Cleaning of oil-polluted bottom sediments of the boreal lake, Samotlor oil field, North Russia: case report

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ABSTRACT

Small lakes in areas of intensive crude oil production may be susceptible to oil pollution arising from accidental spills and leaks, eventually leading to the pollution of bottom sediments. Effective cleaning of aquatic bottom sediments remains a challenge. Flotation is a potentially simple and reliable approach for the cleanup of bottom sediments without their excavation from the water body. Full-scale testing of flotation-based technology using the specially designed airlift plant allowed the cleaning of bottom sediments of an unnamed boreal lake ('the lake') within the Samotlor oil field, North Russia, heavily polluted with crude oil several decades ago. The lake bottom sediments are dominated by peat and unevenly polluted with oil. The average oil content in the lake bottom sediments was 111 g kg^{-1}. During the 1.5 months' field test in July–August 2018, the average total oil concentration in the bottom sediments of the lake was reduced to 1.99 g kg^{-1}. Secondary water contamination was minimal; the content of oil hydrocarbons in the water after completion of work did not exceed 0.09 ± 0.04 mg L^{-1}. This study demonstrates that flotation-based technology can be applied for *in situ* cleaning of oil-contaminated lake bottom sediments including those in boreal climates.

Key words | airlift plant, cleaning, flotation, lake bottom sediments, oil contamination

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HIGHLIGHTS

- Small lakes within the West Siberian oil and gas-bearing basin have become sites of intensive oil pollution.
- The flotation technique has been prooved a reasonable solution for *in situ* cleaning of oil-contaminated lake bottom sediments.
- Full-scale testing of the flotation technology using a specially designed airlift plant enabled the removal of oil from the bottom sediments polluted with oil several decades ago.

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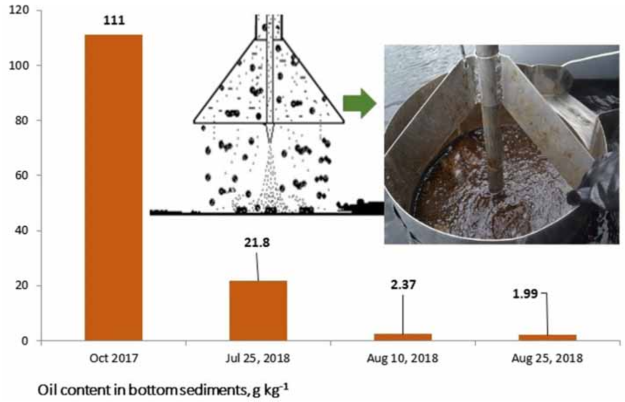
GRAPHICAL ABSTRACT



INTRODUCTION

Oil contamination of water bodies is usually associated with major accidents with serious consequences for marine ecosystems, such as the *Torrey Canyon* supertanker accident in 1967, *Exxon Valdez* tanker spill in 1987, the *Prestige* tanker spill in 2002, and the Deepwater Horizon blowout and subsequent oil spill in 2010. However, inland water bodies are also under pressure due to oil pollution in many instances. Often, marine oil spill accidents reach the shoreline, also polluting associated marshlands (Liu *et al.* 2012). Basin runoff from industrialized areas also represents an important pathway for hydrocarbons to enter freshwater reservoirs (Colella & D'Orsogna 2014). The number of spills from ageing, ill-maintained or sabotaged pipelines has increased over the world, and places like Arctic Russia have become sites of reoccurring oil pollution (Jernelöv 2010).

Russia contains some of the world's richest freshwater resources. In addition to the enormous reserves of surface water, Russia possesses one of the largest reserves of fuel and energy resources, and remains the world's third largest oil producer, accounting for 12% of global output (BP 2019). The giant West Siberian oil and gas-bearing basin occupies the entire territory of the Tyumen area, as well as overlapping neighboring areas (Soromotin 2011).

Significant ecological consequences from accidental oil spills and chronic contamination by petroleum production are typical for rivers and small water bodies such as lakes, ponds and wetlands within the West Siberian basin, which then lose their recreational value and economic importance (Nikanorov *et al.* 2015; Vorobiev & Noskov 2015; Moskovchenko & Babushkin 2017). During the period of rapid development of the mining and oil/gas industry during the Soviet era, many believed natural resources were inexhaustible, typified by the popular slogan 'We can't wait for charity from nature, we must conquer it' (Gladun & Zakharova 2017). Some oil-producing companies in post-Soviet Russia inherited contaminated lakes in this region, resulting from intensive oil production in the second half of the 20th century. Oil pollution arising from accidental spills and leaks in this era has over time led to the pollution of aquatic bottom sediments.

Cleaning such polluted water bodies remains a challenge (NOAA 2017). Technologies for removing oil from the water surface are well advanced. There are a huge variety of devices and materials used for the operational collection of oil including mechanical and bio-based materials for oil spill treatment (Doshi *et al.* 2018). Conversely, there is a large 'gap' in technologies and devices for the cleaning of polluted bottom sediments.

Flotation technologies are based on a combination of physicochemical and physical mechanisms for the separation of hydrophobic pollutants from soil in a gas–liquid–solid system (Lim *et al.* 2016). The principle of the technology is based on differences in the surface properties of the pollutant and bottom sediments, so that lighter particles with a lower sedimentation rate can be separated. The mechanism of flotation operates through the generation of air bubbles which attach to hydrophobic contaminants and rise to the surface of the reservoir, enabling the collection of pollutants using standard mechanical methods (ranging from hand skimmers to high-tech autonomous collectors). The benefits of the flotation method include simplicity, low operating cost and high oil removal efficiency (Lim *et al.* 2016). Furthermore, the flotation technology separates very fine or light particles with a low settling rate.

The use and operating parameters of flotation technology for bitumen recovery from oil sands in *ex situ* systems have been previously studied (Schramm *et al.* 2003; Masliyah *et al.* 2004). This principle can be applied for *in situ* excavation of oil from aquatic bottom sediments. In a pilot experiment, flotation technology was previously tested for decontamination of bottom sediment in Lake Schuchye (Komi Republic, North Russia), which was intensively polluted as a result of a series of accidents on the Harjaga-Usinsk and Vozej-Usinsk oil pipelines in 1994 (Lushnikov *et al.* 2006). However, no specialized equipment has been developed to date that makes it possible to efficiently extract oil pollutants from aquatic bottom sediments in a full-scale field work with no risk of secondary water pollution.

The goal of this work was to undertake a full-scale testing of the flotation technology based on the airlift principle, using a specially designed airlift plant on an oil-polluted lake within Samotlor oil field, North Russia, followed by its comprehensive examination.

MATERIAL AND METHODS

Site description

The Samotlor oil field is located near Nizhnevartovsk in Khanty-Mansiysk autonomous district-Yugra, the Tyumen region of the vast West Siberian Lowland, Russia, and it is the largest of the more than 300 fields that have been discovered in the region (Clarke 1991). The field was discovered in 1965, with production commencing in 1969.

Because of the flat relief, low drainage, and cold and humid continental climate, the West Siberian Lowland is characterized by a great expanse of peatlands (almost 50% of the territory). This region also plays an important role for freshwater accumulation as it contains more than 800,000 lakes (Solomeshch 2005).

As a result of almost five decades of oil production, small lakes and wetlands adjacent to the Samotlor oil field have been subjected to oil contamination. The subject of this study was an unnamed, non-flowing wetland lake located in the catchment area of the Samotlor oil field (Figure 1) that has been heavily contaminated with crude oil since Soviet times. Most of the pollution occurred in the 1980s and 1990s due to a series of pipeline accidents. The surface area of the lake at the time of the study was 12,635 m^2.

Sampling and field studies

Field measurements and the sampling of water and bottom sediments in the lake prior to cleaning were conducted in October 2017 to estimate the initial pollution levels of the water and bottom seidments in the lake. After the conclusion of cleaning with flotation technology in August 2018, a second sampling of water and bottom sediments was undertaken, in order to evaluate the effectiveness of decontamination.

The location of the sampling points was determined based on the morphological features of the lake, and spatially distributed to cover the entire area of the lake. The position of water and bottom sediments sampling points are shown in Figures 3 and 4. The sampling points selected were sampled both prior to cleaning to assess the initial state of the lake, and after the cleanup to monitor the effectiveness of cleaning bottom sediments by flotation technology.

Further field studies at sampling points included water transparency, surface temperature and bathymetry. Water transparency was determined by a standard method using a Secchi disk, where water depth is determined by the depth to which the standard white disk (Secchi disk) remains visible. Water temperature was measured with a meteorological thermometer with an accuracy of 0.1 °C.

Water sampling was performed to analyze total oil content, concentration of Cl^-, pH, electrical conductivity, and biochemical oxygen demand. Three water samples of approximately 1 L were collected in glass bottles and stored at 4 °C for 4–6 hours until laboratory analysis. Bottom sediments were collected for chemical and physico-chemical analysis (measurements of organic matter content, total oil content, electrical conductivity, Cl^- concentration and pH of water extracted). Bottom sediments (approximately 1 kg for each sampling point) were collected using a P 04.09 peat sampler (Eijkelkamp, Netherlands). Samples were removed from the sampler and transferred into glass jars, then stored at 4 °C until analysis. Bottom sediments of the upper horizon (0–25 cm) were collected and analyzed in this study.

Analytical methodology

All the samples were processed at the hydrochemical laboratory of Yugra State University (Khanty-Mansiysk, Tyumen region).

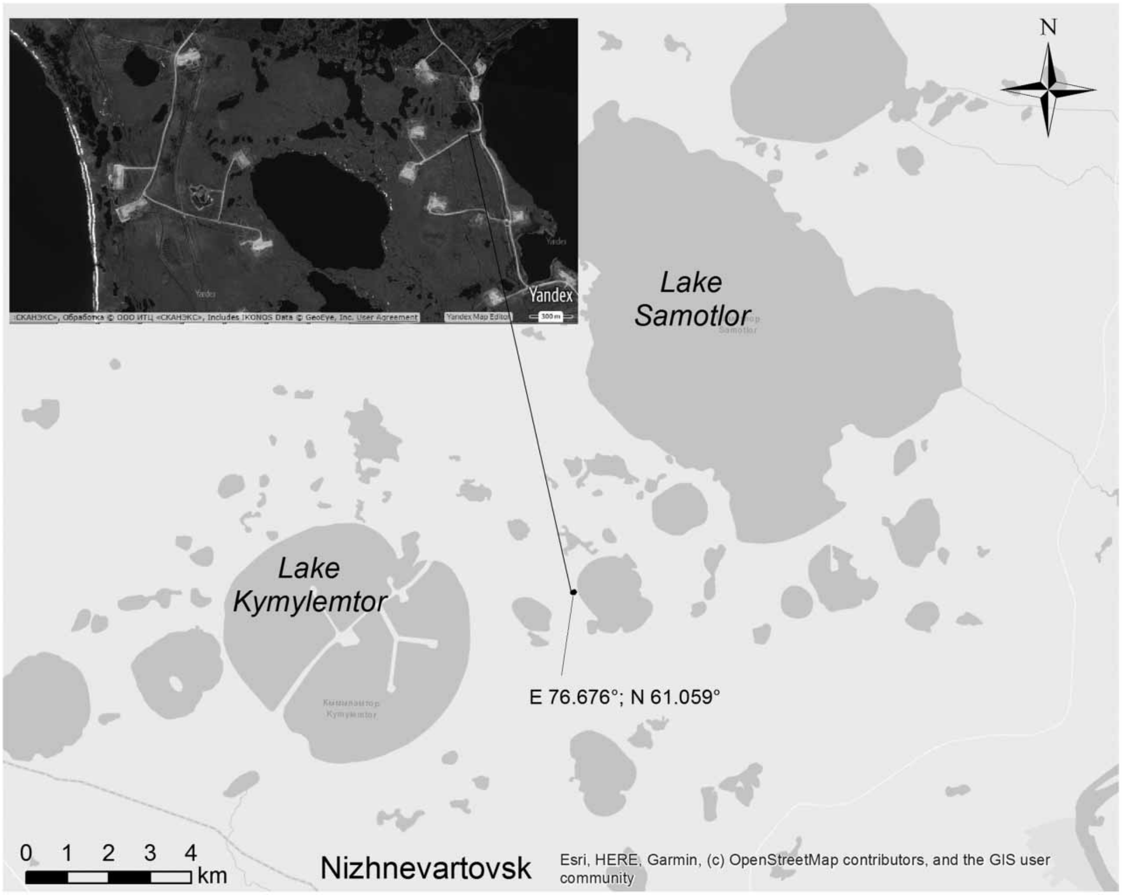


Figure 1 | Location of the boreal lake, Samotlor region.

The total hydrocarbon content in water and bottom sediments was determined by infrared (IR) spectrophotometry (PND F 14.1:2:4.5-95; PND F 16.1:2.2.22-98). The method is based on extraction of the oil with CCl_4 and chromatographic fractionation with aluminum oxide, followed by IR quantitative determination using a Nicolet iS10 Fourier transform IR spectrophotometer, Thermo Fisher Scientific (USA). To calculate the oil accumulation coefficient (bottom sediments: water) we used the ratio of total oil concentration in bottom sediments (mg kg^{-1}) to that in water (mg L^{-1}).

A dual purpose conductivity/pH meter MPC 227, Mettler-Toledo GmbH (Switzerland), was used for measuring pH and electrical conductivity in water and in the water extract of the bottom sediment. The concentration of chloride in water and bottom sediments was determined by ion chromatography (PND F 14.1:2:4.132-98; PND F 16.1.8-98) using a 761 Compact IC, Metrohm (Switzerland). To calculate the coefficient of chloride accumulation (bottom sediments:water) we used the ratio of Cl^- concentration in bottom sediments (mg kg^{-1}) to that in water (mg L^{-1}).

Loss on ignition (LOI) was determined to estimate total organic matter content (%) in the sediments. The method is based on determining the weight loss of a sample after calcination at a temperature of 525 °C (GOST 26213-91, 1992).

Biochemical oxygen demand (BOD_5) was determined by the amount of oxygen, in mg L^{-1}, which was consumed by biochemical oxidation of organic substances in water after 5 days (PND F 14.1:2:3:4.123-97).

ArcGIS-10.2.2 software was used to create a bathymetry depth map and for modelling bottom oil distribution maps.

Field work on cleaning bottom sediments

The flotation technology used was an airlift plant (Figure S1, Supplementary Material) consisting of the following: (1) deck frame module (catamaran, boat engine, base frame for fixing the lifting system); (2) movement and positioning module (four filling containers, winch, rope, four plow anchors); (3) oil lifting module (motor pump, suction hose with mesh, pressure hose, ejector, dismountable supply

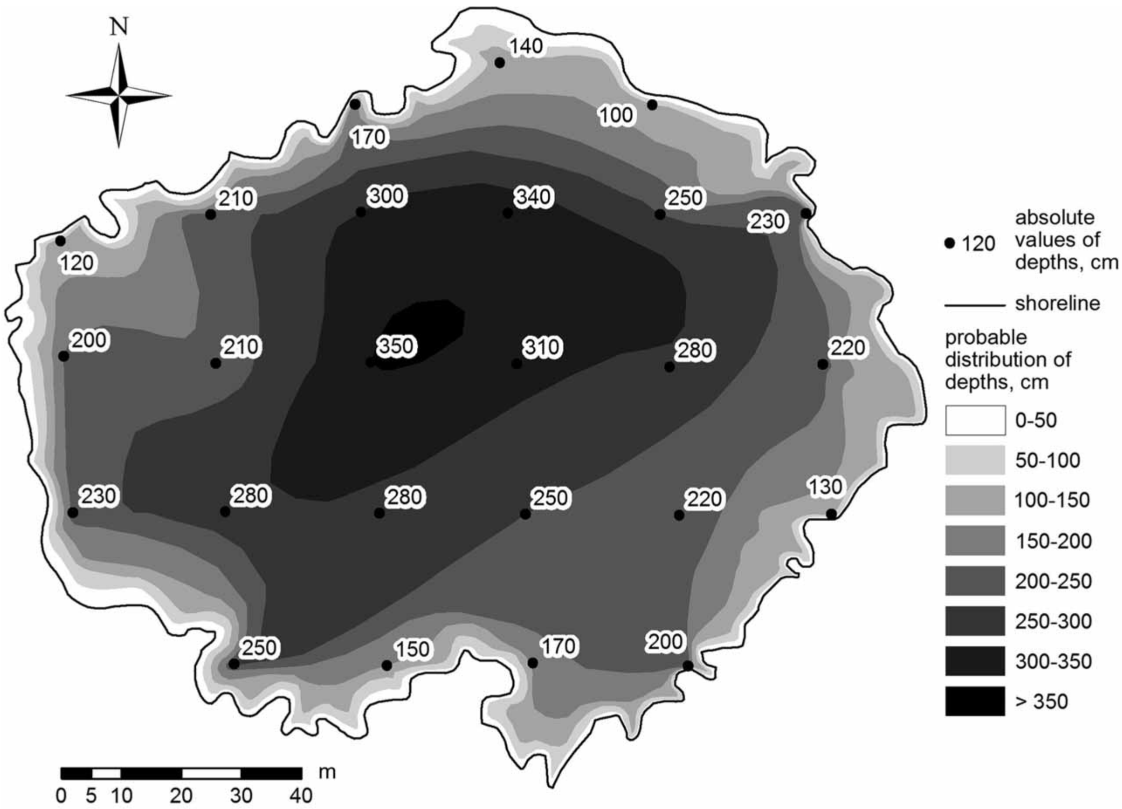


Figure 2 | Absolute values and probabilistic distribution of lake depths (cm).

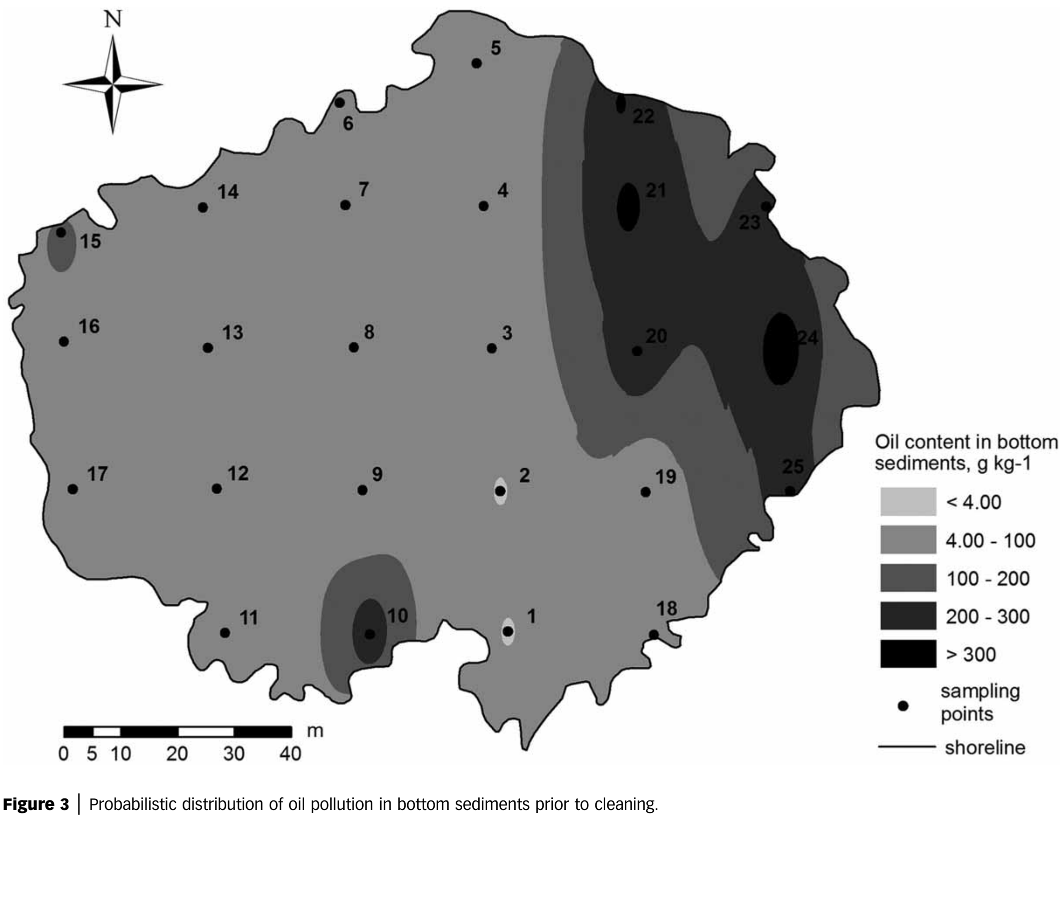


Figure 3 | Probabilistic distribution of oil pollution in bottom sediments prior to cleaning.

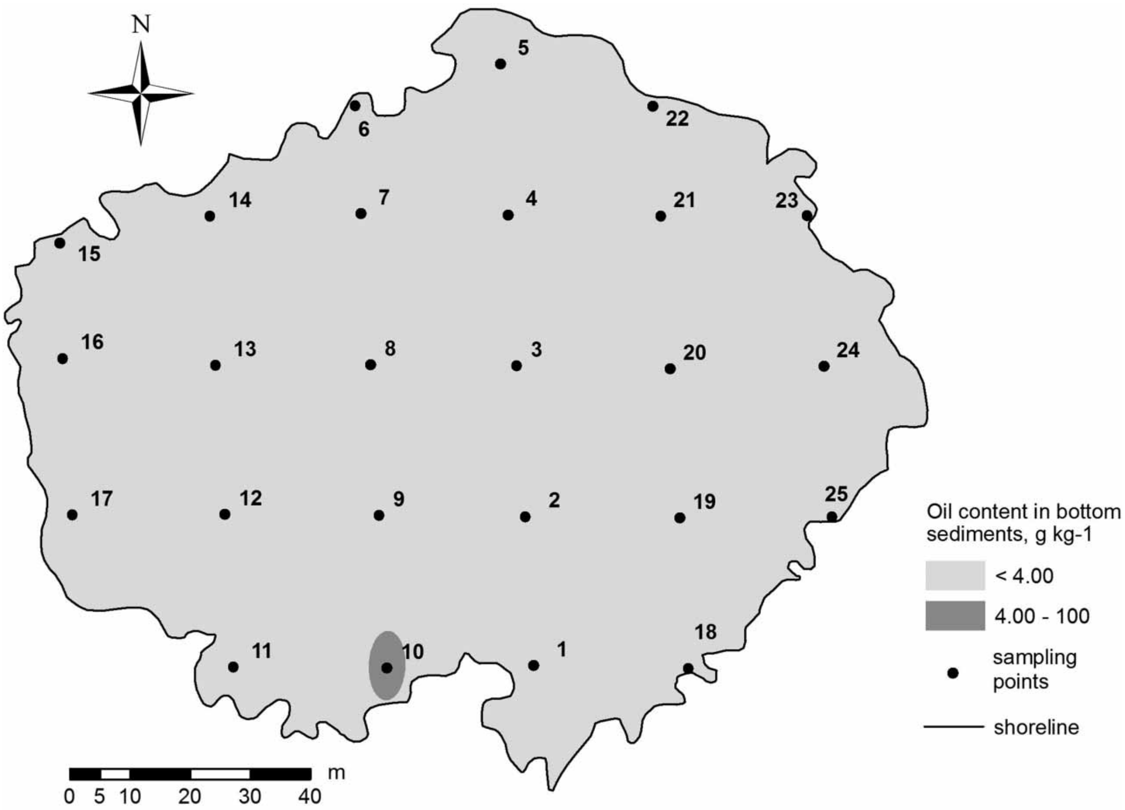


Figure 4 | Probabilistic distribution of oil pollution in bottom sediments after cleanup.

pipe, two sprayers, prefab umbrella, airlift corrugated pipe, primary receiving tank); (4) oil collection module (oil skimmer, oil-collecting tank with cylinders).

When the airlift plant was started, an idle supply of the water–air mixture was maintained until the oil-collecting tank was filled with water so that the light hydrophobic fraction did not enter the drainage pipe. The channel for supplying the water–air mixture was lowered, together with the intake hood, to the required depth of 1.5 m from the bottom of the lake (and when working in shallow areas, to the minimum possible depth) at an angle of 45°. The device was fixed above the bottom sediments and the separation of accumulated oil from bottom sediments commenced. As a result of the operation of the airlift plant oil aggregates in the bottom sediments were covered with air bubbles and rose to the surface in an ascending airlift flow (Figure S2). The oil extracted from the bottom sediments was accumulated in the oil-collecting tank. After cleaning bottom sediments, the residual oil was collected from the surface of the reservoir using a vacuum truck (KAMAZ KO-529-13). The airlift plant was operated within the floating barrier fenced 20×20 m squares (sections No. 1 and No. 4) or entire perimeter of the section (sections No. 2 and No. 3). Cleaning the bottom took 46 days; the work continued from July 10, 2018 to August 24, 2018. The average area of bottom sediment processing per minute was 2.5 m^2, and the average area of bottom sediment processing per day was 1,050 m^2 (Table S1).

For the convenience of cleaning the bottom sediments, the lake was divided into four quadrants (Figure S3): (1) section No. 1 with an area of 3,313 m^2 (bottom sediments with an average oil content of 288 g kg^{-1}); (2) section No. 2 with an area of 352 m^2 (average oil content of 250 g kg^{-1}); (3) section No. 3 with an area of 71 m^2 (average oil content of 157 g kg^{-1}); (4) section No. 4 with an area of 8,899 m^2 (the least polluted bottom sediments with an average oil content of 38 g kg^{-1}).

RESULTS AND DISCUSSION

Initial condition of the lake

A probabilistic model of depths distribution was developed (Figure 2). The depths ranged from 1.0 m near the shoreline to 3.5 m in the central part of the lake. The average depth of the lake, determined based on the model of the probability distribution of depths, was 1.7 m. The water transparency ranged from 1.7 to 1.8 m (Table 1). The temperature of the lake in October 2017 ranged from 2.0–2.5 °C at the surface (Table 1) to 3.0–3.5 °C (data not shown) at the bottom, which was higher due to the establishment of reverse temperature stratification in the autumn.

The water was somewhat acidic, with a pH of 4.7–4.8 (Table 1), attributable to peaty bottom sediments. The specific electrical conductivity of water ranged from 184 ± 9.20 to 291 ± 15.0 μS cm^{-1}; the chloride ion concentration did not exceed 61.5 ± 8.00 mg L^{-1}. Unlike bottom sediments, the water column was not heavily contaminated with oil. Hydrocarbons in the water did not exceed 0.07 ± 0.03 mg L^{-1}, and the bulk of high-density oil was accumulated in bottom sediments, as described below.

The LOI was determined to estimate total organic matter content in the sediments. LOI values in the studied bottom sediments significantly differed, from 13.7% at sampling point No. 11 to 99–99.2% in point Nos. 20 and 23–25 (Table 2). The bottom sediments sampled from the lake were dominated by peat, which is typical for the regions with widespread peatlands due to organic matter from particulate detritus of plants located around the lake and surrounding land (Meyers & Ishiwatari 1995). The mineral component detected in the sediments results from sand infilling from basin roads.

The bottom sediments of the lake were characterized as slightly acidic with a pH of 5.55–6.81 (Table 2). The maximum chloride concentration, 519.8 mg kg^{-1}, was detected in sediments at sample point 3. The minimum chloride concentrations (milligrams per 1 kg of bottom sediments) were found at sample points 21–22 and 24–25. The accumulation coefficient for chlorides in bottom sediments, calculated from the average concentrations in water and bottom sediments, was 1.7. The high concentration of chlorides in the sediments of the lake at the abovementioned sampling points can be attributed to local pollution with highly mineralized formation waters. Specific electrical conductivity in the water extract of the bottom sediments did not exceed 211 μS cm^{-1} and in many cases did not correlate with the concentration of chloride ions.

The lake bottom at the time of the survey in October 2017 was unevenly contaminated with oil (Figure 3, Table 3). The minimum total concentration of petroleum hydrocarbons in bottom sediments at sample point 2 was 2.2 g kg^{-1}. Higher total concentrations of petroleum hydrocarbons were detected at sample points 10 (250 g kg^{-1}) and 15 (157 g kg^{-1}). However, the bottom was most contaminated in the eastern part of the lake (Figure 3, Table 3), where the highest content of petroleum hydrocarbons, 342 g kg^{-1}, was detected in sediments at sample point 24. The average total oil content in the bottom sediments of the lake was 111 g kg^{-1} but varied throughout the lake.

Maximum concentrations of petroleum hydrocarbons were detected in peaty sediments which (1) have a high sorption capacity and (2) contain natural biogenic organic compounds that can be recognized as petroleum hydrocarbons (Kelly-Hooper *et al.* 2013). Previous investigation of bottom sediments in the Ob tributary, Vasyugan River, showed a definite connection between the content of organic substances and petroleum hydrocarbons (r = 0.94) (Vorobiev 2003).

Table 1 | Some physico-chemical characteristics of water

Parameters of water	October 2017 Sampling point 3	October 2017 Sampling point 13	October 2017 Sampling point 19	August 2018 Sampling point 3	August 2018 Sampling point 13	August 2018 Sampling point 19
Water clarity, m	1.70	1.80	1.70	1.40	1.40	1.50
Surface water temperature, °C	2.00	2.50	2.30	15.1	15.5	15.5
pH[a], pH units	4.70 ± 0.20	4.70 ± 0.20	4.80 ± 0.20	4.80 ± 0.10	4.80 ± 0.10	4.70 ± 0.10
Electrical conductivity[a], μS cm^{-1}	248 ± 12.0	291 ± 15.0	184 ± 9.20	na	na	na
Oil content[a], mg L^{-1}	< 0.05	0.05 ± 0.02	0.07 ± 0.03	0.09 ± 0.04	0.09 ± 0.03	0.09 ± 0.04
Chlorides[a], mg L^{-1}	60.5 ± 7.90	61.5 ± 8.00	60.0 ± 7.80	74.7 ± 6.70	73.8 ± 6.60	73.0 ± 6.60
BOD_5^a, mg O_2 L^{-1}	0.70 ± 0.10	0.83 ± 0.12	0.86 ± 0.12	2.00 ± 0.30	4.90 ± 0.70	3.00 ± 0.40

na, not analyzed.
[a]Single measurement result ± error.

Table 2 | Physicochemical characteristics of bottom sediments in October 2017

Sampling point	Parameters of bottom sediments			
	Organic matter[a], %	pH[a], pH units	Electrical conductivity[a], μS cm^{-1}	Chlorides[a], mg kg^{-1}
1	94.9 ± 5.70	6.25 ± 0.10	52.6 ± 3.90	159 ± 39.6
2	95.7 ± 5.70	5.72 ± 0.10	147 ± 11.0	200 ± 49.9
3	93.7 ± 5.60	6.32 ± 0.10	36.2 ± 2.70	520 ± 130
4	92.4 ± 0.45	6.00 ± 0.10	53.2 ± 4.00	66.5 ± 16.6
5	32.6 ± 0.63	5.55 ± 0.10	74.7 ± 5.60	24.8 ± 6.20
6	20.9 ± 0.63	6.20 ± 0.10	107 ± 8.00	112 ± 27.9
7	18.6 ± 0.56	5.95 ± 0.10	70.3 ± 5.30	44.5 ± 11.1
8	23.5 ± 0.71	6.03 ± 0.10	40.0 ± 3.00	22.9 ±5.70
9	78.8 ± 2.40	5.76 ± 0.10	223 ± 17.0	96.4 ± 24.1
10	92.5 ± 5.60	6.21 ± 0.10	38.8 ± 2.90	214 ± 53.6
11	13.7 ± 1.40	6.81 ± 0.10	51.4 ± 3.90	49.8 ± 12.4
12	28.0 ± 0.84	5.87 ± 0.10	68.7 ± 5.20	33.1 ± 8.30
13	17.5 ± 0.52	6.08 ± 0.10	53.7 ± 4.00	26.5 ± 6.60
14	56.9 ± 1.70	5.85 ± 0.10	166 ± 12.0	61.5 ± 15.4
15	93.6 ± 5.60	6.09 ± 0.10	34.2 ± 2.60	354 ± 88.5
16	26.3 ± 0.79	6.10 ± 0.10	64.9 ± 4.90	54.1 ± 13.5
17	27.0 ± 0.81	5.99 ± 0.10	71.7 ± 5.40	32.2 ± 8.10
18	94.3 ± 5.70	5.80 ± 0.10	200 ± 15.0	144 ± 35.9
19	58.1 ± 1.70	5.92 ± 0.10	131 ± 9.80	63.1 ± 15.8
20	99.2 ± 6.00	6.25 ± 0.10	52.1 ± 3.90	19.7 ± 4.90
21	98.8 ± 5.90	5.96 ± 0.10	17.4 ± 1.30	7.10 ± 1.80
22	98.5 ± 5.90	6.25 ± 0.10	3.57 ± 0.27	5.10 ± 1.30
23	99.0 ± 5.90	6.30 ± 0.10	110 ± 8.20	366 ± 91.5
24	99.1 ± 5.90	6.08 ± 0.10	11.0 ± 0.82	6.50 ± 1.60
25	99.0 ± 5.90	6.27 ± 0.10	3.35 ± 0.25	2.80 ± 0.70

[a]Single measurement result ± error.

Table 3 | Oil content in bottom sediments

Sampling point	Oil content in bottom sediments[a], g kg^{-1}			
	October 2017	July 25, 2018	August 10, 2018	August 25, 2018
1	2.80 ± 0.70	10.9 ± 2.73	1.72 ± 0.43	0.78 ± 0.20
2	2.17 ± 0.54	na	na	1.31 ± 0.33
3	2.83 ± 0.71	13.6 ± 3.40	1.51 ± 0.38	1.14 ± 0.28
4	20.3 ± 5.10	na	na	1.55 ± 0.39
5	64.9 ± 16.2	na	na	2.41 ± 0.60
6	71.1 ± 17.8	na	na	1.77 ± 0.44
7	25.7 ± 6.42	na	na	1.98 ± 0.50
8	66.4 ± 16.6	na	na	1.98 ± 0.50
9	30.7 ± 7.68	na	na	1.98 ± 0.49
10	250 ± 62.5	na	na	5.73 ± 1.43
11	34.1 ± 8.53	na	na	1.31 ± 0.33
12	56.7 ± 14.2	na	na	3.42 ± 0.86
13	31.4 ± 7.85	40.8 ± 10.2	4.10 ± 1.03	1.27 ± 0.32
14	18.3 ± 4.58	na	na	3.11 ± 0.78
15	157 ± 39.4	na	na	2.97 ± 0.74
16	86.7 ± 21.7	na	na	1.11 ± 0.28
17	42.9 ± 10.7	na	na	1.17 ± 0.29
18	63.0 ± 15.7	na	na	2.40 ± 0.60
19	27.8 ± 6.94	na	2.80 ± 0.70	1.61 ± 0.40
20	241 ± 60.3	na	na	1.95 ± 0.49
21	316 ± 78.9	na	na	2.47 ± 0.62
22	304 ± 76.1	na	na	1.13 ± 0.28
23	270 ± 67.6	na	na	1.73 ± 0.43
24	342 ± 85.5	na	1.72 ± 0.43	0.77 ± 0.19
25	253 ± 63.2	na	na	2.76 ± 0.69

na, not analyzed.
[a]Single measurement result ± error.

The oil accumulation coefficient, calculated from the average total concentrations of petroleum hydrocarbons in water and in bottom sediments, was high, amounting to 1.7×10^6. This distribution is consistent with long-term contamination. After evaporation and solubilization of lighter oil fractions the remainder forms a stable water-in-oil emulsion known as a 'mousse'. Further weathering by photo-oxidation and biological oxidation produces semi-solid asphaltic residuum (Freedman 1995). The resultant increased-density oil, such as weathered oil and dilbit, has a greater potential for sinking than conventional oils. Evaporation of the diluent with time could increase the oil density, causing the residual oil to submerge or sink. The weathered oil is available for interaction with suspended sediments because little of the bitumen itself can evaporate or biodegrade, further increasing the possibility of sedimentation and sequestration (Winter & Haddad 2014). More than 30 years ago it was obvious that petroleum hydrocarbons can remain for a long time in lake sediments even though partial degradation of petroleum components occur (Meyers 1987). Furthermore, studies from marsh sediments in West Falmouth contaminated in 1969 by oil spilt from the barge *Florida* demonstrate that petroleum residues continue to persist in Wild Harbor sediments after 30 years (Reddy *et al.* 2002).

Results on cleaning of the bottom sediments

The airlift plant (Figure S1), a prototype previously tested in a laboratory model (Vorobiev *et al.* 2016), was applied for decontamination of oil-polluted bottom sediments of the lake in a full-scale field experiment. The operation of the airlift plant is based on the principle of flotation or airlift. This method involves the formation and impact of air bubbles, which are attached to hydrophobic particles (oil) in bottom sediments and uptake them on the water surface. Further, the aggregates of air and pollutant rise, forming a layer on the surface to be collected mechanically. Thus, flotation of oil pollution includes the following stages (Tao 2005): (1) interaction of the contaminant and air bubbles; (2) attachment of the contaminant to the bubbles to form aggregates; (3) flotation (airlift) of gas and contaminant aggregates; (4) separating the contaminant from air bubbles on the surface.

One of the main advantages of the flotation process for cleaning oil-polluted bottom sediments is the formation of a product with a high oil/water ratio. The oil fraction in the fluid recovered with a vacuum suction system varied from 0.16 to 0.5 (Fujita *et al.* 2004), whereas when lifting oil from the bottom using flotation, the oil-to-water proportion under different initial conditions fluctuated from 0.5 to 0.9 (Shi *et al.* 2017). In this research, the oil-to-water ratio in the pumped-out fluid was close to 0.7/0.3. This, firstly, reduces the requirements for the volume of tanks for temporary storage of the collected oil, and, secondly, it makes it possible to obtain a concentrated oil-containing liquid suitable for further use instead of waste (ensuring the principle of non-waste and recycling of natural resources). The oil collected in the current work was not marketable due to the old pollution and could not be processed. All collected oil was transferred to a sludge tank for disposal. From an environmental point of view, the removal of the oil from the environment is always the best solution, but economic aspects may restrict the methods available for oil spill treatment (Doshi *et al.* 2018). Since low operating cost is one of the benefits of the flotation method (Lim *et al.* 2016), it overcomes this limitation.

In the Russian economic reality the cost of cleaning bottom sediments using one airlift unit is USD143/hour in the north of Russia. These costs include salaries, tax payments, employee insurance, equipment and consumables costs, overhead costs, and profitability. Depending on the level of oil pollution of bottom sediments, three groups of lakes for cleaning the bottom sediments may be identified according to labor costs: (1) total oil content up to 50 g per kg of bottom sediments – 159 h per 10,000 m^2 (USD22.8 thousand per 10,000 m^2); (2) total oil content 50.1–100.0 g kg^{-1} – 238 h per 10,000 m^2 (USD34.0 thousand per 10,000 m^2); (3) total oil content more than 100.1 g kg^{-1} – 318 h or USD45.5 thousand per 10,000 m^2.

Evaluation of bottom sediments cleaning efficiency

There is an opinion that the usage of flotation may have restricted efficiency for the removal of aged or weathered oil from soil and sediments (Lim *et al.* 2016). The ageing of oil increases the interaction forces between oil and contaminated substrate, complicating flotation (Wang *et al.* 2010). Despite this, the average concentration of oil in the bottom sediments of the lake was reduced by more than 50 times during the current work.

Before the start of the pilot experiment, the lake bottom was unevenly polluted with oil, and the total oil content in bottom sediments reached 342 g kg^{-1} of bottom sediments (Figure 3, Table 3). The average oil content in the bottom sediments of the lake was 111 g kg^{-1}. As a result of control sampling and analysis of bottom sediment samples at several points 2 weeks after the start of the work, the concentration of oil hydrocarbons was higher than the values of 2017 (Table 3). We associate this with the initial redistribution of oil in bottom sediments as a result of bottom treatment. During the further work on cleaning the bottom of the lake, total content of oil in the bottom sediments steadily decreased; on August 10, 2018 the concentration of petroleum hydrocarbons did not exceed 4.10 g kg^{-1} (Table 3). The final concentration of oil in the bottom sediments sampled at August 25, 2018 averaged over the lake was 1.99 g kg^{-1}; the maximum value was within 5.73 ± 1.43 g kg^{-1} (Figure 4, Table 3).

At present, maximum permissible concentrations of total oil and petroleum products in bottom sediments are not established at the state level either in Russia or, to our knowledge, in other countries. The decree of the Khanty-Mansiysk autonomous district-Yugra Government No. 432-p dated November 23, 2018 recently enforced the regional regulation '*Permissible residual content of oil and petroleum products in bottom sediments after restoration work on water bodies in the Khanty-Mansiysk autonomous district-Yugra*' (Government of Khanty-Mansiysk 2018), which is 4.0 g kg^{-1} for bottom sediments rich in organics (LOI more than 60%) in the case of using IR spectroscopy for quantitative analysis and 1.0 g kg^{-1} in the case of fluorimetry application. Rather high maximum permissible oil concentrations are caused by high 'background' concentrations in northern lakes and rivers reviewed by Vorobiev & Noskov (2015). Inspection of freshwater bottom sediments in the Khanty-Mansiysk autonomous district-Yugra and neighboring regions confirmed a high 'background' content of hydrocarbons up to 3.31 g kg^{-1} (Gendrin *et al.* 2006).

The final concentration of petroleum hydrocarbons in bottom sediments averaged 1.99 g kg^{-1} in the lake, which is two times less than the maximum permissible level. According to the data obtained at 24 sampling points, the achieved concentrations of oil in bottom sediments satisfied this criterion, with the exception of a slight excess at point No. 10 (Figure 4, Table 3). However, this complies with the requirements of the regional regulation, according to which it is allowed to exceed the normative value by no more than two times for <20% of bottom sediments samples. In this instance the excess in one sample corresponds to 4% variation of the normative value.

The consequences of cleaning bottom sediments for water column

The main disadvantages of the flotation technique are the risk of secondary water contamination, the requirement for large amounts of water to aid the process, and high downstream cost for treatment of wastewater in *ex situ* systems (Lim *et al.* 2016). The airlift plant used in the current work was designed to enhance bottom sediment decontamination efficiency while minimizing harmful secondary effects on the entire ecosystem.

To ensure there was no serious secondary water contamination after the operation of the airlift plant and cleaning of sediments, we sampled and analyzed surface water on August 25, 2018. The sampled water was characterized by pH of 4.7–4.8 (Table 1), which corresponds to water pH prior to carrying out activities. A significant increase in BOD_5 was detected (Table 1), which might reflect release of compounds available for oxidation by microorganisms. Some of the elevated hydrocarbons may be degraded by microorganisms in the near-surface layer of water and evaporated from the water surface. However, the bulk of the oil extracted from the bottom sediments was accumulated in the oil-collecting tank and disposed of.

Even so, total oil content in water along with chloride concentration was slightly elevated in August 2018 compared to October 2017, which can be explained by the release of Cl^{-} from bottom sediments during the cleaning, and by the higher lake water level in October. The content of oil hydrocarbons in the water did not exceed 0.09 ± 0.04 mg L^{-1}, which is within the maximum permissible concentration for drinking water in Russia, 0.3 mg L^{-1} (GN 2.1.5.1315-03 2003). The value was also on a par with the oil content in water after cleaning of bottom sediments by the airlift plant prototype in a laboratory experiment, which reached 0.08 ± 0.03 mg L^{-1} (Vorobiev *et al.* 2016).

CONCLUSION

Full-scale testing of the flotation technology using the specially designed airlift plant enabled the removal of oil from the bottom sediments of a boreal lake heavily polluted with oil several decades ago. Overall, the average total oil concentration in the bottom sediments of the lake was reduced by more than 50 times, from 111 to 1.99 g kg^{-1}. Secondary water contamination was minimal; the content of oil hydrocarbons in the water after completion of work did not exceed 0.09 ± 0.04 mg L^{-1}.

The flotation technique proved to be a reasonable solution for *in situ* cleaning of oil-contaminated lake bottom sediments including those in severe climatic conditions. The possibility of cleaning of bottom sediments without their excavation and onshore treatment through the use of flotation technology will save significant time and resources, and minimize the formation of secondary waste.

CONFLICTS OF INTEREST

There are no conflicts to declare.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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