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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-lived halocarbon trends and budgets from atmospheric chemistry modelling constrained with measurements in polar firn" *by* P. Martinerie et al.

Anonymous Referee #2

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Review of Martinerie et al "Long lived halocarbon trends and budgets from atmospheric chemistry modelling constrained with measurements in polar firn" ACPD 2009

This comprehensive work combines models of firn air processes and atmospheric chemistry and transport, together with measurements of firn air concentrations from both polar regions, to understand the atmospheric emission histories of several key synthetic compounds. The complexity but elegance of the task is best explained in figure 1. This is perhaps the most complete way to test atmospheric chemical models and emissions estimates for the past century. Testing of emissions scenarios and atmospheric chemical model concentrations by comparing firn measurements with modelled firn profiles has been done before for several CFCs (Sturrock et al., 2002). This study tests many emissions-sink scenarios through model derivations of atmospheric trends against vertical profiles in the atmosphere and firn profiles including those

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Interactive Discussion
Discussion Paper

of Sturrock et al. The authors bring expertise in each of the steps.

The paper is suitable for this journal and should be published with the following matters addressed.

The possibilities of natural production of CCl4 and SF6 are intriguing questions that have tried to be answered by previous firn air studies. It is not clear whether this work has come closer to an answer. While the mean ages of the deepest firn samples analysed here are quite old, their age spread is relatively wide, apparently compromising their ability to quantify pre-anthropogenic levels. More could be said about the maximum background levels of these compounds (and the CFCs) that can be constrained by this work (ends of sections 6.2.1, 6.2.3 and 6.2.4 and in the conclusions).

The comparison with vertical atmospheric measurements doesn't seem to be a large constraint to the budget determinations. More could be said about this.

Estimates of the uncertainty ranges of the atmospheric concentration records derived from the firn profiles should be made more prominent, in Figure 8 for example. It would be interesting to compare the uncertainties from their combination of several firn sites with the single but narrow air age spread site of Sturrock et al., especially for periods of rapid change and for the oldest (possibly zero) concentrations. It is notable that the quite different scenarios, for CFC 114 especially (fig 8), are not significantly different when viewed in the firn model comparisons with measurements in fig 10. Does this suggest that the sensitivity of these firn sites and their diffusion models in testing atmospheric scenarios is rather low? An exception might be for the deeper (lock in zone) section of Dome C, where CFC 114 seems to agree with the "late start" result from Sturrock, but the gradients of the curves here are very steep and the differences are both large and difficult to quantify in the figure (the figure could be redrawn, or reproduced larger?)

More minor suggestions and points follow:

Abstract
line 17. Explain further what is meant by "insignificant": zero, below detection...?
Introduction
The wording of the sentence at line 15 is confusing.
The concept of the timescale of trace gas diffusion in firn is introduced at line 7. I think it is

ACPD
9, S575–S580, 2009

important to clarify the processes at work in firn, for example, to distinguish between the speed of trace gas diffusion and the trace gas flux. The latter is the main process that transforms signals in the atmosphere into vertical profiles in the firn. As well as the concentration gradient and temperature as mentioned in the text, the flux depends on the diffusivity of the gas in the firn (the product of its diffusivity in air and a factor that takes into account the tortuosity of the firn). The main effect of the snow accumulation rate is to move the firn downwards, rather than to directly influence the diffusion of air within it. The description of firn processes and how they are modelled, in section 2.2, seems to under emphasise the importance of the lowest part of the firn where diffusion is essentially zero yet pores are still open- often referred to as the lock in zone- where air moves down slowly with the firn and most of the atmospheric record can be found. How accurate are the molecular diffusion coefficients (p1000, line29)?
P1001. NOAA CMDL is now called NOAA ESRL (Earth System Research Laboratory).

Section 3.2
Line 21. Firn data of Battle et al 1996 and newer ice core data of MacFarling Meure et al 2006 would provide a more accurate update on the Machida ice core record for the atmospheric N2O input.

Section 5.2.
There are a number of clarifications required in this section. P 1005 Line 1: which “results”- the simulated vertical profiles? Please be specific.
Line 6: There seems to be a circular argument here- the CFCs atmospheric records are to be found from the firn concentration profiles. How can they also be used to constrain the firn diffusivity profiles, at least beyond the earliest atmospheric CFC measurements?
This is also confusing because it is also said in the introduction that “typically” CO2 and CH4 records have been used to constrain the firn model. Then in Section 2.2 (the firn model) says that a trace gas with a “well known atmospheric trend” is used and Table 3 presents scenario ages for CO2 and CH4. It needs to be made clearer in the main text -as well as the supplement- what trace gases were used to establish the firn diffusivity profiles. This will require mention of the analysis technique for the firn air concentrations and reference to the atmospheric records used to compare with.
If CO2 and CH4 from the Law Dome ice and firn records are used to constrain the firn diffusivities for the early part of the record, the CO2 concentration flattening in the mid 1900s might lead to problems. Is this why it was found (mentioned in the supplement) that CH4 has a better

ACPD
9, S575–S580, 2009

match?
Line 8: for firn model validation?
Line 9: experimental data- meaning measured concentration-depth profiles?
Line 22: were melt layers actually observed at this depth? At 27 m porous firn would be expected and shouldn't melt be obvious?
Line 25. Here and elsewhere, "experimental" might be better replaced with "measured" or "observed".
P 1006 Line 2. the atmospheric concentration scenario?
Line 7. The gentle slopes at small depths...
Line 8. Steeper slopes
Line 27. the particular trace gas...
P 1007 Line 7. the firn ages do not always increase with decreasing diffusivity Line 11. Why should the gases of interest here have similar mean and scenario ages?
P 1008 Line 3 globally? Perhaps "generally"?
Line 15. It is unclear what makes DI the "best" site (what does "the regularity of the depth-age profile mean?)
Line 16. Wouldn't impermeable melt layers reduce the air age spread?
Line 22. Time of drilling
Line 26. ...of widening for sites with lower snow accumulation rates...
P1010 Line 25 there are no...
Line 25 delete "trend"
Line 25 which is the "studied" period?
Line 27 which recent data- for what years?
P1011 Line 28 model-data differences
P1012 line 2 slope change (it is continuous)
End of section 6.2.2 I couldn't follow the reasoning behind the lowest CFC 113 concentration argument.
P1014 line 2 and later: abundance is calculated from concentration to determine lifetime calculations
Line 2 loss rate
Line 3 spelling of proportional
Line 28 and p1015 line 2 and elsewhere: "region" is probably better than area or places
P1015 line 5: a homogeneous

ACPD

9, S575–S580, 2009

Interactive Comment

Line 18 what is meant by “normally”?
P 1016 line 23. delete “time”.
P 1017 line 17. +140Line 26. the emissions are equivalent to 48P 1018 line 8: globally, or generally?
Table 5 is a nice summary of the uncertainties in the budget from the main steps of the analysis. Together with the top of P 1019 it also summarises almost the entire cumulative trace gas budget imbalances in comparison with the recent, post Montreal, situation.
P1019 line 25: ...are also shown... It would be useful to say how the total radiative forcing of these compounds compares to the total for long lived greenhouse gases.
P1021 line 23. “...slope reduction in the SF6 trend”- emission or concentration?
P1022 line 15. ...budget histories...
Line 21. Insufficiently...
Is the last sentence in the conclusions rather obvious for any compound that exists in the atmosphere?

Specific responses to the review criteria follow:
1) Does the paper address relevant scientific questions within the scope of ACP? Yes
2) Does the paper present novel concepts, ideas, tools, or data? Yes
3) Are substantial conclusions reached? Yes, though possibly more on the natural sources might be nice
4) Are the scientific methods and assumptions valid and clearly outlined? Yes, with some changes required
5) Are the results sufficient to support the interpretations and conclusions? Yes
6) Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? Mostly- some extra required as identified above
7) Do the authors give proper credit to related work and clearly indicate their own new/original contribution? yes
8) Does the title clearly reflect the contents of the paper? yes
9) Does the abstract provide a concise and complete summary? yes
10) Is the overall presentation well structured and clear? Mostly-some changes suggested
11) Is the language fluent and precise? Needs some improvement and clarity where noted
12) Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

ACPD
9, S575–S580, 2009

Interactive Comment

Hardly used
13) Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? Some reorganisation between text and supplement could reduce and clarify the firn air age and diffusivity discussions
14) Are the number and quality of references appropriate? yes
15) Is the amount and quality of supplementary material appropriate? yes

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 991, 2009.

ACPD

9, S575–S580, 2009

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