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Article

Trace Elements in the Bottom Sediments of the Crimean Saline Lakes. Is It Possible to Explain Their Concentration Variability?

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Abstract: Knowledge of trace elements content and their behavior in aquatic ecosystems is important for their sustainable use. There is a lack of such data for saline and, especially, hypersaline lakes and lagoons. Concentrations of more than 20 elements were evaluated in bottom sediments of 15 saline/hypersaline lakes and Lagoon Sivash in Crimea. An average salinity varied from 4 to 335 g/L in studied water bodies. The concentration of the trace elements varied from lake to lake. The highest variability was recorded for Cd, from 4.13 mg/kg to below the detectable level (CV = 1.463), and for Se, from 5.52 to 0.05 mg/kg (CV = 1.053). The lowest variability demonstrated by Cr, from 368 to 17 mg/kg (CV = 0.463), and by V, from 67.8 to 1.7 mg/kg (CV = 0.481). According to the found content of studied elements, all lakes were separated into three groups, and Lagoon Sivash was not included in these clusters. Salinity affected the concentration of some elements in bottom sediments, and this effect was not linear or unidirectional. In some cases, the action of other factors, often unknown, masked the effect of salinity. The geochemical background affects the structure and functioning of aquatic ecosystems, but the state of these ecosystems can significantly modify this background. An understanding of the differences in the elemental composition of bottom sediments in different lakes is possible only based on an integrated consideration of the interaction of all landscape, intra-ecosystem, and anthropogenic processes and factors that can influence this.

Keywords: saline lakes; bottom sediments; trace elements; ecosystems

1. Introduction

To some extent, geochemical factors determine the formation of the structure and functioning of aquatic ecosystems [1–3]. As an example, some macro- and trace elements (Ca, Mg, K, Fe, Co, etc.) are essential food elements that are critically needed in some quantities for physiological processes in organisms, their proper growth, and development [4,5]. Being essential, they may become toxic at high concentrations [6]. Some other elements, such as Ag, As, Cd, Cr, Hg, Ni, Pb, Sn, and others have no currently known biological functions but lead to toxic effects even at low concentration [7,8]. Geochemical anomalies with such low or high concentrations of different elements form due to the natural geological factors as well as to the different anthropogenic activities [9]. Increased or decreased concentrations of trace elements in aquatic environmental components can pose risks to the normal ecosystem functioning and the health of people that use ecosystem resources [7,8,10]. Trace elements were accumulated by organisms of low trophic levels, and in the aquatic food chains, their concentration may increase in a trophic chain from primary producers to the 3rd consumers.

The top-level predators (fish, birds, and humans) may accumulate the highest concentration, threatening health and livelihood [10,11]. In aquatic ecology, one of the current general goals is to improve our understanding of interactions between geochemical background and ecosystem functioning to include the realities of these mutual interdependencies in environmental management.

There are both natural and anthropogenic sources of trace elements in aquatic ecosystems. Currently, trace element pollution of the biosphere by anthropogenic releases increased with acceleration and significantly exceeded that from natural sources, from 2 to 335 times for different elements [1,12,13]. Direct atmospheric fallout of trace elements on water areas and their watersheds are main anthropogenic sources contributing to pollution of water bodies globally. For a better understanding of ecosystem functioning and prediction of ecosystem possible dynamics, knowledge of the interaction between geochemical and biological processes in water bodies is necessary. To move in this direction, knowledge of the geochemical background of aquatic ecosystems and the natural and anthropogenic factors affecting them are necessary. So, all knowledge of trace elements content and their behavior in different aquatic ecosystems is important for the sustainable use of water bodies. Bottom sediments, as a rule, are the main accumulators of most elements in water bodies. Therefore, a lot of attention is paid to the study of trace elements in them, especially in fresh and marine waters [3,7,8,14–16]. Significantly less attention is paid to the study of trace elements in saline and, especially, hypersaline lakes and lagoons [17–25], which, however, show that such reservoirs are characterized by unique features. Currently, the importance of the sustainable use of hypersaline water bodies is growing, including for aquaculture [26]. This contributes to actuality to study the geochemical background of saline and hypersaline lakes and lagoons worldwide.

Hypersaline waters (salinity above 35 g/L) are among the most extreme habitats of the planet and have their own biological and geochemical features [27,28]. On the Crimean Peninsula, the largest in the Black Sea, there are more than 50 hypersaline lakes and the world's largest hypersaline lagoon Bay Sivash of the Sea of Azov (Figure 1) [29,30].

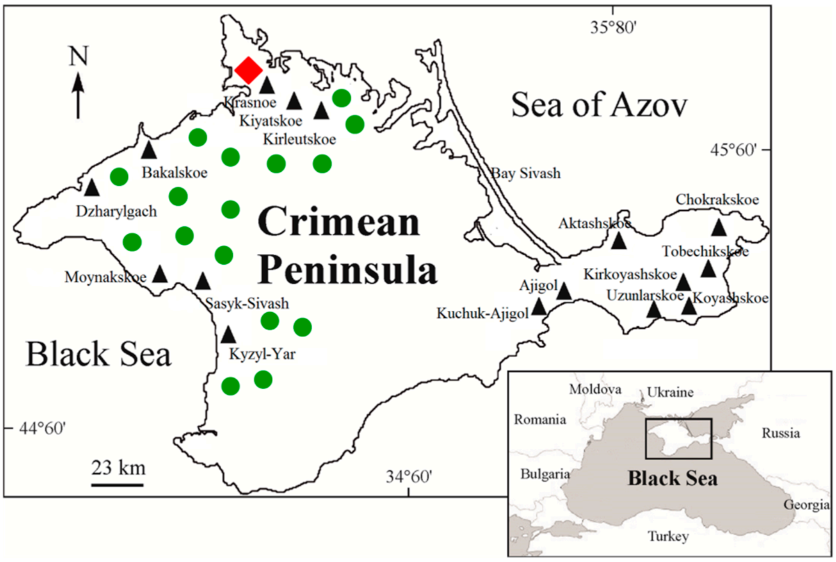

Figure 1. The Crimean map with the studied lakes and Bay Sivash: black triangles—studied lakes; red rhombus—the center of the chemical industry (the cities of Krasnoperekopsk and Armyansk); green circles—agricultural land.

Among the lakes, there are two groups of lakes: marine origin and continental (sulfate). To understand the coupling of lake ecosystem functioning with geochemical background, we need to know this background. It was never studied before in the Crimean saline lakes, and this is the first attempt in such direction. The work aimed to study the content of 26 elements in the bottom sediments of 16 lakes and Bay Sivash and try to understand what factors can influence the variability of their concentrations. It was supposed to check the assumptions: 1. bottom sediments of the Crimean marine and continental lakes have significantly different concentrations of trace elements; 2. Sivash Bay, where, unlike all lakes, there is a very high proportion of bivalve mollusk shells, it will have significantly lower concentrations of many elements in bottom sediments.

2. Materials and Methods

2.1. Study Area

Crimea is the largest peninsula in the Black Sea (area of 27,000 km^2), and due to arid climate in the majority of its territory, there are more than 60 saline and hypersaline lakes and Bay Sivash, the world's largest hypersaline lagoon (Figures 1 and 2) [29–31].

Figure 2. The Crimean studied lakes: (**a**) Lake Koyashskoe; (**b**) Lake Kirkoyashskoe; (**c**) Lake Chokrakskoe; (**d**) Mud volcano on the dried bottom of Lake Tobechikskoe; (**e**) Lake Krasnoe; (**f**) Lake Uzunlarskoe.

There are two types of lakes: marine origin (thalassohaline) and continental (athalassohaline, sulfate), which are located in the caldera of mud paleovolcanoes. The Center of the large chemical industry in the North part of Crimea (the cities of Krasnoperekopsk and Armyansk with different chemical plants) is the main source of atmospheric microelement pollution in the peninsula (Figure 1). It also dumps some poorly treated wastewater in Lake Krasnoe (Figure 2). This center impacts on all lakes in North and West Crimea. It also dumps some poorly treated wastewater in Lake Krasnoe (Figure 2). Agriculture is the second most important microelement pollutant (fertilizers, pesticides, and insecticides). It is most intensively developed in North and West Crimea occupying more than 70% of the total territory here. Near Lake Uzunlarskoe, there is a bombing military polygon, a local polluter (Figure 2). Bottom sediments were collected (one–three times) in 15 saline lakes and two sites of Bay Sivash in 2016 and 2018 (Figure 1, Table 1).

Table 1. General characteristics of the Crimean studied water bodies.

Lakes	Coordinates	Chemical Type	Origin	Dried Periods	Area, km^2	Average Salinity during Sampling, g/L	Lake Belongs to the Cluster Number
Tobechikskoe	45°09′07′′ N–36°22′29′′ E	Thalassohaline	Marine	Yes	19.0	256	3
Chokrakskoe	45°27′50′′ N–36°18′32′′ E	Thalassohaline	Marine	Yes	8.5	248	3
Aktashskoe	45°23′09′′ N–35°50′02′′ E	Thalassohaline	Marine	Yes	25.0	144	3
Adjigol	45°05′09′′ N–35°25′08′′ E	Thalassohaline	Marine	Yes	0.8	155	3
Kuchuk-Adjigol	45°06′08′′ N–35°27′08′′ E	Athalassohaline	Marine	No	0.6	4	3
Koyashskoe	45°02′08′′ N–36°12′03′′ E	Thalassohaline	Marine	No	5.5	280	3
Kirkoyashskoe	45°04′08′′ N–36°13′04′′ E	Athalassohaline	Continental	Yes	0.7	127	2
Uzunlarskoe	45°05′11′′ N–36°07′09′′ E	Thalassohaline	Marine	Yes	20.0	130	2
Krasnoe	45°59′15′′ N–33°53′07′′ E	Thalassohaline	Marine	No	23.4	335	2
Kiyatskoe	45°58′41′′ N–33°54′34′′ E	Thalassohaline	Marine	No	12.5	227	2
Kirleutskoe	45°55′29′′ N–34°02′44′′ E	Thalassohaline	Marine	No	21.0	259	2
Bakalskoe	45°43′55′′ N–33°10′56′′ E	Thalassohaline	Marine	No	8.0	119	3
Dzharylgach	45°34′59′′ N–32°51′30′′ E	Thalassohaline	Marine	No	8.0	152	1
Sasyk-Sivash	45°09′17′′ N–33°30′41′′ E	Thalassohaline	Marine	Yes	75.3	280	1
Kyzyl-Yar	45°03′34′′ N–33°35′22′′ E	Thalassohaline	Marine	No	8.0	6	1
Bay Sivash	45°17′14″ N–35°28′01″ E	Thalassohaline	Marine	No	2560.0	65	–

2.2. Sampling and Sample Processing

For a sampling of bottom sediments, an acrylic tube with an inner diameter of 58 mm and a pointed lower edge was used. The tube was immersed in the thickness of bottom sediments, after which the lower edge of the tube was closed with a rubber stopper-piston and the upper—with a plastic cap. Along with this, the salinity was measured by a manual refractometer WZ212 (Kelilong Electron Co. Ltd., Fuan, Fujian, China). In the laboratory, to reduce the near-wall effect, the 1.5 mm edge of the bottom sediment column was cut off using a pointed ring of smaller diameter (55 mm). After cutting, the samples from sediment depth 0–5 cm were weighed and dried at a temperature of 40–50 °C to constant weight. Samples from depth 0–5 cm of dried bottom sediments were ground in a porcelain mortar to a homogeneous state. To analyze the content of the elements, two-three weighed sub-samples of 1 g were taken from each sample. Then, acid decomposition was performed openly in a glass beaker using a hotplate. Five milliliters of high purity hydrochloric acid, 1 mL of hydrogen peroxide, and 5 mL of double-distilled water were added to 1 g of substrate, then, the sample was placed on a hot electric stove for 5–10 min, until wet salts were formed, and after this, 10 mL of high purity nitric acid and 1 mL of hydrogen peroxide were added. After this, the sample was again placed on a hot electric stove for 5–10 min, and then 10 mL of high purity nitric acid and 90 mL of double-distilled water were added. After the sample was completely dissolved, the resulting mixture was filtered through a “blue ribbon paper” filter into a 50 mL flask; the volume was brought to the mark with double-distilled water. Determination of the content of the following isotopes ^{7}Li, ^{9}Be, ^{11}B, ^{24}Mg, ^{27}Al, ^{39}K, ^{44}Ca, ^{49}Ti, ^{52}Cr, ^{51}V, ^{55}Mn, ^{57}Fe, ^{59}Co, ^{60}Ni, ^{65}Cu, ^{66}Zn, ^{75}As, ^{78}Se, ^{88}Sr, ^{98}Mo, ^{107}Ag, ^{114}Cd, ^{121}Sb, ^{137}Ba, ^{205}Tl, and ^{207}Pb were performed using atomic emission spectrometry (mass spectrometry) on an inductively coupled plasma mass spectrometer PlasmaQuant MS Elite (Analytik Jena AG, Jena, Germany). The spectrometer was calibrated using the appropriate calibration solutions Quality Control Standard 28 in HNO_3/HF, 125 mL was used. As a background, “blank” samples were used. A “blank sample” was prepared in parallel with the analyzed one, and it contained the same reagents and in the same quantities as the analyzed sample. For each sample of bottom sediments, measurements were carried out in triplicate, the result was the average value of measurements (up to six out of one sample). The error of the method used did not exceed 10% of the result. Mass fraction of element in an air-dry sample (Xe, mg/kg) was calculated by the formula:

$$Xe = (Xv \times V)/M \quad (1)$$

where Xv is an element contained in the analyzed solution, mg/dm^3, V is the volume of the prepared solution, dm^3; M is the mass of a sample of air-dry samples, kg.

2.3. Data Processing

Data were processed with standard statistical methods [32]. The author's calculated average values, coefficients of correlations (R), determination (R^2), variability (CV), and standard deviation (SD), also parameters of regression equations using the standard program MS Excel 2007. The significance of average value differences (p) was evaluated by Student's t-test after normality tests before [33]. The confidence level of R was assessed by comparing with R critical values ($p \leq 0.05$) [34]. To calculate Euclidean distances between lakes in the cluster analysis, STATISTICA 6 was used.

3. Results

3.1. Macroelements

The content of elements Al, K, and Mg in the samples was higher (>100 g/kg, dry weight) than could be determined because the device was off the scale. The found concentrations of other macroelements are given in Table 2, from which it can be seen that the average concentration of Ca in thalassohaline lakes was 189 g/kg (CV = 0.318), and in the only studied athalassohaline (sulfate) lake it was significantly lower, 26.1 g/kg. The average concentration of Sr was 146.8 g/kg (CV = 0.907) in thalassohaline lakes, and in the two athalassohaline lakes, it was much lower, 0.3 g/kg (CV = 0.907). The concentration of Mn was more variable, averaging 1.6 g/kg (CV = 0.161), and Fe 23.8 g/kg (CV = 0.381). Concentrations of Ca, Sr, and Mn in the bottom sediments did not correlate with the salinity, and the concentration of Fe significantly correlated with the salinity (R = 0.585, $p = 0.03$) in thalassohaline lakes and the dependence was approximated by the equation:

$$C_{Fe} = 13.39 + 0.06 \times S, \quad (2)$$

where C_{Fe}—Fe concentration, g/kg, S—salinity, g/L.

An analysis of the calculated pairwise correlation coefficients showed that a statistically significant negative correlation existed only for two pairs, namely Ca and Fe (R = −0.786, $p = 0.005$), and also Sr and Fe (R = −0.744, $p = 0.007$). Cluster analysis was performed for lakes, where Ca, Fe, Mn, and Sr were determined; it showed that all lakes are grouped into two main clusters. The first cluster included the Bakalskoe and Dzharylgach lakes, which are part of the Tarkhakut group of lakes. All other considered lakes were included in the second cluster. The cluster analysis for four macroelements showed that according to the nature of the distribution over the lakes, the elements were divided into two groups (Ca and Sr) and (Fe and Mn), which was to be expected. It should be noted that Fe and Mn are more similar in distribution than Ca and Sr, judging by the Euclidean distance, which is three times greater for the second pair than for the first.

Table 2. The concentration of the macroelements in bottom sediments of the Crimean studied water bodies.

Lake	**Element, g/kg (Dry Weight)**			
	Ca	**Mn**	**Fe**	**Sr**
Tobechikskoe	202.7	0.8	34.3	51.6
Chokrakskoe	155.5	0.8	28.0	126.7
Aktashskoe	132.2	0.9	33.9	38.4
Adjigol	*	11.7	23.8	*
Kuchuk-Adjigol	*	0.5	*	0.3 **
Koyashskoe	*	0.5	15.8	*
Kirkoyashskoe	26.1 **	*	*	0.3 **
Krasnoe	186.9	0.7	36.9	175.4
Kiyatskoe	181.9	0.5	33.1	70.2
Kirleutskoe	100.6	0.7	22.9	85.3
Bakalskoe	252.9	0.9	20.5	263.5
Dzharylgach	315.5	0.9	6.8	473.0
Sasyk-Sivash	175.1	0.6	21.8	122.8
Kyzyl-Yar	186.3	0.8	14.0	61.3
Bay Sivash	*	*	17.9	*
Minimum	100.6 ***	0.5	6.8	38.4 ***
Maximum	252.9	11.7	36.9	473.0 ***
Average	189.0 ***	1.6	23.8	146.8 ***
CV	0.318 ***	1.965	0.381	0.907 ***

*—higher than the level of detection; **—in an athalassohaline lake, excluded in the calculation of the average concentration in marine lakes; ***—average only for thalassohaline lakes; CV—coefficient of variation.

3.2. Trace Elements

The content of 19 trace elements was studied in the bottom sediments of 15 lakes and Sivash Bay, the results are given in Table 3. The concentration of four elements (Ag, Cd, Sb, and Tl) in several lakes was lower than the level of detection, while in others, they had a low concentration. As these four elements were not identified in all lakes, they were excluded from further analysis. The concentration of Se (CV = 1.053) showed the highest variability from lake to lake, the concentration of other elements fluctuated less, and CV ranged from 0.463 (Ti) to 0.619 (Mo). When conducting a cluster analysis with the concentration of 15 trace elements, Sivash Bay was allocated into a separate cluster, while all the lakes were in another cluster. This was not surprising since concentrations of 13 elements in its bottom sediments were significantly lower than in lakes. This was probably due to the very high content of bivalve mollusk shells in the bottom sediments. In turn, all the lakes were grouped into two main clusters. One of these two clusters was subdivided into two subclusters. The characteristics of all three clusters of lakes are given in Table 3. The concentrations of all elements, excepting Se and Ba, were significantly lower in the 1st cluster of lakes than in the other two groups of lakes (p = 0.05–0.005), and the Ba concentration was significantly higher in the 1st cluster (p = 0.01). The concentration of Se did not differ significantly in all groups of lakes. It should be noted that the division of lakes into clusters according to macroelements and trace elements did not coincide, although some similarity was visible. Among the studied lakes, all lakes, except Kirkoyashskoe (continental lake), belong to the marine type of lakes. Lake Kuchuk-Adzhigol, being of marine origin, currently does not feed on the sea. It receives only the surface freshwater from its watershed. The bottom sediments of these two lakes were characterized significantly (p = 0.05–0.005) with a higher concentration of several elements, namely Li 2.3 times, Co 2.2 times, Ni 2.2 times, Cu 1.5 times, Zn 2.2 times, and Pb 2.4 times. There were no significant differences in the concentration of other elements between marine and continental feeding lake groups.

Table 3. The concentration of the trace elements in the bottom sediments of the different clusters of the Crimean studied water bodies.

Lake	**Element, mg/kg**																		
	Li	**Be**	**B**	**Ti**	**Cr**	**V**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Se**	**Ba**	**Mo**	**Pb**	**Ag**	**Cd**	**Sb**	**Tl**
First group																			
Dzharylgach	3.61	0.15	72.51	96.34	12.22	12.87	2.80	6.48	3.04	6.70	3.05	0.73	190.22	0.52	2.64	0.01	0.58	0.13	0.18
Sasyk-Sivash	10.06	0.48	77.27	195.21	31.81	38.43	4.29	12.60	8.23	22.06	5.09	1.58	362.24	0.59	3.76	0.01	0.37	0.101	0.28
Kyzyl-Yar	5.85	0.27	43.00	78.22	17.85	21.31	3.84	9.624	9.15	15.80	3.93	1.01	296.88	0.46	3.95	0.01	0.29	0.09	0.15
Average	6.51	0.30	64.26	123.26	20.63	24.20	3.64	9.57	6.81	14.85	4.02	1.11	283.11	0.52	3.45	–	–	–	–
CV	0.503	0.557	0.289	0.511	0.489	0.538	0.210	0.320	0.484	0.520	0.254	0.391	0.307	0.124	0.205	–	–	–	–
Second group																			
Kirkoyashskoe	35.14	0.89	161.93	328.30	62.77	64.63	12.85	39.58	15.66	68.24	10.90	1.36	98.47	1.39	12.34	****	****	****	****
Uzunlarskoe	21.38	0.10	95.43	362.60	47.88	64.62	9.41	29.80	13.01	56.70	7.78	0.05	78.53	1.10	8.054	****	****	****	0.09
Krasnoe	14.81	0.71	80.32	366.53	44.58	51.88	6.23	18.15	11.98	30.25	5.78	5.52	79.20	0.39	6.08	0.01	0.53	0.17	0.35
Kiyatskoe	19.71	0.92	124.10	368.78	56.82	67.83	6.82	22.97	13.50	39.26	7.17	0.59	134.48	0.79	4.37	0.01	0.54	0.22	0.35
Kirleutskoe	20.76	0.88	177.95	323.83	66.08	66.28	6.59	21.35	13.74	45.55	6.30	1.20	133.75	0.67	5.02	0.01	0.43	0.25	0.34
Average	22.36	0.70	127.95	350.01	55.63	63.05	8.38	26.37	13.58	48.0	7.59	1.74	104.89	0.87	7.17	–	–	–	–
CV	0.340	0.493	0.327	0.063	0.167	0.101	0.334	0.323	0.099	0.310	0.265	1.246	0.266	0.446	0.447	–	–	–	–
Third group																			
Tobechikskoe	22.34	0.79	103.26	217.54	50.64	52.82	6.16	21.35	11.72	44.02	9.14	1.30	226.70	0.40	6.611	0.02	0.46	0.16	0.50
Chokrakskoe	15.50	0.42	178.09	195.52	35.54	35.68	4.65	12.64	7.68	27.01	8.74	0.92	110.16	0.29	3.38	0.02	0.57	0.12	0.22
Aktashskoe	13.46	0.74	106.26	287.03	48.08	58.52	7.12	17.63	20.93	32.34	5.68	0.80	161.88	1.35	4.27	0.02	0.43	0.10	0.37
Adjigol	16.23	0.37	57.18	175.08	20.66	29.21	6.59	16.31	6.06	34.19	20.41	0.73	70.16	0.16	6.18	****	****	0.21	****
Kuchuk-Adjigol	30.07	1.04	44.52	229.01	47.74	64.92	14.49	35.12	15.56	65.50	13.64	0.83	100.39	0.33	12.31	****	****	0.13	****
Koyashskoe	9.08	0.34	56.01	230.79	25.92	29.99	6.21	18.17	7.22	24.78	8.74	0.65	62.78	0.78	7.83	****	4.13	0.02	****
Bakalskoe	11.15	0.48	85.48	205.59	27.49	35.30	5.13	14.74	7.47	21.90	5.46	1.42	147.55	0.65	4.65	0.01	0.51	0.13	0.22
Average	16.83	0.60	90.11	220.08	36.58	43.784	7.19	19.42	10.95	36.11	10.12	0.95	125.66	0.57	6.46	–	–	–	–
CV	0.428	0.441	0.507	0.161	0.336	0.334	0.462	0.384	0.503	0.414	0.523	0.310	0.458	0.718	0.463	–	–	–	–
Bay Sivash																			
Bay Sivash	0.75	0.01	9.47	17.04	3.59	1.69	0.22	7.53	0.33	1.47	0.35	0.11	44.83	0.10	0.30	****	0.02	****	0.01
All water bodies																			
Minimum	0.75	0.01	9.47	17.04	3.59	1.69	0.22	6.48	0.33	1.47	0.35	0.05	44.83	0.10	0.30	****	0.02	****	0.01
Maximum	35.14	1.04	178.09	368.78	62.77	67.83	14.49	39.58	20.93	68.24	20.41	5.52	362.24	1.39	12.34	0.02	4.13	0.25	0.50
Average	15.62	0.536	92.048	229.84	37.48	43.50	6.462	19.00	10.33	33.49	7.64	1.17	143.64	0.62	5.73	0.01	0.74	0.14	0.26
CV	0.589	0.601	0.531	0.463	0.492	0.481	0.541	0.492	0.502	0.572	0.608	1.053	0.612	0.619	0.561	0.372	1.463	0.460	0.536

****—concentration was below detectable level; CV—coefficient of variation.

When analyzing the entire set of data, no reliable correlation was found between salinity and the concentration of any element. When analyzing this dependence—the content of elements on salinity—in each group of lakes separately, it turned out that for individual elements this dependence was highly significant. The equations approximating this dependence are given in Table 4. In cluster 1, a reliable positive correlation was found only between the B content and salinity. In the second and third clusters, there was a negative correlation of the contents of Co, Ni, and Zn with salinity. A negative correlation of As and Mo contents with salinity was observed only in the second cluster, and Li and Be—only in the third cluster.

Table 4. Correlation between salinity (X, g/L) and the concentration of a trace element (Y, mg/kg dry mass).

Number of Equation	Y (Element)	Equation	R	*p*
		Cluster 1		
1	B	$Y = 31.45X^{0.163}$	0.998	0.01
2	Mo	$Y = 0.451 + 0.0005X$	0.999	0.01
		Cluster 2		
3	Co	$Y = 125.78e^{-0.523X}$	−0.852	0.03
4	Ni	$Y = 45.46e^{-0.003X}$	−0.903	0.01
5	Zn	$Y = 86.75e^{-0.003X}$	−0.913	0.01
6	As	$Y = 11.46e^{-0.002X}$	−0.816	0.05
7	Mo	$Y = 2.20e^{-0.005X}$	−0.954	0.005
		Cluster 3		
8	Li	$Y = 36.77 - 4.17\ln(X)$	−0.748	0.03
9	Be	$Y = 1.31 - 0.15\ln(X)$	−0.730	0.05
10	Co	$Y = 18.83 - 2.44\ln(X)$	−0.945	0.005
11	Ni	$Y = 43.43 - 5.03\ln(X)$	−0.871	0.005
12	Zn	$Y = 81.01 - 9.49\ln(X)$	−0.815	0.01

R—the correlation coefficient; *p*—the confidence level of correlation coefficient.

The distribution of the content of most elements reliably positively correlated with each other, which is shown by pairwise correlation coefficients (Table 5). The Ba content does not correlate with any element and the Se content—only with the Co content. The cluster analysis with 15 elements showed that according to the nature of the distribution over the lakes, the elements were divided into several groups. Ti and B presented as separate groups; their distribution over the lakes is not like the other elements considered. The nature of the distribution of B is somewhat more than that of Ti and Ba, similar to the distribution of other elements, but also was positioned as a separate group. All other elements were grouped into two clusters, the first included Cr, V, and Zn, and the second included Li, Be, Co, Ni, Cu, As, Se, Mo, and Pb. Moreover, the greatest similarity in the distribution was in the pairs of Li and Ni, As and Pb, as well as Se and Mo.

Table 5. Correlation coefficients between concentrations of different trace elements.

	Li	Be	B	Ti	Cr	V	Co	Ni	Cu	Zn	As	Se	Ba	Mo	Pb
Li	1	0.59	0.77	0.90 **	0.85 **	0.91 **	0.85 **	0.89 **	0.82	0.94 **	0.90 **	no	no	no	0.68
Be		1	0.52 *	0.54 *	0.81	0.78	0.57	0.49 *	0.77	0.57	no	no	no	no	no
B			1	0.81	0.79 **	0.78	0.65	0.50 *	0.71	0.48 *	0.68	no	no	0.49 *	no
Ti				1	0.86 **	0.90 **	0.85 **	0.77	0.78	0.83 **	no	no	no	no	0.55
Cr					1	0.97 **	0.75	0.83	0.89 **	0.90 **	no	no	no	0.49 *	0.47 *
V						1	0.85 **	0.78	0.89 **	0.90 **	no	no	no	0.49 *	no
Co							1	0.93 **	0.96 **	0.93 **	0.92 **	0.42 ***	no	0.39 ***	0.78
Ni								1	0.62	0.94 **	0.61	no	no	no	0.68
Cu									1	0.91 **	0.84 **	no	no	0.49 ***	0.62
Zn										1	0.90 **	no	no	0.37 ***	0.71
As											1	no	no	no	0.75
Se												1	no	no	no
Ba													1	no	no
Mo														1	no
Pb															1

*—p (the confidence level of correlation coefficient) = 0.01; **—p = 0.001; ***—p = 0.05.

4. Discussion

The results showed that the totality of the studied water bodies is not a homogeneous group according to geochemical peculiarities. Of the entire considered group of water bodies, Bay Sivash stood out sharply, where the concentrations of all trace elements in bottom sediments were significantly lower than in lakes. This can be explained by the fact that in the bottom sediments of the bay, there were many mollusk shells, up to 80% of the total mass, which accumulated much smaller amounts of elements than clays or silts. Additionally, the bay has a small water exchange with the Sea of Azov that likely removes some quantity of microelements from the bay to the sea. It is easy to explain the difference in the concentration of several elements in sulfate lakes (Kuchuk-Adzhigol, Kirkoyashskoe), which are fed by surface runoff, and all others are marine, which has mainly marine nutrition with chloride type of water. These two groups of lakes differ in Ca and Sr concentrations and in the ratio of the main anions Cl^{-1}/SO_2^{-2} [29]. The concentration of sulfate ions in seawater is much lower, and Ca and Sr concentrations are much higher. Earlier it was found that seed shrimp Ostracoda, which form shells with Ca, are very abundant in the thalassohaline Crimean saline lakes but don't live in athalassohaline lakes [35]. Such decreased Ca content in those lakes is assumed to explain the absence of Ostracoda in them.

The solubility of the chlorides of Li, Co, Ni, Cu, Zn, and Pb is higher than their sulfates, so the sulfate salts of these metals will be easier to settle. Thus, in sulfate reservoirs, a large proportion of these elements will be in bottom sediments, and not in water. There is an increased concentration of the above elements in the bottom carbonate-sulfate sediments of the Crimean continental lakes compared to marine ones as noted earlier [24,36]. The influence of a lake chemical type on the concentration of elements in bottom sediments was obtained by comparing chloride and sulfate lakes in Mongolia, where the concentration of Li in the water of chloride lakes was 40% higher than in the water of sulfate lakes [37].

The division of lakes into three clusters is currently difficult to unambiguously explain, as many factors have determined this (the ratio of anions, the geological and soil background, the level and nature of anthropogenic influence, the nature of the water exchange state of ecosystems, etc.), but the contribution of different factors cannot be characterized quantitatively yet. The first cluster included lakes with very different salinity but located close to each other, which suggests a similar geological background. Also, they are characterized by similar economic activities and approximately equal remoteness (from 70 to 95 km) from the powerful center of the chemical industry in the north of Crimea. The second cluster includes lakes located in different parts of Crimea; these are lakes with the highest level of industrial pollution and one continental lake located in the caldera of a mud paleovolcano. Three of these lakes are very close to the largest center of the chemical industry (from 5 to 15 km); one of them (Lake Uzunlarskoe) is situated in an area of military bombing training. In the case of Lake Kirkoyashskoe, high concentrations of some elements can probably be explained by the fact that the lake is located in the caldera of a mud volcano, which led to the formation of a geological background with a high content of elements, as shown earlier [38,39]. The data obtained on three hypersaline lakes of the Pamir region, when the concentration of elements was determined in parallel in the sediments of lakes, in the sand along the banks, and a mud volcano located in one of the lakes [17], support such explanation. A significant positive correlation was observed between the element concentration in the bottom sediments of the lake and the sand on the shore ($R = 0.976$, $p = 0.0001$), while the concentration of all compared elements (Co, Ni, Cu, Zn, As, Se, Ba, Mo, Pb, and Sb) was significantly higher in bottom sediments, at 1.7–4.0 times. The concentration of these elements in the sand was 1.7–40 times lower in the sand than in the discharge of a mud volcano. The third group included the marine lakes of the Kerch Peninsula and Lake Bakalskoe, located at the other end of the Crimean Peninsula. The similarity of bottom sediments of the lakes of the Kerch Peninsula was noted earlier, showing that they are characterized by the dominance of sulfate salts and the influence of the powerful iron ore sedimentary rocks of the Cimmerian layer [24,40]. Their weathering products provide rather high concentrations of Fe, Ti, Cr, Mn, Pb, V, Co, and Zn in the bottom sediments of lakes [24]. Lake Bakalskoe has a completely different geological background; agricultural activity is developed on its catchment, and it is moderately affected by chemical plants in the north of Crimea (about 45 km). This probably provides relatively high concentrations of some elements in it. Summarizing all the above, we can conclude that the geological background, the chemical peculiarities of a lake, and anthropogenic activity together determined the concentration of elements in the bottom sediments. The analyses of Table 3 showed that the maximum marked concentrations of Li, Be, B, Ti, Cr, V, Co, Ni, Cu, Zn, Se, Mo, and Pb were noted in the second cluster of the lake. For all lakes, except continental Kirkoyashskoe, this can be explained by the strong influence of anthropogenic pollution. In the case of Lake Kirkoyashskoe, this can probably be explained by geological background. Mud volcanoes can significantly affect the geological background of hypersaline lakes and the concentration of elements in the bottom sediments of lakes. Based on a study of the composition of the liquid phase substances from the vents of volcanoes, it was shown that their mud contains a high concentration of As, Se, B, Ba, Cu, Ni, Zn, Pb, Ag, Sb, etc. [36,38,41]. It should be noted that modern mud volcanism is developed on the Kerch Peninsula, including in some lakes themselves (Figure 2d), with episodic powerful eruptions [41], which can significantly affect the geochemical processes in the lakes. The general conclusion that can be drawn by analyzing the composition of the lake clusters is that high concentrations of various elements in the bottom sediments of the lakes can be due to both natural and anthropogenic causes. Each lake must be approached as an individual unique object to correctly assess the relationship between causes and effects.

Considering the natural factors affecting the elemental composition of the bottom sediments of salt lakes, future studies should take into account not only the composition of the rocks and products of their weathering but also the chemical composition of soils and underground waters. For example, a feature of the Kerch Peninsula is the presence of a large number of sulfates in soils and groundwater ([24], own unpublished data). The accumulation of various elements by bottom sediments is affected by their particle size distribution, which, in turn, is largely determined by the frequency and duration of the drying periods of the lakes [24,29,42]. In many Crimean hypersaline lakes, all water areas or their parts regularly dries up partly or entirely over during summer. The share of dried area and duration of such periods vary from lake to lake and from year to year. High amounts of different elements are lost from a lake during the drying periods due to winds [43–45]. So, to understand the differences of lakes, it is necessary to know the character of the long-term fluctuations between dry and wet periods as well as the lake’s water balance depending on the local climate.

According to our data, the influence of salinity on the concentration of different elements in the bottom sediments of the studied lakes cannot be characterized unambiguously (Table 4). Of course, salinity cannot affect the income of elements into the water bodies. However, it can to some extent determine the distribution of elements between the water mass and bottom sediments [14,42,46–49]. Interannual climatic fluctuations in the water balance affect salinity, which can vary over a very wide range in the same lake [50,51]. For example, in the Kuyalnik lagoon, salinity varied from year to year over a wide range from 70 to 340 g/L, with significant, but multidirectional, correlation of the Pb content in water and bottom sediments with salinity [19]. A similar trend was shown for other elements in different water bodies [14,42,46–49]. The analysis of all available data suggests that this dependence is non-linear; there is a critical salinity when the sign of the dependence may change. This is primarily determined by the solubility of salts at different salinity, and a salinity value when sedimentation begins. In this case, it can be assumed that an increase of solubility with increasing salinity would lead to a decrease in the element concentration in the bottom sediments. This can explain the negative relationship between the concentration of this element in bottom sediments and salinity, which was found for Co, Ni, Zn, As, and Mo in the second cluster of lakes and Li, Be, Co, Ni, and Zn in the third cluster of lakes (Table 4). It is more difficult to explain the positive correlation of B concentration in bottom sediments with salinity in the first cluster of lakes because a relation between B behavior and salinity is more complicated [52]. Mo behaved differently in the three clusters of lakes: in the first cluster, its concentration in bottom sediments correlated positively with salinity, in the second cluster, the relation was negative, and in the third cluster, there was no correlation with salinity. All this can be explained only by the fact that salinity does not determine the concentration of elements in bottom sediments itself, but only coupling with all other factors involved in this. Previously, regarding the behavior of natural and artificial radionuclides in the Crimean saline lakes, it was shown that only taking into account the interaction of many factors, it is possible to get an understanding of the behavior of different isotopes in a lake [42,49]. The distribution of elements between water and bottom sediments is determined, first of all, by coupling the evaporative concentration of saline water and sedimentation, which depend on many external factors for a lake and internal factors (salinity, temperature, pH, Eh, oxygen concentration, nature, and intensity of biological processes) [31,42,49]. In the Crimean saline lakes, ecosystems can be in alternative states [50,51,53]: in highly productive years, there is an intensive development of the floating filamentous green algae mats, which form reduced conditions near the bottom with hydrogen sulfide, and in other years, there is the dominance of phytoplankton forming oxidative conditions near the bottom. This significantly affects the intensity and direction of an element exchange between bottom and water [42,49,54]. The populations of some biological species can selectively accumulate some elements and significantly modify the behavior of these elements in the ecosystems [10,35,55,56]. There is both a direct and reverse relationship between the state of ecosystems and the geochemical background in lakes, which, unfortunately, are still poorly studied. The geochemical background affects the structure and functioning of aquatic ecosystems, but the state of these ecosystems can significantly modify this background. When analyzing the ecosystem role of elements, it should be remembered that the toxicity of the number of elements for various organisms' taxa decreases with increasing salinity [57,58]. In hypersaline waters, organisms are adapted to existence at very high concentrations of toxic elements, which, for example, is shown for As, Se, and Hg [55,59–62]. Due to this, in ecosystems of hypersaline waters, organisms can often accumulate some toxic elements without harm to themselves. In this case, toxic elements may even stimulate an increase in ecosystem productivity. As an example, arsenic is a well-known toxin in the freshwater and marine aquatic environment, but it can serve as an important metabolic energy source to some microorganisms in hypersaline waters modifying the food web [59,63]. At present, the relationship of geochemical processes and the functioning of ecosystems in hypersaline waters is an urgent and extremely interesting field of research.

Summarizing the results of studies of saline and hypersaline lakes in different regions along with closely located fresh ones [17,18,21–23,54,64], the authors can conclude that salinity affects the concentration of some elements in bottom sediments. However, these effects are not-linear and not-unidirectional. In some cases, the action of other factors, often unknown, masked the effects of a salinity [14,42,46–49], sometimes changing the sign of this influence. It is impossible to single out any one main factor affecting the formation of the concentration of elements in all cases. The lakes differ in the composition of driving factors coupling, and this composition can change over time. Probably, the main conclusion from this study is the following: an understanding of the differences in the elemental composition of bottom sediments in different lakes is possible only based on an integrated consideration of the interaction of all landscape, intra-ecosystem, and anthropogenic processes and factors that can influence this. This article may be regarded only as one of the first steps to understand the coupling of geochemical peculiarities and ecosystem functioning of the saline lakes. Currently, we lack understanding of this due to limited our knowledge and available data.

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