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Assessment of Trace and Rare Earth Elements Pollution in Water Bodies in the Area of Rare Metal Enterprise Influence: A Case Study—Kola Subarctic

Eugenia Krasavtseva [1,2,*], Sergey Sandimirov [2], Irina Elizarova [2] and Dmitriy Makarov [2,*]

[1] Kola Science Centre of the Russian Academy of Sciences, Laboratory of Nature-Inspired Technologies and Environmental Safety of the Arctic, Fersman St., 14, 184209 Apatity, Russia
[2] Kola Science Centre of the Russian Academy of Sciences, Institute of North Industrial Ecology Problems, Fersman St., 14a, 184209 Apatity, Russia
* Correspondence: vandeleur2012@yandex.ru or e.krasavtseva@ksc.ru (E.K.); mdv_2008@mail.ru (D.M.); Tel.: +7-951-295-9092 (E.K.)

Abstract: An extended study of the chemical composition of surface waters and lakes bottom sediments, which are affected to varying degrees by an enterprise that mines and processes rare metal ores in the Lovozero tundra, was carried out. Using inductively coupled plasma mass spectrometry, the content of trace elements and rare earth elements was established. Elevated concentrations of trace elements and rare earth elements were revealed in samples of water and bottom sediments of lakes receiving wastewater from the enterprise and polluted due to dusting in the tailings of the processing plant. Thus, the average content of the total REEs in the surface layers of the SR Ilma and Lovozero (at the mouth of the Sergevan River) reaches 561 and 736 mg/kg, respectively, while for the SR of Lake Krivoe this indicator was 74 mg/kg. The enrichment factor (EF_i), geoaccumulation index (I_{geo}), potential ecological risk index factor (E_{ir}) and potential ecological hazard index (RI) were calculated. Assessing the total pollution with trace elements and rare earth elements of bottom sediments of lakes Ilma and Lovozero at the mouth of the Sergevan River, the value of potential ecological risk reaches values corresponding to the level of moderate ecological risk pollution (RI_{lma} = 174, $RI_{Lovozero}$ = 186). The conducted correlation analysis made it possible to establish some of the main phases containing trace elements and rare earth elements in the bottom sediments of lakes Ilma and Lovozero.

Keywords: Lovozero tundra; geochemistry; surface waters; bottom sediments; pollutants; rare earth elements

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1. Introduction

The development of mineral deposits leads to pollution of environmental components with a wide range of substances, in particular, trace elements (TEs) and rare earth elements (REEs) [1–3]. Large mining and processing enterprises concentrated in the Murmansk region are powerful sources of negative environmental impact. Several large mining and processing enterprises are located on the territory of the Murmansk region: Apatit JSC, Kola MMC JSC, Olkon JSC, etc. A huge amount of waste is generated in the process of mining and beneficiation of ores. Stored solid mineral wastes are a source of serious pollution of the atmosphere, soils, surface and ground waters with heavy metals all over the world, especially near metal mining and processing sites [1–3].

Fluctuations in temperature, humidity and wind speed lead to the formation of dust on the surface of tailings, which is transported over long distances and enters soils and water bodies [4].

Thus, the consequences of the activity of a recently abandoned enterprise for the extraction of polymetallic ores in the province of Guangdong, South China, were studied in [5]. High levels of cadmium, zinc, copper and lead were found in agricultural soils. Analysis of tailings and waste waters showed an excess of the same elements. A possible cause of soil contamination is the wind transport of tailing particles and drainage and mine waters passing through the tailings.

Pollution of soils and water bodies with heavy metals and arsenic in the process of leaching by rainwater, wind transfer of fine particles of tailings from the tailings of lead-zinc and gold mining enterprises in China, Mexico and Spain is considered in [6–9].

Crushed tailings of ore dressing are stored in tailings [10–14], and wastewater from enterprises is often not cleaned of pollutants to standardized levels.

The development of deposits of rare metal ores can pose a threat to the environment by increasing the concentration of REEs in air, water and soil [15–17]. Fine particles of tailings can be transported by wind currents over considerable distances, settling on the surface of water bodies, soils and plants [18,19]. Dust particles can cause respiratory and cardiovascular diseases, severe intestinal disorders, keratosis and skin cancer [20].

At different times, employees of various research institutes conducted research on water bodies in the zone of mining enterprises' influence [21–25]. The relevance of such work is due to the vulnerability of water bodies in the Arctic Zone of the Russian Federation (AZRF) and the impact on them of emissions from industrial enterprises and large volumes of wastewater with a complex chemical composition. Significant rearrangements of the water ecosystems of the Russian Arctic inland water bodies are taking place, which integrate all environmental changes occurring in the territory of their watersheds and accumulate most of the pollutants that have entered the watersheds [21].

On the other hand, modern changes in the Arctic climate towards warming have led to research interest in the analysis of the historical development of aquatic ecosystems. Climate warming is one of the modulators of anthropogenic processes of pollution and eutrophication of freshwater ecosystems in areas of intensive nature management in the Arctic Zone [26]. Furthermore, in the lakes of the Russian Arctic, the habitat of hydrobionts is disrupted, leading to a decrease in species diversity in the context of environmental pollution and climate change [27].

In recent years, with the development of the scientific and technical base of research institutes, more and more studies have been conducted on the contamination of environmental components with REEs. Elevated REE contents are found in soil horizons, surface waters and dust in the atmospheric air in industrialized regions [28–31]. Thus, the bottom sediments (BS) of small urban lakes in the Murmansk region and Karelia were studied [28,29], the total REE content and the fractional composition of the samples were determined and the factors influencing the accumulation of REEs in the samples were established. In a previous study on the impact of an enterprise developing a deposit of apatite-nepheline ores, which include REEs, the content of REEs was established not only in bottom sediments but also in water samples and suspended solids [32].

The study goal was to conduct an investigation to assess the TE and REE pollution of surface waters and BS of lakes affected by an enterprise that extracts and processes rare metal ores.

2. Materials and Methods

2.1. Description of the Study Area

The only enterprise in Russia engaged in the extraction and processing of rare metal ores of the cerium group is located in the center of the Kola Peninsula. The Lovozero alkaline massif is a multiphase Paleozoic central-type intrusion that cuts through Archean basement rocks (gneisses of various compositions) and contains Upper Devonian effusive-sedimentary formations (picrite porphyrites, basalts, trachybasalts and their tuffs, shales, sandstones, quartzites). The massif is composed of alkaline rocks of three complexes of loparite-bearing, eudialytic lujavrites and vein rocks [33]. The composition of the developed ores also includes apatite and villiomite (water-soluble NaF), which causes an increased content of fluorine and phosphate ions in the wastewater of the enterprise.

A brief description of the lakes in the study area is given in Table 1.

Table 1. Geographic coordinates of sampling points, morphometric parameters of the studied lakes.

Lake	Geographic Coordinates	Height above Sea Level, m	Maximum Length, km	Maximum Width, km	Lake Area, km^2	Drainage Area, km^2
Ilma *	67.890713° N, 34.603778° E	399	0.8	0.46	0.27	6.26
Lovozero **	67.975035° N, 35.133188° E; 67.943437° N, 35.089683° E	153.9	44.5	8.64	224.5	3770
Krivoe *	67.962042° N, 34.562104° E	199.5	1.58	0.73	0.47	16.2

Note: *—data from [34]; **—data from [35].

The rivers in the territory of the study area belong to the group of small mountain rivers flowing down from mountain heights and emptying into lakes Umbozero and Lovozero [36]. Their width is insignificant, up to 5 m; the depth on the rapids and rifts is 0.1–0.8 m, up to 3 m on the reaches and the flow speed is 0.2–0.7 m/s, up to 2.7 m/s on the rapids. The riverbeds are meandering rapids, and the bottom is sometimes rocky. The banks are predominantly steep and up to 1 m high. Lovozero, Shomiyok (12.1 km) flows into Lake Sikir, and Alluive (6.5 km), Azimuth (8.7 km), Sengisyavr (9.2 km) and Tavayok (11.7 km) flow into Lake Umbozero. In the upper reaches they are characterized by a rapid stream and numerous rapids, while in the lower reaches they flow through forests and swampy areas. The rivers receive numerous tributaries along their entire length.

2.2. Sampling and Analyses

In the spring of 2022, samples of surface water and BS were taken from lakes Ilma, Krivoe and Lovozero (Figure 1). Samples of the BS were taken at the points with the greatest depth (lakes Ilma and Krivoe) and at two points of Lake Lovozero—at the confluence of the Sergevan River and in the northern part of the lake.

Lake Ilma is located to the southwest of the facility and, as previously suggested, is subject to pollution due to loose dusty tailings. Lake Krivoe is located to the northwest of the enterprise. Significant impact on this water body is not expected due to the considerable distance from the enterprise and the absence of wastewater discharge points into the lake. Lake Lovozero receives wastewater from the enterprise after it passes through the Sergevan River. The pollutants in wastewater include suspended solids, aluminum, iron, zinc, manganese, phosphates and fluorides. It should be noted that Lake Lovozero is the third largest lake in the Murmansk region and belongs to the highest category of reservoirs of fishery importance.

Sampling of surface waters was carried out in the near-surface and near-bottom layers. Water samples were analyzed by a set of methods: colorimetric, ion-exchange chromatography, atomic absorption spectrometry and mass spectrometry with inductively coupled plasma. Samples of BS were taken using an open-gravity-type bottom sediment core sampler in duplicate. Samples of BS were disassembled into layers 1 cm high then dried, ground with a rubber pestle in a mortar to a powdery state and submitted for analysis.

Samples were analyzed at the Center for Collective Use of the Institute of Problems of Industrial Ecology of the North, KSC RAS. Samples of BS were subjected to open acid decomposition, and the concentrations of microelements were determined in the resulting solutions with inductively coupled plasma mass spectrometry (ELAN 9000 (PerkinElmer, Waltham, MA, USA)) [37]. The control and quality of the analysis was ensured by the simultaneous decomposition and analysis of certified standard samples of the bottom sediments of Lake Baikal BIL-1 (GSO 7126-94). The measurement error did not exceed 0.5% at p = 0.95.

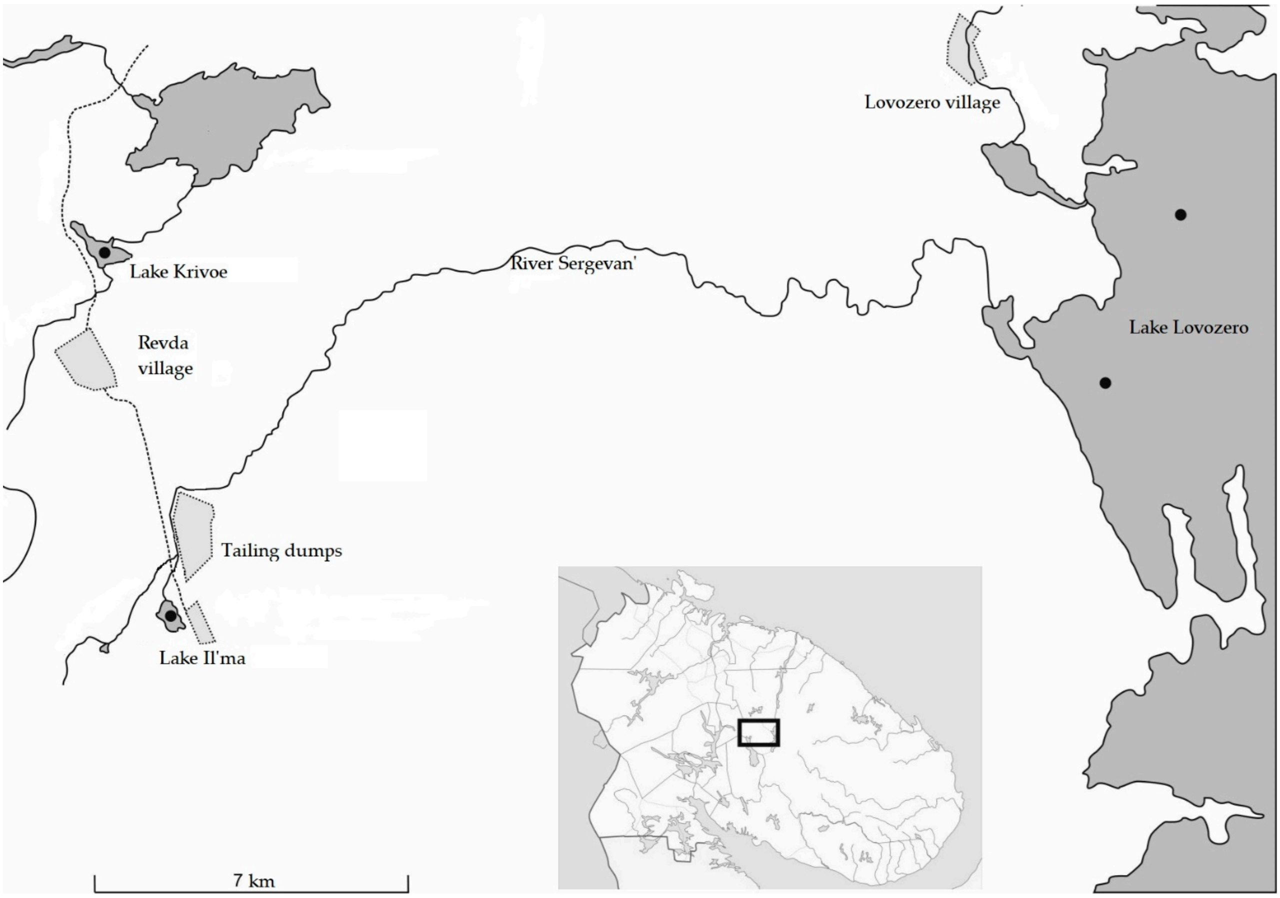

Figure 1. Sampling map—scheme of surface waters and BS of lakes.

The results were statistically processed. Arithmetic mean, standard deviation and median were calculated. In addition, correlation coefficients were calculated to determine the source of entry of a set of elements into the samples. All statistical calculations were carried out in Microsoft Excel 2010 (version 2209, build 16.0.15629.20200, Redmond, WA, USA).

The concentrations of elements in water samples taken from the lakes were compared with the MPC of fishery water bodies (MPC_{fw}) and the limits of concentrations in the water of freshwater ecosystems in accordance with [38]. A certain content of elements in samples of BS was compared, along with Clarke numbers of elements in the Earth's crust according to [39], limits of contents in BS of freshwater ecosystems according to [38] and average contents in BS of Lake Imandra [32].

2.3. TE and REE Pollution and Risk Assessment

The pollution assessment of the studied BS was carried out on the basis of the calculated coefficients and indices: the enrichment factor (EF_i), geoaccumulation index (I_{geo}), potential ecological risk index factor (E_{ir}) and potential ecological hazard index (RI) [40].

The enrichment factor was calculated by the following equation:

$$EF_i = C_i/C_{bi},$$

where C_i is the metal concentration, measured for each investigated sample; C_{bi} is the background concentration of the metal.

The EF values were interpreted as suggested [41,42], distinguishing seven EF classes: <1—no enrichment, 1–3—minor enrichment, 3–5—moderate enrichment, 5–10—moderately severe enrichment, 10–25—severe enrichment, 25–50—very severe enrichment, >50—extremely severe enrichment.

The geoaccumulation index determines and defines metal contamination in sediments [43]. I_{geo} enables an assessment of environmental contamination by comparing differences between current and pre-industrial concentrations of pollutants [43–45].

The geoaccumulation index was calculated by the following equation:

$$I_{geo} = \log_2 (C_n/1.5B_n),$$

where C_n is the measured total concentration of heavy metals (HM), in our case—TE, determined in a soil (mg/kg); B_n is the geochemical background value of the elements.

The constant in the equation—1.5—is used to analyze natural fluctuations in the environment and very small anthropogenic impacts [39]. I_{geo} consists of 7 classes (I_{geo} value–I_{geo} class–pollution level): ≤0–0: unpolluted, 0–1–1: unpolluted to moderately polluted, 1–2–2: moderately polluted, 2–3–3: moderately polluted to highly polluted, 3–4–4: highly polluted, 4–5–5: highly polluted to very highly polluted, >5–6: very highly polluted.

The potential ecological risk index factor and potential ecological hazard index can comprehensively evaluate concentration effects, toxicity and ecological sensitivity of HM [46–48]. The potential ecological hazard index was formulated by Hakanson in 1980, integrating the concentration of HM with ecological effect, environmental effect and toxicology, and was used to assess the HM pollution ecological hazard for sedimentology [49]. According to this method, the E_{ir} of an individual element and the RI of a multi-element element can be calculated using the following equations:

$$E_{ir} = T_i \times C_n/B_n,$$

where T_i is a factor of the toxic reaction of an individual toxic element. The established T_i values are 1 for Zn, 2 for Cr, 5 for Ni, Cu and Pb, 10 for As and 30 for Cd [48].

$$RI = \sum E_{ir}$$

Hakanson proposed the following classification for the E_r value: <40—low ecological risk, 40–80—moderate ecological risk, 80–160—appreciable ecological risk, 160–320—high ecological risk, >320—serious ecological risk; and RI value: <150—low ecological risk, 150–300—moderate ecological risk, 300–600—high ecological risk, >600—significantly high ecological risk [47].

In a recent study, the T_i values of 15 REEs were calculated: La = 1, Ce = 1, Pr = 5, Nd = 2, Sm = 5, Eu = 10, Gd = 5, Tb = 10, Dy = 5, Ho = 10, Er = 5, Tm = 10, Yb = 5, Lu = 20, Y = 2 [50].

In our study, enrichment factor, geoaccumulation index, potential ecological risk index factor and potential ecological hazard index of pollution of the surface layers of BS of lakes Krivoe, Ilma and Lovozero were calculated.

3. Results and Discussion

Since the enterprise is developing a deposit of rare metal ores, special attention was paid to the analysis of REEs in selected samples of water and BS.

3.1. Water Sample Analysis

The content of the main cations and anions in water samples of the near-surface layer is presented in Table 2.

Table 2. The content of the main cations and anions in the water samples of the studied lakes, mg/L.

Lake	pH	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	HCO_3^-	SO_4^{2-}	Cl^-	$NO_2^-+NO_3^-$	∑M *
Ilma	6.68	0.45	0.10	6.95	0.76	13.36	2.26	1.10	0.307	24.98
Krivoe	6.77	5.54	1.92	10.46	2.46	36.31	6.92	4.19	0.068	67.80
Lovozero	6.3	3.01	1.48	3.82	0.54	16.17	2.63	2.54	0.191	30.19

Note: *—mineralization of water, mg/L.

The composition of water in taken samples corresponds to the natural distribution of the main ions: $HCO_3^- > SO_4^{2-} > Cl^-$; $Na^+ > Ca^{2+} > Mg^{2+} + K^+$ and allows us to attribute them to the class of hydrocarbonate waters. Using the value of mineralization, the lakes are classified as low-mineralized and ultrafresh [51]. According to the acid/alkaline conditions, the considered lakes are neutral.

Based on the results of the chemical analysis of the selected lake water samples, the formulas for the ion–salt composition of the lakes Krivoe, Ilma and Lovozero was calculated:

$$\mathrm{M26}\,\frac{\mathrm{HCO3\ 69}}{\mathrm{Na\ 47\ Ca\ 29}}\ \mathrm{pH\ 6.77}\text{—Lake Krivoe;}$$

$$\mathrm{M26}\,\frac{\mathrm{HCO3\ 74}}{\mathrm{Na\ 91\ Ca\ 9}}\ \mathrm{pH\ 6.68}\text{—Lake Ilma;}$$

$$\mathrm{M28}\,\frac{\mathrm{HCO3\ 77\ Cl18}}{\mathrm{Na\ 25\ Ca\ 22}}\ \mathrm{pH\ 6.3}\text{—Lake Lovozero.}$$

The composition according to the content of the main ions is as follows: Lake Krivoe—bicarbonate, sodium-calcium; Lake Ilma—hydrocarbonate, sodium; Lake Lovozero—hydrocarbonate-chloride, sodium-calcium.

The concentrations of TEs and REEs, as well as MPC_{fw} and the limits of the content of elements in the water of freshwater ecosystems [38], are given in Table 3.

In general, the content of pollutants in the samples of the lakes under consideration, in comparison with other lakes of the Murmansk region, experiencing anthropogenic load due to pollution by wastewater from mining enterprises, is relatively low. For instance, Kuetsjärvi, the source of TEs and acid compounds entering the waters of which is the Pechenganickel plant (processing copper-nickel sulfide ores), is one of the most polluted lakes in the Murmansk region [52]. In the lake, elevated contents of nickel (more than 200 times) and copper (20 times) are observed. In the surface waters of the Monchegorsk test site, polluted by wastewater from the Severonickel copper-nickel plant, nickel and copper were also found in concentrations that exceed the conventional background values by 1–2 orders of magnitude [23].

Elevated concentrations of iron, aluminum and manganese in the surface waters of the Arctic Zone of the Russian Federation, as a rule, are explained by the humification of watersheds [53]. However, a comparison of the concentrations of a number of elements in water samples from Lake Lovozero, taken at different points of the lake, suggests that the excess of MPC_{fw} is due to pollution by insufficiently treated wastewater from the enterprise.

In [32], in the water of Lake Imandra, subject to anthropogenic pollution from the enterprise developing the deposit of apatite-nepheline ores, La concentration varied from 0.02 to 0.04 μg/L, Ce from 0.005 to 0.03 μg/L, Pr from 0.004 to 0.009 μg/L and Nd from 0.007 to 0.04 μg/L. In the water samples taken by us, as can be seen from Table 3, the concentrations of elements are at least an order of magnitude higher. Attention is drawn to the increased concentrations of zinc, copper and light group REEs (LREEs) in the near-bottom water layer of lakes in comparison with the surface layer, which may be a consequence of the leaching of the upper layers of BS by lake water.

Table 3. Concentrations of elements in surface water samples, μg/L.

Element	1	2	3	4	5	6	7	8
Cu	1.22 ± 0.08	1.50 ± 0.11	0.62 ± 0.04	1.65 ± 0.12	0.53 ± 0.03	0.98 ± 0.10	1	-
Zn	4.84 ± 0.17	64.58 ± 1.20	1.52 ± 0.11	55.69 ± 1.70	2.01 ± 0.11	39.89 ± 1.30	10	-
Mn	31.49 ± 0.65	109.35 ± 4.17	16.57 ± 0.62	256.41 ± 5.80	1.14 ± 0.08	17.13 ± 0.24	10	-
Al	77.18 ± 1.04	29.09 ± 1.21	124.82 ± 3.11	140.31 ± 2.93	65.93 ± 1.41	115.47 ± 4.71	40	-
La	0.33 ± 0.04	0.35 ± 0.03	1.25 ± 0.09	1.83 ± 0.08	0.91 ± 0.03	0.73 ± 0.02	-	0.06–0.2
Ce	0.50 ± 0.02	0.55 ± 0.07	2.10 ± 0.14	3.59 ± 0.12	0.51 ± 0.02	1.13 ± 0.05	-	0.08–2
Pr	0.08 ± 0.01	0.06 ± 0.01	0.22 ± 0.01	0.45 ± 0.02	0.17 ± 0.01	0.14 ± 0.01	-	0.007
Nd	0.31 ± 0.02	0.20 ± 0.01	0.69 ± 0.02	1.39 ± 0.10	0.45 ± 0.02	0.48 ± 0.01	-	0.040
Sm	0.05 ± 0.01	0.03 ± 0.01	0.10 ± 0.01	0.19 ± 0.02	0.08 ± 0.01	0.08 ± 0.01	-	0.008–1
Eu	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.05 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	-	0.001–0.03
Gd	0.05 ± 0.01	0.03 ± 0.01	0.09 ± 0.01	0.17 ± 0.01	0.07 ± 0.01	0.07 ± 0.01	-	0.0080
Tb	0.01 ± 0.01	0.00	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	-	0.001
Dy	0.04 ± 0.01	0.02 ± 0.01	0.04 ± 0.01	0.09 ± 0.01	0.06 ± 0.01	0.06 ± 0.01	-	-
Ho	0.01 ± 0.01	0.00	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	-	0.0010
Er	0.03 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.05 ± 0.01	0.03 ± 0.01	0.03 ± 0.01	-	0.0040
Tm	0.00	0.00	0.00	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	-	0.0010
Yb	0.03 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.04 ± 0.01	0.03 ± 0.01	0.03 ± 0.01	-	0.004–0.085
Lu	0.01 ± 0.01	0.00	0.00	0.01 ± 0.01	0.01 ± 0.01	0.00	-	0.0010
Y	0.33 ± 0.02	0.13 ± 0.01	0.23 ± 0.02	0.49 ± 0.03	0.38 ± 0.02	0.36 ± 0.01	-	0.03–0.7

Note: The table shows the mean values and standard errors. 1—Lake Lovozero, northern part (surface layer), 2—Lake Lovozero, northern part (bottom layer), 3—Lake Lovozero, the mouth of the River Sergevan (surface layer), 4—Lake Lovozero, the mouth of the River Sergevan (bottom layer), 5—Lake Ilma (surface layer), 6—Lake Ilma (bottom layer), 7—MPC_{fw}, 8—limits of water content in freshwater ecosystems according to [38].

3.2. BS Analysis

Since the average rate of sedimentation in the lakes of the Kola Peninsula is about 0.1 cm per year [54], and the enterprise began its operations in 1951, the content of elements in the deep layers of BS cores of more than 7–8 cm can be considered conditionally background, reflecting the content of elements before industrial development.

3.2.1. TE Pollution

Based on the data of chemical analysis of samples of lakes BS, enrichment factor, geoaccumulation index, potential ecological risk index factor and potential ecological hazard index were calculated to assess the pollution of BS with TEs (Table 4).

Compared to previous work [40], increased values of the enrichment factor of the finely dispersed material of the tailings of the enrichment of loparite ores were noted, including moderate enrichment for Mn, Sr, and these elements were added to the table for calculating the enrichment factor, geoaccumulation index. The factor of the toxic reaction of an individual toxic element has not been established for these elements; therefore, they do not participate in the calculation of potential ecological risk index factor or potential ecological hazard index.

Table 4. Concentrations of TEs in the surface and background layers of BS (mg/kg), enrichment factor, geoaccumulation index, potential ecological risk index factor and potential ecological hazard index.

Lake	Layer, cm	Cu	Ni	Zn	Cr	Cd	Pb	Sr	Mn	RI
Krivoe	0–7	20.50 ± 16.45 / 13.33 (6.92 − 45.24)	21.82 ± 17.14 / 14.83 (7.32 − 51.76)	61.41 ± 47.43 / 36.26 (21.95 − 141.83)	17.08 ± 4.78 / 15.31 (10.96 − 24.04)	0.121 ± 0.20 / 0.01 (0.01 − 0.51)	18.39 ± 6.66 / 16.25 (12.33 − 31.82)	116.94 ± 40.13 / 103.04 (74.92 − 193.58)	665.37 ± 85.15 / 639.35 (567.91 − 801.92)	-
	13–14	11.56 ± 2.77 / 11.56 (9.60 − 13.52)	22.45 ± 14.82 / 22.45 (11.98 − 32.93)	79.17 ± 28.68 / 79.17 (58.89 − 99.45)	15.56 ± 0.04 / 15.56 (15.53 − 15.59)	0.061 ± 0.08 / 0.06 (0.01 − 0.12)	14.13 ± 5.34 / 14.13 (10.36 − 17.91)	86.93 ± 2.45 / 86.93 (85.20 − 88.66)	586.23 ± 143.68 / 586.23 (484.63 − 687.82)	-
	EF	1.77	0.97	0.78	1.10	1.08	1.30	1.35	1.13	-
	I_{geo}	0.07	−0.19	−0.29	−0.14	−0.14	−0.06	−0.05	−0.12	-
	E_{ir}	8.87	4.86	0.78	2.20	32.50	6.51	-	-	55.71
Ilma	0–7	17.70 ± 7.87 / 20.88 (5.48 − 26.02)	22.55 ± 9.11 / 27.89 (8.33 − 30.24)	213.06 ± 3.25 / 212.11 (201.40 − 229.51)	23.02 ± 7.01 / 25.15 (11.54 − 29.86)	0.95 ± 0.28 / 0.97 (0.48 − 1.38)	24.00 ± 8.75 / 26.99 (12.60 − 33.69)	763.89 ± 233.11 / 759.17 (409.73 − 1074.29)	804.11 ± 130.50 / 870.00 (587.52 − 950.00)	-
	15–16	12.53 ± 0.43 / 5.15 (4.84 − 5.45)	27.39 ± 0.76 / 7.18 (6.64 − 7.71)	199.34 ± 7.89 / 202.57 (196.99 − 208.16)	11.57 ± 0.54 / 10.87 (10.49 − 11.25)	1.01 ± 0.35 / 1.20 (0.95 − 1.46)	6.07 ± 1.80 / 9.26 (7.99 − 10.53)	393.46 ± 3.66 / 360.87 (358.28 − 363.45)	427.93 ± 10.65 / 522.69 (515.16 − 530.22)	-
	EF	1.41	0.82	1.07	1.99	0.95	3.96	1.94	1.88	-
	I_{geo}	−0.03	−0.26	−0.15	0.12	−0.20	0.42	0.11	0.10	-
	E_{ir}	7.06	4.12	1.07	3.98	28.36	19.78	-	-	64.37
Lovozero, northern part	0–7	15.90 ± 1.80 / 16.90 (13.46 − 17.85)	30.92 ± 3.50 / 32.07 (26.20 − 36.50)	109.62 ± 4.23 / 110.09 (104.21 − 114.66)	46.15 ± 9.86 / 47.97 (32.45 − 58.19)	1.64 ± 0.57 / 1.72 (0.83 − 2.19)	11.40 ± 4.58 / 14.00 (4.22 − 15.94)	295.11 ± 25.20 / 303.40 (264.71 − 329.79)	777.17 ± 17.60 / 775.00 (755.23 − 807.28)	-
	8–9	17.14 ± 0.09 / 17.20 (17.14 − 17.26)	35.09 ± 1.00 / 35.80 (35.09 − 36.50)	169.61 ± 40.65 / 140.86 (112.12 − 169.61)	52.60 ± 3.95 / 55.39 (52.60 − 58.19)	2.10 ± 0.04 / 2.13 (2.10 − 2.16)	2.34 ± 1.33 / 3.28 (2.34 − 4.22)	286.47 ± 11.97 / 294.93 (286.47 − 303.40)	810.11 ± 12.68 / 801.15 (792.18 − 810.11)	-
	EF	0.93	0.88	0.65	0.88	0.78	4.87	1.03	0.96	-
	I_{geo}	−0.21	−0.23	−0.37	−0.23	−0.28	0.51	−0.16	−0.19	-
	E_{ir}	4.64	4.41	0.65	1.75	23.42	24.35	-	-	59.21
Lovozero, Sergevan	0–7	15.07 ± 3.18 / 14.19 (12.11 − 20.58)	50.42 ± 3.46 / 49.00 (45.73 − 54.56)	117.02 ± 20.76 / 128.39 (85.96 − 133.70)	87.53 ± 9.38 / 86.21 (71.97 − 102.50)	1.90 ± 0.17 / 1.91 (1.59 − 2.15)	15.25 ± 4.48 / 14.16 (10.84 − 22.44)	659.49 ± 77.60 / 648.26 (559.77 − 759.23)	1503.57 ± 191.28 / 1490.00 (1250.00 − 1850.00)	-
	8–9	11.28 ± 1.71 / 11.48 (11.28 − 13.70)	48.89 ± 2.13 / 48.94 (48.89 − 51.90)	168.93 ± 28.95 / 168.93 (127.99 − 168.93)	84.39 ± 4.12 / 84.78 (84.39 − 90.22)	1.51 ± 0.39 / 1.51 (1.51 − 2.06)	10.80 ± 0.90 / 10.92 (10.80 − 12.07)	537.27 ± 46.01 / 541.22 (537.27 − 602.35)	1220.00 ± 49.50 / 1220.00 (1220.00 − 1290.00)	-
	EF	1.34	1.03	0.69	1.04	1.26	1.41	1.23	1.23	-
	I_{geo}	−0.05	−0.16	−0.34	−0.16	−0.08	−0.03	−0.09	−0.09	-
	E_{ir}	6.68	5.16	0.69	2.07	37.72	7.06	-	-	59.39

Note: The numerator is mean value ± standard deviation; the denominator is the median (the minimum and maximum values of the sample). Lovozero, Sergevan—Lake Lovozero, the mouth of the River Sergevan.

According to the value of the calculated EF value, minor enrichment of Lake Krivoe BS was revealed with Cu, Cr, Cd, Pb, Sr and Mn; Lake Ilma BS with Cu, Zn, Cr, Sr and Mn; the northern part of Lake Lovozero BS with Sr and Lake Lovozero at the mouth of the Sergevan River BS with Cu, Ni, Cr, Cd, Pb, Sr, Mn. The level of enrichment Pb of BS samples "moderate enrichment" was found for Lake Ilma and the northern part of Lake Lovozero.

According to the calculated value of I_{geo}, the pollution of the BS of Lake Krivoe can be classified as unpolluted to moderately polluted with Cu, the BS of Lake Ilma with Cr, Pb, Sr and Mn and the BS of the northern part of Lake Lovozero with Pb.

The vertical distribution of the content of some elements in the selected samples of BS of lakes Ilma and Lovozero (northern part) is shown in Figure 2.

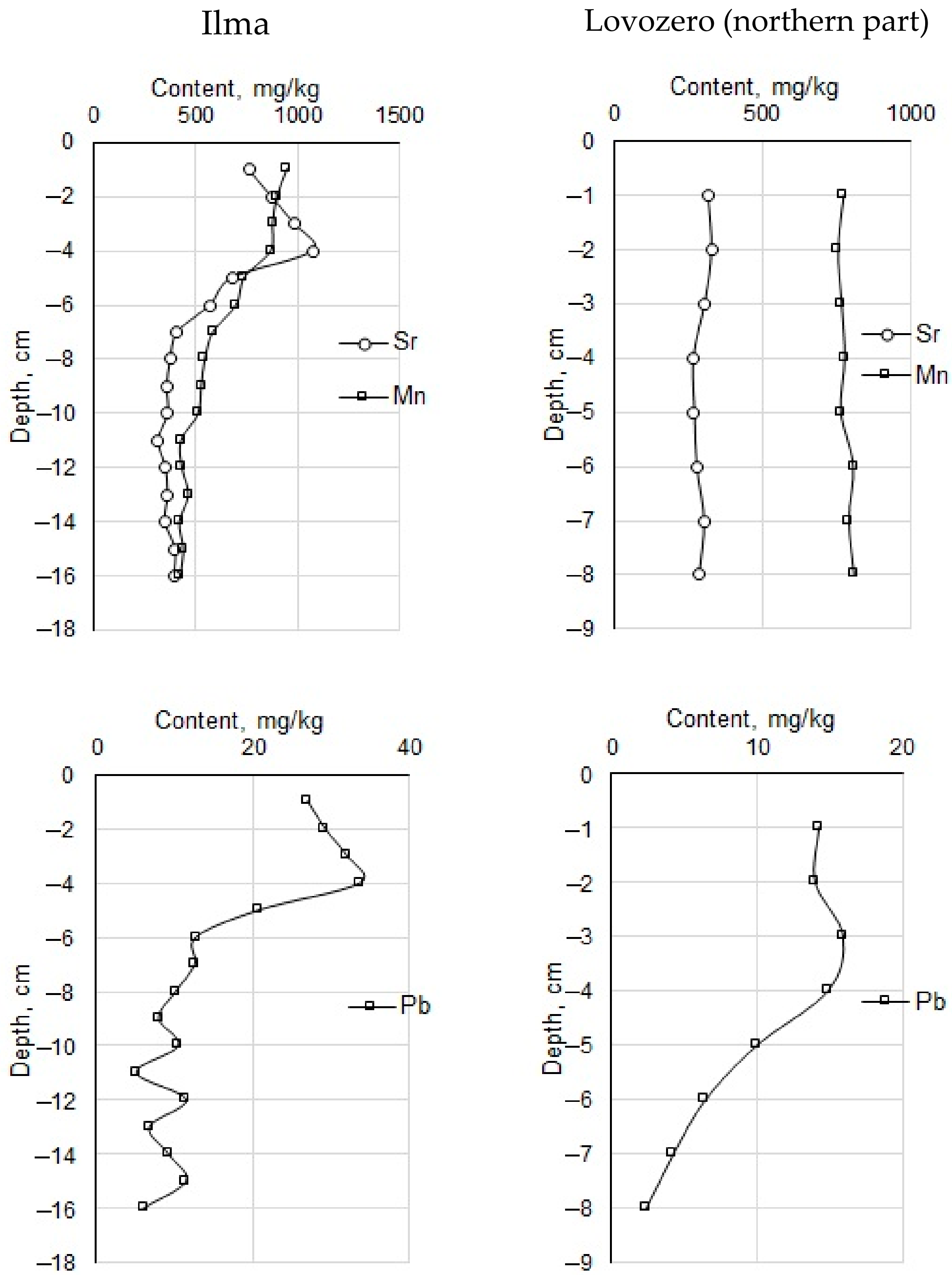

Figure 2. The vertical distribution of the content of some elements in the selected samples of BS of lakes Ilma and Lovozero (northern part).

One can note an increase in the content of a number of elements in the upper layers of BS columns, corresponding to the beginning of the enterprise's activity. Most of the elements indicated in Table 4 are included in the composition of the ores of the developed deposit [40]. Cu and Ni are among the priority pollutants in the Murmansk region, the increased content of which is associated with the activities of copper-nickel production [55]. As has been repeatedly noted in [55–57], an increased content of Pb may be a consequence of active traffic of vehicles. In our case, the road from the settlement of Revda passes by Lake Ilma towards the processing plant, and to the north of the sampling point for BS of Lake Lovozero is the road to the village of the same name.

The calculated values of potential ecological risk index factor and potential ecological hazard index do not exceed the values corresponding to low ecological risk.

3.2.2. REE Pollution

Based on the data of chemical analysis of samples of bottom sediments of lakes, enrichment factor, geoaccumulation index, potential ecological risk index factor and potential ecological hazard index were calculated to assess the contamination of BS with REEs (Table 5).

According to the calculated EF values, minor enrichment of Lake Krivoe BS was revealed with LREEs and part of the heavy group REEs (HREEs) as well as Lake Ilma BS with all the elements listed in the table; however, large calculated values are typical for the LREEs, the northern part of Lake Lovozero BS with LREE light group and Lake Lovozero at the mouth of the Sergevan River BS with all the listed elements. A higher level of enrichment of samples of BS with REEs was noted for Lake Ilma.

According to the calculated value of I_{geo}, the pollution of the BS of Lake Krivoe and Lake Lovozero at the mouth of the Sergevan River can be classified as unpolluted to moderately polluted with La, Ce, Pr, BS of Lake Ilma with higher calculated I_{geo} values for La-Sm.

The calculated values of potential ecological risk index factor and potential ecological hazard index for Lake Ilma and samples taken in Lake Lovozero at the mouth of the Sergevan River approach the lower limit of values corresponding to low ecological risk but do not cross it.

Table 6 shows the contents of REEs in the upper layers of BS (0–9 cm) of lakes Ilma and Lovozero, Clarke numbers of elements in the Earth's crust, limits of contents in BS of freshwater ecosystems and average contents in BS of Lake Imandra, which are subject to anthropogenic pollution from an enterprise developing a deposit of apatite-nepheline ores, which also include REEs [32].

As can be seen, the average values of the REE contents in the BS of the lakes Ilma and Lovozero exceed by many times the Clarke numbers of elements in the Earth's crust, the limits of content in the BS of freshwater ecosystems and the average content in the BS of Lake Imandra. The average contents of REEs in the BS of the studied lakes Krivoe, Ilma and Lovozero are distributed in accordance with the Oddo–Harkins rule: lanthanides with even ordinal numbers are more abundant in the Earth's crust than those with odd ones [58].

The vertical distribution of the content of REEs in the selected samples of BS of lakes is shown in Figure 3.

As can be seen, the lowest REE contents are characteristic of the BS of Lake Krivoe. However, in the last 20–30 years (the upper layers of the column), a sharp increase in these indicators can be noted. This may be associated with the commissioning of the second field of the tailings of the enrichment plant about 30 years ago. In our previous work, we carried out a study devoted to modeling the atmochemical halo of dust dispersion carried outside the first field of the tailing dump [59]. It is logical to assume that with the advancement of potential sources of dusting to the north and an increase in dusty areas due to the storage of pulp in tailings open to the atmosphere, dust can spread over greater distances than in the original model.

Table 5. Concentrations of REEs in the surface and background layers of BS (mg/kg), enrichment factor, geoaccumulation index, potential ecological risk index factor and potential ecological hazard index.

Lake	Layer, cm	La	Ce	Pr	Nd	Sm	Eu	Gd	
Krivoe	0–7	$\frac{16.05 \pm 7.71}{13.15\ (10.09 - 29.51)}$	$\frac{32.67 \pm 15.57}{26.44\ (20.34 - 60.16)}$	$\frac{3.64 \pm 2.36}{2.76\ (1.85 - 7.65)}$	$\frac{10.91 \pm 5.00}{8.98\ (7.17 - 19.65)}$	$\frac{1.86 \pm 0.51}{2.13\ (1.27 - 2.93)}$	$\frac{0.75 \pm 0.18}{0.72\ (0.49 - 1.03)}$	$\frac{2.06 \pm 0.21}{2.17\ (1.71 - 2.28)}$	
	13–14	$\frac{10.30 \pm 0.37}{10.21\ (9.95 - 10.47)}$	$\frac{21.53 \pm 0.02}{20.87\ (20.86 - 20.89)}$	$\frac{2.20 \pm 0.59}{2.09\ (1.67 - 2.51)}$	$\frac{7.64 \pm 1.78}{8.52\ (7.26 - 9.78)}$	$\frac{1.25 \pm 0.61}{2.07\ (1.24 - 2.50)}$	$\frac{0.66 \pm 0.06}{0.63\ (0.59 - 0.67)}$	$\frac{1.40 \pm 0.68}{2.02\ (1.34 - 2.50)}$	
	EF	1.56	1.52	1.65	1.43	1.49	1.13	1.47	
	I_{geo}	0.02	0.01	0.04	−0.02	0.00	−0.12	−0.01	
	E_{ir}	1.56	1.52	8.25	2.86	7.45	11.27	7.33	
Ilma	0–7	$\frac{123.27 \pm 140.73}{64.99\ (33.55 - 431.32)}$	$\frac{244.83 \pm 265.90}{139.75\ (81.11 - 826.18)}$	$\frac{28.42 \pm 31.42}{17.20\ (6.90 - 96.36)}$	$\frac{82.04 \pm 80.80}{53.81\ (25.80 - 257.09)}$	$\frac{14.92 \pm 10.17}{12.63\ (5.89 - 35.40)}$	$\frac{3.44 \pm 2.22}{3.58\ (1.03 - 5.91)}$	$\frac{9.04 \pm 3.10}{8.82\ (5.33 - 14.41)}$	
	15–16	$\frac{41.69 \pm 1.75}{49.22\ (40.98 - 50.46)}$	$\frac{92.41 \pm 2.08}{109.50\ (88.02 - 110.97)}$	$\frac{10.59 \pm 0.37}{12.88\ (10.62 - 13.14)}$	$\frac{37.93 \pm 1.43}{42.81\ (31.80 - 43.82)}$	$\frac{8.95 \pm 0.96}{11.09\ (8.40 - 11.77)}$	$\frac{2.90 \pm 0.07}{3.17\ (2.82 - 3.22)}$	$\frac{7.16 \pm 0.19}{8.18\ (7.05 - 8.31)}$	
	EF	2.96	2.65	2.68	2.16	1.67	1.19	1.26	
	I_{geo}	0.29	0.25	0.25	0.16	0.05	−0.10	−0.07	
	E_{ir}	2.96	2.65	13.42	4.33	8.34	11.86	6.31	
Lovozero, northern part	0–7	$\frac{94.32 \pm 30.70}{73.59\ (71.70 - 144.18)}$	$\frac{178.19 \pm 61.75}{139.42\ (130.63 - 277.79)}$	$\frac{21.83 \pm 6.54}{19.05\ (14.27 - 32.82)}$	$\frac{69.57 \pm 12.90}{64.56\ (56.58 - 90.88)}$	$\frac{14.01 \pm 1.07}{14.23\ (12.59 - 15.86)}$	$\frac{3.92 \pm 0.52}{3.63\ (3.33 - 4.54)}$	$\frac{9.87 \pm 3.81}{8.15\ (6.26 - 14.38)}$	
	8–9	$\frac{72.24 \pm 0.72}{72.55\ (72.04 - 73.26)}$	$\frac{137.80 \pm 1.15}{136.61\ (136.60 - 139.42)}$	$\frac{18.42 \pm 0.10}{18.35\ (18.28 - 18.52)}$	$\frac{62.87 \pm 1.20}{63.72\ (61.87 - 64.56)}$	$\frac{13.65 \pm 0.57}{13.66\ (13.30 - 14.46)}$	$\frac{4.39 \pm 0.10}{4.36\ (4.35 - 4.53)}$	$\frac{14.25 \pm 0.09}{14.24\ (14.19 - 14.38)}$	
	EF	1.31	1.29	1.19	1.11	1.03	0.89	0.69	
	I_{geo}	−0.06	−0.06	−0.10	−0.13	−0.16	−0.23	−0.34	
	E_{ir}	1.31	1.29	5.93	2.21	5.13	8.93	3.46	
Lovozero, Sergevan	0–7	$\frac{163.75 \pm 62.63}{125.61\ (110.56 - 260.19)}$	$\frac{309.60 \pm 121.55}{234.27\ (205.74 - 498.86)}$	$\frac{37.37 \pm 17.55}{25.52\ (22.76 - 61.79)}$	$\frac{124.51 \pm 32.09}{106.07\ (96.06 - 172.52)}$	$\frac{27.50 \pm 5.97}{24.63\ (22.19 - 36.22)}$	$\frac{4.88 \pm 0.24}{4.89\ (4.46 - 5.09)}$	$\frac{14.46 \pm 2.18}{14.37\ (11.45 - 17.69)}$	
	8–9	$\frac{103.30 \pm 9.84}{103.21\ (101.95 - 117.22)}$	$\frac{191.88 \pm 20.57}{190.87\ (188.86 - 220.98)}$	$\frac{18.69 \pm 3.60}{19.09\ (16.67 - 23.79)}$	$\frac{89.32 \pm 7.92}{88.52\ (81.26 - 100.53)}$	$\frac{21.00 \pm 1.27}{20.06\ (19.64 - 22.80)}$	$\frac{4.61 \pm 0.12}{4.63\ (4.59 - 4.78)}$	$\frac{10.18 \pm 1.20}{10.12\ (9.54 - 11.88)}$	
	EF	1.59	1.61	2.00	1.39	1.31	1.06	1.42	
	I_{geo}	0.02	0.03	0.12	−0.03	−0.06	−0.15	−0.02	
	E_{ir}	1.59	1.61	10.00	2.79	6.55	10.58	7.10	
Lake	**Layer, cm**	**Tb**	**Dy**	**Ho**	**Er**	**Tm**	**Yb**	**Lu**	**RI**

Table 5. *Cont.*

Lake	Layer, cm	La	Ce	Pr	Nd	Sm	Eu	Gd	
Krivoe	0–7	$\frac{0.33 \pm 0.08}{0.33\ (0.20-0.42)}$	$\frac{2.09 \pm 0.39}{2.34\ (1.99-3.00)}$	$\frac{0.41 \pm 0.08}{0.43\ (0.37-0.57)}$	$\frac{1.31 \pm 0.27}{1.14\ (1.07-1.68)}$	$\frac{0.17 \pm 0.04}{0.17\ (0.15-0.25)}$	$\frac{1.16 \pm 0.19}{1.14\ (0.96-1.46)}$	$\frac{0.15 \pm 0.03}{0.18\ (0.14-0.22)}$	-
	13–14	$\frac{0.32 \pm 0.12}{0.34\ (0.25-0.42)}$	$\frac{2.19 \pm 0.44}{2.16\ (1.85-2.47)}$	$\frac{0.52 \pm 0.08}{0.47\ (0.41-0.52)}$	$\frac{1.35 \pm 0.08}{1.33\ (1.28-1.39)}$	$\frac{0.18 \pm 0.03}{0.20\ (0.18-0.22)}$	$\frac{1.25 \pm 0.08}{1.22\ (1.16-1.27)}$	$\frac{0.21 \pm 0.00}{0.19\ (0.19-0.22)}$	-
	EF	1.04	0.96	0.80	0.97	0.96	0.93	0.73	-
	I_{geo}	−0.16	−0.20	−0.27	−0.19	−0.20	−0.21	−0.32	-
	E_{ir}	10.39	4.78	8.00	4.85	9.56	4.63	14.52	96.97
Ilma	0–7	$\frac{1.42 \pm 0.45}{1.48\ (0.83-2.16)}$	$\frac{8.50 \pm 2.57}{7.69\ (5.22-11.97)}$	$\frac{1.68 \pm 0.43}{1.73\ (1.13-2.21)}$	$\frac{4.67 \pm 1.00}{5.01\ (3.02-5.72)}$	$\frac{0.73 \pm 0.20}{0.80\ (0.49-0.97)}$	$\frac{4.23 \pm 1.00}{4.36\ (2.90-5.44)}$	$\frac{0.64 \pm 0.15}{0.66\ (0.42-0.85)}$	-
	15–16	$\frac{1.21 \pm 0.14}{1.22\ (1.13-1.32)}$	$\frac{6.94 \pm 0.22}{6.12\ (5.97-8.28)}$	$\frac{1.64 \pm 0.03}{1.70\ (1.58-1.82)}$	$\frac{4.64 \pm 0.04}{4.65\ (4.61-4.76)}$	$\frac{0.64 \pm 0.01}{0.70\ (0.60-0.72)}$	$\frac{3.68 \pm 0.05}{4.01\ (3.57-4.07)}$	$\frac{0.57 \pm 0.01}{0.59\ (0.56-0.62)}$	-
	EF	1.17	1.22	1.02	1.01	1.15	1.15	1.12	-
	I_{geo}	−0.11	−0.09	−0.17	−0.17	−0.12	−0.12	−0.13	-
	E_{ir}	11.68	6.12	10.22	5.03	11.50	5.75	22.36	122.50
Lovozero, northern part	0–7	$\frac{1.56 \pm 0.64}{1.20\ (0.91-2.38)}$	$\frac{8.76 \pm 3.23}{7.10\ (5.82-12.40)}$	$\frac{1.83 \pm 0.81}{1.35\ (1.07-2.78)}$	$\frac{5.20 \pm 2.44}{3.79\ (2.77-8.06)}$	$\frac{0.81 \pm 0.44}{0.50\ (0.45-1.41)}$	$\frac{4.68 \pm 2.23}{3.19\ (2.63-7.29)}$	$\frac{0.73 \pm 0.35}{0.47\ (0.43-1.20)}$	-
	8–9	$\frac{2.27 \pm 0.13}{2.21\ (2.09-2.37)}$	$\frac{12.23 \pm 0.12}{12.21\ (12.15-12.40)}$	$\frac{2.49 \pm 0.19}{2.63\ (2.29-2.66)}$	$\frac{8.24 \pm 0.13}{8.25\ (8.16-8.34)}$	$\frac{1.15 \pm 0.00}{1.14\ (1.14-1.16)}$	$\frac{6.85 \pm 0.08}{6.81\ (6.78-6.92)}$	$\frac{1.09 \pm 0.03}{1.07\ (1.05-1.12)}$	-
	EF	0.69	0.72	0.74	0.63	0.70	0.68	0.67	-
	I_{geo}	−0.34	−0.32	−0.31	−0.38	−0.33	−0.34	−0.35	-
	E_{ir}	6.86	3.58	7.35	3.16	7.03	3.42	13.41	73.07
Lovozero, Sergevan	0–7	$\frac{2.07 \pm 0.15}{2.09\ (1.81-2.28)}$	$\frac{11.18 \pm 1.11}{11.41\ (9.55-12.59)}$	$\frac{2.17 \pm 0.33}{2.13\ (1.79-2.65)}$	$\frac{5.77 \pm 1.14}{5.47\ (4.44-7.61)}$	$\frac{0.86 \pm 0.12}{0.80\ (0.74-1.07)}$	$\frac{5.48 \pm 0.95}{5.16\ (4.23-6.94)}$	$\frac{0.80 \pm 0.12}{0.79\ (0.61-0.97)}$	-
	8–9	$\frac{1.71 \pm 0.10}{1.74\ (1.65-1.86)}$	$\frac{9.67 \pm 1.01}{9.16\ (8.85-11.10)}$	$\frac{2.00 \pm 0.04}{2.01(1.91-2.04)}$	$\frac{5.09 \pm 0.38}{5.13\ (4.98-5.63)}$	$\frac{0.72 \pm 0.04}{0.70\ (0.58-0.78)}$	$\frac{4.60 \pm 0.45}{4.42\ (4.16-5.24)}$	$\frac{0.71 \pm 0.12}{0.70\ (0.69-0.88)}$	-
	EF	1.21	1.16	1.08	1.13	1.19	1.19	1.13	-
	I_{geo}	−0.09	−0.11	−0.14	−0.12	−0.10	−0.10	−0.12	-
	E_{ir}	12.12	5.78	10.80	5.67	11.95	5.95	22.65	115.13

Note: The numerator is mean value ± standard deviation; the denominator is the median (the minimum and maximum values of the sample). Lovozero, Sergevan—Lake Lovozero, the mouth of the River Sergevan.

Table 6. The content of REEs in BS (layer 0–9 cm) of the studied lakes and other objects, mg/kg.

Element	BS Lake Ilma	BS Lake Lovozero (Sergevan)	BS Lake Krivoe	Clarke Number of Elements in the Earth's Crust	Limit of Contents in BS of Freshwater Ecosystems	BS Lake Imandra
La	$\frac{107.31 \pm 125.94}{59.45\ (33.55 - 431.32)}$	$\frac{151.86 \pm 59.25}{122.08\ (103.30 - 260.19)}$	$\frac{14.74 \pm 7.16}{10.47\ (9.89 - 29.51)}$	30.00	19.5–100	103.00
Ce	$\frac{215.80 \pm 237.39}{120.36\ (81.11 - 826.18)}$	$\frac{286.67 \pm 114.91}{231.57\ (191.88 - 498.86)}$	$\frac{30.11 \pm 14.42}{21.86\ (20.34 - 60.16)}$	50.00	43–100	172.00
Pr	$\frac{25.07 \pm 28.01}{14.20\ (6.90 - 96.36)}$	$\frac{33.79 \pm 16.83}{24.44\ (18.69 - 61.79)}$	$\frac{3.38 \pm 2.11}{2.51\ (1.85 - 7.65)}$	5.00	8–8.3	21.00
Nd	$\frac{73.65 \pm 71.94}{46.80\ (25.80 - 257.09)}$	$\frac{117.94 \pm 30.83}{103.73\ (89.32 - 172.52)}$	$\frac{10.38 \pm 4.50}{8.98\ (7.17 - 19.65)}$	23.00	19–44	79.00
Sm	$\frac{14.23 \pm 8.91}{11.87\ (5.89 - 35.40)}$	$\frac{26.25 \pm 5.75}{24.53\ (21.00 - 36.22)}$	$\frac{2.08 \pm 0.48}{2.13\ (1.27 - 2.93)}$	6.50	3.3–30	12.00
Eu	$\frac{3.39 \pm 1.93}{3.33\ (1.03 - 5.91)}$	$\frac{4.84 \pm 0.22}{4.87\ (4.46 - 5.09)}$	$\frac{0.73 \pm 0.16}{0.69\ (0.49 - 1.03)}$	1.00	0.07–12.2	3.10
Gd	$\frac{8.88 \pm 2.70}{8.58\ (5.33 - 14.41)}$	$\frac{13.70 \pm 2.46}{13.24\ (10.18 - 17.69)}$	$\frac{2.10 \pm 0.23}{2.17\ (1.71 - 2.50)}$	6.50	5.0–6.0	12.00
Tb	$\frac{1.38 \pm 0.40}{1.41\ (0.83 - 2.16)}$	$\frac{2.01 \pm 0.19}{2.02(1.71 - 2.28)}$	$\frac{0.35 \pm 0.07}{0.36\ (0.20 - 0.42)}$	0.90	0.3–1.1	1.20
Dy	$\frac{8.50 \pm 2.24}{7.97\ (5.22 - 11.97)}$	$\frac{11.01 \pm 1.08}{11.10\ (9.55 - 12.59)}$	$\frac{2.38 \pm 0.35}{2.34\ (1.99 - 3.00)}$	4.50	1.8–4.5	6.70
Ho	$\frac{1.71 \pm 0.38}{1.78\ (1.13 - 2.21)}$	$\frac{2.12 \pm 0.30}{2.00\ (1.79 - 2.65)}$	$\frac{0.47 \pm 0.07}{0.45\ (0.37 - 0.57)}$	1.00	0.9–1.0	1.10
Er	$\frac{4.70 \pm 0.87}{4.86\ (3.02 - 5.72)}$	$\frac{5.68 \pm 1.01}{5.47\ (4.44 - 7.61)}$	$\frac{1.31 \pm 0.24}{1.20\ (1.07 - 1.68)}$	2.50	2.6–3.0	3.00
Tm	$\frac{0.73 \pm 0.17}{0.77\ (0.49 - 0.97)}$	$\frac{0.83 \pm 0.11}{0.79\ (0.72 - 1.07)}$	$\frac{0.19 \pm 0.03}{0.18\ (0.15 - 0.25)}$	0.25	0.40	0.37
Yb	$\frac{4.26 \pm 0.87}{4.36\ (2.90 - 5.44)}$	$\frac{5.35 \pm 0.87}{5.16\ (4.23 - 6.94)}$	$\frac{1.16 \pm 0.17}{1.14\ (0.96 - 1.46)}$	3.00	1.4–4.4	2.30
Lu	$\frac{0.64 \pm 0.13}{0.66\ (0.42 - 0.85)}$	$\frac{0.80 \pm 0.12}{0.79\ (0.61 - 0.97)}$	$\frac{0.18 \pm 0.03}{0.19\ (0.14 - 0.22)}$	0.70	0.2–0.5	0.29

Note: The numerator is mean value ± standard deviation; the denominator is the median (the minimum and maximum values of the sample).

In BS Lake Ilma there is a regular increase in the content of REEs in the upper layers, which corresponds to the beginning of the enterprise in the 1960s.

Note that Lovozero contains more REEs in its water than Lake Ilma, while the ratio of the contents of REEs to the background values in the BS of Lake Ilma exceeds the same parameter for Lake Lovozero. This difference allows us to assume a different nature of the input of elements into water bodies, with wastewater in dissolved form in Lake Lovozero and due to dusting of fine-grained enrichment tailings in Lake Ilma.

Assessing the total pollution with TEs and REEs of BS of lakes Ilma and Lovozero at the mouth of the Sergevan River, the value of potential ecological risk reaches 174 and 186 units, respectively, which indicates the level of moderate ecological risk pollution.

3.3. Correlation Matrix

In order to establish possible relationships between metals and determine the main sources of metals in the bottom sediments of lakes, the Pearson correlation coefficient was calculated (Table 7) [60,61].

In BS samples from Lake Ilma, there is a strong positive correlation between LREEs (r = 0.97–0.99), which indicates a point source of anthropogenic influence [49]. The same elements are included in the composition of the tailings of the enrichment of loparite ores, which suggests that the lake is polluted due to dusting of the tailings.

A positive correlation between Cu and Ni (r = 0.958) also points to a one-time input of these elements. As mentioned above, this may be due to the activity of large copper-nickel enterprises in the region [55].

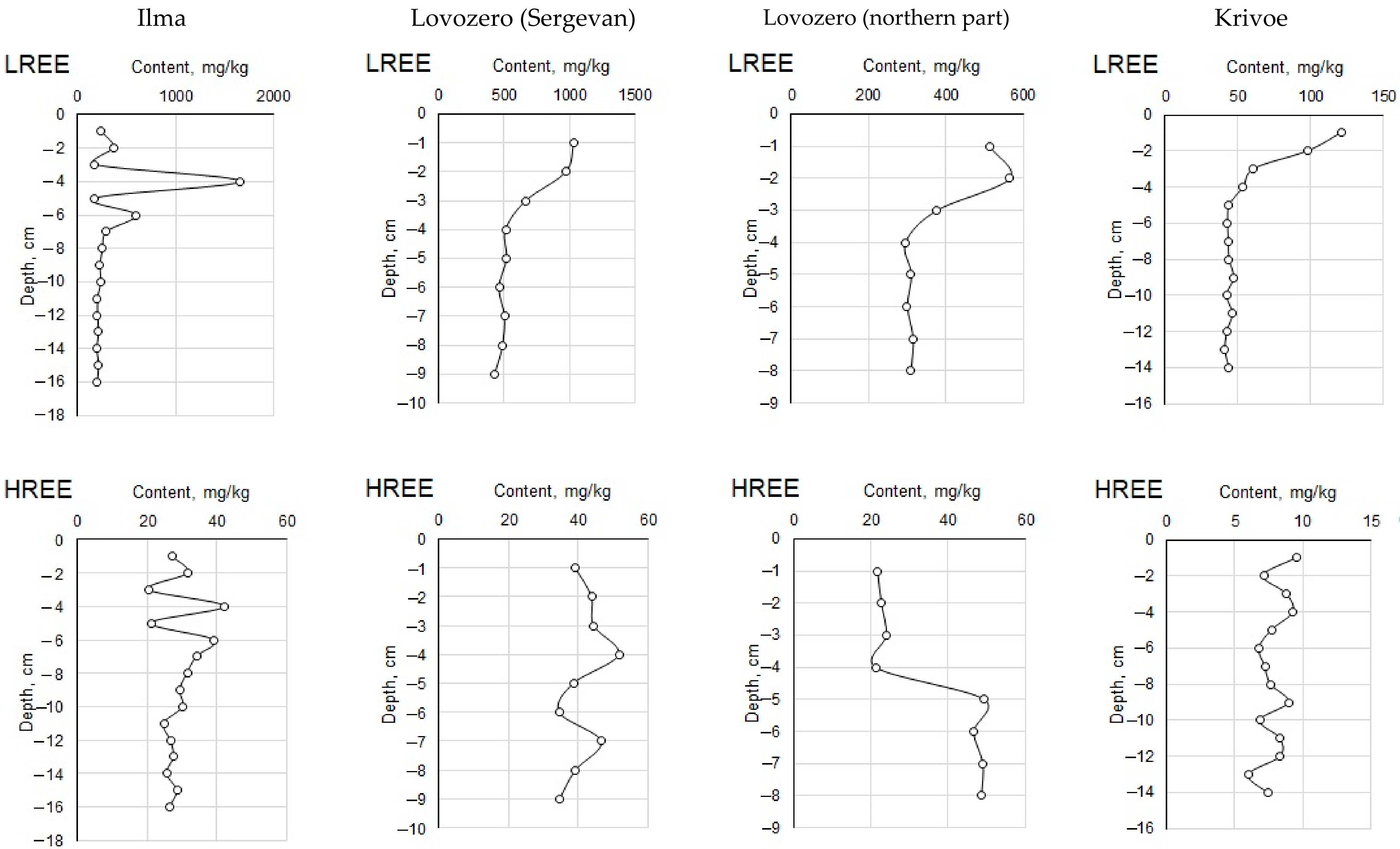

Figure 3. The vertical distribution of the content of REEs (LREEs and HREEs) in the selected samples of BS of lakes.

Table 7. Matrix of Pearson correlation coefficients for BS of lakes Ilma (lower triangle on the left) and Lovozero, Sergevan River (upper triangle on the right).

Variable	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Cu	Ni	Zn	Cr	Cd	Pb	Sr	Mn	Fe	OC
La	1.000	1.000	0.986	0.999	0.958	0.281	0.033	0.075	−0.172	−0.283	−0.204	−0.412	−0.057	−0.091	0.863	−0.045	−0.927	−0.620	−0.570	0.973	0.900	0.874	0.783	0.962
Ce	0.999	1.000	0.986	0.999	0.958	0.277	0.037	0.079	−0.169	−0.278	−0.194	−0.404	−0.052	−0.086	0.861	−0.049	−0.925	−0.624	−0.567	0.974	0.900	0.879	0.782	0.964
Pr	0.997	0.998	1.000	0.985	0.908	0.206	0.048	0.137	−0.100	−0.196	−0.156	−0.396	−0.079	−0.096	0.804	−0.116	−0.873	−0.604	−0.608	0.966	0.872	0.844	0.818	0.958
Nd	0.998	0.998	1.000	1.000	0.962	0.311	0.045	0.085	−0.159	−0.278	−0.208	−0.409	−0.048	−0.080	0.870	−0.023	−0.929	−0.603	−0.579	0.972	0.906	0.868	0.787	0.954
Sm	0.975	0.974	0.983	0.984	1.000	0.432	0.139	0.073	−0.172	−0.308	−0.208	−0.329	0.090	−0.004	0.950	0.173	−0.960	−0.564	−0.602	0.900	0.898	0.836	0.692	0.910
Eu	0.508	0.551	0.525	0.517	0.506	1.000	0.494	0.425	0.370	0.153	0.135	0.246	0.529	0.578	0.422	0.533	−0.350	0.185	−0.204	0.274	0.484	0.343	0.290	0.063
Gd	0.896	0.892	0.915	0.913	0.967	0.417	1.000	0.875	0.858	0.804	0.801	0.842	0.959	0.884	0.115	0.327	−0.031	0.108	−0.550	−0.061	0.103	0.095	0.079	0.014
Tb	0.856	0.845	0.870	0.870	0.928	0.319	0.982	1.000	0.960	0.914	0.821	0.760	0.763	0.779	0.018	0.210	0.104	0.279	−0.393	0.080	0.234	0.233	0.370	0.005
Dy	0.738	0.741	0.773	0.770	0.855	0.418	0.941	0.931	1.000	0.972	0.844	0.834	0.759	0.808	−0.214	0.174	0.313	0.399	−0.289	−0.171	−0.021	−0.031	0.146	−0.236
Ho	0.557	0.559	0.594	0.593	0.722	0.344	0.854	0.857	0.947	1.000	0.889	0.862	0.708	0.741	−0.349	0.069	0.440	0.388	−0.224	−0.281	−0.165	−0.125	0.046	−0.295
Er	0.628	0.628	0.651	0.649	0.757	0.449	0.863	0.888	0.891	0.940	1.000	0.932	0.807	0.855	−0.340	−0.132	0.303	0.079	−0.121	−0.215	−0.171	0.041	−0.083	−0.169
Tm	0.638	0.640	0.663	0.664	0.784	0.425	0.875	0.869	0.899	0.964	0.962	1.000	0.887	0.880	−0.375	0.137	0.422	0.288	−0.067	−0.448	−0.311	−0.179	−0.280	−0.397
Yb	0.289	0.281	0.308	0.312	0.467	0.155	0.620	0.664	0.689	0.874	0.876	0.902	1.000	0.932	0.051	0.314	−0.006	0.069	−0.383	−0.152	0.010	0.068	−0.097	−0.071
Lu	0.304	0.299	0.328	0.328	0.484	0.198	0.640	0.670	0.692	0.874	0.890	0.906	0.987	1.000	−0.125	0.088	0.063	0.063	−0.219	−0.138	−0.035	0.078	−0.100	−0.147
Cu	0.201	0.186	0.143	0.146	0.049	0.072	−0.096	−0.067	−0.388	−0.431	−0.132	−0.216	−0.226	−0.194	1.000	0.422	−0.888	−0.376	−0.630	0.790	0.867	0.699	0.663	0.811
Ni	0.139	0.118	0.078	0.083	−0.040	−0.124	−0.199	−0.168	−0.498	−0.584	−0.334	−0.402	−0.419	−0.399	0.958	1.000	−0.019	0.585	−0.205	−0.082	0.269	−0.040	0.159	−0.160
Zn	0.002	0.033	0.038	0.021	0.048	0.373	0.039	−0.113	−0.012	0.025	−0.004	0.061	−0.052	0.083	−0.040	−0.123	1.000	0.719	0.599	−0.839	−0.764	−0.721	−0.524	−0.908
Cr	0.164	0.150	0.102	0.110	−0.016	−0.029	−0.214	−0.222	−0.518	−0.600	−0.388	−0.391	−0.445	−0.435	0.918	0.960	−0.038	1.000	0.364	−0.521	−0.243	−0.449	−0.065	−0.751
Cd	−0.784	−0.809	−0.792	−0.794	−0.781	−0.755	−0.653	−0.526	−0.556	−0.440	−0.434	−0.552	−0.201	−0.214	−0.034	0.064	−0.306	−0.116	1.000	−0.406	−0.421	−0.250	−0.387	−0.614
Pb	0.363	0.347	0.303	0.308	0.178	0.112	−0.002	0.024	−0.293	−0.428	−0.155	−0.240	−0.349	−0.337	0.947	0.953	−0.185	0.923	−0.147	1.000	0.931	0.924	0.858	0.905
Sr	0.497	0.491	0.446	0.449	0.331	0.298	0.137	0.120	−0.171	−0.304	−0.043	−0.088	−0.263	−0.232	0.921	0.889	0.016	0.907	−0.375	0.962	1.000	0.903	0.911	0.780
Mn	0.162	0.137	0.113	0.111	0.021	−0.143	−0.074	−0.026	−0.365	−0.425	−0.150	−0.266	−0.257	−0.201	0.933	0.938	−0.009	0.827	0.161	0.876	0.815	1.000	0.798	0.806
Fe	0.257	0.242	0.217	0.214	0.142	0.018	0.036	0.028	−0.280	−0.318	−0.067	−0.126	−0.170	−0.086	0.904	0.870	0.277	0.820	−0.063	0.810	0.839	0.941	1.000	0.668
OC	−0.076	−0.111	−0.126	−0.125	−0.184	−0.396	−0.210	−0.109	−0.435	−0.424	−0.171	−0.309	−0.160	−0.126	0.821	0.839	−0.191	0.675	0.466	0.723	0.590	0.934	0.801	1.000

Note: OC—organic carbon, defined as the loss on ignition of the samples.

Moderate or strong positive correlations of organic carbon with a number of metals—Cu, Ni, Cr, Cd, Pb, Sr, Mn, but not REE—indicate the formation of organic complexes with these metals [62]. Fe shows a significant strong correlation with the same metals, which indicates their similarity, identical behavior and the fact that one of the main phases for these metals is complex compounds with iron oxyhydroxide [61,63].

In BS samples from Lake Lovozero (the mouth of the Sergevan River), a similar pattern is observed: there is a strong positive correlation for LREEs (r = 0.97–1.0). However, in contrast to Lake Ilma, judging by the strong correlation of organic carbon with LREEs, it can be concluded that organic carbon is one of the main geochemical carriers of LREEs in this area [61].

Moderate or strong positive correlations of organic carbon were also found for Cu, Pb, Sr and Mn, which indicates the formation of organic complexes with these metals [62]. A significant correlation of the same elements with Fe also indicates their similar behavior and indicates the role of iron oxyhydroxide as one of the main phases for these metals [63].

The toxic effect of lanthanides for marine unicellular algae was established in [64]. In living organisms, REEs can form chelate compounds with substances involved in metabolism, in particular, nucleic acids and amino acids [65]. HREEs, according to the literature data, are capable of replacing calcium ions in organisms [66,67], including enzymes. In addition, rare earth elements have an anticoagulant effect [68]. The question of the carcinogenic effect of REEs has not yet been resolved [69].

4. Conclusions

Based on the results of the study, elevated contents of TEs and REEs, which are part of the tailings of rare metal ores, were revealed in water samples and BS of lakes Krivoe, Ilma and Lovozero. The total content of elements was established both in the surface layers of BS and in deep layers, which were formed in the pre-industrial period and, thus, characterize the geochemical background of the study area.

The obtained results testify to the susceptibility of the lakes Ilma and Lovozero to strong anthropogenic pollution of various nature—dusting of tailings and discharge of wastewater from the enterprise. The conducted correlation analysis confirmed the single source of LREE inflow to the subsidiaries Ilma and Lovozero. At the same time, differences were revealed in the forms of occurrence of a number of TEs and REEs in the selected samples of BS, which may indicate a different route of pollutant entry. An increase in the content of LREEs in the upper layers of the BS of Lake Krivoe is indicative of a relatively recent anthropogenic impact. Of particular relevance to the study is the fact that Lake Lovozero belongs to the reservoirs of fishery value of the highest category, and one of the types of economic activity of the indigenous people of the Lovozero region is fishing.

The problem of technogenic pollution of environmental components with REEs is relevant for Russia, China, the USA, Australia and other countries [70]. Currently, the US Environmental Protection Agency does not classify any rare earth elements as carcinogens; however, there is no conclusion that they are not carcinogenic [69].

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