

Methyl Chloride as a Tracer of Tropical Tropospheric Air in the Lowermost Stratosphere Inferred from CARIBIC Passenger Aircraft Measurements

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CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) is a flying observatory that measures various atmospheric compounds from onboard a Lufthansa A340-600 aircraft. In this study, we present variations of nitrous oxide (N_2O) and methyl chloride (CH_3Cl) in the lowermost stratosphere (LMS) obtained from air samples collected by CARIBIC for the period 2008–2012. To correct for the secular increase of atmospheric N_2O , the CARIBIC N_2O data are expressed as ΔN_2O , deviations from the long-term trend at Mauna Loa, Hawaii (data provided by NOAA's measurements). The ΔN_2O values are used to classify CARIBIC air samples as representing air from either the LMS or the upper troposphere (UT). ΔN_2O undergoes a pronounced seasonal variation in the LMS with a minimum in spring. The amplitude increases going deeper in the LMS (up to potential temperatures of 50 K with respect to the thermal tropopause), as a result of the seasonally varying subsidence of air from the stratospheric overworld. Seasonal variations of CH_3Cl in the LMS are similar in phase to those of ΔN_2O . Significant correlations are found between CH_3Cl and ΔN_2O in the LMS from winter to early summer, both being affected by mixing between stratospheric air and UT air. This correlation however disappears in late summer to autumn. The slope of the CH_3Cl - ΔN_2O correlation observed in the LMS allows us to determine the stratospheric lifetime of CH_3Cl to be 35 ± 7 yr. Finally, we examine the partitioning of tropospheric air, tropical tropospheric air and extratropical tropospheric air in the LMS based on a mass balance approach using ΔN_2O and CH_3Cl . This analysis clearly indicates efficient inflow of tropical tropospheric air into the LMS in summer and demonstrates the usefulness of CH_3Cl as a unique tracer of tropical tropospheric air.

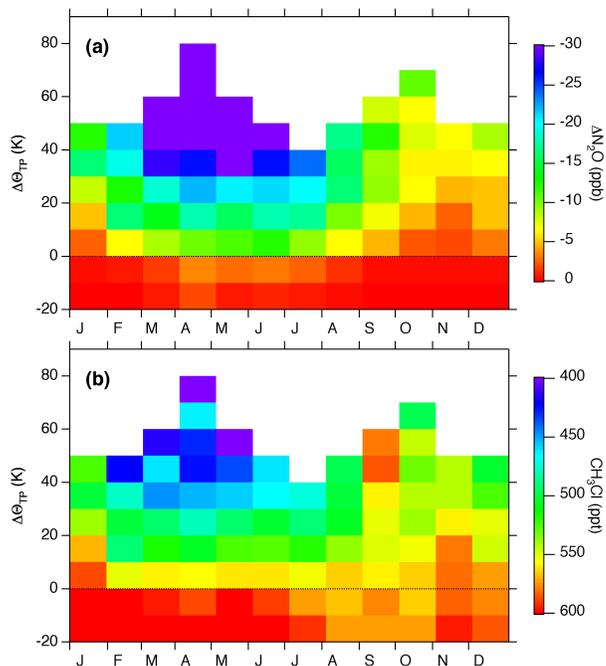


Figure 1. Seasonal variations of (a) ΔN_2O and (b) CH_3Cl at different potential temperature layers with respect to the thermal tropopause ($\Delta\Theta_{TP}$) in the UT/LMS observed by CARIBIC.