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Short-lived halocarbons efficient at influencing climate through ozone loss in the upper troposphere-lower stratosphere

Ryan Hossaini (1), Martyn Chipperfield (1), Steven Montzka (2), Alex Rap (1), Sandip Dhomse (1), and Wuhu Feng (1)

(1) University of Leeds, School of Earth and Environment, Leeds, United Kingdom (r.hossaini@leeds.ac.uk), (2) National Oceanic and Atmospheric Administration (NOAA), Boulder, USA

Halogenated very short-lived substances (VSLS) of both natural and anthropogenic origin are a significant source of atmospheric bromine, chlorine and iodine. Due to relatively short atmospheric lifetimes (typically <6 months), VSLS breakdown in the upper troposphere-lower stratosphere (UTLS), where ozone perturbations drive a disproportionately large climate impact compared to other altitudes. Here we present chemical transport model simulations that quantify VSLS-driven ozone loss in the UTLS and infer the climate relevance of these ozone perturbations using a radiative transfer model.

Our results indicate that through their impact on UTLS ozone, VSLS are efficient at influencing climate. We calculate a whole atmosphere global mean radiative effect (RE) of -0.20 (-0.16 to -0.23) Wm-2 from natural and anthropogenic VSLS-driven ozone loss, including a tropospheric contribution of -0.12 Wm-2. In the stratosphere, the RE due to ozone loss from natural bromine-containing VSLS (e.g. CHBr3, CH2Br2) is almost half of that from long-lived anthropogenic compounds (e.g. CFCs) and normalized by equivalent chlorine is $\sim$4 times larger. We show that the anthropogenic chlorine-containing VSLS, not regulated by the Montreal Protocol, also contribute to ozone loss in the UTLS and that the atmospheric concentration of dichloromethane (CH2Cl2), the most abundant of these, is increasing rapidly. Finally, we present evidence that VSLS have made a small yet previously unrecognized contribution to the ozone-driven radiative forcing of climate since pre-industrial times of -0.02 (-0.01 to -0.03) Wm-2. Given the climate leverage that VSLS possess, future increases to their emissions, either through continued industrial or altered natural processes, may be important for future climate forcing.