Distribution and isotopic compositions of Archaeal membrane lipids within methane hydrate-bearing sediment (IODP exp311)

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Introduction

Investigation of bacterial activities by scientific drilling is important for better understanding of deep biosphere and biophile element cycles within subsurface of the Earth. Chemical structures and isotopic compositions of bacterial lipid biomarkers provide key information on the bacterial biomass and metabolic activities. In the present study, lipid biomarkers of Archaea, which occupies significant biomass in deep sediment, are focused to clarify the bacterial activities within drilled sediment core of Cascadia margin gas hydrate zone, offshore Vancouver Island.

Materials & methods

Sediments used in this study were collected from Site U1327 and U1328 (maximum drilled depth: 300 mbsf). Dried and powdered sediment sample was extracted with organic solvents. Alcohol fraction was separated from the extract by silica gel chromatography. Lipid biomarkers in the alcohol fraction were identified and quantified using a gas chromatograph-mass spectrometer (GC-MS) and a GC. Carbon and hydrogen isotopic compositions were analyzed using a GC/isotope ratio mass spectrometer (IRMS).

Results and discussions

Archaeal biomarkers in the alcohol fraction including archaeol, acyclic and cyclic (containing three pentane rings) biphytane diols (BPD[0], BPD[3-I] and BPD[3-II]) occur in sediments from U1328. Biphytanes (BP[0] to BP[3]) derived from membrane tetraether lipids were also obtained after ether-bond cleavage of the alcohol fraction. Concentrations of BPs (e.g. BP[0]: 14.6-458 ug/gC_{org}) were approximately 10 times higher than those of BPDs (e.g. BPD[0]: 0.7-295 ug/gC_{org}). Depth profiles of these biomarkers are consistent with each other, suggesting the same origin. In addition, a positive correlation (e.g. sigma (BP): r^2 = 0.86) between concentrations of total organic carbon (TOC) and the biomarkers would reflect a heterotrophic origin. One the other hand, there is no correlation between them in Site U1327, although the concentrations of the BPDs (e.g. BPD[0]: 3.0-228.7 ug/gC_{org}) are similar to those of SiteU1328. Increase of the biomarkers above and below bottom simulating reflectors (BSRs) and the methane hydrate accumulations inferred from wire line logging and infrared images indicate that high microbial activities in these regions would contribute to the methane generation.

Carbon isotopic composition of BPs is mostly about -20 permil within the entire core of Site U1328. This value is similar to that of TOC, suggesting a heterotrophic origin. This interpretation is consistent with that from the correlation between concentration of BPs and TOC. However, in some sediment horizons (42 and 142 mbsf), BP[1] is significantly depleted in ^{13}C (~-40 permil). This ^{13}C depletion of BP[1] is attributable to the contribution of methanogen or anaerobic methane oxidizing Archaea. Especially, a reported carbon isotopic fractionation factor (alpha = 1.046) during lipid synthesis by *Methanosarcina barkeri* (Londry et al, 2008) is consistent with that estimated from *in situ* carbon isotopic composition of CO_2 and BP[1] (alpha = 1.043) at 142 mbsf. In addition, hydrogen isotopic composition of the BP[1] (-172permil) is similar to that of methane (-160permil). Hence, the occurrence of these isotopically light BP[1] could indicate methanogen activity, or methanogenesis zone.

Reference

1. Londry et al.,(2008), Organic Geochemistry 39,608-621 .