



Transport and Chemistry of Short-Lived Bromocarbons in the Tropics

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We have developed a detailed chemical scheme for the degradation of the short-lived source gases bromoform (CHBr_3) and dibromomethane (CH_2Br_2) and implemented it in the TOMCAT/SLIMCAT three-dimensional (3D) chemical transport model (CTM). The CTM has been used to predict the distribution of the two source gases (SGs) and 11 of their organic product gases (PGs). These first global calculations of the organic PGs show that their abundance is small. The longest lived organic PGs are CBr_2O and CHBrO , but their peak tropospheric abundance relative to the surface volume mixing ratio (vmr) of the SGs is less than 5%. We calculate their mean local tropospheric lifetimes in the tropics to be ~ 7 and ~ 2 days (due to photolysis), respectively. Therefore, the assumption in previous modelling studies that SG degradation leads immediately to inorganic bromine seems reasonable.

We have compared observed tropical SG profiles from a number of aircraft campaigns with various model experiments. In the tropical tropopause layer (TTL) we find that the CTM run using p levels (TOMCAT) and vertical winds from analysed divergence overestimates the abundance of CH_2Br_2 , and to a lesser extent CHBr_3 , although the data is sparse and comparisons are not conclusive. Better agreement in the TTL is obtained in the sensitivity run using θ levels (SLIMCAT) and vertical motion from diabatic heating rates. Trajectory estimates of residence times in the two model versions show slower vertical transport in the SLIMCAT θ -level version. In the p -level model even when we switch off convection we still find significant amounts of the SGs considered may reach the cold point tropopause; the stratospheric source gas injection (SGI) is only reduced by $\sim 16\%$ for CHBr_3 and $\sim 2\%$ for CH_2Br_2 without convection.

Overall, the relative importance of the SG pathway and the PG pathway for transport of bromine to the stratospheric overworld ($\theta > 380$ K) has been assessed. Assuming a 10-day washout lifetime of Br_y in TOMCAT, we find the delivery of total Br from CHBr_3 to be 0.72 pptv with $\sim 53\%$ of this coming from SGI. Similarly, for CH_2Br_2 we find a total Br value of 1.69 pptv with $\sim 94\%$ coming from SGI. We infer that these species contribute ~ 2.4 pptv of inorganic bromine to the lower stratosphere with SGI being the dominant pathway. Slower transport to and through the TTL would decrease this estimate.