

Artificial Radionuclides ^{137}Cs and ^{90}Sr in the Components of the Ecosystems of the Salt Lakes of the Crimea

Mirzoyeva Natalya*

FSBIS Institute of Marine Biological Research named A.O. Kovalevsky, Russian Academy of Sciences (FSBIS IMBR RAS), Sevastopol, the Crimea, 299011, Russia

Abstract:

In 2016, for the first time a radioecological study was made of 11 salt lakes of the Crimea to investigate the contamination of their ecosystems by ^{137}Cs and ^{90}Sr . There was a positive correlation between the salt content and the concentration of anthropogenic radionuclides ^{90}Sr and ^{137}Cs in the water of the studied groups of lakes. Absorbed doses of ^{137}Cs and ^{90}Sr radiation in aquatic plants in the lakes were $7.7 \cdot 10^{-6}$ and $3.2 \cdot 10^{-6}$ Gy/year, respectively, and lay within the “Uncertainty Zone” according to the scale “Zones of Chronic Exposure to Ionizing Radiation”, proposed by Polikarpov.

Keywords: Crimean salt lakes; Black sea; Chernobyl NPP accident; ^{90}Sr ; ^{137}Cs ; Absorbed doses

Introduction

More than 50 continental and marine salt lakes are located in the Crimea^[1-3]. They contain an almost inexhaustible supply of sodium, magnesium, bromine and other chemical elements^[1] and represent a unique ecosystem with the original hydrochemical regime and structure of communities of living organisms^[3-7]. Many of the salt lakes of the Crimea are used for recreational and economic purposes^[1,3,8]. The high salinity of water in these lakes is maintained mainly due to the intensive evaporation, especially in summer. This can lead to the concentration of many chemical elements, including and radioactive materials^[1,3,4,9].

The entry of artificial radionuclides into the environment is due to both open tests of nuclear weapons and accidents at enterprises with technologies that use nuclear energy. The significance of ^{137}Cs and ^{90}Sr in the environment as a result of the Chernobyl NPP accident (89 and 7.4 PBq, respectively) can be compared with the formation of these radionuclides due to nuclear weapons tests in open media: 1,300-1,500 PBq for ^{137}Cs and 650-1,300 PBq for ^{90}Sr , and also as a result of other nuclear incidents^[10-13].

In 1986, as a result of the Chernobyl NPP accident, the Crimean region was exposed to a primary radioactive contamination by means of transport of air masses from the accident area^[10,14]. In the post-accident period, the radioecological situation in the Crimea was determined by secondary radionuclide contamination, primarily ^{90}Sr . From the beginning of 1986 until the end of May 2014 this radionuclide in dissolved form had been entering with the Dnieper river water through the North-Crimean Canal (NCC) to the Crimean region and into the Crimean inland water reservoirs^[13,15-17].

The main feature of the Chernobyl NPP accident was that the radioactive pollution of the environment took place within time scales considerably smaller than the typical time for the occurrence of biogeochemical processes. Therefore for larger timescales ^{90}Sr and ^{137}Cs , as quality radiotracers, can characterize the behaviour of hydrological and biogeochemical processes occurring in aquatic ecosystems.

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***Corresponding author:** Natalya Mirzoyeva, FSBIS Institute of Marine Biological Research named A.O. Kovalevsky, Russian Academy of Sciences (FSBIS IMBR RAS), Sevastopol, the Crimea, 299011, Russia, E-mail: natmirz@mail.ru

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The aim of this investigation was to conduct a comparative study of the salt lakes of the Crimea in regard to migration and redistribution of the post-accident ⁹⁰Sr and ¹³⁷Cs within the components of the studied aquatic ecosystems, and determination of the rate of the biogeochemical processes in the salt lakes of the Crimea using of ⁹⁰Sr and ¹³⁷Cs as radiotracers.

In accordance with the formulated purpose of investigations the following tasks were performed:

To determine the concentration and peculiarity of redistribution of the ⁹⁰Sr and ¹³⁷Cs in the components of aquatic ecosystems of the lakes;

To conduct a comparative analysis of the content of ⁹⁰Sr and ¹³⁷Cs in the water of the Black Sea ecosystems located in areas close to the location of the salt lakes and the content of these radionuclides in the lake water, and to identify possible sources of intake of ⁹⁰Sr and ¹³⁷Cs into the aquatic ecosystems;

To calculate the exposure dose received by various ecological groups of hydrobionts of the salt lakes of the Crimea from the ionizing radiation of ⁹⁰Sr and ¹³⁷Cs in the post-accident period.

Materials and Methods

Sample sites and samples characteristics

In 2016, for the first time in the history of the salt lakes of the Crimea, as well as for the entire period after the nuclear weapons test and after the Chernobyl NPP accident, a radioecological study was conducted on the contamination of the ecosystems of 11 salt lakes by ¹³⁷Cs and ⁹⁰Sr. The following lakes were investigated: Krasnoye, Kiyatskoye, Kirleutskoye lakes from the Perekopskaya group, Dzarilgach, Bakalskoye lakes from the Tarkhankutskaya group, Sasyk-Sivash, Kyzyl-Yar, Moinakskoe lakes from the Evpatoriyskaya group, and Tobechikskoe, Chokrakskoe, Aktashskoe lakes from the Kerchenskaya group [Table 1, Figure.1]. They have a marine origin (closed lagoons), are drainless, and can be temporally fully or partly dried[1,18-20].

Table 1: Coordinates and Characteristics of the Sampling Stations

Name of objects of study/ (material of research)	Sampling date	Sampling co-ordinates	Salinity, ‰	pH
Perekopskaya group				
Lake Kiyatskoe (water, bottom sediments (0-5 cm))	14.06.2016	45°59.729' N 33°53.310' E	200.0	7.7
Lake Kirleutskoe (water, bottom sediments (0-5 cm), <i>cysts of Artemia</i>)	14.06.2016	45°55.231' N 34°02.681' E	235.0	7.9
Lake Krasnoe (water, bottom sediments (0-5 cm))	14.06.2016	45°59.437' N 33°57.319' E	330.0	9.3
Tarkhankutskaya group				
Lake Dzarilgach (water, bottom sediments (0-5 cm), <i>cysts of Artemia</i>)	18.05.2016	45°33.965' N 32°54.599' E	115.0	8.5
	08.11.2016	45°33.968' N 32°51.582' E	140.0	7.9

Lake Bakalskoe (water, bottom sediments (0-5 cm); water plants <i>Polysiphonia subulifera</i>)	27.06.2016	45° 45.514' N 33° 10.794' E	46.5	8.6
Yevpatoriyskaya group				
Lake Kyzyl-Yar (water, bottom sediments (0-5 cm); water plants <i>Stuckenia pectinata</i>)	18.05.2016	45°03.560' N 33°35.360' E	3.5	7.9
	06.09.2016	45°03.560' N 33°35.360' E	3.1	
Lake Sasik-Sivash (water, bottom sediments (0-5 cm))	27.06.2016	45° 09.151' N 33° 30.447' E	280.0	7.7
Lake Moinakskoe (water)	18.05.2016	45°10.518' N 33°18.597' E	47.0	8.2
Kerchenskaya group				
Lake Chokrakskoe (water, bottom sediments (0-5 cm))	08.06.2016	45°27.508' N 36°18.325' E	226.0	7.9
Kerchenskaya group				
Lake Aktashskoe (water, bottom sediments (0-5 cm), <i>cysts of Artemia</i>)	11.04.2016	45°22.219' N 35°46.421' E	270.0	7.4
Lake Tobechikskoe (water, bottom sediments (0-5 cm))	07.06.2016	45°09.118' N 36°22.490' E	176.0	8.20
Adjacent stations of the Black Sea along the coast of the Crimea				
The sea near Lake Bakalskoye (water)	18.05.2016	45° 47.190' N 32° 59.740' E	17.5	8.4
The sea near Tarkhankut Cape (Water; water plants <i>Cystoseira sp.</i>)	09.06.2016	45° 15.500' N 32° 29.670' E	17.3	8.4
the Black Sea, Yevpatoria Bay (water, bottom sediments)	22.04.2016	44°23.000'N 33°40.330' E	17.3	8.3
Sevastopol bays, the Black Sea (water, bottom sediments)	06.06.2016	44° 36.554' N 33° 28.215' E	17.0	8.2
the Black Sea, the Kerch Strait (water, bottom sediments)	24.04.2016	45°49.978' N 36°00.089' E	17.2	8.5
The Sea of Azov (water)	09.06.2016	45° 29.990' N 36° 00.220' E	14.1	8.1



Figure.1: Map-scheme of the sampling stations in the Crimea region (2016)

We collected and analyzed 36 samples of water, 18 samples of bottom sediments, 6 samples of water plants (*Cystoseira* sp., *Polysiphonia subulifera* (C. Agardh) Harvey, *Stuckenia pectinata* (L.) BÖRNER) and cysts of *Artemia*. Samples of sea water in the Black Sea areas located close to the salt lakes were collected to conduct a comparative analysis on the content of ^{90}Sr and ^{137}Cs with the concentrations of these radionuclides in the water of the salt lakes, and to identify possible sources of entry of ^{90}Sr and ^{137}Cs into the aquatic ecosystems.

^{90}Sr radiochemical procedures

The method of ^{90}Sr determination was based on the following radiochemical procedure. After acid leaching and/or preconcentration of strontium with a carbonate (for water) or an oxalate (for hydrobionts and bottom sediments), purification from interfering elements is performed by hydroxide precipitation. After equilibrium between ^{90}Sr and the daughter product ^{90}Y (at least 14 days), ^{90}Y is separated from the ^{90}Sr solution and measured by Cerenkov's radiation in a low background liquid-scintillation counter (LSC) LKB "Quantulus 1220". The Lower Limit of Detection (LLD) was 0.01-0.04 Bq/kg for hydrobionts and bottom sediments and the limit (in Bq/m³) was similar for water samples. Recoveries are calculated from stable Sr recovery by flame photometry for ^{90}Sr and gravimetrically from yttrium oxalate for ^{90}Y ^[21, 22]. Each result is reported as the mean of the values activity of parallel duplicate samples, which were measured separately. Total relative error of the each result does not exceed 20 %.

The quality of the analytical methods and the reliability of the results were supported by the constant participation in international intercalibrations during 1990-2004 under the aegis of the IAEA (Vienna, Austria). Results of the IBSS participation in the intercalibration were included in the intercalibration report materials^[23, 24] and they were certified as reliable data.

Gamma-spectrometric measurements of ^{137}Cs

The ^{137}Cs content was measured using a "1282-CompuGamma CS" gamma spectrometer (LKB Wallac, Finland) with a NaI (Tl) scintillation detector, as well as the butt semiconductor gamma detectors Canberra-Packard XtRa GX2019 and ORTEC GMX-10 (USA), made on the basis of crystals of ultra-pure germanium, with a relative efficiency of 16-23 %. Analysis of the obtained gamma spectrum was carried out with the help of the Canberra-Packard MCA S100 analyzer, System 100^[13].

Radiological dose calculation

The radiological dose (Gy/y) for the hydrobionts were calculated using individual coefficient DCF (dose-rate conversion factors) and the mean of ^{90}Sr concentrations for each group of hydrobionts, as well for water and bottom sediments from the habitat area of the hydrobionts^[25, 26]. Values of dose conversion factors for calculation of internal and external doses of ^{90}Sr for aquatic organisms were taken from worksheets of the computer program of the RAD-BCG Calculator^[26]. The dose estimates were compared with the dose limits for aquatic organisms from DOE Standard (2001) and with the scale of Zones of chronic dose rates and their effects in the biosphere proposed by Polikarpov (1998)^[27].

Results and Discussion

^{90}Sr and ^{137}Cs in the water of the aquatic ecosystems of the salt lakes of the Crimea

Among all the studied objects the greatest concentration of the artificial radionuclides ^{90}Sr and ^{137}Cs in water in 2016, as well as one of the highest levels of salt concentrations, was observed in the Sasik-Sivash Lake of the Evpatoriyskaya group, exceeding the concentrations of ^{90}Sr 3.7-38 times and of ^{137}Cs 2-120 times those found in the water of all other lakes [Table 2, Figure 2].

Table 2: Concentrations ^{137}Cs and ^{90}Sr in water of the salt lakes of the Crimea

Name of objects of study	Sampling date	Salinity, ‰	Concentration, Bq/m ³	
			^{137}Cs	^{90}Sr
Perekopskaya group				
Lake Krasnoe	14.06.2016	330	37.4 ± 0.2	54.7 ± 21.3
Lake Kiyatskoe	14.06.2016	235	5.3 ± 0.3	60.1 ± 4.7
Lake Kirleutskoe	14.06.2016	200	2.3 ± 0.1	18.7 ± 2.3
Tarkhankutskaya group				
Lake Dzarilgach	18.05.2016	115	22.2 ± 2.4	54.6 ± 4.1
Lake Bakalskoe	27.06.2016	46.5	31.4 ± 2.7	40.0 ± 2.7
Yevpatoriyskaya group				
Lake Kyzyl-Yar	18.05.2016	3.5	0.8 ± 0.04	9.3 ± 1.1
Lake Sasik-Sivash	27.06.2016	280	95.9 ± 8.1	313.6 ± 25.0
Lake Moinakskoe	18.05.2016	47	29.8 ± 1.4	8.3 ± 1.1
Kerchenskaya group				
Lake Tobechikskoe	07.06.2016	176	8.7 ± 0.4	28.9 ± 2.2
Lake Chokrakskoe	08.06.2016	226	49.3 ± 3.1	46.0 ± 3.4
Lake Aktashskoe	08.06.2016	88.5	11.5 ± 0.5	85.8 ± 3.9

In the first months after the Chernobyl NPP accident the maximum concentrations of ^{137}Cs and ^{90}Sr were 134.1 and 53.0 Bq/m³, respectively^[12, 13]. To identify the sources of these artificial post-accident radionuclides entering the salt lakes of the Crimea, we assume that the same concentrations of ^{137}Cs and ^{90}Sr were present in the water of the investigated objects in the first months after the Chernobyl NPP accident [Figure. 1]. Values exceeding these values of concentrations of ^{137}Cs and ^{90}Sr , taking into account their decay by 2016, indicates a secondary source of entry of these pollutants into the studied reservoirs, primarily with the Dnieper water along the North-Crimean canal^[28-30]. We believe that the lower values of the ^{137}Cs and ^{90}Sr concentrations in the water of the salt lakes of the Crimea compared to the maximum concentration of these radionuclides that fell with atmospheric transport on the territory of the Crimea after the Chernobyl NPP accident and corrected for decay by 2016 can be explained either by the absence of secondary sources of post-accident radionuclides, or their redistribution by components of the ecosystems of these reservoirs.

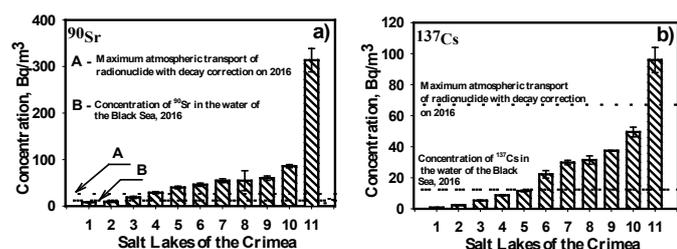


Figure.2: Ranking of the concentration of ^{90}Sr (a) and ^{137}Cs (b) in the water of salt lakes of the Crimea (sampling 2016)

1 – Kyzyl-Yar Lake, 2 – Kirlautskoe Lake, 3 – Kiyatskoe Lake, 4 – Tobechikskoe Lake, 5- Aktashskoe Lake, 6 – Dzarilgach Lake, 7 – Moinakskoe Lake, 8 – Bakalskoe Lake, 9 – Krasnoe Lake, 10 – Chokrakskoe Lake, 11 – Sasik-Sivash Lake; b) 1 - Kyzyl-Yar Lake, 2 – Moinakskoe Lake, 3 – Kirlautskoe Lake, 4 – Tobechikskoe Lake, 5 – Dzarilgach Lake, 6 – Bakalskoe Lake, 7 – Chokrakskoe Lake, 8 – Krasnoe Lake, 9 – Kiyatskoe Lake, 10 – Aktashskoe Lake, 11 – Sasik-Sivash Lake

In 2016 the concentration of ^{90}Sr in the water of practically all salt lakes studied, except for the Moinakskoe and Chokrakskoe lakes, was 1.3-12.3 times higher than that for ^{137}Cs [Table 2, Figure 2]. At the same time, the initial entry of ^{137}Cs was 2.5 times higher than the initial entry of ^{90}Sr on the water area of Crimea's water bodies^[13]. These differences can first of all be explained by the secondary entry of dissolved forms of ^{90}Sr with the waters of the Dnieper through the NCC into the salt lakes of the Crimea, and by the peculiarities of the ^{90}Sr and ^{137}Cs redistribution between the components of the ecosystems of these water objects.

The content of ^{137}Cs and ^{90}Sr in the water of all studied lakes, whose salinity was higher than that of the Black Sea, exceeded by 2-34.5 times the concentration of these radionuclides in the waters of the adjacent areas of the Black Sea [Table 2, Figure. 2]. It is known that alkali metal chlorides and other salts sharply increase the solubility of strontium salts by 25 times^[1]. ^{137}Cs also has a very high solubility in salt water^[31,32]. A positive correlation between the increase of salinity and the retention of radionuclides (^{137}Cs and ^{90}Sr) in the water column of the salt lakes was observed [Figure 3].

Thus, the level of the content of the artificial radionuclides ^{137}Cs and ^{90}Sr in the salt lake ecosystems was primarily determined by the sources of their entry into the water bodies. Subsequently, the redistribution of these radionuclides depended on the levels of salinity of the water in the lakes [Table 2, Figure 3] and their hydrochemical and hydrological properties.

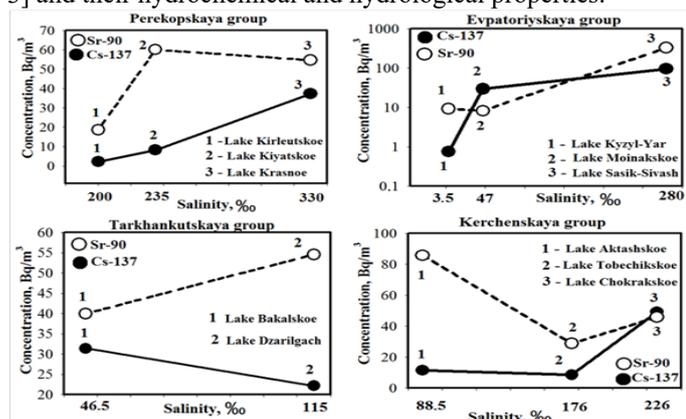


Figure 3: Concentrations of ^{137}Cs и ^{90}Sr depending on salinity of water of the salt lakes of the Crimea (sampling 2016)

It may be noted that in 2016 the concentration of ^{90}Sr and ^{137}Cs in the water of all the investigated salt lakes of the Crimea and the control sampling stations did not exceed the maximum permissible concentration for ^{90}Sr in drinking water (RSS-99/2009).

^{90}Sr and ^{137}Cs in the bottom sediments and water plants of the aquatic ecosystems of the salt lakes of the Crimea

According to the results of our investigations, which were obtained in 2016, no direct correlation was found between the salinity of water and the ^{90}Sr and ^{137}Cs concentrations in the bottom sediments of the salt lakes. The lowest concentrations of ^{90}Sr and ^{137}Cs were observed in the bottom sediments of Lake Sasik-Sivash, the highest concentrations were in Lake Kyzyl-Yar. So the concentrations of both ^{90}Sr and ^{137}Cs in the bottom sediments showed the opposite trend of the concentrations of these radionuclides in the water of these lakes [Table.2, Table 3 and Figure.4].

The concentration of ^{90}Sr in the bottom sediments of 6 out of 10 lakes was lower than that for the bottom sediments of the Black Sea. The ^{137}Cs content in the bottom sediments of all investigated reservoirs was 1.6-32 times lower than the values for marine bottom sediments [Figure.4]. This indicates lower rates of biogeochemical processes in the salt lakes in comparison with the Black Sea. The flow of the radioactive contaminants from the water column to the bottom sediments of the lakes is slowed down. The high salinity of the lakes, exceeding that of the Black Sea water, contributed to keeping the dissolved forms of ^{137}Cs and ^{90}Sr in the aquatic environment, and it reduced the deposition of post-accident radionuclides into the bottom sediments of the studied reservoirs [Figure 4].

Table 3: Concentrations ^{137}Cs and ^{90}Sr in the bottom sediments of the salt lakes of the Crimea

Name of objects of study	Sampling date	Salinity, ‰	Concentrations, Bq·kg ⁻¹ DW	
			^{137}Cs	^{90}Sr
Perekopskaya group				
Lake Krasnoe	14.06.2016	330	below detection level	2.2 ± 0.3
Lake Kiyatskoe	14.06.2016	235	below detection level	3.3 ± 0.4
Lake Kirlautskoe	14.06.2016	200	below detection level	2.6 ± 0.5
Tarkhankutskaya group				
Lake Dzarilgach	18.05.2016	115	6.2 ± 1.2	22.7 ± 1.3
Lake Bakalskoe	27.06.2016	46.5	5.8 ± 1.4	0.7 ± 0.2
Yevpatoriyskaya group				
Lake Kyzyl-Yar	18.05.2016	3.5	24.4±6.7	15.0 ± 2.1
Lake Sasik-Sivash	27.06.2016	280	4.1 ± 2.5	2.0 ± 0.2
Kerchenskaya group				
Lake Tobechikskoe	07.06.2016	176	16.5 ± 1.6	5.0 ± 0.8
Lake Chokrakskoe	08.06.2016	226	13.5 ± 1.3	5.1 ± 0.5
Lake Aktashskoe	08.06.2016	88.5	5.3 ± 0.9	2.6 ± 0.4

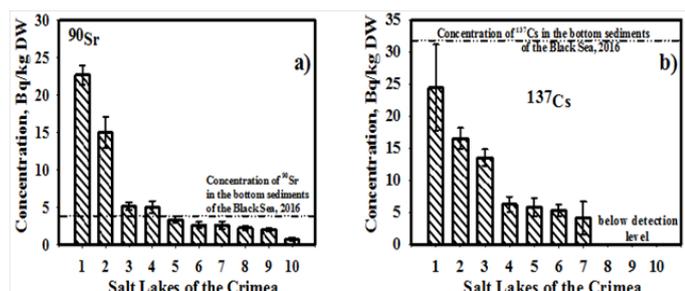


Figure 4: Ranking of the concentration of ^{90}Sr (a) and ^{137}Cs (b) in the bottom sediments of salt lakes of the Crimea (sampling 2016)

a) 1 – Lake Dzarilgach, 2 – Lake Kyzil-Yar, 3 – Lake Chokrakskoe, 4 – Lake Tobechnikskoe, 5 – Lake Kiyatskoe, 6 – Lake Aktashskoe, 7 – Lake Kirluetskoe, 8 – Lake Krasnoe, 9 – Lake Sasik-Sivash, 10 – Lake Bakalskoe; b) 1 - Kyzil-Yar Lake, 2 – Lake Tobechnikskoe, 3 – Lake Chokrakskoe, 4 – Lake Dzarilgach, 5 – Lake Bakalskoe, 6 – Lake Aktashskoe, 7 – Lake Sasik-Sivash

The concentrations of the ^{90}Sr and ^{137}Cs in aquatic plants, as well as in the bottom sediments of the lakes were insignificant [Figure. 4, 5].

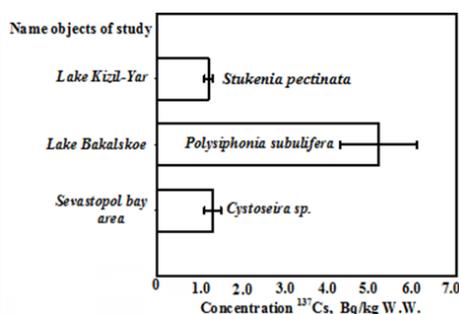


Figure 5: ^{137}Cs in the water plants of salt lakes of the Crimea and of the Black Sea

The concentrations of ^{137}Cs and ^{90}Sr in cysts of *Artemia* which were collected in lakes Kirluetskoye, Dzarilgach and Aktashskoye in 2016 were below the detection levels. *Artemia* cysts are relatively radioresistant to long-lived post-accident radionuclides such as ^{137}Cs and ^{90}Sr .

The absorbed doses from ionizing radiation of ^{137}Cs and ^{90}Sr on the water plants *Stuckenia pectinata* (Lake Kyzil-Yar) and *Polysiphonia subulifera* (Lake Bakalskoe) amounted to $7.7 \cdot 10^{-6}$ Gy per year and $3.2 \cdot 10^{-6}$ Gy per year, respectively. They were within the “Uncertainty Zone” according to the scale “Chronic Exposure to Ionizing Irradiation”, proposed by Polikarpov (1998), *i.e.*, they did not have a noticeable effect on aquatic plants in the period after the Chernobyl NPP accident.

Conclusions

The main sources of the artificial radionuclides ^{137}Cs and ^{90}Sr after the Chernobyl NPP accident have been identified in the salt lakes of the Crimea. The primary entry of radionuclides to waters of the lakes occurred as a result of the atmospheric transport from the site of the Chernobyl NPP accident to a remote region such as the Crimea by May 1986. Subsequently (until 2014), dissolved radionuclides entered with the Dnieper waters through the North-Crimean canal. In 2016, the concentration of ^{90}Sr in the water in virtually all the lakes was 1.3-12.3 times higher than that for ^{137}Cs . At the same time, the

initial entry of ^{137}Cs was 2.5 times higher than the initial entry of ^{90}Sr . The secondary entry of ^{90}Sr into the ecosystems of the salt lakes with the waters of the NCC and the Black Sea is more significant than its atmospheric transport after the Chernobyl NPP accident. For ^{137}Cs this way of entering into the salt lakes is less important than for ^{90}Sr . The content of the artificial radionuclides ^{137}Cs and ^{90}Sr in the salt lake ecosystems was primarily determined by the sources of their entry into water bodies. Subsequently, the redistribution of these radionuclides depended on the levels of salinity of the lakes and their hydrochemical and hydrological properties.

A positive correlation between the increase of salinity and the retention of the radionuclides in the water column of the salt lakes was observed. Their salinity, exceeding that of the Black Sea, contributed to the stability of the dissolved forms of ^{137}Cs and ^{90}Sr in the aquatic environment and reduced their deposition into the bottom sediments of the studied reservoirs. In 2016, the highest concentrations of ^{137}Cs and ^{90}Sr in the water column and the lowest content of these radionuclides in bottom sediments were observed in Lake Sasik-Sivash (water salinity 280 g/L).

The investigated lakes are drain less, so that radioactive material that had entered these aquatic ecosystems accumulate over time and redistribute between the components of these water objects.

In 2016 the concentration of ^{90}Sr and ^{137}Cs in the water of all the investigated lakes and the control sampling stations did not exceed the maximum permissible concentration for ^{90}Sr in drinking water [33].

The cysts of *Artemia* which were collected in lakes Kirluetskoye, Dzarilgach and Aktashskoye in 2016 are relatively radioresistant to such long-lived post-accident radionuclides as ^{137}Cs and ^{90}Sr .

The absorbed doses from ionizing radiation of ^{137}Cs and ^{90}Sr on the water plants *Potamogeton pectinatus* (Lake Kyzil-Yar) and *Polysiphonia subulifera* (Lake Bakalskoe) did not have a noticeable radiation effect on these hydrobionts in the period after the Chernobyl NPP accident.

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