

Assessment of Heavy Metal Pollution of Surface Sediments in Wadi Shu'ayb, Jordan

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ABSTRACT

Stream sediment samples (n = 20) were collected from Wadi Shu'ayb area, extracted and then subjected to Atomic Absorption Spectrometer (AAS) analysis for determining the heavy metal concentration. Calciometry method was used to determine the CO_3^{2-} content and loss on ignition at 555°C was used to determine the total organic matter. The average concentrations of Zn and Cd in the sediment samples were higher than those in uncontaminated sediments, probably due to anthropogenic sources such as fertilizers and pesticides used in agricultural activities as well as the effluent of the Salt and Fuhays treatment plant. Correlation analysis revealed positive correlations among the values of Zn, Cd, Mn, Fe, Pb, TOM and CO_3^{2-} and negative correlations among the values of TOM, Cu, Mn, Fe, Pb and CO_3^{2-} . These results indicate that heavy metals in the sediments of Wadi Shu'ayb have different anthropogenic and natural sources and confirm the complicated behavior of these metals. The sediment pollution assessment was performed by using the geoaccumulation index, pollution load index and enrichment factor. Igeo results revealed that the sediments of Wadi Shu'ayb were uncontaminated with Cu, Zn, Mn and Pb; moderately contaminated with Cd; and heavily contaminated with Fe. The PLI values of <1 indicated that the sediments were unpolluted with Cu, Zn, Cd, Mn, Fe and Pb. The calculation of EF showed that Cu, Zn and Pb were depleted (0.108, 0.031 and 0.379 ppm, respectively), whereas Mn and Cd were enriched (3.48 and 21.12 ppm, respectively) in the sediments.

KEYWORDS: Heavy metal, Pollution, Assessment, Sediments, Wadi Shu'ayb, Jordan.

INTRODUCTION

Pollution of heavy metals is a global issue because these metals have toxic effects on living organisms at high concentrations (Forstner, 1990; Harte et al., 1991; Schuurmann and Market, 1998; MacFarlane and Burchett, 2000; Habes and Nigem, 2006). In addition, heavy metals are of high ecological significance because, as they are not removed from water by self-

purification methods, they tend to accumulate in reservoirs and thereby enter the food chain (Loska and Wiechula, 2003). The elevation of heavy metal levels in a reservoir was shown to be mainly due to an increase in their concentrations in the bottom sediment (Habes and Nigem, 2006). The occurrence of heavy metal in the environment results from anthropogenic activities. In addition, natural processes such as chemical weathering of rocks and volcanic activities, play a role in enriching the water of the reservoirs with heavy metals (Forstner and Wittmann, 1979; Forstner

and Wittmann, 1983; Nriagu, 1989; Veena et al., 1997).

Stream sediments are generally a mixture of several components including different mineral species and organic debris. They represent an ultimate sink for heavy metals discharged into the environment (Gibbs, 1977; Luoma and Bryan, 1981; Bettinetti et al., 2003; Hollert et al., 2003). Heavy metals occur in nature at low concentrations under normal conditions. Anthropogenic activities can raise the levels of these metals in various parts of the ecosystems (Tayel and Anwar, 2001). They accumulate in the sediments through complex physical and chemical adsorption mechanisms, depending upon the nature of the sediment matrix and the properties of the adsorbed compounds (Maher and Aislabie, 1992; Leivouri, 1998; Ankley et al., 1992). Heavy metals of river sediments in hydrological systems are more sensitive than dissolved concentrations and are indicators of contamination (Gaiero et al., 1997). Several processes lead to the association of heavy metals with solid phases, such as direct absorption by fine-grained inorganic clay particles, adsorption of hydrous ferric and manganese oxides, which may in turn be associated with clays; adsorption on or complication with natural organic substances, which may also be associated with inorganic particles, and direct precipitation as new solid phases (Gibbs, 1973; Habes and Nigem, 2006). The adsorption process is influenced by several chemical and physical factors such as pH, oxidative-reductive potential, dissolved oxygen content, organic and inorganic carbon content and the presence in water phase of some anions and cations that can bind or co-precipitate the water-dissolved or suspended pollutants (Di Toro et al., 1991; Calmano et al., 1993; Wen and Allen, 1999). The aim of this study was to focus on heavy metal (Cu, Zn, Cd, Mn, Fe and Pb) concentrations and distribution in sediments from Wadi Shu'ayb area and to assess heavy metal pollution to the sediment.

STUDY AREA

The lithology of the study area was characterized by outcrops of Lower Cretaceous sandstone (Kurnub Sandstone Formation covers the northeast region of the study area), overlain by Upper Cretaceous carbonate rock formations. The formation is divided into a lower and an upper part: the Ajlun group composed of Naur; Fuhays; Hummar; ~~Sih~~, and Wadi As -Sir formations and the Belqa group composed of Wadi Umm-Ghudran and Amman silicified limestone (Shawabkeh, 2004). The Upper Cretaceous formations are exposed at higher elevation and dominated by highly weathered caliches. The two banks along Wadi Shu'ayb consist of limestone with thin layers of chert, marl and chalky marl. Kurnub Sandstone (Lower Cretaceous) covers the upper part of Wadi Shu'ayb with ferruginous interbedded siltstone.

The structure of the study area is affected by the Dead Sea Transform Fault system, as several faults were detected in the investigated area, where the main valleys lie along the main fault systems. The regional fault system (Wadi Shu'ayb) is 25 km long and extends along the eastern flank of Wadi Shu'ayb in the North - Northeast (NNE) direction (Mikbel and Zacher, 1981; Shawabkeh, 2004). The valleys of the area are relatively young with V-shaped morphology and steep escarpments along their paths.

The area study Wadi Shu'ayb is affected by different environmental activities such as agriculture, especially of vegetables and plant, highway network along the Wadi, two wastewater treatment plants (Salt and Fuhays) and human activity. Agriculture in Wadi Shu'ayb area depends on the rainfall in the winter season and irrigation with wastewater in other seasons. The average rainfall here is 346 mm (MWI, 2007) during the winter season, which starts in November and ends in March. Surface runoff is formed in summer due to flushing of pollutants of the wastewater treatment plant into drainage by the rainfall (Duqqah et al., 2005).

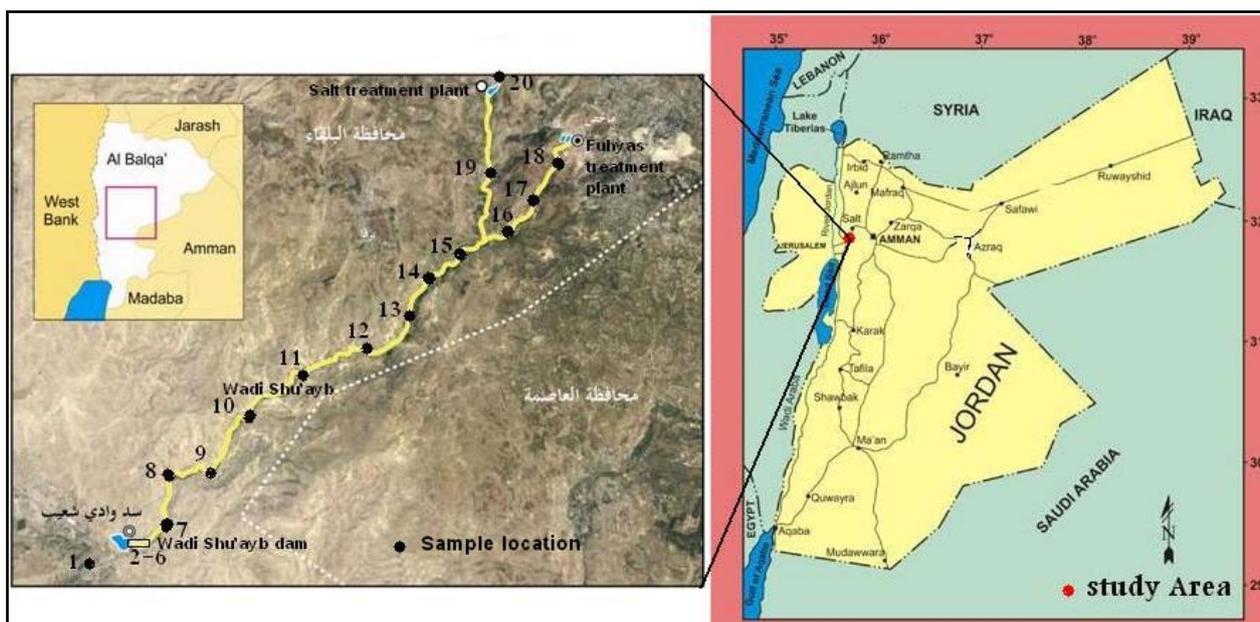


Figure (1): Location map of the study area

SAMPLING AND ANALYSIS

Twenty composite samples from the top stream (1-20 cm) were collected between June and October 2013 from along the main valley of Wadi Shu'ayb (Figure 1). The samples were transported to the laboratory in polyethylene bags and dried in the oven at 55°C for 24 h. All dried samples were sieved, and the particles <60 μm (0.05 mm) in size were analyzed, as it has been proven that this size is the best size for analysis for arid and semi-arid regions (Saffarini and Lahwani, 1992). The extraction method was used in pollution studies to determine the heavy metal contents such as Cu, Zn, Cd, Mn, Fe and Pb, and the best extraction results were obtained with 0.5 M nitric acid (HNO₃). Twenty gram of dry sediment was accurately weighed, digested with 100 ML of HNO₃ and shaken vigorously every 30 min. The final extracts were filtered through 0.45-μm filters. The supernatant contents of heavy metals (Cu, Zn, Cd, Mn, Fe and Pb) were determined by using atomic absorption spectrophotometer (AAS; 2280 Model; Perkin Elmer) at the University of Jordan. Quality control was performed for Cd, Fe, Zn, Mn, Cu, Ni, Pb and Cr by using Merck ICP4 standard solutions. 95% confidence level was used. The total organic matter

(TOM) in each sample was determined by using loss on ignition at 555°C. The calcimetry method (Loring and Rantala, 1992) was used to determine the carbonate (CO₃²⁻) content of the sediments. In this method, the amount of carbon dioxide (CO₂) released from the reaction is dependent on the amount of calcium carbonate (CaCO₃) in the sample. Duplicate analyses were performed and recovery of known additions was used to ensure data quality.

RESULTS AND DISCUSSION

Heavy Metal Distribution

The concentrations of heavy metal determined in this study are shown in Table 1. The variable concentration distribution along Wadi Shu'ayb area (Figure 2) showed TOM, CO₃²⁻, Zn and Cd enrichment in the Wadi Shu'ayb dam sediments. Relatively high concentration of Fe was obtained in the dam sediment and upstream near the outcrops of ferruginous sandstone. The average concentrations of Zn and Cd in Wadi Shu'ayb (0.28 and 0.265 ppm, respectively) were higher compared to those in uncontaminated sediments and soils. According to Bowen (1979), the Zn and Cd

levels in uncontaminated sediments and soils are 0.09 and 0.0003 mg g⁻¹ dry wt., respectively. The elevated concentrations of Zn and Cd in the sediments of Wadi Shu'ayb were probably due to the anthropogenic sources nearby the stream sediment site. These sources include fertilizers and pesticides used in agricultural activities and the effluent of Salt and Fuhays treatment plant. The wastewater treatment plant of Salt and Fuhays was constructed in the upper reaches of Wadi Shu'ayb. The effluent of the treatment plant flood waters along the Wadi Shu'ayb area.

The TOM in the sediments of Wadi Shu'ayb was 0.32–7.20%, with an average of 2.03% (Table 1). The relatively high content of TOM in Wadi Shu'ayb

sediments is related to the high organic content of the plant remains 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 CO₃²⁻ content of sediments of Wadi Shu'ayb was 11–82%, with an average of 53.05% (Table 1). The high CO₃²⁻ content in the sediments is mainly derived from the carbonate rocks exposed at the long Wadi site and in the catchment area. These rocks mainly include limestone, chalky marl and caliches. The carbonate mineral is important for co-precipitation of heavy metals such as Cd and Zn (Alloway, 1990).

Table 1. Concentrations of Cu, Zn, Cd, Mn, Fe and Pb (ppm, dry wt.), TOM and CO₃²⁻ (%) in stream sediment samples collected from Wadi Shu'ayb area

S. No.	%		ppm					
	TOM %	CO ₃ ²⁻	Cu	Zn	Cd	Mn	Fe	Pb
wsh1	1.70	38	0.266	0.12	0.152	4.65	16.40	1.117
wsh2	4.02	82	0.228	0.44	0.320	4.61	7.10	0.95
wsh3	6.12	69	0.241	0.65	0.302	3.95	6.31	0.82
wsh4	7.20	58	0.231	0.84	0.488	3.09	5.40	0.96
wsh5	1.31	71	0.229	0.72	0.528	4.93	6.40	0.83
wsh6	2.34	71	0.227	0.93	0.411	4.3	4.42	1.131
wsh7	0.75	57	0.262	0.12	0.214	2.72	2.23	1.122
wsh8	1.35	38	0.264	0.17	0.278	4.05	19.61	1.123
wsh9	1.10	49	0.269	0.12	0.250	3.95	5.03	1.12
wsh10	1.23	51	0.283	0.11	0.274	4.7	6.04	1.122
wsh11	2.11	32	0.288	0.16	0.212	5.5	7.12	1.11
wsh12	1.30	52	0.287	0.12	0.268	4.37	9.04	1.13
wsh13	1.60	64	0.277	0.12	0.230	4.42	8.25	1.14
wsh14	2.21	59	0.286	0.19	0.222	5.15	7.20	1.112
wsh15	0.61	54	0.281	0.16	0.302	3.57	6.35	1.13
wsh16	0.78	53	0.210	0.13	0.182	2.59	5.12	1.23
wsh17	0.45	11	0.326	0.21	0.172	5.69	25.00	0.82
wsh18	0.32	13	0.178	0.21	0.182	3.38	19.83	0.42
wsh19	0.71	66	0.240	0.13	0.278	2.74	3.52	0.63
wsh20	3.31	73	0.242	0.03	0.087	3.16	5.21	0.85
Maximum	7.20	82	0.326	0.93	0.528	5.69	25	1.23
Minimum	0.32	11	0.178	0.03	0.087	2.59	2.23	0.42
Average	2.03	53.05	0.26	0.28	0.265	4.08	8.78	0.99

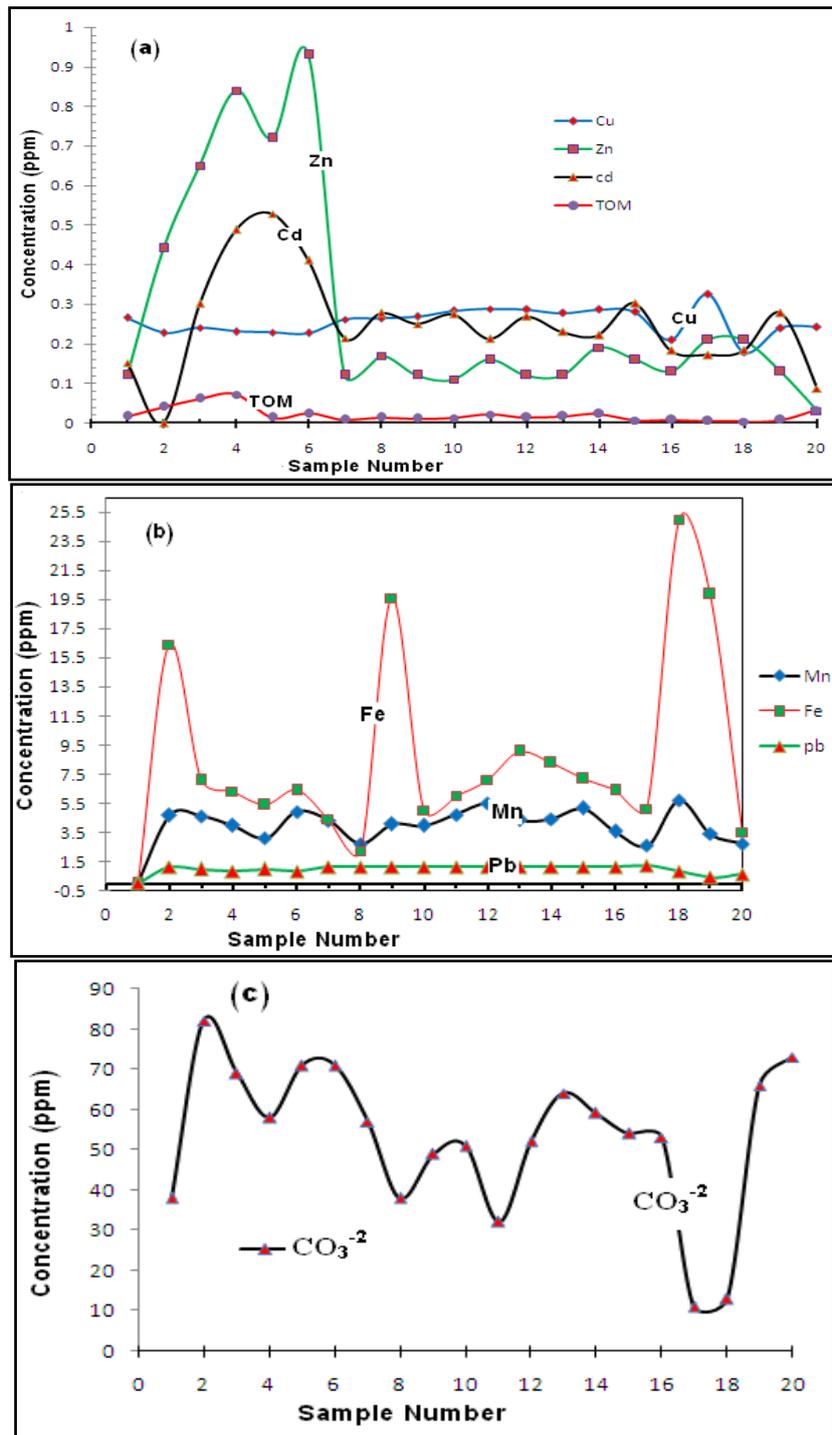


Figure (2): Distribution of heavy metal concentration (ppm) in sediment for Wadi Shu'ayb area (a) TOM, Cu, Zn, Cd (b) Mn, Fe, Pb (c) CO_3^{2-}

Correlation Analysis

The Pearson's correlation coefficients among the contents of TOM, CO_3^{2-} , Cu, Zn, Cd, Mn, Fe and Pb in the sediments of Wadi Shu'ayb are presented in Table 2. Low positive linear correlations among the concentrations of Zn, Cd, Mn, Fe, Pb, TOM and CO_3^{2-} were clearly observed: Zn with CO_3^{2-} ($r = 0.37$), Cd with TOM ($r = 0.337$), Mn with Zn ($r = 0.07$), Mn with Cd ($r = 0.04$), Fe with Cu ($r = 0.2$), Fe with Mn ($r = 0.4$), Pb with CO_3^{2-} ($r = 0.2$), Pb with Cu ($r = 0.45$) and Pb with Mn ($r = 0.16$). Strong positive correlation of Zn with TOM ($r = 0.61$), Cd with Zn ($r = 0.82$) and Mn with Cu ($r = 0.58$) revealed the affinity of these

metals to TOM and the sources of pollution were identified to be fertilizers and pesticides used in agricultural activities and the effluent of Salt and Fuhays City treatment plant. Negative correlations were observed for TOM with Cu, Mn, Fe and Pb; CO_3^{2-} with Cu, Mn and Fe; Cu with Zn and Cd; Zn with Fe and Pb; Cd with Fe and Pb; and Fe with Pb (Table 2). These results indicate that the analyzed heavy metals are not associated with each other and no relationship exists between the variables. Furthermore, these metals have different anthropogenic and natural sources in the sediments of Wadi Shu'ayb (Habes and Nigem, 2006).

Table 2. Correlation matrix of heavy metals in the sediments of the Wadi Shu'ayb area

Variable	TOM	CO_3^{2-}	Cu	Zn	Cd	Mn	Fe
CO_3^{2-}	0.443						
Cu	-0.220	-0.256					
Zn	0.611	0.372	-0.395				
Cd	0.372	0.410	-0.227	0.821			
Mn	-0.067	-0.249	0.582	0.069	0.037		
Fe	-0.279	-0.786	0.193	-0.194	-0.310	0.392	
Pb	-0.054	0.186	0.456	-0.138	-0.006	0.160	-0.297

Sediment Contamination Assessment

Geoaccumulation index (Igeo) is used to assess heavy metal pollution in the sediments of Wadi Shu'ayb. Igeo was introduced by Muller (1981) to measure the degree of metal pollution in aquatic sediment studies. It is calculated as follows: $I_{geo} = \text{Log}_2 (C_n/1.5B_n)$, where C_n is the measured concentration of a heavy metal in stream sediments, B_n is the geochemical background value in average shale (Turekian and Wedepohl, 1961) of element n and 1.5 is the background matrix correction factor due to lithogenic effects. The Igeo consists of seven classes (0–6), ranging from background concentration to extremely heavily polluted. The highest grade (class 6) reflects 100-fold enrichment above the background

values. Igeo value <0 indicates practically unpolluted; $>0-1$ indicates unpolluted to moderately to strongly polluted; $>1-2$ indicates moderately polluted; $>2-3$ indicates moderately to strongly polluted, and >5 indicates very strongly polluted. Thus, the Igeo values indicated that the sediments of Wadi Shu'ayb are moderately polluted with Cd and strongly polluted with Fe (Figure 3a). Cd pollution is contributed by the use of fertilizers and pesticides in agricultural activities and the effluent of Salt and Fuhays City treatment plant, whereas Fe pollution is contributed by high weathering of ferruginous sandstone, including Kurnub Sandstone group covering the study area. The results of Igeo (Table 3) revealed that the sediments are not polluted with Cu, Zn, Mn and Pb.

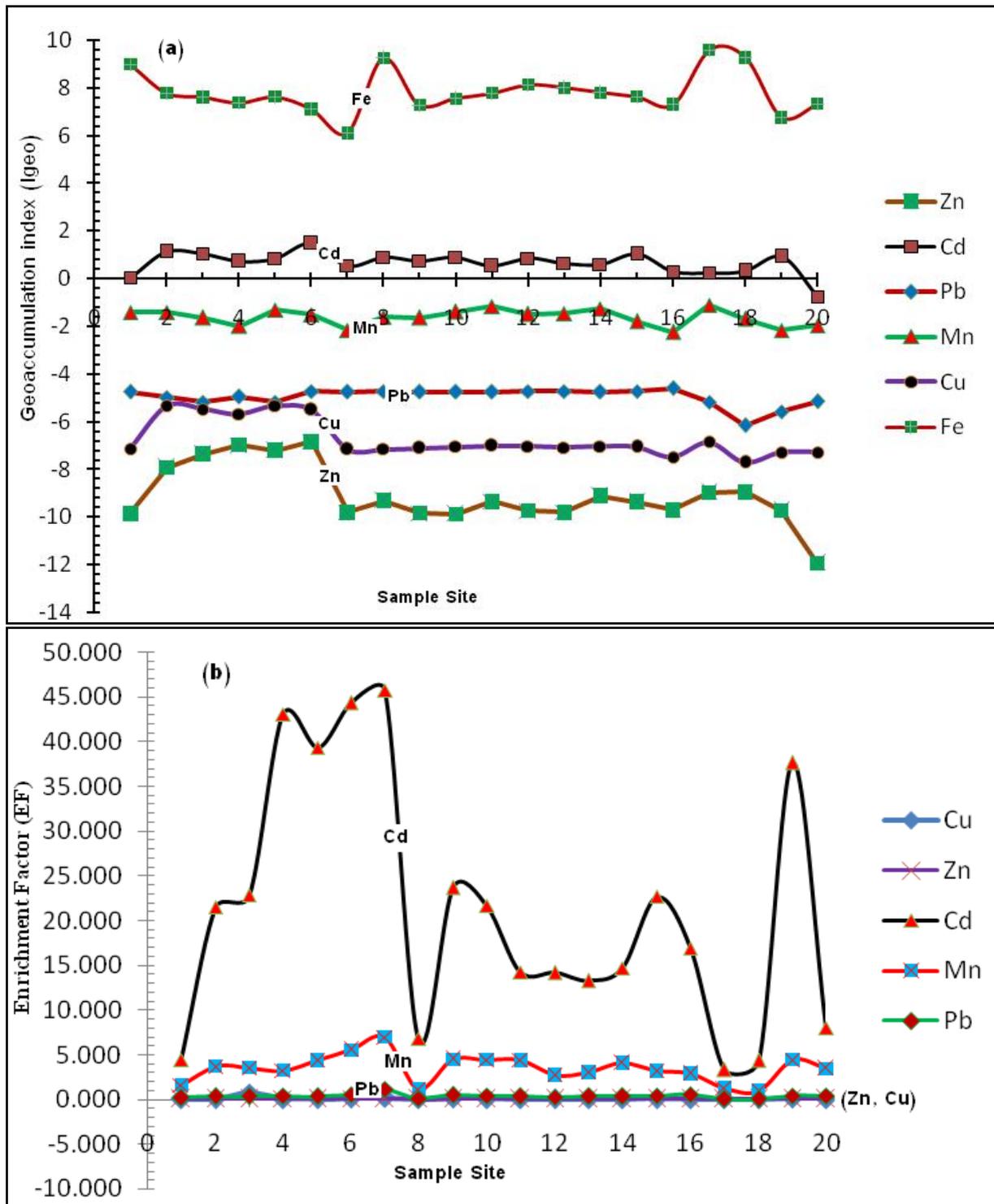


Figure 3): (a) Geoaccumulation index (Igeo) and (b) enrichment factor (EF) for sediments from the Wadi Shu'ayb area

Table 3. Geoaccumulation index (Igeo), contamination factor (CF), pollution load index (PLI) and enrichment factor (EF) of heavy metal sediments of Wadi Shu'ayb area

	Geoaccumulation index (Igeo)						
	Cu	Zn	Cd	Mn	Fe	Pb	
Maximum	-5.33	-6.84	1.48	-1.11	9.60		-4.6
Minimum	-7.72	-11.94	-0.75	-2.24	6.12		-6.15
Average	-6.74	-9.08	0.65	-1.62	7.82		-4.95
	Contamination Factor (CF)						
	Cu	Zn	Cd	Mn	Fe	Pb	PLI
Maximum	0.0130	0.0131	5.380	0.694	0.535	0.0615	<1
Minimum	0.0071	0.0004	0.744	0.316	0.048	0.0210	<1
Average	0.0103	0.0043	2.119	0.497	0.197	0.0497	<1
	Enrichment Factor (EF)						
	Cu	Zn	Cd	Mn	Fe	Pb	
Maximum	0.710	0.140	45.729	6.946	-	1.174	
Minimum	0.016	0.004	3.278	0.971	-	0.050	
Average	0.108	0.031	21.115	3.479	-	0.379	
Background	25	0.095	0.098	8.2	46.70	20	

Pollution load index (PLI) was used to assess the heavy metal pollution in the sediments of Wadi Shu'ayb area. PLI for each site was evaluated as indicated by Tomilson et al. (1980) and calculated as follows: $PLI = (CF_1 * CF_2 * \dots * CF_n)^{1/n}$, where n is the number of metals (six in the present study) and CF is the contamination factor. CF is calculated as follows: $CF = [C_{\text{heavy metal}}] / [C_{\text{background}}]$, where C is the measured concentration of a heavy metal in stream sediments, background value in average shale (Turekian and Wedepohl, 1961). PLI >1 indicates pollution, whereas PLI value <1 indicates no pollution (Chakravarty and Patgiri, 2009; Seshan et al., 2010). The PLI value of Wadi Shu'ayb sediments was <1 (Table 3), revealing that the sediments are unpolluted with Cu, Zn, Cd, Mn, Fe and Pb.

Enrichment factor (EF) was used to assess the degree of contamination and the possible anthropogenic impact on the sediments of Wadi Shu'ayb area (Simex and Helz, 1981). The geochemical normalization of the heavy metals' data was employed for conservative elements such as Al, Fe and Si.

Several studies successfully used Fe (reference) to normalize heavy metal contaminants (Schiff and Weisberg, 1999; Baptista et al., 2000; Mucha et al., 2003). In this study, Fe was used as a conservative tracer to differentiate natural components from anthropogenic ones. EF was calculated as follows: $EF = (M/Fe)_{\text{sample}} / (M/Fe)_{\text{background}}$, where $(M/Fe)_{\text{sample}}$ is the ratio of metal and Fe concentration of the sample, $(M/Fe)_{\text{background}}$ is the ratio of metal and Fe concentration of a background (Ergin et al., 1991; Loska and Wiechula, 2003; Chakravarty and Patgiri, 2009; Seshan et al., 2010; Bentum et al., 2011). The background concentrations of the heavy metal study (Cu, Zn, Cd, Mn, Fe and Pb) were taken from Turekian and Wedepohl (1961). According to Sutherland (2000), five contamination categories were recognized based on the EFs; EF <2 indicating depletion to minimal enrichment; EF = 2–5 indicating moderate enrichment; EF = 5–20 indicating significant enrichment; EF = 20–40 indicating very high enrichment; EF >40 indicating extremely high enrichment. The EF values of Cu, Zn, Cd, Mn and Pb in the Wadi Shu'ayb sediments are

shown in Table 3 and Figure 3b. According to Zhang and Liu (2002), EF values of 0.5–1.5 indicate that the metal is derived entirely from crustal materials or natural processes, whereas EF values >1.5 suggest an anthropogenic source. The results of the present study showed that Cd and Mn were significantly enriched in the Wadi Shu'ayb sediments, because EF values of these two metals were greater than 1.5 (Table 2). The highest average of EF was noted for Cd, with a high enrichment value of 21.11 (average). Mn had the second highest EF, with a moderate enrichment value of 3.48 (average) (Table 2). Cd and Mn at the study site were deriving from anthropogenic sources including fertilizers and pesticides used in agricultural activities and the effluent of Salt and Fuhays City treatment plant. These two metals have been the major metals contaminating the sediments of Wadi Shu'ayb area in the past few years, while Cu, Zn and Pb are minor contaminants. The difference in the EF values may be due to the difference in the magnitude of input for each metal in the sediment or due to the difference in the removal rate of each metal from the sediment.

CONCLUSIONS

Data analyses were performed by several verification methods in order to provide an important tool for a better understanding of the complex dynamics of heavy metal pollution. These methods included correlation coefficient analysis of concentrations among TOM, CO_3^{2-} , Cu, Zn, Cd, Mn, Fe and Pb, which showed weak to strong positive and negative correlations, indicating that these metals have complicated geochemical behaviors. Sediment

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pollution in the present study was assessed by using the Igeo, PLI and EF values. The results of Igeo revealed that the sediments of Wadi Shu'ayb area are unpolluted with Cu, Zn, Mn and Pb; moderately polluted with Cd; and strongly polluted with Fe. The elevated concentrations of Cd and Fe are due to anthropogenic sources, including fertilizers and pesticides used in agricultural activities, effluent of Salt and Fuhays treatment plant and natural weathering of ferruginous sandstone including the Kurnub Sandstone group covering the blank site of the study area. The PLI of Wadi Shu'ayb sediments was <1, indicating that the sediments were unpolluted with Cu, Zn, Cd, Mn, Fe and Pb. The results of EF showed high Cd enrichment (21.11 ppm) and moderate Mn enrichment (3.48 ppm). Natural weathering of rocks exposed at the Wadi Shu'ayb site can be considered as an additional source of heavy metals to the sediments of Wadi Shu'ayb, such as the Kurnub Sandstone group.

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