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Research Article

^{134}Cs, ^{137}Cs, ^{40}K, ^{232}Th, ^{226}Ra in bottom sediments of the Dvina Bay on the White Sea (the Suhoe Sea Gulf)

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Abstract

The Suhoe Sea Gulf is a unique White Sea water body. Taking into account the risk of contamination of the White Sea with radionuclides as a result of the activities of the domestic and foreign nuclear industry and considering the plans to construct a deep-water part of the Arkhangelsk port on the Kuya River, the content and distribution patterns of natural and man-made radionuclides in the bottom sediments of the Suhoe Sea Gulf need to be studied. The specific activity of radionuclides was measured using a PROGRESS-2000 gamma spectrometer. Statistical processing of the data was performed using the STATISTICA (data analysis software system), version 10 software by StatSoft, Inc. (2011). The average specific activity of ^{226}Ra, ^{232}Th and ^{40}K was 6.5 ± 1.4, 14.2 ± 4.3, 416 ± 89, accordingly. ^{134}Cs and ^{137}Cs were detected in 4 and 5 samples with a mean specific activity value of 3.3 ± 1.6 and 3.5 ± 1.1, respectively. The highest specific activity values of ^{40}K are confined to the pelitic deposits. The main driving force in the processes of accumulation and redistribution of ^{232}Th is gravitational water accumulation and mechanical transfer. The measured values of the specific activity of radionuclides do not exceed those previously determined by other authors in the bottom sediments of the White Sea. Correlation analysis showed a significant relationship between the content of ^{134}Cs and ^{137}Cs (0.77, $p = 0.05$), ^{232}Th and ^{40}K (0.67, $p = 0.05$) and ^{137}Cs and ^{40}K (0.84, $p = 0.05$). Factor analysis allowed two groups of radionuclides to be identified, their content being is determined by different processes: ^{134}Cs, ^{137}Cs, and ^{40}K are jointly controlled by the most powerful factor (50%) and ^{232}Th is affected by the weaker factor (29 %). Both factors are based on natural processes of radionuclide receipt and redistribution: the first factor reflects the ability of bottom sediments to adsorb ^{40}K and isotopes of cesium, which are similar in chemical properties, and the second one reflects the natural process of removal by rivers of ^{232}Th with terrigenous material.

Keywords

bottom sediments, White Sea, Suhoe Sea, radioactivity, anthropogenic radionuclides ^{134}Cs, ^{137}Cs; natural radionuclides ^{226}Ra, ^{232}Th, ^{40}K.

Introduction

Among the Arctic seas, the least contribution to technogenic radioactivity in the world ocean is made by the waters of the White Sea owing to slower water exchange and intensive sedimentation of radionuclides (Kiselev et al. 2000).

The main sources of technogenic radionuclides in the bottom sediments of the White Sea are discharges of radioactive waste from the British nuclear centre in Sellafield (Vakulovsky et al. 1988) and flushing from the catchment area of the White Sea rivers (Kiselev et al. 2000). A possible source of man-made radionuclides includes emergency situations at OAO Zvyozdochka Ship-Repairing Centre in Severodvinsk) (Ioyrish et al. 2008). Traces of radioactive contamination from the Chernobyl accident are also noted (Aliyev et al. 2006; Kiselev et al. 2006; Matishov et al. 2007; Kryauchyunas 2008, 2014; Shvartsman et al. 2008; Bazhenov et al. 2010; Grigoriev et al. 2015; Klimovskiy et al. 2017).

As part of state monitoring of technogenic radionuclides on the territory of the Russian Federation, samples of marine soil are taken at ten points on the Dvina Bay of the White sea, confined to its Western part near Severodvinsk, while the Eastern part of the Dvina Bay remains uncovered by the observation network. Taking into account the plans for constructing a deep-water area of the seaport of Arkhangelsk and the risks of accidental contamination by radionuclides of the White Sea, an important task is to obtain new data on the content and distribution of radionuclides in bottom sediments.

Purpose of the research: to establish the content and distribution patterns of technogenic and natural radionuclides in the bottom sediments of the Suhoe Sea Gulf.

Study area

The current state of the Suhoe Sea Gulf, its geomorphological, hydrological and hydrochemical characteristics, as well as the history of research and economic development of the area on the Dvina Bay of the White Sea, are presented in the work (Moseev and Sergienko 2016; Miskevich et al. 2018). The authors show that this area is currently undergoing significant geomorphological changes. The Suhoe Sea Gulf is a vast and shallow lagoon-like bay in the southeastern part of the Dvina Bay of the White Sea. It is part of the hydrographic system of the mouth of the Northern Dvina River and is located on its estuary in the zone of mixing of river and sea waters. The Suhoe Sea Gulf should be attributed to the unique water bodies of the White Sea, with a wide variety of different aquatic ecosystem parameters. The studied area is still relatively poorly studied and studies of it are of particular relevance, given the possibility of construction of the deep sea port in this area in the very near future.

Tidal phenomena generate fluctuations in many elements of the Suhoe Sea ecosystem and, above all, this affects the variability of hydrological parameters. It is formed by tidal half-day, daily and half-month cycles. On the other hand, these are superimposed by a daily cycle associated with the influence of solar radiation, and synoptic cycles. Salinity, in the framework of biogeochemical estuary barriers by regulating the deposition and dissolution of suspensions, has a significant impact on the spatial distribution of water turbidity, depth and accumulation of heavy metals and radioactivity in the bottom sediments. This fact should be taken into account when studying the Suhoe Sea and designing economic and other activities in its waters. In the Suhoe Sea, there is an increased content of suspended solids characteristic of the estuary shallow zones of a tidal sea. This is due to accumulation of mainly bound sediments, represented by silt and silt-clay sediments. Such sediments can be quite easily agitated by strong tidal currents or even small waves (less than 1 m) of significant steepness, if their height becomes comparable with the depth of the shallow areas of the Suhoe Sea. With prolonged storm surges in shallow, silt deposit areas of the Suhoe Sea, concentrations of suspended solids can increase to 200 mg/l or more (Miskevich and Miskevich 2017; Miskevich et al. 2018).

The current recreational load on the Suhoe Sea and adjacent catchment areas is heterogeneous. In particular, Mudyug Island may be attractive for tourism purposes and the mainland coast of the eastern zone of the northern part of the Suhoe Sea is rarely visited by people because of its poor transport accessibility. The species composition of the coastal flora of the Suhoe Sea is represented by two species of plant included in the Red book of the Arkhangelsk Region with the status of biological control (Moseev and Sergienko 2016).

The studied area is a fishing area where it is allowed to catch aquatic biological resources: European delta smelt, White Sea herring, navaga, polar flounder, river flounder. In the construction of a deep sea port on the Suhoe Sea, the geoecological situation in the northern part of the Sea may change dramatically. According to (Miskevich and Miskevich 2017; Miskevich et al. 2018), the optimal way to resolve this will be to transfer the port territory beyond the Suhoe Sea, for example, to the mouth of the Kui River. The design of the seaport should take into account the active lithodynamics of sediments and the coasts of the Suhoe Sea islands, which can cause shifting of them, as well as formation of new islands (drained shoals) and straits and mixing, which can undoubtedly affect migration and redistribution of radioactive elements in bottom sediments.

Materials and methods

As a result of the expedition work in the Suhoe Sea Gulf in the Dvina Bay of the White Sea in 2016 and 2018, nine bottom sediments were tested by drilling from ice with a manual geological drill to a depth of 0.2 m (Fig. 1). The specific activity of the samples was determined in the Marinelli geometry (1 l) with a PROGRESS-2000 gamma spectrometer. The mea-

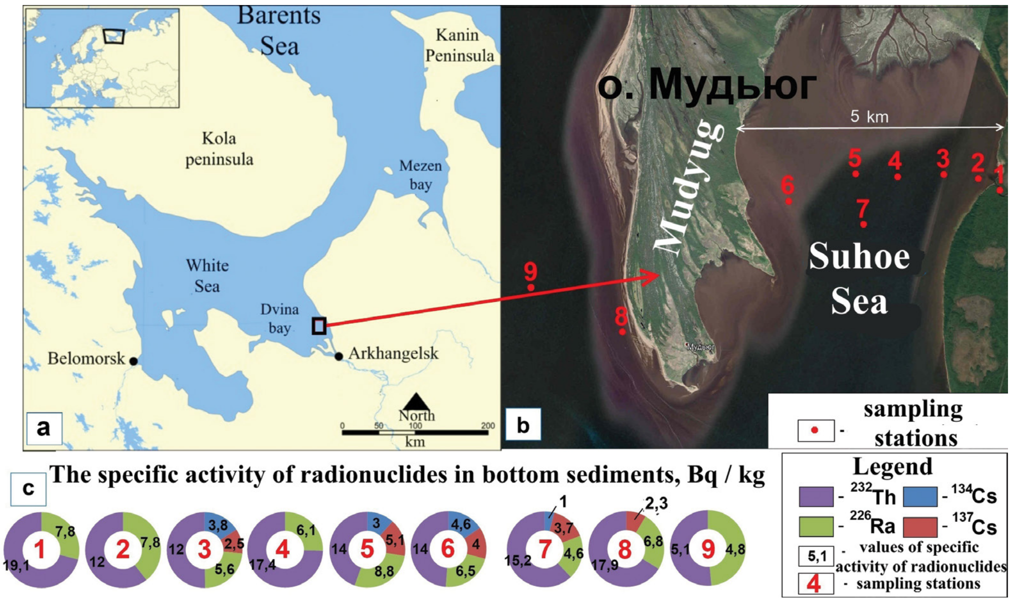

Fig. 1. Study of radioactivity distribution of bottom sediments in the White Sea (Dvina Bay, Suhoe Sea Gulf): a – location of the study area, b – location of sampling points of bottom sediments; c – specific activity of radionuclides in bottom sediments, Bq/kg

Table 1. Specific activity of radionuclides in bottom sediments, Bq/kg

^{134}Cs	^{137}Cs	^{226}Ra	^{232}Th	^{40}K
$\frac{1.0^{*} - 4.6}{3.3 \pm 1.6(3.8)}; 4^{**}$	$\frac{2.3^{*} - 5.1}{3.5 \pm 1.1(3.7)}; 5$	$\frac{4.6^{*} - 8.9}{6.5 \pm 1.4(6.5)}; 9$	$\frac{5.1^{*} - 19.1}{14.2 \pm 4.3(14.4)}; 9$	$\frac{265 - 506}{416 \pm 89(493)}; 9$

Note: * values below the detection limit (excluded from the analysis); **in the denominator are the minimum and maximum values, in the numerator – the mean, standard deviation and median. The number next to the fraction is the number of samples with measured values.

surement error of radionuclides ranged from 10 to 30 % (Antropov et al. 1996; PROGRESS software 1997).

Statistical processing of the data was performed using the STATISTICA (data analysis software system), version 10 software by StatSoft, Inc. (2011).

Results

This paper presents the results of a study of the distribution of technogenic (^{134}Cs, ^{137}Cs) and natural radionuclides (^{40}K, ^{232}Th, ^{262}Ra) in bottom sediments sampled on the lower reaches of the Ulmitsa River, in the Suhoe Sea Gulf and in the coastal part of the White Sea. The location of the sampling points and the measurement results are shown in figure 1 and in the table.

Discussion

In bottom sediments collected at sampling stations: 3, 5, 6, 7 and 8, trace values of long-lived technogenic radionuclides ^{134}Cs (half-life $T_{1/2}$ = 2.062 years), ^{137}Cs ($T_{1/2}$ = 30.174 years) are recorded. The change in the specific activity for ^{134}Cs in bottom sediments ranged from 1.0 to 4.6 Bq/kg, and for ^{137}Cs – from 2.3 to 5.1 Bq/kg. The average value of the specific activity of ^{137}Cs corresponds to the minimum level of the specific activity of this isotope in bottom sediments of the White Sea in 1994, which ranged from 0.9...5.0 to 67.0 Bq/kg with a maximum accumulation in the Dvina Bay (Matishov et al. 1995). The average specific activity of ^{137}Cs obtained is less than the average value of this indicator in the bottom sediments of the Dvina Bay of the White sea for the period of observations from 1998 to 2017 (5.0 ± 2.3 Bq/kg) (Radiation Situation in Russia 2018).

It should be noted that, at sampling stations 3 and 6, the ^{134}Cs content slightly exceeds the activity of ^{137}Cs, which might indicate recent arrival of ^{134}Cs in the bottom sediments of the Suhoe Sea. Natural radionuclides (NRN), unlike man-made ones, have always been part of the seabed sediments. The main source of arrival of NRN in the bottom sediments can be alluvial outflows and erosion of the coastline. Potassium-40 is one of the main NRN in the bottom sediments, its specific activity varying from 264 to 506 Bq/kg (Fig. 2).

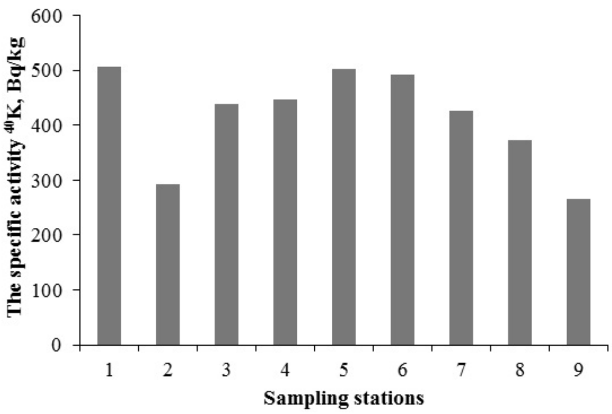

Fig. 2. Specific activity of ^{40}K in bottom sediments at sampling stations.

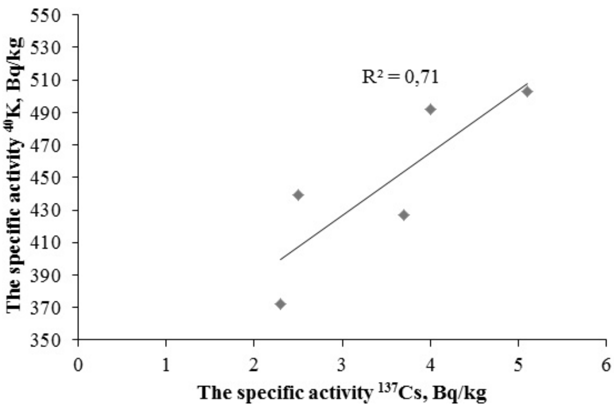

Fig. 3. Specific activity of ^{137}Cs, ^{40}K in bottom sediments.

Analysis of the specific activity of ^{40}K in the samples showed that the highest concentrations of ^{40}K, from 420 to 506 Bq/kg, are typical of pelitic deposits (Yudakhin et al. 2002). The maximum content of ^{40}K was recorded in the fine sediments on the lower reaches of the Ulmitsa River and the "deep" area of the Suhoe Sea, beyond the front of the river delta. A decrease in ^{40}K concentration is observed in sandy bottom sediments in shallow areas of the Suhoe and White Seas Bay, mainly confined to river deltas and the frontal and underwater areas of Mudyug Island beach. Isotopes of cesium and potassium, as elements with the same configuration of the structure of the outer electron shell and similar chemical properties (Ginwood and Earnshaw 1998), are accumulated together (Fig. 3).

^{232}Th, which is characterised by widespread presence, is the second element by quantitative content in the considered bottom sediments, its specific activity varying from 5 to 19 Bq/kg. Thorium is a typical element of hydrolysate and migration in solution is uncharacteristic of it. According to (Titaeva 2000; Rikhvanov et al. 2007), thorium mainly migrates in the form of thin suspensions and in clastic material and, in the future, its deposition is associated with the terrigenous (clay) component. This pattern explains the relatively high content of ^{232}Th in the sediments of station 1, on the lower reaches of Olmeca River (where deep erosion disappears) and station 8, which was laid on the underwater accumulative terrace of Mudyug Island beyond the front of the beach, where wave activity is at is maximum. As a result of wave activity, the coastal part of the land is eroded and the terrigenous material is carried and accumulated in the underwater part of the beach. Increased concentration of ^{232}Th in the sediments of stations 4 and 7 is explained by their location in one of the channels of the river delta of Mudyug Island, which is the main takeaway of terrigenous material. Thus, thorium is inactive under the conditions of underwater hypergenesis, its transportation and re-deposition occurring mainly owing to mechanical transport and underwater gravitational accumulation of fine-dispersed thorium-containing minerals that enrich the pelitic sediments of the coastal part of the White Sea, including the Suhoe Sea.

In turn, the specific activity of ^{226}Ra in bottom sediments ranges from 4 to 9 Bq/kg. The distribution of radium is virtually unrelated to the fractional composition of bottom sediments. Most likely, its distribution in the bottom rocks correlates with the pH parameter. As is known, ^{226}Ra belongs to alkaline earth metals and is characterised by active migration in acidic waters. In alkaline waters, the solubility of the radium compound is significantly reduced and its active accumulation in the bottom sediments is observed, i.e., the element is characterised by concentration on alkaline barriers (Emelyanov 1998; Ivanov et al. 1997).

Correlation analysis showed a significant relationship between the content of ^{134}Cs and ^{137}Cs (0.77, $p = 0.05$), ^{232}Th and ^{40}K (0.67, $p = 0.05$) and ^{137}Cs and ^{40}K (0.84, $p = 0.05$). Factor analysis allowed two groups of radionuclides to be identified, the content of which is determined by different processes: ^{134}Cs, ^{137}Cs, and ^{40}K are jointly controlled by the most powerful factor (50 %), and ^{232}Th is affected by the weaker factor (29 %). Both factors are based on natural processes of radionuclide receipt and redistribution: the first factor reflects the ability of bottom sediments to adsorb ^{40}K and isotopes of cesium, which are similar in chemical properties, and the second one reflects the natural process of removal by rivers of ^{232}Th with terrigenous material.

Conclusion

The water area of the Suhoe Sea Gulf is characterised by constant geomorphological changes, both past and present, which undoubtedly affects the redistribution of radionuclides in the bottom sediments. In the studied area, lithodynamic processes that change the depth of the bays and the configuration of the coastline occur actively. The water area of the Suhoe Sea Gulf is characterised by variability of hydrological and hydrochemical characteristics owing to mixing of river and sea waters.

The measured values of the specific activity of ^{137}Cs in the bottom sediments of the Suhoe Sea Gulf do not exceed the values previously obtained by other authors for the White and Barents Seas.

It was found that high specific activity of ^{40}K was observed in pelitic sediments: on the lower reaches of Ulmitsa River and fine marine sediments of the "deep" area of the Suhoe Sea beyond the front of the river delta.

It has been found that ^{232}Th is immobile in supergene environments and its transportation and redeposition occur mainly by mechanical transfer of gravitational and underwater accumulation.

Insignificant concentrations of man-made radionuclides ^{134}Cs and ^{137}Cs were recorded in the bottom sediments.

It is established that the spatial distribution of ^{226}Ra in bottom sediments is not related to their fractional composition.

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