Data report: carbon isotope compositions of methane in void gas samples from IODP Expedition 315 Site C0001, Nankai Trough, offshore Japan¹

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Abstract

We investigated carbon isotope compositions of methane and molecular compositions in void gas samples from Integrated Ocean Drilling Program Expedition 315 Site C0001. Methane is the predominant gas with traces of ethane and no heavier hydrocarbons detected. The methane/ethane (C_1/C_2) ratios of all samples are higher than 5500. The δ^{13} C values of methane range from -74.9‰ to -73.9‰. These data indicate that methane found in Hole C0001F is predominantly of microbial origin.

Introduction

Nankai Trough is one of the most extensively studied subduction zones in the world and is known as an area of active subsurface fluid circulation. The widely observed bottom-simulating reflectors suggest the presence of gas hydrates (Matsumoto et al., 2004). Many fractures and faults developed within the accretionary prism are most likely providing conduits of nutrient and energy substrates that potentially support indigenous microbial population and activities in the subsurface environment (Saito and Suzuki, 2007). Methane production derived from gas hydrate dissociation and migration of thermogenic gas from deeper depths are also expected. The carbon isotope compositions of methane in combination with gas molecular compositions have been widely used for the characterization of hydrocarbon gas origin (Bernard et al., 1977; Rice and Claypool, 1981). In this study, we measured the carbon isotope compositions of methane and molecular compositions in void gas samples from Site C0001, which will provide critical clues to the origin and migration paths of the hydrocarbon gas in this area.

Materials and methods

Six void gas samples were taken from 156 to 240 meters below seafloor (mbsf) in Hole C0001F. The sections for sampling void gas were restricted because the release of void gas from the core liner was preferred in order to avoid explosion caused by gas expansion. Void gas samples were collected directly through the core liner with a plastic syringe and were transferred into 65 mL vials that were vacuumed beforehand, sealed with rubber stoppers, and crimp-sealed with aluminum caps. Void gas samples

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were analyzed for volatile hydrocarbon contents on board following the method of Pimmel and Claypool (2001). After analyzing the samples, they were preserved at 4°C for later measurement in the onshore laboratory at Hokkaido University, Japan.

In the shore laboratory, concentrations and carbon isotope compositions of methane in void gas samples were measured by gas chromatograph/combustion/isotope ratio mass spectrometer (HP6850GC/Finnigan MAT 252) with the original purge-and-trap system following the method of Hirota et al. (2010). The analytical precision in the determination of the δ^{13} C ([{($^{13}C/^{12}C_{sample}$)/($^{13}C/^{12}C_{PDB}$)] – 1] × 1000) of methane through a single analysis is estimated to be better than 0.07‰ when more than 5.5 nM of methane is introduced.

In the onshore laboratory, we also measured concentration and carbon isotope compositions of methane in headspace gas samples, but these samples were apparently affected by leakage of methane from sample vials or microbial oxidation of methane in the sample and are not discussed further.

Results

Concentrations and carbon isotope compositions of methane and methane/ethane (C_1/C_2) ratios in the void gas samples measured on board and onshore together with headspace gas data measured on board are shown in Table **T1** and Figure **F1** (see also Table **T20** in the "Expedition 315 Site CO001" chapter [Expedition 315 Scientists, 2009]) Methane is the predominant hydrocarbon present in all samples. Concentrations of methane in the void gas samples are 1 order of magnitude higher than those of the maximum range in the headspace gas samples. The C_1/C_2 ratio is >1000 in most samples, suggesting that methane found at Site C0001 is predominantly of microbial origin (Bernard et al., 1977; Rice and Claypool, 1981).

Carbon isotope compositions of methane in void gas samples measured in the onshore laboratory are shown in Table T1 and Figure F1. The δ^{13} C values of methane in void gas samples range from -74.9% to -73.9% with an average of -74.2%, which is consistent with their microbial origin (Fig. F2) (Schoell, 1980; Whiticar et al., 1986).

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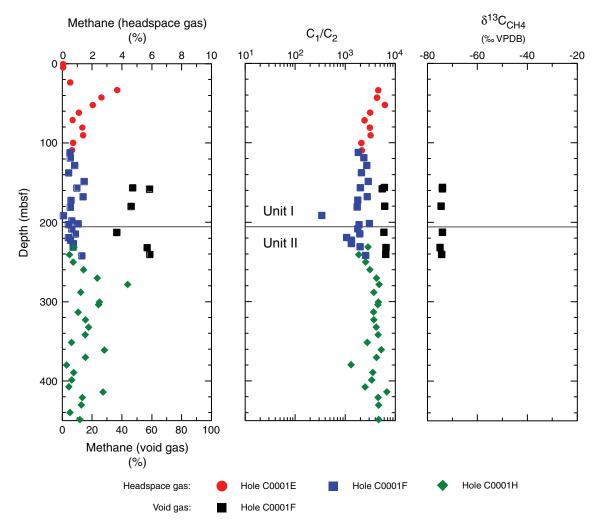
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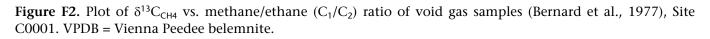
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Figure F1. Plots of methane concentrations and methane/ethane (C_1/C_2) ratios in headspace gas and void gas samples (onboard analysis) and carbon isotope compositions of methane in void gas samples (onshore analysis), Site C0001. VPDB = Vienna Peedee belemnite.







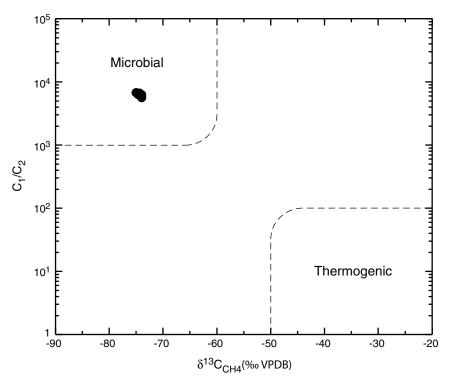




Table T1. Concentrations, carbon isotope compositions, and C_1/C_2 ratios in void gas samples, Site C0001.

Core, section, interval (cm)	Depth (mbsf)	Onboard analysis		Onshore analysis
		Methane (%)	C ₁ /C ₂	δ ¹³ C _{CH4} (‰ VPDB)
315-C0001F-				
6H-1, 104	156.5	47.2	6106	-73.9
6H-3, 26	158.2	58.6	5549	-73.9
9H-1, 19	179.9	46.1	6127	-74.5
14H-6, 4	212.9	36.4	5966	-74.0
20X-2, 78.5	232.0	56.8	6587	-74.9
21X-1, 122	240.6	58.6	6494	-74.3

VPDB = Vienna Peedee belemnite.

