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Interactive comment on "20th Century trends and budget implications of trihalomethanes and dihalomethanes inferred from North GRIP firn air" *by* D. R. Worton et al.

Anonymous Referee #2

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General comments

The paper presents new firn air data and gives new insights about the anthropogenic emissions from trihalomethanes and dihalomethanes, with special emphasis on the chloroform budget. It is well written and clearly organized. After a short review about the role of halogens in the atmosphere, the authors show data from firn air measurements obtained in different polar sites. The very good agreement obtained for the two Arctic and the two Antarctic sites gives good confidence in the data. Section 3.2., the longest of the paper, is dedicated to atmospheric modelling, the main conclusion being

that chloroform anthropogenic emissions are significantly underestimated. By using different emission scenarii, the authors conclude that paper manufacturing, the main anthropogenic source of chloroform, is likely underestimated by a factor of 3-4 and that the overall anthropogenic contribution of the chloroform total budget could have been as high as 40% in 1990. This is a new and important result about the chloroform budget. Conclusions are also driven about the anthropogenic budget of the studied brominated trihalomethanes. Although clearly written, there are still some points which may be confusing and which need to be checked carefully (see comments below).

Specific comments

- The title of the paper could contain the name “chloroform” as most of the discussion is dedicated to this compound.

- Section 2.1. is too short. It only mentions the firn air samples which were collected at NGRIP. It should also give information about sampling at the other sites (DI, Dome C and DML). At that point, it should be clearly stated (i) which measurements were performed where (chloroform at all sites, brominated compounds only at NGRIP) and (ii) which of these measurements are new data (then giving the corresponding references for the data already published). Acronyms for the different sites should also be defined here. Uncertainties associated with the measurements should also be mentioned (or discussed) in this section. On Figure 1, error bars are plotted only for the northern hemisphere measurements, that point should be explained.

- Section 3.2.2: “the CH3Cl anthropogenic emissions are far better constrained relative to natural emissions”. This seems to be contradictory with the conclusion of this paper which claims that anthropogenic emissions are largely underestimated.

- Section 3.2.3. Uncertainties in the estimated emission factor should be discussed in detail, either in this section or in the section 3.2.7, when it is concluded that the emission factor is likely inaccurate.

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- Lots of assumptions, inherent to this kind of study, are made and discussed. However, a very important assumption, i.e. that chloroform natural sources are known and constant, would deserve more discussion. The authors have made a scenario about changing soil surface emission (DSS), however this scenario should also be compared with the scenarii involving anthropogenic changes (figures 9 and 10). Natural source emissions have been divided by 2, based on different calibration scales (section 3.2.1). If these natural emissions were higher than assumed, how would it change the conclusions of the paper? This point must be addressed in the paper.

- Section 3.2.5: Discussion of the scenarii involving constant emissions of WC and OI during the period 1990-2002. Firstly, it is a bit too much of a coincidence that these emission trends would change in 1990, exactly the same year as for the paper manufacture. Secondly, I'm not convinced that this hypothesis brings some conclusive result to the discussion. Therefore I would suggest removing this scenario from figures 8 and 9 and from the discussion.

- Section 3.2.6.: China is responsible for $<$10% of global pulp and paper production. What about Russia?

- Section 3.2.7.: It is not clear why the DSS scenario (which represents an anthropogenic perturbation of the soil emission, therefore affecting mostly the northern hemisphere) would improve the SH simulation.

Technical comments

- P. 708, L. 28: Check the reference of the section.

- P. 309, L. 19, 20, 21: Check all references from Khalil (for example, references from 1998, 1983 are Khalil and Rsamussen and not Khalil et al.)

- P. 715, L. 12: In the paper, scenarii using a factor of 2 for WC and OI and a factor of 5 for PP emissions are used. Here, the numbers are a factor of 2 for PP and 5 for WC and OI. Please clarify.

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- P. 715, L1 6 to L 21: If possible, shorten this sentence.

- P. 719, L 24: Delete “the” (“the these”).

- P. 721, L 23: Delete “the” before “whether”.

-Table 1: Check consistency between the table and its legend (2001/ 2002).

- All Figures (with depth X-axis or CFC12 x-axis): It would be useful to give some date indication on these figures (at least, the year 1990).

- Figure 6: Give the full names of ECF and TCF in the text legend.

- Figure 8: DSS should be defined in the text legend. Should it be (oceans +DSS)/ 5 instead of oceans DSS/5 ?

- Figures 9,13, 14: Reverse the X-scale (depth)?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 701, 2006.