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Does Salinity Affect the Distribution of the Artificial Radionuclides ^{90}Sr and ^{137}Cs in Water of the Saline Lakes? A Case of the Crimean Peninsula

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Abstract: In the framework of radioecological monitoring, the features of the distribution of the post-Chernobyl nuclear power plant (NPP) accident artificial radionuclides of ^{137}Cs and ^{90}Sr in the Crimean saline lakes were studied. Samples were collected from 12 Crimean lakes with a salinity range from 2 to 400 g/L in 2014–2017. Concentration of ^{90}Sr varied from 5.9 to 313.6 Bq/m^3, and ^{137}Cs from 0.8 to 106.7 Bq/m^3. Closing of the North Crimean Canal resulted in a decrease in radionuclide concentration in the lakes. About 61% of the total variability of ^{90}Sr and 33% of ^{137}Cs in lake water can be explained by salinity changes. The salinity affects the behavior of radionuclides in water, mainly influencing their solubility, on isotope exchange between the solution and bottom sediments, and also, probably, through its influence on accumulation of isotopes by aquatic organisms. Salinity is not the alone factor determining the behavior of radionuclides in the lakes, it only acts by interacting with other factors.

Keywords: radioactivity; salinity; ^{90}Sr; ^{137}Cs; Crimea; artificial radionuclides

1. Introduction

The emission of artificial radionuclides into the environment can happen through open tests of nuclear weapons, authorized discharges from nuclear reprocessing plants, and accidents at enterprises using nuclear energy [1,2]. In some developed countries like the United Kingdom, France, United States, and Australia as well as in fast developing countries like India, China, nuclear power stations became an important component of the national energy policy; a “nuclear renaissance” is underway [3]. The accident on the Chernobyl nuclear power plant (the Chernobyl NPP) in 1986 was the largest one, and the accident on the Fukushima-1 nuclear power plant was one of the last of such cases [4]. The catastrophic accidents in Chernobyl and Fukushima have demonstrated that radioactive releases had far-reaching consequences for public risks [5]. Continued use of nuclear energy as a carbon-free, efficient, and reliable source of energy in combination with potential and real nuclear accidents make it necessary to identify the patterns of radionuclide behavior in natural objects, depending on various environmental factors [5–7].

The most dangerous long-lived artificial radionuclides that enter into the environment and determine the radioactive contamination of the biosphere are ^{137}Cs and ^{90}Sr [1,5,8,9]. Radioactive contamination of aquatic ecosystems located both near the site of the explosion and far away from it is associated with the release into the atmosphere and wind transport of radioactive products and aerosol particles.

As a result of the nuclear weapons test, 1500 (1500) PBq of ^{137}Cs and 1300 (1300) PBq of ^{90}Sr were emitted into the environment [1,2]. Atmospheric fallout of radionuclides from a stratospheric (global) source leads to slow pollution of the surface of the entire earth prolonged many years-long. Global fallouts are not completely uniform, most of them, about 75%, occur in the Northern and only 25%—in the Southern Hemisphere. Minimum precipitation of the artificial radionuclides is observed at the poles and equator, the maximum—at latitudes from 30 to 60° in the Northern Hemisphere [10]. The Chernobyl NPP accident (with coordinates: 51°16'24" N, 30°13'20" E) caused release of 89.0 PBq of ^{137}Cs and 7.4 PBq of ^{90}Sr radionuclides into the atmosphere [4,10]. The Black Sea and the Crimean Peninsula are located between 30–40° N, i.e., are in the region of the greatest fallout of radioactive aerosols from the atmosphere both after testing nuclear weapons and after the Chernobyl accident. In the first months after the Chernobyl accident, the waters of the Black Sea and Crimea were subjected to acute radioactive contamination. In 1986, the radioecological situation in the Crimean saline lakes depended primarily on the arrival of artificial ^{90}Sr and ^{137}Cs radionuclides into these water bodies as a result of atmospheric global radioactive fallout and atmospheric transfer of radioactive substances after the Chernobyl accident [11–15]. In May 1986, the 1.7–2.4 PBq of ^{137}Cs and 0.3 PBq of ^{90}Sr precipitated on the area of the Black Sea and the Crimean peninsula after the Chernobyl Nuclear Power Plant accident [11]. Later, the secondary chronic transport of these radionuclides in Crimea, first of all ^{90}Sr, occurred since 1987 until April 2014 because of Dnieper waters through the North Crimean Canal (NCC) [11–16]. The NCC originated from the Kakhovskaya reservoir (lower part of the Dnieper river). The results of our radioecological monitoring showed that the concentration of ^{90}Sr in 1987 in the surface water of the Kakhovskaya reservoir was at order higher (645.7–991.6 Bq/m^3) than that after the atmospheric precipitation of this radionuclides in 1986 (19.6–98.1 Bq/m^3) and in 43–66 times higher than the pre-accident levels concentration of ^{90}Sr in water of the Black Sea (15 Bq/m^3) [16]. The concentration of ^{90}Sr in water of the NCC changed as follows: 59.3 Bq/m^3—in 1986, 522.2 Bq/m^3—in 1987, 105.6 Bq/m^3—in 1995 [11], 54.9 Bq/m^3—in 2011 [17]. At the same time in 2011, the ^{90}Sr concentration was 3.7 Bq/m^3 in water of the Chernorechenskoe reservoir, to which water came from the slopes of the Crimean mountains [17]. All above indicate on the secondary entry of the dissolved radionuclides from the Chernobyl NPP accident region through the cascade of the Dnieper reservoirs and the NCC into such remote areas as the Crimea and the Black Sea.

Crimea, the largest peninsula in the Black Sea, the most part of its area is an arid zone, and there are a lot of the diverse water objects with a salinity range from 0 to 400 g/L [18–21]. The saline lakes, including the Crimean ones, are very valuable ecosystems [22], and knowledge on their ecology and radiological situation is a key element of their sustainable integrated use including for aquaculture development.

The North Crimean Canal (NCC) was built in from 1961 to 1971 to ensure sustainable water supply in the south of Ukraine and Crimea. The total length of the NCC main branch from the Kakhovsky reservoir to Kerch is 403 km, which made it the longest canal in Europe [14,23,24]. The average annual NCC discharge reached 380 m^3/s, the maximum—up to 500 m^3/s, and about 80% of the water supplied through the NCC was used for agriculture in Crimea. Until April 2014, NCC also fed 11 artificial reservoirs in Crimea, the total volume of which is about 200 million m^3. Agricultural water was either discharged through the bypass channels or seeped into the nearby Crimean natural lakes. In April 2014, the supply of Dnieper water to Crimea through the North Crimean Canal was completely stopped because of the prevailing geopolitical situation leading to sharp salinity change in water bodies [14,24].

The relevance of this research is determined by the fact that the first radioecological studies in the Crimean saline lakes were only carried out in 2013. It was found that, despite that the Black Sea and the Crimean saline lakes had the same intake sources of artificial radionuclides ^{137}Cs and ^{90}Sr, their concentration, for example, ^{90}Sr (350.5 Bq/m^3) exceeded in the studied lakes that values found for the Black Sea by 23.4 times [13,16,25–27]. Of cause, the Black Sea and lakes differ in hydrological, hydrochemical, and biological characteristics, which determine the available differences in radionuclide content. Salinity is one of the most important factors affecting the abiotic environment, structure, and functioning of aquatic ecosystems [28–33]. It may be assumed that salinity could be among the key ecological factors affecting the distribution of radionuclides in water and their bioaccumulation by organisms. As an example, salinity can affect the solubility of stable strontium salts in water [31,32]. It is known that total content of ^{90}Sr and ^{137}Cs in the lakes depend on the sources of these radionuclides intake into the water body [16,27], however, the regularities in the dependence of the ^{90}Sr and ^{137}Cs distribution of ^{90}Sr and ^{137}Cs in the water of the Crimean saline lakes on salinity were not analyzed yet.

Previous studies conducted on the Crimean lakes showed that the concentrations of ^{90}Sr and ^{137}Cs in the waters of lakes dropped after closing of the canal and varied in a wide range of magnitudes in 2014–2017 [16,24–27]. Therefore, main question of this study: Does salinity influence the distribution of ^{90}Sr and ^{137}Cs in water of natural water bodies? Trying to answer this, authors analyzed the data collected in 2014–2017 to highlight salinity effect on concentration of ^{90}Sr and ^{137}Cs in water of several Crimean lakes with different salinity.

2. Materials and Methods

2.1. Studied Lakes

In Crimea, there are about 100 saline water bodies located in the arid steppe zone with small hills [19–21]; the study was conducted on 12 of them (Figures 1 and 2, Table 1).

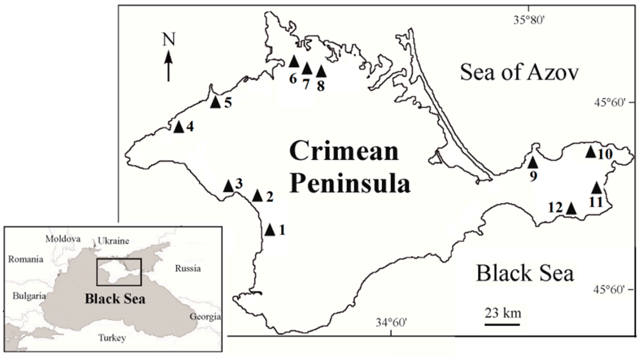

Figure 1. Distribution of the studied saline lakes in the Crimea: 1—Kyzyl-Yar, 2—Sasyk-Sivash, 3—Moynakskoye, 4—Dzharylgach, 5—Bakalskoye, 6—Krasnoye, 7—Kiyatskoye, 8—Kirleutskoye, 9—Aktashskoye, 10—Chokrakskoye, 11—Tobechikskoye, 12—Koyashskoye.

Figure 2. Photoviews of some studied lakes: (**a**)—Lake Tobechikskoye, (**b**)—Lake Kyzyl-Yar, (**c**)—Lake Bakalskoye, (**d**)—Lake Koyashskoye.

Table 1. Measured concentrations of ^{90}Sr and ^{137}Cs in water of the Crimean saline lakes during 2014–2017.

Lake	Date	No.	Coordinates	Salinity, g/L	pH	Concentration, Bq/m^3		Source of Data
						^{90}Sr	^{137}Cs	
Aktashskoye	11.04.2016	1	45°22′22″ N 35°46′42″ E	270	7.4	209.1 ± 13.8	-	[27]
	08.06.2016	2	45°23.483′ N 35°50.515′ E	89	8.5	85.8 ± 3.9	11.5 ± 0.5	[16]
	17.08.2017	3	45°23.144′ N 35°50.036′ E	200	8.1	196.9 ± 9.9	14.9 ± 0.9	new data
Bakalskoye	27.06.2016	4	45°45.514′ N 33°10.794′ E	47	8.6	40.0 ± 2.7	23.6 ± 1.1	[16]
	14.07.2017	5	45°43.921′ N 33°10.936′ E	62	8.2	41.9 ± 3.0	24.4 ± 1.0	new data
Chokrakskoye	08.06.2016	6	45°27.508′ N 36°18.325′ E	226	7.9	46.0 ± 3.4	49.3 ± 3.1	[16]
Dzharylgach	18.05.2016	7	45°33.965′ N 32°54.599′ E	115	8.5	29.7 ± 2.4	22.2 ± 2.4	[16]
	06.09.2016	8	45°33.967′ N 32°54.591′ E	130	8.1	54.6 ± 4.1	-	[16]
	08.11.2016	9	45°33.968′ N 32°51.582′ E	140	7.9	31.6 ± 2.8	34.6 ± 1.8	[16]
	14.07.2017	10	45°34.990′ N 32°51.505′ E	140	8.0	62.6 ± 5.1	36.7 ± 2.0	new data
Kirleutskoye	13.05.2014	11	45°56.572′ N 34°01.828′ E	184	7.6	121.3 ± 7.1	-	[26]
	18.11.2014	12	45°55.368′ N 34°02.385′ E	271	7.6	52.9 ± 8.4	-	[16]
	14.06.2016	13	45°55.231′ N 34°02.681′ E	235	7.9	18.7 ± 2.3	2.3 ± 0.1	[16]
	23.11.2016	14	45°53.753′ N 34°02.043′ E	207	8.2	76.1 ± 7.3	13.3 ± 1.0	[16]
	22.06.2017	15	45°55.479′ N 34°02.730′ E	267	7.2	76.7 ± 5.7	-	new data
Kiyatskoye	13.05.2014	16	46°0.084′ N 33°56.707′ E	107	7.8	98.0 ± 5.2	-	[26]
	18.11.2014	17	45°59.361′ N 35°57.365′ E	154	7.8	129.0 ± 7.5	-	[16]
	14.06.2016	18	45°59.729′ N 33°53.310′ E	200	7.7	60.1 ± 4.7	5.3 ± 0.3	[16]
	23.11.2016	19	45°58.395′ N 33°55.364′ E	224	8.2	209.2 ± 13.8	3.4 ± 0.2	[16]
	23.06.2017	20	45°58.684′N 33°54.572′E	218	7.4	59.0 ± 3.9	6.4 ± 0.4	new data
Koyashskoye	10.04.2016	21	45°02′08″N 36°12′03″E	230	7.9	39.0 ± 2.6	-	[27]
Krasnoye	14.06.2016	22	45°59.437′N 33°57.319′E	330	9.3	54.7 ± 21.3	37.4 ± 0.2	[16]
	23.06.2017	23	45°59.275′N 33°53.110′E	350	9.6	268.8 ± 23.9	46.5 ± 3.0	new data
Kyzyl-Yar	13.05.2014	24	45°03.558′N 33°37.460′E	2	7.1	9.6 ± 0.8	-	[26]
	18.05.2016	25	45°03.560′N 33°35.360′E	4	7.9	6.0 ± 0.8	0.8 ± 0.04	[16]
	06.09.2016	26	45°03.560′N 33°35.360′E	3	-	9.3 ± 1.1	-	[16]
	28.07.2017	27	45°03.982′N 33°37.782′E	4	7.8	5.9 ± 0.8	1.7 ± 0.1	new data
Moynakskoye	18.05.2016	28	45°10.518′N 33°18.597′E	47	8.2	8.3 ± 1.1	29.8 ± 1.4	[16]

Table 1. *Cont.*

Lake	Date	No.	Coordinates	Salinity, g/L	pH	Concentration, Bq/m^3		Source of Data
						^{90}Sr	^{137}Cs	
Sasyk-Sivash	27.06.2016	29	45°09.151′N 33°30.447′E	280	7.7	313.6 ±25.0	95.9 ± 8.1	[16]
	08.11.2016	30	45°12.282′N 33°31.526′E	322	7.8	258.9 ±14.7	83.0 ±7.1	new data
	28.07.2017	31	45°09.280′N 33°30.680′E	295	7.5	49.4 ± 9.2	106.7 ±8.7	new data
Tobechikskoye	07.06.2016	32	45°09.118′N 36°22.490′E	176	8.2	28.9 ± 2.2	8.7 ± 0.4	[16]
Chokrakskoye	17.08.2017 (pool)	-	45°27.835′N 36°18.526′E	400	6.6	below the detectable level	below the detectable level	new data
Tobechikskoye	16.08.2017 (pool)	-	45°09.238′N 36°22.623′E	400	7.2	below the detectable level	below the detectable level	new data

Lake Kiyatskoye is located in the northern part of the Crimea. The lake has an oblong shape, its area is 12.5 km^2, length—11 km, average width—2 km, and average depth is about 0.5 m. The lake is connected by a canal with a drainage system of the North Crimean Canal. This may explain one of the highest values of ^{90}Sr concentration—350.5 ± 15.8 Bq/m^3 in 2013 [26]. Salinity was around 83 g/L in 2013, and in 2014–2017, it fluctuated from 107 to 235 g/L.

Lake Kirleutskoye is also located in the northern part of the Crimea. The lake has an elongated shape, its area is 21 km^2, length—13 km, width—from 1.5 to 3 km, and depth is from 0.2 top 0.8 m. The lake level is 4.3 m below sea level and is the lowest among the lakes of the Perekop group. There is some water exchange with Lake Kiyatskoye. In 2014–2017, salinity fluctuated from 184 to 271 g/L.

Lake Krasnoye is also located in the Perekop lake group as two lakes above. The lake area is 23.5 km^2, the average depth is 1.5 m. It elongated in the meridional direction, length of over 13 km, the maximum width is 2.5 km. The dam divides the lake in two parts, the northern part of the lake is used as a storage tank-evaporator of industrial waste of the Crimean Soda Plant, and heavy metal pollution is very high. In 2016–2017, salinity fluctuated from 330 to 350 g/L.

Lake Bakalskoye is located on the Bakalskaya sand spit in the West Crimea. It is one of the most studied Crimean lakes [34]. The lake has an oval shape, its length from north to south is 4 km, and from west to east is 3.5 km. The area of the lake is very volatile throughout the year, and it varies, amounting to 7–8 km. The depth varies from 0.5 to 1.5 m. In 2000–2017, salinity fluctuated from 16 to 110 g/L. The lake ecosystem fluctuates between two alternative states; in different years, it may be with intensive development of floating filamentous green algae *Cladophora* mats and without mats [34].

Lake Dzharylgach is also included in the Tarkhankut lake group as Lake Bakalskoye. Its area is 8.3 km^2, the length is 8.5 km and the average width is 1 km with the largest 2.3 km. The average depth is 0.5 m, the largest of 1.0–1.6 m. In 2007–2017, salinity fluctuated from 90 to 140 g/L.

Lake Moynakskoye is located on the western outskirts of Evpatoria city. The lake is stretched from the south to the north, perpendicular to the coastline of the sea. The length of the lake is 1.85 km, the largest width is 0.89 km, its depth is from 0.45 m to 1 m, and its area is 1.76 km^2. Until the beginning of 1970, salinity fluctuated in the range of 160–200 g/L in the lake. Then intensification of agricultural activity began, and the flow of fresh water into the lake increased, salinity gradually began to decline [35]. In 1986 salinity dropped to 80 g/L, in 1996 to 60–70 g/L, in 1997 to 55–60 g/L, and in 2007–2018 it fluctuated from 45 to 55 g/L. The lake is used as a famous balneological source of medical mud for more than 150 years. Salinity dropping threatens the loss of medicinal properties of the lake.

Lake Sasyk-Sivash is the largest Crimean lake with an area of 75.3 km^2. The length is 14 km. The average width is 5.5 km, the largest is 9 km. The depth is medium—0.5 m, the largest—1.2 m. It is separated from the sea by a spit with a width of 0.9–1.7 km. In summer the lake area is significantly reduced, and the salinity of the water increases. In 2016–2018, salinity fluctuated between 280 and 340 g/L.

Lake Kyzyl-Yar enters into the Evpatoria group of lakes and is separated from the Black Sea by a narrow sandy spit. The area is 8 km^2, the length is 5.7 km, the average width is 1.4 km, the average depth is 2.0 m, and the largest is 3.7 m. It was a hypersaline lake with salinity higher than 200 g/L before construction of the reservoir of fresh water from the North Crimean canal built upper 20 km in the gully. After that filtration of fresh water began into the lake along the gully, and 20–25 years later, waters in the lake became fresh. In 2010–2013, salinity fluctuated between 1.5 and 2.5 g/L, and in 2014–2018, it fluctuated from 1.9 to 4 g/L.

Lake Tobechikskoye enters into the Kerch group of lakes, the third in area in this group (18.7 km^2), after lakes Aktashskoye and Uzunlarskoye. The length is 9 km. The average width is 2 km, the largest is 4.5 km. The depth is average—1 m, the largest—1.2 m. The lake is separated from the Kerch Strait by a narrow spit. In 2000–2018, salinity fluctuated from 50 to 370 g/L. The lake ecosystem can be in different alternative states [36].

Lake Koyashskoye is also on the Kerch peninsula. The lake was separated from the Black Sea by a narrow sand spit [37]. Its area is 5 km^2, length is 3.84 km. The maximum width is 2.81 km, the average is 1.2 km. The catchment area is 23 km^2, the average depth is 0.5 m, and the largest one is 1.0 m. In 2000–2017, salinity fluctuated between 150 and 370 g/L.

Lake Akhtashskoye is situated on the Sea of Azov side of the Kerch peninsula. The area is 26.8 km^2, the length is 8 km, the average width is 3.0 km, the largest is 3.5 km, and the average depth is 0.7 m, the largest—2 m. It is connected with the North Crimean channel. There were wide spatial and temporal fluctuations of salinity—from 30 to 280 g/L in 2007–2017.

Lake Chokrakskoye is also situated on the Sea of Azov side of the Kerch peninsula. The lake has an oval shape and is separated from the sea by a narrow sand spit, the area is 8.5 km^2, the catchment area is 74 km^2, the length is 4.1 km, the average width is 1.9 km, the largest is 3.6 km, the average depth is 0.5 m, and the largest is 1.5 m. In summer, it often almost completely dries up. In 2006–2017, salinity fluctuated from 180 to 400 g/L.

2.2. Method of Determination of the ^{90}Sr

The water samples of 20 L were taken by a plastic canister in lakes to measure the ^{90}Sr concentration. Simultaneously with biota sampling salinity, temperature and pH were measured using a refractometer Kellong WZ212 (Kelilong Electron Co. Ltd, Fuan, China) and a pH meter PHH-830 (OMEGA Engineering, INC., Norwalk, CT, USA). Collected samples were transported to the laboratory for analysis. The method of ^{90}Sr measurement was based on a following radiochemical procedure. After acid leaching and/or preconcentration of strontium with a carbonate for water, 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) Quantulus 1220 (LKB Wallac Oy, Turku, Finland). The lower limit of detection (LLD) for water samples for water samples was 0.01–0.04 Bq/m^3. Recoveries were calculated from stable Sr recovery by flame photometry for ^{90}Sr and gravimetrically from yttrium oxalate for ^{90}Y [12,38]. Data were reported as the mean of the values activity of parallel duplicate samples, which were measured separately. Variation between the data points 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 [39,40] and they were certified as reliable data.

2.3. Gamma-Spectrometric Measurements of ^{137}Cs

The ^{137}Cs concentration in water was measured using samples of 100 L taken by a plastic canister into the plastic barrels. To determine the dissolved forms of ^{137}Cs (directly the activity of ^{137}Cs in water) the collected water was filtered through Millipore membrane filters with a diameter of nominal pore size of 0.45 µm to remove the suspended matter. Then carried out sorption extraction ^{137}Cs using an installation consisting of two sequentially connected absorbers filled with a granular sorbent. This inorganic composite sorbent is a thin film of mixed ferrocyanide of potassium iron obtained by chemical method from aqueous solutions on a carrier which is delignified wood meal. The approximate composition of the film is K (0.2–1.8) FeO [Fe(0.3–0.7)(CN)1.8–4.2] [41]. The water was pumped through the adsorbers with using a peristaltic pump type 372.C (Elpan, Lubawa, Poland) at a rate of 0.055 L/min, at which the efficiency of extracting ^{137}Cs was between 50 and 90% [42]. Then the sorbent was removed from the adsorbers and was ashing in a muffle furnace at a temperature of 400 °C to reduce the volume. The radiometric measurements were performed using a scintillation gamma spectrometer Compugamma (LKB Wallac Oy, Turku, Finland). Uncertainties for ^{137}Cs activities were propagated from the one sigma counting uncertainty, the uncertainty because of background correction, calibration uncertainties, and the uncertainty associated with the subtraction of activities between the first and second adsorbers [11].

2.4. Data Analysis

Parameters of the regression equations, average values, standard deviations (SD), coefficients of variation (CV), determination (R^2) and correlation (R) were calculated in the standard program MS Excel 2007. The confidence level (p) of the calculated correlation coefficients was determined [43]. Using Student's *t*-test, the significance of differences in mean values was (p) evaluated. In STATISTICA 6, cluster analysis with calculation of the Euclidian distance between samples and building the dendrograms was made.

3. Results

During sampling, temperature in the lakes fluctuated from 19.6 to 33.3 °C and pH—from 7.1 to 9.6; no significant correlations with their value ($p > 0.2$) and other parameters were found. The observed values of salinity and pH are given in the Table 1. Previously, it was also shown that pH did not affect the concentration of artificial radionuclides in water of saline lakes [11].

^{90}Sr. The data on the measured concentrations are given in the Table 1. From a sample to a sample, the ^{90}Sr concentration varied in rather wide limits; there were differences between lakes as well as between the samples taken in one lake in different time. Significant correlation between ^{90}Sr and salinity was found using the whole data array, and the best approximation was given by the equation (R = 0.736, $p = 0.005$):

$$Y = 3.937 \times S^{0.554}, \qquad (1)$$

where Y is the concentration of ^{90}Sr in water, Bq/m^3; S is the salinity, g/L.

Based on the determination coefficient (R^2), about 54% of the total variability of ^{90}Sr concentration in water in total dataset can be explained by salinity changes. In this case, at high salinities, there is a significant variability in the values of strontium concentration in water. In the salinity range of 200–300 g/L, the data are divided into two disjoint groups. Using data on salinity and ^{90}Sr concentrations, cluster analysis of all samples was done. All samples were grouped in two clusters as Figure 3a shows.

There was no significant relation between salinity and ^{90}Sr concentration in the first cluster (Figure 4a). It only may be suggest that there was some positive trend in a salinity range 100–150 g/L and a negative trend when salinity exceeded 150 g/L. There was direct positive relation between salinity and ^{90}Sr concentration in the second cluster (R = 0.970, $p = 0.001$) (Figure 4b). The average concentration of ^{90}Sr in the first cluster was 54.9 Bq/m^3 (SD = 48.58, CV = 0.34), and the average value in the other one was 227.6 Bq/m^3 (SD = 18.45, CV = 0.22). These differences are significant ($p = 0.001$).

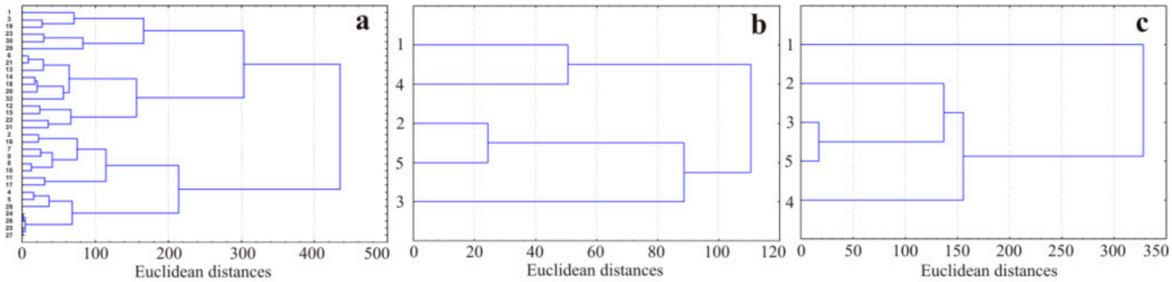

Figure 3. The dendrogram of similarity in relation "salinity—^{90}Sr concentration" between samples taken from the studied Crimean lakes (a sample number was given in Table 1): (**a**) for all samples, (**b**) for samples taken from Lake Kirleutskoye, (**c**) for samples taken from Lake Kiyatskoye.

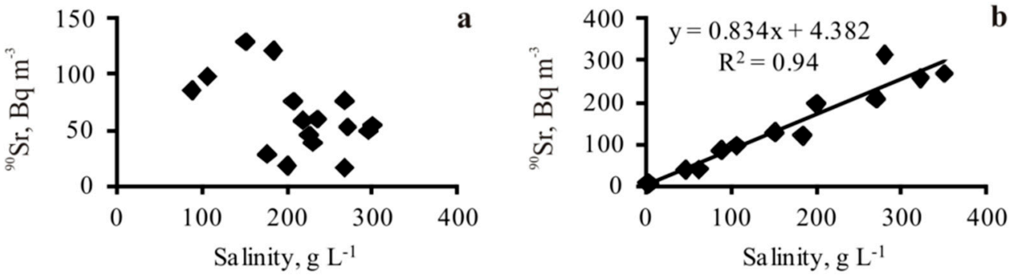

Figure 4. The correlation between salinity and ^{90}Sr concentration in the first (**a**) and second (**b**) cluster of the studied lakes.

Samples from the same lake, collected at different time points, fell into different clusters. Samples collected from Kiyatskoye and Kirleutskoye lakes split into separate clusters (Figure 2b,c). This separation cannot be explained by season or any other accounted factor. Interestingly, when the surface of lakes Tobechikskoye and Chokrakskoye was almost completely covered with deposited salt, in the brine pools (salinity was 400 g/L), ^{90}Sr concentration was below the detection level (Table 1). At a salinity of 400 g/L, when the salts precipitate, radionuclides move into the bottom sediments [16]. According to our measurements, the radionuclides are not detected in salt from lakes of Crimea [11]. So, to identify patterns, we considered data on the salt lakes of Crimea, where salinity was 350 g/L and lower (Table 1, Figure 4).

^{137}Cs. Data on the measured concentrations of ^{137}Cs are given in the Table 1. There is a significant positive correlation between ^{137}Cs concentration and salinity. The best approximation of this relation is given by the equation (R = 0.604, p = 0.005):

$$Z = 0.750 \times S^{0.61}, \tag{2}$$

where Z is the concentration of ^{137}Cs in water, Bq/m^3, S is the salinity, g/L.

Based on the determination coefficient (R^2), about 37% of the total variability of a ^{137}Cs concentration in lake water can be explained by only salinity changes. As in the case of strontium, cluster analysis divided all samples into two separate groups (Figure 5).

No correlation between ^{137}Cs concentration and salinity was found in one cluster, and significant positive relation between these parameters was observed in the second cluster (R = 0.708, p = 0.001) (Figure 6).

Salinity delimited these two clusters: in one cluster, all samples were taken in a salinity range from 3.7 to 89 g/L, in other—from 176 to 350 g/L. Maximum concentration was 107 Bq/m^3 at salinity 295 g/L in one cluster, and 30 Bq/m^3 at salinity 47 g/L in the other one. It is interesting to note that when the surface of the Tobechikskoye and Chokrakskoye lakes was almost completely covered with salt, in the pools of brine (salinity above 370 g/L) the cesium concentration was below the detection level similar as for strontium. There is a significant positive correlation between logarithms of ^{137}Cs and ^{90}Sr concentrations (R = 0.516, p = 0.01). At the same time, no more than 27% of the variability of one isotope correlates with changes of the other.

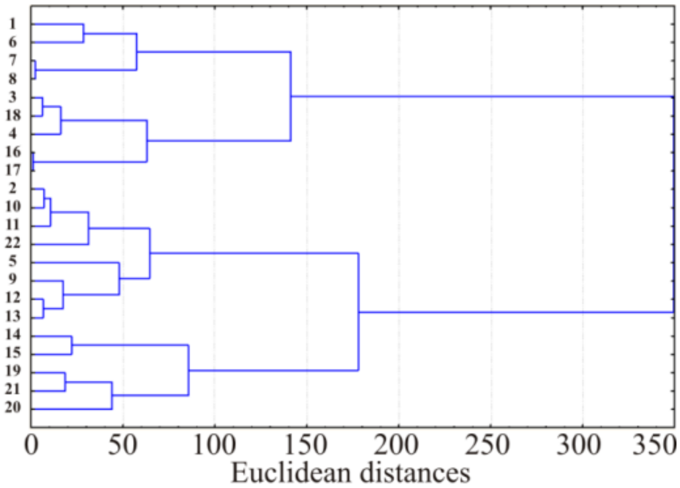

Figure 5. The dendrogram of similarity in relation "salinity—^{137}Cs concentration" between samples taken from the studied Crimean lakes (a sample number was given in Table 1).

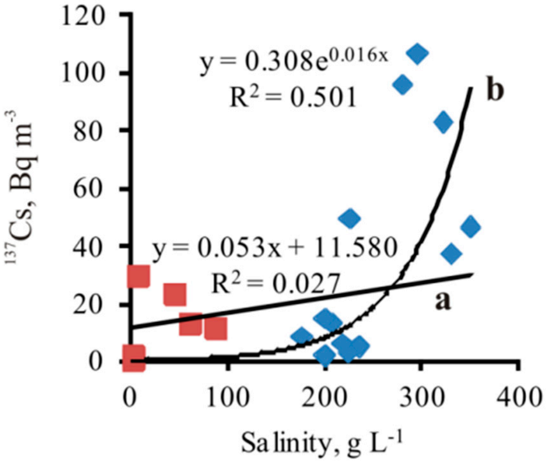

Figure 6. The correlation between salinity and ^{137}Cs concentration in the first (**a**) and second (**b**) cluster of the studied lakes.

4. Discussion

The main reasons for a difference in concentrations of radionuclides ^{90}Sr и ^{137}Cs in water of the lakes are the sources of radioactive contamination and processes of these radionuclides elimination [26,27]. The input of artificial radionuclides into water body depends on the source of anthropogenic pollution, and elimination of these isotopes from hydrological and biogeochemical processes occurring in the aquatic ecosystems [11,27]. Processes of lake metabolism depend on lake morphometry and catchment conditions [44,45]. One of the factors affecting the isotopes concentrations in the water of the studied lakes is a level of connection of a lake with the North Crimean channel, as a source of secondary pollution by radionuclides in the post-Chernobyl accident years before the closure of the canal in 2014 [14,27]. In the years after the accident, the Crimean lakes were polluted by secondary chronic radionuclide contamination of ^{90}Sr and ^{137}Cs from the Dnieper waters coming from the North-Crimean Canal [13,16]. The Crimean lakes had different level of interconnection with the North-Crimean Canal that affects the level of artificial radionuclide contamination [27]. As an example, ^{90}Sr concentration was lower by 5.4 times in water and 9.3 in sediments of Lake Koyashskoye, which never had connection to the Canal, than in Lake Akhtashskoye, connected to the Canal. After closing of the Canal in April 2014, as our data demonstrated, concentrations of ^{90}Sr and ^{137}Cs significantly dropped in some lakes. As an example, in Lake Kiyatskoye, ^{90}Sr concentration dropped from 350.5 (SD = 16) [26] to 111 (SD = 62).

^{90}Sr is chemically close to stable isotopes of strontium (^{84}Sr, ^{86}Sr and ^{87}Sr), total sum of which was studied in the Crimean saline lakes earlier [46]. It was shown that sum of stable strontium isotopes varied from 12 to 295 mg/L in water of the Crimean saline lakes. It also varied in single lake, as an example, in Lake Kiyatskoye—from 99 to 223 mg/L and in Lake Chokrakskoye—from 12 to 38 mg/L. Using the available coefficient [8], the authors transformed ^{90}Sr concentrations from Bq/m^3 into g/m^3. There is a significant positive correlation between ^{90}Sr in 2014–2017 and sum of stable strontium isotopes in a lake (Figure 7) ($R = 0.867$, $p = 0.03$). This relationship can be explained by geochemical similarity of artificial and natural strontium isotopes when a new supply of ^{90}Sr from the canal is absent. Using own and published [46] data, authors evaluated a proportion between sum of stable Sr isotopes and artificial ^{90}Sr for different lakes (concentration of ^{90}Sr/concentration of sum of stable Sr); it was in average 1.8×10^{-10} ($SD = 0.97 \times 10^{-10}$, $CV = 0.537$).

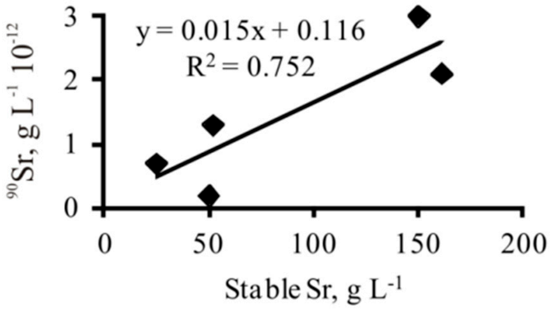

Figure 7. Relation between ^{90}Sr in 2014–2017 and sum of stable strontium isotopes in the Crimean saline lakes (data on stable Sr concentrations were taken from [46]).

Analysis of the above data shows that salinity influences the content of the both isotopes, which also related to the other characteristics of the investigated lakes during sampling. The salinity affects the concentration of radionuclides in water through influencing their solubility and isotope exchange between the solution and bottom sediments, as well as through their accumulation in aquatic organisms. Chemically strontium is close to calcium. The alkali metal chlorides and other salts dramatically increase the solubility of calcium and strontium salts, and the solubility of strontium sulfate increases by a factor of 13 in water with a content of 15% sodium chloride [27]. The solubility of strontium carbonate in distilled water is 8–10 mg/L, calcium carbonate—13–60 mg/L, strontium sulfate—148 mg/L, and calcium sulfate over 2000 mg/L, and this indicates that strontium carbonate and sulphate should crystallize simultaneously with calcium carbonate and gypsum [46]. Calcium precipitates mainly in the form of two salts ($CaSO_4$ and $CaCO_3$), the crystallization of which begins at the total salinity of about 135–155 g/L and the bulk of which precipitates in the salinity range of 180–360 g/L [19,46]. Strontium salts are also precipitated practically in this range. As a result of crystallization and sedimentation of these salts, the concentration of calcium and strontium ions in brine, as a rule, begins to decrease with a salinity increase above 135–150 g/L [46]. Relation "salinity—^{90}Sr concentration" reflects this in the first cluster. In the second cluster, there is an increase of ^{90}Sr concentration in whole salinity range—up to 350 g/L. Authors will try to explain this relation. In the Crimean salt lakes, there are quite often cases when the water is oversaturated with calcium and strontium ions, which can be explained primarily by a drop in the concentration of sulfates in water [46]. The lack of sulfate ions prevents the crystallization of gypsum (Ca_2SO_4) and its sedimentation, and the ^{90}Sr ions remain in water. The concentration of sulfate ions depends on several factors, and microbiological activity is one of the most important factors [19,46,47].

On average, the solubility of NaCl in water at 20 °C is 360 g/L increasing with a temperature rise [48]. At this point, all salts have sedimented. Water pools on salt crust contain rain water or water from freshwater springs. Upper layer of salt crust contains mostly chloride salts ($MgCl_2$, $CaCl_2$, NaCl) and only this salt can dissolve in water reaching a salinity limit shortly. This explains the behavior of the studied isotopes in brine with salinity above 370 g/L, when dissolved radionuclide salts in brine are almost absent. Salinity determines more than 90% of ^{90}Sr concentration variability in the Crimean saline lake water up to the beginning of crystallization (salinity 140–160 g/L) in both clusters. When the process of crystallization occurs, salinity is not the main determinant of ^{90}Sr concentration variability in water/brine.

To explain the salinity impact on ^{137}Cs concentration in brine is more difficult than on ^{90}Sr concentration. Cesium salts have the highest solubility among the alkali metal salts of weak acids [49], and salinity cannot directly influence the ^{137}Cs concentration. The solubility of NaCl, for example, is lower by 6.2 times than CsCl, and the solubility of K_2SO_4 is lower by 23 times than Cs_2SO_4. Such high solubility does not give us a chance to explain the found influence of salinity on ^{137}Cs concentration by processes of crystallization of salts with ^{137}Cs. Clay minerals can absorb cesium [50] as well as there is cesium complexation with particulate organic matter [51]. The concentration of Cs in lake brine and its sedimentation on bottom are also influenced by the quantity and quality of suspended matter (TSM) in lake brine. Content and composition of TSM in brine of the Crimean saline lakes are the variable characteristics. As an example, total concentration varied from 9 to 540 g/m^3 and share of organic matter in SM, from 10 to 88% [52]. The concentration of seston in lakes is determined by the ratio of the production and destruction processes of organic matter as well as the intensity of sedimentation and resuspension. High salinity may influence the sedimentation intensity through crystallization of some Ca salts; sedimenting salts can attach to suspended matter and together sink on bottom. Currently, the positive relation between the concentration of total suspended solids and salinity was found [52,53]. Thus, salinity can also influence the distribution of ^{137}CS indirectly, thereby complicating the overall picture and making necessary more comprehensive studies of this issue.

Salinity is not the only factor determining the behavior of isotopes in the lake water; its values determine only 61% of the total variation in strontium concentration and 33% of the cesium concentration. Found differences in dependence of content of two radioactive isotopes in water from salinity can be explained by differences in the chemical properties of these elements. Differences in the behavior of ^{90}Sr and ^{137}Cs in freshwater ecosystems were noted by Timofeeva-Resovskaya [54]. According to her classification, ^{137}Cs is a pedotope, i.e., belongs to the class of elements, 95–100% of which are accumulated by bottom sediments, whereas ^{90}Sr belongs to equitrops, i.e., elements uniformly distributed in the water column, biota, and bottom sediments in the ecosystems.

Probably, there are several other reasons for fluctuations in the concentration of these isotopes in the lake water. The authors cannot present a representative list of all the causes and quantify their role. However, some assumptions in this direction can be made. The concentration of the isotopes can be affected by the ratio of different salts in water, and not only by the total salinity or NaCl concentration [55,56]. This, in particular, can be determined by the fact that many organisms, massively developing, can selectively accumulate certain isotopes, changing their concentrations in water. When decomposing organic matter, these isotopes can partially return to the water. The bioaccumulation of strontium and cesium by aquatic organisms are known to be inversely and strongly dependent on the concentrations of their metabolic analogues: K, potassium, is for cesium and Ca, calcium, is for strontium [5]. Resuspention of sediments by strong winds in the Crimean shallow saline lakes is a common phenomenon [57], and this influences the re-distribution of radionuclides between water and bottom sediments. Consequently, content of isotopes in water can also depend on the regime of winds, their speed, and direction. In the Crimean hypersaline lakes and lagoons, up to a salinity of 150–170 g/L, the floating mats of filamentous green alga *Cladophora* can develop intensively [24,36]. Their intensive development can completely exclude resuspension of sediments by wind [34]. Floating mats often contribute to the formation of hypoxic or anoxic conditions at the bottom [24,36], which may also affect isotope exchange between the water and the bottom.

All above arguments lead to the conclusion that there are a number of factors and mechanisms affecting the behavior of isotopes in lakes, but our knowledge is still insufficient to make an integral analysis of interaction of these factors. The Crimean saline lake ecosystems can be in different alternative states [32,34,36], and there are also alternative geochemical/biogeochemical lake states as the cluster analysis shows. In different lake states, there are distinct types of relations between artificial radionuclide concentration and salinity. In future, it would be interesting to understand how ecological and geochemical alternative states are connected and interact with each other.

According the Russian Radiation Safety Standards NRB-99/2009 [58], the threshold limit value/the maximum permissible concentration in drinking water for ^{90}Sr is 4.9 Bq/L (4900 Bq/m^3), and for ^{137}Cs is 11.0 Bq/L (11,000 Bq/m^3); these values are much higher than that observed in the Crimean lakes now. The authors conclude that there are no safety risks from ^{137}Cs and ^{90}Sr contamination in the Crimean lakes for human health and aquaculture development.

5. Conclusions

For the first time for the Crimea saline lakes, a positive correlation between concentrations of after-Chernobyl radionuclide ^{90}Sr and ^{137}Cs in water and salinity was revealed, regardless of the source of radionuclides in aquatic ecosystems. The salinity determines only 61 and 33% of the total variability of ^{90}Sr and ^{137}Cs concentrations in lake water, respectively. Salinity cannot influence the sources of radionuclide input in the lakes but it determines a balance in distribution of radionuclides between water and bottom contributes to the retention of radioisotopes in water. Salinity is not the alone factor determining the behavior of radionuclides in the lakes and acts only by interacting with other factors. At the moment, the authors could not provide the complete list of all the interacting factors and causes in the determination of radionuclide behavior in the lakes. Further wider and deeper field and experimental studies are required to create an integral picture of all the factors affecting the concentration of artificial radionuclides in natural waters. There are no safety risks from ^{137}Cs and ^{90}Sr contamination for human health and aquaculture development in the Crimean lakes.

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References

1. Aarkrog, A. Source terms and inventories of anthropogenic radionuclides. In *Radioecology. Lectures in Environmental Radioactivity*; Holm, E., Ed.; World Scientific Publishing: Lund, Sweden, 1994; pp. 21–38.
2. Aarkrog, A. Input of anthropogenic radionuclides into the World Ocean. *Deep Sea Res. Part II* **2003**, *50*, 2597–2606. [CrossRef]
3. Kessides, I.N. The future of the nuclear industry reconsidered: Risks, uncertainties, and continued promise. *Energy Policy* **2012**, *48*, 185–208. [CrossRef]
4. Brumfiel, G. Directly Comparing Fukushima to Chernobyl. Nature News Blog. 2011. Available online: http://blogs.nature.com/news/2011/09/directly_comparing_fukushima_t.html (accessed on 8 August 2018).
5. Brinkmann, L.; Rowan, D.J. Vulnerability of Canadian aquatic ecosystems to nuclear accidents. *Ambio* **2018**, *47*, 585–594. [CrossRef]
6. Hakanson, L.; Andersson, T.; Nilsson, A. Radioactive cesium in fish in Swedish lakes 1986–1988 general pattern related to fallout and lake characteristics. *J. Environ. Radioact.* **1992**, *15*, 207–229. [CrossRef]
7. Fuller, N.; Lerebours, A.; Smith, J.T.; Ford, A.T. The biological effects of ionising radiation on Crustaceans: A review. *Aquat. Toxicol.* **2015**, *167*, 55–67. [CrossRef]

8. Rikhvanov, L.P. *Common and Regional Problems of Radioecology*; Publishing house of Tomsk Polytechnic University: Tomsk, Russia, 1997; 384p. (in Russian)
9. Izrael, Y.A. Radioactive contamination of the Earth's surface. *Her. Russ. Acad. Sci.* **1998**, *68*, 116–131. (In Russian)
10. Gudiksen, P.H.; Harvey, T.F.; Lange, R. Chernobyl source term, atmospheric dispersion and dose estimation. *Health Phys.* **1989**, *57*, 697–705. [CrossRef] [PubMed]
11. Polikarpov, G.G.; Egorov, V.N.; Gulin, S.B.; Stokozov, N.A.; Lazorenko, G.E.; Mirzoyeva, N.Y.; Tereschenko, N.N.; Tsitsugina, V.G.; Kulebakina, L.G.; Popovichev, V.N.; et al. *Radioecological Response of the Black Sea to the Chernobyl Accident*; EKOSI-Gidrofizika: Sevastopol, Russia, 2008; 667p. (In Russian)
12. Mirzoyeva, N.Y.; Kulebakina, L.G. Methods of radiochemical determination of ^{90}Sr in environmental samples. In *Radioecological Response of the Black Sea to the Chernobyl NPP Accident*; Polikarpov, G.G., Egorov, V.N., Eds.; EKOSI-Gidrofizika: Sevastopol, Russia, 2008; pp. 56–61. (In Russian)
13. Gulin, S.B.; Mirzoyeva, N.Y.; Egorov, V.N.; Polikarpov, G.G.; Sidorov, I.G.; Proskurnin, V.Y. Secondary radioactive contamination of the Black Sea after Chernobyl accident: Recent levels, pathways and trends. *J. Environ. Radioact.* **2013**, *124*, 50–56. [CrossRef] [PubMed]
14. Gulin, S.B.; Mirzoyeva, N.Y.; Lazorenko, G.E.; Egorov, V.N.; Trapeznikov, A.V.; Sidorov, I.G.; Proskurnin, V.Y.; Popovichev, V.N.; Bey, O.N.; Rodina, E.A. Contemporary radiological situation related to regime of the functioning of the North-Crimean Canal. *Radiat. Biol. Radioekol.* **2016**, *56*, 1–8. (In Russian)
15. Kuzmenko, M.I.; Volkova, E.N.; Klenus, V.G. Radioactive contamination of the Dnieper and its reservoirs and some measures after the Chernobyl accident. *Hydrobiol. J.* **1992**, *28*, 51–86. (In Russian)
16. Mirzoyeva, N.Y.; Arkhipova, S.I.; Kravchenko, N.V. Sources of inflow and nature of redistribution of ^{90}Sr in the salt lakes of the Crimea. *J. Environ. Radioact.* **2018**, *188*, 38–46. [CrossRef] [PubMed]
17. Mirzoyeva, N.Y.; Egorov, V.N.; Gulin, S.B. Radionuclides^{137}Cs and^{90}Sr in components of the Black Sea ecosystems: Contemporary status and prognosis. In *Diversity in the Coastal Marine Sciences: Historical Perspectives and Contemporary Research of Geology, Physics, Chemistry, Biology and Remote Sensing*; Finkl, C.W., Makowski, C., Eds.; Springer: Dordrecht, The Netherlands, 2018; Chapter 17; pp. 275–294.
18. Kurnakov, N.S.; Kuznetsov, V.G.; Dzens-Lytovsky, A.I.; Ravich, M.I. *The Crimean Salt Lakes*; AN USSR Publ.: Moscow, Russia, 1936; 278p. (In Russian)
19. Anufriieva, E.; Hołyńska, M.; Shadrin, N. Current invasions of Asian Cyclopid species (Copepoda: Cyclopidae) in Crimea, with taxonomical and zoogeographical remarks on the hypersaline and freshwater fauna. *Ann. Zool.* **2014**, *64*, 109–130. [CrossRef]
20. Anufriieva, E.; Shadrin, N. Resting stages of crustaceans in the Crimean hypersaline lakes (Ukraine) and their ecological role. *Acta Geol. Sin. Eng. Ed.* **2014**, *88*, 46–49. [CrossRef]
21. Shadrin, N.V.; Anufriieva, E.V.; Belyakov, V.P.; Bazhora, A.I. Chironomidae larvae in hypersaline waters of the Crimea: Diversity, distribution, abundance and production. *Eur. Zool. J.* **2017**, *84*, 61–72. [CrossRef]
22. Shadrin, N.; Zheng, M.; Oren, A. Past, present and future of saline lakes: Research for global sustainable development. *Chin. J. Oceanol. Limnol.* **2015**, *33*, 1349–1353. [CrossRef]
23. Denisova, A.I.; Timchenko, V.M.; Nakhshina, E.P.; Novikov, B.I.; Ryabov, A.K.; Bass, Y.I. *Hydrology and Hydrochemistry of the Dnieper and its Reservoirs*; Naukova Dumka: Kiev, Ukraine, 1989; 216p. (In Russian)
24. Shadrin, N.V.; Anufriieva, E.V.; Kipriyanova, L.M.; Kolesnikova, E.A.; Latushkin, A.A.; Romanov, R.E.; Sergeeva, N.G. The political decision caused the drastic ecosystem shift of the Sivash Bay (the Sea of Azov). *Quatern. Int.* **2018**, *475*, 4–10. [CrossRef]
25. Gulin, S.B.; Gulina, L.V. Natural and technogenic radionuclides in ecosystem of the salt lake Koyashskoye (south-eastern Crimea). *Mar. Ecol. J.* **2011**, *1*, 19–25. (In Russian)
26. Mirzoyeva, N.; Gulina, L.; Gulin, S.; Plotitsina, O.; Stetsuk, A.; Arkhipova, S.; Korkishko, N.; Eremin, O. Radionuclides and mercury in the salt lakes of the Crimea. *Chin. J. Oceanol. Limnol.* **2015**, *33*, 1413–1425. [CrossRef]
27. Mirzoyeva, N.Y.; Arkhipova, S.I.; Anufriieva, E.V.; Shadrin, N.V. Marine hypersaline lakes of the Kerch Peninsula and the differences in their responses to external influences: ^{90}SR concentration after the Chernobyl accident Indicators of climatic changes. In *Indicators of Climatic Changes in Marine Ecosystems*; Berdnikov, S.V., Yaitskaya, N.A., Lychagina, Y.M., Eds.; SSC RAS Publishers: Rostov-on-Don, Russia, 2016; pp. 52–59.
28. Khlebovich, V.V. *Critical Salinity of Biological Processes*; Nauka: Leningrad, Russia, 1974; 236p. (In Russian)

29. Williams, W.D. Salinity as a determinant of the structure of biological communities in the salt lakes. *Hydrobiologia* **1998**, *381*, 191–201. [CrossRef]
30. Khlebovich, V.V.; Aladin, N.V. The salinity factor in animal life. *Her. Russ. Acad. Sci.* **2010**, *80*, 299–304. [CrossRef]
31. Shadrin, N. Hypersaline lakes as the polyextreme habitats for life. In *Introduction to Salt Lake Sciences*; Zheng, M., Oren, A., Deng, T., Eds.; Science Press: Beijing, China, 2018; pp. 180–187.
32. Shadrin, N.; Anufriieva, E. Ecosystems of hypersaline waters: Structure and trophic relations. *Zh. Obshch. Biol.* **2018**, *79*, 418–427. (In Russian)
33. Shadrin, N. Peculiarities of structure, functioning and dynamics of the salt lake ecosystems. In *Introduction to Salt Lake Sciences*; Zheng, M., Oren, A., Deng, T., Eds.; Science Press: Beijing, China, 2018; pp. 188–197.
34. Shadrin, N.V.; Anufriieva, E.V. Climate change impact on the marine lakes and their Crustaceans: The case of marine hypersaline Lake Bakalskoye (Ukraine). *Turk. J. Fish. Aquat. Sci.* **2013**, *13*, 603–611. [CrossRef]
35. Bondarenko, L.V.; Yakovenko, V.A. Transformation of the crustacean species structure in Lake Moinaki in connection with its desalination. *Visnyk of Dnipropetrovsk University. Biol. Ecol.* **2000**, *8*, 100–105. (In Russian)
36. Golubkov, S.M.; Shadrin, N.V.; Golubkov, M.S.; Balushkina, E.V.; Litvinchuk, L.F. Food chains and their dynamics in ecosystems of shallow lakes with different water salinities. *Russ. J. Ecol.* **2018**, *49*, 442–448. [CrossRef]
37. Khomenko, S.V.; Shadrin, N.V. Iranian endemic *Artemia urmiana* in hypersaline Lake Koyashskoye (Crimea, Ukraine): A preliminary discussion of introduction by birds. *Branta. Trans. Azov Black Sea Ornithol. Stn.* **2009**, *12*, 81–91. (In Russian)
38. Harvey, B.R.; Ibbett, R.D.; Lovett, M.B.; Williams, K.J. Analytical procedures for the determination of strontium radionuclides in environmental materials. In *Aquatic Environment Protection: Analytical Methods*; Ministry of Agriculture Fisheries and Food, Directorate of Fisheries Research: Lowestoft, UK, 1989; Volume 5, pp. 1–33.
39. International Atomic Energy Agency (IAEA). *AQCS: Catalogue for Reference Material Sandinter-Comparison Exercises 1998/1999*; International Atomic Energy Agency (IAEA): Vienna, Austria, 1998; 136p.
40. International Atomic Energy Agency (IAEA). *Reference Materials Catalogue 2004–2005*; International Atomic Energy Agency (IAEA): Vienna, Austria, 2004; 116p.
41. Betenkov, N.D.; Kaftailov, V.V.; Bushkov, E.I. Sorption of cesium from solutions of the seawater type. *Radiochemistry* **1987**, *29*, 127–129. (In Russian)
42. Bey, O.N.; Proscurnin, V.Y.; Gulin, S.B. Measurement of the Cs concentration from its own β-radiation using liquid scintillation spectrometry. *Radiochemistry* **2016**, *58*, 147–149. [CrossRef]
43. Müller, P.H.; Neuman, P.; Storm, R. *Tafeln der Mathematischen Statistik*; VEB Fachbuchverlag: Leipzig, Germany, 1979; 272p.
44. Hakanson, L. Great Lakes—Form and function, as exemplified using data from Lake Vanern, the fourth largest lake in Europe. In *Great Lakes: Ecology, Management and Conservation*; Donovan, J.S., Ed.; Nova Science Publishers: New York, NY, USA, 2010; pp. 1–39.
45. Staehr, P.A.; Baastrup-Spohr, L.; Sand-Jensen, K.; Stedmon, C. Lake metabolism scales with lake morphometry and catchment conditions. *Aquat. Sci.* **2012**, *74*, 155–169. [CrossRef]
46. Ponizovskii, A.M. *Salt Resources of the Crimea*; Crimea: Simferopol, Ukraine, 1965; 166p. (In Russian)
47. Park, N.; Lee, J.; Chon, K.; Kang, H.; Cho, J. Investigating microbial activities of constructed wetlands with respect to nitrate and sulfate reduction. *Desalin. Water Treat.* **2009**, *1*, 172–179. [CrossRef]
48. Glinka, N.L. *General Chemistry*; Mir: Moscow, Russia, 1970; 636p. (In Russian)
49. Fanning, J.C. The solubilities of the alkali metal salts and the precipitation of Cs^+ from aqueous solution. *Coordin. Chem. Rev.* **1995**, *140*, 27–36. [CrossRef]
50. Fan, Q.H.; Tanaka, M.; Tanaka, K.; Sakaguchi, A.; Takahashi, Y. An EXAFS study on the effects of natural organic matter and the expandability of clay minerals on cesium adsorption and mobility. *Geochim. Cosmochim. Acta* **2014**, *135*, 49–65. [CrossRef]
51. Garnier, J.M.; Ciffroy, P.; Benyahya, L. Implications of short and long term (30 days) sorption on the desorption kinetic of trace metals (Cd, Zn, Co, Mn, Fe, Ag, Cs) associated with river suspended matter. *Sci. Total Environ.* **2006**, *366*, 350–360. [CrossRef] [PubMed]
52. Boulion, V.V.; Anokhin, L.E.; Arakelova, E.U. Primary production of the hyperhaline lakes in Crimea. *Proc. Zool. Inst.* **1989**, *205*, 14–25. (In Russian)

53. Shadrin, N.; Kolesnikova, E.; Revkova, T.; Latushkin, A.; Chepyzhenko, A.; Dyakov, N.; Anufriieva, E. Macrostructure of benthos along a salinity gradient: The case of Sivash Bay (the Sea of Azov), the largest hypersaline lagoon worldwide. *J. Sea Res.* **2019**, *154*, 101811. [CrossRef]
54. Timofeeva-Resovskaya, E.A. Distribution of radioisotopes in the main components of freshwater reservoirs. *Tr. Inst. Biol. Ural. Fil. AN SSSR* **1963**, *30*, 1–77. (In Russian)
55. Horne, R.A. *Marine Chemistry. The Structure of Water and the Chemistry of the Hydrosphere*; Wiley InterScience: New York, NY, USA, 1974; 568p.
56. Zheng, M. *Saline Lakes and Salt Basin Deposits in China*; Science Press: Beijing, China, 2014; 321p.
57. Anufriieva, E.V.; Shadrin, N.V. Morphometric variability of *Arctodiaptomus salinus* (Copepoda) in the Mediterranean-Black Sea region. *Zool. Res.* **2015**, *36*, 328–336.
58. RSS-99/2009. Radiation Safety Standards (NRB-99/2009): The Sanitary Rules and Regulations (SanR & R 2.6.1.2523–09): Approved. and Enter. in Force From 1 September 2009 to Replace SanR & R 2.6.1.758-99. Registered with the Ministry of Justice 14 August 2009, Reg. Number 14534. Available online: http://base.garant.ru/4188851/#1000 (accessed on 7 August 2018). (In Russian).