Enhanced dichloromethane in South East Asia during the East Asian winter monsoon

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The success of the Montreal Protocol to phase out the consumption of ozone-depleting substances has facilitated the gradual healing of the ozone layer. However, recent research has revealed that increasing emissions of chlorinated very short-lived substances (Cl-VSLS) threaten to delay this recovery. Historically, VSLS were not considered damaging to ozone as they have relatively short atmospheric lifetimes and are not expected to reach the stratosphere in large quantities. The short lifetime and corresponding low ozone depletion potentials (ODPs) has led them to be excluded from the Montreal Protocol. However, chlorinated VSLS, which are mainly anthropogenic in origin, were found to be increasing (~1.3 ± 0.3 ppt Cl yr-1, 2008-2012) in contrast to the decline of long-lived controlled chlorinated substances (-13.4 ± 0.9 ppt Cl yr-1) over the same period (Carpenter et al., 2014). Importantly, the majority of the change was contributed by dichloromethane (CH_2Cl_2), with an increase of ~60% over the last decade.

This study focuses on observations of CH_2Cl_2 at Bachok, Malaysia (6.0696° N, 102.3972° E) during the Northern Hemisphere winters, 2013/14 and 2015/16. The objectives were (1) to observe the variability of CH_2Cl_2 from one winter to another, (2) to identify any enhancement above background level for CH_2Cl_2 and (3) to investigate the potential source regions that can contribute towards the variation of CH_2Cl_2. We used the UK Met Office's Numerical Atmospheric Modelling Environment (NAME) atmospheric dispersion model to produce footprints of where the air sampled during Bachok campaign had previously been close to the Earth's surface. NAME performed 12-day backward calculations for batches of 30,000 inert particles from 0-100 m above the surface of measurement site. The trajectories were calculated using three dimensional meteorological fields produced by the UK Met Office's Numerical Weather Prediction tool, the Unified Model (UM). At the end of the 12 day travel time, a gridded time integrated particle density was produced. Subsequently, the NAME footprint and particle distribution analysis were compared with the time-series of CH_2Cl_2 measured during the campaigns in order to explain the variations in the mixing ratio of CH_2Cl_2 and identify potential source regions. Additionally, meteorological analysis using ERA-Interim Reanalysis was used (1) to characterize cold surge events (an important meteorological process occurring

during the Northeast Monsoon) using a cold surge index and (2) to explore the influence of cold surges towards the variation of observed CH_2Cl_2.

The range of CH_2Cl_2 observed at Bachok was 47 to 352 ppt. Interestingly, all of the samples already exceed the levels of CH_2Cl_2 typically observed in the marine boundary layer (range = 21.8 - 34.3 ppt). The strongest enhancements of CH_2Cl_2 were observed on days which are linked to the influence of cold surges and high East Asian emissions. During these events, the NAME footprints and particle contribution analysis suggest that the source of CH_2Cl_2 is largely from China. Also, high concentrations of CH_2Cl_2 correspond to the occurrence of Northeast Monsoon cold surge.
In conclusion, this study highlights the important role of Northeast Monsoon cold surges and East Asia emissions toward the enhancement of CH_2Cl_2 in the region of the South China Sea. Importantly, this region is a major source region for air entering the stratosphere. If there are no controls on East Asian emissions, the contribution of Cl-VSLSs such as CH_2Cl_2 and CH_2ClCH_2Cl (1,2-dichloroethane) to stratospheric chlorine loading is likely to increase substantially in the coming years and potentially delay the long-term recovery of the ozone layer.

Keywords: Chlorinated very short-lived substances (VSLS), Dichloromethane