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Assessment of Hydrocarbon State of Soil and Bottom Sediments of Arctic Zone of Yakutia

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***Abstract* – The study of soil samples and bottom sediments from the Arctic region of the Sakha Republic (Yakutia) in the coastal zone of the East Siberian Sea (Nizhnekolymsky region) was conducted. The yield of chloroform extracts and the nature of the IR spectra were used to evaluate the hydrocarbon composition of the samples taken. The localized areas with high levels of pollution were identified. All of them were located within the territories of man-made objects. The remaining territories correspond to the background indicators or have insignificant traces of pollution. Following the results of microbiological studies the degree of enrichment with microorganisms of studied soil and bottom sediment samples belong to the category of poorly enriched. It was possible to identify hydrocarbon oxidizing microorganisms, which differ only by their poor qualitative and quantitative composition in some oil-contaminated samples. The low activity of hydrocarbon oxidizing microflora against poor soil enrichment with microorganisms indicates the sluggishly ongoing self-healing processes of soils of the Arctic region.**

***Keywords* – geochemical background; oil pollution; chloroform extract; bottom river and sea sediments; hydrocarbon oxidizing microorganisms; soil self-regeneration.**

I. INTRODUCTION

High consumption of hydrocarbons by modern society induces the exploration of oil and gas. According to geologists, the Northeast Region of the Arctic is particularly promising in this direction. The oil and gas reserves can be up to 80% of Russia’s hydrocarbon potential as a whole on the shelf of the Northern Seas of the Russian sector of the Arctic [1]. The activities of oil and gas industrial complexes are inevitably associated with various spills and leaks. The prospective development of the Arctic region, mainly associated with the extraction of hydrocarbons, leads to the need to solve environmental problems aimed at protecting the environment from oil pollution [2]. Oil and petroleum products are the most dangerous sources of pollution and represent a threat to the Arctic environment [3]. As a result, among the important tasks there would be timely detection of oil pollution and the development of methods for its elimination, which would minimize the negative effects of anthropogenic impact on the environment.

The study of the bottom sediments of the Arctic zone for the content of hydrocarbons aimed at environmental safety preservation has already been carried out by some researchers [4-6]. The analysis on the hydrocarbon content in the bottom sediments and soils requires a differentiated approach to isolate the anthropogenic component against the background of natural organic matter. As a result, the study of natural geochemical background is of particular value, since it allows to use the results of analytical studies for the purpose of identification of oil pollution, taking into account the contribution of hydrocarbons of natural origin [7-9].

Under the cryolithozone conditions, i.e. low soil temperatures, all metabolic processes associated with the growth and activity of microorganisms and terrestrial vegetation are extremely slow. This predetermines very low rates of soil self-regeneration processes when they are polluted with oil or oil products. The self-regeneration period can last for many decades. Currently, the most effective, environmentally friendly and preferred ones are biological methods of cleaning soils from oil pollution [2, 10, 11]. Different physical and chemical composition of soil and a landscape of soil microflora dictate the need to develop recovery measures separately for each specific region. As a result, it is necessary to know the geochemical background of the hydrocarbon state of soils and bottom sediments, the study of their microbiological activity in order to identify native hydrocarbon oxidizing microorganisms (HOM).

In addition, a comprehensive study of geochemical background of hydrocarbon-containing components and the microbiological activity of soils and bottom sediments has important scientific and practical importance, since it will allow us to assess the current state of soil of the cryolithozone before an intensive man-made intervention.

II. Subject and Methods of Study

The object for research was Nizhnekolymsky District of the Sakha Republic (Yakutia). Nizhnekolymsky District is a potentially oil and gas bearing area located in the Northeast sector of the Arctic coast. It is washed by the waters of the East Siberian Sea (ESS) in the north. The water area of the ESS is not well investigated. The largest river flowing into the ESS is the Kolyma River. Its flow mainly determines the nature of sedimentation in the coastal zone of the sea [8]. In the territory of Yakutia on the Kolyma River there are three large tank farms and a seaport the Green Cape. Transportation and storage of petroleum products increase the risks of environmental pollution by oil, which may get into the river channel and further along the river flow into the coastal part of the sea. The samples for research were taken at the end of August 2018. The study covered the coastal zone of the ESS, i.e. the river and the sea bottom sediments, the coastline of the lower reaches of the Kolyma River, the right bank (soils of clean areas and the areas with high man-made pressure: tank farms, pier and a seaport). Sampling sites and their characteristics are given in Table 1.

In accordance with the atlas of the soil and geographical regionalization of the Sakha Republic (Yakutia), the sampling for the study was made from the territory located in the polar soil-bioclimatic belt; the Eurasian polar soil-bioclimatic region; tundra plant-soil zone, subarctic subzone; in the Indigiro-Kolyma soil province; the northeast flood plain [18]. The flood plain swampy (acidic), tundra glei peaty-humus and taiga glei and glei undifferentiated main soil types are typical for this area.

The soil samples were taken from the depth of 0-20 cm in accordance with the regulatory documents. All samples were taken with a clean instrument. The background samples were taken at a distance of at least 100-150 m from the contaminated area. The bottom sediments were collected using a dredger: The river sediments were taken from the depth of 1-2 m, the sea sediments were taken from the depth of 4-5 m. The binding on the terrain of the observation points, reference sections, sampling points of soil and bottom sediments was carried out using a GPS receiver and landmarks. After the sampling the soil samples were packaged in polymer bags. All packages with the samples of soil and bottom sediments were labeled with the number, date, place and time of sampling. For the purpose of further study, the samples were preserved by freezing. The samples were delivered to the laboratory for geochemical and microbiological tests in the cooler bag. To carry out the test, the delivered samples were divided into two parts. One part, intended for microbiological research, was stored in a freezer. Another part of the samples was prepared for geochemical studies: dried, sifted out from the roots of plants, and ground.

To determine the content of oil and oil products in the soils and bottom sediments, the conventional methods were used [12]; however, as shown by the results of our studies [13] and based on other authors [6, 10, 14] these methods are applicable only to fresh spills, since they do not take into account the contribution of asphaltic-resinous components that are formed in both the soils and the bottom sediments in the processes of oxidative destruction of oil pollution. It is especially important to take this fact into account in order to identify old pollutants where the asphaltic-resinous components predominate [15]. As a result, the soil samples and the bottom sediments were studied for the content and composition of hydrocarbon-containing components according to the methods of studying organic matter of sedimentary rocks adopted in geochemistry [13]. The first stage of these studies was the extraction of samples with chloroform. For the environmental purpose the method of cold chloroform extraction is applied. The level of oil pollution was assessed by the yield of chloroform extracts (CE) in comparison with the geochemical background. In the selected extracts, the structural group composition was studied by the method of IR-Fourier transform spectroscopy (IR-Fourier transform spectrometer Protege 460, Nicolet).

Microbiological studies of the samples were carried out using the following methods of analysis:

- determination of relative humidity and absolute dry weight (ADW) of bottom sediments and soils;
- determination of pH of the soil suspension and bottom sediments;
- determination of the number of microorganisms;
- identification of HOM from the background and oil-contaminated soil samples and bottom sediments;
- determination of HOM activity.
- determination of relative humidity and ADW of bottom sediments and soils.

For the purpose of cultivation and accumulation of HOM the mineral medium of Müntz with the oil was used. The pure cultures were obtained using the Koch and Drigalsky method, considering that one bacterial cell forms one colony on a dense medium [16]. The cultures were stored in a refrigerator at the temperature of +4±1°C with the following frequency of passages: 1 time in 10-14 days.

TABLE I. CHARACTERISTICS OF SAMPLES AND BOTTOMED SEDIMENTS

No.	Types of samples	Sampling point	Yield of CE %	pH	Rel.hum.[a] %	TMN[b]	Fungus	HOM
						ufc/g ADW of soil		
1	soil	Nizhnekolymskaya tank farm, Green Cape vil. 68º47.163'N 161º22.303'E	3.268	5.5	16.5	$2.2\cdot10^5$	$3.3\cdot10^7$	$4.2\cdot10^4$ *Pseudomonas*
2	soil		9.719	5.6	32.0	$2.8\cdot10^5$	$2.1\cdot10^7$	$2.2\cdot10^4$ *Pseudomonas*
3	soil		12.298					
4	soil		7.827	5.6	20.2	$3.6\cdot10^5$	$9.0\cdot10^6$	0
5	b/s[c]		0.051	6.2	24.2	$2.9\cdot10^6$	$3.3\cdot10^4$	0
6	b/s	Green Cape seaport	0.055	6.2	36.5	$2.2\cdot10^6$	$5.8\cdot10^4$	$22.0\cdot10^5$ *Bacillus*
7	b/s		0.037	6.3	36.2	$3.1\cdot10^6$	$1.6\cdot10^4$	$1.0\cdot10^5$ *Aspergillus*
8	b/s	Mooring line Kolyma River, vil. Chersky	0.006	6.3	25.7	$3.3\cdot10^6$	$3.9\cdot10^4$	0
9	soil	oil spill in the boiler room	0.051	5.7	26.1	$3.8\cdot10^5$	$4.2\cdot10^6$	0
10	soil	Green Cape vil.	0.053	5.3	21.4	$1.3\cdot10^6$	$3.9\cdot10^6$	0
11	soil	Gas station	2.722	5.2	9.6	$1.6\cdot10^6$	$6.0\cdot10^6$	0
12	soil	Tank farm Nizhnie Kresty, vol. Petushki	0.176	6.0	12.9	$2.2\cdot10^5$	$1.2\cdot10^6$	0
13	soil		4.815	5.6	13.2	$6.0\cdot10^5$	$4.2\cdot10^6$	0
14	b/s	Kolyma River 68º59.572'N 161º33.211'E	0.039	6.2	56.4	$3.2\cdot10^5$	$4.6\cdot10^4$	0
15	b/s	68º59.572'N 161º33.211'E	0.018	6.1	20.3	$2.0\cdot10^5$	$8.1\cdot10^4$	0
16	soil	69º03.826'N 161º29.725'E	0.011	6.2	20.9	$1.5\cdot10^5$	$2.2\cdot10^4$	0
17	b/s	69º03.826'N 161º29.725'E	0.004	6.3	26.6	$3.3\cdot10^5$	$<10^3$	0
18	b/s	69º08.367'N 161º30.389'E	0.012	6.2	33.4	$3.2\cdot10^5$	$<10^3$	$1.4\cdot10^4$ *Pseudomonas*
19	b/s	69º11.250'N 161º29.940'E	0.013	6.1	21.7	$8.0\cdot10^5$	$<10^3$	0
20	b/s	69º12.429'N 161º26.426'E	0.033	6.2	49.5	$6.4\cdot10^5$	$<10^3$	$1.2\cdot10^4$ *Pseudomonas*
21	b/s	69º15.276'N 161º26.957'E	0.016	6.1	59.6	$6.2\cdot10^5$	$9.0\cdot10^6$	0
22	b/s	ESS 69°47.6783'N 161°22.4390'E	0.006	6.6	31.6	$2.2\cdot10^5$	$5.2\cdot10^4$	$1.8\cdot10^4$ *Penicillium*
23	b/s		0.007	6.5	30.4	$2.8\cdot10^5$	$<10^3$	0
24	b/s		0.008	6.6	38.3	$3.1\cdot10^5$	$2.6\cdot10^4$	0
25	b/s		0.007	6.8	32.4	$2.6\cdot10^5$	$<10^3$	0
26	b/s		0.007	6.6	33.1	$8.2\cdot10^5$	$<10^3$	0

a. a is a relative humidity;

b. b is the total microbial number (TMN);

c. c is the bottom sediments.

III. Results

The results of chloroform extraction showed that in some samples taken from the existing Nizhnekolymsky tank farm and Nizhnie Kresty tank farm in the village of Petushki, the operation of which was discontinued in 2007, the gas stations had 2.722÷12.298% of CE, which is much higher than those for the background soil samples (0.011 ÷ 0.053%) (Table 1). Such a high number of CE over the background ones may indicate the soil contamination with oil or petroleum products. According to the Goldberg classification [17], the identified oil pollution can be classified as high (2.722 ÷ 4.815%) and very high (7.827 ÷ 12.298%). In the remaining samples, the CE outputs are close to the background values (0.004 ÷ 0.055%). To determine the possible contamination of samples with oil, it was necessary to conduct more detailed studies of the chemical composition of the samples. To do this, the CEs were studied by the method of IR-Fourier spectroscopy.

Fig. 1 shows the IR spectra, the most typical for CE samples of the study area. Thus, the spectra “a”, “b”, “c” are the IR spectra of the background samples of soil, river and the sea bottom sediments. Their spectra are typical of modern precipitation. They are characterized by the intense absorption of oxygen-containing groups: absorption bands (a.b.) in the region of 3300-3400 cm^{-1} (hydroxyl groups), a.b. - 1700 cm^{-1} (carbonyl groups), a.b. - 1170 cm^{-1} (ether bonds) and hydrocarbons with a long branch: doublet a.b. - 720 and 730 cm^{-1}. The IR spectra of CE soils polluted with oil are more of hydrocarbon and aromatic character in comparison with the background. For example, the figure shows the IR spectrum “d” of soil samples with a high level of oil pollution. Similar spectra were obtained for another oil-contaminated soil samples from the areas of tank farms. In the IR spectra of the extracts of these samples, the intensity of a.b. is high amounting to 1460, 1380 cm^{-1}, which is typical of the methyl and methylene groups, as well as a.b. amounting to 1600, 810, 750 cm^{-1}, which is typical of aromatic structures. There is no doublet in the region of 720 and 730 cm^{-1}. It should be noted that according to the IR spectra these contaminants should be characterized as partially oxidized or degraded since they preserve a high content of oxygen-containing groups (3300-3400, 1700, 1170 cm-1). On the contrary, the “e” spectrum is typical of contaminations that have not yet been degraded; there are practically no a.b. in the spectrum of oxygen-containing groups. Such a character had the spectra of CE samples from the gas station ana fresh spill in the boiler room. And, finally, the “f” spectrum characterizes bottom sediments with the traces of oil pollution. Such IR spectra were typical of some samples of the river bottom sediments collected near the Green Cape seaport. The spectra of these samples differ from the background ones in a higher content of aromatic structures (a.b. 1600 cm^{-1}), as well as methyl and methylene groups (a.b. 1460, 1380 cm^{-1}). Another samples were clean.

Thus, the method of IR-spectroscopy provides a large amount of information, allowing to identify oil contamination even in the trace amounts when the output of CEs fluctuates at the background level. Besides, based on the IR spectra it is possible to understand the course of oxidative decomposition or degradation, which allows us to give an approximate estimate of the duration of the presence of oil pollution in soils or bottom sediments.

As a result of microbiological studies, the presence of viable microflora with various physiological and biochemical characteristics was established in the studied samples, which indicates the biological activity of microbiocenoses of background samples of permafrost soils and oil-contaminated ones. In terms of qualitative composition, the psychrophilic fungal forms and spore-forming bacterial groups of microorganisms dominate in soils. According to the degree of enrichment, the studied soil samples can be classified as poorly enriched. The studies have shown that the soil moisture, both background and polluted, is small and insufficient for the favorable development of soil microflora (Rel. hum. Is 9.6 ÷ 26.1%).

Near the Green Cape village, in the area of the Nizhnekolymsky tank farm, the background soils were slightly acidic (pH 5.3) (Table 1). The weakly acidic nature of the soil is also a characteristic of oil-contaminated soils of this territory (pH 5.6). Perhaps in this connection, the fungal forms of microorganisms (9.0 106 ÷ 3.3 107 ufc/g ADW) prevailed over the microbial (2.2 105 ÷ 3.6 105 ufc/g ADW) in soils. This is probably due to the fact that the fungal forms of microorganisms prefer a more acidic medium in comparison with microbes.

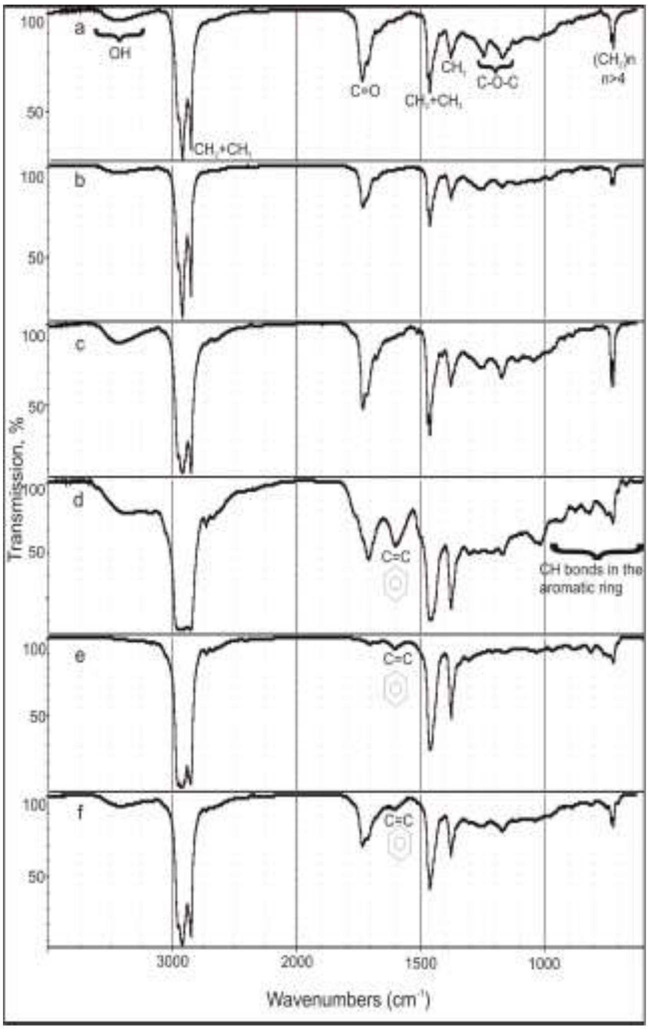

Fig. 1. IR spectra of chloroform extracts of background samples: “a” - soil, “b” and “c” - river and sea bottom sediments and oil-contaminated samples, respectively: “d” - degraded oil pollution; “e” - fresh oil pollution; “F” - traces of oil pollution.

The river and sea bottom sediments near the border of the Kolyma River - the East Siberian Sea are characterized by the medium close to neutral (pH 6.2 ÷ 6.8). In almost all of these samples, the bacterial groups of microorganisms predominated over the fungi (2.2 105 ÷ 3.1 106 ufc/g ADW versus <103 ÷ 5.2 104 ufc/g ADW).

Hydro-oxidizing microflora in the soil samples and the bottom sediments from the territory of the Nizhnekolymsky tank farm, was mostly represented by fungi of the genus *Aspergillus*, spore-forming bacteria of the genus *Bacillus* and nonfermentable bacteria of the genus *Pseudomonas*. From the samples of the bottom sediments on the border of the river Kolyma and the East Siberian Sea, the microscopic fungi of the genus *Penicillium* were identified, and from the bottom sediments of the river Kolyma (lower current, right bank) the nonfermentable bacteria of the genus *Pseudomonas* (P. *aeruginosa*) were identified. It should be noted that the number of identified HOM from the oil-polluted areas is small (about 104 ufc/g ADW). This is not enough for the active process of biodegradation of oil pollution.

The results showed that it was not possible to identify HOM in all soil samples with a high level of oil pollution, i.e the gas station territory, the Nizhnie Kresty tank farms (Petushki village) and some areas of the Nizhnekolymsky tank farms. In the case of long-term oil pollutions, this may be due to the exhaustion of readily available hydrocarbons in the soil because to their oxidative destruction. Over time, as the oil or the oil products are oxidized, the nutrient medium for HOM becomes more scarce and the number of microorganisms decreases. However, the number of HOM is small in polluted soils where an emergency oil spill had occurred relatively recently (2 years ago). As it has beeb noted, in the sample from the territory of the gas station it was not possible to identify HOM despite the fact that this territory is most often exposed to various kinds of leaks. The scarcity of the qualitative and quantitative composition of the identified HOM should be also noted.

The obtained data are in good agreement with the results of studies [9, 19, 20] on self-restoration of soils from oil pollution in the areas with extremely low temperatures. The authors also talk about slow self-restoration of soils of the northern ecosystems and the need to develop effective biological methods for cleaning them from oil pollution.

IV. CONCLUSION

In the Nizhnekolymsky District of the Arctic region of the Sakha Republic (Yakutia) the soil samples (tank form, gas station, seaport, Kamenny Cape) of the river (lower course of the Kolyma river) and the sea (East Siberian Sea) bottom sediments were selected. The sample studies allowed the local sites with high levels of contamination to be identified. These sites are located in the territories of the objects experiencing high man-made pressure (tank farms, gas stations). According to the results of studies using the IR-Fourier spectroscopy, an assessment was made of soil samples and bottom sediments with low CE (at a background level) for contamination with oil and oil products. All samples of the bottom sediments and the soils outside the man-made objects turned out to be clean or had traces of oil pollution.

The HOMs were identified from several soil samples with a high and very high level of pollution (the territory of the Nizhnekolymsky tank farm). However, they are not predominant in the total microflora and are characterized by poor quality diversity. The fungal forms of microorganisms predominate in these soil samples, which almost do not participate in the processes of soil self-purification from the oil pollution. In the remaining contaminated soil samples, it was not possible to identify HOM at all, even where there were fresh oil spills. The insufficient activity of HOM against the background of weak soil enrichment with microorganisms indicates a sluggishly ongoing self-restoration process of soils of the Arctic region.

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