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EVALUATION OF HEAVY METALS ALONG THE MEDITERRANEAN COASTAL WATERS OF THE NILE DELTA REGION, EGYPT

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ABSTRACT: This work focuses on four marine sites in the Mediterranean Sea around the Nile Delta, Egypt. Surface water samples were collected seasonally during 2003. The concentrations of some heavy metals in dissolved form (Fe, Mn, Zn, Cu, Ni and Pb) are evaluated. The levels of heavy metals in the coastal waters were 11.92-30.45μgl^{-1} for Fe; 5.79-17.36μgl^{-1} for Zn; 0.30-0.83μgl^{-1} for Cu; 0.51-2.90μgl^{-1} for Ni and 0.53-10.31μgl^{-1} for Pb. These are compared, with sites in the estuaries and outlets of the Nile Delta. Fe (19.72-60.33μgl^{-1}); Mn (12.63-35.60μgl^{-1}); Zn (2.67-22.00); Cu (0.56-1.67μgl^{-1}); Ni (1.43-3.73μgl^{-1}); Pb (1.72-59.7μgl^{-1}). The results showed a remarkable decrease in the concentrations of different heavy metals with increased salinity.

Comparing the present data with the minimal risk concentration reported by WQC, the distribution of heavy metals was significantly lower in coastal sea water of the Mediterranean Sea off Egypt. The study indicated also that the average contents of Ni and Pb are slightly high in the area of water exchange than those reported by WQC.

KEY WORDS: Heavy metals, Mediterranean Sea. Nile Delta Region.

INTRODUCTION

Trace metal concentrations in the estuaries, outlets and marine waters associated with the Nile Delta Region are the results of both natural and anthropogenic processes. Municipal, agricultural and industrial waste waters are well known to be principal sources of trace metals. Most pollutants end up in coastal and marine waters, potentially causing alteration or loss of habitat and probable reductions in numbers of species. The untreated waste waters of the northern lakes in the Nile Delta region are released directly into the Mediterranean and could have an adverse effect on marine organisms and water quality. At suitable concentrations, some trace metals are essential for enzymatic activity but they also form an important group of enzyme inhibitors when natural concentrations are exceeded (Bryan, 1976). A number of studies have been carried out in the last year on the Egyptian Mediterranean Waters and Nile River tributaries, Rosetta and Damitta as well as the outlets of El-Burullus and El-Gamil. (Abdel-Moati, 1981; Fahmy, 1981; El-Rayis and Saad, 1984; Saad and Abbas, 1985; Saad *et al.*, 1991; Abu El-Khair, 1993).

The objective of the present work is to investigate the present status of some heavy metals in the coastal waters of the Mediterranean adjacent to the Nile Delta and to evaluate the impact of the effluent.

MATERIALS AND METHOD

Sampling analytical methodology:

All reagents were of analytical grade and Milli-Q water was used throughout the study. Glass was soaked in detergent, rinsed with water, washed in 10% HNO_3 for 5 days, rinsed with Milli-Q water and kept in the oven at 110°C until required. Eight-surface water samples were taken seasonally during the period from January to October 2003. These samples were collected to represent four sites of the Mediterranean Sea coast of Egypt (Rosetta and Damitta estuaries as well as El-Burullus and El-Manzala outlets). Two samples were collected (Fig. 1) from each site, one from both the estuaries (V, VII) and the other from the coastal seawater (I, II, III and IV) of the Mediterranean Sea. Extraction, analysis and measurements of trace metals were carried out according to Martin (1972).

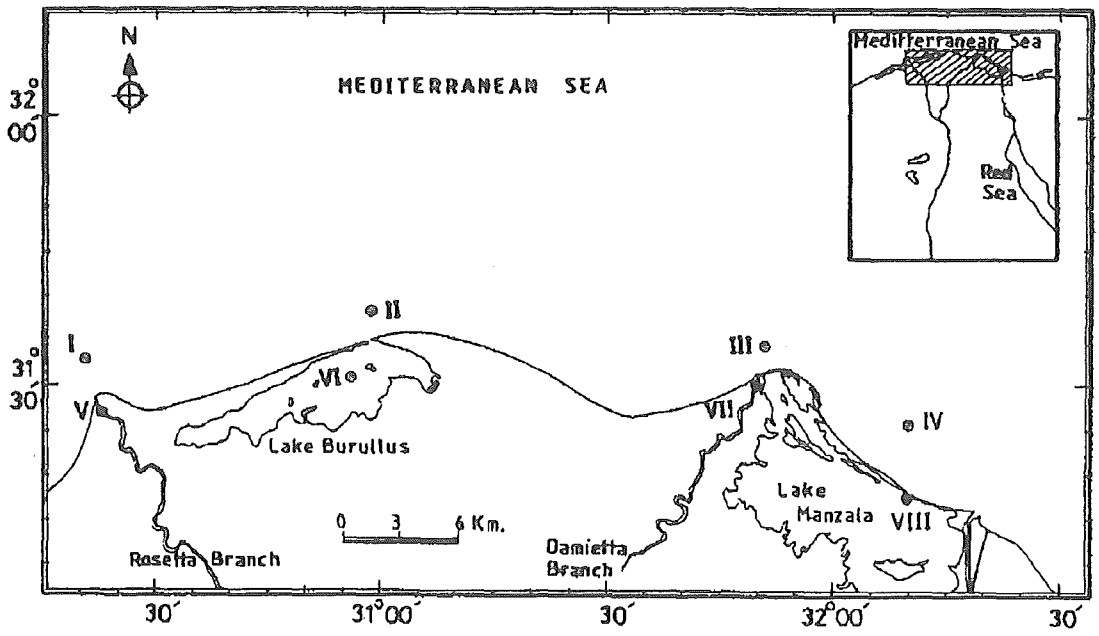

Fig. 1. Map of Sampling Stations.

Water samples were collected using Niskin bottle sampler in previously acid-washed polyethylene bottles. The samples were filtered as soon as possible after collection through a previously acid-washed 0.45μM memberane filter. The pH of filtrates was adjusted to 4-5 with 6N HCI. Metals in the filtered seawater were preconcentrated by complexing the metals with ammonium pyrolidine dithiocarbmate (APDC), extracting the complexed compound into methyl isobutyl ketone (MIBK) and back-extracted into an acidic aqueous solution (Boniforti *et al.*, 1984). Concentrations of different metals in the final acidic extracts were measured using a GBC-932 Ver. 1.1 atomic absorption spectrophotometer in the flame mode.

Quality control samples:

Quality control samples represented 10% of the total analysis load. Precision was determined by three replicate analysis of one sample. Expressed as coefficient of variation (C.V.), precision was 5, 6, 4, 8, 5 and 6% for Cu, Ni, Pb, Mn, Fe and Zn, respectively.

The accuracy of the pre-concentrated technique of dissolved trace elements determination was evaluated by spiking 750ml of seawater, previously stripped of all trace metals using the APDC-MIBK extraction.

Spiked samples were extracted using the previous technique and the concentrations of trace metals were determined. The recovery of metal spiked were 90% for Cu, 94% for Ni, 91% for Pb, 105% for Mn, 91% for Fe ad 95% for Zn.

Quality control samples were determined by using a reference material (NASS-5, national research of council of Canada) (Table 1) and applying the computerized 4.3 quality system program provided by DANIDA from VKI. Two natural samples were analyzed in duplicate in each of 6 batches of samples after spiking by a known concentration from the reference material. The same two natural samples were analyzed without spiking. The highest and lowest percentage recovery for spiked samples were used to determine the accuracy. It ranged from 90 to 110%, while precision agreed within 10%. The limits of detection (μgl^{-1}) were calculated by six determinations (duplicate measurements) in one batch of synthetic seawater. The detection limits were 0.045, 0.009, 0.02, 0.008, 0.007 and 0.03μgl^{-1} for Fe, Zn, Ni, Mn, Pb and Cu respectively. The results for the analysis of heavy metals in the reference material are shown in Table 2.

Table 1. Concentrations (mean $\pm$ SD) for different metals in the reference material (NASS-5, national research of council of Canada).

Metal	Found	Certified
Fe	0.197 ± 0.025	0.207 ± 0.035
Mn	0.910 ± 0.070	0.919 ± 0.057
Zn	0.098 ± 0.047	$0.102 \pm 0.0.39$
Cu	0.305 ± 0.055	0.297 ± 0.046
Ni	0.260 ± 0.057	0.253 ± 0.028
Pb	0.10 ± 0.005	0.008 ± 0.005

Statistical analysis:

A cluster analysis technique was performed on the results relating to the concentration of heavy metals of the water samples collected from the coastal water and the area of water exchange through the outlets or the estuaries. The object is to look for similar groups of stations or variables (heavy metals) which are grouped together in clusters through the application of this technique. Dissolved heavy metals studied can be classified into different categories.

Table 2. Range and Mean ± S.D. of Heavy Metals concentration (μgl^{-1}) in the Mediterranean sea water infront of Nile Delta (Jan. – Oct. 2003).

Station \ Metals		Fe	Mn	Zn	Cu	Ni	Pb
Stations of coastal seawater	I	11.92 - 23.10	5.79 - 10.68	1.28 - 2.93	0.43 - 0.83	0.51 - 1.20	2.40 - 10.31
		14.91 ± 4.73	8.83 ± 1.89	1.94 ± 0.65	0.55 ± 0.10	0.94 ± 0.29	5.02 ± 3.20
	II	15.12 - 30.45	7.12 - 14.24	0.86 - 5.07	0.40 - 0.83	0.80 - 1.40	0.53 - 8.21
		20.16 ± 6.19	10.78 ± 2.75	3.22 ± 1.54	0.55 ± 0.16	1.07 ± 0.24	2.59 ± 3.25
	III	12.00 - 18.44	7.95 - 14.24	1.13 - 7.4	0.30 - 0.67	0.53 - 2.90	0.74 - 5.47
		15.08 ± 2.39	9.88 ± 2.58	3.6 ± 2.32	0.46 ± 0.16	1.27 ± 0.95	2.68 ± 2.01
	IV	16.72 - 30.10	5.84 - 17.36	1.33 - 3.80	0.40 - 0.83	0.75 - 1.34	0.62 - 8.13
		21.56 ± 5.48	13.49 ± 4.49	2.64 ± 0.93	0.59 ± 0.17	0.99 ± 0.24	3.07 ± 2.97
	annual average	11.92 - 30.45	5.79 - 17.36	0.86 - 7.40	0.30 - 0.83	0.51 - 2.90	0.53 - 10.31
		17.93 ± 4.69	10.75 ± 2.93	2.85 ± 1.36	0.54 ± 0.15	1.07 ± 0.43	3.34 ± 2.86
Stations of water exchange	V	19.72 - 43.81	12.63 - 24.92	4.27 - 10.00	1.67 - 2.39	1.43 - 3.73	15.87 - 59.7
		30.66 ± 10.38	16.15 ± 5.08	6.88 ± 2.05	1.93 ± 0.27	2.66 ± 1.02	30.23 ± 17.60
	VI	30.16 - 41.36	14.24 - 35.60	2.67 - 22.00	1.39 - 3.27	1.63 - 3.73	1.72 - 24.53
		36.10 ± 4.06	22.32 ± 7.99	10.73 ± 7.02	2.08 ± 0.72	2.76 ± 0.75	9.54 ± 9.17
	VII	20.1 - 30.11	13.5 - 24.92	3.23 - 13.5	0.56 - 1.90	1.53 - 3.47	3.87 - 26.53
		26.03 ± 3.81	18.82 ± 4.06	9.4 ± 3.29	1.10 ± 0.50	2.42 ± 0.71	13.63 ± 9.43
	VIII	30.46 - 60.333	13.67 - 26.68	2.8 - 10.93	1.52 - 8.10	1.60 - 2.67	1.93 - 15.1
		44.08 ± 10.92	20.69 ± 4.69	6.77 ± 3.55	3.40 ± 2.73	2.05 ± 0.41	6.66 ± 5.03
	annual average	19.72 - 60.33	12.63 - 35.60	2.67 - 22.00	0.56 - 1.67	1.43 - 3.73	1.72 - 59.7
		34.22 ± 7.29	19.49 ± 5.46	8.45 ± 3.98	2.13 ± 1.06	2.47 ± 0.72	15.02 ± 10.31

RESULTS AND DISCUSSION

Distribution of heavy metals:

The regional and seasonal distributions of Fe concentration in the study area are presented in Figure 2. Fe concentration in the coastal seawater fluctuated between 11.92μgl^{-1} in spring at station I and 30.45μgl^{-1} in winter at station II. But a relatively high concentration of Fe was recorded in the mixing water inside the estuaries and outlets, perhaps as a result of the polluted water discharged from the surrounding area.

It should be pointed out that there was a tendency for a decreasing iron concentrations in the coastal seawater at stations (I, II, III and IV). The annual mean values of iron ranged from 26.03±4.06 to 44.08±10.92μgl^{-1} in the sites of water exchange. These values were twice those reported in the coastal seawater in the same regions which ranged from 14.91±4.73 to 21.56±5.48μgl^{-1} (Table 2).

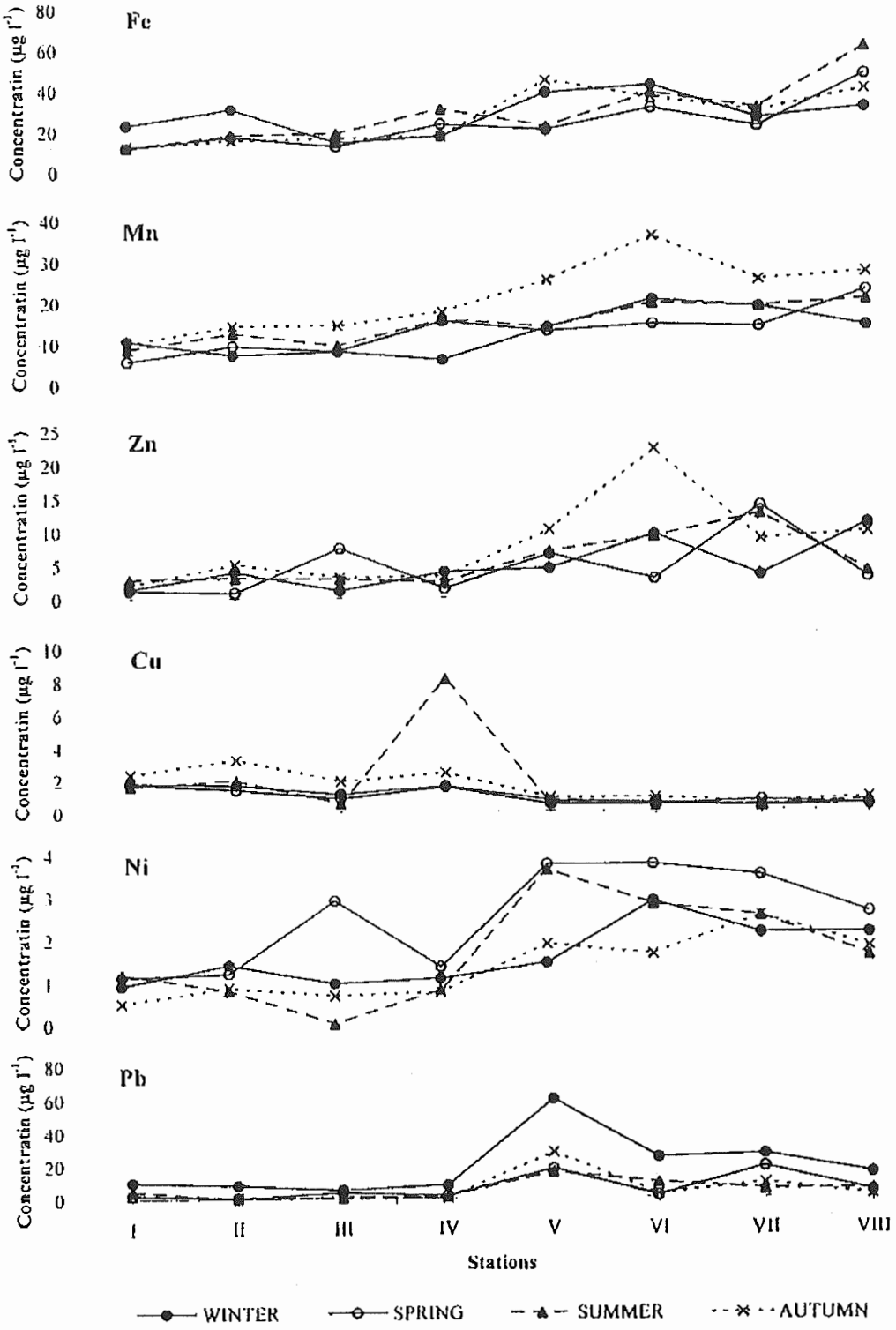

Fig. 2. Regional and seasonal variations of dissolved heavy metals along the Mediterranean Sea in front of Nile Delta, Egypt during 2003.

Table 3. Dissolved heavy metal concentrations (μgl^{-1}) in inland and coastal waters compared to the reported values.

Location	Fe	Mn	Zn	Cu	Ni	Ph	References
Coastal water							
Mediterranean Sea (Egypt)	11.92-30.45	5.79-17.36	0.86-7.40	0.30-0.83	0.51-2.90	0.53-10.31	Present study
Eastern harbour and El-Mex Bey (Egypt)	-	-	-	4.9	-	0.5	Shriadah and Emara (1991)
Red Sea (Egypt) offshore	0.56-4.4	0.06-0.21	0.13-1.17	0.07-0.29	0.05-0.21	0.02-0.68	Shriadah *et al.*, (2004)
Red Sea (coast, Egypt)	16.2	0.83	-	5.1	-	-	Saad and kandeel (1988)
Red Sea (Jeddah, KSA)	-	-	6-14	4.9	-	1.1-27	Hamza and Amireh (1992)
Arabian Gulf (Bahrain)	0.01-0.08	0.03-0.13	0.03-11.25	0.03-0.38	0.13-0.53	0.03-0.23	Al-Sayed *et al.*, (1994)
South Agcan (Greece)	-	0.13-29.12	-	0.16-15.21	0.55-15.98	0.23-9.22	Voutsinou-Talladour *et al.*, (1997)
El-Mex Bey (Egypt)	13.0	-	-	3.0	-	-	Okbah (2000)
Island waters							
Estuaries & outlets (Egypt)	19.72-60.3	12.36-35.6	2.67-22.0	0.56-1.67	1.43-3.73	1.72-59.7	Present study
River Nile (Egypt)	2.5	0.46	8.18	1.3	-	-	El-Rayis & Saad (1985)
Lake Mariut (Egypt)	42.5	-	18.3	10.6	-	-	Saad (1985)
Ora River (Nigeria)	124.7	450	7.5	8	-	5	Mombeshota *et al.*, (1981)
Lake Victoria (Kenya)	-	50-3276	25-125	5-57.0	-	7-93.6	FAO (1992)
Background							
Rivers	40.0	7.0	20.0	7.0	-	0.3	Burton and liss (1976)
Coastal waters	2.0	0.4	2.5	1.0	-	0.03	Martin and Whitfield (1983)
Minimal Risk Concentration	50	20	20	10	2	10	WQC (1972)

The levels of Mn distribution (Fig. 2 and Table 2) in the coastal sites and the sites of water exchange showed a wide variation in its content. For the coastal waters, Mn concentration varied between 5.79μgl^{-1} in spring at station I and 17.36μgl^{-1} in autumn at station IV, while the level of Mn concentration in the sites of water exchange increased 2-3 fold. This reflects the influence of drainage water on the levels of Mn content, which ranged from 12.63μgl^{-1} at station V in spring to 35.6μgl^{-1} at station VI in autumn (Fig. 2). The annual mean concentrations of dissolved Mn showed considerable variations

(Table 2) for the investigated area. It ranged from 16.15±08 to 22.32±7.99μgl^{-1} in the sites of water exchange and ranged between 8.83±1.89 and 13.49±4.49μgl^{-1} for the coastal seawater.

Concentrations of dissolved Zn at the sites of water exchange and the coastal seawater are shown in Table 3 and Fig. 2. The range and mean values revealed wide variation in regard to the coastal seawater stations and the other stations in the area of water exchange at the same region. The distribution pattern of dissolved Zn indicates significantly lower concentration in the study area. Seasonal and regional variations of dissolved Zn concentrations, ranged from 0.86 to 3.87μgl^{-1} for the coastal water and from 2.8 to 22.0μgl^{-1} for the stations of water exchange (stations V, VI, VII and VIII).

The regional and seasonal variations of dissolved Cu values in the investigated area are presented graphically in Fig. 2. In the sites of water exchange, the absolute values of Cu concentration fluctuated between 0.56μgl^{-1} at station VII in summer and 3.27μgl^{-1} at station VI in autumn. In the coastal sites, it ranged between a minimum of 0.30μgl^{-1} in winter at station III and a maximum 0.83μgl^{-1} in autumn at station I, II and IV. The annual average concentrations of dissolved Cu in the investigated area was relatively high in the sites of water exchange, ranged from 1.10±0.58μgl^{-1} to 3.40±2.73μgl^{-1} (Table 2) and decreased in the coastal sites, ranged from 0.46±0.18 to 0.62±0.17μgl^{-1}.

Seasonal variations of dissolved Ni concentration in the investigated area showed that the dissolved Ni at the sites of water exchange is higher than that in the coastal sites (Fig. 2). The dissolved Ni concentration ranged between 0.94±0.29 at site I and 1.27±0.95μgl^{-1} at site III for the coastal water. In the sites of water exchange, it ranged from 2.05±0.41 to 2.76±0.75μgl^{-1} (Table 2).

Regional variations of dissolved Pb concentration in the present study showed a decreasing trend from the sites of water exchange to the coastal sites (Table 2 and Fig. 2). Seasonal distribution of dissolved Pb concentrations in the coastal sites varied between 0.53μgl^{-1} in summer at station II and 10.31μgl^{-1} in winter at station I. The results revealed relatively high concentration of dissolved Pb in the sites of water exchange, ranging from 1.72μgl^{-1} in spring at station VI and 59.7μgl^{-1} in winter at station V (Fig. 2).

Comparing the present concentrations in the coastal water with those reported in the literature (Table 3), the concentrations observed around the Nile Delta are much higher than those recorded in the Red Sea and are similar to the south Agean, Greece (Voutsinou-Tallodour *et al.*, 1997; Shriadah *et al.*, 2004; Al-Sayed *et al.*, 1994). The concentrations of heavy metals in the area of water exchange are much lower than the values of Lake Victoria (FAO, 1992) and much higher than that reported in River Nile (El-Rayis and Saad, 1984). In general, the levels of dissolved heavy metals in the coastal water of the Mediterranean Sea are lower than the minimal risk concentration reported by WQC (1972).

Correlation matrix:

The correlation matrix between different heavy metals revealed correlations for some metals (Table 4). The correlation of Ni and Pb with Cu was poor ($r<0.2$) and between Mn and both Ni and Pb (r-0.2 and 0.06, respectively), reflecting different sources and different biogeochemical behaviours (Shriadah and Emara, 1991). It also indicated the different impacts of the land based sources on this area (Mohamed *et al.*, 2003). However, good positive correlations ($P<0.05$) were found between several pairs of heavy metals.

There were significant relationship between Fe and Cu (r=0.76), Mn and Zn (.066), Mn and Fe (0.66); Mn and Cu (r=0.53); Zn and Ni (0.42); as well as Fe and Zn (0.33). These might indicate the important role of iron and manganese as a common sink for trace elements such as Cu and Zn.

Table 4. Correlation matrix between metals (n=32, p>0.05).

	Cu	Ni	Pb	Mn	Fe	Zn
Cu	1					
Ni	0.192	1				
Pb	0.138	0.287	1			
Mn	0.533	0.202	0.061	1		
Fe	0.76	0.284	0.314	0.664	1	
Zn	0.296	0.421	0.155	0.662	0.332	1

Cluster analysis:

The cluster analysis of heavy metals variations from all stations (Fig. 3) demonstrated that Cu, Fe, Mn and Zn are firt linked together with small distance, then the combined distance increased to reach Ni and Pb. The highest similarity observed is between Cu and Fe, (r=0.76: P>0.05).

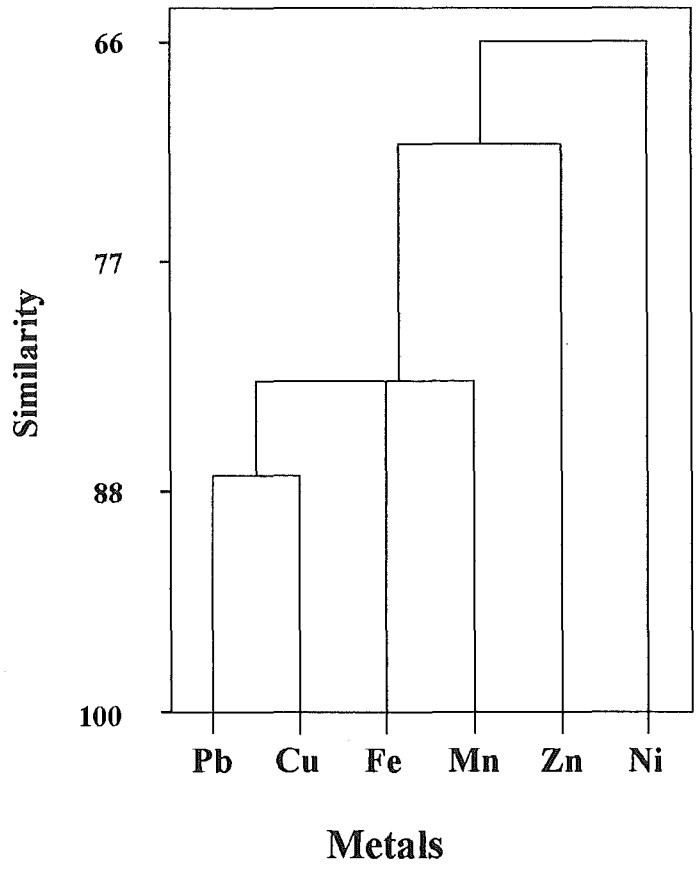

Fig. 3. Dendrogram for different stations of the Mediterranean coastline and water exchange versus selected heavy metals.

The dendrogram for selected heavy metals versus different stations showed three major clusters (Fig. 4). The cluster analysis revealed the highest degree of similarity between stations IV and VIII which are linked together and similarly between stations I and II. Such a pattern indicates that the area of water exchange at station VIII and the coastal water at station IV is more active than the other sites. Also, the cluster analysis showed similarity between stations I and II. These analyses indicate a relationship between stations I and II with VI. Stations VII and III are combined with stations IV and VIII, then the combined distance increased to reach station V. Stations IV and VIII are affected by the wastewater discharged from Lake Manzalla.

The cluster analysis of the investigated area showed that all stations are clustered together at different levels of similarities except station V, which is mostly affected by the drainage water discharged from Rashid Estuary.

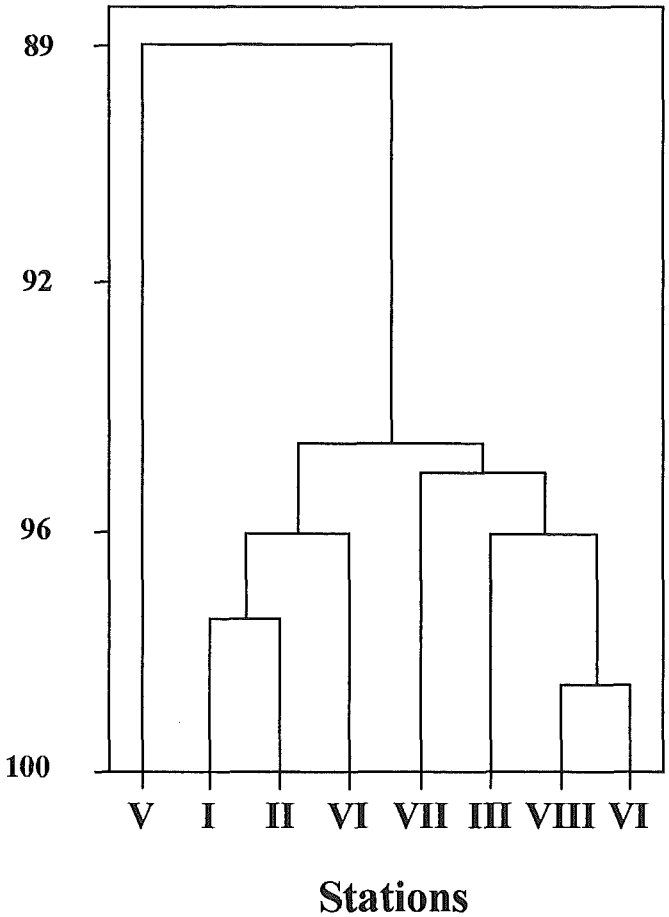

Fig. 4. Dendrogram for selected heavy metals versus different stations of the Mediterranean coastline and water exchange.

CONCLUSION

Lower concentrations of heavy metals occur along the Egyptian coastal water of Nile Delta compared to other areas of the world. The concentrations in the area of water exchange (estuaries and outlets) exhibit no significant effects on the levels of heavy

metals content of the Mediterranean Sea water. The impact of anthropogenic inputs on the distribution of heavy metals along the coastal area was limited.

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