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Understanding 13C(CO2) and 18O(CO2) data obtained on CARIBIC aircraft air samples from the tropical troposphere, and the upper troposphere/ lowermost stratosphere

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The project CARIBIC (http://caribic-atmospheric.com) aims to study atmospheric chemistry and transport by regularly measuring many compounds in the free troposphere (FT) and the upper troposphere/lowermost stratosphere (UT/LMS) by using passenger aircraft. CO2 isotope ratios were measured by CARIBIC-1 (November-1999 to April-2002, mostly d13C(CO2)) and CARIBIC-2 (June-2007 to March-2009, highest precision d13C(CO2) and d18O(CO2) data). (The latter effort has been possible due to the contribution of JRC-IRMM, Geel, Belgium.) This gives a new, unique and fairly extensive data set for the NH free troposphere and the UT/LMS region. In particular, CARIBIC-2 utilizing high-quality and high-resolution sampling gave a detailed distribution of d18O(CO2), with signals not affected by sampling and canister effects. For the first time we can demonstrate how the variability in air mass origin (recognized by using chemical tracers) is recorded by CO2 isotope signals on the inter-continental scale. There are two main mechanisms responsible: (i) UT/LMS mixing and (ii) seasonality of tropospheric CO2 as well as variable degree of mixing background air and air masses affected by CO2 sources/sinks. In the UT/LMS region both d13C(CO2), d18O(CO2) and CO2 are found to correlate well with stratospheric tracers, in particular N2O. The data for the free and upper troposphere show however little latitudinal gradient and are in good agreement with the data of selected NOAA stations in NH tropics. We discuss that CO2 distribution in the NH FT and UT (at CARIBIC flight routes) is regulated by uplift and pole-wards transport of tropical air. Next, d18O(CO2) is thought to be a new, previously not recognized tracer in the UT/LMS region as well as tracer of gross CO2 fluxes, where CO2 exchanges its oxygen with natural water pools.