Geophysical Research Abstracts
Vol. 15, EGU2013-9420, 2013
EGU General Assembly 2013

Uncertainties and constraints regarding the contribution of very short-lived substances to stratospheric bromine loading

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A major factor of uncertainty in the assessment of stratospheric bromine loading is the unclear role of very short-lived substances (VSLS). One of the major obstacles for short-lived source gases in contributing to the stratosphere is generally thought to be the loss of inorganic bromine (Bry) in the tropical tropopause layer due to dehydration. Besides the dehydration process itself, transportation pathways and velocities are also of vital importance as they influence the partitioning between mostly insoluble organic source gases and partly soluble inorganic degradation products. To investigate this complex system we employ an extensive set of sensitivity calculations with a three-dimensional chemistry transport model comprising a consistent parameterization of convective transport and a comprehensive chemistry scheme. The model considers the two most important bromine VSLS, bromoform (CHBr3) and dibromomethane (CH2Br2) assuming a fixed and uniform detrainment mixing ratio of 1 pptv each. Despite our simplified approach our model agrees reasonably well with available observations of bromine source and product gases. We find that source gas injection is the dominant pathway for VSLS into the stratosphere; about 50% of CHBr3 and 93% of CH2Br2 is able to overcome the cold point tropopause at approximately 17 km altitude, modulated by the inter-annual variability of the vertical transport efficiency. In fact, our sensitivity calculations indicate that the extent of source gas injection of CHBr3 is highly sensitive to the strength of convection and large-scale ascent; in contrast, modifying the photolysis or the destruction via OH yields a significantly smaller response. The next important aspect we identified is that the partitioning of available Bry from short-lived sources is clearly shifted away from HBr, according to our current state of knowledge the only member of the Bry family which is efficiently adsorbed on ice particles. This effect is caused by very efficient heterogeneous reactions on ice surfaces which reduce the HBr/Bry fraction below 15% at the tropical tropopause. Under these circumstances there is no significant loss of Bry due to dehydration in the model; VSLS contribute fully to stratospheric bromine. In addition, we conduct several sensitivity calculations to test the robustness of this result. The loss of inorganic bromine is not very sensitive to moderate changes of the involved parameters such as the abundance of water vapor, sedimentation velocity of particles or ice uptake coefficients. However, dehydration may play a minor role for Bry removal under the assumption that HOBr is efficiently adsorbed on ice as well since the heterogeneous reactions alter the partitioning equilibrium of Bry in favor of HOBr (up to 12% loss of bromine from VSLS). Even in the extreme and unrealistic case that adsorbed species on ice particles are instantaneously removed the maximum loss of bromine does not exceed 25%.