様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

# 科学研究費助成事業　　研究成果報告書

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研究課題名（和文）Relationship between crustal degassing and microbial mineralization in active mud volcanoes, hydrocarbon seeps, and other environments

研究課題名（英文）Relationship between crustal degassing and microbial mineralization in active mud volcanoes, hydrocarbon seeps, and other environments

研究代表者
Ｓｎｙｄｅｒ　Ｇｌｅｎ（Snyder, Glen）

東京大学・大気海洋研究所・特任研究員

研究者番号：９０７５１７７７

交付決定額（研究期間全体）：（直接経費）　　3,100,000円

研究成果の概要（和文）：今年、私たちはメタン由来自生炭酸塩（MDAC）とそれらに関連する浸出水のサンプリングと分析を継続しました。日本海沖のサンプルについては、より多くのアーカイブされたサンプルを使用しました。千葉県茂原市の海岸ベースの湧出液でもサンプリングが行われました。前年のサンプリングに加えて、湧出地域でメタンセンサーを使用してリアルタイムのメタン濃度を測定し、実験室で分析された溶存ガスの濃度と比較しました。サンプリングにより、希ガスと炭素の安定同位体を特定し、日本海側と太平洋側のガス湧出量の違いを明らかにすることができました。

研究成果の学術的意義や社会的意義
Methane seeps are an integral part of the Earth's carbon cycle. The anoxic environments associated with them have been used as analogues for early Earth marine habitats. By studying the gas composition and the composition of MDACs we can elucidate the past and present dynamics of these systems.

研究成果の概要（英文）：This year we continued sampling and analyses of methane-derived authigenic carbonates (MDACs) and the seep water associated with them. For offshore Japan Sea samples, we used more archived samples. Sampling was also carried out on shore-based seeps from Mobara, in Chiba Prefecture. In addition to the previous years sampling, we employed methane sensors in the seep area to determine real-time methane concentrations and compare them with those of dissolved gas that was analyzed in the laboratory. The sampling allowed us to determine noble gas and stable carbon isotopes and to highlight the differences between gas seeps in Sea of Japan versus on the Pacific side. We also analyzed samples we had collected from seep sites in Tatar Strait 5 years ago, which provide an indicator of how seep compositions are site specific .

研究分野：Isotope geochemistry

キーワード：Methane　Cold Seep　Noble gas　Sea of Japan　Carbonate　Authigenic　Stable Isotopes　Gas hydrate

１．研究開始当初の背景 Initial background to the study

In a previous study (17K05712) we determined that massive hydrates in Joetsu Basin (Umitaka Spur, Torigakubi Spur, Joetsu Knoll) as well as Oki Trough contained abundant spheroidal microdolomite. The microdolomites we in some cases hollow and contained microbial DNA as well as saline fluid. Gas inclusions were also present, providing the potential for isotopic studies of them as well. Prior to this study, similar spheroidal microdolomites were found in saline lagoons of Brazil, suggesting that the presence of saline waters was essential to the growth and formation of microdolomites.

２．研究の目的 Objectives of the study

A) To search for microdolomites in other methane-rich settings in Japan, including Mobara seeps on land in Chiba Prefecture, and in gas-hydrate settings of Tatar Strait, offshore Sakhalin. To compare these with archived samples from Brazil lagoons.
B) To look at the structure of the microdolomites and elucidate how they were formed
C) To study the internal dark material in some of the microdolomites to see its relationship with microbially mediated precipitation
D) To study the isotopic composition of gas inclusions to determine if they provide valuable information
E) To investigate whether the microdolomite chemistry, inclusions, gas composition, etc. is quantitatively different from methane derived authigenic carbonates (MDACs) which form around methane seeps and gas hydrates, but as large nodules or cemented sediment.

３．研究の方法 Research methods

A) To study the cores and external cements of the microdolomites, thin-foil sections were prepared and analysed using STXM and TEM.
B) Gas inclusion chemistry and isotopic composition was determined by crushing 1g-2g of microdolomites or MDAC chips under a vacuum, separating cryogenically, and analyzing through noble gas mass spectrometry.
C) Mineralogy of MDACs and microdolomites was determined using X-Ray diffraction.

４．研究成果 Research results

A) The microdolomites can be classified into several different types, depending on the internal composition and external cement. These range from polygonal type microdolomites with dark interiors to isopachous-cloudy types which contain internal dolomite polygons. The dark polygonal type contains fibriform pores and amorphous calcium carbonate (ACC). The microdolomites with cloudy cores contain nanocrystalline spherical crusts. Further discussion can be found in Shiraishi et al.(2023).
B) Despite repeat sampling of sediments around gas vent sites associated with springs in Mobara, no microdolomites could be found. Because the seeps are associated with freshwater, apparently the conditions are not appropriate for microdolomite formation, even in shallow subsurface sediments. Because of this, the focus of our study of gas inclusions moved to look primarily at the samples that we had from Joetsu Basin MDACs, and MDACs from Ofshore Sakhalin, Tatar Strait and compared them with the microdolomite samples, also from Joetsu Basin and from Brazil lagoons.
C) Gas chemistry and noble gas isotopic composition of microdolomites and MDACs from Joetsu Basin, Tatar Strait, and Brazil were all significantly different from air or air-saturated-water, demonstrating that even though both types of carbonate exhibit porosity, the permeability is low enough to effectively retain gas that can be analyzed. Furthermore, the gas composition is unique to each site, suggesting a potential to study the carbonates for gas composition in lieu of the many cases where gas hydrate or seafloor gas bubbles cannot be recovered for analysis. In the case of helium isotopes, Joetsu Basin MDACs are similar to those of the gas hydrate gases collected previously. The MDACs have 3He/4He isotopic ratios indicative of a deep mantle source for gas chimney helium. Burial of the authigenic carbonates, however, has resulted in a slight accumulation of radiogenic 4He within the carbonate, lowering the 3He/4He ratios. On the other hand, the shallow MDAC samples from Tater Strait have radiogenic 3He/4He and 40Ar/36Ar ratios, indicative of migration of deep fluids in contact with a radiogenic crustal source. The lagoon dolomites from Brazil show atmospheric 40Ar/36Ar ratios as would be expected, however the 3He/4He ratios are unusually high and may reflect the decay of anthropogenic tritium in shallow groundwater at the time of precipitation.

D) Both MDACs from Joetsu Basin and those from Tatar Strait consist primarily of aragonite. In some cases, minor amounts of barite are also present, showing that precipitation occurred near the Sulfate Methane Interface. Although the Joetsu Basin MDACs were formed in close proximity to the hydrates which host internally the microdolomites, they differ completely in mineralology. Residual saline water trapped in pockets of the massive gas hydrate (sometimes several meters thick), accounts for this difference. Gas hydrate is also present in Tatar Strait, but in thin laminar and granular occurences which have not been observed to host microdolomite.

５．主な発表論文等

〔雑誌論文〕　計5件（うち査読付論文　4件／うち国際共著　4件／うちオープンアクセス　3件）

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|---|---|
| １．著者名<br>Snyder, G.T., Yatsuk, A., Takahta, N., Shakirov, R., Tomaru, H., Tanaka, K., Obzhirov, A., Salomatin, A., Aoki, S., Khazanova, E., Maryina, E., Sano, Y, Matsumoto, R. | ４．巻<br>10 |
| ２．論文標題<br>Ocean Dynamics and Methane Plume Activity in Tatar Strait, Far Eastern Federal District, Russia as Revealed by Seawater Chemistry, Hydroacoustics, and Noble Gas Isotopes | ５．発行年<br>2022年 |
| ３．雑誌名<br>Frontiers in Earth Science | ６．最初と最後の頁<br>1-21 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.3389/feart.2023.1188142 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

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|---|---|
| １．著者名<br>Syrbu, N.S., Snyder, G.T., Shakirov, R.B., Kholmogorov, A.O., Zharkov, R.V., Tsunogai, U. | ４．巻<br>431 |
| ２．論文標題<br>Geochemical distribution of helium, hydrogen, carbon dioxide, and methane in Sakhalin Island mud volcanoes, hot springs, and cold seeps | ５．発行年<br>2022年 |
| ３．雑誌名<br>Journal of Volcanology and Geothermal Research | ６．最初と最後の頁<br>1-11 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.1016/j.jvolgeores.2022.107667 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスではない、又はオープンアクセスが困難 | 国際共著<br>該当する |

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|---|---|
| １．著者名<br>Yatsuk, A., Gresov, A., G.T. Snyder | ４．巻<br>10 |
| ２．論文標題<br>Hydrocarbon Gases in Seafloor Sediments of the Edge Shelf Zone of the East Siberian Sea and Adjacent Part of the Arctic Ocean | ５．発行年<br>2022年 |
| ３．雑誌名<br>Frontiers in Earth Science | ６．最初と最後の頁<br>1-19 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.3389/feart.2022.856496 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

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|---|---|
| １．著者名<br>Shiraishi Fumito、Akimoto Takayuki、Tomioka Naotaka、Takahashi Yoshio、Matsumoto Ryo、Snyder Glen T. | ４．巻<br>11 |
| ２．論文標題<br>Microbial traces found in microdolomite associated with seep-related shallow gas hydrate | ５．発行年<br>2023年 |
| ３．雑誌名<br>Frontiers in Earth Science | ６．最初と最後の頁<br>1-9 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.3389/feart.2023.1188142 | 査読の有無<br>有 |
| オープンアクセス<br>オープンアクセスとしている（また、その予定である） | 国際共著<br>該当する |

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| 1．著者名<br>MATSUMOTO Ryo、HIRUTA Akihiro、OI Takeshi、KAKIZAKI Yoshihiro、SHIMONO Takaya、ISHIDA Naoto、SAEIDI-ORTAKAND Mahsa、SNYDER Glen、TOMARU Hitoshi、FUKUDA Akari、HIROMATSU Mineo、OHKAWA Shiro、YANAGIMOTO Yutaka、TANAHASHI Manabu、KAKUWA Yoshitaka | 4．巻<br>133 |
| 2．論文標題<br>Overview: Distribution, Occurrence, and Origins of Methane Hydrates around the Japan Islands | 5．発行年<br>2024年 |
| 3．雑誌名<br>Journal of Geography (Chigaku Zasshi) | 6．最初と最後の頁<br>63～89 |
| 掲載論文のDOI（デジタルオブジェクト識別子）<br>10.5026/jgeography.133.63 | 査読の有無<br>無 |
| オープンアクセス<br>オープンアクセスではない、又はオープンアクセスが困難 | 国際共著<br>－ |

〔学会発表〕　計3件（うち招待講演　2件／うち国際学会　0件）

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| 1．発表者名<br>Snyder, G., Takahata, N., Bowden, S.A., Zhang, N., Shakirov, R.B., Tomaru, H., Shiraishi, F., Sano, Y., Matsumoto, R. |
| 2．発表標題<br>Hydrocarbon seep dynamics revealed by noble gas and stable isotope analysis of gas inclusions of authigenic carbonates: A comparison of samples from Tatar Strait, Russia; Joetsu Basin, Japan; and Brejo do Espinho, Brazil. |
| 3．学会等名<br>Japan Geoscience Union Meeting, 2022（招待講演） |
| 4．発表年<br>2022年 |

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| 1．発表者名<br>Shiraishi, F., Akimto, T., Tomioka, N., Takahashi, Y., Matsumoto, R., Snyder, G. |
| 2．発表標題<br>Characteristics of microdolomite associated with gas hydrate |
| 3．学会等名<br>Japan Geoscience Union Meeting, 2022 |
| 4．発表年<br>2022年 |

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| 1．発表者名<br>Snyder, G., Matsumoto, R., Tomaru, H., Shiraishi, F., Miki, S., Sano, Y., Nakajima, M.-T. |
| 2．発表標題<br>Project COESS and the tracing of seafloor volatiles: An action plan for the UN Decade of Ocean Science for Sustainable Development |
| 3．学会等名<br>Japan Geoscience Union Meeting, 2023（招待講演） |
| 4．発表年<br>2023年 |

〔図書〕　計0件

〔産業財産権〕

〔その他〕

From Cold Seeps to Hydrothermal Vents
https://www.frontiersin.org/research-topics/47648/from-cold-seeps-to-hydrothermal-vents-geology-chemistry-microbiology-and-ecology-in-marine-and-coastal-environments

6．研究組織

| | 氏名<br>（ローマ字氏名）<br>（研究者番号） | 所属研究機関・部局・職<br>（機関番号） | 備考 |
|---|---|---|---|
| 研究分担者 | 柿崎　喜宏<br>(Kakizaki Yoshihiro)<br>(20570633) | 東京大学・大学院理学系研究科（理学部）・特任研究員<br>(12601) | |
| 研究分担者 | 白石　史人<br>(Shiraishi Fumito)<br>(30626908) | 広島大学・先進理工系科学研究科（理）・准教授<br>(15401) | |
| 研究分担者 | 松本　良<br>(Matsumoto Ryo)<br>(40011762) | 明治大学・研究・知財戦略機構（生田）・研究推進員（客員研究員）<br>(32682) | |
| 研究分担者 | 戸丸　仁<br>(Tomaru Hitoshi)<br>(80588244) | 千葉大学・大学院理学研究院・准教授<br>(12501) | |
| 研究分担者 | 高畑　直人<br>(Takahata Naoto)<br>(90345059) | 東京大学・大気海洋研究所・助教<br>(12601) | |

６．研究組織（つづき）

| | 氏名<br>（ローマ字氏名）<br>（研究者番号） | 所属研究機関・部局・職<br>（機関番号） | 備考 |
|---|---|---|---|
| 研究分担者 | 伊地知　雄太<br>(Ijichji Yuta)<br>(10911258) | 東京大学・大学院理学系研究科（理学部）・特任研究員<br>(12601) | |

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

| 共同研究相手国 | 相手方研究機関 |
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