



Airborne CARIBIC measurements in the UTLS: Distribution, seasonal cycle and variability of acetone

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In the Earth's atmosphere volatile organic compounds (VOCs) have multifaceted functions, e.g. controlling (together with reactive nitrogen) the photochemical production of ozone or influencing directly (via e.g. acetone) or indirectly (via ozone) the Earth's oxidation capacity. The UTLS region (upper troposphere / lower stratosphere) plays a key role in the climate system. Any change in its structure and chemical composition results in changes in the radiative forcing of the atmosphere and thus in climate change. The UTLS is – in spite of the immense importance – still one of the least understood regions of the atmosphere. The need for a better understanding of the dynamics across the tropopause is therefore obvious.

Since May 2005 the CARIBIC passenger aircraft (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container – Lufthansa, Airbus 340-600) measures ~100 trace gases and aerosol components within the UTLS (9-12 km altitude).

Acetone (CH_3COCH_3), as one of the most abundant oxygenated volatile organic compounds (OVOCs) in the atmosphere, can be used to gain more information about the dynamic processes and chemical cycles. The presented measurements have been carried out by the PTR-MS instrument (proton-transfer-reactions mass spectrometry) onboard CARIBIC. Because of the meteorological and geographical variance of the atmospheric layering, the tropopause is crossed up to 20 times during one (quasi-horizontal) long-range flight. Thereby tropospheric and stratospheric air is sampled to fractions of ~60 % and ~40 %, respectively. The stratospheric flight sections largely occur in the extra-tropical tropopause (transition) layer (exTL).

With an average of four long-distance flights per month between Germany and the major destinations in North- and South-America, as well as South-East Asia, the largest airborne dataset of acetone on a nearly worldwide scale has been composed.

The following major findings in the analysis of the collected data are shown:

- A strong seasonal variation of acetone occurs at the mid-latitude tropopause with maxima of approximately 900 pptv (parts per 10^{12} vol) in summer and minima of approximately 200 pptv in midwinter.
- This seasonality propagates into the LMS (lowermost stratosphere) in ~6 weeks with rapidly decreasing concentrations and increasing phase shifts reaching 2 km above the tropopause.
- Probability density functions (PDFs) and the course of the seasonal variation of acetone relative to the tropopause are interpreted regarding the in-mixing and subsequent dispersion of acetone in the LMS.
- The production of HO_x due to the photolysis of acetone is compared with the one following the photolysis of ozone and subsequent reaction of $\text{O}(^1\text{D})$ with water vapour.