

CARIBICで観測された上部対流圏の塩化メチル変動：広域分布と夏季のアジアモンスーンからの流出 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 (CH₃Cl) in the upper troposphere (UT) observed mainly by CARIBIC for the years 2005-2011. The CH₃Cl 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 CH₃Cl mixing ratios increase toward tropical latitudes, and the observed UT CH₃Cl level over these two regions and the Atlantic was higher than that measured at remote surface sites. Strong emissions of CH₃Cl 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 CH₃Cl in the UT. We also observed elevated mixing ratios of CH₃Cl and CO in air influenced by biomass burning in South America and Africa, and the emission ratios derived for CH₃Cl to CO in those regions agree with previous observations. In contrast, correlations indicate a high CH₃Cl 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 CH₃Cl emission in South Asia to be 134±23 Gg Cl yr⁻¹, which is higher than a previous estimate due to the higher CH₃Cl to CO ratio observed in this study.

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