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ORIGINAL ARTICLE

Features of ^{90}Sr behavior in Crimean lakes with different salinity of their water environment

N. Yu. Mirzoeva[1] · S. I. Arkhipova[1] · V. Yu. Proskurnin[1] · O. N. Miroshnichenko[1] · I. N. Moseichenko[1]

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Abstract The comparative study of technogenic ^{90}Sr behavior features in Crimea lakes' ecosystems with different levels of salinity was carried out in 2016–2021. Two sources of ^{90}Sr input were identified for all the studied lakes: the primary source concerned with atmospheric fallout immediately after the Chernobyl NPP accident and the secondary long-term input of this radionuclide by waterway. The half-life of the ^{90}Sr concentration in the water of the hypersaline Lake Sasyk-Sivash was estimated to vary from 0.8 to 1.1 years after the closure of the North Crimean Canal (NCC). Biogeochemical processes in the lake under the absence of the secondary source of the radionuclide input were shown to decrease in the ^{90}Sr residence time in the water column by 131 times. For brackish water bodies, a significant factor influencing the radionuclide concentration in ecosystems of lakes was the pH of their water, while for hypersaline lakes the level of water salinity was the main factor determining ^{90}Sr behavior. The concentration of ^{90}Sr in bottom sediments of studied lakes depended mainly on this radionuclide concentration in a water environment. Calculated ^{90}Sr distribution factors (K_d) for studied lakes' bottom sediments varied in a range of $n \cdot 10^0 \div n \cdot 10^2$ for hypersaline lakes and of $n \cdot 10^1 \div n \cdot 10^2$ for lakes with brackish waters. Due to the closure of the NCC, the ^{90}Sr redistribution took place in lake ecosystems only under the geochemical processes within the water bodies themselves. The results obtained in this work are of particular importance as a starting point or a basis for further radioecological studies of the Crimean inland waters after the reopening of the NCC and the Dnieper waters re-entering the territory of Crimea in 2022 after the 8 years of their absence.

Keywords ^{90}Sr · Salt lakes · Water · Bottom sediments · Chernobyl NPP accident · Crimea

✉ O. N. Miroshnichenko
oksaniya_89@mail.ru

[1] A.O. Kovalevsky Institute of Biology of the Southern Seas of RAS, Sevastopol, Russian Federation

1 Introduction

Resulting of nuclear weapons testing and accidents at facilities using nuclear energy, primarily accidents at nuclear power plants, contamination of the Earth's surface with artificial radionuclides has become global to date. Determination of the behavior features of radionuclides introduced into natural water bodies is one of the important tasks of modern aquatic geochemical studies.

The ^{90}Sr isotope is considered one of the most dangerous technogenic radionuclides for biota and humans. This is due to the effects of its ionizing radiation on living organisms, high toxicity (25 times more than the toxicity of other nuclear fission products), and significant half-life (28.78 years) (Guillen et al. 2010; Budarkov et al. 2000; Vasilenko and Vasilenko 2002; UNSCEAR, 2010, 2011a). ^{90}Sr is a chemical analogue of calcium. It can replace Ca in the bone tissue of living organisms and this way be firmly fixed, causing chronic exposure effects throughout the life of biological species (Vasilenko and Vasilenko 2002; Higley et al. 2003; Guillen et al. 2010; UNSCEAR 2011b; US DOE, 2019; Kryshev et al. 2020; Marx et al.2020).

Radionuclides in natural waters are known to exist in various chemical and physical forms, depending on their release and transfer features, as well as on the properties of the elements involved in these processes (UNSCEAR 2011b; Poinssot and Geckeis 2015; Petrucci et al. 2017; US DOE 2019). According to their ability to be leached from insoluble matrices to the aquatic environment, radionuclides can be ranged in the following row: $^{89}Sr > ^{90}Sr > ^{125}Sb > ^{105}$, $^{106}Ru > ^{137}Cs > ^{141}Ce >$ (^{144}Ce, ^{91}Y, ^{95}Zr, ^{60}Co, ^{54}Mn) with quantitative ratio characterizing such ability as: 25: 1: 0.3: 0.1: 0.004: 0.02: (0.01 $\div$ 0.001) (Polikarpov et al. 2008). Studies on the ^{90}Sr solubility in water (Chudinovskikh et al. 2004; Romanenko 2004; Guillen et al. 2010) showed the following compounds – $Sr(OH)_2$, $SrCl_2$, $Sr(NO_3)_2$ – to be easily soluble in water, while such compounds as $SrCO_3$, $SrSO_4$ and SrO appeared to be insoluble. Speaking about its aquatic geochemistry, the main feature of ^{90}Sr is that in most of its compounds in the aquatic environment it is present in a soluble cationic (Sr^{2+}) form (Romanenko 2004; Polikarpov 2008; Guillen et al. 2010; Poinssot and Geckeis 2015). This contributes to the spread of ^{90}Sr both within the water ecosystem most contaminated with radioactive substances, and its migration along the "waterways" at distances far from the source of contamination (Vasilenko and Vasilenko 2002; Akleev et al. 2006; Polikarpov et al. 2008; Gulin et al. 2013, 2016; Tereshchenko et al. 2014; Mirzoyeva et al. 2018a).

Several studies (Romanenko 2004; Chudinovskikh et al. 2004; Polikarpov et al. 2008; Konoplev et al. 2020; Sazykina et al. 2022) have shown that radioactive products, entering the aquatic environment in any way, are intensively diluted in the water column and then are included into the general cycle of substances in a basin. Then they migrate through water areas as a result of hydrophysical processes, are sorbed and concentrated by living and nonliving matter, and are repeatedly transferred along the trophic chains. Finally, their global biogeochemical cycle ends with their elimination into the bottom sediments, acting in this case as a long-term geological depot (Romanenko 2004; Chudinovskikh et al. 2004; Polikarpov et al. 2008; Bakhvalov et al. 2012; Tereshchenko et al. 2014; Mirzoyeva et al. 2018a; Konoplev et al. 2020).

In 1986, after the accident at the Chernobyl nuclear power plant (Chernobyl NPP), the maximum concentration of ^{90}Sr in surface waters of Crimea and the Black Sea caused by atmospheric precipitation was 98.1 $Bq{\bullet}m^{-3}$ (Polikarpov et al. 2008; Tereshchenko et al. 2014; Mirzoyeva et al. 2018a). During the ensuing years, from the end of 1986 until 2014, the water route of ^{90}Sr entry to the territory of Crimea with the Dnieper waters through the NCC dominated. The maximum concentration of ^{90}Sr in the Crimean surface waters caused by water transport was noted in 1987 and amounted to 991.6 $Bq{\bullet}m^{-3}$ (Polikarpov et al. 2008; Tereshchenko et al. 2014; Mirzoyeva et al. 2018a). Since 2014, the Dnieper water entry to the territory of the Crimean Peninsula through the system of the NCC had been blocked by a dam (Gulin et al. 2016). Water supply along the canal was resumed only in March 2022 (North Crimean Canal unblocked; water went to Crimea, 2022). Thus, for 8 years, such a significant factor for secondary radionuclide contamination of inland water bodies of Crimea as the inflow of the Dnieper water along the bed of the NCC was absent.

There are more than 50 salt lakes, both continental and marine types, on the Crimean Peninsula (Balushkina et al. 2009). The high salinity level of the water environment in these lakes is maintained due to the intensive evaporation of water in the summer. This factor determines the concentration of many chemical elements, including radioactive ones. The first determinations of the ^{90}Sr content in water of the Crimean hypersaline lakes, both after the global fallout and after the Chernobyl NPP accident, were carried out in 2013 on Lake Kiyatskoye of the Perekopsk group. The ^{90}Sr concentration in its water exceeded the Black Sea by 23.4 times (Polikarpov et al. 2008; Mirzoyeva et al. 2015).

Almost all salt lakes of Crimea are used for recreational and economic purposes. Salt, being mined in some of these lakes, is a valuable mineral resource and plays an important role in all areas of human activity (Lisovsky et al. 2004). The water supply along the NCC system sourced from the Dnieper has an impact on the radioecological and hydrochemical state of the inland water bodies of Crimea, including lakes with different salinity levels.

The relevance of this study is caused by the need to identify trends in the ^{90}Sr radioecological behavior in water bodies with different levels of salinity. Radioecological studies for lakes Adzhigol and Kuchuk-Adzhigol (located in the Kerch group of Crimean salt lakes) were carried out for the first time in the history of their existence in 2018. In addition, under the conditions of recommencement of the Dnieper water supply to Crimea after 8 years of its complete absence, the results obtained in this study are the starting point for the following assessment of the degree of the NCC system influence as one of the key factors (Gulin et al. 2016) for the change in the radioecological and hydrochemical state of internal water bodies of the region.

The aim of the study was a comparative analysis (in the period of 2016–2021) and identification of trends in ^{90}Sr concentration changes in the abiotic components of Crimean lakes located in Evpatoria and Kerch groups and differing within the group in terms of salinity and hydrochemical characteristics of their environments.

To achieve this goal, the following tasks were set and solved:

– Carrying out of the radiochemical treatment of the lakes' abiotic components samples followed by ^{90}Sr

activity measurement by its daughter ^{90}Y by using low-level beta-spectrometry;

– Determination of the ^{90}Sr concentration in water and bottom sediments of the lakes of Evpatoria (Sasyk-Sivash and Kyzyl-Yar) and Kerch (Adzhigol and Kuchuk-Adzhigol) geographical groups of Crimean salt lakes;
– Assessment of the hydrochemical and hydrophysical features of the studied objects;
– The comparative analysis of the possible effect of the aquatic environment salinity level on the ^{90}Sr distribution between the abiotic components of the studied water bodies;
– The final task was to reveal possible regularities in the influence of water salinity level on the change in the concentration and distribution of the ^{90}Sr artificial radionuclide in the ecosystems of the studied water objects.

2 Material and methods

2.1 Objects of the study

For research, we chose lakes that are close to each other spatially but differ in the level of salinity of their aquatic environments. The objects of study were lakes Sasyk-Sivash and Kyzyl-Yar, as well as Lake Adzhigol and Lake Kuchuk-Adzhigol, located in the Evpatoria and Kerch groups of Crimean salt lakes, respectively. A schematic map of the location of the lakes is shown in Fig. 1.

Lake Sasyk-Sivash is located between the cities of Evpatoria and Saki and thus belongs to the Evpatoria group of lakes. This reservoir is a part of the complex landscape reserve of regional significance. The greatest depth of the lake is 1.2 m, and the average is 0.5 m (Lisovsky et al. 2004). This lake is separated from the sea by an embankment, with the width varying from 0.9 to 1.7 km. Lake Sasyk-Sivash is located at a distance of 15 km from Lake Kyzyl-Yar and it is the largest lake in the Crimean Peninsula. In summer, the area of Lake Sasyk-Sivash decreases significantly, and the salinity of its water increases due to evaporation. This lake is a place of recreation, as its sulfide silt mud of the seaside type is classified as therapeutic (Lisovsky et al. 2004; Shadrin et al. 2018).

Lake Kyzyl-Yar is located on the western coast of Crimea, and it is a part of the Evpatoria group of salt lakes. The depth of Lake Kyzyl-Yar varies from 2.0 to 3.7 m. This lake is separated from the sea by a narrow sandy barrier (Lisovsky et al. 2004). As a result of the infiltration of the Dnieper water into Lake Kyzyl-Yar from the Mezhgornoye storage pond, which was filled with water from the NCC, the salinity of the lake decreased from 162 in 1985 to $2 \div 3$ g·L^{-1} in 2005, with its subsequent stabilization by this level. For 20 years, the lake has turned from hypersaline to brackish (Shadrin et al. 2018).

Lake Adzhigol is a salt lake of firth origin without a drain, and spatially it is a part of the Kerch group of lakes. The average depth of the lake is 0.35 m, and the greatest is 0.6 m. Lake Adzhigol is separated from the Black Sea by a narrow isthmus as well. Previously, the mud of this lake was classified as therapeutic, but at present, due to pollution by sewage, the lake mud resources have been excluded from this list.

The Kerch group of salt lakes also includes Lake Kuchuk-Adzhigol, which is separated from the Black Sea by a narrow isthmus. It is located at a distance of 800 m from Lake Adzhigol. The lake used to be healing, but as a result of mismanagement of human activity, this water body has lost its healing properties and status (Pasinkov et al. 2014).

Morphometric characteristics of the studied water objects are presented in Table 1 (Lisovsky et al. 2004).

Even though the lakes within the same group are located at a close distance from each other, their hydrochemical characteristics are different.

The salinity, acidity, and water temperature of the studied lakes were measured in-situ using refractometer Kelilong WZ212 and pH-meter PHH-830. The water characteristics obtained in the lakes during sampling for the study are presented in Table 2.

Thus, all the studied lakes have a similar origin, hydrological regime, and type of water supply. All studied water objects are characterized by drainless type of the hydrological regime, have a firth origin and a mixed type of water supply – with infiltrated sea waters, as well as with surface and ground waters of the Black Sea artesian basin (Lisovsky et al. 2004). At the same time, being located at short distances from each other, the studied water bodies of each geographical group are similar in ^{90}Sr input ways and intensities but differ in salinity of the aquatic environment and other hydrochemical characteristics.

2.2 Sampling

For ^{90}Sr water concentration measurement in studied lakes water samples of 20 L volume were taken into plastic canisters. Bottom sediments were sampled in 2016–2019 with a modernized corer with a tube of 210 mm inner diameter equipped with a vacuum valve (Woods Hall Oceanographic Institute, USA) (Polikarpov et al. 2008). Bottom sediment samples were extruded from the tube corer with a plunger of appropriate diameter and sediment was sliced into layers of 1 or 5 cm thickness. ^{90}Sr activity

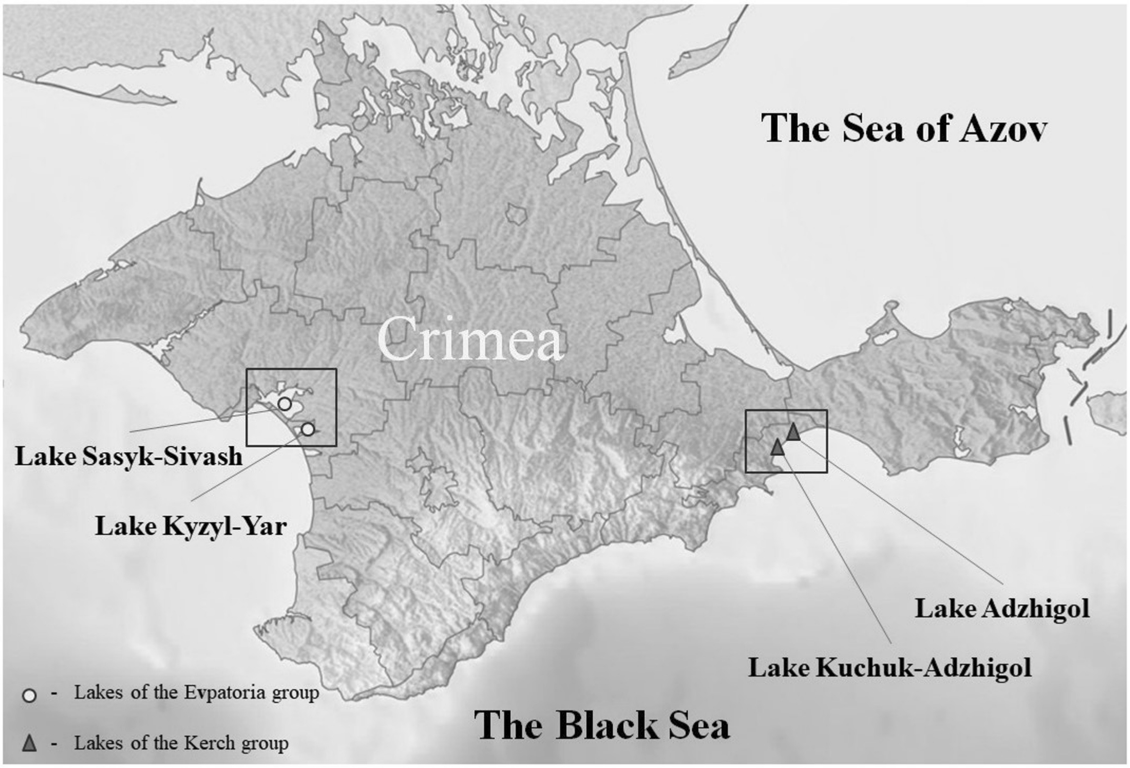

Fig. 1 Schematic map of the studied water objects location

Table 1 Morphometric characteristics of the salt lakes of Crimea

Lake	Area (km^2)	Length (km)	Average width (km)	Catchment area (km^2)	Average rainfall (mm)
Sasyk-Sivash	75.3	14	5.5	1064	400
Kyzyl-Yar	8	5.7	1.4	328	400
Adzhigol	0.5	1.2	0.6	2.5	400–450
Kuchuk-Adzhigol	0.3	0.9	0.8	2.5	400–450

concentration was averaged for surface 0–5 cm sediment layer in all studied lakes.

The sampled material was stored for no longer than 8–10 h, followed by radiochemical treatment. Complete radiochemical processing of samples and measurement of ^{90}Sr concentration in water and bottom sediments of the studied lakes were carried out in a laboratory.

2.3 Samples chemical treatment and measurement

Various ways are known for extracting and concentrating of ^{90}Sr from environmental samples (Grahek et al. 2018). Both ion exchange resins (Wang et al. 2020) and effective adsorbents are used to remove Sr^{2+} from contaminated waters (Zhang et al. 2022). In the actual study radiochemical analysis was used ^{90}Sr isolation from natural objects (Polikarpov et al. 2008; Harvey et al. 1989).

Radiochemical analysis in general included the following steps: pre-treatment of sampled material, the unified radiochemical treatment of samples with counting sources production, determination of chemical yield using stable strontium, and measurements of β-activity of ^{90}Y in counting sources. The flow chart of every step is described below.

Pre-treatment of water samples. Water samples were separated from suspended matter by filtration. In the filtered water sample, ^{90}Sr was concentrated by co-precipitation with mixed strontium ($SrCO_3$) and calcium ($CaCO_3$) carbonates. Into the water sample, acidified with 1 mL of concentrated (37%) hydrochloric acid (HCl) per each 1 L of the sampled material, the solution of stable carriers –

Table 2 Sampling metadata and water characteristics in the studied lakes in 2016–2021

Lake	Sampling location	Sampling date (dd.mm.yyyy)	Salinity (‰)	pH	t (°C)
Evpatoria group					
Kyzyl-Yar	N 45°03.983’ E 33°37.781’	18.05.2016*	3.5 ± 0.2	7.9 ± 0.1	22.1 ± 0.4
	N 45°03.982’ E 33°37.782’	28.07.2017*	3.7 ± 0.2	7.8 ± 0.1	29.8 ± 0.6
	N 45°03.059’ E 33°35.063’	25.07.2018	7.0 ± 0.2	6.0 ± 0.1	27.6 ± 0.5
	N 45°03.057’ E 33°35.070’	13.06.2019	6.0 ± 0.1	9.1 ± 0.1	25.0 ± 0.5
	N 45°03,412’ E 33°37,815’	26.06.2020	8.0 ± 0.2	7.7 ± 0.1	27.5 ± 0.5
	N 45°03.445’ E 33°38.667’	09.04.2021	8.0 ± 0.2	7.9 ± 0.1	12.3 ± 0.2
Sasyk-Sivash	N 45°09.251’ E 33°30.745’	27.06.2016*	280 ± 0.2	7.7 ± 0.1	31.8 ± 0.6
	N 45°09.251’ E 33°30.745’	08.11.2016*	322 ± 0.2	7.8 ± 0.1	6.0 ± 0.1
	N 45°09.033’ E 33°30.680’	28.07.2017*	295 ± 0.2	7.8 ± 0.1	29.8 ± 0.6
	N 45°09.375’ E 33°30.428’	25.07.2018	280 ± 0.2	6.2 ± 0.1	34.5 ± 0.7
	N 45°09.247’ E 33°30.755’	13.06.2019	260 ± 0.2	7.7 ± 0.1	28.0 ± 0.5
	N 45°09.274’ E 33°30.695’	17.06.2020	350 ± 0.5	7.4 ± 0.1	30.8 ± 0.6
	N 45°09.235’ E 33°30.753’	09.04.2021	310 ± 0.5	7.3 ± 0.1	14.0 ± 0.1
Kerch group					
Adzhigol	N 45°06.544’ E 35°28.153’	26.04.2018	150 ± 0.2	7 ± 0.1	25.4 ± 0.5
	N 45°06.546’ E 35°28.152’	05.11.2019	The lake dried up, there was no water		
	N 45°06.318’ E 35°27.596’	01.07.2021	50 ± 0.2	8.7 ± 0.1	32.1 ± 0.5
Kuchuk-Adzhigol	N 45°06.100’ E 35°27.182’	26.04.2018	5 ± 0.2	8.5 ± 0.1	22.1 ± 0.4
	N 45°06.200’ E 35°26.546’	05.11.2019	1.5 ± 0.1	8.4 ± 0.1	17.0 ± 0.2
	N 45°06.166’ E 35°26.515’	01.07.2021	3.0 ± 0.1	9.5 ± 0.1	31.0 ± 0.5

*Data for 2016 and 2017 are taken from work (Mirzoyeva et al. 2018b)

Sr^{2+} and Fe^{3+} – was added. After 2 h of stirring NH_4Cl was added to keep Mg in the solution. After 3–4 series of intensive stirring the precipitant was added – 200 mL of 1.5 M Na_2CO_3. After that, samples were intensively stirred and settled overnight in plastic containers, and then the supernatant was decanted and discarded. The precipitate was subjected to complete dissolution with 6 N HCl, after which the resulting solution was processed according to a unified radiochemical technique for sample analysis described below.

Pre-treatment of bottom sediment samples. Bottom sediments samples were в weighted, dried in porcelain dishes at 100–110 °C to a constant weight, and then about 200 g of material was subsampled and ashed in a muffle furnace at 450–500 °C. 400 mg of Sr^{2+} and up to 600 mL of concentrated HCl were added to the ashed sample and the material was subjected to at least 30 min boiling on a hot plate. The solution was filtered through a glass fiber filter. The insoluble residue was twice resuspended in 200 mL of distilled water and both suspensions were filtered. The combined filtrate was subjected to a unified radiochemical technique.

The unified radiochemical technique for ^{90}Sr measurements is based on the procedure of subsequent purification from interfering elements, followed by settlement for at least 14 days for equilibrium established between ^{90}Sr and its daughter ^{90}Y. After the equilibrium reached ^{90}Y content (equal to ^{90}Sr one) was measured in samples by Cherenkov radiation using an ultra-low-background liquid scintillation counter (LSC) «Quantulus 1220» (LKB Wallac). The lower limit of detection for ^{90}Sr in samples was $0.01 \div 0.04$ Bq·m^{-3} (Bq·kg^{-1}). *^{90}Sr yield* was calculated from the yield of stable Sr measured using SPECTRUM-5 atomic absorption (AA) spectrometer and for ^{90}Y it was obtained by gravimetric analysis from yttrium oxalate (Polikarpov et al. 2008; Harvey et al. 1989). Each result is presented as the average of the activity values obtained from parallel duplicate samples that were measured separately. ^{90}Sr concentration values for sediments are expressed as Bq·kg^{-1} of dry sediment weight (d.w.). The total relative uncertainty of each result did not exceed 20%. The quality of the analytical techniques used, and the reliability of the results obtained were supported by constant participation in international intercalibration measurements during 1990–2004 under the auspices of the IAEA (Vienna, Austria) and by parallel measurement in Risø National Laboratory (Risø, Denmark) in 1993–2001. The results of IBSS participation in intercalibration were included in the intercalibration report documents (IAEA 1998, 2004) and were certified as reliable data.

A total of 16 water samples and 34 sediment samples were taken and analyzed.

2.4 Calculations

Coefficient of ^{90}Sr distribution for bottom sediments (K_d) was determined by the ratio:

$$K_d = \frac{A_s}{A_w} \quad (1)$$

whereA_s –^{90}Sr activity concentration in the bottom sediments, Bq·kg^{-1} (d.w.);

A_w –^{90}Sr activity concentration in lake water taking into account the saltwater density,

Bq·kg^{-1}.

The trend of ^{90}Sr concentration change in lake water was described by an exponential function as:

$C = C_0 \cdot e^{-pt}$, (2). Where C –^{90}Sr concentration at time t, Bq·m^{-3};

C_0 –^{90}Sr concentration at the initial moment of the period considered, Bq·m^{-3};

p – the time constant.

The time constant (p) of this function with a sufficient degree of reliability is an indicator of the half-period of the determined radionuclide concentration in water (Mirzoyeva et al. 2013). Extrapolation of the approximating curve for function (2) allows making predictive estimates of the time period for radionuclide concentrations in water to be decreased down to pre-accident levels (Polikarpov et al. 2008; Mirzoyeva et al. 2013).

3 Results

The results of the ^{90}Sr concentration measurements in the water of the studied Crimean salt lakes are presented in Table 3.

For comparative analysis, ^{90}Sr concentrations were also determined in the Black Sea waters, sampled in the areas adjacent to the studied lakes. The research results obtained from 2016 to 2017 are given from the work of (Mirzoyeva et al. 2018b).

It was noted (Table 3) that among the lakes of the Evpatoria and Kerch groups, the highest average ^{90}Sr concentrations in water were obtained in water bodies with the highest salinity: in Lake Sasyk-Sivash 97.7 ± 7.9 Bq·m^{-3} (salinity 299.6 ‰) and in Lake Adzhigol – 61.0 ± 3.8 Bq·m^{-3} (100 ‰). The average concentrations of this radionuclide in the water of lakes Kyzyl-Yar (6.6 ± 1.0 Bq·m^{-3}, salinity 5.6 ‰) and Kuchuk-Adzhigol (29.1 ± 2.2 Bq·m^{-3}, 3.2 ‰) were 15.8 and 2.1 times lower than those for lakes with higher water salinity in the Evpatoria and Kerch groups respectively.

The averaged activity concentration of ^{90}Sr in the surface layer (0–5 cm) of bottom sediments of the studied water objects is presented in Table 4.

The highest ^{90}Sr concentrations were noted in the bottom sediments of lakes Sasyk-Sivash (25.4 ± 3.2 Bq·kg^{-1} d.w.) with a salinity of 295 ‰ and Adzhigol (18.8 ± 1.9 Bq·kg^{-1} d.w.) with a salinity of 150 ‰. The concentrations of ^{90}Sr in the bottom sediments of the adjacent coastal regions of the Black Sea ranged from 1.8 to 3.4 Bq·kg^{-1} (d.w.) and were of the same level as in the sediments of the brackish water lakes Kyzyl-Yar and Kuchuk-Adzhigol (Table 4).

Table 3 ^{90}Sr concentration in water of the salt lakes of Crimea and adjacent Black Sea areas in 2016–2021

Water area	Sampling date	Salinity (‰)	^{90}Sr ($Bq \cdot m^{-3}$)
Evpatoria group			
Lake Kyzyl-Yar	18.05.2016	3.5 ± 0.2	6.0 ± 0.8
	06.09.2016	3.1 ± 0.2	9.3 ± 1
	28.07.2017	3.7 ± 0.2	5.9 ± 0.8
	25.07.2018	7.0 ± 0.2	10.1 ± 1
	13.06.2019	6.0 ± 0.1	2.9 ± 0.7
	26.06.2020	8.0 ± 0.2	4.3 ± 1.8
	9.04.2021	8.0 ± 0.2	7.8 ± 1.1
	Average	5.6 ± 0.2	6.6 ± 1.0
Lake Sasyk-Sivash	27.06.2016	280 ± 0.2	313.6 ± 25
	08.11.2016	322 ± 0.2	258.9 ± 15
	28.07.2017	295 ± 0.2	49.4 ± 9
	25.07.2018	280 ± 0.2	26.2 ± 0.5
	13.06.2019	260 ± 0.2	17.8 ± 3.3
	17.06.2020	350 ± 0.5	b.d.l.*
	09.04.2021	310 ± 0.5	17.71 ± 2.6
	Average	299.6 ± 0.3	97.7 ± 7.9
Kerch group			
Lake Adzhigol	26.04.2018	150 ± 0.2	107.3 ± 6.0
	05.11.2019	The lake dried up, there was no water	
	01.07.2021	50 ± 0.2	14.6 ± 1.5
	Average	100 ± 0.2	61.0 ± 3.8
Lake Kuchuk-Adzhigol	26.04.2018	5 ± 0.2	33.1 ± 2.0
	05.11.2019	1.5 ± 0.1	39.7 ± 3.0
	01.07.2021	3.0 ± 0.1	14.6 ± 1.5
	Average	3.2 ± 0.2	29.1 ± 2.2
Adjacent Black Sea areas			
Evpatoria Bay	09.06.2016	18.3 ± 0.2	13.3 ± 1.4
	09.07.2017	18.2 ± 0.2	8.6 ± 1.0
	31.08.2018	18.3 ± 0.2	17.8 ± 1.6
	18.04.2019	18.1 ± 0.2	9.8 ± 1.0
	27.06.2020	18.2 ± 0.2	9.1 ± 1.4
	13.07.2021	18.3 ± 0.2	11.6 ± 1.7
	Average	18.2 ± 0.2	11.7 ± 1.4
Feodosia Gulf	30.10.2016	18.3 ± 0.2	12.0 ± 1.7
	18.07.2017	18.2 ± 0.2	6.4 ± 1.0
	12.09.2018	18.3 ± 0.2	12.4 ± 1.5
	02.05.2019	18.4 ± 0.2	14.3 ± 1.3
	11.06.2020	18.3 ± 0.2	7.1 ± 1.2
	07.08.2021	18.2 ± 0.1	9.3 ± 1.7
	Average	18.3 ± 0.2	10.3 ± 1.5

*b.d.l. – below the detection limit

The distribution coefficient (K_d) is a quantitative criterion characterizing the processes of ^{90}Sr transition into the bottom sediments of lakes. Calculated K_d of ^{90}Sr for bottom sediments of the studied lakes varied in the following ranges: $n \cdot 10^0 \div n \cdot 10^2$ for hypersaline lakes, $n \cdot 10^{1-} \div n \cdot 10^2$ (brackish lakes), $n \cdot 10^2$ (the Black Sea). The upper ranges of coefficients of ^{90}Sr distribution between water and the bottom sediments of the studied lakes are similar to ones obtained for the Black Sea. This indicates similar mechanisms of ^{90}Sr elimination from the water column to bottom sediments of the studied water bodies, regardless of the characteristics of the last ones. The lowest

Table 4 Activity concentration of ^{90}Sr and coefficients of its distribution (K_d) for the surface layers (0–5 cm) of the bottom sediments from the Crimean salt lakes and adjacent water areas of the Black Sea in 2016–2019

Water object	Sampling date	Salinity (‰)	^{90}Sr in bottom sediments (Bq kg^{-1} d.w.)	K_d
Sasyk-Sivash	27.06.2016	280 ± 0.2	2.0 ± 0.2	10^0
	28.07.2017	295 ± 0.2	25.4 ± 3.2	10^2
	25.07.2018	280 ± 0.2	19.2 ± 1	10^2
	13.06.2019	260 ± 0.2	7.7 ± 0.9	10^2
Kyzyl-Yar	18.05.2016	3.5 ± 0.2	3.3 ± 1	10^2
	28.07.2017	3.7 ± 0.2	3.1 ± 1	10^2
	25.07.2018	7.0 ± 0.2	2.9 ± 1	10^2
	13.06.2019	6.0 ± 0.1	b.d.l.	–
Adzhigol	26.04.2018	150 ± 0.2	18.8 ± 1.9	10^2
Kuchuk-Adzhigol	26.04.2018	5 ± 0.2	2.6 ± 0.3	10^1
	05.11.2019	1.5 ± 0.1	14.3 ± 1.8	10^2
the Black Sea	28.01.2016	18.4 ± 0.2	1.8 ± 0.2	10^2
	22.04.2016	18.4 ± 0.2	3.4 ± 0.3	10^2
	02.05.2019	18.1 ± 0.2	1.8 ± 0.2	10^2

b.d.l. Below the detection limit

K_d was observed for the bottom sediments of the hypersaline lake Sasyk-Sivash. So, the variability of K_d values must be associated with the features of geochemical factors inherent in each water body.

4 Discussion

4.1 ^{90}Sr sources in the studied water bodies

It is known (Mirzoyeva et al. 2015, 2018b; Shadrin et al. 2018) that the Kyzyl-Yar and Kuchuk-Adzhigol lakes had both atmospheric and long-term water ^{90}Sr input ways into their ecosystems. The extensive freshwater input is additionally evidenced by the low salinity levels of the aquatic environment of their ecosystems: 3.1 ÷ 8 ‰ and 1.5 ÷ 5 ‰ for Lake Kyzyl-Yar and Lake Kuchuk-Adzhigol, respectively. It was revealed (Shadrin et al. 2018) that until 2014, fresh waters of the Dnieper river leaked into Lake Kyzyl-Yar from the Mezhgornoye Reservoir, where they entered through the NCC.

As for Lake Kuchuk-Adzhigol, along with sea and ground waters supplied its water balance, sewage was continuously discharged here, causing the additional freshening and pollution of its ecosystem (Pasinkov et al. 2014).

The technique, used to identify sources of ^{90}Sr input into Lake Sasyk-Sivash, implied a comparison of the average concentrations of this radionuclide in lake water with the maximum values of ^{90}Sr concentrations caused by atmospheric and water transport of radioactivity after the Chernobyl NPP accident (Mirzoyeva et al. 2018b). It was determined that ^{90}Sr was introduced into Lake Sasyk-Sivash both by atmospheric and water routes (Mirzoyeva et al. 2018b. The same approach (Mirzoyeva et al. 2018b) was also applied to identify sources of ^{90}Sr in Lake Adzhigol (Fig. 2). It was obtained (Fig. 2) that the concentration level of ^{90}Sr in Lake Adzhigol, as well as in Lake Sasyk-Sivash, was in the range of radionuclide concentrations corresponding to the atmospheric and waterways of its entry.

Thus, for all the studied lakes, two sources of the ^{90}Sr input were determined: the primary source was concerned with atmospheric fallout immediately after the Chernobyl NPP accident and the secondary long-term source was concerned with waterway entry of this radionuclide. The water route of the radionuclide entry on the one hand was concerned with directly fresh the Dnieper waters input through the NCC system (for Kyzyl-Yar and Kuchuk-Adzhigol lakes), and on the other hand with seepage of polluted waters of the Black Sea into the lakes (for Sasyk-Sivash and Adzhigol lakes).

4.2 ^{90}Sr in the water of lakes with different salinity of the water environment

The ranking of the average ^{90}Sr concentrations in water of the lakes in 2016–2021 is shown in Fig. 3.

It was determined (Table 3, Fig. 3) that in hypersaline lakes Sasyk-Sivash (299.6 ‰) and Adzhigol (100 ‰), with an increase in the salinity of the environment by 2.9 times, the average ^{90}Sr concentration in water increased 1.6 times. In the lakes with brackish water Kyzyl-Yar (5.6 ‰) and Kuchuk-Adzhigol (3.2 ‰), no dependence of the radionuclide concentration on the water salinity was observed. In the lake Kuchuk-Adzhigol, Adzhigol, and

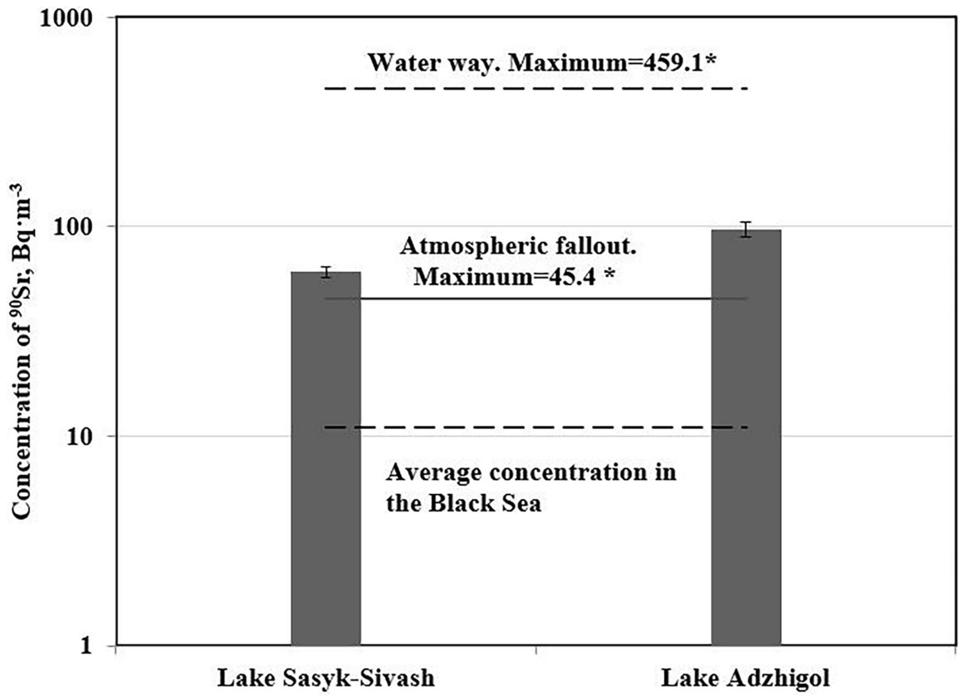

Fig. 2 Comparison of the average ^{90}Sr concentrations in waters of Lake Sasyk-Sivash and Lake Adzhigol with its maximum levels caused by atmospheric and water routes of the radionuclide entry (* –^{90}Sr concentrations are decay-corrected to 2016)

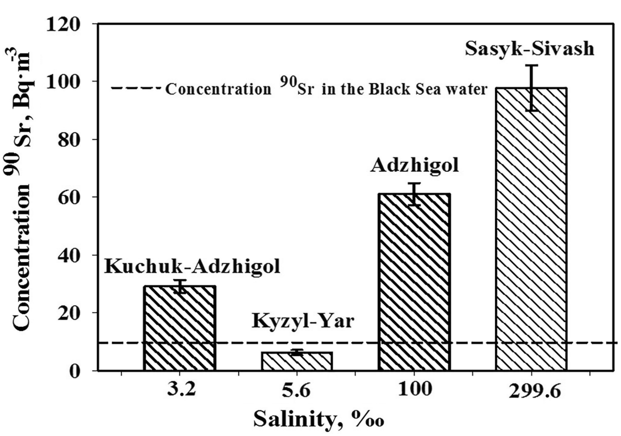

Fig. 3 Distribution of ^{90}Sr concentration in water of the studied Crimean salt lakes in 2016–2021 (presented data are obtained in the actual study and taken from Mirzoyeva et al. 2018b, Mirzoeva et al. 2020)

Sasyk-Sivash, the concentration of ^{90}Sr in water were 2.8, 5.9, and 8.4 times higher, respectively, as compared to the adjacent Black Sea areas. At the same time, the values of water salinity in these lakes were either 5.7 times lower (Lake Adzhigol), or 5.5 times higher (Lake Kuchuk-Adzhigol) and 17.1 times higher (Lake Sasyk-Sivash) than the Black Sea salinity. As it was noted earlier, the level of artificial radionuclides concentration in water depends, first of all, on the source of entry, the frequency and amounts of radionuclides entries to the water objects (Polikarpov et al. 2008).

Since 2014, after the closure of the North Crimean Canal, the input of ^{90}Sr with the Dnieper water into the inland waters of Crimea has almost ceased (Gulin et al. 2016; Mirzoyeva et al. 2018b). At the same time, about 61% of the total variability of ^{90}Sr content in lake water can be explained by salinity changes (Mirzoeva et al. 2020). This value shows a significant influence of such a hydrochemical parameter as salinity on the behaviour of artificial radionuclides, in particular ^{90}Sr, in ecosystems of the studied lakes and on the process of their redistribution over the ecosystem components. In other words, an increase in the salinity of the lake environment contributes to the presence of ^{90}Sr in the aquatic environment in dissolved form.

Lake Sasyk-Sivash was considered a model object for describing the regularity of ^{90}Sr concentration changes in the water of the Crimean hypersaline lakes after the closure of NCC. Figure 4 shows that ^{90}Sr concentration has been decreasing linearly since 2017, regardless of the salinity level of the environment. The minimum ^{90}Sr value is 17.7 times less than the maximum one, while the salinity changed only by 1.3 times. This indicates only a slight change in the salinity of the lake water over the entire period of observation. We believe that such a sharp decrease in ^{90}Sr concentration in the water of Lake Sasyk-Sivash was an ecosystem response to a reduction in the secondary input of this radionuclide by water route, associated with the closure of the NCC (Gulin et al. 2016; Mirzoyeva et al. 2018b). The decrease in ^{90}Sr concentration in the lake water to levels "below the detection limit"

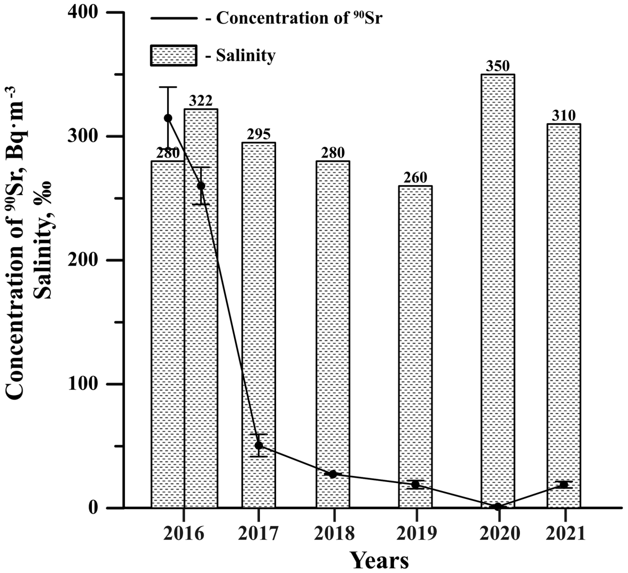

Fig. 4 Dynamics of ^{90}Sr concentration (line) and salinity (columns) in water of Lake Sasyk-Sivash in 2016–2021

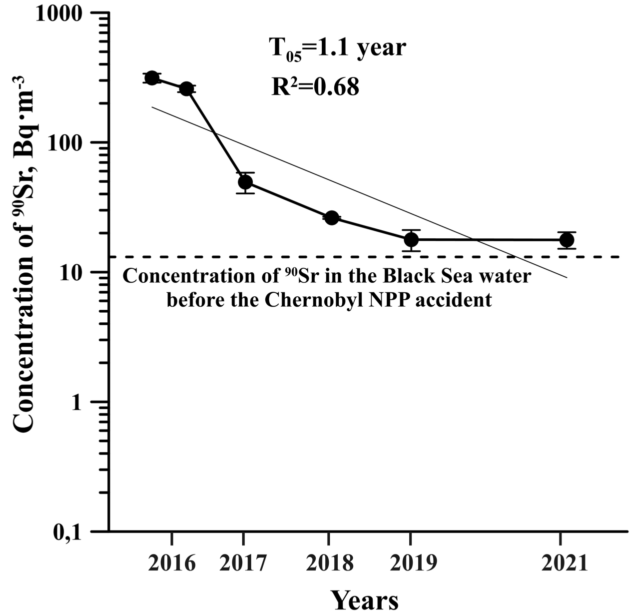

Fig. 5 Determination of the period of decreasing in twice the ^{90}Sr concentration in water of Lake Sasyk-Sivash (2016–2021)

observed in 2020 was highly likely due to the precipitation of radionuclides in the total mass of salts, crystallization of which occurs in hypersaline environments at a salinity of 340 ‰ and higher.

Considering the dynamics of ^{90}Sr concentration in water of Lake Sasyk-Sivash using a logarithmic ordinate scale, it was noted that this dynamic is satisfactorily ($R^2 = 0.75$) described by an exponential function. The parameters of this function can be used to determine the half-period of the ^{90}Sr concentration in the lake water (Fig. 5) (Mirzoyeva et al. 2013).

Since the beginning of radioecological studies in Lake Sasyk-Sivash, the calculated period of decreasing in twice the ^{90}Sr concentration in the water of the lake (Fig. 5) varies in the range of 0.8 years [for the period when the ^{90}Sr concentration was below the detection level (b.d.l.)] and 1.1 years (excluding the period when the ^{90}Sr concentration was b. d. l.).

The obtained values allow us to conclude that biogeochemical processes in Lake Sasyk-Sivash, in the absence of additional ^{90}Sr input, reduce its residence time in the aquatic environment by 131 times, as compared with its elimination only due to radioactive decay. Under the conditions of the NCC being closed, the decrease of ^{90}Sr concentration in the water of Lake Sasyk-Sivash to the pre-accident levels (typical for the Black Sea) occurred already in the middle of 2019.

Speaking about the features of ^{90}Sr behavior in brackish water bodies, Lake Kyzyl-Yar was considered as a model object. The closure of the NCC in 2014 was reflected in the increase of the salinity level in the lake water as a linear function of time ($R^2 = 0.87$), Fig. 6. At the same time, throughout the entire period of the study, ^{90}Sr concentration was on average 1.8 times lower and salinity was 3 times of the values typical for the adjacent waters of the Black Sea (Fig. 6).

There was no long-term regularity in the dynamics of ^{90}Sr concentration in Lake Kyzyl-Yar water revealed for the entire period of the study. However, some features were noted for the change of this radionuclide concentration in the lake water during shorter periods in 2017–2018 and

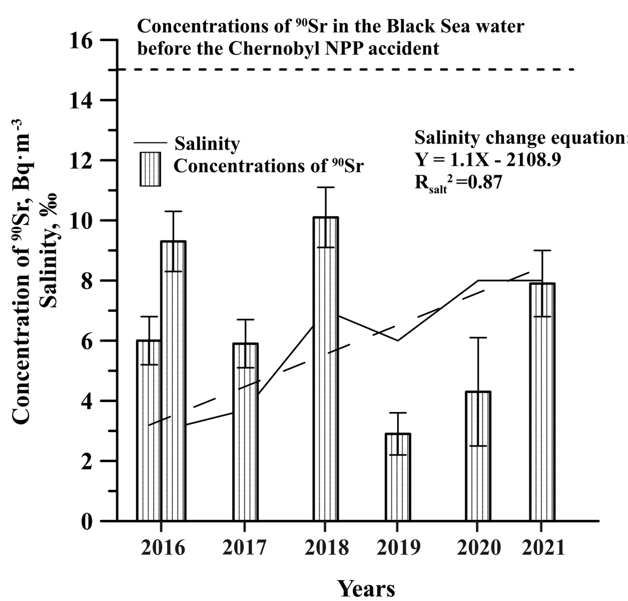

Fig. 6 Dynamics of ^{90}Sr concentration (bars) and salinity (line) in water of Lake Kyzyl-Yar in 2016–2021

2019–2020 (Table 3, Fig. 6). Calculated coefficients of correlation between ^{90}Sr concentrations in lake water and its salinity and pH were 0.007 and 0.82 respectively. This shows the absence of a relationship between ^{90}Sr concentration dynamics in water and changes in the salinity of the environment but reveals a significant negative dependence of ^{90}Sr content on the water pH of this brackish lake (Fig. 7). It was found that the highest concentration of ^{90}Sr in the water of the lake over the entire period of the study was observed under the increase of its acidity (decrease of pH down to 6.0). And in contrast under the increase of water alkalinity (pH $>$ 9.0), the radionuclide concentration decreased by 3.5 times. We believe, that with an increase in water acidity, the process of ^{90}Sr remobilization from bottom sediments into the aquatic environment takes place in brackish waters, and in a more alkaline environment, the radionuclide is removed from the aquatic environment (Tables 2 and 3, Fig. 7).

For the brackish waters of Lake Kuchuk-Adzhigol, a similar dependence of the ^{90}Sr concentration dynamics on the water pH was noted. Intensification of sedimentation processes under the lake water alkalinity increasing up to pH = 9.5 resulted in decreasing of ^{90}Sr concentration in its waters down to the order of values typical for adjacent areas of the Black Sea in 2021. At the same time, as was noted earlier (Mirzoyeva et al. 2018b), there was no dependence of ^{90}Sr concentration dynamics on pH for hypersaline lakes.

Thus, the closure of the NCC led to the fact that ^{90}Sr distribution in lake ecosystems was mainly influenced by geochemical processes within the basins themselves, which ensured both the elimination of ^{90}Sr into the bottom sediments of lakes and the occasional remobilization of this radionuclide from bottom sediments into the aquatic environment. For brackish water lakes, a significant factor influencing the processes of the radionuclide redistribution was the pH of the water environment, while for hypersaline lakes, such a factor was the level of water salinity in these water reservoirs.

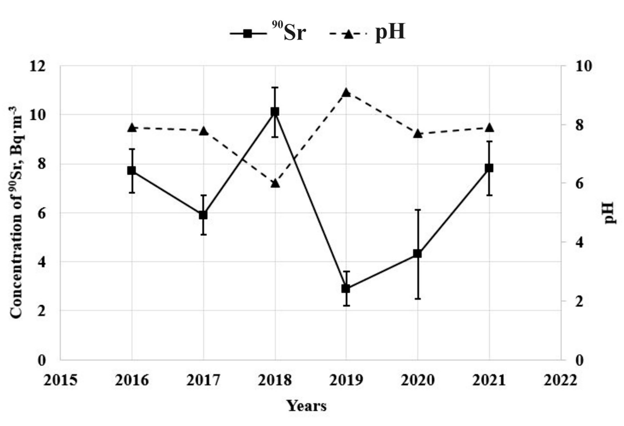

Fig. 7 Dynamics of ^{90}Sr concentrations and pH of water in Lake Kyzyl-Yar in 2016–2021

4.3 ^{90}Sr in bottom sediments of the lakes

The ranking of the average ^{90}Sr concentrations in the bottom sediments of Crimean salt lakes in 2016–2019 is shown in Fig. 8.

Data presented in Table 4 and Fig. 8 shows the activity concentrations of ^{90}Sr in the surface (0–5 cm) sediments to be higher in lakes with higher salinity than those in brackish water bodies. In all lakes, except Kyzyl-Yar, the concentration of ^{90}Sr in their bottom sediments exceeds that value obtained for the Black Sea. This is likely to be due to the different content of ^{90}Sr in the water of lakes, as well as due to the different rates of sediment mass accumulation and sedimentation processes in water columns of different lakes. This issue remains to be carefully studied.

A high positive correlation was determined between ^{90}Sr concentrations in the bottom sediments of brackish water lakes (Kyzyl-Yar and Kuchuk-Adzhigol) and the concentrations of this radionuclide in water (r = 0.7), as well as a negative correlation between these values and salinity of the water environment (r = -0.8). A moderate negative correlation (r = -0.5) is observed between the concentrations of ^{90}Sr in water and bottom sediments in hypersaline lakes (Sasyk-Sivash and Adzhigol). A poor correlation or absence was noted for the concentration of ^{90}Sr in the bottom sediments of all lakes against the pH of the lake's water.

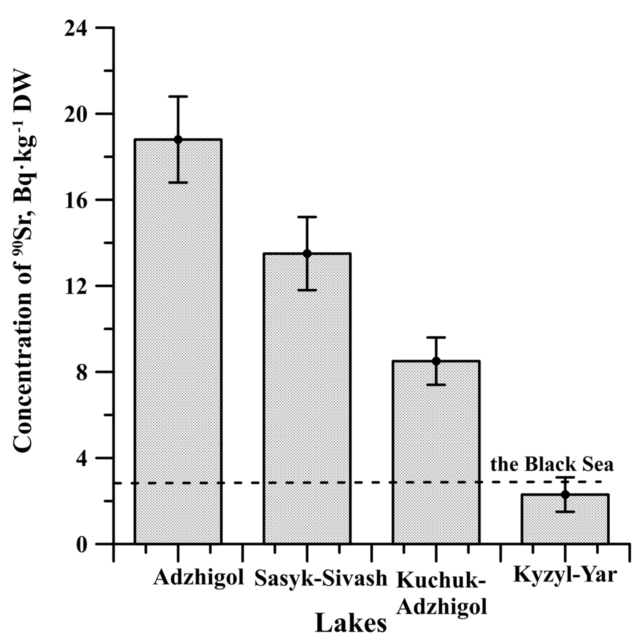

Fig. 8 Average ^{90}Sr activity concentration in bottom sediments of Crimean salt lakes in Evpatoria and Kerch groups in 2016–2019

Thus, the bottom sediments content of ^{90}Sr in the Crimean salt lakes is affected, first of all, by the concentration of this radionuclide in the water of the basins, as well as by the salinity of their environment.

5 Conclusion

From 2016 to 2021, a comparative study of the features of ^{90}Sr distribution in the abiotic components of Crimean salt lakes with different salinity and hydrochemical characteristics of their environment and located in the Evpatoria and Kerch groups has been carried out. Radioecological studies of the content and distribution of ^{90}Sr in the abiotic components of two Crimean salt lakes of the Kerch group Adzhigol and Kuchuk-Adzhigol were carried out for the first time.

Two sources of ^{90}Sr entry were identified for all the studied lakes: the primary one with atmospheric fallout immediately after the Chernobyl accident and the secondary – long-term input of this radionuclide by waterway during further years. The waterway of the radionuclide entry into the lakes was concerned with the Dnieper waters being directly brought in through the NCC (for Kyzyl-Yar and Kuchuk-Adzhigol lakes), as well as with seepage of polluted waters of the Black Sea into the lakes (Sasyk-Sivash and Adzhigol).

Using Lake Sasyk-Sivash as an example, the period of decrease in twice the ^{90}Sr concentration in water of the hypersaline lake after the closure of the North Crimean Canal was calculated. It was found that this period was in a range of 0.8–1.1 years for the whole time of radioecological monitoring from its very beginning. That is, in the absence of a ^{90}Sr input source, biogeochemical processes in the lake reduce the residence time of this radionuclide in its aquatic environment by 131 times. The decrease in ^{90}Sr concentration in the water of Lake Sasyk-Sivash to pre-accident levels, being typical for the adjacent Black Sea, occurred already in 2019.

For brackish water reservoirs, a significant factor influencing the processes of radionuclide retention in water was the pH of the medium; for hypersaline lakes, such a factor was the high level of their water salinity. At the same time, the correlation between ^{90}Sr concentration in the bottom sediments of all lakes and the pH of their medium was very poor or absent.

For brackish water lakes, a high positive correlation ($r = 0.7$) was noted between ^{90}Sr sediment and water concentrations, as well as a high negative correlation ($r = -0.8$) of ^{90}Sr content in sediments and salinity of the lake environment was found. The correlation between ^{90}Sr sediment and water concentrations for hypersaline lakes (Sasyk-Sivash and Adzhigol) was not as high ($r = -0.5$) and can be considered moderately negative. So, the decisive factors determining the ^{90}Sr content in bottom sediments of the salt lakes are the concentration of the radionuclide in lakes water and the salinity of their water environment.

The calculated distribution coefficients (K_d) of ^{90}Sr for the bottom sediments of the studied lakes varied in the following ranges: $n \cdot 10^0 \div n \cdot 10^2$ (hypersaline lakes), $n \cdot 10^1 \div n \cdot 10^2$ (brackish water lakes). The upper ranges of ^{90}Sr K_d between the water and bottom sediments of the studied lakes are similar to such values typical for the bottom sediments of the Black Sea. This, in turn, indicates similar mechanisms of ^{90}Sr elimination from water to bottom sediments of the studied water bodies, regardless of the characteristics of their environment. The lowest K_d value was observed for the bottom sediments of the hypersaline lake Sasyk-Sivash. The variability of K_d values is obviously associated with the features of geochemical factors inherent in each water body.

It was shown that the closure of the NCC led to the fact that ^{90}Sr distribution in lake ecosystems was mainly influenced by geochemical processes within the basins themselves. These processes ensured either the elimination of ^{90}Sr into the bottom sediments of lakes or the occasional remobilization of this radionuclide from bottom sediments back into the aquatic environment.

The results obtained in this work are of particular importance as a starting point and a basis for further radioecological studies of the inland waters of the Crimean Peninsula after the NCC opening and the Dnieper water anew inflow to the territory of Crimea in 2022 after 8 years of its absence.

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Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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