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Heavy Metals Contamination in Sediments of the Western Part of Egyptian Mediterranean Sea

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Abstract: Concentrations of eight heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) in surface sediment from El-Sallum to Sidi-Kreer along western part of Egyptian Mediterranean Coast were determined to evaluate their levels and spatial distribution. The ranges of the measured concentrations in the sediments are as follows: 0.524-0.924 µg/g for Cd, 16.248-34.164 µg/g for Cr, 26.529-33.332 µg/g for Cu, 846.426-1433.933 µg/g for Fe, 32.371-108.915 µg/g for Mn, 31.703-43.592 µg/g for Ni, 20.672-35.624 µg/g for Pb, and 26.267-112.73 µg/g dry weights for Zn. There are no significant correlations among most of these metals, indicating they have different anthropogenic and natural sources. To assess metal concentrations in sediment, Numerical Sediment Quality Guidelines (SQGs) were applied. The concentrations of Cd, Cr, Pb, and Zn in all sediment samples are lower than the proposed TECs indicated that there are no harmful effects from these metals. On the other hand, concentration of Ni exceeded TEC in all samples while Cu was exceeded the TEC at El-Sallum and Sidi-Barrani indicated that these stations were in potential risk. The metal contamination in the sediments was evaluated by applying Index of geo-accumulation and contamination factor.

Key words: heavy metal, surface sediment, contamination, geoaccumulation index, contamination factor, Mediterranean Sea, Egypt.

INTRODUCTION

Metals are natural constituents in nature. In fact, during the last few decades, industrial and urban activities have contributed to the increase of metals contamination into marine environment and have directly influenced the coastal ecosystems (Buccolieri *et al.,* 2006). Although most adsorbed pollutants on the sediments are not readily available for aquatic organisms, the variation of some physical and chemical characteristics (pH, salinity, redox potential and the content of organic chelators) of the overlying water may provoke the release of the metals back to the aqueous phase, hence under changing environmental conditions sediments may become themselves important pollution sources (Soares *et al.,* 1999). All metals are toxic above some threshold bioavailable level. Ag, Hg, Cu, Cd, and Pb are particularly toxic (Kontas, 2008). Heavy metals are regarded as serious pollutants of aquatic ecosystems because of their environmental persistence, toxicity and ability to be incorporated into food chains (Forstner and wittman, 1983. Dmirbas *et al.,* 2005). Exposure to heavy metals has linked to several human diseases such as development retardation or malformation, kidney damage, cancer, abortion, effect on intelligence and behavior, and even death in some cases of exposure to very high concentrations (Banerjee, 2003; Jiries, 2003). Sediments are preferable monitoring tools since contaminant concentrations are orders of magnitude higher and they show less variation in time and space, allowing more consistent assessment of spatial and temporal contamination (Beiras *et al.,* 2003; Caccia *et al.,* 2003).

The aim of this study is to assess the level of metal concentrations in surface sediments in the western part of Egyptian Mediterranean Sea examining the occurrence and distribution of metals and to explore the natural and anthropogenic input of heavy metals and to assess the pollution status on the area and to highlight relationships among metals.

MATERIALS AND METHODS

Samples were taken in January, 2006 from eight sites along western part of the Egyptian Mediterranean coast from El-Sallum to Sidi-Kreer as showing in Figure 1. Latitude and Longitude as well as depth (m) for each site sample were illustrated in Table 1.

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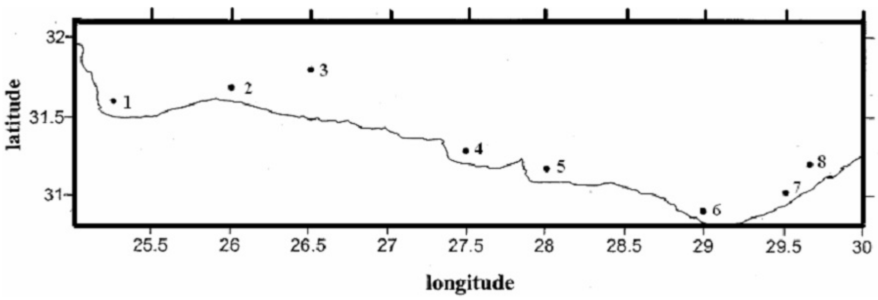

Fig. 1: Map of location samples

Table 1: Location and depth of the sediment samples:

Location	Station no.	Latitude	Longitude	Depth (m)
El-Sallum	1	31° 36.0'	25° 25'	85
Sidi-Barrani	2	31° 40.2'	26° 00'	60
El-Shalliah	3	31° 34.2'	26° 30'	75
Alam El-Roum	4	31° 16.2'	27° 30'	Nd
Fouka	5	31° 09.6'	28° 00'	63
El-Alamein	6	31° 11.2'	29° 00'	50
El-Hammam	7	31° 01.2'	29° 30'	62
Sidi-Kreer	8	31° 10.1'	29° 50'	50

The area of study extends between Alexandria at the east and El-Sallum at the west. The area lies between Longitudes 29°50' E (Sidi-Kreer) and 25° 30' (El-Sallum) including Arab's Gulf. The study area is almost covered with pure carbonate sediments. The bottom sediments are chemically homogeneous and the organic matter content is generally low and is of marine origin (Nasr, 1978).

The surface sediment samples were collected from 5 cm layer bottom sediments using a Van Veen type grap sampler. Eight composite sediment samples (3 graps per sample) were collected from each site. After collection, sediment samples were dried in a vacuum oven (Medline, OV-12) at 70ºC for 3-4 days until constant weight, lightly ground in an agate mortar for homogenization and prepared for analysis.

Approximately 0.2 of dried sample (triplicate samples for each sediment location) was extracted overnight with 2 ml concentrated nitric acid (HNO_3) and evaporated until near dryness and no nitrous vapors were released. After cooling, 5 ml of an acid mixture made of concentrated acids $HF:HClO_4:HNO_3$ 6:2:3 ratio was added. The digestion continued until no coloured vapours were released. The working standard solutions were freshly prepared by diluting an appropriate aliquot of the stock solutions (IAEA-UNEP, 1990).

Flame atomic absorption spectrophotometer (Varian 10 Plus) was used for the determination of the eight heavy metals under investigation (Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn).

For quality control, standard reference materials (SD-M-2/TM, marine sediments, National Research Council, Canada) were prepared, with each batch of samples analyzed. Six replicates of the SRM were digested with the same procedure as the samples. Recovery ratios of heavy metals in the SRM were fluctuated from 97.5% to 104.2% as illustrated in Table 2.

Table 2: Heavy metal concentrations (mg g-1 dry weight) in reference materials analyzed together with sediment samples.

Metal	Certified values	Found values	Standard deviation	Recovery (%)
Cd	0.113	0.101	0.026	104.2373
Co	13.6	12.269	0.569	97.50546
Cr	77.2	79.016	1.371	102.2983
Cu	32.7	33.011	0.827	100.9421
Ni	56.1	54.875	1.392	97.76765
Pb	22.8	24.263	0.521	101.9903
Zn	74.8	76.206	1.492	101.845

RESULTS AND DISCUSSION

The total metal concentrations and average values for each sampling site found in sediments in this study are shown in Table 3. Metal contents were ranging over following intervals: Cd: 0.524-0.924 µg/g; Cr: 16.248-34.164 µg/g; Cu: 26.529-33.332 µg/g; Fe: 846.426- 1433.933 µg/g; Mn: 32.371-108.915 µg/g; Ni: 31.703-43.592 µg/g; Pb: 20.672-35.624 µg/g; Zn: 26.267-112.73 µg/g dry weights. Mean contents of the ecosystem studied were: Cd: 0.721 µg/g; Cr: 24.11 µg/g; Cu: 3.304 µg/g; Fe: 1122.950 µg/g; Mn: 57.953 µg/g; Ni: 38.762 µg/g; Pb: 27.845 µg/g; Zn: 59.717 µg/g dry weights, allowing to arrange the metals from higher to lower mean content in this area as: Fe > Zn > Mn > Ni > Cu > Pb > Cr > Cd.

Table 3: Average concentration of heavy metals (Ug/g dry weight) in sediment samples collected from El-Sallum to Sidi-Kreer along Egyptian Mediterranean coast.

Station number	Average concentration of heavy metals (µg/g dry weight)							
	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
1	0.924	28.634	33.332	1433.933	108.915	31.703	30.057	55.639
2	0.864	20.167	32.512	709.088	32.271	36.657	27.634	46.101
3	0.625	16.852	26.529	1229.085	58.088	35.666	20.672	47.691
4	0.729	27.634	31.210	1200.722	54.861	39.629	29.349	63.588
5	0.524	16.248	28.510	1427.630	74.223	41.610	24.634	81.074
6	0.634	18.634	29.444	999.026	48.407	43.592	26.423	45.306
7	0.807	34.164	30.916	1137.692	37.918	41.610	35.624	112.073
8	0.663	30.471	29.980	846.426	48.944	39.629	28.364	26.267
Mean	0.721	24.101	30.304	1122.950	57.953	38.762	27.845	59.717
Maximum	0.924	34.164	33.332	1433.933	108.915	43.592	35.624	112.073
Minimum	0.524	16.248	26.529	709.088	32.271	31.703	20.672	26.267
TEC	0.99	43.4	31.6	--	--	22.7	35.8	121
PEC	4.98	111	149	--	--	48.6	128	459

Note: TEC: Threshold Effect Concentration
PEC: Probable Effect Concentration

Pearson's correlation coefficient matrix among the selected heavy metals is presented in Table 4. Significant correlations between the contaminants of Cd and Cu (r=0.8683), Cr and Pb (r=0.8705), Fe and Mn (r=0.7873), could indicate the same or similar source input. In most cases, however, there are no significant correlations among most of these heavy metals, suggesting that these metals are not associated with each other and their identical behavior transport in estuarine environment. Furthermore, these metals might have different anthropogenic and natural sources in sediments of the area of study.

Table 4: Correlation matrix between heavy metals in sediment samples from El-Sallum to Sidi-Kreer

	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Cd	1							
Cr	0.5505	1						
Cu	0.8683*	0.5522	1					
Fe	-0.1534	-0.0626	-0.1621	1				
Mn	0.1333	-0.0123	0.1723	0.7873*	1			
Ni	-0.6047	-0.0683	-0.3598	-0.2155	-0.5690	1		
Pb	0.6123	0.8705*	0.6956	-0.0889	-0.1178	0.1270	1	
Zn	0.0755	0.2822	0.0580	0.4529	-0.0300	0.2925	0.5332	1

Note: * is significant at $p<0.05$

Compared with the literature values reported for the heavy metal content of the sediments, the present study recorded concentration value lowers than that reported by Guven and Akinci of Izmir Inner Bay but within the range observed by Yu *et al.*, for sediment samples from Quanzhou Bay, China and that reported by Filgueiras *et al.,* for sediment of Luoro River (Spain) (Guven and Akinci, 2008; Yu *et al.,* 2008; Filgueiras *et al.,* 2004).

The accumulation of heavy metals in sediments can be a secondary source of water pollution, once environmental condition is changed (Chen *et al.,* 1996; Cheung *et al.,* 2003). Therefore, an assessment of heavy metal contamination in sediments is an indispensable tool to assess the risk of an aquatic environment. To assess metal concentrations in sediment, Numerical Sediment Quality Guidelines (SQGs) were applied. SQGs include a threshold effect concentration (TEC) and a probable effect concentration (PEC) (Table 3). If the metals in sediments are below the TEC, harmful effects are unlikely to be observed. If the metals are above the PEC, harmful effects are likely to be observed MacDonald *et al.*, noted in his studies that most of the TECs provide an accurate basis for predicting the absence of sediment toxicity, and most of the PECs provide an accurate basis for predicting sediment toxicity (MacDonald *et al.,* 2000). The concentrations of Cd, Cr, Pb, and Zn in all sediment samples are lower than the proposed TECs indicated that there are no harmful effects from these metals. On the other hand, the concentrations of Ni exceeded TEC in all samples while Cu for stations 1 and 2 exceeded the TEC indicated that these stations were in potential risk.

Assessment of Metal Contamination:

***Assessment According to United States Environmental Protection Agency (USEPA)*:**

The chemical contamination in the sediments was evaluated by comparison with the sediment quality guideline proposed by USEPA. These criteria are shown in Table 5. Mn and Pb in all stations under investigation were belong to unpolluted sediments, while stations 2, 3, 5, 6 are considered as moderately polluted by Cr while the other stations belong to heavily polluted. On the other hand, Cd, Cu, and Ni in all study sediments belong to moderately polluted sediments.

Table 5: EPA guidelines for sediments

Metal	Not polluted	Moderately polluted	Heavily polluted	Present study
Cd	-	-	>6	0.52-0.92
Cr	<25	25-75	>75	16.25-34.165
Cu	<25	25-50	>50	26.53-33.33
Mn	<300	300-500	>500	32.373-108.92
Ni	<20	20-50	>50	31.70-43.59
Pb	<40	40-60	>60	20.67-35.62

Assessment According to Geoaccumulation Index:

To understand the current environmental status and the metal contamination with respect to natural environmental, other approaches should be applied.

A common criterion to evaluate the heavy metal pollution in sediments is the geoaccumulation index (*I*geo), which was originally defined by Muller (1979) to determine metals contamination in sediments, by comparing current concentrations with pre-industrial levels and can be calculated by the following equation (Muller, 1979):

$$Igeo = \log_2[C_n/(1.5B_n)$$

Where, C_n is the measured concentration of the examined metal (n) in the sediment, B_n is the geochemical background concentration of the metal (n), and factor 1.5 is the background matrix correction factor due to lithogenic effects. Muller (1981) has distinguished seven classes of geoaccumulation index (Table 6) (Muller, 1981). In this work, B_n values have been taken equal to metal concentrations of the Mediterranean background sediment according to Adamo *et al.*, (2005). According to the Muller scale, the calculated results of *I*geo values (Table 7) indicate that Cd can be considered as from moderate to strong pollutant at most of the study stations (2<*I*geo<3) with the exception of station 5 which shows moderate pollution (*I*geo=1.8) [20]. Iron and Manganese show unpolluted situation for all stations (*I*geo<0). Unpolluted situation was recorded for Cr, Pb, and Zn for all stations. Nickel can be considered as moderate pollutant at all stations (0<*I*geo<1), but Cu fluctuated between unpolluted to the moderately polluted as shown in Table 7. On the basis of the mean values of *I*geo, sediments are enriched for metals in the following order: Cd > Ni > Cu > Pb > Cr > Zn > Mn > Fe

Table 6: Muller's classification for the geoaccumulation index (Muller, 1981)

I_{geo} value	Class	Quality of sediment
≤ 0	0	Unpolluted
0-1	1	From unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	From moderately to strongly polluted
3-4	4	Strongly polluted
4-5	5	From strongly to extremely polluted
≥ 6	6	Extremely polluted

Table 7: Geoaccumulation index (I_{geo}) values of heavy metals in sediments from Egyptian Mediterranean Coast

Station	I_{geo}							
	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
1	1.06	-0.97	-0.25	-3.99	-3.63	0.25	-0.74	-0.82
2	1.53	-0.71	0.05	-4.78	-4.48	0.29	-0.32	-0.87
3	1.62	-0.20	0.08	-3.77	-2.72	0.08	-0.20	-0.59
4	1.28	-0.26	-0.01	-4.02	-3.71	0.40	-0.23	-0.40
5	0.80	-1.02	-0.14	-3.77	-3.28	0.47	-0.49	-0.05
6	1.08	-0.82	-0.10	-4.29	-3.89	0.54	-0.38	-0.89
7	1.64	-0.34	0.00	-3.83	-3.44	0.44	-0.23	-0.68
8	1.14	-0.12	-0.07	-4.53	-3.88	0.40	-0.28	-1.68

Assessment According to Contamination Factor and Degree of Contamination:

The contamination factor C_f and the degree of contamination are used to determined the contamination status of sediment in the present study. C_f values are suggesting for describe the contamination factor (Hakanson, 1980). $C_f < 1$: low contamination factor; $1 \leq C_f < 3$: moderate contamination factor; $3 \leq C_f < 6$: considerable contamination factor; $C_f \geq 6$: very high contamination factor. The degree of contamination (C_d) was defined as the sum of all contamination factors and these values were given in Table 6. The following terminology is adopted to describe the degree of contamination (C_d values) for the selected eight metals. $C_d < 6$: low degree of contamination; $6 \leq C_d < 12$: moderate degree of contamination; $12 \leq C_d < 24$: considerable degree of contamination; $C_d \geq 24$: very high degree of contamination indicating serious anthropogenic pollution.

Maximum value of contamination factor for cadmium was noticed for sediment of at station 1 while the minimum C_f was recorded at station 5 (Table 8). Station 1 had considerable contamination factor values for cadmium according to the Hakanson's classification, while the rest of the investigated stations recorded a moderate contamination for this metal. All stations in the present study recorded low contamination factor for Cr, Cu, Mn, Ni, Zn except station 7 which exhibited moderate contamination for Zn only. Moderate contamination for Pb was recorded for all stations under investigation. Station 7 recorded the maximum value of degree of contamination while station 3 recorded the lowest degree of contamination as illustrated in Table 8. Stations 1, 2, 4, and 7 recorded moderate degree of contamination while the rest of stations revealed low degree of contamination.

Table 8: Contamination factor (C_f), Degree of contamination (C_d), and metal pollution index (MPI) of sediment collected along the western part Egyptian Mediterranean Coast.

St. No.	C_f							C_d	MPI
	Cd	Cr	Cu	Mn	Ni	Pb	Zn		
1	3.080	0.318	0.741	0.227	0.466	1.503	0.586	6.921	2.200
2	2.880	0.224	0.722	0.067	0.539	1.382	0.485	6.300	2.058
3	2.083	0.187	0.590	0.121	0.525	1.034	0.502	5.041	1.513
4	2.430	0.307	0.694	0.115	0.583	1.467	0.669	6.265	1.805
5	1.747	0.181	0.634	0.155	0.612	1.232	0.853	5.413	1.340
6	2.113	0.207	0.654	0.101	0.641	1.321	0.477	5.515	1.585
7	2.690	0.380	0.687	0.079	0.612	1.781	1.180	7.409	2.018
8	2.211	0.339	0.666	0.102	0.583	1.418	0.276	5.596	1.666

Metal Pollution Index:

An evaluation of the overall metal contamination was carried out by calculating a metal pollution index (MPI) which defined as a linear sum of the contamination factors weighted to take into account the differences in toxicity of the various metals.

$$MPI = \sum_i (W_i / W_t) \times CF_i$$

Where CF_i is the contamination factor for metal I, W_i is the weight for metal I, and $W_t = \sum_i W_i$

The weights of metals Zn, Cu, Cr, Pb, Ni, and Cd were established by Gonçalves *et al.*, (1992). Metal Pollution Index (MPI) for the investigated stations was illustrated in Table 8.

Stations 3, 4, 5, 6, and 8 must be classified as low contamination area where MPI < 2, while stations 1, 2 and 7 have MPI values >2 confirming that it is a considerable contamination for the previous six metal according to the classification of Gonçalves *et al.*, (1992).

***Conclusions*:** The results of this study supply valuable information about metal contents in sediment from different sampling stations from El-Sallum to Sidi-Kreer along the western part of Egyptian Mediterranean Sea. Moreover, these results can also be used to test the chemical quality of the marine sediment, in order to evaluate the possible risk to the marine community of this area. Geoaccumulation Index, contamination factor and degree of contamination, Metal Pollution Index were successfully applied for the assessment of contamination by heavy metals from western part of Egyptian Mediterranean Sea. The results obtained from Igeo method emphasized, to a large extent, the results obtained from the contamination factor. It could be concluded that both Igeo and C_f are powerful tools for the assessment of contamination of heavy metals. The two methods concluded that the Cd concentration fluctuated from moderate to strong contamination in all stations. According to Igeo classification, all stations under investigation are unpolluted by Cr, Pb, Zn and moderately polluted by Ni and fluctuated from unpolluted to moderately polluted by Cu. On the other hand, sediment samples were classified to be low contaminated by Cr, Cu, Mn, Ni, and Zn and moderately polluted by Pb according to the contamination factor calculation. As showing, the two methods of calculations (C_f and Igeo) are useful and successful but Igeo is more specific to determine the degree of pollution than the contamination factor, where Igeo is classified to 6 classes while the later containing four classes only. For the overall assessment of heavy metals in the present study MPI and degree of contamination are applied. According to the degree of contamination; sites 1, 2, 4, and 8 are moderately contaminated while stations 3, 5, 6, and 7 are classified as low contaminated by the eight metals under investigation.

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