

Spatial and temporal variations of atmospheric methane concentration and its carbon and hydrogen isotopic ratios in the upper troposphere/lower stratosphere over the Eurasian continent observed by commercial airliner

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Aircraft observation campaigns over the northern high latitudes have been conducted to clarify the spatial and temporal variations of GHGs concentrations and their sources in the surfaces (Sugawara et al., 1996; Tohjima et al., 1997; Nakazawa et al., 1997; Paris et al., 2008); however, systematic time-series observations using aircrafts are still limited (Matsueda et al., 2002; Haszpra et al., 2012; Umezawa et al., 2012). In addition, there is no systematic and simultaneous observation for the atmospheric CH<sub>4</sub> concentrations and their isotopic ratios ( $\delta^{13}\text{C}$ ,  $\delta\text{D}$ ) in the upper troposphere/lower stratosphere over the northern high latitudes; except for a few studies using balloon flights and aircraft observation campaigns (Sugawara et al., 1997; Rice et al., 2003; Röckmann et al., 2011). In this study, we have conducted monthly air sampling on-board a commercial airliner between Europe and Japan from April 2012, and clarified the spatiotemporal variations of CH<sub>4</sub>,  $\delta^{13}\text{C}$  and  $\delta\text{D}$  in the upper troposphere/lower stratosphere over the Eurasian continent.

In the upper troposphere, CH<sub>4</sub> concentrations,  $\delta^{13}\text{C}$  and  $\delta\text{D}$  showed no clear seasonal variations. In the lower stratosphere, on the other hand, CH<sub>4</sub> and  $\delta^{13}\text{C}$ ,  $\delta\text{D}$  showed clear anti-phase seasonal variations; seasonal maximum (minimum) of the CH<sub>4</sub> concentration ( $\delta^{13}\text{C}$ ,  $\delta\text{D}$ ) was found in November to January and seasonal minimum (maximum) was in spring. They can be explained by effective flushing of the lowermost stratospheric air with the tropospheric air in autumn and subsidence of the middle stratospheric air in spring (Sawa et al., 2015). Moreover, compact correlations of CH<sub>4</sub> with respect to  $\delta^{13}\text{C}$  and  $\delta\text{D}$  were found in the lower stratosphere, suggesting the occurrence of reactions of CH<sub>4</sub> not only with OH, but also with Cl and O(<sup>1</sup>D).

Keywords: Methane, carbon and hydrogen isotopic ratios, upper troposphere/lower stratosphere, Eurasian continent