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Global emissions of short-lived ozone-depleting gases: Quantifying regional variability and trends using long-term measurements and atmospheric modelling

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

(1) Lancaster University, Lancaster Environment Centre, Lancaster, United Kingdom (t.claxton@lancaster.ac.uk), (2) School of Earth and Environment, University of Leeds, Leeds, United Kingdom, (3) National Oceanic and Atmospheric Administration, Boulder, Colorado, United States

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 halocarbons and has been extremely successful to date. However, so-called Very Short-Lived Substances (VSLS), such as dichloromethane (CH2Cl2) and chloroform (CHCl3), are an emerging issue as a potentially significant source of atmospheric chlorine, not controlled by the Montreal Protocol. Due to short lifetimes (less than ∼6 months), VSLS release reactive halogens in both the troposphere and stratosphere, and thereby influence composition in both regions. At present, the emission sources and strengths of these anthropogenic compounds are poorly quantified, and their effects on atmospheric composition (e.g. ozone) are uncertain. Here, we consider long-term (2007-2016) CH2Cl2 and C2Cl4 observations from the global NOAA surface monitoring network. Combined with simulations of the TOMCAT 3-D chemical transport model, we use these data to derive an optimised 1°x1° global emission inventory for CH2Cl2 and C2Cl4, over a 10-year period (2007-2016), through a 'synthesis inversion' approach. Prior emission estimates for both compounds were updated from the Reactive Chlorine Emissions Inventory (RCEI, Keene et al., 1999).

Results from the inversion show a large increase in present-day emissions of CH2Cl2 over Asia, in contrast to decreases over Europe and North America, relative to the 1990s prior. For example, Asian CH2Cl2 emissions are estimated to have increased from 0.13 Tg CH2Cl2/year in the late 1990s to 0.61-0.69 Tg CH2Cl2/year in 2016. We estimate a present-day global CH2Cl2 source of ∼1 Tg CH2Cl2/year, with ∼70% of this total due to anthropogenic Asian emissions, and ∼20% emanating from the ocean.

Over the 10-year period our results show (a) a continual increase of global CH2Cl2 emissions of 37 Gg/year that matches well with independent emissions estimates produced for the 2018 WMO/UNEP Ozone Assessment, and (b) a steady decrease of C2Cl4 emissions of 2.5-5 Gg/year, mostly due to reductions from Europe and North America. Modelled CH2Cl2 and C2Cl4 concentrations using the posterior emissions agree well with independent observations (i.e. not used in the inversion itself) throughout the vertical extent of the troposphere, including data from the recent CAST, CONTRAST and ATTREX aircraft missions over the West Pacific.