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DETERMINATION OF CONTROL LEVELS OF RADIONUCLIDES ENSURING ACCEPTABLE ENVIRONMENTAL RISK IN THE BARENTS SEA WATER AND BOTTOM SEDIMENTS

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To assess the radioecological situation, criteria were developed ensuring acceptable environmental risk – control levels of radionuclides in the components of the natural ecosystem. The method was applied to evaluate the control levels of technogenic radionuclides in the Barents Sea water and bottom sediments. If these levels are not exceeded, marine biota is considered protected from radioactive contamination. Local concentration factors of ^{137}Cs, ^{90}Sr, and ^{239}Pu in the Barents Sea biota were estimated using the data of long-term observations. Moreover, the reference organisms were selected – a fish (cod), mollusc (mussel), aquatic plant (fucus), and marine mammal (harp seal). The values of ^{137}Cs, ^{239}Pu, and ^{90}Sr concentration factors were, respectively, as follows (L·kg^{-1}): in fish, 93, 262, and 12; in molluscs, 51, 1,180, and 21; in aquatic plants, 69, 732, and 19; and in marine mammals, 63, 222, and 14. The values of the water–sediment distribution coefficients of ^{137}Cs, ^{239}Pu, and ^{90}Sr were 426, 189,600, and 443 L·kg^{-1}, respectively. For most radionuclides and the reference organisms from the Barents Sea, the values of radionuclide concentration differ from the global average reference values. For the period of 1992–2020, there is no pronounced temporal trend for the concentration factors of all technogenic radionuclides in the Barents Sea fish; this indicates the establishment of equilibrium in the distribution of radioactivity between the components of the Arctic marine ecosystem. The control levels of radionuclides were as follows: in the Barents Sea water (Bq·L^{-1}), 115 for ^{137}Cs, 439 for ^{90}Sr, and 0.124 for ^{239,240}Pu; in the Barents Sea bottom sediments (kBq·kg^{-1} fresh weight), 48.9 for ^{137}Cs, 194 for ^{90}Sr, and 23.6 for ^{239,240}Pu. The contamination index for both water and bottom sediments of the Barents Sea was calculated using the monitoring data and reference levels. In 2006–2020, its values were several orders of magnitude lower than 1 and did not tend to increase or decrease. In terms of marine biota protection, the main contributor to the contamination index for the Barents Sea water is ^{239,240}Pu (up to 75 %) while the main contributor to the contamination index for the Barents Sea bottom sediments is ^{137}Cs (up to 90 %). To date, the ratio of the contributions of technogenic radionuclides to the contamination index for the Barents Sea water and bottom sediments is stable.

Keywords: Arctic, Barents Sea, water, bottom sediments, biota, control level, concentration factor, distribution coefficient, radionuclide

A correct interpretation of monitoring data requires their comparison with the criteria for assessing the radiation situation which ensure acceptable environmental risk. As such criteria, the control levels of radionuclides in the components of the natural ecosystem can be used since the measurement results can be directly compared with them. For assessing radiation and environmental impact on the ecosystem objects, recommendations were developed; those were based on the radiation situation monitoring (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015) and calculation of control levels of radionuclides in seawater (Poryadok rascheta, 2016). The control levels defined in the recommendations (Poryadok rascheta, 2016) are based on the use of generalized values of concentration factors of radionuclides in biota, water–sediment distribution coefficients (Sediment Distribution Coefficients, 2004), and a standard selection of reference organisms (ICRP Publication 108, 2008); these control levels do not take into account regional specifics.

The values of the concentration factors of radionuclides in the components of the Arctic marine ecosystem may differ significantly from the global average values. The Arctic region is characterized by a harsh climate with low temperatures. In cold waters of the Arctic, the accumulation and excretion of radionuclides in hydrobionts occur slower than in water bodies of a temperate or warm climate. Moreover, a set of reference organisms during radioecological assessments for the Arctic seas differs from the standard one (Kryshev et al., 2022 ; Sazykina & Kryshev, 2011).

The presence of technogenic radionuclides in the Barents Sea is mainly due to the following factors: atmospheric fallout after nuclear weapon tests in the second half of the XX century; transport by currents of radioactive discharges from enterprises of Great Britain and France; presence in the Arctic seas of sunken or submerged nuclear and radiation hazardous facilities; and influx of radionuclides after the accident at the Chernobyl nuclear power plant in 1986 (Sarkisov et al., 2015 ; Sivintsev et al., 2005). Expeditionary surveys of the Barents Sea showed the presence in seawater of such long-lived technogenic radionuclides as ^{137}Cs, ^{90}Sr, and ^{239}Pu (Gwynn et al., 2016 ; Jensen et al., 2016).

The objective of the study is to determine the control levels of the technogenic radionuclide content in the Barents Sea water and bottom sediments, the non-exceeding of which ensures the radiation safety of marine biota. To do this, local concentration factors of technogenic radionuclides in biota and water–sediment distribution coefficients for the Barents Sea were estimated using long-term observational data. Moreover, reference species of the Barents Sea ecosystem were selected, as well as threshold values of the radiation dose rate below which no deterministic radiobiological effects for marine biota occur.

MATERIAL AND METHODS

The control level of the *i*-th radionuclide in seawater was calculated by the formula:

$$C_i = \frac{P}{\varepsilon_{a,i} \cdot \alpha_i \cdot CF_i + \varepsilon_{e,i} \cdot (\tau_w + 0.5 \cdot K_{d,i} \cdot \tau_s)}\,, \tag{1}$$

where P is the threshold value of the dose rate above which deterministic radiobiological effects for a reference organism can occur, mGy·day^{-1};

$\varepsilon_{a,i}$ is the dose coefficient of internal exposure of the organism from the accumulated *i*-th radionuclide, (mGy·day^{-1})/(Bq·kg^{-1} fresh weight);

α_i is the quality factor associated with the relative biological effectiveness of radiation, dimensionless;

CF_i is the concentration factor of the *i*-th radionuclide in a reference organism, L·kg^{-1};

$\varepsilon_{e,i}$ is the dose coefficient of external exposure of the organism from seawater and bottom sediments, (mGy·day^{-1})/(Bq·kg^{-1} fresh weight);

$K_{d,i}$ is the water–sediment distribution coefficient for the *i*-th radionuclide, L·kg^{-1} fresh weight;

τ_w and τ_s are the fractions of time that a marine biota representative spends in a water column and near bottom, respectively, dimensionless.

The control level of the *i*-th radionuclide in marine sediments was calculated by the formula:

$$S_i = \frac{P}{\varepsilon_{a,i} \cdot \alpha_i \cdot CF_{s,i} + 0.5 \cdot \varepsilon_{e,i} \cdot \tau_s}\,, \tag{2}$$

where $CF_{s,i}$ is the ratio of the specific activity of the *i*-th radionuclide in a marine organism to its specific activity in bottom sediments, $kg \cdot kg^{-1}$ fresh weight.

The following criteria are used to select reference objects of the natural ecosystem: ecological significance of the object, accessibility for radioecological monitoring, exposure dose, radiosensitivity, and self-healing ability (Kryshev & Sazykina, 2012). When determining reference organisms of the Arctic seas, the availability of monitoring data is of particular importance since those allow estimating local values of the concentration factors of technogenic radionuclides in marine biota. Based on this, the following organisms were selected as reference ones for the Barents Sea: fish, cod *Gadus morhua* Linnaeus, 1758; bivalve, mussel *Mytilus edulis* Linnaeus, 1758; aquatic plant, *Fucus distichus* Linnaeus, 1767; and marine mammal, harp seal *Pagophilus groenlandicus* (Erxleben, 1777).

To determine the concentration factors of radionuclides in reference organisms of the Barents Sea and water–sediment distribution coefficients, a database was created on the content of radionuclides in its ecosystem components. The total error in determining the specific activity of radionuclides for the Barents Sea components is no more than 25 %. Most of the data were obtained as a result of expeditionary surveys under the Russian–Norwegian monitoring program for the Barents Sea in 2006–2020 (Gwynn et al., 2016 ; Jensen et al., 2016). Data for an earlier period (since 1992) were taken from the literature (Brown et al., 2004 ; Kryshev et al., 2002).

The database includes 107 values of specific activities for ^{137}Cs, 45 values for $^{239,240}Pu$, and 65 values for ^{90}Sr. The data on the radionuclide content in water, bottom sediments, and biota, obtained in one year and at one spot in the Barents Sea, were used to determine the concentration factors and distribution coefficients. Fig. 1 shows a map of the study area during the Russian–Norwegian monitoring and location of reference sampling sites.

To calculate the resulting local values of the concentration factors and distribution coefficients, statistical analysis was carried out – checking the normality of the distribution using the Shapiro–Wilk test (Kobzar, 2006). In the absence of a normal distribution, nonparametric statistics was applied (Kobzar, 2006).

The values of dose coefficients were determined using BiotaDC v.1.5.1 software tool (http://biotadc.icrp.org/) – a complement to (ICRP Publication 136, 2017). Marine organisms were approximated by ellipsoids with the following mass and ratios between the axes: cod, 2 kg, 1 / 0.2 / 0.2; harp seal, 130 kg, 1 / 0.24 / 0.24; mollusc, $1.64 \cdot 10^{-2}$ kg, 1 / 0.5 / 0.5; and aquatic plant, $6.5 \cdot 10^{-3}$ kg, 1 / 0.01 / 0.01. As the threshold values of the dose rate for biota, the following ones were set: 10 $mGy \cdot day^{-1}$ for molluscs; 1 $mGy \cdot day^{-1}$ for fish and aquatic plants (brown algae); and 0.1 $mGy \cdot day^{-1}$ for long-lived marine mammals (ICRP Publication 108, 2008 ; ICRP Publication 124, 2014). The values of the indicators τ_w and τ_s were conservatively taken equal to 0.5 for fish, aquatic plants, and marine mammals; for molluscs, the value $\tau_s = 1$ was accepted.

The value of the indicator α_i (the quality factor associated with the relative biological effectiveness of radiation) for ^{137}Cs and ^{90}Sr is equal to 1. As shown in (Sazykina & Kryshev, 2016), the quality factor of α-emitting radionuclides for biota averages 15. At the same time, it was found that for various α-emitting radionuclides, the values of the quality factor differ significantly: for radium isotopes, $\alpha_i = 5$; for plutonium and americium isotopes, $\alpha_i = 50$. Taking into account this result, when determining the control levels of ^{239}Pu in the Barents Sea water and bottom sediments, we used the value $\alpha_i = 50$.

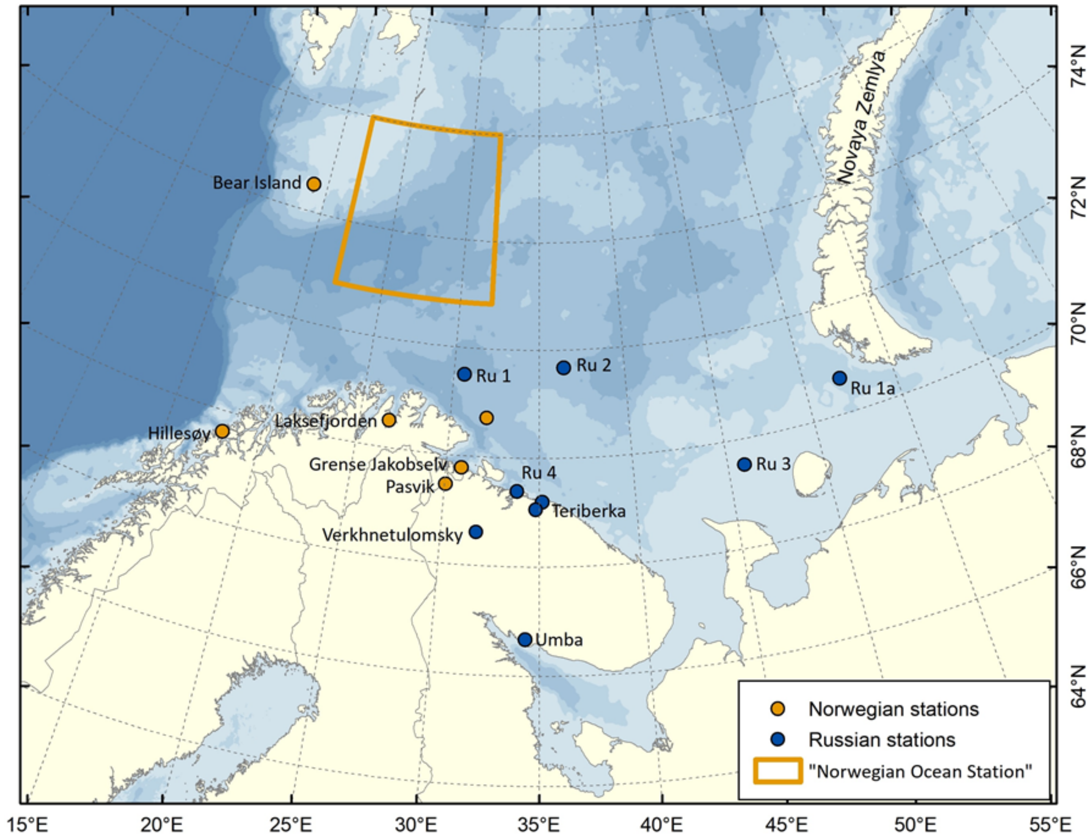

Fig. 1. Map of the study area and location of the reference sampling sites

RESULTS AND DISCUSSION

As a result of the radioecological database analysis, the concentration factors of ^{137}Cs, ^{90}Sr, and ^{239,240}Pu were determined in reference objects of the Barents Sea biota. The calculation results are given in Table 1. For comparison, it includes the values of the concentration factors of these radionuclides from the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015) based on the IAEA publication (Sediment Distribution Coefficients, 2004) and not taking into account regional specifics.

For fish, molluscs, and aquatic plants of the Barents Sea, the values of ^{137}Cs concentration factors are in good agreement (see Table 1) with the generalized global mean values from the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015), while for marine mammals, the local value of ^{137}Cs concentration factor is 3.3 times lower. For all reference organisms of the Barents Sea, the concentration factors of ^{90}Sr are 2–7 times higher than the generalized global average values. In the Barents Sea fish, the concentration factor of ^{239,240}Pu is higher than the value specified in the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015); in molluscs and aquatic plants, it is significantly lower.

Table 1. Long-term mean values of the concentration factors of radionuclides in the Barents Sea biota in 1992–2020

Radionuclide	Concentration factor according to monitoring data, L·kg^{-1}	Two-sided confidence interval (95 %), T1, T2	Concentration factor according to the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015), L·kg^{-1}
Fish			
^{137}Cs	93	76, 129	100
^{90}Sr	12	10, 27	3
^{239,240}Pu	262	197, 1,000	100
Molluscs			
^{137}Cs	51	26, 258	60
^{90}Sr	21	7, 56	10
^{239,240}Pu	1,180	912, 4,333	3,000
Aquatic plants			
^{137}Cs	69	58, 76	50
^{90}Sr	19	10, 31	10
^{239,240}Pu	732	449, 1,724	4,000
Marine mammals			
^{137}Cs	63	36, 86	210
^{90}Sr	14	1, 26	2
^{239,240}Pu	222	111, 333	280

The water–sediment distribution coefficients for the Barents Sea, determined as a result of the analysis of the radioecological database, are given in Table 2 in comparison with the values of this indicator from the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015) which are global mean values for marine ecosystems.

Table 2. Long-term mean values of the radionuclide water–sediment distribution coefficients in the Barents Sea in 2006–2020

Radionuclide	Distribution coefficient according to monitoring data, L·kg^{-1}	Two-sided confidence interval (95 %), T1, T2	Distribution coefficient according to the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015), L·kg^{-1}
^{137}Cs	426	362, 640	3,000
^{90}Sr	443	180, 720	1,000
^{239,240}Pu	189,600	56,360, 318,310	100,000

The average values of the water–sediment distribution coefficients of ^{137}Cs and ^{90}Sr for the Barents Sea according to the monitoring results (see Table 2) are lower than the values specified in the recommendations (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015) by 7 and 2.3 times, respectively. The mean water–sediment distribution coefficient of ^{239,240}Pu for the Barents Sea is 1.9 times higher than the value recommended in (Otsenka radiatsionno-ekologicheskogo vozdeistviya, 2015); however, it has a wide confidence interval.

The concentration factors of technogenic radionuclides in the Barents Sea fish do not have a pronounced trend or a clear tendency to increase or decrease (Figs 2 and 3). This may indicate the establishment of equilibrium in the distribution of radioactivity between the components of the Arctic marine ecosystem.

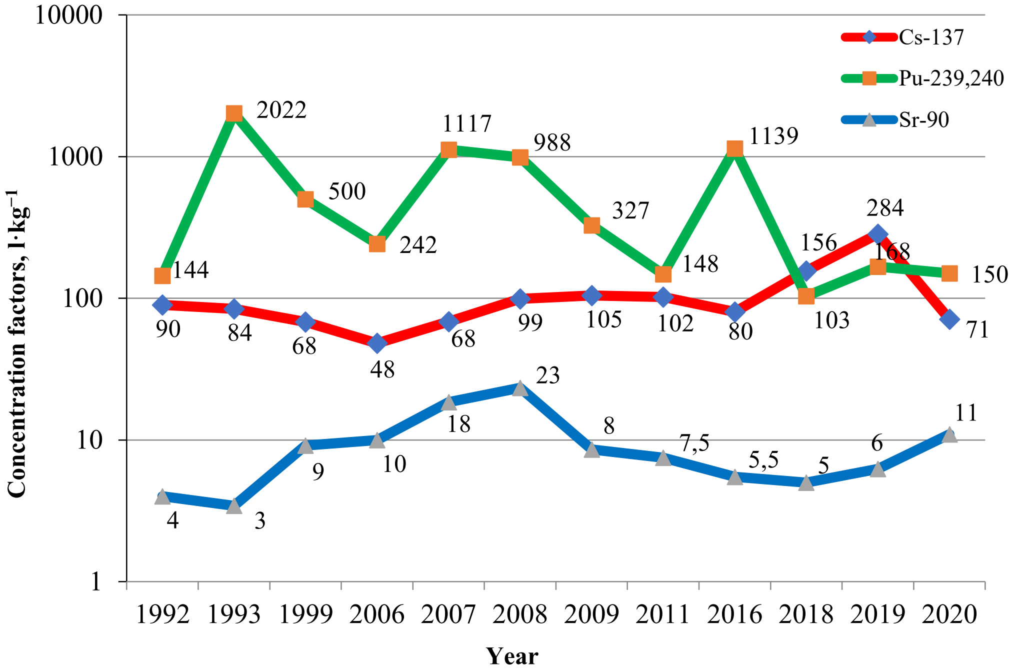

Fig. 2. Dynamics in the concentration factors of radionuclides in the Barents Sea fish in 1992–2020

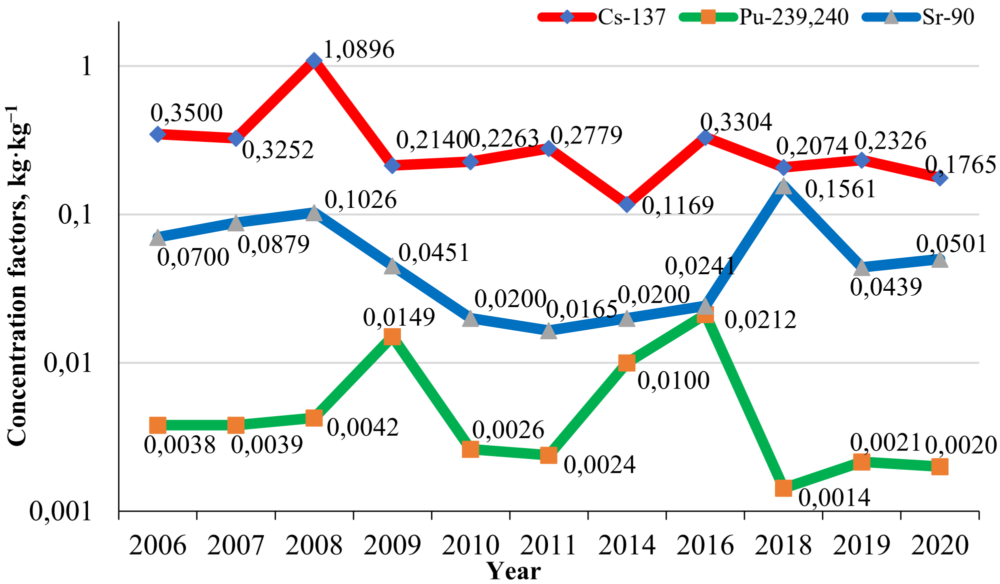

Fig. 3. Dynamics in the concentration factors of radionuclides in the Barents Sea fish from bottom sediments in 2006–2020

The calculated values of the concentration factors and distribution coefficients were used to determine the control levels of ^{137}Cs, ^{90}Sr, and $^{239,240}Pu$ in the Barents Sea water and bottom sediments. Control levels for each reference organism inhabiting the Barents Sea are given in Table 3.

Table 3. Control levels of radionuclides in the Barents Sea water and bottom sediments ensuring the radiation safety of the reference organisms

Organism	Control level in water, Bq·L^{-1}			Control level in bottom sediments, Bq·kg^{-1} fresh weight		
	^{137}Cs	^{90}Sr	^{239,240}Pu	^{137}Cs	^{90}Sr	^{239,240}Pu
Fish	$8{,}78 \cdot 10^{2}$	$4{,}37 \cdot 10^{3}$	$1{,}05 \cdot 10^{0}$	$3{,}75 \cdot 10^{5}$	$1{,}94 \cdot 10^{6}$	$2{,}00 \cdot 10^{5}$
Mollusc	$5{,}53 \cdot 10^{3}$	$1{,}45 \cdot 10^{4}$	$2{,}34 \cdot 10^{0}$	$2{,}35 \cdot 10^{6}$	$6{,}41 \cdot 10^{6}$	$4{,}43 \cdot 10^{5}$
Aquatic plant	$9{,}20 \cdot 10^{2}$	$1{,}36 \cdot 10^{3}$	$3{,}77 \cdot 10^{-1}$	$3{,}93 \cdot 10^{5}$	$6{,}03 \cdot 10^{5}$	$7{,}14 \cdot 10^{4}$
Marine mammal	$1{,}15 \cdot 10^{2}$	$4{,}39 \cdot 10^{2}$	$1{,}24 \cdot 10^{-1}$	$4{,}89 \cdot 10^{4}$	$1{,}94 \cdot 10^{5}$	$2{,}36 \cdot 10^{4}$

As the resulting control level of the radionuclide content in the Barents Sea water, its lowest value among all the values for reference organisms is taken: this ensures the safety of the most vulnerable link in the ecosystem. As seen from Table 3, the lowest values of control levels of all radionuclides are those for a marine mammal (harp seal). The control levels of technogenic radionuclides in the Barents Sea water ensuring the safety of marine biota (Bq·L^{-1}) are as follows: 115 for ^{137}Cs, 439 for ^{90}Sr, and 0.124 for ^{239,240}Pu. Reference levels of technogenic radionuclides in the Barents Sea bottom sediments (kBq·kg^{-1} fresh weight) are estimated as 48.9 for ^{137}Cs, 194 for ^{90}Sr, and 23.6 for ^{239,240}Pu.

For the Barents Sea, the contamination indices for water I_w and bottom sediments I_s were calculated by the formulas:

$$I_w = \sum_i \frac{C_{w,i}}{C_i}; \; I_s = \sum_i \frac{C_{s,i}}{S_i}, \quad (3)$$

where $C_{w,i}$ is the volumetric activity of the *i*-th radionuclide in seawater, Bq·L^{-1};

$C_{s,i}$ is the specific activity of the *i*-th radionuclide in bottom sediments, Bq·kg^{-1} fresh weight.

Fig. 4 shows the dynamics of the contamination indices for the Barents Sea water and bottom sediments by technogenic radionuclides in 2006–2020.

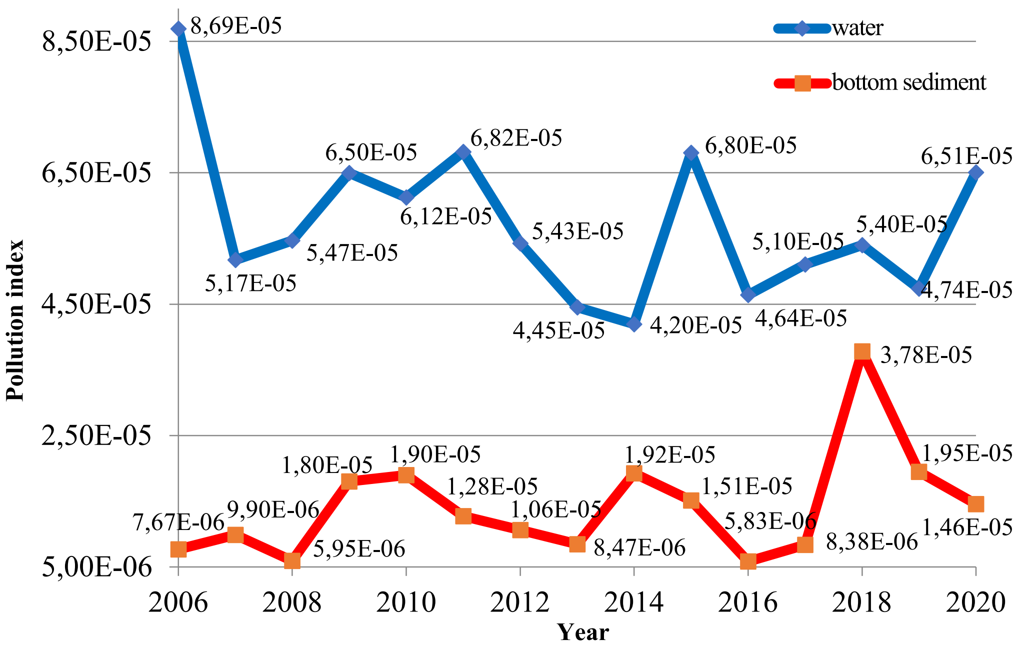

Fig. 4. Dynamics in contamination indices for the Barents Sea water and bottom sediments in 2006–2020

Fig. 4 shows that the contamination index for the Barents Sea water ranged from $4.2 \cdot 10^{-5}$ (2014) to $8.7 \cdot 10^{-5}$ (2006). The contamination index for the Barents Sea bottom sediments varied from $5.8 \cdot 10^{-6}$ (2016) to $3.8 \cdot 10^{-5}$ (2018). The contamination indices for both water and bottom sediments of the Barents Sea in 2006–2020 did not tend to increase or decrease.

The relative contribution of ^{137}Cs, ^{90}Sr, and ^{239,240}Pu to the contamination index for the Barents Sea water in 2006–2020 is shown in Fig. 5. The largest contribution to the contamination index for water was made by ^{239,240}Pu – 49–75 % in different years. Interestingly, the contribution of ^{137}Cs to the contamination index for the Barents Sea water varied within 16–41 %; the contribution of ^{90}Sr varied within 5–15 %. The contamination index for the Barents Sea bottom sediments is almost completely (up to 90 %) determined by the contribution of ^{137}Cs. In 2006–2020, the ratio of the contributions of technogenic radionuclides to the contamination index for the Barents Sea water and bottom sediments remained stable, without a noticeable tendency to change.

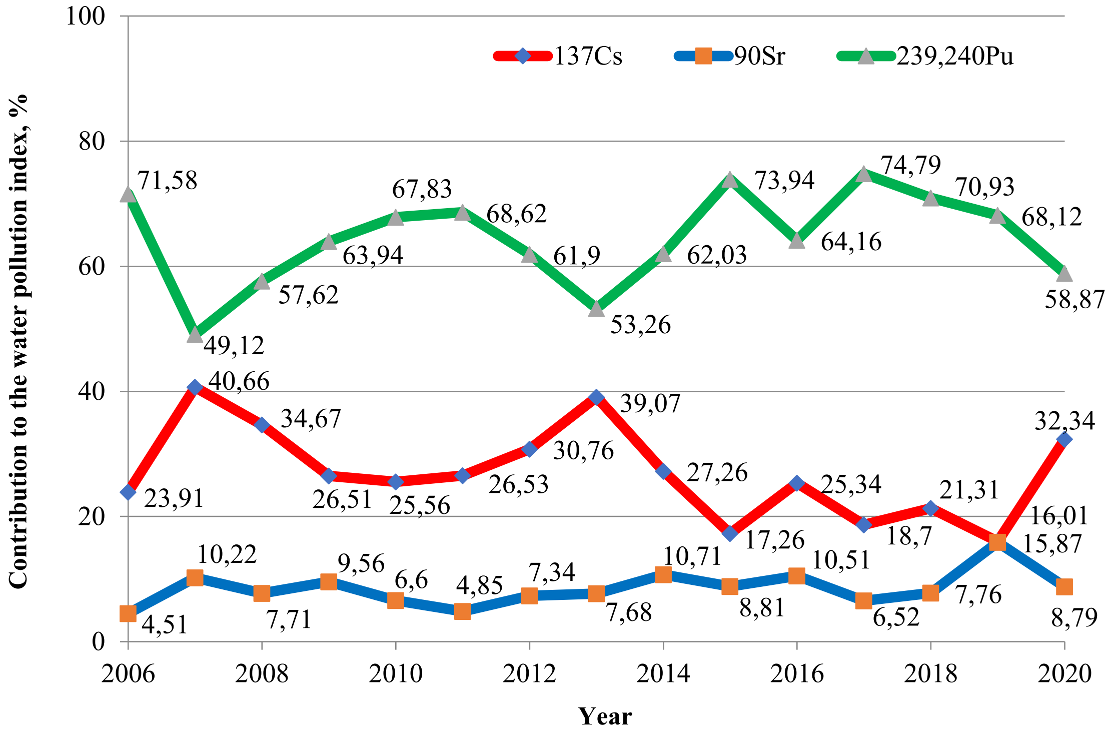

Fig. 5. Dynamics in the relative contribution of radionuclides to the contamination index of the Barents Sea water in 2006–2020

Conclusion. The concentration factors of technogenic radionuclides in the Barents Sea biota and the water–sediment distribution coefficients were determined using long-term observational data. According to the monitoring results, the water–sediment distribution coefficients of ^{137}Cs and ^{90}Sr for the Barents Sea are 7 and 2.3 times lower, respectively, than the global average values. For most of reference organisms of the Barents Sea, the values of the concentration factors of technogenic radionuclides differ from the reference values. The absence of a pronounced temporal trend for the concentration factors of all technogenic radionuclides in the Barents Sea fish in 1992–2020 indicates the establishment of equilibrium in the distribution of radioactivity between the components of the Arctic marine ecosystem.

Control levels of technogenic radionuclides in the Barents Sea water and bottom sediments are calculated, the non-exceeding of which ensures the safety of marine organisms. The control levels of radionuclides in the Barents Sea water ($Bq \cdot L^{-1}$) are 115 for ^{137}Cs, 439 for ^{90}Sr, and 0.124 for $^{239,240}Pu$. In the Barents Sea bottom sediments ($kBq \cdot kg^{-1}$ fresh weight), the values are 48.9 for ^{137}Cs, 194 for ^{90}Sr, and 23.6 for $^{239,240}Pu$.

In 2006–2020, the values of the contamination index for both water and bottom sediments of the Barents Sea, calculated using monitoring data and values of control levels, were several orders of magnitude lower than 1 and did not tend to increase or decrease. In terms of impact on natural biota, the main contribution to the contamination index for the Barents Sea water is made by $^{239,240}Pu$ (up to 75 %); to the contamination index for bottom sediments, by ^{137}Cs (up to 90 %). The ratio of the contributions of technogenic radionuclides to the contamination index of the Barents Sea water and bottom sediments is currently stable.

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ОПРЕДЕЛЕНИЕ В ВОДЕ И ДОННЫХ ОТЛОЖЕНИЯХ БАРЕНЦЕВА МОРЯ КОНТРОЛЬНЫХ УРОВНЕЙ СОДЕРЖАНИЯ РАДИОНУКЛИДОВ, ОБЕСПЕЧИВАЮЩИХ ПРИЕМЛЕМЫЙ ЭКОЛОГИЧЕСКИЙ РИСК

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Разработаны критерии оценки радиационной обстановки, обеспечивающие приемлемый экологический риск, — контрольные уровни содержания радионуклидов в компонентах природной среды. Метод применён для расчёта контрольных уровней содержания техногенных радионуклидов в воде и донных отложениях Баренцева моря, непревышение которых обеспечивает радиационную защиту морской биоты. Для этого выполнена оценка локальных коэффициентов накопления ^{137}Cs, ^{90}Sr и ^{239}Pu в биоте Баренцева моря с использованием многолетних данных наблюдений, а также выбраны представительные виды его экосистемы — рыба (треска), моллюск (мидия), водное растение (фукус) и морское млекопитающее (гренландский тюлень). Значения коэффициентов накопления ^{137}Cs, ^{239}Pu и ^{90}Sr составили соответственно (л·кг$^{-1}$): в рыбе — 93, 262 и 12; в моллюсках — 51, 1180 и 21; в водорослях — 69, 732 и 19; в морских млекопитающих — 63, 222 и 14. Значения коэффициентов распределения радионуклидов ^{137}Cs, ^{239}Pu и ^{90}Sr между водой и донными отложениями составили 426, 189 600 и 443 л·кг$^{-1}$ соответственно. Для большинства представительных организмов Баренцева моря значения коэффициентов накопления техногенных радионуклидов отличаются от справочных значений. Отсутствие выраженного временнóго тренда для коэффициентов накопления всех техногенных радионуклидов в рыбе Баренцева моря в 1992–2020 гг. указывает на установление равновесия в распределении радиоактивности между компонентами арктической морской экосистемы. Контрольные уровни содержания радионуклидов в воде Баренцева моря (Бк·л$^{-1}$) составляют 115 для ^{137}Cs, 439 для ^{90}Sr, 0,124 для ^{239,240}Pu; в донных отложениях (кБк·кг$^{-1}$ сырого веса) — 48,9 для ^{137}Cs, 194 для ^{90}Sr, 23,6 для ^{239,240}Pu. Значения индекса загрязнения как воды, так и донных отложений Баренцева моря, рассчитанные с использованием данных мониторинга и величин контрольных уровней, в 2006–2020 гг. были на несколько порядков ниже единицы и не имели существенной тенденции к повышению или снижению. Основной вклад в индекс загрязнения воды Баренцева моря с точки зрения воздействия на природную биоту вносит ^{239,240}Pu (до 75 %), в индекс загрязнения донных отложений — ^{137}Cs (до 90 %). Соотношение вкладов техногенных радионуклидов в индекс загрязнения воды и донных отложений Баренцева моря в настоящее время является постоянным.

Ключевые слова: Арктика, Баренцево море, вода, донные отложения, биота, контрольный уровень, коэффициент накопления, коэффициент распределения, радионуклид