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Halocarbon trends and budgets over the last century : a model study constrained by firn air data

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The concentrations of 18 halocarbons in polar firn air were analysed using a model of gas diffusion in the firn and a 2D model of tropospheric and stratospheric chemistry run on the century time scale. Model results were validated by comparison with atmospheric data sets: surface trends, total column, and balloon-based measurements. For most species, the predicted trends are consistent with observed trends within the known uncertainty limits.

Halocarbon emission data, when available, were used to constrain the chemistry model. The predicted atmospheric concentration trends were used as input to a model of gas diffusion in interstitial air of polar firn, in order to be compared with Arctic and Antarctic data obtained within the FIRETRACC and CRYOSTAT EC projects. The uncertainties on halocarbon budgets vary largely from one species to another, and we discuss the impact of different factors such as emissions, lifetime, measurement calibration, and diffusivity in polar firn.

Firn measurements provide information on atmospheric trends over longer periods than the atmospheric records. Our results confirm that the natural sources of major CFCs, HCFCs, CCl4 and SF6 are unsignificant. For some compounds, this work produced the first estimate of historical atmospheric concentration trend and/or historical emissions. Finally, the impact of these changes on stratospheric temperature and ozone (through homogeneous chemistry) is estimated.