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Global seasonal distribution of CH_2Br_2 and $CHBr_3$ in the upper troposphere and lower stratosphere

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Halogens from long-lived anthropogenic substances contribute to the depletion of stratospheric ozone. Besides these long-lived substances, chlorinated and brominated substances with lifetimes of less than 6 months are additional sources of stratospheric halogens. These substances, also known as very short-lived substances (VSLSs), have both natural and anthropogenic origins. The contribution of chlorinated VSLSs (Cl-VSLSs) to stratospheric chlorine is a few percent. In comparison, brominated VSLSs (Br-VSLS) contribute to about a quarter of the stratospheric bromine. The relative contribution of VSLSs to stratospheric halogen loading is expected to increase as the Montreal Protocol controlled substances progressively decrease. Due to their short lifetimes, VSLSs rapidly release their halogen content into the lowermost stratosphere, a region where changes in ozone have a relatively large impact on surface climate.

Here we present the global seasonal distribution of the two major Br-VSLSs CH_2Br_2 and $CHBr_3$, which account for about 80 % of total organic Br-VSLS. Measurements from four High Altitude and Long Range Research Aircraft (HALO) missions, the HIAPER Pole-to-Pole Observations (HIPPO) mission, and the Atmospheric Tomography (ATom) mission were used for this purpose. Observational results show a similar seasonality of CH_2Br_2 in the free and upper troposphere of

both hemispheres and less clear seasonality with larger variations for $CHBr_3$. The distribution of CH_2Br_2 in the lowermost stratosphere suggests differences in hemispheric autumn, where the influx of tropospheric air seen in northern hemispheric summer to autumn is not evident in the Southern Hemisphere. However, the southern hemispheric database is insufficient to quantify this difference. The observed distributions were additionally compared to distributions based on model results of TOMCAT and CAM-Chem, both using the emission inventory of Ordóñez et al. (2012). Neither model was able to reproduce the seasonal distribution of CH_2Br_2 in the Southern Hemisphere. In contrast, both models show a pronounced seasonality of $CHBr_3$ in both hemispheres, which is not confirmed by observations. The distributions of both substances in the lowermost stratosphere are overall well captured by the models, except for southern hemispheric autumn with considerably lower mixing ratios in the observations.