

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 #1

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General Comments

Firn air profile concentration data for six halomethanes are presented. Individual species show different concentration changes with depth. These profiles are used to infer changes in past atmospheric concentrations and of occurred changes in possible sources and sinks of these gases. The authors present a nice data set. The analytical quality appears to be very good. The vertical resolution of the profiles is dense enough to give a fine resolution of the temporal changes of the studied species. The comparison of data from four sites gives further confidence in the representativeness

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of these measurements. Observed differences in concentration profiles in the ice cores originating from the Arctic and Antarctica allow deciphering hemispheric differences in sources and sinks.

Next, these observations are used in conjunction with a firn air transport and atmospheric model to develop estimates of the changes in atmospheric sources and sinks, in particular of CHCl₃. The authors undertake a remarkable effort in trying to define contributions from different emission sources. Developing these estimates relies on a plethora of published data and many assumptions that build on these previous, in itself somewhat uncertain data and conclusions. I am somewhat torn between applauding the thoroughness of this effort and doubting the feasibility of this task. Given the large uncertainties of many of the individual contributions, I have doubts that several of the conclusions drawn in this paper are robust and justified.

I would like to propose developing a statistical (as good as possible with available information) error estimate of contributing sources and sink terms. This should include uncertainty estimates of all steps and data that go into the analysis procedure, e.g. firn air sampling procedure, chemical analysis error, firn air transport uncertainties, atmospheric model, atmospheric oxidation sink (including its seasonality) and its potential change over the study period, other contributing sinks and of all considered natural and anthropogenic sources. Next, a statistical error propagation calculation should be performed and from that it could be concluded whether or not the observations indeed disagree (at the statistically significant confidence level) with what the model calculation predicts. If this can be proven in a convincing manner, than I would agree that the (lengthy) speculations enumerated on in chapters 3.2.5 - 3.3 are justified.

Specific and Technical Comments

Title: Since data from four ice cores are presented and discussed, I suggest changing 'from North GRIP firn air' to 'from four firn air profiles'

Page 703/Line 8 - 12: Please explain this statement in more detail or remove it all

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together.

704/21-23: I believe that atmospheric concentrations of several halons are still on the rise, so I would suggest revising this sentence accordingly as in the way it is written now it infers that there are no increases of atmospheric bromine-containing trace gases.

706/18-20 and Fig. 1: I agree that plotting THMs versus CFC-12 is a convincing way of illustration, but it would still be helpful to have an approximate time scale, possibly as a second x-axis.

709/4: Please explain what is meant by 'gravitational settling'.

710/8: Please explain 'RCEI'.

712/25: Instead of '1:1', I suggest to say ' the relationship between ought to be linear and in ...'

720/11: Provide a reference for the given average global concentration (10.2 pptv).

723/10: Be more specific than 'bottom of the firn'.

725/18: ' that show excellent '

Table 1: I suggest listing the references for these data in the table.

Figures 1,2: How were the error bars calculated? Why are they shown on some data series, but missing on others?

Figure 2: For better comparison, it would be nice to have CHCl3 plotted in the same format as for all the other gases (together with the constant emission/seasonally varying sink line).

Figure 4: Use different line patterns so that the ND1 and ND2 lines can be distinguished in B&W format. Show that boxes represent latitude gradient. Can uncertainty ranges be added?

Figure 5: Explain 'FAOSTAT' in figure caption.

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Figure 6: Explain 'ECF' and 'TCF' in figure caption.

Figure 8, 10, 11, 12, 13, 14: Again, please explain all shown abbreviated data series in the figure caption and use line formats that can be distinguished in B&W.

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