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Growth in the stratospheric loading of halogenated very short-lived substances and their impact on ozone and climate

R. Hossaini (1), M. Chipperfield (2), S. Montzka (3), and S. Dhomse (2)

(1) Department of Chemistry, University of Cambridge, Cambridge, UK. (R.Hossaini@leeds.ac.uk), (2) School of Earth & Environment, University of Leeds, Leeds, UK., (3) NOAA, Boulder, CO, USA.

Observations show that very short-lived substances (VSLS), with lifetimes generally <6 months, are an important source of stratospheric halogens. Bromine-containing VSLS are produced naturally by seaweed and phytoplankton, whereas chlorine-containing VSLS are primarily anthropogenic. We have used the TOMCAT/SLIMCAT chemical transport model to quantify the depletion of ozone in the lower stratosphere from VSLS, and a radiative transfer model to quantify the radiative effects of that ozone depletion.

According to our simulations, VSLS-driven ozone loss had a radiative effect nearly half that from long-lived halocarbons in 2011 and, since pre-industrial times, has contributed a total of about -0.02 W m^{-2} to global radiative forcing. We find that natural bromine-containing VSLS exert a $\times 3.6$ larger ozone radiative effect than long-lived halocarbons, normalized by halogen content, and are therefore relatively efficient at influencing climate.

We have also developed a detailed chemical mechanism describing the tropospheric degradation of chlorine-containing VSLS. The scheme was included in the CTM and used to quantify trends in the stratospheric injection of chlorine from anthropogenic VSLS, Cl_y[VSLS], between 2005 to 2013. By constraining the model with surface measurements of chloroform ($CHCl_3$), dichloromethane (CH_2Cl_2), tetrachloroethene (C_2Cl_4), trichloroethene (C_2HCl_3) and 1,2-dichloroethane (CH_2ClCH_2Cl), we infer a 2013 stratospheric Cl_y[VSLS] mixing ratio of 123 parts per trillion (ppt). Stratospheric injection of source gases dominates this supply, accounting for $\sim$83% of the total. The remainder comes from VSLS-derived organic products, phosgene ($COCl_2$, 7%) and formyl chloride ($CHClO$, 2%), and also hydrogen chloride (HCl, 8%). Stratospheric Cl_y[VSLS] increased by $\sim$52% between 2005-2013, with a mean growth rate of 3.7 ppt Cl/yr. This increase is due to recent and ongoing growth in anthropogenic CH_2Cl_2 – the most abundant chlorinated VSLS not controlled by the Montreal Protocol.

We conclude that potential further significant increases in the atmospheric abundance of short-lived halogen substances, through changing natural processes or continued anthropogenic emissions, could (i) be important for future climate and (ii) become relevant for understanding the future evolution and recovery of stratospheric ozone.

References

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