Measurement and simulation of CH_4, O_3, NO_2, BrO, and major brominated source gases during the NASA-ATTREX Global Hawk deployments in 2013: Implications for the photochemistry and total amount of bromine in the TTL and stratosphere

K. Pfeilsticker[1], B. Werner[1], E. Atlas[6], R. Cheung[2], M. P. Chipperfield[3], F. Colosimo[2], T. Deutschmann[1], J. Elkins[5], D. Fahey[5], W. Feng[4], J. Festa[2], Ru-Shan Gao[5], R. Hossaini[3], M. Navarro[6], R. Raecke[1], L. Scalone[1], M. Spolaor[2], T. Thornberry[5], C. Tsai[2], and J. Stutz[2]

[1]*Institute of Environmental Physics, University of Heidelberg, Heidelberg, Germany*
[2]*Department of Atmospheric and Oceanic Science, University of California Los Angeles, Los Angeles, USA*
[3]*Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, UK*
[4]*National Centre for Atmospheric Science, School of Earth and Environment, University of Leeds, UK*
[5]*NOAA Earth System Research Laboratory, Boulder, USA*
[6]*The Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, USA*

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, and 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 for example how, and to what extent, VSLS and inorganic bromine compounds are transported to the TTL are not clear to date. 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 present talk (which complements the talk of Navarro et al. on the GWAS measurements of brominated source gases) reports on 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 assist in an improved concentration retrieval of O_3, NO_2, and BrO from limb scattered sunlight measurements using the well-known Differential Optical Absorption Spectroscopy (DOAS) technique. The model also allows us to attribute observed concentration variations to transport and photochemical processes. When properly accounting for the transport-related concentration variations, measured BrO is found to be mostly larger than model simulations using standard JPL-kinetic data in the model. When these kinetic data, which are mostly available for higher temperatures than those (> 188K) encountered during the NASA ATTREX mission are adjusted to match our temperature observations, this gap can be largely closed, and total bromine in the TTL is quantified to 21±1 ppt in 2013.

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