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Baseline

Assessment of metal contamination in coastal sediments, seawaters and bivalves of the Mediterranean Sea coast, Egypt

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ABSTRACT

In order to assess metal contamination on the Mediterranean coast of Egypt, 45 sediment samples, seawaters and bivalve specimens were collected from Rosetta coastal area for Mg, Al, K, Fe, Sr, Zn, Pb, Mn, As, Ce, Ni, Cr and Zr analyses by Inductively Coupled Plasma-Mass Spectrometer. The Enrichment Factor (EF), the Geoaccumulation Index (Igeo) and the Contamination Factor (CF) indicated that the coastal sediments of Rosetta area were severely enriched, strongly polluted with As, Pb and very highly contaminated with As, Pb, Ni, Ce, mostly as a result of anthropogenic inputs. Comparison with other samples from the Arabian Gulf, Red Sea and abroad coasts suggested that the studied samples have higher concentrations of Fe, Pb, As, Zn and Ni. The natural sources of heavy metals in the study area are attributed to weathering and decomposition of mountain ranges of the Sudan and Ethiopia, while the anthropogenic ones are the metals produced from industrial, sewage, irrigation and urban runoff.

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Metals are introduced into the aquatic system as a result of weathering of soil and rocks, volcanic eruptions and from a variety of human activities involving mining, processing and use of metals and/or substances containing metal contaminants (Karageorgis et al., 2003; Lin et al., 2012; El-Sorogy 2012, 2013a,b). Sediments act as both carriers and sinks for contaminants in aquatic environments. Heavy metal concentrations in sediment are many times greater than the same metals in the water column. Sediments can act as a scavenger agent for heavy metal and an adsorptive sink in aquatic environment. It is therefore considered to be an appropriate indicator of heavy metal pollution (Idris et al., 2007; Gupta and Singh, 2011).

The Egyptian black sands are the end products of the disintegrated materials from the igneous and metamorphic rocks (Dabbour, 1995; Dawod and Abdelnaby, 2007). These deposits comprise huge reserves of the six common economic minerals that include ilmenite, magnetite, garnet, zircon, rutile and monazite (El-Askary and Frihy, 1987; Hedrick, 1989). The Rosetta black sands have been the subject of many articles concerning the mineralogy and economics of these black sands (El-Miligy and El-Azab, 1994; Mahmoud et al., 2013).

Rosetta area as one of the Egyptian coastal areas on the Mediterranean Sea receives different types of pollutants, therefore this study is designed to assess the current status of metal contamination in the coastal sediments, seawaters and bivalves from Rosetta beach,

Mediterranean Sea, Egypt and to identify potential sources of contamination. This evaluation helps developing effective coastal management guidelines and strategies for better management of coastal activities.

Study area is bordered by the north western part of Suez Canal from the east and northern side of Manzala Lake from the south, between longitudes 30° 21'–30° 28' E and latitudes 31° 26'–31° 29' N. It is located northeast of Abu Khashaba village, east of Rosetta and bounded by the Mediterranean Sea shoreline from the north. Rosetta beach area is an open area, nearly flat, very gently dipping to north and occupied mainly by sabkha.

The study area is characterized by a low relief mainly below 2 m above sea level, and slope gently from south to north. Geomorphologically, it is represented by a flat coastal plain dissected by some hummocky sand dunes in the south. The beach face slope increases from zero near the Rosetta mouth and becomes relatively steeper eastwards where it reaches about 15° at a distance of about 20 km from the mouth (Wassef, 1973).

The beach deposits are composed of loose fine sands with a considerable amount of heavy minerals. The sand particles are composed of quartz, feldspars and mafic minerals with specific gravities between 2.65 and more than 5 g/cm³. The heavy minerals of the black sands are ilmenite, magnetite, zircon, garnet, rutile, and monazite. The black sands also contain traces of gold, cassiterite, beryl, chromite, corundum, apatite, collophane, uranothorite and gangue minerals. The latter include hornblende, actinolite, augite, hedenbergite, hypersthene, enstatite and minor amounts of biotite, epidote, staurolite, sphene, tourmaline, sillimanite and olivine (Hammoud, 1966).

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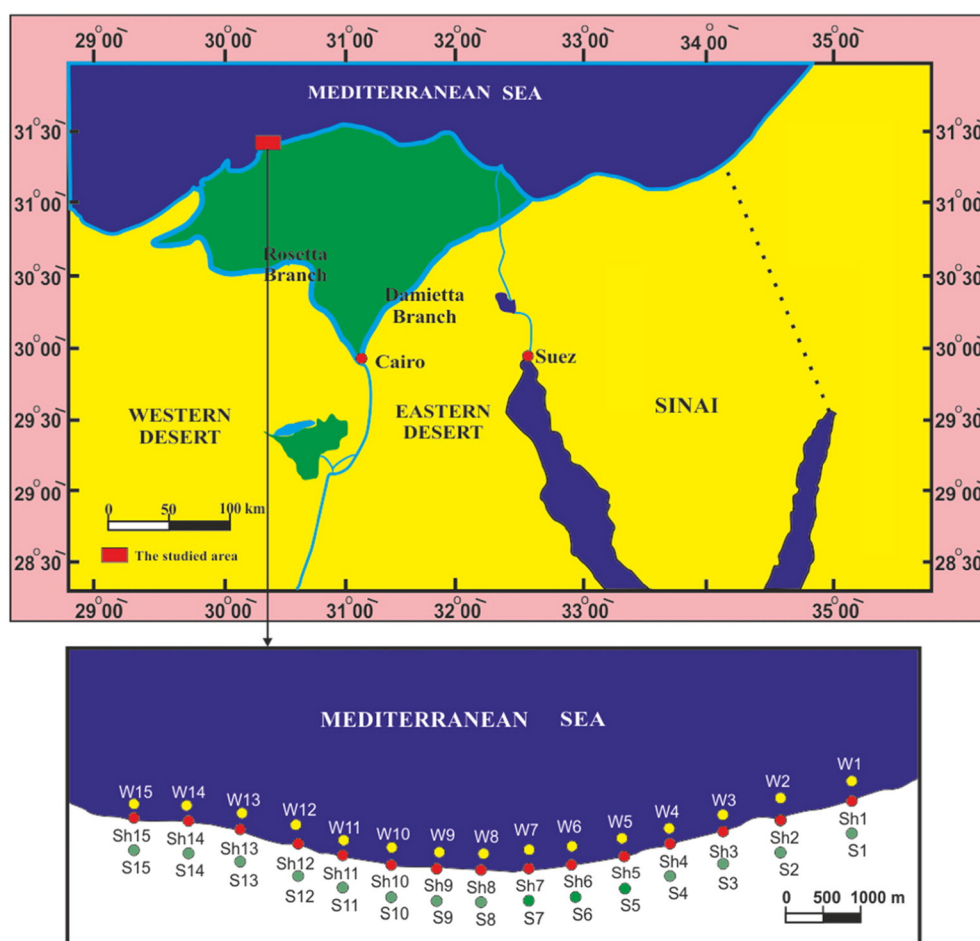


Fig. 1. Location map of Rosetta coastline and sample sites along the coast.

45 coastal sediment samples, seawaters and bivalves were analyzed for Mg, Al, K, As, Fe, Sr, Zn, Pb, Mn, Ce, Ni, Cr, and Zr from Rosetta beach area along the Egyptian Mediterranean Sea (Fig. 1). The sediment samples were prepared by accurately weighing around 100 mg of samples into a dry and clean Teflon microwave digestion vessels, 2 ml of HNO_3 , 6 ml HCl and 2 ml HF were added to the vessels (Trabzuni et al., 2014). Samples were digested using scientific microwave (Model Milestone Ethos 1600). The resulting digest was transferred to a 15 ml plastic volumetric tube and made up to mark using deionized

water. A blank digest was carried out in the same way. The analytical determination of trace metals was carried out by ICP-MS (Inductively Coupled Plasma-Mass Spectrometer): NexION 300D (Perkin Elmer, USA). The bivalve shells (*Ostrea* sp.) were chosen for heavy metals analysis due to their abundance and occurrence all over the studied stations and they are benthic epifauna and easy to sampling. Also they are filter feeders and have the potential to bioconcentrate contaminants, which would normally be present in the water or within sediments.

Table 1
Concentrations of heavy metals in coastal sediments of Rosetta beach, Egyptian Mediterranean coast.

Samples	Al	Fe	K	Mg	Mn	Ce	Cr	Ni	Sr	Zr	Zn	As	Pb
S1	240	193,900	90	570	610	1690	0.26	894.17	162.34	0.21	298.50	352.65	476.45
S2	240	193,100	70	560	600	1740	0.29	819.24	128.88	0.22	314.42	315.86	457.99
S3	240	194,900	100	560	610	1740	0.26	744.32	175.01	0.32	320.91	279.06	465.94
S4	240	189,800	130	590	610	1670	0.26	722.54	143.61	0.28	324.50	325.35	471.47
S5	240	192,600	60	570	610	1720	0.26	877.94	121.10	0.30	347.25	299.06	388.22
S6	240	193,000	110	580	610	1750	0.26	706.91	132.30	0.28	336.46	234.32	338.50
S7	240	195,400	120	580	610	1690	0.25	839.54	148.43	0.35	334.35	309.99	460.57
S8	240	198,100	130	570	620	1680	0.23	844.37	123.42	0.28	348.50	376.14	378.45
S9	240	191,500	110	530	640	1720	0.25	722.74	126.73	0.24	387.91	399.16	364.86
S10	240	196,200	60	540	630	1690	0.27	817.74	118.81	0.34	353.25	344.53	368.23
S11	240	189,600	130	581	602	1653	0.26	732.11	140.16	0.23	394.11	361.31	333.47
S12	240	192,294	82	566	615	1742	0.25	847.71	178.22	0.37	340.19	227.96	388.73
S13	240	193,123	119	579	609	1711	0.26	776.92	151.35	0.27	318.40	282.39	391.54
S14	240	195,233	122	568	613	1682	0.24	849.50	152.48	0.37	339.00	367.91	411.51
S15	240	198,000	119	583	610	1683	0.23	854.70	139.49	0.27	333.50	389.99	391.22
Min.	240	189,600	60	530	600	1653	0.23	706.91	118.81	0.21	298.50	227.96	333.47
Max.	240	198,100	130	590	640	1750	0.29	894.17	178.22	0.37	394.11	399.16	476.45
Average	240	193,791.18	102.47	567.47	614.06	1703.76	0.26	803.03	143.49	0.29	340.23	323.11	405.71

Table 2

Concentrations of heavy metals in seawater samples of Rosetta beach.

Samples	K	As	Sr	Mg	Fe	Ce	Cr	Mn	Ni	Al	Zn	Zr	Pb
W1	412.24	0.406	3.952	1525	0.166	12.64	0.003	0.011	0.005	0.005	0.022	0.002	0.006
W2	411.13	0.344	3.111	1465	0.087	29.49	0.003	0.009	0.005	0.005	0.013	0.002	0.006
W3	411.28	0.342	2.704	1389	0.524	15.49	0.004	0.021	0.012	0.037	0.016	0.014	0.006
W4	411.43	0.312	2.311	1354	0.157	9.31	0.003	0.008	0.005	0.007	0.012	0.004	0.006
W5	412.53	0.196	1.008	1524	0.321	17.15	0.004	0.008	0.006	0.011	0.008	0.004	0.006
W6	411.24	0.409	3.941	1514	0.161	11.66	0.003	0.014	0.005	0.007	0.023	0.003	0.005
W7	411.16	0.273	1.379	1352	0.076	10.64	0.005	0.009	0.005	0.003	0.013	0.004	0.006
W8	411.42	0.249	1.078	1425	0.147	9.47	0.003	0.007	0.004	0.006	0.009	0.003	0.006
W9	419	0.333	2.316	1419	0.147	12.15	0.004	0.006	0.004	0.011	0.01	0.005	0.005
W10	411.12	0.346	2.075	1355	0.211	13.64	0.003	0.008	0.005	0.008	0.013	0.007	0.006
W11	411.24	0.278	1.808	1426	0.249	10.02	0.003	0.008	0.005	0.007	0.013	0.005	0.006
W12	411.16	0.273	1.379	1352	0.076	16.53	0.005	0.009	0.005	0.003	0.013	0.004	0.006
W13	416.1	0.259	1.263	1387	0.419	8.13	0.004	0.011	0.006	0.014	0.009	0.002	0.005
W14	411.16	0.236	1.101	1325	0.141	11.51	0.004	0.006	0.005	0.009	0.005	0.004	0.005
W115	408.11	0.199	1.111	1520	0.31	13.53	0.004	0.009	0.005	0.012	0.011	0.003	0.006
Min.	408.11	0.20	1.01	1325.00	0.08	8.13	0.003	0.006	0.004	0.003	0.005	0.002	0.005
Max.	419.00	0.41	3.95	1525.00	0.52	29.49	0.005	0.021	0.012	0.037	0.023	0.014	0.006
Average	412.20	0.30	2.09	1422.47	0.22	10.06	0.004	0.010	0.006	0.011	0.013	0.005	0.006

As shown in Table 1, Fe was the most abundant major element in the sediment (189,600–198,100 µg/g), followed by Ce (1653–1750 µg/g), Ni (706.91–894.17 µg/g), Mn (600–640 µg/g), Mg (530–590 µg/g), Pb (333.47–476.45 µg/g), Zn (298.50–394.11 µg/g), As (227.96–399.16 µg/g), Al (240 µg/g), Sr (118.81–178.22 µg/g), K (60–130 µg/g), Zr (0.21–0.37 µg/g), and Cr (0.23–0.29 µg/g).

The values of Fe, as example, were much higher than those recorded in the coastal sediments of the Gulf of Aqaba (Al-Taani et al., 2014), the Red Sea coast (El-Sorogy et al., 2015a), the Arabian Gulf (Youssef et al., 2015a), India (Alagarsamy and Zhang, 2010), shale and continental crust backgrounds (Turekian and Wedepohl, 1961; Taylor, 1964), the Arabia Gulf (de Mora et al., 2004a) and the coastal sediments of the Daliao River System, China (Lin et al., 2012). Our Fe values were less than those recorded from Azerbaijan, Iran and Kazakhstan coasts (de Mora et al., 2004b).

Pb, As, Zn and Ni values were much higher than those recorded in coastal sediments of the Gulf of Aqaba, the Red Sea coast, the Arabian Gulf and backgrounds of shale and continental crust (Turekian and Wedepohl, 1961; Taylor, 1964). Also these were higher than those recorded from the Daliao River System, China (Lin et al., 2012) and Azerbaijan, Iran and Kazakhstan coasts (de Mora et al., 2004b).

Table 2 illustrates the concentrations of heavy metals in 15 representative seawater samples. Mg was the most abundant major element in the sediment (1325–1525 µg/g), followed by K (408.11–419 µg/g), Ce (8.13–29.49 µg/g), Sr (1.01–3.95 µg/g), As (0.20–0.41 µg/g), Fe (0.08–0.52 µg/g), Zn (0.005–0.023 µg/g), Al (0.003–0.037 µg/g), Mn

(0.006–0.021 µg/g), Pb (0.005–0.006 µg/g), Ni (0.004–0.012 µg/g), Zr (0.002–0.014 µg/g) and Cr (0.003–0.005 µg/g).

Most of the recorded heavy metals in seawaters, especially Fe, Mg, Zr, Zn and As are less than those recorded in the Aranian Gulf, Gulf of Aqaba and north Atlantic (El-Sorogy et al., 2015b; Al-Taani et al., 2014; Donat and Bruland, 1995). Comparisons made between averages of metal contents in coastal sediments and seawater revealed that sediment samples contain 880,868-fold higher iron, 133,833-fold higher nickel, 67,666-fold higher phosphorous, 61,400-fold higher manganese, 26,153-fold higher zinc, 21,818-fold higher aluminum, 1076-fold higher arsenic, 68-fold higher strontium, 65-fold higher chromium, 58-fold higher zircon, 45-fold higher selenium, 0.40-fold higher magnesium and 0.25-fold higher potassium.

As shown in Table 3, Sr was the most abundant major element in bivalve specimens (2420–12,890 µg/g), followed by Mg (1780–3420 µg/g), K (120–390 µg/g), Fe (10.49–456.53 µg/g), Al (0.087–105.3 µg/g), Mn (0.026–22.162 µg/g), Ni (0.005–9.473 µg/g), Zn (0.022–4.117 µg/g), Zr (0.017–3.363 µg/g), As (0.006–2.496 µg/g), Pb (0.003–0.922 µg/g), Ce (0.001–0.949 µg/g) and Cr (0.003–0.499 µg/g).

The recorded values are situated within those recorded in bivalves from the Indian, the Arabian Gulf and the north Adriatic Sea coasts (Kesavan et al., 2013; Al-Jaberi, 2014; de Mora et al., 2004a, 2004b; Martincić et al., 1984). Comparisons made between averages of metal contents in *Ostrea* specimens and seawater reveal that *Ostrea* specimens contain 2803-fold higher strontium, 1431-fold higher aluminum, 372-fold higher manganese, 320-fold higher nickel, 300-fold higher iron,

Table 3

Concentrations of heavy metals in bivalve specimens of Rosetta beach, Egyptian Mediterranean coast.

Samples	Mg	K	Sr	Cr	Zn	Zr	Pb	As	Mn	Ni	Al	Fe	Ce
Sh1	1780	290	4890	0.152	3.078	1.628	0.922	2.496	0.62	4.25	3.54	10.56	0.354
Sh2	2340	120	7390	0.015	3.435	1.675	0.725	1.024	2.21	3.07	4.39	10.84	0.488
Sh3	2440	140	5910	0.321	0.451	1.538	0.458	1.243	0.12	2.02	3.74	11.34	0.485
Sh4	3220	240	2420	0.011	0.085	0.069	0.011	0.024	0.10	0.02	0.34	10.97	0.002
Sh5	1850	270	5540	0.499	4.117	3.363	0.506	1.253	5.15	0.97	18.93	52.56	0.111
Sh6	2420	150	6840	0.334	2.740	2.235	0.338	0.825	3.40	0.64	12.35	34.42	0.072
Sh7	2890	390	5830	0.244	1.982	1.615	0.246	0.590	2.44	0.46	8.75	24.45	0.051
Sh8	3380	140	3300	0.022	0.172	0.139	0.022	0.049	0.21	0.04	0.70	13.98	0.004
Sh9	2460	280	6180	0.005	0.043	0.035	0.005	0.012	0.05	0.01	0.17	10.49	0.001
Sh10	2220	220	3950	0.214	2.818	1.910	0.707	0.628	2.14	5.83	46.89	143.63	0.293
Sh11	2480	240	6650	0.049	2.084	2.236	0.867	1.242	22.16	9.47	105.03	456.53	0.949
Sh12	3070	160	4390	0.003	0.022	0.017	0.003	0.006	0.03	0.01	0.09	20.25	0.074
Sh13	2670	230	5240	0.009	2.778	1.263	0.736	0.563	3.05	1.24	13.47	40.82	0.386
Sh14	3420	130	12,890	0.067	1.243	1.554	0.802	1.325	13.65	0.76	16.45	75.88	0.589
Sh15	3290	340	6490	0.046	0.365	0.296	0.046	0.104	0.44	0.08	1.47	14.18	0.084
Min.	1780	120	2420	0.003	0.022	0.017	0.003	0.006	0.03	0.01	0.09	10.49	0.001
Max.	3420	390	12,890	0.499	4.117	3.363	0.922	2.496	22.16	9.47	105.03	456.53	0.949
Average	2660	223	5860	0.133	1.694	1.287	0.426	0.759	3.72	1.92	15.75	65.72	0.263

Table 4

Minimum, maximum and average values of enrichment factor, geoaccumulation index and contamination factor of the studied heavy metals.

		Pb	Zn	Cr	Ni	Mn	Sr	As	Zr	Ce	Mg	Fe	Al	K
EF	Min.	4.14	0.76	0.0006	2.54	0.17	0.10	4.30	0.0003	6.78	0.01	–	0.001	0.001
	Max.	5.86	1.03	0.0008	3.20	0.19	0.15	7.57	0.0009	7.25	0.01	–	0.001	0.001
	Aver.	4.95	0.87	0.0007	2.88	0.18	0.12	6.06	0.0005	7.03	0.01	–	0.001	0.001
Igeo	Min.	3.10	1.43	–5.69	2.63	–1.09	1.10	3.55	–10.13	4.22	–5.41	1.42	–8.97	–9.38
	Max.	3.46	1.71	–5.45	2.86	–0.99	1.68	4.36	–9.36	4.31	–5.25	1.48	–8.97	–8.26
	Aver.	3.29	1.56	–5.57	2.75	–1.05	1.36	4.03	–9.72	4.27	–5.31	1.45	–8.97	–8.66
CF	Min.	16.67	3.14	0.003	10.40	0.71	0.40	17.54	0.001	28.02	0.04	4.02	0.003	0.002
	Max.	23.82	4.15	0.003	13.15	0.75	0.59	30.70	0.002	29.66	0.04	4.20	0.003	0.005
	Aver.	20.29	3.58	0.003	11.81	0.72	0.48	24.85	0.002	28.88	0.04	4.11	0.003	0.004

Table 5

Correlation matrix of trace metals in the study area.

	Al	Fe	K	Mg	Mn	Ce	Cr	Ni	Sr	Zr	Zn	As	Pb
Al	1												
Fe	.492 ^b	1											
K	–.369 ^a	–.768 ^b	1										
Mg	.452 ^a	.774 ^b	–.593 ^b	1									
Mn	.337	.827 ^b	–.679 ^b	.855 ^b	1								
Ce	.480 ^b	.992 ^b	–.758 ^b	.732 ^b	.787 ^b	1							
Cr	.507 ^b	.950 ^b	–.806 ^b	.807 ^b	.855 ^b	.932 ^b	1						
Ni	.446 ^a	.962 ^b	–.772 ^b	.767 ^b	.816 ^b	.950 ^b	.906 ^b	1					
Sr	.426 ^a	.815 ^b	–.518 ^b	.793 ^b	.771 ^b	.806 ^b	.795 ^b	.809 ^b	1				
Zr	.431 ^a	.841 ^b	–.672 ^b	.799 ^b	.819 ^b	.815 ^b	.778 ^b	.851 ^b	.770 ^b	1			
Zn	.509 ^b	.974 ^b	–.756 ^b	.809 ^b	.874 ^b	.957 ^b	.930 ^b	.952 ^b	.792 ^b	.886 ^b	1		
As	–.024	.414 ^a	–.245	.446 ^a	.593 ^b	.394 ^a	.423 ^a	.426 ^a	.386 ^a	.280	.407 ^a	1	
Pb	.414 ^a	.387 ^a	–.405 ^a	.475 ^b	.380 ^a	.353	.491 ^b	.367 ^a	.442 ^a	.265	.355	.197	1

^a Correlation is significant at the 0.05 level (2-tailed).^b Correlation is significant at the 0.01 level (2-tailed).

258-fold higher zircon, 130-fold higher zinc, 71-fold higher phosphorous, 33-fold higher chromium, 3-fold higher arsenic, 2-fold higher magnesium, 0.54-fold higher potassium and 0.007-fold higher selenium.

Enrichment Factor (EF) analysis (Simex and Helz, 1981; Zhang and Liu, 2002) suggested that the Rosetta coastal sediments are moderately severe enrichment with Ce and As (EF = 5–10), moderate enrichment with Pb (EF = 3–5) and minor enrichment with Ni (EF < 3) indicating that these heavy metals may be enriched as a result of anthropogenic inputs. The remaining metals showed EF values of less than 2, suggesting that they were originated from natural sources. The minimum, maximum and average EF values for the trace metals are tabulated in Table 4.

Based on Geoaccumulation Index (Igeo) of Muller (1981), the studied sediments are unpolluted with Al, K, Mg, Zr, Mn, and Cr (Igeo < 0), moderately polluted with Sr, Zn and Fe (1 < Igeo < 2), moderately to strongly polluted with Ni (2 < Igeo < 3), strongly polluted with Pb (3 < Igeo < 4) and strongly to very strongly polluted with As and Ce (4 < Igeo < 5). Also according to Contamination Factor (CF) of Hakanson, 1980 and Savvides et al., 1995, the sediments are low contaminated with Al, K, Mg, Mn, Cr, Sr, and Zr (Cf < 1), considerably contaminated with Zn and Fe (3 ≤ Cf < 6) and very highly contaminated with Cd, As, Pb, Ni and Ce (Cf ≥ 6). The Igeo and CF values for the trace metals are tabulated in Table 4.

The results of interrelationships matrices between the studied elements are shown in Table 5. The gathering metal agents Fe and Mn are well associated with each other ($r = 0.827$) and each shows positive association with Al ($r = 0.492$ and 0.337 respectively) referring to be all originated from the same source. Both Ce and Zn are well associated with each other ($r = 0.957$). Each shows good association with the essential components of the sediments Fe and Mn, Mg rather than Al. Zinc and Cerium show no significant relationship with all the studied heavy metals. Each of Zr and Cr are well associated with each other ($r = 0.778$) and each shows positive association with Fe, Mg, Mn, Ce, Ni, Sr, and Zn (Table 4).

The natural source of heavy metals Fe, Al, K, Mn, Mg, Zn, Zr, Sr and Cr, which have an enrichment factor < 2 is believed to be the weathering and decomposition of mountain ranges of the Sudan and Ethiopia, where Egypt's heavy-mineral sands are derived from times of flooding at the upper reaches of the Nile, transported down the river and its tributaries to the Mediterranean, and concentrated by the sea (Wendorf and Schild, 1976). Also eroded alluvial material from the upper reaches of the river has undergone natural separation and sorting by rain and wind and dissolution of the soluble minerals.

Anthropogenic source of heavy metals As, Pb, Ce, and Ni which have an enrichment factor ≥ 2 is believed to be a wide range of potential effects of the coastal ecosystems, particularly from point and non-point sources of pollution. Storm water runoff from hinterland and from industrial, sewage, irrigation, and urban runoff are the main sources of heavy metals in the studied sediments.

Comparison of the heavy metal concentration in the studied coastal sediments, seawaters and bivalves with other samples from the Arabian Gulf, Red Sea and abroad coasts indicated that the studied samples have higher concentrations in certain heavy metals like Fe, Pb, As, Zn and Ni. Enrichment factor, geoaccumulation index and contamination factor indicated an enrichment of As and Pb, Ce and Ni as a result of anthropogenic inputs, the studied sediments are moderately polluted with Sr, Zn and Fe; moderately to strongly polluted with Ni; strongly polluted with Pb; and strongly to very strongly polluted with Ac and Ce. The sediments are low contaminated with Al, K, Mg, Mn, Cr, Sr, and Zr, are considerably contaminated with Zn and Fe; and very highly contaminated with As, Pb, Ni and Ce. The natural source of the heavy metals which have an enrichment factor < 2 is believed to be the weathering and decomposition of mountain ranges of the Sudan and Ethiopia as well as heavy metals driven from erosion of alluvial material in the upper reaches of the river. The anthropogenic source of heavy metals As, Pb, Ce, and Ni which have an enrichment factor ≥ 2 is believed to be the storm water runoff from hinterland and metals produced from industrial, sewage, irrigation, and urban runoff.

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