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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Perfluorocarbons in the global atmosphere: tetrafluoromethane, hexafluoroethane, and octafluoropropane" *by* J. Mühle et al.

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Please note that we provide our replies in bold after each comment from Anonymous Referee 2 (received and published: 18 April 2010).

The paper presents measurement of very long-lived perfluorocarbons in the atmosphere and estimated annual-mean global total emissions using simple inversion methodology. This work is a genuine advancement in terms of measuring capability. Firstly, they are able to make direct measurements dating back to the 1970s, and more remarkable is the unprecendented measurement precision and accuracy achieved. Enjoyed reading the paper, particularly the observational part. The paper

can be published as it is. I only have some simple suggestions, which the authors may like to consider during the revision.

We thank the referee for the very positive evaluation of our analytical and modeling work.

P.6497, the para of L5: I do not think, this statement is still valid "2-D model provides similar results to that of 3-D models". Doesn't matter species of what lifetime is simulated. How does the 2-D models account for synoptic variations or dynamics of shorter timescales, which can influence site level concentrations between hours or days. I am sure dynamics at these time scales are important for many of the sites discussed here. There is also issues with stratosphere-troposphere exchange and interhemispheric transport and their interannual variability.

This in turn will also effect your source inversion results, and lead to over statements like the one in P.6506, the para of L15. Do you really believe that EDGAR emission distribution can be questioned following the inversion methodology presented here?

The used 2-D model has been used successfully in several previous studies to simulate the zonally averaged behavior of gases with lifetimes that are long compared to the interhemispheric exchange time and that show no sharp emissions changes. We have modified the statement about the used model accordingly. We filter out pollution events with the goal to remove synoptical variability and target background air and further reduce effects from synoptical variability by using monthly mean values for the inversion. Compared to the global estimates, semi-hemispheric emissions may be more sensitive to model transport parameter uncertainties and inter-annual transport variations, but the influence of these limitations are accounted for in the uncertainties of the derived emissions by including model parameters uncertainties in the error analysis. We have added this statement to the revised paper. As explained in the paper, we achieved significant error reductions of the emissions during the inversion in all model boxes

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when in situ high-frequency data were available (2006-2009) and in the 30-90°N box for all years. From 2006-2009 the semi-hemispheric emissions for all PFCs were clearly uncorrelated (R^2<0.1). Given that the EDGAR database does not provide all details necessary to understand how the PFC emission estimates are calculated or apportioned, we can only speculate that our measurement derived semi-hemispheric distribution could actually be more accurate.

At the end I congratulate the AGAGE team for being able to produce such a wonderful datasets, and research that will help the atmospheric science community in general.

We thank the referee for the very positive evaluation of our work.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 6485, 2010.