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Stratospheric ozone and HCl trends (2000-2016): how significant is recent growth in chlorinated very short-lived substances?

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Chlorinated very short-lived substances (VSLS), such as dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$) and perchloroethylene (C_2Cl_4) constitute a small, but growing, source of stratospheric chlorine (e.g. WMO, 2014). High-altitude aircraft observations provide some constraint on the stratospheric source gas injection of chlorine from these compounds, though product gas injection – from VSLS-derived phosgene and HCl produced in the troposphere – is uncertain. Furthermore, in addition to an overall paucity of atmospheric VSLS observations, the impact of VSLS growth on stratospheric composition trends in the recent past (e.g. ozone, HCl) remains poorly constrained.

We have used the TOMCAT/SLIMCAT 3-D chemical transport model (CTM) to quantify (a) the stratospheric chlorine injection from VSLS (source and product gas contributions) between 2000 to 2016, and (b) the resulting impact on stratospheric ozone and HCl trends at a range of altitudes over this period. In particular, we consider the role of VSLS in the mid-latitude lower stratosphere, where recent satellite observations show a continuous downward trend in ozone in recent decades.

Based on our model, we estimate that the stratospheric source gas injection of chlorine from CH_2Cl_2, $CHCl_3$, C_2Cl_4, C_2HCl_3 and $C_2H_4Cl_2$ increased from 55 ($\pm$ 10) ppt Cl in 2000, to $\sim$92 ($\pm$ 18) ppt Cl in 2016. The model reproduces well observed amounts of these compounds around the tropopause, including recent high-altitude measurements obtained during the NASA ATTREX missions. We estimate a smaller increase in the stratospheric product gas injection of chlorine from the above compounds, from $\sim$18 ($\pm$ 4) ppt Cl in 2000, to 22 ($\pm$ 6) ppt Cl in 2016, with VSLS-derived phosgene accounting for $\sim$30% of this input and HCl the remainder. Total chlorine from VSLS (i.e. the sum of source and product gas injection) is estimated at $\sim$114 ($\pm$ 23) ppt Cl in 2016, having increased at a rate of $\sim$4 ppt Cl/year over the 2004-2016 period.

Modelled stratospheric HCl trends were calculated with and without the inclusion of chorine from VSLS, and compared to those derived from the ACE satellite instrument over the 2004-2016 period. The inclusion of VSLS leads to significantly better agreement between the modelled and observed HCl trends, particularly in the mid-upper stratosphere. The model overpredicts the rate of HCl decline when VSLS are not included, suggesting that VSLS growth has already begun to offset some of the benefits of the Montreal Protocol in reducing stratospheric chlorine.

We also show that our model simulations capture the downward trend in lower stratospheric ozone that has been observed by merged satellite products over the last two decades. We will present comparisons of modelled ozone versus the satellite data, and sensitivity simulations examining the influence of VSLS growth on the downward ozone trend versus dynamical influences, that are found to be the predominant driver.