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Ecological and Geochemical Investigations in the Ob Estuary at Sites of Hydrocarbon Exploration Drilling

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Abstract—During recent years, the basins of the Kara Sea (Kamennomysskaya, Obskaya, and Chugor'yakhinskaya structures) in the Russian Federation have been considered as promising regions for oil and gas exploration and, simultaneously, as possible paths of relatively cheap pipeline and tanker transportation of hydrocarbons projected for recovery. On the other hand, exploration operations, recovery, and transportation of gas pose a considerable risk of accidents and environmental pollution, which causes a justified concern about the future state of the ecological system of the Gulf of Ob and the adjoining parts of the Kara Sea. Therefore, regular combined environmental investigations (monitoring) are the most important factor for estimating the current state and forecasting the dynamics of the development of estuary systems. The program of investigations (schedule, station network, and measured parameters) is standardized in accordance with the international practice of such work and accounts for the experience of monitoring studies of Russian and foreign researchers. Two measurement sessions were performed during ecological investigations in the region of exploration drilling: at the beginning at final stage of drilling operations and borehole testing; in addition, natural parameters were determined in various parts of the Ob estuary before the beginning of investigations. Hydrophysical and hydrochemical characteristics of the water medium were determined and bottom sediments and water were analyzed for various pollutants (petroleum products, heavy metals, and radionuclides). The forms of heavy-metal occurrence in river and sea waters were determined by the method of continuous multistep filtration, which is based on water component fractionation on membrane filters of various pore sizes. These investigations revealed environmental pollution by chemical substances during the initial stage of drilling operations, when remains of fuels, oils, and solutions could be spilled, and part of the chemical pollutants could enter the environment. Owing to horizontal and vertical turbulent diffusion, wave mixing, and the effect of the general direction of currents in the Ob estuary from south to north, areas are formed with elevated concentrations of the analyzed elements and compounds. However, the concentration levels of chemical pollutants are practically no higher than the maximum admissible concentrations, and their substantial dissipation to the average regional background contents can be expected in the near future. Our investigations allowed us to determine in detail the parameters of anthropogenic pollution in the regions affected by hydrocarbon exploration drilling in the Obskii and Kamennomysskii prospects in the Gulf of Ob and estimate their influence on the ecological state of the basin of the Ob River and the Kara Sea on the whole.

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INTRODUCTION

The examination of environmental conditions in the Arctic basin on a regional scale (Kara Sea basin) is strongly linked to the investigations of local pollution sources, which include primarily the region of the Novaya Zemlya Archipelago with its nuclear test site on the land surface and sites of solid radioactive waste disposal in shallow bays and basins of the great Siberian rivers Ob and Yenisei. In terms of ecology, large Siberian rivers are the next most important source of anthropogenic pollution in the Kara Sea. An investigation of the Kara Sea basin over a long time period has revealed artificial radionuclides and chemical pollutants, including organic substances and heavy metals, which have been transported by the run-off of the Ob and Yenisei into the marine environment from radiochemical and chemical plants and oil and gas production complexes.

During the past few years, the basin of the Ob estuary (Kamennomysskaya, Obskaya, and Chugor'yakhinskaya structures) have received increasing attention as a promising region for oil and gas exploration and, simultaneously, as a possible path for relatively cheap pipeline and tanker transportation of hydrocarbons projected for recovery. Gas exploration in the estuary zone of the Ob River is of prime importance for future economic development of Russia. On the other hand, exploration and extraction operations and gas transportation pose a considerable risk of accidents and environmental pollution, which causes a justified concern as to the future state of the ecosystem of the Gulf of Ob and the adjacent part of the Kara Sea. The input of organic pollutants, especially petroleum products and toxic substances, into water basins activates reduction processes in sediments with the participation of organic matter, changes the sulfur and carbon cycle, causes oxygen deficit in natural water and eutrophication of the basin, and depresses the vitality of biological communities.

Combined ecological and geochemical investigations in such local regions are of prime importance for the assessment of the current state and prediction of the development of environmental conditions in the estuary systems of the whole Arctic basin. The abundance and behavior of pollutants, which are the most important parameters of the anthropogenic impact, are controlled by both the location of their sources and the combined influence of a number of geochemical factors, such as the composition of suspended material, redox conditions in sediments, their mineral and grain-size compositions, hydrochemical parameters, and water circulation induced by global and regional currents. A combined approach to the study of anthropogenic pollution of an aquatic environment with the determination of the natural background parameters provides insight into the regularities in the distribution and migration of pollutants in the water basin in the case of the appearance of polluted zones.

This paper presents the results of ecological and geochemical investigations of a local area in the Ob estuary, undertaken in 2002–2003 at sites of hydrocarbon exploration drilling.

REGIONS AND METHODS OF INVESTIGATION

The investigations were carried out on the R/V *Kaliningradets* in the Ob estuary at the Kamennomysskaya and Obskaya 1 sites (Fig. 1) during the beginning and the last stage of drilling and borehole testing, which were performed from the Amazone platform. Experimental data on the background parameters of the Ob estuary and the adjacent areas of the Kara Sea were invoked for interpretation with our results. These parameters were determined by researchers at the Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, during the 2001–2003 expeditions of the R/V *Akademik Boris Petrov* in the framework of the joint Russian–German project "Run-offs of Siberian Rivers and Their Behavior at the Adjacent Part of the Arctic Basin" [1]. The program of investigations included the determination of the main hydrochemical characteristics of the aquatic environment (t, pH, Eh, O_2, etc.) and the analysis of bottom sediment and water samples for various pollutants: petroleum products (PP), surface-active substances (SAS), heavy metals, and radionuclides. The choice of pollutants for analysis was based on the experience of previous work in various areas of the ocean (Gulf of Mexico, North Sea, Norwegian Sea, and Anadyr Bay [2, 3]).

Petroleum products (PP) are among the most abundant and hazardous contaminants in surface waters. Oil and products of its processing are very complex (variable and diverse) mixtures of substances, including low- and high-molecular weight, saturated and unsaturated aliphatic, naphthenic, and aromatic hydrocarbons; oxygen-, nitrogen-, and sulfur-bearing compounds; and unsaturated heterocyclic compounds, such as resins, asphaltenes, anhydrites, and asphaltene acids. In our investigations, the petroleum products were conditionally restricted to the hydrocarbon fraction (aliphatic, aromatic, and alicyclic hydrocarbons). The contents of PP and anionic SAS were measured in concentrates (after chloroform extraction) using a Fluorat-2 fluorometric liquid analyzer [4].

Heavy metals (HM) were analyzed in bottom sediments by X-ray fluorescence on a Spectroscan analyzer [5] and in water samples by atomic emission spectrometry with inductively coupled plasma (ICP AES) after trace element concentration on chelate sorbents with grafted amino carboxyl groups [6].

Anthropogenic radionuclides were analyzed in bottom sediments by the method of low-background gamma-spectrometry on a high-efficiency instrument with a Ge detector [7].

RESULTS AND DISCUSSION

Field work in the Ob estuary was carried out in the summer–fall period. The investigations in the region of exploration drilling included two measurement sessions in each of the two sites, Kamennomysskaya and Obskaya no. 1, at the beginning of drilling operations and at the last stage of drilling and borehole testing; in addition, more than 40 background samples of bottom sediments and water were analyzed for HM, PP, SAS, and natural and artificial radionuclides. The sampling of water and sediments in the region of the drilling operations was carried out along four azimuths (N, S, W, and E) at varying distance from the platform (50, 100, 250, 500, and 1000 m). In order to determine the current background parameters of the environment, samples were additionally taken at a distance of 5000 m (3000 m) from the platform on the windward side.

The identical conditions of formation of the chemical composition of the river in the territory result in a narrow range of hydrochemical parameters. For instance, at the Kamennomysskaya site the water temperature is 16.2°C at the surface and 16.3°C in the near-bottom layer (5–6 m). The oxygen content is 6.45 mg/dm^3 at the surface and 6.48 mg/dm^3 at deep levels. The same parameters for the surface and near-bottom water layers at the Obskaya 1 site are 17.4 and 17.6°C and 7.68 and 7.72 mg/dm^3, respectively.

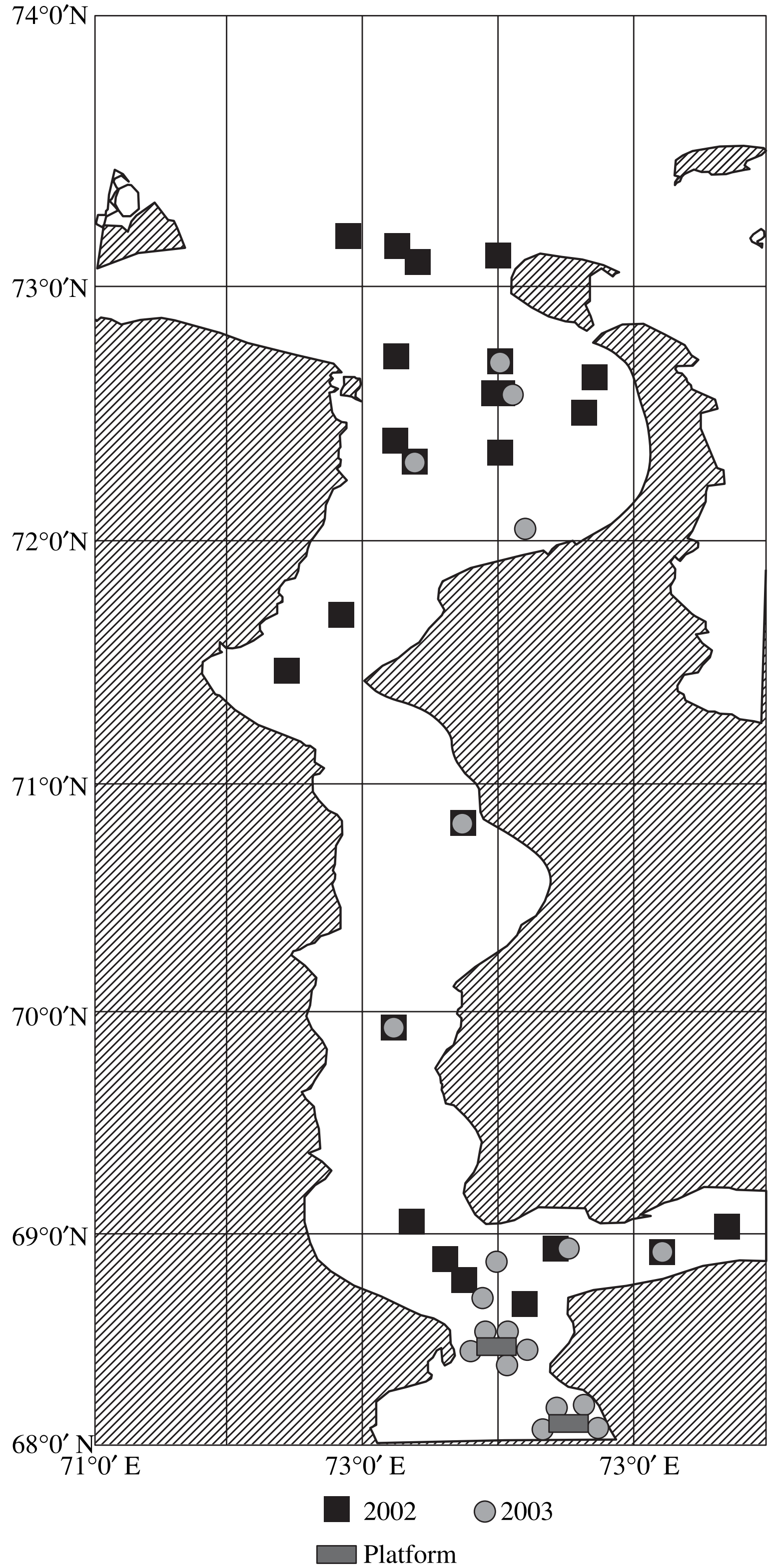

Fig. 1. A sketch map of the region studied.

During the initial period of pollution of the aquatic environment, water masses are more informative, because they can transport elements and compounds in dissolved and suspended forms far from the pollution source at the expense of currents and wind. Bottom deposits are the main long-term indicators of ecological and geochemical conditions in a basin. In order to estimate the general patterns of possible anthropogenic pollution of the water basin during drilling operations, samples of water and bottom sediments were simultaneously collected and analyzed.

The abundance of PP showed the most significant variations in the environment during drilling operations. For instance, the background level of PP in the bottom sediments is about 0.004–0.006 mg/g (data from 2001–2002 before the beginning of the summer-fall period of drilling operations) (Table 1), whereas the PP concentration reached 0.14–0.16 mg/g during drilling at the Obskaya 1 site (Fig. 2). Water samples from the Obskaya 1 site showed PP concentrations no higher than two background values, and the contaminated area extended along the current direction (Fig. 3). More significant increases were observed in the near-bottom water layer, which can be explained by lower water mobility in this zone, especially in the absence of wind waves. It should be noted that the reliability of the interpretation of PP concentration and distribution in bottom sediments and water is largely related to the maintenance of the temperature conditions of sample storage and possible transportation for subsequent analysis in the laboratory. Our investigations of the kinetics of PP and SAS destruction in aqueous solutions at room temperature showed (Fig. 4) that up to 65% PP was decomposed within the first 4–5 days, and their concentration in the solution did not change in the following 4–5 days. Anionic SAS are almost completely decomposed at room temperature already after several days.

Table 1. Contents of petroleum products in the background samples of bottom sediments collected in 2001 and 2002

No.	2001, mg/g	2002, mg/g
1	0.008	0.013
2	0.012	0.010
3	0.011	0.008
4	0.009	0.008
5	0.010	0.010
6	0.006	0.005
7	0.009	0.009
8	0.005	0.004
9	0.013	0.014
10	0.007	0.006
11	0.008	0.012
12	0.012	0.009

Elevated concentrations of HM were also detected. For instance, at Obskaya 1 site, the anomalous zone of HM distribution (for lead) in the bottom sediments was relatively small, and elevated contents were detected at

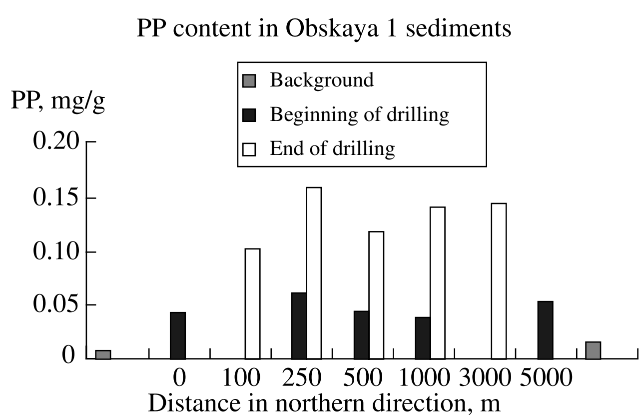

Fig. 2. Distribution of PP in bottom sediments according to the data of fluorometry.

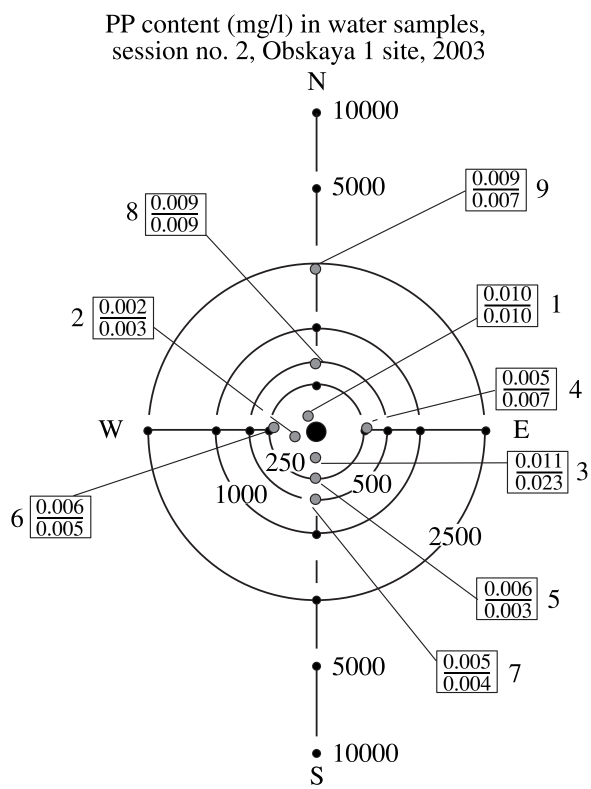

Fig. 3. Concentrations of PP in water samples 1–9 (surface water in the numerator and near-bottom water in the denominator).

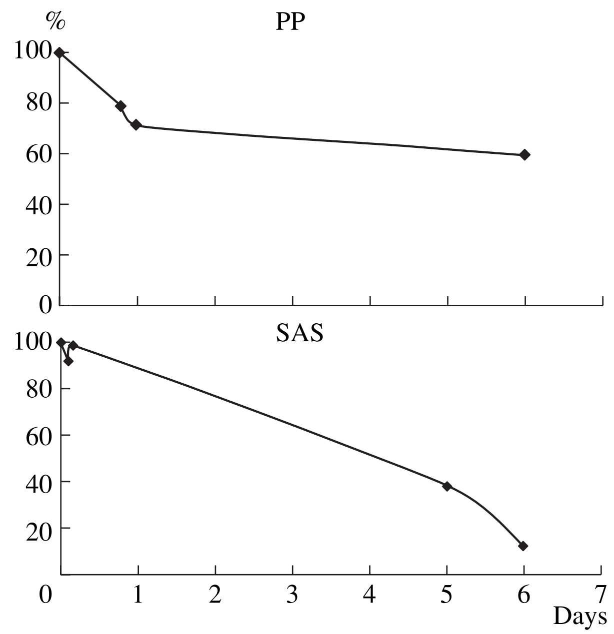

Fig. 4. Behavior of SAS and PP at room temperature.

a distance of up to 100 m from the platform along the current direction (Fig. 5).

Significant deviations from the average content of lead and, to a lesser extent, cadmium were observed in surface water samples. During the initial stage of drilling, elevated concentrations were detected within a radius of 250 m from the platform, and the zone of anomalously high concentrations of these elements extended already for 500 m from the center at the end of operations (Tables 2, 3).

An important problem arising during the investigation of chemical elements in near-bottom waters is the determination of the forms of their occurrence, because the toxicity and biological activity of elements depend not only on their total content but to a greater degree on the proportions of the existing forms. The occurrence forms of HM in the river water and the adjacent portions of the Kara Sea were determined by the method of continuous multistep filtration designed at the Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences [8]. The method is based on the fractionation of water components on membrane filters (ranging from 1 to 0.025 μm in pore size) and the subsequent analysis of the obtained fractions by inductively coupled plasma mass spectrometry. A 400-ml sample was passed through a set-up consisting of cells with a tangential flow of the analyzed liquid relative to a series of membranes with decreasing pore

Table 2. Results of the determination of heavy-metal concentrations in water samples collected at the beginning of drilling operations at the Obskaya 1 site, mg/l

Station coordinate, meters from the drilling platform		Cd	Co	Cu	Fe	Mn	Ni	Pb	Ti	V	Zn
3000 N	surface	0.00002	0.0058	0.0044	0.2381	0.0097	0.0159	0.0058	<0.01	<0.002	0.0509
	bottom	0.00009	0.0021	0.0089	0.2370	0.0100	0.0176	0.0021	<0.01	<0.002	0.0547
1000 N	surface	0.00006	0.0040	0.0063	0.2203	0.0111	0.0164	0.0040	<0.01	0.0038	0.0350
	bottom	0.00001	0.0034	0.0043	0.2198	0.0070	0.0160	0.0037	<0.01	<0.002	0.0551
250 N	surface	0.00011	0.0037	0.0028	0.2094	0.0091	<0.01	0.0119	<0.01	<0.002	0.0418
	bottom	0.00003	0.0026	0.0083	0.2783	0.0272	<0.01	0.0023	<0.01	<0.002	0.0738
100 N	surface	0.00027	0.0032	0.0046	0.2421	0.0127	<0.01	0.0125	<0.01	<0.002	0.0398
	bottom	0.00003	0.0028	0.0087	0.1709	0.0146	<0.01	0.0005	<0.01	<0.002	0.0608
1	surface	0.00016	0.0009	0.0109	0.2268	0.0034	0.0141	0.0114	<0.01	<0.002	0.0783
	bottom	0.00003	0.0005	0.0044	0.2618	0.0097	<0.01	0.0040	<0.01	<0.002	0.0414
250 W	surface	0.00042	0.0038	<0.002	0.2014	0.0061	<0.01	0.0129	<0.01	<0.002	0.0302
	bottom	0.00015	0.0027	0.0058	0.2296	0.0103	<0.01	0.0007	<0.01	0.0021	0.0539
250 O	surface	0.00012	0.0047	0.0046	0.2006	0.0158	<0.01	0.0065	<0.01	<0.002	0.0395
	bottom	0.00010	0.0028	0.0058	0.1765	0.0097	<0.01	0.0090	<0.01	<0.002	0.0487
100 S	surface	0.00008	0.0027	0.0063	0.1533	0.0091	<0.01	0.0090	<0.01	0.0033	0.0348
	bottom	0.00001	0.0027	0.0033	0.2220	0.1614	<0.01	0.0025	<0.01	<0.002	0.0522
250 S	surface	0.00004	<0.01	<0.002	0.2306	0.0091	<0.01	0.0040	<0.01	<0.002	0.0293
	bottom	0.00004	<0.01	0.0053	0.1190	0.0113	0.0158	0.0030	<0.01	<0.002	0.0501
500 S	surface	0.00004	<0.01	0.0038	0.1882	0.0097	<0.01	0.0038	<0.01	<0.002	0.0520
	bottom	0.00009	<0.01	0.0136	0.2832	0.0125	0.0190	0.0030	<0.01	<0.002	0.0462

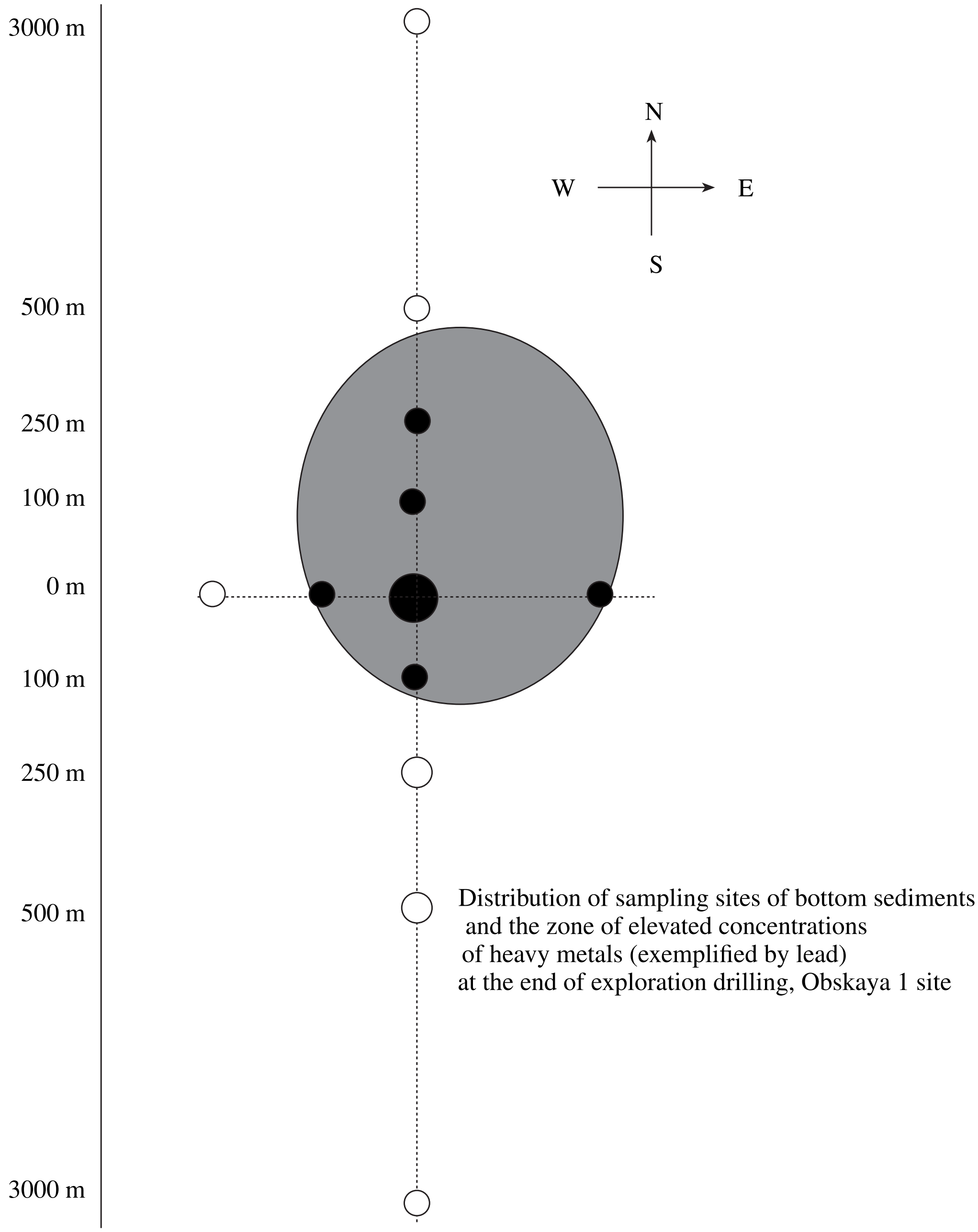

Fig. 5. Outlines of the zone of elevated lead concentration after drilling.

sizes (1.0, 0.45, 0.1, 0.05, and 0.025 μm), in which the liquid phase moved between two filters located one above another. The sample was filtered at a rate of 1.6–2.0 ml/min and a circulation rate of 0.2–0.6 ml/min. After the pumping of the whole sample, it was passed in a closed space for 40 min more. Under such a regime, a five-fold sample volume passed through the set-up, which allows one to maintain equilibrium conditions, when the coefficient of penetration of particles of a given size must be about 1. The separated fractions were accumulated in corresponding chambers, from which liquids were collected by syringes for the subsequent mass spectrometric determination of the concentrations of individual elements. This method provides an opportunity to increase the concentration of a metal by a factor of 40 owing to the separation of water components into smaller volume fractions, which allows reliable investigations of the distribution of trace elements of waters between particles of different sizes even at a very low metal content in the water.

The analysis of water samples by the method of membrane filtration demonstrated that the dissolved form of element occurrence is predominant in the open part of the Kara Sea (Fig. 6). The fraction of suspended

Table 3. Concentrations of heavy metals in water samples collected at the end of drilling operations at the Obskaya 1 site, mg/l

Station coordinate, meters from the drilling platform		Cd	Co	Cu	Fe	Mn	Ni	Pb	Ti	V	Zn
5000 N	surface	–	–	–	–	–	–	–	–	–	–
	bottom	0.00017	0.0008	0.0364	0.2523	0.0236	<0.01	0.0020	<0.01	<0.002	0.0087
1000 N	surface	–	–	–	–	–	–	–	–	–	–
	bottom	0.00035	0.0008	<0.002	0.3159	0.02930	0.0160	0.0085	<0.01	<0.002	0.0354
500 N	surface	0.00008	0.0006	<0.002	0.6954	0.0526	<0.01	0.0180	<0.01	<0.002	0.0682
	bottom	0.00030	0.0009	<0.0114	1.1350	0.0458	0.0231	0.0040	<0.01	<0.002	0.0955
250 N	surface	0.00052	0.0013	0.014	0.05	0.011	<0.01	0.0120	<0.01	0.009	0.046
	bottom	0.00008	0.0003	0.005	0.036	0.007	<0.01	0.0088	<0.01	<0.002	0.083
100 N	surface	0.00020	0.0015	0.0052	0.0603	0.0109	0.0119	0.0124	<0.01	0.0030	0.0530
	bottom	0.00014	0.0005	0.0025	0.0263	0.0039	0.0136	0.00556	<0.01	0.0036	0.0432
50 N	surface	0.00035	0.0010	0.013	0.076	0.012	<0.01	0.0133	<0.01	0.008	0.038
	bottom	0.00011	0.0009	0.012	0.077	0.015	<0.01	0.0066	<0.01	<0.002	0.057
0	surface	0.00024	0.0016	0.01	0.15	<0.001	<0.01	0.0120	<0.01	<0.002	0.01
	bottom	0.00004	0.0007	0.015	0.27	0.012	<0.01	0.0020	<0.01	<0.002	0.036
100 W	surface	0.00006	0.0010	<0.002	0.0587	0.0132	<0.01	0.0176	<0.01	<0.002	0.0440
	bottom	0.00002	0.0001	<0.002	0.0536	0.0068	0.0102	0.0003	<0.01	<0.002	0.0414
50 S	surface	0.0003	0.0013	0.0108	0.0481	0.0152	0.0130	0.0146	<0.01	<0.002	0.0599
	bottom	–	–	–	–	–	–	–	–	–	–
100 S	surface	0.00015	0.0012	0.006	0.033	0.008	<0.01	0.0153	<0.01	<0.002	0.029
	bottom	0.00006	0.0005	0.013	0.17	0.016	<0.01	0.0035	<0.01	0.005	0.079
1000 S	surface	–	–	–	–	–	–	–	–	–	–
	bottom	0.00006	0.0001	0.011	0.048	0.012	<0.01	0.0053	<0.01	<0.002	0.072
5000 S	surface	–	–	–	–	–	–	–	–	–	–
	bottom	0.00008	0.0010	0.01	0.028	0.009	<0.01	0.0060	<0.01	<0.002	0.058

material increased in the samples collected in the Ob estuary at the region of work for many elements, including lead (Fig. 7). No difference was observed in the distribution of trace elements in the background and anomalous samples collected at the area of drilling operation between particles of different sizes, which could be related to the difficulty of registering small effects at very low contents of elements in water.

The concentrations of anthropogenic radionuclides (exemplified by cesium-137) showed no significant variations in the bottom-sediment samples, which in principle could have been expected taking into account

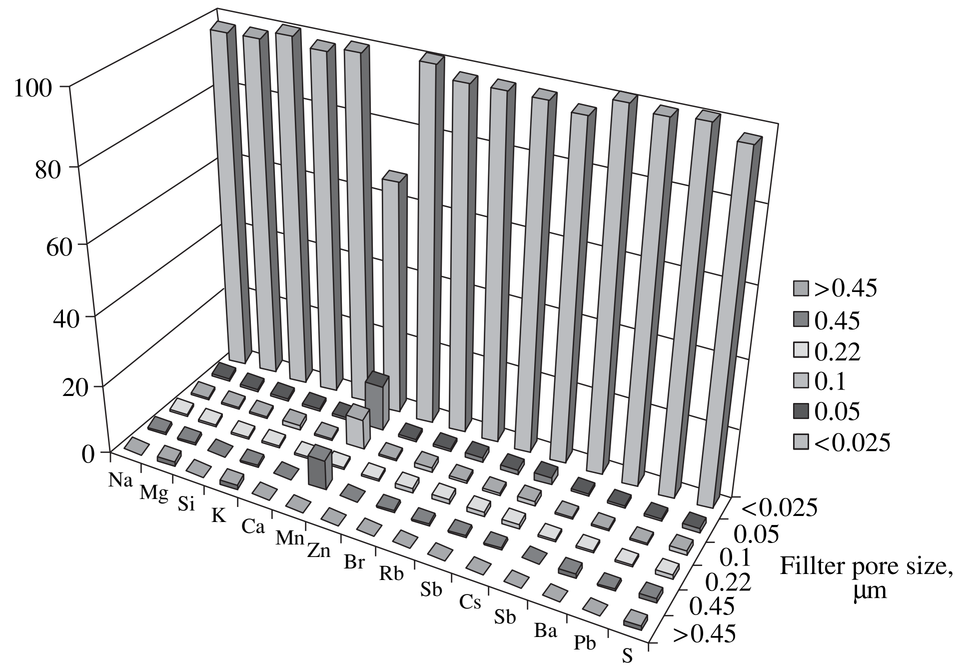

Fig. 6. Forms of heavy-metal occurrence in the open part of the Kara Sea.

that the sediment were roiled owing to the redeposition of silt from the upper interval of boreholes. The level of radiocesium concentration in the bottom-sediment samples collected during drilling operations is comparable with the specific activity of the radionuclide in the bottom sediments sampled long before and after the drilling operations (Fig. 8).

Our investigations provided detailed insight into the scale of anthropogenic pollution in a local region of the aquatic environment at the Kamennomysskaya and Obskaya 1 sites during the drilling operations of 2002 and 2003, and allowed us to estimate the degree of influence of pollutants on the ecological state of the Ob estuary.

In particular, the average PP content in bottom-sediment samples collected before the beginning of drilling operations was about 4–6 mg/kg, which, according to the classification of [9], characterizes them as weakly contaminated. The content of oil hydrocarbons in some samples of bottom sediments of the Ob River collected during drilling at the Obskaya 1 site was 55–205 mg/kg, which classifies them as contaminated. Taking into account that PP contents of 10–100 mg/kg in marine (river) bottom deposits are the minimum values at which biological effects either are lacking or occur as reversible reactions of marine organisms, it is clear that the levels of PP concentration in the bottom sediments of the drilling area at the Obskaya 1 site (Fig. 2) may cause significant impacts on the ecological state of this basin. In the region of the Kamennomysskaya site, the content of PP in sediments is lower (Fig. 9), which is probably related to the fact that the drilling operations at this site are performed on the main course of movement of the Ob River. The content of PP in the water samples is much lower than the maximum admissible concentrations (MAC), which is 0.05 mg/l for lead.

The concentrations of HM in the bottom sediments are close mainly to the average regional values, which obviously pose no hazard for the ecological state of the water basin. The range of specific concentrations in the anomalous sediment samples of even the most important element lead, which is classified as an element of hazard class 1 and the input of which into the environment is related to a large extent to anthropogenic activity, is 25–30 mg/kg, which is only slightly higher than the average values for this region (10–15 mg/kg). During the two seasons of our investigations, these values were relatively stable except for a short-term increase above the average level during the drilling operations (Fig. 10).

It is known that the content of pollutants in the bottom sediments of marine and river basins depends to a large extent on their grain-size and mineral compositions. Bottom sediments containing a significant fraction of pelitic material efficiently accumulate not only HM but also PP [10, 11]. Because of this, the content of pollutants in them may be much higher than in water during the period of contamination. The Gulf of Ob is a typical estuary, which is characterized by a considerable width, length, shallowness, strong wind, and tidal mixing. Bottom areas covered with soft silty deposits are relatively rare, but they are characterized by high contents of various classes of pollutants. The explored

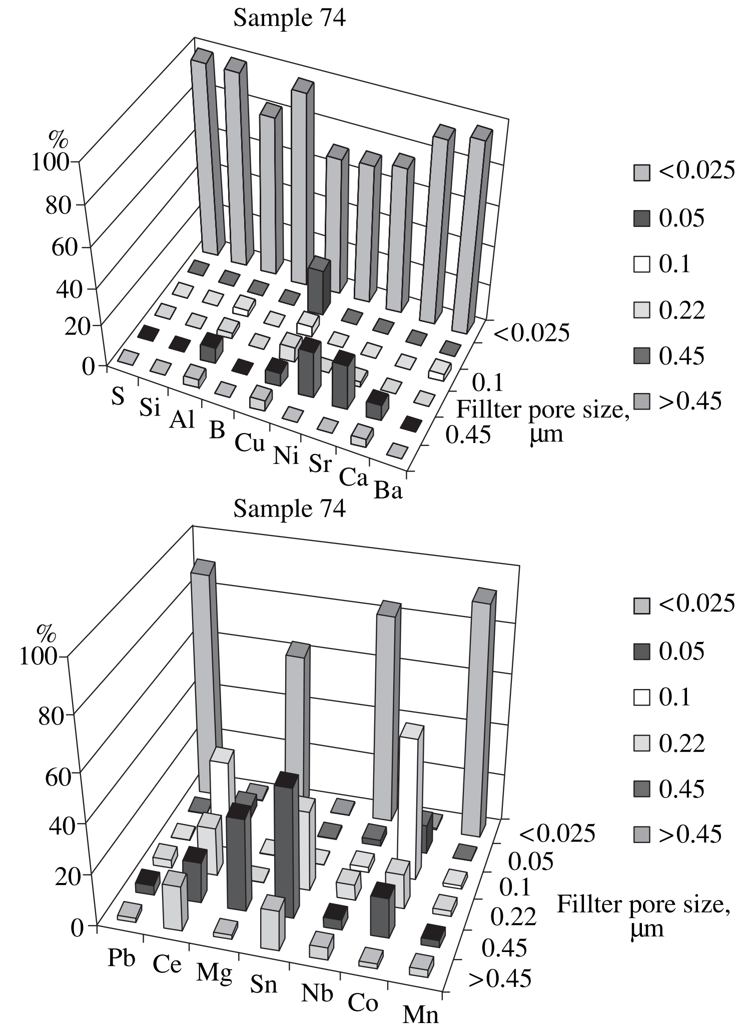

Fig. 7. Forms of heavy metal occurrence in the estuary zone.

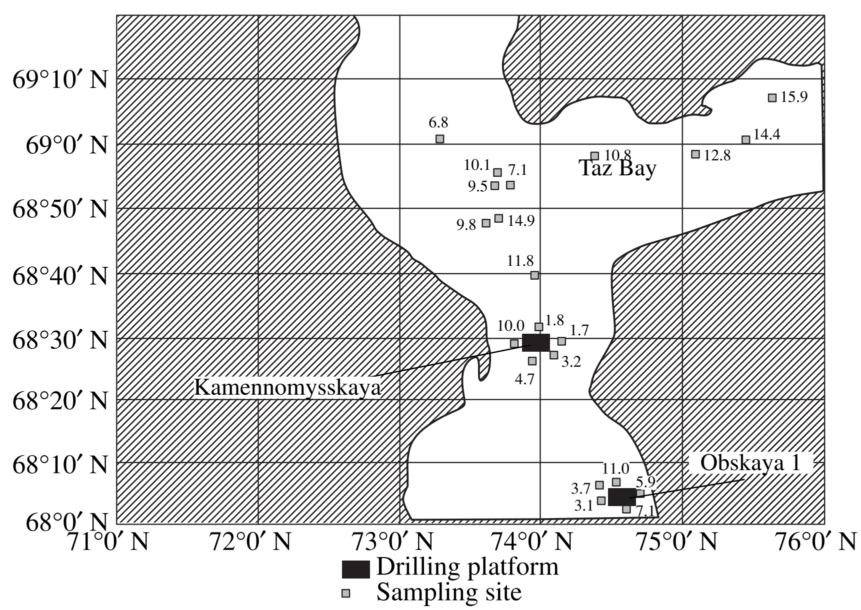

Fig. 8. Distribution of cesium-137 in the bottom sediments from the regions of drilling.

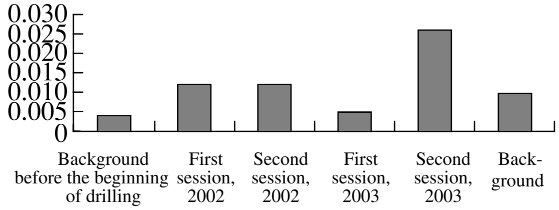

Fig. 9. Variations in PP content (mg/g) in the bottom sediments of the Kamennomysskaya site.

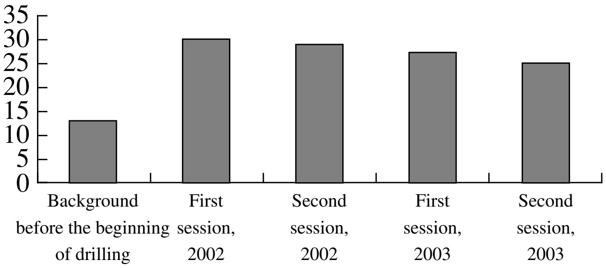

Fig. 10. Variations in lead content (mg/kg) in the bottom sediments of the Kamennomysskaya site.

part of the Gulf of Ob is dominated by sandy ground with variable contents of silty and pelitic fractions. Sediments consisting of silty material with sand grains weakly sorb radionuclides and HM, whereas sediments enriched in pelitic material show high sorption capacity for pollutants. This is illustrated by Table 4, which presents the results on the specific content of radiocesium in samples of different lithologic compositions. These data show that the activity level of cesium-137 is higher in samples with a higher content of pelitic fraction.

An investigation of the rate of sedimentation by the lead method showed that the rates of accumulation of modern sediments for the region are close to the average level for the Ob estuary basin and fall within 0.10–0.13 cm/yr [11]. A more significant source of contamination is the long-distance transport of pollutants with the waters of streams crossing the regions of oil production, entering with the surface and underground runoffs from drilling and processing sites. For instance, the content of PP in the Taz Bay is higher than in the Ob estuary owing to the influence of rivers flowing over the Pur oil and gas producing region and carrying PP into the Taz Bay [12].

CONCLUSIONS

Our investigations of 2002–2003 in the regions of hydrocarbon exploration drilling revealed local areas of chemical pollution (PP and HM) of the environment. Currently, the zones of high concentrations of pollutants occur rather locally and over short time periods. The pollution of water and bottom sediments could be caused by the spills of fuel, oil, and operation solutions. The concentrations of pollutants in anomalous samples were much lower than the MAC values, which is primarily related to the strict maintenance of the main ecological requirements during drilling operations in this region. For instance, during drilling at the Kamennomysskaya and Obskaya 1 sites, all the slime and drilling mud formed during the operation period was collected and, when accumulated in a sufficient amount, given up to a supplementary ship for the subsequent removal and utilization at a special place.

Our investigations showed that the combined approach to the evaluation of ecological conditions in a water basin, which included the monitoring of pollutants and determination of the parameters of natural geochemical background, provided detailed insight into the character of anthropogenic pollution in the region of hydrocarbon exploration drilling at the Obskii and Kamennomysskii prospects in the Ob estuary and allowed us to estimate their influence on the ecological state of the basin of the Ob River and the whole Kara Sea. In geochemical aspects, such investigations based on the use of chemical admixtures and radionuclides as tracers of natural processes can be used to estimate the spatial and temporal scales of the distribution of admixture fields associating with possible accidents.

Table 4. Grain-size composition and activity of cesium-137 in bottom sediments sampled in the Ob estuary in 2002

Station no.	Grain-size (mm) composition, %				Cs-137 activity, Bq/kg
	0.05–0.01	0.01–0.005	0.005–0.001	<0.001	
1 (04)	9.5	9.3	10.9	14.1	1.8 ± 0.3
2 (07)	3.0	5.5	18.9	16.3	5.3 ± 0.7
3 (15)	29.9	13.9	21.3	28.0	7.1 ± 1.0
4 (17)	3.0	36.1	9.6	14.3	4.0 ± 0.6
5 (22)	2.7	14.1	39.4	27.2	9.2 ± 1.2
6 (23)	5.2	21.4	21.2	36.4	7.0 ± 0.8
7 (29)	0.9	10.7	11.7	19.4	2.0 ± 0.4
8 (35)	0.5	15.7	28.0	53.0	4.6 ± 0.6

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