Methane variations observed in the upper troposphere/lowermost stratosphere over the Eurasian Continent and their interpretation based on the carbon and hydrogen isotopic ratio

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Aircraft observation campaigns over northern high latitudes have been conducted several times to elucidate spatial and temporal variations of CH_4 concentration and their sources on the surface. However, simultaneous observations of CH_4 concentration and its isotopic ratios ($\delta^{13}C$ and δD) in the upper troposphere/lowermost stratosphere (UT/LMS) are quite limited, although such observations provide crucial information for quantifying contributions of sources/sinks of CH_4 to its atmospheric variations. In this study, we present spatiotemporal variations of CH_4, $\delta^{13}C$ and δD using monthly on-board commercial airliners in UT/LMS over the Eurasian continent from April 2012. In the LMS, CH_4 and $\delta^{13}C$, δD showed clear aniti-phase seasonal variations; seasonal maximum (minimum) of the CH_4 concentration ($\delta^{13}C$, δD) was found in November to January and seasonal minimum (maximum) was in March to May. The observed seasonal variations can be explained by effective flushing of the LMS air with the tropospheric air (high CH_4 and low $\delta^{13}C$ and δD) from summer to autumn, and by subsidence of the deeper stratospheric air (low CH_4 and high $\delta^{13}C$ and δD) from winter to spring. Backward trajectory analyses with ERA-Interim reanalysis data were conducted for all air samples. By classifying the results into four seasons, it was found in each season that the correlation of $\delta^{13}C$ or δD with potential velocity (PV) at each sampling point is improved by employing the PV values at locations where each air mass is suited 2-3 weeks before. Such an improvement is probably made, reflecting that isotopically heavier CH_4 generally originates in higher altitudes and/or latitudes, and CH_4 with lighter isotopes in lower altitudes and/or latitudes. We also examined the chemical pathways of CH_4 destruction in the extratropical UT/LMS based on correlations between CH_4 and $\delta^{13}C$. The enriched $\delta^{13}C$ values with the lower CH_4 concentrations indicate occurrence of reactions of CH_4 with Cl and $O(^1D)$, in addition to the major destruction pathway via OH.

Keywords: methane, isotopic ratio, UT/LMS region, backward trajectory analyses

