Surveys of gas plumes off Hokkaido, Sea of Okhotsk

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Gas hydrates (GH) are attracting attention as a future energy resource, with projects aimed at their utilization under way in various countries. In Japan, the MH21 R&D project in the Nankai Trough region has entered its production test stage. On the other hand, in 1995, when Japan pioneered a project for the utilization of GH, clear bottom-simulating reflectors (BSR) were confirmed also at the Kitami-Yamato Bank in the Okhotsk Sea offshore of Abashiri, indicating the possible existence of GH there. In addition to this, seismic survey records collected by the National Institute of Advanced Industrial Science and Technology (AIST) during a cruise for their GH01 project in 2001 also confirmed noticeable BSR.
On the zone which GH exists in stability by temperature and pressure conditions (HSZ: Hydrate Stability Zone), GH existed in the upper part of HSZ is called shallow type GH, and that existed in the lower part is called deep type GH. This deep type GH is observed in a zone immediately above a BSR. Therefore, observation of BSR becomes an index of deep type GH existence. This BSR is confirmed in off Okushiri Island, off Hidaka, off Tokachi and off Abashiri in the around of Hokkaido Island.
On the other hand, shallow type GH is found in sediments of the surface layer or the exposed seafloor. That have been recovered off Abashiri in the Okhotsk Sea in the around of Hokkaido. In the area existed shallow type GH, gas plumes are also observed by echo sounder. Therefore, observation of gas plume becomes an index of shallow type GH existence.
In this study, to clarify the distribution of gas plume off Hokkaido in the Okhotsk Sea, a survey using the Oshoro-Maru, the research training ship of the Hokkaido University, was conducted in November 2015, and analysis of the data of quantitative echo sounder that was acquired in the past by research ships of the Hokkaido Research Organization (ORC). As a result, including past surveys, the number of locations where gas plumes have been confirmed is about 300 in the Okhotsk Sea offshore of Hokkaido.

Keywords: Gas hydrate, Gas plume, Marine sediment

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Characteristics of structure I natural gas hydrate encaged thermogenic methane

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Crystallographic structures of natural gas hydrate are usually either structure I or structure II. The latter can encage larger hydrocarbons, for example, propane, isobutane, n-butane, and neopentane. Because the origin of these molecules is thermogenic, methane ascending with them from deeper sediment layer is also thermogenic. Hydrate-bound thermogenic methane has been reported in the world (Gulf of Mexico, offshore Vancouver Island, Caspian Sea, etc.). C1/C2+ of guest gas in these sites are less than 10, indicating that compositions of ethane and propane in hydrate-bound hydrocarbons are in an order of several percent. Therefore, crystallographic structure of gas hydrate composed of thermogenic gas is primarily the cubic structure II.
On the other hand, the structure I gas hydrates retrieved off Joetsu contained thermogenic methane (delta 13C > -50 permil, e.g. Lu et al., 2011). C1/C2+ of hydrate-bound hydrocarbons was more than 2,000, whereas the maximum value of methane delta 13C was -35permil (Hachikubo et al., 2015). It is still unknown how higher hydrocarbons reduced in the sediment. Gas hydrates have been discovered at the southwestern Sakhalin Island in the cruises of LV59 (2012), LV62 (2013), LV67 (2014), and LV70 (2015) on board R/V Akademik M. A. Lavrentyev in the framework of Sakhalin Slope Gas Hydrate (SSGH) project. We reported in the last JpGU meeting that hydrate-bound gas contained 13C-rich methane, suggesting thermogenic origin. In this study, we focus on the gas hydrates of the cubic structure I containing thermogenic methane retrieved from the Tatar Trough, off Sakhalin Island, and compare with those retrieved off Joetsu.
We obtained hydrate crystals from sediment cores, and stored them in liquid nitrogen. Raman spectra of the crystal showed two peaks of C-H stretching mode, correspond to methane molecules in large and small cages of the structure I, and small peaks of hydrogen sulfide were also detected. We also obtained hydrate-bound gas on board and measured their molecular and stable isotope compositions. C1/C2+ of hydrate-bound hydrocarbons ranged between 200 and 800, suggesting that contribution of thermogenic C2+ was low. However, delta 13C and delta D of hydrate-bound methane distributed from -48permil to -42permil and from -200 permil to -170 permil, respectively. According to an empirical classification of the methane stable isotopes (delta 13C and delta D; Whiticar, 1999), hydrate-bound methane obtained at the Tatarsky Trough was mainly thermogenic origin. Characteristics of hydrate-bound methane is similar to those obtained off Joetsu. delta 13C of CO2 in sediment gases was high (+20 permil), suggesting interaction between methane and CO2 through microbial activity.
We appreciate the support of the crew onboard R/V Lavrentyev during the LV59, LV62, LV67, and LV70 cruises off Sakhalin Island. This study was supported by the Grant-in-Aid for Scientific Research (B) 26303021 of the Japan Society for the Promotion of Science (JSPS).
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Keywords: hydrate, methane, Sakhalin Island

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Controls of mud diapirism on gas hydrate systems in the Lower Fangliao Basin, offshore southwest Taiwan

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The accretionary wedge of the incipient arc-continent zone of Taiwan has been identified rich in gas hydrates as inferred from reflection seismic data. We employed 2D and 3D seismic data to understand the interplay of structural development, especially mud diapirism, and gas hydrate formation in the Lower Fangliao Basin, a slope basin situated in the upper accretionary wedge. Seismic reflection data show mud tectonics exerts controls on the formation of bottom-simulating reflectors (BSRs) and the distribution of gas hydrates. Mud diapirs can be recognized on seismic profile in terms of acoustically transparent piercement structures. The formation of mud diapirs in the study area is ascribed to overpressured sedimentary layers, compressional tectonic forces, and gas-bearing fluids. The sedimentary strata on both sides of a mud diapir exhibit dragging and onlapping features due to uplifting of the diapir. Both normal strata and growth strata are discernable, suggesting the dynamics of mud diapiric development through time.
The interplay of mud diapirism, sediment dispersal, and regional convergent tectonics to the gas hydrate system is echoed from seismic facies in the study area. Five seismic facies have been observed, including uneven-truncated, stratified-parallel, chaotic-transparent, strong-parallel-reflection, reflection-free facies and are deciphered as seafloor/erosional surface, hemipelagic sediments, mass transport deposits (MTDs), sandy turbidite sediments, and mud diapirs, respectively. The gas hydrate and free-gas zonation within gas hydrate stability zone (GHSZ) is characterized by (1) high amplitude reflections with the analogous phase of seafloor indicating possible porous turbidite sands reservoir; (2) BSRs showing polarity reversal to that of seafloor, suggesting higher impedance gas-hydrate charged sands overlying lower impedance sands with free gas; (3) those strong reflections in the fault zones as gas-bearing fluid conduits; (4) strong reflections on the sides of mud diapirs (e.g. flank drags) and above buried mud diapir demonstrating the presence of gas hydrates, and (5) high amplitude reflections dragging on diapiric flanks with reversal phase of seafloor indicating free-gas charged sands abutting mud diapirs. Vertical venting governed by mud tectonics is the key to inducing thermogenic gas seepages. When such structure is absent, biogenic gas could be the alternative source for free gas or gas hydrate accumulations. Upward mud intrusion contributes to initiation of brittle deformation for deeply buried gas migration pathways. The low-permeability nature of mud diapirs promotes prominent traps for free gas or gas hydrate preservation along the diapiric flank. Due to its high thermal conductivity, active mud diapirs may act as dewatering catalyst for hitherto preserved gas hydrates, allowing dissociated gas to be accumulated, even within GHSZ.

Keywords: gas hydrates, mud diapirs, seismic reflection, accretionary wedge, Taiwan

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Resource assessment of shallow gas hydrate of Japan Sea: Overview and Preliminary Results of 2013-2015 METI Project

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It had been well documented that shallow gas hydrates occur as a nodular to bedded form of a few cm to a few meters in the hydrate mound, the upper part of gas chimney structure (acoustic blanking zone) along the eastern margin of Japan Sea (e.g., Matsumoto et al., 2005, 2012). On the basis of the Basic Act on Ocean Policy (approved by the Cabinet on April 2013), METI launched 3 years project to assess the resource potential of the shallow gas hydrates in Japan Sea. Gas hydrate laboratory of MU has conducted a regional bathymetric and geologic survey, drilling survey, environmental assessment survey etc. as AIST's sub-commissioned project. Regional survey has focused on regional mapping of potential hydrate-bearing structures by means of MBES and SBP systems along the eastern margin of Japan Sea and around Hokkaido Island, and confirmed 1742 hydrate mounds with gas chimney in 3 years. LWD drilling and pressure coring on selected hydrate mounds and gas chimneys successfully identified gas hydrate concentration zones characterized by high sonic velocity, high resistivity, low natural gamma ray etc, and finally recovered hydrate-bearing sediments including more than several meters thick, massive and bedded pure gas hydrates for the first time in the world. A number of ROV dives observed sea floor manifestations of methane seeps, outcrops of a few meter thick hydrate beds and crater-like depressions formed by a collapse of massive hydrates. Long term monitoring of benthic environments have been also performed under this project. Preliminary results as to the resource assessment will be discussed in the presentation. This study was conducted as a part of the shallow methane hydrate exploration project of METI. We express sincere thanks to personnel from the AIST, JOGMEC and allied Universities and Institutes for their participation in long term sea-going expedition and laboratory experiments.

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Keywords: shallow gas hydrates, Japan Sea, 2015 coring campaign

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Identification of shallow methane hydrate concentrated intervals by LWD within the gas chimney-mound structure, eastern margin of Japan Sea.

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Two LWD, Logging While Drilling, survey cruises were carried out in 2014 and 2015 summer season in the eastern margin of Japan Sea in order to explore the geological characters of the shallow methane gas hydrate within the gas chimney-mound structure, which is well developed in this area. 2014 summer cruise hires natural gamma ray, resistivity, sonic and CMR loggings. Clear anomalies of low natural gamma, high resistivity, high sonic velocity, low NMR porosity were detected at the gas chimney structures in off-Joetsu and Mogami Trough area. Thus, it was interpreted that the methane hydrate concentrated intervals show these anomalies. In 2015, gas chimney structures were explored in more detail with natural gamma ray, resistivity, sonic, CMR and neutron logging tools. The hydrate concentrated intervals were clearly identified with high neutron porosity, low neutron gamma density and low sigma (neutron capture cross section) anomalies in addition to the previously recognized logging anomalies. We will report the detailed methane hydrate distribution within the gas chimney-mound structure according with the correlation of LWD and coring results.
This study was conducted as a part of the shallow methane hydrate exploration project of METI. We express sincere thanks to Mr. Tetsuya Fujii, Mr. Tokujiro Takayama, Mr. Takashi Kotera, JOGMEC and Dr. Shusaku Goto, AIST, for their support and advices.

Keywords: shallow methane hydrate, gas chimney mound structure, Logging While Drilling

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High-resolution 3-D seismic survey (HR3D) of gas chimney structures off Joetsu, Niigata Prefecture

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In the summer of 2015, a high resolution three-dimensional seismic survey (HR3D) was carried out off Joetsu, Niigata Prefecture, to delineate the detailed structure of the gas chimneys which are widely distributed in the area off Joetsu.
In the sections of sub-bottom profiler (SBP), the gas chimneys are characterized by blanking which makes the inner structure of the chimneys invisible. This could be caused by high reflectivity materials existing in shallow layers near the sea floor and the seismic energy could not penetrate to deeper layers. This brings difficulties in tracking of the shallow gas hydrates and BSR as well as formation boundaries in the gas chimneys.
To image the detailed three-dimensional structure within the gas chimneys and its surrounding areas, an HR3D was planned and conducted with short streamer cables with high-density shot and receiver intervals along with high frequency airgun (GI Gun.)
The results of HR3D is good enough to reveal the fine structure in the gas chimneys which are unclear in the SBP data, even though the resolution is inferior to the SBP. The resolution of HR3D data is much higher than that of the existing large-scale 3D surveys which were carried out for petroleum exploration, even though the penetration is not enough compared to the existing 3D in this area.
The HR3D date, along with loggings and other geological data, will be a very useful tool to investigate the spatial distribution of gas hydrates which were confirmed at the wells drilled on the mounds and in the pockmarks.
This study was conducted as a part of the Shallow Methane Hydrate Exploration Project of METI (FY2015.)

Keywords: high-resolution 3D seismic survey, HR3D, shallow gas hydrate, gas chimney, off Joetsu

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Detailed depositional topography in the Toyama Trough revealed by the 7K13 Cruise

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Regional bathymetry and backscatter data were obtained by a multibeam echo sounder and a sub-bottom profiler in the Toyama Trough for estimation of shallow methane hydrate resources during the 7K13 Cruise of R/V Kaiyo-maru No. 7 (Matsumoto et al., 2014). This study reports detailed depositional topography imaged along the upper reach of the Toyama Deep-Sea Channel (TDSC).
Some kinds of bars and terraces were found on the bottom of the TDSC. Bars include point bars, longitudinal bars. Some terraces were formed with translation of the channel. Steps transverse to the channel were associated with pools down channel of the steps.
Large sediment waves develop on the levees on the outer banks of meander bends of the TDSC. Some sediment waves are associated with a large scour on the back slope of the levee, suggesting that they were formed as cyclic steps.
This study uses data obtained by the H25 fiscal year shallow methane hydrate exploration project of METI.

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Type distribution and composed area of “gas chimney structure” around Japan Island

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Distributions of shallow gas hydrates often correlate with “Gas chimney structure”, characterized by well-developed acoustic blanking in the eastern part of Japan Sea. Matsumoto et al. (2015MS) confirmed 971 gas chimneys during the RV MBES & SBP survey in 2013-2014, identified three morphological types (Single, Composite, related with inversion structures) based on the occurrence and distribution pattern. Continuously, 2015 survey was performed from eastern part of Japan Sea to the north Pacific and southern part of the sea of Okhotsk, south of Hokkaido and off Abashiri. This survey was performed in 75 days from May 6th to July 19th. In order to find the existence of “gas chimney structure”, we firstly extracted topographic anomalies from the initial survey based on depth profiles and back-scatter images by MBES (Multi-Beam Echo Sounder) EM302, secondary evaluated the presence of acoustic blanking, “gas chimney structure” from detailed SBP (Sub-Bottom Profiler) TOPAS PS18 survey on the topographic anomalies.
“Gas chimney structures” were identified in all the survey areas from around Oki islands, Toyama trough, Mogami trough, off Nishi-tsugaru, off Okushiri islands, off Hidaka, off Tokachi, and off Abashiri, counting up to 771. The total number of “gas chimney structures” has become 1742 throughout. In this presentation, we introduce the features with morphological types of “gas chimney structure” in each area, focus the number and size distribution.
This research was a part of METI's project entitled “FY2014 Promoting research and development on methane hydrate”.

Reference

Matsumoto et al. (2015MS), Types and distribution of gas chimneys: host structure of shallow gas hydrates, Japan Geoscience Union Meeting 2015.

Keywords: shallow gas hydrate, gas chimney structure, number and size distribution

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Recovery and hydrate estimate of gas hydrate bearing sediments by pressure coring tool PCTB and onboard core handling system PCATS

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Multi-hole drilling/coring was conducted on 3 hydrate mounds/gas chimney structures in Oki trough and off Joetsu in August to November, 2015, with an intention to reveal the distribution and amount of shallow gas hydrates in gas chimney structure. On the basis of the results of 2014 drilling campaign, the pressure coring system of Geotek LTD composed of coring tool PCTB and core handling system, PCATS, installed onboard the drill ship. PCTB is designed to recover pressurized gas hydrate bearing sediment cores of 2.5 m long and 5.1cm in diameter, and 2015 campaign recovered 32 PCTB cores with in situ pressure in 42 deployments. Immediately after the core recovery on deck, PCTB cores were transferred to PCATS (Pressure Core Analysis and Transfer System) for transparent X-ray imaging, Gamma-ray density and Vp logs to roughly identify the lithology and occurrence of hydrates. Then, the pressurized cores were cut into 2 to 5 sections for detailed measurements and for shore-based analysis. Quantitative degassing experiments to measure total amount of hydrate gas has provided the precise volume% of hydrate in the section. After degassing, waters of the section were squeezed to measure chloride and sulfate concentration. Chloride concentration of the pristine IW is calculated from hydrate amount (vol%) and measured water chemistry, assuming that the squeezed water is a mixture of pristine IW, hydrate water (Cl and SO4 = zero) and sea water contamination (Cl=559mM, SO4=28.9mM). Cl of the pristine IW provides the baseline to estimate the amount of hydrate in nearby sections and cores from squeezed “IW” waters. We also report the occurrence, micro-texture and estimated amount of gas hydrate in pressure cores. This study was conducted as a part of the shallow methane hydrate exploration project of METI. We express our thanks for allowing us to present this paper.

Keywords: Pressure coring, gas hydrate amount

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Concentration anomalies of pore waters collected from shallow gas hydrate deposits in the eastern margin of the Japan Sea

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Massive gas hydrates have been observed around the gas chimney structures, excess gas accumulation is responsible for the rapid formation of gas hydrates near the seafloor. Although the dissociation of gas hydrates results in the dilution of pore water due to the release of fresh water from the hydrate crystals, rapid formation of gas hydrates can enclose saline pore waters excluded from the crystals. The in situ pore water geochemistry in the subseafloor environments in the gas hydrate area reflects the dynamic history of formation/dissociation of gas hydrates. We have conducted geochemical analyses of pore waters collected from the shallow gas hydrate occurrences along the eastern margin of the Japan Sea during the PS15 expedition in order to show the geochemical models of shallow gas hydrate formation system.
Although concentrations of chloride dissolved in pore waters are close to the seawater of ~560 mM at the sites where no or small amount of gas hydrates accumulates, those at the dense massive gas hydrate sites are often increased; reaching >1400 mM in the highly gas hydrate accumulated intervals. These high chloride intervals locate in shallower depths at the higher methane flux sites. Our results indicate that the pore water geochemistry is often modified in response to the formation dynamics of massive gas hydrates near the seafloor.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: Shallow gas hydrate, Pore water

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Comparing physical properties of the sediments in the Japan Sea

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In 21 century, most of the methane hydrates distributed in the Japan Sea are by an outer layer type, and it is interpreted that developing in dozens of meters from the seabed of the mound with the gas chimney structure in a recent study (Matsumoto at. al.2014). Because the outer layer type methane hydrates are different from the depths type of the Nankai trough in many respects, we should prepare the new development scheme when producing tests in future. Therefore, this study focuses on the behavior of the sediment physics on the methane hydrate accumulations, and influence of the seafloor environment. When we would take methane hydrate near the seafloor, not only we are concerned about degradation of the seabed environment, but also we are apprehension about the ground sinking by the machine built and the effect of foundation pile exchanging to the poor subsoil by the degradation. On the other hand, it is essential to grasp stress of sediment of the depth profiling by examining the ground strength every depth because various work in the collection of the hydrate is performed in the deep layer.
Therefore, we had geotechnical tests, such as Vane share strength tests, Cone penetrate tests, and Water content tests, at the reference sites around the Oki and the Jyoetsu offing. In addition to study mechanical tests, we compared with MD179 results. This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: The Japan Sea, sediments, physical properties

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Cone penetration tests at shallow gas hydrate exploration sites in 2015

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To develop a production method for natural gas from shallow gas hydrate accumulations, the mechanical strength of shallow sedimentary layers should be investigated. During shallow gas hydrate exploration in 2015, cone penetration tests were conducted at test sites off Joetsu, Japan. The cone penetration resistance, sleeve friction, and pore pressure were measured by using a piezocone penetrometer testing (PCPT) apparatus supplied by Geoquip Marine. The effective section of the cone was 10 cm^2. The penetration rate and the stroke of the PCPT were 2 cm/s and 3 m, respectively. The cone penetration resistance was less than 100 MPa. The data measured by the penetrating cone were transmitted to an onboard display and a recording device in real time. An undisturbed specimen 1 m long was also sampled immediately above and below the PCPT trial zones, and vane shear and undrained triaxial tests were performed. Because test wells for PCPT were drilled at sites near coring wells, the PCPT results were compared with descriptions of the core specimens. The comparisons confirmed that the PCPT responded sensitively to thin sandy layers, granular gas hydrates, and carbonates in a muddy zone. Undrained shear strengths was also estimated from the PCPT results by using a relational expression for subsurface exploration on land. The estimated values of undrained shear strength were similar to those of the undrained shear strengths measured by the triaxial tests on the undisturbed specimen sampled near the PCPT trial zones. By using the estimated undrained shear strength values, the depth profiles were compared.
This study was conducted as part of the shallow methane hydrate exploration project of the Ministry of Economy, Trade and Industry (METI), Japan.

Keywords: Shallow gas hydrate, Piezocone penetrometer testing, Undrained shear strength

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A working hypothesis on the accumulation of methane hydrate in gas chimneys developed in the eastern margin of Japan Sea

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A lot of gas chimney structures are identified by a sub-bottom profiler as acoustic blanking in the eastern margin of Japan Sea. The acoustic blanking is caused by hard stuff such as carbonate nodules and gas hydrate that develop usually on the seafloor or very shallow depths. Therefore, the deeper part of the acoustic blanking or gas chimney structure was remained unknown. The coring of gas chimney structure of three topographic highs by R/V Hakurei in 2014 clarified that gas hydrate occur from the surface to the deep in the gas chimneys.
A series of observation of the recovered cores by necked eyes, X-ray CT photography and experiments such as sieving of carbonate nodules from mud clarified the close association of carbonate nodules and gas hydrate in the sediments of gas chimneys. This combination of carbonate nodules and gas hydrate occurs periodically and the calculated cycles are around 15 ky.
Preceding studies clarified the following subjects.
1. Carbonate nodules in the gas hydrate field of Japan Sea are interpreted to be formed in the sulfate-methane interface (SMI).
2. The carbon and oxygen isotopic ratios of the carbonate nodules demonstrated the thermogenic methane from the deep largely contributes the formation of the nodules.
3. Active faults and folds develop in the mobile belt of the eastern margin of Japan Sea and form many topographic highs.
Combining the above-mentioned preceding studies and acquired data, we propose the following working hypothesis: Methane gas has been periodically supplied from the depth by the movements of active faults, and the gas repeatedly formed both carbonate nodules and methane hydrate at around the depth of SMI that existed at or in the shallow depth of the seafloor.
Further studies are required if the proposed mechanism of the accumulation of shallow gas hydrate would be applied generally to the other gas hydrate-bearing topographic highs that distribute in the eastern margin of Japan Sea.
We express sincere gratitude to all the persons in JOGMEC who engaged to operate R/V Hakurei in 2014. X-ray CT photos of cored samples are taken under the cooperation program between Kochi Core Center and Akihiro Hiruta (14B013). This research is a part of the shallow methane hydrate exploration project of METI.

Keywords: shallow methane hydrate, carbonate nodule, Japan Sea, drilling, active fault

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A rock paleomagnetic study of marine sediments in gas hydrate area of the eastern marging of Japan Sea

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We conducted a rock magnetic study of marine core sediments to clarify relation between shallow gas hydrate and around the sediments.The core samples were taken from around Oki area and off Joetsu, Japan Sea, during PS15 cruise in 2015. We mainly report magnetic susceptibility measurement of whole-round core samples.
This study was conducted as a part of the shallow methane

Keywords: Shallow gas hydrate, Rockmagnetism, marine sediments, Japan Sea

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Estimation of shallow gas hydrate formation age by methanol analysis in the gas hydrates

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Natural gas hydrate, an inclusion compound of natural gas in water cages, is found in deep-sea sediments and permafrost region. In eastern margin of Japan Sea, shallow gas hydrates have been found and recovered by piston coring (Matsumoto et al., 2011; Lu et al., 2011). We have an interest on formation age of the shallow gas hydrates. In our previous study, methanol and formaldehyde might be formed by natural radiation and accumulated in the gas hydrates. It indicates that the concentration of these volatile organic compounds is related to the formation age. Previous measurements of methanol in shallow gas hydrates recovered by piston coring showed that it was difficult to discuss the formation age because the gas hydrate samples were recovered from a few meter below the sea floor and the expected amount of methanol formation by natural radiation was too small. In this study, gas hydrate recovered from much deeper region was sampled in 2015 expedition. Methanol in the hydrate together with pore water samples were analyzed by gas chromatography mass spectrometer (GC/MS). The small amount of methanol was detected even in the deeper gas hydrate samples (from ~100 meter below sea floor). The interpretation will be discussed, considering the sedimentation rate. This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: shallow gas hydrates, formation age, methanol, gas chromatography, natural radiation

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Atmospheric gas concentration anomalies in the ocean: A preliminary report from a shallow gas hydrate exploration project

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We could often notice the gas plumes rising from a seafloor water column at gas hydrate fields. Active gas vents at the seafloor have previously been reported by some researchers. Methane (CH_4) is a major constituent of seep gases. Methane is an important short-lived climate pollutant. It is reported that oil spill at sea bottom and offshore oil/gas platforms affect atmospheric CH_4 concentration. Likewise gas seeps may contribute to atmospheric gas concentration above the sea surface. Our objectives were to investigate the distribution of atmospheric CH_4 distribution over the sea surface of gas hydrate areas by continuous measurement on a research vessel.
We took advantage of topographical survey (7K14, 7K15) for grasping the resources of shallow gas hydrate and for continuously measuring CH_4 concentration. We used the R/V Kaiyo-Maru No.7 (Kaiyo Engineering Co., Ltd., Japan) from April to June 2014 and from May to July 2015. Continuous measurement of atmospheric CH_4 was performed on the ship using a wave-length-scanned cavity ring-down spectrometer (WS-CRDS) (model G2201-i, Picarro Inc., USA). Air sample was collected from an air inlet installed at the compass deck (approximately 8 m above the sea level) of the ship using an air pump placed in an observation room. The ship sailed at approximately 6 knot during the survey periods. Ship-s location data were obtained with a nautical GPS.
There were 2 types of sea areas: (1) areas with gas plumes observed, and (2) areas with no gas plumes observed. Additionally, gas plumes were unevenly distributed in the gas plume area. In some of gas plume areas, the anomalies of CH_4 concentration were coincidently observed around above gas plumes. Atmospheric gas concentration affected by sea water temperature, water depth, and scale of gas plume varied every different sea areas.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: gas hydrate, Methane gas, gas plume

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Shallow methane hydrate outcrops discovered through ROV submersible survey in the Japan Sea

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In the Japan Sea, several outcrops of methane hydrate were discovered through ROV (Remotely Operated Vehicle) submersible survey. Occurrences of the outcropping methane hydrates and their surrounding topographies are described using submersible videos and still images. This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: shallow methane hydrate, ROV, Japan Sea

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Preliminary account of benthic habitat mapping on shallow gas hydrate areas on the eastern margin of Japan Sea.

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This study presents results of an approach for sea floor habitat mapping based on an integrated analysis of multibeam bathymetric data, associated geoscientific information, and benthos data from shallow gas hydrate areas on the eastern margin of Japan Sea.
Six areas, SW of Oki trough, SE margin of Oki trough, Northern Torigakubi spur, off SW of Sado, off Hajikizaki and NE of Torimiguri were investigated. The number of individuals of macrobenthos taken a picture of to the high-definition television camera of ROV "Hyper dolphin" was done and several in total was done at ten seconds in which the position of ROV was recorded. The bottom sediment was recorded at the same time, and the relation between the benthos distribution and the bottom sediment was examined. In addition, the positional data of ROV, the bottom sediment, and the benthos distribution were input to GIS, it reflected in the bathymetric chart, the habitat map was made, and the benthos distribution and the seafloor condition in each area were compared.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: shallow gas hydrate, habitat mapping, benthos

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Analysis of benthic community food web at gas hydrate deposits using stable isotope analysis

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To better understand the ecology of benthic community at gas hydrate deposits, the stable isotopic signatures of carbon, nitrogen and sulfur ($\delta^{13}C$, $\delta^{15}N$ and $\delta^{34}S$) of the surface sediment and benthic fauna collected at Joetsu Basin and Mogami Trough were evaluated. We analyzed their food web and speculated the carbon and sulfur cycles in the benthic water at hydrate deposits.
Sampling of benthic fauna at seeps using a slurp gun and a strainer was conducted during September 2013 and October 2014 at seep and reference sites. We obtained surface sediment sample using MBARI ROV coring system at depths of 0-2.5 and 2.5-5 cm below seafloor. Macrofaunal sample was dissected on board and frozen. Meiobenthos were removed by sieving of sediment samples and frozen. In laboratory, faunal sample was powdered after freeze drying. We measured stable isotope signatures of carbon and nitrogen using IRMS (Flash 2000/Delta V IRMS, Thermo Scientific Inc.) after removal of inorganic carbon using HCl solution and neutralization by NaOH. Similarly, stable isotope signature of sulfur was measured using free dried faunal sample. We collected the precipitates of zinc and barium in pore water extracted from the sediment sample and seawater by filtration for sulfur isotope composition of sulfate and sulfide.
Our result shows that no distinct difference between the isotopic signatures of red snow crab, one species of eelpout, *Bothrocara hollandi*, northern shrimp and amphipods collected both at seep site and reference site. It suggests that their food habitat depends mainly on photochemically-produced organic carbon and sulfur from seawater sulfate even in an individual inhabiting around methane seep.
While, biplots of $\delta^{15}N$ versus $\delta^{13}C$ vs and $\delta^{34}S$ versus $\delta^{13}C$ suggest that some benthic animals such as solemyid clam and frenulata tube worm depend on carbon and sulfur derived from chemosynthetic bacteria.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: methane hydrate, benthic fauna, food web, Stable isotope signatures

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Detailed Topographic survey on basin areas of the Japan Sea and around Hokkaido

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Regional bathymetric and backscatter data were obtained by a multibeam echo sounder (MBES) and a sub-bottom profiler (SBP) in 10 promising basin areas of the Japan Sea and around Hokkaido, for estimation of shallow methane hydrate resources. Survey was conducted on basin areas between West of the Oki Island and the Okushiri Basin of Japan Sea, and in the Hidaka Trough, off Tokachi and off Abashiri area around Hokkaido, using EM302 and TOPAS PS18 of Kongsberg, during the 7K13(43 days), 7K14(62 days) and 7K15(75 days) Cruises of R/V Kaiyo-maru No.7 (Matsumoto et al., 2014, Matsumoto and Satoh, 2015). Topographic shade maps are produced from obtained bathymetric data, and many topographic features concerning sedimentary processes, such as cliffs, slide sheets, submarine channels, are recognized in the survey areas.
Many sets of submarine slide sheet and adjacent cliff are observed in West of the Oki Islands and the Oki Trough areas, and no major submarine channels are found in these areas. In the southwest part of Oki Trough, almost of all trough floor are occupied by a large number of slide sheets. Although several number of slides are found, large submarine channels, flat basin floor are observed, and many topographic highs formed by tectonic inversion (Okamura et al., 1996a, b) are distributed in the Toyama Trough and the Mogami Trough area. Many sets of cliffs and slide sheets are found in the slopes of western and northeastern margin of the Hidaka Torough, and with no major submarine canyons. Two remarkable submarine channels are observed and topographic highs are extended from south to north between two channels off Abashiri area in the Okhotsk Sea.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.
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Keywords: marine topographic map, submarine slide, submarine channel

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Sedimentary Environment and Early Diagenesis of Thin Sandy Sediment Layers below Sea Floor in the Eastern Margin of Japan Sea

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Sediments in most of the sites along the eastern margin of Japan Sea are mainly composed of muddy fine clastics, and can be often found accompanied by a small amount of very fine to medium grained sandy intercalations, which are usually observed as thin layers and laminations in muddy layers. Fine-grained sediment samples below the seafloor were retrieved by the MD179 in 2010, the HR14 in 2014 and the PS15 in 2015 at the Umitaka Spur, Joetsu Channel, Toyama Trough, Japan Basin, Nishi Tsugaru and Okushiri Ridge areas. It is important to clarify the relationship between burial depths and absolute porosities of the argillaceous sediments in relation to early diagenesis. They consist of silt- to clay-grained particles, and they sometimes contain very fine- to medium-grained thin sandy layers. Average porosities of these fine-grained sediments are 50 % in all study areas, which quickly reduce from 60% to less than 50% within 10 meters and gradually decrease to the depth. However, mean pore sizes in the Nishi Tsugaru are around 1000 nm while 100 nm in the other areas, which tend to decrease with depth. It is suggested that repacking of the muddy particles gradually advances by mechanical compaction, which may crucially influence permeability.
This research is a part of the METI project entitled “FY2015 promoting research and development on methane hydrate.

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Characteristics of sedimentation rates around hydrate seep area in the eastern part of Japan Sea

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Background

It is difficult to estimate the sedimentation age in methane hydrate area of eastern parts of the Japan Sea due to unable to find TL (thin laminated) layers, suitable for correlation with Japan sea stratigraphy, from core sediments. Oi et al. (2015MS) anticipated that sediment expands by hydrate growth caused to remarkable differences of sediment age with a reference core and a hydrate bearing core.

Purpose

One purpose is to clear the Chronostratigraphy based on radiocarbon dating and age from diatom fossils of several meter drilled cores in hydrate bearing area, Umitaka Spur and Oki Trough during the cruise from August to October 2015. The other purpose is to compare the sedimentation rate changes with approximately 60 m long core from Mogami Trough off Tobishima West (Oi et al., 2015MS).

Method

For radiocarbon dating, we used shells, woods and foraminiferal picked from undisturbed mud sediments. $\delta^{13}C$ correction and calibration (Reimer et al. 2013; Bronk Ramsey 2009) were dated by the 3MV AMS at the Institute of Accelerator Analysis Ltd. as the basis samples (HOxII) after serial oxidation and reduction reactions. For Analysis of diatom fossils, we made untreated spraying slides using mud samples of core sediments or included massive hydrate. Certifications of Diatom zones followed two standard stratigraphic classifications applied in North Pacific wide area (Yanagisawa and Akiba, 1989) and the upper Quaternary in Japan Sea (Akiba et al., 2014).

Discussions

30 radiocarbon dates of 2015 samples indicate that upper parts of sedimentation rates off Joetsu were 2 to 3 times as fast as those of Oki Trough and Mogami Trough during the last 50 kyr. These difference might have relevance to the distance from Japan Island. Furthermore, we found the basic boundary (300ka) by the distinct of diatom fossils, *Proboscia curvirostris*, from 20-25mbsf of Oki Trough cores and about 88mbsf of Umitaka Spur. These dates and previous data of Mogami Trough (about 42mbsf) also describe both remarkable slow depositions in Oki Trough and fast depositions in Umitaka Spur. In this presentation, we discuss stratigraphy of other cores including methane hydrate.

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Keywords: methane hydrate, sediment age, microfossils

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Ecology and stable isotope composition of benthic foraminifera associated with cold seeps on the Hidaka Trough, northwestern Pacific

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Benthic foraminifera at cold seeps on the Hidaka Trough, northwestern Pacific were studied to investigate the effects of methane on the geochemistry and faunal characteristics of benthic foraminifera assemblages and to discuss potential applications of foraminifera for reconstruction of methane release in the past and present. Sediment cores for this research were collected from eight chimney sites and one reference site with gravity corer in July 2015. Calcareous forms dominate benthic assemblages, accounting for 90 percent or more of the benthic populations for most samples. Results from Rose Bengal staining method indicate that certain species inhabit seep sites in the study area. *Rutherfordoides cornuta*, which is related to high methane gas content of the sediments and reported as methanophilic taxa from methane seepages at Sagami Bay (Akimoto et al., 1994; Kitazato, 1996), found alive (cytoplasm containing specimens) within surface sediments at four chimney sites in our study area including cores at the center of chimney. Other calcareous foraminiferal assemblages associated with chimney sites were typically infaunal species including *Brizalina pacifica*, *Bolivina spissa*, *Chilostomellina fimbriata*, *Globobulimina auriculata*, *Nonionella globosa*, *Nonionella stella*, *Stainforthia fusiformis*, and *Uvigerina akitaensis*, which can inhabit below water-interface and are also abundant in organic-rich oxygen-depleted environments. Geochemical analyses of living (stained) benthic foraminifera in our research sites doesn't show highly negative $\delta^{13}C$ values comparable to those fossil (unstained) benthic foraminifera that are reported from seep sites such as the Gulf of Mexico (Sen Gupta and Aharon, 1994) or Monterey Bay (Martin et al., 1999); however differences in $\delta^{13}C$ values for living benthic foraminifera of a given species were observed within a single core or between cores at chimney sites, which are unusual. For instance chimney site cores contain live specimens of *B. spissa* with $\delta^{13}C$ values ranging from -0.43‰ to -1.07‰, -0.71‰ to -1.97‰, and -0.37‰ to -0.94‰, respectively. In contrast, at reference core the $\delta^{13}C$ composition of *B. spissa* varies little and remains approximately constant around -0.70‰ over the length of the core. Variable carbon isotope values are also evident in other species such as *U. akitaensis*. These results suggest that $\delta^{13}C$ values of foraminifera tests are influenced by methane seepage and different pore-water chemistry. Therefore, variations in isotopic composition can suggest temporal variations in seep activities and the differences in carbon isotope values will be expect to increase with the activity of the seeps. A good comprehending on ecology and stable isotope composition of modern benthic foraminifera at cold seeps may help identify paleo-seeps and will enhance our knowledge of climatic and oceanographic changes. This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: Benthic foraminifera, Cold seeps, Hidaka Trough, Methanophilic taxa, Northwestern Pacific, Stable isotopes

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Isotopic composition and U-Th age of methane derived authigenic carbonates

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Shallow gas hydrates often occur in close association with carbonate concretions. The precipitation of carbonates is considered as the results of sulfate reduction-methane oxidation (AOM) at SMT at around 2 to 5 mbsf in high methane flux fields. Methane derived authigenic carbonates (MDAC) of shallow gas hydrate fields in Japan Sea range in size from a few mm to a few meters. They are mostly composed of high mangesian calcite (HMC) and/or aragonite (AR). Oxygen isotopic signatures seem to indicate that HMC were formed in waters slightly depleted in O-18 while AR were from slightly enriched in O-18, or alternatively, HMC from higher temperature and AR from lower temperature. Frequency and amount of MDAC should reflect the intensity of AOM, that is, the intensity of methane fluxes. Therefore, the secular variation of methane flux in the past can be referred from the age distribution of MDAC. Ages of the precipitation of MDAC were determined by means of the U-Th isotope disequilibrium method. Previous studies (Watanabe and Nakai, 2006; San-no, 2008MS; Suzuki, 2010MS) have revealed that the age of the MDAC collected from surface sediments (< 4 mbsf) tends to concentrate around 20ky, and discussed the relation with glacio-eustasy. MDAC of this study ranges from the sea floor to about 80 mbsf, covering the entire depth range of shallow gas hydrate. Precise age determination of the MDAC should reveal the time constraint and conditions of the formation of shallow gas hydrate of Japan Sea. This study was conducted as a part of the shallow methane hydrate exploration project of METI. We express our thanks for allowing us to present this work.

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Keywords: MDAC methane derived authigenic carbonates, shallow gas hydrates, U-Th isotopic disequilibrium

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Isotopic compositions of Carbon, Oxygen, and Strontium in authigenic carbonates from Umitaka Spur, off-Joetsu, southeast of Japan Sea

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We present isotopic compositions of Carbon ($\delta^{13}C$), Oxygen ($\delta^{18}O$), and Strontium ($^{87}Sr/^{86}Sr$) in authigenic carbonates from Umitaka Spur, off-Joetsu, southeast of Japan Sea.
The carbon isotopic values in authigenic carbonate from Umitaka Spur are higher than those of Joetsu Knoll and west off-Tobishima Island. This range corresponds to the $\delta^{13}C$ values of thermogenic methane (Bernard et al., 1978). Range of the $\delta^{18}O$ values of authigenic carbonates from Umitaka Spur is mostly equal to those of Joetsu Knoll and west off-Tobishima Island.
The $^{87}Sr/^{86}Sr$ ratios in authigenic carbonates from shallow depth of Umitaka Spur is equal to those of modern surface seawater in off-Joetsu. The Sr-isotopic ratios in authigenic carbonate from deeper depth are lower ratios. This Sr-isotopic trend can be correlated to the global Sr-isotopic trend in seawater from late Pleistocene to present. It indicates that Sr-isotopic ratio of authigenic carbonate reflects the Sr-isotopic ratio of seawater at the time of deposition. The Sr-isotopic ratios in pore water are parallel lower than those of authigenic carbonate. It indicates that pore water includes light Sr by diagenetic procedure.
Acknowledgments: This study was conducted as a part of the shallow methane hydrate exploration project of METI. Isotopic measurement of Strontium was performed in Kochi Core Center, by visiting scientist program of JAMSTEC.

Keywords: authigenic carbonates, shallow gas hydrate, Japan Sea

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Geochemistry of pore water, dissolved gas, and sediment from offshore Hidaka area, Hokkaido

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Precise acoustic investigations resulted in the wide distribution of gas chimney structures accompanying high backscatter anomalies, which indicates strong methane migration from deep sediments to the seafloor and the potential formation of gas hydrate/carbonate near the seafloor in the offshore area of Hidaka, Hokkaido. We have deployed gravity corer and collected sediments to analyze compositions of sediment, pore water, and dissolved gas for characterizing geochemical system related to the high methane environments near the seafloor.
Concentrations of sulfate dissolved in pore waters just on the gas chimney show rapid decrease with depth. Concentration of methane and the depth of sulfate-methane interface, indicative of methane flux, locates at 74 to 420 cmbsf, equivalent to the shallow gas hydrate area in the eastern margin of the Japan Sea. Carbonates are observed only at the western slope region where the moderate methane flux is estimated. Geochemical environments are different among the locations reflecting subseafloor structures.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

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Geochemistry of dissolved gas around gas chimney structures in the Mogami Trough, Japan

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In the Mogami Trough, eastern margin of the Japan Sea, active gas venting has not been observed on the seafloor, however, distribution of mounds and pockmarks on the gas chimney structure (vertical acoustic blanking zone of shallow sediment) indicates the strong flux of gas-rich fluid from deep sediments and formation of gas hydrates near the seafloor. Concentrations and isotopic compositions of headspace gases collected inside and outside the well-developed gas chimneys were analyzed in order to characterize geochemical structure across the gas chimney and surrounding sediments. Distributions of methane (C1) and ethane (C2) concentrations and C1/C2 ratios show gradual increase toward the center of gas chimney particularly in shallow sections, reflecting higher biogenic methane production in the higher gas migration zone within the chimney. The stable carbon isotopic compositions of methane are also high in the center of the chimney due to higher contribution of thermogenic gas derived from deep section. The higher concentration and upward flux of gas are observed in the mound area rather than pockmark area, reflecting that the activity of gas chimney may control the distribution of gas hydrate responsible for shallow topographic anomalies.

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Geochemistry of pore waters around gas chimney structures in the Mogami Trough, Japan

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Shallow gas hydrates have been often found on the gas chimney structures in the eastern margin of the Japan Sea. Although the strong gas flux through the gas chimney is a key for the massive accumulation of gas hydrates near the seafloor, geochemical nature inside/across the gas chimney has not been discussed well. We have collected pore waters from shallow sediment inside/outside the gas chimney in the Mogami Trough and analyzed major ion concentrations to understand the special change of pore water geochemistry reflecting the activity of gas chimney.We found significant relation between the distance from the center of gas chimney and the gradient of alkalinity/depth of sulfate-methane interface, indicative of methane flux; they decrease similarly in response to the distance, particularly outside the chimney. Although the activity of gas chimney indicated by the chimney size and methane flux varies in the same area of the Mogami Trough, gas hydrates potentially accumulate near the center of gas chimneys.

Keywords: Gas Hydrate, Gas chimney

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Geochemistry of seawater and interstitial water from Tsushima Basin and Oki Trough, Japan

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The characteristic seafloor topography associated with gas hydrates in shallow sediments was reported in the seafloor of the SE margin of Tsushima Basin and Oki Trough, Japan. Interstitial water and seawater collected from these areas during the UT14 cruise were analyzed for characterizing the fluid geochemistry responsible for methane migration toward the seafloor and formation of hydrates. In the eastern margin of Tsushima Basin, high concentrations of sulfate and alkalinity in interstitial water reflect very shallow SMI depths (~1.7mbsf), strong methane fluxes, and methane generation due to the decomposition of organic matters in shallow sediments. The low concentrations of silicate dissolved in seawater indicate that the buried old organic matters are responsible for the formation and distribution of gas hydrates near the seafloor.

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Time-series analysis of pore water from shallow gas hydrate area on the Umitaka Spur, Japan Sea

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Active gas venting (seepage) from the seafloor and outcropping of massive gas hydrate have been observed on the Umitaka Spur in the eastern margin of the Japan Sea. Submersible observation revealed that the strength and location of gas venting had changed within a few days, geochemical environment near gas venting including gas hydrate might have also changed within a short period compared to geological time scale. We have deployed a long-term osmotic fluid sampler (OsmoSampler) near the gas venting site on the Umitaka Spur from September 2013 to March 2014 (160 days, NT13-E02 and NT14-E03 cruises) and collected pore waters continuously to examine the potential changes of pore water geochemistry and the impacts on the near-surface environments.
Concentrations of dissolved ions change shortly, for 3~5 days, through the entire sampling period, which are controlled by the input of saline water from gas hydrate formation and of fresh water from gas hydrate dissociation. Gas venting was observed 10 m away from OsmoSampler, however, concentrations of methane dissolved in pore water were low during the first 20 days and occasional high methane concentrations were observed only from the 20th to 40th day. Rapid gas hydrate formation caused by high gas flux might plug the path delivering gas-rich fluids to the seafloor, contrary, the reduced gas flux (venting) caused the dissociation of gas hydrate. After the 70th day on, methane concentration was constant at low level, <1 mM, indicating that the location of gas venting had been moved due to the gas hydrate plugging. Significant concentrations of ethane during that period also indicate the gas hydrate plugging and subsequent change of major gas source from biogenic-rich to thermogenic-rich. Geochemistry of pore water has changed dynamically and shortly in response to the change of gas venting activity.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: Time-series analysis, pore water, Shallow gas hydrate, gas venting

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Distribution of methane in seawater from shallow gas hydrate areas in the Japan Sea

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We have analyzed the concentrations of methane dissolved in the bottom seawaters collected from shallow gas hydrate occurrences including active gas venting and the associated distribution of carbonates/bacterial mats during the NT15-E03 expedition in the Japan Sea. Methane concentrations are close to the normal bottom water level over the mud seafloor, however, they increase typically near the gas venting, carbonate, and bacterial mat sites; gas venting had only been active during the formation of carbonates and bacterial mats. Contrary, the number of benthos does not correlate with the concentration of dissolved methane, it probably reflects the location and seafloor condition, not the present concentration of methane. The concentration of methane rapidly decreases with shallowing depth due to the oxidation and diffusion of methane ejected from the seafloor in water column, reaching normal seawater level at the intermediate depth. Environmental impacts of the gas venting, possibly gas explosion on the seafloor, are very limited near the seafloor.This study was conducted as a part of the shallow methane hydrate exploration project of METI.

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Characteristics of natural gas hydrate retrieved off Abashiri, the Sea of Okhotsk

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In the area of southwestern margin of the Sea of Okhotsk, an existence of natural gas hydrate has been expected using seismic data. Yamamoto et al. (2002) suggested existence of natural gas hydrate off Shiretoko, the Sea of Okhotsk, by a long piston coring. Recovery of sediment core for gas hydrate started in the cruise of TK11 in 2011 in the framework of joint research between Kitami Institute of Technology and University of Tokyo, and obtained gas-rich sediment cores off Abshiri. Gas hydrate crystals were first retrieved in the cruise of UT12 (T/S Umitaka-Maru) in 2012. New hydrate-bearing sites were also discovered in the cruise of NT13-20 (R/V Natsushima in 2013. We planned operations using T/S Oshoro-Maru, and obtained sediment cores and gas hydrates at this area in the cruise of OS249 (2012), OS263 (2013), and C020 (2015).
Gas hydrate crystals were obtained and stored in liquid nitrogen for Raman spectroscopic analysis and calorimetry. Samples of hydrate-bound gas were obtained onboard and stored in 5-mL vials, and sediment gas were also obtained using a headspace gas method. We measured molecular and stable isotope compositions of these samples. We summarized the results as follows:
1) Gas hydrates belong to the cubic structure I, containing methane (more than 99%) and hydrogen sulfide (less than 1%). C1/C2+ ranges from 5,500 to 5,800.
2) Hydration number is estimated as 6.03±0.04, agrees well with 6.04±0.03 for synthetic methane hydrate (Sum et al., 1997).
3) Dissociation heat from hydrate to gas and water is estimated as 55.1±0.3 [kJ/mol], agrees well with 54.19±0.28 [kJ/mol] for synthetic methane hydrate (Handa, 1986).
4) Hydrate-bound gas is of microbial origin according to C1/C2+ and stable isotopes of hydrocarbons, however, delta 13C of ethane seems relatively large.
5) Gas hydrate in the NT13-20 PC06 core contains trace amount of ethane (several ppm), whereas that in the NT13-20 PC02 core contains about 100ppm of ethane.
6) SMI (sulfate-methane interface) depth of the sediment cores are less than 1m, indicating high methane flux off Abashiri.
We appreciate the support of the crew onboard R/V Natsushima during the NT13-20 cruise, and T/S Oshoro-Maru during the cruises of OS249, OS263, and C020. This study was supported by the Grant-in-Aid for Scientific Research (B) 25289142 and 26303021 of the Japan Society for the Promotion of Science (JSPS).
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Keywords: hydrate, the Sea of Okhotsk, stable isotope

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Isotopic fractionation of ethane at the formation of sII gas hydrate composed of methane and ethane

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Methane and ethane mixed-gas hydrate of the cubic structure II exists at the central and southern Baikal Basin. We found "double structure gas hydrate" composed of the structure I and II in a same sediment core. The structure II gas hydrate contained 13-15% of ethane, on the contrary, the structure I has only several % of ethane. Subramanian et al. (2000a; 2000b) reported that a structure II forms in appropriate gas composition of methane and ethane. Hachikubo et al. (2009) showed that delta D of hydrate-bound ethane in the structure II is smaller than that in the structure I, whereas delta 13C of methane and ethane, and delta D of methane are the same between the structure I and II. It has been unknown how the structure II concentrates light ethane in delta D (hydrogen isotope).
In this study, synthetic mixed-gas (methane and ethane) hydrates were formed and checked isotopic fractionation between phases of hydrate and residual gas. We made a hydrate sample from methane and ethane mixed-gas (85% C1; 15% C2) in a pressure chamber (volume: 120 mL). Before the retrieval of gas hydrate sample, residual gas was also sampled. We measured isotopic compositions (13C and D) of methane and ethane using CF-IRMS. Crystallographic structure of gas hydrate was determined using a Raman spectrometer.
The Raman spectra of C-C stretching mode of ethane in hydrate phase indicated that the sample belonged to structure II. Delta D of hydrate-bound ethane was several permil smaller than that of residual ethane, similar to the behavior of methane delta D in the structure I. Although the mechanism of ethane fractionation at the formation process of the structure II is not fully understood, the results agree with the observation at the Kukuy K-2 mud volcano reported by Hachikubo et al. (2009).
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Effect of water depth on hydration number of natural gas hydrate in Lake Baikal

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Natural gas hydrates exist under the deep sea/lake or permafrost are considered to be a potential natural gas resource. Hydration number "n" of methane hydrate (CH_4 nH_2O) decides the amount of methane in an unit volume/weight of crystal. In the case of ideal full-occupation of hydrate cages, the value of hydration number is 5.75 (Sloan and Koh, 2008). However, researchers have reported that the hydration number is around 6, because small amount of empty cages decrease the free energy and stabilize the crystal. Natural gas hydrates have been retrieved from lake-bottom sediment at Lake Baikal, where the water depths ranged from 450m to 1400m, and their main gas component is methane. Hydration number may change under various pressure condition, but it has not been examined yet. In this study, we report the pressure effect on hydration number of synthetic methane hydrate and natural gas hydrate of Lake Baikal.
Methane hydrate was synthesized under the pressure range between 3 MPa to 20 MPa. Natural hydrate samples were retrieved at the southern Baikal basin (Malenky, Bolshoy, Peschanka P-2, and Goloustnoye G-1) and central Baikal basin (Kukuy K-1, K-2, K-8, K-9, K-10, and Novosibirsk). Raman spectroscopic measurements were made to assess the hydration numbers of samples. Raman spectra were obtained at 123 K in the range 2,800-3,000 cm^{-1} for the C-H stretching peaks of methane, and fitted using a Voigt function to obtain the integrated intensities of the two peaks corresponding to methane encaged in large and small cages of the cubic structure I. The cage occupancies and the hydration numbers were estimated from these peak intensities using a statistical thermodynamic model (Sum et al., 1997). Hydration number of synthetic methane hydrate decreased with pressure, from 6.05 (2.7 MPa) to 5.97 (20.9 MPa), and those of natural gas hydrate also decreased slightly with water depth.
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Keywords: hydrate, hydration number, Lake Baikal, Raman spectroscopic analysis

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Volume proportion of gas hydrate evaluated from oxygen isotope of water in locus sub-samples

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Gas hydrate distributed offshore around the Japanese Island is actively studied for evaluating its resource. However, the evaluation is not always easy because gas hydrate rapidly decomposes in a low-pressure onboard condition. We contrive a new method using oxygen isotope ratio of locus sub-samples of muddy sediment and hydrate. Together with the isotope ratio of the bulk pore-water, we can calculate volume proportion of gas hydrate.
This study was conducted as a part of the shallow methane hydrate exploration project of METI. During the expedition with the research vessel Poseidon in August-October 2015, we analyzed mixed sediment of hydrate and mud from 26 core sections drilled from Oki trough and Joetsu Basin. From each core section, we quickly encapsulated locus sub-samples of mud and hydrate (3-5 sub-samples for each) in 4.5-ml vials. Air in the vial was later substituted in laboratory by He for the mud sub-sample, by He-CO2 for the hydrate and bulk pore-water sub-samples. After leaving more than 2 days in a constant temperature (at 23 degrees), me measured the isotopic ratio by Finnigan DeltaPlus with GasBench II (2SD = 0.15‰).
Mud sub-samples generally recorded larger deviation in oxygen isotopic ratio than hydrate sub-samples. This is because of difficulty in avoiding contamination of hydrate-molten water. After excluding the contaminated sub-samples, we found that oxygen isotopic ratio was always higher in hydrate than in mud, likely reflecting isotopic fractionation during hydrate formation. Difference between the hydrate value and the mud value was site-specific ranging from 1 to 4 permil, and tended to decrease with increasing the volume proportion of hydrate. The oxygen isotope ratios of water in locus sub-samples successfully provided the hydrate volume proportion from 28 core sections, which appear consistent with images observed onboard.

Keywords: Oxygen isotope, Gas hydrate

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Isolation of methanogenic archaea and distribution of methanogenic and methanotrophic archaea in subseafloor sediment

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Shallow gas hydrates are estimated to be buried around Japan, especially in Japan Sea. The methane trapped in those hydrates are produced by biogenic (microbial) or thermogenic system. But the relationship between shallow gas hydrates and the methanogens are yet to be confirmed. So this study focuses on isolation and diversity of methanogenic and methanotrophic archaea.
Sediment samples were collected from the subseafloor (with or without specific structure) by the MBARI push corer, during an environment assessment cruise. Samples were collected from the top, middle, bottom of the recovered sediments of each push core. The samples were stored in different temperature for the microbiological cultivation experiment and microbiological diversity analysis, respectively.
For the methanogenic archaea isolation, cultivation was carried out by enrichment culture using methanogen medium. The cultures were cultivated by 15℃ and 30℃, respectively. We successfully isolated several methanogenic archaea from the surface sediment. The result of the 16S rRNA gene sequence analysis showed the isolated strains identified as one of the order of the methanogen, Methanomicrobiales.
For the methanogenic and methanotrophic archaea diversity analysis, DNA was extracted from the sediment samples, using ISOIL kit. The methane related functional gene, the *mcr*A gene of methanogenic and methanotrophic archaea was choosen as the target gene. The genes were amplified by PCR method. The PCR products were purified by FastGene Gel/PCR Extraction Kit. The purified products were analyzed by clone library method. The result of the clone library analysis indicated that specific structure of the surface of the subseafloor have specific methanogenic and methanotrophic archaea structure.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: shallow gas hydrate, methanogenic archaea, methanotrophic archaea

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Pyrosequencing of planktonic and benthic biota above the sediment-water interface in methane hydrate-bearing areas in the eastern margin of the Japan Sea

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In this study, we have conducted pyrosequencing analysis of eukaryotic community structures from the water columns and the sediment-water interfaces at sites associated with and without methane seeping and bacterial mats in the area associated with the shallow methane hydrate deposits in the Joetsu Basin, Japan Sea. Dominant 18S rRNA gene sequences from shallow water columns at water depths of ~50 m and ~200 m were related to marine dinoflagellates of the genera *Gyrodinium*, copepods of the genera *Centropages*, radiolarians of the subclass Acantharia. From bottom seawater at water depths of ~1000,m, the community structures were distinct at the sites associated with methane seeping and bacterial mats from that at the reference site. The former structures were abundantly composed of the genera *Nanomia* and *Pantachogon* of Cnidaria phylum. Although marine dragon nematodes of the family Chromadorea were dominantly detected from shallow sediments at the site associated with methane seeping and bacterial mats, the overall community structures from shallow sediments were similar at all investigated sites.
This study was conducted as a part of the shallow methane hydrate exploration project of METI.

Keywords: methane hydrate, 18S rRNA, Pyrosequencing, Eukaryotic communities, DNA

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Investigation microbial community for gas-hydrate site off-Sakhalin Island

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Gas hydrates (GH) are widely spread in the sediments under the subsea floor and form at high pressure and low temperature. GH oriented gas were grouped to thermogenic and microbial gas according to composition carbohydrate gas and isotopic molecular weight of methane. The role of microbial communities in the GH sites have been already investigated by several research groups. Isotopic composition of gas hydrate is often use to determine gas derivation such as thermogenic or microbial gases. However, combination study both isotopic gas analysis and microbial diversity have not been performed at all. Recently we successfully obtained mix gas-derived GH core sediment in the western Sakhalin slope off Sakhalin Island by exploration using ultrasonic wave sonar and gravity coring, in SSGH 15 project by using Russian research vessel *Akademik M. A. Lavrentyev*. We will demonstrate that results of investigating molecular and isotropic composition of the sediment gas, sulfate and sodium compositions, and microbial composition of a GH-bearing sediment core obtained in SSGH15.

Molecular composition ratio C_1/C_2+C_3 below SMI were in the range between 116 and 225, while $d^{13}C$ and dD values of methane were in the range of -48.9 and -45.7 permil, and of -165 and -149 permil, respectively. These results indicated that the gas compose of large amount of thermogenic gas and small amount of microbial gas.

Forty two individual clones have successfully analyzed, until we submit this article. Three *Aciduliprofundum* related clones, and three *Methanobrevibacter* related clones were detected. These sequences were clustered into oceanic methanogen in the phylum *Euriarcheota*. This result implied that these archaeon generate microbial methane in the core, and may correspond to decrease isotopic ^{13}C ratio of methane and increase the C_1/C_2+C_3 ratio. Slight amount of sequence in *Crenarcheota*, which may be involved anaerobic methane oxidation (annamox). Interestingly, heterotorophic bacteria in cluster of *Deharococcoidetes*-related *Chroloflexi*, of Candidatus *artibacteria* (named as division JS1/OP9), and of *Planctomycetes* were frequently widespread in the core. The phylum *Chloroflexi* is a lineage for which the class '*Dehalococcoidetes*' was proposed to accommodate the tetrachloroethane respiring coccus *Deharococcides* (1). These bacteria may contribute to decomposition of difficultly degradable organic matters accumulated on deep sea floor. *Planctomycetes* have been often detected, and widespread in methane-seep (2), but the functional characters have been unknown. Recently, candidatus *artibacteria* have been revealed to play significant role as synbiotropic scavenger in artificial methanogenic bioreactor, by using single cell genome analysis (3). According to the study, *atribacteria* may support methanogen and *chloroflexi* through the by-product generation such as acetate, butyrate, and H_2. These results implied that thermogenic and microbial mixed-derived gas composition may be formed by symbiotic metabolism of those species, but not simply generated from inorganic gases such as CO_2 and H_2 by methanogen.

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