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Methyl chloride in the upper troposphere observed by CARIBIC: large-scale distributions and Asian summer monsoon outflow

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CARIBIC is a flying observatory onboard a Lufthansa A340-600 aircraft that observes various atmospheric compounds at almost monthly intervals. In this study, we present spatial and temporal variations of methyl chloride (CH_3Cl) in the upper troposphere (UT) observed mainly by CARIBIC for the years 2005-2011. The CH_3Cl mixing ratio in the UT over Europe was higher than that observed at a European surface baseline station throughout the year, indicative of a persistent positive vertical gradient at NH mid latitudes. A series of flights over Africa and South Asia show that CH_3Cl mixing ratios increase toward tropical latitudes, and the observed UT CH_3Cl level over these two regions and the Atlantic was higher than that measured at remote surface sites. Strong emissions of CH_3Cl in the tropics combined with meridional air transport through the UT may explain such vertical and latitudinal gradients. Comparisons with carbon monoxide (CO) data indicate that non-combustion sources in the tropics dominantly contribute to forming the latitudinal gradient of CH_3Cl in the UT. We also observed elevated mixing ratios of CH_3Cl and CO in air influenced by biomass burning in South America and Africa, and the emission ratios derived for CH_3Cl to CO in those regions agree with previous observations. In contrast, correlations indicate a high CH_3Cl to CO ratio of 2.9 ± 0.5 ppt ppb^{-1} in the Asian summer monsoon anticyclone and domestic biofuel emissions in South Asia are inferred to be responsible. We estimated the CH_3Cl emission in South Asia to be 134 ± 23 Gg Cl yr^{-1}, which is higher than a previous estimate due to the higher CH_3Cl to CO ratio observed in this study.

Keywords: CARIBIC, aircraft observation, methyl chloride, upper troposphere