Izura carbonate concretions showing enormous natural gas seep in the Lower Miocene

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Enormous amount of carbonate concretions of the Lower Miocene Kokozura Formation, the Takaku Group, are cropped out along the Izura coast, north Ibaraki, Japan. The Izura carbonate concretions are stratified and distributed widely in the Kokozura Formation. Fossils of symbiotic bivalves at cold methane seeps often occur with the concretions, suggesting their methane-seep origin. However, small-scale methane seeps cannot be responsible for the formation of Izura carbonate concretions, since their total volume is so enourmous up to more than 6.0×10^6 m^3.

The $\delta^{13}C$ and $\delta^{18}O$ values of carbonates are from −30 to −3‰ and from −5 to +2‰, respectively, showing the isotopic similarity to those related to thermogenic methane. The major component in residual gas obtained by the pulvalization of carbonate concretion is CO_2, that constitutes about 90 vol%. CH_4, C_2H_6 and C_3H_8 are also detected significantly. The comparatively higher abundance of C_2H_6 and C_3H_8 suggests the thermogenic origin of hydrocarbon gases. The $\delta^{13}C$ and δD values of CH_4, are from −57‰ to −42‰, and from −209‰ to −111‰, respectively, suggesting that CH_4, is thermogenic and mainly derived from marine sapropelic organic matter. The $\delta^{13}C$ values of CO_2 from −39 to −15‰ are generally higher than those of $CaCO_3$. The difference between $\delta^{13}C$ values of CO_2 in residual gas and $CaCO_3$ appear to be nearly constant within approximaterly 10±1.7‰.

Izura carbonate concretions can be morphologically classifed into three types, which are located in the upper, middle and lower parts of Kokozura Formation. Carbonate concretions in the upper part are characterized by comparatively lower CO_2 concentrations and larger $\delta^{13}C$ values of CO_2. Archaeal biomarker pentamethylicosane (PMI) with $\delta^{13}C$ values from −104 to −44‰ was detected mainly from the upper part. These observations show the significant generation and contribution of biogenic methane in the later stage of carbonate formation, while the formation of carbonate concretions in the middle and lower parts are much influenced by thermogenic gas. The widespread Izura carbonate concretions are geological evidence of enormous natural gas seep in the Lower Miocene.