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ИнБЮМ – IBSS

Морской биологический журнал
Marine Biological Journal
2021, vol. 6, no. 1, pp. 41–57
https://doi.org/10.21072/mbj.2021.06.1.04

UDC [504.5:620.267](261.243)

ASSESSMENT OF RADIATION STATE OF MARINE ENVIRONMENT IN THE LENINGRAD NPP AREA ACCORDING TO LONG-TERM MONITORING DATA (1973–2019)

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Received by the Editor 05.11.2020; after reviewing 17.12.2020;
accepted for publication 11.03.2021; published online 23.03.2021.

The aim of the study was to conduct a radioecological assessment of the Leningrad NPP marine cooling reservoir – Koporye Bay of the Gulf of Finland. According to the international basic safety standards, accepted at the IAEA General Conference, this issue is of particular relevance due to the need to justify protection from technogenic radiation exposure both to humans and the environment. The assessment was based on the long-term radioecological monitoring data (1973–2019) within the Leningrad NPP observation area: radionuclides concentration in seawater, bottom sediments, and hydrobionts. The reference levels of radionuclides content in seawater and bottom sediments were used as indicators of the radiation state of the marine environment; their calculation procedure is defined in the Recommendations R 52.18.852-2016 and R 52.18.873-2018, issued by the Federal Service for Hydrometeorology and Environmental Monitoring (the Ministry of Natural Resources and Environment of the Russian Federation). These recommendations, developed by RPA "Typhoon" specialists, contain a methodology for assessing the radioecological state of the marine environment by the level of radionuclides activity, based on the principles, ensuring the maintenance of favorable environment, safety of marine hydrobionts, and radiation protection of humans. In the presence of various radionuclides in the marine environment, the sum of technogenic radionuclide activity ratios in seawater (bottom sediments) to the corresponding reference levels shall be below 1. According to monitoring data in the early period of NPP operation (1973–1985), a wide spectrum of technogenic radionuclides was observed in the marine ecosystem components. Along with ^{137}Cs, significant contributors to the contamination of seawater and bottom sediments were ^{54}Mn and ^{60}Co. In contrast to reference levels for ^{137}Cs, reference levels for ^{54}Mn and ^{60}Co in seawater are determined by an environmental criterion, not a radiation-hygienic one. The presence of technogenic radionuclides in algae was registered at distances, exceeding 10 km from the NPP. Biogenic transfer of corrosion radionuclides (^{54}Mn, ^{60}Co, and ^{65}Zn) by fish into rivers, flowing into the Koporye Bay, was noted. The Chernobyl disaster led to a noticeable increase in the pollution of the Koporye Bay with technogenic radionuclides. In May – December 1986, the sum of technogenic radionuclide activity ratios in seawater to the reference levels exceeded the pre-accidental level by 100 times, and in bottom sediments – by 30 times. In 1986, ^{137}Cs and ^{134}Cs were the main contributors to the marine ecosystem radioactive contamination. Currently, the technogenic radioactivity of seawater and bottom sediments of the Koporye Bay is mainly determined by ^{137}Cs; its level is relatively constant, which indicates the stability of the radioecological situation in the Leningrad NPP marine cooling reservoir.

Keywords: Koporye Bay of the Gulf of Finland, Leningrad NPP, radioecological monitoring, seawater, bottom sediments, radionuclide reference levels, environmental and radiation-hygienic criteria, integral indicator of pollution, long-term dynamics

In accordance with the modern paradigm of radiation safety, not only humans, but also other organisms must be protected from radiation risks; it is necessary to confirm (not to assume) that the environment is protected from industrial radiation exposure (Kryshev & Sazykina, 2013, 2014, 2018 ; Radiation Protection…, 2014).

The Leningrad NPP (hereinafter LNPP) is located 80 km west of Saint Petersburg, on the Koporye Bay coast (the Gulf of Finland, the Baltic Sea). Koporye Bay is used as a cooling reservoir for the LNPP since 1973. Currently, the nuclear power plant operates two power units with RBMK-1000 reactors and one power unit with a WWER-1200 reactor. In 2021, it is planned to launch another power unit with a WWER-1200 reactor. Two power units with RBMK-1000 reactors were shut down for decommissioning. LNPP operation is accompanied by releases of radioactive material into the atmosphere and discharges into the marine environment. In this area, other enterprises of the nuclear-industrial complex are also located: A. P. Aleksandrov Scientific Research Technological Institute with a complex of experimental nuclear installations and Leningrad Office of "North-Western Territorial District" branch of "Federal Ecological Operator", which processes and stores radioactive waste.

To date, a unique experience of environmental radioecological monitoring has been accumulated in the LNPP area, including marine cooling reservoir, which is exposed to the effect of radioactive material discharge and a set of non-radiational factors (thermal discharge, chemical pollution, eutrophication, and mechanical injury of organisms in the NPP water intakes) (Kryshev, 2017 ; Kryshev & Ryazantsev, 2010 ; Ekologo-geofizicheskie aspekty…, 1992). Territorially, the monitoring object was the marine cooling reservoir and the adjacent zone of the catchment area with rivers, flowing into the Koporye Bay.

The Koporye Bay of the Gulf of Finland with an area of 255 km^2 is of shallow semi-enclosed water bodies type, with an extended watershed boundary with the main water area. Salinity varies 2 to 4 ‰. The bottom is mainly sandy, and sometimes silt and stones are found. Mean depth is about 12 m. Maximum depths (up to 27 m) are registered on the border with the main water area. The LNPP discharges heated water to the eastern Koporye Bay with the area of about 50 km^2 and mean depth of 5 m. Three rivers (Sista, Kovashi, and Voronka) flow into the eastern Koporye Bay; their total mean annual flow rate is about 10 $m^3 \cdot s^{-1}$. Taking into account their pollution by industrial and domestic wastewater, it can be considered that it is the eastern Koporye Bay (which is used as a cooling reservoir), that is the most affected by anthropogenic factors in the LNPP area.

The aim of our work was to assess the radioecological state of the Leningrad NPP marine cooling reservoir – the Koporye Bay of the Gulf of Finland – based on long-term radioecological monitoring data (1973–2019) and criteria, providing the preservation of a favorable environment and hydrobionts, as well as human radiation protection.

MATERIAL AND METHODS

Radioecological monitoring. For more than 47 years, environmental departments of Roshydromet (Russian Federal Service for Hydrometeorology and Environmental Monitoring), Scientific Research Technological Institute, V. G. Khlopin Radium Institute, and the LNPP are carrying out monitoring of the technogenic radionuclide content in seawater, bottom sediments, and marine biota of the Koporye Bay (Blinova, 1998 ; Bondarenko et al., 2013 ; Vakulovskii & Nikitin, 1984 ; Kryshev, 2017 ; Kryshev & Blinova, 1991 ; Kryshev & Ryazantsev, 2010 ; Radiatsionnaya obstanovka…, 1992–2020 ; Radioaktivnoe zagryaznenie raionov AES, 1990). To analyze radionuclide content, standard methods of sampling, radiochemical analysis, and radiometric and spectrometric measurements are used (Metodicheskie rekomendatsii…, 1980, 1986 ; Nastavlenie gidrometeorologicheskim stantsiyam…, 2015 ; Rukovodstvo po organizatsii…, 1990).

The most detailed radioecological monitoring was carried out in the period, when LNPP with RBMK-1000 reactors reached maximum power (1973–1985), as well as in the first years after the Chernobyl disaster (Kryshev & Blinova, 1991 ; Kryshev & Ryazantsev, 2010). Monthly, radionuclide content was determined in the samples of the NPP intake and discharge channels. Monthly during the growing season of water plants (April to October) at 5 points of the coastal water area and year-round in the discharge channels of heated water, specific activity was determined in the samples of seawater, bottom sediments, and water plants. Annually, specific activity was measured in 20–30 fish samples from the nets of the NPP water intakes and from the catches of fishermen (Blinova, 1998). The results of the NPP cooling reservoir monitoring in the pre-Chernobyl period are summarized for further analysis (Table 1).

In addition to radionuclides (see Table 1), ^{51}Cr, ^{58}Co, ^{95}Zr, ^{95}Nb, and ^{134}Cs were determined in bottom sediment samples in 1–5 % of cases (Kryshev & Blinova, 1991). In the samples of marine plants in the Koporye Bay, along with radionuclides, a wider range of technogenic radionuclides was recorded, than in seawater and bottom sediment samples, in 4–28 % of cases: ^{51}Cr, ^{58}Co, ^{59}Fe, ^{95}Zr, ^{95}Nb, ^{131}I, ^{134}Cs, ^{141}Ce, and ^{144}Ce (Radioaktivnoe zagryaznenie raionov AES, 1990).

Table 1. Technogenic radionuclides content in components of the marine cooling reservoir at different distances from the Leningrad NPP (1973–1985)

Sampling location	Radionuclides				
	^{90}Sr	^{137}Cs	^{54}Mn	^{60}Co	^{65}Zn
Seawater, mBq·L^{-1}					
Discharge channels	30 ± 3 (100 %)	32 ± 6 (100 %)	38 ± 14 (5 %)	33 ± 11 (7 %)	< MDA
1–3 km from the LNPP	27 ± 2 (100 %)	18 ± 2 (100 %)	31 ± 13 (4 %)	30 ± 9 (4 %)	< MDA
3–10 km	23 ± 2 (100 %)	11 ± 2 (100 %)	< MDA	< MDA	< MDA
Over 10 km	23 ± 2 (100 %)	10 ± 2 (100 %)	< MDA	< MDA	< MDA
Reference level in seawater (Poryadok rascheta…, 2016)	16 800	1220	270	610	116

Continue on the next page…

Sampling location	Radionuclides				
	^{90}Sr	^{137}Cs	^{54}Mn	^{60}Co	^{65}Zn
Bottom sediments, Bq·kg^{-1} of wet weight					
Discharge channels	3.5 ± 1.6 (100 %)	4.0 ± 1.7 (100 %)	8 ± 3 (25 %)	8 ± 3 (12 %)	4.4 ± 2.1 (6 %)
1–3 km from the LNPP	2.0 ± 0.9 (100 %)	2.1 ± 1.0 (100 %)	14 ± 6 (10 %)	10 ± 4 (12 %)	11 ± 5 (5 %)
3–10 km	2.1 ± 1.1 (100 %)	2.0 ± 1.0 (100 %)	6 ± 3 (12 %)	7 ± 3 (7 %)	< MDA
Over 10 km	1.5 ± 0.8 (100 %)	1.6 ± 0.7 (100 %)	0.2 ± 0.1 (7 %)	< MDA	< MDA
Reference level in bottom sediments (Poryadok rascheta…, 2019)	2500	1500	810	1800	2400
Water plants, Bq·kg^{-1} of wet weight					
Discharge channels	1.8 ± 0.6 (100 %)	5.2 ± 1.4 (100 %)	18 ± 6 (43 %)	17 ± 5 (41 %)	13 ± 4 (12 %)
1–3 km from the LNPP	1.7 ± 0.7 (100 %)	4.8 ± 1.5 (100 %)	15 ± 5 (31 %)	15 ± 5 (33 %)	10 ± 4 (11 %)
3–10 km	1.7 ± 0.7 (100 %)	3.3 ± 1.5 (100 %)	5.2 ± 1.9 (30 %)	7 ± 3 (19 %)	10 ± 4 (11 %)
Over 10 km	1.2 ± 0.5 (100 %)	1.6 ± 0.7 (100 %)	5.2 ± 2.0 (10 %)	4.4 ± 1.8 (19 %)	7 ± 3 (7 %)
Fish, Bq·kg^{-1} of wet weight					
Discharge channels					
Common roach *Rutilus rutilus* (Linnaeus, 1758)	2.3 ± 0.7 (100 %)	1.7 ± 0.5 (100 %)	18 ± 6 (30 %)	11 ± 3 (52 %)	70 ± 27 (43 %)
Koporye Bay					
Common roach *R. rutilus*	1.8 ± 0.7 (100 %)	1.5 ± 0.5 (100 %)	1.5 ± 0.6 (7 %)	10 ± 3 (14 %)	60 ± 21 (28 %)
Baltic herring *Clupea harengus membras* Linnaeus, 1760	1.3 ± 0.5 (100 %)	2.0 ± 0.7 (100 %)	0.3 ± 0.1 (4 %)	5.4 ± 1.9 (12 %)	12 ± 5 (8 %)
Common perch *Perca fluviatilis* Linnaeus, 1758	1.5 ± 0.5 (100 %)	2.7 ± 0.8 (100 %)	6.3 ± 2.5 (20 %)	10 ± 3 (20 %)	14 ± 5 (10 %)
Rivers					
Common roach *R. rutilus*	1.7 ± 0.7 (100 %)	1.3 ± 0.5 (100 %)	3.7 ± 1.3 (17 %)	1.9 ± 0.7 (17 %)	34 ± 11 (17 %)

Note: in parentheses, the frequency of radionuclide detection above MDA according to monitoring data is indicated; MDA is the minimum detectable activity (5 mBq·L^{-1} for seawater; 0.2 Bq·kg^{-1} for bottom sediments; 0.6 Bq·kg^{-1} for water plants and fish).

The Chernobyl disaster had a significant effect on the radiation state in the LNPP area, including the Koporye Bay (Blinova, 1998 ; Kryshev & Blinova, 1991 ; Kryshev & Ryazantsev, 2010 ; Radioaktivnoe zagryaznenie raionov AES, 1990). The radioactive cloud of the accidental release reached the LNPP area on 28.04.1986. An atmospheric fallout resulted in the pollution of the Koporye Bay catchment area, seawater, bottom sediments, and biota. After a decay of ^{131}I (half-life of 8.04 days) and other short-lived radionuclides, the main radioecological significance in this part of the distant Chernobyl trace was acquired by ^{134}Cs (half-life of 2.06 years) and in particular the long-lived ^{137}Cs (half-life of 30 years); thus, ^{137}Cs content in the marine ecosystem components increased by 6–250 times, compared with the pre-accidental level (Table 2).

Table 2. Dynamics of ^{137}Cs average annual content in the coastal marine ecosystem components in the Leningrad NPP area (1985–1990)

Ecosystem component		Average data for monthly observations					
		1985	1986*	1987	1988	1989	1990
Seawater, mBq·L^{-1}		10 ± 3	1300 ± 570	290 ± 110	130 ± 40	56 ± 18	52 ± 15
Bottom sediments, Bq·kg^{-1} of wet weight		1.5 ± 0.6	49 ± 19	19 ± 8	10 ± 4	10 ± 3	8 ± 3
Water plants, Bq·kg^{-1} of wet weight		0.8 ± 0.3	200 ± 97	17 ± 7	25 ± 8	14 ± 6	12 ± 5
Fish, Bq·kg^{-1} of wet weight	Baltic herring *C. harengus membras*	1.7 ± 0.5	27 ± 11	36 ± 15	19 ± 7	14 ± 5	25 ± 11
	Common perch *P. fluviatilis*	3.5 ± 1.1	22 ± 10	123 ± 41	126 ± 39	113 ± 31	116 ± 39

Note: * denotes averaged data for May – December 1986.

The highest levels of ^{137}Cs contamination for almost all marine ecosystem components (except for predatory fish species) were recorded in 1986. For predatory fish, the effect of trophic levels was observed since 1987, which was manifested in an increased accumulation of radiocesium a year after the accidental pollution, compared with the accumulation by non-predatory species. For most marine ecosystem components, there was a gradual decrease in ^{137}Cs content; however, it remained 5–30 times higher, than the pre-accidental level, even in 1990.

The current values of radionuclides content in the coastal marine ecosystem components of the Koporye Bay of the Gulf of Finland were determined (Table 3) (Bondarenko et al., 2013 ; Radiatsionnaya obstanovka…, 1992–2020).

According to the long-term monitoring data, more than 30 years after the Chernobyl disaster, ^{137}Cs content in bottom sediments, seawater, and fish tissues (common perch) in the Koporye Bay still exceeds the pre-accidental level. At the same time, the content of corrosion radionuclides of the LNPP origin in seawater and bottom sediments has noticeably decreased. Tritium and carbon-14, both of natural and technogenic origin, have acquired particular significance.

Table 3. Radionuclide content in the coastal marine ecosystem components in the Leningrad NPP area (2000–2019)

Ecosystem component		Radionuclide					
		^{90}Sr	^{137}Cs	^{60}Co	^{54}Mn	^{14}C	^{3}H
Seawater, mBq·L^{-1}		12 ± 2	20 ± 4	26 ± 7*	< MDA	–	21 000 ± 10 000
Reference level for seawater		16 800	1220	610	270	137	86 800 000
Bottom sediments, Bq·kg^{-1} of wet weight		0.70 ± 0.16	19 ± 3	1.5 ± 0.7	0.9 ± 0.4	–	–
Reference level for bottom sediments		2500	1500	1800	810	41	–
Fish, Bq·kg^{-1} of wet weight	Common roach *R. rutilus*	0.20 ± 0.08	1.2 ± 0.3	–	–	150 ± 50**	56 ± 17**
	Common perch *P. fluviatilis*	0.30 ± 0.10	6.2 ± 1.3	–	–	63 ± 31**	38 ± 11**

Note: a dash (–) denotes no data; * denotes data for 2002–2004 (Radiatsionnaya obstanovka…, 1992–2020); ** denotes data for 2012 (Bondarenko et al., 2013).

Methods for assessing the radiation state of the marine environment. The reference levels of technogenic radionuclides in seawater and bottom sediments were used as criteria for assessing the radiation state of the LNPP marine cooling reservoir; their calculation procedure is defined in the Roshydromet Recommendations (Poryadok rascheta…, 2019, 2016). There, for the first time in domestic and world practice, a methodology is given for assessing the quality of the marine environment on the basis of environmental and radiation-hygienic principles, that provide the preservation of a favorable environment and biological diversity, protection of aquatic ecosystems, and human radiation protection.

The environmental criterion for assessing the reference levels of radionuclide content in seawater is considered to be the maximum permissible dose (P_{max}) on marine biota objects. Its values are as follows: 1.0 mGy·day^{-1} for marine vertebrates and 10 mGy·day^{-1} for marine invertebrates and water plants (Otsenka radiatsionno-ekologicheskogo vozdeistviya…, 2015 ; Environmental protection…, 2009).

Due to huge species diversity of the biosphere and the practical impossibility of assessing the radiation effect on each flora and fauna component, the methodology of environmental radiation safety has been developed for a relatively small number of representative biota species (Otsenka radiatsionno-ekologicheskogo vozdeistviya…, 2015 ; Environmental protection…, 2009). The reference level of the *i*-th radionuclide in seawater for the *k*-th representative marine biota object $A_{i,k,ec}$, Bq·L^{-1}, is calculated by the formula (Kryshev et al., 2017 ; Poryadok rascheta…, 2016):

$$A_{i,k,\mathrm{ec}} = \frac{P_{max,k}}{(DCF_{i,k,1} \cdot CF_{i,k,2} + DCF_{i,k,2} \cdot \alpha'_{k,2} + 0.5 \cdot DCF_{i,k,2} \cdot K_{d,i,3} \cdot \alpha'_{k,3}) \cdot \tau}\,, \quad (1)$$

where $P_{max,k}$ is criterion of the maximum permissible dose on the *k*-th representative marine biota object, mGy·day^{-1};

$DCF_{i,k,1}$ and $DCF_{i,k,2}$ are dose conversion factors for internal and external exposure, respectively, of the *k*-th representative marine biota object to the *i*-th radionuclide, (μGy·hour^{-1})/(Bq·kg^{-1} of wet weight);

$CF_{i,k,2}$ is accumulation coefficient of the *i*-th radionuclide in the *k*-th representative marine biota object, L·kg^{-1};

$K_{d,i,3}$ is distribution coefficient of the *i*-th radionuclide between seawater and bottom sediments, L·kg^{-1};

$\alpha'_{k,2}$ and $\alpha'_{k,3}$ are the ratios of time, that the *k*-th representative marine biota object spends in the water and at the bottom, respectively, dimensionless coefficients;

τ is conversion factor, equal to 2.4·10^{-2} (mGy·day^{-1})/(μGy·hour^{-1}).

In accordance with regulatory documents (Otsenka radiatsionno-ekologicheskogo vozdeistviya…, 2015 ; Environmental protection…, 2009), the following marine biota objects were selected as reference ones: marine fish, molluscs, crustaceans, algae, and mammals. Non-exceeding the minimum $A_{i,k,ec}$ value for all representative organisms in the marine ecosystem guarantees sustainable functioning and preservation of the ecosystem species diversity as a whole. For most of technogenic radionuclides, detected by the radioecological monitoring system in the LNPP area, fish are critical organisms; they are characterized by the lowest values of the reference levels of radionuclides content in seawater (Poryadok rascheta…, 2016). As representative fish species of the marine cooling reservoir, common roach *Rutilus rutilus* (Linnaeus, 1758) and common perch *Perca fluviatilis* Linnaeus, 1758 were chosen (Radioaktivnoe zagryaznenie raionov AES, 1990), since they are available for catching throughout the year both in areas of discharge water distribution and in other areas, *inter alia* basins of rivers, flowing into the sea bay. These species spawn in the cooling reservoir, including the discharge channels; juveniles live in the channels constantly in significant quantities; large specimens accumulate in the area of heated water distribution, where they are fished in commercial quantities. It is essential that these species breed in thickets of aquatic plants, that accumulate radionuclides. Roe and juveniles of common roach and common perch experience increased dose loads and high temperature (above +30 °C in summer).

When calculating the reference levels of radionuclides in seawater, it is taken into account that it is not consumed for drinking purposes. As a radiation-hygienic criterion, radiation dose limit from seafood consumption is used for a critical population group, which is characterized by significant seafood consumption (Kryshev et al., 2017 ; Poryadok rascheta…, 2016 ; Sazykina & Kryshev, 1999).

The reference level of the *i*-th radionuclide in seawater according to the radiation-hygienic criterion $A_{i,human}$, Bq·L^{-1}, is calculated by the formula:

$$A_{i,human} = \frac{DL_{10\%}}{E_i \sum CF_{i,k,2} \cdot R_k}\ , \qquad (2)$$

where $DL_{10\%}$ is a part of the dose limit (DL) for the critical population group from consumption of seafood, containing the *i*-th radionuclide, equal to 10 %, or 0.1 mSv·year^{-1} (according to Sanitary rules and regulations SanPiN 2.6.1.2523, DL is of 1 mSv·year^{-1});

E_i denotes dose coefficients for the *i*-th radionuclide, Sv·Bq^{-1}, when assessing the dose per person from seafood consumption according to SanPiN 2.6.1.2523;

$CF_{i,k,2}$ is accumulation coefficient of the *i*-th radionuclide in the *k*-th seafood, L·kg^{-1};

R_k denotes values of the annual consumption of the *k*-th seafood by the critical population group.

In order to provide environmental safety, it is recommended to take the minimum value $A_{i,min}$, calculated according to the radiation-hygienic and environmental criteria, as a reference level of radionuclide content in seawater:

$$A_{i,min} = min\{A_{i,human}, A_{i,k,ec}\} . \quad (3)$$

In the presence of a mixture of radionuclides in seawater, it is necessary to fulfill the condition of non-exceeding the environmentally safe level to provide the radiation protection of humans and marine biota (Poryadok rascheta…, 2016):

$$\sum_i \frac{A_{v,i}}{A_{i,min}} \leq 1 , \quad (4)$$

where $A_{v,i}$ is activity concentration of the *i*-th radionuclide in seawater, Bq·L^{-1}.

The fulfillment of the condition (4) provides both environmental and radiation-hygienic safety, since the total contamination of seawater by technogenic radionuclides in this case will not result in exceeding the safe radiation level for biota and human radiation safety standards.

The reference technogenic radionuclide activity ratios for marine bottom sediments are determined in a similar way (Poryadok rascheta…, 2019).

The sum of technogenic radionuclide activity ratios, observed in seawater and bottom sediments, to the values of the reference levels will be further interpreted as an integral indicator of pollution of a reservoir with technogenic radionuclides. The use of ratios, similar to (4), and the corresponding indicators (indices) of pollution is widespread in the practice of radioecological monitoring (Radiatsionnaya obstanovka…, 1992–2020).

RESULTS

The distribution of the integral indicators of seawater and bottom sediments pollution with technogenic radionuclides in the Koporye Bay at different distances from the LNPP, calculated based on the long-term monitoring data (see Table 1) in the pre-Chernobyl period, indicates that the contamination values both of seawater and bottom sediments were the highest in the NPP discharge channels and the lowest at a distance over 10 km (Fig. 1). With the distance from the NPP, there is a gradual decrease in the level of contamination by technogenic radionuclides of the coastal zone of the eastern Koporye Bay. The main contributors to seawater contamination in the Koporye Bay coastal zone, close to the NPP, are ^{137}Cs (52–55 %), ^{54}Mn (14–17 %), and ^{60}Co (7–14 %). The contribution of ^{90}Sr to seawater contamination in this area is 4–8 %. At a distance over 10 km, the contribution of ^{137}Cs to technogenic radioactive contamination increases up to 85 %, and of ^{90}Sr – up to almost 15 %. Approximately the same radionuclide composition and spatial distribution of technogenic radionuclide contamination are registered for bottom sediments.

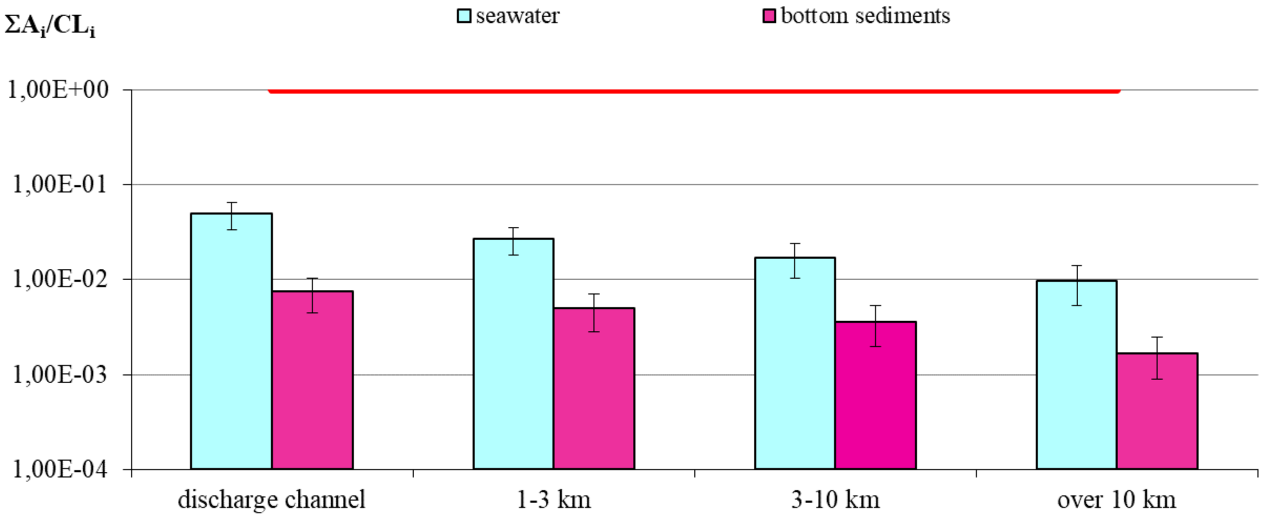

Fig. 1. Sum of technogenic radionuclide activity ratios, observed in coastal seawater and bottom sediments of the Koporye Bay in the Leningrad NPP area, to the reference levels (1973–1985). The upper horizontal line corresponds to the fulfillment of the condition (4), providing the environmental and radiation-hygienic safety of the marine environment

Taking into account the non-equilibrium of radioecological processes after the Chernobyl disaster and following the recommendations (Poryadok rascheta…, 2019, 2016), the sum of observed technogenic radionuclide activity ratios to the reference levels in 1986–1990 was calculated for the studied marine ecosystem on the basis of the monitoring data (Blinova, 1998 ; Kryshev & Ryazantsev, 2010 ; Radioaktivnoe zagryaznenie raionov AES, 1990). The dynamics of variations of this indicator is characterized by reaching the maximum values in May – December 1986 (Fig. 2), close to the value for seawater,

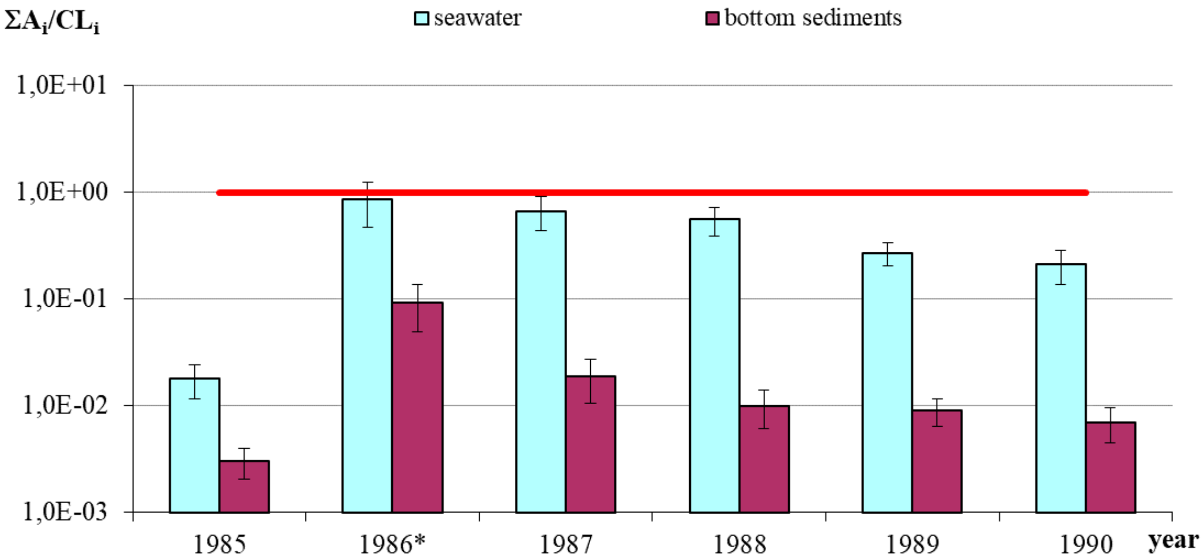

Fig. 2. Dynamics of changes of the sum of technogenic radionuclide activity ratios, observed in seawater and bottom sediments of the Koporye Bay in the Leningrad NPP area, to the reference levels before and after the Chernobyl disaster (1985–1990); * – according to data for May – December 1986. The upper horizontal line corresponds to the fulfillment of the condition (4), providing the environmental and radiation-hygienic safety of the marine environment

which corresponds to the fulfillment of the condition (4), providing environmental and radiation-hygienic safety. The main contributors to the radioactive contamination of the reservoir in 1986 were ^{137}Cs and ^{134}Cs, the reference levels for which are limited by the radiation-hygienic criterion. The rest of the technogenic radionuclides (^{95}Zr, ^{95}Nb, ^{131}I, ^{90}Sr, *etc.*) made a total contribution of less than 2 %. In subsequent years, there was a decrease in the level of contamination, but even in 1990 the value was an order of magnitude higher than the pre-accidental one.

The current dynamics of radioactive contamination of seawater and bottom sediments of the Koporye Bay coastal zone in the Leningrad NPP area is generally characterized by a relatively stable level of technogenic radionuclide activity ratios (Fig. 3).

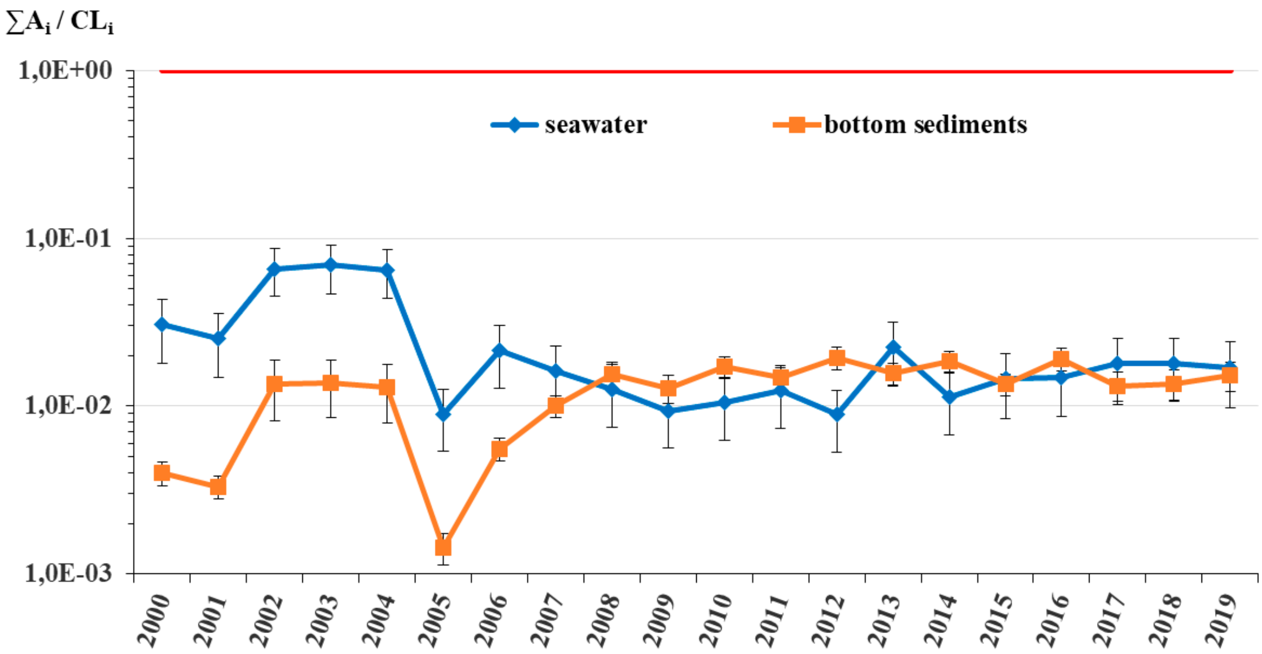

Fig. 3. Dynamics of changes of the sum of technogenic radionuclide activity ratios, observed in seawater and bottom sediments of the Koporye Bay in the Leningrad NPP area, to the reference levels (2000–2019). The upper horizontal line corresponds to the fulfillment of the condition (4), providing the environmental and radiation-hygienic safety of the marine environment

In 2002–2004, an increase was noted in the sum of technogenic radionuclide activity ratios, observed in seawater and bottom sediments of the Koporye Bay coastal zone, to the reference levels; that was associated with ^{60}Co presence in these marine ecosystem components in amounts, comparable to those of ^{137}Cs. In subsequent years, seawater contamination was mainly due to ^{137}Cs and was approximately at the same level. Similar dynamics was observed for bottom sediments.

DISCUSSION

Before the recommendations R 52.18.852-2016 2016 (Poryadok rascheta…, 2016) and R 52.18.873-2018 (Poryadok rascheta…, 2019) were published, there were no regulatory and methodological documents in the country and in the world regarding the criteria for assessing the radiation state of the marine environment, non-exceeding of which provides radiation safety and preservation of a favorable environment, as well as hydrobionts and humans. This created difficulties in solving environmental problems and using atomic energy at sea.

The main source of additional exposure of population due to contacts with the marine environment is seafood consumption. Seawater is practically not consumed in Russia for drinking purposes. For this reason, intervention levels of separate radionuclides content in drinking water cannot be applied to assess the radiation state of seawater (these levels are published in the radiation safety standards NRB-99/2009 (Appendix P-2a); if they are exceeded, certain protective measures should be taken).

Currently, the priority is to provide human safety, but the scientific community also shows considerable interest in regulation and scientific substantiation of the radiation state of the environment (Kryshev & Sazykina, 2013, 2018 ; Environmental protection…, 2009 ; Radiation Protection…, 2014). With radioactive contamination of the sea, hydrobionts can receive higher radiation doses, than humans, in a number of cases, for example with external exposure from bottom sediments. In addition, separate groups of organisms accumulate radionuclides from the environment with high accumulation rates, which are not typical for humans.

The methodology for assessing the indicators of the radiation state of the marine environment based on environmental and radiation-hygienic criteria is developed in the number of publications (Gusev, 1975 ; Kryshev et al., 2017 ; Sazykina & Kryshev, 1999, 2001 ; Kryshev & Sazykina, 2002 ; Sazykina & Kryshev, 2002a, b). Values of the reference levels in seawater for radionuclides, detected by radioecological monitoring systems in different periods of the LNPP operation, are given in Table 4 (Poryadok rascheta…, 2016).

For ^{54}Mn, ^{60}Co, ^{95}Zr, ^{95}Nb, ^{141}Ce, and ^{144}Ce, the limiting levels are the reference ones, calculated according to the environmental criterion. For ^{137}Cs, ^{90}Sr, ^{3}H, and ^{14}C (the most common in the environment), the values of the reference levels in seawater are determined by radiation-hygienic limits. Radionuclides, that are characterized by high accumulation rates in marine biota (^{14}C and ^{65}Zn) and in bottom sediments (^{54}Mn, ^{60}Co, ^{95}Zr, and ^{95}Nb), have the lowest values of the reference levels.

The reference levels of technogenic radionuclide activity ratios in the marine environment components can be directly compared with the data of direct measurements, which makes it possible to use them in the practice of radioecological monitoring. An integral indicator of the radiation state is the sum of technogenic radionuclide activity ratios, observed in seawater and bottom sediments, to the values of the reference levels. Based on the long-term radioecological monitoring data (1973–2019) for the Koporye Bay of the Gulf of Finland in the LNPP area, this indicator was calculated for the first time. Analysis of monitoring data allows us to draw the following conclusions.

During the period, when LNPP with RBMK-1000 reactors reached maximum power (1973–1985), its influence on radionuclides activity in seawater was traced in the discharge channels and the adjacent water area at a distance up to 3 km (Table 1). Only in rare cases (up to 4 %), trace amounts of corrosion radionuclides were determined in the observation area, adjacent to the NPP. Constant ^{137}Cs and ^{90}Sr presence in seawater is explained by the global processes of technogenic radiation background formation. An increase in these radionuclides specific activity was noted in the discharge channels of the 1st and 2nd stages of the LNPP and the adjacent water area.

Table 4. Reference levels of radionuclide content in seawater according to environmental and radiation-hygienic criteria

Radionuclide	Reference levels of activity concentration in water, Bq·L^{-1}		
	according to environmental criterion	according to radiation-hygienic criterion	minimum value, that meets both environmental and radiation-hygienic criteria (Poryadok rascheta…, 2016)
^{137}Cs	180	1.22	1.22
^{134}Cs	70	0.838	0.8382
^{90}Sr	490*	16.8	16.82
^{54}Mn	0.27	1.91	0.272
^{60}Co	0.61	0.628	0.612
^{65}Zn	11	0.116	0.1162
^{95}Zr	0.31	62.7	0.312
^{95}Nb	0.74	83.7	0.742
^{131}I	420	8.04	8.042
^{141}Ce	2.0	14.2	2.02
^{144}Ce	0.72	1.94	0.722
^{3}H	1 000 000*	86 800	86 800
^{14}C	72	0.137	0.1372

Note: * – according to the criterion of classification as liquid radioactive waste.

The LNPP effect on radionuclides activity in sea bottom sediments was traced at a distance up to 10 km; a decrease in the technogenic radionuclides activity in bottom sediments and a decrease in the frequency of detection of radionuclides of the power plant origin with distance from the NPP were recorded.

Due to the accumulation processes, the probability of detecting technogenic radionuclides in water plant samples is significantly higher than in water samples. The LNPP effect on radionuclides activity in marine plants was traced at a distance over 10 km. A decrease in radioactive contamination of marine plants with a distance from the LNPP was recorded. In general, water plants are an informative bioindicator; that makes it possible to trace the spread of technogenic radionuclides in the NPP area.

The highest levels of technogenic radionuclide activity ratios in fish tissues were registered in the LNPP discharge channels. Corrosion radionuclides (^{54}Mn, ^{60}Co, and ^{65}Zn) were recorded in several cases in fish tissue samples from the Koporye Bay, along with the constantly present ^{137}Cs and ^{90}Sr. The biological transfer of trace amounts of these radionuclides into the rivers, flowing into the Koporye Bay, is of certain interest.

The Chernobyl disaster led to a noticeable increase in the Koporye Bay contamination with technogenic radionuclides. The value of the sum of observed technogenic radionuclide activity ratios of Chernobyl origin in seawater in May – December 1986 to the values of the reference levels increased in comparison with that of the pre-accidental period by about 100 times. During this period, the indicator was close to the value, at which the condition (4), providing the preservation of a favorable environment, is violated. The increase in the value was mainly associated with cesium radioisotopes, the reference levels for which in seawater are limited by the radiation-hygienic criterion.

The formation of current dynamics of contamination of the marine ecosystem components with technogenic radionuclides is still affected by the consequences of the Koporye Bay contamination by the "Chernobyl" ^{137}Cs. At the same time, a decrease in the LNPP discharges to the bay is registered. According to the long-term monitoring data, more than 30 years after the Chernobyl disaster, ^{137}Cs concentration in bottom sediments, seawater, and fish tissues of the Koporye Bay still exceeds the pre-accidental level, while the content of corrosion radionuclides of the LNPP origin in bottom sediments and seawater has significantly decreased. Carbon-14 and tritium, both of natural and technogenic origin, are of particular importance in the practice of radioecological monitoring in this area.

Conclusion. Analysis of the long-term radioecological monitoring data (1973–2019) in the Leningrad NPP area allows to conclude that technogenic radionuclide activity ratios in the marine ecosystem components of the Koporye Bay of the Gulf of Finland under normal NPP operating conditions do not exceed the reference levels, which are presented in the Roshydromet Recommendations and provide the preservation of a favorable environment. Several characteristic periods can be distinguished in the dynamics of the radiation state in this area. In the early period of NPP operation (1973–1985), a wide range of technogenic radionuclides was registered in the marine ecosystem components. Along with ^{137}Cs, significant contributors to the contamination of seawater and bottom sediments were ^{54}Mn and ^{60}Co, the reference levels for which in seawater, in contrast to those for ^{137}Cs, are determined by an environmental criterion, not a radiation-hygienic one. The widest range of technogenic radionuclides is recorded in algae – an informative bioindicator, that allows tracing the spread of technogenic radionuclides in the NPP area at a distance over 10 km. Biogenic transfer of corrosion radionuclides (^{54}Mn, ^{60}Co, and ^{65}Zn) by fish into rivers, flowing into the Koporye Bay, was traced.

After the Chernobyl disaster, there was a significant increase in cesium radioisotopes content in the marine ecosystem components. After a decay of relatively short-lived "Chernobyl" radionuclides, ^{137}Cs acquired the main radioecological significance in this area; its content increased in May – December 1986, compared with the pre-accidental level: in seawater – by 130 times, in algae – by 250 times, and in bottom sediments – by 30 times. For predatory fish, the effect of radiocesium accumulation at trophic levels was observed. The maximum specific activity of ^{137}Cs in fish muscles was recorded a year after the accidental pollution – in 1987; the values remained high, and even in 1990 they were 3–8 times higher than the activities of non-predatory species.

Currently (2010–2019), the radioactive contamination of the marine ecosystem components of the Koporye Bay is determined mainly by ^{137}Cs presence; according to the monitoring data, its level is relatively constant, which indicates the stability of the radiation state in the LNPP marine cooling reservoir. Taking into account the NPP potential radiation hazard, as well as the state of joint effect on the marine biota of technogenic ionizing radiation and a set of non-radiational factors (thermal discharge, chemical pollution, and injury of hydrobionts in the NPP water intakes), it can be concluded that it is necessary to continue and develop radioecological monitoring of the LNPP marine cooling reservoir.

The work was carried out within the framework of Roshydromet research and technological work, theme 4.3.1 "Modernization and development of state monitoring of the radiation state in the Russian Federation territory" (2020–2024).

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ОЦЕНКА РАДИАЦИОННОЙ БЕЗОПАСНОСТИ МОРСКОЙ СРЕДЫ В РАЙОНЕ РАСПОЛОЖЕНИЯ ЛЕНИНГРАДСКОЙ АЭС ПО ДАННЫМ МНОГОЛЕТНЕГО МОНИТОРИНГА (1973–2019)

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Целью работы было оценить радиоэкологическое состояние морского водоёма — охладителя Ленинградской АЭС — Копорской губы Финского залива. Согласно международным основным нормам безопасности, принятым на генеральной конференции МАГАТЭ, этот вопрос имеет особую актуальность в связи с необходимостью обоснования защиты от промышленного радиационного воздействия не только человека, но и окружающей среды. Как исходные материалы для оценки использованы данные многолетнего мониторинга (1973–2019) содержания радионуклидов в морской воде, донных отложениях и гидробионтах в зоне наблюдения Ленинградской АЭС. В качестве показателей радиационного состояния морской среды применены контрольные уровни содержания радионуклидов в морской воде и донных отложениях, порядок расчёта которых определён в Рекомендациях Росгидромета Минприроды России Р 52.18.852-2016 и Р 52.18.873-2018. В этих рекомендациях, разработанных специалистами ФГБУ НПО «Тайфун», представлена методика оценки радиоэкологического состояния морской среды по уровню активности радионуклидов на основе принципов, обеспечивающих сохранение благоприятной окружающей среды и морских гидробионтов, а также радиационную защиту человека. При наличии в морской среде смеси радионуклидов должно быть выполнено условие непревышения единицы для суммы отношений наблюдаемых концентраций техногенных радионуклидов в морской воде (донных отложениях) к соответствующим значениям контрольных уровней. По данным мониторинга в ранний период эксплуатации АЭС (1973–1985), в компонентах морской экосистемы присутствовал широкий спектр техногенных радионуклидов. Наряду с ^{137}Cs заметный вклад в загрязнение морской воды и донных отложений вносили ^{54}Mn и ^{60}Co, контрольные уровни для которых в морской воде, в отличие от таковых ^{137}Cs, определяются по экологическому, а не радиационно-гигиеническому критерию. Зарегистрировано присутствие техногенных радионуклидов в водорослях на расстоянии свыше 10 км от АЭС. Отмечен биогенный перенос коррозионных радионуклидов (^{54}Mn, ^{60}Co, ^{65}Zn) рыбой в ре́ки, впадающие в Копорскую губу. Чернобыльская авария привела к заметному увеличению загрязнения Копорской губы техногенными радионуклидами. По сравнению с доаварийным периодом значение суммы отношений наблюдаемых концентраций техногенных радионуклидов в морской воде к соответствующим значениям контрольных уровней в мае — декабре 1986 г. возросло в 100 раз, а в донных отложениях — в 30 раз. Основной вклад в загрязнение компонент морской экосистемы в 1986 г. вносили ^{137}Cs и ^{134}Cs. В настоящее время загрязнение морской воды и донных отложений Копорской губы определяется в основном ^{137}Cs и находится, по данным наблюдений, на относительно постоянном уровне, свидетельствующем о стабильности радиоэкологической обстановки в морском водоёме — охладителе АЭС.

Ключевые слова: Копорская губа Финского залива, Ленинградская АЭС, радиоэкологический мониторинг, морская вода, донные отложения, контрольные уровни радионуклидов, экологический и радиационно-гигиенический критерии, интегральный показатель загрязнения, многолетняя динамика