Geophysical Research Abstracts
Vol. 18, EGU2016-15743, 2016
EGU General Assembly 2016

A new approach to detect local correlations of tropospheric acetone and carbon monoxide sampled onboard the IAGOS-CARIBIC passenger aircraft

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Since 2005 a Lufthansa passenger aircraft is regularly used as a platform for in-situ measurements in the upper troposphere and lowermost stratosphere (UTLMS). Accommodated in a modified airfreight container 15 instruments are deployed in the cargo bay of the aircraft on four selected intercontinental flights per month measuring ∼100 species and aerosol parameters. In contrast to other projects of this scope, using a chemical mass spectrometer also volatile organic compounds like acetone (CH3COCH3) and acetonitrile (CH3CN) are detected enabling an investigation of their relationship with other tracers.

On a global scale acetone is predominantly emitted from the biosphere (∼37 Tg/a; MEGAN-MACC, Sinderarova et al. 2014) and comparably small amounts are directly emitted from biomass burning (∼2 Tg/a; GFED3, Van der Werf et al. 2010) and other anthropogenic sources (∼1 Tg/a; MACCity, Granier et al. 2011). However, at local levels the contributions from the different sources can strongly differ. Acetone is also secondarily produced in the atmosphere by the oxidation of various precursors, e.g. pinene and propane. The emissions of these precursors and their contribution to the total acetone source are not well known and a topic of ongoing discussions. In this context it is initially surprising that generally a good correlation between acetone and carbon monoxide (CO) has been observed in the lower atmosphere by different authors (e.g. de Reus et al. 2003). As a product of incomplete combustion CO is regularly used as a tracer for anthropogenic pollution and biomass burning.

In this study we present an improved method to detect local correlations in IAGOS-CARIBIC flights instead of mixing data from different flights or measured over great distances. Furthermore, a cluster analysis is applied to prevent the consideration of artificial correlations between two well separated clouds of data points. We use the concept of enhancement ratios (EnR) and a simple box model to interpret the correlations. Based on this model we investigate the temporal evolution of EnR and find out that an increase of acetone-CO-EnR with time is common for many scenarios and does not necessarily require secondary production of acetone. Applying our method and criteria to the full IAGOS-CARIBIC dataset reveals that 50 per cent of the available tropospheric acetone data shows a local correlation (correlation coefficient $r \geq 0.5$) with CO. In the Northern hemisphere subtropics and midlatitudes (23.5°N - 66.5°N) we find mean EnR of (17.8 $\pm$ 9.1) pptv/ppbv in summer and (9.4 $\pm$ 4.0) pptv/ppbv in winter. In air masses sampled above or originating from North America the seasonal cycle is more pronounced with a median EnR of 28.9 pptv/ppbv in summer and 10.1 pptv/ppbv in winter. This is in good agreement with the ratio of the total North American emissions of acetone and CO according to inventories from the ECCAD database. For East Asia EnR are low throughout the year and show only a weak annual cycle with a median of 12.4 pptv/ppbv in summer and 10.9 pptv/ppbv in winter. This can be tracked back to higher CO emissions from anthropogenic pollution. The presented findings point out that tropospheric EnR vary considerably depending on season, region and age of air mass, but have great potential to assess the ensemble of regional acetone and CO sources.