

# Temporal and spatial variations of the mole fraction, carbon and hydrogen isotope ratios of atmospheric methane in the Western Pacific region

\*Yasuda Mahito<sup>1</sup>, Shinji Morimoto<sup>1</sup>, Shuji Aoki<sup>1</sup>, Ryo Fujita<sup>2</sup>, Taku Umezawa<sup>3</sup>

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

To understand atmospheric CH<sub>4</sub> variations in the Western Pacific region, we have conducted systematic observations of the CH<sub>4</sub> mole fractions and its carbon and hydrogen isotope ratios (d<sup>13</sup>C and dD) using air samples collected on board container ships that regularly sail between Japan and Australia/New Zealand. In this study, we analyzed the observation data from 2006 to 2019.

In the temperate zones of both hemispheres, clear seasonal cycles in the CH<sub>4</sub> mole fractions, d<sup>13</sup>C and dD were observed. Using an atmospheric four-box model with prescribed CH<sub>4</sub> sink, we estimated the contribution of CH<sub>4</sub> sources to the seasonal CH<sub>4</sub> change. The results suggest that the seasonal change in the northern hemisphere is caused by the balance between CH<sub>4</sub> release from biogenic CH<sub>4</sub> sources such as wetlands in summer and CH<sub>4</sub> destruction by the reaction with OH radicals that become maximum also in summer. On the other hand, the seasonal CH<sub>4</sub> change in the southern hemisphere is almost determined by the seasonal changes in the amount of the CH<sub>4</sub> destruction by OH radicals.

In the Western Pacific, the CH<sub>4</sub> mole fractions, d<sup>13</sup>C and dD showed clear latitudinal gradients, with CH<sub>4</sub> mole fraction increasing and d<sup>13</sup>C and dD decreasing from south to north throughout a year. These observational facts suggest that there are isotopically lighter CH<sub>4</sub> sources (microbial origin) in the northern hemisphere. The latitudinal distributions of the CH<sub>4</sub> mole fractions and d<sup>13</sup>C obtained in this study were compared with those observed by the National Oceanic and Atmospheric Administration (NOAA/GMD) at background sites far from CH<sub>4</sub> source region (hereafter called as NOAA sites). During the boreal summer, the CH<sub>4</sub> and d<sup>13</sup>C in the Western Pacific showed lower and higher values, respectively, compared to those at the NOAA sites, suggesting that the Western Pacific region could be more strongly affected by the destruction by OH radicals. On the other hand, in the boreal winter, the CH<sub>4</sub> mole fractions were higher in the Western Pacific than the background NOAA sites. This would be due to the prevailing westerly winds in the winter season, which transported the air-mass strongly influenced by land CH<sub>4</sub> sources into the Western Pacific region.

The CH<sub>4</sub> mole fractions observed in the Western Pacific, at Syowa Station(69.0° S, 39.4° E) and Ny-Alesund(78.6° N, 11.6° E) showed increasing trends after 2006. In contrast, d<sup>13</sup>C and dD decreased after 2006. We employed the four-box model to investigate the cause of the long-term CH<sub>4</sub> variability. When the CH<sub>4</sub> destruction rate by reaction with OH radical is fixed after 2006, an increase in CH<sub>4</sub> emission after 2006 and decreases in d<sup>13</sup>C and dD of aggregated CH<sub>4</sub> sources in all latitudinal bands after 2007 are required to reproduce the observed CH<sub>4</sub>, d<sup>13</sup>C and dD values. This suggests that biogenic CH<sub>4</sub> emission has increased since 2006.

Keywords: Methane, Isotopic ratio, Western Pacific