Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 6, 2847-2863, 2006
www.atmos-chem-phys.net/6/2847/2006/

20th century trends and budget implications of chloroform and related tri-and dihalomethanes inferred from firn air

D. R. Worton[1], W. T. Sturges[1], J. Schwander[2], R. Mulvaney[3], J.-M. Barnola[4], and J. Chappellaz[4]
[1]School of Environmental Sciences, University of East Anglia, Norwich, UK
[2]Physics Institute, University of Berne, Berne, Switzerland
[3]British Antarctic Survey, Natural Environment Research Council, Cambridge, UK
[4]CNRS Laboratoire de Glaciologie et Geophysique de l'Environnement, Saint Martin d'Heres, France

Abstract. Four trihalomethane (THM; $CHCl_3$, $CHBrCl_2$, $CHBr_2Cl$ and $CHBr_3$) and two dihalomethane (DHM; CH_2BrCl and CH_2Br_2) trace gases have been measured in air extracted from polar firn collected at the North Greenland Icecore Project (NGRIP) site. $CHCl_3$ was also measured in firn air from Devon Island (DI), Canada, Dronning Maud Land (DML), Antarctica and Dome Concordia (Dome C), Antarctica. All of these species are believed to be almost entirely of natural origin except for $CHCl_3$ where anthropogenic sources have been reported to contribute ~10% to the global burden. A 2-D atmospheric model was run for $CHCl_3$ using reported emission estimates to produce historical atmospheric trends at the firn sites, which were then input into a firn diffusion model to produce concentration depth profiles that were compared against the measurements. The anthropogenic emissions were modified in order to give the best model fit to the firn data at NGRIP, Dome C and DML. As a result, the contribution of $CHCl_3$ from anthropogenic sources, mainly from pulp and paper manufacture, to the total chloroform budget appears to have been considerably underestimated and was likely to have been close to ~50% at the maximum in atmospheric $CHCl_3$ concentrations around 1990, declining to ~29% at the beginning of the 21st century. We also show that the atmospheric burden of the brominated THM's in the Northern Hemisphere have increased over the 20th century while CH_2Br_2 has remained constant over time implying that it is entirely of natural origin.

Final Revised Paper (PDF, 566 KB) Discussion Paper (ACPD)

Citation: Worton, D. R., Sturges, W. T., Schwander, J., Mulvaney, R., Barnola, J.-M., and Chappellaz, J.: 20th century trends and budget implications of chloroform and related tri-and dihalomethanes inferred from firn air, Atmos. Chem. Phys., 6, 2847-2863, 2006. Bibtex EndNote Reference Manager