

Biogeochemical processes in gas hydrate-bearing mud volcano sediments from the Kumano forearc basin, Japan

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We investigated biogeochemical processes in mud-volcano subsurface sediments down to 20 meters from the summit, obtained from the Kumano forearc basin in the Nankai Trough during the CK09-01 D/V Chikyu training cruise in 2009.

The cored sediments contained several methane hydrates. The stable isotopic compositions of the pore fluid samples exhibit ¹⁸O-enrichment and D-depletion in proportion to the depletion of the chlorinity indicating that a mixing between seawater and the fresh water components derived from the dehydrate reaction of clay minerals at 60-160°C previously occurred in the deeply buried sedimentary layer. In contrast, hydrate water samples are strongly enriched in deuterium and slightly deviated from Cl- vs. oxygen isotopic composition trend indicating the pore fluid freshening is not governed by gas hydrate dissociation. High B (up to 13094 micoro-M) and Li (up to 188 micoro-M) concentrations in the pore fluid suggest the leaching from clay minerals at >150°C, and indicate that the fluid in the mud volcano was supplied from the accretionary prism (>4000 mbsf) beneath the forearc sediments probably through faults.

The carbon isotopic composition of dissolved inorganic carbon increased with the coring depth, reaching +40 per mil at 3 meters below the seafloor (mbsf). The highly ¹³C-enriched values are possibly due to strong microbial reduction of DIC to ¹²C-enriched products. The hydrogen isotopic composition of methane (ca. -180 permil) and magnitude of the carbon isotopic fractionation between DIC and methane (ca. 76 per mil) bellow 3 mbsf suggest the significant contribution of hydrogenotrophic methanogenesis as the source of methane. The carbon isotopic composition value of acetate was appeared to increase with the sediment depth (from -41 to -22 per mil), synchronous to the increase of carbon isotopic composition of DIC. The significant isotopic fractionation between DIC and acetate (ca. 54 per mil) indicates that the principal process producing acetate is homo-acetogenesis via the reductive acetyl-CoA pathway. Radioactive tracer experiments exhibited relatively high activities of homo-acetogenesis (14~34,900 pmol/cm³/day) compared to that of hydrogenotrophic methanogenesis (0.6~128 pmol/cm³/day). In ordinary marine sediments, homo-acetogenesis from H₂ and CO₂ is thermodynamically inhibited because H₂ concentrations are maintained at low levels less than several nM. However, the homo-acetogenesis was thermodynamically favorable in the cored sediments because of the high concentration of H₂ (>560nM). These results showed that homo-acetogenesis is dominant in the sediments around the summit of the mud volcano, while active hydrogenotrophic methanogenesis has been occurred in a deeper depth, and most portion of methane formed methane hydrates was supplied from the deep active methanogenic zone.

Keywords: Mud volcano, Methane hydrate, Deep origin fluid, homo-acetogenesis, methanogenesis, CO₂ reduction