Chemical processing and transport of O_3, CO, H_2O, acetone, and acetonitrile in the UT/LMS inferred from long-term, high-resolution observations onboard the CARIBIC passenger aircraft

Poster M09

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The extratropical tropopause transition layer (ex-TL)

… names a layer sandwiched at the lower border by the tropopause and at the upper border by the unperturbed lowermost stratosphere (LMS) that is not influenced by recent in-mixing of tropospheric air. It is usually identified by linear or slightly curved correlations between a tropospheric tracer and a stratospheric tracer.

The nature as well as many properties of the ex-TL (seasonal variation, turnover time, short-term variability, convection-induced impact) are badly quantified, yet. Likewise, its relation to the recently discovered and meteorologically defined tropopause inversion layer (TIL) is not known, yet.

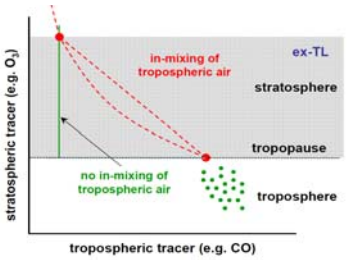

Fig. 1: *Schematic view of a typical tracer correlation around the extra-tropical tropopause*

CARIBIC

In CARIBIC a measurement container with 18 instruments is deployed once a month for four long-distance flights onboard an A340-600 of **Lufthansa**.

Almost 100 trace gases and aerosol parameters are measured at typical flight altitudes of 9-12 km. Since May 2005, more than 100 measurements flights were carried out.

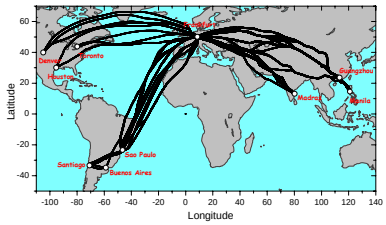

Fig. 2: *Flight paths May 2005 – July 2008*

The ex-TL during CARIBIC

Fig. 3 shows a typical O_3-CO correlation observed between China and Germany in February 2007.

The mixing line or ex-TL, resp., is clearly visible with a weak spatial dependence (red line observed at 42°E, green line at 69°E, both at ~50°N). The mixing reservoirs are well-defined.

The vertical extension (thickness) of the ex-TL shows a seasonal variation, with ~1.6 km in winter and ~2.6 km in summer (not shown).

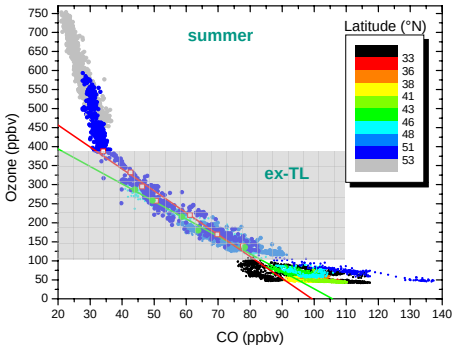

Fig. 3: *Correlation between O_3 and CO for a 11-hour flight (~10000 km) between South China and Germany at 10-12 km altitude*

Fig. 4 indicates a typical H_2O-CO correlation pattern observed in August 2006. The picture demonstrates that the mixing across the tropopause occurred along isentropic surfaces (lines). The blue data points characterize dry subtropical air, the black ones more humid high-latitude air. Note, the high H_2O m.r. in the ex-TL.

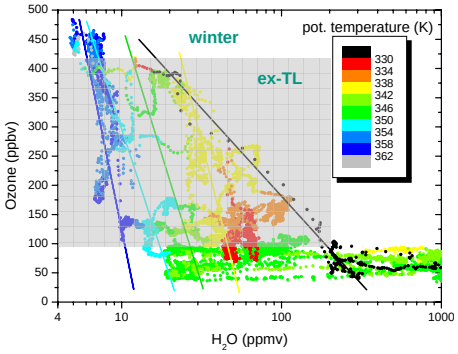

Fig. 4: *Correlation between O_3 and H_2O for a 11-hour flight from China to Germany.*

Fig. 5 shows the vertical profile of acetone relative to the tropopause for 4 flights in different seasons. There is a strong seasonal variation with highest acetone values and the highest variability in summer.

Small acetone concentrations around the top of the ex-TL in summer indicate a transport time in the order of the local (chemical) life time of acetone of ~4 weeks (not shown)

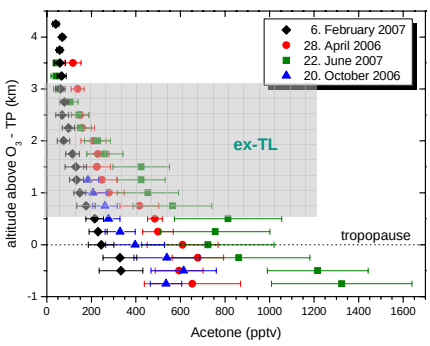

Fig. 5: *Vertical profiles of acetone observed in the mid-latitudes in different seasons.*

Climatology of acetone in the ex-TL

A strong seasonal variation of acetone is observed in the ex-TL (Fig. 6). In autumn the gradient across the tropopause is weak. Elevated acetone values remain in the lowermost stratosphere until ~November when the subsidence of clean, O_3-rich stratospheric air sets in.

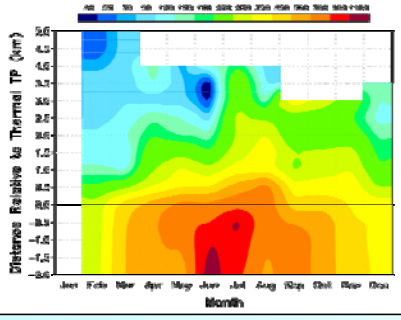

Fig. 6: *Seasonal variation of acetone around the tropopause inferred from the CARIBIC data 2005-2008.*

Seasonal variation of OH-production around tropopause

In Fig. 7 the seasonal cycle of the production of OH due to acetone is compared with the one caused by the photolysis of O_3 and subsequent reaction of $O(^1D)$ with H_2O (also measured in CARIBIC). Both OH sources show a similar seasonal variation with maxima in summer. Except of June (which appears to be an outlier), at the tropopause the photolysis of acetone exceeds the OH production due to the photolysis of ozone. Above the tropopause, this behavior quickly turns around, due to the step decrease of acetone in the stratosphere.

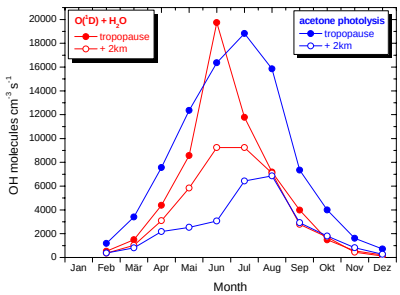

Fig. 7: *Seasonal cycle of the OH production due to the photolysis of O_3 (red) and acetone (blue) at the TP (full circles) and 2 km above it (open circles). Photolysis rates of ozone and acetone were taken from the Messy model (MPI-C, Mainz), which are based on JPL recommendations.*

Conclusion

Year-around tropospheric air mixes into the extra-tropical lowermost stratosphere and forms a mixing layer, the ex-TL, sandwiched between upper tropospheric and clean stratospheric air. Our H_2O data demonstrate that this in-mixing primarily occurs along isentropic surfaces (Fig. 4). Based on the lifetime of acetone a mixing time to altitudes at 2-3 km above the tropopause of ~4 weeks was inferred. Further constraints on the chemical processing and the ventilation of the extra-tropical tropopause region deliver trace gas climatologies, e.g. Fig. 6 for acetone. The seasonal variation of the production of OH near the mid-latitude tropopause was derived from the high-resolution CARIBIC acetone, O_3, and H_2O measurements. Both OH sources indicate a similar seasonal variation. At the tropopause, the photolysis of acetone even seems to dominate as OH source.

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