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Bromoform and Dibromomethane Emission During the SHIVA Western Pacific 2011 Field Campaign: A 3-D Model Case Study

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Halogenated very short-lived species (VSLS) with atmospheric lifetimes of <6 months can be transported to the stratosphere, particularly in regions experiencing rapid vertical transport due to deep convection. Once in the stratosphere bromine released from VSLS contributes to ozone depletion. While the Montreal Protocol has controlled the emission of longer-lived anthropogenic halogenated species, the quantitative impact of naturally sourced VSLS remains unclear and requires further investigation.

We have used the TOMCAT offline global 3-D chemical transport model (CTM) to test different VSLS emission scenarios. In this study, TOMCAT is forced using 6-hourly European Centre for Medium-Range Weather Forecasts analyses, has 60 vertical levels from the surface to ∼60 km and a horizontal resolution of 2.8°x2.8°. Previous work using TOMCAT into halogenated VSLS emission and transport has involved the use of fixed surface mixing ratios of 1.2 pptv bromoform and dibromomethane in the bottom two layers of the model surface in the Tropics (Hossaini et al., 2010). Although an accurate representation of surface mixing ratios of these VSLS, the use of spatially varying emission fluxes should allow for improved accuracy in model predictions.

The EU-funded SHIVA Malaysia 2011 field campaign provided a comprehensive VSLS dataset obtained in a region where these source gases have the potential to reach the stratosphere and deplete ozone. Observations of VSLS were collected during November and December 2011 on board the DLR Falcon aircraft during sixteen local flights. Fourteen of these flights have been used in this study due to technical difficulties experienced on the remaining two flights. Four emission scenarios, including both top-down and bottom-up approaches derived from airborne measurements and ocean fluxes of VSLS, were used in TOMCAT and each scenario was compared to observations of bromoform and dibromomethane collected during the SHIVA campaign. The mean bias of each emission scenario against the SHIVA observations was calculated for all fourteen flights considered.

Results indicate that the bottom-up emission scenario, derived from measured oceanic fluxes of bromoform and dibromomethane, matches the SHIVA observed values of both major VSLS source gases more closely than that of the top-down emission scenarios (0.16 pptv total mean bias for both bromoform and dibromomethane for all fourteen flights considered). Although slightly underestimating the SHIVA observed values, all other emission scenarios overestimate the observations of bromoform by a greater magnitude. Dibromomethane is underestimated by two emission scenarios and overestimated by one, all by a greater extent than that of the bottom-up emission scenario derived from measured VSLS oceanic fluxes. Global mixing ratio maps highlight the difference in global distribution of VSLS emissions between each emission scenario.

Our work suggests that bottom-up derived emission estimates may be more accurate than those derived from airborne measurements, but that all methods produce surface level values within the WMO ranges given for MBL bromoform and dibromomethane. A truly global emission dataset is required to accurately represent VSLS in global models, with particular emphasis on the Southern Hemisphere.

References: Hossaini, R., Chipperfield, M. P., Monge-Sanz, B. M., Richards, N. A. D., Atlas, E., and Blake, D. R.: Bromoform and dibromomethane in the tropics: a 3-D model study of chemistry and transport, Atmos. Chem. Phys., 10, 719-735, doi:10.5194/acp-10-719-2010, 2010