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# THE DISTRIBUTION OF SOME LEACHABLE AND TOTAL HEAVY METALS IN CORE SEDIMENTS OF MANZALA LAGOON, EGYPT

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## ABSTRACT

Five sediment cores from Manzala lagoon were collected in December 2005 in order to determine the total and leachable copper (Cu), zinc (Zn) and cadmium (Cd) beside grain size, total organic carbon (TOC) and total carbonate ($CaCO_3$). The results showed that, the sediment were composed of sand and mud enriched with carbonate (8.45 – 59.31%) and TOC (0.89 – 2.88%). The concentrations of leachable forms ranged from 5.86 to 36.19, 15.29 to 68.85 and 0.65 to 3.66 µg/g for Cu, Zn and Cd respectively, and varies between 12.03 to 95.24, 45.08 to 229.31 and 2.33 to 7.45 µg/g for total forms of the three metals respectively. The study revealed that most of Cd (64%) is dominant in the leachable form (labile) i.e available for any biotic process, while the reverse was observed for Zn where 66% of the total form is present in residual form (non-labile). In case of Cu, the leachable and residual forms are mostly the same. The metal pollution index (MPI) showed that the contamination is restricted to eastern part of the lagoon. The total fraction of Cu and Zn were lower than Effect Range - Low (ERL), while Cd showed a reveres trend.

## 1-INTRODUCTION

Manzala lagoon is located in the northeastern sector of the Nile delta between Damietta Nile branch and the Suez Canal between 31° 45′ and 32° 20′ E longitude and 31° 00′ to 31° 30′ N latitudes Fig (1). It is a shallow-brackish water coastal lagoon with an average depth 1 m, covers an area of about 265.512 acres. It is separated from the Mediterranean Sea by a narrow coastal sand bar, higher in the west than in the east. Three communications outlets with the sea are kept namely; El-Boughdady (1.8 m deep and 36 m wide), the new El-Gamil (6 m deep and 55 m wide) and old El- Gamil (6.5 m deep and 100 m wide). In addition, the lagoon is connected at its east side with the Suez Canal through El-Kabouty channel, and at its northwest side with Damietta Branch of the Nile River by El- Souffara and El-Ratama Canals. Most recently El-Salam Canal (Fig. 1) is constructed to serve agriculture development in Sinai.

Manzalla lagoon receives about 5388.4 × $10^6$ drainage waters annually via several drains. From east to west these are: Bahr El-Baqar, Ramsis, Hadous, El-Sirw, El-Matariya, Faraskur and Inaniya (Fig.1). They discharge agricultural drainage. The major part of drainage water (~ 85%) is discharged by Hadous, Ramsis and Bahr El-Baqar Drains (Khalil, 2003). The latter collects sewage and industrial wastes from different districts through its way from Cairo. Stanley (1988 & 1990) estimated an average long-term sedimentation rate for Manzala lagoon of ≈ 0.5 cm/yr.

Heavy metals are one of the serious pollutants due to their toxicity, persistence and non-degradability in the environment (Tam and Wong, 2000; Yuan *et al*., 2004).

Metals are derived from natural inputs and anthropogenic emissions, such as parent rock weathering, industrial wastewater, transportation, agriculture and climate (El Nemr *et al*., 2006; Luo *et al*., 2006). Metals in the sediments may be classified as lattice (residual fraction) which is the part tightly held to the matrix and is hardly released. The non-lattice held is the part of the metal that resides outside the crystal structure and is more or less mobile (Waldichuk, 1985). Pardo *et al*. (1990) pointed out that the metal present as residual fraction can be taken as guide to show the over- all pollution of the system.

Metals of anthropogenic origin are more loosely bound in sediments and thus are more readily available to organisms (Schropp and Windrom, 1988). Analysis of the leachable (labile) metal fraction of the sediment may be more useful, in terms of discovering its biological significance and the new inputs, than analysis of the total metal fraction (Puente *et al*., 1996; El Nemr, 2003).

The present research includes two metals (Cu & Zn) that are widely affected by anthropogenic inputs (Scoullos and Constandianos, 1988) beside Cd (toxic metal) which classified as carcinogenicity (probable human carcinogen) (USEPA, 1996). The concentration of leachable (labile) and total fraction of these metals were determined in the sediments collected from Manzalla lagoon to evaluate mobility, bioavailability of the metals.

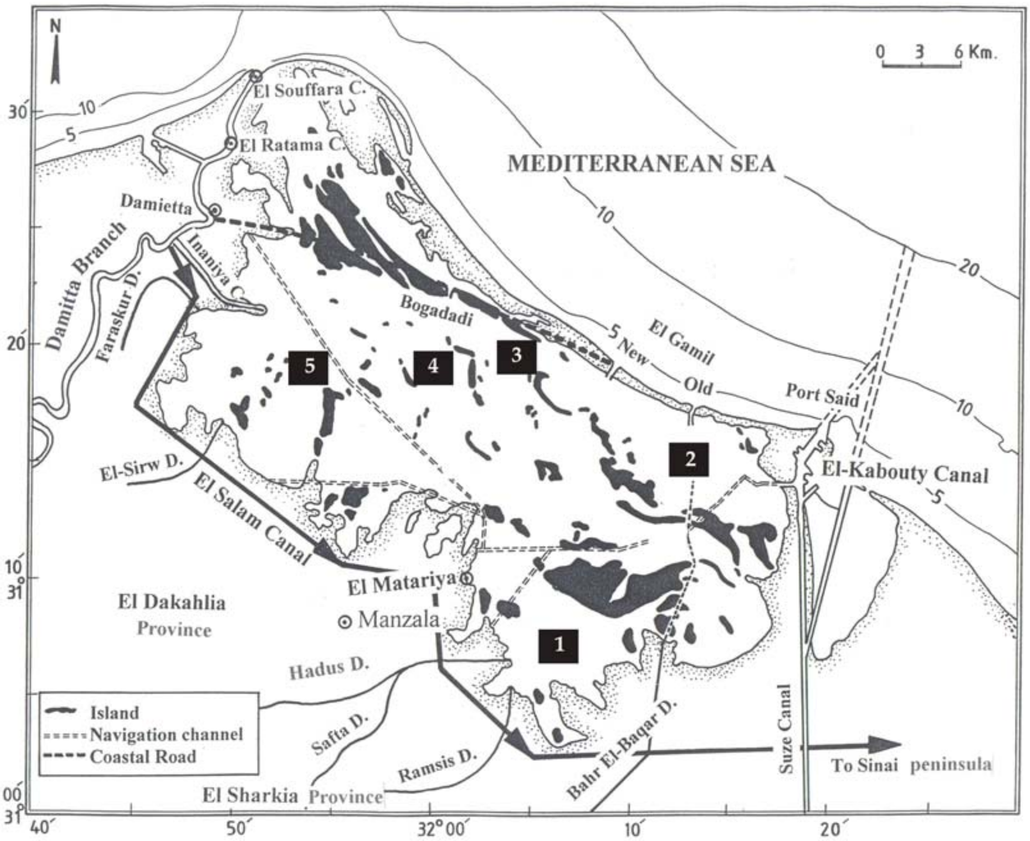

Fig. 1. Sampling locations in Manzala lagoon.

## 2. MATERIALS AND METHODS

Short core sediment 30 cm long was collected by pressing along PVC-tube (5 cm inner diameter) from five stations (Fig. 1) in a way to represent the different lagoon parts; cores 1& 2 from the eastern part and cores 3, 4 & 5 from the western part. The sediment of each core is split every 5 cm interval.

For total heavy metal concentrations, the powder sediment samples are digested in closed Teflon vessels with hydrofluoric acid (HF) in combination with aqua regia (3 HCl : 1 $HNO_3$ v/v). The leachable fraction was extracted from 1 g of non-powdered sediment by maintaining it in suspension during 12 hours in 25 ml of 0.5M HCl solution (Agemian & Chau, 1976). Concentration of metal in the extracts were measured using Flame-Atomic Absorption Spectrophotometer (AAS Perkin-Elmer Model 2380).

The organic carbon contents were determined by the dichromate wet oxidation method of Gaudette and Flight (1974). The carbonate contents were determined by treating the sediment with dilute HCl followed by back titration of the excess acid. Particle size was carried out according to the standard sedimentological method as described by Folk (1974).

### 2.1. Quality Assurance

The accuracy of the metal determination every extraction batch of 9 samples included a blank extraction and reference material (QMD 1 – 6), marine sediments which was digested by HF/HCl /$HNO_3$ solution.

Analytical results of the quality control samples indicated a satisfactory performance of heavy metals determinations and lied within the range of certified values with 95.7 and 101.6% recovery for all metals studied (Table 1).

To prevent contamination, all used plastic lab-ware were previously washed in 10% nitric acid and deionized water (Bugrer *et al.*, 2001) as well as all chemicals used in the experimental were Merck with high purity. The standard deviations for the leachable metals were ± 0.76, ± 1.57 and ± 0.26 for Cu, Zn and Cd respectively, and the coefficient of variation was 5.33, 3.51 and 9.23% respectively.

### 2.2. Data treatment:

- Excel program version 5.0 is used to calculate the concentrations of the leachable and total metals. Also used to compute the correlation matrixs.
- The vertical distribution (contour lines) is drawn using surfer 16 program.
- The accuracy of the analytical methods is determined.
- The results compared with the biological effects of metals, ERL (Effects-Range Low) and ERM (Effects-Range Median) in Table (2) reported by CEQG (2001); Long and Morgan (1990) and Long *et al.* (1995). The incidence of effects increased from 20% to 30% for most trace metals when concentrations exceeded ERL value, but were lower than the ERM values. When concentrations exceeded the ERM value the incidence of adverse effects increased from 60 to 90 % for most trace metals.
- Metal pollution Index (MPI) for the total and leachable metals of the surface sections at the sites investigated in this study were compared using MPI calculated according to Usero *et al.* ( 1996 and 1997) with the formula

$$MPI = (Cu \times Zn \times Cd)^{1/3}$$

**Table (1): Accuracy test results.**

| Element | Cu | Zn | Cd |
|---|---|---|---|
| Certified values | 9.79 | 51.70 | 215.00 |
| Found values | 9.88 | 52.52 | 205.75 |
| Standard deviation | ± 0.28 | ± 0.71 | ± 3.77 |
| Recovery% | 100.9 | 101.6 | 95.7 |

**Table (2): ERL and ERM values.**

| | Cu | Zn | Cd |
|---|---|---|---|
| ERL | 34 | 150 | 1.2 |
| ERM | 270 | 410 | 9.6 |

## 3. RESULTS AND DISCUSSION

### 3.1. Sediment characteristics

The distribution of sand and mud (%) in the vertical section extending along the short cores sediments of Manzalla lagoon are shown in Table (3). The sand and mud content varies between 7 & 72 %, and 28 & 95% respectively. Generally, the sand fraction is more abundant for most of the lagoon sediments, which could be attributed to the abundance of mollusk organisms in such brackish-water environment. Core 1 is characterized by mud sediments due to entrapment of considerable amounts of fine materials in agricultural soils from Bahr El-Baqar drain.

TOC content varies between 0.89 and 2.88 %. The high values were restricted in the upper intervals of core 2 (Table 3), may reflect the time when the modern Man-made extent ion of Bahr El-Baqar drain began to dump waste- materials northwards in the lagoon (Khalil, 2003). Also the high levels of TOC in the western side were mostly attributed to the high amount of the plant detritus contribution.

In general, most core sediment show high values of $CaCO_3$ (8.45 to 59.31%) and reflecting the predominance of brackish-water conditions.

### 3.2. Trace metals

Distribution of trace metals, copper (Cu), zinc (Zn) and cadmium (Cd) as total and leachable in the vertical sediment section are shown in Table (4).

### 3.2.1 Copper (Cu)

The concentration of total Cu ranged from 12.03 µg/g and 212.24 µg/g (Table 4). The levels of TCu were mostly lower than the Effect Range - Low (ERL) value for the cores samples with little exception (Tables 2 & 4). Leachable Cu concentration showed lowest value 5.86 µg/g in core 2 and highest value 36.19 µg/g in core 1. Leachable Cu is representing about 52% of the total form, which indicated that Cu was considered to be more bioavailable in these sediments.

The vertical distribution of total and leachable forms are mostly the same and similar to that of mud. Table (5 shows positive correlation between mud and both of total and leachable Cu (r = 0.64 & 0.93, respectively).

In general, the concentrations of the two forms increased from western to eastern side of the lagoon. High levels were recorded at the surface layer of core 2 and in most intervals of core 1 (in front of Bahr El-Baqar drain).This may reflect the deoxygenated conditions of the sediments at this part of the lagoon which increase the precipitation of Cu.

### 3.2.2. Zinc (Zn)

The concentration of total Zn showed lowest value 45.08 µg/g in core 2 and highest value 229.31 µg/g in core 1. The concentration of Zn as total fraction were lower than ERL except that of core 1 (5 – 30 cm) intervals the values were over the ERL.

Leachable Zn concentration varies between 15.29 µg/g and 68.85 µg/g. It represents 34 % of the total form. This indicates that most of Zn in the sediments is present in non-labile form i.e. unavailable for any biotic process. This phenomenon agrees with Yuan *et al.* (2004).

The vertical distributions of the two forms of Zn are mostly similar to each other and also to that of mud. As in cupper the high levels of zinc were observed in the eastern part of the lagoon mainly due to the discharged water in this side of the lagoon. Table (5) shows positive correlation between the two leachable forms of Cu and Zn (r = 0.90) and that of total forms (r = 0.72). Positive correlation between mud and both total and leachable Zn (r = 0.81 and 0.87, respectively) were showed as well.

### 3.2.3. Cadmium (Cd)

Cd was found in relatively high concentrations in the sediments of Manzalla lagoon. The total Cd varies between 2.33 µg/g (core 3) to 7.45 µg/g (core 1). Concentrations of Cd were higher than ERL value for all studied sediment samples. The high Cd values may reflect the source of the metal in the industrial wastes and /or the agricultural drainage water in association with the phosphate fertilizers, which are extensively used in the agriculture.

The leachable Cd ranges between 0.65 µg/g (core 1) and 3.66 µg/g (core 4). The large proportion of the total Cd (64%) suggests that Cd is mostly mobile element.

The vertical distribution of total and leachable Cd shows two points: First the leachable Cd shows higher values in the western side of the lagoon than the eastern. This is reverse to that found in total Cd. Khaled (1997) and Ahdy (1999) pointed out that in sediments of Abu-Kir Bay, the concentration of Cd increased far from the discharged water. Ahdy (1999) found that Cd is the only metal, which exhibits enrichment as carbonate. Second, leachable Cd is generally decreased with increasing depth, but the reverse was found for total Cd especially in core 1. This leads to the precipitation of Cd with the fine particles of subsurface layers as residual Cd. Bruland (1980) found that Cd is fixed by phytoplankton in the surface water and transport toward the floor with the remains of these organisms, when organic detritus decomposed Cd is liberated with other mineralization products.

Table (5) shows positive correlation between leachable Cd and $CaCO_3$ (r = 0.77). Also it gives negative correlations with all forms of Cu and Zn.

### 3.3. Metal pollution Index (MPI)

The total and leachable forms of the 10 cm, 15 cm and 20 cm surface sections of all core sediments were compared using MPI Table (6). The results in all cases pointed out that the eastern side of the lagoon specially in front of Bahr El-Baqar drain has the highest level of pollution index for total and leachable metals, while the western side has the lowest values.

**Table (3): Grain size compostion, total organic carbon (TOC) and carbonate content ($CaCO_3$).**

| Core No. | intervals (cm) | Mean (ph) | Sand % | Mud % | TOC % | $CaCO_3$ % |
|---|---|---|---|---|---|---|
| 1 | 0 - 5 | 4.2 | 49 | 51 | 1.24 | 51.37 |
| | 5-10 | 7.1 | 13 | 87 | 1.3 | 17.94 |
| | 10-15 | 7.7 | 7 | 93 | 1.76 | 10.03 |
| | 15 - 20 | 7.9 | 5 | 95 | 1.26 | 8.45 |
| | 20 - 25 | 7.9 | 7 | 93 | 1.3 | 12.29 |
| | 25 - 30 | 7.6 | 7 | 93 | 1.5 | 12.34 |
| 2 | 0 - 5 | 4.7 | 35 | 65 | 2.71 | 36.25 |
| | 5-10 | 4.7 | 28 | 72 | 2.68 | 35.95 |
| | 10-15 | 3.8 | 55 | 45 | 1.8 | 42.45 |
| | 15 - 20 | 2.2 | 72 | 28 | 1.07 | 56.98 |
| | 20 - 25 | 2.4 | 64 | 36 | 0.89 | 57.21 |
| | 25 - 30 | 2.4 | 68 | 32 | 0.95 | 59.31 |
| 3 | 0 - 5 | 2.3 | 64 | 36 | 1.4 | 57.78 |
| | 5-10 | 3 | 57 | 43 | 1.48 | 52.81 |
| | 10-15 | 2.3 | 69 | 31 | 1.11 | 58.12 |
| | 15 - 20 | 3.2 | 57 | 43 | 1.52 | 52.58 |
| | 20 - 25 | 3.5 | 51 | 49 | 1.62 | 47.38 |
| | 25 - 30 | 3.6 | 45 | 55 | 1.86 | 46 |
| 4 | 0 - 5 | 2.9 | 65 | 35 | 2.08 | 58.64 |
| | 5-10 | 4.7 | 36 | 64 | 2.52 | 43.77 |
| | 10-15 | 4.5 | 35 | 65 | 2.88 | 39.27 |
| | 15 - 20 | 3.7 | 47 | 53 | 2.37 | 46.7 |
| | 20 - 25 | 2.8 | 56 | 44 | 2.59 | 44.12 |
| | 25 - 30 | 3.9 | 41 | 59 | 2.16 | 32.72 |
| 5 | 0 - 5 | 4.3 | 37 | 63 | 1.96 | 46.73 |
| | 5-10 | 4.3 | 36 | 64 | 1.9 | 40.67 |
| | 10-15 | 4.1 | 39 | 61 | 2.27 | 44.51 |
| | 15 - 20 | 3.9 | 42 | 58 | 1.89 | 42.59 |
| | 20 - 25 | 4.5 | 34 | 66 | 2.11 | 34.79 |
| | 25 - 30 | 4.6 | 34 | 66 | 2.11 | 37.06 |

**Table (4): The concentration (ug/g) of total (T) and Leachable (L) copper (cu), zinc (Zn) and cadmium (Cd)in core sediments of Manzala lagoon (2005).**

| Core No. | Intervals (cm) | Cu | | Zn | | Cd | | L/T % | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | T | L | T | L | T | L | Cu | Zn | Cd |
| 1 | 0 - 5 | 29.46 | 17.01 | 105.24 | 51.90 | 2.91 | 3.09 | 58 | 49 | 94 |
| | 5-10 | 59.27 | 31.57 | 185.09 | 68.85 | 5.69 | 1.71 | 53 | 37 | 30 |
| | 10-15 | 70.95 | 34.59 | 208.68 | 59.34 | 5.37 | 0.65 | 49 | 28 | 12 |
| | 15 - 20 | 60.33 | 36.19 | 186.10 | 65.27 | 3.37 | 0.79 | 60 | 35 | 23 |
| | 20 - 25 | 62.03 | 35.92 | 178.20 | 65.40 | 7.45 | 1.01 | 58 | 37 | 14 |
| | 25 - 30 | 60.50 | 31.74 | 229.31 | 51.44 | 6.10 | 0.88 | 52 | 0 | 14 |
| | average | | | | | | | 55 | 31 | 31 |
| 2 | 0 - 5 | 212.24 | 13.88 | 98.59 | 52.90 | 3.02 | 2.89 | 15 | 54 | 96 |
| | 5-10 | 24.32 | 16.21 | 108.52 | 50.09 | 1.80 | 2.62 | 67 | 46 | 69 |
| | 10-15 | 18.88 | 12.79 | 73.64 | 41.64 | 2.45 | 2.81 | 68 | 57 | 87 |
| | 15 - 20 | 12.72 | 5.90 | 45.08 | 15.29 | 2.18 | 4.03 | 46 | 34 | 54 |
| | 20 - 25 | 14.11 | 6.52 | 45.55 | 18.50 | 3.43 | 3.80 | 46 | 41 | 64 |
| | 25 - 30 | 12.33 | 5.86 | 90.21 | 15.48 | 3.13 | 3.61 | 48 | 17 | 60 |
| | average | | | | | | | 48 | 42 | 72 |
| 3 | 0 - 5 | 12.03 | 9.34 | 68.03 | 33.91 | 2.81 | 3.58 | 78 | 50 | 78 |
| | 5-10 | 87.36 | 8.73 | 164.49 | 25.88 | 2.06 | 3.16 | 10 | 16 | 65 |
| | 10-15 | 12.23 | 11.01 | 79.82 | 34.62 | 4.04 | 3.65 | 90 | 43 | 90 |
| | 15 - 20 | 25.07 | 8.34 | 85.54 | 25.27 | 1.84 | 4.03 | 33 | 30 | 46 |
| | 20 - 25 | 23.36 | 9.93 | 89.92 | 33.87 | 1.66 | 2.33 | 43 | 38 | 71 |
| | 25 - 30 | 23.47 | 6.85 | 93.21 | 26.55 | 2.62 | 2.63 | 29 | 28 | 86 |
| | average | | | | | | | 47 | 34 | 73 |
| 4 | 0 - 5 | 20.31 | 7.16 | 84.25 | 21.70 | 3.26 | 3.54 | 35 | 26 | 92 |
| | 5-10 | 28.04 | 15.04 | 99.63 | 33.15 | 3.83 | 3.66 | 54 | 33 | 96 |
| | 10-15 | 33.20 | 17.76 | 107.58 | 40.36 | 2.32 | 2.63 | 53 | 38 | 88 |
| | 15 - 20 | 27.09 | 16.59 | 91.63 | 41.52 | 3.68 | 2.68 | 61 | 45 | 73 |
| | 20 - 25 | 30.29 | 16.24 | 100.44 | 36.52 | 2.69 | 3.14 | 54 | 36 | 86 |
| | 25 - 30 | 43.03 | 16.72 | 158.66 | 40.92 | 4.60 | 2.34 | 39 | 26 | 51 |
| | average | | | | | | | 49 | 34 | 81 |
| 5 | 0 - 5 | 27.89 | 19.83 | 90.08 | 37.75 | 3.15 | 3.89 | 71 | 42 | 81 |
| | 5-10 | 30.66 | 18.92 | 115.06 | 35.74 | 1.69 | 2.90 | 62 | 31 | 58 |
| | 10-15 | 37.21 | 22.42 | 176.50 | 42.67 | 3.24 | 2.56 | 60 | 24 | 79 |
| | 15 - 20 | 28.62 | 16.63 | 100.88 | 32.02 | 4.68 | 2.81 | 58 | 32 | 60 |
| | 20 - 25 | 33.34 | 20.28 | 119.48 | 38.72 | 3.38 | 2.22 | 61 | 32 | 66 |
| | 25 - 30 | 41.51 | 21.17 | 153.34 | 41.45 | 0.91 | 2.79 | 51 | 27 | 33 |
| | average | | | | | | | 61 | 31 | 63 |
| | Total average | | | | | | | 52 | 34 | 64 |

**Table (5): Correlation matrix of the studied parameters.**

| | *Mean (ph)* | *Sand %* | *Mud* | *TOC %* | *CaCO3* | *Tcu* | *Lcu* | *TZn* | *LZn* | *TCd* | *LCd* |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Mean (ph) | 1.00 | | | | | | | | | | |
| Sand | -0.97 | 1.00 | | | | | | | | | |
| Mud | 0.97 | -1.00 | 1.00 | | | | | | | | |
| TOC | 0.03 | -0.19 | 0.19 | 1.00 | | | | | | | |
| CaCO3 | -0.96 | 0.96 | -0.96 | -0.09 | 1.00 | | | | | | |
| Tcu | 0.64 | -0.64 | 0.64 | 0.14 | -0.65 | 1.00 | | | | | |
| Lcu | 0.94 | -0.93 | 0.93 | 0.04 | -0.93 | 0.57 | 1.00 | | | | |
| TZn | 0.82 | -0.81 | 0.81 | 0.02 | -0.83 | 0.72 | 0.83 | 1.00 | | | |
| LZn | 0.89 | -0.87 | 0.87 | 0.11 | -0.88 | 0.61 | 0.90 | 0.74 | 1.00 | | |
| TCd | 0.56 | -0.47 | 0.47 | -0.36 | -0.54 | 0.32 | 0.57 | 0.51 | 0.45 | 1.00 | |
| LCd | -0.66 | 0.63 | -0.63 | 0.22 | 0.70 | -0.44 | -0.60 | -0.66 | -0.49 | -0.31 | 1.00 |

**Table (6):**

| core | upper 10 cm | | upper 15 cm | | upper 20 cm | |
|---|---|---|---|---|---|---|
| | T | L | T | L | T | L |
| 1 | 61 | 30 | 104 | 43 | 138 | 56 |
| 2 | 52 | 24 | 69 | 36 | 85 | 41 |
| 3 | 54 | 17 | 72 | 29 | 93 | 36 |
| 4 | 40 | 20 | 62 | 33 | 83 | 45 |
| 5 | 43 | 24 | 72 | 38 | 96 | 49 |

## 4. CONCLUSION

This investigation has clearly shown that the heavy metal contents in the total fraction of Cu and Zn were lower than ERL, while for Cd it was higher than ERL but lower than ERM values for all studied samples. The MPI show that the eastern side of the lagoon specially in front of Bahr El-Baqar drain has the highest level of pollution for total and leachable metals. Analysis of the acid leachable fraction showed that (66%) of Zn are present in non-labile form i.e. unavailable for any biotic process. Cd and Cu were the most mobile element since its leachable fraction represented by 64% and %52% of the total concentration.

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