Tracing sources and behaviors of excess methane in the Ise Bay water by using stable isotopes as tracers

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Methane (CH_4) is a major greenhouse gas and can have a significant impact on global climate change. Coastal marine areas are often rich in CH_4 and are known as one of the major sources of atmospheric CH_4, but their sources (where are they formed?) and their behaviors (whether they are released to the atmosphere or oxidized?) have not been fully understood yet. For example, it is known that marine sediments in the coastal areas are reductive and the production of CH_4 proceeds actively, but the river water, which flowing into the coastal areas, are also rich in methane. So the sources and behaviors of the coastal areas are not fully understood. In order to understand the impact of the marine environment on current and future global environmental and climate changes, it is extremely important to elucidate the sources and behaviors of methane in coastal oceans and accurately estimate the fluxes of methane between the oceans and the atmosphere, including time-varying changes.

In this study, we measured the concentration of dissolved CH_4 and stable isotope ratios ($\delta^{13}C$ and δD) of the water column, the interstitial water of sediments in the Ise Bay and Mikawa Bay and the inflowing rivers (Kiso, Nagara and Yahagi River). Using the stable isotope ratio as an indicator, we tried to clarify the origins of CH_4 and its behaviors in the aquatic environments. Sampling was performed in each of 2012, 2013, 2016, 2017 and 2019.

As a result, the maximum concentration of CH_4 (60 to 299 nmol/kg) was observed in the surface water near the estuaries of the main inflow rivers in the Ise Bay, and the supersaturation rate could reach 2700% to 13000%. The isotope ratio of CH_4 ($\delta^{13}C$: -60 to -56 ‰, δD: -215 to -190 ‰) in the surface water of Ise Bay is almost as the same as the isotope ratio of CH_4 ($\delta^{13}C$: -57 ‰, δD: -206 ‰) in the inflow rivers. Therefore, the maximum concentration of CH_4 which was observed in the surface layer of the Ise Bay is derived from the inflowing rivers, and the oxidation of CH_4 in the water column is negligible. Consequently, the concentration of supersaturated CH_4 that has flowed in from the river has decreased due to either dilution or release to the atmosphere. Since the CH_4 emission flux to the atmosphere of the Ise Bay was calculated to be 3.2 to 52.3 $\times 10^3$ mol / d, which is almost consistent with the CH_4 inflow flux from the inflowing river to Ise Bay (4.3 to 73.4 $\times 10^3$ mol / d). That is to say, the most of the CH_4 flowing from the river into Ise Bay was released to the atmosphere immediately after flowing. In conclusion, it is important to clarify the origin and behavior of CH_4 in inflowing river water in considering the amount of CH_4 released into the atmosphere in coastal areas.

Keywords: coastal marine areas, methane, material circulation, biochemistry