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Probing the tropical tropopause layer for organic and inorganic bromine

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Bromine chemistry impacts the levels of ozone in the upper troposphere and the stratosphere. An accurate quantitative understanding of the sources, sinks, and chemical transformation of bromine species is thus important to understand the photochemistry and budget of bromine in the tropical upper troposphere, tropopause layer and lowermost stratosphere (UT/TTL/LS). These regions are also known to serve as a gateway for delivery of ozone depleting gases to the stratosphere. CH_3Br, halons, short-lived organic bromine precursors (VSLS), such as $CHBr_3$, CH_2Br_2, and possibly inorganic product gases have been identified as the main bromine gases delivered to the stratosphere. However, many important details of the transport and delivery of VSLS and inorganic bromine compounds through the TTL are still uncertain. Moreover, a number of chemical processes, including the transformation of the source gases and cycling of inorganic bromine species at low ambient temperature and on ice particles are also poorly understood.

The presentation reports measurements of CH_4, O_3, NO_2, and BrO performed by different instruments and techniques during the 2013 NASA-ATTREX flights in the TTL and LS. The interpretation of our measurements is supported by chemical transport model (SLIMCAT) simulations. SLIMCAT results, in conjunction with extensive radiative transfer calculations using the Monte Carlo model McArtim, also are used to improve retrieval of O_3, NO_2, and BrO concentrations from limb scattered sunlight measurements made with the Differential Optical Absorption Spectroscopy (DOAS) technique during ATTREX. The chemical transport model also allows us to attribute observed concentration variations to transport and to photochemical processes. When properly accounting for the transport-related concentration variations in methane and ozone, we find that measured BrO mostly agrees with model simulations. An exception are regions where the contribution of the short-lived CH_2Br_2 or the partitioning of $BrONO_2$ plays an important role. The present observations confirm previous findings on the formation of $BrONO_2$ of our workgroup.

When the errors and uncertainties in the involved photochemical reaction rates, which are mostly available for higher temperature than those (>188K) encountered during the NASA ATTREX mission, are accordingly considered in the calculation of the bromine partitioning, a total bromine budget (Br_y) in the tropical tropopause layer of 20.3 ppt to 22.3 ppt is inferred, depending on the flight, or region, respectively.