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Methane flux and gas hydrate accumulations in the Sea of Okhotsk and their ecological aspects

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ABSTRACT

Methane is one of the important representatives of the organic substances in the atmosphere (for example, an increase of methane content in the atmosphere can affect enhancing the greenhouse effect). Gas hydrates are an essential part of links in the methane cycle and the accompanying fluxes of other gases. The research object in this paper is the Sea of Okhotsk, where gas hydrate fields and active submarine gas discharge areas were found. The study of methane fluxes is relevant both for the Sea of Okhotsk and the World Ocean. From 1984 to 2015, the background and anomalous methane fields were explored in the Sea of Okhotsk as a result of gas geochemical research carried out by scientists of the Gasgeochemistry Laboratory (POI FEB RAS). The flux of natural gas became stronger from the sources to the seafloor, from the seafloor to the water, and, finally, to the atmosphere due to renew of fault zones. Moreover, the amount of methane vents on the Sakhalin slope has increased from 2-3 vents to more than 400. The most representative hydroacoustic anomalies "flares" mapped direct methods study methane hydrates. The received outcomes indicate the urgency of the study of methane fluxes and the formation-dissociation mechanism of gas hydrates, and the influence of natural gas from hydrocarbon sources on the environment.

Keywords: Gas hydrates, methane, Sea of Okhotsk, gas discharge, greenhouse gas.

1. Introduction

It is presented the explore results of methane background and anomalous fields, methane sources (gas hydrates, oil and gas deposits, etc.) in the Okhotomarine Region, and their influence on the environment received by the Gasgeochemistry Laboratory during 1984-2015. For the period, many gas geochemical fields and methane anomalies have been revealed in the Sea of Okhotsk. They are referred to as the zones of submarine gas discharge. From 1998 to 2004, the complex geological, geochemical and geophysical research of the Okhotsk Sea Area as a common natural system was carried out in collaboration with Marine Center GEOMAR (Germany) within the framework of Russian-German program "KOMEX." The basic direction of research was to study gas hydrates, methane vents, their interaction with sedimentary complexes, geological structures,

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and tectonics of the region. In 2003, 2005, 2006, the more detailed studies of gas hydrate distribution on the north-eastern Sakhalin slope in the Sea of Okhotsk were carried out within the frameworks of Russian - Japanese - Korean projects “CHAOS-1”, “CHAOS-2” and “CHAOS-3”, accordingly. From 2007 to 2015, the investigations for the duration, the complex geophysical, gas geochemical, and geological research were performed within the frameworks of the Russian-Japanese-Korean “Sakhalin Slope Gas Hydrate project”.

The basic purpose of the research was the study of methane fluxes and methane concentrations in the water and in the bottom sediments and gas hydrates in the Sea of Okhotsk, the elucidation of the annual variability of methane submarine discharge, and the definition of methane sources and their roles during the change of the state of surrounding natural environment also. Special attention has been given to the all-around exploration of the mechanism of gas hydrate formation and decomposition.

In the Sea of Okhotsk, there are known 3 areas of gas hydrate occurring (Fig. 1) in the bottom sediments (Zonenshain et al., 1987; Ginsburg et al., 1993; Ginsburg, Soloviev, 1994; Biebow, Hutten, 1999; Biebow et al., 2002; Obzhirov et al., 2003; Matveeva, Soloviev, 2003; Matveeva et al., 2005; Dullo et al., 2004; Mazurenko et al., 2006; Jin et al., 2007; Jin et al., 2008; Shoji et al., 2010; Jin et al., 2011; Jin et al., 2013; Shoji et al., 2014; Jin et al., 2015): the eastern Sakhalin slope (the north-western part of Derugin Basin; for the first time they were discovered in 1991, hypothesized in 1988 as a result of the reveal of submarine gas fluxes) and the north-western slope near Paramushir Island (1986, the south-eastern part of Golyginskii flexure) and the Kuril Basin (2012, they are revealed in the bottom sediments on the western slope of the Kuril Basin on the Terpeniye Gulf side). These geological structures are characterized by the sufficiently high hydrocarbon generation potential (Veselov et al., 2004). Gas hydrate accumulations are part of the Okhotsk sedimentary basin containing thick Mesozoic and Cenozoic sediments disrupted by numerous faults (Ginsburg et al., 1993); some of them may be conduits for migrating gas. Gas hydrates are associated with gas vents located near fluid conductors like active deep faults, diapers, and, possibly, mud volcanoes. Gas hydrates are revealed near seafloor layers of sediments in various forms like lenses, layers, interlayers, and fragments.

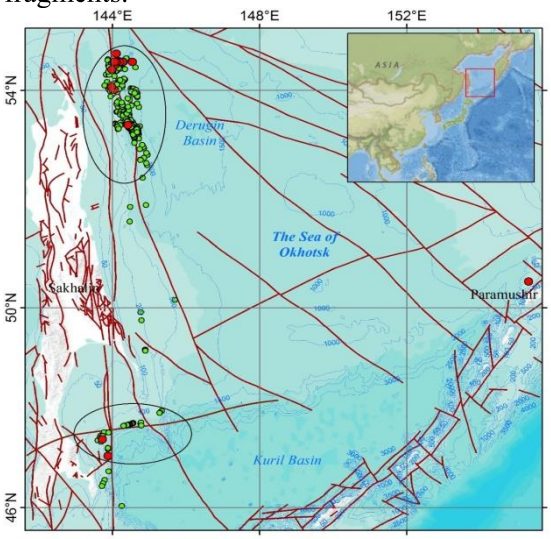

Figure 1. Map showing the location of revealed gas hydrate accumulations (red dots) and some gas vents (green dots) in the north-eastern Sakhalin slope, in the western slope of the Kuril Basin, near Paramushir Island, and there refers to the fault zones (red line) in the Sea of Okhotsk

It is important to note that in one of the structures (KOPRI (Fig. 2)), the thickest gas hydrate layer (up to 34 cm) was revealed at a depth of 48-82 cm below seafloor (Mazurenko et al., 2006). Such thick gas hydrate layer (Fig. 3) in this part of the World Ocean was observed for the first time. Moreover, earlier gas hydrates were sampled at a water depth of 480 m and 530 m in the Caspian Sea and the

Gulf of Mexico. In 2005 the most shallow gas hydrate accumulations were revealed in the Sea of Okhotsk -390 and 385 m (Mazurenko et al., 2006).

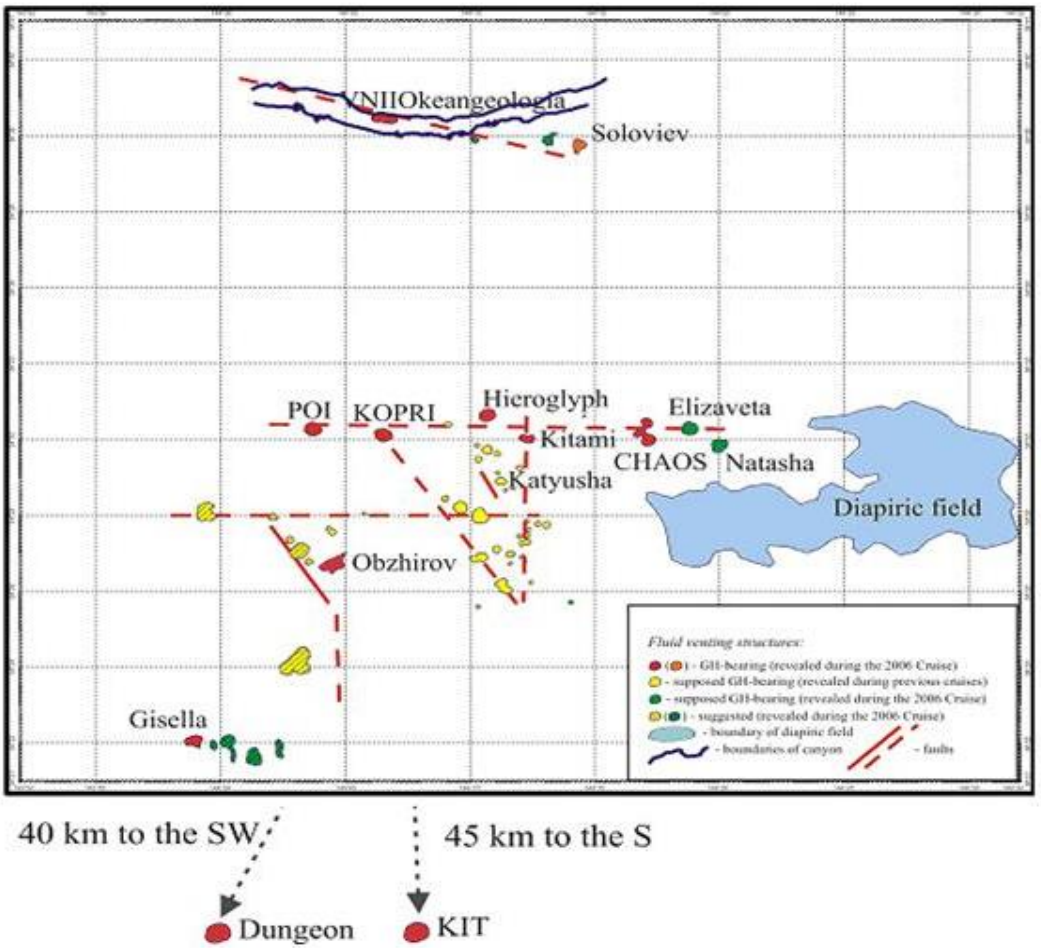

Figure 2. Map showing the location of revealed and suggested gas hydrate accumulations in the north-eastern Sakhalin slope, and they are referred to as the fault zone in the Sea of Okhotsk (Jin et al., 2007)

←*Figure 3*. Gas hydrate layer - 34 centimeter. The sample was taken from structure "KOPRI" (north-eastern Sakhalin slope) during cruise 36 (RV Akademik M.A. Lavrentyev) (Mazurenko et al., 2006)

2. Methods

Water samples to measure methane concentration were taken from Niskin bottles of the Rosett without contact with air. Gas is extracted from the water by a vacuum line,

and then it is analyzed by a gas chromatography system aboard the ship. Gas from sediment samples is extracted by the HeadSpace method (Obzhirov et al., 1999; Obzhirov, 1993). The sensitivity of the hydrocarbon analyses is 0,000001%. To check the correction of analyses, we used the gas standard with concentrations of methane 10, 100, 1000 ppm, and 1% CO_2, ethane ("Altech", USA). The measuring error was less than 5%.

3. Results and discussions

3.1. Background and anomalous methane fields in the Sea of Okhotsk

Research of the gas parts of sediments in the Sea of Okhotsk shows: in the cores where gas hydrates are not found, as a rule, methane concentrations are low in the surface layer (0-200 cm) and increase from a depth of 200 cm (0,1-1 ml/l and more). Probably, it is explained by the presence of the sulfate-reduction zone over the depth. In the sediments containing gas hydrate, this regularity is broken because gas hydrate intervals can sufficiently contain a huge volume of methane and locate at different subbottom depths. In the cores where gas hydrates are found, methane concentrations increase up to 500 ml/l and more independently of depth, sometimes reaching 3000 ml/l (Obzhirov et al., 2007).

Regarding the methane concentration in the water of the north-western part of the Sea of Okhotsk, it sharply increases if gas hydrates are in the sediments. Gas bubbles direct to the sea surface and create anomalies with maximum values 20 000-200 000 nl/l near the seafloor and 500-1000 nl/l near the sea surface. Whereas in the areas of oil-and-gas structures, as a rule, methane concentrations in the bottom water are about 2500 nl/l when background values in the The region is surface seawater - 70-90 nl/l, subsurface seawater (75-100 m) - 100-120 nl/l, and water nearby seafloor (1000 m and deeper) - about 15-20 nl/l (Obzhirov et al., 2007).

In the course of research (2012), according to the announced theme, the following results were received (Cruise 59, R/V "Akademik M.A. Lavrentyev" (Jin et al., 2013)):

For the first time, gas hydrates are discovered in the Kuril Basin (the southern part of the Sea of Okhotsk).

In the gas discharge areas:

The anomaly methane concentrations in the bottom sediments are fixed (up to 100 ml/l), which 1000 and more times exceeds the background. Besides methane, there are ethane, propane, and carbon dioxide in small amounts in the gas composition.

The methane concentrations in the bottom layer of seawater are 1000-2000 nl/l. Toward up methane concentrations reduce to the background values (30-40 nl/l); in the sea surface water, methane concentrations increase to 120-150 nl/l. In the areas without submarine gas discharge, methane concentrations in the seawater correspond to the background values.

In 2013 (Cruise 62, R/V "Akademik M.A. Lavrentyev" (Shoji et al., 2014)), gas hydrates are also revealed in the bottom sediments on 2 stations in the Terpeniye Gulf in the Sea of Okhotsk. As previous expedition research explores the Sea in Okhotsk, the numerous gas vents (mainly methane composition) are exposed from the seafloor to the water.

The main purpose of the expedition 2014 (Cruise 67, R/V "Akademik M.A. Lavrentyev" (Jin et al., 2015)) was a detailed study of gas hydrate areas found in the two previous expeditions of 2012-2013 and a search for new sources of methane.

Two sediment cores were taken in the area of "Kurile flare". A background level of methane content was observed in these sediment cores. Ethane was the most representative of methane homologs. He was not seen at the surface horizons with low

methane content. With the increase of the depth below the seafloor, ethane and methane concentration grew. The distribution curves of ethane and methane in the sediment column were nearly identical.

The sampling depth of seawater was about 2500 m in the Kurile Basin. The methane distribution in the seawater was uneven (as in previous years). Virtually all CTD-stations was observed a maximum of methane concentrations near the seafloor. However, high concentrations were recorded at various depths.

The presence of anomalous methane concentrations in the sea surface water makes a real methane emission from the sea surface to the atmosphere. As methane is a greenhouse gas, its increase in the atmosphere inevitably can be affected by the intensification of the greenhouse effect. In the region's ecological significance of gas, hydrates are related to the display of landslide processes and the impact of gas vents on biota in gas hydrate structures.

3.2. Methane flux in the Okhotsk Sea Area as result of seismotectonic activation

The north-western part of Derugin Basin (the north-eastern Sakhalin slope), where gas hydrate accumulations are revealed directly, is the most noticeable and investigated now. Many researchers recognize the existence of a strong seismo-active submedial fault zone here (Ginsburg, Soloviev, 1994; Kharakhinov. 1998; Bessonova et al., 2001; Bessonova, 2003). Probably, due to the seismotectonic activation of the north-western part of the Okhotsk Sea Area, there are discovered about 400 natural gas vents (mainly, methane composition) from bottom sediments to the seawater and 11 structures (Fig. 4) where gas hydrates were sampled (Jin et al., 2007).

Data analysis (1998-2006) shows that any regularity of the quantitative changes of gas hydrates at a subbottom depth is not traced. However, the marked approach of sediment levels containing gas hydrates to the water-sediment interface when the approach to the central part of gas vents is, and, vice versa, their deepening when gas fluxes are far. We suppose that it is related to secondary formation in the crippling zones of initial gas hydrates located near active deep faults, conductors, and methane flux generators. At the time of seismotectonic activation, the faults open, heat flow intensifies, and as a result, the disturbance of the thermobaric conditions of gas hydrate stability occurs (Obzhirov et al., 2007).

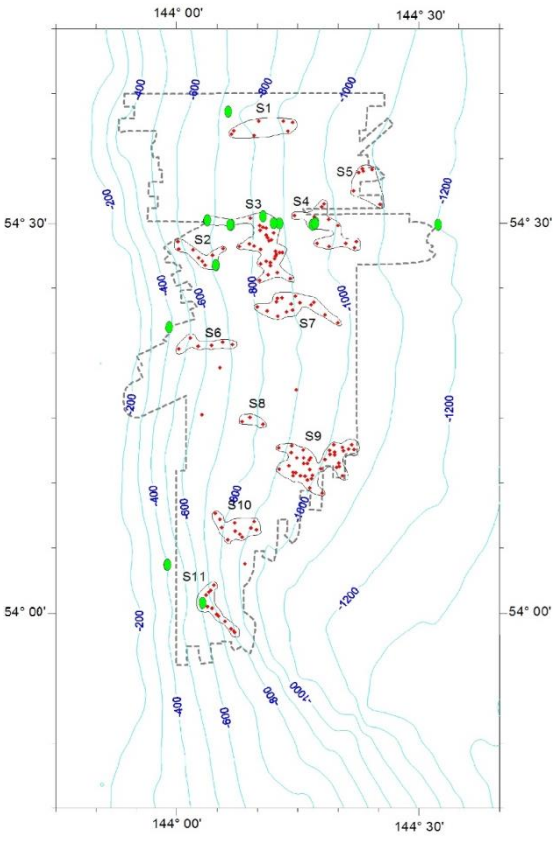

Figure 4. Distribution of seepage structures (red diamonds). Green circles - gas-hydrate accumulations. Close numbered lines mark seep fields. The closed dashed line indicates SSS (Side Scan Sonar) mapping area. The Contour interval is 50 meters (Jin et al., 2008)

Gas hydrates and related methane fluxes are controlled by the Eastern-Sakhalin fault system and the Hokkaido-Sakhalin fault system, which are pathways for deep-buried gas migration. Although the carbon isotopic composition of methane and ethane from gas hydrates is a discussion point, C1 isotopic compositions ranging from -73‰ to -63‰ indicate a mixture of thermogenic and microbial methane. C2 isotopic compositions, almost in every case, indicate a thermogenic source of ethane.

The most intensive methane anomaly fields in the water of the Sea of Okhotsk, exceeding a background in 1000 and more times (Fig. 5), are formed on the north-eastern shelf and slope of Sakhalin. Gas bubbles create anomalies with maximum values over 20 000 nl/l near the seafloor and 500-1000 nl/l near the sea surface.

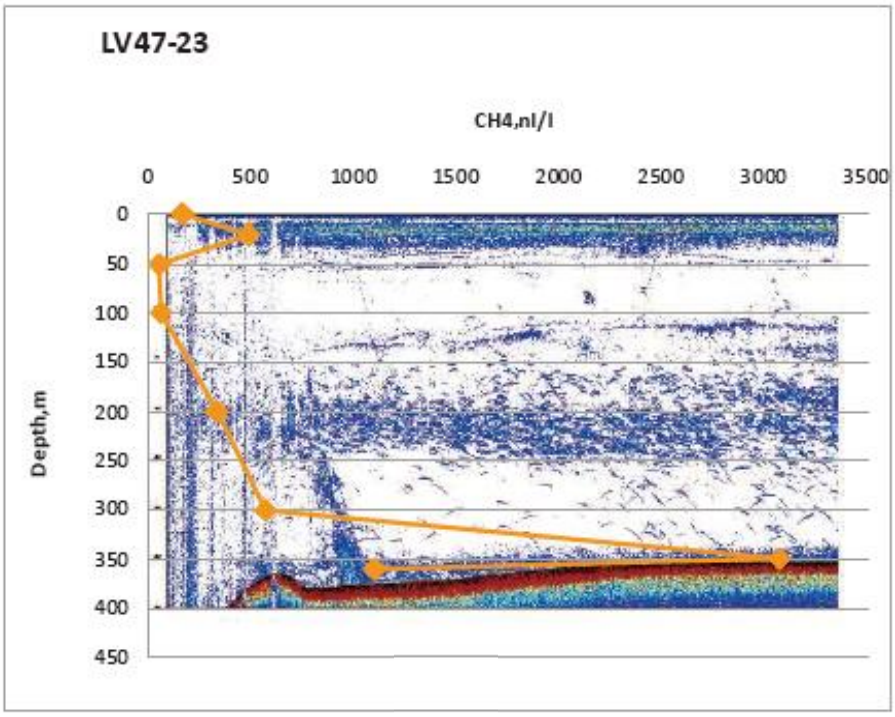

Figure 5. Gas distribution in the seawater on Station LV47-23 (Shoji et al., 2010). Yellow points with the line - methane concentration (nl/l). The base of the picture is a hydroacoustic image. Redline is a surface sea bottom

Expedition research from 1998 to 2002 points to that all these natural gas discharges are the intersection zones of the faults of the north-western and north-eastern direction controllable by the East Sakhalin fault zone, influencing the seismo-tectonic activity of the north-eastern shelf and slope of the Sea of Okhotsk.

Mainly, the methane discharge in the northwestern part in the Sea of Okhotsk is the result of the decomposition of gas hydrates (and free gas under the gas hydrate layer), located nearby active deep faults (for instance, the vents of natural gas "Chaos," Fig. 6, Fig. 1-2). The process of methane discharge resulting from gas hydrate decomposition for the first time was found in 1986 on the north-western slope of Paramushir Island (Obzhirov et al., 1999). Here at a depth of about 800 m, the gas bubbles (main methane) went from

seafloor sediments to the water column and created a sound-scattering flare (acoustic anomaly) in an echogram, which height was 200-300 m. As a result of the research conducted by the Institutes of Volcanology and Oceanology (Zonenshain et al., 1987), gas hydrates were found in the methane vent area seafloor sediments.

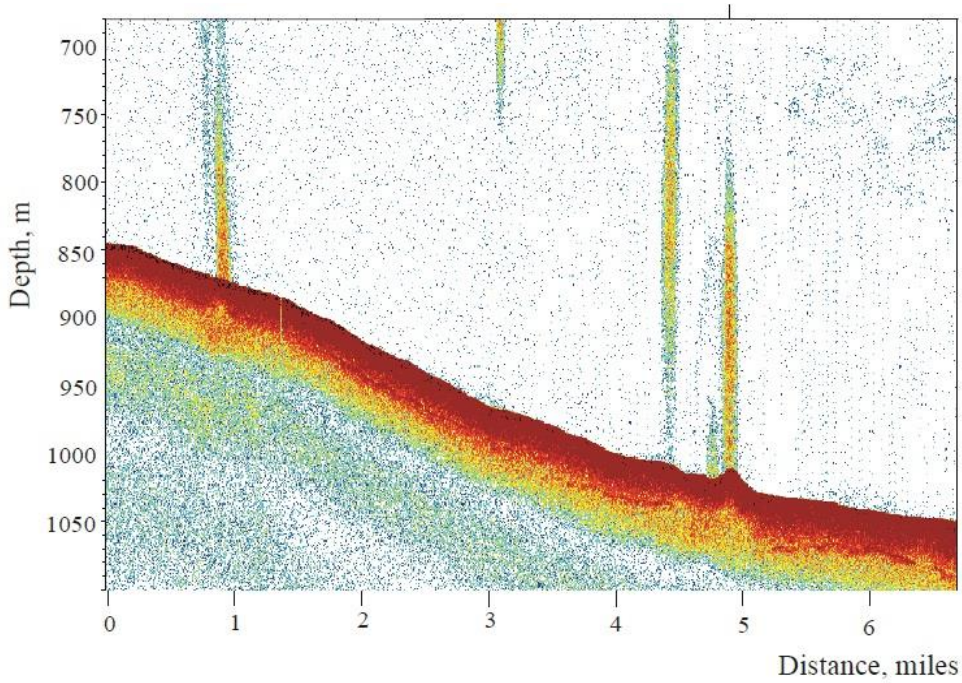

Figure 6. Hydroacoustic anomalies. Author A.S. Solomatin (Matveeva et al., 2005)

The study of gas geochemical fields in the Sea of Okhotsk has shown that on the boundary of 1988-1989, there was a sudden increase of methane concentration in bottom seawater (Obzhirov et al., 1999). The sharp increase of methane concentration in the water of the Sea of Okhotsk, probably, is connected with seismotectonic activation in the Okhotsk Sea area. As a result, the thermal flow has amplified through fault zones, the sediment layer, containing gas hydrates and blocking free gases, has broken. Natural gas has begun to act to the seafloor surface both from under gas hydrate covers and during gas hydrate decomposition with the escape of methane bubbles from seafloor sediment to the water and in someplace from the sea surface to the atmosphere. Methane fluxes from gas hydrate decomposition created sound-scattering flares (the acoustic anomalies of various forms) in an echogram. Methane migrated via fault zones to the water column like seep without acoustic anomaly from oil-gas bearing layers. Originally 2 hydroacoustic anomalies describing methane fluxes have been discovered. By 1995 their amount has increased (about 30). The same year there was an earthquake in the northeast Sakhalin, in the Neftegorsk area. By 2002 the amount of methane vents in the area of the Sea of Okhotsk has exceeded 100. As research shows, all these natural gas vents are referred to as the intersection zones of the faults of the north-western and north-eastern direction controllable by the East Sakhalin fault zone, influencing the seismotectonic activity of the north-eastern shelf and slope of the Sea of Okhotsk (Obzhirov et al., 2004).

Investigations point to the continuation of the activation of seismotectonic processes in the Okhotsk Sea region in 2002 and, consequently, the increase of the area of gas hydrate decomposition and the amounts of methane vents from sediments to the water. Obtained in 2003 - 2005 data confirm the aforesaid and show the further intensification of the process of seismotectonic activation. The monitoring of methane content in the area of gas bubble vents has shown that methane concentration (May-June 2005) remained so high during research and the previous expedition (2003).

The research has been carried out in a gas hydrate field on the slope of Paramushir Island shows that in the bottom seawater in the place of methane vents, since 1985, methane concentration grew up constantly from 120 nl/l in 1985 to 500 nl/l in October 1994. The measurements of gas contents after the Kunashir earthquake show the methane concentration increase by 2 times and more. The earthquake epicenter was situated at a depth of 300 km near Paramushir Island. Still, due to the sharp increase of methane concentration in the water column in the gas vent area, it is possible to assume the seismotectonic activation of the region of the Kuril Islands arch in October 1994 (Obzhirov et al., 2003).

3.3. Ecological aspects of gas hydrate accumulations in the Sea of Okhotsk

Since the time it has been established that gas hydrates are a component of the continental slopes within various parts of the World Ocean, scientists show interest in environmental risk and climate change. As gas hydrates can contain a huge volume of methane (one volumetric unit of hydrate contains gas which occupies about 160-170 volumetric units under normal conditions), it is clear that sediments containing gas hydrates play a vital part in the sedimentary material mobility of continental slopes, sea biota communities, and also the global processes of climate change.

The amount of methane preserved in the gas hydrate accumulations is estimated approximately 8×10^8 m^3/km^2 for the northwestern part of the Sea of Okhotsk and 2×10^{12} m^3 for the entire Sea of Okhotsk (Matveeva, Soloviev, 2003). T. Ludmann, H.K. Wong (2003) denote still more amount: 15×10^{13} m^3 for the entire Sea of Okhotsk. Considering the relation of methane fluxes with the seismotectonic processes in the Okhotsk Sea area (Obzhirov et al., 2003), it is possible to point out the main ecological consequences connected with submarine methane discharge caused by gas hydrate decomposition in the region.

By special research in the Sea of Okhotsk with use of submersible inhabited craft in the area nearby Paramushir Island (Ginsburg, Soloviev, 1994) and Ocean Floor Observation Systems (Biebow, Hutten, 1999; Biebow et al., 2002; Dullo et al., 2004) in the northeastern Sakhalin slope, the seafloor surface was studied in detail. Received data show that methane fluxes impact the ecosystem structure. The rapid development of bacterial mates and sharply expressed prevalence of some benthos (mollusks, crustaceans) have been fixed compared to neighbor territories where seawater has background methane concentrations.

As a result of data analysis, the following features have been allocated:

- Development of bacterial mates occurs in the central parts of gas discharge. Sometimes the fields with a white patch cover seafloor up to 10-15 meters in diameter.
- The abundance of alive Calyptogena mollusks and/or Conchocele mollusks is observed to approaching the central parts of gas vents.
- Sometimes, there are areas with sediments containing gas hydrates where the zones with the character development of the seafloor disturbance and the obvious absence

of gas entrance to the water are remarked. In these territories, the shell remains and fragments are also fixed. It is zones of the last localizations of gas vents and shell finds-bioindicators of the gas activity.

As a result of the seafloor research, it was discovered that there is a bottom deformation with the formation of hills and holes which have a diameter of more than 10 meters and a height of more than 5 meters when gas hydrates destruct. On the slopes, the disturbance and the moving of sediments down are observed (Fig. 7). It is the significant change of bottom morphology. Therefore, it is necessary to consider the opportunity of gas hydrate decomposition with the subsequent bottom destruction because the consequences of this event may be unpredictable. It is concerned, for instance, with engineering-technical works: the lining of oil pipelines, the construction of derricks, etc.

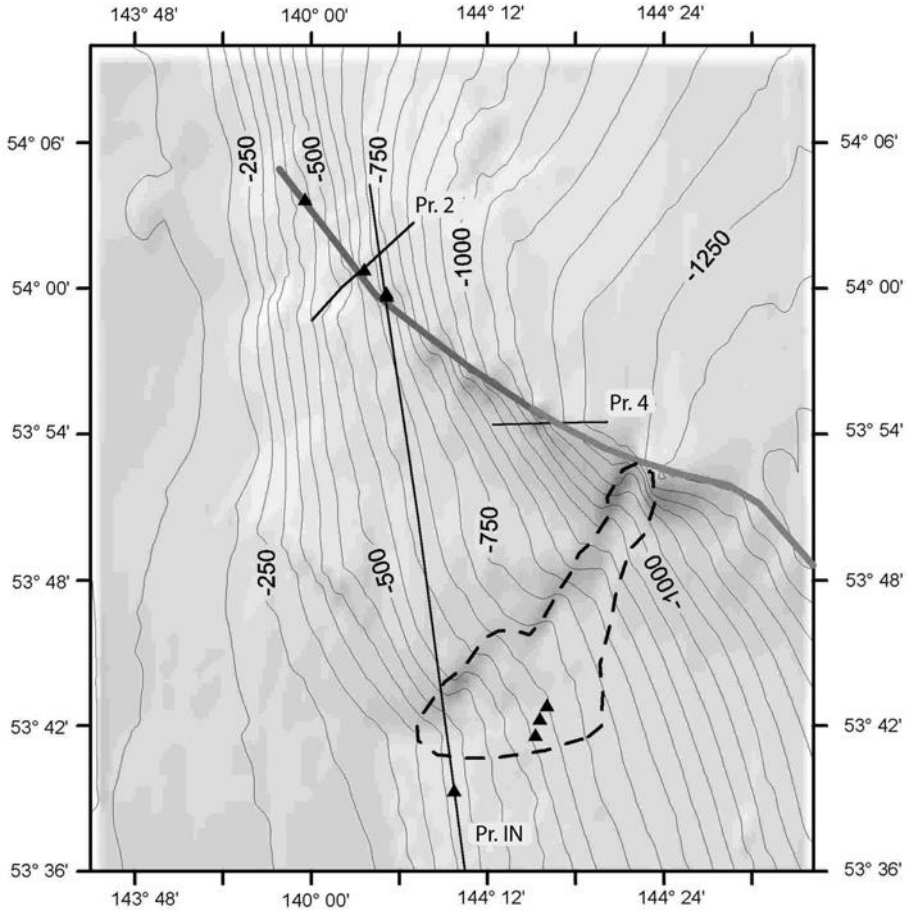

Figure 7. Bathymetric map of the "RV Lavrentiev Fault" area. Solid bold gray line shows the steps in the bottom relief topography. Dashed line bounded south-eastern end of a possible slide block. Triangles are hydroacoustic anomalies. Thin lines represent the position of the profiles. Isobaths are spaced at 25 m; Pr. - profile. Author B.V. Baranov (Matveeva et al., 2005)

As regards the aspect of climate change: a huge amount of methane is released from the seafloor to the water and then from the sea surface to the atmosphere (methane is a strong "greenhouse gas") when gas hydrates decompose. And vice versa, when gas hydrate

accumulations are stable (seismotectonic activation is reduced), they can delay methane entrance to the environment.

Thus, at present, it is essential to understand and determine the role of gas hydrate accumulations in the global and regional nature processes.

4. Conclusions

There are definite geostructural and geodynamic conditions in which gas hydrate accumulations are found in the Okhotsk Sea Area: liquid and gas hydrocarbons, thick sedimentary strata, the presence of breaking disruptions, seismic activity of the respective territories, etc. They determine the occurrence of submarine gas discharge and anomalous methane fields in the Sea, which can influence the environment.

The most powerful gas vents in the Sea of Okhotsk are referred to as the area with gas hydrate accumulations and fault zones. Methane escapes via the fault zone from the oil-gas deposits in sediment layers and sediments containing gas hydrates. Methane bubbles come to the water column and create an acoustic anomaly.

In the Sea of Okhotsk, a significant amount of anomalous methane fields is revealed. The most intensive methane anomaly fields in the water of the Sea of Okhotsk, exceeding a background in 1000 and more times, are fixed on the north-eastern shelf and slope of Sakhalin. Probably, it is connected with the increase of seismotectonic activation in the area.

As a result of the research of the relation of methane fluxes with the seismotectonic processes in the Okhotsk Sea Area, the periods of methane concentration growth accompany the earthquake events in this Region. The gas geochemical data (methane anomalies) can be used to forecast natural catastrophes (earthquakes, landslides, etc.).

The isotopic composition of gas hydrates and carbonate formations of the upper intervals of bottom sediments and their confinement to active fault zones allow us to consider gas hydrates in the Sea of Okhotsk as mixed formations (thermogenic gas in combination with microbial) in the formation of which the role of methane from oil and gas and coal-bearing sediments of the region is not excluded.

Acknowledgments

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