Temporal variations of the global CH_4 sources estimated by mole fraction, carbon and hydrogen isotope ratios of atmospheric CH_4, and an atmospheric chemistry transport model

*藤田 遼[1]、Maksyutov Shamil[2]、森本 真司[1]、青木 周司[1]、中澤 高清[1]、Kim Heon-Sook[3]、梅澤 拓[2]、後藤 大輔[4]、笹川 基樹[2]、町田 敏暢[2]
*Ryo Fujita[1], Shamil Maksyutov[2], Shinji Morimoto[1], Shuji Aoki[1], Takakiyo Nakazawa[1], Heon-Sook Kim[3], Taku Umezawa[2], Daisuke Goto[4], Motoki Sasakawa[2], Toshinobu Machida[2]

1. 東北大学大学院理学研究科大気海洋変動観測研究センター、2. 国立環境研究所、3. 釜山大学、4. 国立極地研究所
1. Center for Atmospheric and Oceanic Studies, Graduate School of Science, Tohoku University, 2. National Institute for Environmental Studies, 3. Pusan National University, 4. National Institute of Polar Research

There still remains a large uncertainty on the relative contributions of CH_4 sources, broadly defined as biogenic (wetlands, rice paddies, ruminants, termites, and landfills), fossil fuel (coal, oil, natural gas, and geological seepage), and biomass burning, to the global CH_4 budget (e.g., Saunois et al., 2016). Carbon and hydrogen isotope ratios of atmospheric CH_4 ($\delta^{13}C$ and δD) allow for better source apportionment and therefore help reduce the uncertainty. In this study, we performed inverse modeling of the observed atmospheric CH_4 mole fractions to estimate surface CH_4 fluxes for 1995–2013, using the NIES global atmospheric tracer transport model (NIES-TM) with a priori CH_4 fluxes and CH_4 sink fields. Forward simulations of the CH_4 mole fraction, $\delta^{13}C$, and δD were further conducted by using the a posteriori (optimized by the inversion) CH_4 fluxes. The $\delta^{13}C$ and δD, thus obtained, were compared to those observed at two polar surface stations, Ny-Ålesund, Svalbard (78˚55'N, 11˚56'E; site code: NAL) and Syowa Station, Antarctica (69˚00'S, 39˚35'E; site code: SYO).

Variations of the atmospheric CH_4 mole fraction simulated using the a posteriori CH_4 fluxes reproduce the observational results fairly well, not only at the sites where the CH_4 data were incorporated into the inversion (104 sites in the global, including NAL and SYO) but also at other independent sites, for example, in the Western Pacific. This suggests that the CH_4 fluxes are well constrained by this inverse modelling at least regional to global scales. However, forward simulations of $\delta^{13}C$ and δD using the a posteriori CH_4 fluxes and the respective isotopic source signatures significantly underestimate the observed $\delta^{13}C$ and δD values globally. It is indicated that the present a posteriori CH_4 fluxes from biogenic sources and those from fossil fuel and/or biomass burning were overestimated and underestimated, respectively. By constraining the CH_4 fluxes by $\delta^{13}C$ and δD values observed at NAL and SYO, the agreements between simulated and observed CH_4, $\delta^{13}C$, and δD are much improved not only at the two sites, but also in the Western Pacific. The relative contributions of biogenic, fossil fuel, and biomass burning sources to the global CH_4 emissions are 62, 30, and 8% for 2003–2012. These values are not in complete agreement with the range of recent top-down estimates, but comparable to an estimate based on global atmospheric $\delta^{13}C$ data (Schwietzke et al., 2016). Our model infers that biogenic and biomass burning CH_4 emissions decreased in the first half of the 2000s and that biogenic CH_4 emissions increased after 2006, which could be responsible for the complicated behavior in global atmospheric CH_4 growth in recent decades, i.e., plateau in the early 2000s and re-rise after 2006.

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