An Assessment of Heavy Metals Contamination in Surface Sediment of Hodeidah Coast and Khor Katheib, Red Sea, Yemen

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Abstract:

The Creek Khor Katheib encompasses Al-Hodeidah commercial seaport, the main Yemeni seaport on the Red Sea and receives domestic and industrial untreated wastewaters from Al-Hodeidah city. The purpose of the present study was to obtain a preliminary assessment of the level and distribution of nine heavy metals (Cu, Cd, Zn, Co, Ni, Cr, Mn, Fe and Al) in surface sediments collected from 25 stations in and outside the creek. Significant spatial variation in concentrations was observed for all metals. Concentration data were processed using correlation analysis and factor analysis. The correlation analysis of the results showed negative correlations between heavy metals and $CaCO_3$, and no correlations between total organic carbon and heavy metals indicating that these metals have complicated biogeochemical behaviours. Pollution status was evaluated using the enrichment factor, and contamination factor. Based on the enrichment factor, Khor Katheib sediments were treated as a moderately severe enrichment with Cd, moderate enrichment with Cr whereas, Mn and Ni had minor enrichment.

Keywords: Heavy metal; Assessment; Sediments; Statistical analysis; Enrichment; Khor Katheib; Red Sea; Yemen.

Introduction

Coastal sediments are important hosts for heavy metal pollutants and play an important role in determining the fate and effects of a wide variety of contaminants (Chester and Voutsinou 1981). Urban and industrial activities contribute to the introduction of significant amounts of pollutants (among them trace metals) into the marine environment and affect directly the coastal systems where they are quite often deposited to sediment through physical, chemical or biological mechanisms (Dassenakis, et al. 2003; Calace, et al. 2005; Choi et al. 2007; Naji and Ismail 2011; Zhan et al. 2010; Zhang et al. 2012; Yuan et al. 2012).

The distribution of metals in sediments is very important, because sediment concentrates metals from aquatic systems and represents an appropriate medium to monitor contamination of heavy metals in an aquatic environment (El-Rayis and Abdallah 2005; Bettinetti et al. 2003; Hollert et al. 2003; Sarkar, et al. 2004). The sediments are usually the ultimate sink of heavy metals discharged into the aquatic environments, therefore, analysis of heavy metals (and other contaminants) in the sediments offers convenient and accurate means of detecting and assessing the degree of pollution (FAO 1994; Chester and Voutsinou 1981). For this reason, several studies aimed at evaluating heavy-metal contamination in harbors have focused on the sediment compartment (Denton et al. 2005; Guerra-Garcia and Garcia-Gomez 2005; Huerta-Diaz et al. 2007; Zonta et al. 2007).

The elevation of metal levels in an aquatic environment is shown mainly by an increase in their concentrations in the recent surface sediments. Their occurrence in the environment results primarily from anthropogenic activities. In addition, the natural processes, such as weathering of rocks and volcanic activities play a noticeable role in enriching the water of reservoirs with heavy metals (Forstner and Wittmann1983; Nriagu1989; Veena et al.1997; Zhang et al. 2003). Under natural condition, some elements have sometimes been found in anomalously high concentrations in the marine environment, apparently in relation to naturally-occurring deposits (Bryan 1976).

The aim of the present study is to investigate the distribution of heavy metals (Cu, Cd, Zn, Co, Ni, Cr, Mn, Fe and Al) and to assess the contamination levels of the surface sediments in Khor Katheib on the Red Sea coast of Yemen.

Study Area

The study area covers the Red Sea coastal zone of Yemen from All-Mandar Village south of Al-Hodeidah the north of Al-Gabana including the Creek Khor Katheib, (Fig. 1). It also includes Al-Hodeidah commercial seaport, which is located at the blind end of the V-shape Creek. The creek lies between the main land on the east and the side arm or spit on the east, called Ras Katheib Spit. Therefore, Ras Katheib protrusion is considered as natural break water. Inside the creek, the navigational canal for commercial ships is the axial line that is extending along the creek with a depth of >10 m, and is about 30 km long. The banks or littorals that are lying east and west of the axial line are shallow. The cross section at any part of the creek almost takes a V shape. The width of the creek generally varies between 2 km near the behind end (the harbor) and widens northwards to about 7 km at the mouth. The surface areas of each of the harbor part and the inner of the creek are respectively 3 and 47 million m^2, with general average depth 3.5 m (Al-Edresy 2012).

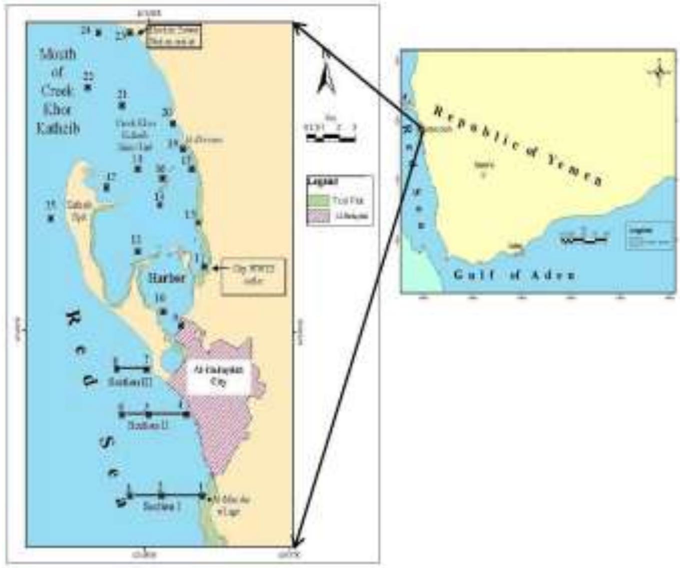

Fig. 1 Study area and location of sediments sampling stations in Khor Katheib, Red Sea, Yemen

Material and Methods

Sample collection and preparation:

Twenty five stations were selected to cover the entire area of the Creek Khor Katheib and the coastal water outside the Creek and southward along the coast of Al-Hodeidah City. The sites of these stations are shown in Figure. 1. The sediment samples were collected using a Van-veen Grab sampler. After retrieval of the sampler, each sediment sample was placed in a self-sealed plastic bag and kept in an ice box before transferring to the laboratory and storing in a deep freeze for later analysis.

In the laboratory, the sediment samples were first thawed before drying in an oven at 50 C° for overnight. Part of the sediment was grind finely in an agate mortar and finally sieved through a 63-µm mesh sieve, before storage in a Patterson Jars to be ready for subsequent chemical analysis. The other part was used for grain size analysis.

Determination of carbonate and total organic matter:

The total carbonate was determined by titration technique described by (Black 1965). The method used for the determination of total organic matter

(TOM) was the loss in weight by heat ignition described by Byers et al. (1987).

Determination of Total Heavy Metals**:**

After drying the sediment samples and grinding, a 0.5 g subsample of each sample was weighed and placed in a Teflon cup and the analysis commenced using the same method for TSM described in UNEP/IAEA (1986). 2 ml of concentrated nitric acid was added to each sample and evaporated to near dryness at 80°C and 6 ml of mixed HNO_3 – $HClO_4$ – HF (3:2:1) was added to each sample. After complete digestion, the sample was evaporated to near dryness and 5 ml of 0.1N HCl was added to each sample and completed to 25 ml in a volumetric flask. The heavy Metals including Cu, Cd, Zn, Co, Ni, Cr, Mn, Fe and Al were analyzed by using Atomic Absorption Spectrophotometer (Perkin-Elmer Model A Analyst 800 series). Blanks were included in each batch of analysis. Calibration standards were regularly performed to evaluate the accuracy of the analytical method.

Statistical analysis

All statistical aspects (Pearson's correlation coefficients matrix and principal component analysis) were calculated. Statistical Program for Social Sciences (SPSS program version 16.0 for Windows) was used. Moreover, Surfer-8 software was utilized to show the distribution of the heavy metals accumulation in the area. Evaluation of heavy metal pollution in the surface sediments of the Creek Khor Katheib was carried out using the enrichment factor (EF) (Li and Schoonmaker, 2005) and the contamination factor (Cf) (Hakanson, 1980).

Results and Discussion

Sediment characteristics

The results of the granulometric analysis of the surface sediment samples of the Creek Khor Katheib and Al-Hodeidah coastal area are listed in Table 1. The mud fraction of the sediments ranged between 0.2 - 88.90 %, with an average of 21.83%. The mud fraction in the sediment samples represented less than 50 % of total sediment except for those at the stations 10, 11, 16 and 19, inside the Creek where this fraction reached values higher than 50 %. The mud content was > 88 % at the station 16. This means that the sediments in the present study area are mainly sandy.

The content of $CaCO_3$ was 0.0-59.9 %, with a general average of 18.1 %. This relatively high carbonate content is mainly derived from the shell fragments, calcareous tests of organisms, and diluted biogenic calcite by detrital material in the sediments. TOC content of sediments was 1.10 - 3.60 %, with a general average of 2.02 %, The relatively high content of TOM (Table 1 and Fig. 2) is mainly related to the high organic matter flux to sediments due to direct discharge of domestic and industrial wastewaters.

Metal concentrations in sediments

The metal concentrations in the sediments are shown in Table 2. The levels of Cu, Cd, Zn, Co, Ni, Cr, Mn, Fe and Al in the sediment samples were 0.0 - 6.20 ppm, 0.0 -1.30 ppm, 1.0 -40.0 ppm, 0.0 -23.0 ppm, 0.0 -67.80 ppm, 0.0 -210.0 ppm, 0.0 -641.0 ppm, 1.4 -22.8 % and 2.9 -27.6 %, respectively. The highest concentrations of Cr, Ni and Co were observed at station 16, whereas the highest concentrations for Cu and Zn were detected at stations 11 and 20. Cd concentrations were high at stations. 15 and 9 located in the entrance of Al-Hodeidah seaport harbor where most shipping activities occur. Therefore, enrichment in those sites is most probably due to the result of ship maintenance and the corrosion of metallic materials, as well as untreated domestic and industrial wastewaters from Al-Hodeidah city.

Previous studies of the surface seawater and sediment samples from the coastal region of the Red Sea coast of Yemen showed that natural with some anthropogenic inputs are the sources of trace metals to region. This is indicated by the low concentrations of all metals except for Cd, Co and Pb in seawater and sediments, which designate possible anthropogenic sources (Al-Shiwafi et al., 2005).

The spatial distributions of the metals concentrations in surface sediments of the study area are illustrated in Figure 3. The distribution of heavy metals generally showed nearly similar patterns for Ni, Co, Cr, Zn and Cu, which were low to high from north to south direction. The concentration of Ni, Co and Cr showed high trend in both south-easterly and east of inner creek (along costal line) but Zn and Cu showed high trend in a northeasterly direction. However, Cd showed a different spatial distribution with increasing trend towards the southwest direction in the creek. On the other hand, Mn, Fe and Al, which, are used as markers of natural metals displays quite similar pattern of distribution (Fig. 3).

Generally, the main natural sources of trace metals are from rock weathering, mineral dissolution in sediments and regional dust transport, as well as to minor anthropogenic inputs of local wastes from coastal facilities and human and developmental activities in main coastal cities (Al-Shiwafi et al., 2005).

Table 1 Mean concentration level of sediment characteristics and heavy metals in sediment of Khor Katheib, Red Sea, Yemen.

Metal range	Al %	Fe %	Mn ppm	Cr ppm	Ni ppm	Co ppm	Zn ppm	Cd ppm	Cu ppm	TOC %	$CaCO_3$	Mud
Min.	2.90	1.40	70.0	0.0	0.0	0.0	1.0	0.0	0.00	1.10	0.0	0.20
Max.	27.60	22.80	641.0	210.0	67.80	23.0	40.0	1.30	6.20	3.60	59.90	88.90
Mean	20.18	11.68	258.12	67.63	16.56	10.54	23.60	0.35	1.75	2.02	18.11	21.83

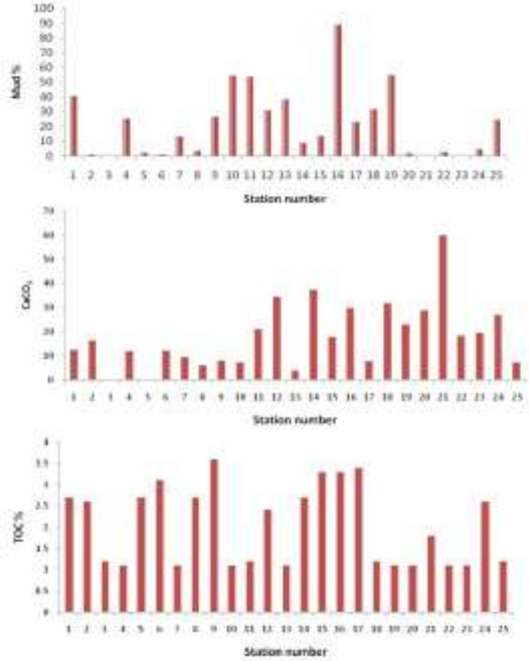

Fig. 2 Distribution of Mud, $CaCO_3$ and TOC (%) in the sediments of Khor Katheib, Red Sea coast, Yemen

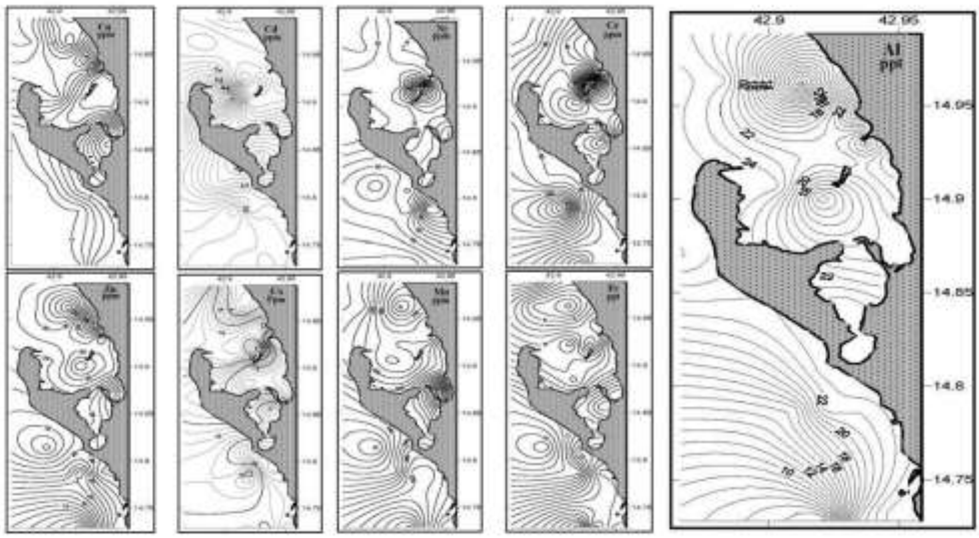

Fig. 3 Areal distributions of heavy metals (Cu, Cd, Ni, Cr, Zn, Co, Mn, Fe and Al) in the sediments of Khor Katheib and Al-Hodeidah coast, Yemen.

Assessment of heavy metals contamination in sediments

Enrichment factor

In the present study, the enrichment factor (EF) is calculated to assess the level of contamination and the possible anthropogenic impact in the sediments of the Creek Khor Katheib. The EF is a good tool to differentiate between the anthropogenic and natural sources of metals in environmental samples (Morillo et al. 2004; Selvaraj et al. 2004; Adamo et al. 2005; Vald'es et al. 2005; Chen et al. 2007; Hung et al. 2009; Fang and Chen, 2010). To identify anomalous metal concentrations, the geochemical normalization of the metal data to a conservative element, such as Al and Fe is employed. Several authors successfully used Al for normalizing the metal concentration in sediments (Huang and Lin, 2003; Woitke et al., 2003; Hung et al., 2009; Fang and Chen, 2010; Chen et al., 2007, 2013; Pekey 2006; Xu, et al. 2014). In the present study, Al has been chosen as a normalization element because of its origin being exclusively lithospheric (Bloundi et al.2009; Jayaprakash et al. 2010).

The EF was calculated according to the following formula:

$$EF = (element/Al)_{sample} / (element/Al)_{background}$$

where $(element/Al)_{sample}$ is the ratio of metal and Al concentrations of the sample, and $(element/Al)_{background}$ is the ratio of metal and Al concentrations of the continental crust background. In this study, the background concentrations of Cu, Cd, Zn, Co, Ni, Cr, Mn, Fe and Al were taken from Taylor and McLennan (1995).

According to Zhang and Liu (2002), EF values between 0.5 and 1.5 indicate that the metal is entirely from crustal materials or natural processes; whereas EF values greater than 1.5 suggest that the sources are more likely to be anthropogenic. The estimated maximum, minimum and mean values of EF are presented in Table 3. The heavy metals of Cd, Cr and Co were found to have EF values greater than 1.5 in most sediment samples, indicating that these heavy metals may be enriched as a result of anthropogenic inputs. The EF values estimated in this work were interpreted as suggested by Birch (2003). EF<1 indicates no enrichment, EF<3 is minor enrichment, EF=3–5 is moderate enrichment, EF=5–10 is moderately severe enrichment, EF=10–25 is severe enrichment, EF=25–50 is very severe enrichment, and EF>50 is extremely severe enrichment.

The calculation of EF showed that Cd, Co, Cr and Ni, were enriched in sediments of Khor Katheib (Table. 2). Cd had the highest EF values among the seven metals studied and it has a moderately severe enrichment (average value 8.59). Cr was moderate enrichment with average values of 3.15. Co, Zn and Mn were minor enrichment (average values were 2.82, 1.01 and 1.33, respectively). Ni and Cu exhibited the lowest EF values among metals studied (average values of 0.93 and 0.14, respectively) with no enrichment (Fig. 4). Generally the difference in EF values for the different metals in the Khor Katheib sediments may be due to the difference in the magnitude of input for each metal into the sediment and/or the difference in the removal rate of each metal from the sediment. Metals can be released into the water phase when changes occur to its physiochemical conditions like pH, redox potential, ionic strength, and the concentration of organic complexing agents (Calmano et al.1990 and Ghrefat et al. 2011; Almasoud et al. 2014).

Contamination factor

The contamination factor (Cf) was also used to assess the level of contamination and the possible anthropogenic impact of contaminants in sediments (Singh et al.2002; Gonzales-Macias et al. 2006; Farkas et al.2007; Cevik et al. 2009). To describe the contamination of a given toxic substance in a basin, Hökanson (1980) proposed the contamination factor (Cf), which is expressed as**;**

$$C_f = C_o / C_b$$

where C_o is the sediment metal content in the sample and C_b is the normal background value of the metal. The Cf was classified into four groups (Hökanson 1980; Savvides et al.1995). Cf <1: low contamination factor; 1≤Cf <3: moderate contamination factor; 3≤Cf <6: considerable contamination factor; Cf ≥6: very high contamination factor.

The results of the Cf (Table 3), demonstrated that Cd could be classified as moderate contaminants. This can be interpreted as a result of cadmium release from the lower sediment layer to the water at the low redox potential with the presence of organic-bound metal. The Cf values indicated that there were no contamination by Cu, Zn, Co, Ni, Cr and Mn (Fig. 5). The result is in agreement with the works of Al-Shiwafi et al. (2005), Nomaan et al. (2012) and Sagheer (2013).

Table 2 Enrichment factor (EF) values for mean metal concentrations in sediments of the Creek Khor Katheib, Red Sea, Yemen.

Metal	**Fe**	**Mn**	**Cr**	**Ni**	**Co**	**Zn**	**Cd**	**Cu**
Min.	0.27	0.67	0.00	0.00	0.00	0.29	0.00	0.00
Max.	1.99	3.16	11.01	3.61	23.67	2.04	36.36	0.57
Mean	1.02	1.33	3.15	0.93	2.82	1.01	8.59	0.14

Table 3 Contamination factor (CF) values for mean metal concentrations in sediments of the Creek Khor Katheib, Red Sea, Yemen.

Metal	**Al**	**Fe**	**Mn**	**Cr**	**Ni**	**Co**	**Zn**	**Cd**	**Cu**
Min.	0.04	0.03	0.08	0.00	0.00	0.00	0.01	0.00	0.00
Max.	0.35	0.49	0.75	2.33	1.00	1.21	0.42	6.50	0.14
Mean	0.25	0.25	0.30	0.75	0.24	0.55	0.25	1.74	0.04

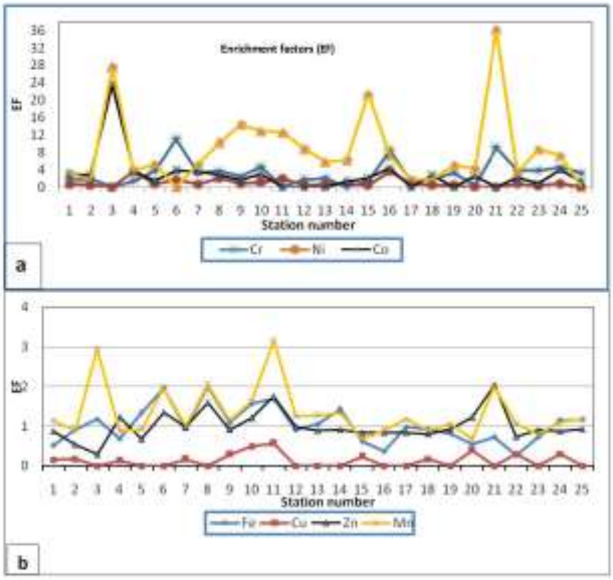

Fig. 4 Enrichment factors (EF) for (a) Cr, Ni and Co (b) Fe, Cu, Zn and Mn in sediments from Khor Katheib, Red Sea coast, Yemen.

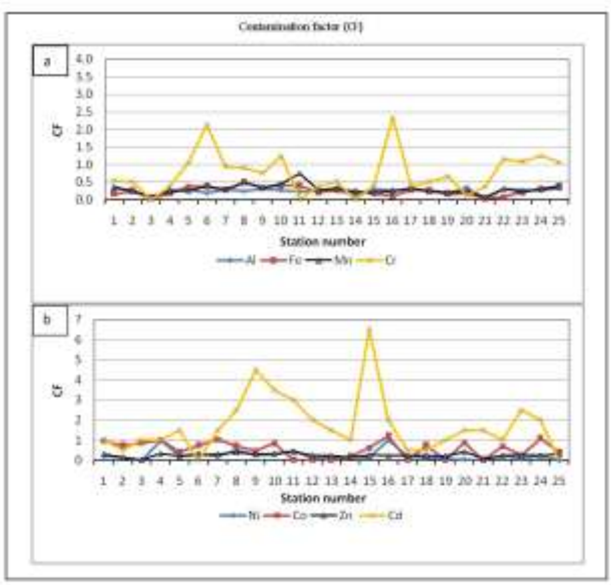

Fig. 5 Contamination factor (CF) for (a) Al, Fe, Mn and Cr, (b) Ni, Co, Zn and Cd in sediments from Khor Katheib, Red Sea coast, Yemen.

Multivariate statistical analysis results

Correlation analysis

The interrelationships matrices between the studied elements in the sediments of the Creek Khor Katheib were calculated using SPSS Statistical Program and the results are shown in (Table 4). The gathering metals agents Fe and Mn are well associated will each other (r = 0.73) and each shows positive association with Al ($r \leq 0.38$) referring to be all originated from same source. Both Cu and Zn are well associated with each other (r = 0.53). Each shows good association with the essential components of the sediments Al and Mn rather than Fe. The association with Mn actually is better than with Al. Zinc is associated with Fe while Cu shows no significant relationship with Fe. The well association with Mn refers to its possibilities to gather metal ions positively changed, as Mn oxy-hydrates are negatively changed, which is opposite to the Fe-oxy-hydroxides (Zabetoglou et al., 2002).

The correlation matrix of organic matter that act as a metal carrier showed a poor positive correlation for most of the metals, except for Cr and Cd which showed a weak positive correlation ($r=0{\cdot}26, P<0{\cdot}05$). In the case of carbonate content there was a negative correlation with heavy metals reflecting the terrestrial source of these metals.

Principal component analysis (PCA)

The factor analysis was applied to obtain more reliable information about the relationships among the variables (Facchinelli et al. 2001; Loska and Wiechula 2003; Bartolomeo et al. 2004; Glasby et al.2004; Ghrefat and Yusuf 2006; Varol 2011; Attia and Ghrefat 2013).

In the present study, PCA (Varimax with Kaiser Normalization) identified four components in sediments of the Creek Khor Katheib, which are listed in Table 5. Four significant principal components, whose eigenvalues are higher than 1 accounting for 70.45 % of the cumulative variance, were distinguished for the analysed data.

PC-1 explains 31.51% of the total variance and is mainly characterized by high levels of Al, Fe, Mn and Zn This element association is considered to represent the lithology of the study area, and a natural input; i.e., they are derived from terrigenous detritus material transported by surface runoff (Krishna et al. 2011; Likuku et al. 2013 and Okbah et al., 2014).

PC-2 accounts for 15.73 % of the total variance, has a positive factor loading for Ni and Cd, which suggests similar sources of anthropogenic origins.

PC-3 accounting for 12.09 % of the total variance and shows a positive loading of Co, Cr. Ni. Significant positive correlations are observed between these elements (Table 4), which suggests a lithogenic control over the distribution of Cr, Co, and Ni. For instance Facchinelli et al. (2001), Gallego et al. (2002), Rodr´ıguez Mart´ın et al. (2006), and Dou et al (2013) also showed that Ni, Cr, and Co were grouped in the common factor, and a lithologic control for their distributions. PC-4 accounting for 11.12% of total variance was correlated with Cu.

Ni emerges at both PC2 and PC3, indicating that some of the Ni originated from source rocks and some from pollution sources. Generally, the results of correlation analysis and factor analysis coincide with each other. Their results demonstrate the lack of clustering between Cu, Cd, Zn, Co, Ni, Cr, Mn, and the sediment components including organic matter and carbonate content confirm the complicated behaviour of these pollutants, which can be influenced by many factors.

Table 4 Pearson's matrix interrelationships between the studied parameters in the sediments of Khor Katheib, Red Sea, Yemen.

	Mud	**$CaCO_3$**	**TOC**	**Cu**	**Cd**	**Zn**	**Co**	**Ni**	**Cr**	**Mn**	**Fe**	**Al**
Mud	1											
$CaCO_3$	0.02	1										
TOC	-0.01	0.00	1									
Cu	0.10	0.00	-0.20	1								
Cd	0.26	-0.04	0.10	0.46	1							
Zn	0.23	-0.21	-0.10	0.53	0.35	1						
Co	0.00	-0.21	0.06	0.30	-0.04	0.16	1					
Ni	0.47	-0.10	0.19	0.03	0.20	0.37	0.42	1				
Cr	0.23	-0.14	0.26	-0.15	0.03	0.11	0.41	0.40	1			
Mn	0.33	-0.33	0.00	0.42	0.37	0.72	-0.09	0.28	0.10	1		
Fe	0.02	-0.47	0.16	0.17	0.28	0.58	-0.05	0.10	0.21	0.73	1	
Al	0.19	-0.34	0.09	0.41	0.12	0.68	0.28	0.20	0.24	0.36	0.38	1

Table 5 Total variance explained and rotated component matrices for heavy

Parameter	Component 1	Component 2	Component 3	Component 4
Al	**0.61**	0.11	0.43	0.22
Fe	**0.90**	0.08	-0.10	-0.17
Mn	**0.78**	0.45	-0.11	0.07
Cr	0.14	0.14	**0.63**	-0.47
Ni	0.07	0.57	**0.58**	-0.22
Co	0.02	-0.08	**0.90**	0.16
Zn	**0.71**	0.40	0.20	0.34
Cd	0.26	**0.64**	-0.14	0.14
Cu	0.32	0.33	0.14	**0.75**
Eigen value	3.78	1.89	1.45	1.33
Total of variance (%)	31.51	15.73	12.09	11.12
Cumulative loading (%)	31.51	47.24	59.33	70.45

metals

Conclusion

The Multivariate statistical analysis methods including correlation matrix analysis and PCA were successfully applied for the assessment of pollution by heavy metals in sediments from Creek Khor Katheib along the Red Sea coast of Yemen. The results of the correlation analysis of the data shows negative correlations among Mn, Fe, Al, Cu, Zn, Cd, Ni, and $CaCO_3$ indicating that these metals have complicated geochemical behaviors. The use of statistical factor analysis also confirms these results.

The Sediment pollution of the area (i.e., Creek Khor Katheib) was assessed using contamination and enrichment factors. The results of contamination factor reveal that the sediments are not contaminated with Cu, Zn, Co, Ni, Cr and Mn, moderately contaminated with Cd. Some of elevated concentration of Cd and Cr are probably resulting from anthropogenic activities in the area.

The calculation of enrichment factor showed that Cd had the highest EF values among the other studied metals and it has a moderately severe enrichment, Cr had moderate enrichment. Mn and Ni had minor enrichment. Despite the human activities in the commercial Al-Hodeidah Harbor, its sediments showed no exceptional high metal contamination. This could be due to the repeatedly dredging processes for accumulated sediments (plus possibly that the spoiled ones are disposed at a place outside the study area). Therefore, the disposed sediments need monitoring to see if it represents a potential risk.

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تقييم تلوث العناصر الثقيلة في الرواسب السطحية لساحل مدينة الحديدة وخور الكتيب، البحر الاحمر، اليمن

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المستخلص :-

خور الكتيب (Katheib) يشمل الميناء التجاري للحديدة، ويعد الميناء الرئيسي لليمن على البحر الأحمر. ويستقبل مياه الصرف الصحي غير المعالجة المنزلية والصناعية من مدينة الحديدة. وكان الغرض من هذه الدراسة الحصول على تقييم أولي لمستوى وتوزيع لتسعة معادن ثقيلة (النحاس ، الكادميوم ،الزنك، الكوبلت، النيكل، الكروم، المنغنيز، الحديد و الالمنيوم) في الرواسب السطحية التي تم جمعها من 25 محطة داخل وخارج الخور. وقد لوحظ الاختلاف المكاني كبير في التراكيز لجميع المعادن. وتمت معالجة البيانات باستخدام تحليل الارتباط وتحليل العوامل. و أظهر تحليل الارتباط للنتائج ارتباطات سلبية بين المعادن الثقيلة وكربونات الكالسيوم $CaCO_3$، ولم توجد علاقة بين الكربون العضوي الكلي والمعادن الثقيلة مشيرا إلى أن هذه المعادن قد يكون لها سلوك بيولوجي وكيميائي معقد.

تم تقييم حالة التلوث باستخدام عامل الإثراء، وعامل التلوث. واستنادا إلى عامل الإثراء، أظهرت النتائج أن رواسب خور الكتيب (Katheib) لديها إثراء متوسط الشدة مع الكادميوم، وإثراء معتدل مع الكروم والكوبلت ، في حين كان المنجنيز والنيكل لهما إثراء طفيف.