Clarifying sources of methane enriched in oxic water columns by using stable carbon and hydrogen isotopes as tracers

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Although methane is one of the representative greenhouse gases in atmosphere, there are still many uncertainties on the emission sources on earth surface. The hydrosphere is the major source of atmospheric methane, and thus slight changes on the hydrospheric environment will cause a drastic change on the atmospheric methane concentration and thus on global warming. Consequently, we must increase our knowledge on the sources of methane enriched in hydrosphere, together with the behaviors of methane in hydrosphere.

Generally, methane is produced in anoxic environments, while being decomposed in oxic environments. Nevertheless, supersaturation of methane in water relative to that in equilibrium with atmospheric concentration have been frequently found in oxic oceans and lakes. Various studies had been done in past to clarify the reasons of this contradiction, being called as 'methane paradox'.

So as to clarify the sources and behaviors of methane in such oxic hydrosphere, the $\delta^{13}C$ values of methane have been frequently used as a tracer in past studies, while the studies using δD values of methane enriched in oxic hydrosphere were limited in past. By using both $\delta^{13}C$ and δD values of methane simultaneously as tracers, the changes in the values of $\delta^{13}C$ and δD during oxidation (partial removal of methane in oxic water column) can be corrected. In this study, we used stable isotopes ($\delta^{13}C$ and δD) of dissolved methane in the water columns of Lake Biwa, Ise Bay, and Mikawa Bay as the tracers to specify the sources of methane supersaturated in the oxic water columns and to clarify the behavior of methane in the oxic water columns. Besides, I defined a new parameter $\Delta(2,13)(= \delta D - 11 \times \delta^{13}C)$ in which variations in both $\delta^{13}C$ and δD values during methane oxidation had been corrected and used this parameter as the tracer to differentiate the sources for each methane dissolved in the water columns. To clarify the sources in the studied fields, I assumed following three methane sources as the possible sources of methane supersaturated in the oxic water columns, (1) river input, (2) lake-floor (or seafloor) sediments, and (3) sinking particles in the water columns, and determined the stable isotope ratios of methane supplied from each. Then, I compared them with those in the water columns, and concluded that methane supersaturated in Lake Biwa and Ise Bay surface water columns is likely to be supplied via river. On the other hand, it was impossible to assume river input as the major source of methane in Mikawa Bay water showing the δD values (-327~-287‰) substantially lower than those dissolved in river input (-175‰). I concluded that the seafloor sediments should be highly responsible for the source of methane enriched in the Mikawa Bay water columns.

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