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Top-down emission estimation of European sources of halogenated hydrocarbons using a Kalman-filter based inversion

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Halogenated hydrocarbons in the atmosphere are mostly synthetic products of the chemical industry designed for a wide range of applications. The first generation of compounds, the bromine- and chlorine-containing halons and chlorofluorocarbons (CFCs), were shown to be harmful to the stratospheric ozone layer. This motivated the international community to initiate the Montreal Protocol in 1987 to phase out their production globally. In the industrialized countries CFCs were consequently replaced by the shorter-lived hydrochlorofluorocarbons (HCFCs) during the 1990s and thereafter by the completely chlorine-free HFCs. Although not harmful to the ozone layer anymore, some of the HFCs are potent greenhouse gases and are therefore regulated under the Kyoto Protocol.

The high-alpine station Jungfraujoch and the coastal station Mace Head are two of only four sites of the European SOGE network (System for Observation of Halogenated Greenhouse Gases in Europe) with high-frequency measurements of halogenated compounds. Based on observations at these two sites, we here present a combined measurement – model analysis of the distribution of European emissions for a selection of compounds, and trace their evolution with time since measurements started in 2000. For the spatial allocation of sources, the measurements were combined with detailed transport simulations.

For a qualitative allocation of sources in Europe we employed the trajectory statistics method of Seibert et al. (1994) and Stohl (1996). For Mace Head trajectories were computed with the FLEXPART model driven by ECMWF analyzed winds at 1°x1° resolution. For the station Jungfraujoch, however, we used the model COSMO-TRAJ driven by high-resolution wind fields (7 km x 7 km) of the weather forecast model COSMO of MeteoSwiss in order to better represent the transport in complex topography over the Alps. The method allows identifying the major source regions of the different compounds in Western and Central Europe. The pesticide methyl bromide (CH3Br), for example, was applied primarily in southern Europe to protect vegetable and strawberry plantations. Its production was banned by the Montreal Protocol which is reflected by a strong reduction in emissions between 2003 and 2008 as seen from Jungfraujoch. A contrasting example is the cooling agent HFC-125 belonging to the second generation of replacement compounds not regulated under the Montreal Protocol. During the same period, HFC-125 exhibited a marked increase with sources more homogeneously spread over Europe than those of CH3Br.

For a more quantitative analysis for the years 2007-2009, we applied the Lagrangian Particle Dispersion Model FLEXPART using meteorological input data of the IFS model of ECMWF at 0.2° x 0.2° resolution, together with a new source inversion method based on sequential Kalman filtering. Different from other approaches the method is essentially independent of an a-priori and adjusts both the emission field and the trace gas background levels in an iterative fashion. In this study, we will contrast results of the trajectory statistics method with the more advanced source inversion, address uncertainties in the methods, and show the evolution of European emissions of a selection of compounds in comparison to official numbers reported by the individual countries to the Montreal and Kyoto protocols, respectively.