Article

Spatial Variability of Metals in Coastal Sediments of Elćkie Lake (Poland)

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Abstract: This study aimed to determine the content and spatial distribution of metals (Ca, Mg, Fe, Na, K, Mn, Zn, Cr, Cu, Pb, Co) in sediments in the coastal zone of Elćkie Lake located in the area of "Green Lungs of Poland" in the north-eastern part of the country, depending on the land use (urban area, agricultural and forest area, and beaches). The concentration of metals was determined using atomic absorption spectrometry. The average contents of major elements in 28 sediment samples occurred in the following order: Ca > Mg > Fe > Na > K > Mn. The order of these elements in the coastal sediments located within the different parts of the catchment was identical. These elements may originate from natural sources such as the Earth's crust, soil, and wind-blown dust from unpaved roads. The average contents of potentially toxic elements (PTEs) in the sediments were as follows: Cr > Zn > Pb > Cu > Co in agricultural and forest areas and beaches (the exception was Cu for beach B, which occurred at the end of the series). A different pattern occurred in urbanized areas: Zn > Cr > Cu > Pb > Co. The spatial distribution of heavy metals in the sediments indicated the highest contents in the shoreline adjacent to the urbanized part of the catchment. The primary sources of metals in sediment are transportation, coal burning, sanitary sewage from unsewered developments on the lakeshore, and storm runoff from roads. This was confirmed by positive correlations of Zn with Cu ($r = 0.58$), Pb ($r = 0.90$), Fe ($r = 0.40$). No correlations between the studied metals and organic matter were found, which may indicate its insignificant influence on metal content in the sediments. Pearson correlation coefficients also showed no relationship between sediment pH and the presence of metals. Factor analysis (FA) indicated that lithogenic (geogenic) and anthropogenic factors have almost equal shares in the distribution of most of the metals studied. The analysis of variance (ANOVA) showed that the average contents of Zn, Cu, Co, and Na in the sediments from urbanized areas are statistically significantly higher than the sediments from other areas (rural/forest, beaches).

Keywords: coastal sediments; metals; sources of pollution

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1. Introduction

The ongoing intensification of human activity and scientific and technological progress have increased the number of processes affecting the environment. These often lead to adverse changes in water, soil, or air quality, thus affecting humans, animals, and plants [1,2]. Aquatic ecosystems located in urbanized areas are the final link in the transport of pollutants such as heavy metals and biogenic elements [3]. Lakes in depressions of the land are natural receivers of pollutants from the catchment area. Lake sediments have a high capacity to retain various pollutants, whether organic, chemical, mineral, or physical, and they also carry these pollutants and can be their secondary source [4,5]. For this reason, they are excellent indicators of the state of the aquatic environment, and they also play a significant role in shaping the chemical composition of surface waters [6]. Heavy metals are among the most common pollutants that adversely affect aquatic ecosystems [7], causing permanent biogeochemical effects [8]. They are not biodegradable, and their mobility and

bioavailability depend on many factors—including temperature, pressure, pH, salinity, chemical form, redox conditions, and organic matter content [9]. Considering water pollution, the crucial trace and toxic elements are Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn. Some of these metals (e.g., Co, Cr, Cu, Ni, and Zn) are also essential for the development and functioning of organisms but are toxic at high concentrations [10].

The content of heavy metals in sediments is an excellent tool to estimate changes in catchment areas and allows assessment of the severity of anthropopressure within the reservoir [11]. Contamination of lakes with metals depends considerably on the degree of urbanization of the area, the intensity of reservoir use, agricultural production, and emission of gases and dust, also from natural sources, introduced into the sediments at a point or in a diffuse manner [12]. The most polluted lakes are urban ones. Crucial metal sources in such lakes include vehicle emissions, atmospheric pollution, stormwater runoff [13], coal combustion [14], and accompanying resuspension and surface runoff. Roads and vehicle traffic are likely some of the most significant sources of metals in the environment [15]. Urban dust is more contaminated with metals than highway dust due to different driving conditions (there is a higher frequency of braking in cities, which causes additional contamination of road dust with metals through, among other things, wear of brake pads) [16]. High loading of heavy metals in urban environments notably occurs among impervious surfaces (urban sidewalks, roadways) [17].

The subject of this paper was the study of sediments collected in the coastal zone of Elckie Lake, located in the north-eastern part of Poland, in the area of the “Green Lungs of Poland”. The human activities associated with tourism and the impact of urban development expose the lake ecosystem. To our knowledge, there are no comprehensive studies on the content of metals in the sediments of this lake. Analyses of the contents of nutrients in water and sediments have already been performed [18]. The pressure from the tourist traffic and the influence of the urbanized part of the catchment may cause degradation of the natural values of the lake.

Due to the unquestionable importance of water resources, the protection and sustainable management of aquatic ecosystems is a priority in environmental programs worldwide. Polish water resources are below the European average, and they are exposed to pollution from agriculture, industry, and tourism; hence, it is essential to protect the existing water reservoirs. The crucial issue in the protection of freshwater reservoirs is the implementation of appropriate protection measures within the catchment area, allowing for a radical reduction in pollution loads introduced not only from point sources but also from non-point sources.

The objectives of the study were to: (1) determine the content and spatial distribution of metals (Ca, Mg, Fe, Na, K, Mn, Zn, Cr, Cu, Pb, Co) in sediments in the coastal zone of Elckie Lake depending on land use (urbanized area, agricultural–forest area, and beach); (2) determine the potential environmental risk of heavy metals (Zn, Cr, Cu, Pb, Co) based on sediment classification using geochemical indices: geoaccumulation index (Igeo), contamination factor (CF) and pollution load index (PLI); and (3) attempt to identify the sources of metal pollution by multivariate statistical analysis and correlation analysis. This study also compared the metal pollution of Elckie Lake with other lakes in the world by catchment type. The results provide new information on the heavy metal contents that will allow the identification and classification of the sources of the studied metals within the lake, thus contributing to the knowledge and better management of this region in the future. The study may be applicable to finding a proper solution between economic development, population needs, and protection of the natural values of the lake ecosystem.

2. Materials and Methods

2.1. Study Area

Elckie Lake (5348′24″ N 2220′58″ E) (Figure 1) is in the central part of the Elk Lake District, within the Elk municipal district (Poland). There are three parts of the lake: the upper, the middle, and the southern. An artificial peninsula and a road bridge isolate

the upper and middle sections, whereas the middle and southern parts are separated by narrowing and shallows. The reservoir area is 382.4 hectares.

Figure 1. The location of 28 research points on Elckie Lake, Poland.

In general, reservoirs with a large surface area are less susceptible to degradation than small ones. Furthermore, the higher the water volume in a lake, the less susceptible it is to degradation. The volume of Elckie Lake is 57,420.3 thousand m^3 . In addition, the resistance to biodegradation of a lake increases with the reservoirs' depth. The average depth of Elckie Lake is 15.0 m. The length of the shoreline determines the area of contact between the lake and the catchment; the longer the shoreline, the more pollutant migration paths generally connect the catchment and the reservoir. The shoreline length, the total catchment area, and the immediate catchment area of Elckie Lake are 18,650 m, 979.8 km^2 , and 315.1 hectares, respectively.

The land-use pattern of the catchment influences the degree of threat to the lake from external factors. The percentage shares of the individual land use forms in the Elckie Lake catchment area are as follows: urbanized land—37.3%, arable land—31.4%, forests—16.6%, crops and allotments—9.7%, arable land with a high proportion of natural vegetation—3.3%. Farmlands and forests lie on the western side of the lake, and it is clear that agriculture can significantly transform the natural environment. Non-point pollution reaches the water reservoir from cultivated fields along with surface runoff. On the eastern shoreline of the lake sit urbanized areas, i.e., the buildings of the city of Elk (population 61,523), from which most of the pollution probably originates. Elk is one of the most prominent tourist centers in the "Green Lungs of Poland" and is an excellent base for trips to surrounding towns. There are two municipal beaches by the lake, in the vicinity of which there are campsites,

sports fields, guarded bathing beaches, and water equipment rentals. Elk is a typical tourist town with only a small industry (wood, meat, and food processing). The eastern side of the lake is polluted mainly by the sanitary sewage from the unsewered residential estates located on the shores of the lake, rain sewage discharges, and traffic pollution (street dust from the abrasion of tires, brake linings, and asphalt, containing heavy metals).

The main climate characteristics are long winters, short springs, a relatively short vegetation season, high humidity (average annual 80%), and frequent fog. The average annual air temperature ranges from 4.8 to 8.3 °C. There is also relatively high precipitation, averaging 600–700 mm per year [19].

2.2. Sample Collection and Preparation

The study subjects were sediments (28 samples) collected in August 2019 in the coastal zone of Elckie Lake. The littoral zone of urban lakes is the most liable to the migration of pollutants from the catchment. Metals from the topsoil of the catchment are transported to waters by surface runoff and erosion and then accumulated on the sediment surface [20]. The number of samples and their location depended on land use; accessibility and anthropogenic activities around the lake were the main criteria for choosing research points [21]. There are four groups: (1) urbanized areas (points 1–6); (2) agricultural–forest areas (points 7–11), (3) beach B (points 12–18), and (4) beach A (points 19–28) (Figure 1). The points of the first group sit on the eastern shore—within the city, near shoreline outlets of storm drains, roads, and parking lots. The second includes points from the western, less urbanized shore of the lake. There are mainly fields, forested areas, and also single-family dwellings nearby. Most of the samples come from the beaches due to the prominent use of the lake’s tourist potential. Both beaches are infilled and have similar morphodynamics. Beach B is characterized by a much higher number of visitors due to the amenities of a large parking lot, water equipment rentals, playing fields, and changing rooms, which are not available at beach A.

Sediments were sampled in the shoreline area. Ten individual samples were collected from a depth of 0–5 cm, from beneath the water’s surface, at each designated point. The crucial layer where water–sediment interactions occur and biological activity is relatively high is that of surface sediments [22]. The collected material was blended to obtain a representative sample (approximately 1000 g) from each point. After the transportation of the samples to the laboratory, pH in the water was measured by the potentiometric method and then dried in an “air-dry” condition and stored for further analyses. Before determination organic matter content by ignition loss at 550 °C, samples were dried at 105 °C.

The analysis of the granulometric composition of the sediments was performed using the sieve method, which consisted of separating the sample of raw, air-dry sediments into granulometric fractions using an appropriate set of sieves (mesh sizes: 2, 1, 0.2, 0.1, and 0.063 mm) connected to a mechanical shaker. After sieving, the fractions from each sieve were weighed, and the percentage of sample masses from the respective sieves to the total sample mass was calculated.

Due to the lack of binding criteria in Poland, the quality of bottom sediments was assessed based on geochemical criteria and the classification of local sediments made by Bojakowska and Sokołowska [23]. The data they used came from studies of sediments in the <0.2 mm fraction.

One gram of dry sample (fraction <0.2 mm) was placed in Teflon dishes; 8 mL of HNO₃ and 2 mL of 30% H₂O₂ were added, and the sample was digested in a microwave digestion system (Ethos Easy, Milestone, Italy) according to the instructions. The samples were filtered and quantitatively transferred into 50 mL volumetric flasks. All solutions were prepared using ultrapure water. Before starting the procedure, laboratory glassware was drenched in nitric acid (8%), washed with tap water, then rinsed with deionized water. Metal concentrations were determined using flame atomic absorption spectrometry on a Thermo Scientific AAS ICE 3500 spectrometer (Thermo Scientific Portable Analytical Instruments Inc., Tewksbury, MA, USA). All measurements were performed in triplicate.

Analysis results were verified using the certified reference material for sediments NCS DC 73317a. The measurement results of the standard reference material showed good agreement with the certified values (Zn = 96%, Cu = 101%, Pb = 95%, Cr = 80%, Mn = 97%, Fe = 98%, Co = 93%, Ca = 99%, Na = 89%, Mg = 97%, K = 95%). Reference material values were measured at the beginning and after each series of analyzes.

2.3. Assessment of Sediment Contamination

The geochemical background of Poland [23], global background [24], and selected indices—geochemical index (I_{geo}), contamination factor (CF), pollution load index (PLI)—were applied to assess the level of contamination of lake sediments.

The geochemical index (I_{geo}) is applied to assess pollution levels by individual heavy metals. I_{geo} has a precise scale to determine the degree of contamination of the matrix under study and is widely used for soil and sediment [25]. The I_{geo} value was calculated using the formula [26]:

$$I_{geo} = \log_2[C_n / (1.5 \times B_n)] \quad (1)$$

where C_n means the measured content of the analyzed metal ($\text{mg} \cdot \text{kg}^{-1}$), B_n is the geochemical background concentration [23]. The constant 1.5 is the background matrix correlation coefficient due to lithological variability.

A contamination factor (CF) is applied to assess soil and sediment quality, especially concerning the presence of toxic elements. It assesses the difference between the metal concentration in the sample and reference values. The application of this index is simple, and the result is related to a precise scale, which allows determining the contamination of the matrix [25,27].

$$CF = C_{\text{metal}} / C_{\text{background}} \quad (2)$$

where C_{metal} means metal concentration in the sample and $C_{\text{background}}$ is the geochemical background.

The pollution load index (PLI) is a complex index, allowing comprehensive assessment of the contamination of soil/sediment with heavy metals. It combines any number of analyzed elements and allows the comparison of contamination at different locations in the soil [25,28].

$$PLI = (CF_1 \times CF_2 \times \dots \times CF_n)^{1/n} \quad (3)$$

where n is the number of metals analyzed.

Table 1 presents interpretations of the values of the used indices.

Table 1. Interpretation of values and classes of used indices.

Geoaccumulation Index (I _{geo})		Contamination Factor (CF)		Pollution Load Index (PLI)	
Value	Sediment Quality	Value	Sediment Contamination	Value	Pollution Status
I _{geo} ≤ 0	uncontaminated	CF < 1	low	PLI = 0	denote perfection
0 < I _{geo} < 1	uncontaminated to moderately contaminated				
1 < I _{geo} < 2	moderately contaminated	1 ≤ CF < 3	moderate	0 < PLI ≤ 1	only baseline levels of pollution
2 < I _{geo} < 3	moderately to strongly contaminated	3 ≤ CF< 6	considerable		
3 < I _{geo} < 4	strongly contaminated				
4 < I _{geo} < 5	strongly to extremely contaminated	CF ≥ 6	very high		
I _{geo} ≥ 5	extremely contaminated			PLI > 1	deterioration of sediment quality

The concentrations of the studied metals (Ca, Mg, Fe, Na, K, Mn, Zn, Cr, Cu, Pb, Co) refers to air-dry sediments. The results were compared with literature data for different lakes worldwide, considering the type of catchment: urbanized, agricultural–forest, and beaches.

2.4. Statistical Analysis

STATISTICA 13.1 (TIBCO Software Inc., Palo Alto, CA, USA) was used to perform all statistical analyses. The Shapiro–Wilk test was applied to assess the distribution normality of the tested variables. When the test showed statistically insignificant values at $p > 0.05$, the distribution of the variables was considered similar to normal. In all tests, the statistical significance was assumed to be 0.05. Firstly, descriptive statistics were used to determine mean, maximum, and minimum values. The Pearson correlation coefficient was applied to study the relationships between metals in sediments and identify their sources. One-way analysis of variance (ANOVA) showed significant differences between the study groups. Levene’s test was applied to test for homogeneity of variance. Tukey’s post hoc test was used to examine the differences between the means of the groups. The logarithmic version of the Box–Cox transformation was used to transform the data not having a normal distribution to a normal distribution. Then, the Kaiser–Meyer–Olkin (KMO) criterion was calculated, and Bartlett’s test of sphericity was performed. The KMO criterion value (0.6) enabled the use of factor analysis (FA). Factor analysis (FA) is the most common multivariate statistical method applied in environmental studies and is used to extract a small number of latent factors to analyze the relationships between observed variables [29]. FA was applied to identify sources of metals in sediments. This study’s number of factors was based on the Kaiser’s criterion and the scree plot criterion. In order to interpret FA results, it was assumed that the relations of the original variable with the factor are strong when the absolute values of its charges are higher than 0.70. The purpose of FA is usually the interpretation of individual factors’ influence on the variables. Cluster analysis (CA) via Ward’s version was used to investigate possible sources of heavy metals in sediments based on data classification.

3. Results

3.1. Selected Parameters Shaping Metal Content in Coastal Sediment

Metal contamination in sediments is an increasingly global problem [30,31]. Only a fraction of free metal ions is dissolved in water [32], whereas over 90% of the heavy metal load in aquatic systems is associated with suspended particles and sediments [33–35]. Thus, the distribution of metals in the coastal sediments of lakes with diverse catchment characteristics may provide evidence of anthropogenic impact on an ecosystem. Various factors, including sediment nature, granulometric composition, pH, and organic matter content, determine the metal concentrations in the sediments.

Sediment pH affects the solubility of metals [36]. High pH values promote adsorption and precipitation, whereas low pH can weaken the binding strength of metals and hinder their retention by sediments [37]. The pH values ranged from 7.8 to 8.3 pH (Table 2). There was no significant variation in the areas analyzed.

Table 2. Selected parameters of coastal sediments of Elckie Lake.

	Urban Areas (n = 6)	Rural/Forest Areas (n = 5)	Beach A (n = 10)	Beach B (n = 7)
pH (-)	7.9–8.1	7.8–8.1	7.9–8.2	8.0–8.3
OM (%)	0.4–2.2	0.7–1.9	0.4–1.4	0.3–1.0
granulometric fractions (%)				
>2.0 mm	15.91	28.81	10.01	12.23
1.0–2.0 mm	15.76	6.81	13.82	10.48
0.2–1.0 mm	61.23	50.94	50.43	55.79
0.1–0.2 mm	6.11	12.05	22.16	19.22
0.063–0.1 mm	0.72	1.04	2.83	1.67
<0.063 mm	0.27	0.35	0.75	0.61

Organic matter (OM) strongly impacts the bioavailability and toxicity of metals in sediments [38]. Its presence arises from the decomposition of dead biota under bacterial

activity [21,39]. The high OM content may increase the adsorption of pollutants in the aquatic environment [40]. The intensive application of agricultural fertilizers, population growth, and sewage inflow may increase OM content [41]. The highest average amounts of OM were in agricultural and forestry areas (1.14%), slightly lower in urban areas (0.98%), and the least amount was on beaches (A—0.88% and B—0.67%) (Table 2). However, the variation in OM content was minor.

Sediment grain size is a crucial determinant of adsorption and considerably influences metal bioavailability [42]. Particle size is an essential property of sediments that affects their physicochemical properties, erosion, and sedimentation [43]. In general, we suppose that the finer fraction holds the most considerable load of examined metals due to its much higher specific surface area, which causes much greater heavy metals' adsorption on sediments. However, the role of the coarser sediment fractions should not be excluded [44–46]. The surface sediments of Elćkie Lake are mainly sandy fractions (>98%). Results clearly showed that the 0.2–1.0 mm fraction (average 54.2%) prevails in the studied sediments. In addition, in most research points, the following fractions also have a significant share: 0.1–0.2 mm (average 16.2%), >2 mm (average 15.2%) and 1.0–2.0 mm (average 12.1%). The agricultural and forestry areas had a high proportion of fractions >2.0 mm (average 28.8%) compared to the other groups, which potentially reduced the sorption capacity of the sediments. As an example, Lake Guařba has a similar granulometric composition [47]. The coarse-grained fraction mainly derives from erosion of the rocks present in the catchment area, and only a small part is of anthropogenic origin. It is dominated by the products of physical weathering of the underlying rocks, whereas components derived mainly from chemical weathering prevails in finer fractions [23]. The two most important factors affecting metal enrichment are sediment grain size distribution and organic carbon content [48].

3.2. Concentrations of Major Elements and Five Potentially Toxic Elements (PTE) in Sediments

The study of the coastal sediments of Elćkie Lake based on the average metal contents showed the following order of elements in examined areas (Table 3):

1. urbanized: Ca > Mg > Fe > Na > K > Mn > Zn > Cr > Cu > Pb > Co,
2. agricultural and forest: Ca > Mg > Fe > Na > K > Mn > Cr > Zn > Pb > Cu > Co,
3. beach A: Ca > Mg > Fe > Na > K > Mn > Cr > Zn > Pb > Cu > Co,
4. beach B: Ca > Mg > Fe > Na > K > Mn > Cr > Zn > Pb > Co > Cu.

The average concentrations of major elements were identical in all regions and followed the descending order: Ca > Mg > Fe > Na > K > Mn. These may come from natural sources such as the Earth's crust, soil, and wind-borne dust from unpaved roads. Natural factors such as rock weathering and leaching processes are of minor importance but can cause locally elevated concentrations of some elements [49].

Elements such as Ca, Mg, K, Na, Fe, and Mn are common in regional soils and mineral sediments. Analysis of coastal sediments in Elćkie Lake showed the highest average Ca content (31,415.4 mg·kg^{−1}), which occurred in agricultural and forestry areas, which may be related to liming of fields by farmers. The highest Mg concentration was on the beaches (A—4410.2 mg·kg^{−1}, B—4865.2 mg·kg^{−1}). In urban areas, the average concentration of Fe was the highest (2609.5 mg·kg^{−1}). Higher Fe contents in cities may be related to pollutants from vehicular transport [50]. For instance, the abrasion of vehicle brake pads constantly used to reduce vehicle acceleration may cause relatively high emissions of this element [51]. In urbanized (560.0 mg·kg^{−1}) and forested areas (523.2 mg·kg^{−1}), average Na concentrations were similar. In sediments close to beaches, the content of Na was slightly lower. The highest concentration of K was found in urbanized areas (302.4 mg·kg^{−1}), whereas the contents were similar on both beaches (A—217.7 mg·kg^{−1}, B—234.6 mg·kg^{−1}). The highest amount of Mn occurred in the urbanized areas (97.8 mg·kg^{−1}), although there was not much variation between the studied localizations. Mn originates from the soil, and significant amounts of this element may also come from vehicle emissions such as tire wear or dust resuspension [52,53]. In addition, Mn content in soil is related to methylcyclopentadienyl

manganese tricarbonyl (MMT), which has displaced tetraethyl lead used as an antiknock additive in gasoline and oils [54].

Table 3. Descriptive statistics of metal concentrations in lake sediments including catchment land-use pattern (urbanized, agricultural and forest areas, beaches).

	Ca	Mg	Fe	Na	K	Mn	Zn	Cr	Cu	Pb	Co
GGB [24]	22,100	15,000	47,200	9600	26,600	850	95	90	45	20	19
LGB [23]	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	48	5	6	10	2
Urban Area											
min	15,841.8	1506.9	1931.8	283.1	160.9	68.7	9.6	10.1	0.7	2.3	0.8
max	47,718.1	4772.3	3659.0	747.3	436.1	120.9	61.6	15.5	51.7	22.2	1.6
average	28,633.1	2697.4	2609.5	560.0	302.4	97.8	30.4 ^a	13.1	11.1 ^b	9.3	1.2 ^{de}
SD	11,584.9 ^f	1207.5	590.7	200.6	109.3	20.2	20.6	2.0	20.0	9.7	0.3
Rural/Forest Area											
min	23,774.7	973.1	1169.4	328.0	92.9	18.4	7.5	8.7	1.6	2.3	0.5
max	50,447.3	4412.8	3767.0	864.7	210.0	136.4	16.0	16.2	2.6	3.9	0.7
average	31,415.4	2292.5 ^h	1922.0	523.2	132.4	94.3	10.8	12.7	2.0 ^c	3.4	0.6 ^d
SD	11,123.8	1383.5	1055.7	237.5	48.4	46.4	3.6	3.6	0.5	0.7	0.1
Beach A											
min	21,153.9	2619.1	1706.2	291.2	59.7	60.1	4.6	10.0	0.3	2.2	0.4
max	42,402.2	8814.0	3953.9	370.8	568.5	109.3	28.1	14.2	1.9	4.0	1.3
average	27,400.3 ^{fg}	4410.2	2182.9	320.7	217.7	89.9	10.4 ^a	12.4	1.1	2.7	0.7 ^e
SD	6468.5	1801.9	679.9	28.2	147.6	15.5	6.6	1.1	0.5	0.5	0.3
Beach B											
min	23,198.5	3792.3	2026.8	339.6	190.9	47.8	7.9	11.3	0.1	2.8	0.4
max	35,672.2	5998.3	2999.3	697.6	276.1	72.8	14.9	12.9	2.3	4.0	1.3
average	28,946.2 ^g	4865.2 ^h	2457.2	468.8	234.6	65.1	11.2	11.8	0.7 ^{bc}	3.2	0.9
SD	4078.8	910.2	392.9	150.7	33.6	8.4	2.3	0.5	0.7	0.4	0.3

Numbers with the same letter (a–h)—statistically significant difference at $p < 0.05$ ANOVA. Numbers without letters—no statistical difference ANOVA. GGB—global geochemical background, LGB—local geochemical background, SD—standard deviation, n.a.—not available.

The study focused on PTEs because they pose a risk to the environment and living organisms. PTEs concentrations in sediments from agricultural and forestry areas and beaches follow the order: $\text{Cr} > \text{Zn} > \text{Pb} > \text{Cu} > \text{Co}$. The exception was Cu in the case of beach B, which occurred at the end of the series. The order of PTE concentrations in sediments from urbanized areas differs from the above and was $\text{Zn} > \text{Cr} > \text{Cu} > \text{Pb} > \text{Co}$. Studies [55] showed the influence of urban activities as a source of heavy metals in lake sediments. The metals most commonly found in the highest concentrations in lake sediments from urbanized catchments are Zn, Cr, and Cu [56], whereas Cu, Pb, and Zn are most commonly found in high concentrations in urban runoff [12]. The same relationships occurred in this study. The average metal contents in Elćkie Lake sediments from urbanized areas are dominated mainly by Zn. The occurrence of heavy traffic in the vicinity of research points 1–6 (Figure 1) is the main source of heavy metal contamination of sediments there. Heavy metals accumulated on paved surfaces due to vehicle emissions are washed away during rain and enter the lake through storm runoff. Moreover, sanitary wastewater from unsewered housing developments located along the lakeshore can be a source of metals. The mean content of Zn ($30.4 \text{ mg} \cdot \text{kg}^{-1}$) and Pb ($9.3 \text{ mg} \cdot \text{kg}^{-1}$) in sediments from urbanized areas was about three times higher than in agricultural and forest areas (10.8 and $3.4 \text{ mg} \cdot \text{kg}^{-1}$, respectively) and beaches (A— 10.4 and $2.7 \text{ mg} \cdot \text{kg}^{-1}$, B— 11.2 and $3.2 \text{ mg} \cdot \text{kg}^{-1}$, respectively). Tire abrasion during vehicle exploitation emits high Zn amounts [15,57]. Tires are believed to contain approximately 1.3–1.7% [58] to 4.3% [59] Zn. Moreover, the presence of Pb in lake sediments may be related to vehicle emissions [60].

Most vehicles use unleaded gasoline, but the element remains in the environment that is especially exposed to heavy traffic [61]. Pb can also enter lake surface sediments from coal combustion and associated dust resuspension. High Cu concentrations ($11.1 \text{ mg}\cdot\text{kg}^{-1}$) were also recorded in sediments from urbanized areas, about ten times higher than from beaches (A— $1.1 \text{ mg}\cdot\text{kg}^{-1}$, B— $0.7 \text{ mg}\cdot\text{kg}^{-1}$) and five times higher than from agricultural–forest areas ($2.0 \text{ mg}\cdot\text{kg}^{-1}$). The presence of Cu in sediments from urbanized areas may also be related to communication. Cu is common in car bearings, brake linings, and other engine parts [62]. Car exploitation causes wear and tear of the metal, and Cu is released into the environment [57]. The contents of Co ($1.2 \text{ mg}\cdot\text{kg}^{-1}$) and Cr ($13.1 \text{ mg}\cdot\text{kg}^{-1}$) in sediments from urbanized areas occurred at only slightly higher levels than in beaches (A— 0.7 and $12.4 \text{ mg}\cdot\text{kg}^{-1}$, and B— 0.9 and $11.8 \text{ mg}\cdot\text{kg}^{-1}$, respectively) or agricultural–forest areas (0.6 and $12.7 \text{ mg}\cdot\text{kg}^{-1}$, respectively). Cr may be present in road dust in urbanized catchments, which occurs due to the friction of tires and road surfaces, from marking paint and anti-corrosion coatings on vehicles and safety barriers [15]. Analysis indicates that sediments from urbanized localizations were more contaminated with heavy metals than beaches or agricultural–forest areas (Table 3). Urbanization has led to heavy metal contamination of urban soils and surface sediments of urban lakes due to anthropogenic sources such as transportation and coal burning [14].

3.3. Contamination Indices (I_{geo} , CF, PLI)

Figure 2 shows I_{geo} , CF, and PLI values calculated for PTEs in coastal sediments. The geoaccumulation index is a well-known parameter used to determine sediment contamination by toxic elements [25,63,64]. The average I_{geo} values for metals followed the order $\text{Cr} > \text{Co} > \text{Pb} > \text{Zn} > \text{Cu}$. The highest average I_{geo} values occurred for Cr, indicating moderate contamination by this element. According to the standard of the degree of contamination [26], the mean I_{geo} values suggest that the studied sediments are uncontaminated by Co, Pb, Zn, and Cu. The maximum I_{geo} values for Pb (0.6, point 6) and Cu (2.5, point 1) show the highest amounts of these metals occurred at these sampling sites. These points sit on the urbanized part of the lake shoreline. This observation is consistent with the metal concentration analysis and suggests that the heavy traffic intensity, high population densities, and sanitary sewage from unsanitized residential areas located on the lakeshore affect the distribution of metals.

The average values of CF followed the order $\text{Cr} > \text{Cu} > \text{Pb} > \text{Co} > \text{Zn}$. The index calculated for beaches and agricultural and forestry areas proved low contamination by Zn, Cu, Pb, and Co. Points 1 and 6 (urban area) were moderately contaminated by Zn and Pb; significant contamination by Cu was detected at point 1 (urban area), suggesting significant anthropogenic contributions of these elements at the above sites. In addition, points 5, 6 (urban area), 9, and 10 (agricultural and forestry area) were significantly contaminated by Cr. Contamination caused by this element was moderate in all other points. Cr contamination in the studied sediments may come from geogenic and anthropogenic factors, especially road dust.

PLI is a simple tool to compare the pollution status at different sites. The PLI values assessed from Zn, Cu, Pb, Cr, and Co contents in the sediments ranged from 0.3 to 1.8. The highest values occurring in the urbanized area indicate the most polluted sites in the sediments of Elckie Lake (points 1 and 6, PLI equal to 1.8 and 1.3, respectively). Both of these points sit close to the outlets of the stormwater drainage system, near high-traffic intersections. A PLI value >1 indicates contamination and suggests an anthropogenic source of pollution. In points in agricultural and forestry areas and beaches, PLI values were low.

3.4. Spatial Distribution of Metals

The spatial distribution of metals (Figure 3) in coastal sediments helps distinguish sites with high metal content and shows the general distribution trend of potentially toxic elements in sediments. The most contaminated sediments are found in point 6 (Zn $61.6 \text{ mg}\cdot\text{kg}^{-1}$, Pb $22.2 \text{ mg}\cdot\text{kg}^{-1}$, and Cr $15.5 \text{ mg}\cdot\text{kg}^{-1}$). This point is located near a street that is one of the

main transportation routes of the city. It suggests that the high contents of these elements originate from anthropogenic sources related to transportation and communication. High Cu concentration ($51.7 \text{ mg}\cdot\text{kg}^{-1}$) was found at point 1. This point is located close to the outlet of the stormwater drainage system, and there is a junction connecting three streets with heavy traffic nearby. In addition, the highest concentration of Co ($1.6 \text{ mg}\cdot\text{kg}^{-1}$) was at point 5. The study showed the highest amount of Mn ($136.4 \text{ mg}\cdot\text{kg}^{-1}$ —point 9), Na ($864.7 \text{ mg}\cdot\text{kg}^{-1}$ —point 7), Fe ($3770.0 \text{ mg}\cdot\text{kg}^{-1}$ —point 11), and Ca ($50,447.3 \text{ mg}\cdot\text{kg}^{-1}$ —point 11) in the agricultural and forest areas. Typically, higher amounts of Mn and Fe in forest catchments, and Ca and Na in agricultural catchments, originate due to eluting these elements from soils periodically deprived of plant cover. The highest amounts of Mg ($8814.0 \text{ mg}\cdot\text{kg}^{-1}$) and K ($568.5 \text{ mg}\cdot\text{kg}^{-1}$) occurred at point 19 (beach A).

Figure 2. I_{geo} , CF and PLI values for the PTEs analyzed.

Figure 3. Spatial distribution of the analyzed metals (letter C on charts in the bottom right corner means concentration).

The results of the study were compared with metal concentrations in sediments in other lakes around the world (Table 4). Comparing metal concentrations within the beaches, other authors obtained similar or lower results, except for Cr and the macronutrients: Ca, Na, K, and Mg. In this study, these elements occurred in higher amounts. Heavy metal contents in the sediments of lakes with urbanized and agricultural–forest catchments were generally noticeably higher in other studies.

Table 4. Means/ranges of metal concentrations in coastal lake sediments in different areas of the world (dash “–” —not available).

Lake, Localisation	Zn	Cu	Pb	Co	Cr	Mn	Fe	Ca	Na	Mg	K	Ref.
Rural/Forest Areas												
Lere Lake, Chad	29.0	13.5	5.1	1.4	20.5	-	14,410	-	-	-	-	[21]
Lake Guaíba, Brazil	33.6	1.9	4.7	-	3.4	600	12,200	850	330	560	260	[47]
Elckie Lake, Poland	30.4	11.1	9.3	1.2	13.1	97.8	2609.5	28,633.1	560	2697.4	302.4	this study
Urban Areas												
Lake Koumoundourou, Greece	91.7	24.6	46.1	-	76.1	-	-	-	-	-	-	[65]
Lake Guaíba, Brazil	9.3–34.58	1.7–10.9	2.2–14.6	-	1.5–5.7	30–270	3300–6500	160–870	220–370	90–570	70–250	[47]
Sapanca Lake, Turkey	62.0	26.7	15.2	-	19.1	337.8	-	-	-	-	-	[66]
Kielce City Lake, Poland	11–98	1.4–35	56–324	-	3.7–168	122–940	2500–18,320	-	-	-	-	[67]
Elckie Lake, Poland	10.8	2.0	3.4	0.6	12.7	94.3	1922.0	31,415.4	523.2	2292.5	132.4	this study
Beaches												
Lake Manyas, Turkey	224.7	33.6	119.1	-	-	760.7	23,419	-	-	-	-	[68]
Lake Guaíba, Ipanema Beach, Brazil	11.9	1.8	4.3	-	2.7	60	3600	250	300	300	170	[47]
Lake Guaíba, Itapua Beach, Brazil	8.1	0.9	3.0	-	2.1	100	4100	290	290	160	60	[47]
Elckie Lake, beach A, Poland	10.4	1.1	2.7	0.7	12.4	89.9	2182.9	27,400.3	320.7	4410.2	217.7	this study
Elckie Lake, beach B, Poland	11.2	0.7	3.2	0.9	11.8	65.1	2457.1	28,946.2	468.8	4865.2	234.6	this study

4. Discussion

Identification of Pollution Sources Using Statistical Analyses

Pearson correlation coefficients between elements provide valuable information about the sources of elements in the environment [69–71]. Positive correlations of Zn with Cu ($r = 0.58$), Pb ($r = 0.90$), and Fe ($r = 0.40$) were found (Table 5). These relationships mean that these metals tend to accumulate together, and the result is their co-occurrence and interdependence; they come from similar sources and migrate together [72–74]. Sources of Cu, Pb, Zn, and Fe can include domestic and industrial wastewater, motor vehicle traffic, and human activities, including tourism [75]. Mn shows weak correlations with other trace metals, indicating its independent variability in the studied sediments [76]. Correlations of Fe with Ca ($r = 0.47$), Mg ($r = 0.37$), and K ($r = 0.60$) were also detected. Increased concentrations of Fe and Mn in the surface layer of sediments may not result as much from pollution as from their accumulation due to redox processes at the sediment–water interface. There were no correlations found between the metals and organic matter, which may indicate its small contribution to shaping the metal content of the sediments. The lack of correlation may also be related to the absence or low amount of fulvic or humic acids in coastal sediments. These compounds have a high metal adsorption capacity [77,78]. Correlation analyses also did not show an association between sediment pH and the presence of metals.

Table 5. Spearman correlation matrix between heavy metal concentrations and sediment properties.

	Zn	Cu	Pb	Co	Cr	Mn	Fe	Ca	Na	Mg	K	OM	pH
Zn	1.00												
Cu	0.58	1.00											
Pb	0.90	0.72	1.00										
Co	0.19	0.01	0.08	1.00									
Cr	0.21	0.09	0.31	0.07	1.00								
Mn	0.12	0.02	0.13	0.03	0.35	1.00							
Fe	0.40	0.08	0.32	0.31	0.16	−0.57	1.00						
Ca	0.03	−0.17	−0.20	−0.03	−0.27	−0.55	0.47	1.00					
Na	0.17	0.29	0.09	0.07	−0.57	−0.13	−0.09	0.17	1.00				
Mg	−0.21	−0.08	−0.25	0.18	−0.44	−0.54	0.39	0.24	−0.02	1.00			
K	0.33	0.37	0.26	0.53	−0.18	−0.27	0.60	0.15	0.28	0.58	1.00		
OM	0.06	0.28	0.14	−0.31	−0.12	0.25	−0.34	−0.16	−0.07	−0.04	−0.15	1.00	
pH	−0.10	0.02	0.05	−0.04	−0.08	−0.32	0.11	−0.35	−0.12	0.42	0.08	−0.03	1.00

One-way analysis of variance (ANOVA) was applied to test the significance of the relationship between area type and metal distribution in the coastal sediments of Elćkie Lake. Calculations showed a statistically significant (ANOVA) difference in mean contents of Zn, Cu, Co, and Na in sediments from urbanized areas compared to agricultural and forest areas, beach A and beach B. Statistically significant differences in mean contents of Cu, Co, and Mg were found in sediments from agricultural and forest areas compared to other locations. In this study, we analyzed metal contents in sediments from two beaches of the city of Elćk. The ANOVA showed a statistically significant difference in mean Na content in sediments from beach B and beach A. The mean Na content in sediments from beach B was higher than in the sediments from beach A. Infrastructure construction works in the vicinity of the recently established beach B may be the reason for the increased Na content in the coastal sediments. Overall, the results of the ANOVA analysis showed that the type of area bordering Elćkie Lake has a statistically significant relationship with the metal content in coastal sediments.

To identify metal sources, factor analysis with varimax rotation was undertaken. The relationships between metals were analyzed based on three factors (Table 6). Factors with eigenvalues greater than 0.70 were obtained, which explain 62% of the total variance. Factor F1 explains 25% of the total variance. It is related to Mn and geogenic processes in the study areas, which involve natural erosional processes and non-point agricultural sources. Factor F2 explains 23% of the total variance. It is associated with concentrations of Pb, Cu, and Zn. The study shows that Pb, Cu, and Zn may come from sewage, motor vehicle traffic, technical and construction transformations of urbanized areas, and waste management. These activities are particularly intensified during tourist seasons. Factor F3 showed only 14% of the total variance. Soil weathering and modification activities in tourist and recreational areas may increase Na content in sediments. All three factors explain 62% of the total variance, which indicates that lithogenic (geogenic) and anthropogenic factors make almost equal contributions to the distribution of most of the metals studied.

Table 6. Spearman correlation matrix between heavy metal concentrations and sediment properties (pH and OM).

	Factor 1	Factor 2	Factor 3
Zn	0.01	0.91	−0.07
Cu	0.13	0.81	0.27
Pb	0.14	0.93	−0.10
Co	−0.40	0.27	−0.27
Cr	0.29	0.23	−0.69
Mn	0.77	0.11	−0.20
Fe	−0.69	0.39	−0.32
Ca	−0.56	−0.11	0.17
Na	−0.10	0.25	0.74
Mg	−0.68	−0.14	0.23
K	−0.66	0.51	0.13
OM	0.46	0.12	0.40
pH	−0.25	−0.05	−0.01
% of total variance	25	23	14

5. Conclusions

The use of multivariate statistical techniques in combination with elemental content analysis is an effective tool for identifying metal sources in the coastal sediments of Elćkie Lake. The average major element contents of the 28 sediment samples occurred in the order $\text{Ca} > \text{Mg} > \text{Fe} > \text{Na} > \text{K} > \text{Mn}$, and were identical regardless of the kind of adjacent land development (urbanized, agricultural–forest, beaches). These elements may come from natural sources, such as the Earth’s crust, soil, and wind-borne dust from unpaved roads.

The average content of potentially toxic elements (PTEs) in the sediments followed the order $\text{Cr} > \text{Zn} > \text{Pb} > \text{Cu} > \text{Co}$ in agricultural and forest areas and beaches. The exception was Cu for beach B, which occurred at the end of the series. A different pattern occurred in urbanized areas, i.e., $\text{Zn} > \text{Cr} > \text{Cu} > \text{Pb} > \text{Co}$. The spatial distribution of heavy metals in the sediments indicated the highest contents in the shoreline within the urbanized part of the catchment (city of Elćk). Urbanization has led to heavy metal contamination of the coastal sediments of Elćkie Lake due to anthropogenic activities, primarily transport, coal combustion, and sanitary sewage from unsanitary residential areas located on the lakeshore. Metals may also enter the lake through storm runoff from roads. This was confirmed by positive correlations of Zn with Cu ($r = 0.58$), Pb ($r = 0.90$), and Fe ($r = 0.40$). No correlations between the studied metals and organic matter were found, which may indicate its insignificant contribution to metal content in the sediments. Correlation analyses also showed no relationship between sediment pH and metal presence.

Factor analysis indicated that lithogenic (geogenic) and anthropogenic factors have almost equal shares in the distribution of most of the metals studied. Analysis of variance (ANOVA) showed that the mean contents of Zn, Cu, Co, and Na in the sediments from urbanized areas are statistically significantly higher compared to the sediments from other areas (agro-forestry, beaches). Lake Elćkie sits in the area of the Green Lungs of Poland, and its ecosystem is little affected by human activities, despite the development of tourism and the impact of urban development. The PLI for Lake Elćkie due to potentially toxic elements was below 1.0.

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