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Observations of atmospheric CH4 and its carbon and hydrogen stable isotopic ratios in the upper troposphere over the western Pacific

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As part of the CONTRAIL project, Automatic air Sampling Equipment (ASE) has been used on board commercial airliners operated by Japan Airlines (JAL) for atmospheric trace gas measurements. The collection of air samples with ASE was made at least once a month between Sydney or Brisbane, Australia and Tokyo, Japan (Dec. 2005–Mar. 2009), between Tokyo and Guam (Apr. 2009–Mar. 2010), and between Honolulu and Tokyo (Apr.–Sep. 2010). In addition to CH4 concentration, we newly started to analyze the air samples for carbon and hydrogen isotopic ratios (δ13C and δD) of CH4 in Dec. 2006. In the upper troposphere (UT) of the northern hemisphere (NH) (>10°N), the seasonal minimum and maximum of the CH4 concentration appeared in boreal winter–spring and summer, respectively. Influences of the stratospheric air intrusion event were observed in the CH4 concentration, δ13C and δD in the former season. The summertime high CH4 concentrations were observed when the lower tropospheric (LT) CH4 reaches a seasonal minimum in the NH, which implies that air masses with high CH4 concentrations were transported to the NH-UT. Since such high CH4 concentrations were often accompanied by low δ13C and δD values, the air masses would be substantially influenced by isotopically depleted sources. By examining the relationships between δ13C or δD and the CH4 concentration observed in summer, δ13C and δD values of possible sources were estimated to be -51.7$\pm$1.2‰ and -262$\pm$43‰ respectively. These values are higher than those reported for biogenic CH4 but lower than those for CH4 from fossil fuels and biomass burning. To examine where the sources for the high NH-UT CH4 concentrations exit, we made tagged tracer experiments using an atmospheric chemistry transport model, assigning 15 source regions to the model surface. By inspecting the model results, which capture general features of the seasonal CH4 cycles observed in the NH-UT, it was suggested that China and South Asia play important roles in the summertime high CH4 concentrations in the NH-UT. A possible transport process is deep convection associated with Asian summer monsoon. In the UT over the tropics, temporal variations similar to those in the LT (our unpublished data) were observed not only for the CH4 concentration but also for δ13C and δD, presumably due to strong convective activity in that region. The seasonal cycles of the CH4 concentration observed in the SH-UT (>10°S) were similar to those observed in the LT; seasonal CH4 minimum and maximum appeared in austral summer and winter–spring, respectively. On the other hand, temporal variations of δ13C and δD were relatively irregular, compared to those in the LT which vary in almost opposite phase with the CH4 concentration. The CH4 concentration in the SH was higher in the UT than in the LT, which is ascribed to transport of the NH air to the SH through UT.