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Deriving an atmospheric budget of total organic bromine using airborne in-situ measurements of brominated hydrocarbons in the Western Pacific during SHIVA

Stephan Sala (1), Harald Bönisch (1), Timo Keber (1), Dave Oram (2,3), Graham Mills (3), and Andreas Engel (1) (1) Goethe University Frankfurt, Institute for Atmospheric and Environmental Sciences, Frankfurt / Main, Germany (s.sala@iau.uni-frankfurt.de), (2) National Centre for Atmospheric Science, Centre for Oceanic and Atmospheric Science, University of East Anglia, Norwich, United Kingdom, (3) Centre for Oceanic and Atmospheric Science, University of East Anglia, Norwich, United Kingdom

Halogenated hydrocarbons play a major role as precursors for stratospheric ozone depletion. Released from the surface in the troposphere, the halocarbons reach the stratosphere via transport through the tropical tropopause layer. The contribution of the so called very short lived species (VSLS), having atmospheric lifetimes of less than half a year as sources gases for stratospheric bromine is significant. Source gas observations of long-lived bromine compounds and VSLS have so far not been able to explain the amount of bromine derived in the stratosphere from observations of BrO and modeling of the ratio of BrO to total bromine. Due to the short lifetimes and the high atmospheric variability, the representativeness of the available observations of VSLS source gases remains unclear, as these may vary with region and display seasonal variability.

During the SHIVA (Stratospheric Ozone: Halogen Impacts in a Varying Atmosphere) project an extensive dataset with over 700 samples of ambient air of all halogen species relevant for the atmospheric budget of total organic bromine (long lived halocarbons: H-1301, H-1211, H-1202, H-2402 and CH3Br, very short lived substances: CHBr3, CH2Br2, CHBr2Cl, CHBrCl2 and CHBrCl) have been collected from onboard the FALCON aircraft in the West Pacific region. Measurements were performed with the newly developed fully-automated in-situ instrument GHOST-MS (Gas chromatograph for the Observation of Tracers – coupled with a Mass Spectrometer) by the Goethe University of Frankfurt and with the onboard whole-air sampler WASP with subsequent ground based state-of-the-art GC/MS analysis by the University of East Anglia.

We will present the datasets, compare these to other observation, derive a bromine budget for the West Pacific and derive an estimate of the amount of bromine from VSLS reaching the stratosphere.

Using the mean mixing ratios in the upper troposphere of the halocarbons mentioned above, the calculated budget of the total organic bromine is (20.01 ± 1.77) ppt and the total amount from VSLS is (4.35 ± 0.59) ppt. Regarding the entry of bromine in the stratosphere, the estimation of the bromine content at the LZRH gives a value of (18.54 ± 1.78) ppt and a value of (2.88 ± 0.60) ppt from VSLS, reflecting a fraction of 16% of the total bromine budget.