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The Budget of Halogen Compounds in the tropical UTLS

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Inorganic halogen compounds play a major role in the context of stratospheric ozone loss. Most of the bromine and chlorine present in the stratosphere is injected from the troposphere in the tropics. While the greater part is transported in the form of long-lived natural (e.g. CH_3Cl, CH_3Br) and anthropogenic compounds (e.g. CFCs, Halons) and converted to inorganic forms in the stratosphere, a sizeable contribution of halogen transport in the form of inorganic and very short lived organic substances (VSLSs) that can be processed before reaching the stratosphere has been recognised in recent years (WMO 2006). However, this contribution has not yet been fully quantified. It may make up as much as $\sim$30 % of the total source of stratospheric bromine and thus could have severe implications for the stratospheric bromine budget and ozone loss by catalytic cycles involving bromine oxide (BrO). The relative contribution of VSLSs to the chlorine budget is much smaller, but may be important in determining the actual amount of inorganic chlorine present in the UTLS. This has implications for chlorine activation and ozone loss induced by heterogeneous chemistry on background aerosol and cirrus particles in this region (Solomon et al., 1997; von Hobe et al., 2006).

Here we present calculations using the chemistry module of the Chemical Lagrangian Model of the Stratosphere (CLaMS) to investigate different source, transport and chemical processing scenarios to estimate the amount of inorganic bromine and chlorine compounds in the tropical UTLS. An upper limit of inorganic bromine from VSLSs is deduced, which is compared to estimates derived from balloon-borne DOAS and airborne in-situ measurements of BrO carried out during several field campaigns, including HIBISCUS, TROCCINOX, SCOUT-O_3 and SCOUT-AMMA, as well as es-

timates based on SCIAMACHY satellite data. For chlorine, we attempt to constrain the amount of inorganic chlorine that can realistically be present in UTLS air masses with a primarily tropospheric origin, and discuss the results in the context of unexpectedly high amounts of chlorine oxide (ClO) following cirrus events observed during TROCCINOX and SCOUT-O_3 (von Hobe et al., 2006).

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