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Assessment and Distribution of Heavy Metals Pollutants in Manzala Lake, Egypt

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Abstract

Contamination with heavy metals is one of the most serious problems in the aquatic environments. In Egypt, Manzala Lake is suffering from this problem. The objective of this work was to assess heavy metals pollutants and their spatial distribution in Manzala Lake using GIS technique. Geo-referenced water and sediment samples were randomly collected from the lake. The detected heavy metals were: Fe, Pb, Cu, Cd, Cr, Zn and Co. The obtained results indicated that the highest concentrations of heavy metals were observed in the northeastern and the southern parts of the lake nearby drains. This could be attributed to industrial, agricultural and municipal wastes coming through the drains especially Bahr El-Baqar drain and the industrial wastes coming from Port Said drains. From the geo-accumulation index, it was noticed that the lake is more polluted with cadmium and lead in the hydrosoils samples. All metals in water are within the EPA standard limit except for cadmium. Geostatistics provides effective methods to quantify the contaminated waters and sediments which support decision-making about redevelopment scenarios or remediation techniques.

Keywords

Manzala Lake, Heavy Metals, Indices, Pollution, GIS

1. Introduction

The contamination by heavy metals in the aquatic environments has drawn particular attentions due to their toxicity, persistence and biological accumulation. Heavy metals have low solubility and primarily get absorbed and

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accumulated on bottom sediments. Bottom lake sediments are sensitive indicators for monitoring pollutants as they act as a sink and a carrier for contaminants in aquatic environment. The most anthropogenic sources of metals are industrial, petroleum contamination and sewage disposal [1]. Manzala Lake, the largest of Egypt's Mediterranean wetlands and the most productive for fisheries, is suffering from land reclamation, industrial and nutrient pollution and overgrowth by water hyacinth. In the last six decades Manzala Lake was subjected to various threats: agriculture drainage, municipal sewage and industrial waste water. These pollutants have turned the lake into polluted, unhealthy ecosystem affecting fish production and natural resources that are distributed within the lake. A total of fresh water (mostly from agricultural drainage) flow annually into Manzala Lake from nine major drains and canals. It receives and carries the greatest part of wastewater into the lake through a very densely populated area of the Eastern Delta passing through Qalubyia, Sharkia, Ismailia and Port Said Governorates, and contributing much to the deteriorating water quality of the lake. Due to the toxicity of heavy metals, accurate information about their concentrations in aquatic ecosystem is needed. Accordingly, the objective of this study is to assess and evaluate the heavy metals pollutants in Lake Manzala using heavy metals indices with integrating GIS techniques for distribution of heavy metals in water and hydrosoils and joining them with their sources.

2. Materials and Methods

2.1. Study Area

Manzala Lake is the largest lake in the northern region of Egypt and the most productive for fisheries. The lake, as illustrated in **Figure 1**, lies between 31˚45', 32˚15'E and 31˚00', 31˚35'N. It covers an area of about 52,611 hectares. It is bordered by Suez Canal from east, Nile-Damietta branch from west and Mediterranean Sea from north. It is a shallow lagoon where about 50% of the lake area has a depth ranged between 0.5 and 1.0 m. The lake receives polluted water from different drains namely; Bahr El-Baqar, Ramsis, El-Matria, Hadous, Faraskur, El-Serw and Lissa El-Gamalia.

2.1.1. Climate

The climate of the Mediterranean coastal region of Egypt is belonging to the dry arid climatic zone. The study area lies in Meig's warm coastal deserts in which summer is warmest season with mean temperature less than 30˚C, and winter is the coldest season with mean temperature above 10˚C. The mean minimum air temperature at Manzala Lake ranged between 8.4˚C in January and 21.4˚C in August with mean annual temperature of about 15.4˚C, and the mean maximum air temperature varies from 18.3˚C in January to 31˚C in August with mean annual temperature of 24.9˚C. The relative humidity ranges from 68% in May to 76% in August with a mean annual value of 72%. The total annual rainfall attains a value of 106.7 mm. while the evaporation rate varies from

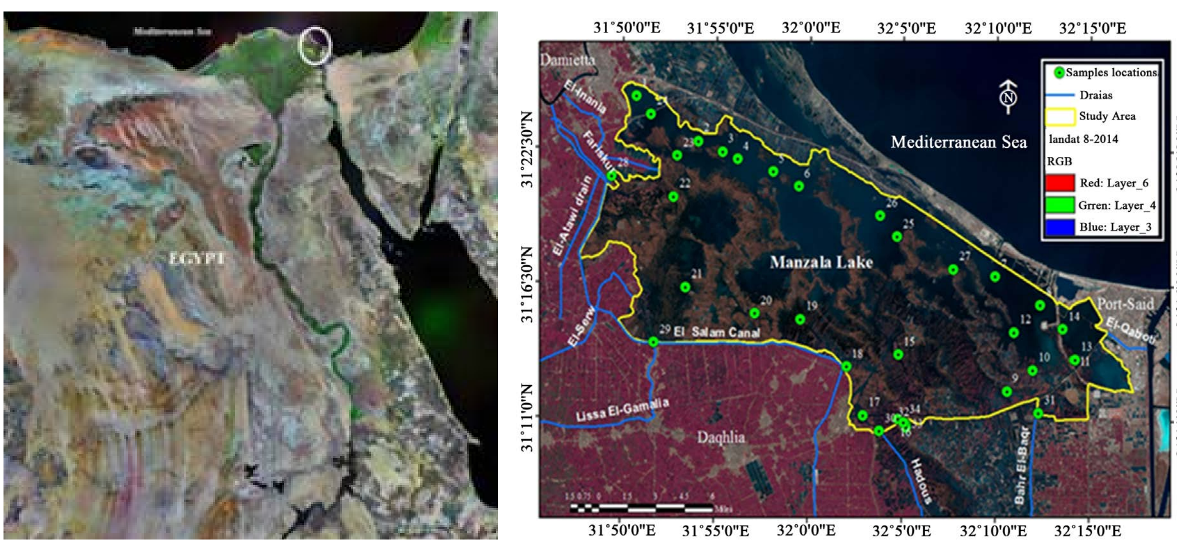

Figure 1. Location map of Manzala Lake and sampling locations.

2.8 mm/day in December and July to 5.4 mm/day in June. The prevailing wind is South westerly in January and February; North and north west in April, May, June, July, August and September; North easterly in October and November; and South westerly in December.

2.1.2. Water Resources

The lake is connected to the Mediterranean Sea via three outlets permitting exchange of water and biota between the lake and the sea, these outlets are El-Gamil, El-Boughdady and the new El-Gamil [2]. There is a small canal (El-Qaboty canal) that link between the lake and Suez Canal. There are major drains and canals flow into Lake Manzala as follows:

- Faraskur: Agricultural drain, serves 20,000 feddans, an area of about 44.48 km^2 (4% of total inflow).
- El-Sirw: Agriculture drain, serves 68,700 feddans (152.8 km^2), 13% of total inflow.
- Matariya: It serves 50,000 of land under agricultural reclamation (2%).
- Ramsis: Discharges a relatively small amount of water to Manzala Lake (24 km^2).
- Hadous: Is the largest drain in the eastern delta, serving some of agricultural land of about 1756.96 km^2 (49 %).
- Bahr El-Baqar: Serves an agricultural area of about 119.2 km^2, and receives about 300 million m^3/year of treated and untreated sewage from Cairo (25% of total inflow).

2.2. Samples

Thirty-four hydrosoil samples and thirty one water samples were randomly collected from different sites within the lake as illustrated in (**Figure 1**). Geographic positions of these samples were appointed using the GPS device (Garmin, etrex model). The collected samples were sent to the laboratory for heavy metal analysis. Seven essential heavy metals were studied in this work. These elements were Fe, Pb, Cu, Cd, Zn, Cr and Co.

2.3. Analytical Methods

2.3.1. The Analysis of the Dissolved Heavy Metals in Water Samples

Surface water samples were collected and then stored in acid-washed polyethylene bottles for analyses. Then, these samples were filtered using 0.45 um membrane filters. All the precautions occurred to minimize risks of sample contamination were followed during collection and treatment of samples. Solvent extraction was utilized using ammonium pyrrolidinedi-thiocarbamate (APDC) and methyl isobutyl ketone (MIBK). Where, water samples were pre-concentrated with APDC-MIBK extraction procedure according to the standard methods [3]. Heavy metals in the obtained solution were measured using the Flame Atomic Absorption Spectrophotometer (AAS: Perkin Elmer Analyst 100).

2.3.2. The Analysis of Heavy Metals in Hydrosoils Samples

Hydrosoil samples were collected using a Van-Veen grab coated with polyethylene. Sub-samples were taken from the central part of the grab to avoid contamination. The samples were kept in self-sealed acid pre-cleaned plastic bags, rinsed with metal-free water. The samples were deep-frozen until analysis. The samples were dried in the oven at 70˚C and sieved using 0.75 mm plastic sieve and digested for about two hours in a mixture of 3:2:1 HNO_3, $HClO_4$ and HF acids, respectively as described by Oregioni and Astone [4].

2.4. Geostatistics

The Geostatistical analyst in ArcGIS (ver. 10.1) software package was used to develop the ordinary Kriging method and the semivariogram between each pairs of points versus their separation distances [5]. This semivariogram was used in predicting the studied heavy metals in both waters and sediments of Manzala Lake.

2.5. Heavy Metals Indices in Hydrosoil Samples

2.5.1. Enrichment Factor (EF)

Enrichment Factor is considered as an effective tool to evaluate the magnitude of contaminants in the environment. Iron (Fe) was chosen as the controlling element [6]. The EF values < 2 indicate that the metal is entirely from crustal materials or natural processes; whereas EF values > 2 suggest that the sources are more likely to be anthropogenic [7].

$$\text{Enrichment Factor} = (\text{M/Fe})\text{sample}/(\text{M/Fe})\text{background} \tag{1}$$

where, M is the concentration of metal. The background value is that of avera.ge shale. Six categories are recognized: ≤1 background concentration, 1 - 2 depletion to minimal enrichment, 2 - 5 moderate enrichment, 5 - 20 significant enrichment, 20 - 40 very high enrichment and >40 extremely high enrichment.

2.5.2. Contamination Factor (CF)

The CF is the ratio obtained by dividing the concentration of each metal in the sediment by the baseline or Background value [8].

$$\text{Contamination Factor}(\text{CF}) = \text{C metal/C background} \tag{2}$$

The following terminologies are used to describe the contamination factor: CF < 1 (low contamination factor); 1 ≤ CF < 3 (moderate contamination factors); 3 ≤ CF < 6 (considerable contamination factors) and CF ≥ 6 (very high contamination factor).

2.5.3. Pollution Load Index (PLI)

The PLI of a single site is the root of number (*n*) of multiplied together Contamination Factor (CF) values.

$$\text{PLI} = (\text{CF}_1 * \text{CF}_2 * \text{CF}_3 * \cdots * \text{CF}_n)^{1/n} \tag{3}$$

where, *n* is the number of metals (seven in the present study) and CF is the contamination factor. A PLI value of zero indicates perfection; a value of one indicates the presence of only baseline levels of pollutants, and values above one would indicate progressive deterioration of the site quality [9] [10].

2.5.4. Degree of Contamination (DC)

The degree of contamination (DC) defined as the sum of all contamination factors for a given site:

$$\text{Cd} = \sum_{i=1}^{n} \text{CF}i \tag{4}$$

where CF is the single contamination factor and n is the count of the elements present. DC values less than *n* would indicate low degree of contamination; $n \leq \text{DC} < 2n$, moderate degree of contamination; $2n \leq \text{DC} < 4n$, considerable degree of contamination; and DC > 4*n*, very high degree of contamination. Where, n = 7 = the count of the studied heavy metals.

2.5.5. Geo-Accumulation Index (Igeo)

An index of geo-accumulation (Igeo) was originally defined by Müller [11] to determine and define the metal contamination in sediments by comparing current concentrations with pre-industrial levels.

$$\text{Igeo} = \text{Log2}\left(\frac{\text{Cn}}{1.5\text{Bn}}\right) \tag{5}$$

where, Cn is the measured concentration of heavy metals in sediments, Bn is the geochemical background value in average shale of element n and 1.5 is the background matrix correction due to anthropogenic influences. The geo-accumulation index (Igeo) was distinguished into seven classes by Buccolieri *et al.* [12]: Igeo ≤ 0, class 0, unpolluted; 0 < Igeo ≤ 1, class 1, from unpolluted to moderately polluted; 1 < Igeo ≤ 2, class 2, moderately polluted; 2 < Igeo ≤ 3, class 3, from moderately to strongly polluted; 3 < Igeo ≤ 4, class 4, strongly polluted; 4 < Igeo ≤ 5, class 5, from strongly to extremely polluted; and Igeo > 5, class 6, extremely polluted.

4. Results and Discussion

Metals generally enter the aquatic environment through erosion of the geological matrix, or due to anthropogenic activities caused by industrial effluents, domestic sewage, and mining wastes. Lead and Cadmium are toxic to living organisms even at quite low concentrations, whereas others, such as Zn and Cu, are biologically essential and natural constituents of aquatic ecosystems, and generally only become toxic at very high concentrations.

4.1. Heavy Metals in Water of Manzala Lake

The concentrations of dissolved heavy metals in water samples of Manzala Lake between different habitats are as shown in **Table 1**. **Figures 2(a)-(g)** illustrates the spatial distribution of dissolved heavy metals in the water samples collected from Manzala Lake. The mean concentration of the measured metals in water was found to be in the following sequence; Fe > Zn > Pb > Co > Cu > Cd > Cr.

Table 1. The concentrations of dissolved heavy metals in the water of Manzala Lake.

Site no.	X	Y	Fe	Zn	Pb	Cu	Co	Cr	Cd
1	31.402	31.845	13.65	5.773	3.808	1.229	1.269	0.33	0.932
2	31.372	31.901	34.73	7.423	6.463	2.728	1.597	0.63	0.887
3	31.365	31.923	17.57	8.612	4.712	1.868	1.896	0.416	0.891
4	31.360	31.936	24.58	8.519	4.871	1.892	3.077	0.492	0.902
5	31.352	31.968	17.71	7.292	4.954	1.617	1.673	0.454	0.941
6	31.342	31.990	20.16	6.741	2.694	1.683	1.645	0.951	0.725
7	31.282	32.166	14.06	9.976	5.482	1.468	1.621	0.721	0.785
8	31.262	32.206	20.89	9.506	3.794	2.113	1.689	0.359	0.802
9	31.204	32.177	11.405	5.375	5.5	1.15	0.975	1.27	0.225
10	31.218	32.200	41.52	3.265	3.93	0.655	0.775	1.1	0.23
11	31.225	32.237	32.115	5.58	9.15	0.98	1.395	1.98	5.5
12	31.244	32.183	20.56	3.935	5.225	1.38	1.765	1.66	4.67
13	31.225	32.237	14.17	5.98	3.75	1.265	1.775	22.1	3.93
14	31.246	32.226	17.4	3.15	5.19	1.24	1.925	4.015	9.15
15	31.228	32.080	12.315	1.18	5.95	0.64	0.85	0.46	2.09
16	31.184	32.080	19.87	1.635	1.925	1.14	1.005	0.3	3.75
17	31.186	32.049	17.05	1.305	1.805	0.72	0.995	1.84	5.19
18	31.220	32.034	12.395	1.365	2.21	0.605	0.805	0.095	0.245
19	31.251	31.993	7.38	1.495	1.595	0.515	0.705	0.34	0.27
20	31.255	31.952	4.785	1.14	3.25	1.03	0.685	0.45	0.29
21	31.273	31.890	9.055	1.17	2.505	0.695	0.71	0.24	0.28
22	31.334	31.879	11.4	1.34	4.56	1.05	2.324	0.23	0.275
23	31.362	31.882	12.98	1.32	5.63	1.65	0.86	0.25	0.275
24	31.390	31.858	13.34	1.12	6.46	1.76	1.552	0.35	0.285
25	31.308	32.078	15.62	5.605	8.234	1.776	1.765	0.65	0.924
26	31.322	32.063	17.95	6.34	3.992	1.392	2.103	0.86	0.802
27	31.286	32.129	21.48	4.56	7.374	3.793	2.369	1.23	1.212
28	31.348	31.823	183.3	11.77	6.12	4.879	6.557	0.973	0.987
29	31.235	31.862	167.4	11.91	6.73	4.775	5.937	0.633	0.325
30	31.176	32.064	42.73	7.069	3.58	2.996	6.557	0.252	0.805
31	31.189	32.205	47.74	9.465	4.77	2.947	7.15	0.592	0.748
	Mean		29.59	5.19	4.72	1.73	2.13	1.49	1.59
	SE (±)		7.24	0.61	0.33	0.20	0.33	0.70	0.37
	EPA, 2002		300	5000	50	50		100	2.37

EPA (2002): Environmental Protection Agency limit for water & values in (μg/l).

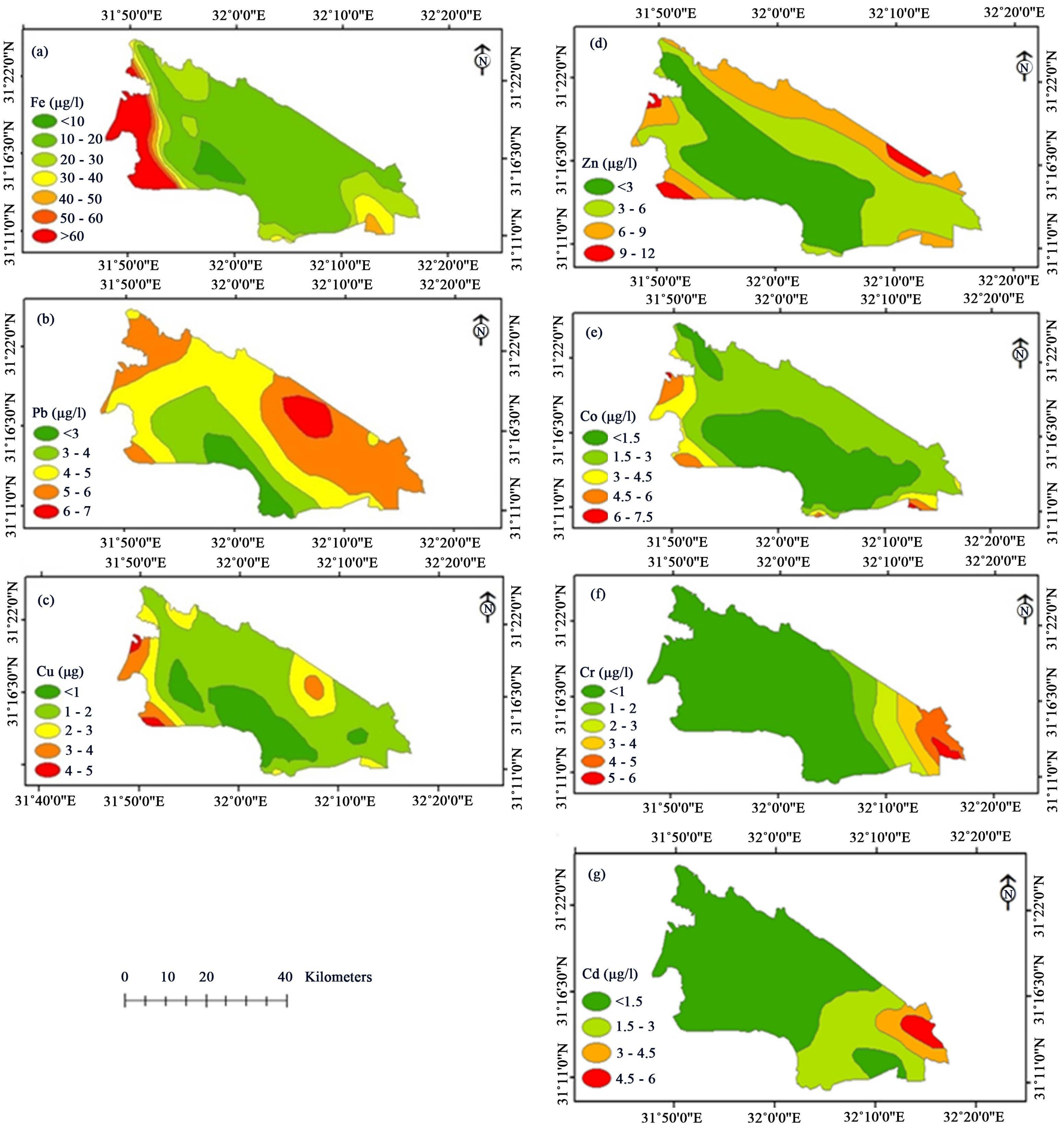

Figure 2. The spatial distribution of (a) iron; (b) lead; (c) copper; (d) zinc; (e) cobalt; (f) chromium and (g) cadmium in the water samples of Manzala Lake using GIS technique.

Iron varied from 4.78 μg/l nearby El-Gamalia city to 183.3 μg/l at Faraskur drain. This result is lower than those (1420 μg/l - 696 μg/l) recorded by Saeed and Shaker [1] and El-Alfy [13]. While the concentration of copper varied from 0.51 to 4.87 at Faraskur drain. The highest concentrations of iron and copper are distributed at the southwestern parts of lake especially nearby drains, where there are more agricultural wastes as revealed by Arain *et al*. [14]. The concentrations of dissolved Fe in the oxygen depleted waters were markedly higher than those found in the oxic northern waters of Manzala Lake "*i.e.* nearby El-Boughaz area" [9]. Higher values of Cu in Manzala Lake were observed in both water and hydrosoil by Ali and Abdel-Satar [15].

Zinc is one of the most abundant toxic heavy metals, the oral toxicity of humans to most zinc compounds is relatively low. High concentrations of zinc in the marine environment may exist from the discharge of industrial wastes especially electroplating and synthetic fiber production. It ranged between 1.12 and 11.91 μg/l. It's noticeable that the highest concentrations of zinc were observed as well at the drains and nearby industrial areas as a result to direct discharge of industrial wastes in these areas. However, the lowest concentration of zinc is distributed in the middle part of the lake far from drains. These values are lower than that recorded by Bahnsawy *et al.* [16].

Dissolved lead in water ranged between 1.59 to 8.23 µg/l. The lowest concentration of lead was recorded far away from drainage areas, but the highest concentration was observed at northeastern side nearby the industrial compound. These areas receive huge quantities of sewage and industrial wastes, as well as from spill of leaded petrol from fishing boats and dust which holds a huge amount of lead [10]. The present result of lead in water is within the limit of EPA [10]. The United States Environmental Protection Agency EPA [18] has classified Pb as being potentially hazardous and toxic to most forms of life.

Cobalt was in the range between 0.68 to 7.15 µg/l. The lower concentrations of cobalt are distributed at the middle and northern parts of the Lake. The highest concentration of cobalt was recorded nearby drains at the southern parts as a result of agricultural wastes especially Bahr El-Baqar drain. This result agreed with Nagpal, [19].

The sources of chromium entering the aquatic environment are from paint and chemical works, oil drilling and recovery rigs. Large quantities of chromium may be released from petrochemical industries and cement, fertilizer, power, and chlor-alkali plants. It varied from 0.09 to 22.10 µg/l. The highest concentration of Cr is found at Ashtoum El-Gamil area, near tourist villages and Company of Natural gas Petroget. Furthermore, untreated domestic sewage was discharged into the sea from activities at the coastal area [20]. The lowest concentration of Cr is obtained nearby Hadous drain. The highest concentration in Manzala is higher than the threshold limit (1.5 µg/l) of Environment Canada [21], but is within EPA limit [10].

Cadmium is one of the black listed elements, it can be considered as one of the most dangerous elements on marine life and humans. The dissolved cadmium in water of Manzala Lake is in the range between 0.22 to 9.15 µg/l. The lowest concentration was recorded at open water far from drains, but the highest concentration was recorded nearby El-Qaboty area south to port said city as a result of urban extension and agricultural disposal especially the phosphatic fertilizers, this is in agreement with Bahnasawy *et al.* [17]; El-Serehy *et al.* [20] and Hamed *et al.* [22]. The estimated Cd value in this study is more than the EPA limits [10]. It is one of the most toxic elements with widespread carcinogenic effects in humans and considered to be toxic if its concentration exceeds 0.01 mg/L both in drinking and irrigation water.

4.2. Heavy Metals in the Hydrosoils of Manzala Lake

Hydrosoil contamination poses one of the worst environmental problems in ecosystems, acting as sinks and sources of contaminants in aquatic systems. Hydrosoil analyses play an important role in assessing the pollution status of the environment. The concentrations of heavy metals in the hydrosoils of Manzala Lake are as given in **Table 2** and the spatial distribution is illustrated in **Figures 3(a)-(g)**. The concentrations of heavy metals in the hydrosoils of Manzala Lake take the following sequence; Fe > Cr > Zn > Pb > Cu > Co > Cd, which except Fe differs from these of the lake's water.

Iron in the hydrosoils of Manzala Lake was in the range between 250 and 666.37 µg/g. Where copper varied from 1.25 to 52.68 µg/g. The highest value of iron was recorded at Bahr El-Baqar drain. For copper the highest concentration was recorded at Hadous drain. These drains have agricultural drainage, industrial and domestic wastes. These results are in agreement with those obtained by Saeed and Shaker [1] and El-Alfy [13]. It was found that those drains are rich in organic carbon and some authors found a correlation between the concentration of heavy metals in sediment and the abundance of organic matter [13]. Whereas the lowest values of iron were recorded far away from drains, nearby El-Boughaz area and the northern parts of the Lake. For copper, the lowest value was recorded at El-Temsah area far away from drains as well. The values of copper are higher than that recorded by Hamed *et al*. [22]. The maximum values of iron and copper in hydrosoils exceed the limits set by EPA [10] where the guideline values are 15 and 25 µg/g for iron and copper, respectively.

The range of lead in the hydrosoils of Manzala Lake was between 3.44 and 65.53 µg/g. The maximum concentration of lead was recorded nearby the drains, as they receive huge quantities of sewage and industrial wastes, beside agricultural drainage water via Bahr El-Baqar drain and may attributed to the decaying of plankton and precipitation of organic matter associated with Pb and Cd to the sediment [17]. The lowest concentration of lead was noticed at El-Temsah area (at the northern parts of the lake far from drainage water). The maximum value of lead is higher than EPA limit [10], but within the limit of EU [23].

Table 2. The concentrations of heavy metals in the hydrosoils of Manzala Lake.

Site no.	X	Y	Fe	Zn	Cr	Cu	Pb	Co	Cd
1	31.402	31.845	301.60	26.85	27.51	8.80	11.27	8.51	1.40
2	31.372	31.901	250.00	4.76	4.52	1.25	3.44	2.97	0.66
3	31.365	31.923	306.10	6.75	66.24	8.21	14.11	13.91	1.46
4	31.360	31.936	271.70	5.75	8.80	2.21	5.53	3.32	0.74
5	31.352	31.968	305.40	12.79	42.72	10.72	12.18	12.34	1.37
6	31.342	31.990	298.50	22.22	27.86	6.59	9.62	7.50	1.16
7	31.282	32.166	290.50	2.98	54.46	10.05	19.01	9.70	2.04
8	31.262	32.206	306.50	78.06	39.73	7.08	14.36	14.48	1.56
9	31.204	32.177	310.30	3.38	38.80	30.14	17.27	20.24	1.31
10	31.218	32.200	294.10	75.65	12.41	9.72	65.53	6.79	1.19
11	31.225	32.237	306.80	4.07	62.98	2.50	3.80	19.01	1.23
12	31.244	32.183	307.20	28.10	28.76	9.46	63.12	14.36	2.65
13	31.225	32.237	303.20	6.01	5.78	3.46	12.52	12.52	1.91
14	31.246	32.226	306.40	3.85	67.49	9.46	4.70	15.16	2.71
15	31.228	32.080	308.80	3.18	21.19	28.61	15.36	17.20	1.94
16	31.184	32.080	306.90	3.02	16.27	24.40	23.70	15.92	2.17
17	31.186	32.049	307.50	3.02	16.88	26.33	36.51	18.04	1.78
18	31.220	32.034	304.90	2.51	9.41	21.63	22.29	16.37	2.38
19	31.251	31.993	308.10	2.59	9.31	25.52	19.97	17.91	2.56
20	31.255	31.952	301.50	3.35	2.64	16.85	21.21	14.05	2.30
21	31.273	31.890	309.20	3.51	24.88	33.25	14.82	19.88	2.33
22	31.334	31.879	305.40	3.08	22.23	17.62	21.10	15.13	1.96
23	31.362	31.882	305.30	2.43	22.62	17.14	12.50	14.88	1.49
24	31.390	31.858	305.20	2.84	21.87	21.59	20.66	5.51	2.06
25	31.308	32.078	308.80	69.36	31.50	12.64	24.52	16.61	2.82
26	31.322	32.063	309.10	77.13	29.73	13.85	25.57	15.65	2.82
27	31.286	32.129	309.30	157.70	37.85	14.14	23.36	16.02	2.73
28	31.348	31.823	630.44	50.84	37.48	10.81	22.06	6.56	0.56
29	31.235	31.862	661.21	85.77	86.09	42.47	26.45	5.94	0.66
30	31.176	32.064	654.77	72.61	101.50	52.68	20.44	6.56	0.88
31	31.189	32.205	666.37	71.21	101.40	50.36	20.70	7.15	1.07
32	31.180	32.087	299.50	19.00	14.51	18.80	33.50	7.59	1.16
33	31.178	32.087	301.30	19.50	16.00	14.44	47.54	11.76	2.62
34	31.182	32.085	299.40	54.81	15.23	12.14	18.65	7.49	1.26
	Mean		342.98	29.08	33.14	17.50	21.39	12.27	1.73
	SE (±)		19.84	6.34	4.51	2.20	2.44	0.86	0.12
	Average Shale		47200	95	90	45	20	19	0.30
	EPA, 2002		15	123	25	25	10	-	6
	EU, 2002		-	300	150	140	300	11.6	3

EPA: Environmental Protection Agency for sediment samples in (μg/g); EU: European Union Standard in (μg/g).

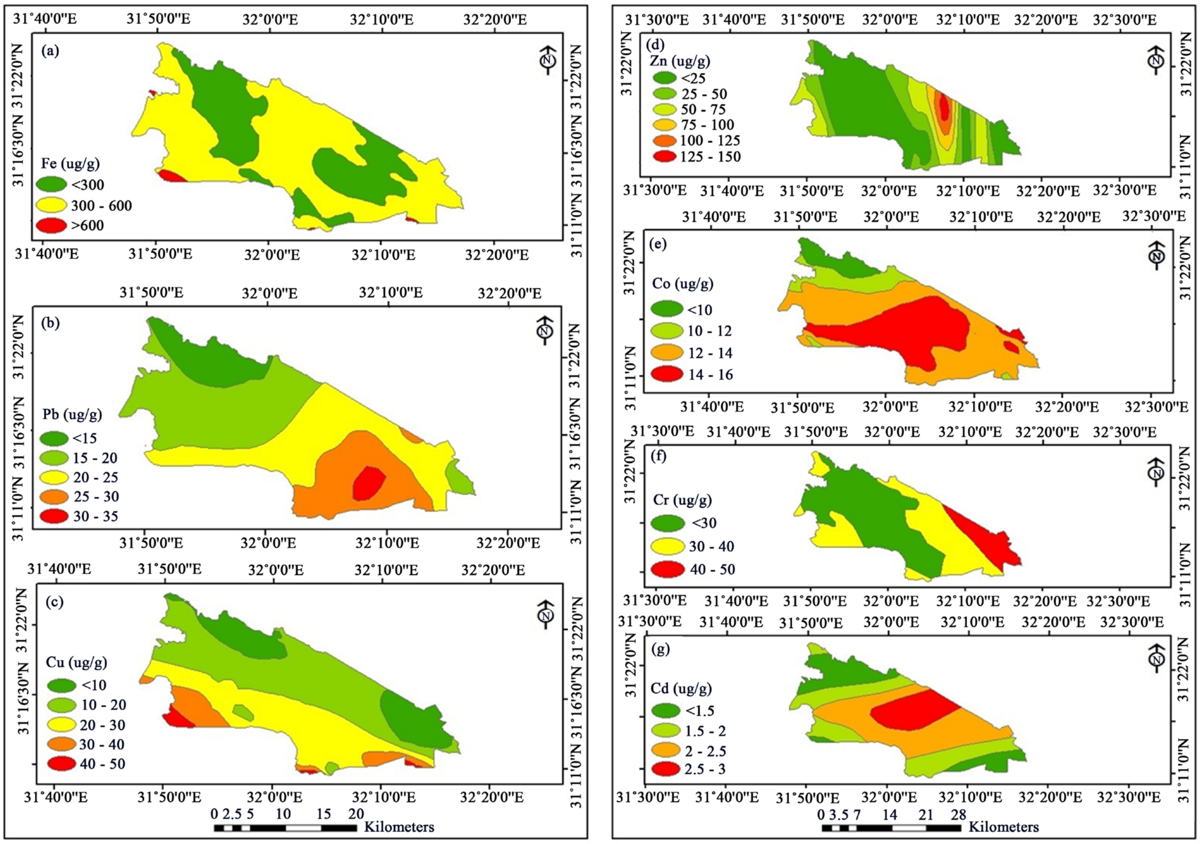

Figure 3. The spatial distribution of (a) iron; (b) lead; (c) copper; (d) zinc (e) cobalt; (f) chromium and (g) cadmium in the hydrosoils of Manzala Lake.

Zinc varied from 2.43 to 157.70 µg/g, its highest concentration may be as a result of industrial disposal spilled directly or indirectly to the lake. The maximum concentration of Zinc in hydrosoils was obtained nearby the industrial compound northern to Lake Manzala. This result is higher than those recorded by Hamed *et al*. [22] and El-Alfy [13]. The lowest value of Zn was estimated at the western part of the lake far from industrial wastes. It's obvious that Cd and Pb accumulation decreased in saline medium while the accumulation of Zn increased so the highest concentration of zinc in the hydrosoil especially in the northern parts where sea water intrusion occurs [24]. The maximum values of zinc are more than the EPA limit, but within the limit of EU [23].

The concentration of cobalt in the hydrosoils of Manzala Lake was in the range of (2.79 to 20.24 µg/g). Its maximum concentration was observed nearby Damietta city which may due to the irrigation of agricultural lands with untreated water and uses of agricultural fertilizers. Also high concentrations of Co were observed nearby Bahr El-Baqar drain. Its lowest value was recorded at the northwestern part of the lake far away from drains. These values were lower than (54.29 to 80.30 µg/g) recorded by El-Bady [25] at Bahr El-Baqar region. Cobalt in the hydrosoils of Manzala Lake recorded values more than the EU limit [23].

Chromium varied from 2.64 to 101.50 µg/g. The lowest concentration of Cr was recorded far away from drains. While the highest concentration was observed nearby drains (especially, Bahr El-Baqar and Hadous drains), the areas south to Port Said city near the industrial activities [19]. Also, high concentrations of Cr were found at Ashtoum El-Gamil area and near tourist villages and Company of Natural gas Petroget. Furthermore, untreated domestic sewage was discharged into this area from activities at the coastal area, the combustion of coal and oil. Chromium is within the limit of EU [23], but its maximum value is more than what be revealed by the EPA limit [10].

Cadmium in the hydrosoils varied from 0.56 to 2.82 µg/g. The lowest concentration was recorded at Faraskur drain. However, the higher concentrations were concentrated in the area nearby the industrial activities. The previous results are in agreement with those obtained by Bahnasawy *et al*. [17] and Hamed *et al*. [22]. They reported that the high concentrations in this area were due to the paint factories disposal without treatment directly to the lake. Cadmium recorded higher values than the sediment quality guidelines and agreed with Saeed and Shaker [1]. The values of cadmium in hydrosoils are within EPA [10] limit of a guide value (6 µg/g). Cadmium in uncontaminated soils must not exceed 0.7 ppm.

4.3. Indices of Heavy Metals in the Hydrosoils of Manzala Lake

4.3.1. The Enrichment Factor

The Enrichment Factors of heavy metals in the hydrosoils of Manzala Lake are as shown in **Table 3**; it's indicated that the EF of Cu ranged between 5.24 and 112.79. For Zn, the EF varied from 3.95 to 253.32 and EF of Cr varied from 4.59 to 115.53. The EF of Pb varied from 32.51 to 484.91 and for Co, it varied from 22 to 160 and EF of Cd varied from 140.25 to 1435.26. From the previous results, it's obvious that the EF of all metals is in the high significant categories and the sources of these metals in Lake Manzala are almost from anthropogenic activities. The sequence of EF for heavy metals in the hydrosoils of Manzala Lake is: Cd > Pb > Zn > Co > Cr > Cu. From that, cadmium is more abundant than other metals; whereas Cu showed the lowest appearance.

4.3.2. Contamination Factor

From **Table 4**, the CF values of Fe showed low contamination factor. It ranged between low at all sites for copper and moderate CF at Hadous and Bahr El-Baqar Drains as (CF > 1). For Cr, CF is lower than 1 (low CF) and >1 only in the northern part of the Lake. CF of lead varied from 0.17 to 3.28 (low to considerable CF). The values of CF for cobalt are <1 in all sites (low CF) and >1 only nearby Bahr El-Baqar drain (moderate CF). The CF of cadmium showed moderate, considerable and very high CF.

4.3.3. Pollution Load Index (PLI) and Degree of Contamination (DC)

The PLI and DC are indicated in **Table 4**, where the PLI in this study area of Manzala Lake ranged between 0.11 and 0.57. This showed that there is no appreciable pollution in Lake Manzala with those metals. From the results of contamination degree (**Table 4**), it ranged between low to moderate degree of pollution. The spatial distribution of DC is illustrated in **Figure 4**.

4.3.4. The Geo-Accumulation Index (Igeo)

The results of Igeo are as shown in **Table 5** and **Figure 5**. The negative values of Fe, Cr, Co, Cu and Zn and according to the classification of Muller [26], it indicated that the Lake isn't polluted with those metals. From the geo-accumulation index results, Igeo values of lead showed unpolluted to moderately polluted categories nearby the biological treatment project east to Port Said City, at the southern part of the Lake nearby Drains and at Ibn-Salam area (nearby Hadous drain). The Igeo results of cadmium also showed moderately polluted category in all locations.

4.4. The Correlation between Heavy Metals in Water and Hydrosoils

From **Table 6**, there are significant positive correlation between Fe in hydrosoil with Cu, Cr in hydrosoil (r = 0.7); Fe, Zn and Co in water (r = 0.7, 0.5 and 0.9) and correlates negatively with Co and Cd in hydrosoil as r = −0.4 for each. Copper in hydrosoil showed a positive significant correlation with Cr in hydrosoil and Co in water where (r = 0.5 for each). Zinc in hydrosoil correlated with Cr in hydrosoil, Fe, Co and Zn in water (r = 0.4) and with Cu in water (r = 0.6). Cr in hydrosoil correlated with Fe in water (r = 0.37), Cu in water (r = 0.49), Zn in water (r = 0.54) and Co in water (r = 0.7). Cobalt in hydrosoil showed only significant positive correlation with Cd in hydrosoil (r = 0.6) and negative with Fe, Cu, Zn and Co in water as (r = −0.44, −0.53, −0.54 and −0.53 respectively). Also cadmium in hydrosoil showed negative significant correlation with Fe, Cu, Zn and Co in water (r = −0.54, −0.47, −0.58 and −0.51 respectively). There are a significant positive correlation between iron in water and Cu, Zn and Co in water (r = 0.8, 0.6 and 0.7). Cu in water and Zn, Pb and Co in water (r = 0.7, 0.4 and 0.8). Between Zn and Co in water (r = 0.6) and Cr and Cd in water (r = 0.36). Other relations between elements showed low or no significant correlations. It's obvious that the heavy metals with positive correlation were considered to have similar sources as estimated by Dan *et al.* [27].

Table 3. The enrichment factor (EF) of heavy metals in the hydrosoils of Manzala Lake.

NO	Enrichment Factor (EF)						
	Fe	Cu	Zn	Cr	Pb	Co	Cd
1	1.00	30.60	44.23	47.84	88.19	70	727.72
2	1.00	5.24	9.46	9.49	32.51	30	412.84
3	1.00	28.13	10.96	113.49	108.79	113	748.37
4	1.00	8.53	10.51	16.98	48.02	30	427.35
5	1.00	36.82	20.81	73.36	94.12	100	705.78
6	1.00	23.16	36.98	48.95	76.05	62	609.83
7	1.00	36.29	5.10	98.32	154.43	83	1103.23
8	1.00	24.23	126.54	67.98	110.57	117	799.24
9	1.00	101.88	5.41	65.58	131.35	162	664.72
10	1.00	34.67	127.80	22.13	525.84	57	638.75
11	1.00	8.56	6.59	107.66	29.23	154	629.23
12	1.00	32.32	45.45	49.11	484.91	116	1357.10
13	1.00	11.99	9.86	9.99	97.49	103	991.53
14	1.00	32.40	6.25	115.53	36.19	123	1391.96
15	1.00	97.18	5.12	35.99	117.43	138	986.39
16	1.00	83.39	4.89	27.80	182.25	129	1112.97
17	1.00	89.81	4.88	28.79	280.21	146	909.21
18	1.00	74.41	4.09	16.19	172.53	133	1230.18
19	1.00	86.88	4.18	15.84	152.97	144	1304.73
20	1.00	58.62	5.52	4.59	166.02	116	1202.31
21	1.00	112.79	5.64	42.20	113.12	160	1183.56
22	1.00	60.52	5.01	38.17	163.05	123	1011.28
23	1.00	58.89	3.95	38.86	96.63	121	766.83
24	1.00	74.20	4.62	37.58	159.76	45	1062.98
25	1.00	42.93	111.60	53.50	187.39	134	1435.26
26	1.00	47.00	123.98	50.44	195.23	126	1433.87
27	1.00	47.95	253.32	64.18	178.24	129	1390.21
28	1.00	17.99	40.07	31.18	82.58	26	140.25
29	1.00	67.37	64.45	68.28	94.41	22	157.28
30	1.00	84.39	55.10	81.30	73.67	25	210.25
31	1.00	79.27	53.10	79.80	73.31	27	252.87
32	1.00	65.84	31.52	25.41	263.97	63	610.42
33	1.00	50.27	32.16	27.85	372.37	97	1365.51
34	1.00	42.53	90.95	26.68	147.01	62	659.50

Table 4. The contamination factor (CF), pollution load index (PLI) and degree of contamination (DC) for heavy metals in the hydrosoils of Manzala Lake.

NO	Contamination factor (CF)							PLI	DC
	Fe	Cu	Zn	Cr	Pb	Co	Cd		
1	0.006	0.20	0.29	0.30	0.56	0.45	4.65	0.28	6.45
2	0.005	0.03	0.05	0.05	0.17	0.16	2.19	0.08	2.65
3	0.006	0.18	0.70	0.08	0.71	0.73	4.85	0.29	7.25
4	0.006	0.05	0.09	0.06	0.28	0.17	2.46	0.11	3.12
5	0.006	0.24	0.45	0.14	0.61	0.65	4.57	0.29	6.66
6	0.006	0.15	0.29	0.25	0.48	0.39	3.86	0.24	5.43
7	0.006	0.22	0.57	0.03	0.95	0.51	6.79	0.26	9.09
8	0.006	0.16	0.42	0.87	0.72	0.76	5.19	0.38	8.12
9	0.007	0.67	0.41	0.04	0.86	1.07	4.37	0.31	7.42
10	0.006	0.22	0.13	0.84	3.28	0.36	3.98	0.35	8.81
11	0.007	0.06	0.66	0.05	0.19	1.00	4.09	0.19	6.05
12	0.007	0.21	0.30	0.31	3.16	0.76	8.83	0.43	13.58
13	0.006	0.08	0.06	0.07	0.63	0.66	6.37	0.18	7.87
14	0.006	0.21	0.71	0.04	0.23	0.80	9.04	0.26	11.04
15	0.007	0.64	0.22	0.04	0.77	0.91	6.45	0.28	9.03
16	0.007	0.54	0.17	0.03	1.19	0.84	7.24	0.28	10.01
17	0.007	0.59	0.18	0.03	1.83	0.95	5.92	0.30	9.50
18	0.006	0.48	0.10	0.03	1.11	0.86	7.95	0.25	10.54
19	0.007	0.57	0.10	0.03	1.00	0.94	8.52	0.26	11.16
20	0.006	0.37	0.03	0.04	1.06	0.74	7.68	0.20	9.93
21	0.007	0.74	0.26	0.04	0.74	1.05	7.75	0.31	10.59
22	0.006	0.39	0.23	0.03	1.06	0.80	6.54	0.27	9.06
23	0.006	0.38	0.24	0.03	0.63	0.78	4.96	0.23	7.02
24	0.006	0.48	0.23	0.03	1.03	0.29	6.87	0.24	8.94
25	0.007	0.28	0.33	0.77	1.23	0.87	9.39	0.47	12.88
26	0.007	0.31	0.31	0.86	1.28	0.82	9.39	0.47	12.98
27	0.007	0.31	0.40	1.75	1.17	0.84	9.11	0.54	13.59
28	0.013	0.24	0.39	0.56	1.10	0.35	1.87	0.34	4.53
29	0.014	0.94	0.91	0.95	1.32	0.31	2.20	0.52	6.66
30	0.014	1.17	1.07	0.81	1.02	0.35	2.92	0.55	7.34
31	0.014	1.12	1.07	0.79	1.04	0.38	3.57	0.57	7.97
32	0.006	0.42	0.15	0.21	1.68	0.40	3.87	0.30	6.74
33	0.006	0.32	0.17	0.22	2.38	0.62	8.72	0.37	12.42
34	0.006	0.27	0.16	0.61	0.93	0.39	4.18	0.31	6.56

Table 5. The geoaccmulation index (Igeo) of heavy metals in the hydrosoils of Manzala Lake.

NO	Geoaccmulation index (Igeo)						
	Fe	Cu	Zn	Cr	Pb	Co	Cd
1	−2.37	−0.88	−0.72	−0.69	−0.43	−0.53	0.49
2	−2.45	−1.73	−1.48	−1.48	−0.94	−0.98	0.16
3	−2.36	−0.91	−1.32	−0.31	−0.33	−0.31	0.51
4	−2.42	−1.48	−1.39	−1.19	−0.73	−0.93	0.21
5	−2.37	−0.80	−1.05	−0.50	−0.39	−0.36	0.48
6	−2.38	−1.01	−0.81	−0.69	−0.49	−0.58	0.41
7	−2.39	−0.83	−1.68	−0.39	−0.20	−0.47	0.66
8	−2.36	−0.98	−0.26	−0.53	−0.32	−0.29	0.54
9	−2.36	−0.35	−1.62	−0.54	−0.24	−0.15	0.46
10	−2.38	−0.84	−0.28	−1.04	0.34	−0.62	0.42
11	−2.36	−1.43	−1.54	−0.33	−0.90	−0.18	0.44
12	−2.36	−0.85	−0.71	−0.67	0.32	−0.30	0.77
13	−2.37	−1.29	−1.37	−1.37	−0.38	−0.36	0.63
14	−2.36	−0.85	−1.57	−0.30	−0.81	−0.27	0.78
15	−2.36	−0.37	−1.65	−0.80	−0.29	−0.22	0.63
16	−2.36	−0.44	−1.67	−0.92	−0.10	−0.25	0.68
17	−2.36	−0.41	−1.67	−0.90	0.09	−0.20	0.60
18	−2.37	−0.49	−1.75	−1.16	−0.13	−0.24	0.72
19	−2.36	−0.42	−1.74	−1.16	−0.18	−0.20	0.75
20	−2.37	−0.60	−1.63	−1.71	−0.15	−0.31	0.71
21	−2.36	−0.31	−1.61	−0.73	−0.31	−0.16	0.71
22	−2.37	−0.58	−1.67	−0.78	−0.15	−0.28	0.64
23	−2.37	−0.60	−1.77	−0.78	−0.38	−0.28	0.52
24	−2.37	−0.50	−1.70	−0.79	−0.16	−0.71	0.66
25	−2.36	−0.73	−0.31	−0.63	−0.09	−0.23	0.80
26	−2.36	−0.69	−0.27	−0.66	−0.07	−0.26	0.80
27	−2.36	−0.68	0.04	−0.55	−0.11	−0.25	0.78
28	−2.05	−0.80	−0.45	−0.56	−0.13	−0.64	0.10
29	−2.03	−0.20	−0.22	−0.20	−0.05	−0.68	0.17
30	−2.03	−0.11	−0.29	−0.12	−0.17	−0.64	0.29
31	−2.03	−0.13	−0.30	−0.12	−0.16	−0.60	0.38
32	−2.37	−0.56	−0.88	−0.97	0.05	−0.57	0.41
33	−2.37	−0.67	−0.86	−0.93	0.20	−0.38	0.76
34	−2.37	−0.75	−0.41	−0.95	−0.21	−0.58	0.45

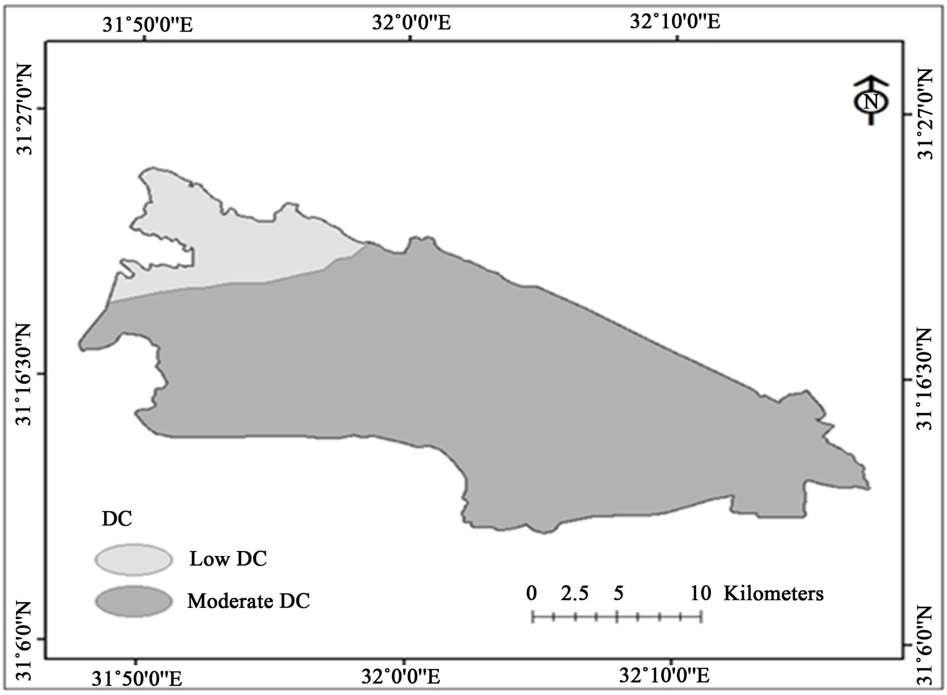

Figure 4. The spatial distribution of contamination degree (DC) of heavy metals in the hydrosoils of Manzala Lake.

Table 6. Pearson-moment correlation (r) between the concentrations of heavy metals in the hydrosoils and water of Manzala Lake.

Variables		Hydrosoil (μg/g)							Water (μg/l)						
		Fe	Cu	Zn	Cr	Pb	Co	Cd	Fe	Cu	Zn	Cr	Pb	Co	Cd
Hydrosoil (μg/g)	Fe	1													
	Cu	0.669**	1												
	Zn	0.426*	0.180	1											
	Cr	0.714**	0.495**	0.362*	1										
	Pb	0.046	0.119	0.268	−0.154	1									
	Co	−0.352*	0.057	−0.185	−0.109	−0.014	1								
	Cd	−0.445**	−0.080	−0.022	−0.269	0.239	0.641**	1							
Water (μg/l)	Fe	0.755**	0.202	0.367*	0.372*	0.116	−0.449*	−0.549**	1						
	Cu	0.721**	0.193	0.614**	0.498**	−0.073	−0.539**	−0.479**	0.800**	1					
	Zn	0.510.9**	−0.079	0.395*	0.546**	−0.181	−0.541**	−0.589**	0.606**	0.709**	1				
	Cr	−0.084	−0.240	−0.108	−0.156	−0.089	0.035	0.081	−0.063	−0.085	0.036	1			
	Pb	0.108	−0.211	0.272	0.309	−0.151	−0.125	−0.152	0.285	0.432*	0.336	−0.048	1		
	Co	0.934**	0.480**	0.481**	0.703**	−0.010	−0.536**	−0.510**	0.723**	0.805**	0.660**	−0.044	0.216	1	
	Cd	−0.149	−0.222	−0.214	0.113	−0.018	0.285	0.254	−0.100	−0.206	−0.170	0.368*	0.077	−0.120	1

**Correlation is significant at the 0.01 level. *Correlation is significant at the 0.05 level.

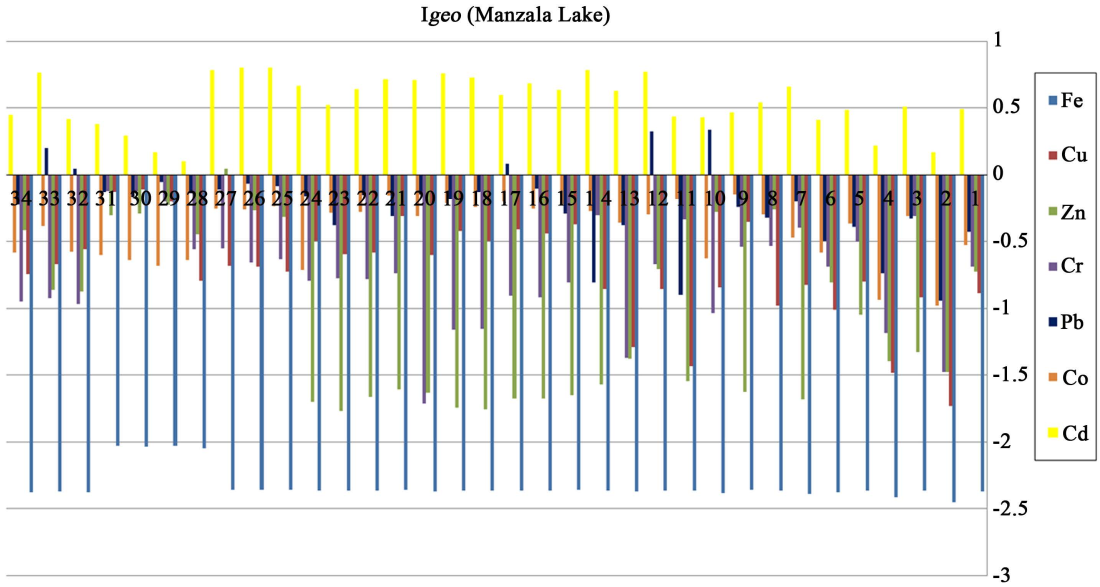

Figure 5. The geoaccumulation index for the heavy metals in the hydrosoils of Manzala Lake.

5. Conclusion

It could be concluded that the geospatial tools such as ordinary Kriging could be very helpful in evaluating and studying the spatial distribution of heavy metals in both water and sediment of Manzala Lake in Egypt. The obtained results clearly demonstrate that Manzala Lake is highly contaminated with Fe, Cd, Pb and Cr due to the continuous discharge of different pollutants into it. It can also be concluded that the southern drains namely, Bahr El-Baqar, Ramsis, El-Matria, Hadous, Faraskur, El-Serw and Lissa El-Gamalia play an important role in causing a severe pollution in Manzala Lake. This is especially in the southeastern parts of the lake, which receive great quantities of effluents from Bahr El-Baqar, Hadous and Ramsis drains. From the sequence of enrichment factor (EF), for heavy metals in the hydrosoils of Manzala Lake, cadmium is more abundant than other metals, whereas Cu shows the lowest appearance. Contamination factor (CF) showed that only nearby Bahr El-Baqar drain (moderate CF) and the CF of cadmium showed moderate, considerable and very high CF there. From the pollution load index (PLI) and degree of contamination (DC), it is showed that there is no appreciable pollution in Lake Manzala. Geo-accumulation index (Igeo) results of lead and cadmium showed moderately polluted category in all locations. Great efforts and cooperation between different authorities are needed to protect the lake from pollution and reduce the environmental risk. This can be achieved through the treatment of agricultural, industrial, and sewage discharge. Regular evaluation of pollutants in the lake is also very important.

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