

## Seasonal variations and trends of greenhouse gases in the upper troposphere/lowermost stratosphere by flask-based aircraft measurements between Europe and Japan

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As part of the CONTRAIL project, we have conducted measurements of greenhouse gases ( $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ , and  $\text{SF}_6$ ) by monthly air sampling in the upper troposphere/lowermost stratosphere (UT/LMS) onboard commercial airliners between Europe and Japan since April 2012. The observed mixing ratios showed sharp gradients around the dynamical tropopause defined by potential vorticity calculated from the meteorological reanalysis fields. In the UT north of 50°N,  $\text{CH}_4$  and  $\text{SF}_6$  were higher and seasonal phase of  $\text{CO}_2$  were earlier than in the lower latitudes. In the LMS up to potential temperature of 50 K above the tropopause,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ , and  $\text{SF}_6$  exhibited seasonal variations with maxima in November/December and minima in April/May. The remarkable seasonal variation in the LMS is explained by the subsidence of air from the deeper stratosphere in spring and by the efficient flushing of the LMS with tropospheric air in autumn. We observed persistent increasing trends of the all greenhouse gases over the past 5 years both in the UT and LMS. Our measurements constitute a unique data set in the UT/LMS useful for investigating temporal and spatial variations of these radiatively and chemically important greenhouse gases.

Keywords: Greenhouse gases, Upper troposphere/Lower Stratosphere, Aircraft measurement