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Environmental Assessment of Heavy Metal Pollution in Bottom Sediments of Aden Port, Yemen

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Abstract

The aim of the present study is investigate the distribution of heavy metals (Mn, Zn, Cu, Pb, Co, Cr and Ni) and to evaluate the contamination levels of the Aden Port sediments. In this study, the concentrations of heavy metals were measured, using Atomic Absorption Spectrophotometer (AAS) for 21 sediment samples collected at different stations in Aden Port during 2004.

The range and average concentrations measured in $\mu g\ g^{-1}$ were 138.23 - 658.87 (335.5) for Mn, 21.85 - 263.49 (128.59) for Zn, 8.06 - 111.00 (19.89) for Cu, 14.8 - 138.06 (77.28) for Pb, 13.8 - 33.64 (23.97) for Co, 17.00 - 233.93 (82.19) for Cr and 16.17 - 48.07 (34.54) for Ni. To evaluate the levels of sediment contaminations, the background values of the different elements were defined, depending on the international standards. In case of Pb, Cr, Zn and Co at most of sites, their concentrations in the sediments exceeded the background levels. However, the average values for Cu, Ni and Mn were less than the background values.

It is noted that the contamination factors in the investigated sediments were 0.31-3.39 for Zn, 0.4-5.55 for Cu, 0.74-6.9 for Pb, 1.06-2.59 for Co, 0.57-7.80 for Cr, 0.43-1.27 for Ni and < 1 for Mn. In general, the contamination factors of heavy metals in the present study could be arranged as following:

$$Pb > Cr > Zn \geq Co > Ni > Cu > Mn$$

The relatively high levels of Pb, Cr, Zn and Co in the sediments of the Aden Port are due to the discharges of untreated wastewater of desalination plant, electrical power station, refinery plant, textile industry, oil spills from the oil pipes, as well as domestic wastewater.

Keywords: Gulf of Aden, Trace metals, Contamination factor.

Introduction

Marine sediments can be sensitive indicators for monitoring contaminants in aquatic environments [1 and 2]. The bottom sediments serve as a reservoir for heavy metals and therefore deserve special consideration in the planning and design of aquatic pollution research studies. If a sufficiently large and stable sediment sink can be located and studied, it will allow the investigators to evaluate the geochemical changes over time and possibly to establish baseline levels against which current conditions can be compared and contrasted. Heavy metals, such as cadmium, mercury, lead, copper and zinc, are regarded as serious pollutants of aquatic ecosystems because of their environmental persistence, toxicity and ability to be incorporated into food chains [3].

The Gulf of Aden, due to its strategic position, links with the Indian Ocean through the Arabian Sea and the Red Sea. The seawaters from the Indian Ocean and the Red Sea have effects on the composition of the seawater in the Gulf of Aden. According to Boström *et al.*, [4], the heavy metals entering the marine environment are transported by prevailing currents. The main sources of pollution in Yemen are power and desalination plants, sewage treatment facilities, industrial facilities, port facilities, agricultural activities, coastal constructions, mining and quarrying activities [5].

The study by Szefer *et al.,* [6] in the Golf of Aden sediments showed significant regional variations concerning with both total and leachable metals. The total concentrations of Cd and Pb were greatest in the sediments of the Labour Island. Cu, Cr and Fe occurred in the highest levels in the Bandar Fuqum sediments, while maximum levels of Mn, Co and Ni were observed in the sediments of Sahel Abyan and Bandar Fuqum. The labile easily extractable species of metals, such as Cd and Pb, like their total concentrations and additionally Cu, Zn, Cr, Ni and Fe were found in the highest concentrations in the sediments of the Labour Island. The sediments from Sahel Abyan and Khawr Bir Ahmed were characterized by a maximum accumulation of bioavailability species of Co and Mn

EPC [7] studied heavy metal concentrations in the sediments and mollusks. They recommended a continuous monitoring programme for the Gulf of Aden / Arabian Sea beaches to ensure that the concentrations of heavy metals remain within the baseline levels established during the present survey. The aim of the present study is to investigate the distribution of heavy metals (Mn, Zn, Cu, Pb, Co, Cr and Ni) and to evaluate their levels in the Aden Port sediments.

Study area

Aden is located along the southern coast of Yemen. It lies at 12° 47' latitude and 44° 58' longitude (Fig. 1). It is a semi island and consists of rocks. Aden's population is 800000, with a land area of 35846 hectares. The convenient location of Aden's natural Port on the major sea route between the Far East and Europe has resulted in a rich history as a trading center. Aden's importance as shipping center peaked in early 1960s, when it was the fourth busiest port in the world [8].

Material and methods

Sample collection and analysis

Twenty one surficial sediment samples were collected from the area of Aden Port, using a grab sampler for the bottom sediments, while the beach samples were collected by hand (Fig 1). The samples were left to dry in Petri dishes at room temperature. The dry samples were divided into two portions. The first portion was used for grain size distribution by dry sieving and pipit analysis. The second portion was used for chemical analysis. Sub sample of the air dried sediments was homogenized with a pestle and mortar in order to normalize for variations in grain size distribution. The dried homogenized sediment samples were sieved through a 63µm screen and kept in clean plastic containers ready for analysis. The powder samples, less than 63µm were used for heavy metal analysis; 0.3g were digested with HNO_3, HF, Aqua regia (HNO_3 : HCl) and $HClO_4$ [9]. The samples were separately analyz ed by using Parkin Elmer Atomic Absorption Spectrophotometer (2830). The results of trace metals concentrations were determined on a dry weight basis in µg g^{-1}.

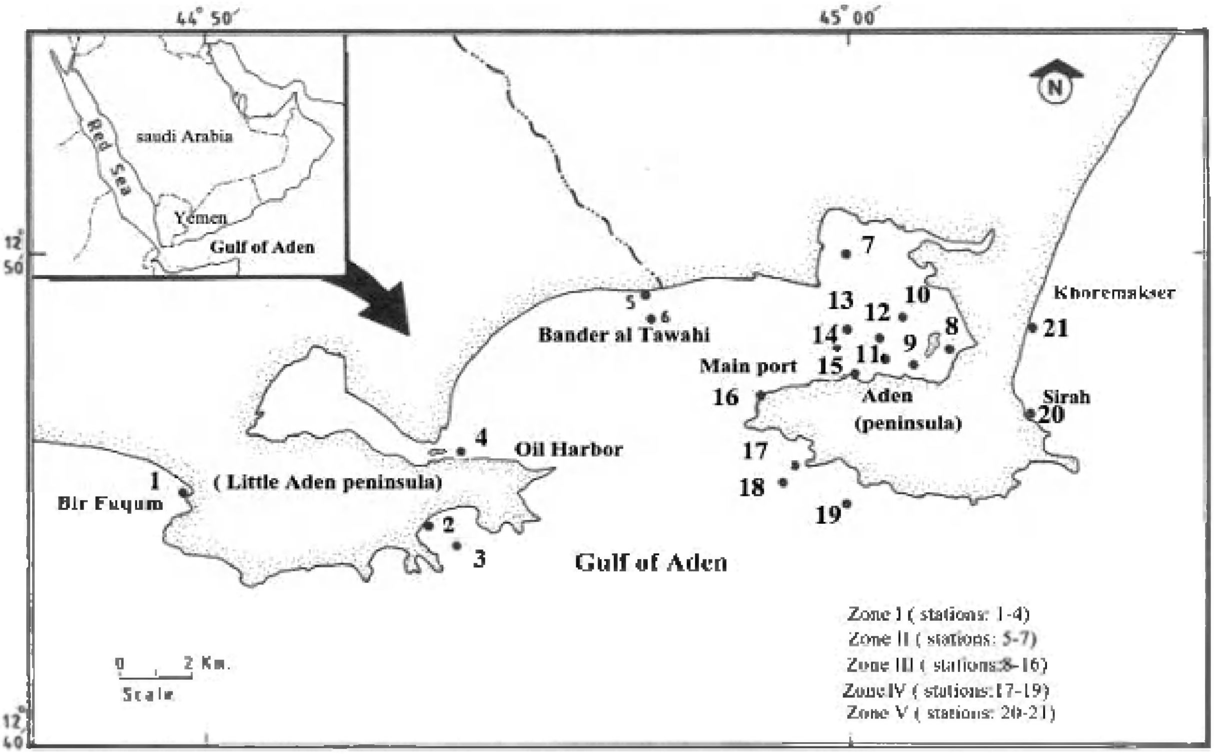

Figure1. Sampling Sites of Aden Port.

The total carbonate content was determined, using the method described by Dean [10]. The organic carbon of the investigated sediments was determined, using the wet oxidation method as mentioned by El-Wakeel and Riely [11].

The accuracy of the analytical procedure used was reputedly cheeked by analyzing reference samples (IAEA-365, Monaco).

The quality control samples represented 10% of the total analytical load. The duplicates, spikes and blanks were treated identically, using the same reagents for testing the precision, respectively. The percentage recovery for spiked samples ranged from 93 to 105%, while precision agreed within 5%.

Results

Sediment characterization

The particle size distribution, calcium carbonate and organic carbon of the study area are given in Table 1. The relative percentage of clay, silt and sand in the sediments of Aden Port were in the range (0-25%) for clay, (0.89-62.22 %) for silt and (14.44-86%) for sand. The concentrations of organic carbon (OC) were from of 0.32 to 2.97% and the average value was 0.86%. The maximum concentration was found at station15 (Fishing Harbor) and the minimum was at station17 (Cornish Q. Mohur). The concentration of carbonate in the sediment samples ranged from 5.67 to 78.67% at station 21 (Sabel Abyan) and station 19 (Dunafah), respectively, with a mean value of 31.87%.

Table 1. Characterization and heavy metals concentration (µg g -1) in the sediments of Aden Port, Yemen.

Zones	Stations	Grain size analysis			$CaCO_3$	OC	Mn	Zn	Cu	Pb	Co	Cr	Ni
		Sand	Silt	Clay									
		-	%	-	%	%							
I	1	73	27	-	29.67	0.69	239.16	62.58	22.76	96.71	31.66	141.37	41.39
	2	86	3	11	58.00	0.76	391.47	21.85	13.3	78.15	25.54	40.69	23.78
	3	77	13	10	18.83	1.19	138.41	51.16	18.08	76.44	28.03	119.37	38.53
	4	82	7	11	20.33	0.84	407.25	44.49	15.49	75.76	27.66	168.86	37.86
II	5	86	1	13	10.17	0.61	421.45	64.72	20.23	64.03	33.64	233.93	47.77
	6	73	8	19	49.67	0.79	658.87	30.28	14.48	67.33	27.52	101.69	38.11
	7	77	13	10	18.33	1.21	304.87	52.19	21.96	88.74	29.52	129.47	41.62
III	8	77	1	22	8.67	0.77	416.9	188.87	9.52	46.26	16.88	46.47	24.93
	9	62	18	20	50.00	1.13	438.94	160.78	17.76	87.03	22.84	56.79	33.28
	10	53	21	26	31.83	0.88	277.11	164.21	18.39	60.27	16.92	56.28	28.95
	11	72	17	11	33.33	0.83	374.15	155.45	13.53	42.27	17.2	53.5	31.76
	12	79	11	10	32.00	0.85	329.93	238.7	15.27	98.65	28.39	59.8	37.86
	13	62	21	17	38.33	0.79	258.55	263.49	14.81	121.43	25.15	58.77	35.94
	14	52	36	12	39.83	1.32	293.45	171.46	20.4	43.30	19.82	66.49	39.24
	15	15	62	23	25.00	2.97	415.49	233.21	111.00	136.24	23.92	65.27	48.07
	16	83	5	12	6.33	0.33	479.15	181.35	9.97	64.71	15.16	46.96	26.13
IV	17	84	1	15	51.33	0.32	418.6	129.09	13.68	94.55	27.92	153.58	39.53
	18	71	6	23	50.33	0.42	573.67	89.01	14.27	97.51	26.58	37.06	33.13
	19	74	1	25	78.67	0.38	239.89	237.57	12.84	138.06	22.68	30.23	29.52
V	20	82	2	16	13.00	0.54	145.07	26.13	8.09	14.80	13.75	17	16.17
	21	79	2	19	5.67	0.46	138.23	133.82	11.91	30.77	22.63	42.53	31.9
Background level							790	70	20	20	13	30	38

Heavy metal distribution

The regional variations of total concentration of heavy metals (Mn, Zn, Cu, Pb, Co, Cr and Ni) in the sediments of the Aden Port are given in Table 1. The range and average concentrations (µg g^{-1}) were 138.23 - 658.87 (335.5) for Mn, 21.85 - 263.49 (128.59) for Zn, 8.06 - 111.00 (19.89) for Cu, 14.8 - 138.06 (77.28) for Pb, 13.8 - 33.64 (23.97) for Co, 17.00 - 233.93 (82.19) for Cr and 16.17 - 48.07 (34.54) for Ni. The high values of Mn were found at all stations, except at stations 3, 20 and 21, which revealed the lowest content of Mn ranging from 138.2 to 145.1 µg g^{-1}. The greater amounts of Zn were recorded at four stations; station 12 (239.0 µg g^{-1}), station 13 (264.0 µg g^{-1}), station 15 (233.2 µg g^{-1}) and station 19 (238.0 µg g^{-1}). The highest levels of Pb were observed at most of stations, (more then 60 µg g^{-1}), while stations 8, 11, 14, 20, and 21 revealed lower contents of Pb, ranging from 14.8 µg g^{-1} at station 20 to 46.3 µg g^{-1} at station 8. The highest levels of Co, Cr and Ni were detected at station 5 (33.6 µg g^{-1} for Co, 233.9 µg g^{-1} for Cr and 47.8 µg g^{-1} for Ni). On the other hand, the lowest concentrations of these metals (13.8, 17.0 and 16.2 µg g^{-1} for Co, Cr and Ni, respectively.) were recorded at station 20. The high concentration of Zn (238 µg g^{-1}) was found at station 19. This is related to the high $CaCO_3$ content, which reached 79%, while the highest level of Cu was detected at station 15 (Fishing Harbor). Its content reached 111.0 µg g^{-1};

increased five fold than reported in the other sites. This station was characterized by a low content of organic carbon (about 3%).

Table 2. Contamination factors (Cf) of the sediments in Aden Port, Yemen during 2004

Zones	Stations	Contamination factors (Cf)						
		Mn	Zn	Cu	Pb	Co	Cr	Ni
I	1	0.30	0.89	1.14	4.84	2.44	4.71	1.09
	2	0.50	0.31	0.67	3.91	1.96	1.36	0.63
	3	0.18	0.73	0.90	3.82	2.16	3.98	1.01
	4	0.52	0.64	0.77	3.79	2.13	5.63	1.00
II	5	0.53	0.92	1.01	3.20	2.59	7.80	1.26
	6	0.83	0.43	0.72	3.37	2.12	3.39	1.00
	7	0.39	0.75	1.10	4.44	2.27	4.32	1.10
III	8	0.53	2.70	0.48	2.31	1.3	1.55	0.66
	9	0.56	2.30	0.89	4.35	1.76	1.89	0.88
	10	0.35	2.35	0.92	3.01	1.3	1.88	0.76
	11	0.47	2.22	0.68	2.11	1.32	1.78	0.84
	12	0.42	3.41	0.76	4.93	2.18	1.99	1.00
	13	0.33	3.76	0.74	6.07	1.93	1.96	0.95
	14	0.37	2.45	1.02	2.17	1.52	2.22	1.03
	15	0.53	3.33	5.55	6.81	1.84	2.18	1.27
	16	0.61	2.59	0.50	3.24	1.17	1.57	0.69
IV	17	0.53	1.84	0.68	4.73	2.15	5.12	1.04
	18	0.73	1.27	0.71	4.88	2.04	1.24	0.87
	19	0.30	3.39	0.64	6.90	1.74	1.01	0.78
V	20	0.18	0.37	0.40	0.74	1.06	0.57	0.43
	21	0.17	1.91	0.60	1.54	1.74	1.42	0.84
Reference "unpolluted"		1.00	1.00	1.00	1.00	1.00	1.00	1.00

$Cf < 1$ law contamination factors; $1 \leq Cf < 3$ moderate contamination factors
$3 \leq Cf < 6$ considerable contamination factors and $Cf \geq 6$ very high contamination factors.

Table 3. The Enrichment Factors (EF) for each metal in the five zones

Zones	Mn	Cu	Zn	Pb	Cr	Ni	Co
I	8.51	10.44	17.18	177.76	31.21	12.69	40.29
II	12.88	10.98	18.22	154.45	40.21	14.85	41.69
III	10.09	14.72	73.40	163.66	14.90	11.91	28.69
IV	11.86	8.36	61.06	246.04	19.62	12.50	37.34
V	4.42	6.44	31.55	52.02	8.43	9.17	27.66

Discussion

Grain size analysis of the sediments illustrates that the study area was covered with coarser sediments, which had higher sand content at most of stations, except at station 15 (zone III), which contained more fine material and relatively high content of organic carbon (around 3%). In most samples, the value of organic was lower than 1% in the sediments. The carbonate content was variable, ranging from 5.7 to 78.7% and most of the samples contained < 40%.

The highest mean values of Zn (195.3 µg g^{-1}) and of Cu (25.6µg g^{-1}) were recorded in the main port (Zone III) and were higher than those reported in the other zones; increased four times than the mean values recorded in zones I and II. This may be attributed to the various sources, i.c. sewage outfall from Hugaif through the pipelines extending into the sea, as well as the industrial effluents, such as fishery refrigerator fishing and fiberglass boat factory. The anthropogenic input was an important source for these elements, through the extensive use of the antifouling paints by shipping activities [12].

Föstner and Wittman [13] showed that the distribution of heavy metals in marine deposits was influenced by sediment texture, clay content, organic carbon, iron hydrous oxides and carbonates. In the present study, the high levels of heavy metals were recorded at station 15, which was characterized by relatively high content of organic carbon (3%) and fine material (85% of silt and clay).

The high concentration of lead at stations 13, 15 and 19 (ranging from 121-138 µg g^{-1}) might be related to the high amounts of calcium carbonate content (ranging from 25 to 79%). The visual logging indicates that most of carbonate was comprised from fragmentary mollusks. A significant concentration of Pb was recorded at most of stations, except at stations 20 and 21 (means of 22.8 µg g^{-1}). The highest content of Pb was reported in zone IV (110 µg g^{-1}), while zones I, II and III had content around 80 µg g^{-1}. The high levels of Pb content was probably due to several factors, which were domestic and industrial effluents, municipal runoff and atmospheric deposition, as well as the contribution of leaded fuel in the out board boat engines and automobiles and car batteries. Kishe and Machiwa [3] showed that the elevated levels of trace metals in the aquatic systems were related to the effluents from the urban areas.

The highest mean concentration of Cr (155 µg g^{-1}), Co (30.2 µg g^{-1}), Ni (42.5 µg g^{-1}) and Mn (461.7 µg g^{-1}) were found in Al Hiswah and Caltix (Zone II).This might be related to the industrial wastes from the Power and Desalination Plants and the run-off from Lahij governorate, which included disposal of liquid effluents and terrestrial runoff, as well as the atmospheric deposition [1,14]. Cr and Ni were enriched mainly by industrial inputs [15]. Also, the relationship between Pb and carbonate (r = 0.53) is clearly evident in Fig. 2B, where the variations in Pb concentration were linked with the variations in carbonate content.

The comparison between the present concentrations with those reported in the literature concluded that the concentrations observed in the Aden Port were lower or higher than those recorded in the other studies. The average concentration of Zn obtained in the present study was much higher than those reported for the sediments of Al-Hodiedah [16] and Hadramout coast, Yemen [17] and Red Sea in front of Egypt [18]. The level of Copper was similar to those reported by Szefer *et al.,* [6] and lower compared with those found by Al-Kahali, *et al.,* [17]. The average concentration of Pb obtained in the present studied was much higher than those reported by EPC [7]; Hadramout Yemen [17] and Dead Sea coast in Jordan [19] and within the range recorded by Szefer *et al.,* [6]. The average concentration of Co in the present study were in the range recorded by Szefer *et al.,* [6] and EPC, [7] in the Gulf of Aden and higher than the Hadramout coast in the Arabian sea, Yemen [17]. The data of Cr content was much higher than those reported by EPC [7], Al-Adrise [16], Al-Kahali, *et al.,* [17] and the concentration of Ni was similar with those reported in other studies [5 and 15].

Evaluation of sediment pollution

It is difficult to make an overall assessment of the degree of metal contamination in estuarine and marine sediments [20]. This is a consequence of variations in analytical procedures between studies and the presence of an unknown natural background in the sediments. In the present study, three approaches were employed to evaluate the sediment pollution; comparison with the background value, sediment quality guidelines and contamination factors (Cf).

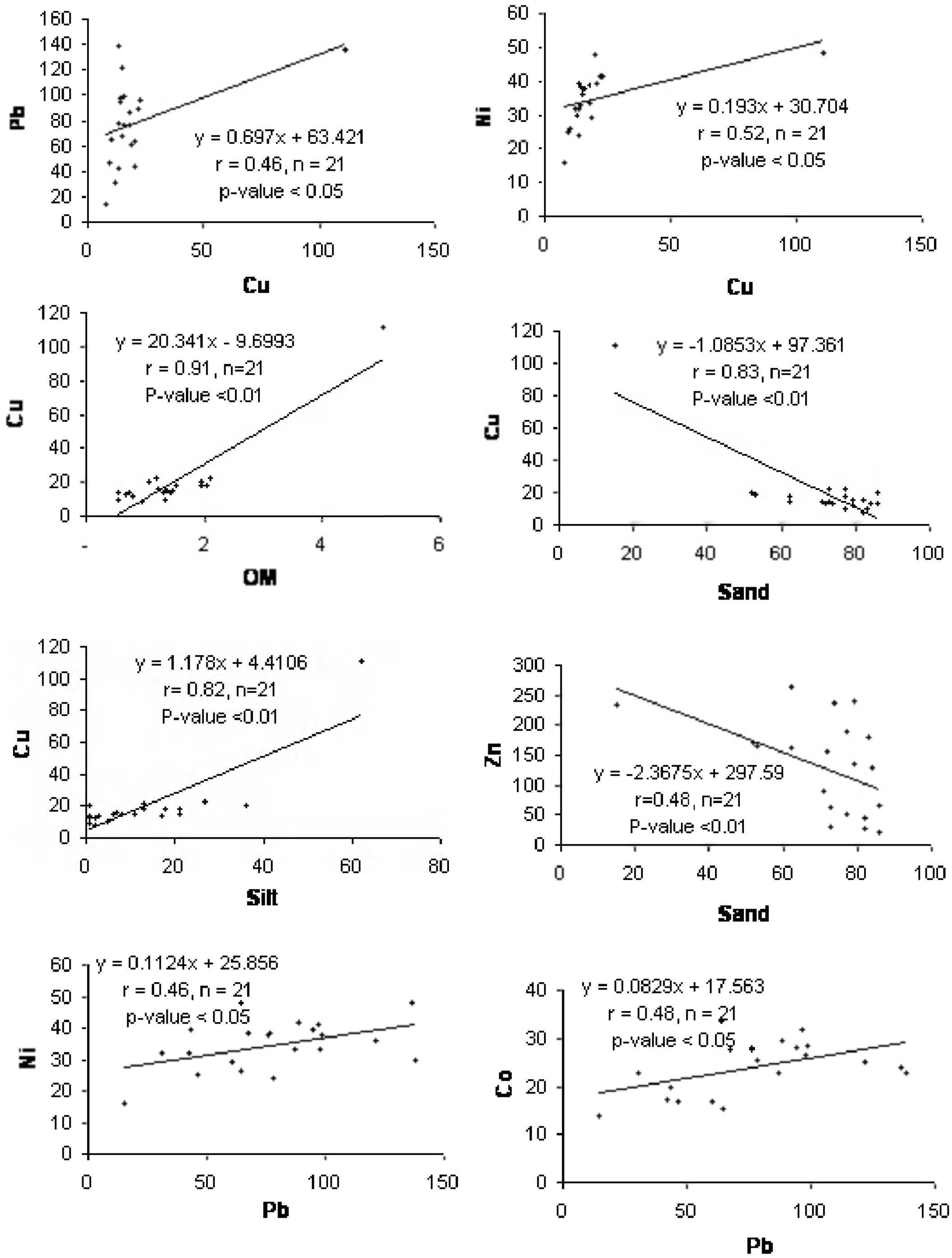

Figure 2A. Relationships between pairs of variables; Cu and Pb, Cu and Ni, Cu and OM, Cu and Sand, Cu and Silt, Zn and Sand, Pb and Ni, Pb and Co.

The background values of the different elements were defined, depending on the international standards: Zn, Cu and Pb [21], Co and Ni [22], Cr [21, 23] and Mn [21]. The concentrations of Pb, Cr and Co were greater than the elemental background concentrations at all stations, except at station 20. At zones III and IV, the concentrations of Zn exceeded the background value from 2 to 3 folds and decreased at zone I and II. The concentrations of Cu at all stations were equivalent or smaller than the background value, except at station 15 which revealed a higher amount of Cu. Ni was equivalent or smaller than the background value at most of stations, except at stations 5 (47.8 $\mu g\ g^{-1}$) and station 15 (48.1 $\mu g\ g^{-1}$). The concentrations of Mn in the sediments of the investigated area were less than the background value.

The level of contamination expressed by the contamination factor (Cf) [1, 24, 25] and it was calculated as follows:

Cf = (metal content in the sediment) / (metal content in natural reference sediment)

The contamination factor was classified into four groups [1, 24, 25]; $Cf < 1$ refers to the low contamination factor $1 \leq Cf < 3$ refers to the moderate contamination factor, $3 \leq Cf < 6$ refers to the considerable contamination factor and $Cf \geq 6$ refers to the very high contamination factor. The values of contamination factor (Cf) are shown in Table 2. Very high contamination was recorded at stations 13, 15 and 19 for Pb and Cr at station 5 and considerable contamination for zones I and II, while between considerable and moderate contamination was found at the other stations, except at station 20. The values of contamination factor showed low levels for Cu at all stations, except at station 15, which revealed considerable contamination factor. Low contamination factor was recorded at all stations for Mn. This contamination factor was between low and moderate for Ni, between low and very high for Cr and between low and considerable for Zn.

Enrichment factors (EF)

The enrichment factors (EF) were evaluated by computing the ratios of metal concentrations to Fe concentration [26]. The enrichment factor for each metal was calculated from the formula stated by Rubio *et al.,* [20], Saad *et al.,* [27] and Rule [28].

$$EF = \frac{(\text{Metal / Fe})\ \text{Sample}}{(\text{Metal / Fe})\ \text{Crust}}$$

The enrichment factors (EF's) of trace metals in the Aden Port sediments at the five zones (Table 3) revealed that they were highly enriched in Pb, especially at zone IV. The EF's varied in the range 52-246 for Pb, 17-73 for Zn, 27-42 for Co, 9-15 for Ni, 8-40 for Cr, 6-15 for Cu and from 4 to 13 for Mn. In general, the lowest enrichment factor for all trace metals in the sediment samples were recorded at zone V, while the EF for zone II was highly with Mn, Cr, Ni and Co. The sediments of zone III was enriched with Cu and Zn, as well as Pb and Co. These high levels of the enrichment factors might be related to the anthropogenic effect from several sources.

Statistical Analysis

The statistical analysis as shown in Fig. 2A illustrates a significant relationship ($p<0.01$ or $p <0.05$) between Cu-Pb, Cu-Ni, Cu-OM, Cu-Silt, Pb-Ni, Pb-Co (positive) and Cu-Sand, Zn-Sand (negative). Fig. 2B revealed presence of significant relationship ($p < 0.01$ or $p < 0.05$) between Pb-CO_3, Co-Ni, Co-Cr, Cr-Ni, Ni-OM (positive), except Cr-Clay (negative). It is evident that the relationships between heavy metals and sediment properties are linked with variations in the particle size and carbonate concentration. Copper revealed significant correlation with organic matter (r=0.9) and fine material, silt fraction (0.62) and reverse correlation was observed with the coarsest fraction, (r=0.48). The relationships between pairs of metals were observed (Fig. 2A and

2B). This could suggest that the association of pair elements have the same source of contamination.

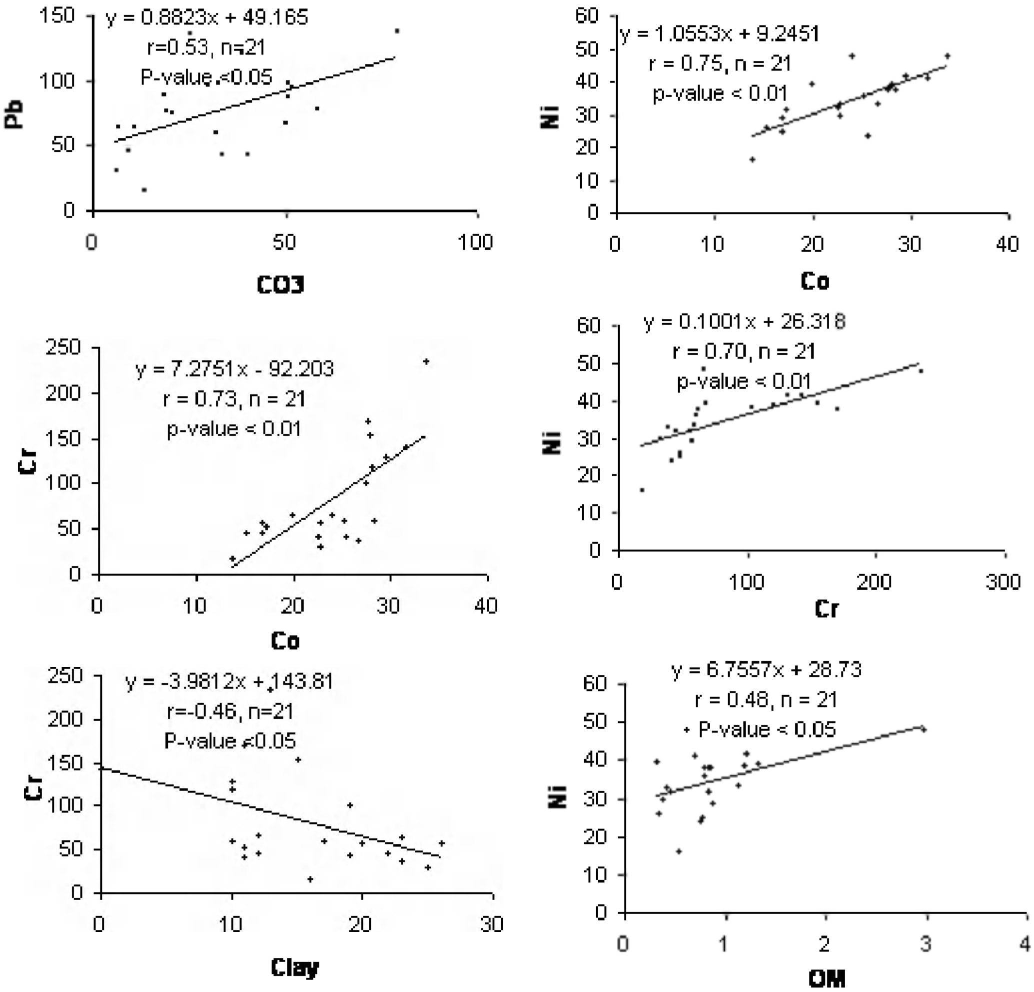

Figure 2B. Relationships between pairs of variables; Pb and CO3, Co and Ni, Co and Cr, Cr and Ni, Cr and Clay, Ni and OM.

Conclusion

In general the spatial distribution of heavy metals in the sediments of Aden Port was controlled by the association of heavy metals with carbonate, fine particles (silt-clay) and organic matter content. The present study reflects the impact of anthropogenic input as a source for heavy metals to this port.

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