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Radiogeochemical Studies of the Features of Radionuclide Distribution at the Sites of Dumping of Radioactive Wastes of the Novaya Zemlya Archipelago

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Abstract—This paper presents the results of the studies performed from aboard the R/V *Akademik Boris Petrov* of the Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, in 2002–2004 aimed at surveying the sites of dumping of potentially dangerous objects in Stepovoy, Abrosimov, and Tsivol'ka bays of the Kara Sea. These studies coupled the instrumental facilities of acoustic survey with the visual examination of the objects recovered and also included sampling of the bottom sediments and near-bottom water in the vicinity of the objects and away from them. Subsequently, the samples collected were analyzed using direct gamma spectrometry and radiochemical concentrating selected radionuclides. This allowed us to obtain a statistically reliable database on the specific concentrations of radionuclides (cesium, strontium, cobalt, and plutonium) in the water column and in the bottom sediments. In selected parts of Stepovoy and Abrosimov bays, at the sites where containers were located, local areas with contaminated bottom sediments were registered in the immediate vicinity of the objects, in which significant concentrations of Cs-137 were detected. We also carried out experiments with the samples of the collected bottom sediments on the determination of the kinetic parameters of sorption in the sediment–solution system using the method of radioactive indicators. Taking into account the hydrological conditions, the results obtained allow one to explain particular features of radioactivity distribution in selected aquatic areas. Based on a comparison between the results of the studies performed in 1992–1994 and 2002–2004, we estimated the dynamics of the changes in the mean level of specific concentrations of radionuclides in the regions of the location of sunken objects in shallow-water bays of the Novaya Zemlya Archipelago. The registration of elevated concentrations of corrosive elements in the surface layers of the bottom sediments in these zones suggests a necessity of a multidisciplinary study consisting of regular repeated measurements at reference points in different areas aimed at before-the-fact prevention of possible leakages of radioactive substances from the sunken objects, which might result in a significant contamination of the environment.

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INTRODUCTION

The radioecological condition of the aquatic environment of the Arctic Basin is strongly related to the existence of sources of contamination such as the products of the global fall-outs related to nuclear weapon tests, the dumpings of solid and liquid radioactive wastes off the Novaya Zemlya Archipelago, the transboundary transfer of radioactivity from the plants for nuclear waste reprocessing located in Cellafield (Great Britain) and Cape Ahr (France), and the radionuclide runoff by the Ob and Yenisei rivers from the PO Mayak and the Krasnoyarsk Mining and Chemical Industrial Complex.

Domestic and foreign radioecologists suggest that, at present, the sites of dumping of solid radioactive wastes located in shallow-water bays of the Novaya Zemlya Archipelago represent the most dangerous source for technogenous pollution of the northern areas. The total activity of the wastes is estimated at 2.3 mln. curie. From the end of the 1950s to 1992, the Soviet Union had sunken wastes with a total activity of 2.5 mln. curie, including 13 reactors from damaged nuclear submarines (six of them with unremoved nuclear fuel) and three reactors and an assembly with partly unremoved nuclear fuel from the icebreaker *Lenin.* The principal area of dumping was located in the eastern part of the Novaya Zemlya shelf; here, at eight sites, wastes were dumped with an activity comprising 70% of the total activity of the marine wastes dumped by the USSR. The wastes mostly consist of extremely active substances thus representing the greatest ecological hazard [1]. The strength of the protecting cases of the sunken objects, including those with spent nuclear fuel, as well as that of numerous containers with solid wastes causes anxiety and requires regular comprehensive field and laboratory studies on the radioactivity distributions at various dumping sites and elaboration of prognostic estimates concerning the changes of the ecological situation in these areas.

The distribution and behavior of artificial radionuclides, which represent the most important parameters of the anthropogenic impact, are defined both by the localization of their sources and by the combination of geochemical factors such as the composition of the particulate matter in the water column, the redox conditions in the sediments, the mineral and grain-size compositions of the sediments, the hydrochemical parameters, and the hydrodynamics of the water masses. Biogeochemical factors also play an important role in the processes of migration of radionuclides in the sediment–solution system.

The studies of the influence of the characteristics and composition of the sediments on the sorption–desorption processes of various radionuclides at the sites of local contamination recovered are especially interesting for understanding the behavior of radionuclides in the marine environment of different dumping areas. One of the principal parameters that characterize the condition of radionuclides in the sediment–solution system is the coefficient of equilibrium distribution (Kd). The use of this parameter allows one to conclude on a quantitative basis about the features of behavior and migration of individual radionuclides at dumping sites.

In this paper, we present the results of studies performed by us in 2002–2004 in Tsivolka, Stepovoy, and Abrosimov bays and in the adjacent part of the Kara Sea. We paid significant attention to the study of the influence of environmental parameters on the coefficients of radionuclide distribution at specified sites of dumping of radioactive wastes. These studies comprised a part of the research program at the Vernadsky Institute aimed at the studies of the present-day condition of the radioactive pollution of marine environment at the sites of dumping of solid radioactive wastes in shallow-water bays of the Novaya Zemlya Archipelago; selected preliminary results of them were published earlier [2].

MATERIALS AND METHODS

The studies were performed from aboard the R/V *Akademik Boris Petrov* and partly from the R/V *Professor Shtokman* at the sites available to the vessels (with respect to the sea depth) in Stepovoy, Abrosimov, and Tsivolka bays (Fig. 1).

Sampling of the bottom sediments was performed with the use of a box corer and a Niemiste tube. CTD probing of the water column was carried out with a

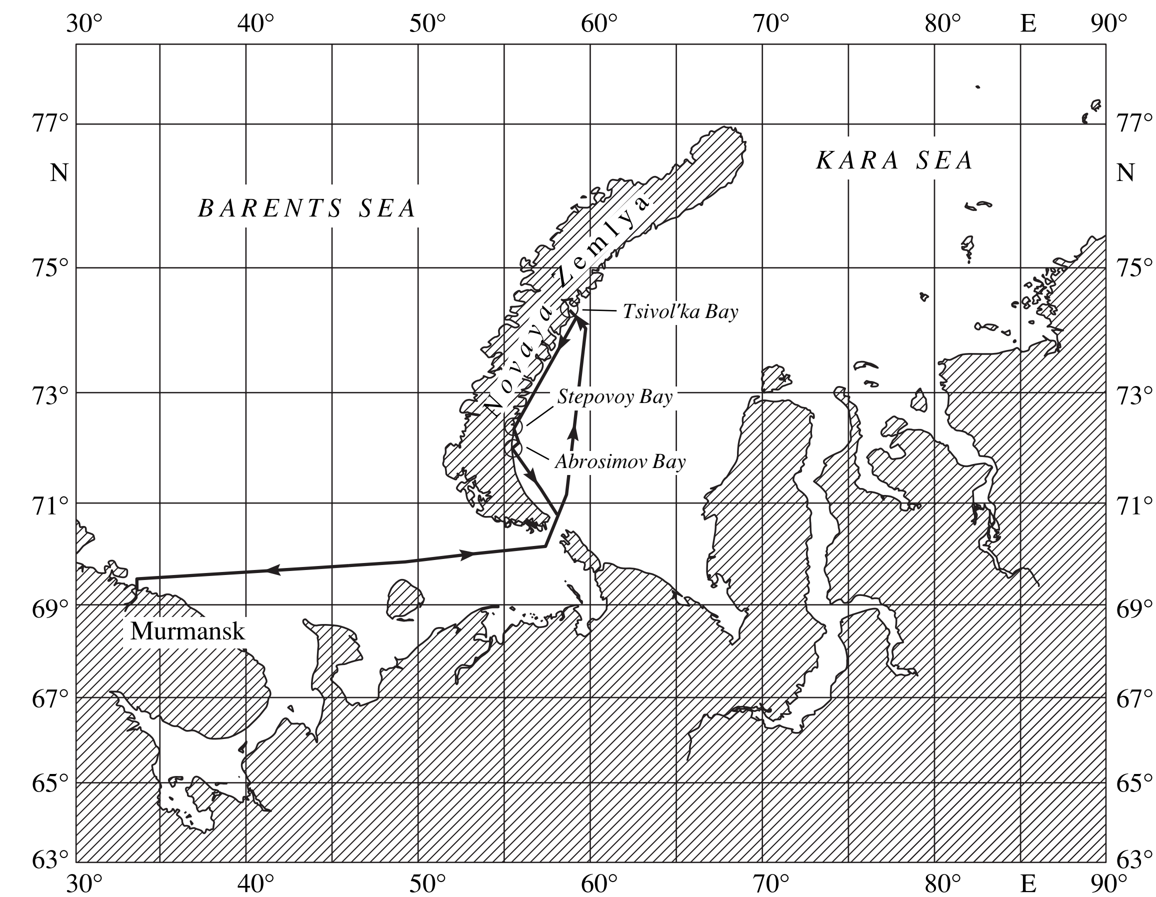

Fig. 1. Map of the study area.

MARK 3B probe equipped with a Rozett set of plastic bottle samplers. Water samples for subsequent radionuclide determination were collected with a Malysh subsurface pump supplied with a PVC hose; samples for hydrochemical examinations were collected with the Rozett bottle samplers.

The hydrochemical studies of the sediments included determinations of pH and Eh followed by extraction of interstitial waters and their hydrochemical analyses (total alkalinity and phosphate concentration); they were performed using fresh samples collected immediately after the sampler lifting on board the vessel.

The determination of gamma-emitting radionuclides in the bottom sediment samples was performed using direct gamma spectrometry with no sample decomposition. Radiochemical method was used to determine the cesium-137, strontium-90, and plutonium-239 and -240 radionuclides in the water samples and isotopes of radiostrontium and radioplutonium in the bottom sediment samples [3]. The specific activity of lead-210 in the sediment cores was determined through measuring the total activity of bismuth-210 with respect to the line $E = 41$ keV with subtraction of the supported lead-210 according to the measurements of the gamma lines of daughter products of decay of radon-226 [4].

Low-background facilities for measuring beta- and gamma-preparations were certified and installed at a special laboratory for radiation monitoring, which was accredited by the Gosstandart of the Russian Federation (Federal Registration no. 41438-2000).

The sorption activity of the bottom sediments with respect to radionuclides was studied in the surface sediments collected in Stepovoy and Abrosimov bays at sites with elevated concentrations of radiocesium. The experiments were performed at a special laboratory on board the R/V *Akademik Boris Petrov* and at laboratories at the Institute.

The experiments were performed in the following way. Cesium-137, strontium-85, and cobalt-60 radionuclides (in selected experiments, americium-241 was used) were introduced into a water sample 200 ml in volume with a salinity of 26‰ (characteristic of the near-bottom water in Stepovoy Bay during the expeditionary studies). The volume of the radionuclide added never exceeded one or two drops and virtually required no correction in the pH value of the solution. The solutions were held over a day in order to reach an isotopic equilibrium; then, a portion of bottom sediments ($V/m = 20$) was inserted. The samples were kept under the conditions of intensive mixing (electric mixer) over a few days; at fixed intervals, aliquote samples were taken. They were separated into different phases, the activity of the liquid phase was measured, and then the coefficient of distribution of the radionuclide between the liquid phase and the sediment was estimated. The aliquote part of the sample was retained in order to determine its grain-size composition.

The species of radionuclides in the sediment were studied using the method of sequential processing by solutions of various reagents. The experiments with the method of sequential extraction may provide information on reversible and irreversible bonds of radionuclides and their potential mobility in ecosystems. The following solutions were used in order to determine different forms: seawater (water-soluble forms), 1 N CH_3COONH_4 (exchangable and easily-soluble forms), 1 N HCl (forms with an insoluble fraction to solution ratio of 1 : 5), and 6 N HCl (acid-soluble forms with a phase ratio of 1 : 2) [5].

RESULTS AND DISCUSSION

Hydrochemical Characteristics of the Bays Studied

In contrast to the open areas of the Kara Sea, the bays of Novaya Zemlya are, to a great extent, protected from the wind and storm action by mountainous coasts and are virtually not subjected to the influence of the runoff of the Ob and Yenisei rivers. In the summertime, in the bays of the Southern Island, the surface seawater layer is episodically desalinated due to the local streams produced by the melting snow cover in the mountains; in the bays of the Northern Island, the effect of desalination is also produced by icebergs.

As one can see from Table 1, a typical two-layered structure of the water column is observed only in depressions of the bottom topography where the salinity can reach 34‰. In the upper part of the water column (layer 20–30 m thick), the changes in the *T*–*S*-para-meters are gradual and the pycnocline is mostly poorly manifested.

Among the three bays studied, Stepovoy Bay is distinguished by the relatively high contents of soluble phosphates in the water, which can be compared to their concentrations in the Kara Sea in the zone of the influence of riverine runoff. These phosphates might be supplied with the fresh waters of the local streams that drain the poorly developed soil cover of the Southern Island. The same streams deliver to the bays the greater part of suspended sedimentary matter consisting of the products of abrasion of ancient sedimentary rocks of the land. In the shallow-water bays of Novaya Zemlya, the bulk of the sedimentary matter is represented by dark gray clayey particles, which have been hydrodynamically sorted from the materials of abrasion of Proterozoic and Paleozoic rocks, including black schists, which had undergone stages of deep katagenesis and metamorphism under the conditions of greenschist and amphibolite facies [6–8]. The organic matter (OM) in these materials is strongly recycled and has actually lost its reducing potential in the course of its geological evolution.

The sufficient amount of nutrients in semi-enclosed bays provides the development and blooming of phytoplankton. In October 2002, we collected net plankton samples from the upper water layer of Stepovoy Bay

Table 1. Hydrochemical characteristics of the water masses in the bays of the Novaya Zemlya Archipelago (August 2003)

Sea depth, m	Sampling depth, m	S, ‰	pH	Oxygen		Alkalinity, mM	PO_4^{3-}, mM
				mg/l	% of saturation		
Stepovoy Bay							
32	0–2	28.4	8.25	10.32	83.2	2.4	0.50
	5	28.7	8.28	10.53	85.2	2.4	0.00
	20	29.9	8.18	10.16	82.2	2.4	0.66
49	0–2	29.5	8.27	11.46	92.1	2.4	0.66
	26	31.4	8.05	–	–	2.6	1.33
	40	33.4	7.91	–	–	2.7	0.83
Tsivol'ka Bay							
61	0–2	26.4	8.30	12.96	96.8	2.2	0.00
	5	28.5	8.28	10.53	83.5	2.3	0.17
	46	31.2	8.24	11.03	87.3	2.6	0.00
	52	33.6	8.04	10.89	80.6	2.6	0.66
65	0–2	29.5	8.26	11.26	91.2	2.4	0.17
	38	31.4	8.23	10.80	85.0	2.6	0.00
	43	33.0	8.12	10.41	79.5	2.6	0.00
32	0–2	15	8.24	12.01	92.8	1.8	0.25
	25	29.6	8.25	10.30	84.4	2.4	0.25
Abrosimov Bay							
35	0–2	12.3	8.28	12.48	93.5	1.2	0.33
	12	29.6	8.30	14.91	93.1	2.4	0.00

and separated refined phytoplankton preparations in order to analyze the isotopic composition of the biomass. The typical marine (oceanic) diatomaceous species *Chaetoceros* and *Thalassionema* and the peridinium species *Ceratium arcticum* dominated. The isotopic composition of the biomass of the preparations ranged from –28.2 to –26.7‰ PDB depending on the species composition of algae; the total isotopic composition of the sources of carbon dissolved in seawater varied from –2.5 to +1.0‰ PDB.

An increase in the phosphate concentrations in the near-bottom waters was also noted; it proceeded against the background of the dynamics of the oxygen concentration and pH values. The consistent trends in the variations of the chemical parameters of the water with depth suggest the proceeding of biogeochemical processes of synthesis and destruction of autochtonous OM; this was best manifested in Stepovoy Bay, where an autumn alga bloom was observed.

The trends of the geochemical parameters over depth registered in the water column were also revealed in the recent bottom sediments. By the example of Stepovoy Bay (2002), we detected the changes in the pH and Eh values and in the nutrient concentrations with depth (Table 2). This fact together with the presence of carbonate–clayey nodules at depths of a few centimeters beneath the surface represents a geochemical indication of the early beginning of the development of the processes of anoxic diagenesis in the sediments of the bay immediately under the thin (2 cm) layer of oxidized sediments. As follows from the data of the measurements performed on board the vessel, already at a depth of less than 7 cm below the floor surface, the sediments feature a reducing regime with Eh = –100 mV. The bacterial destruction of labile OM in the sediments is related to the reduction of sulfates accompanied by an increase in the alkalinity and phosphate concentration in interstitial waters. We argue that the source of labile OM in the sediments of the bay is represented by the

Table 2. Geochemical parameters of the bottom sediments in the aquatic area off the Novaya Zemlya Archipelago (October 2002)

Sampling depth, cm	Moisture content, %	pH	Eh, mV	Alkalinity, mM	PO_4^{3-}, mM
Stepovoy Bay					
Near-bottom water	–	8.12	+373	2.4	2.22
2	63.8	7.24	+380	–	–
2–7				2.6	4.2
7	44.4	7.64	–70	–	–
7–12	–	–	–	3.0	2.4
12	38.5	7.71	–120	–	–
Novozemel'skii Trough, western slope					
Near-bottom water	–	8.08	+445	2.4	1.84
0	68.8	7.31	+530	–	–
2	–	7.19	+530	–	–
7	–56.5	7.40	+470	2.7	1.6
12	53.5	7.90	+120	–	1.6
20	51.4	7.74	+50	3.1	5.6
30	45.6	7.71	0	–	–
35	46.7	7.68	–20	3.2	0.0

autochtonous biomass synthesized by phytoplankton. The bays of the Southern Island are characterized by conditions favorable for phytoplankton growth and blooming such as the relatively calm regime of the water mass and the sufficient level of nutrients and irradiation.

For the sake of comparison, in Table 2 we present corresponding data on the sediment core taken in the open sea at the latitude of Stepovoy Bay from the western slope of the Novozemel'skii Trough at a sea depth of 154 cm. Earlier [9] we noted that, in this part of the Kara Sea, strongly oxidized sediments are developed with an upper dark brown crust of oxidized clayey silts up to 20 cm thick. In the sediment core studied, a negative value of Eh = –20 mV was encountered at a depth of 35 cm. In the oxidized sediments of the Novozemel'skii Trough, the zone of reducing diagenesis accompanied by hydrogen sulfide release and appearance of dark-colored reduced sulfur compounds (monosulfides, hydrotroilite) in the sediment is located significantly lower than in the more shallow-water semi-enclosed bay. The reducing conditions in the upper layers of the sediments in the bay may be regarded as the manifestation of a more aggressive environment capable of initiating processes of corrosion of metallic protective screens in the regions of nuclear dumping and possible processes of mobilization of radiocesium from the bottom sediments.

Radionuclide Distribution in the Sediment–Solution System

The results of the recent studies on radionuclide determination in the bottom sediments of the bays considered are presented in Table 3. An analysis of the data of the studies of 2002–2004 showed that, in Tsivol'ka Bay, at the site of burying of a reactor unit of the nuclear icebreaker *Lenin*, no supernormal values of specific concentrations of individual radionuclides exceeding their mean background were observed. In Stepovoy Bay, in the region of sink of a nuclear submarine with spent nuclear fuel (outer part of the bay), we did not found any significant excess of the specific radionuclide concentrations observed over their background concentrations. These facts prove the reliability of the protective screens used on the objects with the nuclear fuel studied.

Meanwhile, in Stepovoy and Abrosimov bays, in the immediate vicinity of the sunken containers, we recovered areas with significant contamination of bottom sediments. In the samples of the surface sediments, significant cesium-137 contents were detected and in a selected area of Stepovoy Bay, in addition to cesium-137, the presence of cobalt-60 was also registered.

The samples from the sites of local contamination of marine environment were subjected to a more detailed examination in order to study the parameters of the

Table 3. Results of determination of specific concentrations of radionuclides in the bottom sediments (R/V *Professor Shtokman*, 2004)

Station no.	Date	Time	Cesium-137 activity in the layer 0–2 cm, Bq/kg	Cesium-137 activity in the layer 2–4 cm, Bq/kg
Tsivol'ka Bay				
1	07.09.04	18:24	8 ± 1	10.0 ± 1.5
2	07.09.04	19:20	11 ± 2	19.0 ± 1.6
3	09.09.04	11:01	8.8 ± 1.7	17.0 ± 1.7
4	09.09.04	12:36	8.7 ± 1.5	
5	12.09.04	16:51	5.3 ± 1.6	5.7 ± 1.6
6	13.09.04	09:06	2.8 ± 1.2	–
7	13.09.04	10:02	1.9 ± 1.0	–
8	13.09.04	10:19	2.3 ± 1.0	–
9	13.09.04	10:39	2.0 ± 1.0	–
10	13.09.04	14:49	5.0 ± 1.5	–
Stepovoy Bay				
1	15.09.04	14:30	58 ± 9	–
2	15.09.04	15:48	687 ± 48	681 ± 52
3	16.09.04	14:30	284 ± 31	–
4	16.09.04	15:20	16.5 ± 4.0	–
5	17.09.04	16:10	15.3 ± 1.8	–
6	17.09.04	16:25	12 ± 2	–
Abrosimov Bay				
1	19.09.04	15.20	8 ± 2	–
2	19.09.04	16.20	6.8 ± 2.6	6.3 ± 1.8
3	19.09.04	16:37	19 ± 3	28 ± 4
4	19.09.04	17:40	26 ± 5	41 ± 5
5	19.09.04	17:57	26 ± 7	–
6	21.09.04	15:12	24 ± 6	41 ± 5
7	21.09.04	16:29	27 ± 2	52 ± 4
8	21.09.04	16:48	27 ± 5	33 ± 5
9	21.09.04	17:07	10 ± 2.4	–

sorption activity of the bottom sediments with respect to radionuclides.

The principal parameters that define the sorption properties of the sediments with respect to various radionuclides are the mineral and grain-size compositions of the sample. It is known that, at significant contents of clayey particles in the sample, bottom sediments are active accumulators of radionuclides and heavy metals [10, 11]. Therefore, in the periods of contamination, the contents of pollutants in the sediments may significantly exceed their concentration in the water. We confirmed the direct dependence of the activity of ^{137}Cs in the upper layer of the bottom sediments on the content of the clayey fraction in the sample [11]. The results of determination of the grain-size composition of the sediments from Stepovoy and Abrosimov bays under consideration are listed in Table 4.

We noted a certain distinction in the grain-size compositions of the two bays: in the sediments of Abrosimov Bay, large particles (from 1 to 0.01 mm) dominate (40–50%), while in Stepovoy Bay, the proportion of these particles comprises 22–32%. In the sediments of Stepovoy Bay, clayey particles with smaller sizes (from 0.01 to 0.001 mm) prevail.

We plotted kinetic profiles of the coefficients of distribution for principal radionuclides; they show that the equilibrium is reached rather quickly in less than two days (Fig. 2). The averaged values of the coefficients of equilibrium distribution for the radionuclides under study (at a salinity of the solution of 26‰) are presented in Table 5. These results agree well with the results previously obtained in Stepovoy and Abrosimov bays by other researchers [12] and lie within the limits prescribed by the IAEA regulations for near-shore aquatic areas [13].

It should be noted that, in Stepovoy and Abrosimov bays, the values of the distribution coefficients for kinetic profiles of radionuclide absorption (by the example of ^{137}Cs) are significantly different (Fig. 3). The experiments with ^{137}Cs at a salinity of 34‰ showed that, in the salinity interval 26–32‰, salinity variations only slightly influence the changes in the absolute values of the equilibrium distribution coefficient; the differences observed in the values of the equilibrium *K*d may be related, first, to the different lithological com-

Table 4. Grain-size composition (%) of the upper layer (0–2 cm) of the bottom sediments

Ordinal no. (station no.)	Fraction (mm) content, %					
	1–0.25	0.25–0.05	0.05–0.01	0.01–0.005	0.005–0.001	<0.001
1 (No 1)	1.17	6.40	16.28	34.21	24.43	17.51
2 (No 3)	1.66	8.83	22.98	13.71	35.68	17.14
3 (No 4)	0.64	10.88	43.03	13.53	19.40	12.52
4 (No 7)	1.60	0.10	41.00	12.80	26.42	18.08

Note: 1, 2—Stepovoy Bay (Sample nos. 1, 3; see Table 3); 3, 4—Abrosimov Bay (Sample nos. 4, 7; see Table 3).

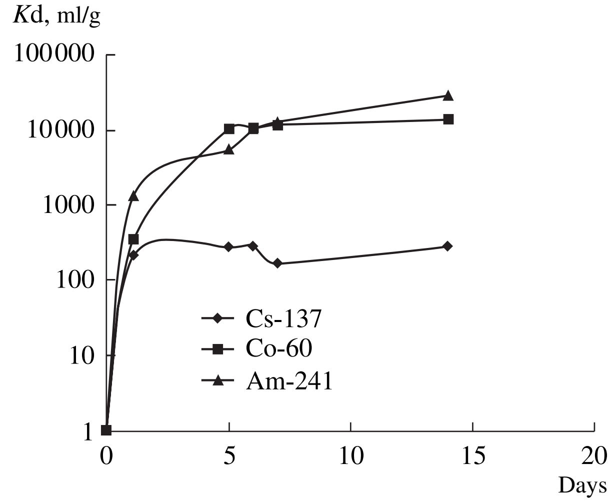

Fig. 2. Kinetic curves of the radionuclide distribution coefficients.

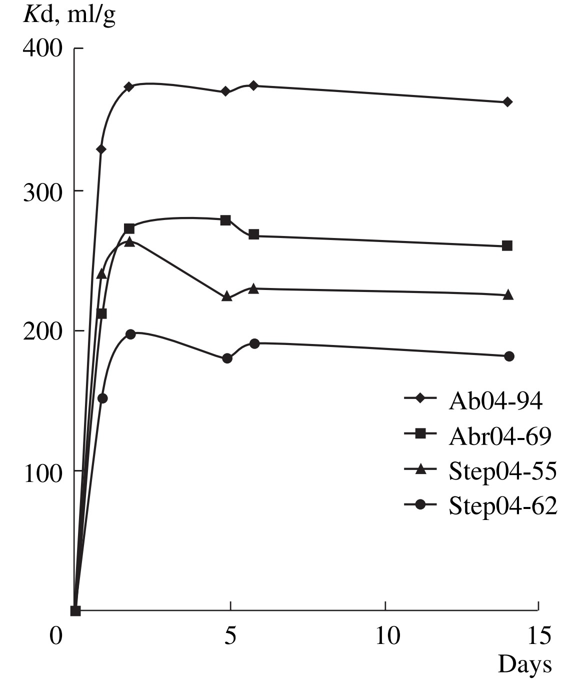

Fig. 3. Kinetic curves of the Cs-137 distribution coefficients in the sediments of Stepovoy and Abrosimov bays.

position of the samples. Since the greater part of the cesium activity is related to the middle grain-size fraction of the sediments, it is evident that, in this case, the differences in the lithological composition of the sediments of the two bays considered represent the most important factor affecting the equilibrium distribution coefficient. In contrast to other radionuclides, cesium-137 not only participates in the ion exchange and sorption interactions with clayey particles but is also capable of selective penetration into interlayer spaces of layer clay minerals such as illite, smectite, and montmorillonite. The positions of cesium inside crystals are very sustainable; therefore, the sediments that contain mica minerals represent traps for cesium-137. The observed difference in the values of the equilibrium distribution coefficient in the sediments from Stepovoy and Abrosimov bays may suggest that, at the sites of dumping, all other factors being the same, the distances of propagation of portions of cesium-137 from the source of radioactive contamination in Stepovoy Bay are greater than those in Abrosimov Bay.

Laboratory experiments on desorption of radionuclides from the bottom sediments using various solutions (Fig. 4) show that, for cobalt-60, easily soluble mobile forms are mostly characteristic, while the greater part of cesium-137 occurs in an acid-soluble form or in insoluble residuum; this is confirmed by the results of sorption experiments on binding cesium-137 in the bottom sediments.

Table 5. Mean values of the coefficients of distribution of different radionuclides

No	Cs-137	Co-60	Am-241	Sr-85
Stepovoy Bay	2.2×10^2	1.4×10^4	2.1×10^4	1.8
Abrosimov Bay	3.1×10^2	3.9×10^3	1.9×10^4	2.5

An assessment of the data on the cesium-137 activity measurements performed in all the sediment samples from the two upper layers (0–2 and 2–4 cm) shows that, in virtually all the samples, the specific radioactivity of radiocesium in the lower layer is higher than that in the surface layer (Fig. 5). The estimates of the sedimentation rate, which were obtained as a result of an analysis of the sediment cores collected from noncontaminated sites of all the three bays using the lead method, show no noticeable differences and lie within the limits from 1.0 to 1.2 mm/yr. The vertical distributions of cesium-137 in the cores from Stepovoy and Abrosimov bays feature a single significant peak; its temporal parameters relate it to the period of mass weapon nuclear tests [14, 15]. Based on these data, one can suggest that there was no significant supply of radioactive contamination from the sunken containers to the upper layer of the bottom sediments formed over 20–30 years after the waste dumping.

As to the water column, elevated values of specific activity of cesium-137 were registered only in the near-bottom water layer of Stepovoy Bay at one of the sites of the waste dumping (Table 6).

An analysis of the data obtained together with the results of the sorption experiments allows us to suggest that these anomalous regions might be related to a partial radioactivity transfer from the solid phase to the water owing to diffusion processes at bottom sediment disturbing in a shallow-water bay or due to possible

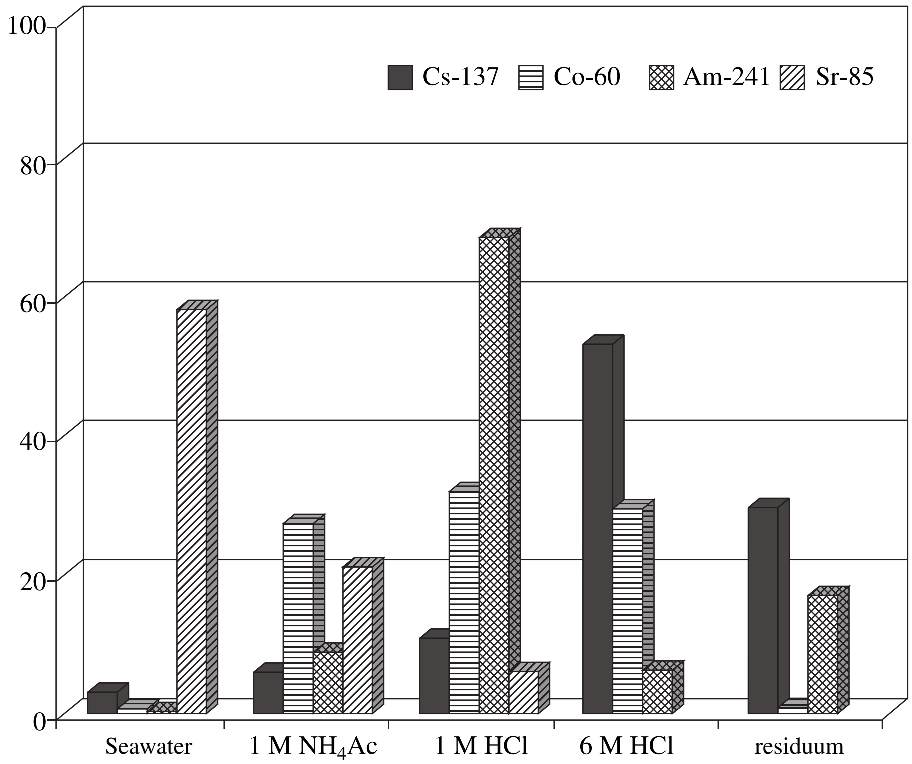

Fig. 4. Forms of occurrence of radionuclides (percentage of the total content in the sample).

variations in the geochemical parameters of the environment. Alternatively, it also seems probable that fractures or more serious damages have appeared in selected containers, which might result in a radioactivity exchange in the solid waste–water system. The hydrological conditions in the inner part of Stepovoy Bay, namely, the existence of two water layers with different hydrophysical characteristics of the water column, prevent the deep waters of different parts of the bay from exchange and result in localization of the contaminated deep waters in the inner part of the bay. The results obtained on the cesium-137 activity in the surface waters show no signs of radionuclide supply to Stepovoy Bay with local water streams from the archipelago.

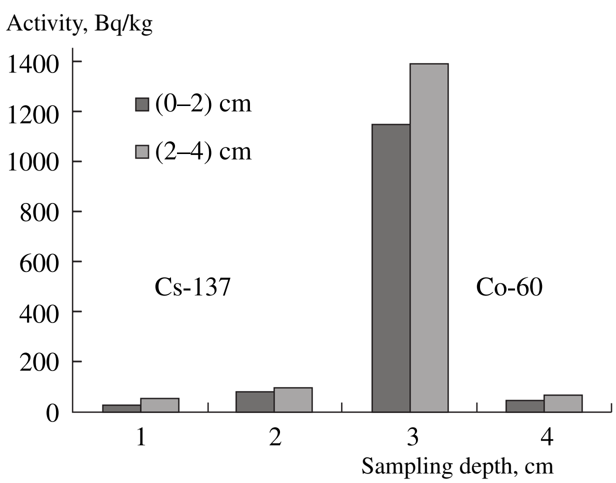

Fig. 5. Contents of Cs-137 and Co-60 in the surface layers of the sediments (0–2 and 2–4 cm): 1, 2, 3—Cs-137; 4–Co-60).

We performed a comparison of the averaged data obtained by us on the contents of various radionuclides in the upper layer of the bottom sediments at the sites of dumping of solid radioactive wastes in Stepovoy and Abrosimov bays with the results of previous surveys performed by the NPO Taifun of the Russian Hydrometeorological Agency together with Norwegian scientists in the same areas in 1992–1994 [16]. The results of this comparison, though rather approximate, allow us to reveal tendencies in the radioactivity changes in the areas under consideration over the past decade (Table 7).

As one can see from the data presented in Table 7, we observe a decrease in the total radionuclide activity in the upper layer of the bottom sediments. One of the reasons of the decrease in the radionuclide activity is their relatively short half-life period, especially that of cobalt-60. Another, and probably more important, reason of the activity drop may lie in the absence of new delivery of contaminants to the upper layer of the bottom sediments during the recent years.

Meanwhile, during the expeditions of 2002–2004, at the sites of the container location, elevated concentra-

Table 6. Averaged values of cesium-137 activity in the near-bottom water layer of the bays under consideration (at the sites of local contamination of the sediments)

Ordinal no.	Number of samples	Bay	Specific activity of cesium-137, Bq/m^3
1	10	Tsivol'ka Bay	3.3 ± 0.9
2	24	Stepovoy Bay	39 ± 14
3	16	Abrosimov Bay	4.2 ± 1.8

Table 7. Changes in the total radioactivity of the upper layers of the bottom sediments with time (Bq/kg)

Period of studies	Radionuclide	Stepovoy Bay	Abrosimov Bay
1992–1994	Cesium-137	260–5450	200–8400
1992–1994	Cobalt-60	31–3150	1–50
2002–2004	Cesium-137	20–1800	5–44
2002–2004	Cobalt-60	1–26	1–5

tions of corrosive elements in the upper layers of the bottom sediments were registered [2]. This implies the proceeding of the process of corrosion of protecting barriers and generates a need for more comprehensive studies in order to more correctly estimate this process.

CONCLUSIONS

In the opinion of the ecologists, at present, solid radioactive wastes buried in the shallow-water bays of the Novaya Zemlya Archipelago represent the most important and dangerous sources of contamination of the environment of the Arctic region. Generally, the aftereffects of the possible release of radioactive wastes into the seawater because of the corrosion of protective barriers of the buried objects (containers, nuclear submarines, reactor units of the icebreaker *Lenin*, and others) may be compared to those of the Chernobyl' accident of 1986. The corresponding line of research implies carrying out wide-scale scientific studies of the physicochemical processes and behavior of radionuclides and corrosion chemical elements in the water–bottom system and formulation of justified forecasts of the ecological conditions of the environment for a long-term period. In parallel, fundamental studies with the use of up-to-date technical facilities for analyses of the horizontal and vertical structures of the bottom sediments and the water column should also be performed. Selected initial results in this field are presented in this paper.

Indeed, since the sedimentary matter in the bays of Novaya Zemlya Archipelago is mostly represented by the products of abrasion of ancient metasedimentary rocks containing strongly recycled OM, the processes of reducing diagenesis in the sediments might develop only within the limits provided by the autochtonous biological production. The development of reducing processes in the sediments, which proceeds at a necessary participation of labile OM, shifts upward the position of the geochemical barrier related to the zone where the parameters pH and Eh change and many elements that occur in bound condition in the sediments acquire migration ability. For example, in the case of radiocesium, the development of reducing diagenesis may result in demobilization of this radionuclide from the bottom sediments owing to its replacement by the ammonium ion, which is a product of microbial destruction of labile OM.

The studies of 2002–2004 show that, in the aquatic areas of Stepovoy and Abrosimov bays, at the sites of location of buried containers, several zones with a significant increase in the radioactivity (with respect to cesium-137) in the bottom sediments and water column (the latter is true for Stepovoy Bay) were registered. The drop in the total activity in the upper layer of the sediments and the decrease in the specific concentration of radiocesium may indicate the absence of noticeable leakages of radioactivity from sunken objects that might result in a significant contamination of the environment.

The general pattern of the radioecological condition of the environment in the aquatic areas of Stepovoy and Abrosimov bays is rather clear; meanwhile, additional studies are required to analyze the situation in other bays. For example, in Tsivol'ka Bay, elevated contents of cesium-137 in the bottom sediments and near-bottom water layer were registered at selected sites, against the background of the *T*–*S*-diagram monotonously changing with depth. In Oga Bay, during recent years, no studies with the use of modern hydroacoustic and contact methods have been performed.

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REFERENCES

1. *Facts and Problems Related to the Burial of Radioactive Wastes in the Seas Surrounding the Territory of the Rus-*

sian Federation (Administr. Prez. Ros. Fed., Moscow, 1993) [in Russian].

2. E. M. Galimov, N. P. Laverov, O. V. Stepanets, and M. V. Vladimirov, "Radiogeochemical Investigations of 2002 in Shallow Bays of the Novaya Zemlya Archipelago," Geokhimiya, No. 1, 3–14 (2004) [Geochem. Int. **42**, 1–10 (2004)].
3. O. V. Stepanets, A. P. Borisov, N. Yu. Kremlyakova, et al., "Application of Artificial Radionuclides for the Geochemical Investigations of the Ob and Yenisei Estuaries and Adjacent Regions of the Kara Sea," Geokhimiya, No. 11, 1091–1098 (1998) [Geochem. Int. **36**, 983–990 (1998)].
4. B. Hagedorn, S. Harwart, M. M. R. van der Loeff, and M. Melles, "Lead-210 Dating and Heavy Metal Concentration in Recent Sediments of Lama Lake (Norilsk Area, Siberia)," in *Land–Ocean Systems in the Arctic* (Springer, Berlin, 1999), pp. 361–376.
5. F. I. Pavlotskaya, *Migration of Radioactive Products of Global Falls in Soils* (Atomizdat, Moscow, 1974) [in Russian].
6. B. A. Klubov and E. A. Korago, "Nature of Liquid Bitumens of the Northern Novaya Zemlya Archipelago," Dokl. Akad. Nauk **315**, 925–928 (1990).
7. E. N. Korago, G. N. Kovaleva, B. G. Lopatin, and V. V. Orgo, "The Precambrian Rocks of Novaya Zemlya," Geol. Soc. Mem., No. 30, 135–143 (2005).
8. N. N. Dunaev, O. V. Levchenko, L. R. Merklin, and Yu. A. Pavlidias, "Novaya Zemlya Shelf in the Late Quaternary," Okeanologiya **35**, 440–450 (1995).
9. E. M. Galimov, N. P. Laverov, O. V. Stepanets, and L. A. Kodina, "Preliminary Results of Ecological and Geochemical Investigations of the Russian Arctic Seas (Data obtained from Cruise of the R/V *Akademik Boris Petrov*)," Geokhimiya, No. 7, 579–597 (1996) [Geochem. Int. **34**, 521–538 (1996)].
10. G. I. Ivanov, *Methodology and Results of Ecogeochemical Studies in the Barents Sea* (VNIIOkeangeologiya, St. Petersburg, 2002) [in Russian].
11. O. V. Stepanets, A. P. Borisov, V. M. Komarevskii, et al., "Radiogeochemical Studies of the Aqueous Environment in the Central Part of the Kara Sea and Ob and Yenisei Estuaries in 1999–2000," Geoekologiya, No. 5, 415–421 (2004).
12. J. Carroll, F. Boisson, S. W. Fowler, et al., "Radionuclide Adsorption to Sediments from Nuclear Waste Dumping Sites in the Kara Sea," Mar. Poll. Bull. **35** (7), 296–304 (1997).
13. *Sediment Kds and Concentration Factors for Radionuclides in the Marine Environment*, IAEA Techn. Rep., No. 247, (1985).
14. H. W. Jeter, "Determining the Ages of Recent Sediments Using Measurement of Trace Radioactivity," Terra Aqua **78**, 1–11 (2000).
15. O. V. Stepanets, A. P. Borisov, A. N. Ligaev, et al., "Estimation of the Sedimentation Rate for Modern Sediments of the Kara Sea: Evidence from Radioactive Tracers," Geokhimiya, No. 7, 752–761 (2001) [Geochem. Int. **39**, 683–691 (2001)].
16. P. Strand, A. I. Nikitin, B. Lind, et al., *Dumping of Radioactive Waste and Radioactive Contamination in the Kara Sea. Results from 3 Years of Investigations (1992–1994) Performed by the Joint Norwegian–Russian Expert Group for the Investigations of Radioactive Contamination of the Northern Areas* (NRPA, Osteras, 1997).