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The features of methane fluxes in the western and eastern Arctic: A review. Part I

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Abstract. The article provides a review of the modern researches on methane content and its emissions into the atmosphere in the Arctic region. We discussed various methane sources and summarized the certain existing data of its origins as well as driving forces of the methane upward and lateral migration. The greenhouse gas flux of methane from the Arctic marginal seas plays a significant climatic, geopolitical, and social role, but remains one of the most debated topics in ocean sciences.The Arctic seas are presented today in the literature both as a threat of a global ecological catastrophe due to methane emissions, and as sources of gigantic deposits of the fossil carbon, including coal, permafrost strata, oil-gas and gas hydrates storages, rivers runoff, and as the most sensitive indicator of regular (evolutionary) processes of climate change. Large amounts of organic matter are stored in permafrost on land and under the sea that have been partly and further will be degraded to CO_2 and CH_4. Reviewed studies suggested that the Arctic is a substantial source of CH_4 to the atmosphere (between 32 and 112 Tg(CH_4) yr^{-1}), primarily because of the large area of wetlands throughout the region. A recent assessment of the Arctic region identified thousands of gigatonnes (1 Gt = 10^{15} g) of stored carbon, including unresearched deposits of methane, stored within permafrost and as gas hydrate. We concluded that methane sources and the pathways of its transportation in sediments and into the water column of the Arctic seas are characterized by the extreme ambiguity of existing estimates, due to the complexity of natural gas genesis and its migration mechanisms (diffusion, filtration, bubble gas fluxes). These differences illustrate that we currently cannot predict changes of the methane emissions from the Arctic, as too many unknowns and too large uncertainties persist. Although release of CH_4 to the ocean and atmosphere has become a topic of discussion, the region remains sparingly explored. Submarine permafrost is still poorly studied, mainly due to the lack of direct observations. Objective assessment of the methane distribution and dynamics of its oxidation patterns in sediments and water column in the Arctic seas requires further studies based on the integrated marine expeditions, remote sensing and onland gas monitoring stations. Authors are experienced in methane flux and resources research in Arctic region since 1976th. The study is one of the important topic for planning of future research in the Arctic region, since Russian Federation will be in charge of International Arctic Council (a high level intergovernmental forum) for 2021–2023.

Keywords: methane, climate change, gas hydrate, permafrost, microbial methane turnover, microbial methane oxidation, seismo-tectonic pathways, methane emission, Arctic seas.

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Особенности потоков метана в западной и восточной Арктике: обзор. Часть I

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Реферат. В статье приведен обзор современного состояния исследований содержания метана и его выхода в атмосферу в Арктическом регионе. Представлены различные источники метана, и рассмотрены немногие существующие данные о его происхождении. Поток парникового газа метана из окраинных арктических морей играет значительную климатическую, геополитическую и социальную роль и остается одной из наиболее обсуждаемых тем в науках об океане. Арктика является наиболее чувствительным индикатором регулярных (эволюционных) процессов изменения климата. В настоящее время арктические моря представляют угрозу глобальной экологической катастрофы из-за эмиссии метана вследствие глобального потепления и таяния вечной мерзлоты. В Арктике сосредоточены огромные запасы углерода. В вечной мерзлоте на континенте и под водой содержится большое количество органического вещества, которое подвержено процессам разложения до газов CO_2 и CH_4. Существенный вклад в содержание углерода вносит речной сток. Важными источниками метана являются ископаемые углеводороды, включая уголь, нефть, газ, газогидраты, запасы которых, вероятно, огромны. Рассмотрены различные пути поступления метана в окружающую среду, механизмы вертикальной и горизонтальной миграции. По литературным данным, в Арктике возможно выделение CH_4 в атмосферу в диапазоне 32–112 Tg(CH_4) год$^{-1}$, преимущественно благодаря большому количеству болот в регионе. Недавняя оценка позволила выявить в Арктическом регионе тысячи гигатонн (1 Гт = 10^{15} г) накопленного углерода, включая неразведанные залежи метана в вечной мерзлоте и газогидратах.

Очевидно, что существующие оценки метановых источников и путей его переноса в осадках и толще вод Арктического региона характеризуются крайней неоднозначностью, обусловленной сложностью генезиса природного газа и механизмов его миграции (рассеяния, фильтрации, пузырькового переноса). Хотя выход CH_4 в океан и атмосферу является предметом обсуждений, регион мало исследован. Вечная мерзлота недостаточно изучена из-за отсутствия прямых наблюдений. Из-за недостатка данных и большого количества неопределенностей в настоящем невозможно предсказать изменения в эмиссии метана в Арктике. Объективная оценка структуры распределения и динамики окисления метана в отложениях и водной толще в арктических морях требует дальнейших исследований, основанных на изучении региона в комплексных морских экспедициях, дистанционном зондировании и организации станций газового мониторинга на суше. Авторы исследуют поток метана и ведут поиск ресурсов в Арктике с 1976 г. Представленное в статье направление является одной из важных целей для будущих исследований в Арктике в связи с грядущим председательствованием Российской Федерации в Международном Арктическом совете (экологический форум на высоком уровне) в 2021–2023 гг.

Ключевые слова: метан, изменения климата, газогидраты, вечная мерзлота, микробный оборот метана, сейсмотектонические зоны проницаемости, эмиссия метана, арктические моря.

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Introduction

Currently, the discussion about the role of the Arctic seas in the climatic and resource aspects of the planet occupies a leading place. The greenhouse gas flux of methane from the Arctic marginal seas plays a significant climatic, geopolitical, and social role, but remains one of the most debated topics in ocean sciences. The Arctic seas are presented today in the literature both as a threat of a global ecological catastrophe due to methane emissions, and as sources of gigantic deposits of gas hydrates and as the most sensitive indicator of regular (evolutionary) processes of climate change. Different views on the scale and genesis of methane fluxes in the lithosphere–hydrosphere–atmosphere system are common: regardless of the point of view, all studies agree that there are huge, still far from an objective assessment, hydrocarbon resources in the depths of the Arctic basin. A recent assessment of the Arctic region identified thousands of gigatonnes (1 Gt = 10^{15} g) of stored carbon, locked in permafrost, oil-gas reserves, and likely in gas hydrates [Schuur et al., 2015].

However, it is certain that present day warming is amplified in the Arctic Ocean and that the Arctic contains potentially large deposits of methane stored within permafrost and as gas hydrate. There is concern that warming of overlying waters may melt these deposits, releasing CH_4 into the ocean and atmosphere systems [IPCC… , 2013; State of the Climate… , 2017, 2018].

Considering the faster increase of Arctic temperatures compared to the global average, these deposits may constitute important greenhouse gas emissions due to the climate change in the next 100 years. When thawed, these deposits can be released relatively quickly into the atmosphere as greenhouse gases CO_2 and CH_4. The magnitude and timing of these releases have the potential to accelerate climate change beyond what we project from human activities alone [q.v. e.g. Schuur et al., 2015]. Although there are large quantities of methane stored in the Arctic, it is a current debate whether and how fast methane might be released. Using multiple climate models [Lamarque, 2008], predicted an upper estimate of the global seafloor flux of between 560 and 2140 Tg(CH_4) yr^{-1} (Tg = 10^{12} g), mostly in the high latitudes [IPCC… , 2013; State of the Climate… , 2017, 2018; Jackson, 2000].

It was found that hydrates residing in a typical deep ocean setting (4 °C and 1000 m depth) would be stable and in shallow low-latitude settings (6 °C and 560 m) any seafloor CH_4 fluxes would be oxidized within the sediments. The recent discovery of active methane gas venting along the landward limit of the gas hydrate stability zone (GHSZ) on the shallow continental slope west of Svalbard suggests that this process may already have begun, but the source of the methane has not been determined. Both gradual and rapid warming is simulated, and localized gas release is observed for both cases [IPCC… , 2013; Reagan, Moridis, 2009]. Only in cold-shallow Arctic settings (0.4 °C and 320 m) CH_4 fluxes would exceed rates of benthic sediment oxidation. In the longer term there are estimates that between 35 and 940 PgC could be released over several thousand years in the future following a 3 °C seafloor warming. Cold water column temperatures in the high latitudes lead to buildup of hydrates in the Arctic and Antarctic at shallower depths than is possible in low latitudes. A critical bubble volume fraction threshold has been proposed as a critical threshold at which gas migrates all through the sediment column. This hydrate model, embedded into a global climate model, predicts ≈0.4–0.5 °C [Archer et al., 2009].

It was also found [Hunter et al., 2013] that 21st century hydrate dissociation in shallow Arctic waters was comparable in magnitude to [Biastoch et al., 2011], although maximum CH_4 seafloor fluxes were smaller than [Lamarque, 2008], with emissions from 330 to 450 Tg(CH_4) yr^{-1} for RCP4.5 to RCP8.5 [IPCC… , 2013]. The stability of marine hydrates is sensitive to changes in temperature and pressure and once destabilised, hydrates release methane into sediments and ocean and potentially into the atmosphere, creating a positive feedback with climate change. The results indicate that a warming-induced reduction is dominant even when assuming rather extreme rates of sea level rise (up to 20 mm yr^{-1}) under moderate warming scenarios (RCP 4.5). Over the next century modelled hydrate dissociation is focussed in the top ~100 m of Arctic and Subarctic sediments beneath <500 m water depth. Predicted dissociation rates are particularly sensitive to the modelled vertical hydrate distribution within sediments. Under the worst case business-as-usual scenario (RCP 8.5), upper estimates of resulting global sea-floor methane fluxes could exceed estimates of natural global fluxes by 2100 (>30–50 Tg (CH_4) yr^{-1}), although subsequent oxidation in the water column could reduce peak atmospheric release rates to 0.75–1.4 Tg (CH_4) yr^{-1} [Hunter et al., 2013]. Arctic bottom water temperatures and their future evolution, projected by a climate model, were analyzed. Within the next 100 years, the warming affects 25 % of shallow and mid-depth regions containing methane hydrates. Release of methane from melting hydrates in these areas could enhance ocean acidification and oxygen depletion in the water column. The impact of methane release on global warming, however, would not be significant within the considered time span [Biastoch et al., 2011]. Simulations of heat penetration through the sediment suggest that changes in the gas hydrate stability zone will be small on century time scales, except for the high-latitude regions of shallow ocean shelves [Fyke, Weaver, 2006].

Results and discussions

1. The atmospheric CH_4 concentrations

Direct atmospheric measurements of CH_4 of sufficient spatial coverage to calculate global annual means began in 1978. Values for time series of globally averaged CH_4 mole fractions as analyzed by the WDCGG, using statistical methods, show an increasing tendency except for the early 2000s. Atmospheric methane was 1803.2 ppb (1801.2 to 1805.2) in 2011; this is 150 % greater than before 1750. Globally averaged 'pre-industrial' CH_4 in 1750 was 722 ± 25 ppb [IPCC… , 2013]. The increase in annual mean CH_4 from 2016 to 2017 was 6.9 ± 0.9 ppb, comparable to the average growth rate over the past 10 years (+7.1 ± 2.6 ppb yr^{-1}; the uncertainty is the standard deviation of annual increases). Since 1750, CH_4 has increased by ~1128 ppb from 722 ± 15 ppb [State of the Climate… , 2017]. More recently since 2007, atmospheric CH_4 is observed to increase again. Results of measurements from the Advanced Global Atmospheric Gases Experiment (AGAGE) and the Australian Common-wealth Scientific and Industrial Research Organisation (CSIRO) networks were presented by [Rigby et al., 2008]. Values have shown an increase again since 2007, but the mechanism behind this trend is not fully understood. Measurements of atmospheric CH_4 from air samples collected weekly at 46 remote surface sites show that, after a decade of near-zero growth, globally averaged atmospheric methane increased during 2007 and 2008. During 2007, CH_4 increased by 8.3 ± 0.6 ppb. CH_4 mole fractions averaged over polar northern latitudes and the Southern Hemisphere increased more than other zonally averaged regions. In 2008, globally averaged CH_4 increased by 4.4 ± 0.6 ppb; the largest increase was in the tropics, while polar northern latitudes did not increase. The most likely drivers of the CH_4 anomalies observed during 2007 and 2008 are anomalously high temperatures in the Arctic and greater than average precipitation in the tropics [Dlugokencky et al., 2009].

The atmospheric CH_4 concentrations near the ocean surface measured along the two cruise tracks between Qingdao, China and Iceland during July–September 2012 were studied by [Zhang et al., 2017].

CH_4 was observed at very high latitudes, up to 87° N. The mean CH_4 concentration increased from 1849 to 1866 ppbv after the ship passed through the North Pacific Ocean to the Arctic Ocean during cruise track I. Compared with cruise track I, relatively higher CH_4 concentrations during cruise track II were observed, with the mean CH_4 concentration of 1882 ppbv for the whole cruise track. As the ship sailed over the remote water relatively far away from terrestrial and continental shelf regions, methane emissions from degradation of shelf permafrost, destabilization of marine hydrates and wetlands cannot reasonably explain this phenomenon [Zhang et al., 2017].

2. Organic matter and methane in the Arctic

Considering the Arctic Ocean, organic matter and methane from various sources contribute to its carbon content. Several studies brought attention to the storage capabilities of permafrost

[Tarnocai et al., 2009; Schuur et al., 2008; Zimov et al., 2006]. For example, 1700 billion tones of organic carbon have been estimated to be stored in terrestrial soils in the northern permafrost zone [Schuur et al., 2015].

A recent assessment of the Arctic region identified 1000–2000 gigatonnes (Gt = 10^{15} g) of stored carbon, mainly locked in buried plant matter but also including methane bound in gas hydrate that is vulnerable to climate change over the next century [McGuire et al., 2009]. Old and long-term buried material consists of organic matter and pre-existing methane in terrestrial and subsea permafrost, permafrost associated and continental slope gas hydrates, and commercial relevant oil and gas reservoirs. Large amounts of organic matter are stored in permafrost on land and under the sea that have been partly and further will be degraded to CO_2 and CH_4 [Schuur et al., 2015]. Released organic matter from permafrost that degraded by methanogens under anaerobic conditions might have generated a considerable pool of methane. It is unknown how much methane from that source is still trapped within and beneath permafrost soil and sediment. Additional organic matter and methane are transported by some of the largest rivers on Earth and by groundwater discharges into the Arctic Ocean.

Finally, less ice-cover of the ocean leads to extended phytoplankton blooms generating additional organic matter and most likely methane. There are significant uncertainties associated with those stocks [McGuire et al., 2009].

Studies suggest that the Arctic is a substantial source of CH_4 to the atmosphere (between 32 and 112 Tg(CH_4) yr^{-1}), primarily because of the large area of wetlands throughout the region. Analyses to date indicate that the sensitivity of the carbon cycle of the Arctic during the remainder of the 21st century is highly uncertain [McGuire et al., 2009].

The Arctic now represents about 4 % of the global methane budget; 23 vs. 568 Tg(CH_4) yr^{-1} for 2012, according to [Saunois et al., 2016]. This budget is lower than bottom-up estimates (range 37–89 Tg(CH_4) yr^{-1}, according to the review by [Thornton et al., 2016]). Information on the magnitude of methane flux from the Arctic basin to the atmosphere is extremely contradictory: models from insignificant emissions to explosive release, leading to a global catastrophe, are considered. Investigations led by [Shakhova et al., 2010] estimated the total East Siberian Arctic Shelf (ESAS) emissions from diffusion, ebullition, and storm-induced degassing at 8–17 Tg(CH_4) yr^{-1}. A subsequent measurement campaign led by [Thornton et al., 2016], though not made during a stormy period, failed to observe such high rates and estimated an average flux of 2.9 Tg(CH_4) yr^{-1} instead. Other studies supported the lower methane fluxes.

Berchet et al. [Berchet et al., 2016] also found that such high values as reported by [Shakhova et al., 2010] at the ESAS were not supported by atmospheric observations, and suggested the range of 0.0–4.5 Tg(CH_4) yr^{-1} instead. A reference scenario with the ESAS emissions of 8 Tg(CH_4) yr^{-1}, in the lower part of previously estimated emissions, is found to largely overestimate atmospheric observations in winter, likely related to overestimated methane leakage through sea ice [Berchet et al., 2016]. Fenwick et al. observed low sea air fluxes of methane across the western part of the Arctic Ocean. They investigated ~10,000 km transect across contrasting hydrographic environments, from the oligotrophic waters of the deep Canada Basin and Baffin Bay to the productive shelves of the Bering and Chukchi Seas [Fenwick et al., 2017]. The percent saturation relative to atmospheric equilibrium ranged from 30 to 800 % for CH_4, with the highest concentrations occurred in the northern Chukchi Sea.

The differences between the first Arctic estimates of the ESAS and later estimates there and in other regions of the Arctic illustrate large variabilities and thus uncertainties associated with methane fluxes from the Arctic region. This difference demonstrates that we currently cannot predict changes of the methane emissions from the Arctic as too many unknowns and too large uncertainties persist. Although these various sources are generally known, the scaling of the sources appears challenging.

3. The sources of methane in the Arctic

3.1. The permafrost

Permafrost landscapes in northern high latitudes are an important, but poorly known, component of the global carbon cycle [Kittler et al., 2017]. Permafrost is defined as soil, rock, and any other subsurface earth material that exists at or below 0 °C continuously for two or more consecutive years [Osterkamp, 2001]. Permafrost zone soils have accumulated over hundreds and thousands of years.

Subsequent inundation of this area at the Pleistocene/Holocene transition put this loess permafrost carbon under water and also started thawing the permafrost surface [Rachold et al., 2007].

Undersea permafrost carbon initially formed on land as the continental shelf was exposed by sea levels that were 120 meters lower during the last glacial period [Walter et al., 2007].

The exposed organic carbon started to decompose potentially under anaerobic conditions. This would have converted a portion of the carbon pool to CO_2 and CH_4 in the past, leaving an unknown quantity of organic carbon remaining both in the sediment and in permafrost that persists under the ocean.

To put permafrost into perspective: soils from the rest of Earth's biomes (excluding Arctic and boreal biomes) contain 2050 petagrams (Pg = 10^{15} g) of organic carbon in the surface's top 3 meters. Soils from the northern circumpolar permafrost region, that have been quantified, add another 50 % (1025 Pg) to the 0–3 m inventory, even though they occupy only 15 % of the total global soil area [Schuur et al., 2015].

Both terrestrial and sub-seafloor permafrost started to thaw at increasing rates during the last 30 years due to global warming releasing organic matter available for degradation. Arctic temperatures rise faster than the global average [IPCC… , 2013; Overland et al., 2014] and climate models also predict a strong high-latitude warming for the future [IPCC… , 2013]. Arctic temperature rise will affect the local carbon cycle and might liberate an unknown volume of methane via biodegradation of organic matter and dissociation of methane hydrates currently stored within and beneath permafrost as well as along the continental margin. For example, permafrost temperature has increased by +1 to +2 °C in northern Russia during the last 30 to 35 years [State of the Climate… , 2017, 2018]. In 2016, the average annual surface air temperature (SAT) over land north of 60° N was the highest value since reliable records began in 1900. For example, in August 2016, sea surface temperatures (SSTs) were up to 5 °C higher than the 1982–2010 average in regions of the Barents and Chukchi Seas and off the east and west coasts of Greenland [State of the Climate… , 2017, 2018].

3.2. The gas hydrate

While it is clear that there are substantial stocks of carbon in the Arctic, there are significant uncertainties associated with the magnitude of organic matter stocks contained in permafrost and the storage of methane hydrates beneath both subterranean and submerged permafrost of the Arctic [McGuire et al., 2009].

In contrast to terrestrial permafrost, there are no reliable published estimates of total organic carbon inventory for the subsea permafrost pool [State of the Climate… , 2017, 2018]. Substantial quantities of methane are believed to be stored within submarine hydrate deposits at continental margins. Hydrates consist of cages of water molecules that are stabilized by mainly methane. These structures are stable under low temperature and high pressure conditions that define the gas hydrate stability zone (GHSZ). Gas hydrate concentrates CH_4 within its cage-like molecules, with 1 m^3 of gas hydrate sequestering a maximum of 180 m^3 of methane as measured at standard temperature and pressure (STP).

Models and geophysical data indicate that large areas of the Arctic shelves are underlain by subsea permafrost. As a result of their exposure during the last glacial maximum, the shelves are thought to be almost entirely underlain by permafrost from the coastline down to a water depth of about 100 m. Subsea permafrost is still poorly understood, mainly due to the lack of direct observations. Large volumes of methane in gas hydrate form can be stored within or below the subsea permafrost and the stability of this gas hydrate zone is sustained by the existence of permafrost. Degradation of subsea permafrost and the consequent destabilization of gas hydrates could significantly if not dramatically increase the flux of methane to the atmosphere [Rachold et al., 2007]. Ruppel and Kessler believe gas hydrate to be widely distributed in the sediments of marine continental margins and permafrost areas, locations where ocean and atmospheric warming may perturb the hydrate stability field and lead to release of the sequestered methane into the overlying sediments and soils [Ruppel, Kessler, 2017].

Dissociation of gas hydrate deposits may likely accelerate global warming, increase ocean acidification, and exacerbates oxygen loss [Biastoch et al., 2011]. Approximately 1 % or more of global gas hydrates occurs in high northern latitude permafrost areas [Ruppel, 2015]. These permafrost associated gas hydrates (PAGH) occur both onshore beneath tundra (e.g., Russia, Canada, and the U.S.) and on continental shelves of the Arctic Ocean whose permafrost has been inundated by sea level rise since ~15 ka [q.v. e.g. Rachold et al., 2007].

Many permafrost associated gas hydrates (PAGH) formed by a process that can be described in the vernacular as “freezing in place” of gaseous CH_4 that has presumably migrated to shallower depths from underlying conventional gas reservoirs containing thermogenic gas [Ruppel, 2015]. Lacking better well distribution, it is not possible to determine the absolute seaward extent of ice-bearing permafrost, nor the distribution of permafrost beneath the present-day continental shelf at the end of the Pleistocene [Ruppel et al., 2016]. In contrast, ice-bearing subsea permafrost patches were detected during geophysical investigations offshore in the Laptev Sea [Rekant et al., 2015] and reach to 60–100 m isobath in the Canadian Beaufort Sea [Riedel et al., 2017].

Deep-water marine gas hydrates associated with a bottom-simulating reflectors (BSR) were identified in the Canadian Beaufort Sea [Riedel et al., 2017] and in the SW Barents Sea [Vadakkepuliyambatta et al., 2017].

Seismic observations of BSRs revealed significant thermogenic gas input into the hydrate stability zone throughout the SW Barents Sea [Vadakkepuliyambatta et al., 2017]. The Barents Sea is a major part of the Arctic where the Gulf Stream mixes with the cold Arctic waters. Late Cenozoic uplift and glacial erosion have resulted in hydrocarbon leakage from reservoirs, evolution of fluid flow systems, shallow gas accumulations, and hydrate formation throughout the Barents Sea [Vadakkepuliyambatta et al., 2017].

Continental slopes north of the East Siberian Sea potentially hold large amounts of methane (CH_4) in sediments as gas hydrate and free gas. Gas seepage offshore Svalbard was postulated to result from gas hydrate dissociation, possibly triggered by anthropogenic ocean warming. Observations of CH_4 release along the Svalbard margin seafloor [Westbrook et al., 2009] suggest observed regional warming of 1 °C during the last 30 years is driving hydrate dissociation, an idea supported by modelling [Reagan, Moridis, 2009]. However, large-scale leakage, reported by Mau et al., is not caused by anthropogenic warming. The much broader seepage area, extending from 74° to 79° N, from 5° to 25° E, where more than a thousand of gas discharge sites were imaged as acoustic flares, occurs in water depths at and shallower than the upper edge of the gas hydrate stability zone [Mau et al., 2017].

Miller et al. [Miller et al., 2017] presented results of chemical analysis of pore water from 32 sediment cores taken during Leg 2 of the 2014 joint Swedish–Russian–US Arctic Ocean Investigation of Climate–Cryosphere–Carbon Interactions (SWERUS-C3). The cores come from depth transects across the slope and rise extending between the Mendeleev and the Lomonosov ridges, north of Wrangel Island and the New Siberian Islands, respectively. Miller et al. noted that abundant CH_4, including gas hydrates, do not characterize the East Siberian Sea slope or rise along the investigated depth transects, except for one station on the Lomonosov Ridge [Miller et al., 2017].

To date, the northen most submarine gas hydrate are found in the upper part of the western slope of the Chukchi Plateau within the deep-water eastern margin of the East Siberian Sea [ARA07C Cruise Report…, 2017]. They were recovered at a depth of 610 meters on the 3 meters bsf in two sediment cores (station section ARA07C GC13, coordinates 75.6795° N, 169.7379° E) on local morphostructures (mounds). Such morphostructures, as a rule, are formed above gas-saturated channels (gas chimney) in the upper part of sedimentary strata above gas-bearing or oil-and-gas bearing structures. The gas pockmarks and methane anomalies were mapped earlier in this region [Savvichev et al., 2004]. This gashydrate accumulation is, likely, related to the the rift.

3.3. The coal deposits

Another hardly known source of methane are coal deposits that extend over large regions deposits. The studies of Russian scientists established the patterns of relationship between permafrost degradation in the regions of continental arctic coal deposits, including degradation caused by migration of methane from coal-bearing column (fig. 1) [Obzhirov, 1979; Gresov et al., 2014, Gresov et al., 2017]. The six basic regularities of the distribution of natural gas and permafrost, gas cryological zonality, gas composition and gas permeability of coal formations in permafrost have been determined.

Methane content trapped in coal basins is projected up to 11 trillion tons in Russian Far East. According to e.g. [Gresov et al., 2014], some of the coal basins continue under the seafloor of the Eastern Arctic.

Gresov et al. established that the areas of projected gas-oil, oil and gas deposits on the shelf of the East Siberian Sea are characterized by minimal methane and hydrocarbon content of the bottom sediments (less than 0.05 and 0.001 cm^3/kg, respectively) as well as great thickness of the sedimentary cover within the North Chukotka (more than 10 km), South Chukotka, and Aion Basins (more than 3 km) [Gresov et al., 2017].

They determined that abnormal gas geochemical fields are formed within rises of small thickness of the sedimentary cover and active faulting and tectonic disturbance; these fields are, by nature, regions of gas discharge.

They showed that the main geochemical markers and indicators of the oil and gas content of the East Siberian Sea shelf are the molecular mass of the hydrocarbon fractions, the carbon isotopic composition of methane in the bottom sediments, and the sedimentary cover thickness.

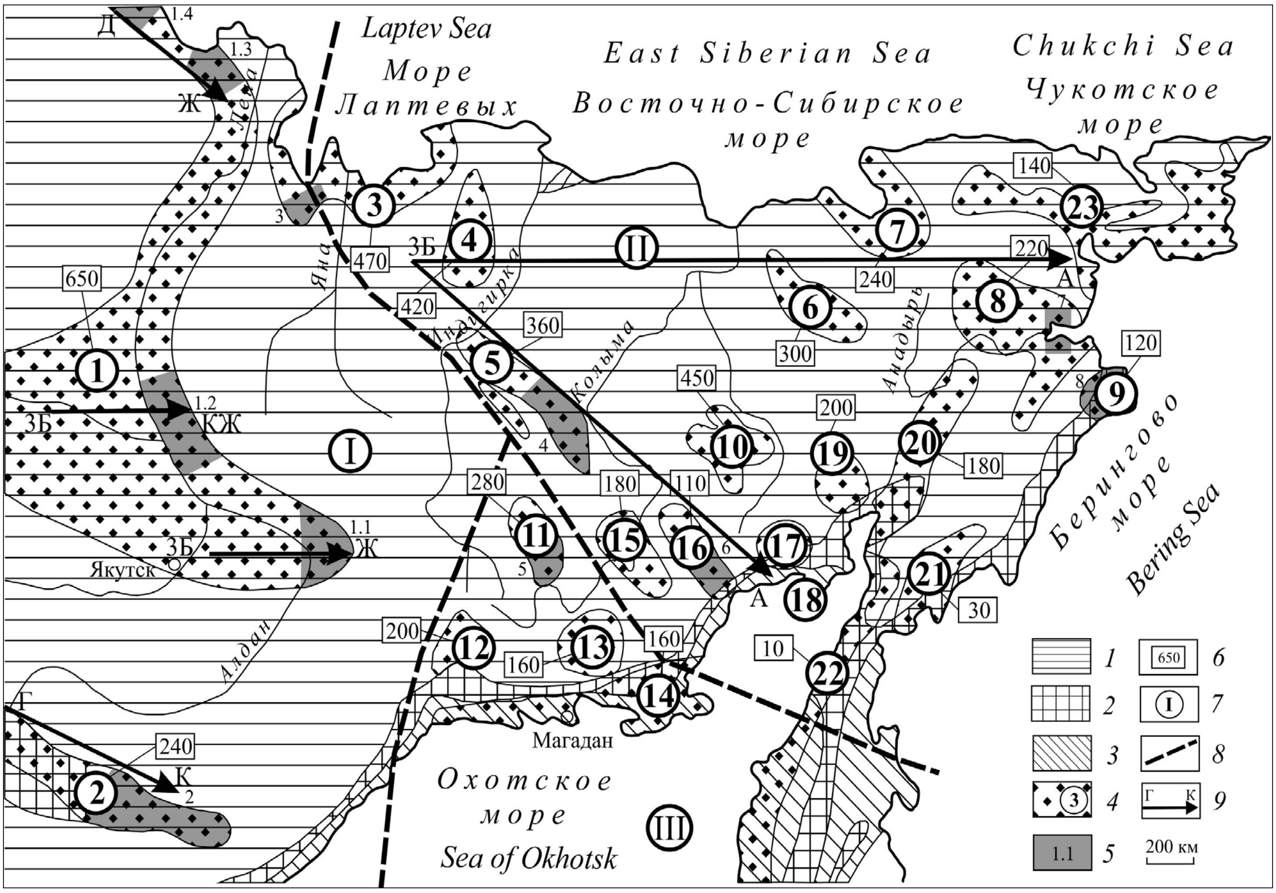

Fig. 1. Permafrost types and their distribution in the North-Eastern Russia, Arctic. 1–3 – permafrost: 1 – continuous type, 2 – faltering type, 3 – massive-island type; 4 – coal basins: 5 – coal-methane active areas; 6 – thickness of permafrost, m; 7 – lithosphere plates; 8 – lithosphere plates borders; 9 – directions of coal methamorphic changes. [Gresov et al., 2014]

More than 15 lignite and brown coal beds with thickness ranging from 7 to 10 m and methane content of 2 m^3/ton were established at depths of 200–250 m in the Chaun Lowland of the Aion Basin. Here, in seven wells, ten appearances of gas were registered, related to drilling into brown coal beds and Cretaceous intrusions in the tectonic zones with methane concentrations ranging from 20.7 to 77.5 % and a gas rate up to 0.05 m^3/min. Within the south part of the East Siberian Sea, the bottom sediments of the central part of the Aion Basin are characterized by gas geochemical indicators of oil- and gas content with MHC > 19 g/mol and carbon isotopic composition of methane ranging from –42.7 to –53.4 ‰ VPDB. The characteristics of the hydrocarbon gases of the bottom sediments of the Chauna Lowland are close to the characteristics of the coal-gas and magmatic formations [Gresov et al., 2017].

3.4. The oil-gas deposits

In addition to permafrost and gas hydrates, many of world's largest gas fields are north of the Arctic cycle. Oil and gas seepages were found offshore Scott Inletin Baffin Bay [Grant et al., 1986]. Gautier et al. estimated that about 30 % of the world's undiscovered gas and 13 % of undiscovered oil may be preserved in the Arctic. Gautier et al. suggest that two-thirds of the undiscovered gas is in just four AUs: South Kara Sea (607 TCF), South Barents Basin (184 TCF), North Barents Basin (117 TCF), and the Alaska Platform (122 TCF). The South Kara Sea, the offshore part of the northern West Siberian Basin, contains almost 39 % of the undiscovered gas and is the most prospective hydrocarbon province in the Arctic. Although substantial amounts of gas may be found in Alaska, Canada, and Greenland, the undiscovered gas resource is concentrated in Russian territory [Gautier et al., 2009].

3.5. River removal of methane

3.5.1. Fresh water

Additional organic matter and methane are transported by some of the largest rivers on Earth and by groundwater discharges into the Arctic Ocean. The Arctic Ocean is the most river-influenced and landlocked of all oceans [Charkin et al., 2017]. Huge rivers empty into the Arctic Ocean, carrying vast amounts of sediment that can be seen from space as immense swirls in the coastal region [Parmentier et al., 2017].

About 10 % of global runoff flows into the large areas of shallow Arctic shelf seas [Lammers et al., 2001].

One of the most obvious implications of the observed warming is that river run off will increase [Peterson et al., 2002]. Discharge was correlated with changes in both the North Atlantic Oscillation and global mean surface air temperature. Fresh water (FW) supply and together with organic matter from the continental land mass is of special importance to the Arctic Ocean, which contains only 1 % of global ocean water, yet it receives 11 % of global river run off [Shiklomanov et al., 2000]. During 1964–2000, the discharge to the Arctic Ocean has increased by 5.6 $km^3\ yr^{-1}$, mostly due to a large increase from the Eurasian rivers [McClelland et al., 2006]. The average annual discharge of FW from the six largest Eurasian rivers to the Arctic Ocean increased by 7 % from 1936 to 1999. The average annual rate of increase was 2.0–0.7 $km^3\ yr^{-1}$. The observed large-scale change in FW flux has potentially important implications for ocean circulation and climate [Peterson et al., 2002]. There have been observations of a 7 % increase in organic matter discharge from Eurasian rivers to the Arctic shelf over recent decades.

Although, river borne export of CH_4 via the Bykovskaya Channel, which is the main outflow of the Lena River, was not observed [Shakhova et al., 2010], creeks draining from the permafrost in the same region were found to contain 1000 times higher concentrations of methane [Bussmann, 2013] than in the open ocean (2 nM).

The HRS data also revealed abundance of the gas seeps in the study area. Most of them mark the local permeable zones within the permafrost, which are most likely former thermokarst depressions (lakes/taliks). The "fuzzy"-facies of the gas seep anomalies are concentrated along the Lena and Yana paleoriver valleys and therefore may relate to river taliks [Rekant et al., 2015].

According to these few results, rivers transport organic matter as a prerequisite for methane production and creeks from permafrost directly add methane to the atmosphere and to the river system. How large either source is, is currently unknown.

3.5.2. The submarine groundwater discharge

Another important transport pathway of organic matter and pre-formed methane to the sea might be submarine groundwater discharge (SGD). SGD is a mixture of fresh groundwater and seawater that recirculates through the subterranean estuary as a result of tides and wave action, and then discharges to the ocean [Moore, 1999].

Active SGD was documented in the vicinity of the Lena River delta. Groundwater (GW) currently comprises almost one-quarter of Yukon River water discharged to the Bering Sea, which subsequently is transported into the Arctic Ocean via the Bering Strait [Walvoord, Striegl, 2007].

In the coastal areas of the shallow Siberian Arctic seas, where permafrost was submerged most recently, taliks (layers or columns of thawed sediments within permafrost) might form as a result of the combined effect of geothermal flux from fault zones, the warming effect of rivers and overlying seawater. These taliks could be gas-charged and connected to SGD [Shakhova et al., 2017], which could be manifested as point sources of methane to the coastal waters.

SGD discharge in the Siberian Arctic seas depends on the thermal state of the permafrost as well as on the geological and tectonic structure of the shelf. The geological prerequisites for subpermafrost GW discharge include the presence of lithological conditions (sand, gravel, cracks and fissures in rocks) and channels (taliks) between the subpermafrost GW (confined aquifer) and the marine water column. At fault crossings: (1) there is an increased crushing or jointing of rock masses, which is favorable for uprising SGD transport; and (2), the impact of geothermal heat flux is increased, which thaws the permafrost [Charkin et al., 2017].

4. The origin of methane

The origin of methane of all above sources is either biogenic or thermogenic. Methane is either generated under reduced conditions in anoxic marine sediments, predominantly through microbially mediated CO_2 reduction and disproportionation of methylated substrates [Hinrichs, Boetius, 2002] or formed by thermal breakdown of organic matter at high temperature and pressure.

Isotopic observations suggest a biogenic origin (either terrestrial or marine) of the methane in air masses originating from the ESAS during late summer, 2008 and 2009 [Berchet et al., 2016].

In the Beaufort Sea, the sources of methane available for release into the water column are primarily from microbial degradation of sedimentary organic matter and secondarily from thermogenic gas seepage [Lorenson et al., 2016].

In the offshore Prudhoe Bay and the Mackenzie River delta, microbial methane sources predominate with minor influxes from thermogenic methane and may include methane from gas hydrate [Collet, 2014].

Offshore western Svalbard, stable carbon isotopic compositions ($\delta^{13}C$) of methane coupled with a virtual lack of any higher hydrocarbons point to microbial generated methane in water samples from the so-called Svalbard plume, but a thermogenic origin cannot be ruled out [Mau et al., 2017].

Both, stable hydrogen and carbon isotope data revealed the predominant occurrence of biogenic methane being dissolved in pore water of partially thawed subsea permafrost of the ESAS [Sapart et al., 2017]. Sapart et al. demonstrate that at locations where a thick marine clay layer is present, this CH_4 is partially oxidized before reaching the seawater. However, at locations where ebullition was observed from the seabed, no oxidation was identified in the stable isotope surface sediment profile. In that case, and considering the very shallow water column (<10 m) in this area, this microbial gas will likely reach the atmosphere when sea ice is absent. Triple isotope dataset of CH_4 from the sediment and water of the shallow ESAS reveals the presence of CH_4 of microbial origin formed on old carbon with unexpectedly low stable carbon ($\delta^{13}C$ as low as 108 ‰) and hydrogen (D as low as 350 ‰) isotope signatures down to about 50 m under the seabed in the thawed permafrost. Sapart found high concentrations (up to 500 μM) of CH_4 in the pore water of the partially thawed subsea permafrost of this region. For all sediment cores, both hydrogen and carbon isotope data reveal the predominant occurrence of CH_4, that is not of thermogenic origin as it has long been thought, but resultant from microbial CH_4 formation [Sapart et al., 2017].

In contrast, $\delta^{13}C$ of methane in the sediments of 45 station of the coastal ESS revealed clear presence of thermogenic methane and ethane. It appears, that biogenic methane sources are dominant in the Arctic [Shakirov, 2018].

5. Methane production and methane consumption

Physical changes such as warming surface waters, reduced permanent ice cover, and thawing of permafrost have been linked to increased net primary productivity [q.v. e.g. Arrigo, van Dijken, 2015]. These changes may increase organic matter export from the surface waters of the Arctic Ocean [Boetius et al., 2013], potentially accelerating the microbial processes that produce CH_4 under low O_2 conditions.

At the Arctic shelf, methane production was so far identified as a source contributing 20 % of methane to the Chukchi Sea methane load [Li et al., 2017].

In this region, emissions from the sediment-water interface and the in situ production of CH_4 were estimated to account for 75 % and 20 %, respectively [Li et al., 2017]. They calculated the budget of CH_4 using a mass balance model and estimated that the emissions from the sediment-water interface and the in situ production of CH_4 are the main sources of CH_4, and that the sea-to-air flux and oxidation of CH_4 (which accounts for 52 % and 43 % of the exports of CH_4) are the major outputs. Analysis of the spatial distribution of CH_4 in the western Arctic Ocean during the summer of 2014 revealed an increasing trend northward toward the shelf break stations and a decreasing trend toward the Canada Basin stations. The surface waters at all of the stations are oversaturated with CH_4, and the mean sea-to-air CH_4 flux in the CSS is 10.08 μmol m^{-2} day^{-1}, although the CSS accounts for 0.16 % of the surface area of the world ocean, it accounts for 0.30 % of total global CH_4 emissions [Li et al., 2017].

Study results indicate that the in the South Chukchi Basin diffusive methane fluxes at the sediment-water interface within the southern and northern sites were estimated to be 14.5 μmol dm^{-2} day^{-1} and 0.7 nmol dm^{-2} day^{-1}, respectively [Matveeva et al., 2015].

Although methane production [Karl et al., 2008] in the water column is commonly a small source of the greenhouse gas, increased primary production in the Arctic seas might accelerate this source adding to the other sources. An additional, though poorly understood, source of methane is methanogenesis within the aerobic water column, which is thought to occur in sinking particles [Karl, Tilbrook, 1994], within the digestive tracts of organisms [Angelis, de, Lee, 1994], and through the metabolism of methylated substrates [Damm et al., 2010; Karl, Tilbrook, 1994].

This aerobic water column CH_4 production is often masked by biological oxidation, which acts to maintain CH_4 concentrations near atmospheric equilibrium [Hanson R., Hanson T., 1996].

5.1.1. Turnover of methane by microbial oxidation in the sediment and water column and the temperature influence on this process

It is thought that this organic material is vulnerable to biodegradation, but Arctic microorganisms are used to low temperatures and need to adapt to elevated temperature and increased organic matter from land and from the surface ocean for degradation. Due to the increased sedimentation rate, most organic matter might be buried and slowly anaerobically biodegraded. Anaerobic oxidation can account for up to 80 % of methane consumption in sediments [Reeburgh, 2007]. The global methane budget of the ocean is well balanced by anaerobic and aerobic microbial methane oxidation (at deeper sites >100 m). For example, data collected offshore Svalbard in the summer of 2015, revealed that 0.02–7.7 % of the dissolved methane was aerobically oxidized by microbes and a minor fraction (0.07 %) was transferred to the atmosphere during periods of low wind speeds [Mau et al., 2017].

Oxidation rates in the Arctic vary considerably, which is to a small extent due to ice cover. Specific oxidation rate constants for methane, found in the Beaufort Sea are comparable to estuarine and oxic/anoxic boundary layer values. While these rates are generally higher than typical open ocean rates, water column oxidation rates would account for only 1–2 % of the methane pool available in the water column. Besides, results show that during ice covered periods methane oxidation rates are much higher than in ice-free periods when rates were undetectable. Although the restriction to oxic zones is challenged as aerobic, methane-oxidizing bacteria (MOB) were found to oxidize about 32 % of the methane in anoxic zones in lakes [Martinez-Cruz et al., 2017].

The simple model of a 10 m thick sea water surface layer, proposed by [Kitidis et al., 2010], suggests that methane oxidation accounts for ~37 % of the methane loss during ice-free conditions and up to 46 % during ice-covered periods. Oxidation of CH_4 accounts for 43 % of the exports of CH_4 in the Southern Chukchi Sea shelf [Li et al., 2017].

Depending on the origin and flow path of the water mass, a stock of methanotrophic bacteria might be already in the water and thus might easily adapt to a sudden methane increase. However, the microbial turnover depends also on temperature [Bussmann et al., 2015] and if the bacteria are psychrophiles and thus capable of growth and reproduction in cold temperatures or not.

Most of the seafloor flux of CH_4 is expected to be oxidised in the water column into dissolved CO_2. This fraction depends on the depth of water and ocean conditions [Mau et al., 2007a]. Elliott et al. demonstrated significant impacts of such seafloor release on marine hypoxia and acidity, although atmospheric CH_4 release was small [Elliott et al., 2011].

6. Investigation of methane emission intensity depending on tectonic and seismic conditions

Tectonics and seismicity can significantly affect pathways of methane [Mau et al., 2007a, 2017]. Faults have been found to be major pathways of methane from shallow and deep reservoirs. Tectonic movements along faults are thought to close or open fluid and gas migration pathways. Therefore, correlation between tectonics (passive and active) and gas emissions could identify sensible areas where large amounts of methane can be rapidly emitted [Mau et al., 2007a, 2017].

During analyzing the relationships between the gas fluxes in the marginal seas and seismic activity, consideration must be given to earthquakes occurring not only in the transition zone "continent–ocean", but also on the continent. Shakirov et al. showed that there is a naturally-determined "gas-geochemical response" of geodynamic and seismotectonic processes in the interaction of the lithospheric plates at great distances [Shakirov et al., 2017]. A regular "gas-geochemical response" of seismotectonic processes was revealed in the interaction of lithospheric plates at large distances, using the example of the gas outlets of the Seas of Japan and Okhotsk and Lake Baikal.

Using the example of one of the lineaments of eastern Asia, it was shown that the lineament geotectonic structures composing the regmatic net of the Earth control the formation and activity of the largest centers of methane emission; these centers are indicators of the tectonosphere activity and hydrocarbon accumulation. Changes in the activity of these centers are informative signals of seismic fluctuations. The centers of the heaviest submarine fluxes of natural gas (methane up to 99 vol %), which are removed from each other in the Far East and the Eastern Arctic seas, can be controlled by the same lineaments and by the zones of junction, where lineaments of various geodynamic nature are interlinked.

Tectonic structures evidently influence the continuous methane release from the seafloor in the Eastern Arctic and Sub-Arctic [Shakirov et al., 2017] (fig. 2). Changes in the activity of these centers are informative signals of seismic fluctuations.

In the eastern Arctic sector there are promising areas at depths from 400 m located along the foot of ridges, the sides of tectonic troughs, and the continental slope (areas modified by rift-related structures and by nodes of disjunctive intersections, etc.). In addition to the flares, which were discovered by Russian researchers in the Laptev Sea [Chernykh, 2014], methane in concentrations, sufficient to form hydrates in sediments, is revealed in the Chukchi Plateau [Matveeva et al., 2015] and the East Siberian Sea [Shakirov et al., 2013]. These structures, in terms of permeability of the lithosphere, are

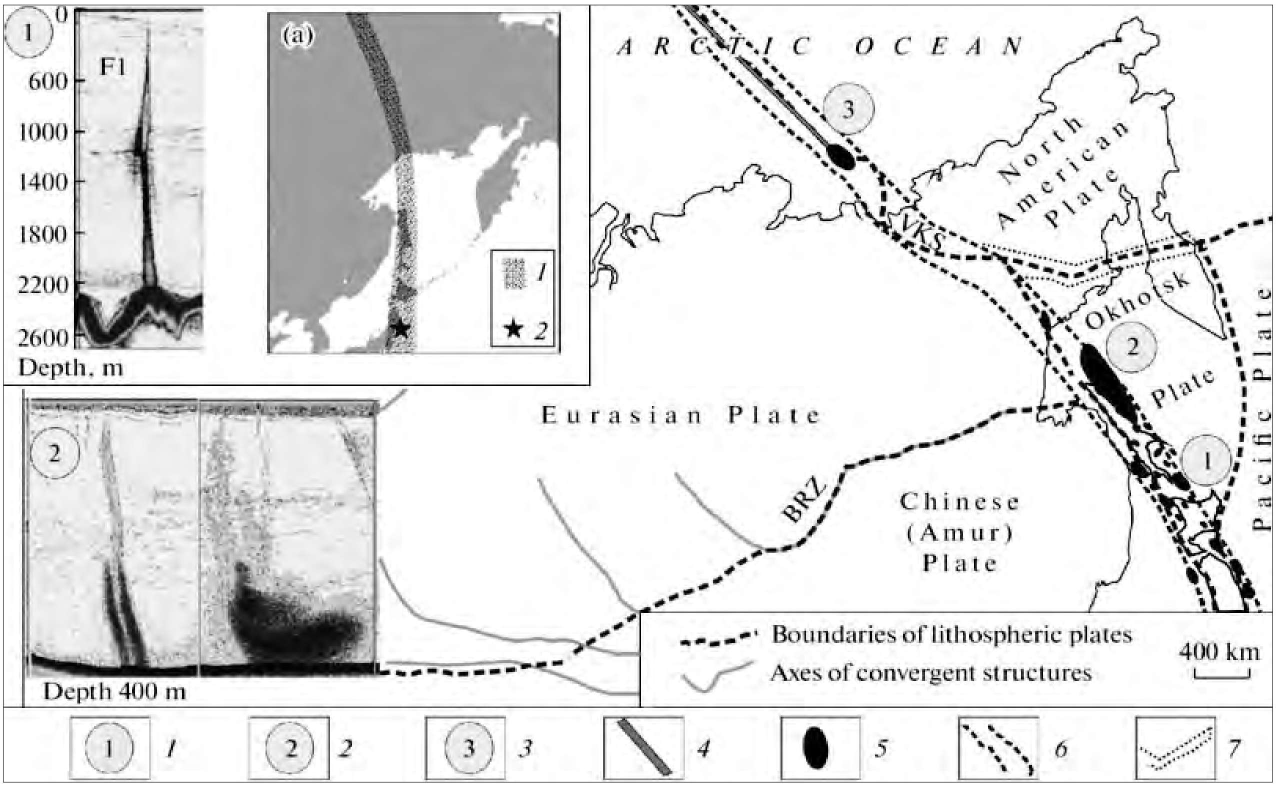

Fig. 2. The map of earthquakes and lineament control of some active centers of natural gas emission. (1) F1 gas flare in the Sea of Okhotsk, (2) "Gizella" gas flare in the Sea of Okhotsk, (3) the field of methane flares in the Laptev Sea [Chernykh, 2014], (4) the Middle Arctic Ridge, (5) the areas of the natural gas flares, (6) the zone of the Okhotsk-Japan Seas lineament and its extension into the Laptev Sea, (7) the northern boundary of the Okhotsk Plate [Ulomov, 2007]. VKS – Verkhoyansk–Kolyma System. BRZ – Baikal Rift Zone. Insert (a): position of the Tohoku Earthquake epicenter (2011) on the scheme of the lineament (1 – lineament, 2 – Tohoku event). [Shakirov et al., 2017]

favorable to form methane hydrates in sedimentary deposits under the appropriate lithological, thermobaric, and geochemical conditions, which have been studied by Russian researchers [Geology and mineral… , 2004; Shakirov et al., 2013; Matveeva et al., 2015].

The article [Shakirov et al., 2013] presented the results of integrated gas-geochemical studies of bottom sediments from comprehensive study of sediments from East-Siberian Sea along the profile from Billings Cape to the Mendellev Ridge. The revealed methane anomalies in sediments up to 2.4 % are controlled by neo-tectonic faults. Isotopic composition of ^{13}C–CH_4 reveals influx of deep fluid. Study of sediment chemical composition allowed distinguishing zones in tectonic faults, revealed by methane anomalies in sediments, where conditions are favourable for concentration of Mn, Cu, and Ag. Figure 3 demonstrates the scheme of gas geochemical studies in the East Siberian Sea [Gresov et al., 2017].

In the south and west of the Chukchi Sea a zone of sediment accumulation was found with a high content of organic carbon, increased background content and anomalies of sulfonic metals (Mo, Zn, Hg, Ag, Au), iron group metals (V, Ni, Co) and some platinoids (Ru, Pt). This zone is confined to the neotectonically active rift system, stretching from the Bering Strait and Eastern Chukotka to the continental slope, where it borders the Cenozoic rift-bearing basin of the Charlie Canadian Basin. The geochemical features of carbon-rich sediments indicate their formation under conditions of lack of oxygen, and, in some cases, in suboxide and anoxide environments near water and gas endogenous sources. The high content of carbon and certain metals make it possible to consider fine-grained sediments of the rift-induced troughs of the Chukchi Sea as a possible analogue of some types of ancient high-carbon sediments attributable to black-shale sediments [Astakhov et al., 2013].

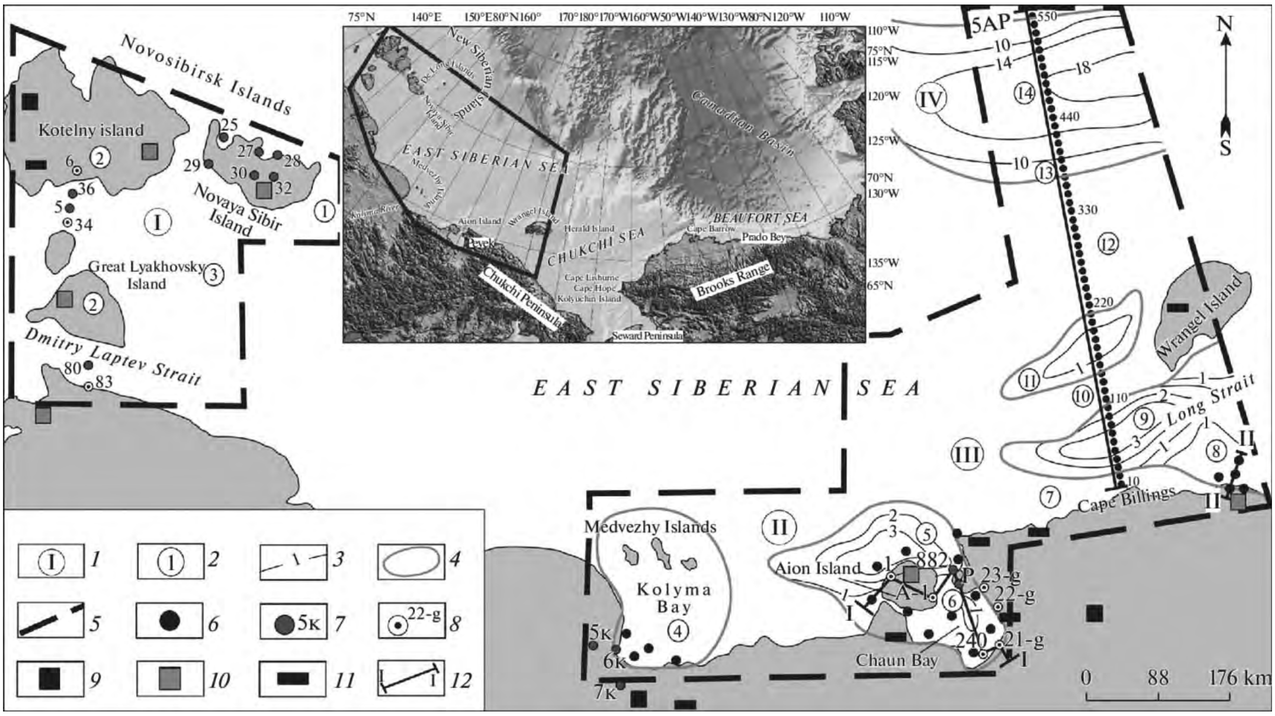

Fig. 3. Location scheme of gas geochemical studies in the East Siberian Sea. 1 – sedimentary basin: I – Novosibirsk, II – Aion, III – South Chukotka, IV – North Chukotka; 2 – geostructural elements:
(1) Novosibirsk Trough, (2) Kotelny-Svyatoi Nos Rise, (3) Blagoveshchensk structural terrace, (4) Medvezhinsk Rise, (5) Aion Lowland, (6) Chaun Lowland, (7) Kuul Anticlinorium, (8) Valkarai Lowland, (9) Long Lowland, (10) Wrangel Arch, (11) Wrangel Graben, (12) North Shelagi Rise, (13) Kolyuchin Graben-Rift, (14) North Chukotka Lowland; 3 – structural contour lines at sedimentary cover bottom, km; 4 – geostructure boundaries; 5 – boundaries of gas geochemical study areas; 6 – gas geochemical stations (2008, RV Akademik MA Lavrentyev); 7 – well and its number; 8 – gas-emitting well and its number. Coal fields and appearances: 9 – hard coal; 10 – brown coal; 11 – bitumen manifestations; 12 – gas geochemical section. [Gresov et al., 2017]

7. Traps and pathways of methane on its way from the sediment to the atmosphere

If methane is generated in abundance, then its buoyancy force drives it towards the seafloor, through the water column into the atmosphere.

Sufficient amounts of methane from preformed gas beneath or within permafrost, gas hydrates, and gas fields can cause oversaturation and, thus, gas emission via bubbles from sediments (ebullition) [e.g. McGinnis et al., 2006].

In general, Frederick and Buffett's modeling suggested that SGD may play a large role in submarine permafrost evolution and gas hydrate stability [Frederick, Buffett, 2015].

Most of the observed BSR occur close to the SII GHSZ indicating significant thermogenic gas input into the hydrate stability zone throughout the SW Barents Sea. The distribution of BSR is controlled primarily by fluid flow focusing features, such as gas chimneys and faults [Rekant et al., 2015].

Methane can be transferred to the atmosphere either directly via bursting gas bubbles or indirectly via wind speed dependent dissolved gas transfer. The former was predicted to be only important in seas with water depth <100 m while the latter was observed to be limited due to ocean stratification.

Vigorous bubbling events (1.5 to 5.7 bubbles per second) were observed at some sites of the ESAS [Shakhova et al., 2013]. Vigorous bubbling events as well as seepages of thermogenic CH_4 [Cramer, Franke, 2005] indicating that part of the water column supersaturation of the ESAS result from a seabed source [Sapart et al., 2017].

Geissler et al. found evidence of wide-spread methane venting also at the Northern Svalbard shelf in close vicinity to the Hinlopen/Yermak Megaslide slide scar [Geissler et al., 2016]. In the SW Barents Sea, gas flares were mapped along a segment of the Ringvassøy Loppa Fault Complex near the Snøhvit and Albatross gas field [Chand et al., 2014]. Bubble release was also documented at the Haakon Mosby Mud Volcano, located in 1270 m water depth at the center of the Bjørnøya slide scar on the SW Barents Sea slope [Sauter et al., 2006]. Thousands of gas emission sites were discovered along the western margin of Svalbard generating a hundreds of km-long methane plume [Mau et al., 2017], and generates a dissolved methane plume that is hundreds of kilometer in length.

Ice related processes have contributed to the widespread development of indurated (low-permeability) sediments that could be particularly effective at trapping CH_4 beneath some Arctic Ocean shelves [Ruppel, Kessler, 2017]. For example, high-amplitude up-dip truncations suggest impermeability (cap) at the BGHSZ in the Canadian Beaufort Sea, preventing further migration of any free gas.

Structural geology controls numerous methane related processes: BSR distribution, groundwater discharges from thawing permafrost, and gas emissions into the water column. For example, primarily focused fluid flow features, such as gas chimneys and faults, control the distribution of BSR in the SW Barents Sea [Vadakkepuliyambatta et al., 2017].

Along the western Svalbard margin from Bjørnøya to Kongsfjorden most gas emissions detected, as acoustic flares were found in the vicinity of the Hornsund Fracture Zone [Mau et al., 2017]. The methane discharges on bathymetric highs are characterized by sonic hard grounds, whereas glaciomarine and Holocene sediments in the troughs apparently limit seepage.

Offshore northern Svalbard, an amphitheatre-shaped slide scar area with head and side-walls up to 1600 m high, indicates that at least 1250 km^3 of shelf sediments were excavated, and up to 2400 km^3 of sediment were finally involved in the slide. Large blocks with lateral dimensions of up to 4 km and taller than 300 m can be observed in the depositional area. The failure event was dated to 30 cal kyr B.P. The Hinlopen shelf failed coincidently with rapidly falling sea-level during the last glaciation [Geissler et al., 2016].

To date, there has been no clear evidence for the presence of gas hydrates, free gas or degassing features, which led Winkelmann and Stein [Winkelmann, Stein, 2007] to argue that hydrate dissociation and gas overpressure are not among the main preconditions for slope failure initiation. Instead, they favoured tectonic control, related to the development of a forebulge as the glaciation intensified.

Gas hydrate, a frozen, naturally-occurring, and highly-concentrated form of methane, sequesters significant carbon in the global system and is stable only over a range of low-temperature and moderate-pressure conditions. The gas discharge occurs in water depths at and shallower than the upper edge of the gas hydrate stability zone throughout.

A link of seismotectonics and methane seepage was also revealed to account for variable methane emissions offshore Costa Rica; apparently an earthquake in 2002 sealed pathways of methane and lowered methane discharge at seep sites situated 300 km apart [Mau et al., 2007b].

Methane rich fluids feeding the Svalbard plume appear to migrate either along faults, along stratigraphic boundaries or through a combination of these two structures. Most of the flares mapped in this study are located in the vicinity of the Hornsund Fracture Zone (HFZ); a few single flares were found in Kongsfjordrenna, near the Knølegga Fault Zone, and along the northern edge of the Kveithola Trough. Knies and Damm et al. had previously noted a relationship between high methane concentrations and the HFZ at the western Spitsbergen shelf [Knies et al., 2004; Damm et al., 2005].

In contrast, Rajan et al. suggested that, since the fluids expelled at ~250 m water depth offshore Prins Karls Forland (PKF) align with the outcrop of a glacigenic sequence, fluid migration is likely occurring along dipping strata in the prograding sequence. Since the Barents Sea Ice Sheet extended to the slope edge from northern Norway to northern Svalbard, glacigenic stratigraphy could provide a pathway for ascending fluids not only at the PKF, but also farther south at Hornsundbanken, Sørkappbanken, and Spitsbergenbanken. Our surveys could not identify fluid migration along stratigraphic boundaries; but our data indicate that the majority of the gas emissions follow the HFZ [Rajan et al., 2012; Ingólfsson, Landvik, 2013].

8. Methane transfer to the atmosphere

Although methane seepage has been discovered at numerous seep sites around the world, quantitative estimates of the fate of methane in the water column remain rare. For example, large discrepancies exist in the data of methane fluxes from the sea to the atmosphere. The estimates in vertical sections: Lena River Delta – continental slope (the Laptev Sea), Taimyr Peninsula – Voronin Trough, and along the Novaya Zemlya Archipelago fluxes of methane in 2015 show a small contribution (1–400 mol km^{-2} day^{-1} (52 mol km^{-2} day^{-1} in average)) [Vetrov et al., 2018].

Methane transfer to the atmosphere is limited in the ocean water column by stratification of water masses. Offshore Svalbard, density stratification resulting from salinity changes limits vertical eddy diffusion of methane to the surface mixed layer. The methane released from gas emission sites in the lower water column remains in the ocean where most is microbially oxidized while being transported [Gentz et al., 2014].

The Arctic Ocean is markedly stratified between 0 m and 150 m. For the winter period, dissolved CH_4 concentrations beneath the sea ice were 5 to 10 times higher than in the summer at the ESAS [Shakhova et al., 2010]. Stratification results in low diffusion and heat transfer rates between water masses and is a fundamental reason why the Arctic Ocean is consistently ice covered.

Typically, a large fraction of the gas dissolves during the bubbles' transit through the water column. Its quantity depends on release depth, volume of the bubbles, and the buoyancy force of the plume [Greinert, McGinnis, 2009].

[Fenwick et al., 2017] noted that the freshwater layer at the surface often acted to dilute the concentrations of the gases within the mixed layer and limiting the associated sea-air fluxes.

However, it has been shown that, when the water depth is <100 m, a significant fraction of the methane in the bubble might be directly transported to the atmosphere [e.g. Römer et al., 2017]. Because the ESAS average depth is only 45 m, the water column provides a short conduit for bottom-released CH_4 to be vented to the overlying atmosphere [Shakhova et al., 2010].

Zhang et al. [Zhang et al., 2017] observed clearly discernible peaks of atmospheric CH_4 near the surface of ocean that coincided with the location of the areas of confluence of the warm saline and cold less saline waters in the Arctic.

Therefore, methane transfer appears to be confined to a few areas where oceanographic settings do not hamper methane transfer to the atmosphere.

9. Sea-air flux of dissolved methane

However, while indurated sediments, stratification in the water column, ice cover and microbial methane oxidation in the sediment and water column limit the quantity of methane reaching the atmosphere, migration paths along faults and ebullition bypasses these obstacles. While stratification and ice cover hinder methane transfer to the atmosphere, increasing wind speed amplifies the gas transfer velocity [Mau et al., 2017].

In the works on a large amount of data and in a large area of the northwestern Pacific Ocean, the effect of pulsating emissions of methane is observed: if wind speed is low and there is a methane flux from the underlying deep waters, an increase takes place in the concentration of methane in the surface water due to its transport (fig. 4). Methane concentration dramatically drops to the equilibrium values as wind speed increases, because the methane flux from deep horizons does not have time to compensate the methane outflow from the surface [Obzhirov et al., 2016].

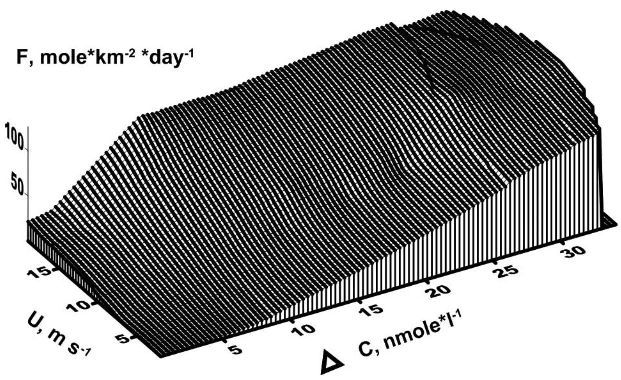

Fig. 4. Variations of the maximum methane flux F on the water–air interface depending on wind speed U and difference ΔC between the measured and equilibrium concentration of methane in the sea water [Obzhirov et al., 2016].

Conclusion

Although release of CH_4 to the ocean and atmosphere has become a topic of discussion, the subject remains sparingly explored.

The rate of methane release, additional to existing atmospheric methane burden, remains difficult to predict. Seismic reports suggest a widespread gas hydrate occurrence, however, gas hydrates, which were also suggested to occur along the western Svalbard margin and at the East Siberian Sea slope, have not been found. In contrast, gas hydrate was found in the upper part of the western slope of the Chukchi Plateau within the deep-water eastern margin of the East Siberian Sea. Potential oil-gas occurrences are based on geophysical data, but have not been approved by geochemical and other direct methods. According to few results, rivers transport organic matter as a prerequisite for methane production, and creeks from permafrost directly adds methane to the atmosphere and to the river system. How large either source is, is currently unknown.

Because the methane stable isotopic signature cannot be easily distinguished from Arctic wetland emissions, hypothesis, if gas hydrates or wetland methane accelerated climate warming in the past of the Earth, remains debatable.

Although microbial methane oxidation is a comparably slow process in contrast to physical movements within the water (i.e., eddy diffusion and advection), the sink might be strong enough to hamper methane sea–air flux away from bubble emission sites. How these methane sinks (anaerobic and aerobic methane oxidation) change due to global warming is uncertain. And if the bacteria are psychrophils and thus capable of growth and reproduction in cold temperatures or not and how long the lag time, i.e. the time needed to adapt to the changes, will be uncertain.

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