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Inverse Modelling to Constrain Global Emissions of Short-Lived Chlorocarbons

Tom Claxton (1), Ryan Hossaini (1), Martyn Chipperfield (2), Chris Wilson (2), and Stephen Montzka (3)

(1) Lancaster Environment Centre, Lancaster University, Lancaster, United Kingdom, (2) School of Earth and Environment, University of Leeds, Leeds, United Kingdom, (3) National Oceanic and Atmospheric Administration, Boulder, Colorado, USA

Halogens play an important role in atmospheric chemistry in both the troposphere and stratosphere. In the stratosphere, it is well established that halogenated source gases (e.g. chlorofluorocarbons, CFCs, halons etc.) have caused widespread depletion of the ozone layer in recent decades. The Montreal Protocol was implemented to prohibit production of these long-lived chemicals and has been extremely successful to date. However, so-called Very Short-Lived Substances (VSLS) are also a significant source of atmospheric bromine, and an emerging source of chlorine not controlled by the Montreal Protocol. Due to short lifetimes (less than $\sim$6 months), VSLS release reactive halogens in both the troposphere and stratosphere, and thereby influence composition in both regions.

Major chlorinated VSLS include dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$) and perchloroethylene (C_2Cl_4) and have mixed sources. For example, $CHCl_3$ is emitted naturally from the ocean, while anthropogenic emissions from industrial processes are thought to be most important for CH_2Cl_2 and C_2Cl_4 (e.g. through their use as solvents). At present, the emission sources and strengths of these compounds are poorly quantified, and their effects on tropospheric and stratospheric composition are uncertain.

The TOMCAT 3-D chemical transport model (CTM) was used to produce an optimised 1˚x1˚ global emission inventory for CH_2Cl_2, $CHCl_3$ and C_2Cl_4 in 2014, through a synthesis inversion modelling approach. Prior emission estimates for these compounds were adapted from the Reactive Chlorine Emissions Inventory (RCEI, Keene et al., 1999) and a variety of observations have been used in the inversion calculation, including (i) global surface observations from the NOAA network, and (ii) recent aircraft observations obtained over SE Asia (CAST and CONTRAST campaigns).

Relative to the 1990s prior, results from the inversion show a large increase in present-day emissions of CH_2Cl_2 over Asia, in contrast to decreases over Europe and North America. For example, CH_2Cl_2 emissions from temperate Asia are estimated to have increased from 0.14 Tg CH_2Cl_2/year in the late 1990s to 0.46 Tg CH_2Cl_2/year in 2014. We estimate a present-day global CH_2Cl_2 source of $\sim$1 Tg CH_2Cl_2/year, with $\sim$60% of this total due to Asian emissions.

Using the posterior emissions in a forward TOMCAT simulation, the modelled CH_2Cl_2 (and $CHCl_3$ and C_2Cl_4) mixing ratios agree well with a range of recent observational data, including independent observations (i.e. not used in the inversion itself) throughout the vertical extent of the troposphere. However, the strength of oceanic emissions remains uncertain, particularly for CH_2Cl_2, with our results showing a net ocean source at low latitudes, and a net ocean sink at high latitudes. The implications for this additional VSLS chlorine for changes in tropospheric composition (e.g. impact on Cl atom concentrations) will be briefly discussed.