Egypt. J. Aquat. Biol. & Fish., Vol.7, No.4 : 47 - 66 (2003) ISSN 1110 - 6131

HEALTH RISK ASSESSMENT IN RELATION TO HEAVY METALS POLLUTION OF WESTERN MEDITERRANEAN SEA, EGYPT

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Keywords: heavy metals, risk assessment, Mediterranean Sea sediment pollution

ABSTRACT

Concentrations of heavy metals (Mn, Fe, Zn, Cu, Pb, Co, Cr, Ni and Cd) were measured in the sediment collected from 7 stations along west Mediterranean Sea (Sidi- Krir ; open sea , east & west Sidi Krir, Sidi Barrani and El-sallum 1, 2, 60m). Also, health risk has been performed in relation to recreational activities at these places, using an exposure assessment model. The results of this study indicated a general moderate or absence of serious pollution in these places due to heavy metals; whereas the concentration of elements found could be mainly due to geological and atmospheric sources. However, the health risk evaluation is useful as a screening methodology for assessing the urgency of sediment remediation actions.

INTRODUCTION

In the last decade, studies on the occurrence, distribution and accumulation of toxic pollutants have significantly increased, mainly due to the global effects of environmental pollution on aquatic ecosystem. Heavy metals have been shown to be an important group of water contaminants, because of their high toxicity and persistence in all aquatic systems (Oscaramin, 1996). Metal contaminants can enter the environment in excess of natural amounts from industrial and mining effluents, from the combustion of fossil fuels, discharge of sewage and sewage sludge from fertilizer and pesticide residues förstner and Wittmann, 1979). The potential impacts of heavy metals are generally restricted to locations adjacent to major cities or industrialized areas on the coastal fringe (Batley, 1995) and to site draining areas of intensive agriculture. Once introduced into marine environment, heavy metals have the potential to affect sediment nutrient cycling, cell growth and regeneration as well as reproductive cycles and photosynthetic potential of marine organisms (Peters *et al.*, 1997). Heavy metals such as Pb, Cd and Zn are good markers of contamination from human activity (Ravanelli *et al.*, 1997). Oral ingestion of contaminated soils and dust by children is the principle cause of lead absorption (Xintaras, 1992). Moreover, Cd is contained in some phosphate-based fertilizers; such sources could constitute a major source of Cd absorption by humans. In addition, sewage sludge from wastewater treatment can contain significant quantities of Cd. It is also a side product of zinc smelting (Tran, 2002). The leachable metal fraction is defined as the anthropogenic fraction of metals involved with the sediment particle. Diluted acids have often extracted the leachable fraction (Malo, 1977; Salomons and Förstner, 1980; Hamilton *et al.*, 1984; Queirazza and Guzzi, 1987; Angelidis and Aloupi, 1995). The objective of this study is to determine concentration and distribution of some metals in a series of sediments collected from different sections along Mediterranean Sea, to monitor environmental changes and their health risk.

MATERIAL AND METHODS

Sediment samples were collected during spring 2003, from seven stations along the west cost of Mediterranean Sea (Sidi-Krir open sea, west Sidi Krir, east Sidi Krir, Sidi Barrany and Sallum at 1, 2 and 60 m depth) (Fig. 1). The homogenized samples were stored in clean polyethylene jars. In the laboratory, the sediment samples were separated into sub-samples for the respective treatment procedures.

Determination of total heavy metals

The sediment samples were dried at 105 °C, sieved and subsequently ground in a mortar. From each dried sample, 0.2 g of sediment was completely digested, using HNO_3, HF and $HClO_4$ (Trefry and Mettz, 1984). The final solution was diluted to 25 ml with distilled deionized water. Metal concentrations were determined, using Perkin-Elmer model atomic absorption spectrophotometer (AAS). Reagent and blanks were analyzed to check for metal contamination during processing.

Determination of leachable heavy metal

An aliquot of 0.5 g of dry sediment was treated with an excess of 160 ml of 0.3N HCl and stirred for two hours at room temperature, the suspension was filtered on Nuclepore 0.4 µm polycarbonate filters, previously treated with 6M HCl and ultra pure water, then with 1M CH_3CO_2NH and later with 100 ml ultra pure water. The analytical procedure has been validated on a certified sample of IAEA SD-M-2/TM marine sediment which was totally digested by $HNO_3/HF/HClO_4$ solution. Results obtained for the IAEA-SD-M-2/TM marine sediment are given in Table (1). All metal concentrations were within acceptable ranges.

Risk assessment

For quantification of exposure in relation to sediment contamination by heavy metals, a multiple pathway exposure model (SEDISOIL) was used (Bocking *et al.*, 1996). The model was developed by the National Institute of Public Health and Environmental Protection and includes six exposure routes: the ingestion of sediment, surface water included suspended matter, dermal contact via surface water and sediment, and fish consumption. The following equations were incorporated into the model to calculate total exposure.

Ingestion of contaminated sediment (mg kg^{-1} day^{-1})

$= (Cs \times IRs \times EF \times AF) \div BW$

where Cs = Concentration of the contaminant in sediment (mg kg^{-1} day^{-1}); IRs = Ingestion rate of sediment (kg dw/ exposure day), EF = exposure frequency (days/365days), AF = Absorption factor (unitless), and BW = Body weight (kg)

Dermal contact with contaminated sediment (mg kg^{-1} day^{-1})

$= (Cs \times SAs \times AD \times Ass \times Mf \times EDs \times EF \times AF) \div BW$

where, SAs = dermal surface area for sediment exposure (m^2), AD = dermal adherence rate for sediment (mg cm^{-1}), Ass = dermal absorption rate for sediment (liter/hour), Mf = matrix factor (unitless), and EDs = exposure duration from dermal exposure to sediment (hour/day)

Daily exposure is calculated according to the following equation

$[(6 \times \text{daily exposure}_{child}) \div 70] + [64 \times \text{daily exposure}_{adult}) \div 7$

If the hazard index is below 1, no health risk may occure

RESULTS AND DISCUSSION

The analysis of environmental matrices such as water or sediments provides a picture of the total contaminants load rather than of that fraction of direct ecotoxicological relevance. (Campanella *et* *al.*, 2001). Table (2) shows total and leachable contents of Mn, Pb, Zn, Fe, Cd, Cu, Co, Cr, Ni in sediment samples collected from seven stations along Mediterranean Sea weastern coast. The sediment values for total metals were 26.85 for Mn, 60.05 for Pb, 76.6 for Zn, 222.5 for Fe, 5.14 for Cd, 12.3 for Cu, 27.73 for Co, 20.06 for Cr and 38.98 mg/kg for Ni. The corresponding values of leachable metals were 24.73 for Mn, 51.66 for Pb, 17.2 for Zn, 190.06 for Fe, 4.43 for Cd, 8.25 for Cu, 17.27 for Co, 17.49 for Cr and 29.11 mg/kg for Ni. Mann-Whitney U-test analysis showed a significant difference between total and leachable metals (Zn, Co, Ni) P-values were 0.004, 0.004, 0.003, respectively. The total concentration of Zn in the sediment was 76.6 ± 23.59 mg/kg. The contamination of coastal regions including estuaries and marginal seas are attributed to a number of causes (Förstner *et al.*, 1978). The most important are direct input of effluents from industries and communities dumping of wastes from ships and through atmospheric fallout. The concentration of leachable Zn (non-residual Zn) shows the same pattern as the total Zn in sediments, indicating that most of Zn is in a leachable form [$Zn(OH)_2$]. This indicates that Zn is derived from anthropogenic source (El-Sayed, 1988). Meanwhile, the total Cu was 12.3 mg/kg which is lower than at any places along Alexandria coastal region. The distribution of leachable Cu shows the same pattern of total sediment, which seems to be affected by the anthropogenic activities. Therefore, it is the non–residual metal concentration, which truly reflects the extent to which the sediments have subjected to heavy metal pollution (Chester, 1981; Chester 1985). On the same manner, the lowest concentration of total Pb in this area indicates that this area receives little or no flow or sediment. This indicates that this area is protected from industrial inputs.

The Concentration of total and leachable Pb decreases in the sea ward direction, which may be due to the remoteness from the source of pollution as well as the desorption of Pb from sediment facilitated by increasing salinity (De-Groot *et al.*, 1976). On the other hand, a higher concentration of Cd was found at this region than that found by Khaled (1997) (Table 3). The increased concentration of Cd in the sea ward direction may be due to carbonate content in the sediment, where Cd is the only metal which exhibits enrichment as carbonate. The association of Cd with carbonate was reported by many investigators (Deure *et al.*, 1978; Förstner *et al.*, 1981), who found that Cd is usually co-precipitated with organic minerals and the occurrence of Cd in sediment is mainly due to the formation of

$CdCO_3$. Cd in sediment is less mobile than other metals due to the formation of $CdCO_3$ phases in alkaline media. Also, the highest concentration of Cd may be due to the use of Cd as pigments in ship painting (Moore *et al.*, 1985). The concentration of Mn in sediment may reflect atmospherically transported material (Voutsinou *et al.*, 2000). Meanwhile, the concentration of Ni may be due to the dissolved load by natural chemical weathering process (Gaiero *et al.*, 2001). Grain size analysis indicated that the pollution at this region may be due to geological changes (Table 5).

Risk assessment

The results of this health risk assessment indicated that sediment contamination by Cd, Zn in this area may present a health hazard, if the risk was calculated on the basis of the standard exposure model. Dermal absorption depends on the dermal absorption rate for the individual compounds. Ingestion of sediment was considered an important route of exposure if exposure, was calculated based on location-specific input data. However, no specific data on the sediment ingestion rate by children and adults were available for risk assessment purposes. Therefore, sediment ingestion rates were based on soil ingestion rates in relation to contaminated sites. Other exposure routes were of minor importance in relation to total exposure levels for heavy metals, while swimming contributed less than 1% to the total average exposure levels.

CONCLUSION

This study showed that the level of heavy metals were low in most of sampling station. The application of the standard exposure model, which is currently used by the Dutch government, indicates that sediment contamination by Zn and Cd may cause a health 'risk in relation to recreational activities. In general, exposure risk assessment of present type can be used for screening purposes to identify important exposure pathways and to determine the urgency of sediment remediation actions.

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Table 1. Trace metal concentrations (mean ± SD) for reference material SD-M-2/TM marine sediment.

Metal	unit	SD-M-2/TM	Confidence interval n =5
Mn	g/kg	12.1	5.2-15.6
Pb	mg/kg	22.8	20.1-25.6
Zn	mg/kg	74.8	72.0-78.3
Fe	g/kg	27.1	25.0-28.5
Cd	mg/kg	0.113	0.108-0.149
Cu	mg/kg	32.7	31.7-34.2
Co	mg/kg	13.6	13.1-14.2
Cr	mg/kg	77.2	64.0-82.8
Ni	mg/kg	56.1	53.3-58.5

Data expressed on a dry weight (dw) basis; 95% confidence intervals of the median; n = number of accepted laboratory means which were used for calculation of recommended, information values and confidence intervals.

Table (2): Mean concentration of total and leachable heavy metals at different stations along Med-sea (mg/Kgdw)

Stations \ Metal	Mn			Pb			Zn			Fe			Cd		
	T	L	%	T	L	%	T	L	%	T	L	%	T	L	%
- Sidi Krir (open sea)	21.76	19.66	90.0	61.23	56.19	91.7	162.02	15.38	9.4	189.87	151.89	80	4.8	3.48	73
- Sidi Krir (west)	20.47	15.39	75.0	57.04	47.64	83.5	29.49	15.21	51.5	368.83	290.86	79	6.8	5.9	87
- Sidi Krir (east)	12.58	12.05	95.7	47.93	37.30	77.8	150.32	21.69	14.4	200.06	177.92	89	4.7	3.85	82
- Sidi Barrany	17.08	17.18	100.0	68.42	53.19	77.7	110.66	16.99	15.4	133.51	126.87	95	5.65	4.76	84
- El-Sallum (1-1)	12.12	10.08	83.1	63.48	62.42	98.3	18.96	12.61	66.5	159.67	153.13	96	5.48	4.78	87
- El-Sallum (1-2)	90.21	85.24	94.5	55.24	48.87	88.4	36.4	23.18	63.6	389.41	349.67	89	4.31	4.03	94
- El-Sallum (60m)	13.77	13.56	98.4	67.01	55.98	83.5	28.35	15.32	54	116.24	80.11	69	4.27	4.24	99
Mean	26.85	24.73	91	60.05	51.66	85.8	76.6	17.20	39.3	222.51	190.06	85.3	5.14	4.43	86.6
± SE	10.66	10.16	-	2.7	3.03	-	23.59	1.45	-	41.97	36.08	-	0.34	0.30	-
Mann-whitney-U.test		19.0			10.0			2.0			18.0			12.0	
P-value		0.482			0.064			0.004			0.406			0.110	

Table (2): Cont.

Metal / Stations	Cu			Co			Cr			Ni		
	T	L	%	T	L	%	T	L	%	T	L	%
- Sidi Krir (open sea)	22.41	8.00	36	30.39	11.62	38	13.81	12.67	92	37.46	31.82	85
- Sidi Krir (west)	17.78	8.14	46	27.52	26.59	97	28.58	25.78	90	36.61	35.61	97
- Sidi Krir (east)	8.04	7.96	99	29.73	17.35	58	24.32	21.03	86	42.76	31.66	74
- Sidi Barrany	12.44	11.9	98	28.94	19.24	66	22.44	16.81	75	40.16	24.10	60
- El-Sallum (1-1)	7.04	6.53	93	23.27	16.59	71	18.22	16.09	88	43.15	25.36	59
- El-Sallum (1-2)	11.23	9.54	85	24.75	12.13	49	13.84	13.22	96	33.37	29.89	90
- El-Sallum (60m)	7.43	5.69	77	28.34	17.36	61	19.23	16.83	88	39.33	25.33	64
Mean	12.30	8.25	76.3	27.56	17.27	62.9	20.06	17.49	87.9	38.98	29.11	75.6
± SE	2.20	0.77	-	0.99	1.88	-	2.06	1.73	-	1.31	1.62	-
Mann-whitney-U. test		14.0			2.0			16.0			1.0	
P-value		0.180			0.004			0.277			0.003	

Table 3. Mean concentrations of some heavy metals at different places alonge Alexandria coastal sea (mg/ kg)

Stations/ Metals	Zn		Cu		Pb		Cd	
	T	L	T	L	T	L	T	L
Abu- Qir bay	99.8	57.1	77.02	40.35	84.3	54.3	3.5	1.6
Easten Harbur	52.6	18.6	38.6	17.9	114.9	72.36	4.8	3.1
EL- Mex bay	88.7	49.9	59.4	17.24	84.6	49.3	3.5	2.2

Table 4. Avarage total exposure levels of total and leachable heavy metals (mg/kg/day).

	Fraction	Total		Leachable	
Metal		Adult	Child	Adult	Child
Mn	Ingestion of sediment	4.03×10^{-3}	53.7×10^{-3}	3.71×10^{-3}	55.48×10^{-3}
	Dermal contant	72.49×10^{-3}	338.3×10^{-3}	66.8×10^{-3}	311.7×10^{-3}
	Total	76.52×10^{-3}	392.01×10^{-3}	70.51×10^{-3}	367.22×10^{-3}
	Daily exposure	0.732		0.657	
Pb	Ingestion of sediment	9.01×10^{-3}	120.1×10^{-3}	7.75×10^{-3}	103.3×10^{-3}
	Dermal contact	162.14×10^{-3}	756.6×10^{-3}	139.49×10^{-3}	650.8×10^{-3}
	Total	171.14×10^{-3}	876.7×10^{-3}	147.2×10^{-3}	754.1×10^{-3}
	Daily exposure	1.635		1.465	
	St. range of hazard	1.7-4.4			
Zn	Ingestion of sediment	11.49×10^{-3}	153.2×10^{-3}	2.58×10^{-3}	34.43×10^{-3}
	Dermal contact	206.82×10^{-3}	965.52×10^{-3}	46.44×10^{-3}	216.72×10^{-3}
	Total	218.31×10^{-3}	1118.4×10^{-3}	49.02×10^{-3}	251.1×10^{-3}
	Daily exposure	1.99		0.471	
	St. range of hazard	1.08-1.2			

Table 4. cont.

	Fraction	Total		Leachable	
Metal		Adult	Child	Adult	Child
Co	Ingestion of sediment	4.134×10^{-3}	55.12×10^{-3}	2.59×10^{-3}	34.54×10^{-3}
	Dermal contact	74.412×10^{-3}	347.26×10^{-3}	46.63×10^{-3}	217.6×10^{-3}
	Total	57.17×10^{-3}	402.38×10^{-3}	49.22×10^{-3}	252.14×10^{-3}
	Daily exposure	0.752		0.471	
Cr	Ingestion of sediment	3.01×10^{-3}	40.12×10^{-3}	2.62×10^{-3}	34.98×10^{-3}
	Dermal cotact	54.16×10^{-3}	252.76×10^{-3}	47.23×10^{-3}	220.37×10^{-3}
	Total	57.17×10^{-3}	292.88×10^{-3}	49.85×10^{-3}	255.35×10^{-3}
	Daily exposure	0.547		0.476	
Ni	Ingestion of sediment	5.85×10^{-3}	77.96×10^{-3}	4.37×10^{-3}	58.22×10^{-3}
	Dermal Contact	105.25×10^{-3}	491.15×10^{-3}	78.6×10^{-3}	366.78×10^{-3}
	Total	111.09×10^{-3}	569.11×10^{-3}	82.96×10^{-3}	425×10^{-3}
	Daily exposure	1.064		0.795	

Table 4. Cont.

	Fraction	Total		Leachable	
Metal		Adult	Child	Adult	Child
Fe	Ingestion of sediment	33.37×10^{-3}	445.02×10^{-3}	28.51×10^{-3}	380.12×10^{-3}
	Dermal contant	598.08×10^{-3}	2803×10^{-3}	513.16×10^{-3}	2394.47×10^{-3}
	Total	631.45×10^{-3}	3248.02×10^{-3}	541.67×10^{-3}	2774.82×10^{-3}
	Daily exposure	6.0514		4.976	
Cd	Ingestion of sediment	0.77×10^{-3}	10.28×10^{-3}	0.66×10^{-3}	8.86×10^{-3}
	Dermal contact	13.88×10^{-3}	64.76×10^{-3}	11.96×10^{-3}	55.82×10^{-3}
	Total	14.65×10^{-3}	75.04×10^{-3}	12.62×10^{-3}	64.68×10^{-3}
	Daily exposure	0.1394		0.1205	
	St. range of hazard	0.11-0.16			
Cu	Ingestion of sediment	1.85×10^{-3}	24.6×10^{-3}	2.59×10^{-3}	34.54×10^{-3}
	Dermal contact	33.21×10^{-3}	154.98×10^{-3}	46.63×10^{-3}	217.6×10^{-3}
	Total	35.06×10^{-3}	179.58×10^{-3}	49.22×10^{-3}	252.14×10^{-3}
	Daily exposure	0.336		0.471	
	St. range of hazard	0.02-0.05			

Table 5. Grain size analysis of sediment.

Stations	Type of sediment (Mz)	Sorting
Sidi krir (open sea)	2.6 F fine sand	0.32 very well sorted
Sidi-Krir east	3.4 F very fine sand	0.59 moderately well sorted
Sidi-Krir west	1.9 F medium sand	0.325 very well sorted
Sidi-Barrany	2.2 F fine sand	0.325 very well sorted
Sallum 1-1	2.5 F fine sand	0.32 very well sorted
Sallum 1-2	2.5 F fine sand	0.32 very well sorted
Sallum (60 m)	2.3 F fine sand	0.32 very well sorted

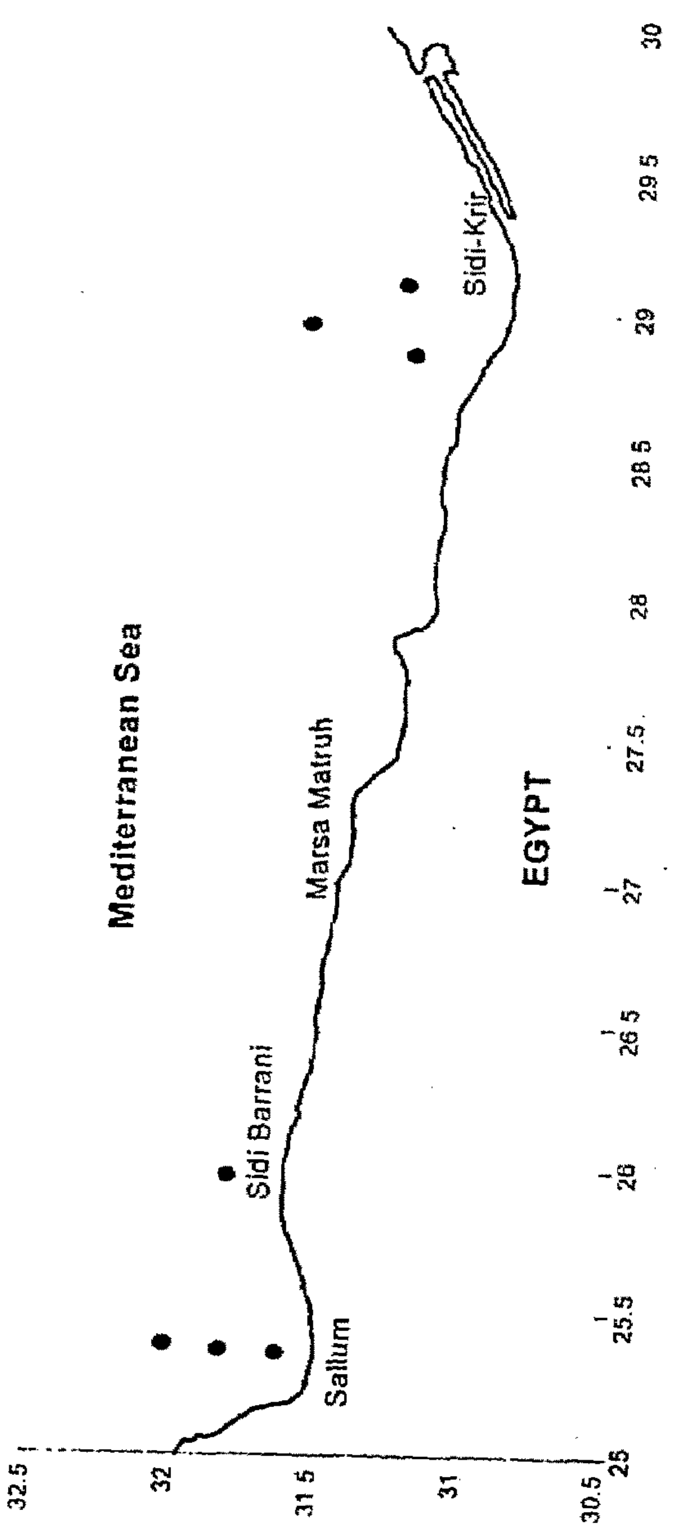

Fig 1. Sample locations.

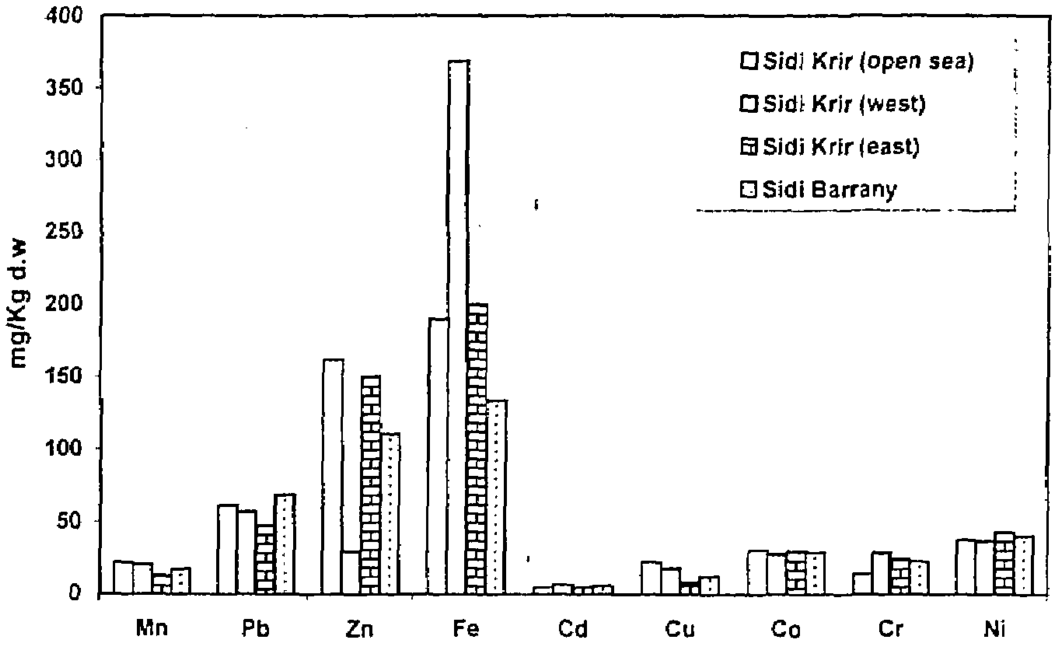

Fig (2-a) : Concentration of total heavy metals (mg/Kg d.w) in sediment samples from Sidi Krir region. The statistical significance of differences were evaluated by the Mann-Whitney-test

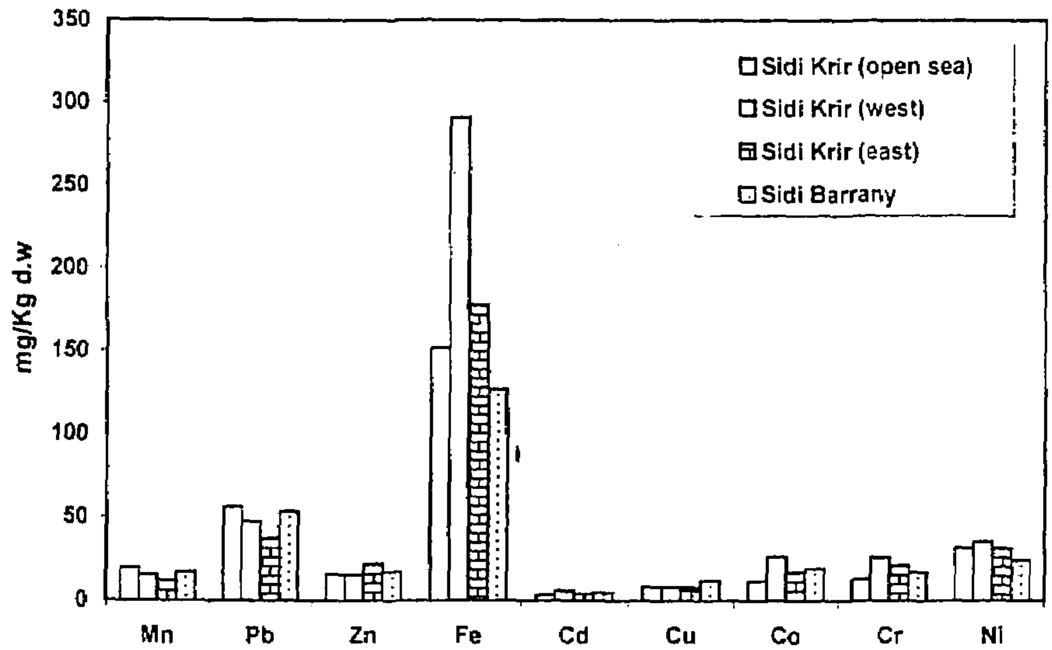

Fig (2-b) : Concentration of leachable heavy metals (mg/Kg d.w) in sediment samples from Sidi Krir region. The statistical significance of differences were evaluated by the Mann-Whitney-test

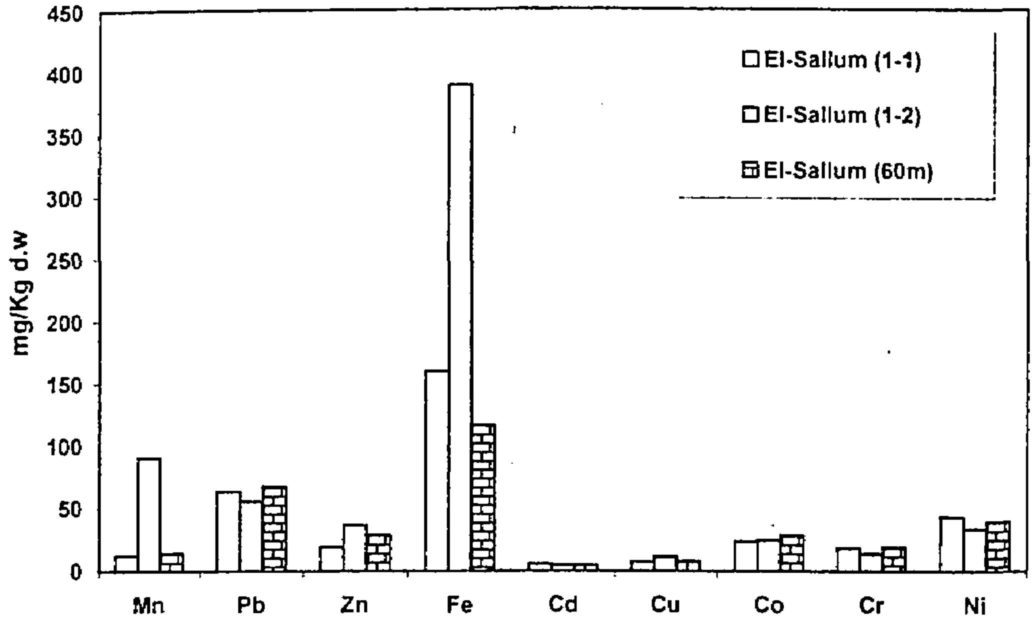

Fig (3-a) : Concentration of total heavy metals (mg/Kg d.w) in sediment samples from Salum region. The statistical significance of differences were evaluated by the Mann-Whitney-test

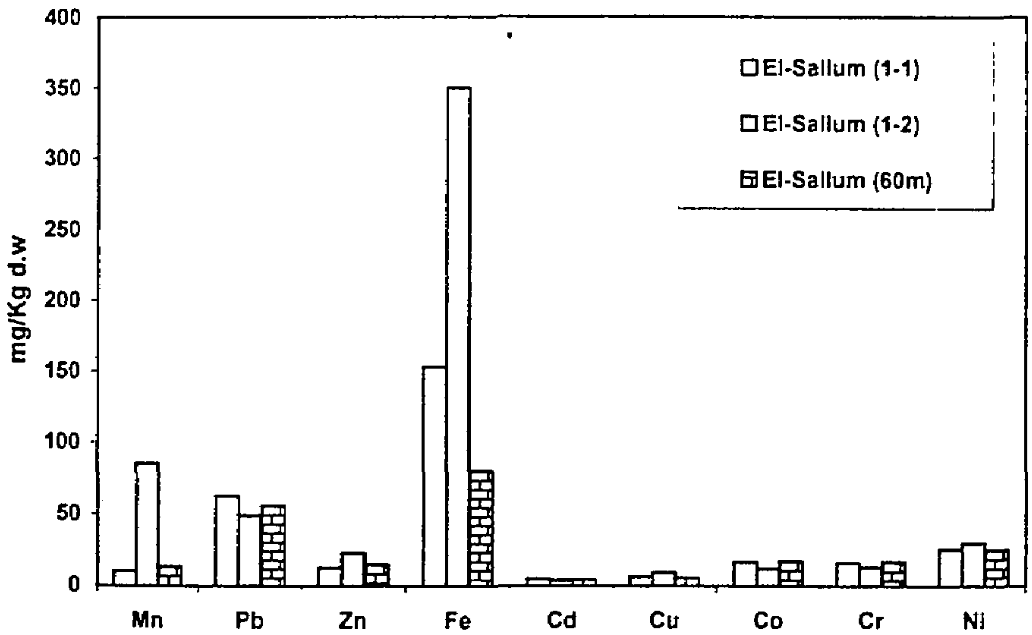

Fig (3-b) : Concentration of leachable heavy metals (mg/Kg d.w) in sediment samples from Salum region. The statistical significance of differences were evaluated by the Mann-Whitney-test