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Radionuclides Distribution and its Sorption Behavior in the Surface Layer of the Kara Sea Bottom Sediments

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Abstract—The sorption characteristics of the Kara sea bottom sediments were investigated. The sediments were collected during scientific expedition of the R/V *Akademik Mstislav Keldysh* (Cruise AMK-66). The mineral and fraction composition of the sediments was determined. It has been found that ^{137}Cs is sorbed mainly by an ion exchange mechanism because of the clay minerals presence in sediments. At the same time, ^{243}Am and ^{90}Sr sorbed from seawater by the surface complexation mechanism. It has been established that the sorption kinetics of ^{243}Am, ^{137}Cs, and ^{90}Sr is about 20 hours, 2 hours, and less than an hour, respectively. The Cs and Am sorption isotherms in experiments with sea and deionized water are described by the Henry equation. The Sr behavior in the deionized and sea water is described by the Langmuir and Freindlikh equations, respectively.

Keywords: AMK-66, bottom sediments, Novaya Zemlya, Sedova bay, radionuclides sorption, ^{137}Cs, ^{90}Sr, ^{243}Am
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INTRODUCTION

Solid radioactive wastes (SRW) have been disposed in several bays of the Novaya Zemlya archipelago since 1950s to 1992 (Bilashenko, 2013). The volume of dangerous dumped wastes, including dangerous reactors of nuclear submarines and Lenin nuclear ice-breaker with nuclear fuel containers that were not completely released before burial, accounts for 70 vol % of all Soviet marine dumps (Stepanets, 2010). The disposal sites are located in the Novaya Zemlya bays of the Kara Sea, including the Techenii, Tsivol'ki, Abrosimova, Stepovogo, Oga, Blagopoluchiya, and Sedova bays, and deep-water area of the Eastern Novaya Zemlya trench. This is confirmed by the previously published studies, which revealed the presence of highly active radionuclides in the bottom sediments and soils of Novaya Zemlya (Volosov et al., 2011; Aliev, 2006; Stepanets et al., 2010; Leverov et al., 2016; Miroshnikov, 2017). This is related to the previous burial of SRW and dumps of technological wastes of nuclear enterprises in riverine systems. Therefore, it is required to estimate the sorption properties of bottom sediments and soils in the coastal Arctic zones, which could be involved in a zone of potential pollution during an emergency situation.

The problem of retention of radionuclides by bottom sediments of the Arctic seas has received interest since the end of the 20th century. The sorption and desorption of ^{137}Cs and ^{90}Sr were experimentally studied in the bottom sediments of the Kara and Barents seas. Radionuclide immobilization was regarded as a new potential source of the Arctic pollution. It was shown that cesium is strongly immobilized on the surface of clay minerals of bottom sediments sampled near the Novaya Zemlya coast, that the Cs sorption is mainly controlled by ion exchange mechanism, and the partition coefficients (*Kd*) for fresh water are two orders higher than those of sea water. This indicates the high probability of radionuclide mobilization from bottom sediments when fresh water enters the ecosystem (Oughton, 1997; Borretzen, 2000; 2002).

The radioactivity of the bottom sediments of the Kara Sea has been studied for a long time by Russian and foreign scientists. The Abrosimova, Stepovogo, and Tsivol'ki bays were studied in detail (Kvasha, 2001; Sarkisov, 2011; Gwynn, 2015; Laverov et al., 2016; Usacheva et al., 2016). At the same time, Sedova Bay has been poorly studied up to now. Thereby, the activity of SRW disposed in it exceeds the total activity of SRW containers buried in these three relatively well studied bays: 1100 containers with nuclear and radioactively dangerous wastes with activity estimated at 111.8 TBq were disposed in Sedova Bay (Sivintsev, 2005).

The aim of this work was to establish the peculiarities in localization of ^{137}Cs pollution of surface sediments taken during cruises of the R/V *Mstislav Keldysh* in 2015–2016, to determine their component composition and specific activities of ^{137}Cs and ^{90}Sr, and to estimate the sediments sorption potential of Sedova bay (Novaya Zemlya) relative to the Cs(I), Sr(II), and Am(III) radionuclides.

EXPERIMENTAL

Marine sediments were collected during Cruise of the R/V *Akademik Mstislav Keldysh* using geological equipment (box corer and bottom grab) and were prepared for further on-board studies. Sample preparation included drying, weighing, and homogenization.

^{137}Cs was determined on the Canberra GR 3818 gamma spectrometer. After preliminary heating of bottom sediment aliquot (450°C, 8 hours) and dissolution in 8 M HNO_3 ^{90}Sr was extracted and concentrated on an SR Resin (Triskem Int.). Then, it was coprecipitated with $Ca_3(PO_4)_2$, and its activity was determined and calculated.

The quantitative mineral analysis of sampled marine sediments was carried out with X-ray powder diffraction using the Rietveld method (Bish and Post, 1989; Doebelin et al., 2015; Krupskaya and Zakusin, 2017) on an X-ray Ultima-IV Rigaku diffractometer (Cu-K_α, D/Tex-Ultra detector, 3°–65° 2θ scanning area).

Sorption experiments were carried out for a bottom sediment sample, which was taken from the upper horizon (0–2 cm) in station 5374 core and preliminarily analyzed. The amount of organic carbon was determined using Tyurin's method (Vorob'eva, 2011), the element composition was determined by X-ray fluorescence using AXIOS Advanced (PANalytical B.V., Holland) spectrometer, while the grain-size composition was determined by laser diffraction.

The sorption was studied from modeled deionized and sea water simulating the composition of the Kara seawater: NaCl (21.03 g/L), Na_2SO_4 (3.52 g/L), KCl (0.61 g/L), KBr (0.088 g/L), $Na_2B_4O_7 \cdot 10H_2O$ (0.034 g/L), $MgCl_2 \cdot 6H_2O$ (9.5 g/L), $CaCl_2 \cdot 2H_2O$ (1.32 g/L), $SrCl_2 \cdot 6H_2O$ (0.02 g/L), $NaHCO_3$ (0.17 g/L) (Short-Term Methods, 2002).

The contents of ^{137}Cs, ^{90}Sr, and ^{243}Am in the solution were determined by Tri-Carb 2700TR liquid–scintillation counter. The dependence of ^{137}Cs, ^{90}Sr and ^{243}Am sorption on the bottom sediments of the Kara Sea on pH value was calculated on the basis of sorbent concentration of 2 g/L; the sorption kinetics of ^{137}Cs, ^{90}Sr, and ^{243}Am was determined through definite time intervals: 1, 2, 3, 4, 8, 12, 24, 48, and 96 hours. The activities of ^{137}Cs ^{90}Sr, and ^{243}Am in the solutions were 7.6, 10.8, and 6.3 Bq/mL.

The experimental study of sorption dependence on the radionuclide concentrations was carried out using CsCl, $SrCl_2$, and $Eu(NO_3)_3$. The europium nitrate was used in the experiments as analog of americium. Concentrations of the carriers varied from 10^{-1} to 10^{-6} mol/L. Basic-acid conditions in aqueous aliquots of bottom sediments corresponded to natural ones: 7.0 ± 0.5. The experiment was also carried out for two environments: model sea and deionized waters.

The experimentally obtained values of count rate of pulses of background (Ib), initial (I0), and equilibrium (Ip) solutions were used to calculate the degree of sorption (S, %) and solid–liquid radionuclide partition coefficient (*Kd*, mL/g).

RESULTS AND DISCUSSION

Present-Day Radioecological State of the Surface Sediments of the Kara Sea

In spite of the fact that anthropogenic wastes are presently not dumped in the Kara Sea, it is necessary to monitor the modern state of the radionuclide distribution in marine deposits. The measurement of ^{137}Cs specific activities for samples taken during expeditions in 2015–2016 and studied in detail in this work (12 points) shows the low radioactivity level of the surface layer of the bottom sediments of the Kara Sea, including the Ob and Yenisei estuaries, as well as the Novaya Zemlya bays (Fig. 1).

It has been established that the ^{90}Sr specific activity is no more than 0.5 Bq/kg in all studied bottom sediments. The highest values of ^{137}Cs specific activities were mainly found at a depth of 6–8 cm (from 10 to 40 Bq/kg), which indicate a sufficiently high sedimentation rate in sampling localities and, on the other hand, a significant decrease of their basin area in recent years.

Sediments from stations 5313 and 5309 are redeposited sediments of the Ob and Yenisei buried paleovalleys (Levitan et al., 2005; Stepanets, 2010). Sediments in this area practically are not accumulated at present, which leads to the extremely low activities of radiocesium (0.1–0.2 Bq/kg). Station 5306 is located in the transit zone between the St. Anna Trough and the Kara Sea shelf, and radiocesium activity in this area is also very low of 0.2 Bq/kg. The sediments of the Ob and Yenisei estuarine zones differ in the high contents of clay minerals with good sorption parameters: smectites and mixed-layer minerals of the illite–smectite series with high content of smectite interlayers (Table 1). During quantitative analysis, smectites and illite-smectites with the predominance of smectite interlayers were calculated together and their total contents are given in Table 1, as for illites and illite–smectites with the predominance of illite interlayers. The detailed interpretation of mixed-layer minerals was beyond the scope of this study.

The increase of clay component in the estuarine zone is accompanied by the increase of ^{137}Cs concentration in the surface sediments up to 9.0 Bq/kg for the Ob estuarine zone and up to 20.1 Bq/kg for the Yenisei estuarine zone. This was also previously found by

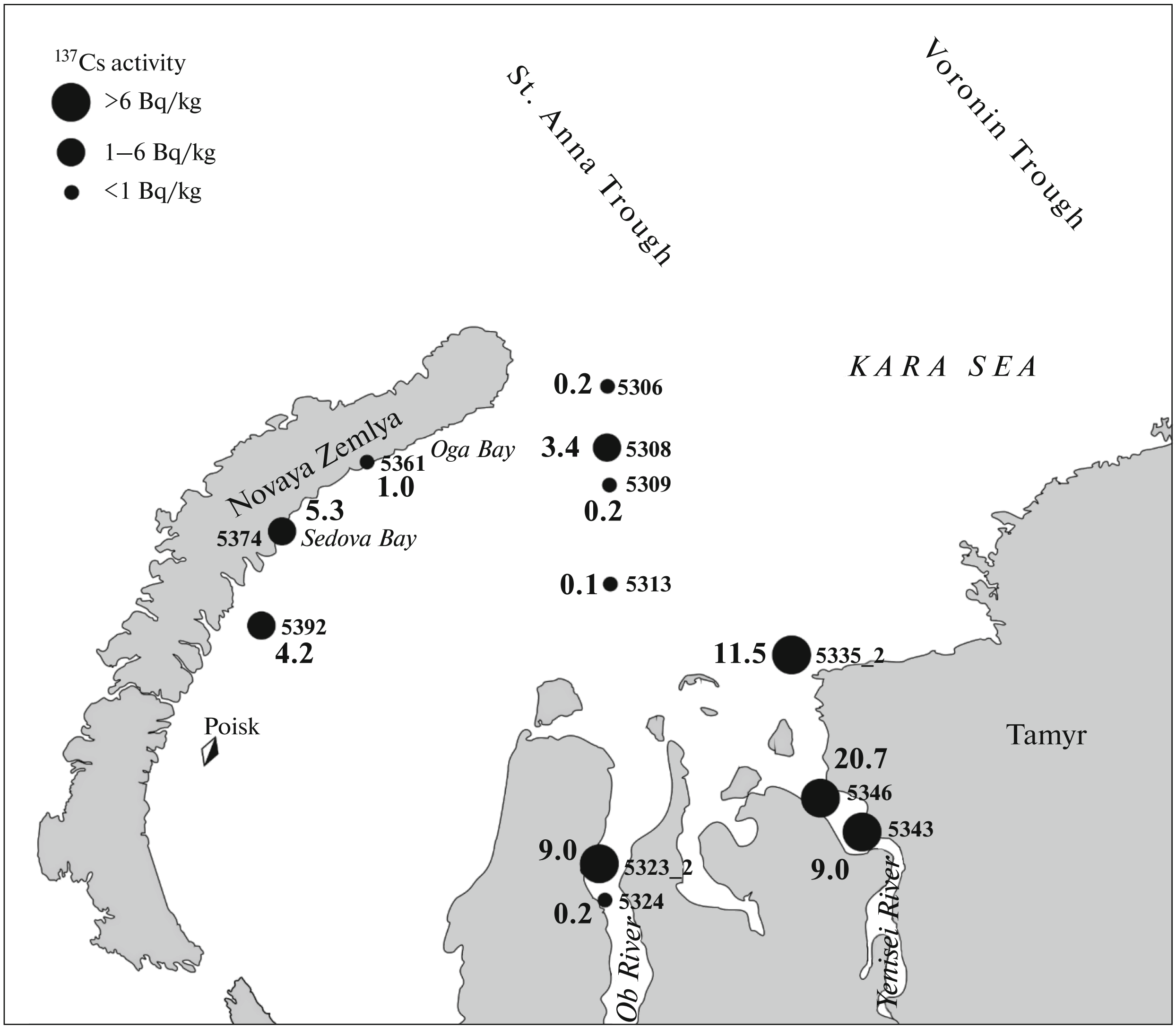

Fig. 1. Modern radioecological situation in the Kara Sea surface bottom sediments, Ob and Yenisei estuaries, and Bays of Severnyi Island (Novaya Zemlya Archipelago).

researchers working in this basin (Stepanets, 2010). The specific activities of ^{137}Cs sharply decrease with decreasing fraction of clay component in the flushing zones of estuaries with high sedimentation rates, which is clearly traced by the example of surface sediments from station 5324. The ^{137}Cs activities in the surface sediments of the Yenisei estuarine zone (stations 5343 and 5346) and its fan (station 5335_2) are higher than in other facies zones of the Kara Sea. In the Novaya Zemlya bays and Eastern Novaya Zemlya Trough, the content of clay minerals (illite) is sufficiently low. These areas also preserve the low ^{137}Cs activities no more than 5.3 Bq/kg. Thereby, the surface sediments in the vicinity of the nuclear test site, in Sedova Bay and slope of the Eastern Novaya Zemlya trough, have slightly higher activities (4.2–5.3 Bq/kg) as compared to Abrosimova Bay (1.0 Bq/kg), which is located beyond the main wind rose of the aerosol distribution after tests (Pavlov, 1995).

The ^{137}Cs activity was correlated with contents of different clay minerals in sediments of the Kara Sea. The influence of each mineral on cesium sorption was calculated using coefficients from the modified Levitan equation the Levitan formula: $E = (20\mathrm{Ill}_1 + 135\mathrm{Sm}_1 + 30\mathrm{Chl}_1 + 10\mathrm{Kaol}_1) \cdot \mathrm{C}_1$, where E is the sorption potential (mg-equiv/g); Ill_1, Sm_1, Chl_1, and Kaol_1 are contents of illite, smectite, chlorite, and kaolinite, respectively, mm (rel %); C_1 is the content of fraction less than 0.001 mm in sediment (wt %). Numerical coefficients before the contents of separate groups of clay minerals denote their average sorption capacities (mg-equiv/g) (Geodekyan et al., 1997). The correlation matrix was constructed for the dependence of ^{137}Cs activity on each mineral, multiplied by corre-

Table 1. Mineral and grain-size composition, ^{137}Cs activity of surface layer of the bottom sediments of the Kara Sea

Component	Station no.										
	5306	5308	5309	5313	5323_2	5324	5335_2	5343	5346	5361	5374
Non-clay minerals (min. composition, wt %)											
Quartz	42.4	24.1	67.6	68.9	30.4	54.9	22.2	18.4	17.5	26.1	22.8
Microcline	10.9	3.7	6.7	11.3	4.6	11.6	5.7	10.4	7	3.5	4.1
Albite	16.5	11	12.3	6.7	12.3	11.7	12.4	26.4	14.2	6.2	14
Calcite	–	0.7	–	–	0.5	0.7	–	1.1	0.6	12	–
Dolomite	0.3	–	–	–	–	0.5	–	–	–	7.5	1.7
Ankerite	–		–	1.5	1.4	–	1.1	–	1.9	–	
Pyrite	0.8		–	–	–	–	–	–	–	–	–
Halite	–	3.2	–	–	–	–	3.1	0.7	1.8	–	
Total	70.9	42.7	86.6	88.4	49.1	79.3	44.4	56.9	43	55.2	42.6
Clay minerals (min. composition, wt %)											
Illite	11.0	30.5	9.3	6.4	23.7	6.2	32.7	20.9	25.3	38.3	39.4
Smectite	6.0	22.0	–	–	21.9	8.5	18.6	16.2	27.5	–	–
Chlorite	8.0	4.0	1.0	3.3	2.6	3.3	1.6	3.7	2.7	5.2	14.6
Kaolinite	4.0	0.8	3.0	1.8	2.8	2.7	2.7	2.2	1.6	1.4	3.4
Total	29.1	57.3	13.3	11.6	50.9	20.7	55.6	43.1	57.0	44.8	57.4
Modern activity, Bq/kg											
Cs-137	0.2	3.4	0.2	3.4	9.0	0.2	11.5	31.7	20.7	1.0	5.3
Grain-size composition, wt %											
<0.005 mm	21.0	15.2	11.6	19.7	20.3	21.2	34.9	15.3	26.1	36.3	35.9
0.005–0.05 mm	49.1	24.2	20.8	27.4	73.7	47.5	64.9	72.3	72.9	62.6	60.0
0.05–0.5 mm	29.9	58.1	61.8	47.1	6.0	31.3	0.1	12.4	1.0	1.1	4.1
0.5–2 mm	0.0	2.5	5.8	5.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 2. Chemical composition of surface sediments of Sedova Bay

Major element composition of sample 5374										
Major element	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	MnO	K_2O	CaO	MgO	Na_2O	P_2O_5
Content, %	54.8	17.06	0.92	8.12	1.00	2.84	0.85	3.37	2.66	0.26

Trace-element composition of sample 5374											
Trace element	Sr	Y	Zr	Nb	Ba	Pb	Co	Ni	Cu	Zn	Rb
Content, %	0.013	0.003	0.018	0.0016	0.092	0.0014	0.0024	0.0092	0.0035	0.0123	0.0116

sponding coefficient. The maximum correlation coefficient of 0.59 was established for radiocesium and smectite. For other minerals, the correlation coefficient was very low (0.19) or negative. We can conclude that smectite has the maximum influence on the cesium sorption among clay minerals for the studied Kara Sea. This conclusion need to be verified on a much larger number of samples studied by above mentioned methods.

Characteristics of Surface Sediments of Sedova Bay (Severnyi Island, Novaya Zemlya Archipelago)

The surface sediments in Sedova Bay from station 5374 (Fig. 1) are represented by liquid incoherent greenish-black silty-pelitic muds with scarce fragments of poorly rounded shales and siltstones. The ^{37}Cs and ^{90}Sr activities are 5.3 Bq/kg and less than 0.5 Bq/kg, respectively. The content of organic carbon is 1.5%.

The bottom sediments have nearly Clarke contents of most studied elements (Table 2) and are characterized by the elevated contents of Fe and Mn, as well as P, S, V, and the lowered Ca content. The elevated Fe content in the bottom sediments of the Kara Sea has been noted previously, while the low Ca content is typical of sediments supplied by the Ob River (Asadulin et al., 2013).

Dispersion coefficients usually vary in a narrow range (Cv < 50%), reaching 131, 95, and 70% only for Ca, Mn, and Zr, respectively. The elevated Ca content is related to the involvement in calculation of samples 5343, 5346, and 5361 enriched in calcitic shells. The elevated Zr contents were likely related to its residual accumulation in the coarsest material.

The study of fraction composition revealed the predominance of silt and pelite-sized components in the surface layer, which are characterized by the highest sorption ability: sand (2.0–0.05 mm) 4.1%, silt (0.05–0.005 mm) 60.0%, and pelite (<0.005 mm) 35.9% (Table 1).

In terms of mineral composition, the surface sediments of Sedova Bay represent the polymineral mixture with the high contents of quartz, feldspars, and clay minerals, which are dominated by components with relatively low sorption capacity, illite and chlorite (Table 1). At the same time, numerous studies show that illite is characterized by the high selective sorption relative to ^{137}Cs (Comans, 1992; Volosov, 2011; Zachara, 2002).

Sorption of Radionuclides by Surface Sediments of Sedova Bay (Severnyi Island, Novaya Zemlya Archipelago)

The study of sorption kinetics of radionuclides on the bottom sediments of the Kara Sea. The study of sorption kinetics of radionuclides showed that ^{137}Cs rapidly reaches equilibrium, approximately for 2 hours (Fig. 2a), ^{90}Sr reaches equilibrium for less than one hour (Fig. 2b), while ^{243}Am reaches equilibrium approximately for 20 hours (Fig. 2c). Based on these results the experiment was optimized to reach sorption quasi-equilibrium in all cases.

Dependence of radionuclide sorption on the pH value of aqueous phase. Data obtained during sorption experiments (Fig. 3) allowed us to estimate the influence of solution pH on the radionuclide sorption by bottom sediments. The great difference in obtained values between model sea and deionized waters indicates that the degree of sorption decreases with the appearance of competitive ions. It is seen from Fig. 3a that ^{137}Cs sorption does not depend on pH value, which indicates the predominant ion-exchange mechanism of interaction of cesium ions with mineral surface, as was previously determined in (Churakov, 2013; Eliason, 1966; Ngouana and Kalinichev, 2011, and others). The sorption coefficient in sea water is much lower (<40%), which is related to the presence of competitive Na in a model water. A decrease of Cs sorption is explained by the rapid sorption of highly hydrated sodium with lower ionic radius, which results in the occupation of all ion exchange sites.

Different results were obtained for strontium and americium. A positive correlation was found between the degree of ^{90}Sr sorption and pH value of model deionized water. This indicates the predominance of mechanism of ^{90}Sr complexation on the surface of clay minerals. The sorption of ^{90}Sr ions in the model deionized water significantly increases with an increase of pH value (Fig. 3b). This is supposedly related to a change of properties of both adsorbate and adsorbent. The Sr^{2+} ions begin to pass into less soluble species SrOH^{+}, on the one hand, and, on the other hand, the negative charge of the adsrobent surface increases. It is pertinent to mention that the experiment with strontium in seawater led to opposite con-

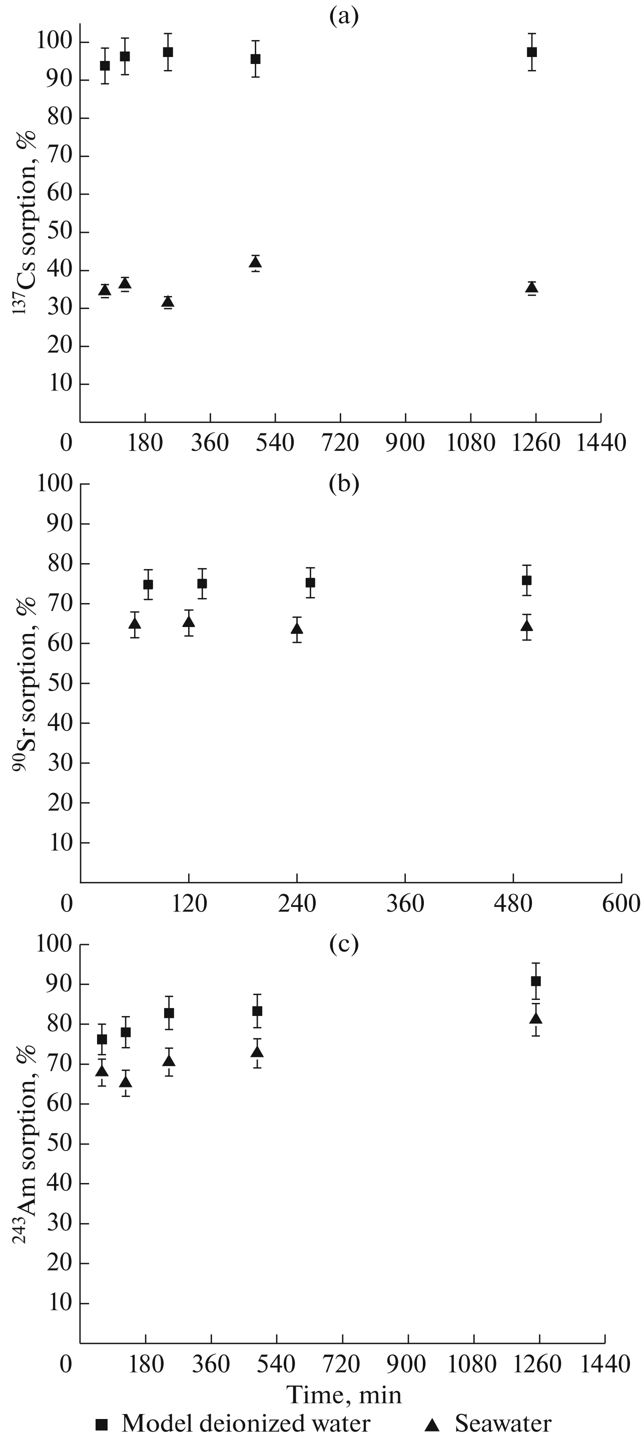

Fig. 2. Kinetics of Cs (a), Sr (b), and Am (c) sorption on the bottom sediment of Sedova Bay.

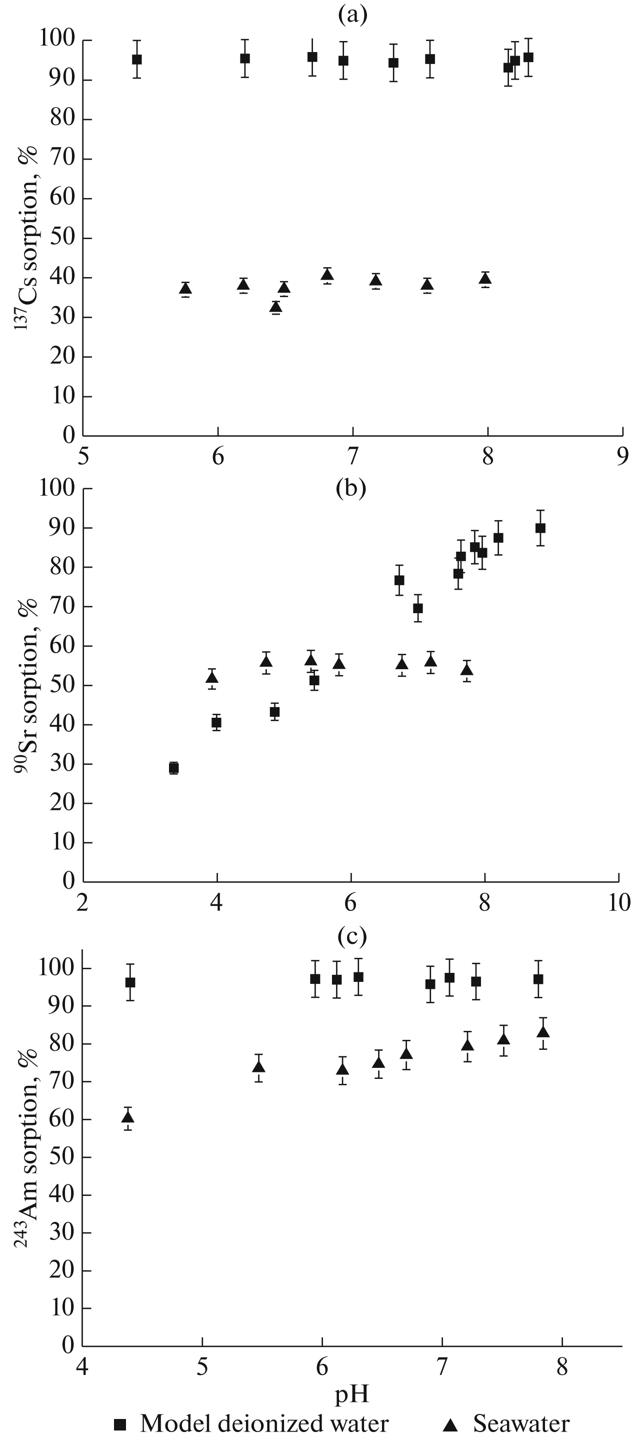

Fig. 3. Variations of Cs (a), Sr (b), and Am (c) sorption on pH value.

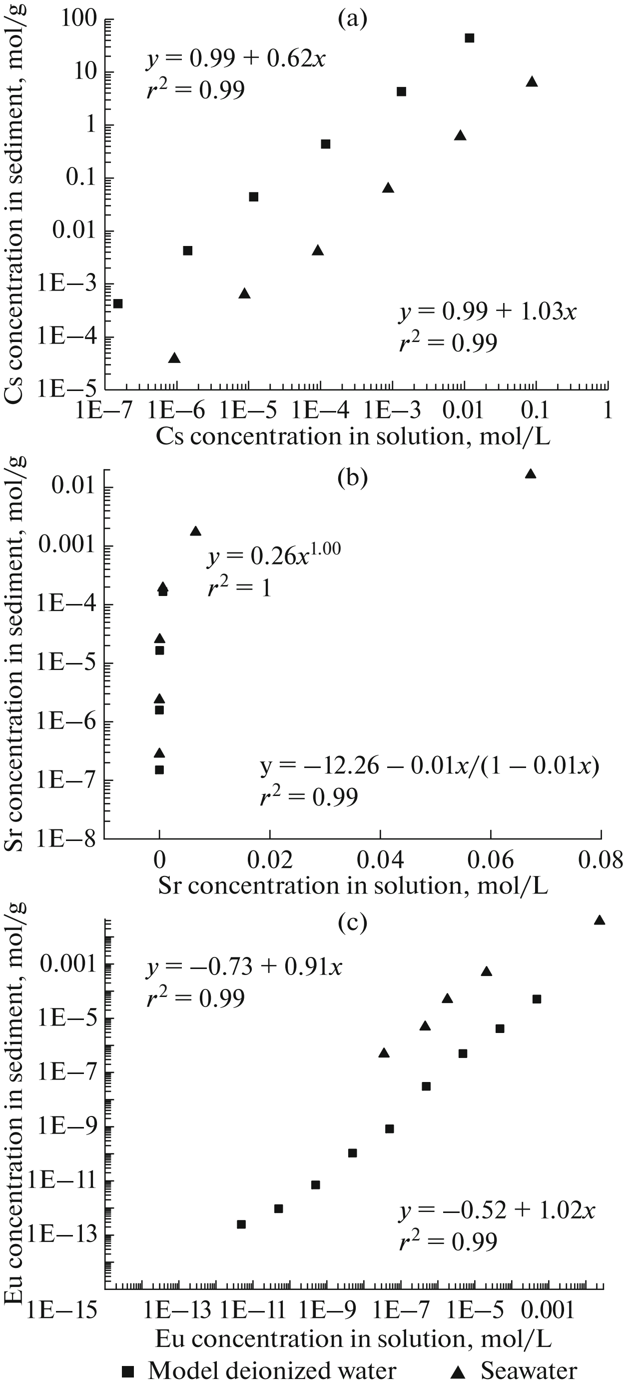

Fig. 4. Cs (a), Sr (b), and Am (c) sorption isotherms for sea and model deionized waters for the bottom sediment from Sedova Bay.

clusions, finding no pH dependence. The sorption mechanism in this case is similar to the ion exchange one, as for cesium.

It has been found that the degree of Am sorption depends on the pH value (Fig. 3c). This radionuclide in sea water is prone to complexation on the surface of clay mineral surface in the bottom sediments of Sedova Bay. Based on experimental data (Fig. 3b), the americium sorption is controlled by complexation with surface groups, since the pH dependence implies the participation of proton in the reaction. The absence of this dependence for model deionized water suggests ion–exchange as the main sorption mechanism.

The study of isotherms of radionuclide sorption on the bottom sediments of the Kara Sea. Concentrations unfavorable for the precipitation of the studied radionuclides were determined during experiment on studying the dependence of sorption effect on the radionuclide concentration and calculation of partition coefficients using a Hydra software (KTH Royal Institute of Technology).

Isotherm of Cs sorption is described by the Henry equation, *Kd* for model deionized and sea water was 3404 and 61.6, respectively (Fig. 4a). The isotherm of Sr sorption for sea water is described by the Freindlikh's equation, $Kd = 236$. For model deionized water, the curve is described by the Langmuir equation: the maximum sorption is 214 ± 28 mol/g, *Kd* varies from 200 to 600 (Fig. 4b). The isotherm of Eu sorption for model deionized and sea water is described by the Henry equation. Kd in sea water reaches 4187 owing to the high affinity of adsorbate to adsorbent (Fig. 4c).

CONCLUSIONS

The sorption properties of the bottom sediments of the Kara Sea, which were sampled in the framework of the complex research expedition of the R/V *Akademik Mstislav Keldysh* (cruise AMK-66), were studied. The mineral and fraction composition of the sediments was determined and great amount of clay minerals (smectite and ilite-smectite mixed-layered minerals) was found.

It was established that the sorption of ^{137}Cs on sediments owing to the presence of clay minerals is mainly controlled by the ion-exchange mechanism. The sorption of ^{243}Am and ^{90}Sr in sea water is controlled by compexation on sediment surface. The study of sorption kinetics showed that equilibrium was reached for 20 hours for ^{243}Am, for 2 hours for ^{137}Cs, and less than 1 hour for ^{90}Sr. Isotherms of Cs and Am sorption from model sea and deionized waters in experiments are described by the Henry equation. The Sr sorption from the model deionized and sea water is described by the Langmuir and Freindlikh equations, respectively.

The sorption coefficients (*Kd*) of the studied radionuclides are as follows: 61.6 and 3404 for ^{137}Cs from sea and model deionized water, respectively; 236 and from 200 to 600 for ^{90}Sr from sea water and model deionized water, respectively; and 4187 for ^{241}Am from sea water.

This research is a pilot work on studying the sorption of radioecologically important radionuclides on the bottom sediments of the Arctic Ocean.

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