Spatial variation and environmental risk assessment of heavy metal in the surficial sediments along the Egyptian Red Sea coast

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ABSTRACT
The concentrations of certain heavy metals (Fe, Mn, Zn, Cr, Ni, Pb, Cu, and Cd) of sandy sediment samples collected from eight sites along the Egyptian Red Sea coast were investigated in order to evaluate the pollution status and environmental risk assessment of the study area. The results of the partitioning study showed that the average concentrations of the heavy metals analyzed in investigated sediment exhibited the following decreasing order Fe > Mn > Zn > Cr > Ni > Pb > Cu > Cd. The degree of surface sediment contamination was computed using Metal Pollution Index (MPI) and Geo-accumulation index (I geo). Metal Pollution Index showed that station 5 (Qusier Middle) suffered with metal pollutions. Association with adverse effects to marine organisms was determined using the classification of the sediments according to Sediment quality guidelines (SQGs). Sediment quality guidelines based on the consensus approach revealed that Pb and Cu had no adverse ecological effects.
Key words: Heavy Metal, Red Sea, Metal Pollution Index (MPI), Geo-accumulation Index (I geo) and Sediment Quality Guidelines (SQGs).

INTRODUCTION

Rapid industrialization and economic developpment in coastal regions have undoubtedly introduced heavy metals into these regions all over the world and it is more so in Red Sea, where in the industrial growth has been phenomenal during the last decade. Coastal sediments act as ultimate sinks for metals that are discharged into this environment. Recent studies have shown that sediment from coastal milieu near industrial and urban areas have been contaminated to some extent by heavy metals, which are toxic to biota (Feng *et al.* 2004; Romano *et al.* 2004; Santos *et al.* 2005).

Thus, studies of metals in the sediment are important to decipher the source, to evaluate their enrichment pattern and to assess any probable localized influence. This leads to better understanding of their behaviour in an aquatic environment and also helps in detecting the source of the pollution (Jayaprakash *et al.* 2008; Pekey 2004; Buccolieri *et al.* 2006). Sediments are also important carriers of metals in the hydrological cycle, because metals are partitioned with the surrounding waters, and thus they reflect the quality of an aquatic system. Metals accumulated in this process may be subsequently released to the overlying water column as a result of either physical disturbance or diagnosis and the sediments may persist as a source of pollutants long after the cessation of direct discharges.

Moreover, digenetic reactions are also important near the sediment–water interface responding to redox changes and affecting metal concentrations in vertical sediment profiles. Thus, this study provides some information on the surface sediment geochemistry of the investigated area to improve the sediment data inventory, characterize the geochemistry of heavy metals in surface sediments as well as understand the influence of anthropogenic activities and distribution of metals in sediments along Red Sea coast.

MATERIALS AND METHODS

Study Area

The Red Sea is one of the most important repositories of marine biodiversity on a global scale and features a range of important coastal habitats. Red Sea is still one of the least ecologically disturbed seas relative to other closed water bodies. There is a growing risk of marine pollution, and environmental degradation from urban expansion, tourism and industrial development; this causes excessive sedimentation, which in turn leads to suffocation of benthic communities and ecosystem damage (Gesamp, 2001). Coastal habitat is being converted for urban and industrial development. Tourism and recreation usage are growing quickly. The Red Sea remains one of the busiest marine transport regions of the world, making this multi–national sea prone to major metals and oil pollution. It is extended from Latitude 22° 15 to 27° 15 N and from Longitude 33 ° 50 to 37 ° 50 E. (Fig.1).

### Sample collection and analyses

Surface sediment samples were collected from eight stations (Table 1) in the depth 3-5 m after tidal zone using Hydro-Bios stainless steel grab sampler along Egyptian Red Sea Coast (Fig.1). Each sample was placed in polyethylene plastic bags and they were then kept in an ice box. As soon as the field work was finished, samples were brought to laboratory and stored

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at -20 freezers for future analysis. The samples for assessment were dried using room temperature and kept in an acid washed container for future use.

The total metals were extracted from the sediment samples according to standard operating procedures for trace metals (UNEP/IAEA, 1986). An exact weight (1g) of dry sample (dried at room temperature) of sediment was completely digested in Teflon vessels using a mixture of HNO_3, HF and $HClO_4$ (3:2:1) at $70^{\circ}C$ (UNEP/IAEA, 1986 and Ajay and Van Loon, 1989).

The final solution was diluted to 10 ml with double deionized distilled water. All digested solutions were analyzed in duplicate using an atomic absorption spectrophotometer (AAS Perkin Elmer analyst, Model 100) and the results were expressed in µg/g; dry weight. Reagents of analytical grade were utilized for the blanks and calibration curves. Precision was checked against standard reference material and lied within the range of certified values with 90-97% recovery for all metals studied.
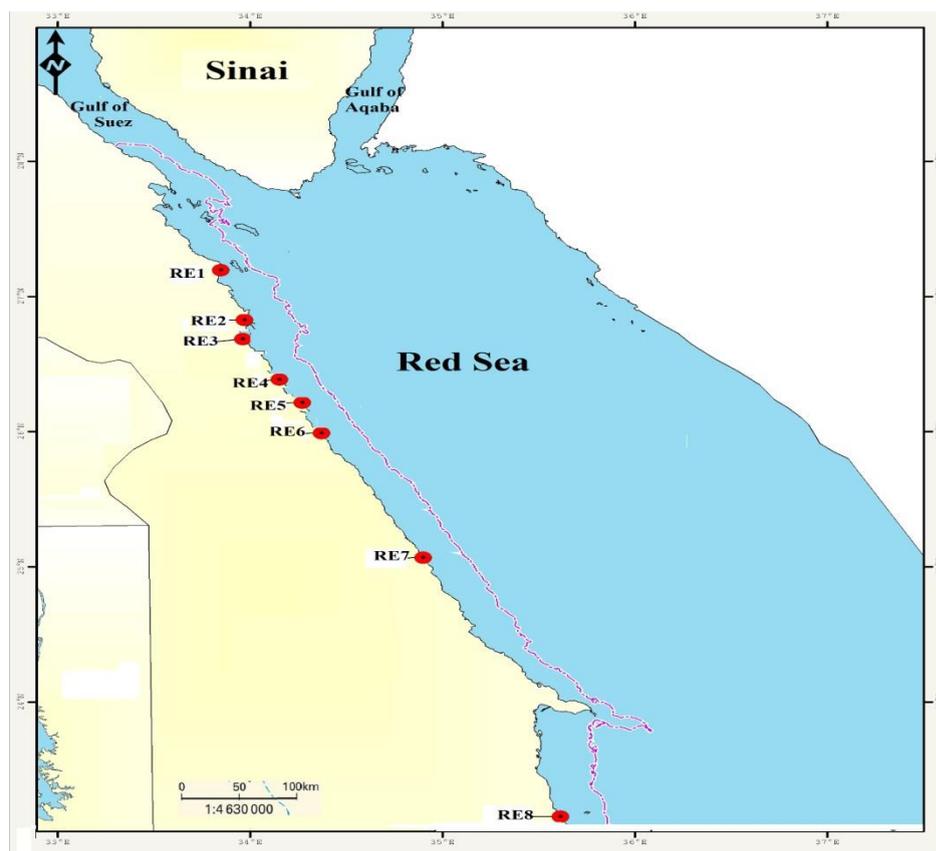
Figure (1): Sampling Sites of the Study Area.

Table (1): List of names, sites codes and sampling locations of the Red Sea Coast.

Name	Site Code	Latitude	Longitude
Red Sea			
1- Hurghada (Sheraton Hotel)	RE-1	27° 1' 37.5"	$33^{o}50'$ 48.4"
2- Safaga North (Suma Bay)	RE-2	26° 47' 34.9"	33° 56' 12.5"
3- Safaga Middle (Phosphate Company)	RE-3	26° 30' 20"	34° 0' 20"
4- El Hamarawein (North Port)	RE-4	26° 15' 9"	34° 12' 5"
5- Qusier Middle (Phosphate mining)	RE-5	26° 8' 30"	34° 14' 30"
6- Qusier South (Port phosphate)	RE-6	25° 55' 48"	34° 36' 36"
7- Marsa Alam (Port)	RE-7	25° 4 ' 6.1"	34° 45' 0 .4"
8- Bir Shalatin (Fishing Port)	RE-8	23° 9' 9.9"	35° 36' 48.3"

Water content (WC %) measurement is carried out gravimetrically after drying at a maximum temperature of 105ºC over night to constant weight. Controlled period of time, is sufficiently high to eliminate "free" forms of water and sufficiently low not to cause a significant loss of organic matter and unstable salts by volatilization (Pansu and Gautheyrou, 2006). The carbonates were determined by destroyed using hydrochloric acid and the volume of released carbon dioxide is measured at controlled temperature and pressure.

This method is part of the international standard (NF ISO, 10693) an improvement of this method using the pressure calcimeter. The pressure change caused by the reaction between HCl and the soil or sediment (0.5g dry and sieved) sample is measured with a digital plunge-in Manometer through a silicone–rubber septum placed on a screw-capped tube (Balazs *et al.* 2005). The result is generally expressed as a percentage of limestone:

$$CaCO_3 \% = \frac{V \times M \times 2.28 \times 100}{1.000 \times P}$$

V: volume of evolved CO_2 per sample (ml), M: mass in g of 1L of CO_2 in titration pressure and temperature conditions (M= 1.773), P: weight of sample (g) and 2.28 is the ratio between the molecular weight of $CaCO_3$ to that of CO_2.

A grain-size analysis was carried out using the conventional method (Folk, 1974). About 30g of washed and quartered dried sample was subjected to the combined technique of dry sieving and pipette analysis. Granulometric analysis was carried out manually to describe the sediment. Organic carbon was determined using acid/dichromate titration method as described by (Gaudette *et al.* 1974). The results of the analysis were calculated by the following equation:

Organic Carbon% = 10 (1-T/S) [1.0N (0.003) (100/W)]

Where T= sample titration, ml ferrous solution, S= standardization blank titration, ml ferrous solution. N= normality of $K_2Cr_2O_7$, and W= weight of sediment sample (g).

Metal Pollution Index (MPI), Geoaccumulation Index (I_{geo}), Sediment quality guidelines (SQGs) were calculated to determine the degree of metal pollution as well as to predict adverse biological effects in contaminated sediments of the investigated area. Person's correlation was applied to determine correlation matrix between metal concentrations, TOC%, $CaCo_3$% and grain size in surface sediments of Red Sea coast using Statistical Package for Social Science (SPSS) version 15.

RESULTS AND DISCUSSION

Sediments characterization

The water content is an important factor in controlling the early digenetic processes of sediments. It affects the rate of reactions particularly, the redox processes and the pH. The result of water content of the collected samples showed that, the Red Sea sediments have water in its pore ranged from 19.8 % at Marsa Alam site to 9.06 % at Bir Shalatin site (Table 2).

Water content reflects the ability of sediment to hold water molecules between its particles, which are mainly a function of particle size and mineral composition (Mehrem., 2002). Maxwell (1968) classified sediments based on carbonate content to high carbonate (>80%), impure carbonate (80 – 60%), transitional (60 – 40%), terrigenous (40 – 20%) and high terrigenous (<20%). The results of the present study showed that, sediments from Safaga North (Suma Bay) and Safaga Middle are characterized by high percentage of CaCO3 with value 91.50 % and 90 % respectively. The major sources of carbonate in these stations are weathering of limestone rock on the sea floor in addition to the detrital fragmental molluscal shells and skeletons (Madkour, 2004).Transitional carbonate was found in Hurghada (Sheraton Hotel) and Marsa Alam with value 50 % and 59.10 % respectively.

While El Hamarawein , Qusier Middle , Qusier South and Bir Shalatin sites belonging to terrigenous carbonate with value 40 % ,40 % , 30 % and 30 % respectively.

Table 2: Sediments characterization along the area of study.

stations	sand	silt	clay	TOC	WC	CaCO$_3$	sediment type
Re1	99.76	0.24	0	1.453	14.53	50	sand
Re2	99.62	0.38	0	1.483	14.93	91.5	sand
Re3	100	0	0	1.356	13.56	90	sand
Re4	100	0	0	1.571	15.71	40	sand
Re5	99.95	0.05	0	0.79	15.83	40	sand
Re6	99.39	0.61	0	1.77	17.68	30	sand
Re7	100	0	0	0.66	19.8	59.1	sand
Re8	83.23	14.58	2.19	0.91	9.06	30	sand

Total Organic Carbon (TOC) contents in the investigated sediments ranged from 1.77 % Qusier South to 0.66 % Marsa Alam area because of the presence of the Qusier port and activities there. It was noticed that the sediments in the study area are relatively low in TOC content. This reflects the high productivity of the water column that contributes organic detritus to bottom sediment. It also reflects, terrestrial input from land-based sources. The organic matter is mainly derived from the autolysis of dead cells or actively excreted by diverse organisms as benthic algae, copepods, sea urchins and planktonic species (Kenneth, 1988).

TOC is composed of light-weight materials, and there is a close connection between the presence of fine sediments and the contents of organic matter (El-Askary et al., 1988). In the present study, Coars sediments contained higher organic matter than finer ones this is because of the presence of alkaline feldspars which provide an environment conducive to increasing organic materials. distribution of organic matter in the sediments does not clearly depend on particle size but it predominantly depends on the conditions of the layer formation. Grain size analysis of the Egyptian Red Sea coastal sediments revealed the dominance of the sand fraction in all of the investigated samples (Table 2).The mean size of sediments ranged from -0.64 Φ at Safaga Middle to 2.14 Φ at Bir Shalatin (i.e. from very coarse sand to fine sand). Sorting changed from 0.48 at El Hamarawein to 1.91 at at Bir Shalatin (i.e. from well sorting to poorly sorting). Skewness varying from -0.62 at Qusier South to 0.49 at Safaga Middle (i.e. from strongly coarse skewed to strongly fine skewed). Kurtosis varying from 0.35 at Safaga Middle to 1.26 at El Hamarawein (i.e. from very platykurtic to leptokurtic). Samples characterized by the presence of quartz, feldspar, aragonite, epsomite, dolomite and calcite. Types of flux of clastic sediments, diversity of skeletal grains and the effectiveness of currents and wave actions produce this variation in the sediment characteristics.

The relative abundance of the fine sediments in some samples of the different areas is not only due to the abundance of terrigenous fine sediments but also due to the land filling and dredging in these areas. Our results are in agreement with (Madkour et al., 2008) and (Mansour et al. 2000).

Spatial distribution of total heavy metals

The total concentrations of Fe, Mn, Pb, Zn, Ni, Cr, Cd and Cu in surface sediments of the Egyptian Red sea coast are given in Table (3). Iron (Fe) concentrations were ranged between 207.96 to 276.9 mg /g stations 1 and 7 with average concentration 261.5163 mg /g. Fe concentrations of the shallow marine sediments in different transects along the Red Sea coast are relatively high indicating the terrigenous contamination (Mansour et al. 2000). The manganese content in the investigated sediments ranged between 32.94 mg /g to 153.6 mg /g at stations 1 and 7 with average concentration 102.175 mg /g. Iron was the most abundant metal measured followed by Mn and there are wide variations in the levels of the two metals among the sites of the study area. A significant correlation with $r = 0.81$ ($p = 0.05$) was found between the concentration of Fe and Mn reflecting the occurrence of these two metals is mainly controlled by natural processes such as coastal erosion deposition (Nasr et al., 1990). The concentrations of lead (Pb) in this study fluctuated from 11.16 to 29.66 mg /g with average value 20.8688 mg /g This may be attributed to anthropogenic activities in Bir Shalatin such as fishing boats, resulting in the release of large amount of Pb to the surrounding environment.

Zinc (Zn) concentration values recorded in this study ranged between 0.0193 to 207.6 μg/g with an average value 62.4350 μg/g. Zinc showed a common trend of increase towards El Hamarawein due to the disposal of municipal wastewaters at El Hamarawein harbour. The range concentrations (mg /g) of Ni, Cr and Cu were 6.033 to 45.53, 5.674 to 52.65 and 1.895 to 17.81 with average concentrations (mg /g) were 22.5029, 30.438 and 8.9381(mg /g).

Table (3): Metal concentrations (μg/g) in sediments of the Red Sea Coast and their risk assessment.

Stations	Fe	Mn	Pb	Zn	Ni	Cr	Cd	Cu	MPI
1	207.96	32.94	11.16	6.32	6.033	5.674	0.752	1.895	8.3472
2	261.03	61.84	21.25	13.49	11.51	15.86	1.649	3.462	16.2333
3	268.18	117.1	21.48	0.0211	16.47	27	1.11	7.991	9.3038
4	270.91	130.9	15.7	187.1	30.52	36.49	1.233	9.67	32.9314
5	270.87	124.7	24.99	207.6	24.06	42.16	2.274	11.88	38.4749
6	270.21	123.7	17.97	84.91	25.44	40.35	1.374	7.607	29.3270
7	276.9	153.6	24.74	0.0193	45.53	52.65	1.993	17.81	14.2788
8	266.07	72.62	29.66	0.0197	20.46	23.32	2.453	11.19	10.5010
Mean	261.52	102.18	20.87	62.44	22.50	30.44	1.61	8.94	19.9247
Aver. Shale[a]	4.7	805	20	95	68	90	0.30	45	-
TEL [a]*	n.a.	n.a.	30	124	18	52	0.7	19	-
*TET [d]	n.a.	n.a.	112	271	61	160	4.2	108	-

TEL [d:] Threshold Effect Level., MPI: Metal Pollution Index, TET [d] : Toxic Effect Threshold., a.Turekian and Wedepohl (1961)., d. MacDonald et al. (2000).

Heavy Meatals Contamination and Risk Assessment

Metal Pollution Index (MPI):

The overall metal contents at the investigated sites were compared using the metal pollution index (MPI) calculated according to (Usero et al. 1996 and Masoud et al. 2007b).to estimate the degree of pollution with the formula:

$$MPI = (M_{Fe} \times M_{Mn} \times M_{Zn} \times M_{Pb} \times M_{Cd} \times M_{Cu} \times M_{Cr} \times M_{Ni})^{1/8}$$

The MPI where values of M_{Fe}, M_{Mn}, M_{Pb}, M_{Zn}, M_{Ni}, M_{Cr}, M_{Cd} and M_{Cu} in the collected samples are summarized in Table (3). MPI indicated metal pollution load at the each site. The highest MPI values were measured at the site 5 (Qusier Middle) suffered from metal pollutions with values of 38.4749.This may be due to impact of human related activities such as tourism, Fishing, Boats.

Geoaccumulation Index (Igeo)

The Geoaccumulation Index (Igeo) was calculated to determine metals contamination in sediments of Egyptian Red Sea Coast. This expression was proposed by Müller (1979) in order to calculate metals concentration in sediments by comparing current concentrations with undisturbed or crustal sediment (control) levels.

Müller (1981) has classified Igeo in relation to contamination levels into seven classes, Unpolluted (Class 0, Igeo<0), unpolluted to moderately polluted (Class 1, 0< Igeo<1), moderately polluted (Class 2,1< Igeo<2), moderately to strongly polluted (Class 3, 2< Igeo<3), strongly polluted (Class 4, 3< Igeo<4), strongly to very strongly polluted (Class 5, 4< Igeo<5) and very strongly polluted (Class 6, Igeo>5). The highest grade reflecting a 100-fold enrichment above baseline values. The Geoaccumulation Index (Igeo) is expressed by the following pattern:

Igeo =log2 (Bn/1.5 * Cn)

Where Cn is the measured concentration of the sediment for metal (n), Bn is the geochemical background value of metal (n) and factor 1.5 is the possible variations of background data due to lithogenic impacts. The values of geoaccumulation index (Igeo) of investigated metals were shown in table (4). This suggests that the Fe concentrations were very strongly polluted (Igeo > 5) except station 1 was strongly to very strongly polluted (4<Igeo<5).

While the investigated sediments were Unpolluted (Igeo<0) at all stations by Mn, Pb, Ni, Cr and Cu this due to these metals were probably due to influxes of some non-lithogenic and natural sources into the surface sediments. All stations were Unpolluted (Igeo<0) at all stations except stations 4 and 5 which were unpolluted to moderately polluted (0< Igeo<1) by Zn. The concentration of Cd had wide variations recorded at many classes , unpolluted to moderately polluted (0< Igeo<1) at station 1, moderately polluted (1< Igeo<2) at stations 2, 3, 4 and 6 but at stations 5,7 and 8 was moderately to strongly polluted (2<Igeo<3).

Sediment Quality Guideline (SQGs)

In order to predict adverse biological effects in contaminated sediments, numerous sediment quality guidelines (SQGs) have been developed over the past decade (MacDonald et al., 2000). In the present study, we compared the concentration of investigated metals with SQG of threshold effect level (TEL), which described the threshold effect concentrations, and toxic effect threshold (TET), which described the probable effect concentrations (PEC) in order to assess the possible biological consequences of these metal concentrations in the surface sediments (Table 3). Threshold effect concentrations should be used to identify sediments that are unlikely to be adversely affected by sediment-associated contaminants, while the PECs should be used to identify sediments that are likely to be toxic to sediment-dwelling organisms (MacDonald et al., 2000). The obtained results of the present showed that the concentrations of Pb and Cu at all sediment samples are lower than TEL indicated that there are no harmful effects from this metal. Concentration of Zn in sediment samples at all stations lower than TEL except stations 4 (El Hamarawein) and 5 (Qusier Middle) with values 187.1 µg/g and 207.6 µg/g respectively, which higher than TEL and lower than TET. But concentration of Ni recorded values between TEL and TET except stations 1(Hurghada), 2 (Safaga North) and 3 (Safaga Middle) with values (6.033, 11.51 and 16.47 µg/g respectively) lower than TEL. This indicated that these stations were in potential risk. Cr was lower than TEL at all sediment stations except station 7(Marsa Alam) with value 52.65 µg/g higher than TEL and lower TET. On the other hand, the concentration of the Cd at all sediment stations has values between TEL (0.7 µg/g) and TET (4.2 µg/g).

Table (4): Geoaccumulation Index (Igeo) values in surface sediments of Red Sea coast.

Stations	Fe	Mn	Pb	Zn	Ni	Cr	Cd	Cu
1	4.90	-5.29	-1.43	-4.51	-4.09	-4.59	0.74	-5.17
2	5.23	-4.38	-0.50	-3.41	-3.16	-3.10	1.88	-4.30
3	5.27	-3.46	-0.48	-12.77	-2.64	-2.33	1.31	-3.09
4	5.28	-3.30	-0.94	0.39	-1.75	-1.89	1.46	-2.81
5	5.28	-3.37	-0.26	0.54	-2.09	-1.68	2.35	-2.51
6	5.28	-3.38	-0.74	-0.75	-2.01	-1.75	1.62	-3.16
7	5.31	-3.06	-0.28	-12.89	-1.17	-1.36	2.15	-1.93
8	5.26	-4.15	-0.02	-12.86	-2.33	-2.54	2.45	-2.60

Correlation Matrix

Pearson's correlation matrix was used to determine the relationships between Fe , Mn , Pb , Zn ,Ni , Cr, Cd , Cu , $CaCo_3$%, TOC% and grain-size of surface sediments (Table 5).The result indicated that most metals including Fe , Mn , Ni , Cr , Cd and Cu have positive significant association but there are negative correlation between Pb and Zn (r = - 0.11).

On the other hand, $CaCo_3$ recorded negative correlation with Fe , Mn , Pb , Zn ,Ni and Cr (r = -0.02, -0.13, -0.03, -0.46, -0.29 and -0.29 mg/g respectively). There are negative correlations between TOC and metals except Zn, Cd and Cu were significant positive correlation (r = 0.05, 0.76 and 0.74 mg/g respectively).There were negative correlation between grain size and TOC, CaCo3 ,Fe, Mn , Ni and Cr except Pb , Zn, Cu and Cd were significant positive correlation with grain size (r = 0.41 ,0.03, 0.01 and 0.61 mg/g respectively). The sediments of the study area were mainly sand, thus the total metal concentrations were not controlled by the mean size of each sediment sample. Significant correlations between most of these metals were probably due to influxes of some non-lithogenic and natural sources into the surface sediments.

Table (5): Pearson's correlation coefficient between heavy metals, $CaCo_3$%, TOC% and grain-size.

	Fe	Mn	Pb	Zn	Ni	Cr	Cd	Cu	CaCo₃%	TOC%	Grain Size
Fe	1										
Mn	0.81	1									
Pb	0.65	0.29	1								
Zn	0.30	0.43	-0.11	1							
Ni	0.69	0.87	0.36	0.24	1						
Cr	0.79	0.96	0.39	0.43	0.93	1					
Cd	0.57	0.26	0.92	0.12	0.42	0.44	1				
Cu	0.70	0.79	0.65	0.17	0.91	0.86	0.65	1			
CaCo₃%	-0.02	-0.13	-0.03	-0.46	-0.29	-0.29	-0.29	-0.28	1		
TOC%	-0.28	-0.27	-0.73	0.05	-0.46	-0.40	-0.76	-0.74	0.09	1	
Grain Size	-0.10	-0.35	0.41	0.03	-0.19	-0.1	0.61	-0.01	-0.50	-0.31	1

CONCLUSIONS

This study showed that heavy metal concentrations in the sediments along the Egyptian Red Sea coast were in the following decreasing order Fe > Mn > Zn > Cr > Ni > Pb > Cu > Cd.

The degree of surface sediment contamination was computed using Metal Pollution Index (MPI) and Geoaccumulation index (I geo). Metal Pollution Index showed that station 5 (Qusier Middle) suffered with metal pollutions. The Geoaccumulation Index (Igeo) exhibited the concentrations of Fe were very strongly polluted (Igeo>5) except station 1 was strongly to very strongly polluted (4< Igeo<5). While the investigated sediments were Unpolluted (Igeo<0) at all stations by Mn,Pb,Ni,Cr and Zn except stations 4 and 5. Sediment Quality

Guidelines (SQGs) showed that the concentration of Pb and Cu at all sediment samples are lower than TEL. On the other hand, the concentration of the Cd and Ni at all sediment stations have values between TEL and TET.The source of pollution of the investegated area may be due to impact from human related activities such as tourism, urbanization, dumping of solid waste, raw sewage near these sites were main sources of metal pollution in pore water sediment samples, alkaline feldspar which product reducing layer, hard skeleton of corals, marine paints, construction residuals and corrosion of steel constructions. the discharge of untreated sewage and industrial waste water, remnants of ships and fishing, accumulation of oil and sludge, pipeline of petroleum construction and industrial activities.

RECOMMENDATIONS

According to the results of the present study and based on the assessment of the state of pollution and evaluations of the hot spots, the following recommendations should be taken into consideration:

• Implementation of environmental laws NO. 4 (1994) and modified with NO. 9 (2009) for protection of the aquatic environments is strongly recommended.
• Industrial, domestic and agricultural drainages wastewater must be completely treated.
• Development of simple, cheap and fast techniques for the treatment of waste water is recommended.
• Continuous monitoring programs like the environmental information and monitoring program (EIMP) for Egyptian Red Sea coasts should be formulated and conducted to evaluate continuously the state of pollution.
• Since the Egyptian Red Sea Coasts had showed high concentrations of Cd. Restriction on the usage of Cd particularly in painting industry, is obligatory recommended.

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