figure S3?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 4073, 2012.