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

*Ryo Fujita¹, Shinji Morimoto¹, Yoichi Inai¹, Shuji Aoki¹, Toshinobu Machida², Yousuke Sawa³, Hidekazu Matsueda³, Yosuke Niwa³, Kazuhiro Tsuboi³, Keiich Katsumata², Taku Umezawa², Takakiyo Nakazawa¹

1. Center for Atmospheric and Oceanic Studies, Graduate School of Science, Tohoku University, 2. National Institute for Environmental Studies, 3. Meteorological Research Institute

Aircraft observation campaigns over northern high latitudes have been conducted several times to elucidate spatial and temporal variations of CH₄ concentration and their sources on the surface. However, simultaneous observations of CH₄ concentration and its isotopic ratios (δ¹³C 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 CH₄ to its atmospheric variations. In this study, we present spatiotemporal variations of CH₄, δ¹³C and δD using monthly on-board commercial airliners in UT/LMS over the Eurasian continent from April 2012. In the LMS, CH₄ and δ¹³C, δD showed clear anti-phase seasonal variations; seasonal maximum (minimum) of the CH₄ concentration (δ¹³C, 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 CH₄ and low δ¹³C and δD) from summer to autumn, and by subsidence of the deeper stratospheric air (low CH₄ and high δ¹³C 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 δ¹³C 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 CH₄ generally originates in higher altitudes and/or latitudes, and CH₄ with lighter isotopes in lower altitudes and/or latitudes. We also examined the chemical pathways of CH₄ destruction in the extratropical UT/LMS based on correlations between CH₄ and δ¹³C. The enriched δ¹³C values with the lower CH₄ concentrations indicate occurrence of reactions of CH₄ with Cl and O(¹D), in addition to the major destruction pathway via OH.

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