Current state and trends in the content of ^{137}Cs and ^{90}Sr in abiotic and biotic components of Arctic ecosystems (Barents and Kara Seas case study)

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Current state and trends in the content of ^{137}Cs and ^{90}Sr in abiotic and biotic components of Arctic ecosystems (Barents and Kara Seas case study)

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Abstract. This publication summarizes long-term data on radioactive contaminants in seawater, bottom sediments, and marine fish and describes current patterns in radionuclide distribution using data collected by the Murmansk Marine Biological Institute during cruises to the Barents and Kara Seas in 2015–2017. During the study period, no short-lived artificial radionuclides were found in the Barents and Kara Seas. ^{137}Cs and ^{90}Sr isotopes with a longer half-life are still registered in Atlantic waters that are the main source of radioactive contamination in these Arctic seas. Volumetric activity of ^{137}Cs and ^{90}Sr was compared at different depths. A uniform distribution of ^{137}Cs in the vertical column of water and a tendency to increase the volume activity of ^{90}Sr from surface to bottom were observed in the Barents Sea. Bottom sediments in the Kara Sea contain on average 2 times more ^{137}Cs than the Barents Sea. Atlantic cod is characterized by an extremely low concentration of ^{137}Cs in muscular tissue, about an order of magnitude lower than in the period of maximum contamination of Barents Sea waters observed in 1982. Currently there is a decreasing trend in concentrations of anthropogenic radioisotopes in the environment and biota. Processes of ^{137}Cs and ^{90}Sr removing from biotic and abiotic components are described with exponential functions. Due to natural marine purification processes, the decrease of ^{137}Cs and ^{90}Sr concentrations in the Barents and Kara Seas occur 2–4 times faster than the physical radioactive decay of these radionuclides.

1. Introduction

Since the radioactive contamination has started historically, technogenic radionuclides enter the Barents and Kara seas from various sources: atmospheric deposition of nuclear weapons test products in the 1950s–1960s, accidents at the Chernobyl Nuclear Power Plant in 1986 and at the Fukushima-1 NPP in 2011, income of artificial radionuclides with the discharge of large rivers, discharge of liquid and disposal of solid radioactive waste, emergencies in the operation of ships with nuclear power plants, etc. [1, 2, 3, 4].

After the ban on the testing of nuclear weapons in three environments, the main focus of studying the radioecological state of the environment and assessing possible risks in the Arctic region is the issues of spatial and temporal dynamics of regional pollution formed during the nuclear era. Despite many atmospheric and sub-sea nuclear tests conducted by different countries in the middle of the

XXth century, concentrations of technogenic radioisotopes in the Barents and Kara Seas in the 1950s–1970s were relatively low. A significant increase in the level of the radioactive contamination of both seas occurred in the late 1970s and early 1980s. The cause of the pollution was an influx of high-level waste discharged into the Irish Sea into the complex of the Sellafield nuclear fuel processing plant (Great Britain), which, through the current system, flowed into the Barents and Kara Seas.

In the 1990s, due to the introduction of sewage treatment facilities in radiochemical plants located in Western Europe, the content of anthropogenic isotopes in the waters of the Arctic seas decreased down to background values. However, ^{137}Cs and ^{90}Sr are continued to be found in bottom sediments and biota, which indicates the occurrence of these elements, which are not typical for marine ecosystems, in the total cycle of matter.

Analyzing publications on radio-ecological studies conducted in the Barents and Kara seas in different years we have distinguished two types of studies that significantly differ in their geographical scale.

The first type is local studies performed in the areas of the Barents and Kara seas associated with high potential danger and public concern, such as (1) Chernaya Bay area – the bay in the Barents Sea at the southwestern tip of Yuzhny (Southern) Island of Novaya Zemlya where nuclear weapon was tested in the 1960s [5], (2) sites for disposal of solid radioactive waste in the Kara Sea [6], (3) the continental shelf area of the Murman Coast where the submarine «Kursk» sunk [7, 8], (4) Andreeva Bay where a temporary radioactive waste storage facility is located [9, 10].

The second type is regional studies conducted in the Barents and Kara seas at a considerable distance from sources of radioactive contamination [1, 2, 11, 12, 13].

The purpose of this study is to assess the current state of environmental pollution, including biota, in the Barents-Kara Region, to describe patterns of redistribution of ^{137}Cs and ^{90}Sr, and to identify vectors of radionuclide accumulation in the Arctic water bodies.

2. Materials and methods

2.1. Field sampling

Water samples from different water layers and surface sediments (0–2 cm layer) were collected during cruises to the Barents and Kara seas of the research vessel «Dalnie Zelentsy» and ships of opportunity (diesel-electric ships «Talnakh», «Nadezhda», and «Norilsk Nikel») in 2015–2017. 132 water samples and 71 samples of the bottom sediments were collected in the Barents Sea, 26 and 15 samples in the Kara Sea, respectively. Anfezh cellulose inorganic sorbent was used for the concentration of ^{137}Cs from seawater. The sample volume was at least 100 litres [14].

Cod specimens were caught on July 24, 2018, in the coastal area of Kola Bay (near Polarny city, Murmansk Oblast, Russia), 69º10′05″ N, 33º28′49″ E. Fish were caught on the bait. Sex, age, length, and weight were determined for each specimen. The sex was determined according to the gonad state. Fish age was determined by otolith analysis [15]. The specific radionuclide activity of ^{137}Cs has been determined for 40 specimens.

2.2. *Radiometric analysis*

Radiological analysis of samples was performed in the laboratory of MMBI. ^{137}Cs and ^{90}Sr were measured in the samples of water and bottom sediments, in fish, it was ^{137}Cs only. Measurements of the radionuclide activity were made on the γ-spectrometer «InSpector-2000» and the multichannel γ-spectrometer «b13237» with pure germanium detectors («Canberra», USA) for measuring X-ray and gamma radiation. Spectra were analyzed using the basic software «Genie-2000». Measurements were carried out in the 1-L Marinelli-style containers, as well as in the 0.1-L and 0.01-L containers. The measurement time for each sample was at least 24 hours. Analysis of the ^{90}Sr isotope activity was carried out by a radiochemical method [16]. The ^{90}Sr isotope activity was determined by measuring the activity of the equilibrium ^{90}Y in the multi-purpose scintillation counter «LS-6500» («Beckman Instruments Inc.», USA).

2.3. Long-term data analysis

Data on long-term (1962–2005) dynamics of the volumetric activity of ^{137}Cs and ^{90}Sr in waters were obtained from published sources [3, 17, 18, 19, 20, 21, 22, 23, 24] and from an MMBI database (2005–2017).

Bottom sediments of the Barents and Kara seas have been studied for the specific activity of ^{137}Cs and ^{90}Sr since 1982 [17]. To construct curves of the long-term variability of levels of these isotopes in bottom sediments we used MMBI data collected from 1993 to 2017 during cruises to the Barents and Kara seas carried out annually.

The analysis of long-term dynamics of radioactive contaminants in the Atlantic cod, an indicator species for the Barents Sea, was carried out using published [22] and MMBI data (2001−2017).

3. Results and discussion

3.1. Radioactive contamination of seawater

In 2015−2017, no short-lived artificial gamma-nuclides were detected in waters of the Barents and Kara Seas, which may be evidence of absence of recent radionuclide releases from pollution sources. The volumetric activity of ^{137}Cs in the Barents Sea varied from 0.1 Bq·m^{-3} to 4.3 Bq·m^{-3}. The range of measured activities of ^{90}Sr was 0.1–9.2 Bq·m^{-3} (Figs. 1, 2). It was found that the Atlantic waters still contain maximum concentrations of radionuclides. The main stream of cesium along with the Atlantic waters enters the Barents Sea through the North Cape–Medvezhiy Cape. The maximum volumetric activity of ^{137}Cs was observed in the upper layers of the branches of the North Cape Current: the Northern, Central and Murmansk branches. This phenomenon is apparently explained by the peculiarities of the hydrodynamics of water and the discharge of pollutants into the Irish Sea. The discharge from the Sellafield plant comes from the Irish Sea into the upper layers of the high-saline oceanic North Atlantic Current and spread further on. On the Kola Meridian transect, in the area located closer to the center of the Barents Sea, the dilution of the incoming flow of cesium is observed. The volumetric activity of ^{137}Cs is reduced by 0.5–1.0 Bq·m^{-3} due to the admixture of the Atlantic waters in the total volume of the Barents Sea waters.

The range of ^{137}Cs activity in the Kara Sea varied from the minimum detected level (<0.4 Bq·m^{-3}) up to 4.3 Bq·m^{-3}. The content of ^{90}Sr, in contrast to ^{137}Cs, varies over a wider range from 1 to 9.7 Bq·m^{-3}. Allocation of spatial differences in concentrations of ^{137}Cs in waters is difficult according to available data. A decrease in the ^{137}Cs content in water was observed in areas characterized by the shallow depths. It is known that river runoff can significantly affect radionuclides distribution in the Kara Sea. In this regard, the highest concentrations of ^{90}Sr and the lowest concentrations of ^{137}Cs are noted in shallow parts of the sea exposed to the Yenisei River flow.

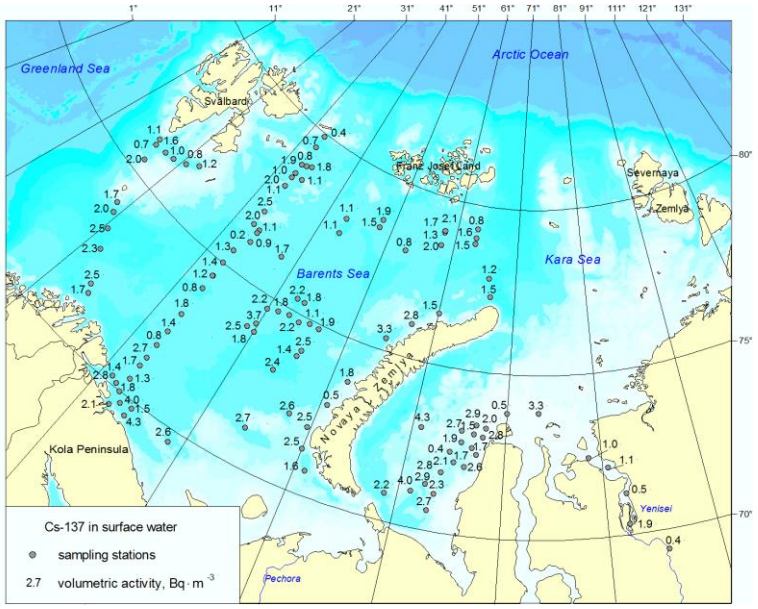

Figure 1. Distribution of ^{137}Cs in surface waters of the Barents and Kara seas, 2015–2017.

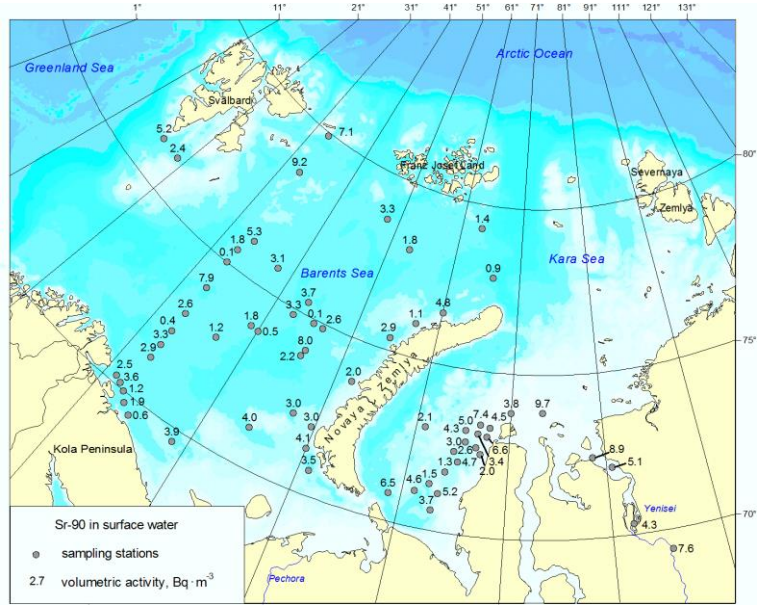

Figure 2. Distribution of ^{90}Sr in surface waters of the Barents and Kara seas, 2015–2017.

Using the Barents Sea example, the volume activity of ^{137}Cs and ^{90}Sr was compared at different depths (Table 1). In the Barents Sea, there is a uniform distribution of ^{137}Cs in the vertical column of water and a tendency to increase the volume activity of ^{90}Sr from a surface to bottom. The average level of ^{137}Cs in surface waters of the studied seas is almost the same in contrast to ^{90}Sr, which concentration in the Kara Sea is about 1.5 times higher.

Table 1. Volumetric activity of ^{137}Cs and ^{90}Sr in the Barents and Kara seas (seawater) at different depths in 2015–2017

Area	Depth, m	^{137}Cs, Bq·m^{-3}			^{90}Sr, Bq·m^{-3}		
		n	min-max	mean	n	min-max	mean
Barents Sea	0	81	0.1–4.3	1.7	41	0.1–9.2	3.0
	20–100	15	0.6–3.1	1.8	14	0.5–9.8	3.7
	100–200	14	0.9–4.7	2.2	12	0.2–8.9	3.2
	200–300	15	0.9–2.7	1.7	7	0.8–6.9	4.7
	300–400	6	1.2–2.3	1.7	3	0.8–9.2	6.1
	> 400	2	2.0–2.5	2.3	–	–	–
Kara Sea	0	26	0.4–4.3	2.1	24	0.5–9.7	4.5

n – number of samples

Multi-year data on levels of artificial radionuclides in seawater, bottom sediments, and fish in the Barents and Kara Seas are summarized in Figure 3. An increase in ^{137}Cs in Barents Sea waters (up to 45 Bq·m^{-3}) occurred in 1979–1980 [1, 22]. Since that time, long-term dynamics of changes in ^{137}Cs volumetric activity has been declining exponentially and corresponds to the known trends of the overall decrease of pollution of the Barents Sea by artificial radionuclides. The exponential function, with a high degree of confidence (R^2 = 0.91) describing the observed trend, is $y = 36.906e^{-0.099x}$, hereafter x is the number of years that have passed after the maximum pollution (Fig. 3a). The corresponding average environmental half-life ($T_{1/2}$) of ^{137}Cs in water of the Barents Sea is about 7.0 years. The decrease of the volumetric activity of ^{90}Sr after the maximum pollution is less clearly expressed (R^2 = 0.58) and is described by the equation $y = 9.9354e^{-0.04x}$, $T_{1/2}$ is 17.3 years (Fig. 3a).

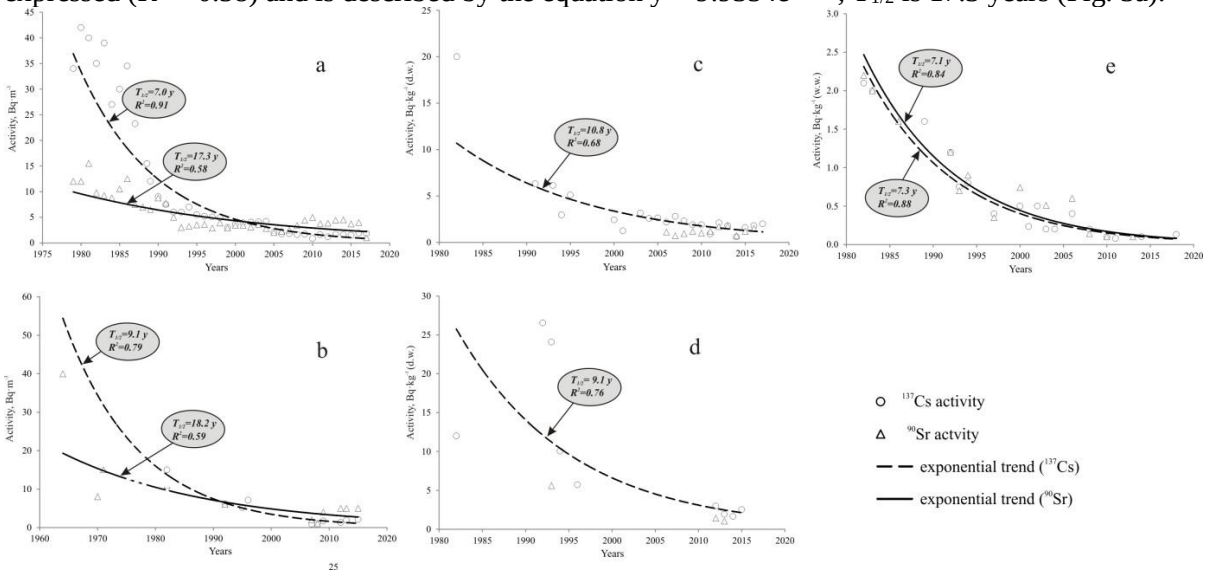

Figure 3. Long-term dynamics of ^{137}Cs and ^{90}Sr activities in: a – Barents Sea waters, 1979–2017, Bq·m^{-3}; b – Kara Sea waters, 1964–2015, Bq·m^{-3}; c – Barents Sea, surface layer of bottom sediments, 1982–2017, Bq·kg^{-1}; d – Kara Sea, surface layer of bottom sediments, 1982–2015, Bq·kg^{-1}; e – Barents Sea, Atlantic cod, 1982–2018, Bq·kg^{-1}(w.w.).

There was also a decrease in the volume of ^{137}Cs and ^{90}Sr activities compared with previous years in Kara Sea waters in 2015–2017. Analyzing long-term data on levels of artificial radionuclides in seawater we have found that ^{137}Cs and ^{90}Sr concentrations tend to decrease, which can be described by

equations $y = 54.441e^{-0.076x}$ and $y = 19.321e^{-0.038x}$, respectively (Fig. 3b). Estimated environmental half-lives of ^{137}Cs and ^{90}Sr are 9.1 and 18.2 years in the Kara Sea.

3.2. Radioactive contamination of the bottom sediments

No short-lived radionuclides were found in bottom sediments of the Barents and Kara Seas from 2015 to 2017. ^{137}Cs and ^{90}Sr isotopes with a longer half-life were found at very low concentrations (Fig. 4, 5), which is fully consistent with previous studies [11, 12]. Pollution of bottom sediments by ^{137}Cs in open areas of the Kara Sea is on average about 2 times higher than in the Barents Sea.

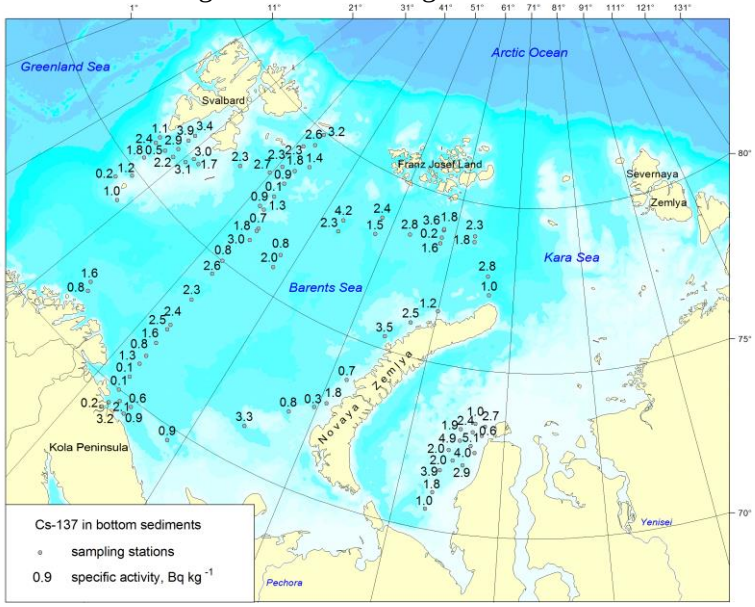

Figure 4. Distribution of ^{137}Cs in bottom sediments (0–2 cm) of the Barents and Kara seas, 2015–2017

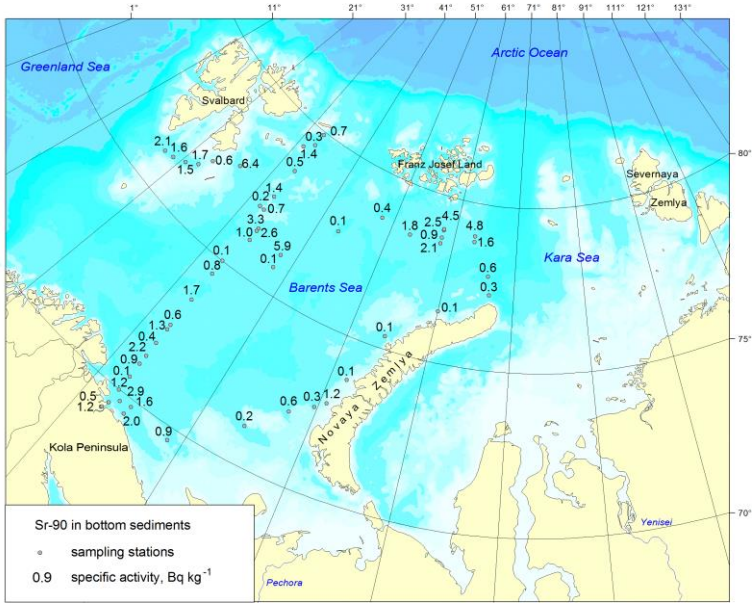

Figure 5. Distribution of ^{90}Sr in bottom sediments (0–2 cm) of the Barents and Kara seas, 2015–2017

The isotopic composition and artificial radionuclides activities in bottom sediments of the Barents and Kara seas is the subject of active study since the 1980s. Mathematical analysis of long-term data

on average ^{137}Cs activities in bottom sediments of the Barents Sea showed an exponential decrease, satisfactorily described by the equation $y = 10.688e^{-0.064x}$, $T_{1/2}$ is 10.8 years (Fig. 3c). A similar dependence for the top layer of the Kara Sea sediment has the form $y = 25.75e^{-0.076x}$, $T_{1/2}$ is 9.1 years (Fig. 3d).

The amount of ^{90}Sr measurements in precipitation of the studied seas is limited [6, 13, 17] and according to the available data, the concentrations of ^{90}Sr remain virtually unchanged in the last decade.

3.3. Radioactive contamination of biota (fish)

^{137}Cs was found in the samples of the one year old cod (0.16±0.05 Bq·kg^{-1} wet weight, further on, w.w.) and in the two year old cod (0.22±0.04 Bq·kg^{-1} w.w.); in remaining samples, the value of specific activity of the radionuclide was less than the minimum detectable value (Table 2).

Table 2**.** Specific activity of ^{137}Cs of the cod.

Age	Sex	L (min-max)/ mean, cm	W (min-max)/ mean, g	Number of samples	Specific activity of ^{137}Cs, Bq·kg^{-1} w.w.
1+	female (26.7 %)	(17.0–25.2)/19.73	(36–140)/67.25	8	< 0.10
	male (26.7 %)	(18.2–21.5)/20.11	(44–102)/69.88	8	< 0.10
	juvenile (46.7 %)	(14.9–24.0)/17.87	(27–81)/44.50	14	0.16 ± 0.05
2+	female (70.0 %)	(20.4–29.5)/25.24	(66–264)/159.86	7	0.22 ± 0.04
	male (30.0 %)	(20.6–26.2)/23.77	(61–141)/112.33	3	< 0.07

Atlantic cod is typically used as a reference object to assess the long-term dynamics of radioactive contaminants in the Barents Sea ecosystem [24]. The increase in ^{137}Cs content in the muscle tissue of cod is due to the pollution of the Barents Sea waters by discharges from the Sellafield plant (England) in the mid-1970s. The concentration of ^{137}Cs in the water reached its maximum of 45 Bq·m^{-3} in 1979. The average content of ^{137}Cs in the Barents Sea cod increased significantly only in 1982 (2.2 Bq·kg^{-1}). After reducing the discharges of artificial radionuclides by plants in Western Europe, there was a decrease in ^{137}Cs concentrations in the water of the Barents Sea with a 5-year shift, followed by a 3-year delayed decrease of its concentration in fish [22].

Fig. 3e shows average ^{137}Cs and ^{90}Sr concentrations in Barents Sea cod from the period of maximum levels of these contaminants in fish (1982) to the present. Changes are described by exponential curves (for ^{137}Cs, it is $y = 2.4692e^{-0.095x}$, for ^{90}Sr, $y = 2.3148e^{-0.097x}$). The environmental half-lives of ^{137}Cs and ^{90}Sr are 7.1 and 7.3 years, respectively (Fig. 3e).

4. Conclusions

The present-day activities of the anthropogenic isotopes in the elements of the Arctic marine ecosystems are very low. There is a stable decrease in concentrations of anthropogenic radioisotopes ^{137}Cs and ^{90}Sr in the environment and biota compared with the period of maximum pollution. The influence of global, regional, and local contamination sources (accidents at nuclear power plants, discharges from West-European radiochemical plants, nuclear-powered fleet bases, burials at Novaya Zemlya), active in the past, has become almost indistinguishable. Processes of removing ^{137}Cs and ^{90}Sr from the biotic and abiotic components are described with exponential functions with a high degree of confidence. Due to natural marine purification processes, such as dilution, sorption by sediments and suspended solid material, accumulation by aquatic inhabitants, the decrease of ^{137}Cs and ^{90}Sr concentration in the Barents and Kara Seas occurs 2–4 times faster than the physical radioactive decay of these radionuclides.

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