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CARIBIC passenger aircraft measurements in the UTLS: Distribution and seasonal variation of acetone and its impact on the production of HO_x

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Volatile Organic Compounds (VOCs) are key players in tropospheric chemistry. VOCs e.g. control (together with NO_x) the photochemical production of ozone or they contribute directly (e.g. via acetone) or indirectly to the oxidation capacity of the atmosphere.

The UTLS region (upper troposphere / lower stratosphere) plays a key role in the climate system and any change of its structure and chemical composition can lead to changes in the Earth's radiative forcing.

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 $\sim$100 trace gases and aerosol components in the UTLS (9-12 km altitude) on four long-distance flights per month. Acetone is measured using a PTR-MS (proton-transfer-reaction mass spectrometer). Over the years the largest airborne in-situ dataset of acetone on a nearly worldwide scale has been composed. Acetone (CH_3COCH_3) with a typical lifetime of $\sim$3-6 weeks and an estimated source strength of 40-100 Tg/year is one of the most abundant oxygenated volatile organic compounds (OVOCs) in the atmosphere.

The coverage of the acetone dataset was expanded by folding the CARIBIC data with 5-day back trajectories from the ECMWF model. In winter acetone mixing ratios of around 300 pptv were observed on average between 8 km altitude up to the tropopause with hints for biomass burning over North Africa. In summer elevated values $>$1500 pptv can be observed especially between 35° - 70°N.

The data were used to estimate the rate of OH production due to acetone photolysis and the findings were contrasted to the rate of OH production from ozone (O_3+ hυ –$>$ O(^{1}D) + O_2 and O(^{1}D) + H_2O –$>$ 2 OH). It is shown that around the tropopause the OH production from acetone photolysis can compete with OH production from ozone photolysis. Due to the vertically strongly decreasing acetone concentration the OH production from acetone photolysis however becomes a minor channel above the tropopause, i.e. in the lowermost stratosphere.