ISSN 2411-3336; e-ISSN 2541-9404

JOURNAL OF MINING INSTITUTE
Zapiski Gornogo instituta
Journal homepage: pmi.spmi.ru



Research article

Evaluation of the efficiency of sorbents for accidental oil spill response in the Arctic waters

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***How to cite this article*:** Vasileva Z.V., Vasekha M.V., Tyulyaev V.S. Evaluation of the efficiency of sorbents for accidental oil spill response in the Arctic waters. Journal of Mining Institute. 2023. Vol. 264, p. 856-864. EDN NVFPPM. DOI: 10.31897/PMI.2023.14

Abstract. The development and operation of new oil fields on the Arctic shelf increases the risks of oil spills, which require the use of oil spill response tools and materials that are efficient in harsh climatic conditions. The question of actual efficiency of sorbents for reducing the level of oil pollution in the conditions of the Arctic and subarctic waters is relevant. The work is aimed at a comparative study of the efficiency of sorbents of mineral and organic origin used by coastal enterprises in the Kola Bay as well as a sorbent based on chitin in model systems simulating real conditions in sea waters. The characteristics of sorption agents were determined applying ASTM F716-18 procedure. Sorption capacity was evaluated in respect of ARCO grade oil, diesel fuel and marine oil. The efficiency of sorbents was estimated in model systems "sea water – oil" at water temperature corresponding to the average annual surface temperature in the Barents Sea. Actual data on sorption capacity of commercial sorbents "Lessorb", "Novosorb", a sorbent based on vermiculite and chitin sorbent in relation to potential pollutants of waters were obtained. The dynamics of sea water saturation with oil products at –0.5 (±1) and 10 (±1) °C was determined. It is shown that at higher temperature the concentration of oil products in sea water column (in the presence of an oil film on the surface) is on average four times higher than at low temperature. Kinetic dependences were obtained that describe the content of oil products in water column and near-surface layer of sea water in the presence of the studied sorbents at 5 (±1) °C, corresponding to the average annual temperature in the Kola Bay of the Barents Sea. A method is proposed for evaluating the efficiency of sorbents from the values of regression coefficients characterizing the dependence of oil products content in sea water on the character of sorbent and duration of its action.

***Keywords*:** oil and oil products spills; sorbents; Barents Sea; chitin; evaluation of sorbents efficiency

Acknowledgment. The work was supported by the Scientific and Educational Centre "Russian Arctic: new materials, technologies and research methods".

Received: 11.11.2022 ***Accepted: 19.01.2023*** ***Online: 23.03.2023*** ***Published: 25.12.2023***

Introduction. The development and operation of oil fields on the Arctic shelf is associated with severe climatic conditions, low temperatures, polar night, ice drift, etc. [1]. These factors increase the risks of emergencies and, as a result, pollution of sea waters [2]. The greatest likelihood of accidental spills of Arctic oil is recorded when it is shipped to shuttle tankers (the Prirazlomnaya platform, the "Varandey" stationary offshore ice-resistant offloading berth, the "Arctic Gates" oil offshore berthless loading terminal) as well during its transhipment in the Kola Bay of the Barents Sea to storage tankers "Umba" and "Kola". The volume and number of shipments along the Northern Sea Route are actively increasing, which potentially rises the risk of emergencies in difficult navigation conditions [3-5].

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At the same time, measures on spill response in the Arctic and subarctic waters are more complicated than in seas with a temperate climate [6-9]. The publication [10] states that response measures in the Arctic and subarctic waters as well as in case of oil spill clean-up in water areas with a temperate climate are aimed at quick localization[1] and removal of spilled oil from sea water, minimizing damage to environmental and socio-economic resources and reducing the recovery time of affected resources by achieving an acceptable level of clean-up. However, unlike oil spill response in countries with temperate climate, response in fragile but harsh conditions of the Arctic requires special measures to prevent natural disasters [11-13].

Arctic climatic factors change the efficiency of oil spill response tools and technologies. The publication [14] indicates the need to determine the efficiency of each oil spill response method in different weather conditions taking into account sharp climate changes occurring in the Arctic and predicted for the future.

Many sorbents of mineral and organic origin are used for collecting oil and oil products (OP) from water surface [15, 16]. Sorption is one of the most efficient, environmentally, and economically acceptable methods of oil pollution control; it allows oil film removal, advanced water treatment from dissolved, emulsified OP [16, 17]. Mineral sorbents are produced from materials with a high specific surface area based on silicates [18] and aluminosilicates (zeolites) [19], vermiculite [20], bentonite [21], etc. [15, 22, 23]. Biodegradable sorbents of organic origin based on peat, coal, wood waste, cellulose, grain products, etc. are considered promising and not causing additional harm in oil and OP spill response [24-26]. At the same time, the question of efficiency of oil and OP spill response using these sorbents in waters of the Arctic seas remains open [27]. According to V.E.Kogan [28], the issue of removing oil pollution from water surfaces in the Arctic was not yet resolved. Article [4] indicates the lack of efficient and reliable methods and technologies in terms of the result obtained in oil spill response in waters of the High North. Thus, the issue of studying the efficiency of available sorption materials to reduce the level of oil pollution in the conditions of the Arctic and subarctic waters is topical.

In publications, numerous studies are described on the use of chitosan as an OP sorbent in case of oil spills on soil, water surface, and for wastewater treatment [29-31]. However, chitosan has a high cost, a complex production process, and is in demand in more high-tech branches of medicine and food industry [32, 33]. Due to economic reasons, it is rarely used on an industrial scale as a sorption material for oil spill response. Unlike chitosan, chitin can be obtained using a simpler technological scheme and at lower economic expenditures [34]. It is cheap and can have similar properties in terms of efficiency of the OP sorption in the conditions of emergency oil spills.

The purpose of the work is to study the sorption activity of mineral and organic sorbents used in emergency oil and OP spill response in the Barents Sea waters. Sorption properties of chitin obtained from crustacean processing waste were studied in the conditions of the OP spill in the Arctic waters.

Methods. Absorbent materials of organic origin – "Lessorb" and chitin as well as of mineral origin – "Novosorb" and a sorbent based on heat-treated vermiculite were chosen as the objects of study. Sorption materials "Lessorb", "Novosorb" and the sorbent based on vermiculite selected

[1] Regulation of the Government of the RF of December 31, 2020 N 2451 "On approval of the Rules for organizing measures to prevent and clean-up oil and OP spills in the territory of the Russian Federation, with the exception of inland sea waters of the Russian Federation and territorial sea of the Russian Federation as well as on recognizing as invalid some acts of the Government of the Russian Federation". URL: https://base.garant.ru/400170332 (accessed 20.09.2022).

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EDN NVFPPM
DOI: 10.31897/PMI.2023.14

as the objects of study are used by coastal services and enterprises engaged in maritime activities in waters of the Kola Bay of the Barents Sea in emergency oil and OP spill response.

"Lessorb" sorbent (TU 9010-002-35615057-99) is commercially produced from sphagnum moss and peat; "Novosorb" (TU 6418-001-57901390-02) is a mineral-based sorbent. Commercially produced sorbents based on expanded vermiculite are a heat-treated, modified mineral from the group of hydromicas. Chitin used for the study was obtained from processing waste of commercial crustaceans – crab Paralithodes Camtschaticus (Department of Technosphere Safety, Murmansk State Technical University). Chitin was obtained by successive stages of demineralization and deproteinization of the crushed shells of cephalothorax (carapace). The obtained chitin is white-cream-coloured flakes with a diameter of 2-5 mm.

The characteristics of sorption materials were determined according to ASTM F716-18 procedure[2]. Comparison of sorption capacity of commercial sorbents with the values declared by the manufacturers was carried out both in relation to ARCO oil (dynamic viscosity, η = 29 mPa·s) and other OP which can become pollutants of the water area: low-viscosity diesel marine fuel (η = 2.0 mPa·s), marine hydraulic oil (η = 0.5 mPa·s). The choice of OP as pollutants is based on their most probable spills during bunkering of ships and transhipment of Arctic oil in waters of the Kola Bay.

The efficiency of using sorbents to reduce the OP concentration in sea water was studied in model systems "sea water – oil" in static conditions, where ARCO oil was used as a model pollutant. Oil was introduced in a volume of 10 ml into containers with sea water taken in the Kola Bay. The area of water surface pollution in all model systems was 0.12 m^2. Water temperature in a series of experiments was maintained at 5 (±1) °C simulating the average annual surface temperature in the Barents Sea at entrance to the Kola Bay. When modelling the emergency oil spill response, the sorption material was evenly placed on samples of polluted water at a mass ratio of sorbent to OP equal to 1:1. Sampling of sea water after treatment with a sorbent was carried out after 1; 1.5; 2.5; 3; 4; 4.5 h. Samples were taken from different water layers – near-surface layer at 2-3 cm from the surface with an oil film and from the water column at a depth of 20 cm. Time frame for conducting a model experiment is set on the basis of time of oil and OP spill localization determined by the Regulations of the Government of the RF[3].

Concentration of the OP in the water column was measured at ambient temperature –5 °C and sea water temperature –0.5 (±1) °C simulating the conditions of the OP oil spill in winter season as well as at ambient temperature 16 °C and sea water temperature 10 (±1) °C simulating the conditions of the OP spill in summer. The experiment continued for four days. Mass ratio of the OP to water is 1:1,000.

Content of the OP in sea water was determined according to the procedure[4] using a standard fluorimetric method on Fluorat-02 liquid analyser.

[2] ASTM F716-18, Standard Test Methods for Sorbent Performance of Absorbents for Use on Chemical and Light Hydrocarbon Spills, ASTM International, West Conshohocken, PA, 2018. URL: https://www.astm.org/Standards/F716 (accessed 30.10.2022).

[3] Regulation of the Government of the RF of December 31, 2020 N 2451 "On approval of the Rules for organizing measures to prevent and clean-up oil and OP spills in the territory of the Russian Federation, with the exception of inland sea waters of the Russian Federation and territorial sea of the Russian Federation as well as on recognizing as invalid some acts of the Government of the Russian Federation". URL: https://base.garant.ru/400170332 (accessed 20.09.2022).

[4] Quantitative chemical analysis of waters. Method for measuring mass concentration of oil products in samples of natural, drinking, waste water by the fluorimetric method on Fluorat-02 liquid analyzer. PND F 14.1:2:4.128-98. URL: https://docs.cntd.ru/document/1200079424 (accessed 20.09.2022).

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The efficiency of reducing pollution of model solutions from the OP was determined from the formula

$$G = \frac{C_0 - C_f}{C_0} \cdot 100\ \%,$$

where C_0, C_f are initial and final OP concentrations in water sample, mg/l.

Discussion of results. The characteristics of sorption materials were determined with respect to different OP under standard conditions and compared with the values declared by sorbent manufacturers and published data (see Table).

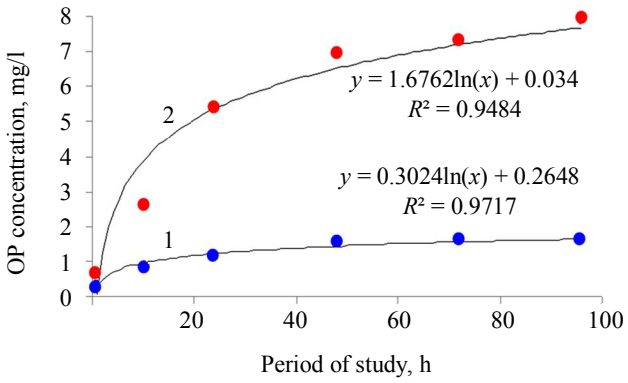


Fig.1. Change in OP concentration in water column during oil spill at –0.5 (±1) (1) and 10 (±1) °C (2)

Sorption capacity, g/g

OP sample	"Lessorb"		Chitin		"Novosorb"		Vermiculite	
	Results of study	Declared by manufacturer[5]	Results of study	Published data [35, 36]	Results of study	Declared by manufacture[6]	Results of study	Declared by manufacture[7]
Oil	6.25	4.0-15.0	5.59	3.9-4.5	4.72	6	4.62	8-12
Diesel fuel	5.63	3.3-9.5	5.31	–	4.53	–	4.28	–
Marine oil	6.35	–	5.61	–	5.28	–	5.48	–

Analysis of experimental data showed that sorption capacity of each material with respect to different OP varies within 10-20 %. All the studied materials showed the highest sorption capacity for marine oil, and the lowest one for diesel fuel. The obtained results on sorption capacity differ from the values declared by the manufacturers being both higher and lower. Difference between the experimental and manufacturer-declared values can be explained by various reasons: breakage of package tightness during transportation and long-term storage; difference in the composition and quality of sorbed OP, methods, and conditions of determination; viscosity and chemical composition of OP.

Under actual spill conditions, the results of OP sorption can be significantly affected by temperature. Under conditions of the Kola Bay and the Barents Sea, the ranges of winter and summer temperatures of sea water differ significantly from the temperatures at which sorption capacity of sorbents is usually determined. Thus, it is necessary to take into account temperature as one of the key factors affecting the sorption properties. Temperature affects surface phenomena, rheological characteristics of the OP, kinetics of emulsification, evaporation, sedimentation, and other processes occurring during the action of sorbents. The response function showing the effect of temperature on the OP spread in the water column is its concentration. To assess the effect of temperature on the spread of the OP in water, the dynamics of the OP concentration value (Fig.1) was studied under conditions of simulating an oil spill in winter and summer seasons in waters of the Kola Bay at ambient temperatures –5 and 16 °C and sea water temperature –0.5 (±1) and 10 (±1) °C, respectively.

[5] "Lessorb" sorbents. URL: https://lessorb.ru/products/sorbenty/vidy-sorbentov/sypuchie-sorbenty/sorbenty-lessorb/ (accessed 30.10.2022).

[6] "Novosorb" sorbent. URL: http://specudm.ru/production/catalog_4/sorbenty/sorbent_novosorb.html (accessed 30.10.2022).

[7] Detailed data on expanded vermiculite. URL: https://agroru.com/news/podrobnye-svedeniya-o-vspuchennom-vermikulite-114768.htm (accessed 21.09.2022).

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Journal of Mining Institute. 2023. Vol. 264 P. 856-864
EDN NVFPPM
DOI: 10.31897/PMI.2023.14
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It follows from the results that the OP content in water column in the presence of an oil film on the surface increases throughout the observation period. At water temperature in winter season –0.5 (±1) °C, the increase in concentration is not so rapid; the concentration curve reaches saturation in 3-4 days at the OP value in water 1.7-1.8 mg/l. Water in the summer season is more saturated with the OP, and during the first day the OP concentration reaches 5.2 mg/l. Further contact of oil film with water leads to an increase in the concentration in water column to 7.9 mg/l on the fourth day of observation. Such an effect of temperature on the distribution of pollutant should be taken into account in oil spill response in different seasons of the year.

Under actual spill conditions, the results of OP sorption by sorption materials can also differ significantly from the established characteristics, since in marine environment, sorption proceeds in a multiphase system that includes such components as water, OP, air (oil vapour), and surface of the sorbent. This entails a number of heterogeneous equilibria between the phase interfaces.

Comparative evaluation of the efficiency of using different sorbents in the model systems "sea water – oil – sorbent" and "sea water – oil" made it possible to obtain data on the kinetics of sorption processes at temperature conditions 5 (±1) °C simulating average annual water temperature in the Kola Bay and initial level of the OP pollution of the surface layer of the offshore zone 1.24 mg/l (Fig.2). Modelling in the "sea water – oil" system was performed for taking into account the contribution of natural OP degradation in marine environment under the influence of different natural factors [8].

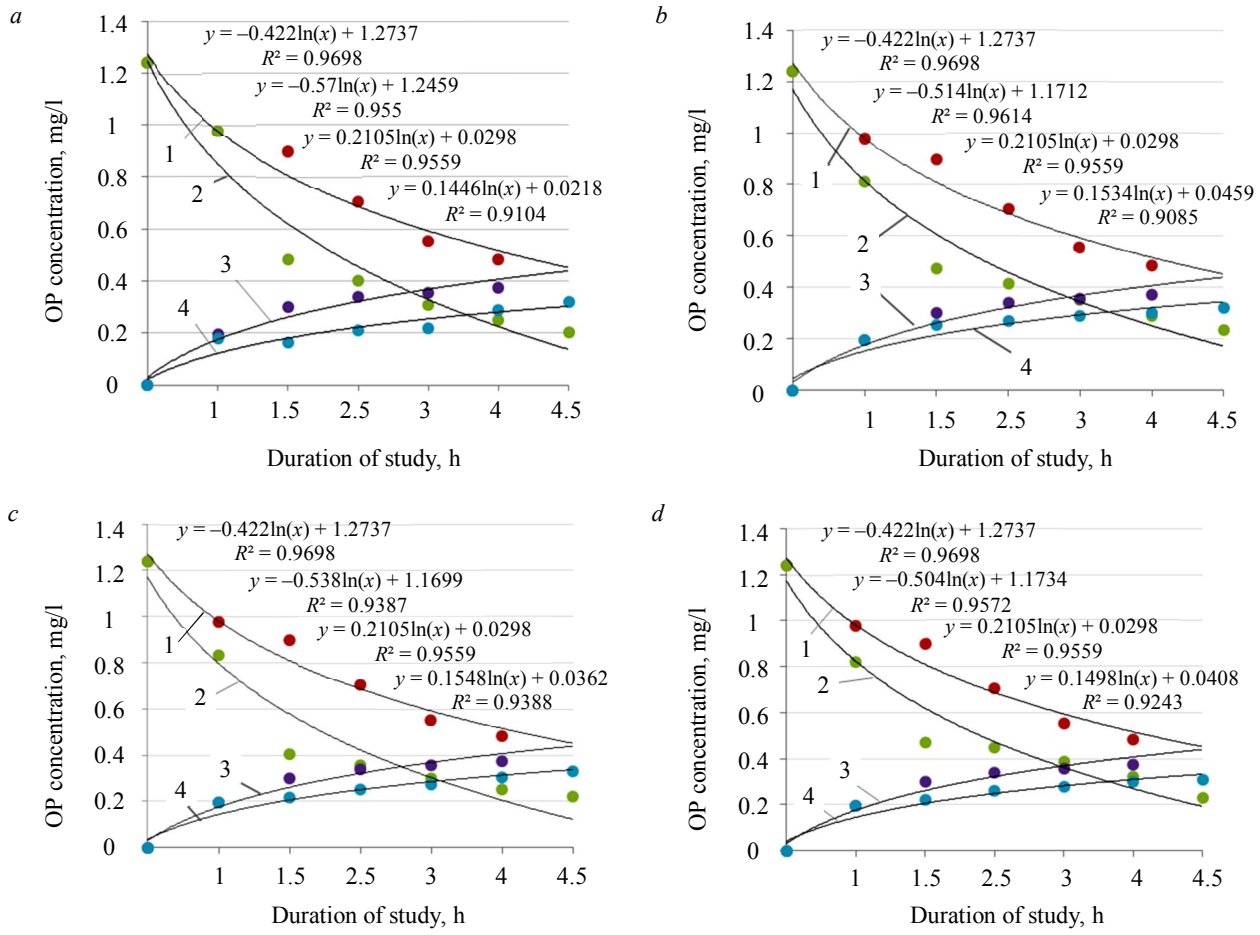


Fig.2. Change in OP concentration in model systems "sea water – oil" with addition of sorbent and without it:
a – chitin; *b* – "Novosorb"; *c* – "Lessorb"; *d* – vermiculite

1 – near-surface layer without sorbent; 2 – near-surface layer with sorbent; 3 – in water column without sorbent; 4 – in water column with sorbent

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As a result of experimental data processing, regression equations and general equations $y = b\ln(x) + a$ were obtained characterizing the dependence of the OP content in the water column and near-surface layer of sea water under the established experimental conditions (Fig.2).

The equation for describing the dependence of a change in the OP concentration in the near-surface layer during an oil spill in the absence of a sorbent (Fig.2, curve 1) looks as follows:

$$y = -0.422\ln(x) + 1.2737.$$

The approximation certainty value for the established dependence was $R^2 = 0.9698$.

The equation for describing the dependence of a change in the OP concentration in water column during oil spill in the absence of a sorbent (Fig.2, curve 3):

$$y = 0.2105\ln(x) + 0.0298.$$

The approximation certainty value for the established dependence was $R^2 = 0.9559$.

Logarithmic dependences of a change in the OP concentration in the near-surface layer of sea water were determined in case of using sorbents (Fig.2, curve 2): chitin, “Novosorb”, “Lessorb”, sorbent based on vermiculite:

$$y = -0.57\ln(x) + 1.2459;$$

$$y = -0.514\ln(x) + 1.1712;$$

$$y = -0.538\ln(x) + 1.1699;$$

$$y = -0.504\ln(x) + 1.1734.$$

Approximation confidence factor for the established R^2 dependencies was 0.955; 0.9614; 0.9387; 0.9572 respectively.

Similarly, the dependences of the OP content in water column on the time of spill when sea water was polluted with oil with the use of (Fig.2, curve 4): chitin, “Novosorb”, “Lessorb”, sorbent based on vermiculite are determined:

$$y = 0.1446\ln(x) + 0.0218;$$

$$y = 0.1534\ln(x) + 0.0459;$$

$$y = 0.1548\ln(x) + 0.0362;$$

$$y = 0.1498\ln(x) + 0.0408.$$

Approximation confidence factor for the determined dependencies R^2 was 0.9104; 0.9085; 0.9388; 0.9243, respectively.

Curves 1 and 2 (Fig.2) show a change in the OP concentration in the near-surface layer without the use of sorbents and in their presence. During the entire observation period, the OP concentrations in the near-surface layer decrease both in the first, and in the second case. The decrease in the OP concentrations in the water of model systems occurs due to evaporation, dissolution, emulsification, and other physical and chemical processes [8] as well as due to the action of sorbents in case of their presence in the system. At the same time, the intensity of a decrease in the OP concentration in the presence of sorbents is higher. We estimate the rate of a decrease in the OP concentration in the near-surface layer from the absolute value of the regression equation coefficient. The regression coefficient b reflects the rate of change in the OP concentration in marine environment – the higher its absolute value, the more intensely the content decreases. Thus, after 4 h of experiment, the OP content

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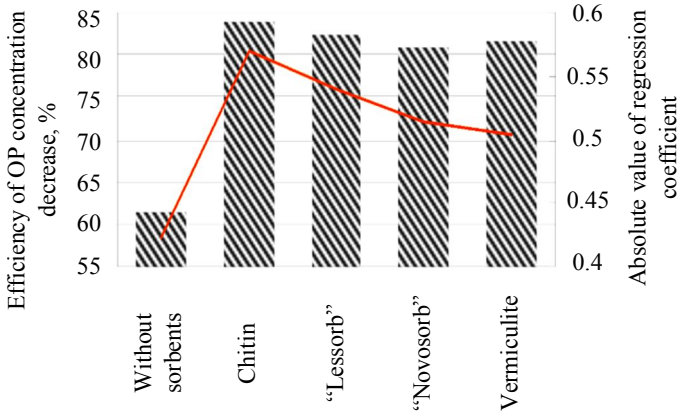


Fig.3. Efficiency of OP content decrease and absolute value of regression coefficient

in the near-surface layer of water without sorbent was 0.48 mg/l (b = 0.422), in the presence of chitin sorbent 0.25 mg/l (b = 0.57); when using "Novosorb" 0.29 mg/l (b = 0.514), when using "Lessorb" 0.25 mg/l (b = 0.538), in case of the action of vermiculite 0.32 mg/l (b = 0.504). From the presented values of the regression coefficients, it follows that the highest rate of oil absorption is recorded in the "sea water – oil – chitin" system. The course of curves 2 (Fig.2) indicates a similar kinetics of the processes for the studied cases of changes in the OP concentration in the near-surface layer of sea water.

The efficiency of reducing the OP concentration with the use of sorbents G, estimated by referring the difference between the initial and final OP concentrations to the initial concentration of pollutants, does not fully correlate with the assessment of the sorption process using regression coefficients. Figure 3 shows a diagram of comparison of the efficiency indicators for reducing the OP concentrations over the studied time interval of 4.5 h and the corresponding regression coefficients of the established dependencies. The highest value of the efficiency of reducing the OP 83.7 % manifested in the presence of chitin, corresponds to the highest regression coefficient of 0.57. The second highest efficiency value of 82.3 %, determined for "Lessorb" sorbent, corresponds to a regression coefficient of 0.538. But the lowest value of the efficiency of reducing the OP concentration 80.9 % recorded in the presence of "Novosorb" corresponds to a slightly larger coefficient than for the sorbent based on vermiculite – 0.514 versus 0.504. At the same time, for the specified time interval, the sorbent based on vermiculite showed a high efficiency of reducing the OP – 81.5 %. This can be accounted for by different physical meanings of the compared values. Thus, the efficiency G depends on the values of initial and final OP concentrations and does not take into account the behaviour of the system in other time intervals, in which the efficiency can assume other values. The regression curve takes into account all the experimental data obtained over the entire observation period, so it is advisable to evaluate the efficiency of sorbents by both criteria. In case of chitin, the maximum value of efficiency G corresponds to the maximum value of regression coefficient b.

Curves 3 and 4 (see Fig.2) show a change in the OP concentration in water column without the use of sorbents and in their presence. During 4 hours of observations in the sea water column (Fig.2, curve 3), the concentration gradually increases due to the processes of dissolution, emulsification of oil and distribution of oil components in the "sea water – oil" model system. An increase in the OP concentration in water column in the absence of sorbent occurs with the regression coefficient 0.2105, while in the presence of sorbents, the regression coefficients have the following values: 0.1446 for chitin; 0.1534 for "Novosorb"; 0.1548 for "Lessorb"; 0.1498 for vermiculite.

Difference in the values of regression coefficients for curves 1 and 2, corresponding to the OP content in the near-surface layer, is significantly greater than the difference between the regression coefficients for curves 3 and 4. This indicates that the effect of sorbent on the surface on the OP concentration in water column is less noticeable than in the near-surface layer.

After 4.5 h of observations, curves 1 and 3 converge at a concentration value of 0.42 mg/l. This means that in the given time interval in the studied model systems and conditions, the OP concentrations became equal in the entire volume.

It is not possible to evaluate the effect of sorbents on water column by calculating G, since the OP content in this case increases with time even in the presence of sorbents. In this case, the effect of sorbent can be determined only by the value of the regression coefficient.

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Data on the efficiency of sorbents in relation to oil and the OP obtained under standard conditions (see Table), differ from the behaviour of sorbents in real conditions of oil spill response due to a number of natural and anthropogenic factors that are not taken into account by researchers and manufacturers of sorption materials. Therefore, the most adequate results can be obtained in real conditions or models simulating them. Under the conditions of model systems "sea water – oil – sorbent" at 5 °C for the studied time interval, the efficiency of sorbents was, %: 83.7 for chitin; 82.3 for "Lessorb"; 80.9 for "Novosorb"; 81.5 for vermiculite.

Conclusion. Experimental data on the sorption capacity of commercial sorbents "Lessorb", "Novosorb", chitin, sorbent based on vermiculite with respect to potential pollutants of the Barents Sea waters – ARCO oil, marine diesel fuel and marine hydraulic oil were obtained. The obtained results on the sorption capacity were compared with data declared by the manufacturers.

Dynamics of sea water saturation with the OP at –0.5 (±1) and 10 (±1) °C was established. It was shown that at a higher temperature, the OP concentration in the sea water column in the presence of an oil film on the surface is, on average, four times higher than at a low temperature.

Kinetic dependences were obtained that describe the OP content in water column and near-surface layer of sea water in the presence of the studied sorbents in the conditions of a model experiment at 5 (±1) °C simulating the average annual temperature in the Kola Bay of the Barents Sea. The kinetics of the processes under study is described by logarithmic equations with values of the approximation certainty factor R^2 greater than 0.9. The character and course of the curves for the near-surface layer and the water column differ. A methodology for evaluating the efficiency of sorbents by comparing the absolute value of the regression coefficient b, which reflects the rate of change in the OP concentration in the marine environment, was proposed. The higher the absolute value of b, the lower the OP content under the action of sorbent. In the near-surface layer, the regression coefficient has a negative value, which reflects a decrease in the OP concentrations. In water column, the coefficient has a positive value and reflects an increase in the OP content throughout the time interval of the spill.

The efficiency of sorbents "Novosorb", "Lessorb", vermiculite and chitin obtained from processing waste of commercial crustaceans was estimated based on results of changes in the OP concentrations in the conditions of simulating an oil spill at an average annual temperature in waters of the Kola Bay. It was shown that the efficiency of using chitin as a sorbent is comparable to that of industrial analogues.

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The authors declare no conflict of interests.

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