

The Speciation and Potential Mobility of Pb, Cd, Cu and Zn in Lake Qarun Bottom Sediments, Fayioum, Egypt

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Abstract. The total concentrations and the solid phase speciation of lead (Pb), cadmium (Cd), copper (Cu) and zinc (Zn), in the bottom sediments from Lake Qarun, Fayioum, Egypt were determined. Besides, the total concentrations of the scavenging metals Fe and Mn were determined. The crustal enrichment factor (EF_{crustal}), the contamination factor (C_f), the modified degree of contamination (mC_d), the metal pollution index (MPI), as well as the geoaccumulation index (I_{geo}). The aim of this work is to investigate the solid phase fractionation distribution of certain hazardous heavy metals and to assess their potential mobility from sediment to water. The concentrations' ranges of Pb, Cd, Cu, Zn, Mn and Fe were 14.0-40.4 $\mu\text{g/g}$, 0.5-7.6 $\mu\text{g/g}$, 18.4-62.6 $\mu\text{g/g}$, 46.5-232.5 $\mu\text{g/g}$, 131.9-587.3 $\mu\text{g/g}$ and 6.4-40.2 mg/g , respectively. The total concentrations of heavy metals showed levels as normal as uncontaminated sediments, except Cd which was slightly higher than the natural levels. Most of the concentrations of Pb (~70%), Cu (~66%) and Zn (~58%) were in the immobile fractions of the solid phase (inert fraction) and they had little hazardous impact on the environment. On the other hand, Cd is enriched in the mobile phase of the sediment (reactive fraction) and strict measures should be applied to reduce or prevent the cadmium-bearing discharges into the lake. The trace metals flux to Lake Qarun sediments is mainly from allochthonous origin. The most probable discharge sources are the agricultural drains, the wastes of fish farms and the municipal wastewaters from the surrounding urban and rural communities.

Keywords: Heavy metals, chemical speciation, saline lake sediments, mobility and bioavailability, Lake Qarun, Egypt.

Introduction

Human activities have brought numerous potentially hazardous trace elements to the environment, particularly in the industrial period (Nriagu,

1996). During the last decades, the studies on heavy metal concentrations in sediments and their bioavailability have received increased scientific attention. The total concentrations of heavy metals indicate the extent of contamination, but they provide little information about the forms in which heavy metals are present, or about their potential for mobility and bioavailability in the environment (Lake, 1987). The mobility of heavy metals strongly depends not only on the characters of pore water, but also on their specific chemical forms or ways of association. It is necessary to identify and quantify the forms in which a metal is present to gain a better understanding of potential and actual mobility of metals under changing environmental conditions (Tessier and Campbell, 1987).

Many investigators used the speciation of heavy metals to study the potential mobility of metals (Tessier *et al.*, 1979; Salomons and Förstner 1984; Banat *et al.*, 2005; and Hu *et al.*, 2006), the solubilization by the decrease in pH, and the metal biosorption by the microbial biomass or the metal complexation with the newly formed humic substances or other factors (Hsu and Lo, 2001; and Zorpas *et al.*, 2003). The toxicity of certain metals depends particularly on their chemical forms rather than their total contents and, therefore, speciation studies gain increasing importance (Liu *et al.*, 2007; Zerbe *et al.*, 1999; Li *et al.*, 2001; Saad and Ahdy, 2006; Huang *et al.*, 2007; Mingbiao *et al.*, 2008; and Jos and Vink, 2009).

Although, Lake Qarun attracts the attention of many authors because of its historical and scientific importance as a unique ecosystem (Elewa, 1994; Ibrahim, 1996; Abdel-Satar *et al.*, 2003 and Ali and Fishar, 2005), the studies on the accumulation of heavy metals in its sediments and their speciation are scarce. The aim of this work, therefore, is to investigate the solid phase fractionation distribution of certain hazardous heavy metals and to assess their potential mobility from sediment to water.

Area of Study

Lake Qarun (Fig. 1) is the third largest lake in Egypt and is situated between longitudes of 30° 24' & 30° 49' E and latitudes 29° 24' & 29° 33' N. It is an inland isolated saline water basin of about 40 km length and 5.7 km width and its water depth ranges from 1 to 8 m meters. Its surface area is about 215 km² and has a volume about 1,100,000,000 m³. It lies in

the arid zone and is considered as a tropical lake. The water level in the lake depends on the balance between the inflow of drainage water coming from the cultivated land of Faiyoum Province (Dardir and Wali, 2009) and evaporation rate. The lake water is generally turbid, especially in the eastern and southern parts that are directly influenced by drainage water discharges. It was considered as an extensive Oasis situated in the Western Desert (Shata *et al.*, 2002).

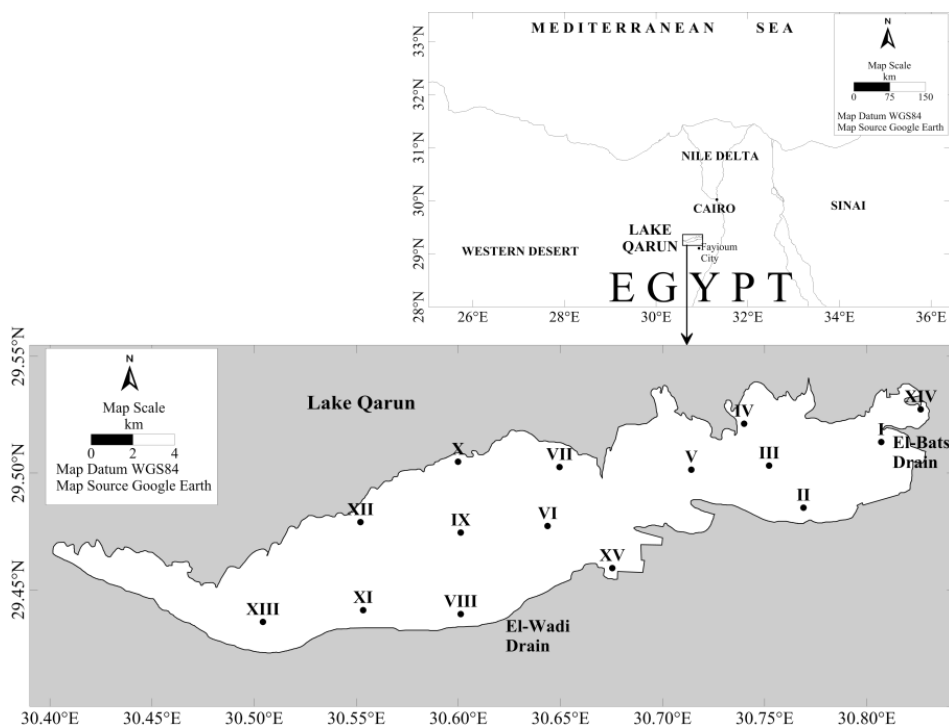


Fig. 1. A map showing the study area and locations of sediment sampling stations.

During the last 2000 years the water level in the Lake has dropped from 20 meters above mean sea level to about 45 meters below mean sea level (El-Abd *et al.*, 1997). The lake has no outlet and loses water mainly through evaporation which is subsequently controls dissolved metals concentrations (Shata *et al.*, 2002). Meshal and Morcos (1981) determined the total annual evaporation to be 190.1 cm, while the net water gain through seepage to the lake is $65.2 \times 10^6 \text{ m}^3$. The lake is bordered by cultivated lands at its southern and southeastern sides, and by an uninhabited desert at the northern side. It receives drainage waters

mainly from two large drains, namely, El-Bats and El-Wadi drains joining the lake at its eastern extremity and at the middle of its southern borders (Fig. 1). The bottom of Lake Qarun is characterized by the presence of great accumulations of calcareous shells and fragments of mollusks (Shata *et al.*, 2002). The sediments near the drains contain a higher percentage of silt and clay, whereas they are blanketed with a wind-blown layer of sand at the northern side of the lake (Shata *et al.*, 2002).

Materials and Methods

Sampling

Thirteen surficial sediment samples, at depths ranging from 1 to 8 m, were collected from Lake Qarun, and two from the outlets of El-Bats and El-Wadi Drains. Sampling locations were selected to cover the study area as shown in Fig. (1).

In the laboratory, samples were stored in polypropylene centrifuge tubes and kept in the freezer at (-20°C). Prior to processing and analysis sediment samples were dried at room temperature, sieved using the -1 phi mesh sieve to separate stones and shells, and then grounded with a pestle and agate mortar (Che *et al.*, 2003).

Determination of Total and Forms of Heavy Metals in Sediments

A modified five-step sequential extraction procedure (Tessier *et al.*, 1979; Steinberg and Tayarani-Dastmalian 1993; Perin *et al.*, 1997; and Akcay *et al.*, 2003) was used to determine the distribution of heavy metals in the following operational fractions:

Step 1: To extract the exchangeable metal (F1), which is mainly affected by adsorption-desorption processes.

Step 2: To extract the metal, bound to carbonate phase (F2), which is affected by solid solution reactions and changes of pH.

Step 3: To extract the metal bound to Fe-Mn oxide phase (F3), which is unstable under variable redox condition.

Step 4: To extract the metal bound to organic matter and sulphide (F4).

Step 5: To dissolve the residue from step 4 (F5) which is occluded in the crystal lattice of resistant minerals.

Trace metal concentrations were determined using Atomic Absorption Spectrophotometry (Spectr AA-10 plus Varian) and expressed as ($\mu\text{g/g}$). Copper, zinc, iron and manganese were determined using Flame-AAS while lead and cadmium were determined by Graphite furnace-AAS. The accuracy of the studied metals was conducted using increments of the solutions. The recovery percentages ranged from 90 to 100 for the studied metals. The precision of the technique was tested, by replicate analysis of the studied metals, using Standard Reference Material of IAEA-SL-1 (International Atomic Energy Agency), as shown in Table 1. All laboratory glassware were washed in 10% HNO_3 solution and rinsed with deionized water prior to each use (Burger *et al.*, 2001).

Table 1. Replicate analysis for IAEA-SL-1 standard reference material of metals $\mu\text{g/g}$.

IAEA-SL-1	Pb = 37.7	Cd = 0.26	Cu = 30.0	Zn = 223	Fe = 67400	Mn = 3460
	35.2	0.31	28.8	231	65900	3278
	34.8	0.27	33.9	228	66800	3303
	41.2	0.28	28.2	225	67001	3309
	40.6	0.25	32.7	220	65904	3471
	34.1	0.24	32.1	235	63840	3485
Mean	37.18 \pm 3.4	0.27 \pm 0.025	31.14 \pm 2.5	227 \pm 5.7	65889 \pm 1251	3369 \pm 100
CV%	9.14	9.26	8.03	2.51	1.90	2.97

CV% = Coefficient of variation in percent.

Metal Pollution Index

Metal Pollution Index (MPI) is used to determine the most polluted locations (Usero *et al.*, 1996; 1997) by applying the formula:

$$\text{MPI} = (M_1 \times M_2 \times M_3 \times \dots \times M_n)^{1/n}$$

Where, M_n is the concentration expressed in $\mu\text{g/g}$ dry weight (dw) of each investigated metals.

Enrichment Factor (EF)

A common approach to assess the enrichment of an element relative to its source is to calculate the normalized enrichment factor (EF) (Hörnung *et al.*, 1989; and Deely and Fergusson 1994).

The normalized enrichment factor (EF) was calculated using the formula

$$\text{EF}_{\text{source}} = (\text{E/I})_{\text{sample}} / (\text{E/I})_{\text{source}}$$

Where,

$(E/I)_{\text{sample}}$ = ratio of the concentration of the element E to the concentration of the indicator in the sample

$(E/I)_{\text{source}}$ = ratio of the concentration of same element E to the concentration of the indicator in the source material

Source indicator or marker I, is usually a conservative element that is derived predominantly from one specific source (Martin and Meybek, 1979).

Geoaccumulation Index (I_{geo})

A common approach to estimate the enrichment of metal concentrations above background or baseline concentrations is to calculate the geoaccumulation index (I_{geo}). This index is calculated as follows:

$$I_{\text{geo}} = \log_2 C_n / 1.5B_n$$

Where,

C_n is the concentration of the element in the enriched samples, and

B_n is the background or pristine value of the element (Bowen, 1966).

The factor 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic variations in the sediments (Stöffers *et al.*, 1986).

Modified Degree of Contamination (mC_d)

The modified degree of contamination (mC_d) is defined according to Håkanson (1980) and modified by Abraham (2005). The modified degree of contamination is given by:

$$mC_d = C_d / n$$

Where,

n = number of analyzed elements and

C_d is the degree of contamination and defined as the sum of the contamination factor for the four studied metals

$$C_d = \sum_{i=1}^n (C_f^i)$$

Where,

i = the element (or pollutant) and

C_f is defined as contamination factor of each individual metal and is given by:

$$C_f = M_x / M_b$$

Where,

M_x and M_b , are the mean concentration of a pollutant in the contaminated sediments and the preindustrial (baseline) sediments (Bowen, 1966).

Potential Mobility (PM)

The potential mobility (%) of a metal strongly depends on its specific form(s) of association with the host sediment (Huang *et al.*, 2007) and is given by:

$$PM = 100 \cdot \Sigma C_{F1} + C_{F2} + C_{F3} + C_{F4} / C_T$$

Where,

C_{Fn} is the partial concentration of the metal in the fractions F_1 to F_4 ,

C_T is the total concentration of the same metal in the sediment

Results and Discussions

Total Heavy Metals

The total concentrations of the investigated metals (Pb, Cd, Cu, Zn, Fe, and Mn) and the available threshold effect level (TEL) and probable effect level (PEL) are given in Table 2. Canadian Council of Ministers of the Environment (CCME) considered the threshold effect level (TEL), as the concentration above which some adverse biological effect (or response) will be produced and below which it will not. The probable effect level (PEL) is the concentration which is likely to cause an adverse biological. Generally, toxicity rarely occurs below the threshold effect level (TEL) and frequently occurs above the probable effect level (PEL) (MacDonald *et al.*, 1996).

Lake Qarun showed a gradual increase in salinity during the last century, due to its closed nature and extensive evaporation. Therefore, salts, heavy metals and pesticides, carried by agricultural drainage water, accumulate in the lake components. The interpretation of the distribution of metals in this study has to consider two different aspects:

1. The relative position of the sampling station to the pollutant sources;

2. Environmental characteristics by agricultural drainage water.

Table 2. Total concentration, means and MPI of heavy metals in Lake Qarun sediments.

St.	Pb $\mu\text{g/g}$	Cd $\mu\text{g/g}$	Cu $\mu\text{g/g}$	Zn $\mu\text{g/g}$	Fe mg/g	Mn $\mu\text{g/g}$	MPI
I	36.75	5.87	58.62	208.93	32.84	500.59	187.49
II	28.59	3.62	46.02	155.07	19.57	558.19	141.62
III	31.75	5.02	51.69	175.63	24.78	577.18	165.69
IV	21.56	1.21	36.02	148.99	17.12	517.49	103.63
V	30.37	4.06	47.91	162.29	21.44	468.53	145.86
VI	24.62	1.98	37.98	139.09	19.51	426.06	113.51
VII	20.16	4.67	38.98	173.32	25.58	415.25	137.49
VIII	22.87	2.17	41.23	148.99	15.81	536.32	117.14
IX	29.77	3.09	44.56	154.82	14.61	304.45	118.87
X	18.24	1.51	30.76	141.13	11.96	277.52	85.72
XI	33.82	0.98	35.88	62.98	10.46	230.19	75.16
XII	14.76	1.07	18.42	46.53	6.43	131.90	47.50
XIII	14.05	0.53	20.43	73.67	9.51	206.02	52.91
XIV	40.42	7.62	62.61	232.54	40.25	587.31	217.54
XV	16.97	4.02	22.90	144.26	24.96	404.37	114.68
Mean	25.65	3.16	39.60	144.55	19.66	409.42	121.66
S.D.	± 8.16	± 2.06	± 13.03	± 50.41	± 9.1	± 146.57	
Sum	384.70	47.42	594.01	2168.24	294.77	6141.37	
Min.	14.05	0.53	18.42	46.53	6.43	131.90	47.50
Max.	40.42	7.62	62.61	232.54	40.25	587.31	217.54
TEL	35.00	0.60	36.00	123.00			
PEL	91.00	3.50	197.00	315.00			

Lead is a non-essential element, being a toxic metal that can affect humans when ingested or inhaled in high doses. In this study the concentrations of lead were within the range of 14.05-40.42 $\mu\text{g/g}$ with the lowest at station XII and the highest at station XIV, with a mean value 25.65 $\mu\text{g/g}$. Lead concentrations are generally below TEL values, which indicates that the adverse biological effects on the fauna and flora of the lake are expected to occur rarely (except at stations I and XIV), located close to El-Bats Drain.

Cadmium is a hazardous environmental contaminant that is also transported atmospherically. In fish, it can cause anaemia, vertebral fractures, decreased digestive efficiency and biochemical effects (Burger *et al.*, 2002). Cadmium concentrations in Lake Qarun sediments ranged between 0.53 $\mu\text{g/g}$ at station XIII and 7.62 $\mu\text{g/g}$ at station XIV, with a mean value of 3.16 $\mu\text{g/g}$. All Cadmium concentrations showed values higher than (TEL), except at station XIII, while 47% of studied locations were higher than (PEL). These results indicated that the lake sediments are contaminated with Cd. The highest Cd concentration was about five

times the TEL. The increase in the level of Cd in Lake Qarun sediments might be the result of the intensive application of phosphate fertilizers that are later washed out to the lake through El-Bats drain. Murphy and Doskey (1975) stated that Cd is known to be linked to phosphate fertilizers.

Copper is an essential element for growth of aquatic organisms. At high concentration level, it accumulates in blood, liver and kidneys and causes anemia, renal and intestinal irritations. The current study reveals that the concentration of copper fluctuated between 18.42 at station XII to 62.61 at station XVI, with an average of $39.60 \pm 13.03 \mu\text{g/g}$. 60% of stations were higher than TEL, but still lower than PEL.

Zinc is an essential micronutrient in all marine organisms, being a cofactor in nearly 300 enzymes. Therefore, marine animals are able to regulate Zn at the concentrations in sea water and sediments from normal ambient levels to incipient lethal levels (França, *et al.*, 2005). High levels of Zn may cause pancreatitis, anemia, muscle pain, acute renal failure (Pais and Benton Jones, 1997). In the present study, the concentrations of Zn ranged from 46.53 at station XII to 232.54 at station XIV, with an average of $144.55 \pm 50.41 \mu\text{g/g}$. The concentrations of zinc at all stations (except at stations XI, XII and XIII) were higher than TEL, but still lower than PEL.

Iron is an important trace element required for all biological life activities. At high concentrations, iron II is toxic for most organisms (Abdel-Halim, 2004). In the present study, Fe ranked first in the order of abundance compared to the other five metals. The concentrations of Fe fluctuated between 6.43 at station XII and 40.25 at station XIV, with a mean value of $19.66 \pm 9.1 \text{ mg/g}$.

Manganese is one of the more bio-chemically active transition metals in aquatic environment. The concentrations of manganese ranged from 131.90 to 587.31 $\mu\text{g/g}$ with a mean value of $409.42 \pm 146.57 \mu\text{g/g}$ (Table, 2). It occupied the second order of abundance of studied metals. The higher concentration of Mn in the lake sediments may be due to the precipitation of Mn-oxides under lower Eh conditions in the lagoonal environment (Eaton, 1979). The solubility of Mn-sulfide compounds is the highest among all studied sulfide-forming metals. Part of the dissolved Mn may be trapped in the sediments and the other part may diffuse to the water column.

Table 2 shows that the distributions of studied heavy metals in Lake Qarun have highest values in the eastern side (Stations I and XIV), due to the discharges of polluted water from El-Bats Drain and other anthropogenic sources in this part of the lake. Westward the levels of pollution are decreased relatively. These results are in agreement with those of Ali and Fishar (2005) and Ali (2002). The presence of trace metals in Lake Qarun is mainly of allochthonous origin, due to either agricultural influx, wastes of fish farms or sewage *via* surrounding cultivated lands.

Metal pollution index (MPI)

As shown from Table 2, the lowest MPI value was recorded at station XII, while the highest was found at station XIV and followed by station I, which indicated that the lowest polluted area was at station XII. Meanwhile, the highest polluted areas in the studied locations were at stations XIV and I. This might be reasonable, due to their location close to El-Bats Drain that drains the Lake with large amounts of industrial and agricultural wastes.

Enrichment Factor (EF)

Enrichment factor is a good tool to differentiate the metal source between anthropogenic and naturally occurring. Table 3 presents the relation between the present content of a given metal in bottom sediments and its pre-industrial one. The enrichment factors of cadmium in certain European lakes are as follows: 2.6 in Lake Varnen (Hakanson, 1980); 2.9 in Lake Constance (Förstner and Müller, 1974) and 4.6 in Lake Geneva (Vernet, 1976). These values are very low compared with the present study, confirming that the bottom sediments' loading with cadmium is considerably higher ($EF > 3$). It could be inferred that Lake Qarun sediments can be regarded as unpolluted with Mn while it is slightly enriched with Zn and Cu and extremely enriched with Cd.

Geoaccumulation Index (I_{geo})

In Lake Qarun both (I_{geo}) and (mC_d) are calculated for Cd, Pb, Cu and Zn. The values of preindustrial reference (B_n) have been taken from the background lakes' sediments reference values according to Hakanson (1980). The geoaccumulation index (Table, 4) consists of 7 grades or classes (Müller, 1981).

The I_{geo} for individual elements at the investigated stations are presented in Table 5. The negative I_{geo} values found in Table 5 reveals low levels of contamination for Pb, Cu and Zn, which fall in class 0, *i.e.* practically uncontaminated. For Cd, 26.67 % of samples falls in class 0 (uncontaminated), 20% of samples falls in class 1 (uncontaminated to moderately contaminated), 46.67% of samples falls in class 2 (moderately contaminated) and only one sample falls in class 3 (station XIV), which indicated moderately to heavily contaminated.

Table 3. The enrichment factors (EF) for Pb, Cd, Cu and Zn in Lake Qarun sediments.

Station	Pb	Cd	Cu	Zn	Mn	Means
I	2.51	32.08	2.00	1.80	0.76	7.83
II	3.28	33.20	2.64	2.24	1.42	8.56
III	2.87	36.36	2.34	2.00	1.16	8.95
IV	2.83	12.69	2.36	2.46	1.51	4.37
V	3.18	34.00	2.51	2.14	1.09	8.58
VI	2.83	18.22	2.18	2.02	1.09	5.27
VII	1.77	32.77	1.71	1.92	0.81	7.80
VIII	3.26	24.64	2.93	2.66	1.69	7.04
IX	4.57	37.96	3.42	3.00	1.04	10.00
X	3.42	22.66	2.89	3.34	1.16	6.69
XI	7.26	16.82	3.85	1.70	1.10	6.15
XII	5.15	29.87	3.21	2.05	1.02	8.26
XIII	3.32	10.00	2.41	2.19	1.08	3.80
XIV	2.25	33.98	1.75	1.63	0.73	8.07
XV	1.53	28.91	1.10	1.63	0.81	6.80
Mean	3.34	26.94	2.49	2.19	1.10	7.21
Min.	1.53	10.00	1.10	1.63	0.73	3.80
Max.	7.26	37.96	3.85	3.34	1.69	10.00

Table 4. The geoaccumulation index consisting of 7 grades or classes (Müller, 1981).

I_{geo} value	I_{geo} class	Designation of sediment quality
≥ 5	6	Extremely contaminated
4-5	5	Heavily to extremely contaminated
3-4	4	Heavily contaminated
2-3	3	Moderately to heavily contaminated
1-2	2	Moderately contaminated
0-1	1	Uncontaminated to moderately contaminated
≤ 0	0	Practically uncontaminated

Table 5. Geoaccumulation indexes (I_{geo}), contamination factors (C_f), degree of contamination (C_d) and modified degree of contamination (mC_d) of Pb, Cd, Cu and Zn in Lake Qarun.

Station	Index of geoaccumulation (I_{geo})				Contamination factors (C_f)				(C_d)	mC_d
	Pb	Cd	Cu	Zn	Pb	Cd	Cu	Zn		
I	-1.51	1.97	-0.36	-0.33	0.53	5.87	1.17	1.19	8.76	2.19
II	-1.88	1.27	-0.70	-0.76	0.41	3.62	0.92	0.89	5.84	1.46
III	-1.73	1.74	-0.54	-0.58	0.45	5.02	1.03	1.00	7.50	1.88
IV	-2.28	-0.31	-1.06	-0.82	0.31	1.21	0.72	0.85	3.09	0.77
V	-1.79	1.44	-0.65	-0.69	0.43	4.06	0.96	0.93	6.38	1.60
VI	-2.09	0.40	-0.98	-0.92	0.35	1.98	0.76	0.79	3.88	0.97
VII	-2.38	1.64	-0.94	-0.60	0.29	4.67	0.78	0.99	6.73	1.68
VIII	-2.20	0.53	-0.86	-0.82	0.33	2.17	0.82	0.85	4.17	1.04
IX	-1.82	1.04	-0.75	-0.76	0.43	3.09	0.89	0.88	5.29	1.32
X	-2.53	0.01	-1.29	-0.90	0.26	1.51	0.62	0.81	3.20	0.80
XI	-1.63	-0.61	-1.06	-2.06	0.48	0.98	0.72	0.36	2.54	0.64
XII	-2.83	-0.49	-2.03	-2.50	0.21	1.07	0.37	0.27	1.92	0.48
XIII	-2.90	-1.50	-1.88	-1.83	0.20	0.53	0.41	0.42	1.56	0.39
XIV	-1.38	2.34	-0.26	-0.17	0.58	7.62	1.25	1.33	10.78	2.70
XV	-2.63	1.42	-1.71	-0.87	0.24	4.02	0.46	0.82	5.54	1.39
Mean	-2.11	0.73	-1.00	-0.97	0.37	3.16	0.79	0.83	5.15	1.29

Modified Degree of Contamination (mC_d)

The mC_d for investigated area generally lies in the range 0.39–2.70. According to classification system proposed for the modified equation, about 60% of samples recorded very low degree of contamination, 26.67 % of samples revealed low degree of contamination and 13.33 % of samples recorded moderate degree of contamination. In considering the C_f results, cadmium is significantly enriched at some stations (particularly at stations XIV, I and III (Table 5), which may be due to the use of fertilizer. The input of phosphate fertilizer, which contains some Cd, is yet a source for the accumulation of Cd in sediments, such as seen in Lake of Yangtze in China. (Yang *et al.*, 2009).

Speciation of Metals

Depending on chemical and geological conditions, heavy metals can be partitioned into different chemical forms that are associated with a variety of organic and inorganic phases (Arain, *et al.*, 2007). Thus, speciation analysis of heavy metals might provide much useful information regarding the chemical nature or potential mobility and bioavailability of a particular element, which consequently can offer a more realistic estimate of actual environmental impact (Akçay *et al.*, 2003; and Cuong and Obbard, 2006).

It is clear from Table 6 and Fig. (2) and (3) that the fractionation of the studied metals among the solid phase, except cadmium, are lowest in the exchangeable fraction; their ranges are (0.38-1.36, 0.26-0.43 and 0.55-1.32 $\mu\text{g/g}$) for lead, zinc and copper, respectively. These results were in agreement with those of Saad and Ahdy (2006); Huang *et al.*, (2007); and Mingbiao *et al.*, (2008). High percentage of exchangeable Cd (38.92%) was observed; this value was associated with the higher capacity of principal polarization, resulting in strong adsorption of Cd onto colloids in the sediments. Generally, the **F1** fraction had the greatest tendency toward remobilization from the sediment phase to the more bioavailability pore water phase (Li *et al.*, 2007). Also, significant high concentrations of exchangeable Cd in sediments have been reported by Yang *et al.*, (2009) and Yuan *et al.*, (2004). The second form of Cd is **F2**, the mean percentage recorded 35.47, ranging from 34.55 to 36.41% (Table 6), may be due to the increase in concentration of carbonates in the sediments (Mansour and Sidky, 2003). Förstner and Wittmann (1983) reported that the occurrence of cadmium in sediments was mainly related to the formation of CdCO_3 . Lin and Chen (1996); Saad and Ahdy (2006) observed that cadmium was strongly associated with carbonate. The cadmium in **F2** is the most liable; hence it might be available for uptake by biota. The mean percentage residual phase (**F5**) of the same element was 13.02%, followed by **F3** (6.59%) and the least was **F4** (6.00%). Overall, the speciation of Cd in the sediments from this lake exhibited high bioavailability and consequently this element posed a high ecological risk. Accordingly, it could be concluded that Lake Qarun is suffering from the impact of anthropogenic and agricultural waste discharges.

The mean percentages of **F5** fraction accounted for about 66.05% for Cu and about 57.7% for Zn. The next important phase of copper was the organic fraction, due to the high stability of organic-Cu compounds. This result is in agreement with Li *et al.*, (2001) and Stumm and Morgan (1981). The percentages of **F4**-Cu ranged from 11.39 to 31.62%, which were higher than those of Zn (3.06-8.95%). This is probably due to the well-known tendency of copper to form organic complexes *e.g.* with humic acids and the various metabolites deriving from algal biomass decomposition and other biochemical reactions (Leal *et al.*, 1999). The **F5**-Cu is likely bound to ferric oxides, which would be released when environmental conditions changes (Yang *et al.*, 2009). The percentages

range of F2-Zn and F3-Zn were 15.67-26.70% and 5.08-12.19%, respectively, which were greater than those of Cu 6.25-9.71% and 4.55-6.98%, respectively. These data suggest that Cu and Zn in the sediments from lake might pose some ecological risk.

In the study area, the F5-Pb fraction is 56.77–78.96%. The fractions bound to F3 reducible oxides were about 5.67-28.00%. These values were related with Fe and Mn dioxide and crystal ferric oxides (Ramos, *et al.*, 1994). When the environmental conditions changed, this fraction was easily released from the sediment phase into the water. Additionally, F1-Pb, which had the highest mobility, reached 0.38–1.36%. Pb can affect the procreation ability of a biological system.

Table 6. Solid speciation data of heavy metals ($\mu\text{g/g}$) in Lake Qarun.

Metal	Station	F1	%	F2	%	F3	%	F4	%	F5	%	TSE	TD	
Pb	I	1.36	3.30	1.84	4.47	4.53	11.01	0.97	2.36	32.46	78.86	41.16	36.75	
	II	0.95	3.11	1.59	5.20	4.09	13.37	1.23	4.03	22.73	74.31	30.59	28.59	
	III	1.12	3.21	1.67	4.78	2.65	7.59	3.68	10.54	25.81	73.89	34.93	31.75	
	IV	0.67	2.99	1.27	5.66	3.74	16.68	0.66	2.94	16.08	71.72	22.42	21.56	
	V	1.05	3.17	1.63	4.92	4.35	13.14	0.94	2.84	25.13	75.92	33.10	30.37	
	VI	0.79	3.03	1.40	5.36	3.18	12.18	1.55	5.94	19.18	73.49	26.10	24.62	
	VII	0.81	3.90	1.21	5.83	4.89	23.55	1.10	5.30	12.75	61.42	20.76	20.16	
	VIII	0.72	3.00	1.33	5.54	2.87	11.95	1.64	6.83	17.45	72.68	24.01	22.87	
	IX	1.00	3.11	1.63	5.07	2.32	7.22	3.05	9.49	24.15	75.12	32.15	29.77	
	X	0.54	2.90	1.11	5.97	3.40	18.28	0.67	3.60	12.88	69.25	18.60	18.24	
	XI	1.22	3.25	1.73	4.61	2.83	7.54	3.80	10.12	27.96	74.48	37.54	33.82	
	XII	0.41	2.81	0.92	6.30	3.99	27.31	0.41	2.81	8.88	60.78	14.61	14.76	
	XIII	0.38	2.76	0.88	6.39	3.66	26.58	0.65	4.72	8.20	59.55	13.77	14.05	
	XIV	1.21	2.65	1.37	3.00	2.59	5.67	4.44	9.72	36.06	78.96	45.67	40.42	
	XV	0.49	2.86	1.05	6.13	4.80	28.00	1.07	6.24	9.73	56.77	17.14	16.97	
	Max.		1.36	3.90	1.84	6.39	4.89	28.00	4.44	10.54	36.06	78.96		
	Min.		0.38	2.65	0.88	3.00	2.32	5.67	0.41	2.36	8.20	56.77		
	Mean		0.85	3.07	1.38	5.28	3.59	15.34	1.72	5.83	19.96	70.48		
Cd	I	2.95	40.52	2.63	36.13	0.47	6.46	0.34	4.67	0.89	12.23	7.28	5.87	
	II	1.71	39.68	1.53	35.50	0.25	5.80	0.35	8.12	0.47	10.90	4.31	3.62	
	III	2.28	36.95	2.22	35.98	0.43	6.97	0.27	4.38	0.97	15.72	6.17	5.02	
	IV	0.53	38.41	0.48	34.78	0.09	6.52	0.10	7.25	0.18	13.04	1.38	1.21	
	V	1.95	39.71	1.75	35.64	0.36	7.33	0.21	4.28	0.64	13.03	4.91	4.06	
	VI	0.91	39.57	0.81	35.22	0.14	6.09	0.13	5.65	0.31	13.48	2.30	1.98	
	VII	2.28	40.00	2.04	35.79	0.45	7.89	0.32	5.61	0.61	10.70	5.70	4.67	
	VIII	0.99	38.98	0.89	35.04	0.14	5.51	0.14	5.51	0.38	14.96	2.54	2.17	
	IX	1.44	39.45	1.29	35.34	0.27	7.40	0.18	4.93	0.47	12.88	3.65	3.09	
	X	0.67	38.51	0.61	35.06	0.08	4.60	0.14	8.05	0.24	13.79	1.74	1.51	
	XI	0.42	38.18	0.38	34.55	0.09	8.18	0.09	8.18	0.12	10.91	1.10	0.98	
	XII	0.46	38.02	0.42	34.71	0.07	5.79	0.11	9.09	0.15	12.40	1.21	1.07	
	XIII	0.22	37.29	0.21	35.59	0.04	6.78	0.03	5.08	0.09	15.25	0.59	0.53	

Metal	Station	F1	%	F2	%	F3	%	F4	%	F5	%	TSE	TD
	XIV	3.18	40.66	2.84	36.32	0.52	6.65	0.35	4.48	0.93	11.89	4.82	7.62
	XV	3.61	37.88	3.47	36.41	0.65	6.82	0.45	4.72	1.35	14.17	9.53	4.02
	Max.	3.61	40.66	3.47	36.41	0.65	8.18	0.45	9.09	1.35	15.72		
	Min.	0.22	36.95	0.21	34.55	0.04	4.60	0.03	4.28	0.09	10.70		
	Mean	1.57	38.92	1.44	35.47	0.27	6.59	0.21	6.00	0.52	13.02		
Cu	I	0.29	0.58	4.65	9.22	3.52	6.98	5.74	11.39	36.21	71.83	50.41	58.62
	II	0.31	0.76	3.54	8.64	2.53	6.18	6.47	15.80	28.11	68.63	40.96	46.02
	III	0.31	0.69	4.05	9.01	2.89	6.43	5.91	13.14	31.81	70.74	44.97	51.69
	IV	0.43	1.27	2.56	7.56	1.82	5.38	7.92	23.39	21.13	62.40	33.86	36.02
	V	0.26	0.62	3.77	8.94	2.68	6.36	6.07	14.40	29.38	69.69	42.16	47.91
	VI	0.38	1.08	2.79	7.90	1.98	5.61	7.36	20.84	22.81	64.58	35.32	37.98
	VII	0.35	0.98	2.90	8.09	2.06	5.74	7.02	19.58	23.53	65.62	35.86	38.98
	VIII	0.32	0.85	3.10	8.26	2.25	6.00	6.85	18.26	25.00	66.63	37.52	41.23
	IX	0.32	0.80	3.39	8.45	2.43	6.06	6.84	17.06	27.12	67.63	40.10	44.56
	X	0.41	1.39	2.18	7.38	1.55	5.25	7.29	24.69	18.10	61.29	29.53	30.76
	XI	0.40	1.17	2.64	7.74	1.87	5.49	7.53	22.09	21.65	63.51	34.09	35.88
	XII	0.29	1.59	1.14	6.25	0.83	4.55	5.14	28.18	10.84	59.43	18.24	18.42
	XIII	0.30	1.50	1.41	7.04	1.02	5.09	6.33	31.62	10.96	54.75	20.02	20.43
	XIV	0.28	0.53	5.17	9.71	3.67	6.90	6.23	11.71	37.87	71.16	53.22	62.61
	XV	0.32	1.44	1.56	7.02	1.11	5.00	3.05	13.73	16.17	72.81	22.21	22.90
	Max.	0.43	1.59	5.17	9.71	3.67	6.98	7.92	31.62	37.87	72.81		
	Min.	0.26	0.53	1.14	6.25	0.83	4.55	3.05	11.39	10.84	54.75		
	Mean	0.33	1.02	2.99	8.08	2.15	5.80	6.38	19.06	24.05	66.05		
	Zn	I	0.77	0.39	33.01	16.63	24.81	12.50	9.05	4.56	130.84	65.92	198.48
II		1.18	0.73	36.76	22.79	23.99	14.88	9.37	5.81	89.97	55.79	161.27	155.07
III		0.82	0.48	31.36	18.41	22.13	12.99	5.21	3.06	110.84	65.06	170.36	175.63
IV		1.05	0.64	34.96	21.33	23.05	14.06	8.60	5.25	96.23	58.72	163.89	148.99
V		0.86	0.52	31.64	19.11	21.82	13.18	10.74	6.49	100.48	60.70	165.54	162.29
VI		1.11	0.69	35.53	22.02	23.28	14.43	8.70	5.39	92.72	57.47	161.34	139.09
VII		0.74	0.43	30.46	17.75	21.91	12.77	9.56	5.57	108.92	63.48	171.59	173.32
VIII		1.26	0.78	37.73	23.45	24.73	15.37	10.16	6.31	87.03	54.09	160.91	148.99
IX		0.94	0.57	32.62	19.88	22.02	13.42	6.98	4.25	101.55	61.88	164.11	154.82
X		1.32	0.82	38.92	24.19	25.51	15.86	12.19	7.58	82.95	51.56	160.89	141.13
XI		0.69	0.91	19.62	25.96	12.78	16.91	6.75	8.93	35.74	47.29	75.58	62.98
XII		0.55	0.97	15.16	26.70	9.89	17.42	5.08	8.95	26.09	45.96	56.77	46.53
XIII		0.75	0.86	21.85	25.14	14.24	16.38	7.06	8.12	43.03	49.50	86.93	73.67
XIV		0.74	0.34	33.88	15.67	26.34	12.18	8.91	4.12	146.39	67.69	216.26	232.54
XV		0.99	0.60	33.80	20.62	22.61	13.80	7.38	4.50	99.11	60.47	163.89	144.26
Max.		1.32	0.97	38.92	26.70	26.34	17.42	12.19	8.95	146.39	67.69		
Min.		0.55	0.34	15.16	15.67	9.89	12.18	5.08	3.06	26.09	45.96		
Mean		0.92	0.65	31.15	21.31	21.27	14.41	8.38	5.93	90.13	57.70		

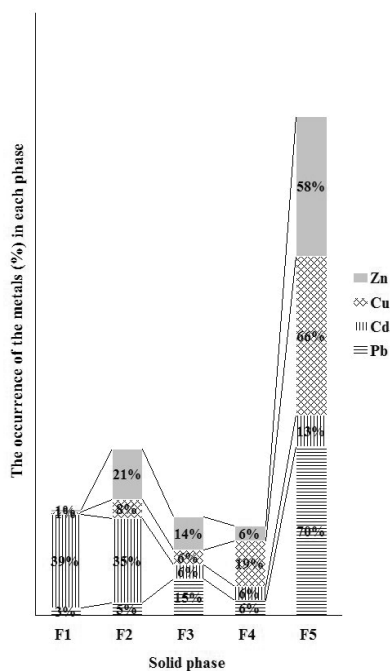


Fig. 2. Comparison of the occurrence of the studied metals (%) in each phase.

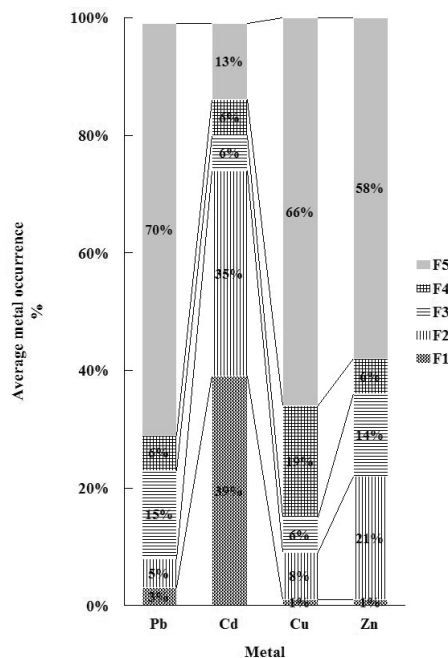


Fig. 3. The average metal occurrence (%) in the mobile forms (F1-F4) and the inactive form F5.

The studied metals content in the chemical phase gave the following order for the sediments: F5 > F3 > F4 > F2 > F1 for lead; F1 > F2 > F5 > F3 > F4 for cadmium; F5 > F4 > F2 > F3 > F1 for copper and F5 > F2 > F3 > F4 > F1 for zinc (Figs. 2 and 3). The studied metals' speciation was in excellent agreement with those mentioned in Yang *et al.*, (2009). Metals bound to these different phases will behave differently in the sedimentary environment and thus have different potentials for re-mobilization and uptake by biota.

The potential mobility of a metal (the probability of a metal to move from sediment to water) strongly depends on the specific chemical forms of association. The potential mobility of the studied metals is illustrated in Fig. 4, which shows that the mobility of the four metals had the following order: cadmium (86.98%) > zinc (42.30%) > copper (33.96%) > lead (29.52%). As the percentages of the mobile phases increase, the active fractions might be easily released and become hazardous to the environment and the human beings through the food chain (Mingbiao *et al.*, 2008). Cadmium might cause serious

environmental harm as it is present mainly in the reactive fractions of the solid phase and might be released to the water column. On the other hand, the average solid phase occurrences (%) of lead, copper and zinc were in the residue (inert fraction), and amounted to ~70%, 66% and 58%, respectively, and they have little impact on the lake's environment.

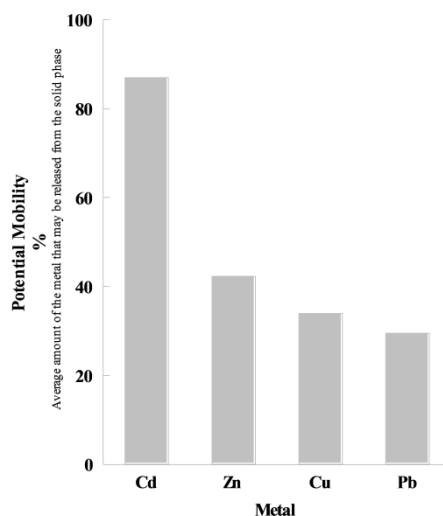


Fig. 4. The potential mobility (%) of metals.

Conclusions

The impact of heavy metal flux into the sedimentary environment of Lake Qarun was evaluated using Enrichment Factors (EF) and geoaccumulation indexes (I_{geo}) to assess the degree of contamination of each individual metal. The modified degree of contamination (mC_d) and metal pollution index (MPI) are also used to assess potential mobility of heavy metals.

The trace metals flux to Lake Qarun sediments is mainly of allochthonous origin. The discharges are agricultural, wastes of fish farms and municipal wastewaters from the surrounding urban and rural communities.

It was found that the total concentrations of the studied heavy metals are in the same concentration level of natural sediment, except cadmium, which has a higher concentration.

Considering the enrichment factor (EF), Lake Qarun can be regarded as unpolluted with Mn. It is highly polluted with Cd followed by $Pb > Cu > Zn$. The geoaccumulation index (I_{geo}), for Pb, Cu and Zn showed practically uncontaminated levels, while $I_{geo}Cd$ fluctuated from moderately to heavily contamination. The modified contamination factor (mC_d), showed that stations I and XIV are classified as considerably moderately contaminated and the rest of stations are between low and very low degree of contamination. Metal pollution index (MPI) showed that station XIV had the highest MPI, followed by station I, while station XII had the lowest MPI.

The results from the sediment fractionation indicated that Cd was mainly bounded to the exchangeable phase of sediments, followed by carbonate fraction. Accordingly, the speciation of Cd in the sediments from the Lake Qarun showed higher bioavailability compared to the other metals (Pb, Cu and Zn) and consequently, it posed a greater ecological risk. The order of mobility of the four metals is $Cd > Zn > Cu > Pb$.

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التوزيع التجزيئي وقابلية الحركة للرصاص والكاديوم والنحاس والزنك في الرواسب السطحية لقاع بحيرة قارون - الفيوم، مصر

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الأنفوشي ٢١٥٥٦ - الإسكندرية - مصر

المستخلص. تم تعيين التركيزات الكلية والتوزيع التجزيئي لعناصر الرصاص، والكاديوم، والنحاس، والزنك في بعض الرواسب السطحية من قاع بحيرة داخلية، وكذلك التركيزات الكلية للحديد، والمنجنيز، والتي تعتبر من العناصر المتحركة في ترسيب الفلزات في البيئة المائية، وتم حساب معاملات زيادة التركيز والتلوث ودرجة التلوث المطورة، ومؤشرات التلوث الفلزي، والتراكم الأرضي للتمييز بين مصادر الدفق الطبيعية، ومصادر الملوثات وتقدير الحالة البيئية لرواسب البحيرة، وتراوحت قيم التركيزات الكلية لعناصر الرصاص والكاديوم والنحاس والزنك و المنجنيز والحديد بين ١٤-٤٠،٤ ميكروجرام/جرام، ٥-٠،٦ ميكروجرام/جرام، ٤-٢٣٢،٥ ميكروجرام/جرام، ٤-١٨،٤ ميكروجرام/جرام، ٩-١٣١،٣-٥٨٧،٣ ميكروجرام/جرام، ٤-٦،٢-٤٠،٢ ملليجرام/جرام على الترتيب، ويمكن ترتيب العناصر تبعاً لتركيزاتها في الرواسب ترتيباً تنازلياً كالاتي حديد < منجنيز < زنك < نحاس < رصاص < كاديوم ، وتبين أن تركيزات هذه العناصر تقع ضمن النسب الطبيعية المسجلة في رواسب المناطق غير الملوثة، ما عدا الكاديوم والذي زادت تركيزاته عن المعدلات الطبيعية بدرجة ملحوظة، وقد أظهرت القياسات أن النسب الأكبر من تركيزات عناصر الرصاص (٧٠٪) والنحاس

(٦٦٪) الزنك (٥٨٪) توجد فى الحالة المستقرة أو الخاملة فى الرواسب وليس لها تأثير ضار على البيئة. أما الكاديوم فقد تبين أن معظم وجوده فى الرواسب يقع فى الحالة النشطة غير المستقرة (٨٧٪) مما يحدونا إلى الإشارة إلى ضرورة اتخاذ الإجراءات اللازمة نحو حظر أو تقليل صرف المخلفات إلى البحيرة لمنع زيادة تدهور الوضع البيئي لها.