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The questions of environmental monitoring and rehabilitation of oil –contained soils in the Arctic zone of Yakutia.

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Abstract

In the study of permafrost soils to characterize the features of the natural background, the identification of oil pollution of soil, its composition and characteristics of the boundaries, it was used a complex of analytic methods: IK-Fyrie spectroscopy, gas-liquid chromatography, chromatography mass-spectrometry and geochemical approach to the interpretation of the data. The results of an experiment of the study of the degradation of oil pollution in the soils of permafrost under the effect of oil destructors on native micro flora of hydrocarbon.

Keywords: *oil pollution, biodegrading, permafrost soil, micro flora, the natural background.*

On the shelf of the northern seas of the Russian sector of Arctic oil and gas reserves, according to geologists, may be up to 80% of the hydrocarbon potential of Russia as a whole. Development of these resources in harsh environments is a difficult technical problem, including the creation of conditions required for environmental management, industrial and environmental safety. Prior to the implementation of large-scale projects on hydrocarbon production is important to assess the ecological status of Arctic shelf to oil pollution (OP).

A significant contribution to pollution made by the seaports, transportation - sea and river fleet, oil facilities and vessels. Bottom sediments of port facilities in coastal bays of the Arctic seas are characterized by high concentrations of petroleum hydrocarbons. Of great importance is monitoring and evaluation of coastal ecological status of the territories in relation to human activities, leading to a change in the environment.

In arctic and subarctic regions, in contrast to regions with favorable climatic conditions the processes of biodegradation of spilled oil and NP occur much more slowly, which determines a lower capacity of permafrost soils to cleanse itself. The presence of closely overlies the permafrost, low temperatures and short growing season - all this leads to lower rates of transformation of oil pollution.

The state of the environment of the Arctic and subarctic zones of Yakutia is practically unknown. Oil and NP are the most dangerous sources of pollution and are a particular risk of pollution and environmental threat to the Arctic environment. They are delivered to the Arctic regions along the rivers. Most oil depots are located on the banks of rivers and coastal zone of the Laptev Sea. As a result of spills during transportation and storage depots in the operation is contaminated water and the shore of the land. International experience of events shows that in the arctic conditions it is possible to collect and dispose of only 10-15% of spilled oil. The residual oil contaminations in permafrost, surviving for many years, it is source of petroleum hydrocarbons in runoff into the sea and its coastal part. Thus, initially the local oil pollution can spread over large areas. Specific features of the vast arctic and subarctic regions are the presence of permafrost soils, long-term freezing of ponds, marshes and wide distribution of wetlands [1, 1993]. The soils of northern latitudes, which have a low biogenic poor talent to heal itself. When oil spills, soaks the moss surface, penetrates to the zone of thawing and continues to spread laterally over this area, undergoing a slow transformation. Most of the spilled oil remains in the top surface (10 cm) layer of the soil [2, 2003].

In this regard, on the northern territories of environmental monitoring facilities NGK is one of the priorities in order to minimize damage as a result of human impact on natural ecosystems in the Arctic.

In the Republic of Sakha (Yakutia) in the last 5 years there have been several major accidents involving oil spills, and NP, including in the Arctic and subarctic zones. In 2008 there was an emergency spill of diesel fuel "Arctic" in the stock of fuel and lubricants in Moma district, in 2009, according to official data, in the waters of the Gulf Bulunkan Laptev Sea (Tiksi n) as a result of the accident got 37 tons of oil. Many northern regions of Yakutia for heating homes using crude oil. Her delivery and storage, as seen from the examples, pose a threat to the environment.

All of this is the solution of tasks, which include: prolonged monitoring of areas of the Arctic zone in the modern analytical level, especially in areas placing oil and gas facilities. An important direction is to study the ability of northern ecosystems to recover when hit by oil components and the NP. Of particular importance is research aimed at developing technologies for the elimination of oil pollution and the creation of oil destructors, effective in extreme climatic conditions of the subarctic zone.

In our opinion, the issues of monitoring oil pollution in the area and the NP and scientific support for the rehabilitation of contaminated territories were more likely to promote the use of methods of organic geochemistry.

One of the key tasks of geo – chemical monitoring is the research of informative and reliable parameters for the diagnosis of soil and sediment of oil and NP to evaluate the quality of the sewage treatment works and the choice of destructors, the most effective for arctic and subarctic conditions. As practice has shown environmental studies, existing methods for determining oil pollution in the soils of the methods of fluorescence and infrared spectroscopy [3, 4] should be considered as an express definition for oil pollution in case of fresh spills and mass analysis of a large number of samples to identify areas of possible contamination.

Their weakness is a significant understatement of the content of NP in soil samples was due to incomplete definition of a light hydrocarbon (HC) - gasoline and kerosene fractions and high HC and getero taking components - resins and asphaltenes [5, 2007]. Analyst in determining the content of NP in soils should be clear that the extracts selected for their composition is a mixture of hydrocarbon compounds of man-made, i.e., the actual oil pollution, and natural origins - organic matter (OM) of soils. Each of these components is characterized by a set of large number of hydrocarbon compounds, whose content may vary over a wide range.

One way to assess anthropogenic component (residual oil pollution, or NP) is proposed in the above-mentioned methods [3, 4], in which the contribution of natural soil OM is minimized through the use of the lightest of the solvent - hexane and adsorption of heavy hydrocarbons and asphalt-resin components columns with a aluminum dioxide. These techniques are widely used to assess oil-contaminated soils and sediments in the light spills NP and fresh oil spills, but they do not meet the objectives of long-term monitoring of contaminated areas and evaluating the work of rehabilitation, as well as spills of heavy NP.

When monitoring contaminated areas the researcher faces the task definition of oil pollution on the background of the presence of native soil OM and sediment. On the non-standard and

complexity of the problem and possible ways to solve a lot of useful information can be found in [6-8]. Because of the complexity and diversity of pollutants and the characteristics of soil OM question should be decided in each case, specifically, in accordance with the task. There may be a useful experience in geochemical studies of rocks and RH naphthides.

In the study of organic geochemistry of recent sediments OM contains information on research findings bituminous component agents (bitumen), the features of their structure for the agents of different types of soils and sediments are reviews of modern methods of analysis of organic compounds. Significant variations in the content of bitumens in the OM of modern sediments and diversity of their composition leads to an understanding that the contribution of the native bitumens OM in the total content of the extract of the analyzed samples from contaminated areas can not be reduced to simple arithmetic subtraction "average natural background" as defined to break bitumoid in background samples [9]. Moreover, as shown by geochemical studies, background samples themselves, which are selected on the "clean" areas may be contaminated with NP. Knowledge of the regional natural background, the specific composition of the RH soils or sediments must, from the stage of selecting background samples, which requires specialized geochemical studies.

On the other hand, in organic geochemistry provides extensive and detailed information on the composition of the condensates and oil characteristics of the distribution of individual hydrocarbons, the presence in their composition of hydrocarbon biomarkers. The data in each case can be very useful in the identification of a particular type of pollutant that requires a special study of a pollutant source.

The petroleum geochemistry with a high degree of detail and studied the issues of oil degradation in terms of deposits, and the results of the model experiment. The data on the processes of transformation of oil as a result of bacterial oxidation of hydrocarbons under aerobic conditions, the selectivity and the phasing of these processes can be very useful in studying the characteristics of the degradation of oil pollution in time. The results can serve as a basis for the creation of new efficient oil destructors, using indigenous micro flora in the development of methods of oil spill.

However, it is necessary to take into account that under natural conditions, the influence of factors such as the contribution of native soil OM, and the possible imposition of secondary pollutants may actually bio degradatsionnyh to snivelirovat effect of changes in the composition of oil pollution, and thus lead to underestimation of the effectiveness of oil destructors. The criteria for a correct evaluation of the effectiveness of developed oil destructors, and quality control for the remediation of contaminated soils remidiatsii is an independent scientific challenge that requires special features of the transformation studies of oil pollution in different contexts, taking into account the influence of natural background and secondary factors.

The Institute of Oil and Gas Problems SB RAS for 11 years of environmental studies are conducted to monitor different areas of oil and gas complex of contamination of soils and sediments by oil and PP in the subarctic zone of Yakutia. One of the ways is to increase the effectiveness of monitoring through the development of informative and reliable parameters to assess the

contamination of soils and sediments by oil and NP. The results obtained may be useful in connection with the planned research on the environmental assessment of the Arctic zone of Russia.

In our studies used a set of analytical methods of organic geochemistry: FT-IR spectroscopy, gas-liquid chromatography, gas chromatography-mass spectrometry and geochemical approach to the selection of analytical parameters, which has enabled to take into account the specifics of contaminants and the native soil OM and sediment - natural background. The proposed geochemical parameters have several advantages over the methods of [3, 4] and allow: 1) differentiate the oil pollution from the native soil OM and sediments, 2) to determine the residual hydrocarbon oil spills as a light - gasoline, kerosene, condensate, and heavy NP - oil, fuel oil, butter, which is especially important for long-term monitoring of contaminated areas, and 3) identify and define the nature of the distribution of individual hydrocarbons in the oil pollution [10, 2004, 11, 2007].

In our view, it is possible to improve the quality of monitoring and at the present level of the analytical approach to the study of important environmental problems. These include the definition of a regional natural background, the study of the current state of soil and sediment contamination in the oil and NP in the oil and gas facilities, the identification of man-made anomalies, determine the level and composition of the pollution, the study of the transformation of oil pollution in natural conditions and under the influence of oil destructors, performance evaluation of the quality of remediation work in contaminated areas.

In this paper the results of studies on the degradation oil pollution native soil micro flora of hydrocarbon (VDM) with regional sorbents - zeolite deposits Khonguruu (Suntarsky District, Western Yakutia) and vermiculite deposits Inagli (Aldan district, South Yakutia). The aim of research was to evaluate the technology being developed for the treatment of permafrost soil from pollution by oil and PP in the subarctic conditions of Yakutia.

The experiment was conducted under natural conditions in the storage of fuels and lubricants in a. Hon, where the September 16, 2008 there was an emergency spill of diesel fuel "Arctic." The total area of contamination was 0.8 hectares. For frozen soils is characterized by the maximum temperature of -9 to -11 ° C. The average temperature in July from 6 to 14 ° C. The materials for the study were the sample of soil contaminated with diesel fuel, "Arctic" from the accident site and background samples taken at a distance from the spill with non-contaminated sites.

The experiment was conducted from June 16 to August 21, 2009, and consisted of two phases. In the first phase in June were selected contaminated soil samples in the experimental plots and then held tillage VDM immobilized on sorbents - zeolite and vermi-kulite. Immobilized on the surface of minerals, micro-organisms have made to the contaminated sites at the rate of 200 g per 1 m 2 - for the zeolite and 50 g per 1 m2 - for vermiculite. In the second phase in August at these sites were re-sampled to study the degradation of diesel fuel under the influence of oil destructors under natural conditions.

In addition, the two segments were selected from the control samples of contaminated sites, where soil treatment was not carried out with the introduction of drugs that allowed us to estimate the effectiveness of the impact of oil destructors by VDM on the processes of degradation of oil pollution in comparison with control samples.

Analytical studies included cold extraction of hydrocarbon compounds with chloroform from the soil samples, quantification of the extract yield (residual petroleum hydrocarbons), study group composition of extracts: The number of oils, resins and asphaltenes, as well as qualitative structural-group composition of the extracts by FT-IR spectroscopy and determination of the individual composition of saturated hydrocarbons by gas chromatography-mass spectrometry.

Instruction results of samples of permafrost soil contaminated with diesel fuel, and background samples are presented in Table 1. When evaluating oil pollution of soil samples using data on natural background. They were used as diagnostics of oil pollution in the soil samples, and in assessing the extent of degradation of oil pollution. It was assumed that the composition of the contaminated samples after the effective biological treatment should be changed in a direction approaching the composition of the background samples.

Table 1

Degradation of oil pollution in the frozen soils contaminated with diesel fuel "Arctic"

Point sampling	The conditions of experiment	№ sample	exit Xb*, g/kg	Degradation of oil – pollution june - august , %	The group component XB, %		
					UV	RESIN	ASFALTENI
BACKGROUND SAMPLE			0–1,516		6,23–13,79	58,22–69,34	21,94–32,70
Control samples	*without УОМ and sorbents JUNE AND AUGUST*	1 2	25,823 21,247	**17,80**	92,64 81,99	6,87 13,23	0,49 4,77
Х –У3-09	*UOM +vermikulit вермикулит* to make - June after objection - August	3 4	23,420 2,450	**89,53**	81,53 23,05	14,22 65,29	4,25 11,66
Х –У7-09	*UOM+ ZEOLIT* to make - June after objection - August	5 6	18,35 1,521	**91,71**	80,72 21,50	16,37 50,86	2,91 26,91
Х –У8-09	*UOM+ ZEOLIT* to make - June after objection - August	7 8	19,803 2,875	**85,48**	93,13 17,05	6,51 71,14	0,36 11,81

* – the oil content determined by the output of the chloroform extract - bitumoid chloroform (CB) in soil samples.

The background of the sample. As can be seen from Table 1, the content of natural hydrocarbons in background samples ranged from trace to high values of 1.516 g / kg. The peculiarities of the chemical structure of the background samples correspond to the CB soil samples collected from natural sites studied previously [9, 2009]. This is indicated by a close nature of the IR spectra of samples and component composition of the group with a predominance of asphalt-resin components of the hydrocarbon compounds (Table 1). However, according to gas chromatography-mass spectrometry in the saturated alkane hydrocarbons were found relatively low molecular weight alkanes C12-C15, which is not typical for alkane hydrocarbon organic matter in recent sediments. In this case, it indicates the presence of traces of diesel fuel contamination in the

background samples. Since the study area belongs to the oil and gas complex, such deviations from the natural background typical of man-made objects. The analyzed samples can be considered as background for the territory object location.

The first phase (June 2009). In all samples studied (1, 3, 5, 7) with contaminated sites, selected in June 2009 before making the soil VDM, the yield of CB ranged from 18.350 to 25.823 g / kg, which corresponds to a high level of contamination on the classification of [12, 2001]. CB samples are characterized by a close structural-group composition of hydrocarbon compounds with a predominance of compounds with oxygen-containing groups and bonds, as indicated by the low absorption in the 1100-1300, 1700-1740, 3300 cm-1. The IR spectra of extracts are typical of oil-contaminated areas. The group is also a component of the characteristic oil - gruntovyh samples contaminated with oil or NP. This is indicated by the predominance of hydrocarbons (oil) 80,72-93,13% over the amount of asphalt-resin components (Table 1). According to gas chromatography-mass spectrometry of the saturated hydrocarbon extracts of the relevant pollutant - diesel fuel, results in the predominance of relatively low molecular weight alkanes of high molecular (Table 2, sample 1, 2, 7). Hydrocarbon composition of oil-polluted samples is characterized by a high content of alkanes of normal structure, represented by the homological series of n-C12 n-C29. Among them, 97% is a relatively low-molecular structure of n-C12 n-C20, which accounts for the high value of the ratio of their relatively high-molecular homologues. The maximum of the distribution of n-alkanes account for n-C15, 16, the coefficient of lf / h close to unity. Among the saturated hydrocarbons n-Heptadecan and n-octadecane and pristane prevail over fitanom, the ratio (Pr + Ph) / (n-C17 + n-C18)) is less than unity, which is typical for oil polluted objects.

Table 2

Individual composition of saturated hydrocarbons

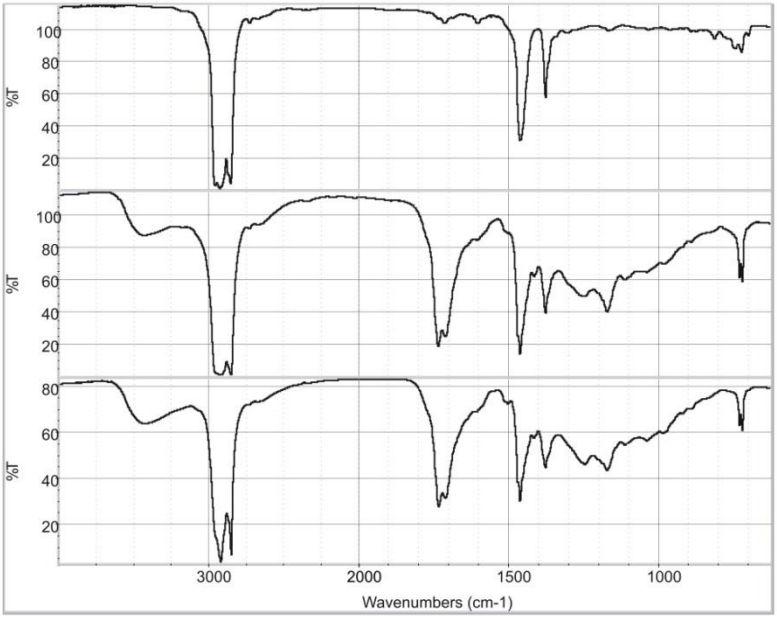
Параметры	№ SAMPLE				
	1	2	7	8	BACK-GROUNG
The conditions of experiment	Control samples (without UOM)		With UOM and zeolits UOM+zeolit		
	JUNE	AUGUST	BEFORE PUTTING – JUNE	AFTER PUT-TING– AU-GUST	
нК.-нC_{20}/нC_{21}-к.к.	34,13	26,57	33,65	1,19	0,59
ALKANOV	н-$C_{16,17}$	н-$C_{15,16}$	н-$C_{15,16}$	н-$C_{17,18,27,29}$	н-$C_{27,29,31}$
Coefficient IN THE WHOLE	0,99	0,95	1,08	1,74	2,04
iZOPRENOIDI/н-ALKANI	0,45	0,54	0,46	0,88	0,41
iC_{19}/нC_{17}	0,74	0,95	0,71	2,70	2,45
iC_{20}/нC_{18}	0,72	0,85	0,79	2,67	2,46
iC_{19}+iC_{20}/нC_{17}+нC_{18}	0,73	0,93	0,75	2,69	2,46

These results indicate that the composition of soil samples analyzed, selected almost a year after the accidental spill of diesel fuel, there are no visible signs of biodegradation of contamination. This confirms the very low ability to cleanse itself of permafrost soils in cold regions.

The second phase (August 2009). The results of the study of soil samples from experimental plots in two months after the introduction of VDM showed a decrease in residual petroleum hydrocarbons on the order, which corresponds to 85,5-91,7% degradation of oil pollution in

comparison with control samples (without making VDM), where the degradation was 17, 8% (Table 1).

The high degree of degradation of oil pollution of soil installed on the results of studying the composition and chemical structure of the selected extracts. As seen in the IR spectrum (Fig 1/?sample7 in the chemical structure of the original extract soil samples is dominated by hydrocarbon groups and communications connections with longmethyline chains (720 -1 cm)aromatic hydrocarbons (750,810 and 1600 cm - 1). Oxygen-containing groups and links are present in small numbers (1170 and 1710 cm-1). The group composition of these samples (Table 1) is dominated by hydrocarbons, and the share of resins and asphaltenes have less than 20%.

ХБ, г/кг	**ИК-спектры хлороформенных экстрактов проб почв (до опыта- проба 7, после опыта проба 8, фоновая проба)** Спектры записаны на ИК-Фурье спектрометре "Protégé -460", Nicolet, USA) Усл-я съемки: диапазон 4000 - 600 см-1 в кювете с окошками NaCl, толщина кюветы 33 микрона	Групповой состав ХБ,%		
		масла	смолы	асф. комп.
до опыта(7) *загрязнение* **19,803**		**93,1**	**6,5**	**0,4**
после опыта (8) ***деградация загрязнения*** 85,5% **2,875**		**17,0**	**71,1**	**11,8**
Фоновая проба **1,431**		**13,8**	**58,2**	**28,0**

Pic.1. The IR spectra of chloroform extracts of soils

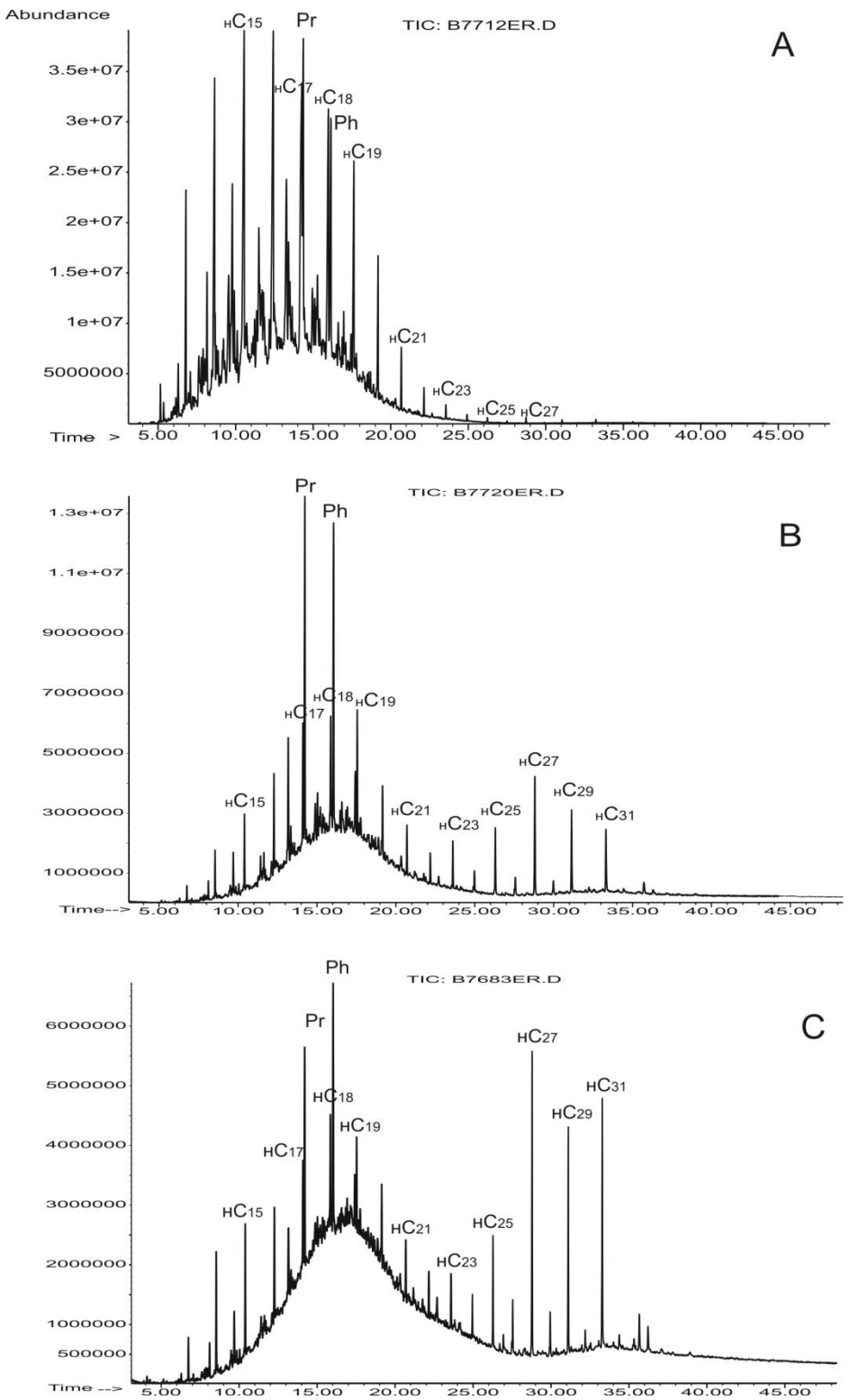
According to the group composition (Table 1) we can conclude that in the process of biodegradation by microorganisms in the first part of the hydrocarbon contamination is utilized, ie, oil. Their number in the extracts of soil samples with the introduction of VDM drops from 80,72-93,13 23,05-17,05% up. As expected, after the Bioremediation of the "residual contamination" has changed dramatically in the direction of increasing the amount of asphaltenes and especially the resinous components (Table 1). However, the results of FT-IR spectroscopy showed that the structural-group composition of extracts of samples with the introduction of VDM more in line with the natural soil OM rather than residual oil pollution (Fig. 1, sample 8). The findings suggest that deep biochemical transformation of oil pollution, which resulted in the studied samples from the IR spectra is practically difficult to detect the presence of petroleum hydrocarbons. The nature of the IR spectrum is typical of CB agents' bitumens recent sediments. The dominant absorption band in the spectrum in the 1700-1740 cm-1 indicates the highest amount of carbonyl groups, a strong absorption at 1170 cm-1 - the high number of ester bonds. Clearly marked absorption in the region 3200-3600 cm-1 due to the presence of hydroxyl groups belonging to the carboxylic

acids, ketones and esters. In the chemical structure substantially involved long methylene chains (doublet of 720 and 730, 1380 and 1460 cm-1) and low - compounds with aromatic rings (750 and 1600 cm-1). All these features are characteristic of immature OM of continental facies, and are set for different types of agents permafrost soils of Yakutia [9, 2009].

After making the VDM in the CB samples show significant changes in the composition of acyclic saturated hydrocarbons. Almost 10% decreased the proportion of n-alkanes, which are longer homological series of n-C13 to n-C33 (Table 2, sample 8, Fig. 2, B). In their structure compared with the initial oil pollution (Fig. 2, A) is significantly increased relative content of high molecular homologues of n-C21 n-C25, which is expressed in a decrease in the ratio is relatively low, relatively high-molecular n-alkanes.. Changed the distribution of n-alkanes: it has become bimodal, with peaks at n-C17, 18 and n-C27, 29. Was found between the redistribution of normal alkanes and isoprenoid structure: pristane and fit prevail over the next elution of n-and n-geptadekanom oktadekanom. The value of the ratio iC19 + + iC20/nS17 nS18 8 in the sample increased to 2.69 compared to 0.75 with a sample of 7 (Table 2).

The changes in the composition of the hydrocarbon fraction of the treated samples indicate that VDM is actively running processes of biodegradation. As a result of the chemical, structural-group and component composition of the group (and especially the distribution of individual saturated hydrocarbon extracts of soil samples) after treatment with oil destructors become almost identical with that of the background samples. These results indicate that, when introduced into soil contaminated with native VDM immobilized on local sorbents, in just one summer season, you can achieve a high degree of degradation of the permafrost soil-pollution diesel fuel.

High rates of degradation of 85 to 92% in soil samples with high levels of soil contamination in comparison with control samples without the introduction of VDM, in which the degradation rate was only 18 % indicate the effectiveness of oil destructions in a cold sub_ Arctic permafrost soils of the regions Thus, the results of research on the development of indigenous oil destructors by VDM, immobilized on a local sorbents showed that this trend is promising during bioremediation of contaminated soils in the permafrost of various oil and gas facilities.



Pic. 2. Mass chromatograms of hydrocarbon fractions chloroform extracts of samples: A - Sample 7 (to experience), B - Sample 8 (after the experience), C - Sample Background.

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