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Evaluation of Heavy Metals Pollution in Seawater, Suspended Particulate Matter and Sediment from Abu-Qir Bay, Alexandria, Egypt

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ABSTRACT

The level of heavy metals (Mn, Fe, Co, Ni, Cu, Zn, Pb, and Cd) in seawater, suspended particulate matter (SPM) and sediment along Abu-Qir Bay coast, Alexandria, Egypt, were evaluated by selecting nine stations as hot spots during winter 2020. Resuls showed that, the average concentrations of the investigated heavy metals followed the following order: Fe > Zn > Mn > Ni > Pb > Co > Cu > Cd in seawater, Fe > Zn > Mn > Pb > Cu > Ni > Co > Cd in SPM, and Fe > Mn > Zn > Ni > Co > Cu > Pb > Cd in sediment. Moreover, the distribution coefficient (Kd) revealed that, Fe was highly stable in sediments, and the log (Kd) value was 6.521 in all water bodies. In addition, geo-accumulation index (I_{geo}), pollution load index (PLI), modified degree of contamination (mC_d), potential ecological risk index (RI), as well as the mean effect range median quotient ($MERM-Q$) were calculated. Calculations recorded that, the values of I_{geo} for Cd in surface sediments ranged between 1 and 3 indicating that, the study sites were moderate to strongly contaminated with Cd. The mC_d values of Ni, Cu, Zn, Pb, and Cd ranged between 1.32 and 4.24, reflecting a low to moderate degree of contamination. The studied metals presented a moderately toxic ecological risk ($150 \leq RI < 300$) except for St **5** & St **8,** that showed a considerable toxic ecological risk ($RI > 300$). Based on $MERM-Q$ classification, all samples had medium-low toxicity; Ni, Cu, Zn, and Pb may be originated from anthropogenic sources, whereas Cd emerged from natural sources with some contribution of anthropogenic source. Redundancy analysis (RDA) revealed that the TOC % and fine sand % of the sediment had a certain influence on the Fe enrichment. The data of the water quality index (WQI) revealed that St **2**, St **3**, St **4**, St **5** & St **6** were slightly polluted, and could be considered hot spots.

INTRODUCTION

The contamination of coastal and marine environments by heavy metals is a worldwide problem not only in the developing countries, but also in the developed ones as well (**Boran *et al.*, 2010**). Industrial and agricultural activities, vehicle emissions, domestic waste, and shipping traffic, especially in and close to harbors, are causes of heavy metal pollution (**Masindi & Muedi, 2018**). Heavy metals are toxic, long persistent, not easily oxidized, degraded, taken away, or converted to less harmful components through biological or chemical processes, which severely damage marine environments (**Sun *et al*., 2020**).

Coastal systems like bays receive considerable quantities of heavy metals (**Cao *et al*., 2018**), among which is Abu-Qir Bay, a shallow semi-circular basin situated 35 km east of Alexandria city. It extends for about 50 km from Abu-Qir Head-Montazah in the west to Rosetta mouth of the Nile River in the east, and northwards to about 40 km in the Mediterranean Sea (Fig 1). The surface area of the Bay is about 360 km^2 and its water volume is 4.3 km^3, while the maximum depth reaches 22 m (**Abdel Ghani *et al*., 2013**). It is noteworthy that the Bay receives several discharges as; freshwater from Rosetta mouth of the Nile River loaded by nutrients, industrial and domestic wastes from El-Tabia pumping station (TPS), and the outlet of Lake Edku (Boughaz El-Maddya) carrying trace metals, pesticides, humic acids, and nutrients. In addition, it receives industrial wastewaters from 22 different factories of paper, fertilizer, food processing and canning, and textile manufacturing industries. The Bay is also exposed to pollution from the activities of gas production and Abu-Qir Electrical Power Station (EPS) as well as fishing boats.

Hence, sediment should be analyzed to assess levels of contamination by heavy metals in the aquatic environment because heavy metal concentrations in water are sometimes lower than the detection limits (**Salati and Moore, 2010**). Therefore, sediments are considered as sensitive indicators to monitor contaminants in the aquatic systems (**Pekey *et al*., 2004**). Furthermore, high concentrations of heavy metals in sediment can indicate anthropogenic sources (**Tylmann, 2011**).

The main objective of the current study was to the assess the heavy metals contamination in Abu-Qir Bay sediment. That target was achieved by identifying the sources of pollution, evaluating the environmental impact of contaminated sediments, and setting an application of principal component analysis and RDA to investigate the distribution and source relationship of the heavy metals in seawater, particulate matter, and sediments. Moreover, the current study was conducted to evaluate the water quality index (WQI) of Abu-Qir Bay through measuring some physical and chemical parameters; the temperature of water, salinity, pH, dissolved oxygen (DO), oxidizable organic matter (OOM), and total alkalinity (T. Alk.) in the surface water as well as heavy metals, (Mn, Fe, Co, Ni, Cu, Zn, Pb, and Cd) in water and suspended particulate matter (SPM). This task was managed by studying nine stations (St **2** to St **10**) in front of Abu-Qir city, Abu-Qir EPS, Boughaz El-Maddya, TPS, and the paper and fertilizer factories in Abu-Qir area.

MATERIALS AND METHODS

1. Sampling

Triplicate surface sediment (7 samples), and seawater (9 samples) were collected from nine stations during winter 2020, representing the hot spots influenced by human activities; industrial and agriculture discharge along Abu-Qir Bay, Alexandria (Figure 1). The Sediment samples were collected using Van-Veen grab sampler, packed in clean polyethylene bags, and preserved at 4°C in an ice box. Before analysis, the samples were dried at room temperature and grinded using an agate mortar. Seawater samples were collected using PVC Niskin's bottle, packed in pre-washed plastic bottles, and stored at 4°C for preservation and laboratory analysis to evaluate physicochemical parameters and heavy metals.

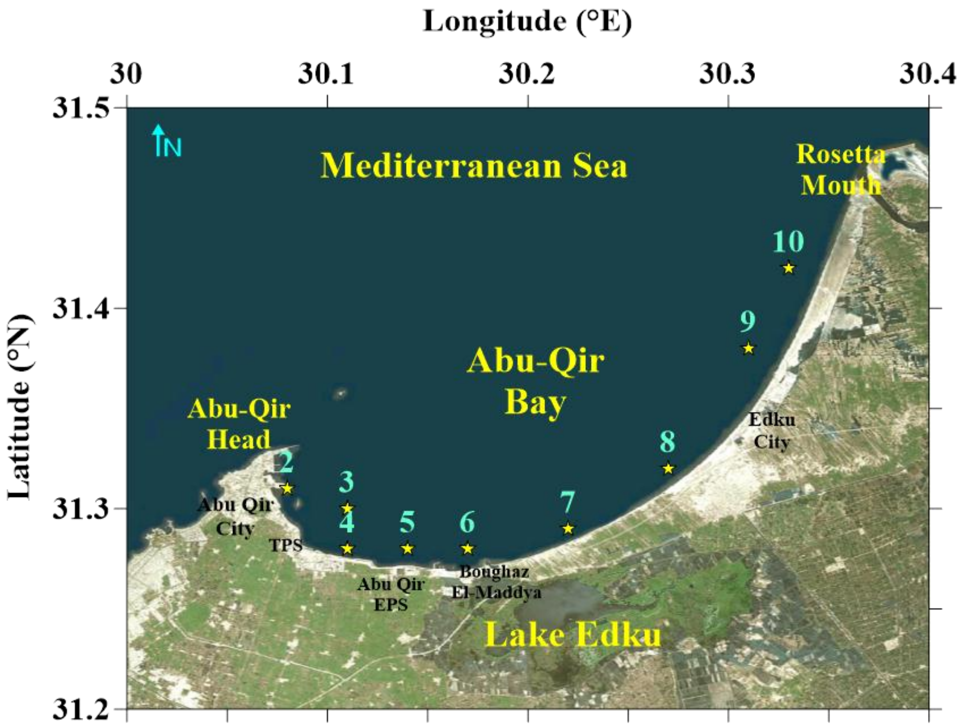

Figure 1. Abu-Qir Bay area and sampling locations.

2. Analytical methods

2.1. Physicochemical parameters of seawater

The water sample was sub-sampled and immediately the following parameters were measured in-situ i**)** water temperature, pH and salinity using calibrated CTD device, model, (YSI 6000). **ii)** Fixing of dissolved oxygen (DO). Then, at laboratory DO was determined by using Winkler's method (**Grasshoff *et al*., 1999**), Oxidizable organic matter (OOM) was determined using the method depicted by **FAO (1976),** and **t**otal alkalinity (T. Alk.) was determined according to **APHA (1995)**.

2.2. Dissolved and particulate heavy metals in seawater

The volume of water samples (3-4 L) was filtered using 0.45 µm membrane filter paper. The dissolved metal ions were determined after preconcentration using the cation exchange resin (chelex-100 in ammonia form) following the decription of **Gao *et al*. (2002)**. The membrane filters with their content of suspended particulate matter (SPM) were washed several times, and then dried at 65 °C for 48 hrs to a constant weight. Each dried membrane filter was placed in a Teflon cup, a mixture of HNO_3-$HClO_4$-HF (3:2:1) (6 ml) was added, and the Teflon cup was then heated. After complete digestion, the sample was evaporated until dryness, then 6N HNO_3 (1 ml) was added and the volume was completed with 0.1N HCl to the final volume of 25 ml (**Abdullah & Royle, 1974**). The concentrations of Mn, Fe, Co, Ni, Cu, Zn, Pb, and Cd were determined by ICP-OES (Agilent). The results were expressed in µg/L for both seawater and SPM.

2.3. Heavy metals in Sediments

The heavy metals in surface sediments were digested according to the method described by **Oregioni and Astone** (**1984**). Exact weight of each dried sediment sample (about 0.25-0.3 g) was placed in a Teflon beaker. Concentrated HNO_3 (3 ml) was added dropwise to the sample. The outcome was then heated at 80 °C until dryness, then a mixture of HNO_3/$HClO_4$/HF (3:2:1) (5 ml) was added to the sample. After complete digestion, temperature was increased gradually to 120 °C to evaporate the $HClO_4$ residue. Afterwards the sample was cooled at room temperature, and the residue was rinsed with 0.1N HCl (5 ml). The sample was filtered and washed several times with deionized water, and the final volume was adjusted to 25 ml. The concentrations of Mn, Fe, Co, Ni, Cu, Zn, Pb, and Cd were measured by ICP-OES (Agilent). The results were expressed in mg/kg dry weight.

2.4. Geochemical parameters of sediments

The grain size analysis was determined by the dry sieving the method of **Folk (1974**), and the sediment texture was classified according to **Folk** (**1980**). The total organic carbon (TOC) was determined according to the method reported by **Gaudette and Flight** (**1974**). According to that method, the sediment sample was oxidized by $K_2Cr_2O_7$ and concentrated H_2SO_4, then the excess dichromate was titrated against $(NH4)_2Fe(SO4)_2 \cdot 6H_2O$ solution and diphenylamine was used as an indicator.

2.5. Quality control and quality assurance

The reagents used during the present study were of analytical grade (Merck), triplicate analysis for samples showed good accuracy. All glassware and plastic vessels were washed with solution of diluted HNO_3 and deionized water, then they were dried. To test the precision, blanks were treated using the same reagents as samples. IAEA certified reference material, for marine sediment (SD-M-2/TM), was employed for quality control. In this study the precision of measurements for all metals was within 10 % RSD. In terms of preparing the solution, seawater certified reference materials CASS-4 from National Research Council (NRC) in Canada, were treated in a way similar to that used for the other samples. The recovery of the selected metals ranged from 92 to 110 % and the precision was within 10 % RSD.

2.6. Statistical analysis

Principal component analysis (PCA), as a varimax rotated was conducted with IBM-SPSS program (version 22) applying Kaiser normalization.

RESULTS AND DISCUSSION

1. Seawater physicochemical parameters

The horizontal distributions of physicochemical parameters of Abu-Qir Bay seawater are presented in Figure (2). Abu-Qir seawater temperature ranged from 16.10 to 17.16 °C, and the max value was found near Abu-Qir EPS (St **5**). The recorded temperature values flowed the behavior of winter season (**Radwan, 1996**; **Zakaria *et al*., 2019**). The salinity ranged from 37.24 to 38.21, and the max value was observed at St **5**. Changes in salinity may be attributed to fresh - marine water mixing. The salinity of Abu-Qir Bay coastal water was influenced by the discharge of wastewater from the surrounding urbanized area (**Zakaria *et al*., 2019**). The pH values of seawater ranged between 7.13 and 8.61, indicating that it was in the slightly alkaline side. This may be because of increasing phytoplankton and the photosynthesis processes that resulted in higher pH values as well as an increase in the dissolved oxygen concentrations (**Das *et al*., 1997**). Moreover, the values of DO ranged between 8.64 mg/L at Sts **2**, **3**, **4** & **5** and 10.24 mg/L at Sts **9** & **10**. Based on temporal variations, the DO in surface water recorded the highest values that could be related to the entry of oxygen from the atmosphere into the surface water and then to the algal photosynthesis (**Das *et al*., 1997**). Whereas the low values of DO may be due to the decomposition of organic matter and respiration of marine organisms as well as the biochemical reactions (**Das *et al*., 1997**). It was noticed that, the average value of OOM was 2.92, with a maximum value of 10.24 mgO_2/L at St **2** and a minimum of 0.32 mgO_2/L at Sts **5**, **7** & **8**. The high value of OOM at St **2** could be attributed to the impact of huge organic pollutants flow from TPS and Rosetta mouth **(Alam El-Din & Al-Hogaraty, 2001)**. The values of T. Alk. ranged between 3.00 mmol/L at St **8** and 4.80 mmol/L at St **6**, with an average value of 3.29 mmol/L. The change in pH, salinity, DO, and alkalinity may be due to the water flow of TPS (Figure 2).

2. Heavy metals distribution in seawater

The fluctuations of heavy metals concentration in seawater of Abu-Qir Bay are presented in Table (1). The range and average concentrations (µg/L) were 1.467-12.738 (5.716) for Mn, 7.242-22.594 (14.502) for Fe, 0.508-3.119 (1.632) for Co, 1.808-9.398

(5.103) for Ni, 0.458-3.738 (1.432) for Cu, 4.600-18.333 (12.014) for Zn, 1.625-9.325 (4.733) for Pb, and 0.333-2.090 (0.874) for Cd. Though the highest concentrations of Mn, Co, Cu and Pb were detected at St **3**, yet the highest levels of Zn and Cd were detected at St **10** & St **2,** respectively. Remarkably, St **5** showed the highest concentrations of Fe and Ni. On the other side, St **8** presented the lowest levels of Mn, Co, Ni, Cu, Zn, Pb and Cd, whilst the lowest concentration of Fe was recorded at St **4**.

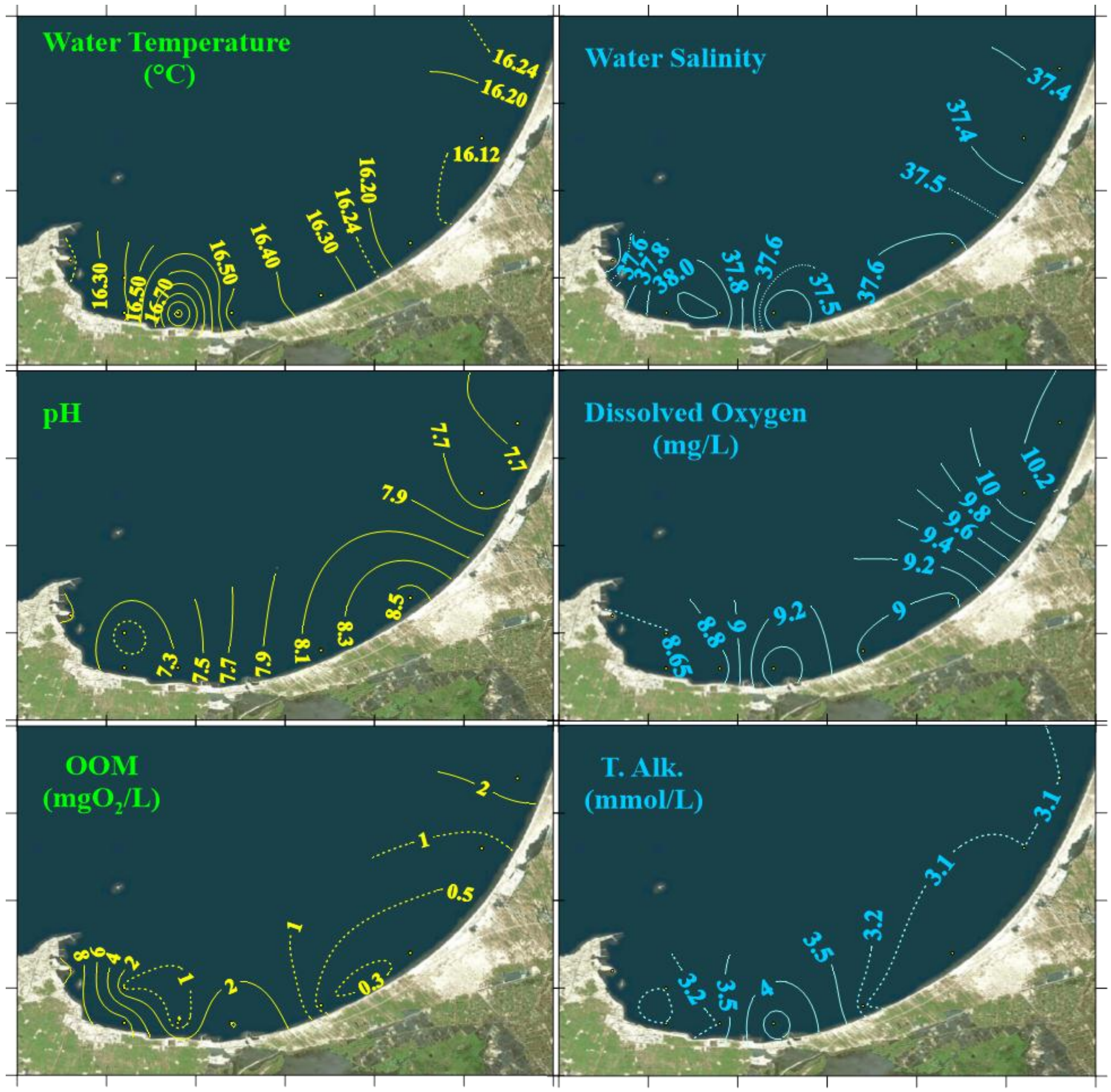

Figure 2. Horizontal distribution of some physicochemical parameters of Abu-Qir Bay seawater.

High values of Ni (8.783 µg/L), Pb (7.079 µg/L) and Cd (1.421 µg/L) were detected at St **7**, St **10** & St **3,** respectively. Besides, high concentrations of Zn (18.111 & 16.159 µg/L) were measured at St **3** & **6,** respectively. Low values of Mn (1.483 µg/L), Cu (0.542 µg/L) and Zn (4.658 µg/L) were recorded at St **4**. Moreover, low concentrations of Cd (0.458 & 0.471 µg/L) were measured at St **4** & St **6**, respectively. In Abu-Qir Bay seawater, Fe and Zn were the prevailing metals, whilst Cd was the least abundant (Table

1). The average concentrations of the studied heavy metals followed the successive sequence of Fe > Zn > Mn > Ni > Pb > Co > Cu > Cd.

3. Heavy metals distribution in SPM

The fluctuations of heavy metal concentrations in SPM of Abu-Qir Bay seawater were presented in Table (1). The range and average concentrations (µg/L) were 1.811-12.645 (5.502) for Mn, 118.243-414.754 (277.282) for Fe, 0.092-0.858 (0.440) for Co, 1.115-3.575 (1.733) for Ni, 0.542-4.833 (1.886) for Cu, 3.325-26.652 (10.377) for Zn, 0.542-4.659 (2.182) for Pb, and 0.008-0.292 (0.110) for Cd. The highest concentrations of Mn, Cu, Zn and Pb were detected at St **6**, whereas the highest levels of Co and Ni were observed at St **4**. Meanwhile, St **2** & St **7** showed the highest values of Fe and Cd, respectively. On the other side, St **7** presented the lowest concentrations of Co, Cu, Zn and Pb, while the lowest values of Mn and Fe were recorded at St **9**. In addition, St **2** & St **5** presented the lowest values of Ni and Cd, respectively. Markedly, high values of Zn (19.000 µg/L) and Pb (4.413 µg/L) were detected at St **10**. Besides, high concentrations of Fe (345.635, 394.167 & 338.281 µg/L) were recorded at St **3**, St **4** & St **5**, respectively. The average concentrations of the studied heavy metals in SPM followed the sequence of: Fe > Zn > Mn > Pb > Cu > Ni > Co > Cd.

Table 1. Heavy metals concentrations in seawater and SPM of Abu-Qir Bay.

St No	Heavy metals concentrations (µg/L)															
	Mn		Fe		Co		Ni		Cu		Zn		Pb `		Cd	
	D	P	D	P	D	P	D	P	D	P	D	P	D	P	D	P
2	3.434	6.639	16.311	414.754	2.139	0.418	4.631	1.115	1.303	2.279	8.730	7.148	5.541	1.418	2.090	0.164
3	12.738	4.413	16.960	345.635	3.119	0.262	5.802	1.349	3.738	2.611	18.111	12.937	9.325	3.135	1.421	0.087
4	1.483	6.050	7.242	394.167	1.025	0.858	4.125	3.575	0.542	1.183	4.658	5.067	2.708	0.900	0.458	0.183
5	5.328	6.359	22.594	338.281	2.078	0.227	9.398	1.773	1.375	0.703	13.367	3.773	5.219	0.586	0.828	0.008
6	7.580	12.645	12.514	299.638	0.993	0.659	4.536	2.181	1.261	4.833	16.159	26.652	2.652	4.659	0.471	0.029
7	3.792	2.658	16.925	220.417	1.533	0.092	8.783	1.317	1.083	0.542	11.558	3.325	3.975	0.542	0.667	0.292
8	1.467	3.283	13.125	160.833	0.508	0.417	1.808	1.442	0.458	1.867	4.600	10.042	1.625	2.508	0.333	0.025
9	9.074	1.811	10.709	118.243	1.709	0.554	3.439	1.162	1.466	1.230	12.608	5.446	4.473	1.480	0.628	0.095
10	6.548	5.659	14.135	203.571	1.579	0.476	3.405	1.683	1.659	1.722	18.333	19.000	7.079	4.413	0.968	0.103
Min	1.467	1.811	7.242	118.243	0.508	0.092	1.808	1.115	0.458	0.542	4.600	3.325	1.625	0.542	0.333	0.008
Max	12.738	12.645	22.594	414.754	3.119	0.858	9.398	3.575	3.738	4.833	18.333	26.652	9.325	4.659	2.090	0.292
Average	5.716	5.502	14.502	277.282	1.632	0.440	5.103	1.733	1.432	1.886	12.014	10.377	4.733	2.182	0.874	0.110
***Marine Guideline**					1.0		70.0		1.3		15.0		4.4		5.5	
The Criterion Continuous Concentration (CCC)**							7.7		2.6		76.6		8.1		8.7	
*****Minimal Risk Conc.**	20.0		50.0				2.0		10.0		20.0		10.0			

D: Dissolved heavy metals, P: Heavy metals in SPM, * Australian water quality guideline values suitable for the protection of 95 % of all species (**ANZECC/ARMCANZ, 2000**), ** National Recommended Water Quality Criteria (**US EPA, 2006**), ***World quality criteria (**WQC, 1972**).

4. Sediment characterization

The results of sediment grain size and total organic carbon (TOC %) of surface sediment of Abu-Qir Bay are listed in Table (2). The results of grain size showed a little variation and a uniform distribution from fine sand to silt sand. Finding the fine sediment can be due to human and terrestrial activities (**El Nemr & El-Said, 2017**). As shown in table (2), sand content varied from 99.89 % at St **8** & St **9** to 23.74 % at St **2**. Whereas silt content varied from 68.35 % at St **5** to 0.10 % at St **9**. TOC % in Abu-Qir Bay sediment, recording rangee from 0.59 % at St **9** to 1.95 % at St **7,** with an average value of 1.26 %. Except for St 5 & St 9, the TOC % recorded for all sediment samples was > 1, which is slightly higher than LEL and SEL (**Persaud *et al.*, 1992**).

5. Heavy metals in sediments

The variations of heavy metal concentrations in the surface sediment of Abu-Qir Bay are given in Table (2. The range and average concentrations (mg/kg) were 416.477-1803.088 (862.857) for Mn, 29318.380-81203.242 (52549.293) for Fe, 29.642-68.579 (40.733) for Co, 47.341-112.765 (71.957) for Ni, 14.288-49.594 (30.021) for Cu, 63.975-204.539 (115.805) for Zn, 17.384-46.065 (28.186) for Pb, and 1.208-4.599 (2.570) for Cd. The highest concentrations of Zn, Pb, and Cd were detected at St **5**, while St **8** revealed the highest contents of Fe, Co, and Ni. The highest levels of Mn and Cu were recorded at St **6** & St **10**, respectively. On the other side, St **7** showed the lowest values of Mn, Co, Ni, Cu, Zn and Cd. The lowest concentrations of Fe and Pb were recorded at St **6** & St **10**, respectively.

Table 2. Heavy metal concentrations, textural parameters, and TOC in sediment of Abu-Qir Bay.

Station number	Heavy metal concentrations								Textural parameters			
	Mn	Fe	Co	Ni	Cu	Zn	Pb	Cd	Sand	Silt	Clay	TOC
	mg/kg								%			
2	583.658	58657.879	38.721	70.252	31.756	97.963	25.614	2.397	23.74	66.78	9.48	1.25
5	559.107	32795.537	31.665	55.941	26.689	204.539	46.065	4.599	25.06	68.35	6.59	0.86
6	1803.088	29318.380	46.606	85.494	36.338	139.601	34.008	2.986	77.42	20.74	1.84	1.46
7	416.477	41135.911	29.642	47.341	14.288	63.975	17.913	1.208	94.39	5.06	0.55	1.95
8	1300.655	81203.242	68.579	112.765	29.847	129.130	38.809	3.351	99.89	0.11	0.00	1.72
9	528.130	77345.483	33.043	65.531	21.632	73.455	17.512	1.268	99.89	0.10	0.01	0.59
10	848.886	47388.621	36.875	66.376	49.594	101.972	17.384	2.182	99.64	0.36	0.00	1.02
Min	416.477	29318.380	29.642	47.341	14.288	63.975	17.384	1.208	23.74	0.10	0.00	0.59
Max	1803.088	81203.242	68.579	112.765	49.594	204.539	46.065	4.599	99.89	68.35	9.48	1.95
Average	862.857	52549.293	40.733	71.957	30.021	115.805	28.186	2.570	74.29	23.07	2.64	1.26
Log *Kd*	5.213	6.521	4.455	4.190	4.388	3.982	3.819	3.503				
ERL[1]				20.900	34.000	150.000	46.700	1.200				
ERM[1]				51.600	270.000	410.000	218.000	9.600				
TEL[2]		20000.000		15.900	16.000	124.000	30.200	0.700				
PEL[2]				42.000	108.000	271.000	112.000	4.200				
SEL[2]		40000.000		75.000	110.000	820.000	250.000	10.000				
Continental Crust Average[3]		35000.000		50.000	25.000	71.000	16.000	0.098				
Shale Average[4]		47200.000		68.000	45.000	95.000	20.000	0.300				
***LEL**												1.00
****SEL**												10.00

1**Long *et al*. (1995)**; [2]**MacDonald *et al*. (2000)**; [3]**McLennan and Taylor (1999)**; [4]**Turekian and Wedepohi (1961)**. *LEL and **SEL refer to the lowest, and severe effect levels, respectively (**Persaud *et al*., 1992**).

In addition to the highest levels of Fe, Co, and Ni, St **8** revealed high values of both Pb (38.809 mg/kg) and Cd (3.351 mg/kg) as well. Whereas low values of Zn (73.455 mg/kg) and Cd (1.268 mg/kg) were recorded at St **9**. Moreover, low concentrations of Pb (17.512 & 17.913 mg/kg) were measured at St **9** & St **7**, respectively. It was noticed that Mn and Fe were the most abundant heavy metals, while Cd was the least one (Table 2). The average concentrations of the investigated metals in sediments of Abu-Qir Bay followed the sequence presented as: Fe > Mn > Zn > Ni > Co > Cu > Pb > Cd.

Comparing heavy metal levels in sediments of Abu-Qir Bay with those of other studies is presented in Table 3. Results showed that, Mn and Fe metals recorded the highest levels in the present study compared to the mentioned values in Table (3). The exception was only for Mn values reported in Abu-Qir Bay (**Faragallah *et al*., 2004**) which were higher than those found in the current study. The observed concentrations of Cu were lower than those recorded in Abu Qir-Bay (**Faragallah *et al*., 2004**), the Egyptian coast along the Mediterranean Sea, the Eastern Harbour and the Western Harbour. On the other hand, they were higher than those recorded in Abu-Qir Bay (**Abdel Ghani *et al*., 2013**), eastern coast of Alexandria, eastern Mediterranean coast of Egypt, western part of the Egyptian Mediterranean Sea and El-Max Bay. The recorded concentrations of Zn were lower than those observed in Abu-Qir Bay (**Faragallah *et al*., 2004**), the Eastern Harbour, the Western Harbour and El-Max Bay, while they were higher than those observed in Abu-Qir Bay (**Abdel Ghani *et al*., 2013**), eastern coast of Alexandria, eastern Mediterranean coast of Egypt, western part of the Egyptian Mediterranean Sea and Egyptian coast along Mediterranean Sea. Additionally, the Pb gave concentrations lower than those mentioned in eastern coast of Alexandria, Egyptian coast along Mediterranean Sea, the Eastern Harbour, the Western Harbour and El-Max Bay. However, the upper- mentioned concentrations were higher than those mentioned in Abu-Qir Bay (**Abdel Ghani *et al*., 2013**), eastern Mediterranean coast of Egypt and western part of the Egyptian Mediterranean Sea. Markedly, the Cd levels in Abu-Qir exceeded those recorded in the eastern Mediterranean coast of Egypt, western part of the Egyptian Mediterranean Sea, the Eastern Harbour and the Western Harbour. The Cd concentrations were lower than those recorded in Abu-Qir Bay (**Abdel Ghani *et al*., 2013**), Egyptian coast along Mediterranean Sea and El-Max Bay.

6. Evaluation of the pollution environmental impact

6.1. Distribution coefficient (Kd)

The distribution coefficient is one of the important parameters for evaluating the potential migration of heavy metals in a sediment or the suspended matter that contacted with water column. Quantitatively, the distribution coefficient Kd is calculated as the ratio between concentrations of heavy metal in sediments and water according to following equation:

$$Kd = [Metal\ (S)]/[Metal\ (W)] \qquad (1)$$

Where, Kd (L/kg) is the distribution coefficient, $Metal\ (S)$ is the concentration of the heavy metal in sediments (mg/kg) and $Metal\ (W)$ is the concentration of the heavy metal in water column (mg/L). **Nabelkova and Kominkova (2012)** mentioned that Kd provides useful information about risk assessment. In case the $Log(Kd)$ > 5, the propability that the metal has affinity to bind in sediments is indicated. While the values in the range 3 < $Log(Kd)$ < 4 would indicate that the metal is released from the sediment into the water body, and $Log(Kd)$ < 3 indicates prevailing heavy metal in a water body.

Table 3. Comparing heavy metal values in sediment of Abu-Qir Bay with those of other studies in mg/kg.

Location	Mn	Fe	Co	Ni	Cu	Zn	Pb	Cd	Reference
Abu-Qir Bay	416.477-1803.088	29318-81203	29.642-68.579	47.341-112.765	14.288-49.594	63.975-204.539	17.384-46.065	1.208-4.599	Present study
Abu-Qir Bay	115.030–479.600	90–35890	-----	-----	10.240–22.850	25.230–104.080	1.900–16.790	0.310–4.890	**Abdel Ghani *et al.* (2013)**
Abu-Qir Bay	800–2520	12920–14480	-----	-----	38–561	118–1779	-----	-----	**Faragallah *et al.* (2004)**
Eastern coast of Alexandria	20.880-590.930	385-12501	-----	-----	5.480-37.590	4.150-198.060	16.980-81.750	-----	**El Zokm *et al.* (2020)**
Eastern Mediterranean coast of Egypt	10.030–119.370	50.630–2182.100	-----	-----	0.550–17.990	1.860–14.030	8.540–19.250	0.100–0.310	**Khaled *et al.* (2009)**
Western part of the Egyptian Mediterranean Sea	32.270–108.920	709.100–1433.900	-----	-----	26.529–33.330	26.270–112.070	20.670–35.620	0.524–0.920	**Ahdy and Khaled (2009)**
Egyptian coast along Mediterranean Sea	13.480–1384.270	124–36868.800	-----	-----	6.940–192.500	16.610–166.540	49.900–109.770	3.700–53.410	**El Nemr *et al.* (2007)**
The Eastern Harbou	7.800–188.970	10–30240	-----	-----	3.800–129.200	2.900–206.890	1.300–112.090	0.300–1.830	**Abdel Ghani *et al.* (2013)**
The Western Harbour	139–317	8819–36140	-----	-----	39–207	58.500–382	38–1070	0.610–2.440	**Mostafa *et al.* (2004)**
El-Max Bay	159.310–361.980	512.860–2490.590	-----	-----	3.290–47	51.350–448.340	8.800–88.680	2.540–7.540	**Abdallah (2008)**

The overall mean of $Log(Kd)$ for each metal had been estimated considering the water bodies: Mn (5.213), Fe (6.521), Co (4.455), Ni (4.190), Cu (4.388), Zn (3.982), Pb (3.819) and Cd (3.503), Table 2. The following order: Fe > Mn > Zn > Co > Cu > Ni > Zn > Pb > Cd has been observed. Fe showed the highest $Log(Kd)$ values in all water bodies, indicating that Fe is highly stable in sediments. Whereas $Log(Kd)$ for the Cd values indicated that the Cd is mainly contained in water body, suggesting that the studied water bodies can pose higher risk of Cd. To a certain degree of stability ($Log(Kd)$ > 4), metals can cause a threat to bottom organisms (**Li *et al*., 2017**). Generally, the metals may be released to water under certain conditions, and may endanger the entire aquatic ecosystem, given their toxicity profile (**Li *et al*., 2017**).

6.2. Geo-accumulation index (I_{geo})

Geo-accumulation index (I_{geo}) was proposed by **Müller** (**1981**) to evaluate the degree of metals contamination in sediments by comparing the concentrations of metal with the pre-industrial levels and calculated using the successive equation (2):

$$I_{geo} = Log_2\,[C_n/(1.5)B_n] \qquad (2)$$

Where, C_n is the metal (n) concentration measured in sediment, B_n is the geochemical background value of the metal (n), and factor 1.5 is a factor concerning the background level variations due to lithogenic impacts. In the present study, the average shale values stated by **Turekian and wedepohl** (**1961**) were used as the background levels of heavy metals. Seven classes of (I_{geo}) index ranging from 0-6 were classified by **Müller (1981),** and organized from the uncontaminated to the highly contaminated sediments as described in Table (4).

It can be deduced from Tables (4 & 5) that, with reference to Müller scale, most of the I_{geo} values of Cd in surface sediments through all study areas were between 1 and 3 (1 < I_{geo} < 3; class 3), indicating that those sites are moderately to strongly polluted with Cd. On the other hand, I_{geo} values of Ni, Cu, Zn, and Pb in surface sediments of most study areas were apparently lower than 0 (I_{geo} < 0; class 0), indicating a practically unpolluted status.

Table 4. Classification of I_{geo} (Müller, 1981). The modified degree of contamination (mC_d) (Abrahim & Parker, 2007), and potential ecological risk index, RI (Hakanson, 1980).

I_{geo}	I_{geo} class	Designation of sediment quality	mC_d	mC_d class	Contamination degree	RI values	Risk Intensity
< 0	0	Unpolluted	< 1.5	0	Unpolluted	RI < 150	Low ecological risk
0-1	1	Unpolluted to moderately polluted	1.5 ≤ mC_d < 2	1	Slightly polluted	150 ≤ RI < 300	Moderate ecological risk
1-2	2	Moderately polluted	2 ≤ mC_d < 4	2	Moderately polluted	300 ≤ RI < 600	Considerable ecological risk
2-3	3	Moderately to strongly polluted	4 ≤ mC_d < 8	3	Moderately to heavily polluted	RI > 600	Very high ecological risk
3-4	4	Strongly polluted	8 ≤ mC_d < 16	4	Heavily polluted		
4-5	5	Strongly to extremely polluted	16 ≤ mC_d < 32	5	Severely polluted		
> 5	6	Extremely polluted	≥ 32	6	Extremely polluted		

6.3. Pollution Loading Index (PLI)

Pollution loading index (PLI) was proposed by **Tomlinson *et al*. (1980)** and **Satyanarayana *et al*. (1994)** to evaluate the integrated pollution status of synergetic groups for a particular station. If the PLI value > 1, a polluted condition is indicated, while PLI < 1 would signify no metal pollution. This parameter is expressed in the following equation:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots\ldots\ldots. \times CF_n)^{1/n} \qquad (3)$$

Where, n is the number of selected metals, and CF is the contamination factor. CF can be calculated from the following equation (4):

$$CF = \frac{\text{Metal concentration in the sediments}}{\text{Background value of the metal}} \qquad (4)$$

The contamination factor (CF) is used to evaluate the anthropogenic input of metals to sediment. The average shale value is the background concentration of the selected metal as given by **Turekian and wedepohl** (**1961**).

The calculated PLI values for the studied metals are listed in Table (5) and represented in Figure (3). Most of PLI values of surface sediments fall above 1 (PLI > 1), indicating that they were polluted sites. On the other hand, St **7** was an unpolluted site (PLI < 1).

Table 5. Evaluation of heavy metals pollution in sediments from Abu-Qir Bay based on I_{geo}, PLI, mC_d, RI, and $MERM\text{-}Q$ indices.

Station number	I_{geo}					PLI	mC_d	RI	$MERM\text{-}Q$
	Ni	Cu	Zn	Pb	Cd				
2	-0.54	-1.09	-0.54	-0.23	2.41	1.50	2.41	255.79	0.42
5	-0.87	-1.34	0.52	0.62	3.35	2.06	4.24	480.64	0.47
6	-0.25	-0.89	-0.03	0.18	2.73	1.91	3.04	318.87	0.52
7	-1.11	-2.24	-1.16	-0.74	1.43	0.88	1.32	131.06	0.27
8	0.14	-1.18	-0.14	0.37	2.90	2.01	3.36	357.77	0.63
9	-0.64	-1.64	-0.96	-0.78	1.49	1.06	1.46	139.16	0.35
10	-0.62	-0.44	-0.48	-0.79	2.28	1.49	2.26	234.05	0.41
Min	-1.11	-2.24	-1.16	-0.79	1.43	0.88	1.32	131.06	0.27
Max	0.14	-0.44	0.52	0.62	3.35	2.06	4.24	480.64	0.63
Average	-0.55	-1.26	-0.40	-0.19	2.37	1.56	2.58	273.91	0.44

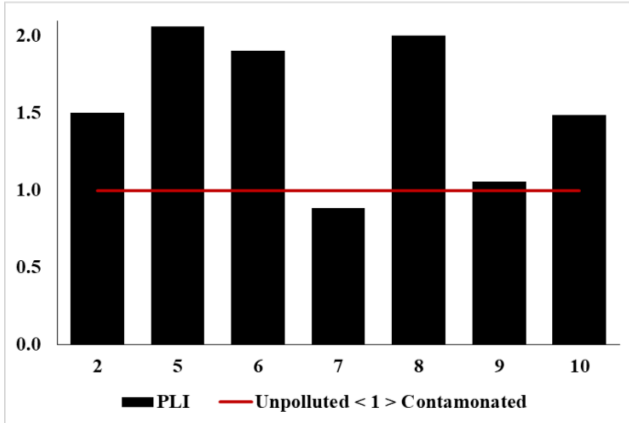

Figure 3. Pollution Loading Index (PLI) of surface sediments from Abu-Qir Bay.

6.4. Assessment according to modified degree of contamination (mC_d)

Abrahim and Parker (**2007**) suggested a modified degree of contamination (mC_d) to evaluate the overall contamination of sediment sample in the presence of multimetal rather than the contamination caused by one metal ion for one sample. According to equation (5) mC_d can be calculated.

$$mC_d = \frac{\sum_{i=1}^{n} C_f^i}{n} \quad (5)$$

Where, n is the number of selected metals and C_f^i refers to the contamination factors proposed by **Hakanson** (**1980**). Based on mC_d values, seven degrees of contamination were classified by **Abrahim and Parker** (**2007**) as shown in Table (4). The mC_d values of the selected metals through all study areas are represented in Table (5) and Figure (4). The values of mC_d for Ni, Cu, Zn, Pb, and Cd ranged between 1.32 and 4.24, reflecting a low to moderate degree of contamination.

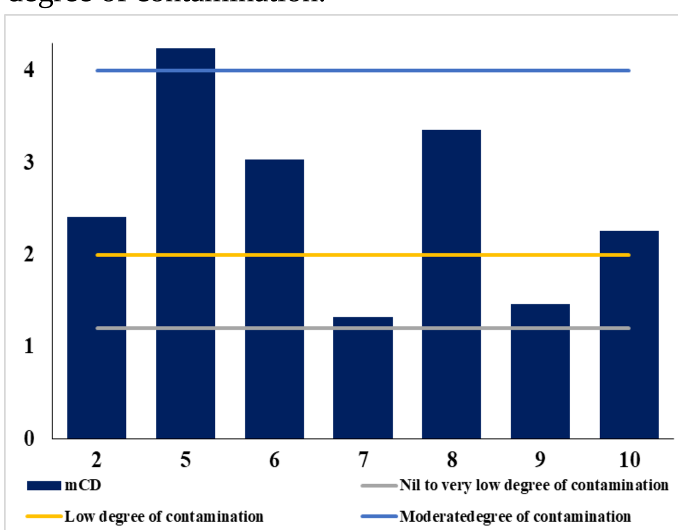

Figure 4. Modified degree of contamination (mC_d) index of surface sediments from Abu-Qir Bay.

6.5. Assessment according to ecological risk index (RI)

Hakanson (**1980**) proposed the ecological risk index (RI) to evaluate the ecological risks for aquatic organisms. According to the equations (6) and (7), the ecological risk index (RI) was calculated as a sum of five heavy metals (Ni, Cu, Zn, Pb, and Cd):

$$E_r^i = T_r^i \times C_F^i \qquad (6)$$

$$RI = \sum E_r^i \qquad (7)$$

Where RI is the sum of the potential ecological risk factor (E_r^i), T_r^i is the metal toxic response factor for a given metal, and C_F^i refers to the contamination factor. The calculated RI values for Ni, Cu, Zn, Pb and Cd in all study areas are listed in Table (5) and shown in Figure (5). Those metals presented moderate ecological risk (150 ≤ RI < 300) for all studied sites, except for St **5** and St **8** that showed a considerable ecological risk (RI > 300). The values for each investigated metal followed the sequence: Zn = Ni =1 < Cr = 2 < Cu = Pb = 5.

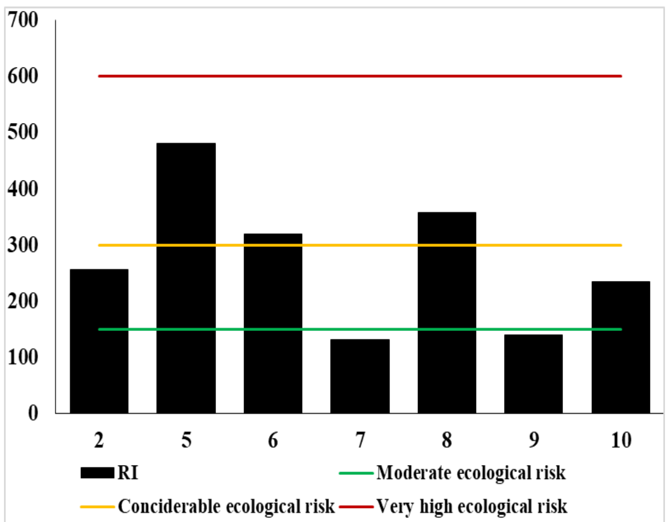

Figure 5. Potential risks (*RI*) index of surface sediments from Abu-Qir Bay.

6.6. Mean effect range median quotient ($MERM\text{-}Q$)

The mean ERM quotient ($MERM\text{-}Q$) is a method used to evaluate the possible biological effects of all the investigated metals and PAHs by comparing their observed concentrations with their limit concentrations and calculating the values using the equation (8) suggested by **Long *et al*.** (**1998**):

$$MERM\text{-}Q = \sum (C_i \,/\, ERM_i) \,/\, n \qquad (8)$$

Where, C_i refers to sediment concentration of metal i, ERM_i is the effect rang medium of metal i, and n refers to number of metals. The ERM_i values for each investigated metal were: Ni = 51.0, Cu = 270, Zn = 410, Pb = 218 and Cd = 9.60.

According to **Long *et al*. (2000)** and **Gao and Chen (2012)**, $MERM\text{-}Q$ can be classified into four categories: $MERM\text{-}Q$ < 0.1 has 9 % probability of being toxic (low), between 0.11-0.51 represents 21 % probability of being toxic (low to medium), and between the range 0.51-1.5 indicates 49 % probability of being toxic (high- medium). While $MERM\text{-}Q$ > 1.5 indicates 74 % of toxicity (high-priority sites). The calculated $MERM\text{-}Q$ values for the sampling sites for Ni, Cu, Zn, Pb, and Cd varied from 0.27 to 0.63 and the average value was 0.44 (Figure 6 & Table 5). Based on this classification, all samples had low to medium toxicity (21 % probability of being toxic).

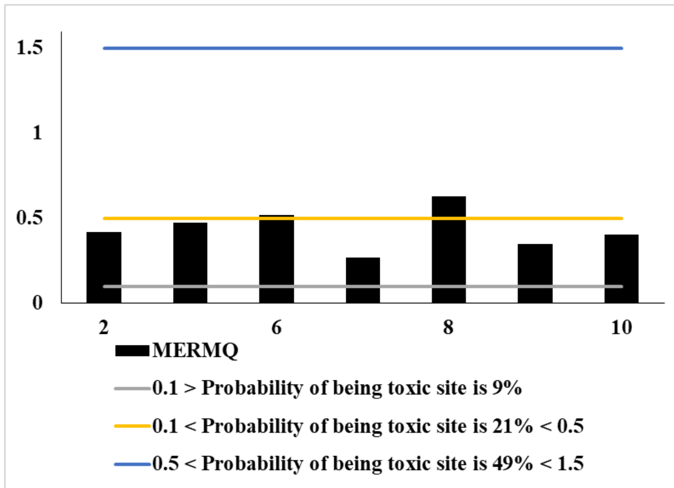

Figure 6. Mean ERM quotient (MERM-Q) index of surface sediments from Abu-Qir Bay.

Comparing the $MERM\text{-}Q$ calculated values with ERM individual value, one can say that, the $MERM\text{-}Q$ is a more effective index for reducing the large data to a single number to assess the extent of each metal. In general, according to the results of I_{geo}, PLI, mC_d, and RI indices as well as $MERM\text{-}Q$, a considerable and intense heavy metals pollution of sediments through all study areas was detected. This may be due to human activities; industrial effluents of Petrojet company, paper and fertilizer factories, Abu-Qir EPS, and fishing boats, as well as the outlet of Lake Edku (Boughaz El-Maddya) which contains different types of pollutants as pesticides, humic acids, and trace metals.

7. Application of principal component analysis (PCA) and redundancy analysis (RDA)

Principal components analysis (PCA) is a useful tool used to understand the distribution and the sources of heavy metals and their relationship (**Varol, 2011**). In the present research, PCA was conducted on basis of the determined concentrations of heavy metals in seawater, SPM and sediments with varimax-rotation. The PCA results are represented in Table (6), the PC1 with a variance of 31.06 % exhibited a significant positive correlation of the congeners Fe (0.92), Co (0.80), and Ni (0.81) of seawater, indicating a common source that may be due to the exogenous discharge (**Mao, 2013**). The PC2 of the variance 23.70 % exhibited a correlation between Mn (0.94), Ni (0.83), and Cu (0.72) of SPM as well as Zn (0.72) of sediment samples, that indicated a common source as well as the sedimentation-re-suspension process. The mentioned metals either in particulate or sediments were strongly affected by the alkalinity of seawater (0.75). The PC3 results also indicated a good positive correlation between Mn (0.73), Cu (0.90), Zn (0.71), and Pb (0.72) of seawater samples. PC4 and PC5 of variance 12.69 % and 8.09 %, respectively showed a good correlation between Cd (0.74) of seawater samples and OOM (0.87) as well as between Cd (0.63) of particulate samples and TOC (0.74%). In the pollution assessment, Cd had higher I_{geo} values ($1 < I_{geo} < 3$; class 3), indicating that these sites were moderately to strongly contaminated with Cd. Therefore, Cd may be originated from natural sources with contributions from agricultural wastes (**El-Nemr & El-Said, 2017**).

Table 6. Rotated components matrix and water quality index for studied parameters of Abu-Qir Bay.

PCA				Variables and loading values	Water Quality Index	
Component	**Total**	**Variance (%)**	**Cumulative (%)**		**St**	**WQI**
PCA1	10.87	31.06	31.06	Fe-w (0.92), Co-w (0.80), and Ni-w (0.81)	2	0.460161
PCA2	8.29	23.70	54.75	Mn-p (0.94), Ni-p (0.83), Cu-p (0.72), and Zn-s (0.72) & alkalinity (0.75).	3	0.060052
PCA3	7.07	20.20	74.95	Mn-w (0.73), Cu-w (0.90), Zn-w (0.71), and Pb-w (0.72).	4	0.707431
PCA4	4.44	12.69	87.64	Cd-w (0.74) & OOM (0.87)	5	0.160326
PCA5	2.83	8.09	95.73	Cd-p (0.63) & TOC (0.74)	6	0.322984
PCA6	1.49	4.27	100.00	Cd-s (0.63)	7	-0.277910
					8	-0.755040
					9	-0.445220
					10	-0.232790

To investigate the relationship between the environmental factors and the distribution of heavy metals in sediment, the RDA was used. The result of RDA for seawater and particulate matter as well as for sediment are represented in Fig. (7A & 7B). The angle between the heavy metal and environmental factors reflects their relationship. Metals in seawater such as Mn, Fe, Co, Ni, Cu, Zn, Pb, Cd and temperature, salinity, as well as TDS can be considered as significant parameters that reflect the water quality of Abu-Qir Bay coastal waters. Strong positive correlation of metals (Mn, Fe, Co, Ni, Cu, Zn, Pb, and Cd), pH, DO and T.alk. in particulate matter was also observed. The concentration of Fe in sediment was positively correlated to TOC and fine sand % (Fig. 7B) which accorded with the result of Kd, whereas the RDA analysis explained that Zn, Pb, and Cd contents in sediments are strongly related.

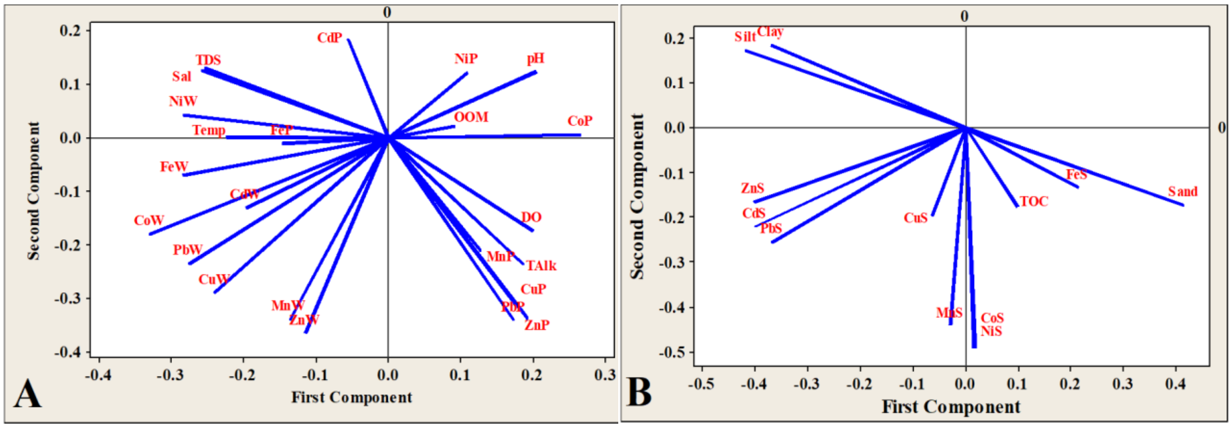

Fig. 7. RDA diagram between heavy metals and: A) Seawater variables; W: dissolved & P: particulate and B) sediment variables; S.

8. Water quality index (WQI)

In the current study, principle component analysis was employed to assess the water quality index (WQI) at each station and detect the hot spot stations. WQI can be calculated according to equation (9)

$$WQI = \sum_{n=1}^{n} \left(\frac{\lambda_n}{\Sigma\lambda}\right) \times PC_n \qquad (9)$$

Where, n and λ_n refer to the number and the Eigenvalues of effective components, respectively. Whereas $\Sigma\lambda$ is the sum of the Eigenvalues and PC_n that refers to the n critical principal component scores (**MacDonald *et al.*, 2003**). The data of water quality index (WQI) of Abu-Qir Bay are shown in Table (6). They depicted that, St **7** to St **10** were relatively good and varied between -0.23 at St **10** and -0.75 at St **8** meanwhile, water quality index of the other stations (St **2** to St **6**) showed that they were slightly polluted to different degrees (0.060-0.707). Thus, they could be considered as hot spots. St **2** exists in front of Abu-Qir city and can be affected by human activities and domestic effluents. While St **3** to St **5** are exposed to pollution caused by the activities of gas production (Petrojet company) and Abu-Qir EPS. St **6** is located at the outlet of Lake Edku (Boughaz El-Maddya) which contains a different type of hazard materials as pesticides, humic acids, and trace metals as well as fishing boats.

CONCLUSION

The marine ecosystem is threatened by the discharge of industrial effluents which affects the sustainability of living organisms and hence public health. Identification of the sources of the heavy metals (Mn, Fe, Co, Ni, Cu, Zn, Pb, and Cd) as well as their distribution between water body and sediment of Abu-Qir Bay area were studied. The present study provided a detailed on heavy metal concentrations in seawater, SPM, and sediment. The level of contamination in Abu-Qir Bay and their ecological risks to aquatic organisms showed that the anthropogenic activities are the fundamental source of heavy metals in Abu-Qir Bay. However, among the eight examined heavy metals in the surveyed area, the content of Cd was the highest in sediment and according to PCA, Cd may be originated from natural sources with some effects from agricultural wastes.

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الملخص العربي

"تقييم التلوث بالمعادن الثقيلة في مياه البحر والجسيمات العالقة والرواسب من خليج أبو قير ، الإسكندرية ، مصر"

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الغرض من هذه الدراسة هو تحديد مصادر المعادن الثقيلة وتوزيعها بين المسطح المائي والرواسب في منطقة خليج أبوقير. تم تقييم مستوى المعادن الثقيلة (Mn، Fe، Co، Ni، Cu، Zn، Pb، Cd) في الماء والجسيمات العالقة (SPM) والرواسب على طول ساحل خليج أبو قير ، الإسكندرية ، مصر ، خلال شتاء 2020. يتبع متوسط تركيزات المعادن الثقيلة المدروسة التسلسل: Fe> Zn> Mn> Ni> Pb> Co> Cu> Cd في ماء البحر ، Fe> Zn> Mn> Pb> Cu> Ni> Co> Cd في SPM ، و Fe> Mn> Zn> Ni> Co> Cu> Pb> Cd في الرواسب. أظهر معامل التوزيع (Kd) أن Fe مستقر بدرجة عالية في الرواسب وكانت قيم Log (Kd) تساوي 6.521 في جميع water bodies. تم حساب معامل التراكم الجغرافي (I_{geo}) ، ومعامل حمل التلوث (PLI) ، ودرجة التلوث المعدلة (mC_d) ، ومؤشر المخاطر البيئية المحتملة (RI) ، ومتوسط حاصل المدي الفعال $MERM-Q$. كانت قيم I_{geo} لـ Cd في الرواسب السطحية بين 1 و 3 مما يشير إلى أن هذه المواقع ملوثة بشكل معتدل إلى قوي بـ Cd. تراوحت قيم mC_d لـ Ni و Cu و Zn و Pb و Cd بين 1.32 و 4.24 ، مما يعكس درجة تلوث منخفضة إلى متوسطة. قدمت المعادن المدروسة مخاطر بيئية معتدلة ($150 \leq RI < 300$) باستثناء محطة 5 ومحطة 8 اللتان أظهرتا مخاطر بيئية كبيرة ($RI > 300$). وفقًا لتصنيف $MERM-Q$ ، فإن جميع العينات ذات سمية متوسطة منخفضة. وفقًا لتحليل المكونات (PCA) فمن المحتمل ان تكون العناصر ؛ Ni و Cu و Zn و Pb قد نشأت من مصادر بشرية المنشأ بينما نشأ الكادميوم من مصادر طبيعية. كشف تحليل التكرار (RDA) أن نسبة الكربون العضوي الكلي ونسبة الرمل الناعم للرواسب لها تأثير معين على غني الحديد. أظهرت بيانات مؤشر جودة المياه (WQI) أن المحطات 2 ، 3 ، 4 ، 5 ، 6 كانت ملوثة قليلاً ، ويمكن اعتبارها hot spots. أظهر حجم التلوث والمخاطر البيئية على الأحياء المائية أن الأنشطة البشرية هي المصدر الرئيسي للمعادن الثقيلة في خليج أبو قير.