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Assessment of Metal Pollution of the Surface Sediments along the Wadi Al Rayyan area, Jordan

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Abstract

Seventy surface sediment samples were collected from Wadi Al Rayyan area. These samples were extracted by Ammonium-Acetate-EDTA, and analyzed by Atomic Absorption Spectrometer to determine their metal concentrations of Cu, Zn, Fe, Mn, Pb, Cd, Cr and Ni. XRF was used to analyze CaO wt%, and XRD was used to determine the mineral composition. The chemical analyses of the samples show an enrichment of Cu, Mn, Pb, Cd, Cr and Ni along the Wadi Rayyan from upstream to downstream, but valuable concentrations of Zn and Fe were found along the Wadi. To evaluate the level of contamination for the surface sediments of Wadi Al Rayyan in this study, the Enrichment Factor (EF), Geoaccumulation index (Igeo), Pollution Index (PI) and Pollution Load Index (PLI) have been used. EF shows the surface sediments to be moderately-to-strongly polluted and strongly-to-extremely polluted with Cu, Zn, Mn, Pb, Cd, Cr and Ni. In regard to Zn, the sediments are moderately polluted, and significantly polluted with Mn and Cr. I_{geo} values for Zn, Fe, and Mn reveal the sediments to be unpolluted ($0 < Igeo \leq 1$), while they are moderately polluted ($1 < Igeo \leq 2$) with Cu, Cd, and Cr, and strongly polluted ($2 < Igeo \leq 3$) with Ni and Pb. PI for Zn, Fe, and Cd shows a low level of pollution ($PI < 1$), a high level of pollution ($PI \geq 3$) with Cu, Pb, Cr and Ni, and a middle level pollution ($PI \leq 1$) with Mn. The PLI values for Wadi Al Rayyan sediments were found to be less than 1 ($PLI<1$), thus, revealing that the surface sediments are unpolluted with respect to Cu, Zn, Fe, Mn, Pb, Cd, Cr, and Ni. These metals can be derived from anthropogenic sources, such as fertilizers and pesticides used in agricultural activities.

Keywords: Heavy Metals, Pollution, Assessment, Surface Sediments, Wadi Al Rayyan, Jordan.

1. Introduction

The various kinds of heavy metal pollution constitute a problem in modern societies. Toxic heavy metals are a major concern, since these elements are not biodegradable, but their elevated uptake by crops may affect food quality and safety. Heavy metals at high concentrations have toxic effects on living organisms (Schuurmann and Market, 1998; MacFarlane and Burchett, 2000; Habes and Nigem, 2006). Heavy metals are very important in the ecological system, because they are not removed from water by self-purification processes, but they tend to be bio-accumulated during the food chain (Loska and Wiechula, 2003, Al-Khashman, 2004). Heavy metals enter into aquatic systems mainly through natural inputs, such as chemical weathering of rocks, volcanic activities, industrial processes and mining activities. The anthropogenic and manufacturing sources include industrial processes, energy production, industry, vehicle exhausts, waste disposal, coal combustion, and agricultural activities as well as the use of synthetic products (e.g. pesticides, paints, batteries, industrial waste, and land applications of industrial, terrestrial runoff and sewage disposal or domestic sludge (Martin et al., 1998; Li et al., 2001; Gray et al., 2003; Bin Chen et al., 2005; Biasioli et al., 2006). These activities can increase the levels of the metals in various parts of the ecosystems (Auburn, 2000; Bilos et al., 2001; El-Hasan and Jiries, 2001; Komarek and Zeman, 2004; Moller et al., 2005; Çevik et al., 2009, Lu et al., 2009; Yang et al., 2011).

Stream sediments and soil are generally mixtures of several components, including different mineral species and organic constituents and debris. The sediments accumulate through complex physical and chemical adsorption mechanisms, depending on the nature of the sediment matrix and the properties of the adsorbed compounds (Maher and Aislabie, 1992; Ankley et al., 1992; Leivouri, 1998, Al-Khashman and Shawabkeh, 2006). Heavy metals discharged into the aquatic systems have been immobilized within the stream sediments by main processes such as adsorption, flocculation, precipitation and co-precipitation. Therefore, sediments in aquatic environments serve as a pool that can retain metals or release metals to the water column by various processes of remobilization (Caccia et al., 2003; Pekey, 2006; Marchand et al., 2006). The main processes lead to the association of heavy metals with solid phases, such as direct absorption by fine-grained, clay particles, adsorption of hydrous ferric and manganese oxides. This may in turn be associated with clays; adsorption on or complication with natural organic substances, and direct precipitation as new solid phases (Habes and Nigem, 2006; Cao et al., 2011).

Numerous studies have demonstrated that the concentrations of heavy metals in sediments can be sensitive indicators of contaminants in the aquatic systems (Bellucci et al., 2002; Bloundi et al., 2009; Suthar et al., 2009; Al-Khashman and Shawabkeh, 2009; Mhamdi et al., 2010, Al-

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Khashman, 2013). Several approaches have been applied in order to assess the severity of sediment contamination and to understand the natural and anthropogenic inputs in the river system. The metal assessment indices include the Enrichment Factor (EF), Geo Accumulation Index (I_{geo}), pollution Index (PI) and Pollution Load Index (PLI). The sediment quality guidelines were often used to screen the potentiality of contaminants within sediments. The aims of this study focus on the concentrations of heavy metals (Cu, Zn, Fe, Mn, Pb, Cd, Cr and Ni) and their distribution in the sediments of Wadi Al Rayyan area in Jordan. This study provides an assessment of heavy metal pollution in the sediments using the Enrichment Factor (EF), Geoaccumulation Index (I_{geo}), Contamination Factor (CF), Pollution Index (PI) and Pollution Load index (PLI).

2. Geological Setting

The study area is covered by Late Cretaceous carbonate rocks. The dominant outcropped rocks belong to Ajlun and Belqa Groups (Table 1). The oldest rocks cropped out in the area belong to the Mesozoic – Cenozoic (Cenomanian, Turonian, Campanian and Maastrichitian) periods, as limestone Formation within Ajlun and Belqa Group (Fig. 1a). Quaternary recent sediments as wadi deposits and soils overlie the Late Cretaceous rocks (Abu Qudaria, 2005).

The study area of Wadi Al Rayyan was affected by different environmental activities such as agriculture in addition to the road networks along the Wadi. Agriculture activities in Wadi Al Rayyan area depend on rainfall throughout the winter season and irrigation processes utilizing the effluents of Arjan springs in other seasons.

Table 1. Geological Classification of Rock Units in the Study Area (Abu Qudaria, 2005).

Era	Period	Epoch	Stage	Group	Formation	Description
Cenozoic	Quaternary	Recent	Holocene -Recent		Soil (S) Calcrete (Cl)	Soil, Calcrete and Gravel
Mesozoic	Late Cretaceous	Upper	Maastrichitian/ Paleocene	Belqa	Muwaqqar Marl (B3)	The formation consists of massive marl and chalk, with bitumen lenses in the lower part, and limestone embedded within clay marl at the top.
			Campanian/ Maastrichitian		Amman Silicified Limestone / Al Hisa Phosphorite (B2) (ASL/AHP)	The formations consists of chert, limestone, phosphates, coquina limestone and marl
		Middle	**Turonian**	Ajlun	Wadi As-Sir (A7) (WSL)	Hard crystalline limestone, yellowish dolomitic limestone and dolomite, over lained by marly limestone well bedded massive micrite and chert nodules.
			Cenomanian		Shuayb (Sh)	Light grey limestone interbedded with marls alternating with chalky limestone.
					Hummar (H)	Hard dense limestone and micritic limestone with bedded clay, overlain by dolomite limestone, the formation highly fractured and cavenous

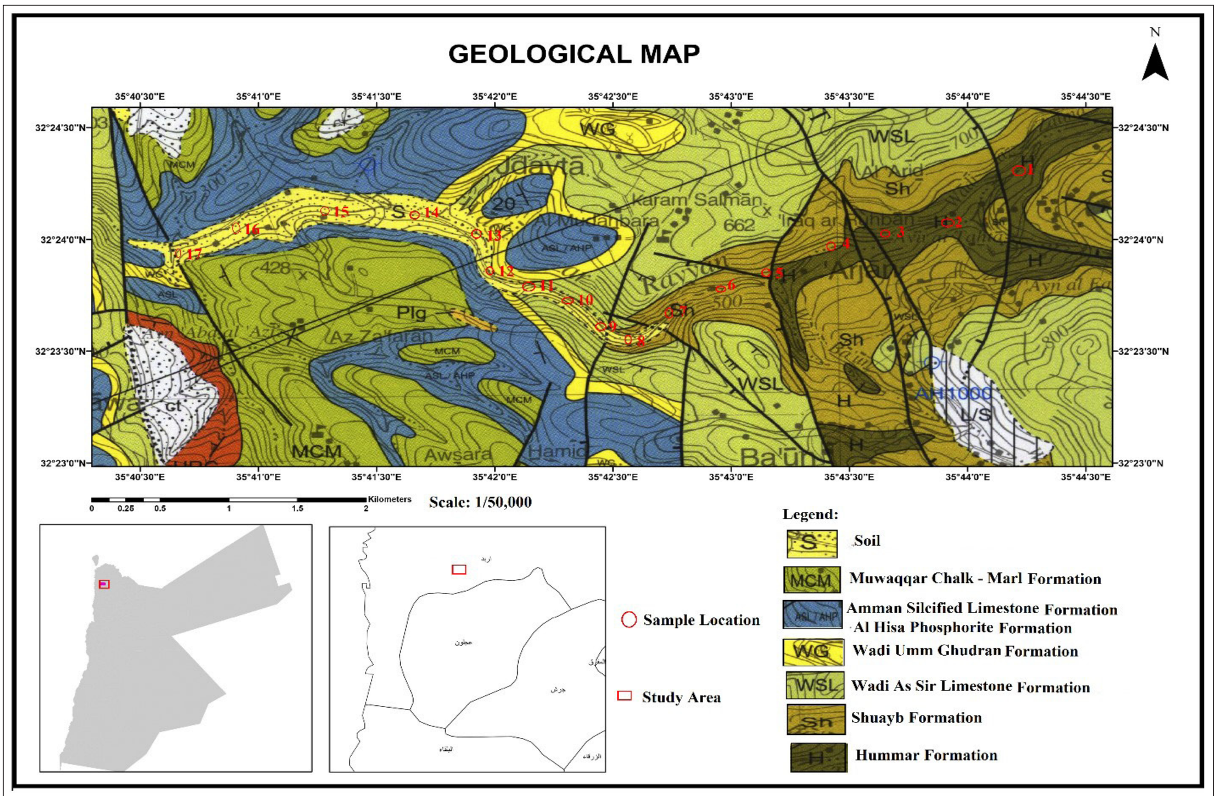


Figure 1. A geological map showing the area under study, and sample location (Modified After Abu Qudaria, 2005)

3. Sampling and Analytical Techniques

Seventy composite stream sediment samples were collected along the main valley of Wadi Al Rayyan area (Fig. 1b). The samples were taken from depths of 5–20 cm depending on the sediment condition. The samples were collected in May, 2016 and were, then, transported to the laboratory of Al-Al-Bayt University using polyethylene bags. The samples were dried in the oven at 50°C for twenty-four hours. All the samples were sieved (1/256 mm) and particles <63 µm (0.05-mm) in size. This size has been provided to be the best size for analysis for arid and semi-arid regions (Saffarini and Lahwani, 1992; Singh et al., 1999).

The extraction method was used to assess the heavy metal pollution contents of Cu, Zn, Fe, Mn, Pb, Cd, Cr and Ni. The best extraction results were obtained with Ammonium-Acetate-EDTA. The extraction solution was prepared by adding 0.5M NH4Ac 0.5M+ HAc + 0.02M EDTA (pH 4.65). Then, 38.5gm of NH4Ac was dissolved in 500 ml H_2O+ 25ml Acetic Acid and 5.8gm EDTA was added, then the volume was increased to one liter with distilled water. Thereafter, 20 gm of the air-dried sediment samples (<63 µm) were placed in a 300 ml Erlenmeyer flask and 100 ml of the extracting solution was added. Then, it was shaken mechanically for thirty minutes. After shaking, suspension materials were filtered through 0.45-µm filters, and the clear solution was collected in polyethylene bottles (Albanese, 2008). Heavy metals were determined using Atomic Absorption Spectrophotometer (AAS) (2280 Model; Perkin Elmer) at the University of Jordan. The quality control was performed for Cu, Zn, Fe, Mn, Pb, Cd, Cr, and Ni using Merck ICP4 standard solutions. A 95 % confidence level was used. The results of the studied samples are shown in Table 1.

The organic matter (OM %) in each sample was determined using loss on ignition (LOI) at 550°C for two hours at the Institute of Earth and Environmental Sciences Lab, Al-al-Bayt University (Dean 1974). The pH measurements were performed using 2gm of dry sediment samples (< 63 µm), with the beaker kept undisturbed for about five minutes, and the solution was stirred with a glass rod (Dean 1974). After that, the pH was measured by pH electrode (Table 1). The chemical analysis of the CaO wt % for the samples was carried out using Phillips X-ray Florescence (XRF) Majex PW-2424 model. In addition, the mineralogical compositions of all the samples were evaluated by X-ray Diffraction (XRD) using a Phillips diffractometer with Cu Ka radiation at Water Environment and Arid Regions Research Center labs, at AL al-Bayt University.

4. Assessment of Contamination Level

To quantify the level of metal contamination in the surface sediments for the present study, many calculation methods have been proposed. In this study, the Enrichment Factor (EF), Geoaccumulation index (Igeo), Contamination Factor (CF), Pollution Index (PI) and Pollution load index (PLI) have been used to assess the metal contamination level in the surface sediments collected in the investigated area (Shi et al., 2008; Yang et al., 2011).

The Enrichment factor (EF) was used to compare the metals originating from anthropogenic activities to those of natural origins, and to determine the degree of contamination and the possible anthropogenic impact on the sediments of Wadi Al Rayyan area. The EF analysis was first presented by Mason, (1966) and Simex and Helz (1981) to evaluate trace element concentration as follows: EF = $(M/Fe)_{sample}$/ $(M/Fe)_{background.}$ Where, M/Fe sample is the ratio of metal and Fe concentration in the sample, M/Fe background is the ratio of metal and Fe

Table 2. Chemical analyses of the surface sediment samples collected from Wadi Al Rayyan area.

			Ppm									µs cm^{-1}
S. No.	OM %	CaO wt%	Cu	Zn	Fe	Mn	Pb	Cd	Cr	Ni	PH	EC
1	1.10	14.2	244	20.5	30.5	2475	1650	61.5	299	415	8.36	240
2	0.28	25.3	278	45.5	35.5	6300	1800	84.5	331	426.5	8.47	310
3	0.57	21.1	182	35	32	1618.5	1400	63	207.5	445.5	8.14	280
4	0.20	23.2	212	32	26.75	4870	1350	64.5	250.5	500	8.54	340
5	0.34	27.3	214.5	69.5	26.05	1458.5	850	57.5	250.5	453	8.45	425
6	0.11	23.5	193	26	25	2094.5	1300	55.5	248	518	8.32	335
7	0.84	24.8	248.5	23.5	25.5	1832	1150	60.5	257	560.5	8.32	310
8	0.10	38.2	261	34	65	1772	1846	60.5	265.5	571	8.33	860
9	0.39	24.1	279.5	156	20	2749	1538	81	262	704	8.50	450
10	0.40	23.2	237	42	29.5	2790.5	1692	62	257.5	648	8.47	361
11	0.30	26.3	211.5	14.5	25	2374.5	1615	64	254.5	590	8.46	610
12	0.36	25.2	208	25	29.55	3242	1625	57	269	503	8.54	540
13	0.25	18.3	209	18	23.5	1283.5	1618	53.5	251.5	473	8.73	280
14	0.55	23.3	198	23	120	1213.5	1628	50	250.5	435.5	8.41	300
15	0.33	22.8	251.5	25.5	30	3419.5	1769	68.5	252	413	8.17	290
16	0.41	24.8	194	44.5	25	2243	1355	50.5	188.5	261	8.37	400
17	0.17	20.2	234	21.5	30	2926	1700	58.5	304	401	8.45	310
Max	1.10	38.2	279.5	280	156	120	6300	1846	84.5	331	8.73	860
Min	0.10	14.2	182	14.5	20	120	850	50	84.5	261	8.14	240
Average	0.42	24.1	227.21	50.0	40.78	2363.3	1739	155.2	240.39	468.9	8.42	407

concentration of a background (Ergin et al., 1991; Chakravarty and Patgiri, 2009; Seshan et al., 2010; Bentum et al., 2011). The geochemical normalization of heavy metal data was employed for conservative elements such as Fe, Al and Si. According to Schiff and Weisberg, (1999) and Mucha et al. (2003), Fe was used to normalize heavy metal contaminants. In this study, Fe was used as a conservative tracer to differentiate natural components from anthropogenic ones. The background concentrations of the heavy metal study are taken from Turekian and Wedepohl (1961). The classification of pollution degree based on the enrichment ratio methodology was divided into a five-category system (Sutherland et al., 2000); If (EF < 2), this implies a depletion to minimal enrichment suggestive of no or minimal pollution. If EF is within the range of 2–5, this indicates a moderate enrichment, suggestive of a moderate pollution. If EF is within the range of 5–20, it means a significant enrichment, suggestive of a significant pollution. If EF is within the range of 20–40, it means a very high enrichment, indicative of a very strong pollution,, and if (EF > 40), it means an extreme enrichment, indicative of an extreme pollution signal.

The Geoaccumulation index (I_{geo}) was used by Muller, (1981) and Wei et al. (2009) to assess heavy metal pollution. The I_{geo} was used to assess metal contamination in urban soils by comparing current and pre-industrial values (Wei et al. 2009; Al-Khashman, 2013). In this study, the geoaccumulation index (I_{geo}) was used to determine the extent of heavy metal pollution in Wadi Al Rayyan surface sediments. The following equations are used in the calculation: Igeo = log2 (Cn/1.5Bn), (Rath et al., 2005). Where Cn is the concentration of the element 'n', and Bn is the geochemical background value of the element n, and 1.5 is the background matrix correction factor due to lithogenic effects. The geochemical background value in average shale is used to calculate I_{geo} (Turekian and Wedepohl, 1961). The geo-accumulation index (I_{geo}) scale consists of seven grades from 0 to 6, ranging from unpolluted to highly polluted (Rath et al., 2005, Faiz et al., 2009). The Geoaccumulation index value class designation of sediment quality is as follows:

$I_{geo} < 0$ unpolluted environments
$0 < I_{geo} \leq 1$ unpolluted-to-moderately polluted
$1 < I_{geo} \leq 2$ moderately polluted
$2 < I_{geo} \leq 3$ moderately-to-strongly polluted
$3 < I_{geo} \leq 4$ strongly polluted
$4 < I_{geo} \leq 5$ strongly to- extremely- polluted, and
Igeo >5 extremely polluted

The pollution index (PI) and Pollution load index (PLI) were used to assess heavy metal pollution in the soil, sediments, and dust. The pollution index (PI) was defined by the following equation: PI = C_n / B_n where Cn and Bn are the measured and background concentrations of metal n, respectively, in the surface sediment samples. PI is classified by (Faiz et al., 2009) as follows: PI ≤ 1 indicates a low level of pollution; 1< PI ≤ 3 indicates a middle level of pollution, and PI > 3 a high level of pollution. The pollution Load Index (PLI) for each sample was calculated following the method by Tomilson et al., (1980) and Soares et al., (1999). It was defined by the following equation:

$$PLI = \sqrt[n]{CF1 + CF2 + CF3 + \cdots + CFn} \quad (1)$$

where n is the number of metals and CF is the contamination factor. Contamination Factor (CF) is calculated from the following relationship:

$$CF = \frac{(Cn) Concentration\ of\ the\ heavy\ metal\ in\ the\ soil\ sample}{(Bn) Background\ concentration\ of\ the\ same\ metal} \quad (2)$$

where C_n is the measured concentration of a heavy metal in the surface sediments, and B_n is the background value. According to Harikumar et al., (2009), Faiz et al., (2009), and Seshan et al., (2010), the PLI value (PLI ≤ 1) indicates a low level of pollution, while 1< PLI ≤ 2 indicates a middle level of pollution and PLI >2 a high level pollution.

5. Results and Discussion

5.1. Physicochemical Properties of the Surface Sediments

The concentrations of different metals in the surface sediments along Wadi Al Rayyan area were studied according to the strength of anthropogenic, natural sources and PH values. The pH values show a relatively equal distribution in the investigated area. The pH-values range between 8.14 and 8.73 (Table 2), suggesting alkaline-prevailing conditions for all the sediment samples. The concentrations of metals in the sediments can vary greatly depending on the organic matter content and pH-values. Usually, pH-values influence the sediment composition, which in turn affects the heavy metal mobility and distribution in the sediment samples (Odat and Alshammari, 2011).

The high pH value of the samples was attributed to the high percentage of carbonate materials in the surface sediments (Murray and Hendershot 2000). Kim et al. (2003) explain that the changes of soil acidity can influence neutral compositions in the soils by removing the bivalent base such as $Ca^{+2}$and Mg^{+2} from soil distribution in the investigated area. The electrical conductivity (EC) values range between 240 and 860 µs.cm^{-1}. A high electrical conductivity (EC) value was found at the human activity site, while the lowest value of electrical conductivity was found at the low human activity site (240 µs.cm^{-1}). The percentage of organic matter (OM%) in the surface sediments of Wadi Al Rayyan range between 0.1 and 1.1 %, with an average of 0.42 % (Table 2). The content of organic matter in Wadi Al Rayyan sediments is related to the organic content of plant remains located adjacent to the Wadi site at various stages of decomposition. Cells and tissues of plant organisms and substances from plant roots, and soil microbes are considered as additional sources.

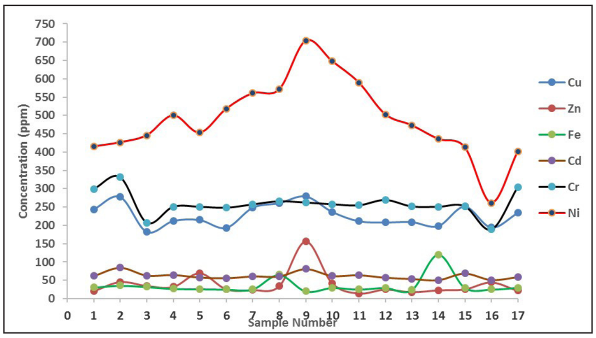
The value of the CaO wt% contents of the surface sediments range from 14.2 to 38.2 wt%, with an average of 24.12 wt% (Table 1). The percentage of CaO content in the sediments is mainly derived from the carbonate rocks exposed along the Wadi site and in the catchment area. These rocks mainly include limestone, chalky marl and caliches. These results are documented for the XRD analysis for all the surface sediment samples. The main mineral composition analysis for all the samples reveals that calcite, dolomite, quartz gypsum and clay minerals are dominant minerals. The carbonate mineral is important for the co-precipitation of heavy metals such as Cd and Zn (Alloway and Ayres, 1997).

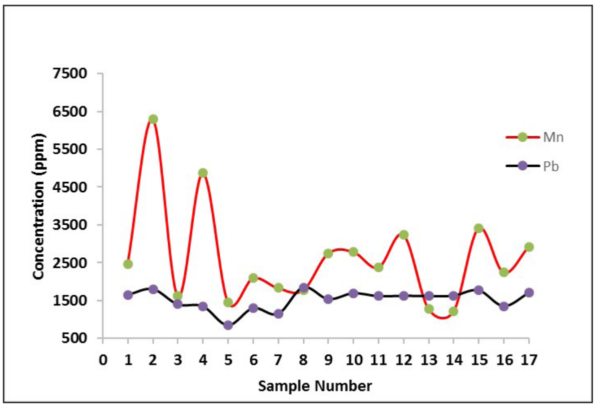
5.2. Heavy Metal Distribution

The chemical concentrations of the heavy metals determined for the study sample are listed in Table 2. The variable concentration of heavy metal distributions along

Wadi Al Rayyan area are shown in Fig. 2. The high level of Cu, Mn, Pb, Cd, Cr and Ni was found along the Wadi Rayyan surface sediments from upstream to downstream, but valuable concentrations of Zn and Fe were found along the Wadi. The enrichment concentration for these elements are located near the roads across the stream and in areas of human activity along the Wadi, as shown in samples 2, 4, 9, 15 and 17 (Fig. 2). The average concentrations of Cu, Zn, Fe, Mn, Pb, Cd, Cr, and Ni in Wadi Al Rayyan sediments are (227.21, 43.5, 38.9, 2746, 1504, 62.47, 258.84 and 488.58) respectively and have been compared with uncontaminated sediments and soils. Zn and Cd levels in uncontaminated sediments and soils contain 0.09 and 0.0003 mg g−1dry wt., respectively (Bowen, 1979). The elevated concentrations of heavy metals in the sediments of Wadi Al Rayyan were probably due to the anthropogenic sources as well as human and agricultural activities near the wadi surface sediments' site (Calace et al., 2005). The sources include fertilizers and pesticides used in agricultural activities and sewage wastewater discharges and seepage for running of the surface water near areas of human activities along the Wadi Al Rayyan. According to Zarei et al., (2014), the elevated values of Pb and Cd might be related to human activities such as wastewater discharges in the surface water along the Wadi, which mixes with the surface running water discharge from the Arjan spring.

The Pearson's correlation coefficients for the contents of Cu, Zn, Fe, Mn, Pb, Cd, Cr and Ni in the surface sediments of Wadi Al Rayyan are presented in Table 3. A positive correlation was found for Cu with Cd (R^2 = 0.75) and Cr (R^2 = 0.66); Zn with Cd (R^2= 0.54); Mn with Cd (R^2 = 0.68) and Cr (R^2= 0.53). The results revealed that metal concentrations rise unnaturally due to the anthropogenic sources of pollution which have been identified to be the human, industrial, and agricultural activities such as sewage effluent, waste combustion, steel processing, fertilizers and pesticides (Wuana et al., 2011). Low positive linear correlations among the concentrations of Cu, Zn, Pb, Cd, and Cr were clearly observed: Cu with Zn (R^2 = 0.43), Mn (R^2 = 0.45), Pb (R^2 = 0.41), Ni (R^2 = 0.38); Zn with Ni (R^2 = 0.42); Pb with Cd (R^2 = 0.28) and Cr (R^2 = 0.45); Cd with Cr (R^2 = 0.45), Ni (R^2 = 0.37); Cr with Ni (R^2 = 0.15) respectively. These values revealed the same sources for all of the abovementioned elements. The results indicate that heavy metals are not associated with each other, and that no relationship exists between the variables (Bany Yaseen and Al-Hawari, 2015). Furthermore, these metals have different anthropogenic and natural sources in the sediments of Wadi Al Rayyan (Habes and Nigem, 2006; Bany Yaseen and Elaimat, 2016).





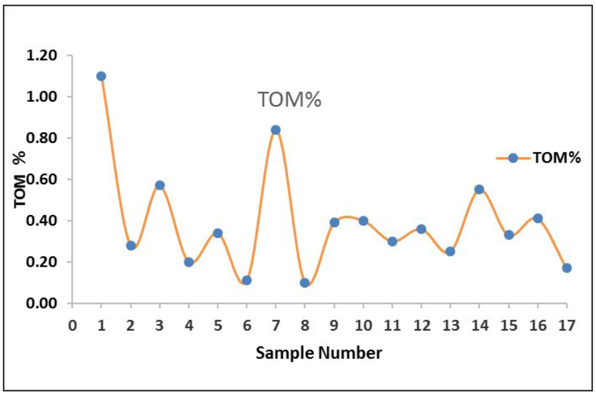


Figure 2. Distribution of metal concentration (ppm) for Cu, Zn, Fe, Cr, Ni, Cd, Mn and Pb and percentage of organic matter (OM%) and CaO wt% for the surfaces sediment samples from Wadi Al Rayyan area.

Table 3. Correlation matrix between metals in the surface sediment samples, from the Wadi Al Rayyan area.

	ppm							
	Cu	**Zn**	**Fe**	**Mn**	**Pb**	**Cd**	**Cr**	**Ni**
Cu	1							
Zn	0.433	1						
Fe	-0.104	-0.194	1					
Mn	0.447	0.069	-0.248	1				
Pb	0.406	-0.181	0.279	0.324	1			
Cd	0.747	0.537	-0.292	0.687	0.286	1		
Cr	0.658	-0.035	0.025	0.535	0.449	0.496	1	
Ni	0.378	0.424	-0.107	-0.053	0.074	0.372	0.153	1

5.3. Enrichment Factor (EF):

Enrichment factor (EF) was used to assess the degree of contamination and the possible anthropogenic impact on the sediments of Wadi Al Rayyan area. The EF analysis was first presented by Simex and Helz (1981) to evaluate trace element concentration as follows:

EF = $(M/Fe)_{sample}$/ $(M/Fe)_{background.}$ Where (M/Fe) sample is the ratio of metal and Fe concentration in the sample. (M/Fe) background is the ratio of metal and Fe concentration of a background (Ergin et al., 1991; Chakravarty and Patgiri, 2009; Seshan et al., 2010; Bentum et al., 2011). The geochemical normalization of the heavy metal data was employed for conservative elements such as Fe, Al, and Si. According to Schiff and Weisberg (1999) and Mucha et al., (2003), Fe (reference) was used to normalize heavy metal contaminants. In this study, Fe was used as a conservative tracer to differentiate natural components from anthropogenic ones. The background concentrations of the studied heavy metals are taken from Turekian and Wedepohl (1961).

The classification of pollution degree based on the enrichment ratio methodology was in a five- category system (Sutherland et al., 2000). If EF is less than 2 (EF <2), it implies depletion to minimal enrichment suggestive of no or minimal pollution. If EF is within the range of 2–5, this indicates a moderate enrichment, suggestive of a moderate pollution, and if EF is in the range 5–20, it means a significant enrichment, suggestive of a significant pollution signal. When EF is in the range 20–40, it means a very high enrichment, indicating a very strong pollution signal, and if EF is more than 40 (EF>40), it means an extreme enrichment, indicating an extreme pollution signal.

The values of EF in the study area are shown in Table 4 and Fig. 3b. The results of EF indicate that high concentrations of Cu, Pb, Cd, Cr, and Ni, are present in the analyzed samples (Table 2). The sediments of Wadi Al Rayyan are moderately-to-strongly polluted and strongly-to extremely polluted. This may result in a risky pollution with Cu, Zn, Mn, Pb, Cd, Cr and Ni, while the values for Zn indicate a minimal pollution in all of the samples excluding sample 9 which shows moderate pollution (Table 4) and Fig.2d (EF ranges between 0.13-5.13 with an average of 0.94). As for Cu, there was significant pollution except in sample 14 which showed a moderate pollution (EF ranges between 3 and 26.12 with an average of 14.42). There was minimal pollution with Mn in the samples 3, 8, and 14, and moderate pollution with Mn in all of the other samples (EF ranges between 0.46 and 8.25 within an average of 4.1). As for Pb, there was significant pollution in the samples 3, 5, 7, 8, and 14, whereas other samples showed a very strong pollution (EF ranges between 6 and 34.21 with an average of 22.59). There was significant pollution with Cd in all of the samples except in sample 9 which revealed a very strong pollution. As for Cr, there was moderate pollution in samples 8 and 14, and significant pollution in all of the other samples (EF ranges between 2.27 and 14.23 with an average of 9.5). There was significant pollution with Ni in the samples 1,2,3, 8, 14, 15, 16, and 17, while other samples revealed a very strong pollution. The strong pollution of the sediment samples with Cu, Pb, Cd, Cr, and Ni depends on the location of the sample and the contributors to the elements' concentrations. The fluctuation in the EF values can be attributed to the changes in the amount of contribution of each metal in the sediments, or to the variance in the removal rate of each metal from the sediments. These heavy metals can be derived from anthropogenic sources, such as fertilizers and pesticides used in agricultural activities. These dangerous metals can be derived from industrial and agricultural activities such as waste, gasoline additives used in industries and automobiles (Mwamburi, 2003). In addition, they might be derived from corrosion agricultural activities in the wadi.

Table 4. Enrichment Factor (EF) for the heavy metals of the surface sediments of Wadi Al Rayyan area.

	Enrichment Factor (EF)						
S. No.	Cu	Zn	Mn	Pb	Cd	Cr	Ni
1	14.95	0.44	3.68	24.07	13.28	10.65	18.69
2	14.64	0.84	8.05	22.56	16.24	10.13	16.50
3	10.63	0.72	2.29	19.46	13.10	7.04	19.12
4	14.81	0.79	8.25	22.45	16.75	10.17	25.67
5	15.39	1.76	2.54	14.51	11.89	10.44	23.89
6	14.43	0.68	3.80	23.13	11.06	10.77	28.46
7	18.22	0.61	3.26	20.06	14.20	10.95	30.19
8	7.51	0.34	1.24	12.63	5.72	4.44	12.07
9	26.12	5.13	6.23	34.21	27.40	14.23	48.35
10	15.02	0.94	4.29	25.51	14.70	9.48	30.17
11	15.81	0.38	4.31	28.74	18.11	11.06	32.42
12	13.16	0.56	4.97	24.46	10.80	9.89	23.38
13	16.62	0.50	2.48	30.63	11.76	11.62	27.65
14	3.08	0.13	0.46	6.03	2.14	2.27	4.98
15	15.67	0.56	5.17	26.23	16.20	9.12	18.91
16	14.50	1.17	4.07	24.11	11.63	8.19	14.34
17	14.58	0.47	4.42	25.21	10.80	11.01	18.36
Max	26.12	5.13	8.25	34.21	27.40	14.23	48.35
Min	3.08	0.13	0.46	6.03	2.14	2.27	4.98
Average	14.42	0.94	4.09	22.59	13.28	9.50	23.13

5.4. Assessment of Pollution Level

In the present study, Geoaccumulation index (I_{geo}) and Pollution load index (PLI) are reported by Muller, (1981) and Wei et al. (2009). The I_{geo} values for the heavy metals are presented in Table 5, and the distribution I_{geo} of the studied samples are shown in Fig. 3a. The values of I_{geo} range between (1 and 4) according to the type of metal and its location along the Wadi Al Rayyan area. The surface sediments of Wadi Al Rayyan are unpolluted to moderately to strongly polluted with Zn, Fe, Mn, Cd, Cr, Ni, and Pb respectively. As for the metals of Zn, Fe, and Mn, the sediments were shown as unpolluted ($0 < Igeo \leq 1$), and are moderately polluted ($1 < I_{geo} \leq 2$) with Cu, Cd and Cr. The sample sediments were strongly polluted with Ni and Pb ($2 < I_{geo} \leq 3$) (Table 5). While there was a high level pollution with Pb and Ni, depending on the location of the sample and the contributors to the element concentrations. The highest I_{geo} value was attributed to Pb and Ni being ($2 < I_{geo} \leq 4$) in some places such as in samples 1, 2, 7, 8, 9, 10, 11, 12, 13, and 14 respectively. These dangerous metals can be derived from industrial and agricultural activities such as waste, gasoline additives used in industries and automobiles as well as from fertilizers and pesticides (Mwamburi, 2003).

The Pollution Index (PI) and Pollution Load Index (PLI) were calculated for all metals andthe statistical values are given in Table 6. PI is classified by (Faiz et al. 2009). PI ≤ 1 indicates a low level of pollution; 1< PI ≤ 3 indicates a middle level of pollution, and PI > 3 a high level of pollution. PI of Zn and Fe for all of the samples correspond to the low level (PI < 1), except for samples 8 and 9 which show a middle level of pollution. There was a low level of pollution with Cd in the sample 5, 6, 12, 13, 14, 16, 17, while the the other samples showed a middle level of pollution (Table 5) and (Fig. 3d). PI for Cu, Pb, Cr and Ni shows a high level of pollution (PI ≥ 3) in all of the samples. PI for Mn shows middle pollution (PI ≤ 1) except in samples 2, 4, 15 which had a high level of pollution (PI ≥ 3). Therefore, according to the values of I_{geo} and PI for Pb and Ni there is a high level of pollution; these reflect the same sources of metals in the study area. The average PI for Zn is (0.61), Fe (0.83) and for Cd it is is (1.04). These metals cause a low level pollution (PI <1); Cu (9.09), Pb (14.35), Cr (6.02) and Ni (14.37) shows a high level of pollution (PI ≤ 3); Mn (2.67) shows a middle level of pollution. The PLI value for all the surface sediment samples of Wadi Al Rayyan was less than 1 (PLI<1) (Table 6). The results reveal that the sediments are unpolluted with respect to Cu, Zn, Fe, Mn, Pb, Cd, Cr, and Ni.

Table 5. Geoaccumulation index (I_{geo}) for heavy metals in the surface sediments of Wadi Al Rayyan area.

	Geoaccumulation Index (I_{geo})							
S. No.	Cu	Zn	Fe	Mn	Pb	Cd	Cr	Ni
1	1.96	0.06	0.13	0.48	3.14	1.74	1.39	2.38
2	2.23	0.13	0.15	1.23	3.44	2.48	1.54	2.44
3	1.46	0.10	0.14	0.32	2.68	1.80	0.97	2.55
4	1.7	0.09	0.11	0.95	2.58	1.92	1.17	2.87
5	1.72	0.20	0.11	0.28	1.62	1.33	1.17	2.60
6	1.54	0.07	0.11	0.41	2.48	1.19	1.16	2.97
7	2.28	0.07	0.11	0.36	2.20	1.56	1.20	3.21
8	2.1	0.10	0.28	0.35	3.53	1.60	1.24	3.27
9	2.24	0.44	0.09	0.54	2.94	2.35	1.22	4.04
10	1.9	0.12	0.13	0.54	3.23	1.86	1.20	3.72
11	1.69	0.04	0.11	0.46	3.09	1.95	1.19	3.38
12	1.66	0.07	0.13	0.63	3.11	1.37	1.26	2.88
13	1.67	0.05	0.10	0.25	3.09	1.19	1.17	2.71
14	1.59	0.07	0.52	0.24	3.11	1.11	1.17	2.49
15	2.02	0.07	0.13	0.67	3.38	2.09	1.18	2.37
16	1.55	0.13	0.11	0.44	2.59	1.25	0.88	1.50
17	1.88	0.06	0.13	0.57	3.25	1.39	1.41	2.30
Max	2.28	0.44	0.52	1.23	3.53	2.48	1.54	4.04
Min	1.46	0.04	0.09	0.24	1.62	1.11	0.88	1.50
Average	1.83	0.11	0.17	0.55	2.94	1.70	1.22	2.87
Background	25	71	46.7	1030	105	0.098	43	34

Table 6. Pollution Index (PI) and Pollution load index (PLI)for the heavy metals in the surface sediments of Wadi Al Rayyan area.

	Pollution Index (PI)								PLI
S. No.	Cu	Zn	Fe	Mn	Pb	Cd	Cr	Ni	PLI
1	9.76	0.29	0.65	2.40	15.71	1.03	6.95	12.21	<1
2	11.12	0.64	0.76	6.12	17.14	1.41	7.70	12.54	<1
3	7.28	0.49	0.69	1.57	13.33	1.05	4.83	13.10	<1
4	8.48	0.45	0.57	4.73	12.86	1.08	5.83	14.71	<1
5	8.58	0.98	0.56	1.42	8.10	0.96	5.83	13.32	<1
6	7.72	0.37	0.54	2.03	12.38	0.93	5.77	15.24	<1
7	9.94	0.33	0.55	1.78	10.95	1.01	5.98	16.49	<1
8	10.44	0.48	1.39	1.72	17.58	1.01	6.17	16.79	<1
9	11.18	2.20	0.43	2.67	14.65	1.35	6.09	20.71	<1
10	9.48	0.59	0.63	2.71	16.11	1.03	5.99	19.06	<1
11	8.46	0.20	0.54	2.31	15.38	1.07	5.92	17.35	<1
12	8.32	0.35	0.63	3.15	15.48	0.95	6.26	14.79	<1
13	8.36	0.25	0.50	1.25	15.41	0.89	5.85	13.91	<1
14	7.92	0.32	2.57	1.18	15.50	0.83	5.83	12.81	<1
15	10.06	0.36	0.64	3.32	16.85	1.14	5.86	12.15	<1
16	7.76	0.63	0.54	2.18	12.90	0.84	4.38	7.68	<1
17	9.36	0.30	0.64	2.84	16.19	0.98	7.07	11.79	<1
Max	11.18	2.20	2.57	6.12	17.58	1.41	7.70	20.71	<1
Min	7.28	0.20	0.43	1.18	8.10	0.83	4.38	7.68	<1
Average	9.09	0.61	0.83	2.67	14.33	1.04	6.02	14.37	<1

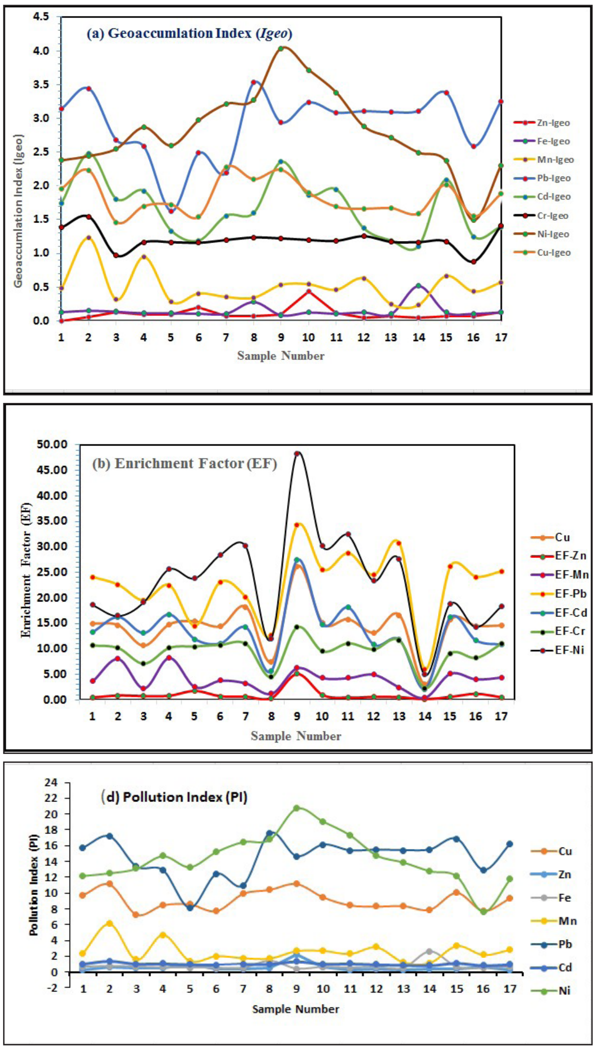


Figure 3. Distribution for (a) Geoaccumulation index (I_{geo}), (b) Enrichment Factor (EF) and (d) Pollution Index (PI) for the surface sediments samples Wadi Al Rayyan area.

6. Conclusions

The data analyses have been performed for this study using several verification methods in order to provide an important tool for a better understanding of the complex dynamics of heavy metal pollution in Wadi Al Rayyan area. These methods include, concentration (ppm) of the heavy metals (Cu, Zn, Fe, Mn, Pb, Cd, Cr, Ni), OM%, CaO wt%, pH and EC µs cm^{-1} and correlation coefficients analyses. Sediment contamination assessment was used for the Enrichment Factor (EF), Geoaccumulation Index (I_{geo}), Pollution Index (PI), and Pollution Load Index (PLI). The main conclusions of this study can be summarized as follows:

1- The heavy mineral concentration in the sample distributions along Wadi Al Rayyan area show the concentration Enrichment of Cu, Mn, Pb, Cd, Cr and Ni along the Wadi Rayyan sediments from upstream to downstream, but a valuable concentration of Zn and Fe was found along the Wadi. The enrichment concentration of these elements are located near the roads across the stream and in areas of human activity along the Wadi. The elevated concentrations of the heavy metals in the sediments of Wadi Al Rayyan may be ascribed to the anthropogenic sources as well as the human and agricultural activities (Calace et al., 2005). The sources include fertilizers and pesticides used in agricultural activities and sewage for untreated municipal sludge, wastewater discharges and seepage of the running of the surface water for transport and deposition by precipitation of solid particles from suspension (Zhang et al., 2011).

2- The Pearson's correlation coefficients for the contents of Cu, Zn, Fe, Mn, Pb, Cd, Cr and Ni for the surface sediments of Wadi Al Rayyan show a positive correlation for Cu with Cd and Cr; Zn with Cd; Cr, Mn with Cd and Cr. The results revealed that the metal concentration rises due to the anthropogenic sources of pollution identified to be the human, industrial, and agricultural activities such as sewage effluents, waste combustion, steel processing, fertilizers and pesticides. Low positive linear correlations have been found among the concentrations of Cu, Zn, Pb, Cd and Cr. These results indicate that these metals have complicated geochemical behaviors and different sources.

3- The content of OM% in Wadi Al Rayyan sediments is related to the organic content of the plant remains, farm residues and accumulation of animal manures, adjacent to the Wadi site at various stages of decomposition, such as cells and tissues of plant organisms as well as substances from plant roots and soil microbes which have been considered as additional sources.

4- The Enrichment Factor (EF) results reveal the sediments to be moderately-to-strongly polluted and strongly-to extremely polluted. This may result in risky significant-to-strong pollution with Cu, Mn, Pb, Cd and Ni, while the values for Zn and Cr indicate a moderate level of pollution. The strongly polluted sediments with Cu, Mn, Pb, Cd and Ni, depend on the location of the sample along the Wadi Rayyan area and the contributors to the element concentrations.

5- The I_{geo} values of the surface sediments of Wadi Al Rayyan show a strong level of pollution with respect to Zn, Fe, Mn, Cd, Cr, Ni and Pb respectively. As for Zn, Fe, and Mn the values show no pollution ($0 < Igeo \leq 1$), and the I_{geo} values of Cu, Cd, and Cr show a moderate level of pollution ($1 < Igeo \leq 2$), and those for Ni and Pb show a strong level of pollution ($2 < Igeo \leq 3$). The strong level of pollution with respect to Pb and Ni, depends on the location of the sample along the wadi.

6- The PI values of Zn, Fe, and Cd for all of the samples show a low level of pollution ($PI < 1$). The PI of Cu, Pb, Cr and Ni shows a high level of pollution ($PI \geq 3$) for all of the samples. PI for Mn shows a middle level of pollution ($PI \leq 1$) except in samples 2, 4, 15 which had a high level of pollution ($PI \geq 3$). The PLI value for all of the surface sediments of Wadi Al Rayyan were found to be less than 1 (PLI<1). These results reveal that the sediments are unpolluted with respect to Cu, Zn, Fe, Mn, Pb, Cd, Cr, and Ni.

7- The results of the statistical analysis and distribution of the metal contamination suggest that human activities, as well as industrial and agricultural activities are the most important pollution sources in the study area.

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