Concentration and isotopic ratio of dissolved methane in sediment interstitial waters and seawater, Japan sea

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Sea-going geochemical surveys were performed in 2004 through 2008 in the Joetsu basin, eastern margin of Japan Sea. We report the methane concentration and the delta $^{13}C_{CH4}$ of dissolved methane in sediment interstitial water and seawater obtained for the last five years.

Sediment samples were retrieved 68 times by piston corer and 50 times by push corer attached with ROV. Seawater samples were retrieved 25 times by 24-port rosette multi sampler of Niskin bottles together with a CTD and 78 times by Niskin bottles attached with ROV. Dissolved methane in sediment interstitial water and seawater were analyzed to understand the source of methane plumes and its influence to marine and atmospheric environment.

Delta $^{13}C_{CH4}$ in the area has a wide range from -100 to -30 permil. This is because that the methane in this region has two sources, one is from thermogenic and the other is biogenic.

The delta$^{13}C_{CH4}$ values of methane hydrate is in the range of thermogenic methane. The delta $^{13}C_{CH4}$ values of methane hydrate retrieved from the Umitaka spur are -30˜-40 permil, while those from the Joetsu knoll are -43˜-51 permil. This seems to suggest a difference in the mixing ratio of thermogenic and biogenic, and also the methane flux in Umitaka spur is relatively stronger than that in Joetsu knoll.

Dissolved methane in sediment interstitial waters are enriched in delta $^{13}C_{CH4}$, centered at around -55 permil delta $^{13}C_{CH4}$near the methane plume sites, while non-plume sites are depleted in delta $^{13}C_{CH4}$, averaging -88 permil. These results signify the source of methane plumes is thermogenic and biogenic methane is distributed widely through out sediments in this area.

Delta $^{13}C_{CH4}$ upper SMI (sulfate-methane interface) become lighter(5˜10 permil) than that below SMI. The difference may reflect the fractionation by microbial consumption of isotopically light methane.

The methane concentrations in the seawater column of the Joetsu Basin were higher than that in reference sites. High concentration anomaly of methane in the seawater was observed in the plume sites. Visual observations were performed at the seafloor by ROV. During the ROV surveys, we discovered the discharge of methane hydrate-coated methane bubbles and Methane hydrates in the sea floor sediments. This phenomenon suggests that methane bubbles can be preserved well in seawater column and reach up to the shallow level. High methane concentrations of seawater are well explained by the fact that methane-hydrate-coated bubbles are to be large amounts of methane to shallow water. Carbon isotopic signatures of seawaters strongly suggest this model.