Hydrogen and carbon isotopic compositions of Archaeal biomarkers in a marine gas hydrate deposit

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Archaea play an important role in carbon cycle via early diagenesis within anaerobic deep marine sediment. Especially, methanogenic Archaea produces methane which forms a vast quantity of gas hydrate in continental margin accretionary sediments. Clarification of methanogen's activity and biomass is required to understand mechanisms of gas hydrate formation, which potentially contribute to energy resource and global warming. However, methanogen's activity is less undersood than anaerobic methane-oxidizing Archaeal (AOM Archaea) activity. In the present work, we developed hydrogen isotopic analysis for Archaeal biphytane released from ether-bond cleavage of archaeal tetraether lipids. This technique provides much information from stable isotopes (δ13C and δD) in archaeal biomarkers. We applied this technique to gas hydrate-bearing sediment within the northern Cascadia margin. Sediment cores were collected from two IODP sites characterized by deep (120-130 mbsf) gas hydrate accumulation (U1327) and active methane seep with shallow (0-40 mbsf) gas hydrate accumulation (U1328). Sediments from both sites contained much amount of Archaeal biomarkers near the gas hydrate layers and estimated bottom simulating reflectors (BSRs): acyclic and cyclic (containing two or three pentane rings) biphytane diols (BPD[0], BPD[2] and BPD[3]), 0.29-474 μg/gCorg; Biphytanes (BP[0] to BP[3]), 0.07-1460 μg/gCorg. Depth profiles of these biomarkers are consistent with each other, suggesting the same origin. Carbon isotopic composition of BPs is mostly about -20‰ within the entire core at both sites. This value is similar to that of TOC, suggesting a heterotrophic origin. However, in some sediment horizons, commonly below the gas hydrate layers, BP[1] is significantly depleted in 13C (-50 to -40‰). This 13C depletion of BP[1] is attributed to the contribution of methanogen or AOM Archaea . Especially, a reported carbon isotopic fractionation factor (α = 1.046) during lipid synthesis by Methanosarcina barkeri (Londry et al, 2008) is consistent with that estimated from in situ carbon isotopic composition of CO2 and BP[1] (α = 1.043) at 142 mbsf. In addition, hydrogen isotopic composition of the BP[1] (-172‰) is similar to that of methane (-160‰). Hence, the occurrence of these isotopically light BP[1] could indicate methanogen activity, or methanogenesis zone.