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Modelling the effective formulation of sorbents for remediation of oil-contaminated Arctic coastlines

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Abstract

The paper presents a scientific approach to developing and evaluating the efficiency of composite sorbents for remediation of oil-contaminated soils along the Arctic coastlines. The authors use a chitin-chitosan complex derived from crab processing waste as the sorption base. Activated sludge – a by-product of wastewater treatment facilities in the Murmansk Region – serves as a source of oil-oxidizing microorganisms adapted to northern conditions. The study reports results of initial material characterization, including comparative spectral analysis of the chitin-chitosan complex, analytical control of heavy metal content in the activated sludge, and its microbiological assessment. The composite sorbent formulation is developed via mathematical modelling using orthogonal central composite design. The variable factors are the component contents in the sorbent; the response function is the residual petroleum content in soil after remediation. Researchers run the experiments at 10±2 °C, matching the average daily temperature along the Kola Peninsula coastlines during summer. The resulting mathematical models describe how sorbent efficiency, assessed by residual petroleum content in soil, depends on component ratios; these models guide the selection of the optimal formulation. 3D models of oil pollution degradation are built to visualize and compare the performance of sorbents with different formulations. The correctness of the optimal sorbent formulation, selected based on the mathematical models, is additionally verified through phytotoxicity test. The paper also demonstrates the high remediation efficiency of the developed sorbent for oil-contaminated soils compared to the commercial analogue. The study finds that the residual petroleum content stays 2-4 times lower with the composite sorbent throughout the entire research period. Based on the research conducted, the authors propose engineering principles for producing the composite sorbent from organic wastes and for its application in effective treatment of oil-contaminated coastlines under Arctic conditions.

Keywords

oil and petroleum product spills; sorbents; chitin-chitosan complex; activated sludge; microorganisms; Arctic coastline; soils; Arctic

Funding

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Introduction

According to the Northern Sea Route Development Plan, the main growth in cargo traffic along the Northern Sea Route in the coming years will relate to sea transportation of oil under the Rosneft's[1] Vostok Oil project. Experts expect a multiple increase in oil transportation volumes in both western

[1] Order of the Government of the Russian Federation dated 01.08.2022 N 2115-r "On Approval of the Development Plan for the Northern Sea Route for the Period until 2035". URL: http://government.ru/docs/46171/ (accessed 12.02.2026).

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and eastern directions [1]. The first stage of the project is scheduled for commissioning in 2026 with full design capacity of 115 million tons per year to be reached by 2033 [2]. The logistics arrangement for sea oil transportation requires construction of offshore transshipment complexes in the Kola Bay area (Murmansk Region) to transfer oil from ice-class shuttle tankers to conventional tankers for further delivery to consumers. Currently, the bulk of Arctic oil transportation relates to operation of the Prirazlomnoye, Novy Port, and Varandey fields; transshipment occurs in the Kola Bay of the Barents Sea. Against the background of steady growth in shipping along the Northern Sea Route, the probability of emergencies objectively rises due to the region's challenging navigation conditions [3].

Despite the set of preventive measures underway, the risk of oil spills cannot be fully eliminated [4]. Among possible scenarios, a spill in the coastal zone poses the greatest environmental hazard: it involves large-scale damage to vulnerable ecosystems and complicates response and cleanup efforts [5].

Degradation of petroleum products (PP) reaching the Arctic coast proceeds significantly slower due to insufficient natural light, low temperatures, and low self-cleaning capacity of soils and water bodies [6]. This drives the need to develop feasible and environmentally sustainable technologies and materials capable of ensuring efficient remediation of oil-contaminated soils along the Arctic coastlines under harsh Arctic conditions [7].

For remediation of oil-contaminated coastal soils, the sorption method ranks among the priority engineering solutions. The specificity of coastlines – combining high environmental value of the area with elevated vulnerability to oil pollution – dictates specific requirements for the cleanup methods: they must ensure not only efficient removal of petroleum products but also minimize the risk of secondary contamination. The use of sorbents allows localization and immobilization of oil pollution, preventing its further migration into adjacent aquatic and terrestrial ecosystems [8].

Based on the nature of raw materials, researchers distinguish three main groups: mineral, organic, and synthetic [9]. Mineral sorbents derive from silicates, aluminosilicates, zeolites, vermiculite, and bentonite and feature a high specific surface area and considerable sorption capacity, ensuring rapid absorption of liquid hydrocarbons [10]. Another advantage of such materials is their relatively low cost. Disadvantages include the impossibility of incineration and biodegradation; they are often hazardous to inhale and require personal protective equipment. Their application in northern ecosystems also carries the risk of residual accumulation of inert particles in the soil [11].

Synthetic sorbents, produced from compounds obtained via chemical synthesis, possess high sorption capacity, pronounced hydrophobicity, and buoyancy; they allow repeated use after mechanical pressing [12]. Such sorbents are expensive and require subsequent collection, which is highly labour-intensive and demands specialized equipment. Synthetic sorbents do not undergo biological decomposition; therefore, spent materials require careful disposal or energy-intensive regeneration (incineration) [13]. When applied excessively, they can become a source of secondary contamination [14].

In this regard, biodegradable organic sorbents gain particular relevance. They not only efficiently accumulate PP but also gradually integrate into natural biogeochemical cycles without disrupting soil structure and fertility. Manufacturers produce them from renewable bio-raw materials – peat, sapropel, plant residues (sawdust, straw, coconut coir, husks), and various organic waste (wood processing products, grain and oilseed agricultural waste, etc.) [15, 16]. Key advantages of such sorbents include biodegradability, which eliminates secondary contamination, and low production cost through the use of industrial waste [17]. They are environmentally safe for soil and aquatic ecosystems and undergo

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disposal via composting or biodegradation. Owing to this, organic sorbents see wide application in remediation of soils and water bodies, especially in areas with high environmental sensitivity [18].

A number of publications focus on the effect of microorganisms on accelerating oil spill degradation. Study [19] describes the most effective methods and technologies for applying microorganisms to remediate oil-contaminated soils under various climatic conditions. In [20], researchers assess the hydrocarbon-oxidizing potential of psychrophilic and psychrotolerant microorganisms isolated from soils of the Yamalo-Nenets Autonomous Okrug at temperatures of 5-15 °C and develop microbial biopreparations for soil remediation. The study shows that a consortium of indigenous microorganisms reduces petroleum product concentration by 78.4 % per season when sorbents and biostimulants are used together. Authors [21] demonstrate the positive role of biostimulants – such as humates, siderates, and mineral fertilizers – for oil biodegradation in contaminated soils.

A promising direction for remediation of oil-contaminated soils and coastal areas is the combined application of organic biodegradable sorbents and oil-oxidizing microorganisms. In this case, organic sorbents serve as a carrier: they rapidly localize oil pollution, reduce environmental toxicity, and create a structural porous matrix [22]. Oil-oxidizing microorganisms catalyse biodegradation of hydrocarbons into nontoxic compounds. The synergistic effect consists in combining the action of the sorbent, which promptly immobilizes oil and reduces its migration into adjacent ecosystems, with the action of hydrocarbon-oxidizing microorganisms, which ensure long-term cleanup through biodegradation of the pollutant. This approach combines the advantages of physical sorption and biological remediation, enabling effective restoration of oil-contaminated areas, including environmentally vulnerable coastal zones [23].

Among organic sorbents, chitosan – a natural biopolymer possessing biocompatibility, biodegradability, nontoxicity, and high adsorption properties – holds particular interest [24, 25]. Its sorption activity includes an equal ability to bind both hydrophilic and hydrophobic compounds [26]. Several authors observed the high efficiency of chitosan for sorption of petroleum products from soils [27, 28].

Chitosan is an N-deacetylated derivative of chitin. In Russia, processing waste of the red king crab (*Paralithodes camtschaticus*) serves as a large-scale source of chitin-containing raw material; the volume for the Barents Sea basin alone amounts to approximately 10 thousand tons annually. Industrial production of chitin and chitosan in Russia was organized in the 1970s-1980s at enterprises in Murmansk and Vladivostok [29]. However, the widespread use of chitosan as a sorbent for oil and petroleum products is limited by its excessive cost [30]. Its production is rather labour-intensive, resource- and energy-consuming, due to the need for multistage operations to remove the protein and mineral components (deproteinization and demineralization) using expensive reagents [31]. Depending on the degree of deacetylation, the commercial cost of chitosan ranges from 80 to 200 thousand roubles per kg, making its use as an oil sorbent economically impractical [24]. Chitin, the precursor of chitosan, is considerably cheaper but is inferior in sorption capacity by 20-25 % compared to chitosan due to high crystallinity and a dense network of hydrogen bonds [32].

As an economically feasible alternative to chitosan, it is relevant to consider an intermediate product of crab raw material processing obtained via a procedure with fewer deproteinization and demineralization stages. Such a sorbent for remediation of oil-contaminated soils has not been previously considered; publications devoted to this issue in domestic and foreign literature were not found. In this study, the intermediate product of crab raw material processing is termed “chitin-chitosan complex” (Chit). It is assumed that it can potentially possess the key advantages of chitosan (sorption activity, biodegradability, biocompatibility) at a substantially lower production cost, ensuring effective remediation of oil-contaminated coastlines.

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Activated sludge (AS) – a large-tonnage waste product of biological wastewater treatment – represents a promising source of oil-oxidizing microorganisms. It contains associations of microorganisms adapted to the degradation of organic compounds, including hydrocarbons [33, 34]. Activated sludge contains consortia of bacteria with pronounced oil-oxidizing activity, fungi and actinomycetes that complement the spectrum of biodegrading enzymes, and extracellular matrix components that promote cell adhesion to the sorbent [35, 36]. Earlier literature already noted the potential for using activated sludge in soil bioremediation [37-39]. A procedure using a mixture of a polymer complex [chitosan – whey protein] – activated sludge was proposed, in particular, in [40]. The use of activated sludge offers a number of advantages – low cost as a production waste, high biological activity due to natural adaptation to pollutants and ambient temperature, and structural compatibility with porous sorbents (including chitin-chitosan complexes).

The development of sorption materials that combine efficient sorption of oil pollution with its microbiological oxidation represents an urgent scientific and practical task of modern environmental engineering. The aim of this work is to develop a composite sorbent based on a chitin-chitosan complex and activated sludge, to optimize its formulation, and to experimentally evaluate its efficiency for remediation of oil-contaminated soils along the Arctic coastlines.

Methods

Soil samples for the series of experiments were collected from the coastal zone of Kola Bay in areas characterized by minimal anthropogenic impact. Sampling was carried out from the topsoil layer (depth 0-10 cm). At each sampling point, a composite sample was formed by mixing at least five individual subsamples taken by the envelope method over a 10×10 m area. Then, under laboratory conditions, mechanical inclusions (stones, roots, and undecomposed plant residues) were removed; samples were dried at room temperature in dark conditions to an airdry state, sieved, and used for a series of experiments modelling artificial oil contamination. The pollutant was ARCO crude oil with a density of 910 kg/m^3, kinematic viscosity of 19.3 mm^2/s (at 20 °C), pour point of –30 °C, and mass content of sulphur and paraffins of 2.3 and 1.8 %, respectively.

The basis of the composite sorbent was a chitin-chitosan complex obtained from the carapace of the red king crab via a shortened chitin conversion procedure with a reduced number of processing stages compared to pure chitosan production, and a reduced mass ratio of raw material to reagents at the deproteinization and demineralization stages. The resulting sorbent consists of white flakes with a particle size of 2-3 mm (Fig.1). The total production cost of the chitin-chitosan complex obtained from red king crab processing waste via the shortened procedure amounts to 0.95 thousand roubles/kg.



Fig.1. Chitin-chitosan complex flakes

The formulation of the obtained chitin-chitosan complex was evaluated using IR spectral analysis by comparison with a reference chitosan sample (Sigma-Aldrich, degree of deacetylation not less than 75 %). Measurements were performed using an FSM2202 Fourier-transform infrared spectrometer (Infraspek) with an ATR-Diamond attachment in the wavenumber range of 450-4000 cm^{-1}, in transmission mode with 20 scans.

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Excess activated sludge served as the bioactive component of the composite sorbent. It was collected from the biological treatment facilities of the Murmashi wastewater treatment plant (Murmansk Region), operated by GOUP Murmanskvodokanal. Activated sludge was sampled at a wastewater temperature of 7-9 °C. Before use, the collected samples were preliminarily decanted to achieve a sludge concentration of 15 g_{dry}/l (dry matter) and a sludge index of 110 cm^3/g.

Heavy metal content in the activated sludge was assessed in accordance with PND F 16.1:2.3:3.11 and PND F 16.1:2.23, using an EXPEC PlaAr AES V 2023 inductively coupled plasma atomic emission spectrometer and an RA-915M mercury analyser.

The number, biomass, and morphology of bacterial cells in the activated sludge were determined by epifluorescence microscopy using nuclear filters with a pore diameter of 0.22 μm and the DAPI fluorochrome at ×1000 magnification [41]. The number of cultivable saprotrophic and hydrocarbon-oxidizing bacteria (HOB) was determined by the limiting dilution method using liquid nutrient media [42, 43]. When determining HOB, sterilized diesel fuel was added to the medium as the sole source of carbon and energy [44]. The presence of pathogenic microorganisms, including salmonella, was assessed at the Testing Laboratory Centre of the Federal Budgetary Healthcare Institution "Centre for Hygiene and Epidemiology in the Murmansk Region" in accordance with MUK 4.2.3695-21 "Control Methods. Biological and Microbiological Factors. Methods for Microbiological Control of Soil".

The effective formulation of composite sorbents for remediation of oil-contaminated soil was simulated using mathematical modelling with orthogonal central composite design (OCCD) [45]. The variable factors subject to optimization were the component contents in the sorbent; the response function was the residual petroleum content in soil after remediation.

Base levels and variation intervals for the mass ratios of components in the sorbent were derived from a series of preliminary experiments (Table 1).

Table 1

Base levels and variation intervals

Factor	Level			Interval
	–1	0	+1	
X_1 – content of the chitin-chitosan complex, g/kg	3.0	3.7	4.4	0.7
X_2 – content of the activated sludge suspension, g_{dry}/kg	1.4	2	2.6	0.6

According to the design matrix for the two-factor experiment (Table 1), nine parallel tests were carried out with varying ratios of the chitin-chitosan complex and activated sludge in oil-contaminated soil. The experiments were conducted at 10±2 °C, corresponding to the average daily temperature along the Kola Peninsula coastlines. For each test, a 1.5 kg sample of pre-prepared soil was placed in an open plastic container. Oil was added to each soil sample under mixing to achieve a mass ratio of 20 g of oil per 1 kg of soil. The experiment duration was 56 days. The choice of this experimental duration at a temperature of 10±2 °C is caused by the need to simulate the Arctic region conditions, where the growing season is short and oil biodegradation is possible only during the limited warm period of the year.

The residual content of petroleum products in the soil was determined by the fluorimetric method using the Fluorat-02-3M liquid analyser in accordance with PND F 16.1:2.21-98. Measurements were taken after 1, 7, 14, 28, 42, and 56 days.

Phytotoxicity test of the developed sorbent's effectiveness for remediation of oil-contaminated soils passed in accordance with FR 1.39.2006.02264[2] and GOST R ISO 22030-2009[3]. This method

[2] FR. 1.39.2006.02264. Procedure for measuring seed germination and root length in seedlings of higher plants to determine the toxicity of anthropogenically contaminated soils (M-P-2006). URL: https://fgis.gost.ru/fundmetrology/registry/16/items/295381 (accessed 12.02.2026).

[3] GOST R ISO 22030-2009. Soil quality. Biological methods. Chronic phytotoxicity in higher plants.

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relies on plant response to chemical substances affecting growth and morphological functions. Test crops included a monocotyledonous and a dicotyledonous species: common oat (*Avena sativa*) and garden radish (*Raphanus sativus*). Dry seeds of the test crops were sown in containers with prepared soil samples. An oil-contaminated soil sample without sorbents served as the control. The response test parameters were seedling height, mm; root length, mm; and plant mass, g. Sampling for phytotesting was performed on days 1, 7, 14, 21, 27, 42, and 56 of the experiment. Estimation of the phytotoxic inhibition effect (phytotoxicity) was performed in accordance with FR 1.39.2006.02264 using the specified formula

$$\text{PhE} = \left(1 - \frac{L_{\text{exp}}}{L_{\text{cont}}}\right) \cdot 100\ \%,$$

where L_{exp} is the mean value of the test function in the experimental group, mm; L_{cont} is the mean value of the test function in the control group, mm.

Results discussion

Within the study, chitin was converted from the carapace of *Paralithodes camtschaticus* into a chitin-chitosan complex to enhance its sorption characteristics relative to pure chitosan. IR spectral analysis of the chitin-chitosan complex, used as the base of the composite sorbent, showed (Fig.2) that the sample obtained via the shortened chitin conversion procedure contains absorption bands characteristic of both chitin and chitosan. Amide groups were identified (1653 cm^{-1} for amide I and 1558 cm^{-1} for amide II), indicating the presence of β1.4-linked N-acetyl-D-glucosamine (GlcNAc). A band corresponding to amino groups (–NH_2) at 1374 cm^{-1}, typical for chitosan, was also observed. However, unlike the reference Sigma chitosan, the sample derived from the carapace lacks a broad absorption band at 3295 cm^{-1}, characteristic of free OH groups formed during deacetylation, which indicates a lower degree of deacetylation. The absence of the broad band at 3295 cm^{-1} may suggest lower hydrophilicity compared to chitosan, potentially affecting the sorption mechanism. At the same time, the IR spectrum of the developed sorbent shows an absorption band at 1418 cm^{-1}, which, according to literature data, is associated with deformation vibrations of the free O–H bond not removed during acid treatment. This indicates the preservation of certain functional groups that may participate in sorption processes. The presence of amide and amino groups in the obtained chitin-chitosan complex suggests its capacity to interact with petroleum products [46].

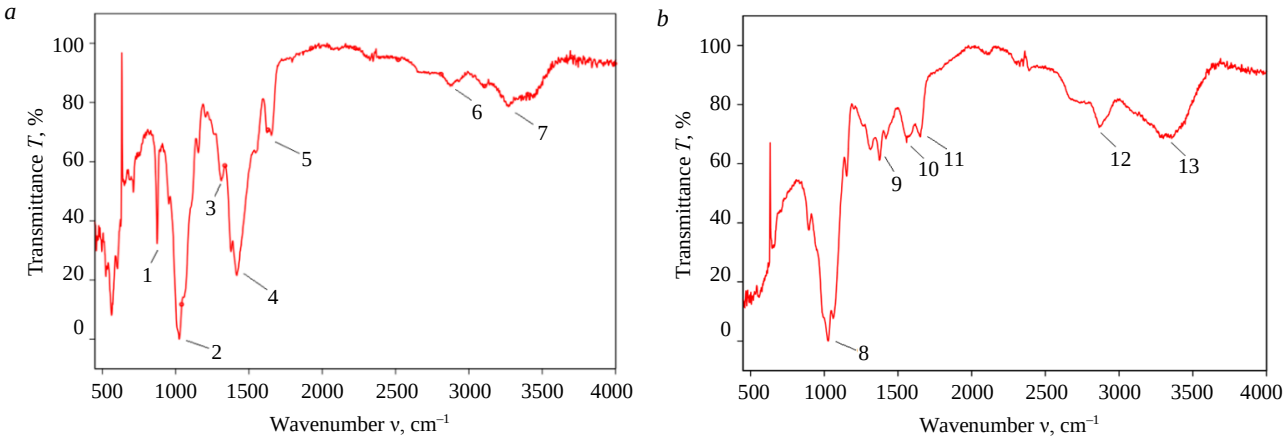


Fig.2. Obtained IR spectra of the produced chitin-chitosan complex (*a*); of the standard chitosan sample Sigma (*b*)
1 – 873; 2 – 1024; 3 – 1374; 4 – 1418; 5 – 1653 and 1558; 6 – 2880; 7 – 3449-3356; 8 – 1027; 9 – 1374; 10 – 1558; 11 – 1653; 12 – 2878; 13 – 3393-3297 cm^{-1}

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Based on the obtained data, we can conclude that the synthesized chitin-chitosan complex is an intermediate product in the transformation of chitin into chitosan. The presence of amide, amino, and partially retained hydroxyl groups accounts for its potential as a sorbent for petroleum products.

When justifying the use of activated sludge as a component of the composite sorbent for remediation of oil-contaminated coastlines, we considered the regulatory requirements specified in GOST R 54534-2011. The standard classifies activated sludge as a wastewater treatment residue and sets limits on heavy metal content to prevent their migration into the remediated soil. The performed analytical control of heavy metal content in the AS sample (Table 2) demonstrates that the actual concentrations of heavy metals in it are significantly below the set regulatory limits.

Table 2

Heavy metal content in the activated sludge

Parameter to be determined	GOST R 54534-2011 requirements	Measurement results	Test method
Zinc, mg/kg	3500	370±70	PND F 16.1:2.3:3.11
Nickel, mg/kg	400	130±50	PND F 16.1:2.3:3.11
Copper, mg/kg	750	103±21	PND F 16.1:2.3:3.11
Arsenic, mg/kg	20	0.8±0.4	PND F 16.1:2.3:3.11
Cadmium, mg/kg	30	1.6±0.8	PND F 16.1:2.3:3.11
Lead, mg/kg	500	10.9±2.7	PND F 16.1:2.3:3.11
Chromium, mg/kg	1000	42±8	PND F 16.1:2.3:3.11
Mercury, μg/kg	15	5.4±2.1	PND F 16.1:2.23

It should be noted that in this study, activated sludge is not considered as a soil substrate (for which GOST R 54534-2011 regulates additional parameters), but solely as a source of active microbial biomass. This shift in functional role renders a number of parameters standardized for soil substrates irrelevant and allows the research focus to move to the bioactive potential of the microorganisms in the activated sludge. Thus, the obtained data indicate that the studied AS complies with environmental requirements regarding heavy metal content, does not introduce additional contamination into the oil-contaminated soil, and can be used as a remedial agent in coastal remediation.

Microbiological analysis showed that the total bacterial count (TBC) in the activated sludge amounted to 1.98×10^8 cells/ml (Fig.3). The bacterial biomass reached 10,900 (10,888) mg/m^3. The number of cultivable saprotrophic bacteria was 17 million cells/ml. The study also detected microorganisms capable of using petroleum hydrocarbons as the sole source of carbon and energy; their abundance was 2500 cells/ml (Fig.3). The activated sludge bacterial community is predominantly represented by large cells 1.0-2.5 μm long. The average volume of bacterial cells in the AS community reached 0.055 μm^3.

According to the literature, the microbial population in AS can vary and depends on the conditions under which the sludge operates. According to various data, the number of bacteria can range from tens of thousands to billions of cells per 1 ml of sludge [47-49]. In this case, the total bacterial count per 1 ml of sludge reached hundreds of millions of cells. This corresponds to the maximum values obtained by Ewert and Paynter [48] when working with bacteriophages in activated sludges. The presence of bacteria capable of oxidizing petroleum hydrocarbons suggests the potential applicability of activated sludge for cleaning soils and grounds from oil contamination.

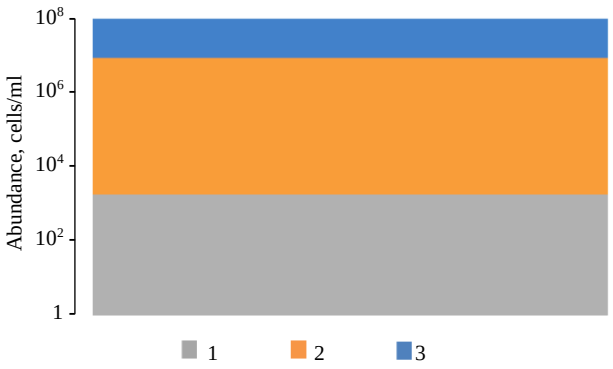


Fig.3. Total bacterial count, number of cultivable saprotrophic bacteria, and hydrocarbon-oxidizing bacteria in activated sludge
1 – HOB; 2 – saprotrophic bacteria; 3 – TBC

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An additional assessment of microbiological safety confirmed the absence of pathogenic microorganisms in the activated sludge, indicating its safety for use in coastal remediation.

Thus, the microbiological analysis confirmed the promising potential of using activated sludge as a source of hydrocarbon-oxidizing microorganisms within the composite sorbent for remediation of oil-contaminated soils.

Further research focused on modelling and identifying an effective formulation of the composite sorbent based on the chitin-chitosan complex and activated sludge, in accordance with the adopted OCCD. The experimental design and the results of its implementation are presented in Table 3.

Table 3

Results of modelling the formulation of a sorbent based on a chitin-chitosan complex and activated sludge

Experiment number	Experimental design				Response function (remediation period)						
	Chitin-chitosan complex content		Activated sludge content		PP content in soil samples *Z*, mg/g						
	Matrix value, *X*	Actual value, g/kg	Matrix value, *Y*	Actual value, g/kg	Day 1	Day 7	Day 14	Day 21	Day 27	Day 42	Day 56
1	–1	3.0	–1	1.4	5.734	1.528	1.439	1.127	1.061	1.056	1.045
2	0	3.7	–1	1.4	5.504	1.354	1.001	0.926	0.913	0.915	0.908
3	+1	4.4	–1	1.4	3.475	1.324	0.794	0.661	0.683	0.653	0.640
4	–1	3.0	0	2.0	3.465	0.842	0.611	0.454	0.448	0.449	0.444
5	0	3.7	0	2.0	2.854	0.780	0.516	0.434	0.412	0.431	0.416
6	+1	4.4	0	2.0	2.796	0.683	0.522	0.386	0.350	0.334	0.319
7	–1	3.0	+1	2.6	4.736	1.635	1.468	0.984	0.885	0.848	0.838
8	0	3.7	+1	2.6	2.690	1.222	1.214	0.949	0.85	0.796	0.779
9	+1	4.4	+1	2.6	2.457	0.925	0.864	0.761	0.693	0.674	0.661

Based on the experimental results (Table 3), regression equations were obtained for each remediation period. These equations describe the dependence of the residual PP content in the soil samples (*Y*) on the mass ratio of the chitin-chitosan complex and the activated sludge suspension,

$$Z_1 = 24.587 - 2.6431X - 13.0865Y + 0.1929X^2 - 0.0119XY + 2.9472Y^2;$$

$$Z_7 = 6.997 - 0.2194X - 5.2589Y + 0.0765X^2 - 0.3012XY + 1.5639Y^2;$$

$$Z_{14} = 9.2565 - 0.9614X - 6.4518Y + 0.0803X^2 + 0.0244XY + 1.612Y^2;$$

$$Z_{21} = 6.3557 + 0.1471X - 5.837Y - 0.0833X^2 + 0.1446XY + 1.3241Y^2;$$

$$Z_{27} = 5.8281 + 0.1984X - 5.4084Y - 0.0782X^2 + 0.1107XY + 1.2338Y^2;$$

$$Z_{42} = 5.6213 + 0.2422X - 5.2449Y - 0.0918X^2 + 0.1363XY + 1.1639Y^2;$$

$$Z_{56} = 5.6716 + 0.2121X - 5.2433Y - 0.0881X^2 + 0.1357XY + 1.1634Y^2.$$

Visualization of the regression equations using Statistica 14.0 software yielded 3D models of the oil-contaminated soil remediation for each time interval. Figure 4 presents the 3D model for the 56-day remediation period, in which the region of the optimal combination of variable factors (chitin-chitosan complex and activated sludge content) resulting in the lowest residual petroleum product content in the soil can be identified. The range of optimal combinations of variable factors across the models for the entire experimental period varies within 4.2-4.4 g/kg of chitin-chitosan complex and 1.9-2.2 g/kg of activated sludge. The extremum of the mathematical model for the 56-day remediation period corresponds to a point with the following values of the variable factors: $X_1 = 4.3$ g/kg; $X_2 = 2.0$ g/kg.

The effectiveness of the composite sorbent was confirmed through phytotesting, which involved soil samples with component ratios according to the experimental design (Table 3). The results of the phytotoxicity test of the composite sorbent's effectiveness are presented (Fig.5) for the monocotyledonous test crop – common oat.

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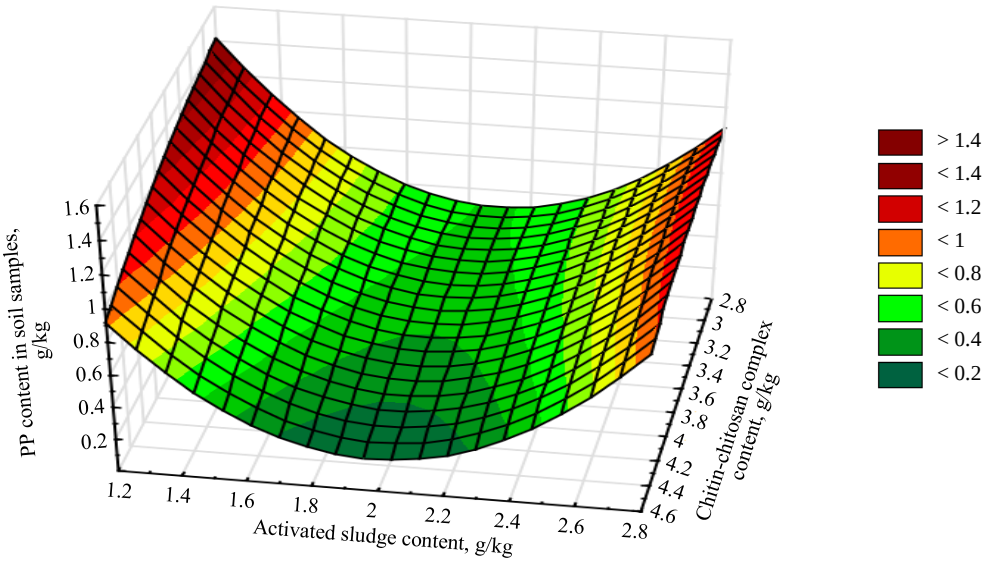


Fig.4. Model of the petroleum product sorption process in soil on day 56 at a temperature of 10 °C

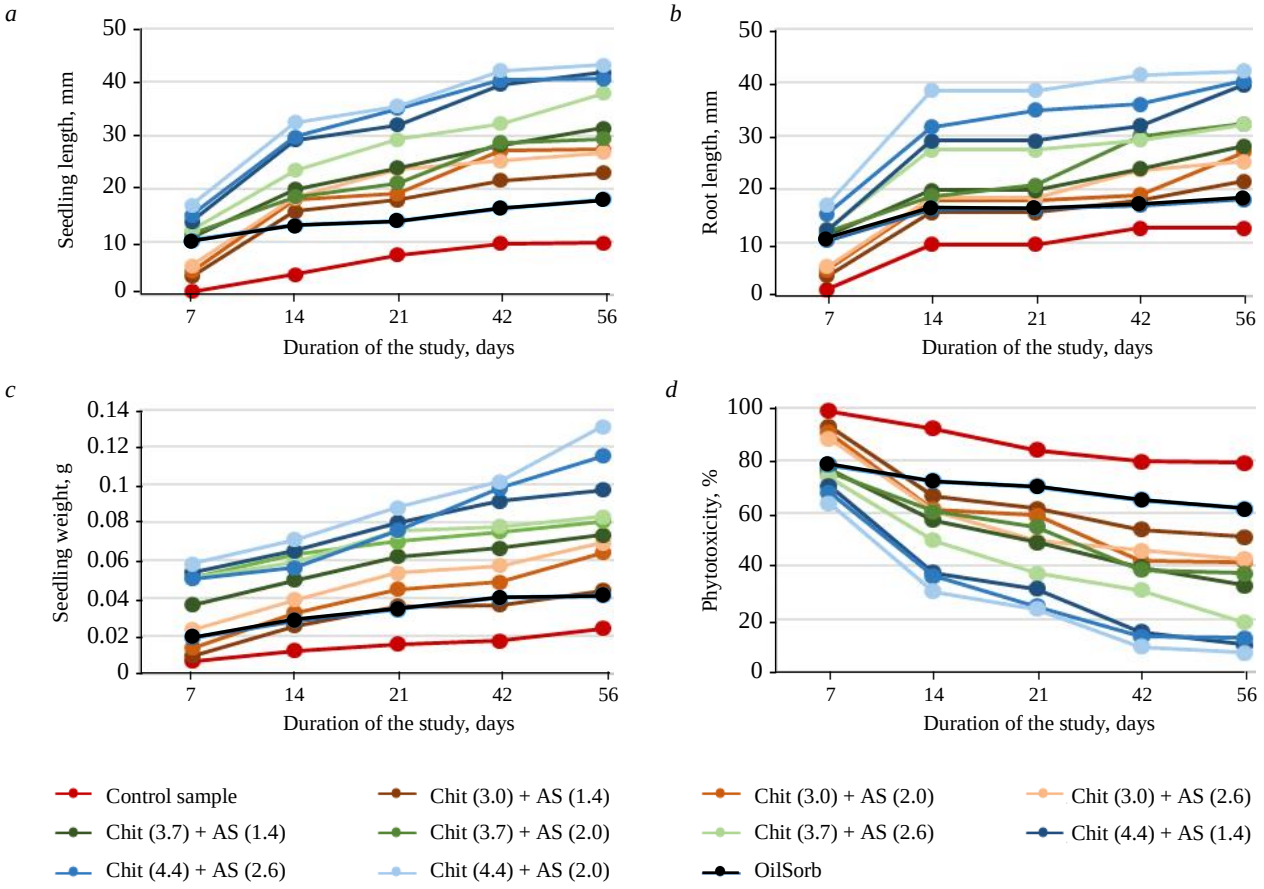


Fig.5. Dynamics of the test function indicators: seedling length (*a*); root length (*b*); plant mass (*c*); phytotoxic effect (*d*)

The test function values “seedling length”, “root length”, and “seedling mass” of the test crop showed substantial growth during the first 14 days of the experiment and a decrease in growth dynamics in the subsequent period. The lowest values of the test functions, reflecting the soil condition during remediation, were recorded for the control sample of oil-contaminated soil without any sorbent added. The best values of the test functions were observed for the sample with a component ratio of 4.4 g of chitin-chitosan complex and 2.0 g_{dry} of activated sludge per 1 kg of soil.

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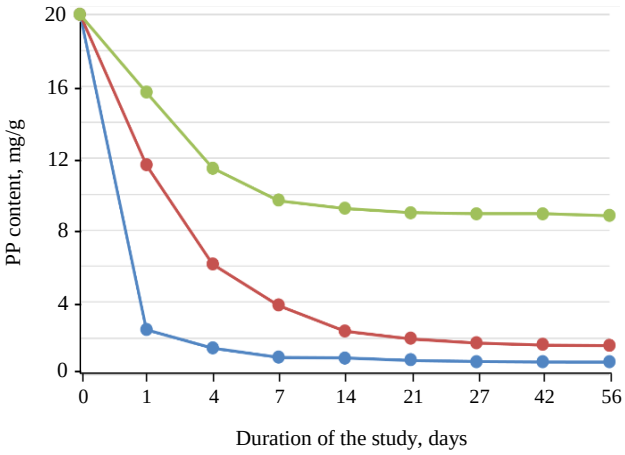


Fig.6. Dynamics of petroleum product content reduction in remediated soil using the developed and commercial sorbent

1 – Chit (4.4) + AS (2.3); 2 – OilSorb; 3 – control sample

The test function values of the commercial sorbent OilSorb during the 7- and 14-day periods of the experiment were comparable to those of the developed sorbent, but after day 21 and onwards demonstrated lower growth dynamics, indicating lower effectiveness of OilSorb for remediation of oil-contaminated soils compared to all combinations of composite sorbent components.

These patterns were confirmed upon assessment of the phytotoxicity parameter of the remediated soils: the highest phytotoxicity values were demonstrated by the control sample and the sample with the OilSorb sorbent, while the lowest phytotoxicity was shown by sorbent sample N 6 with a ratio of 4.4 g of chitin-chitosan complex and 2.0 g_{dry} of activated sludge per 1 kg of soil. This sorbent component ratio is the closest to the optimum found using the mathematical model.

To evaluate the optimal sorbent component ratio determined by means of the mathematical model (4.3 g of chitin-chitosan complex and 2.0 g_{dry} of activated sludge per 1 kg of soil), a comparison of its effectiveness for remediation of oil-contaminated soils was conducted against the commercial sorbent OilSorb, which is in use by coastal oil spill response services (Fig.6).

The reduction in residual PP content in soil without sorbent addition during day 1 of the experiment amounted to 21.6 %, which is associated with the first stage of petroleum product degradation caused by physicochemical weathering, during which gaseous and light volatile components of oil are removed from the soil (Fig.6). The reduction in residual PP content during the first day in soil with the commercial sorbent OilSorb was 41.8 %, with the sorbent's contribution to this value amounting to 20.2 % (equal to the difference between 41.8 and 21.6 %). In the oil-contaminated soil sample with the composite sorbent, the PP content reduction during the first 24 h was 87.5 %, with the sorbent's contribution to this value amounting to 65.9 % (equal to the difference between 87.5 and 21.6 %). Such a sharp reduction in PP content over such a brief period can be explained by the synergistic action of the sorption material and microorganisms.

Starting from day 7, the PP content dynamics in the soil sample without sorbent addition (control sample) remained virtually unchanged until the end of the experiment, in contrast to the other two samples, indicating attenuation of the soil self-purification.

The effectiveness of the developed sorbent at all subsequent sampling points remained higher than in the other model samples. By day 56 of the study, the residual oil content in the sample with the composite sorbent decreased by 96.7 %, with the residual PP content being 2-4 times lower than in the case of OilSorb.

Conclusion

The article presents a scientific approach to the development of a composite sorbent for remediation of oil-contaminated soils in Arctic coastal areas.

The sorbent is based on a chitin-chitosan complex obtained via a shortened, economically feasible chitin conversion procedure. Infrared spectral analysis of the chitin-chitosan complex revealed the presence of functional groups characteristic of both chitin and chitosan, indicating the intermediate nature of the studied product and confirming its potential effectiveness as a sorbent for petroleum products.

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The source of oil-oxidizing microorganisms adapted to northern conditions is activated sludge – a waste product from the biological treatment facilities of wastewater treatment plants in the Murmansk Region, in which the abundance of hydrocarbon-oxidizing bacteria capable of utilizing petroleum components as the sole source of carbon and energy amounts to 2500 cells/ml. The compliance of the activated sludge with environmental standards for heavy metal content was also confirmed, eliminating the risk of secondary soil contamination upon sorbent application.

The optimal ratio of the chitin-chitosan complex and activated sludge was substantiated experimentally within the framework of mathematical experimental design methodology using orthogonal central composite design as the key optimization tool. We obtained the regression dependences that quantitatively describe the dynamics of petroleum product concentration reduction in soil at a temperature of 10±2 °C. Visualization of these equations in a 3D coordinate system showed that the minimum residual petroleum product content in soil is achieved at optimal mass ratios of the chitin-chitosan base and activated sludge equal to 4.3 and 2.0 g/kg, respectively.

The effectiveness of the developed sorbent was verified by the following methods. Phytotoxicity test confirmed the adequacy of mathematical optimization of the sorbent formulation for elimination of oil contamination in Arctic coastal areas. Comparative tests of the remediation effectiveness of oil-contaminated soils with respect to the composite sorbent with optimal component content and the commercial sorbent OilSorb confirmed the advantage of the developed formulation. During the experiment, we found that the residual petroleum product content when applying the composite sorbent is 2-4 times lower throughout the entire observation period of the remediation process.

Thus, the conducted research enabled the development of proposed process background for manufacturing an effective composite sorbent based on a chitin-chitosan complex and activated sludge with an optimal component ratio for remediation of oil-contaminated coasts in Arctic conditions. The obtained results open up opportunities for scaling the procedure and its implementation in the practice of oil spill response in Arctic conditions, combining environmental friendliness with economic feasibility.

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