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*Luchsheva L., Kononov Yu., Obzhirov A.*

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## НАУКИ О ЗЕМЛЕ

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### STUDIES OF MERCURY AND METHANE IN THE SHELF WATERS OF NORTHEASTERN SAKHALIN DURING THE PERIOD OF SEISM TECTONIC ACTIVATION

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#### ABSTRACT

In the period 1998-2000 observations were made of changes in the levels of methane and mercury in the water column of the oil and gas bearing northeastern Sakhalin shelf. A comparative analysis showed significant differences in the levels of methane and mercury concentrations in different parts of the shelf: in the shallow zone of the shelf, at the edge of the shelf, and in the upper part of the continental slope. The maximum concentrations of methane in water (more than 6000 n/l) were recorded in the area of discharge of a cold methane source (gas seep) above the gas hydrate deposit on the shelf edge.

The maximum concentrations of the dissolved form of mercury (up to 960 ng/l) were observed in the shallow part of the shelf and above methane seeps. The maximum concentrations of the suspended form of mercury (up to 247 ng/l) were found within the frontal zone of the East Sakhalin Current. It has been established that the revealed differences in the distribution of methane and mercury concentrations are due to the destruction of gas hydrate deposits during periods of seism tectonic activation of oil and gas structures. Stretching of these structures contributes to intense emanations of endogenous highly toxic dissolved mercury into shelf waters, leading to ecological disasters on the Sakhalin shelf. **Keywords:** monitoring of methane; methane seeps; gas hydrates; forms of mercury; seismicity; ecological disasters; northeastern shelf of Sakhalin.

#### INTRODUCTION

Interest in studying the distribution of mercury concentrations in the waters of the unique oil and gas bearing northeastern shelf of Sakhalin is due to its location in the zone of interaction between the Eurasian, Okhotsk and Pacific lithospheric plates. Therefore, Sakhalin is characterized by high seismicity, which is due to the subduction of the Pacific Plate under the continental lithosphere of Eurasia [1]. The modern subduction of the Pacific Plate is accompanied by earthquakes and intense volcanism, has a global character, and is the defining tectonic process on the eastern margin of Asia [2].

Earth degassing processes associated with tectonics, in particular, with deep faults in the earth's crust lead, as is known, to the formation of deposits of oil, combustible gases, mercury, and other minerals [3, 4]. Therefore, oil and gas fields confined to deep faults are characterized by an increased content of mercury due to its intensive modern degassing from the earth's interior. The primary source of mercury is assumed to be the substance of the Earth's mantle, during the degassing of which mercury, together with superheated water vapor, is transported to the upper part of the earth's crust [5, 6].

Migration of mercury from the bowels of the Earth is carried out together with "mantle" helium, hydrogen and other gases along the mantle fault zones. Therefore, the main mercury deposits are localized within the

planetary mercury belts, controlled by systems of deep tectonic faults in the earth's crust. The increased depth of faults contributes to active mercury emanations from the depths, especially during periods of seism tectonic activation [7]. The degassing processes of mercury coming from land faults have been studied for quite a long time, and the features of its migration in the hydrosphere and bottom sediments are practically not studied [8].

The shelf of northeastern Sakhalin is the only area in the Far East where commercial oil and gas production is carried out. All oil and gas fields on the Sakhalin shelf are connected to the western side of the deep-water Deryugin basin by a series of deep over thrusts that steeply go under the basin bed, where they cut through the rocks of the "basalt" layer and the upper mantle [9]. This zone is associated with crustal earthquakes and anomalously high heat fluxes [10]. During periods of compression, the Pacific oceanic plate goes under the continental plate of the Sea of Okhotsk and methane is released from the deposits of buried organic deposits. In the subduction zone, under the influence of high pressure and low temperature, gas hydrates are formed, consisting mainly of methane, which form vast underwater fields of gas hydrate deposits and zones of free methane discharge in the form of cold gas seeps in the top of the sedimentary cover of the Sakhalin shelf [11].

In 1998-2000 within the framework of the Russian-German COMEX project, we carried out monitoring studies of the distribution of methane fluxes in the waters of the northeastern Sakhalin shelf (fig. 1) in order to explore gas hydrate deposits [12]. As is known, methane monitoring within the sea shelves is intended to accumulate knowledge not only about the formation of oil and gas fields. It is also necessary to assess the gas geochemical situation in oil and gas fields as a prerequisite for future environmental studies.

During the period of methane monitoring on the Sakhalin shelf, there was also a unique opportunity to determine the levels of mercury in water and study the features of its behavior in the water column. Such studies have not been previously carried out in this area and are of great scientific importance due to the fact

that large emissions of methane and other reduced gases can significantly reduce the level of oxygen in sea water, which has a negative impact on marine fauna. Mercury is a dangerous toxicant and in large quantities can have a negative impact on marine biota. In particular, on the populations of the most habitat-sensitive species: gray whales and Pacific herring, for which the water area of the northeast Sakhalin shelf is the most important area for feeding and breeding. The northeastern shelf of Sakhalin is characterized by high productivity of benthic communities, which feed on the gray whales of the Okhotsk-Korean population during the annual summer-autumn feeding. Currently, gray whales are included in the category of endangered species [13].

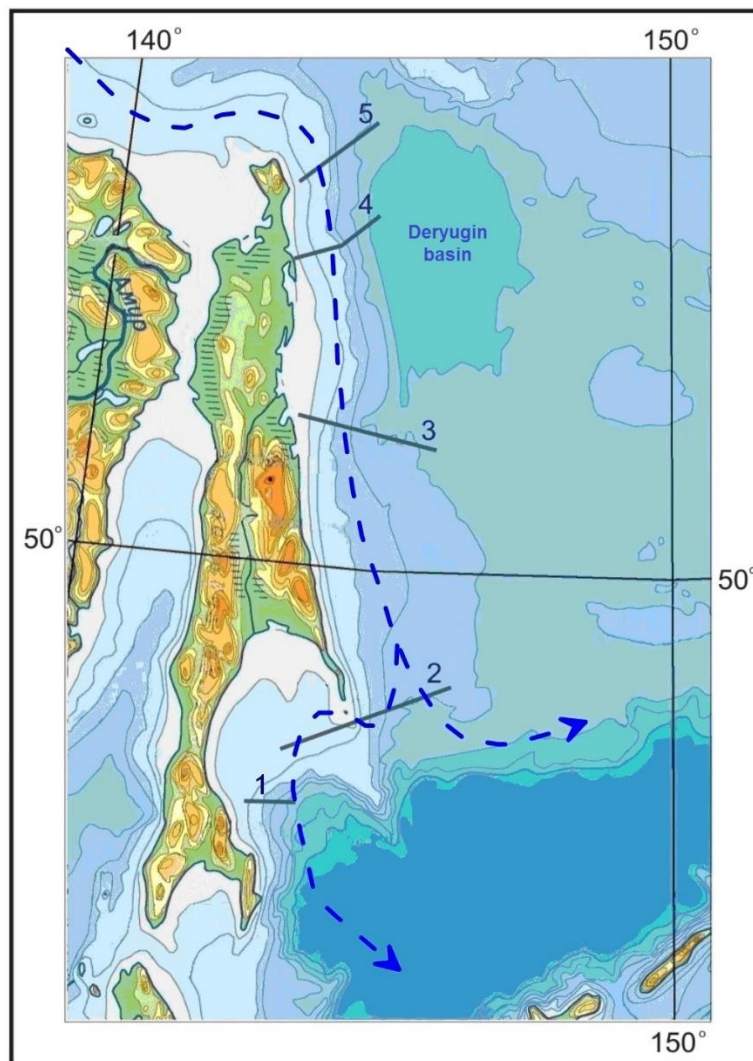


Figure 1. Map of the study area and the location of profiles of methane monitoring stations, made on the R/V Professor Gagarinsky in 1998-2000 on the eastern shelf and the continental slope of Sakhalin; Symbols: the station profiles are marked with numbers [12], the dotted line shows the East Sakhalin Current.

On the Sakhalin shelf, there is also feeding and spawning of another important commercial object - the Pacific herring. In recent years, the ecological situation has sharply worsened in this area, causing mass kills of herring and other species of commercial aquatic organisms. Thus, in June 1999, an ecological disaster occurred in the Piltun Bay, associated with the mass

death of herring that came to spawn [14]. In connection with these facts, it became necessary to clarify the causes of the existing obvious ecological trouble in this area. Therefore, in this work, we have made an attempt to find out the reasons for its occurrence, in particular, using our data on mercury. For a better understanding of the patterns of distribution of mercury

concentrations and its forms, as well as methane and other hydro chemical and hydro physical indicators of the water column of the shelf ecosystem, it is necessary to consider their interannual variability. This paper presents information on the distribution of mercury concentrations, its forms of occurrence, as well as methane concentrations and other hydro chemical and hydrological parameters in the waters of the northeastern Sakhalin shelf in the period 1998–2000. This article proposes a new approach to interpreting the data obtained in terms of assessing their possible relationship with environmental disasters that occurred in the area.

#### MATERIALS AND METHODS

The materials were obtained during three expeditions on research vessels conducted in 1998–2000 within the shelf and continental slope of eastern Sakhalin. The determination of methane concentrations in water was carried out at 21 stations on five hydrological profiles for three years (1998, 1999 and 2000), and mercury: for two years (October 1998 and June 2000). All hydrological parameters (temperature, electrical conductivity, and salinity profiles) were obtained using the Sea Bird-91 and Mark-3 CTD probes, which make it possible to build graphs of the vertical distribution of parameters in the water column directly during the sounding process. Sea water samples were taken in the water column from the seabed to the surface at the given horizons by using the Rosett system and Niskin bathometers.

*Definition of methane.* The gas phase was extracted from sea water samples using a vacuum unit and analyzed on LKhM-80 gas chromatographs. The content of methane, heavy hydrocarbon gases, carbon dioxide, oxygen and nitrogen was determined. The sensitivity of the analysis of hydrocarbon gases was 0.00001%, of other gases - 0.01%.

*Definition of mercury.* To analyze mercury, water samples taken from given horizons by using the Rosett system were passed through membrane filters with a pore diameter of 0.45  $\mu\text{m}$  to separate suspended matter. Then the samples were acidified with nitric acid and stored in polyethylene bottles until analysis at the coastal laboratory [15]. In total, 230 samples of sea water were analyzed, in which the content of dissolved and suspended forms of mercury was determined. Dissolved mercury was determined by preconcentration using the complexing agent thioxin [16].

The determination of suspended mercury was carried out according to the method used for the analysis of bottom sediments [15]. The concentrations of mercury forms in the samples were determined by flameless atomic absorption spectrophotometry [17] using the Hiranuma HG-1 and Yulia-2 mercury analyzers. To calibrate the instrument and control the quality of the analysis, a certified Japanese standard sample of mercury was used: Standard solution (0.05 mg Hg/ml) Kanto Chemical Co, Japan.

#### RESULTS

The study of the distribution of methane and mercury concentrations in sea water in the areas of natural gas emissions on the northeastern shelf of

Sakhalin was carried out in the period 1998-2000 during monitoring studies of methane within the framework of the Russian-German project COMEX [12]. The main goal was to study seasonal changes in methane concentrations in the water column of the Sakhalin shelf and the adjacent part of the Sea of Okhotsk. For research, 5 profiles of sampling points were selected (fig. 1) in zones of intensive methane degassing. The main sources of methane were coastal oil and gas fields and collapsing gas hydrate deposits. A joint study of the distribution of methane and mercury concentrations in the waters of the oil and gas bearing Sakhalin shelf was carried out for the first time in October 1998 and June 2000 on cruises on board the R/V Professor Gagarinsky [18].

The formation of background concentrations of methane and mercury in sea water occurs, as is known, as a result of the impact of a single complex of various natural environmental factors. Therefore, the distribution of the concentrations of these substances is subject to the empirical laws of the distribution of chemical elements in background media. It has been established that if the concentrations of chemical elements quite common in nature fluctuate within a few percent, then their distribution usually obeys the normal distribution law [19]. In this work, a new approach to the interpretation of the obtained data is applied, which is based on the study of N.R. Amundson and O. Bilous on the so-called parametric sensitivity of chemical processes. It is shown that in the components of the natural environment, the values of the concentrations of chemical impurities, arranged in ascending order, are combined into separate sets if their formation occurred under the influence of certain factors: either an averaged complex of environmental factors, or of single local emanations of impurity substances [20].

In accordance with this, in the annual series of conjugate associations of mercury and methane concentrations, separate associations are distinguished, which are approximated by different trends corresponding to the normal and exponential distribution laws. In this case, the background concentrations of mercury and methane form associations with a normal type of distribution, and anomalous concentrations form associations with an exponential type of distribution. The normal distribution of methane and mercury concentrations in water, which is typical for background concentrations, is the result of the summed impact of averaged natural factors on a given water area. This impact is formed under the influence of a single complex of natural processes on a global scale [19].

The exponential distribution of concentrations of substances is a reflection of the periodic effects of single factors of the same event, regularly repeating in time. In particular, the formation of series of parametric sets of methane and mercury concentrations in sea water with an exponential type of distribution could occur during periodic hydrocarbon emanations from the oil and gas deposits. At the same time, the formation of individual associations with an exponential type of distribution with different levels of

concentrations occurred apparently as a result of intermediate dilutions of emanated substances [21].

**Distribution of methane.** In 1998, four parametric sets were identified in the numerical series of the complete sample of methane concentrations in water (table 1). The largest proportion (30%) of the samples were samples with a background methane content with

a normal distribution of concentrations. The remaining 70% of the concentrations belonged to four parametric sets with an exponential type of distribution. Two samples were also recorded with "hurricane" methane contents (6248 nl/l and 11076 nl/l), which exceeded the background level by 93-165 times.

Table 1.

**Parametric sets of concentrations of the complete numerical series (152 samples) of methane concentrations in the waters of the northeastern Sakhalin shelf in October 1998**

Parametric sets	Share of complete number series, %	Concentration of methane, nl/l	Function of approximation
I (background)	30	67 (12-98)	normal
II	20	117 (102-130)	exponential
III	22	222 (139-318)	exponential
IV	21	671 (360-1263)	exponential
V	7	2598 (1350-4094)	exponential

Table 2.

**Parametric sets of concentrations of the complete numerical series (194 samples) of methane concentrations in the waters of the northeastern Sakhalin shelf in May and September 1999**

Parametric sets	Share of complete number series, %	Concentration of methane, nl/l	Function of approximation
I (background)	19	60 (15-80)	normal
II	21	133 (83-200)	exponential
III	19	330 (202-520)	exponential
IV	19	845 (525-1425)	exponential
V	11	2157 (1808-2540)	exponential
VI	11	5375 (2720-9080)	exponential

In 1999, six parametric sets were identified in the numerical series of the complete sample of methane concentrations in water (table 2). The number of samples with background methane content with a normal distribution was only 19%. The main amount of methane concentrations (81%) belonged to five

parametric sets of concentrations with an exponential type of distribution. Two samples were recorded with "hurricane" concentrations of methane (15370 nl/l and 23760 nl/l), which exceeded the background level by 256 and 396 times.

Table 3.

**Parametric sets of concentrations of the complete numerical series (154 samples) of methane concentrations in the waters of the northeastern Sakhalin shelf in June 2000**

Parametric sets	Share of complete number series, %	Concentration of methane, nl/l	Function of approximation
I (background)	20	61 (20-81)	normal
II	28	164 (83-286)	exponential
III	18	460 (360-570)	exponential
IV	10	625 (575-690)	exponential
V	9	943 (765-1165)	exponential
VI	10	2090 (1300-2895)	exponential
VII	5	4430 (3465-5510)	exponential

In 2000, seven parametric sets were identified in the numerical series of the complete sample of methane concentrations in water (table 3). The percentage of samples in the background set with a normal distribution, as well as in other sets with an exponential distribution, was approximately the same as in 1999. The main difference between the 2000 sample and the 1999 sample was a significant decrease in the level of maximum methane concentrations and absence its "hurricane" values.

**Conclusions.** In 1998, the average level of background concentrations of methane (as in 1999 and 2000) was approximately the same. However, in 1998 the proportion of samples with background methane content was 1.5 times higher than in 1999 and 2000. In 1998, the average level of maximum methane concentrations was the lowest over the observation period. At the same time, "hurricane" methane concentrations (6248 and 11076 nl/l) were recorded this year, which exceeded the background by 93 and

165 times. It can be assumed that in 1998 the seismic tectonic activation of the area began.

In 1999, the proportion of samples with background methane content in the total set decreased by 1.5 times compared to that in 1998. At the same time, the average level of maximum concentrations in that year was twice as high as in 1998. In 1999, there were "hurricane" concentrations of methane were recorded (15370 nl/l and 23759 nl/l), which exceeded the background by 256 and 396 times. In 1999, in our opinion, the maximum stage of seismic activation of the region occurred. In that year, obviously, the greatest compression of structures of faults occurred and maximum emanations of methane and liquid petroleum hydrocarbons from deposits. The appearance of water samples with a "hurricane" content of methane during the compression of blocks of the earth's crust, we associate with a certain type of geo deformation, in which the compressible surfaces experience a cylindrical bend. At the front of the shearing blocks and in the compression zones in their rear, extensional structures are formed, as a result of which the Earth's crust becomes well permeable to oil and gas emanations[22].

In 2000, the proportion of sample samples with background methane content was approximately the same as in 1999. However, in 2000 the average level of maximum methane concentrations decreased by 1.2 times compared to 1999. In 2000, "hurricane" methane

concentrations not were observed, and the maximum concentrations (4740 nl/l and 5510 nl/l) were included in the VII parametric set. It can be assumed that this year belonged to the period of decline in the seismic tectonic activity of the region.

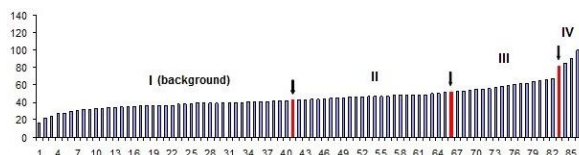
**Distribution of mercury.** In 1998, four parametric populations were identified in a numerical series of a complete sample of mercury concentrations in water. The largest proportion of the samples (45%) were samples with a background mercury content with a normal distribution of concentrations. The remaining samples (55% of the total set of concentrations) belonged to three parametric sets with an exponential type of distribution.

The levels of average mercury concentrations in I, II and III parametric sets were relatively low. They were only 4-6 times higher than 10 ng/l - maximum allowable concentration (MAC) of mercury for the waters of fishery reservoirs (table 4 and histogram of concentration distribution). The IV parametric set included the most anomalous concentrations, the proportion of which (7%) was the smallest in the total sample. This parametric set was characterized by the predominance (80%) of mercury in suspended form. One "hurricane" concentration of mercury (263 ng/l) was also recorded, which was 94% represented by its weighted form. The content of mercury in this sample was 7 times higher than the background level and 26 times the MAC level.

Table 4.

**Parametric sets and histogram of distribution of the complete numerical series (88 samples) of mercury concentrations in the water column of the northeast Sakhalin shelf in October 1998**

Parametric sets	Share of complete number series, %	Average concentrations of total mercury and percentage of its forms			Function of approximation
		Total Hg, ng/l	Dissolved Hg, %	Weighted Hg, %	
I (background)	45	36 (16-42)	85	15	normal
II	28	46 (43-51)	83	17	exponential
III	20	61 (52-67)	81	19	exponential
IV	7	104 (80-129)	20	80	exponential



In 2000, seven parameter sets were identified in the number series of the complete sample of mercury concentrations in water (table 5 and histogram of concentrations). The proportion of sample samples with background mercury content with a normal distribution of concentrations was only 13%. The remaining samples (87% of the total) belonged to six parametric sets with an exponential distribution. In I, II

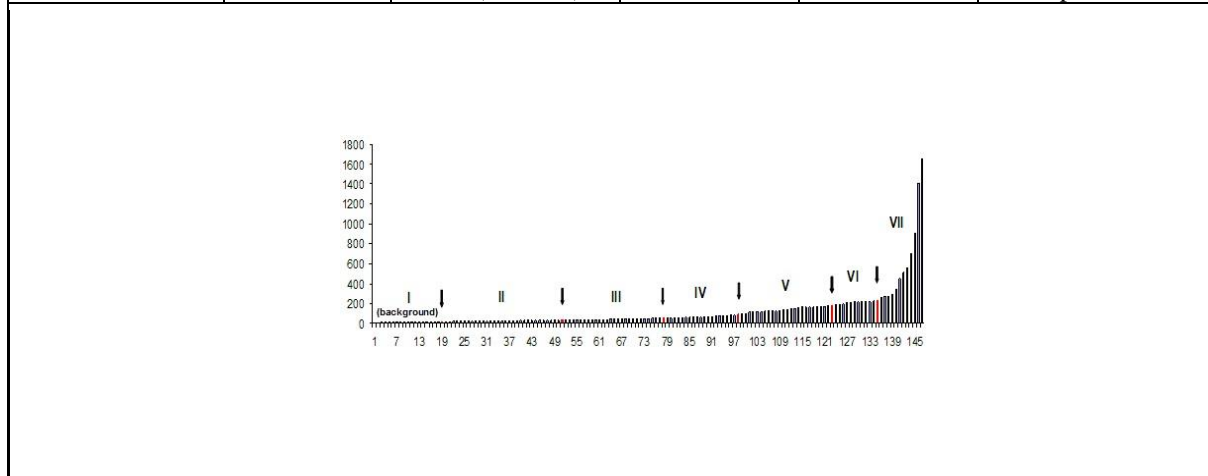
and III parametric sets the levels of average mercury concentrations were relatively low: they were only 1.5-4 times higher than the MAC level. In the IV set, the level of average concentrations of mercury exceeded the norm by 5-9 times. In V and VI sets, the levels of average mercury concentrations were abnormally high and exceeded the norm by 10-26 times. In the VII set (constituting 7% of the total sample), the levels of



mercury concentrations, which exceeded the MAC up to 90 times, can be classified as "hurricane". In this set the most "extreme" concentrations of mercury reached 1412 ng/l and 1650 ng/l.

Table 5.

Parametric sets	Share of complete number series, %	Average concentrations of total mercury and percentage of its forms			Function of approximation
		Total Hg, ng/l	Dissolved Hg, %	Weighted Hg, %	
I (background)	13	15 (6-18)	58	42	normal
II	23	28 (20-37)	70	30	exponential
III	17	44 (38-52)	70	30	exponential
IV	14	68 (53-91)	75	25	exponential
V	17	135 (100-177)	90	10	exponential
VI	9	230 (186-235)	87	13	exponential
VII	7	900 (264-905)	90	10	exponential



Parametric sets and histogram of distribution of the complete numerical series (147 samples) of mercury concentrations in the water column of the northeast Sakhalin shelf in June 2000

**Conclusions.** In 1998, the size I set of background concentrations of total mercury with a normal distribution of concentrations was quite large (45% of the numerical series). In I, II and III parametric sets the levels of average mercury concentrations were relatively low and only 4-6 times exceeded the MAC level. In these sets, the main form of mercury was dissolved its form (~80%). In the IV set (7% of the numerical series), the average level of concentrations of mercury jumped sharply and exceeded by 10 times the MAC level. In this set, the main form of finding mercury (80%) was the weighted its form.

One "hurricane" concentration of mercury (263 ng/l) was also recorded, which exceeded the background level by 7 times. In this sample, 94% of mercury was represented by a volatile suspended elemental form. In 1998, "hurricane" concentrations of methane (6248 nl/l and 11076 nl/l) were also recorded, which could be formed by methane escaping together with the gas phase from the deposits. Therefore, it can be assumed that in 1998 the seism tectonic activation of this region began.

In 2000, size of I set (background mercury concentrations) with a normal distribution was only 13%, which is almost 3 times less than in 1998. The parametric sets of the remaining mercury concentrations (except for the background set) were approximated of exponential function. This may

indicate about the rhythmic flow of mercury emanations into shelf waters from oil and gas deposits as a result of a sharp increase in seismicity. Relatively low average concentrations of mercury in water belonging to II - IV sets (28-68 ng/l) were represented on 70-75% of its dissolved form. They were 1.2-2 times higher than the background level of 1998 and more than 4 and 6 times the MAC level.

However, the average levels of mercury concentrations in V-VII sets increased sharply (by 4-9 times), and the proportion of the dissolved form of mercury in them increased to almost 90%. The level of mercury MAC in these samples was exceeded by 13.5-23 times. In VII set, which accounted for only 7% of the total sample, all levels of average concentrations could be classified as "hurricane". In our opinion, the entry of a large amount of mercury into shelf waters in 2000 (presumably also in 1999) undoubtedly had an extremely negative impact on the ecological situation in the Sakhalin shelf.

**Hydrological section of the water column.** The study of the distribution of methane and mercury concentrations and other hydrological and hydro chemical parameters of the water column was carried out on five profiles. The most representative section turned out to be 4th profile surveyed in 2000, which was located abeam the Piltun Bay (see fig. 1). This section covered all four zones of the shelf: the shallow

water zone (station G00-2 on the depth 25 m), the edge of the shelf (station G00-3 on the depth 100 m), the upper part of the continental slope (station G00-4 and G00-5 on the depth 450 and 600 m) and the deep water zone (station G00-6 on the depth 1200 m) [12].

In this section, the maximum concentrations of methane (up to 5500 nl/l) were recorded in the bottom water layer near the shelf edge, where the release of a powerful cold methane seep with a gas plume height of more than 40 m was activated. Above this seep, the maximum levels of methane concentrations are more than 100–180 times higher than its background level (30–50 nl/l) [23]. The maximum concentrations of mercury were noted mainly in the waters in the water column of the upper part of the continental slope, a well-defined temperature of the shallow part of the shelf, where its average content (322 ng/l) is 3 times

higher than at the shelf edge (103 ng/l). At the same time, the levels of maximum mercury concentrations in the same areas (1032 ng/l and 993 ng/l) were almost the same. Frontal zone of the East Sakhalin Current was recorded, formed by two converging water flows: cold methane-containing waters descending from the shelf, and deep warm sea waters that rose up the slope (fig. 2).

The frontal zone of flow was traced to a depth of 500 m. In the zone of its greatest intensification (at a depth of 200–240 m) a local maximum of suspended mercury was observed, the percentage of which was almost 3 times higher than its background content. Another local maximum of suspended mercury of approximately the same magnitude was observed at the sea surface, directly above the frontal zone (fig. 2A).

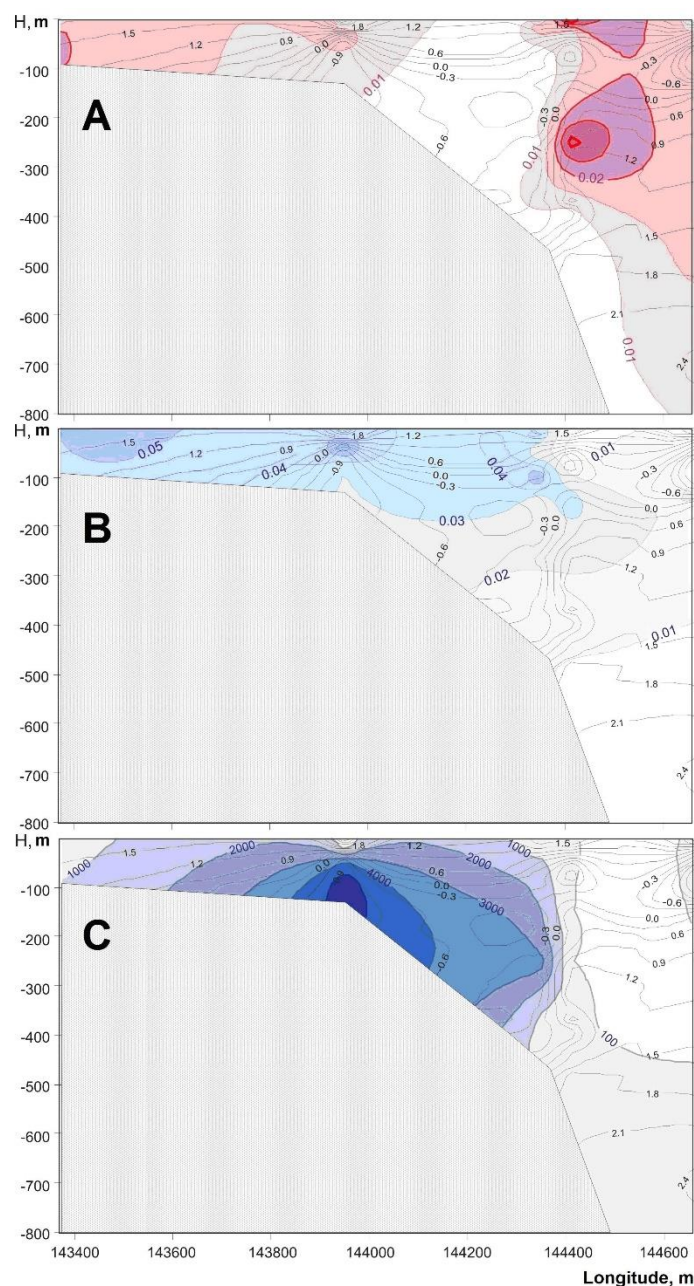


Figure 2. Distribution of isolines of water temperature, concentrations of particulate form of mercury (A;  $\mu\text{g/l}$ ), dissolved form of mercury (B;  $\mu\text{g/l}$ ) and methane (C; nl/l) on the vertical section of the water column of the eastern shelf of Sakhalin on 4th profile of stations.

In our opinion, in the zone of aggravation of the frontal flow zone, a sharp In the zone of aggravation of the frontal zone of the current, the level of content of the dissolved form of mercury was significantly (in 3 times) lower than in the surrounding waters (fig. 2B). In the frontal zone, there was a sharp decrease (15 times) in methane concentrations, almost to the background level (fig. 2C). Decrease in the concentrations of methane and dissolved mercury occurred as a result of their involvement in intensive microbiological processes.

This is possible due to the formation of numerous eddies within the frontal flow zone due to the intensive mixing of heterogeneous layers of water flows. These eddies appear to be local micro niches for a specific microbial community based predominantly on methane.

The functioning of the frontal zone of the current similar, obviously, to biofilters with a fluidized bed for wastewater treatment. In these biofilters, as a result of intensive mixing, strong turbulent flows are created, in which the activated sludge microorganisms are maintained in suspension by ascending flows [24]. In the process of microbial oxidation of methane in natural biofilters, intensive formation of the main exometabolites: carbon dioxide and water, occurs through intermediate products - methanol, formaldehyde and formate. At the same time, microorganisms of the methan trophic community also produce highly toxic methyl mercury during their metabolism [25]. Under the influence of bacteria and fungi, mercury and its compounds are oxidized, as well

as various kinds of transformations leading to mercury detoxification. Its neutralization occurs as a result of the restoration of ionic form of mercury to the elemental form, followed by its removal from the water column [26].

Within the frontal zone of the current, a significant amount of dissolved mercury is consumed and accumulated, obviously, in the tissues of methan trophic bacteria, the accumulations of which form local maxima of suspended mercury in water. In the zone of aggravation of the frontal flow zone at a depth, the maximum of suspended mercury was formed, apparently, by mercury contained in living bacteria, and in the near-surface layer - in dead micro plankton, as a result of its floating and of accumulation. There is even such a phenomenon as "anti-rain of corpses", which is due to the surfacing of dead organisms of the neuston to the water surface. Over time, part of this neuston settles, while the other part dissipates [27]. Obviously, the frontal zone of the East Sakhalin current is a natural biofilter for the purification of shelf waters from methane and mercury. Thus, the natural self-purification of Sakhalin shelf waters from pollutants occurs.

**The cold methane springs.** The study of the distribution of mercury and methane concentrations in the hydrological section of the water column on the 4th profile showed that the source of elevated concentrations of these substances are coastal oil and gas deposits, as well as cold methane seeps which located on the edge of shelf and in the upper part of the continental slope.

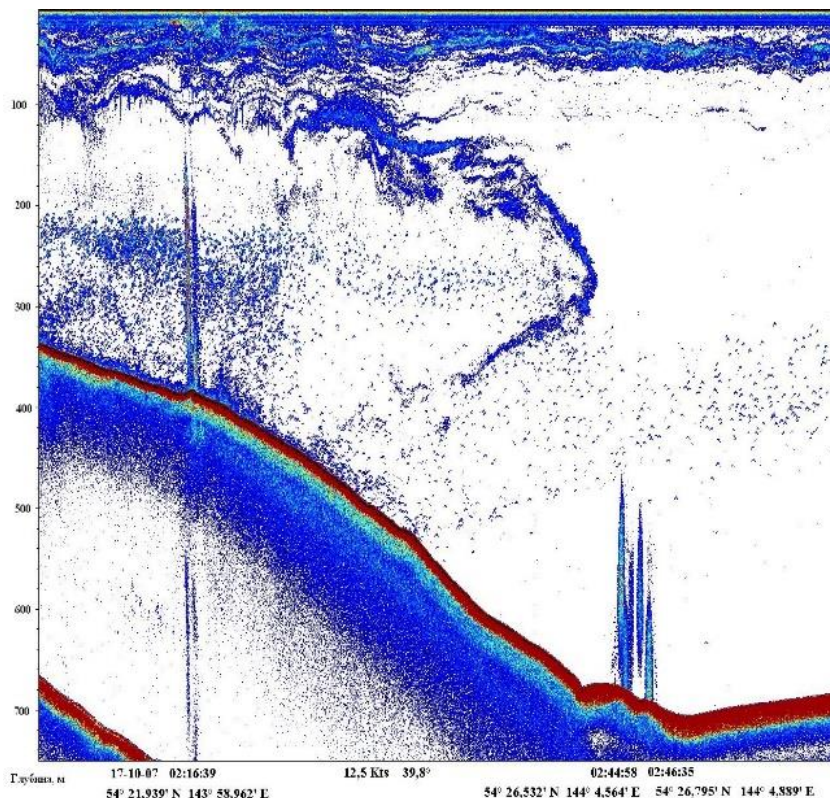


Figure 3. Acoustic echograms of methane fluxes in the water column above the gas seeps "Gizella"(1) and "Fakel Obzhirov"(2) on the northeastern slope of Sakhalin in Oct. 2007 [11].



On the northeastern shelf of Sakhalin during the monitoring studies of methane in 1998-2000 and in other years, a large number of contrasting acoustic anomalies were recorded in the water column, formed as a result of the unloading of underwater methane seeps [11]. Most seeps have a permanent location, while the mode and intensity of their eruptions are not constant over time. During periods of activation, gas seeps release a large amount of methane into the shelf waters, forming contrasting sound-scattering acoustic

anomalies in the water column (fig. 3). In the lower part of the water column, in the area where the most active methane seeps are located, which include the "Fakel Obzhirov" methane seep, the levels of anomalous methane concentrations in the water reach 11,000-24,000 nl/l. This methane seep located at a depth of 700 m on the continental slope of northeastern Sakhalin, was well studied in marine expeditions of the Russian-German KOMEX project [12, 23].

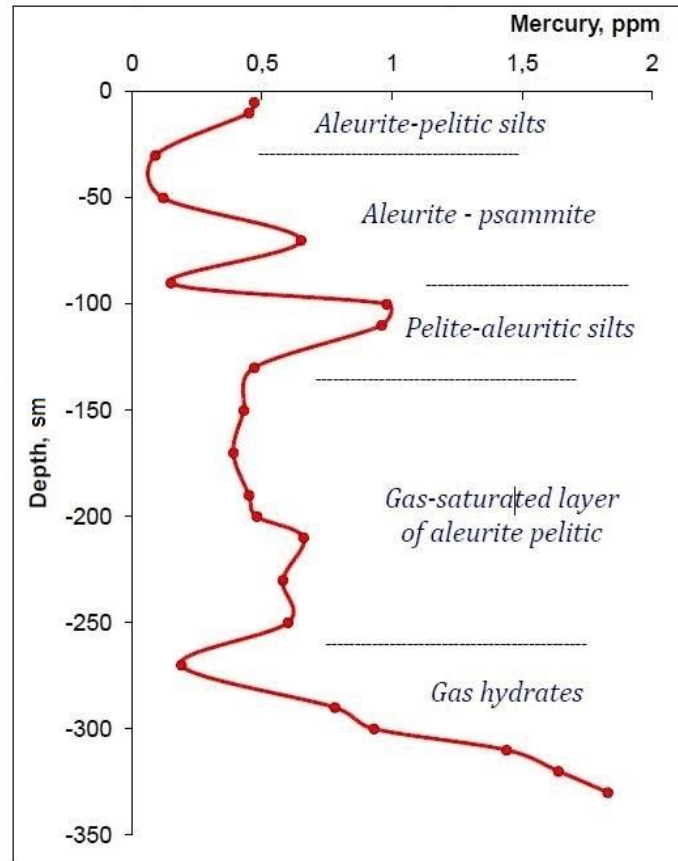


Figure 4. Distribution of mercury (ng/g) in the bottom sediments of the lithological column (Ge 99-29; Sept. 1999) from the area of the methane seep "Fakel Obzhirov" [28].

In the lithological core of bottom sediments from the area where the gas seep "Fakel Obzhirov" is located, a study of the distribution of mercury concentrations was carried out, which revealed an abnormally high level of its content in all granulometric fractions of sediments (fig. 4). The sharp change in mercury concentrations in the core was apparently due to the presence of lithological screens in the sedimentary sequence, which prevent free migration of mercury up to the bottom surface. In the upper layer of the sediment core, the mercury content (470 ng/g) was high and exceeded the background (25 ng/g) by almost 30 times. In the core at a depth of 2.5 m, a layer of sediments with gas hydrates was recorded, directly in which and below it the mercury content was anomalously high and reached 1830 ng/g, which exceeded the background level by more than 70 times.

These data may confirm that gas hydrate deposits shield endogenous mercury flows, accumulate it and are one of the potential sources [29]. Our studies have

confirmed that, at the initial stage of activation of methane seeps, they release an increased amount of mercury in suspended form into the bottom water layer [30]. Metal mercury, as is known, is very volatile and enters the atmosphere in the form of vapors during the degassing of the earth's interior. At the same time, the most significant mercury emanations from oil and gas fields and gas hydrate deposits occur during periods of seismic tectonic activation. Tectonic movements in the upper parts of the lithosphere lead to the opening of zones of deep mantle faults and the destruction of gas hydrate deposits. During these periods, intense discharge of methane and mercury into the shelf waters of Sakhalin takes place.

#### DISCUSSION

On the Sakhalin shelf, the activation of gas sources and their intensity depend on the seismic tectonic movements of the earth's crust within the Hokkaido-Sakhalin folded region. This area is part of the Pacific folded continental marginal belt, in the subduction

zones of which the Pacific Plate subsides under the continent [31]. The structural and dynamic organization of the geological environment in this area is determined by the system of the deep lithospheric faults of the Okhotsk sea plate, which has continental

and subcontinental crust [2, 32]. On the basis of the discovered gas geochemical anomalies, A.I. Obzhurov suggested that seismic tectonic activity on the Sakhalin northwestern margin of the Sea of Okhotsk began in 1988 [33].

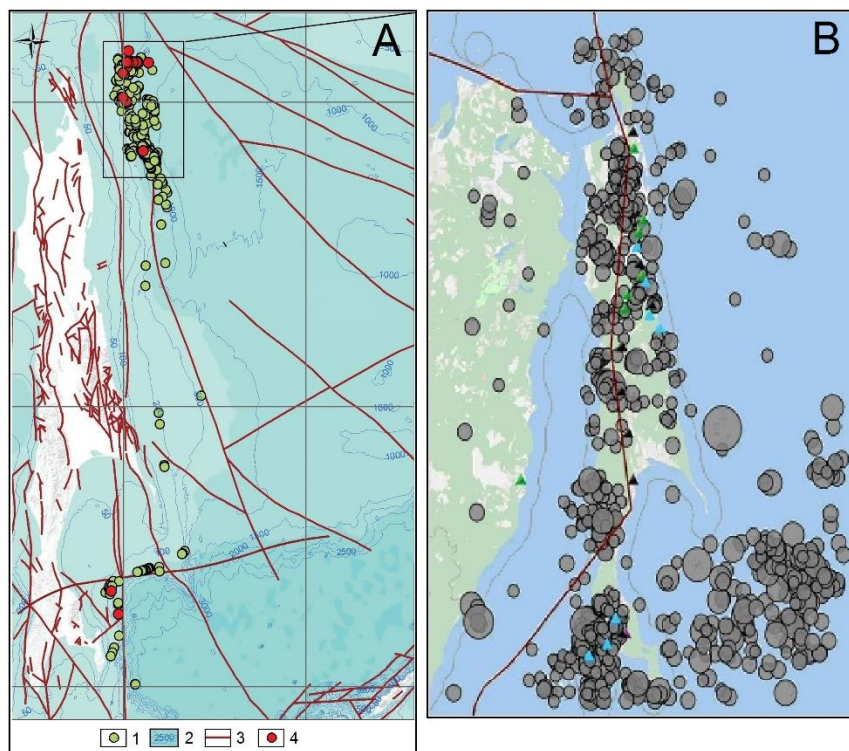


Figure 5. A. Map of faults and outflows of natural gas from the bottom sediments of the Sakhalin region. The Lavrentiev fault zone is highlighted in the upper left corner.

1 - gas flares, 2 - isobaths, 3 - faults, 4 - gas hydrates.

B. Map of epicenters of earthquakes with magnitude  $M > 3$  for the period 2003–2020. (according to the site <http://www.eqalert.ru>). The boundaries of lithospheric plates are indicated according to [38]. Triangles show the location of seismic stations [39].

At present, the tectonic system of Sakhalin is apparently developing under conditions of a right-sided submeridional strike-slip. The entire region of Sakhalin and the water area adjacent to it from the sea are subject to seismic manifestations. Earthquake sources in the form of a wide band are located along submeridional lithospheric faults, as well as at the points of their intersection with faults predominantly northeastward [34, 35]. This can be seen on the map of earthquake Sakhalin epicenters and adjacent territories (fig. 5).

During periods of seismic tectonic activation within the Hokkaido-Sakhalin fold system, earthquakes occur, which are caused by global geological and tectonic processes. Most earthquakes occur on the margins of lithospheric plates. So, in November 2015, a series of seismic events occurred within the mobile Pacific belt, indicating the movement of the Pacific plate, which was accompanied by tremors in California, Chile, Peru, Colombia, Venezuela, Brazil, Indonesia, Papua New Guinea, as well as volcanic eruptions in Indonesia and Peru [36]. Among the most powerful episodes of the El Niño phenomenon, the periods of 1982-1983, 1997-1998 and 2015-2016 can be distinguished [37].

All these episodes reached their maximum power in the eastern part of the Pacific Ocean (in the region of

the East Pacific Rise) and belonged to the "canonical" type. However, along with powerful episodes of El Niño, there are also weakly pronounced and often recurring phases of El Niño, called «Modoki», whose formation is associated with global warming [40, 41]. Relatively recently, a new model for the occurrence of powerful El Niño events has been proposed, which suggests a clear correlation between a periodic sharp increase in seismicity caused by an increase in magmatic activity in local areas located inside the Pacific Plate, near some hot spots (the archipelagos of Samoa, Fiji and Tuamotu). This causes plate movements and heating of their boundaries due to the formation of telluric electric currents, which leads to a significant increase in seawater temperature [42].

It is noteworthy that the events that took place during our research on the Sakhalin shelf in 1998 were preceded by a powerful El Niño event. As is known, during the years of the strongest El Niño of the "canonical" type in the Pacific Ocean, the seismic activity of the East Pacific Rise sharply increases. At the same time, the surface layer of water is significantly warmed up due to earthquakes and due to the release of a large amount of endogenous gases, especially hydrogen and methane [43, 44, 45]. El Niño

phenomenon was first described by Gilbert Thomas Walker [46]. It is a complex combination of external atmospheric processes caused by a complex of geophysical factors. It's leading to the development of the lunar-solar nutation oscillations in the atmosphere and the World Ocean, as well as internal processes occurring in the depths of the Earth, which form periodic pulses of increasing degassing and seism tectonic activity.

The strongest El Niño events cause not only thermal and mechanical fluctuations in the Earth's atmosphere, but also affect the frequency and intensity of earthquakes and environmental disasters within the entire Pacific mobile belt. An example of such phenomena is the devastating earthquake in the city of Uglegorsk on Sakhalin in 2000, as well as environmental disasters: the critical state of the gray whale population and the mass death of the Pacific herring that occurred during our research on the Sakhalin shelf. As is known, the summer-autumn feeding of Okhotsk-Korean gray whales occurs on the northeastern shelf of Sakhalin due to the high productivity of benthic communities in this area. According to 1998-2000 data the total number of western gray whales has decreased markedly and they are now listed as endangered species. In 1999, between 14-47% of whales surveyed were considered emaciated. Many of the gray whales died in the lagoons of the Gulf of California and while migrating to them. Almost twice as many whales died in 2000 than in 1999 [13].

Off the northeast coast of Sakhalin, there is also feeding and spawning of another important commercial object - the Pacific herring. During the breeding season, herring comes to spawn in the Piltun Bay and other desalinated bays of eastern Sakhalin. However, in recent years, the ecological situation has sharply worsened in this area, causing mass kills of herring and other commercial species of the hydrobionts. In June 1999, an ecological disaster occurred in the Piltun Bay, connected with the mass mortality of Pacific herring. Then, more than 5 thousand tons of dead herring were found on the coast of the bay. Official state bodies explained the reason for the death of the fish as a lack of oxygen in the waters of the bay due to unfavorable ice conditions. However, public environmental organizations came to the conclusion that fish poisoning was caused by oil products and dispersants used in oil spills. This could have happened as a result of the activities of Sakhalin Energy, which began commercial oil production on the Sakhalin shelf in 1998 [14].

Usually, the mass death of fish occurs as a result of the release of reactive reduced fluids (hydrogen sulfide, ammonia, hydrogen) from the lithosphere, which leads to the chemical consumption of oxygen dissolved in water [22, 47]. Mercury compounds have a high bioaccumulation coefficient and are capable of exerting the highest toxic effect on cells that form biological barriers. Mercury penetration into glial cells can initiate neuronal degeneration and lead to behavioral abnormalities in fish in a toxic environment [48]. In our opinion, these large-scale environmental

disasters on the Sakhalin shelf could be caused by earthquakes, deformation and destruction of rocks containing oil and gas deposits. As is known, during the compression of rocks from the gas cap of oil fields, first, an intensive release of hydrocarbon gases enriched in atomic mercury occurs, then - gas condensate with liquid fractions of light hydrocarbons and a highly toxic water-methanol solution enriched with organic mercury compounds are released [49].

During periods of earthquakes, large emanations of methane, mercury and other natural toxicants regularly enter the shelf waters of Sakhalin. Therefore, the natural mechanisms of microbiological self-purification of waters from toxic substances, which are subjected to biodegradation by adapted microflora, have obviously developed in the shelf ecosystem. A good example of the natural self-purification of waters is the distribution of methane and mercury considered by us within the frontal zone of the East Sakhalin current. However, during intensive oil production, which began on the Sakhalin shelf in 1998, too many different xenobiotics and toxicants enter the shelf waters. Therefore, the load on the natural processes of self-purification of shelf waters is excessive [50].

Recently, due to the sharp aggravation of the environmental situation on the northeastern shelf of Sakhalin, the question arose: "oil or fish". A few years ago, a similar dilemma was encountered in Norway. The Union of Fishermen demanded a ban on seismological surveys by oil producers in the areas of habitat and migration of fish. Some compromise was finally found, as a result of which the area of territories confiscated from fishers for oil and gas production was reduced, and the amount of harmful waste in the marine environment was also significantly reduced. The essence of the problem was expressed by the representative of the Association of Norwegian Seafarers R. Nielsen, who stressed that the in the future fishing and oil and gas industries are simply doomed to cooperation. Developing these industries, it is necessary to avoid mutual damage, since the world needs food, and it will be needed even after the oil and gas reserves on the sea shelf run out [51].

#### **CONCLUSION**

Our hydrological and gas geochemical studies on the northeastern shelf of Sakhalin revealed the following regularities:

1. During periods of stress-strain state of the subsoil and seism tectonic activation, significant amounts of mercury from oil and gas deposits periodically enter the shelf waters of Sakhalin. The levels of these emanations can be more than 100 times higher than the MPC level of mercury for the waters of fishery reservoirs, which include the Sakhalin shelf.

2. In 1998, gas and hydrothermal activity began to intensify on the Sakhalin shelf. During this period, the background concentrations of mercury with a normal distribution in the numerical series of concentrations prevailed in the waters of the shelf. In 1999 - 2000 there was an activation of oil and gas bearing tectonic structures of Sakhalin. At the same time, the level of mercury content in sea water reached abnormally high values, and the numerical series of

concentrations was characterized by an exponential type of distribution.

3. During periods of seism tectonic activation of the oil and gas bearing structures of Sakhalin, methane gas hydrate deposits are destroyed and cold methane seeps are activated. At the same time, cold shelf waters saturated with methane descend along the continental slope, where they come into contact with rising warm deep sea waters. As a result, a contrasting frontal zone of the East Sakhalin Current is formed on the slope, in the zone of exacerbation of which there is a sharp decrease in the methane content in water to the background level and intensive evaporation of the volatile atomic form of mercury, which leads to the purification of shelf waters from it.

In our opinion, the large-scale discharge of methane from cold methane seeps on the Sakhalin shelf triggers a complex mechanism of natural self-purification of shelf waters from methane and mercury. At the same time, the most toxic soluble form of mercury is converted into its low-toxic suspended form, which occurs due to the functioning of a specific methanotrophic microbiological community within the frontal zone of flow. The main accumulation of suspended mercury occurs in the surface layer of water directly above the frontal zone, from where it either evaporates or disperses in the deep part of the sea.

4. As a result of our studies in the shelf waters of eastern Sakhalin, a previously unaccounted for significant natural source of mercury was identified, which is associated with oil and gas fields. The above examples of environmental disasters that occurred in this region coincided in time with the start of commercial oil production on the northeastern shelf of Sakhalin, as well as with the period of seism tectonic activation of this region.

In our opinion, the increase in seismicity on the Sakhalin shelf could be indirectly related through a complex set of cause-and-effect relationships with planetary scale events: the activation of the Pacific Plate and the unusually strong episode of the El Niño phenomenon of 1997-1998. The superimposition of anthropogenic stress during this period, caused by intensive oil production, on natural intense emanations of mercury in water Sakhalin shelf, is, in our opinion, the main cause of environmental disasters. Such catastrophes can be repeated, especially during periods of significant seism tectonic activation of the interior.

5. Since during periods of seism tectonic activity there is a high probability that significant mercury emanations from oil and gas fields will enter the shelf waters of Sakhalin, it is necessary to inform the oil industry about the real threat of a recurrence of such environmental disasters. The solution to the environmental problem associated with mercury may be to change the existing regulations governing offshore oil and gas production. It is also necessary to submit the information received for public discussion and take special environmental measures in this area of activity, up to the cessation of oil and gas production on the Sakhalin shelf.

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