ANTHROPOGENIC TRANSFORMATION OF THE ECOSYSTEM

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THE BASIC PATTERNS OF THE DISTRIBUTION, MIGRATION AND ACCUMULATION OF RADIONUCLIDES IN THE BOTTOM SEDIMENT OF THE BALTIC SEA

This paper focuses on the impact of certain factors on the contemporary distribution of natural (226Ra, 232Th, 40K) and anthropogenic (137Cs, 60Co) radionuclides in the sediments of the Baltic Sea. The results of the study suggest that the distribution of 137Cs is determined by the content of hydromica of silty-clay and clay grain-size fractions, while radiocaesium is mainly accumulated by silty fractions. The accumulation of 226Ra by bottom sediments is mainly determined by the pH geochemical barrier at the water-seafloor boundary. The accumulation of 232Th occurs mainly in clayey fractions of the sediment. The distribution and accumulation of 40K is predominantly determined by the ratio of potassium contained in hydromica minerals. Significant 60Co activity was registered only in a few samples.

Key words: radionuclide distribution patterns, Baltic Sea, hydromica, pH barrier, sediment fractions, grain-size composition.

A number of researchers have studied the features of migration and accumulation of natural and anthropogenic radionuclides. In particular, one of the fundamental works is *Artificial radionuclides in marine environment* by V. V. Gromov and V. I. Spitsin [6] dedicated to the problems of migration and sorption of anthropogenic radionuclides in oceans. A significant contribution to the study of this issue has been made by I. Ye. Starik, A. P. Lisitsyn, N. M. Strakhov, G. G. Matishev and D. G. Matishev, V. P. Tishkov, V. V. Anisimov, and many others. However, these researchers have focused mainly on radionuclide distribution and behaviour in oceans, at significant depths, in certain sedimentation conditions, and in the presence of long-lasting processes of radionuclide migration in the aquatic environment. The conditions of migration and accumulation of main gamma-emitting radionuclides in the bottom sediment in a relatively shallow basin of the Baltic Sea with dramatic changes in the bathymetric level, the composition of bottom sediment, redox and acid-base characteristics and a significant amount of substances coming from the shore have specific features and are of special interest. Another important aspect of the issue is the immediate influence of the "Chernobyl trace" on the water area of the Baltic Sea.

We made an attempt to consider the influence of a number of factors on the conditions of migration, accumulation and patterns of the contemporary distribution of natural (^{226}Ra, ^{232}Th, ^{40}K) and anthropogenic (^{137}Cs, ^{60}Co) radionuclides in the Baltic Sea sediment. We used data obtained within the typical sedimentation basins of the Baltic area: the Gulf of Finland, the Bornholm and Gotland basins and the Skagerrak.

General regional characteristics

The distribution of isotopes under consideration over the main studied areas falls into a certain pattern. The table below shows a generalisation of statistical parameters of radionuclide distribution.

Gamma-emitting radionuclide activity distribution in the main studied areas (Bq/kg)

Area	^{226}Ra		^{232}Th		^{40}K		^{137}Cs		^{60}Co		N
	A_b	σ	A_b	σ	A_b	σ	A_b	σ	A_b	σ	
Gulf of Finland	52	31	75	34	805	303	560	458	5	11	411
Gotland basin	48	22	56	16	963	152	163	32	—	—	29
Bornholm basin	47	19	52	11	837	143	63	39	3	2	49
Skagerrak	25	8	32	6	615	108	14	8	—	—	28

Comments: 1. In case of the Gulf of Finland, the data relate to aleuropelitic sediment in the central and western parts of the gulf. The gaps in the statistical parameters of ^{60}Co distribution indicate that almost all activity values are below the detection limit. 2. A_b — background median value of radionuclide activity distribution, σ — standard deviation, N — number of samples.

There is an evident gradual decrease in the average activity of ^{137}Cs, ^{232}Th and, to a lesser extent, ^{226}Ra from the east to the west in the following order: the Gulf of Finland — Gotland and Bornholm basin — the Strait of Skagerrak. This pattern in the distribution of ^{137}Cs is linked to the spreading of the "Chernobyl trace" from the Gulf of Finland, which the radioactive cloud moved across. As to Th and Ra, one can assume that such regional distribution of their activity in the bottom sediment relates, to a degree, to the radiogeochemical features of rocks. The increased concentrations of ^{226}Ra and ^{232}Th in the bottom sediment of the Gulf of Finland are influenced, on the one hand, by the high content of uranium and radium in the Vendian and Ordovician rocks of the southern shore of the gulf and, on the other hand, by the vicinity of U (Ra) and Th based granitoid massifs on the south-eastern rim of the Baltic shield [14, 15]. Apparently, as the distance from the main sources of these isotopes increases, their concentration in bottom sediment decreases.

The detailed study of the patterns of radionuclide behaviour and distribution in the bottom sediment was carried out with the help of factor analysis through the method of R-modification main components. We used samples covering radionuclide activity, percentage of different grain-size fractions of bottom sediment, the values of physicochemical parameters (pH, Eh) of near-bottom silted water, and the depth of basins.

The central and western Baltic area (the Baltic Sea proper)

The results of factor analysis of data (69 stations) on the Baltic Sea, characterising mainly its principal sedimentation basins (Gotland and Bornholm basins) and the adjacent seabed areas, showed the following (fig. 1).

The joint analysis of factor loading distribution shown in figure 1 helps identify the four associations, closely connected parameters.

The first, "deep water", association includes grain-size fractions of pelitic dimension, the depth of the sea and ^{232}Th activity. The second, "shallow water" association includes sand fractions, of which the accumulation of radioactive elements is not characteristic. The third association covers the ^{137}Cs activity, the grain-size fractions of aleuritic dimensions, the Eh value. The fourth one concerns with ^{226}Ra and pH. ^{40}K does not belong to any of the above groups but, to a degree, gravitates toward the pelitic and, partially, aleuritic associations. Thus, the pelitic fraction is clearly linked only to the accumulation of ^{232}Th. At first sight, this result is quite surprising, since it is a standard assumption that the accumulation of all four radionuclides is characteristic of the pelitic fraction.

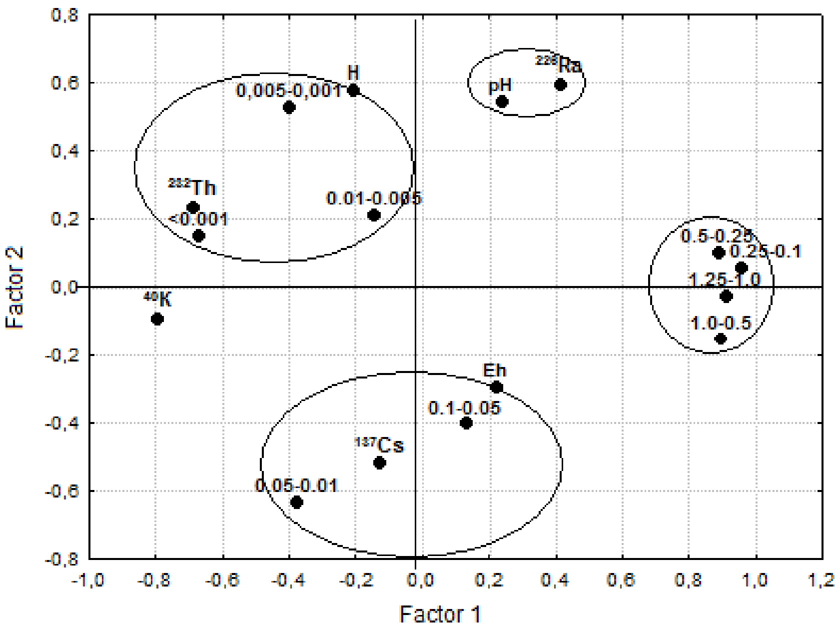

Fig. 1. The diagram of factor loading for radionuclide activity, pH, Eh, depth, and granulometry

Comment: here and below, in factor diagrams, digits stand for grain-size fractions of bottom sediment samples (mm), H is the depth of the sea.

The analysis of the results obtained suggests the following interpretation. The close thorium-pelite fraction bond is not surprising. It is consistent with the data given in A.I. Blazhchishin's work [2] demonstrating the prevailing accumulation of thorium in the pelite fraction of bottom sediment of the Baltic

Sea proper. Thorium has low mobility in most conditions [11], thus, its transportation and resedimentation takes place mostly due to mechanical transfer and gravitational accumulation. V. V. Gordeev and A. P. Lisitsyn [5] come to a conclusion that 97.5 % of the total mass of thorium entering the sedimentation basins with river discharge is in the suspension state. All the above makes it possible to arrive at a conclusion that thorium enrichment of the Baltic Sea pelitic sediments occurs through fine-dispersed thorium-containing minerals, which are transported in the aquatic environment within micromineral suspension. Long transportation involves considerable mechanical processing, which, to a great degree, accounts for its pelitic dimensions.

The distribution of radium is hardly connected with the fraction composition of bottom sediment. On the other hand, the results of factor analysis show a close link between ^{226}Ra and the pH parameter. It is supported by the results of correlation analysis: bottom sediments demonstrate a weak but significant positive correlation (R= +0.36) between the pH of interstital water and ^{226}Ra activity, i. e. at a slightly higher alkalinity, the radium concentration increases. As we know, radium — an alkaline-earth metal — migrates actively in acid waters, while the solubility of radium compounds in alkaline waters is much lower [7; 11]. Concentration on alkaline barriers is characteristic of alkaline-earth metals migrating in solutions in the form of simple cations, in particular, radium [12]. According to the measurements carried out, the near-bottom waters of the Baltic Sea have a neutral or a slightly alkaline medium: pH of the near-bottom waters ranges from 7.24 to 8.07 with a mode value of pH=7.4, which, in general, is hardly favourable for radium migration. However, the alkalinity of interstitial waters medium is slightly higher (pH =7.6—8.5) with a mode value of pH=8.0. Thus, in this case there is a slight alkaline barrier at the water-bottom boundary — the site of radium deposition. The highest radium concentrations are characteristic of the areas with a more alkaline medium.

As it was earlier believed, radiocaesium is accumulated most intensively in the pelitic fraction of the bottom sediments [2; 6; 8—10]. However, the results of the factor analysis within the Baltic Sea proper shows, that ^{137}Cs binds more closely with the aleuritic (0.1—0.01 mm) rather than pelitic fraction.

The possible reasons are as follows. As the works [6, 11] show, hydormicas intensively sorb casesium, and the sorbtion, is to a degree, irreversible: caesium ions, being absorbed, form a stable chemical compound, i. e. chemical adsorption takes place. At the same time, as V. T. Frolov shows [17], in many cases, hydormicas have aleuritic dimensions. Another research [3] offers data proving that hydromicas account for a significant percent of aleuritic fraction of the bottom sediments of the Baltic Sea.

The lower boundary of fin aleuritic silts in the Baltic Sea reaches a depth of 110—130 m [3], i.e aleuritic silts cover mostly the slopes of basins. The results of factor analysis also demonstrate (see fig. 1) that the aleuritic fraction occupies an intermediate position between shallow and deep water sediments, which confirms their prevalent distribution on the slopes. Belonging to the same association as aleuritic silts and radiocaesium, the reduction potential Eh shows that the given material is accumulated in stable oxidative conditions, which is characteristic of basin slopes. In its turn, the pelitic fraction gravitates to the deepest areas, i. e. the basin floors.

The fact of prevalent distribution of hydromica enriched aleuritic fractions at the medium depth of basin slopes and that of the rich in illite pelitic fraction at the significant depths of basin floors makes it possible to explain the prevalent accumulation of ^{137}Cs by the aleuritic rather than pelitic fractions. Probably, the following mechanism of radiocaesium accumulation was involved. As a result of the Chernobyl NPP accident, the waters of the Gulf of Finland were exposed to a significant radiocaesium contamination — the fallout from the Chernobyl cloud. After 1986, large masses of radiocaesium-enriched surface waters were being forced from the Gulf of Finland to the west causing the radiocaesium enrichment of the waters of the central part of the sea. At the same time, there is a striking difference between the surface and bottom waters. The latter are enriched with ^{137}Cs to a much lesser degree, which is especially characteristic of trenches, where radiocaesium activity in the near-bottom layer comprises 27—32 % of that in the surface layers, which is explained by water stratification. However, in 1992, the situation stabilised and the radiocaesium accumulation process in the waters of the Baltic Sea proper ceased [1, p.16]. Thus, in the period of maximum radiocaesium concentration in the waters of the Baltic, the water layers at basin floors contained much less radiocaesium that the slop area waters. Thus, the accumulation of ^{137}Cs through sorption by the slope aleuritic fraction hydromicas was more active than through sorption by the basin floor pelitic fraction hydromicas.

^{40}K is in equilibrium with the non-radioactive ^{39}K isotope. Some studies [4; 13] show that the most wide-spread clay mineral of the Baltic Sea bottom sediment is illite and other potassium-containing hydromicas ($K_{1—1,5}Al_4[Si_{7—6,5}Al_{1—1,5}O_{20}](OH)_4 \cdot nH_2O$). They compose almost the whole medium-pelitic (0.005—0.001 mm) and subcolloidal (<0.001 mm) fractions [3]. On the other hand, in many cases, hydromicas have aleuritic dimensions, as it was mentioned above; thus, ^{40}K, although not related to any of indentified groups, gravitates to the pelitic and, partially, to the aleuritic ones. Its closer connection with the deep water pelitic association indicates that the amount of hydromicas in the pelitic fraction exceeds that in the alueritic fractions of bottom sediments.

The Skagerrak

Interesting results were obtained in the course of factor analysis of samples from the Skagerrak. The analysis of distribution of factor loading shows an almost complete absence of links between the radionuclide activities, the grain-size composition of bottom sediments, and the redox and acid-base situation in the studied area (fig. 2).

Thus, the distribution of radionuclides in the Skagerrak is very specific and differs significantly from the general situation in the Baltic Sea. It can be explained as follows. The studied area within the Strait of Skagerrak is characterised by great depths (198—220 m), while the geomorphological conditions are almost identical throughout the area [4]. The grain-size and lithologic composition is also quite stable and is represented mostly by the pelitic fraction of clay sediments [3].

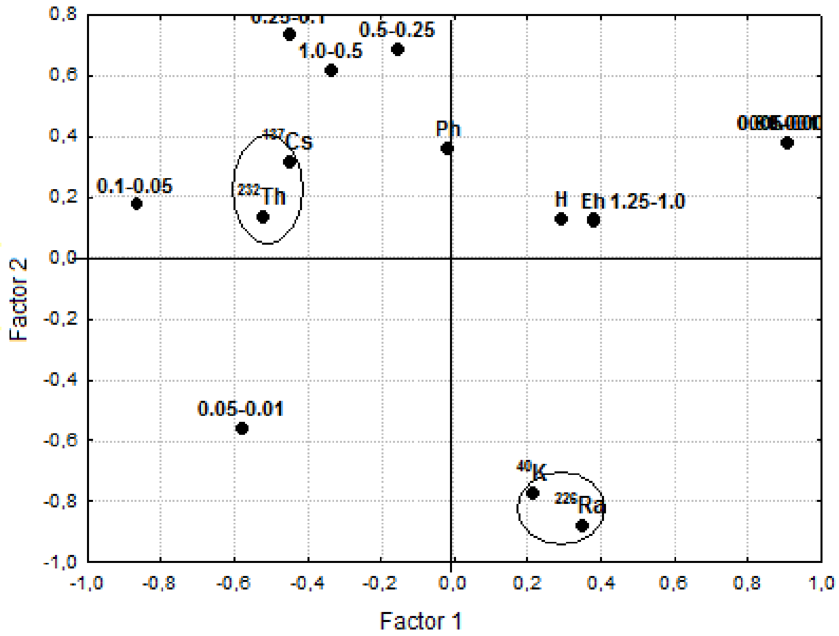

Fig. 2. The diagram of factor loading for radionuclide activity, Ph, Eh, depth and granulometry

The Eh and pH parameters are stable. The concentrations of radionuclides are moderate. Given such low variability of the studied parameters, one can hardly assume that there exists a significant connection between them, which was shown by the factor analysis. The Skagerrak has strong bottom currents resulting from the water inflow from the North Sea — maybe, it is this hydrodynamic factor that plays a decisive role.

Conclusions

In the regional aspect, we established a gradual decrease in the average activity of ^{137}Cs, ^{232}Th and, to a lesser degree, ^{226}Ra from the east to west in the typical sedimentation basins in the following order: the Gulf of Finland — the Gotland and Bornholm basin — the Skagerrak. The tendency in the thorium and radium distribution is a product of regional geological and geochemical factor, that of ^{137}Cs is determined by the spreading of "Chernobyl caesium" from the Gulf of Finland into the other parts of the Baltic Sea. The principal patterns of distribution and accumulation of ^{137}Cs in bottom sediments of the Baltic Sea are determined, to a great extent, by its bond with hydromicas comprising the aleuropelitic and pelitic fractions. At the same time, radiocaesium is mostly accumulated in the aleuritic fraction due to its sorption from the bottom waters. The ^{226}Ra accumulation by bottom sediments is linked, first of all, to the alkaline geochemical barrier at the water-bottom boundary. The ^{232}Th accumulation takes place predominantly in the pelitic fraction of sediments through fine-dispersed thorium-containing minerals, which are transported in the aquatic environment in the form of micro-mineral suspension. The ^{40}K distribution and accumulation is determined by the correlation of potassium contained by hydromica minerals in the bottom sediment. ^{60}Co significant activities were registered in a small number of bottom sediment samples, thus, the patterns of its distribution could not be identified. Probably, the registered ^{60}Co activities can be explained by the local pollution.

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