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Assessment the Leachable Heavy Metals and Ecological Risk in the Surface Sediments inside the Red Sea Ports of Egypt

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Abstract The concentrations and distributions of the leachable heavy metals (Co, Cu, Zn, Ni, Cd, Mn, Pb and Fe) were investigated in the fine sediment fractions (Ø3, Ø4 and Ø5) collected from the Egyptian Red Sea Harbors at Hurghada, Safaga and Qusier. The three fractions were the essential heavy metal carriers and were formed the main constituent of the marine sediments with percentages exceed 50% at the most of studied stations. The accumulation sequence of the metal carriers at Hurghada was Ø5 >Ø4> Ø3, however at Safaga and Qusier, it was Ø5 >Ø3> Ø4. Fe and Mn showed the highest values at Safaga (7483 and 306.3 µg/g, respectively) due to the high terrigenous inputs from the different shipment operations and the wastewater effluents. The highest values of Zn, Cu, Ni and Pb were recorded in the marine area off Hurghada ports (330.38, 298.40, 91.4 and 101.02 µg/g, respectively), which attributed to the coastal based activities at Hurghada shipyard and fishing berth. Meanwhile the highest levels of Co and Cd were observed at the old port of Qusier (5.85 and 4.19 µg/g, respectively). The correlation coefficient and principal component analysis (PCA) indicated the anthropogenic sources of Cu, Zn and Pb in front of Hurghada ports. Based on the SQGs, the concentrations of Co, Fe, Mn, Zn and Pb were below the lowest effect level (LEL) with limited severity for Cd, Cu and Ni at Hurghada shipyard but lower than the sever effect level (SEL). The principal component analysis (PCA) showed correlations between Fe, Mn and partially Cd as well as the strong positive correlations for Cu with Zn and Pb suggesting common source of contamination that is likely originated from the terrestrial materials associated with the shipping of ores and coastal activities. According to the Enrichment factor (EF) and the geo-accumulation factor (Igeo), Cu and Pb were the highest enriched elements due to anthropogenic contamination; consequently the studied ports were classified as moderately to highly contaminated by Cu and Pb at Hurghada.

Keywords Leachable heavy metals; Sediment fractions; PCA; Ecological risk; Red Sea ports

Introduction

The quantity and types of contaminants have rapidly increased in the marine environment during the last decades. Most near-shore marine habitats worldwide have become under increasing pressure (McClanhan et al., 2000). Pollution with heavy metals is one of the global environmental problems because of their toxicity, environmental persistence, non-biodegradable nature and incorporation into food chains (Förstner and Wittman, 1983; Gargouri et al., 2011). For that reason, a lot of effort is expanded to assess their availability, toxicity and ecological risk to marine organisms (Salem et al., 2014).

The Red Sea belongs to the category of land-locked seas that have very restricted water exchange (Idris et al., 2007); this partial isolation condition makes this body of water vulnerable to impact of land-based pollution. For long time, the Red Sea environment was regarded relatively unpolluted (Hanna and Muir, 1990). In recent years, heavy metals contamination was observed in many locations associated with oil production, wastewater discharge, building and landfilling along the coastline, navigation and shipping operations (Madkour and Dar, 2007; El-Metwally et al., 2017). However, the most significant source of anthropogenic pollution in marine system usually related to ports due to dredging, repairing, raw materials shipping, loading, effluent discharge, and other land based and maritime activities (Galkus et al., 2012).

Sediments are the principal sink of heavy metals in the aquatic coastal system, but under certain chemical and physical conditions metals can readily release into the water column again and become a source of metal to marine organisms (Dickinson et al., 1996; Dar, 2014). Therefore, the distribution of heavy metals in the sediments can serve as environmental indicator of current and past condition of the pollutant discharged in the surface marine sediments (Förstner and Salomons, 1980; Abu-Hilal and Badran, 1990). Because of metals are not equally distributed in the different sediment fractions, heavy metals were widely determined in the finer grains in order to minimize the variation effect (Salomons and Förstner, 1984). Finer particles usually contain high concentrations of heavy metals due to possessing large surface area and high association with clay and organic matter (Irvine and Birch, 1998; Yu et al., 2012). It is important to evaluate the ecological risk of heavy metals in fine sediments, not only because of their high content of heavy metals but also because they can readily move by the wave winnowing and currents to adjacent places which may host biologically sensitive communities (Dar et al., 2016a). Assessment of heavy metal levels only cannot describe the ecological risk to marine environment or evaluate the contribution ratio from terrestrial and anthropogenic sources (Zhu et al., 2012). Therefore, some methods were adopted to evaluate the biological and ecological risk of heavy metals in marine sediments such as comparing to sediments quality guidelines (SQGs), and applying several quantitative geochemical indices like pollution load index, enrichment factor and geo-accumulation Index (El Zrelli et al., 2015).

Maritime activities in the Egyptian ports of the Red Sea proper were concentrated in three main locations at Hurghada, Safaga and Qusier. Three main activities involved in the port area of Hurghada; passengers, fishing berth and tourist marina. These activities in addition to effluents of desalination plant and sewage seepage as well as Hurghada shipyard are the main sources of pollution. Safaga has the largest port in the Egyptian Red Sea proper; it plays an important role in the Egyptian international trade. It is characterized by intense shipping traffic with frequent loading and dumping operations. The port involves passenger terminal, terminals for cargo, bauxite, coal, grains, quartz and orthoclase shipment. Qusier has the oldest phosphate port in the Red Sea. The marine area surrounding the ports was frequently affected by wastewater seepage and anthropogenic terrestrial runoff. During the last years, a number of studies have assessed the levels of heavy metals in the Red Sea coastal sediments (e.g., Madkour, 2005; Salem et al., 2014, El-Metwally, 2015; Dar et al., 2016a; 2016b; El-Metwally et al., 2017) and harbours (Madkour, 2004; Madkour and Dar, 2007; Mansour et al., 2011; 2013). However, regardless the rapid expansion in ports and harbour areas of the Red Sea, but the ecological risk of different port facilities on sediments quality has not been evaluated before.

The objective of the present study is to determine the contribution of maritime and land-based activities at the main ports of the Egyptian Red Sea on the levels and distribution of heavy metals in the fine sediments in order to evaluate their environmental risk in the marine habitats.

1 Materials and Methods

1.1 Study areas

Thirty three sampling sites were selected in front of the main ports along the Egyptian Red Sea proper (Figure 1). The sediments samples were collected using a Van Veen grab sampler and small boat in the marine area off the main ports at Hurghada, Safaga and Qusier with depth variation between 5 and 17 m. These sampling sites were grouped in eleven stations for covering the different maritime activities (passengers, shipyard, tourist activities, gains shipment, phosphate shipment, bauxite shipment, coal shipment, fishing, quartz and orthoclase shipments). At Hurghada Port, 4 stations were selected covering the whole marine area off the port; passenger wharf (sites H1 –H3), shipyard (H4), fishing berth (H5-H8) and the touristic marina (H9-H11). Five stations at Safaga Port; grains platform (S1-S2), passenger wharf (S3-S5), bauxite platform (S6-S7), coal platform (S8-S9) and the platform of quartz and orthoclase represented by the stations (S10, S11 and S12). At Qusier Port, station I (Q1-Q4) was located in marine area off of the old port; meanwhile station II (Q5-Q10) was covered the fishing basin.

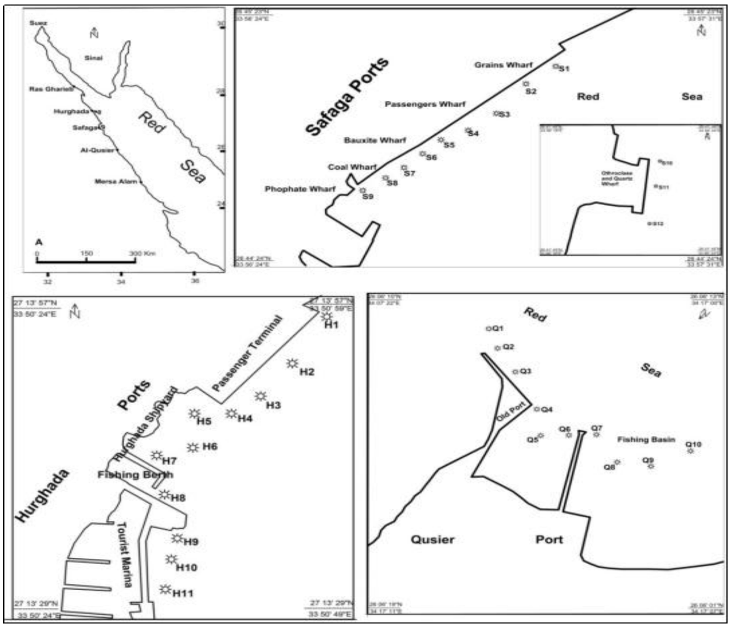

Figure 1 Location map and the sampling stations of the studied ports

1.2 Laboratory analyses

1.2.1 Granulometric analysis

The collected samples were packed in labeled polyethylene bags and immediately transported in ice-cooled box to the laboratory. At the laboratory, these samples were air-dried, disaggregated, and about 100 g of the pre-dried samples were sieved each one phi (Ø) interval to obtain textural properties according to Folk (1974). Seven fractions were obtained: gravel (Ø−1 > 2.00 mm), very coarse sand (Ø0 = 2.00 to 1.00 mm), coarse sand (Ø1 = 1.00 to 0.50 mm), medium sand (Ø2 = 0.50 to 0.25 mm), fine sand (Ø3 = 0.250 to 0.125 mm), very fine sand (Ø4 = 0.125 to 0.063 mm) and mud “silt & clay” (Ø5 < 0.063 mm).

1.2.2 Geochemical analysis

For analysis of carbonate content and total organic matter, about 10 g of pre-dried bulk sediments were completely grinded using agate mortar. The carbonate contents were determined by the method described by Gross (1971) depending on acid-treatment weight-loss. One gram of each powdered sample was treated with 1N HCl acid then the remaining insoluble residue was washed, dried at 60 ℃ in oven, reweighted and the weight loss was converted into carbonate percentage. The total organic matter (TOM) was determined by the ignition loss of one gram from powdered samples at 550 ℃ (Dean, 1974). The difference in weight was calculated as organic matter percentage.

The leachable forms of heavy metals were determined in the finest fractions of the collected samples (Ø3, Ø4 and Ø5). From each fraction, 0.5 g was digested with a mixture of concentrated HNO3 and HClO3 (3:1) then evaporated to near dryness. The digested samples were filtered and diluted with de-ionized water (Chester et al., 1994). This method is adopted to determine the leachable heavy metals (oxides, hydroxides, carbonates and sulphides). The concentrations of: Co, Cu, Zn, Ni, Cd, Mn, Fe and Pb were determined using flame Atomic Absorption Spectrophotometer (AAS, GBC-932) at the National Institute of Oceanography and Fisheries (NIOF), Hurghada, Egypt. To insure maximum accuracy, precision of the methods was confirmed by analysis of replicate measurements for each metal in the sediments sample. The obtained results showed a satisfied precision of 3.9 –16.1. In addition, the AAS was adjusted to provide mean value of triplicate measurements of each metal and the results were expressed in µg/g. the zero level of the AAS were adjusted by blanks and the quality control tools were adopted to avoid possible contamination, meanwhile, the chemical reagents are of high analytical grade and all of glassware were washed with diluted acid and later rinsed with double-distilled water before use.

1.3 Sediment quality guidelines and Ecological Risk indices

Biological adverse effects were evaluated by comparing levels of the measured heavy metals with the numerical sediment quality guidelines (SQG's) proposed by Persuad et al. (1992). Two levels of risk were estimated; the lowest effect level (LEL) and the sever effect level (SEL). The contamination status was assessed by comparing the results of present study with available data given in previous literatures for leachable heavy metals in sediments from various ports worldwide.

Several indices have been applied to assess the ecological risk of heavy metals in various aquatic environments (Fujita et al., 2014) including; metal pollution load index (MPI) (Tomlinson et al. 1980), enrichment factor (EF) (Salomons and Förstner, 1984), and the geo-chemical index (Igeo) (Müller, 1979). The interpretation of these indices is depending on the comparison with a background levels. Rubio et al. (2000) have concluded that the use of regional background values gives more appropriate results than the global background values, consequently the mean results of Hanna (1992) for the sediments collected from the Red Sea during 1943 were used to calculate the background values.

1.4 Statistical analysis

All statistics were performed using Statistica 10 and Wingraph Prism 7. One-way analysis of variance (ANOVA) was applied to test significant variation in metal distribution between sites, and comparing levels of metals between different fractions. To explore potential association between heavy metals and relationship among variables, Pearson's correlation matrix and principal component analysis (PCA) were used.

2 Results and Discussions

2.1 Sediments characteristics

It has been reported that many characteristics of the marine sediments such as texture, organic matter and carbonate content may influence the distribution of metals in sediments (Chen et al., 2007). The granulometry of marine sediments at the different ports is illustrated in Figure 2. It can be noticed that the texture of all studied stations was sandy. At Hurghada, the total of the finest fractions (Ø3, Ø4 and Ø5) was varied between 36.26% in fishing port and 83.47% in front of passenger port; at Safaga, the fine fractions percentage was fluctuated between 46.32% in passenger wharf and 73.17% in the marine area off bauxite wharf with significant for Ø4; meanwhile at the old port of Qusier, these percentage was varied from 59.83% to 64.69% with nearly equal occurrences for Ø3 and Ø4. The recorded high percentages of the fine fractions Ø3, Ø4 and Ø5 at the different ports indicating to different sources of depositions mostly from the maritime activities, terrestrial runoff, phosphate shipments and the nearby land based activities. Dar et al. (2016b) found the average percentages of finest sediments (Ø3, Ø4 and Ø5) between 24.21% and 88.88 % with strong occurrence for Ø4 at Hurghada, and from 37.47% to 48.14% at Safaga with strong occurrence for Ø3. They indicated that the high occurrence of fine sediments at Hurghada and Safaga to the deposition under calm conditions due to the natural protections by the shallow coral terraces and the frontal islands. Madkour and Dar (2007) reported that Ø4 has significant occurrence at the tidal flat area off Hurghada Harbour. Mansour et al. (2013) attributed the high fine sediment contents in front of Hurghada to the terrigenous inputs, landfilling and dredging operations.

2.2 Geochemical characteristics

2.2.1 Carbonates and total organic matter (TOM)

Table 1 shows the measured carbonate and total organic matter (TOM) percentages at the different stations. The studied stations at Hurghada ports recorded the highest percentages of carbonates which was varied between 51.62 and 76.65% indicating that the biogenic source materials constituted significant portion of the sea sediments; Safaga wharfs recorded significant variations in the carbonate percentages between less than 10% at grains and passenger wharfs and 59.52% at quartz and orthoclase wharf; while the carbonate percentage was less than 50% at the old port of Qusier. These data illustrated that the biological productions in the marine area off Hurghada and Qusier have strong contribution in the high carbonate percentage; while the effects of maritime activities and terrestrial runoff of terrigenous materials were the main reasons in carbonate percentage declining at Safaga ports. Significant variations were observed in TOM contents at Hurghada and safaga ports due to the local effects of the anthropogenic effluents. At Hurghada it was varied between 2.48% in marina and 8.78% in the fishing port, while at Safaga varied between 1.91% in grains wharf and 10.09% at coal wharf. On the other hand, the old port of Qusier recorded insignificant variation in TOM percentages which ranged from 2.79 to 3.73%. The recorded variation in the total organic matter (TOM) in bottom sediments of the studied ports at Hurghada and Safaga is usually related to the local hydrodynamics, algal and seagrass flourishing, the terrigenous and domestic wastewater. Mansour et al. (2013) attributed the recorded TOM at Hurghada to the seagrass patches and the algal bottom faces.

Table 1 The averages of carbonates (%), TOM (%) and leachable heavy metals (g/g) in different sediment fractions (Ø3, Ø4 and Ø5) at the studied stations of the Red Sea ports

Stations		CO_3%	TOM%		Fe	Mn	Zn	Cu	Ni	Pb	Co	Cd
H-I	Passenger port	51.62	3.83	$Ø_3$	1385	30.88	37.87	14.58	15.13	15.35	BDL	1.3
				$Ø_4$	1620	49.58	35.62	22.58	19.78	19.6	BDL	1.48
				$Ø_5$	2425	78.37	55.32	40.52	17.2	19.52	BDL	1.92
H-II	Shipyard	56.28	4.6	$Ø_3$	1946	25.3	191.9	125.68	14.15	77.37	BDL	0.01
				$Ø_4$	601	9.27	76.22	43.5	13.92	50.58	BDL	0.01
				$Ø_5$	6556	108.17	330.38	298.4	91.4	101.02	0.1	0.01
H-III	Fishing berth	68.33	8.78	$Ø_3$	2900	24.73	25.91	15.09	28.96	BDL	1.66	BDL
				$Ø_4$	4201	40.05	39.64	30.85	55.7	7.35	2.28	0.09
				$Ø_5$	4511	57.03	89.24	91.9	61.96	19.95	2.43	BDL
H-IV	Marina	76.65	2.48	$Ø_3$	551	14.98	12.38	10.08	24.4	5.33	1.5	BDL
				$Ø_4$	797	25.8	20.18	19.75	19.93	13.43	BDL	0.1
				$Ø_5$	799	28.7	36.03	51.15	20.33	11.03	1.1	0.3
S-I	Grains platform	9.02	1.91	$Ø_3$	3876	155.75	47.1	23.3	22.35	8.98	2.63	0.43
				$Ø_4$	6813	168.63	47.85	30.93	27.28	8.3	1.18	0.68
				$Ø_5$	7483	206.77	66.25	62.58	27.03	15.25	3.5	0.6
S-II	Passenger terminal	8.81	2.63	$Ø_3$	4246	275.71	35.73	17.72	26.97	8.22	2.98	0.43
				$Ø_4$	4417	271.43	54.78	32.35	29.1	14.93	0.62	1.08
				$Ø_5$	5995	306.3	69.32	54.7	31.35	5.45	3.07	1.03
S-III	Aluminum platform	21.86	8.81	$Ø_3$	4543	291.82	126.6	18.35	31.73	12	0.9	1.48
				$Ø_4$	3721	240.85	79.83	20.8	33.28	16.4	3.35	1.78
				$Ø_5$	4788	248.07	109.17	43.1	23.48	7.48	5.5	1.6
S-IV	Coal platform	32.32	10.09	$Ø_3$	4347	161.57	23.93	17.5	36.13	9.48	3.85	0.93
				$Ø_4$	3476	134.5	20.65	22.7	17.03	9.55	5.03	0.58
				$Ø_5$	4479	175.77	20.1	35.23	31.63	15.48	2.38	1.05
S-V	quartz platform	59.52	2.79	$Ø_3$	2988	130.03	21.98	13.12	24.13	13.9	5.17	0.63
				$Ø_4$	3194	142.52	9.07	16.33	22.63	11.98	1.7	1.25
				$Ø_5$	3432	145.77	12.15	35.87	27.62	22.52	4.75	1.2
Q-I	Old platform	48.63	3.73	$Ø_3$	3189	154.27	21.24	17.33	38.1	22.75	4.19	2.09
				$Ø_4$	4371	200.36	27.7	19.08	46.88	26.79	4.96	2.41
				$Ø_5$	4968	233.7	39.78	34.45	43.95	32	4.11	4.19
Q-II	Basin	45.14	2.84	$Ø_3$	5601	183.53	72.78	17.48	73.3	21.12	5.11	1.55
				$Ø_4$	5095	171.27	61.38	17.86	61.0	13.22	5.85	1.84
				$Ø_5$	6207	214.94	80.53	33.76	62.11	25.23	4.71	2.54
LEL (lowest effect level)					20,000	460	120	16	16	31		0.6
SEL (Sever effect level)					40,000	1100	840	110	75	250		10
Red Sea background value, 1943 (Hanna, 1992)					3000	116	24	17.6	16	3	3	0.4
Average shale (Förstner et al., 1982)						850	95	45	68	20	19	0.3

Note: (BDL) Below detection limit

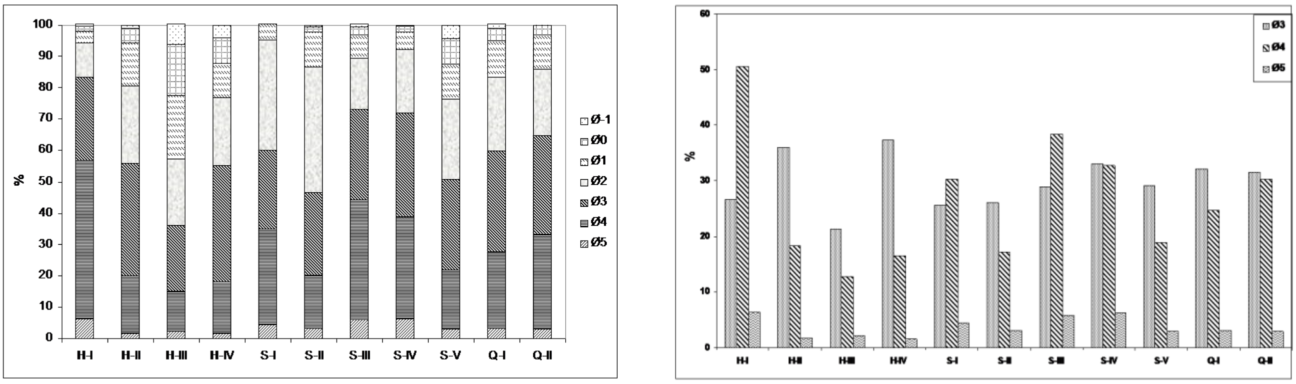

Figure 2 Distribution patterns of the different size fractions and the percentages of the finest fractions Ø3, Ø4 and Ø5 at the studied ports

2.2.2 Distribution of leachable heavy metals and evaluation the biological risk

At Hurghada, the finest fraction Ø5 was the essential heavy metal carrier followed by Ø4 at the different ports except passenger port, which was followed by Ø3. The marine area off Hurghada shipyard recorded significantly high Fe, Mn, Zn, Cu, Ni and Pb in the finest fraction Ø5 (6556, 108.17, 330.38, 298.4, 91.40 and 101.02 µg/g, respectively), meanwhile tourist marina recorded the lowest values. Fishing berth was followed the shipyard in the heavy metal contents in the different fractions (Table 1). The metal accumulation at Hurghada ports was follow the sequence Ø5> Ø4> Ø3. Dar et al. (2016b) and Madkour and Dar (2007) attributed the high accumulation of heavy metals in the marine sediments off the shipyards to the repairing, maintaining, antifouling paint remains and ship constructing. At Safaga, Fe showed relatively high values at the different wharfs relative to Hurghada and Qusier due to the high amount of terrestrial runoff. The highest Fe and Cu were recorded in Ø5 (7483 and 62.58 µg/g, respectively) followed by Ø4 (6813 µg/g) for Fe at the grain wharf and Ø5 (54.70 µg/g) for Cu at passenger terminal; the highest Mn was observed at passenger warf in Ø5 (306.3 µg/g) followed by bauxite wharf in Ø3 (291.82 µg/g). The highest Zn in Ø3 (126.6 µg/g) followed by Ø5 (109.17 µg/g) was showed in front of bauxite wharf. The highest Ni was in Ø3 (36.13 µg/g) in front of coal wharf, Pb was in Ø5 (22.52 µg/g) at orthoclase and quartz wharf, meanwhile Co was observed in Ø5 (5.5 µg/g) at the navigation basin of bauxite wharf. The recorded Cd at all wharfs was less than 2 µg/g. Dar et al. (2016b) concluded that the high Zn and Mn at Safaga marine area attributed to the high quantities of pollutants rich in Zn from antifouling paint remains and Mn from the terrestrial runoff from the phosphate shipments. At Qusier, the highest Fe and Zn was showed at the fishing basin in Ø5 (6207 and 80.53 µg/g, respectively) followed by Ø3 (5601 and 72.78 µg/g, respectively), the highest Mn and Cu was showed in Ø5 (233.7 and 34.45 µg/g, respectively) in front of the old port. The highest Ni was recorded in Ø3 (73.30 µg/g) and Co was in Ø4 (5.85 µg/g) at the fishing basin, meanwhile Cd recorded the highest content in Ø5 (4.19 µg/g) at the old port. The marine area off the old port and fishing basin were highly affected by the terrestrial runoff from the subsurface wastewater and coastal activities. The accumulation sequences of metals at Safaga and Qusier were; Ø5> Ø3> Ø4.

It was interesting to observe that the distribution of Fe and Mn were generally lower in Hurghada area (significant in most cases at $p \leqslant 0.05$) comparing to Safaga and Qusier stations (Figure 3). In consistence with our results, previous work showed general elevated levels of Fe and Mn in sites which were used for shipping of ores, especially phosphate and bauxite as reported in Safaga, Hamrawin and Qusier (Mansour et al., 2011; Madkour et al., 2012; El-Metwally, 2015; Dar et al., 2016a; 2016b). Mn and Fe are essential elements and have relatively low toxicology to aquatic organisms. The concentrations of Fe and Mn in all ports were very low compared with the values of LEL (20,000 and 460 µg/g, respectively) and SEL (40,000 and 1100 µg/g, respectively). The values of Fe in the present study were significantly lower than Port Kemblaharbour, Australia (He and Morrison, 2001), while the recorded Mn values were comparable with those reported in Sydney Harbour (Irvine and Birch, 1998) and Victoria Harbour (Wong et al., 1995; Tang et al., 2008) but were much lower than Hamilton Harbour (Poulton et al., 1996). The Fe availability in this study was below that recorded in other harbours around the world (Table 2).

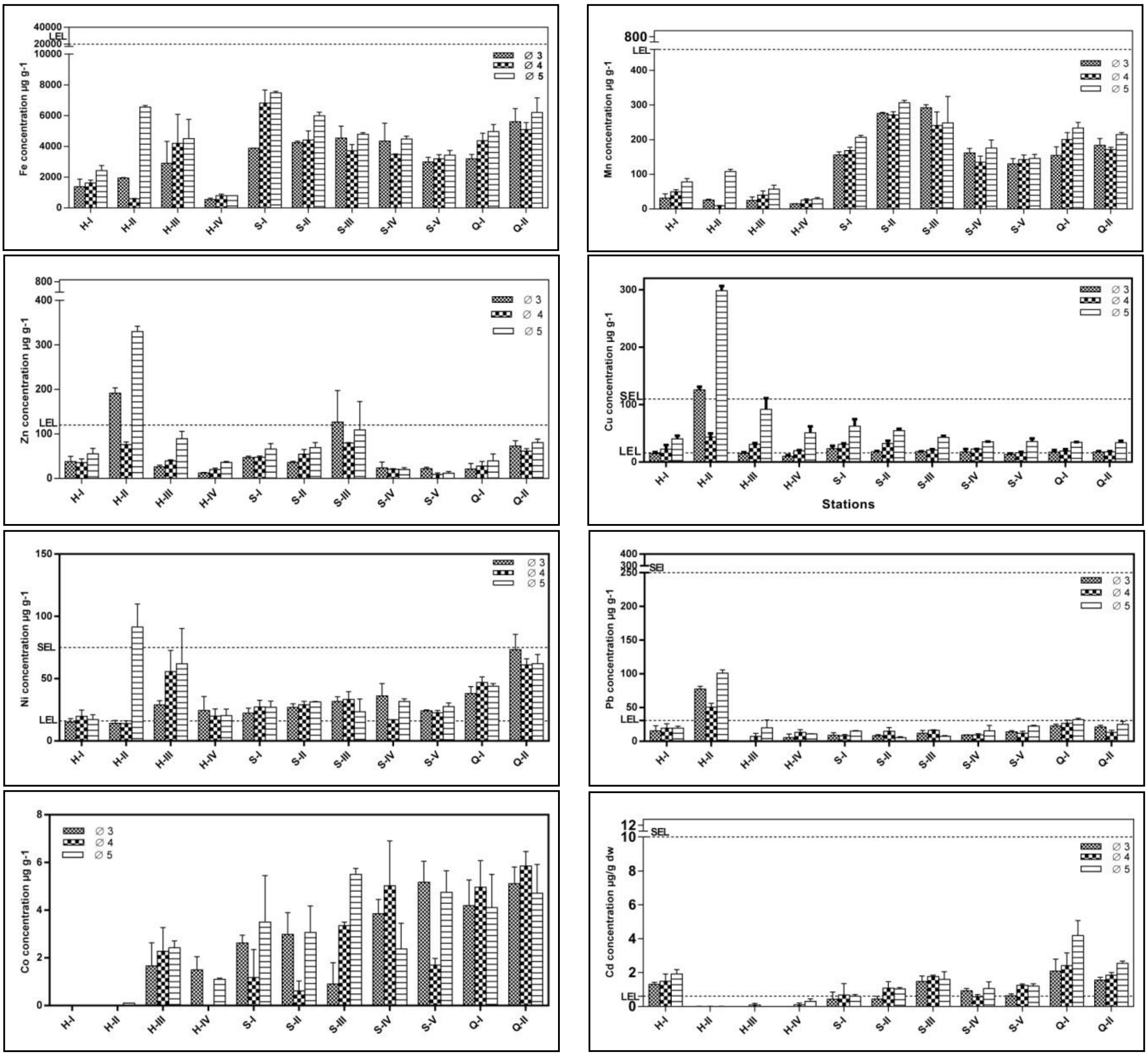

Figure 3 Leachable heavy metals concentrations in fine sediments (Ø3, Ø4 and Ø5) with references to the lowest effect level (LEL) and the sever effect level (SEL) at different stations of the Red Sea ports

The significantly high concentrations of Zn, Cu and Pb at Hurghada shipyard were not surprised, since this area serves the activities of ships maintenance and repairing in Hurghada. High amounts of Zn and Cu are continuously dumped to surface sediments from scraped antifouling paints. It is also believed that, considerable amounts of Pb released from accidental oil spill and seeping of fuel during ships maintenance and from mooring boats (Madkour and Dar, 2007). Cu showed significantly ($p<0.05$) higher levels in the mud sediments (Ø5 fraction). Similar result was observed by El-Said and Youssef (2013) in mangrove sediments of the Red Sea. Among 6 studied metals, they found that Cu showed distinctive high levels in Ø5 relative to the other fractions. Regarding the SQGs, Figure 3 shows the leachable metals rates between LEL and SEL at the different stations. Cu concentrations in all stations exceeded the values of LEL (16 µg/g) and only at the shipyard was exceeded the value of SEL (110 µg/g), meanwhile the recorded concentrations of Zn and Pb in all ports were below LEL (120 and 31 µg/g, respectively) except the shipyard, which were exceeded the guidelines (76.2 ± 5.7 to 330.4 ± 11.5 µg/g for Zn and 50.6 ± 5.8 to 101 ± 4.9 µg/g for Pb). On the other hand, the contamination status of Cu, Zn and Pb was much higher than the Trade Harbours of South Korea (Choi et al., 2012) and lower than those recorded in Port Kemblaharbour, Australia (He and Morrison, 2001), Sydney Harbour (Irvine and Birch, 1998), Kaohsiung Harbour (Chen et al., 2007) and Hamilton Harbour (Poulton et al., 1996) (Table 2).

Table 2 Comparison between the recorded heavy metals levels (mg/g) in the studied ports with other previous studies worldwide

	Co	Cu	Zn	Ni	Cd	Mn	Pb	Fe	extraction	reference
Red Sea Hrabours	<0.01-5.85	10.1-298	9.1-330	13.9-91.4	<0.01-4.19	9.3-306	<0.01-101	551-7483	leachable	present study
Trade Harbours (south Korea)	-	8.6-28.2	47-112	10.3-28.8	0.03-0.22	-	9.9-41.2	-	leachable	Choi et al., 2012
Kaohsiung Harbour, Taiwan	-	5-946	52-1369	-	0.1-6.8	-	9.5-470	-	leachable	Chen et al., 2007
Sydney Harbour, Australia	3.0-60	13-1078	46-2246	17-86	1.0-10.0	30-408	44-1319	10,000-78,000	leachable	Irvine and Birch, 1998
Suva Harbour, Fiji	-	21.4-143	40.2-269	-	-	-	22.1-93.5	14000 48700	leachable	Maata and Singh, 2008
harbour of Ceuta, Spain	-	5-865	29-695	8-671	-	61-332	10-516	3060-41,100	leachable	Guerra-Garcia and Garcia-Gomez 2005
Hamitonharbour, Canada	-	8-135	338-5930	8.0-61	-	42-1152	18-1250	12,200-204,000	leachable	Poulton et al., 1996
Port Kemblaharbour, Australia	-	95-1468	1209-2220	-	-	-	151-484	73,000-100,429	total	He and Morrison, 2001
Bergen harbour, Norway	-	25-1090	26-2900	-	-	-	24-1920	-	total	Paetzel et al., 2003
Victoria Harbour, Hong Kong	-	45.2-3789	97.9-610.4	23.6-177.1	2.3-3.3	373.9-568.8	47.4-138.1	28900-34100	total	Wong et al., 1995
Victoria Harbour, Hong Kong	-	19-280	52-221		-	-	21-85	-	leachable	Tang et al., 2008
Hamraween Bay, Red Sea	-	20.1	99.6	-	-	283.5	73.35	1270	leachable	Dar et al., 2016a
Darwin Harbour (Australia)	1.9-7.65	1.4-14.9	5.8-28.4	3.3-11.1	0.06-0.12	254-411	3.11-13	7,790- 58,200	leachable	Padovan et al., 2012

The spatial distribution of Ni was more homogenous between ports with significant high levels in fishing berth of Hurghada (28.96±3.0 to 61.96±28.0 µg/g) and Qusier old port and fishing basin (38.1±5.0 to 73.3±12.0 µg/g). Salem et al. (2014) stated that the locations recorded high levels of Ni and partially Co related to anthropogenic discharges. The levels of Ni in most studied ports were higher than the LEL (16 µg/g) but lower than SEL (75 µg/g) except Ø5 (91.40 µg/g) fraction that exceeding SEL limit at Hurghada shipyard. The overall concentrations of Ni much higher than the Trade Harbours of South Korea (Choi et al., 2012), within the range recorded in the Sydney Harbour (Irvine and Birch, 1998) and Hamilton Harbour (Poulton et al., 1996) and were lower than Victoria Harbour (Wong et al., 1995; Tang et al., 2008) and Ceuta Harbour (Guerra-Garcia and Garcia-Gomez, 2005) (Table 2).

The concentration of Co recorded significantly very low concentrations ($p \leqslant 0.01$) at Hurghada passenger port and Hurghada shipyard, meanwhile the highest availability were observed in Qusier old port and fishing basin (Figure 3). There were no standard guidelines for Co in the marine sediments, but the overall Co levels in current study were below the standard background of average shale (Förstner et al., 1982) and below the recorded values in other contaminated sites such as in Sydney Harbour (Irvine and Birch, 1998) and Darwin Harbour (Padovan et al., 2012).

Cadmium showed significantly low concentrations ($p \leqslant 0.05$) at Hurghada ports and was below the lowest effect levels (LEL), meanwhile Safaga and Qusier ports recorded Cd concentrations higher than LEL (0.6 µg/g) but still below SEL value (10 µg/g) (Figure 3). Comparing to the levels of Cd with worldwide Harbours (Table 2), Qusier old port and fishing basin recorded levels similar to Hamilton Harbour (Poulton et al., 1996) and Victoria Harbour (Wong et al., 1995; Tang et al., 2008) but it is lower than the levels in Sydney Harbour (Irvine and Birch, 1998).

2.3 Associations and sources of the leachable heavy metals

Correlation matrix and Principal Component Analysis (PCA) estimated the statistical relationship among heavy metals as well as between sediment characteristics and the heavy metals. Additionally, PCA was used to infer the hypothetical sources of heavy metals contamination (Dou et al., 2013; Qiao et al., 2013; Fujita et al., 2014; Yang et al., 2015).

The correlation matrix for the different heavy metals in sediment fractions showed strong association between heavy metals in the mud fraction Ø5, followed by Ø3, and the least association was in Ø4. Two significant associations were observed; the first association was strong positive correlation between metal pairs of; Cu, Zn, Pb and Ni in Ø5 to lesser extents of; Cu, Zn, Pb in Ø3. The second association was common in all fractions, competed Mn with Fe and Cd in strong correlations coincided with a negative correlations of carbonate and with Fe and Mn (Table 3).

As shown in Figure 4, components in Ø3, Ø4 and Ø5 between metals, the obtained results of the PCA showed wide accordance with the correlation matrix. In Ø3 (Figure 4A), two main components with accumulative account for 55.44% of the total variance were found. In the first component (31.98% of the total variance), Fe, Mn and Ni were grouped with positive loading and carbonate content with negative loading. The second component (23.46% of the total variance) grouped positive loading of Cu, Pb and Zn. In Ø4 (Figure 4B), two main components were identified, accounted for 45.32% of the total variance. The first PCA (27.5% of the total variance) was associated with positive loading of Mn, Fe, Cd and Co accompanied with negative loading of carbonate. The second PCA (17.83% of the total variance) showed positive loading between Cu and TOM as well as negative loading of Cu with Cd. In Ø5 (Figure 4C), three components (with total account of 72.49%) were recognized. The first component (32.73% of total variance) included strong positive loadings of Cu, Zn, Pb and Ni. The second component (24.35% of total variance) showed positive loading between Mn and Fe associated with negative loadings toward carbonates. The third PCA component (15.42% of total variance) showed positive loading of Cd with Co and negative loading of Cd and Co toward TOM.

Table 3 Correlation coefficient between the leachable metals and the geochemical characteristics in the different sediment fractions

	$Ø_3$	Co	Cu	Zn	Ni	Cd	Mn	Pb	Fe
$Ø_3$	Co	1.00							
	Cu	-0.18	1.00						
	Zn	-0.09	0.64**	1.00					
	Ni	0.42*	-0.08	0.26	1.00				
	Cd	0.11	-0.20	0.06	0.33	1.00			
	Mn	0.31	-0.13	0.27	0.40*	0.40*	1.00		
	Pb	-0.02	0.79**	0.59**	0.07	0.31	0.01	1.00	
	Fe	0.48**	-0.02	0.30	0.73**	0.20	0.66**	0.01	1.00
	CO_3%	-0.03	-0.02	-0.23	-0.18	-0.20	-0.80**	0.04	-0.58**
	TOM%	-0.09	0.02	0.08	-0.06	-0.09	-0.04	-0.19	0.28
$Ø_4$	Co	1.00							
	Cu	-0.35*	1.00						
	Zn	0.13	0.28	1.00					
	Ni	0.09	0.25	0.18	1.00				
	Cd	0.40*	-0.41*	-0.01	0.26	1.00			
	Mn	0.34	-0.14	0.24	-0.10	0.56**	1.00		
	Pb	-0.14	0.15	0.20	0.14	0.36*	0.00	1.00	
	Fe	0.49**	0.05	0.20	0.14	0.12	0.50**	-0.45**	1.00
	CO_3%	-0.04	-0.17	-0.33	0.03	-0.28	-0.73**	0.04	-0.46**
	TOM%	0.10	0.06	0.07	0.09	-0.20	-0.14	-0.20	0.22
$Ø_5$	Co	1.00							
	Cu	-0.27	1.00						
	Zn	-0.07	0.83**	1.00					
	Ni	-0.07	0.60**	0.63**	1.00				
	Cd	0.19	-0.41*	-0.25	-0.02	1.00			
	Mn	0.40*	-0.29	-0.01	-0.04	0.51**	1.00		
	Pb	-0.11	0.72**	0.62**	0.61**	0.13	-0.14	1.00	
	Fe	0.26	0.10	0.31	0.28	0.06	0.53**	0.04	1.00
	CO_3%	-0.04	0.18	-0.01	0.15	-0.09	-0.72**	0.34	-0.58**
	TOM%	-0.08	0.00	0.04	-0.09	-0.22	-0.16	-0.30	0.09

Note: Two-tailed correlation significant at (*) $p \leq 0.05$ and (**) $p \leq 0.01$

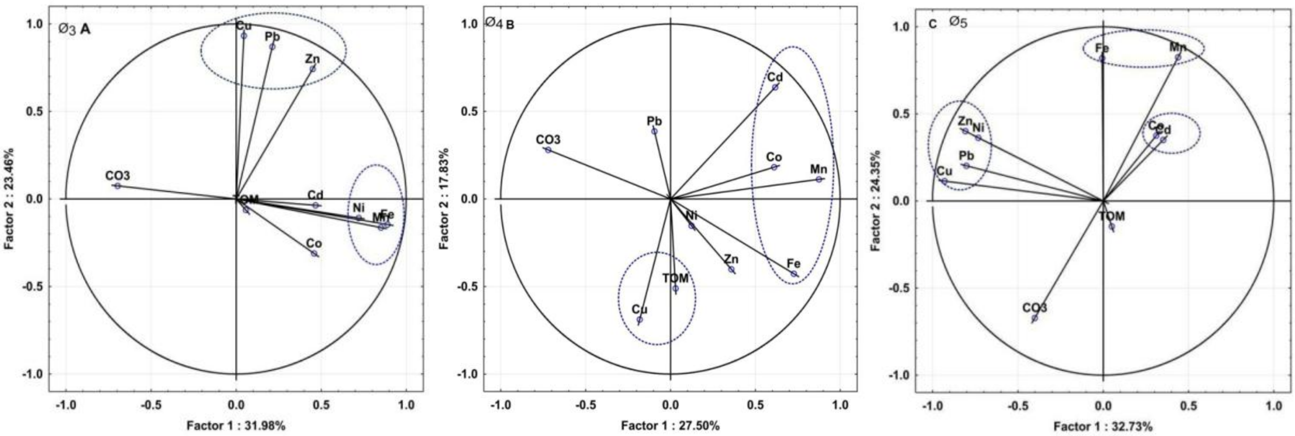

Figure 4 Heavy metals associations in the fine fractions Ø3 (A), Ø4 (B) and Ø5 (C) according the PCA between metals

The overall multivariate analyses showed two major patterns. The first pattern suggested that the metals Fe, Mn and partially Cd were originated from the trrigenous sources and were negatively correlated with carbonate, this mainly due to phosphate ores sedimentation and coastal activities. Previous work in many locations of Red Sea area showed high levels of these metals (Fe, Mn and Cd) associated with terrestrial inputs (e.g., Mansour et al., 2011; Madkour et al., 2012; El-Metwally, 2015; Dar et al., 2016a; 2016b; El-Metwally et al., 2017). The second pattern showed that high portion of Cu, Zn and Pb that mostly have anthropogenic sources since the highest impacts were observed at Hurghada shipyard.

2.4 Assessment the ecological risk

Metal pollution load index (MPI) is a simple method to describe the integrated effect of metal contamination. The values (>1) indicate to the progressive deterioration in sediments quality (El-Said and Youssef, 2013). MPI was obtained from the following formula:

$$MPI = (CF_{Co} \times CF_{Cu} \times CF_{Zn} \times CF_{Ni} \times CF_{Cd} \times CF_{Mn} \times CF_{Pb} \times CF_{Fe})^{1/n}$$

Where, the contamination factor (CF) is the concentration of metal in obtained sample (Cmetal) divided by the background concentration (Cbackground) of the same metal and (n) is the number of measured metals. The calculated values of MPI were ranged between 1.08 and 1.50 (Table 4). The highest MPI values were found in Hurghada stations (1.11 to 1.50) followed by Qusier (Table 4) and the lowest MPI values (1.08 to 1.22) were at Safaga stations. Similar pattern for MPI was recorded by Salem et al. (2014) at Qusier and Safaga.

The enrichment factor (EF) represents the actual contamination level in sediments since it is probably differentiates between natural and anthropogenic sources of metals (Chen et al., 2007; Amin et al., 2009). EF was calculated from the equation:

$$EF = {(Metal/Fe)_{sample}} \Big/ {(Metal/Fe)_{background}}$$

The calculated averages of EF showed that the heavy metals were followed the enrichment order; Pb>>Cu≥ Zn>Cd>Mn=Ni>Co. Zhang and Liu (2002) reported that the lowest values (EF<1.5) indicating to natural sources of heavy metals (crustal materials) and the highest values (EF>1.5) indicating to the significant anthropogenic sources. In general, the calculated EF values illustrated moderate enrichment for Cu (2.45), Zn (2.37), Ni (2.03), Cd (2.23) and Mn (2.04), meanwhile Pb was highly enriched (6.89). With respect to the studied stations, Hurghada shipyard and tourist marina showed high EF enrichment by Cu (6.43 to 8.76), Zn (3.99 to 8.22), and Pb (10.03 to 25.15) attributed to anthropogenic sources from ships repairing, antifouling paints and fuels leakage as well as boats mooring. The studied stations at Safaga and Qusier were enriched by Mn (1.99 to 4.06) and partially Cd (1.55 to 5.2) due to the different shipment operations (Table 5).

Table 4 Geo-accumulation indices (I_{geo}) for sediment samples at the different stations of the studied ports

Stations	Co	Cu	Zn	Ni	Cd	Mn	Pb	Fe
H-I	-12.13	-0.02	0.25	-0.46	1.39	-1.72	2.01	-1.31
H-II	-7.07	2.56	2.47	0.73	-6.91	-1.87	4.08	-0.56
H-III	-1.08	0.79	0.52	1.03	-4.36	-2.10	1.01	-0.21
H-IV	-2.37	0.03	-0.65	-0.15	-2.17	-2.91	1.14	-2.65
S-I	-0.88	0.56	0.57	0.09	-0.08	0.02	1.26	0.42
S-II	-1.01	0.40	0.56	0.28	0.50	0.71	1.08	0.12
S-III	-0.46	0.05	1.54	0.29	1.43	0.58	1.41	-0.05
S-IV	-0.26	-0.07	-0.74	0.24	0.50	-0.14	1.35	-0.13
S-V	-0.21	-0.27	-1.32	0.05	0.77	-0.32	1.84	-0.49
Q-I	-0.02	-0.16	-0.28	0.84	2.27	0.17	2.59	-0.11
Q-II	0.21	-0.19	0.99	1.45	1.72	0.12	2.14	0.32

Table 5 The calculated enrichment factor (EF) and metal pollution load index (MPI) in sediments of the studied ports

Stations	EF_{Co}	EF_{Cu}	EF_{Zn}	EF_{Ni}	EF_{Cd}	EF_{Mn}	EF_{Pb}	MPI
H-I	<0.01	2.44	2.96	1.80	6.49	0.76	10.03	1.33
H-II	0.01	8.76	8.22	2.46	0.01	0.68	25.15	1.50
H-III	0.55	2.02	1.67	2.37	0.06	0.58	2.35	1.11
H-IV	1.21	6.43	3.99	5.65	1.40	0.33	13.87	1.39
S-I	0.40	1.10	1.11	0.79	0.70	2.53	1.79	1.08
S-II	0.45	1.22	1.36	1.12	1.30	4.06	1.95	1.09
S-III	0.75	1.07	3.02	1.27	2.79	3.72	2.75	1.13
S-IV	0.91	1.05	0.66	1.29	1.55	2.25	2.80	1.14
S-V	1.21	1.16	0.56	1.45	2.41	1.99	5.03	1.22
Q-I	1.06	0.96	0.89	1.93	5.20	2.80	6.51	1.26
Q-II	0.93	0.70	1.59	2.18	2.63	2.71	3.52	1.17

Geo-accumulation index (Igeo) has been proposed by Müller (1969) to evaluate the contamination level in sediments by comparing current status with the pre-industrial levels according to the formula:

$$I_{geo} = Log_2 \left({C_n}/{(1.5 \; x \; B_n)} \right)$$

Where Cn is the current concentration of metal, Bn is the geochemical background value of the same metal, and the factor 1.5 is the matrix correction factor of the background. According to Müller (1981), Igeo is estimated over seven categories; uncontaminated sediments (Igeo≤0), uncontaminated to moderately contaminated (0< Igeo≤1), moderately contaminated (1< Igeo≤2), moderately to strongly contaminated (2< Igeo≤3), strongly contaminated (3< Igeo≤4), strongly to extremely contaminated (4< Igeo≤5) and extremely contaminated (Igeo≥5).

As calculated in Table 4, the Igeo values of the studied heavy metals at the different stations were classified the marine sediments as unpolluted to moderate polluted; except at Hurghada shipyard, the marine sediments were classified as moderately to highly polluted by Cu (2.56), Zn (2.47) and Pb (4.08).

3 Conclusions

Distribution and ecological risk of heavy metals were investigated in finest fractions of the surface sediments of Red Sea ports at Hurghada, Safaga and Qusier Cities. The total of the finest fractions (Ø3, Ø4 and Ø5) was varied between 36.26% and 83.47% with considerable high percentages of the different stations indicating to different sources of depositions mostly from the maritime activities, terrestrial runoff, phosphate shipments and the nearby coastal based activities. Carbonate percentage showed significant decline with increasing the terrigenous inputs at Safaga followed by Qusier, however, Hurghada stations showed high carbonate percentages due to the high marine productivity. TOM recorded variable percentages at the different stations may attribute to the local effects of the anthropogenic effluents. At Hurghada, the finest fraction Ø5 was the essential heavy metal carrier followed by Ø4 with reference to Hurghada shipyard that recorded significantly high Fe, Mn, Zn, Cu, Ni and Pb in the finest fraction Ø5. At Safaga, Fe showed relatively high values at the different wharfs relative to Hurghada and Qusier due to the high amount of terrestrial runoff. At Qusier, the highest Fe and Zn was showed at the fishing basin in Ø5 followed by Ø3. The distribution of Fe and Mn were generally lower in Hurghada stations ($p \leq 0.05$) comparing to Safaga and Qusier stations with distinctive high levels of Cu ($p<0.05$) in Ø5. A strong correlation observed between Fe, Mn and partially Cd, it was associated with the process of shipping of raw materials from Safaga and Qusier ports. The organic matter content showed no significant correlation with heavy metals in the surface sediments, and carbonate was negatively correlated with Cu, Zn and Pb. The levels of heavy metals in present study were similar/or below the levels recorded in other ports worldwide. According to sediments quality guidelines (SQGs), the concentrations of heavy metals in sediments of the studied ports were not expected to have biological adverse effects, except the metals Cu, Zn, Pb and Ni may pose some ecological risks to marine organisms near the shipyard of Hurghada.

Authors' contributions

Mohamed E.A. El-Metwally contributed to the paper by collecting samples, analyses, processing the data and writing the manuscript. Amany G. Madkour and Rasha R. Fouad contributed to the paper by collecting samples, laboratory treatment and data analysis. Lamiaa I. Mohamedein contributed to the paper by collecting samples, analysis and coordination. Hamada A. Nour Eldine contributed by collecting samples. Mahmoud A. Dar and Khalid M. El-Moselhy contributed by editing and revising the manuscript.

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