

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

J. Mühle et al.

jmuhle@ucsd.edu

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Please note that we provide our replies in bold after each comment from C. Bayliss (received and published: 12 April 2010).

Thank you very much for the opportunity to comment on this important paper, which clearly demonstrates, through advanced measuring and modelling methods, a gap between atmospheric concentrations of CF_4 , C_2F_6 and C_3F_8 and available historical national and sectoral bottom-up emission inventories.

We thank the commentator for the positive assessment of our analytical and modeling work.

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However, the variability in bottom-up data quantity and quality and lack of clarity in inventory methodologies does not allow for sources of the “missing emissions” to be identified. The authors’ discussion of such sources is out of place in this paper – given that the discussion detracts from the quality of science behind the atmospheric concentration and emission trend data - and their conclusions conjectural and without basis.

p6489, lines 10-12 Uncertainties in the magnitude and temporal evolution of emission factors from non-aluminium industry sources is a limiting factor of greater importance than uncertainties in the aluminium sector data. The methodology employed by the IAI anode effect survey to develop global annual sector wide emission factors is transparent and consistent (ref.). No such bottom up methodology exists for semiconductors/electronics and extrapolation of EDGAR data (a hybrid of bottom up and top down sources – p6506, line 5) for these industries will realise emission factors as indicative of global electronics/semiconductor industry emissions as using EDGAR data for aluminium industry emissions (i.e. uncertain, under estimated and tautological).

Therefore, in the absence of a comparable methodology for calculating non-aluminium industry emissions bottom up, the task of estimating contributions from sectors is currently impossible. A highly uncertain route, though one which would be more certain than that employing linear extrapolation of EDGAR hybrid data would be to subtract the bottom up aluminium industry data from atmospheric measurements to develop estimates of emissions from other (non-reporting) industries. I could understand why the authors would not want to do this, though I cannot see why they might follow the converse route of subtracting extrapolated EDGAR based (uncertain) electronics data, plus annual IAI (more certain) anode effect and PFC measurement data, from atmospheric measurements and apportioning the remaining emissions to unreported aluminium industry sources.

Ergo, there is no place in this document for apportioning emissions by sector.

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We understand the sensitive nature of source apportioning of global perfluorocarbon (PFC) emissions and acknowledge the continuing, long-term effort of the International Aluminium Institute (IAI) to identify and reduce PFC emissions. We now also point out that the report of the World Semiconductor Council (WSC) on the voluntary perfluorocompound emission reduction program of the semiconductor manufacturers contains only very limited information. The discussions in the manuscript are valuable as they demonstrate that the sum of available PFC bottom-up emission estimates from the aluminum and semiconductor/electronics industries is significantly lower than global emissions inferred from our atmospheric measurements, and that this emission gap has been increasing. We point out in the revised manuscript that the missing CF₄ emissions likely stem from the primary Aluminum production *and/or* the semiconductor/electronics industries. We have made changes to the revised manuscript to point out the shortcomings of the various inventories more clearly and to avoid any bias in the discussion.

p6507, lines 4-8 China has only been PFPB-only producer since the mid 2000s. It should be made clear that the average China emission factor (median) derived from measurement data can only be applied to the industry as a whole since China became a 100

We assume that the commentator intended to say "... became a 100% PFPB-based Al producer". We have followed the information given by J. Marks and have corrected our calculations accordingly. All numbers and figures in the text have been updated. The changes are small and the conclusions remain unchanged.

p6507, lines 18-30, p6508, lines 1-5 The order in which potential causes of error in CF₄ emissions are discussed does not reflect their relative probability, but rather a value judgement of the authors. "Indications" of technology upgrades in the semiconductor/electronics industry should not be given more credibility than published mea-

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surement data from Chinese smelters and transparent, consistently reported annual, bottom up survey data. Nor should the linear extrapolation of methodologically "unclear" EDGAR data be rated more credible and accurate than IPCC methodologies that are subject to a regular review process.

We have rewritten the section to point out that the emission gap *could* be due to fundamental problems with the IPCC methodologies *and/or* due to underestimation of PFC emissions from semiconductor/electronics manufacture in EDGAR v4. We also point out that the report of the World Semiconductor Counsel (WSC) on the voluntary perfluorocompound emission reduction program of the semiconductor manufacturers contains only very limited information. We also stress that the EDGAR database does not provide all details necessary to understand how the CF₄ and C₂F₆ emission estimates are calculated or apportioned among source categories.

p6507, line 30 Please expand on potential mis-application of IPCC methodologies by IAI, or delete.

Recently significant PFC emissions during the startup of reduction cells were found which may not be accounted for by IPCC methodology (Maltais, J.-N., Ross, J., Marcoux, A., and Gaudreault, G.: Application of a Method for the Determination of PFC Emissions during Aluminum Pot Start-up, in: Light Metals 2010, edited by: Johnson, J. A., 2010.). We have added this information and clarified the section.

p6508, lines 11-15 IAI Anode Effect Survey reported data has almost 100

We assume that the commentator meant to say that IAI Anode Effect Survey reported data "has almost 100% coverage". However, according to the 2008 Anode Effect survey, the world wide coverage is only 64%, mostly due to the lack of data from China.

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Independently of this, we find it is surprising that UNFCCC Annex I CF₄ emissions from AI and non-AI production agree so well with global AI production related CF₄ emissions estimated from the IAI reports (Annex I and non-Annex I AI production). This would require Annex I non-AI based CF₄ emissions to be large and comparable to non-Annex I AI based CF₄ emissions. We have clarified the section accordingly.

P6511, lines 2-6 I don't believe the authors can go further than this statement, other than to point out the relative certainty in the databases and therefore where one might look for the "missing" data/emissions. There is an "inherent underestimation" in the bottom up inventory of emissions sources, but why this should be apportioned to the IAI anode effect survey (the global aluminium industry) and not to other sources is not clear.

We have rewritten the section and now point out that the emission gap *could* indicate an inherent underestimation of CF₄ emissions by the IAI Anode Effect survey *and/or* that semiconductor/electronics manufacture emissions in EDGAR v4 are underestimated. As pointed above, recently Maltais et al. (2010) discovered significant PFC emissions during the startup of reduction cells which may not be accounted for by IPCC methodology.

P6514, lines 20-24 There is a missing element in the list of emission estimates (UNFCCC, EDGAR, IAI) and that is the bottom up estimation of semiconductors/electronics. The fact that such an estimation/methodology does not exist does not exclude it from scrutiny.

We now point out that the report of the World Semiconductor Council (WSC) on the voluntary perfluorocompound emissions reduction program of semiconductor manufacturers contains only very limited information. We also state that a combined effort by the IAI, WSC, global PFC suppliers, and EDGAR to improve estimate of CF₄ and C₂F₆ emissions and emission factors would be very valuable

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to find the source of the missing emissions.

If we look at (relative) availability of data and transparency of methodology it is clear that there are other places to look than the IAI's database for "missing emissions" (see Fig 1).

See above.

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

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