Nuclear Engineering and Technology 54 (2022) 3034–3042

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Influence of operation of thermal and fast reactors of the Beloyarsk NPP on the radioecological situation in the cooling pond. Part 1: Surface water and bottom sediments

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ARTICLE INFO

Article history:
Received 6 July 2021
Received in revised form
15 September 2021
Accepted 2 March 2022
Available online 5 March 2022

Keywords:
Nuclear power plant
LWGR
FNR
Surface water
Bottom sediments
Radioecological monitoring
^{60}Co
^{90}Sr
^{137}Cs
^{3}H
Long-term dynamics

ABSTRACT

The results of radioecological monitoring of the cooling pond Beloyarsk NPP (Russia) have been presented. The influence of waste technological waters of thermal and fast NPP reactors on the content of artificial radionuclides in surface waters and bottom sediments of the Beloyarsk reservoir has been studied. The long-term dynamics of the specific activity of ^{60}Co, ^{90}Sr, ^{137}Cs and ^{3}H in the main components of the freshwater ecosystem at different distances from the source of radionuclide discharge has been estimated. Critical radionuclides (^{60}Co and ^{137}Cs), routes of their entry and periods of maximum discharge of radioisotopes into the cooling pond have been determined. It is shown that the technology of electricity generation at Beloyarsk NPP, based on fast reactors, has a much smaller effect on the flow of artificial radionuclides into the freshwater ecosystem of the reservoir. During the entire period of monitoring studies, the decrease in the specific activity of radionuclides from NPP origin in surface waters was 4.3–74.5 times, in bottom sediments 10–505 times. The maximum discharge of artificial radionuclides into the reservoir was noted during the period of restoration and decontamination work aimed at eliminating emergencies at the AMB thermal reactors of the first stage of the Beloyarsk NPP.
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1. Introduction

The rapid development of nuclear power in the second half of the 20th century, associated with the construction of nuclear power plants, led to an increase in the release of artificial radionuclides into the environment. It was especially significant during the operation of the first types of reactors or as a result of radiation accidents. The imperfection at the initial stage of the formation of the nuclear industry in electricity generation technologies, numerous abnormal and emergency situations at NPPs, determined the importance of ensuring radiation safety of nuclear power plant staffs, the population and the environment in the area of radiation hazardous facilities [1–5].

Cooling reservoirs of natural or man-made origin are often built into the technological cycle of NPP reactor plants: rivers, lakes, reservoirs, seas [6–8]. Russia, being a one of the leading countries in the world in the area of nuclear energy, pays great attention to radiation safety issues. Thus, the permissible discharges of artificial radionuclides into water bodies from NPP operating in a normal mode are set at levels at which they will not form a radiation dose of the population over 50 μSv/year [9]. During normal operation, the discharge of radionuclides into water bodies from Russian NPPs is a few percent of the current standard [10]. However, there is a certain risk of such an excess in the event of deviations from the NPP conditions of normal operation and increased discharge of radionuclides into the cooling pond. Therefore, an actually direction in radioecology is the study of the accumulation and migration of artificial radionuclides in aquatic ecosystems exposed to the effects of nuclear fuel cycle enterprises [11,12]. The solution of these problems is possible only on the basis of the results of long-term radioecological monitoring, defined by the IAEA as control of the

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https://doi.org/10.1016/j.net.2022.03.004
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content of radionuclides in the environment [13–15].

For the study, the Ural region of Russia has been chosen, that is historically characterized by multicomponent pollution with artificial radionuclides due to nuclear weapons tests at the Novaya Zemlya test site, long-term discharges and accidents at the Mayak Production Association ("Mayak" PA) during the production and processing of nuclear materials, conducting nuclear explosions for peaceful purposes, mining uranium ore, Beloyarsk NPP operation [16–18].

Beloyarsk nuclear power plant named after I.V. Kurchatov is one of the oldest in Russia and the only one with power units based on different operating types. The first stage of the NPP includes two small prototype Light Water Graphite-moderated Reactors AMB-100 and AMB-200 ("Atom Peaceful Big"). The second line stage consists of two reactors BN-600 and BN-800 ("Fast Sodium"), operating on fast neutrons. The objects of research have become the main components of the freshwater ecosystem: surface water and bottom sediments of the Beloyarsk NPP cooling pond. Artificial radionuclides polluting the freshwater ecosystem as part of NPP discharges are unique markers that allow assessing the impact of a nuclear power plant on humans and the environment [12]. The aim of the work was to assess the impact of the operation of thermal and fast reactors Beloyarsk NPP on the radiation situation in the cooling pond based on the analysis of the results of long-term (1976–2019) radioecological monitoring of the reservoir.

2. Description of research objects

2.1. Description of the Beloyarsk NPP

Reactors of the AMB series of Beloyarsk NPP have helped further develop the concept of boiling channel reactors AM-1 with a graphite moderator and water cooling. The technologies tested at the AMB reactors have been used to create more powerful reactors of the RBMK and EGP-6 series. Sodium-cooled fast neutron reactors (BN) have significantly increased the using resources of nuclear fuel. The BN series of reactors has also been aimed at expanding the list and technologies for using nuclear fuel: uranium-plutonium fuel, afterburning of high-background plutonium, neptunium, americium, curium. At the same time, the formation of radioactive waste has been minimized, which brings nuclear power closer to the closure of the nuclear fuel cycle [19,20]. Currently, a new generation of this series of reactors is being designed BN-1200 (Table 1).

At the AMB reactors, for the first time, technologies and operating modes for nuclear superheating of steam, new design solutions, equipment and materials were tested. In this regard, the operation of both AMB reactors and technological equipment of the NPP was accompanied by a significant number of deviations and disruptions in operation, including emergency situations. All of them ultimately led to wear and tear of the metal structures of power units, deformation of the cells of the fuel elements and partial destruction of the graphite stack. This significantly reduced the life of the reactors, that was 17 and 21 years for AMB-100 and AMB-200, respectively. Emergency situations were also noted at the BN-600 reactor [19,21,22]. Accidents and deviations in the operation of the AMB and BN-600 reactors became the reasons for their unscheduled shutdown, repair and restoration work, decontamination of equipment and the NPP site (Table 2).

During the design and construction of the BN-600 reactor, the design flaws of the previous generation of reactors and the accumulated experience in eliminating the consequences of emergencies have been taken into account. In the BN-600 reactor the defense in depth concept has been implemented, it is based on an integrated system of barriers and technical measures to minimize the impact of emissions and discharges from nuclear power plants on humans and the environment [19,23].

Close to the Beloyarsk NPP site there is the adjoined territory of the Institute of Reactor Materials (IRM), where the pool-type reactor IVV-2M with a capacity of 15 MW has been operating since 1966. The radionuclide composition of the discharges from the IRM and the Beloyarsk NPP is similar, and the waste water system of these radiation hazardous objects forms a single complex. Therefore, when assessing the impact on humans and the environment, NPP and IRM are usually considered together [10].

Numerous deviations in operation and emerging emergencies in the initial period of work of the first two reactors of the Beloyarsk NPP led to the need to create a radioecological monitoring system for objects located on the adjacent territory and ecosystems, the most important of which are aquatic [24,25]. The largest freshwater ecosystem in the area where the NPP is located is the cooling pond of the nuclear power plant (Beloyarskoe reservoir).

2.2. Description of the cooling pond of the Beloyarsk NPP

To create a cooling system for reactors and technical water supply for Beloyarsk NPP in 1959–1963 on the river Pyshma the Beloyarsk reservoir (cooling pond) was created by building a dam in the riverbed. The cooling pond is 20 km in length with 3 km in width and located near the Beloyarsk NPP. The maximum depth of the reservoir is 24 m with 7 m on average, its water area is 38.6 km^2. The total volume of the reservoir is $2.65 \cdot 10^8$ m^3, the rate of natural runoff from it is 2.8 m^3/s [10]. The cooling pond is covered with ice from November to April, with the exception of the points of discharge of technological waters of the nuclear power plant.

Bottom sediments in the reservoir are represented by silty sapropel (mainly in the deep-water part), flooded soil, sandy-silty, silty-sandy and sandy soils (mainly in the coastal part).

2.3. The impact of liquid discharges from the Beloyarsk NPP on the environment and humans

Beloyarsk NPP is located on the left bank of the cooling pond. The water used at the nuclear power plant for the decontamination of equipment, premises, staffs and overalls is refined from radionuclides and part of it (unbalanced water) is released into the environment: through an industrial water canal into the Beloyarsk reservoir and through a household sewage system into the Olkhovka River. At a distance of 0.7 km from the industrial water canal downstream of the Beloyarsk reservoir, there is a water intake

Table 1
Description of Beloyarsk NPP reactors.

Reactor type	Unit	Coolant	MWe net, each	Start or commercial operation	Licensed to, or scheduled close	Year of fuel unloading
Light water graphite reactor (LWGR)	AMB-100	Water	108	1964	1981	1986
	AMB-200		160	1967	1989	1993
Fast neutron reactor (FNR)	BN-600	Liquid sodium	560	1980	Planned, 2030	–
	BN-800		882	2016	Planned, 2056	–
	BN-1200		1220	Planned, 2030	–	–

Table 2
Description of incidents and accidents at the reactors of the Beloyarsk NPP [19,21].

Place of incident	Date	Reason	Influence on radiation situation and environment
AMB-100	1964 −1979	Repeated destruction of fuel assemblies in the reactor core.	An increase in the background radiation at the power unit led to the need for multiple decontamination of equipment.
AMB-200	29.05.1976	Malfunctioning in the reactor cooling system. Melting of 50% of the fuel assemblies in the reactor core.	The restoration work took about 9 months. Increase in collective exposure doses for NPP staffs during repair work.
Turbine hall of the first stage of the nuclear power plant	30.12.1978	A fire in the turbine hall that caused the roof to collapse. Damage to the power supply and control system of the AMB-200 reactor.	While organizing the supply of cooling water to the emergency reactor, eight people of the NPP staffs were irradiated.
BN-600	21.01.1987	Leakage of 1 ton of sodium coolant. Temperature rise in the reactor core.	Partial destruction of 12 fuel rods. A fire at the reactor. On the INES scale - 4 hazard level.
Storage facility for liquid radioactive waste (LCW) of NPP	22.12.1992	Flooding of the room for servicing the pumps of liquid radioactive waste during pumping of liquid radioactive waste.	Part of the radioactive water got into the ground under the LCW and through the drainage system into the Beloyarsk reservoir. The total activity is - 6 Ci,^{137}Cs - 6 mCi. On the INES scale - the 3 hazard level.
BN-600	07.10.1993	Depressurization of the primary circuit auxiliary system, leakage of about 1 ton of sodium coolant.	Increased background radiation in the exhaust ventilation system. The release of radioactivity through the ventilation pipe. On the INES scale - 1 hazard level.
BN-600	06.06.1994	Leakage of sodium coolant from the secondary circuit of the reactor during overhaul.	Combustion of the coolant. On the INES scale - 1 hazard level.
BN-600	09.09.2000	Failure of the cooling system of the 3rd circuit of the reactor due to an emergency power outage.	Release of steam from the 3rd circuit of the reactor into the environment.

canal for water supply to the cooling system of the first stage of the NPP (reactors AMB-100 and AMB-200). After 2 km downstream, there is a discharge canal through which water from the NPP cooling system is discharged into the Warm Bay of the reservoir. The temperature of the waste water in the summer months is +23 … 26°C, which exceeds the temperature of water in other parts of the cooling pond by 6 … 7°C.

To exclude the discharge of liquid radioactive waste into the environment, all the water that is used in the operation of the Beloyarsk NPP and contains radionuclides is collected, purified and returned to the circulating water supply system. At the same time, during the operation of the first stage NPP (AMB-100 and AMB-200 reactors), leaks were periodically formed in this system due to both imperfect technologies and emergency situations [19]. In the first years of NPP operation in waste waters, the most radiologically significant radionuclides were ^{137}Cs (45–50% of the total activity), ^{134}Cs (30–35%), ^{60}Co (15–20%) and to some extent ^{90}Sr. The volumes of annual discharges of radionuclides of NPP origin were determined by the operating mode of power units, emergency situations and reactor repairs (Fig. 1). On the basis of the data [19], it was calculated that in some years artificial radionuclides found in the NPP waste waters determined the exposure dose to the population, which exceeded the current standard in Russia (50 μSv/year) up to 3.6 times. Artificial radionuclides were also found in bottom sediments, aquatic plants, plankton, and fish in the cooling pond. A similar radionuclide composition of waste water released into aquatic ecosystems was typical for other types of reactors, for example, on Three Mile Island Nuclear Station in USA [26].

During the operation of three power units of the Beloyarsk NPP, radioactive contamination of unbalanced waters was determined by discharges from reactors of the first stage (AMB-100 and AMB-200), but contribution in the pollution from BN-600 did not exceed 1% or 0.1–0.5 Ci/year [19,22]. Taking into account the radionuclide composition of NPP discharges, the accumulation and migration of the main radiologically significant radioisotopes in the components of the freshwater ecosystem was studied in the created system of radioecological monitoring of the cooling pond: ^{3}H ($T_{½}$ = 12.32 years), ^{60}Co ($T_{½}$ = 5.27 years), ^{90}Sr ($T_{½}$ = 28.78 years) и ^{137}Cs ($T_{½}$ = 30.07 years) [27,28]. Being discharged into the cooling pond, these radionuclides, first of all, pollute the surface water. And if tritium remains in the water to a greater extent, then the rest of the radionuclides are sorbed by bottom sediments and hydrobionts, being involved in various migration processes in the reservoir.

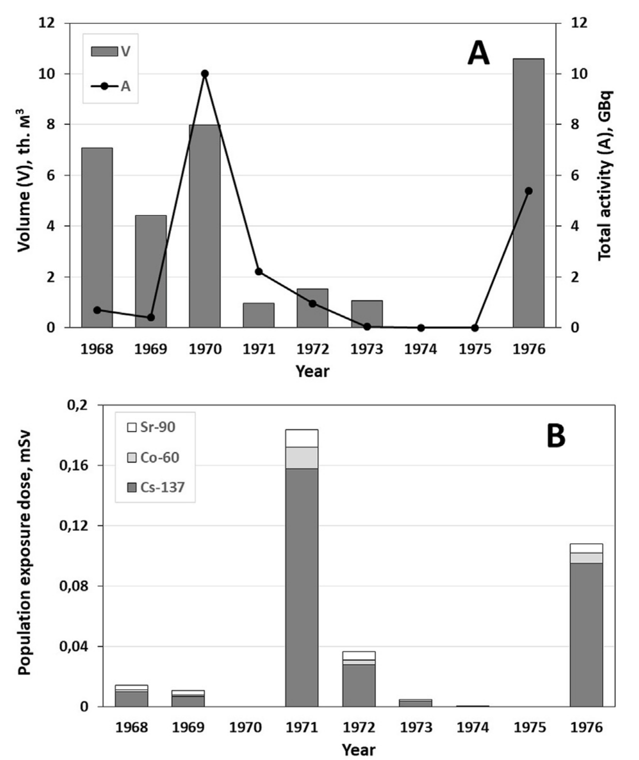

Fig. 1. Description of discharge water of Beloyarsk NPP (volume and activity) in cooling pond (A) and population exposure dose from its radionuclides (^{60}Co, ^{90}Sr и ^{137}Cs) in period 1968–1976 (B) [19].

3. Material and methods

3.1. Sites description

When creating a network of radioecological monitoring of the Beloyarsk reservoir on the cooling pond, 6 control sites were selected (Fig. 2), allowing to obtain a comprehensive information of the content of radionuclides in the components of the freshwater ecosystem and take into account the influence of the nuclear power plant:

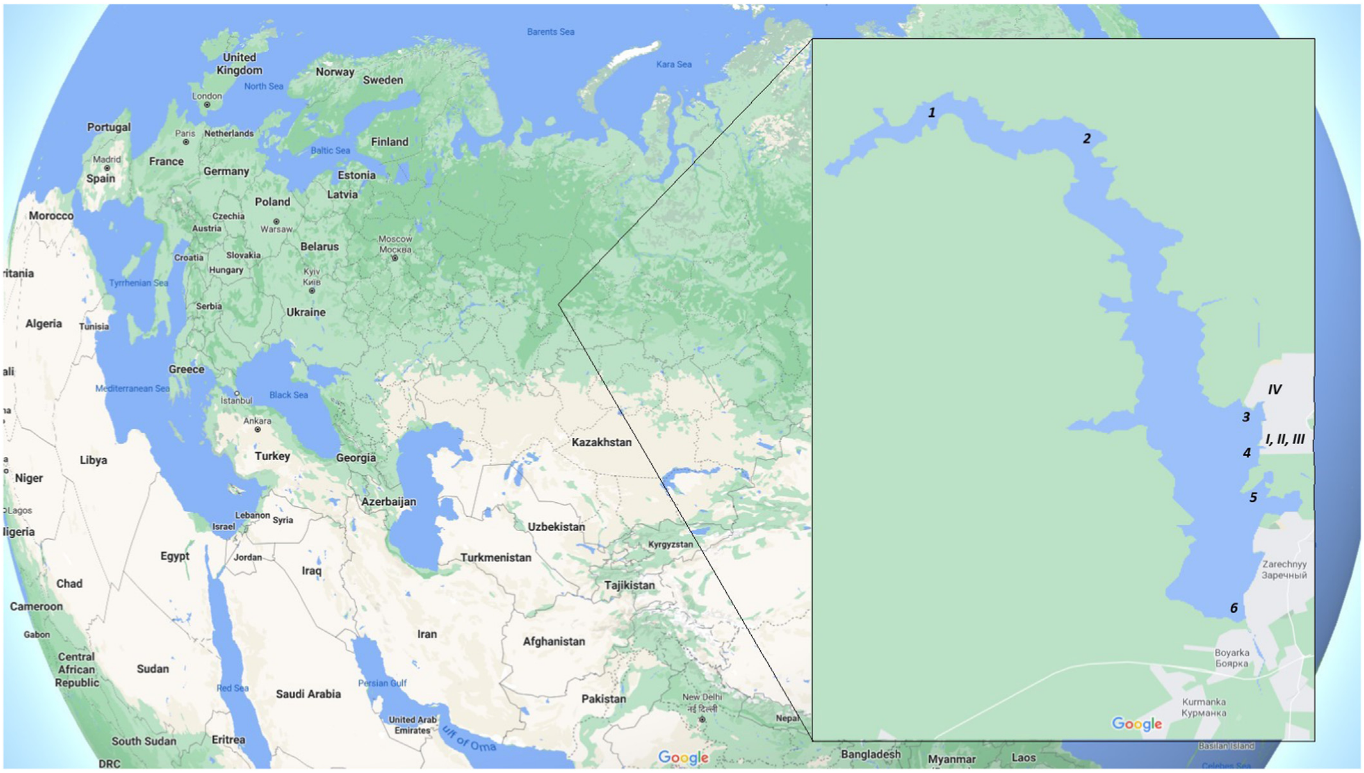

Fig. 2. Schematic map of radioecological monitoring network of cooling pond freshwater ecosystem in the vicinity of Beloyarsk NPP: *1* – The upper reaches of the reservoir, *2* – Pike Bay, *3* – Biophysical Station Area, *4* – Industrial Water Canal of NPP, *5* – Warm Bay, *6* – Dam part of the reservoir. Reactors of Beloyarsk NPP: *I* – AMB-100, *II* – AMB-200, *III* – BN-600, *IV* – BN-800 (schematic map has been created with Google Maps).

S1- Upper reaches of the reservoir outside the observation area of the Beloyarsk NPP to assess the background content of radionuclides in the cooling pond;
S2- Pike Bay for monitoring the temperature regime of the Warm Bay and assessing the effect of dilution of radionuclides upstream. At present, the closest source of pollution is the BN-800 reactor discharge canal;
S3- The area of the Biophysical Station, 300 m upstream of the Industrial Water Canal and near the Bypass Canal, which drains the area around the Beloyarsk NPP from the north. The bypass canal, 1.5–2 km long, originates near the water treatment facilities of the nuclear power plant, that discharge low-level radioactive wastewater into the Olkhovka River. Periodic leaks in the pipeline of the NPP water treatment facilities led to the releasing of radionuclides into the water of the Bypass Canal and their subsequent penetration into the cooling pond;
S4- Industrial Water Canal through which drainage and surface water flows from the site of the Beloyarsk NPP and JSC Institute of Reactor Materials;
S5- Warm Bay, where heated water from the NPP cooling system is discharged;
S6- Dam area to assess the integral discharge of radionuclides from the Beloyarsk reservoir into the river system.

3.2. Sampling of freshwater components

The sampling of the components of the freshwater ecosystem, sample preparation and measurement of the content of radionuclides in them were carried out in accordance with the regulatory and methodological documents in force in Russia [29–32]. Samples were taken in 2–3 replicates in the summer-autumn period of the year [24].

Surface water was sampled in a volume of 200 L for repetition. Water samples were acidified to prevent the sorption of radionuclides on the vessel walls, filtered, and then evaporated. The dry residue was ashed at t = 450 °C in a muffle furnace for 8 h. After cooling, the residue was ground with a pestle to a fine powder.

Bottom sediments were sampled using a special sampler with a cross-sectional area of 38.5 cm^2 to a depth of 20–30 cm. Samples of 2–3 kg of wet weight were dried to an air-dry state, ground, sifted through a sieve with a diameter of 1 mm, and then ashed at t = 400–500 °C in a muffle furnace for 6 h to remove organic matter.

3.3. Measurements

In the components of the freshwater ecosystem, γ-emitting radionuclides were determined by instrumental methods. In the 70–80s, measurements were carried out on an AI-256-6 multichannel amplitude analyzer with a scintillation NaI(Tl) detector of the “Lemon” type with a counting error of 5–10%. Further, on a low-background semiconductor gamma-spectrometer from Ortec (USA) with a coaxial detector system based on high-purity germanium (HPGe) and a gamma-spectrometer from Canberra Packard (USA) with a germanium semiconductor detector with an error measurement no more than 15% and a lower detection limit of 1 Bq/kg.

The method for the determination of ^{90}Sr is based on the leaching of the radionuclide with 6 N hydrochloric acid, the isolation of ^{90}Sr in the form of oxalates, the separation of ^{90}Y from ^{90}Sr, and the radiometric measurement of the obtained preparations. The chemically pure precipitate of strontium oxalate was dried, calcined, weighed, and β-activity was measured in the 70–80s on a counting device of the PST-100 and UMF-1500 type with an SBT-13 end counter with a counting error of 5–10%. Further, on a low-background device UMF-2000 (Russia) with a lower detection limit of 0.4 Bq/kg and a statistical measurement error of no more than 10%.

In the quantitative determination of ^{3}H, water samples were preliminarily enriched by the method of one-stage electrolysis with one or two toppings. The method is based on a significant difference in the rate of release of light (protium) and heavy (deuterium and tritium) hydrogen isotopes during the discharge of ions at the cathode during the electrolytic decomposition of water. The molecular hydrogen released in this case is enriched in protium, and the electrolyte, respectively, in tritium and deuterium. There is little deuterium in the samples, so it can be neglected in the quantitative determination of tritium. Samples were counted on a Delta-300 device (USA). The tritium concentration was determined by a relative method, by comparison with a standard solution.

Calculation of the quantitative content of radionuclides in bottom sediments was carried out on a dry basis, as is customary in the literature.

3.4. Data analysis

Radioecological monitoring of the Beloyarsk reservoir was carried out in the most detailed way in 1976–1988 and in 2002–2019. A total of 834 statistically processed results of measurements of the content of radionuclides in the components of the freshwater ecosystem were analyzed, including:

- surface water - 573 measurements (^{60}Co – 14%, ^{90}Sr – 24%, ^{137}Cs – 28% и ^{3}H – 34%);
- bottom sediments - 261 measurements (^{60}Co – 33%, ^{90}Sr – 22%, ^{137}Cs – 45%).

The reliability of the results was achieved by parallel selection and examination of all samples in 2–3 replicates. Statistical processing of the data obtained was to determine the arithmetic mean and standard deviation of the arithmetic mean. The measurement results were processed using the t-criterion and other generally accepted methods (Mann - Whitney *U* test) and were considered reliable at $p < 0.05$. The results obtained were verified with the official data of the periodic radiation monitoring of Roshydromet carried out in the area of the Beloyarsk NPP [10]. The results of the analysis of monitoring data are presented in the form of the dynamics of the specific activity of radionuclides in surface water and bottom sediments separately for each sampling site.

4. Results and discussion

4.1. Dynamics of radionuclides specific activity in surface water

Surface waters are the main component of the freshwater ecosystem. Artificial radionuclides of NPPs as part of discharges first are released into the water of the cooling pond, then are transferred with it to various parts of the reservoir both downstream and upstream, then accumulate in bottom sediments and are accumulated by hydrobionts (aquatic plants and ichthyofauna). To assess the impact of the Beloyarsk NPP on the freshwater ecosystem, the long-term dynamics of the content of ^{60}Co ^{90}Sr, ^{137}Cs and ^{3}H in the surface water of six control sites of the cooling pond was assessed (Fig. 3). In the period 1988–2002, not all the radionuclides were measured in the samples taken.

4.1.1. Radiocobalt in surface water

The ^{60}Co content in the surface water of the cooling pond was measured in the period 1976–2011. Subsequently, the radionuclide concentration in water samples decreased to the detection threshold by devices. The maximum level of specific activity of ^{60}Co (5.6 Bq/L) was recorded in 1976 in the waste water of the Industrial Water Canal (S4) and changed little for the next two years (Fig. 3-A). During this period of time, emergency situations occurred at the AMB-200 reactor and in the turbine hall of the first stage of the Beloyarsk NPP, which led to the need for lengthy repair work (Table 2). Industrial waters containing increased activity of radionuclides after decontamination of the equipment were fed through the industrial water canal into the cooling pond. Since 1979, the concentration of ^{60}Co in the water of this site of the reservoir began to gradually decrease. However, it remained higher than in other parts of the monitoring network until 1985, when the indicator stabilized at the level of 0.2–0.3 Bq/L. In the waste waters of the Warm Bay (S5), coming from the cooling system of the first stage of the NPP, the ^{60}Co content was significantly lower (1.1–1.3 Bq/L) than in the Industrial Water Canal and approached the results of radionuclide measurements in the area of the Biophysical Station (S3), where such levels were in the range of 0.6–0.8 Bq/L. And if in 1976–1977 the difference in the content of ^{60}Co in the waters of the Warm Bay compared to the Industrial Water Canal was 3–5 times, then in the period 1978–1983 increased up to 10–30 times. This pattern suggests that the ^{60}Co contamination of the freshwater ecosystem in the initial period of the NPP operation was to a greater extent associated with numerous repairs at the NPP site and, to a lesser extent, with radionuclide discharges from the reactor cooling system. Analyzing the spatial distribution of ^{60}Co in surface water, it should be noted that the radionuclide concentration over the entire considered time interval decreased with distance from the NPP in the series of observation sites S4 > S5 ≈ S3 > S2 > S1. In 1976–1978 the ^{60}Co content in the water of the Industrial Water Canal (S4) in comparison with the Upper Reservoir (S1), i.e. technogenic background, it was 105–170 times higher. Later (1979–1988) this difference was reduced to 15–60 times. For the waste waters of the Warm Bay (S5), such differences from the background (S1) were 10–30 and 1–10 times for the two periods under consideration, respectively. In general, during the observation period (1976–2011), the specific activity of ^{60}Co decreased in the water of the Industrial Water Canal (S4) by 5.6 thousand times, in the waste waters of the Warm Bay (S5) by 350, in the area of the Biophysical Station (S3) at 80, in Pike Bay (S2) at 50, in the upper reaches of the reservoir (S1) by 5 times. In recent years, the concentration of ^{60}Co in the surface water of the Beloyarsk reservoir is at a fairly low level in the range of 0.001–0.02 Bq/L. The decrease in the specific activity of ^{60}Co in the surface water of the upper reaches of the reservoir over 35 years also indicates that at a distance of up to 15 km from the Beloyarsk NPP upstream, the nuclear power plant had a certain effect on the contamination of the freshwater ecosystem with radionuclides in the initial period of its operation.

4.1.2. Radiostrontium in surface water

The maximum concentrations of ^{90}Sr in the surface waters of the Beloyarsk reservoir were recorded in the area of the Biophysical Station (S3) in 1976–1978 and amounted to 0.3–0.5 Bq/L, which was significantly lower compared to ^{60}Co (Fig. 3-B). At the same time, such levels of specific activity of ^{90}Sr in water were more than 10 times higher than those of samples taken in the upper reaches of the reservoir (S1). The source of the radionuclide in the area of the Biophysical Station was the waters of the Bypass canal draining the site of the Beloyarsk NPP from the north, starting from the water treatment facilities of the nuclear power plant. As in the case with ^{60}Co, the ^{90}Sr input into the drainage system of the NPP site was due to the decontamination of structures and equipment of the first stage of the nuclear power plant after emergency situations. It is

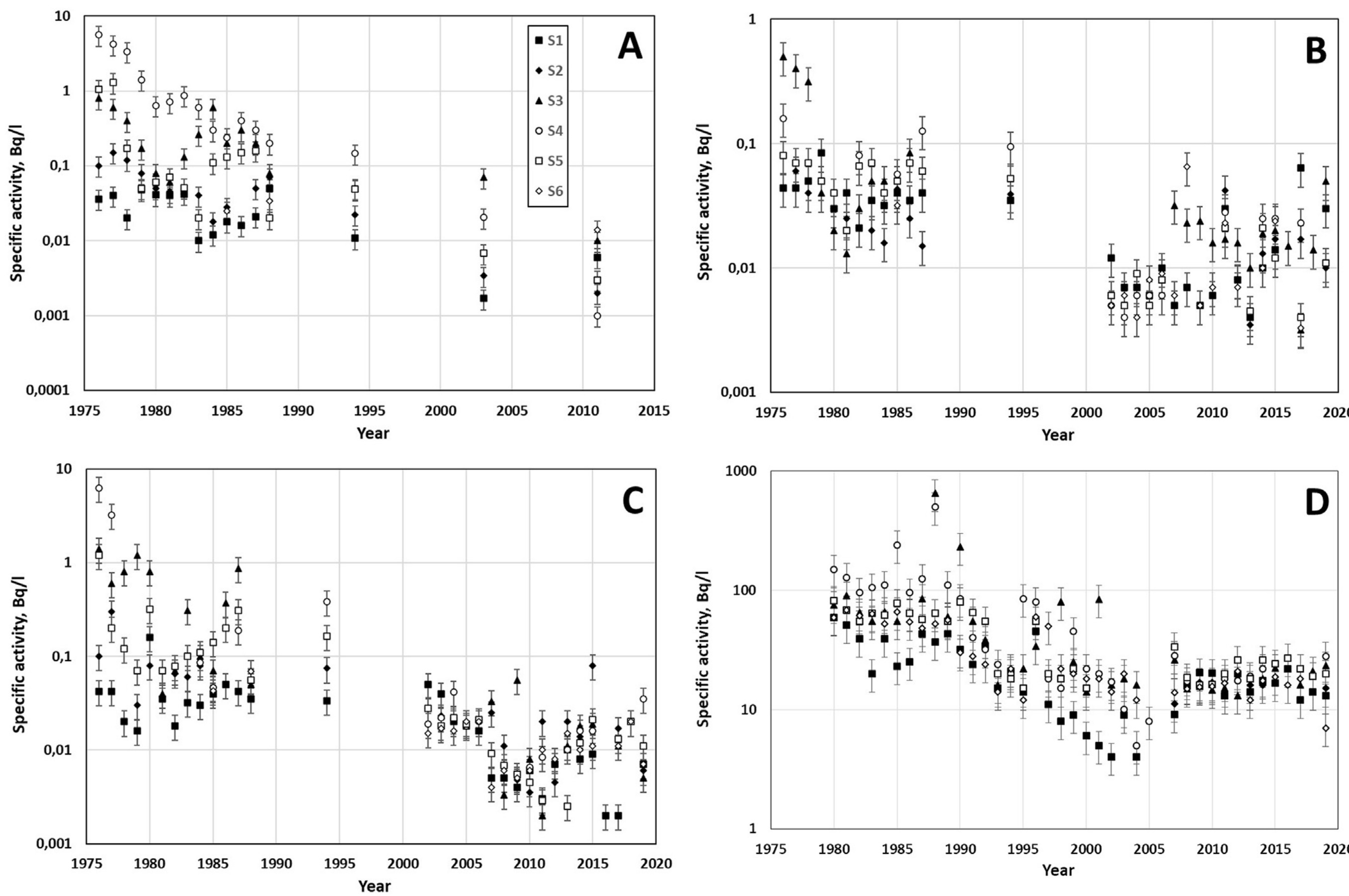

Fig. 3. Dynamics of radionuclides specific activity: A – ^{60}Co, B – ^{90}Sr, C – ^{137}Cs, D – ^{3}H in surface water of the control sites of Beloyarsk NPP cooling pond.

possible that the water treatment facilities did not fully cope with the purification of process water from radioactivity and part of it was released into the drainage system of the Bypass Canal. In the Industrial Water Canal (S4), ^{90}Sr was also recorded, although to a lesser extent (0.15 Bq/L). In other parts of the monitoring network, the radionuclide was recorded in the range of 0.02–0.08 Bq/L, while it was slightly higher in the water of the Warm Bay (S5). In general, over the observation period (1976–2019), the content of ^{90}Sr in the surface water of the cooling reservoir decreased from 1.5 times in the upper reaches of the reservoir (S1) to 10 times in the area of the Biophysical Station (S3), where it was initially maximum. At present, the specific activity of ^{90}Sr in the water of the Beloyarsk reservoir is at a consistently low level in the range of 0.01–0.05 Bq/L.

4.1.3. Radiocaesium in surface water

^{137}Cs is the most radiologically significant radionuclide from the point of view of the formation of human exposure dose. Its maximum concentration was recorded in the waste water of the Industrial Water Canal (S4) in 1976 and amounted to 6.3 Bq/L (Fig. 3-C), which is comparable to that for ^{60}Co at this sampling site. In the same year, the highest content of ^{137}Cs in the range of 1.2–1.4 Bq/L was also noted in the water of the Warm Bay (S5) and the area of the Biophysical Station (S3). However, if since 1977 the concentration of the radionuclide in the water of the Warm Bay dropped sharply to 0.1–0.3 Bq/L, then in the area of the Biophysical Station it was higher in the range of 0.6–1.2 Bq/L until 1980. This fact suggests that after emergencies at the first stage of the Beloyarsk NPP, the ^{137}Cs discharge from the reactor cooling system was quickly reduced to a level only 2–4 times higher than the background level at the site (S1). At the same time, in the water of the Bypass canal (S3) draining water from the wastewater treatment facilities of the Beloyarsk NPP, and where there were leaks of radioactive process waters, the content of ^{137}Cs exceeded the background (S1) by 15–75 times until 1980, and only then there was a tendency to decrease radionuclide concentration. To date, the content of ^{137}Cs in the surface water of the Beloyarsk reservoir is at the level of 0.005–0.035 Bq/L, while the maximum values are preserved in the water of the Industrial Water Canal (S4), and the minimum are recorded in the upper reaches of the reservoir (S1). Over the entire observation period (1976–2019), the content of ^{137}Cs decreased in the surface waters of the cooling pond in the area of the Biophysical Station (S3) by 280 times, the Industrial Water Canal (S4) by 180, Warm Bay (S5) by 110, Upper reservoir (S1) and near the dam part of the reservoir (S6) up to 6 times. In this respect, the Pike Bay (S2) occupies an intermediate position between the sources of discharging process waters from the NPP (S3–S5) and the upper reaches of the cooling reservoir (S1). In Pike Bay, the decrease in the specific activity of ^{137}Cs in surface water over the 45-year period under consideration was 17 times. Since at a distance of 3 km downstream from Pike Bay (S2) there is a discharge channel of the cooling system of the BN-800 reactor, which began operation in 2016, the low concentration of ^{137}Cs in the surface water of this site of the reservoir over the past 5 years indicates the absence of the recorded effect of the new power unit of the Beloyarsk NPP on the radioecology of the cooling pond [33,34]. For other radionuclides in the water of Pike Bay, no increase in their concentrations has been observed since 2016, when the BN-800 reactor of the Beloyarsk NPP began to operate.

4.1.4. Tritium in surface water

Tritium (^{3}H) is characterized by high mobility both in water and air. During the operation of reactors of nuclear power plants, more than 80% of this radionuclide is formed as a product of ternary fission of uranium and plutonium nuclei in fuel rods. Among other radionuclides, tritium is discharged into the cooling pond of the nuclear power plant and, upon evaporation of water, easily passes into the surface atmospheric air, being a source of additional human exposure. At the Beloyarsk reservoir, according to printed matter, ^{3}H began to be controlled since 1980 [35,36]. In the initial observation period, the maximum levels of radionuclide content in the surface water, as in the case of ^{60}Co and ^{137}Cs, were noted in the area of the Industrial Water Canal (S4), amounting to 95–240 Bq/L (Fig. 3-D). At the same time, they exceeded the values in the control area (S1) by 2.5–10 times. And if in other parts of the monitoring network the concentration of tritium in water was at a relatively constant level of 20–80 Bq/L, then in the water of the Industrial Water Canal it increased, reaching a maximum (500 Bq/L) in 1988. A sharp increase in the specific activity of ^{3}H (up to 650 Bq/L) this year was also noted in the water of the Bypass Canal in the area of the Biophysical Station (S3), which is associated with the accident at the BN-600 reactor in 1987 and the ingress of the radionuclide into the reservoir as a result of the discharge of decontamination water after long-term maintenance works. Subsequently, the content of tritium in surface water in all monitored sites of the reservoir began to decrease to a level of 10–80 Bq/L. In recent years, the specific activity of the radionuclide in the water of the cooling reservoir is 5–25 Bq/l, approaching with its minimum values the average technogenic background of freshwater ecosystems in the Ural region. In the surface waters of the Industrial Water Canal (S4), the specific activity of ^{3}H was and remains the highest among the identified ranges. In general, over the observation period (1980–2019), the decrease in the content of ^{3}H in the surface water of the Beloyarsk reservoir was 3.2–8.4 times in the areas of the monitoring network.

4.2. Dynamics of radionuclides specific activity in bottom sediments

Artificial radionuclides from Beloyarsk NPP discharges, entering the cooling pond, are redistributed in the water column and accumulate in bottom sediments, where they become a source of additional irradiation of aquatic biota, in particular, bottom organisms. At the same time, bottom sediments, adsorbing radionuclides, play an important role in the processes of purification of the reservoir, acting as a natural filter. Considering that radionuclides of NPP origin for a long time were being released into the cooling pond of the Beloyarsk NPP mainly through the Bypass (S3) and Industrial Water (S4) canals, it can be assumed that the maximum levels of accumulation of radioisotopes in bottom sediments will be observed precisely in these areas.

Previous studies to assess the degree of accumulation of artificial radionuclides in bottom sediments of the Beloyarsk reservoir showed that they accumulate to the greatest extent in silty sapropel, to a somewhat lesser extent in flooded soil and minimally in sandy soil [12]. Based on a conservative approach, this work presents the results of the analysis of the content of ^{60}Co, ^{90}Sr and ^{137}Cs in the silty sapropel of the control sites of the monitoring network of the Beloyarsk Reservoir, as the maximum accumulator of radionuclides (Fig. 4).

4.2.1. Radiocobalt in bottom sediments

The study of ^{60}Co accumulation in the bottom sediments of the Beloyarsk reservoir, started in 1976, showed that, on average, over the observation sites, the concentration of the radionuclide in the silty sapropel at that time was at the level of 860 Bq/kg. The minimum content of ^{60}Co in bottom sediments was noted in the Upper River (S1) and Near Dam (S6) of the reservoir-cooler, where it was 310–340 Bq/kg (Fig. 4-A). The intermediate position in the accumulation of radionuclide (610–630 Bq/kg), caused by the discharges of the NPP process waters, was occupied by the Industrial Water (S4) and Bypass (S3) canals. The maximum concentrations of ^{60}Co were recorded in the bottom sediments of the Warm Bay (S5) and varied within 3–6 thousand Bq/kg in the first years of observations. In addition to its content in water, the temperature factor has a great influence on the sorption of the radionuclide by bottom sediments. Thus, in laboratory experiments to assess the transfer factors of radionuclides (T_f) in bottom sediments, it was shown that an increase in the temperature regime in water by 28 °C leads to an increase in T_f ^{60}Co up to 8 times [24]. In the conditions of the Beloyarsk Reservoir, the increased temperature of the Warm Bay discharge waters (on average by 6 ... 7°C) also served as a factor influencing the increase in the sorption of ^{60}Co by silty sapropel. By 1987–1988 the content of the radioisotope in all control sites of the reservoir leveled off to levels of 280–350 Bq/kg. After the decommissioning of the AMB series reactors, the ^{60}Co input into the cooling pond sharply decreased, and its relatively rapid decay compared to other studied radionuclides ($T_{1/2}$ = 5.27 years) led to an even greater decrease in the specific activity of the radionuclide in the bottom sediments. To date, the content of ^{60}Co in the silty sapropel of the cooling reservoir of the Beloyarsk NPP has become the minimum since the beginning of observations and amounts to 4–15 Bq/kg. At the same time, the maximum levels of the specific activity of the radionuclide in the bottom sediments are recorded in the area of the NPP Industrial Water Canal. Thus, over the observation period, the accumulation of ^{60}Co in the silty sapropel of the Beloyarsk reservoir decreased from 40 (in the S4 site) to 670 (S5) times.

4.2.2. Radiostrontium in bottom sediments

The ^{90}Sr content in the silty sapropel of the cooling reservoir of the Beloyarsk NPP in all parts of the monitoring network was quite stable during the 45-year period of monitoring studies and reflected the dynamics of the specific activity of this radionuclide in the surface waters (Fig. 4-B). So, in 1976–1987 the maximum levels of ^{90}Sr content in silty sapropel were noted in the area of the Bypass Canal (S3) and Warm Bay (S5) in the range of 35–45 Bq/kg, and the minimum in the upper reaches of the reservoir (S1), in the range of 10–18 Bq/kg. To date, this ratio has changed little. Near the sources of discharging process waters of the Beloyarsk NPP (S3–S5), ^{90}Sr accumulates in silty sapropel at a level of 15–18 Bq/kg, and at a distance from the nuclear power plant (S1–S2, S6), its specific activity in this type of bottom sediments decreases to 1–10 Bq/kg. In general, over the entire observation period, the content of the radionuclide in the silty sapropel of the Beloyarsk reservoir decreased from 1.2 to 8.0 times. It should be noted that the accumulation of ^{90}Sr in bottom sediments is minimal among other studied radionuclides due to its low concentration in the surface water of the cooling reservoir.

4.2.3. Radiocaesium in bottom sediments

In 1976, the maximum levels of ^{137}Cs in the bottom sediments of the Beloyarsk reservoir were recorded in the silty sapropel of the Industrial Water Canal (S4), where they amounted to 4.37 thousand Bq/kg (Fig. 4-C). This was 80 times higher than in the background site of the Upper Reservoir (S1). In the area of other sources of waste water from the Beloyarsk NPP into the cooling pond, high levels of ^{137}Cs accumulation in silty sapropel were also recorded: 1790 Bq/kg in Warm Bay (S5) and 900 Bq/kg in the area of the

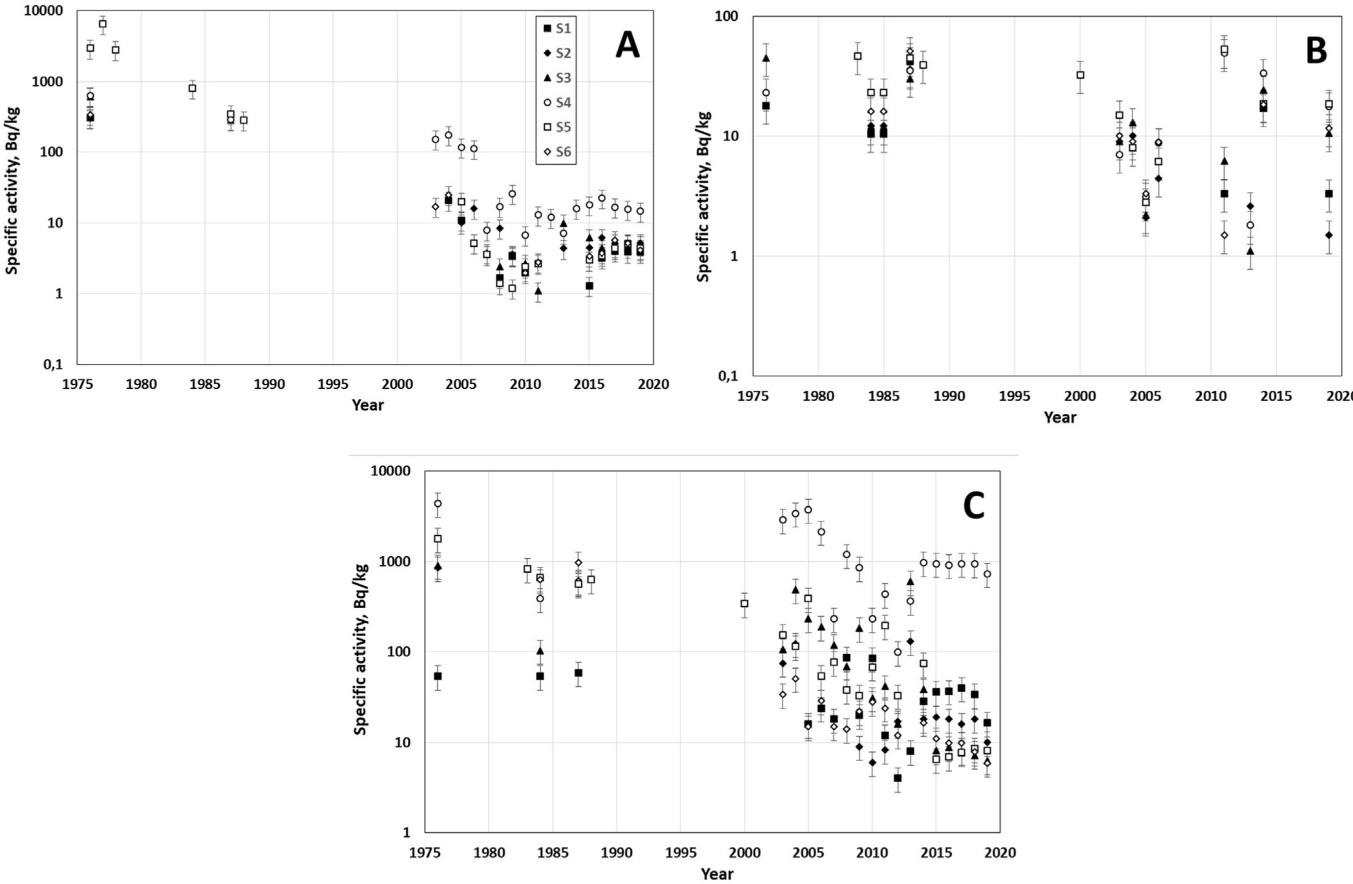

Fig. 4. Dynamics of radionuclides specific activity: A – ^{60}Co, B – ^{90}Sr, C – ^{137}Cs in bottom sediments (silty sapropel) of the control sites of Beloyarsk NPP cooling pond.

Biophysical Station (S3). By 1987, with the exception of the Industrial Water Canal, the radionuclide content in these bottom sediments decreased from 1.5 to 3.0 times, depending on the source of pollution, while in the background site of the reservoir (S1), the ^{137}Cs concentration in the silty sapropel practically did not change remaining at the level of 55–60 Bq/kg. In the silty sapropel of the Industrial Water Canal, high levels of ^{137}Cs remained throughout the observation period up to the present time, amounting to about 900 Bq/kg. These indicators of radionuclide accumulation in bottom sediments were influenced by a number of reasons. First, these are abnormal and emergency situations at the AMB-100, AMB-200 reactors and in the turbine hall of the first stage of the Beloyarsk NPP in 1976–1979. After long-term repair work and equipment decontamination, process waters containing increased activity of ^{137}Cs were discharged into the Industrial Water Canal and then into the cooling pond. The second factor was long-term effluents into the Industrial Water Canal of process waters containing radionuclides from the NPP industrial site. The third important factor was the high absorption capacity and strong binding of ^{137}Cs radioisotopes by silty sapropel. All this together influenced the accumulation of radionuclide in bottom sediments in this site of the cooling reservoir. At present, at the Beloyarsk reservoir, the content of ^{137}Cs in the silty sapropel (except for the Industrial Water Canal) is in the range of 5–15 Bq/kg, and on other types of bottom sediments (flooded soil and sandy soils) it is even lower.

5. Conclusion

From the data presented here, one can see that over the 45-year period of operation of the Beloyarsk NPP there has been a significant improvement in the radioecological state of the cooling pond. The content of artificial radionuclides in the main components of the freshwater ecosystem has decreased by tens and hundreds of times. These changes have been made possible for a number of reasons. First, this is the decommissioning of thermal reactors of the first stage of the nuclear power plant (AMB-100 and AMB-200) in the 80s, which were characterized by an increased intake of artificial radionuclides into the environment and, in particular, into the cooling pond as a result of their work. Second, this is the result of the mechanisms of self-purification of the freshwater ecosystem of the reservoir from radionuclides due to the decay of radioisotopes, their redistribution from the water phase to other components of the ecosystem, primarily into bottom sediments, as well as runoff into the river system. The presented data also show that the commissioning of the BN-800 fast neutron power unit with maximum power in 2016 did not lead to an increase in radioactivity in the Beloyarsk reservoir. The total decrease in the content of artificial radionuclides in the main components of the freshwater ecosystem of the Beloyarsk reservoir over the entire period of monitoring observations (1976–2019) was 4.3–74.5 times in surface waters, 10.1–505 times in bottom sediments (silty sapropel). This indicates the absence of a significant additional input of radioisotopes into the cooling pond after the change in electricity generation technologies due to the transition of the Beloyarsk NPP from thermal to fast reactors and the commissioning of new, higher capacities.

The Russian legislation regulates the levels of intervention for the content of the investigated radionuclides in drinking water:

^{60}Co – 40 Bq/L, ^{90}Sr – 4.9 Bq/L, ^{137}Cs–11 Bq/L, ^{3}H–7600 Bq/L. With the theoretically considered use of the water of the Beloyarsk reservoir as drinking water, in accordance with the current standards, no excess for all the studied radionuclides for the entire period of research in the water of the Beloyarsk reservoir was registered.

In the second part of the study, the results of long-term monitoring will be presented to assess the impact of waste technological waters of thermal (AMB-100 and AMB-200) and fast (BN-600 and BN-800) reactors of Beloyarsk NPP on the accumulation of artificial radionuclides in macrophytes and ichthyofauna of the cooling pond. Considering that various types of reactors were operated during the operation of the Beloyarsk NPP, the analysis of the monitoring results will identify six periods of the plant's impact on the freshwater ecosystem of the cooling reservoir and provide an integral assessment of the accumulation of radioisotopes in all its components (surface water, bottom sediments, macrophytes, ichthyofauna).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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