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Differences in Mercury Concentrations in Water and Hydrobionts of the Crimean Saline Lakes: Does Only Salinity Matter?

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Abstract: Of significant scientific and public concern is the high toxicity, significant bioaccumulation, and magnified concentration within the food web of mercury (Hg). Hg content both dissolved and in suspended forms in water as well as in biomass of different hydrobiont taxa was studied in 18 saline lakes in Crimea from 2012 to 2021. The impact of different factors (salinity, seasonality, anthropogenic activities, geological background, etc.) was analyzed. The generalization of data for all lakes showed that the average concentration of Hg in dissolved form was 129 ng L^{-1}, varying over a wide range. The content of Hg in total suspended substrates was an average of 151 ng L^{-1}, and the total content of Hg in lake water averaged 291 ng L^{-1}. Geological background and anthropogenic activities can determine the total Hg content in lakes. In most lakes, a significant positive correlation was noted between the concentration of one, two, or all three indicators of Hg content and the month of the year, with indicators increasing from winter through to autumn. When analyzing the entire data array, a significant positive correlation was found between the concentration of the suspended form of Hg and salinity; such correlation between the concentration of the dissolved form of Hg and salinity was absent. The highest average Hg concentrations in biomasses were noted in *Artemia* and were the lowest in plants. Geological background and human activity contribute to high Hg content in lakes. Hydrobionts can significantly influence the distribution and behavior of Hg, being an important factor of its cycle in the lakes.

Keywords: mercury; saline lakes; hydrobionts; filamentous algae; *Artemia*

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1. Introduction

Hg concentration has been increasing in the atmosphere, water bodies, and soil since the middle of the nineteenth century [1–4]. Today, various natural sources, such as volcanoes, are responsible for approximately half of the Hg emissions in the atmosphere, and the other half is human-generated [5]. Currently, human activities have produced Hg emissions at a rate 3 to 5 times higher than before the Industrial Revolution in the 1800s [4,6]. There is scientific and public concern regarding Hg due to its high toxicity, significant bioaccumulation, and magnification of concentrations during up the food web [7,8]. Organisms at higher trophic levels accumulate larger concentrations of Hg than lower ones [7,9]. Among different land ecosystems, lakes act as integrators of their watersheds and airsheds and accumulators of contaminants including Hg [9–11]. Direct atmospheric deposition and watershed runoff are the main sources of input Hg in lakes [11,12]. As an example, atmospheric deposition was the dominant Hg input source in Great Salt Lake (84% of a year's total input), and cumulative riverine Hg load was only 16% [12].

Lakes represent important habitats for fish and other valuable hydrobionts, which can accumulate Hg to risk levels for humans and birds [8,9]. Consequently, scientific attention is increasing on all issues of Hg distribution and accumulation in the lake ecosystem components as well as factors influencing these. Even though the total volume of saline lakes on the planet is close to that of fresh lakes, much less attention is paid to different issues of their study than in fresh lakes, including Hg behavior.

Saline lakes play an important natural role and provide people with a wide range of ecosystem services [13,14]. Among other things, they are important areas for nesting, foraging, wintering, and resting during migrations of a wide variety of birds [15,16]. In the hypersaline Great Salt Lake, there is a high Hg concentration, which creates risks for many birds [9]. As an example, Goldeneye ducks accumulate high Hg concentration due to a magnification of 270 times that found in food organisms [9]. The need for more economical use of freshwater leads to the fact that in recent years, the use of various saline and hypersaline lakes for the development of aquaculture has been expanding [17]. This dictates the need to intensify the study of the behavior of Hg in salt lakes with different salinity, both for the development of the ecosystem and biogeochemical concepts and for topical issues of the social and economic development of society, especially in arid regions.

In Crimea, the largest peninsula in the Black Sea, 50 relatively large lakes and many small saline lakes exist [14,18,19]. Lake Sasyk-Sivash has the largest surface area of 75 km^2. All lakes are shallow with a maximum depth of no more than 2 m. Salinity fluctuates in a wide range; the annual average salinity varies from 3 to 300 g L^{-1}; interannual differences are also considerable. The Crimea peninsula lies in the flight paths of important bird migration routes between Western and Eastern Palearctic, Africa, and Asia [15]. The Crimean saline lakes provide homes and food for many bird species nesting here or having a seasonal settlement of many thousands of migrating birds. *Artemia* and other crustaceans are among the main food items for many of them. Due to its diversity of lakes, Crimea is a good area to look for general patterns in Hg behavior within them. The distribution of Hg within the lakes has been studied since 2012, but only a small portion of obtained data was previously published [20,21].

The objectives of this study are: 1. to present new long-term study data; 2. to analyze a coupling of different factors in the determination of Hg behavior in saline lakes, and 3. to prove or disprove the assumptions that: (a) salinity is an important factor in Hg cycling but no one general pattern of this exists, and (b) the crustacean *Artemia* may be the most important factor of Hg cycling in Crimean saline lakes.

2. Materials and Methods

2.1. Study Area

Due to climate aridity, there are more than sixty saline and hypersaline lakes in Crimea, the largest peninsula in the Black Sea (the area is about 27,000 km^2 [18,19,22]. All lakes are shallow polymictic (depth < 2 m) and differ in area, depth, total salinity, geochemical peculiarities, biotic composition, and productivity [18,19,22–27]. Of them, 18 were studied in the period from 2013 to 2021 (Figure 1, Table 1).

2.2. Field Studies

In total, 88 samples of water, 25 samples of hydrobiont biomass, and 12 quantitative zooplankton samples were collected by wading. All methods used have been previously described in more detail [25,28], and they are given below. In total, 1 L water samples were taken by bathometer during every survey. At sampling sites, temperature, salinity, and pH were determined by a PHH-830 electronic thermometer (OMEGA Engineering, INC., Norwalk, CT, USA), a WZ212 refractometer (Kelilong Electron Co., Ltd., Fuan, Fujian, China), and a PHH-830 pH meter (METTLER TOLEDO, Greifensee, Switzerland), respectively. The concentration of total suspended solids (TSS) was assessed by the biophysical complex "Condor" (produced by Akvastandart-Yug, Sevastopol, Russia), which was applied in "point" mode [25]. Zooplankton quantitative samples were taken by filtration of 50–100 L of water through a plankton net (a mesh size of 110 microns). Zooplankton samples were fixed with 4% formalin to identify animal species and to calculate their abundance.

Table 1. The studied Crimean saline lakes and their characteristics.

Lake	Coordinates of the Sampling Sites	Area, km^2	The Total Period of Study, and the Number of Sampling Times	Ranges of Changes in the Environmental Characteristics during the Study Period			
				Salinity, g L^{-1}	Temperature, °C	pH	TSS, g L^{-1}
Perekop lake group							
Kiyatskoe	4,600,107 N 3,394,477 E	12.5	2012–2021, 10 times	78–310	4–41	7.2–8.2	0.02–2.77
Kirleutskoe	4,595,895 N 3,402,173 E	21.0	2012–2021, 11 times	190–318	5–38	6.0–8.2	0.02–0.77
Krasnoe	4,598,811 N 3,388,328 E	23.4	2017–2018, 2 times	340–350	30–31	9.5–9.6	0.14–0.22
Tarkhankut lake group							
Bakalskoe	4,573,171 N 3,318,016 E	8.0	2013–2021, 7 times	18–65	6–26	7.0–9.5	0.002–0.01
Donuzlav	4,544,084 N 3,319,460 E	48.2	2013–2014, 4 times	16–17	5–29	8.6–8.7	0.002–0.01
Dzharylgach	4,557,029 N 3,285,658 E	8.0	2014–2021, 9 times	110–188	15–30	6.0–8.5	0.02–0.25
Yarylgach	4,556,690 N 3,285,635 E	1.6	2019–2021, 4 times	70–135	15–32	7.5–8.3	0.03–0.11
Evpatoria lake group							
Kyzyl-Yar	4,505,731 N 3,363,034 E	8.0	2014–2021, 9 times	2.5–9	12–30	6.0–9.1	0.005–0.29
Moynaki	4,518,052 N 3,331,621 E	1.8	2020, 1 time	62	28	8.0	0.12
Sasyk-Sivash	4,515,406 N 3,351,261 E	75.3	2016–2021, 7 times	260–350	13–35	6.2–7.8	0.27–1.41
Novofedorovskoe	4,508,797 N 3,356,268 E	0.2	2021, 1 time	4	15	8.0	0.03
Kerch lake group							
Uzunlarskoe	4,504,601 N 3,611,014 E	20.0	2014, 1 time	390	31	7.4	0.58
Koyashskoe	4,505,137 N 3,619,191 E	5.5	2014–2021, 4 times	245–370	19–39	7.0–8.6	0.04–1.10
Tobechikskoe	4,518,943 N 3,638,163 E	19.0	2016–2021, 4 times	174–364	24–32	7.2–8.3	0.35–4.78
Aktashskoe	4,538,562 N 3,583,383 E	25.0	2016–2021, 5 times	88–220	18–29	6.5–8.5	0.08–0.69
Chokrakskoe	4,546,408 N 3,630,907 E	8.5	2014–2021, 5 times	245–420	22–32	6.6–7.9	0.15–2.68
Feodosia lake group							
Adzhigol	4,510,577 N 3,546,112 E	0.8	2018–2021, 3 times	50–120	25–32	7.0–8.7	0.06–0.34
Kuchuk-Adzhigol	4,509,982 N 3,544,860 E	0.6	2018–2021, 3 times	3–5	17–31	8.4–9.4	0.001–0.03

Note: The data for 2012–2018 were obtained by the staff of the Department of Radiation and Chemical Biology of the IBSS with the support of the RFBR grant 16-05-00134 A (Mirzoyeva et al., 2015; Stetsiuk et al., 2018); TSS is the total suspended solids.

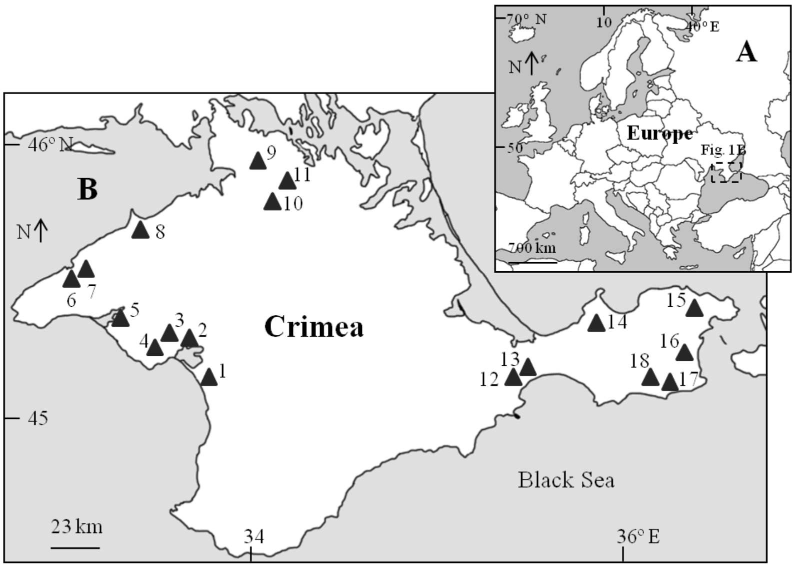

Figure 1. Distribution of the studied saline lakes in Crimea (1—Novofedorovskoe, 2—Kyzyl-Yar, 3—Sasyk-Sivash, 4—Moynaki, 5—Donuzlav, 6—Yarylgach, 7—Dzharylgach, 8—Bakalskoe, 9—Krasnoe, 10—Kirleutskoe, 11—Kiyatskoe, 12—Kuchuk-Adzhigol, 13—Adzhigol, 14—Aktashskoe, 15—Chokrakskoe, 16—Tobechikskoe, 17—Koyashskoe, 18—Uzunlarskoe).

2.3. Laboratory Studies

For separation of the forms of Hg, water samples were filtered through nuclear 0.45-µmpore size filters, which were pre-weighed by an analytical balance VIBRA HT 224RCE (Shinko Denshi Co., Itabashi, Tokyo, Japan), and water was fixed by concentrated nitric acid (10 mL of HNO_3 per 1 L of water) [21,25]. The dissolved Hg form was evaluated in the filtered solution, and the Hg suspended form was determined on the filters. Potassium permanganate solution was added to the filtered water for oxidation in a volume of 15–20 mL and then 5 mL 1/2 sulfuric acid, reducing all dissolved Hg forms to ions. Solids on the filters were acid burned (10 mL 1/2H_2SO_4 and 5 mL HNO_3 per sample). When analyzing wet biomass of hydrobionts, two drops of H_2O_2 were added instead of HNO_3. In the next stage, the samples were reduced at no more than 60 °C and then cooled. A potassium permanganate solution was added into the samples in an amount ensuring complete oxidation of the analyzed sample (from 15 to 20 mL), and the samples were filtered after 10–15 min. Before chemical analyses on a device, 5 mL sulfuric acid (1:1) was added to 100 mL of the filtered sample. A hydroxylamine solution of up to 5 mL was put into all samples to remove excess potassium permanganate. For chemical analyses, 10 mL of a reducing agent ($SnCl_2$) was added. After these manipulations, prepared samples were analyzed by flameless atomic absorption on a Hiranuma-1 mercury analyzer (Hiranuma Sangyo Co., Ltd., Mito, Ibaraki, Japan). The Hg concentration was determined at a wavelength of 253.7 nm. For calibration of the Hiranuma-1 mercury analyzer, a standard solution of mercury (II) ions was used. At the first stage, a 'blank calibration' was done (100 mL of distilled water + 5 mL of H_2SO_4 (1:1)), and then calibration with a series of calibration solutions: 0.2; 0.4; 0.6; 0.8; 1 µg dm^{-3} (10 replicates for each concentration). The Hiranuma-1 sensitivity was 0.01 with the detection limit of 0.5 ng L^{-1} of Hg. To determine Hg in hydrobionts, their wet biomass samples were processed in the same way as described above for suspended matter. All obtained values were re-calculated for dry biomass with known coefficients (% dry mass in wet biomass): for *Artemia*—11%, *Artemia* cysts—40%; *Cladophora* and *Polysiphonia*—16%, *Ruppia*—15%, *Potamogeton*, *Stuckenia* and *Myriophyllum*—11%. The zooplankton samples were processed using Olympus SZ-ST (Olympus, Tokyo, Japan) and LOMO MBS-9 (LOMO, St. Petersburg, Russia) stereo microscopes.

Coefficients of Hg accumulation by total suspended solids (TSS), also known as distribution coefficient, or by hydrobiont biomass (bioconcentration factor) may be calculated [29,30]:

$$K_{sus(biom)} = Hg_{sus(biom)}/Hg_{dis} \quad (1)$$

where $K_{sus(biom)}$ is a coefficient of Hg accumulation by TSS (or biomass), L kg^{-1}; $Hg_{sus(biom)}$ is the Hg concentration in TSS (or biomass), ng kg^{-1} (dry weight); Hg_{dis} is the concentration of Hg in the dissolved phase, ng L^{-1}.

All results were subjected to statistical processing in MS Excel 2007 and Statistica 6.0. Mean values, standard deviations (SD), and coefficients of variability (CV) were calculated as well as parameters of regression equations, coefficients of correlation ®, and determination (R^2). After normality tests [31], the significance of differences between mean values of the data subsets was assessed by Student's t-test. The confidence levels (p) for the correlation coefficients were evaluated [32].

3. Results

3.1. Mercury in Water

As can be seen from Table 1, the physicochemical parameters of the lakes varied within a very wide range: the total range of salinity in the studied lakes was from 2.5 to 420 g L^{-1}, temperatures varied from 4 to 41 °C, pH from 6 to 9.5, and the concentration of TSS in the water from 0.001 to 4.78 g L^{-1}. When analyzing the entire array of data obtained (Figure 2a), a significant positive correlation was found between salinity and total suspended solids concentration (R = 0.780, p = 0.0001), and the dependence is approximated by an exponential equation:

$$TSS = 0.021e^{0.01S} \quad (2)$$

where TSS is total suspended solids, mg L^{-1}; S is salinity, g L^{-1}.

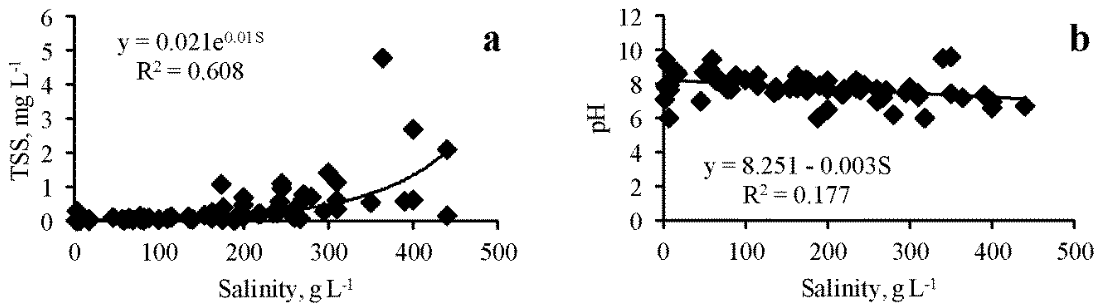

Figure 2. Dependence of the total suspended solids (TSS) and pH on salinity in the Crimean saline lakes ((**a**)—TSS dependence, (**b**)—pH dependence).

When analyzing the relationship between the temporal variability of these two parameters in individual lakes, their correlation as a rule, is even stronger, as in Lake Kiyatskoe (R = 0.872, p = 0.001),for example, although in some other lakes, the relationship between these values was unreliable. When analyzing the entire data array (Figure 2b), a significant negative dependence of pH on salinity was noted (R = 0.420, p = 0.0005):

$$pH = 8.251 - 0.003S \quad (3)$$

Table 2 shows rather high spatial and temporal variability of Hg forms in Crimean saline lakes. If the one outlier with a very high value (3635 ng L^{-1}, Lake Kyzyl-Yar, 11/18/2014) is excluded, then generalization for all lakes shows that the average concentration of Hg in dissolved form was 129 ng L^{-1}, varying over a wide range (coefficient of variation, CV = 0.983). The content of Hg in TSS (excluding the value of 3150 ng L^{-1}, Lake Chokrak, 08/17/2017) was an average of 151 ng L^{-1} (CV = 1.020), and the total content of Hg in lake water, excluding the above cases, averaged 323 ng L^{-1} (CV = 1.230). Of these three indicators, the concentration of dissolved Hg was the least variable, and that of total Hg was the most variable.

Table 2. The dissolved and suspended Hg content in the Crimean saline lakes.

Lake	Dissolved Mercury Content (Hg_{dis}), ng L^{-1}			Suspended Mercury Content (Hg_{sus}), ng L^{-1}			Total Mercury Content in Water (Hg_t), ng L^{-1}			Proportion Hg_{dis}/Hg_{tss}	Hg Concentration in TSS, ng g^{-1}	Coefficient of Hg Accumulation by TSS (K_{sus}), 10^3 L kg^{-1}
	Average	min/max	CV	Average	min/max	CV	Average	min/max	CV	Average/CV	Average/CV	Average/CV
Perekop lake group												
Kiyatskoe	143.7	11/400	0.911	167.2	3/450	0.850	325.7	20/850	0.694	0.441/0.489	655.0/0.721	6.3/0.733
Kirleutskoe	109.8	7/350	1.093	74.7	5/200	0.844	184.5	19/550	0.938	0.595/0.370	987.1/1.259	10.3/0.938
Krasnoe	145.0	140/150	0.049	300.0	200/400	0.471	445	350/540	0.302	0.326/0.401	1629.6/0.157	11.3/0.204
Group average	132.8	52.7/300	0.150	180.6	69/350	0.627	318.4	129.7/646.75	0.410	0.454/0.297	1090.6/0.454	9.3/0.288
Tarkhankut lake group												
Bakalskoe	117.8	55/238	0.594	93.5	3/300	1.213	226.4	63/460	0.639	0.520/0.511	1944.7/0.657	19.7/0.640
Donuzlav	195.3	70/373	0.810	23.4	2/79	1.584	225.9	76/380	0.673	0.085/0.322	3064.4/0.819	27.8/0.678
Dzharylgach	180.4	20/550	1.014	139.0	45/300	0.656	319.5	70/700	0.702	0.565/0.399	2960.9/1.276	29.2/0.925
Yarylgach	40.0	20/50	0.433	46.7	30/70	0.446	86.7	80/90	0.067	0.461/0.449	1372.4/0.894	54.4/1.289
Group average	133.4	40/195	0.530	75.7	23/139	0.678	214.6	75/390	0.447	0.408/0.538	2335.6/0.350	32.8/0.458
Evpatoria lake group												
Kyzyl-Yar	518.6 * (129.1 **)	10/500	2.271 * (1.230 **)	144.5	4/320	0.726	663.1 * (278.4 **)	50/3740 * (700 **)	1.770 * (0.815 **)	0.464/0.725	5224.6/0.910	92.4/1.355
Moynaki	70.0	-	-	50.0	-	-	120.0	-	-	0.583	431.0	6.2
Sasyk-Sivash	82.5	50/150	0.478	155.0	50/300	0.630	237.5	105/450	0.557	0.347/0.299	345.31.145	3.7/0.763
Novofedorovskoe	20.0	-	-	50.0	-	-	70.0	-	-	0.286	1470.6	73.5
Group average	75.4	30/325	0.413	99.9	27/310	0.548	176.5	77/575	0.554	0.420/0.313	1868/1.229	44.0/1.040
Kerch lake group												
Uzunlarskoe	83.0			769.0			852			0.097	1332.8	16.1
Koyashskoe	41.0	20/63	0.525	202.5	30/540	1.472	241.0	50/603	1.301	0.170.664	722.2/0.284	19.5/0.271
Tobechikskoe	265.0	70/450	0.710	202.5	80/250	0.410	467.5	150/680	0.532	0.567/0.324	358.7/0.963	1.9/1.160
Aktashskoe	174.0	20/500	1.119	202.0	30/400	0.756	376.0	50/700	0.731	0.463/0.521	1478.1/1.091	12.6/1.174
Chokrakskoe	126.0	55/200	0.474	851.6	70/3150	1.535	977.6	220/3300	1.344	0.129/0.775	917.9/0.835	9.2/1.110
Group average	137.8	41/296	0.629	446.4	52/1085	0.752	583.0	117.5/1321	0.544	0.285/0.752	961.9/0.473	11.9/0.057
Feodosia lake group												
Adzhigol	87.5	50/125	0.606	275.0	50/500	1.157	362.5	100/625	1.024	0.241/0.601	1130.0/0.405	13.9/0.229
Kuchuk-Adzhigol	66.7	50/100	0.433	153.3	20/400	1.395	220.0	70/450	0.919	0.303/0.659	1254.9 ***/ 0.089	19.2 ***/ 1.720
Group average	77.1	50/112.5	0.191	214.2	153/275	0.402	292.6	220/365	0.351	0.272/0.161	1192.4 ***/ 0.236	16.6 **/ 0.548
Total average	119.2 **	10.5/500 **	0.983	151.2	2/688 **	1.020	322.7	19.1/850 **	1.231	0.369/0.470	1592.5 ***/ 1.615	22.9 ***/ 2.032

Note: TSS is the total suspended solids; *—with the highest value (3634.6 ng L^{-1}, Lake Kyzyl-Yar, 11/18/2014); **—without the highest value (3634.6 ng L^{-1}, Lake Kyzyl-Yar, 11/18/2014); ***—with excluding the anomaly highest value (400,000 ng g^{-1}, Lake Kuchuk-Adzhigol, 04/26/2018).

The coefficients of variation (CV) of the concentration of the dissolved form of Hg, as well as others, in one lake are indicators of the range of temporal variability of the parameters under consideration, and all other CVs characterize spatial variability. Based on the comparison of CV values (Table 2), it can be concluded that the spatial and temporal variability for all indicators is close. Comparison of variability between lakes within a group and between groups shows that they are approximately of the same order (Table 2). At the same time, the average indicators differ for lakes of different groups. According to the average concentration, lake groups are divided into two clusters (Table 2). The average concentration of dissolved forms of Hg does not differ significantly between the

Perekop, Tarkhankut, and Kerch lake groups, averaging 135 ng L^{-1} (CV = 0.201), and in the Evpatoria and Feodosia groups, on average 76 ng L^{-1} (CV = 0.164). In terms of suspended and total Hg, the differences between all lake groups are more pronounced (Table 2). On average, the lowest concentration of suspended forms of Hg was noted in the Tarkhankut lake group, and in the Evpatoria group, it is slightly higher (Table 2), but the differences are not statistically significant. In the Perekop and Feodosiya groups of lakes, it is significantly higher (p = 0.01) by two times (Table 2). The highest concentration of suspended forms of Hg was noted in the lakes of the Kerch group (Table 2), on average 554 ng L^{-1} (CV = 0.752). The total average content of Hg in the water of different groups of lakes varied from 177 ng L^{-1} (CV = 0.554) in the Evpatoria lake group to 583 ng L^{-1} (CV = 0.544) in the Kerch group (Table 2). The average proportion of dissolved forms in the total Hg content in water differed little in lakes of different groups, from 27% (CV = 0.161) and 29% (CV = 0.752) in the Feodosia and Kerch lake groups, respectively, to 45% (CV = 0.401) in the Perekop group (Table 2). The coefficient of Hg accumulation in suspension averaged 24,523 (CV = 2.053), ranging from 9341 (CV = 0.291) in the Perekop group of lakes to 44,321 (CV = 1.042) in the Evpatoria group (Table 2). The Kerch and Perekop groups of lakes do not differ significantly in this parameter, as do the Evpatoria and Tarkhankut groups of lakes, but these two pairs of groups differ significantly from each other (p = 0.005).

The data series obtained for several lakes (Table 1) made it possible to analyze seasonal changes in various forms of Hg in these lakes. In most lakes, a significant positive correlation was noted between the concentration of one, two, or all three indicators of Hg content and the season of the month, i.e., indicators increased from winter to autumn. In particular, for dissolved forms, this was noted for lakes Kiyatskoe (R = 0.710, p = 0.008) (Figure 3a) and Bakalskoe (R = 0.896, p = 0.007) (Figure 3b), as well as Sasyk-Sivash (R = 0.736, p = 0.05) and Tobechikskoe (R = 0.996, p = 0.005). There was no dependence in lakes Dzharylgach and Kyzyl-Yar. There were significant seasonal changes in the Hg total content and in suspended form in the lakes Kiyatskoe and Kirleutskoe (Figure 4).

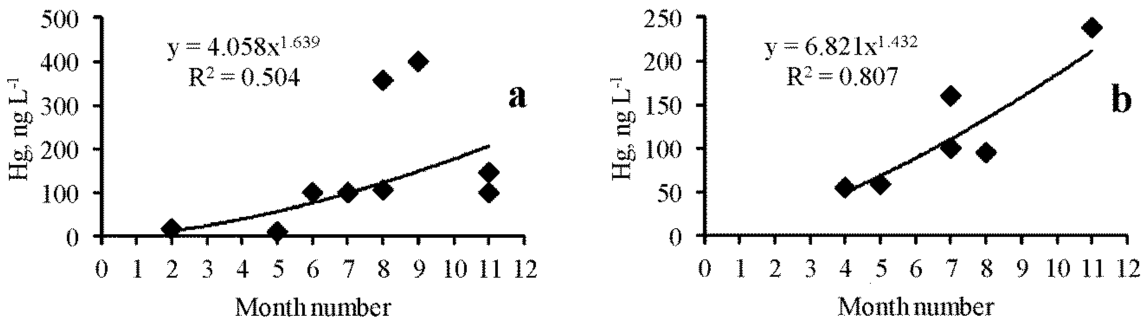

Figure 3. Seasonal changes in the concentration of the Hg dissolved forms of mercury in the lakes Kiyatskoe (**a**) and Bakalskoe (**b**).

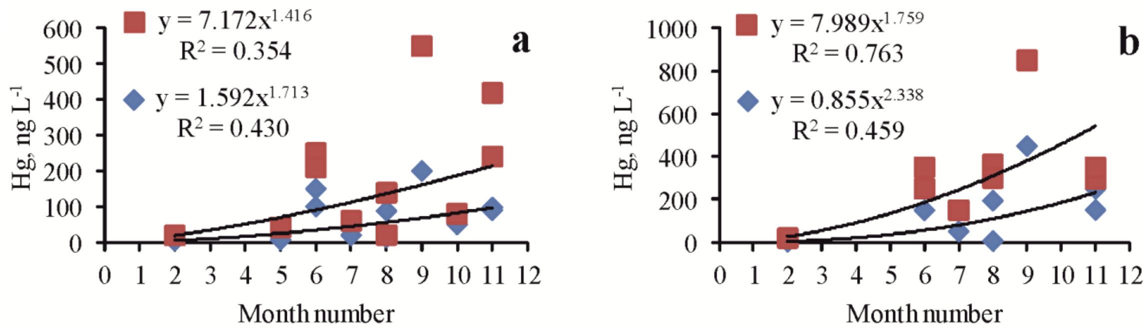

Figure 4. Seasonal changes in the Hg total content (red squares) and suspended form (blue rhombus) in the lakes Kiyatskoe (**a**) and Kirleutskoe (**b**).

The concentrations of Hg dissolved and in suspended forms do not significantly correlate with each other in the total data set; however, in some lakes, a significant positive correlation was observed, as, for example, in Lake Bakalskoe (R = 0.840, p = 0.01), Lake Kirleutskoe (R = 0.805, p = 0.01), Kyzyl-Yar (R = 0.755, p = 0.02), Sasyk-Sivash (R = 0.831, p = 0.01), and others. In some lakes, for example, Dzharylgach or Akhtashskoe, this dependence did not exist.

When analyzing the entire data array, no correlation was found between the concentration of the dissolved form of Hg and salinity. However, for some lakes, such a significant positive relationship was found, for example, in Lake Bakalskoe (R = 0.839, p = 0.01), but in most lakes there was no such relationship. When analyzing the entire data array (Figure 5a), a significant positive correlation was found between the concentration of the suspended form of Hg and salinity (R = 0.581, p = 0.0005):

$$Hg_{sus} = 105.72 + 0.003S^2 - 0.492S \quad (4)$$

where Hg_{sus} is Hg content in TSS, ng L^{-1}.

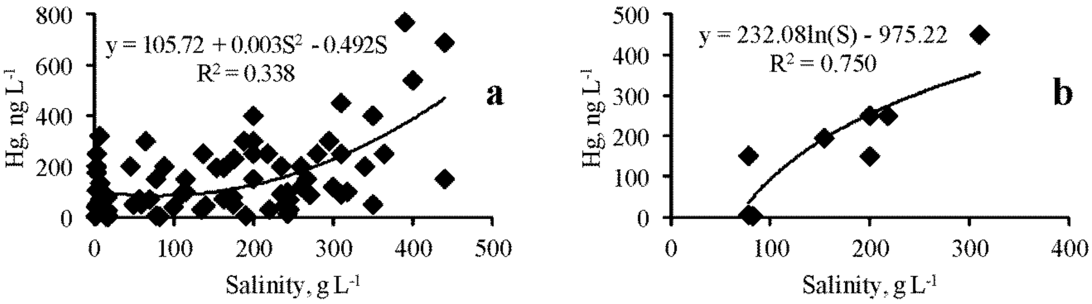

Figure 5. Dependence of the Hg suspended form concentration on salinity ((**a**)—for the entire collection of lakes in general, (**b**)—Lake Kiyatskoye).

In some lakes, this dependence was more pronounced than in the total data set (Figure 5b), for example, in lakes Kiyatskoe (R = 0.866, p = 0.0001), Bakalskoe (R = 0.839, p = 0.005), Kirleutskoe (R = 0.723, p = 0.01). In other lakes, for example, Dzharylgach, Sasyk-Sivash, Tobechikskoe, and Akhtashskoe, there was no such significant dependence. There was no correlation between the concentration of the dissolved form of Hg in water and TSS. Between the concentration of the suspended form of Hg and TSS, a positive significant correlation was noted when analyzing the entire data set (R = 0.450, p = 0.0005), which can be approximated by the equation:

$$Hg_{sus} = 182.47\ TSS^{0.361} \quad (5)$$

When analyzing this dependence in individual lakes, it was not found in several lakes; in others, only a similar trend was noted. A significant correlation was noted only in Lake Bakalskoe (R = 0.821, p = 0.004):

$$Hg_{sus} = 2785.8\ TSS - 20.9 \quad (6)$$

There was no significant correlation between the proportion of the dissolved form of Hg in its total content and salinity in the analysis of the total data set, but it was significantly manifested in most lakes. A significant negative dependence was noted, for example, in lakes Kiyatskoe (R = −0.715, p = 0.04), Kirleutskoe (R = −0.715, p = 0.04), Bakalskoe (R = −0.869, p = 0.007), and Akhtashskoe (R = −0.867, p = 0.008). In Lake Sasyk-Sivash, a significant positive correlation was noted between these parameters (R = 0.883, p = 0.005), whereas in Lake Dzharylgach, there was no correlation at all. The parameters of the approximating equations for different lakes differ significantly. There was no correlation between the proportion of the dissolved form of Hg in its total content and TSS in the analysis of all data and individual lakes, except for Lake Bakalskoe, where a significant negative relationship was noted (R = 0.798, p = 0.007):

$$Hg_{dis}/Hg_t = 0.963e^{-11.75TSS} \quad (7)$$

where Hg_t is a total Hg, ng L^{-1}.

When analyzing the total sample, a significant negative correlation was found between the coefficient of Hg accumulation in suspended matter and TSS (R = −0.483, p = 0.0005):

$$K_{sus} = 3134\ TSS - 1451 \quad (8)$$

where K_{sus} is the coefficient of Hg accumulation by TSS, L kg^{-1}.

A similar dependence as (8), expressed to varying degrees, was also noted in individual lakes. When analyzing the total data set, a weak but significant negative correlation was found between the coefficient of Hg accumulation in TSS and salinity (R = −0.425, p = 0.0005):

$$K_{sus} = 61{,}300 - 8710\ \ln(S) \quad (9)$$

In some lakes, in the relationship of these parameters, both a positive and a negative trend and its absence were noted, but it was not significant. A significant positive dependence was noted only in Lake Bakalskoe (R = 0.961, p = 0.0005):

$$K_{sus} = 65{,}301 - 8710\ \ln(S) \quad (10)$$

The concentration of Hg in suspended matter was variable, ranging from 343 to 7269.6 ng g^{-1} (dry weight) (Table 2), excluding the anomalous highest value (400,000 ng g^{-1}, Lake Kuchuk-Adzhigol, 04/26/2018). Graphical analysis showed that this value varies from month to month (Figure 3). However, the average concentration for all lakes does not change significantly, but the maximum values change significantly ($p < 0.005$) (except for two anomalously low values in September and October, when there were only 2–3 points) (Figure 3, Table 3).

Table 3. The concentration of Hg in the suspended matter of the Crimean saline lakes in different months of the year.

Month	Hg Concentration in TSS, ng g^{-1}				
	Minimum	**Maximum**	**Average**	**CV**	**The Number of Measurings**
January	-	-	-	-	-
February	166	1238	563	0.880	4
March	-	-	-	-	-
April	723	3846	2004	0.504	7
May	80	6136	1472	1.282	9
June	73	4000	1239	0.846	19
July	18	12,500	3289	1.369	10
August	24	12,500	2302	1.433	13
September	3390	739	2064	0.908	2
October	211	1786	754	1.185	3
November	85	23,622	2636	2.410	13
December	-	-	-	-	-

Note: TSS is the total suspended solids.

3.2. Hg in Hydrobionts

The obtained data on the content of Hg in the biomass of filamentous green algae, vascular plants, and *Artemia* (Crustacea, Anostraca) are presented in Table 4. The highest average concentrations of Hg in the biomass were noted in *Artemia* (average 111 ng g^{-1} (dry weight), CV = 1.074), and the lowest in plants (average 22 ng g^{-1}, CV = 0.641). In plants, the average concentration of Hg in the salinity range from 2.4 to 17.0 g L^{-1} was 33.2 ng g^{-1} (CV = 0.432), and in the salinity range from 64 to 89 g L^{-1}, it was 11.3 ng g^{-1} (CV = 0.720). The differences are statistically significant (p = 0.01). At the same time, at low salinities (from 64 to 89 g L^{-1}), a significant positive correlation of Hg concentration with salinity was found (R = 0.906, p = 0.01):

$$Hg_{plant} = 17.014 + 1.815\ S \tag{11}$$

where Hg_{plant} is Hg content in plant biomass, ng g^{-1} (dry weight).

Table 4. The concentration of Hg in hydrobiont biomass in the Crimean saline lakes *.

Taxon	Lake	Date	Hg Content in Dry Biomass, ng g^{-1}	The Coefficient of Hg Accumulation by Biomass, L g^{-1}	Total ** *Artemia* Population Abundance Ind. L^{-1}/ Wet Biomass mg L^{-1}	Salinity, g L^{-1}
Vascular plants (Embryophytes)						
Stuckenia pectinata (L.) Böerner (*Potamogeton pectinatus* L.)	Kiyatskoe	19.02.2013	2.5	0.14		83
	Donuzlav	19.02.2013	51.3	-		16
	Donuzlav	10.08.2014	42.7	0.30		15
	Kuchuk-Adzhigol	01.07.2021	14.6	9.10		3
Potamogeton crispus L.	Kyzyl-Yar	13.05.2014	26.4	0.42		3
Average/CV for *Potamogeton*			27.5/0.725	2.49/1.770		
Ruppia maritime L.	Moynaki	26.06.2020	12.7	0.18		64
	Sivash	16.09.2020	18.7	0.93		89
Average/CV for *Ruppia*			15.7/0.270	0.56/0.956		
Myriophyllum verticillatum L.	Kyzyl-Yar	13.06.2019	30.9	3.10		6
Average/CV for vascular plants			22.0/0.744	2.02/6.25		
Green filamentous algae (Chlorophyceae)						
Cladophora spp.	Kiyatskoe	19.02.2013	10.0	0.10		83
	Kiyatskoe	13.05.2014	7.2	0.68		83
	Bakalskoe	22.08.2018	137.5	1.38		46
	Dzharylgach	17.06.2020	38.1	0.54		140
	Sivash	17.09.2020	20.0	1.00		86
Average/CV for *Cladophora*			42.6/1.279	0.74/0.652		
Polysiphonia spp.	Bakalskoe	13.05.2014	6.2	0.10		18
	The Black Sea	13.05.2014	3.7	0.04		18
	Bakalskoe	10.08.2014	14.7	0.15		19
	Bakalskoe	18.11.2014	13.8	0.14		45
	Bakalskoe	14.07.2017	50.7	0.32		26
	Donuzlav	10.08.2014	5.1	0.05		17
Average/CV for *Polysiphonia*			7.2/0.628	0.08/0.547		
Average/CV for green filamentous algae			4.13/1.680	0.45/1.151		

Table 4. *Cont.*

Taxon	Lake	Date	Hg Content in Dry Biomass, ng g^{-1}	The Coefficient of Hg Accumulation by Biomass, L g^{-1}	Total ** *Artemia* Population Abundance Ind. L^{-1}/ Wet Biomass mg L^{-1}	Salinity, g L^{-1}
			Animals (*Artemia*)			
Artemia spp., cysts	Kirleutskoe	22.06.2017	96.7	1.16	12.6/24.0	235
Artemia spp., adults	Aktashskoe	29.08.2017	29.1	1.45	179.5/18.5	89
	Dzharylgach	14.07.2017	83.6	0.76	240.0/14.6	137
	Dzharylgach	22.08.2018	318.2	0.80	6.3/2.5	188
	Aktashskoe	06.11.2019	38.6	1.93	20.7/0.6	220
	Dzharylgach	17.06.2020	85.5	1.22	33.0/25.9	140
Average for *Artemia*/CV			111.0/1.069	1.22/0.357	82.0/14.4	

Note: * The data for 2012–2018 were obtained by the staff of the Department of Radiation and Chemical Biology of the IBSS with the support of the RFBR grant 16-05-00134 A (Mirzoyeva et al., 2015; Stetsiuk et al., 2018); ** including all stages (with cysts); CV—coefficient of variation.

In general, the dependence of Hg concentration in biomasses on salinity was most likely dome-shaped. The average concentration of Hg in green filamentous algae was significantly different in *Cladophora* and *Polysiphonia* ($p = 0.01$); in *Cladophora,* it was about six times higher. However, the algae existed in different salinity ranges. When the data for both genera of filamentous green algae were analyzed together, a significant positive correlation was found between the content of Hg in biomass and salinity (Figure 6). Among all the studied organisms, the highest Hg accumulation coefficients in biomass were also noted in plants (average 1.80, CV = 0.462), and the lowest in filamentous green algae (average 0.45, CV = 1.933).

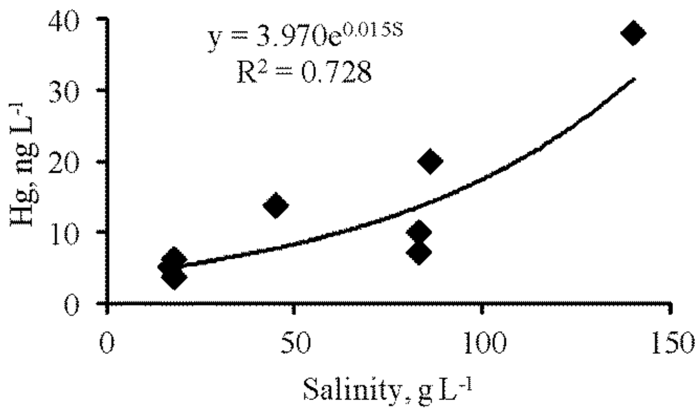

Figure 6. Dependence of Hg concentration in the biomass of filamentous green algae on salinity in Crimean saline lakes.

There was no significant correlation between the Hg concentration in the *Polysiphonia* biomass and salinity; a negative significant correlation was found in *Cladophora* (R = −0.972, $p = 0.001$):

$$Hg_{clad} = 422.6e^{-0.889S} \quad (12)$$

where Hg_{clad} is Hg content in *Cladophora* biomass, ng g^{-1} (dry weight).

For Potamogetonaceae (*Potamogeton* + *Stuckenia*), a significant negative correlation was also found between the concentration of Hg in the biomass and salinity (R = −0.824, $p = 0.05$):

$$Hg_{pot} = 38.12e^{-0.032S} \quad (13)$$

where Hg_{pot} is Hg content in Potamogetonaceae plant biomass, ng g^{-1} (dry weight).

A significant correlation between the concentration of Hg in the plant and algal biomass and the Hg content of the dissolved form in water was not found.

In the salinity range from 88 to 188 g L^{-1} in adult *Artemia*, a significant positive dependence of Hg body concentration with salinity was found (R = 0.996, p = 0.01):

$$Hg_{art} = 0.357e^{0.024S} \quad (14)$$

where Hg_{art} is Hg content in brine shrimp biomass, ng g^{-1} (dry weight).

At higher salinity, judging by the data in Table 4, there was a sharp decrease in the Hg concentration in the body, but the conclusion is preliminary as there is little data. The concentration of Hg in *Artemia* cysts is higher than in the adult *Artemia* (Table 4). The Hg accumulation coefficient did not show a significant correlation with salinity. There was a significant positive correlation between the Hg concentration in *Artemia* and the concentration of its dissolved form in water (R = 0.849, p = 0.03):

$$Hg_{art} = 0.350 + 0.074\ Hg_{dis} \quad (15)$$

A similar correlation between the Hg concentration in biomass of adult brine shrimp with the total content of Hg in water and the concentration of its suspended form was not found.

The concentration of Hg in the biomass of *Artemia* was lower than its concentration in the suspended matter by an average of 188.7 times (CV = 0.844). The concentration of Hg in *Artemia* biomass with its concentration in the particulate matter was positive and significant (R = 0.970, p = 0.003):

$$Hg_{art} = 3.178 + 0.002\ Hg_{sus}/TSS \quad (16)$$

A significant negative correlation was also found between the coefficients of Hg accumulation by suspended matter and *Artemia* biomass (R = −0.915, p = 0.001):

$$K_{art} = 1963e^{-0.022Ksus} \quad (17)$$

where K_{art} is the coefficient of Hg accumulation by *Artemia* biomass, L kg^{-1}.

4. Discussion

Comparison of the obtained values with data on other water bodies [11,33] shows that the new data obtained do not go beyond the general limits of previously determined values. At the same time, they are quite high. For example, Great Salt Lake, being the lake with the highest concentrations of total Hg in water in the USA, has an average Hg concentration of about 45 ng L^{-1} with maximum values < 100 ng L^{-1} [34,35], which is significantly lower than the average concentration for all saline lakes in Crimea and each separately (Table 2). Thus, it can be assumed that the saline lakes in Crimea have a large amount of Hg in the water. As in our studies, the absence of a significant correlation between the concentrations of dissolved and suspended Hg fractions was previously noted, and this may indicate that the suspended form of Hg is not in chemical equilibrium with dissolved one [36].

The Hg concentration in particulate matter increased from spring to autumn (Table 3), which is probably associated with an increase in the proportion of organic matter in TSS due to primary productivity. This is consistent with the fact, as previously shown in the Crimean saline lakes [21], that the concentration of Hg in the suspended matter (ng g^{-1}) significantly directly correlates with the level of primary production in the lake (R = 0.907, p = 0.0001). Based on the value of the coefficient of determination (R^2 = 0.822), it can be concluded that the variability of the Hg concentration in TSS was determined at 82% by primary productivity in the lakes.

Differences in Hg content in different lake groups are determined first of all by the geological features of their drainage basins, as was also noted in other regions [1,11,37]. First of all, one must take into account the rocks and products of their weathering composition as well as the chemical composition of soils and underground waters. The highest total Hg content in the lakes of the Kerch Peninsula, as previously shown for other trace elements [24,38], is determined by several geological factors. The geological background of the Kerch Peninsula includes the massive deposits of Fe-Mn ore as well as a lot of sulfates in groundwater and soils [24,38,39] that can significantly impact Hg behavior. Many mud volcanoes exist in the Kerch peninsula around saline lakes and even on their bottom [40,41]. They can significantly affect the geochemical background of saline lakes, due to the liquid phase from the volcanoes' vents containing high concentrations of different elements including Hg [24,40,41]. In clay from mud volcanoes, Hg is 10–100 times more concentrated than in the upper part of the continental crust [42].

Anthropogenic local and/or global factors can also play a big role in this [34,43]. This, in particular, in the opinion of the authors, is especially significant when considering the lakes of the spatially closely situated Perekop and Tarkhankut lake groups. There is a large chemical industry center in the North part of Crimea, and three lakes of the Perekop lake group are situated from 5 to 15 km from the biggest plant of the center. Possibly due to this, the highest Hg concentrations were observed in those lakes. Lake Krasnoe is used as a reservoir for the toxic effluents of this plant; because of this, it contains not only high concentrations of Hg but several other trace elements [24]. Lake Kiyatskoe is connected with Lake Krasnoe by a narrow channel and by filtration. It is less impacted by the chemical plant, and the total Hg concentration within it is 1.4 times lower than in the brine of Lake Krasnoe (Table 2). The distance between Lake Kiyatskoe and the nearest Lake Kirleutskoe is not more than 2 km and showed a total Hg concentration lower by 1.8 times than in Lake Kiyatskoe. The lakes of the Tarkhankut group are located at a distance of 50 km (Bakalskoe) to 86 km (Yarylgach) from the chemical industry center. It can be assumed that atmospheric emissions from the center still reach them, but with a lower concentration of Hg. Differences in the content of Hg between the lakes of the group may be determined by local anthropogenic impacts (agricultural, villages, and tourism). The impact of people on lakes is minimal in the case of Lake Yarylgach, where the minimum total amount of Hg in water was noted for this lake group. Significantly higher than the background for the region, indicators of Hg content in water are determined, as can be seen from the example of the Crimean saline lakes, by the interaction of geological and intra-water factors (salinity, biotic factors, etc.), as well as the character and power of anthropogenic impacts. The proportions in the contribution of different causes the deviation from the background values of Hg content in different regions and lakes are not the same, and it is very difficult to estimate them. Great Salt Lake (USA), with average salinity near 150 g L^{-1}, has high Hg content, and the different Hg issues of this have been extensively studied over many years [1,3,34,44–46]. However, even in this most studied case, many questions remain unanswered.

A positive correlation between the content of total and suspended Hg and salinity was also noted for lakes of other regions [11,12,34,47,48] and Lagoon Sivash [25]. There is the same positive correlation between TSS and salinity (Figure 2), which also was noted in other cases [11,24,25,39], which can help to partly explain the general trend of the increased suspended Hg form with increasing salinity in a water body. The atmospheric Hg deposition is one of the major Hg sources to different aquatic environments, and high salinity can also enhance Hg input to the lake surface [34,49]. Higher salinity may increase Hg turnover with lowering of Hg sedimentation on the bottom [47] as well as determine a rate of Hg methylation and other processes regulating Hg behavior in the lakes [34,45]. Consequently, Hg concentrations in water may be impacted by many direct and indirect effects of salinity on the ecosystem.

The Hg concentrations ound in the green algae and vascular plant biomass are much lower than the maximum values found earlier in other regions [30,50]. For example, the concentration of Hg in *Potamogeton crispus* L. increased with growth, with the natural Hg content reaching up to 132 ng g^{-1} and exceeding 30,000 ng g^{-1} (dry weight) in experiments [51]. Filamentous green algae *Cladophora* create bottom and floating mats in the Crimean saline and hypersaline lakes, the mass of which can exceed 500 g L^{-1} (dry weight) [52,53]. Calculated with this in mind, data in Table 4 values show that the total Hg content in *Cladophora* biomass in 1 L of water can exceed 5000 ng L^{-1}, which can significantly exceed the total amount of dissolved and suspended forms of Hg in lake water (Table 2). At the end of autumn, filamentous algae and plants begin to die off, and bacteria intensively develop and absorb oxygen, making the environment acidic [54]. This may explain the anomalous increase in the dissolved form of Hg in Lake Kyzyl-Yar from an average of 80 ng L^{-1} (CV = 0.861) in the summer months to a value of 3635 ng L^{-1} in November, where filamentous algae and aquatic plants intensively develop in summer [23]. Based on data in Tables 2–4, we suggest that that plants and filamentous algae play a significant role in the Hg cycle in saline lakes, but experiments are needed to test this hypothesis.

The Hg content in the *Artemia* biomass in the Crimean lakes (Table 4) was significantly lower than in the Great Salt Lake, where it reached 1400 ng g^{-1} (dry weight), averaging about 500 ng g^{-1} [46]. At first glance, this may seem strange, because Hg concentration in Crimean lakes is higher (Table 2) than in Great Salt Lake [34,35,46]. An analysis of our data (see above) led to the conclusion that the dependence of the accumulation factor on salinity has a dome shape, which could partially explain this paradox. In addition, it was found that the accumulation coefficient/bioaccumulation factor for Hg decreases with increasing Hg content in water [46]. A similar inverse relationship between Hg concentration and a rate of Hg uptake has been observed also in other hydrobionts [55]. At the same time in Great Salt Lake, Hg concentration in brine fly (*Ephydra gracilis* Packard, 1871) larvae was on average slightly higher than in *Artemia*, averaging 659 ng g^{-1} (dry weight) [44]. These differences can probably be explained by differences in the diet of *Artemia* and *Ephydra*, the former filtering phytoplankton, whereas the latter consume periphyton [44,46].

Hg in *Artemia* biomass averages 0.6% (CV = 1.270) of its total amount in water (Tables 2 and 4). The mass dying off of *Artemia*, whose average lifespan is 10–30 days, leads to the removal of Hg from the water column, but this does not determine the main role of *Artemia* in the Hg cycle in hypersaline lakes. *Artemia* is an obligate filter feeder and can effectively consume both organic and inorganic particles [56]. Assuming that the daily filtration rate of a brine shrimp averages at least 40% of its body weight [57], the authors determined what minimum percentage of the available suspension (Table 2) it filtered per day. On average, for all cases with *Artemia* (Table 3), it was 4.1% (CV = 0.969). According to our and literary [44,46] data, the Hg concentration in the *Artemia* body did not correlate with the amount of Hg suspended in water, and it can be assumed that the suspended Hg as *Artemia* faecal pellets mainly settles to the bottom. It is known that in water bodies with high *Artemia* abundance and the process of their formation of fecal pellets significantly accelerates the process of sedimentation of suspended matter, and their pellets make up the bulk of bottom sediments [58–60]. Taking into account observed *Artemia* abundance (Table 4), the calculation showed that in this way all suspended Hg can be removed by *Artemia* from the water column to bottom sediments in 5–15 days. In faecal pellets of planktonic crustaceans, including *Artemia*, concentrations of Hg, lead, and other heavy metals can be significantly higher than the average content in the suspended matter [61,62]. Consequently, the turnover of suspended Hg is very fast, and brine shrimp are important factors in Hg cycling in hypersaline water bodies, which, however, was noted earlier [60].

5. Conclusions

The available data show that Hg behavior and variability of its concentration in lakes is determined by a large set of factors, both intra-lake (salinity, composition, functioning of biota, etc.) and external (geological, climatic, anthropogenic). Quantitatively, the role of each does not remain constant; it varies both in space and in time. Biological factors play an important role in the spatial and temporal distribution and behavior of Hg. The presence of large populations of *Artemia* in hypersaline lakes, as well as filamentous algae, can accelerate Hg cycling, but this important issue requires more detailed study.

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