

Relative contributions of different methane emission categories in the continental East Asia inferred from stable carbon isotope measurements at Hateruma Station

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Methane (CH₄) is a radiatively and chemically important atmospheric trace gas. Accurate understanding of its global, regional and national-scale budgets is needed for efficient mitigation actions against climate change under the Paris Agreement. To better understand distribution and magnitude of individual CH₄ emission sectors in East Asia, we started measurements of stable carbon isotope ratio ($\delta^{13}\text{C}$) of atmospheric CH₄ at Hateruma Station, Japan, using a new cryogen-free measurement system (Umezawa et al. 2020). Wintertime atmospheric CH₄ variations at Hateruma Island, located ~220 km east of Taiwan, is characterized by frequent increases due to advection of air mass from the continental East Asia (Tohjima et al. 2014). During winter of 2017 to 2019, we collected series of air samples during 15 high-CH₄ events and analyzed them for $\delta^{13}\text{C}$. By using relationship between CH₄ concentration and $\delta^{13}\text{C}$, $\delta^{13}\text{C}$ signatures contributing to these high-CH₄ events were estimated, which ranged from -57.9 to -37.8 ‰. These values are considered to be result of CH₄ source mixture of different emission sectors in northeastern China, the region which our footprint calculation mainly point to for such high-CH₄ events. Combining FLEXPART-based footprint (<http://db.cger.nies.go.jp/metex/flexcpp.html>) and CH₄ emission map (Ito et al. 2019), we examined contributions of different CH₄ emission sectors on the high-CH₄ events and possible corresponding variations in $\delta^{13}\text{C}$. The comparison showed that the source $\delta^{13}\text{C}$ signatures estimated by observations were in most cases higher than those estimated by simulations. One possible explanation is underestimate of relative contribution of fossil fuel sectors in the simulation. More number of $\delta^{13}\text{C}$ data and more detailed information on $\delta^{13}\text{C}$ signatures of individual CH₄ source sectors are needed for better evaluation of CH₄ emission databases.

Umezawa et al. (2020) A cryogen-free automated measurement system of stable carbon isotope ratio of atmospheric methane, *Journal of the Meteorological Society of Japan*, <https://doi.org/10.2151/jmsj.2020-007>.

Tohjima et al. (2014) Temporal changes in the emissions of CH₄ and CO from China estimated from CH₄/CO₂ and CO/CO₂ correlations observed at Hateruma Island, *Atmos. Chem. Phys.*, <https://doi.org/10.5194/acp-14-1663-2014>.

Ito et al. (2019) Methane budget of East Asia, 1990–2015: A bottom-up evaluation, *Science of the Total Environment*, <https://doi.org/10.1016/j.scitotenv.2019.04.263>.

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