

MIS027-08

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## Molecular and isotopic compositions of gas hydrate-bound hydrocarbons retrieved from off Joetsu, Japan Sea

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Gas hydrates are crystalline clathrate compounds composed of water and gas molecules that are stable at low temperature, high partial pressure of each gas component, and high gas concentration. Recently, natural gas hydrates were obtained at the Umitaka Spur and the Joetsu Knoll on the eastern margin of the Japan Sea (Matsumoto *et al.*, 2009; Hiruta *et al.*, 2009). We investigated the molecular and stable isotope compositions of hydrate-bound gas and dissolved gas in pore water in sediments of these areas. The purpose of this study is to clarify the gas origin and to understand the migration system of the shallow gas hydrate accumulation in this area. Hydrate-bearing sediment cores were retrieved during the cruise onboard *R/V Marion Dufresne* in June 2010. The Calypso piston coring system enabled us to take long sediment cores (up to 40m). Hydrate-bound hydrocarbons are primarily thermogenic at the Umitaka spur ( $C_1$   $\delta^{13}C$ : -36 permil-VPDB,  $C_1$   $\delta D$ : -165 permil-VSMOW), on the contrary, those at the Joetsu knoll (about 30m depth from the sea floor) partly contain microbial methane because  $^{13}C$  and deuterium are both depleted ( $C_1$   $\delta^{13}C$ : -55 permil-VPDB,  $C_1$   $\delta D$ : -194 permil-VSMOW) and close to the field of microbial methane via  $CO_2$  reduction. Depth profiles of methane in sediment showed the minimum depth in  $\delta^{13}C$ , corresponded to the SMI depth where hydrogen sulfide was concentrated. Methane concentration in sediment increased drastically beneath the depth of SMI.  $\delta^{13}C$  of methane increased slightly along with depth, whereas  $\delta D$  of methane decreased. Thermogenic ethane and propane are detected both in hydrate-bound gas and dissolved gas in pore water. Heavy propane ( $\delta^{13}C$ : +5 permil-VPDB) indicated its microbial oxidation. The molecular composition of thermogenic ethane in hydrocarbons was relatively high at the gas hydrate sites, whereas microbial ethane is dominant in the peripheral area. High concentration of neopentane (2,2-dimethylpropane) was detected in the dissolved gas around the depths of gas hydrate at the both sites of Umitaka Spur and Joetsu Knoll. Since neopentane cannot be engaged to the crystallographic structure I of gas hydrate, it might be discharged from the hydrate crystal at the formation process and remained in the sediment.

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Hiruta, A., G.T. Snyder, H. Tomaru, R. Matsumoto (2009) Geochemical constraints for the formation and dissociation of gas hydrate in an area of high methane flux, eastern margin of the Japan Sea. *Earth Planet. Sci. Lett.*, **279**, 326-339, doi:10.1016/j.epsl.2009.01.015.

Matsumoto, R., and 28 others (2009) Formation and collapse of gas hydrate deposits in high methane flux area of the Joetsu Basin, eastern margin of Japan Sea. *J. Geogr.*, **118**(1), 43-71.

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