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Assessment of some Heavy Metals Contamination in Mediterranean Sea Sediments at Jerpoly Coast Libya Using Pollution Indices

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ABSTRACT

The main objective of this study was to evaluate the levels of some trace metals.The state of heavy metal pollution and the mobility of Fe, Mn, Cu, Zn,pb and Cd were studied in Jerpoly regions in the Mediterranean Sea, five surface sediment samples were collected from each region. Methods used to characterize heavy metals in solid phase of the sediments include physical and chemical extraction. using atomic absorption spectrophotometer and to compare metal concentration. The distribution pattern of heavy metals in the sediment follows the sequence: Fe>Pb>Mn> Zn> Cu> Cd revealed that the sediments of Study area have higher concentrations of fe, Zn,Pb while, , Mn,Cu and Cd have lower concentrations than those of other regions. In general, the results revealed very high contamination factor.The results of contamination factors (Table 5) indicated that in thas study Zn, Cu, Mn possess the lowest CFs reflecting lowest contaminated sediments (CF<1). Both Pb and Cd showed low to moderate contamination level of contamination. On the other hand, Sediments of are highly contaminated (CF>6) with Fe. The major role of carbonate as metal carrier is reflected by the positive correlations between Cu, fe with sand . On the other hand, the positive correlation of OM with Cu suggested that OM has an important role in the binding of these elements. The negative correlation of sand with Pb (r = -0.174) indicate that these elements can be easily released by ion exchange processes due to the electrostatic interaction of trace metals as they are weakly bound and is bioavailable to the liquid phase. The study point out that although there were slight variations in the results of the three indices,

الملخص العربي

الهدف الرئيسي لهذه الدراسة كان لتقييم مدى تلوث شاطيء البحر الأبيض المتوسط ببعض المعادن الثقيلة مثل (الحديد والمنغنيزوالنحاس والزنك والرصاص والكادميوم). حيث تم تجميع خمسة عينات من محطات الدراسة ونقلت الى المعمل وأجريت عليهم التحاليل الفيزيائية والكيميائية وتم قياس العينات فى جهاز الأمتصاص الذرى لمعرفة مستويات العناصر الثقيلة المذكورة سابقا .ومن خلال الفحص تبين زيادة تركيز

عنصر الحديد والزنك والرصاص وقلة تركيز المنجنيز والنحاس والكادميوم . وتم اجرى تحليل احصائى للنتايج المتحصل عليها لكل من CF، EF+Igeo، RACو PLI . تبين ان الزنك والنحاس والمنغنيز بالعينات التربة منخضة التلوث)وعند حساب عامل التلوث والرصاص والكادميوم متوسطة التلوث وزيادة تركيز الحديد دلالة عن تلوثه للتربة .(CF<1) (CF>6) .وهناك علاقة أيجابية بين النحاس والحديد مع الرمل والمادة العضوية مع النحاس والعلاقة السالبة للرمل مع الرصاص مؤشر ان هذه العناصر تتبدل وتتفاعل استاتيكى مع بعضها البعض وتتغيير بسبب التلوث. وأستخدمت برامج أخرى لحساب التلوث بالمعادن الثقيلة مثل Multivariate analysis (PCA/CA) and correlation matrix

INTRODUCTION

Rapid industrialization and uncontrolled urbanization around many cities and coastal area have brought alarming level of pollutions to the aquatic environments because of their anthropogenic inputs. Heavy metals are considered as serious inorganic pollutants because of their toxic effects on life in aquatic system, having a high enrichment factor and slow removal rate (Alloway and Ayres, 1997). As a consequence, it is assumed that the equilibrium balance between the metals in sediments and surface water is disrupted that might be a reason to increase water contamination (Rahman et al., 2013). Sediment as a compartment is more conservative than water, as it accumulates historical data on processes within water bodies and the effect of anthropogenic factors on these processes. For these reasons, sediment quality parameters have been used as environmental indicators and their ability to trace and monitor contamination sources is largely recognized (Vallejuelo et al., 2010).

Libyan coastline is considered as the longest African coastline on the Mediterranean Sea. In addition to its length, it is in the same time very rich in its natural resources in the field of fish wealth or energy and mineral raw resources. A study of the distribution, enrichment and accumulation of metals in the Libyan Mediterranean coastal sediments is important to the assessment of the possible influence of anthropogenic activities on sea water. However, the recent available information lacked sufficient and adequate accuracy and details (Hamouda and Wilson, 1989 and Hasan and Islam, 2010). An assessment is thus necessary to appraise the concentration of metals in Mediterranean Sea sediments at Jerpoly coast Libya so as to understand the present condition of the coastline and to compile the baseline data for future monitoring.

The objectives of this study were: to examine the spatial variations of the heavy metal concentration in the surface sediment of the Libyan Mediterranean coast,in gerpoly) to define the natural and/or anthropogenic sources of these metals using multivariate statistical technique, iii) to explore the degree of heavy metal contamination using different pollution indices to assess environmental risks of heavy metals in the study area by comparison with sediment quality guidelines (SQGs).

MATERIALS AND METHODS

Study area

Libya has a unique position in the middle of North Africa. It is a junction between its eastern and western seashores. It embraces the southern coasts of the Mediterranean Sea, forming the heart for this great sea. The Libyan Coast extends to about 1900Km between Beir Al Ramla on the Egyptian borders to the east and Ras Gidier on the Tunisian borders to the west (Fig. 1). This distance is equal to around 37% of the total Arab Coasts on the Mediterranean Sea. Mediterranean Sea sediments at Jerpoly coast Libya extend 60 km eastern Tripoli on coasts of the Mediterranean Sea . In addition to its length, it is in the same time very rich in its natural resources be they in the field of fish wealth or energy and mineral raw resources (El Haddad, 2012). An assessment is thus necessary to appraise the concentration of some metals in Jerpoly coast so as to understand the present condition of Study area and to compile the baseline data for future monitoring.

Five surface sediment samples (5) (0-5 cm) were collected from Mediterranean Sea sediments at Jerpoly coast Libya,The sampling stations were chosen carefully to provide good area coveragein in march 2018. After sampling, the sediments were stored in a plastic vessel and frozen at -20ºC and samples were defrosted at room temperature, dried at 40ºC to constant weight, and ground and homogenized in a mortar to a fine powder and converted to analysis in laboratory at National Institute of Oceanography and Fisheries, Alexandria

"Fig. 1,"**:-Show Station study**

Collection an preparation of Samples :

The samples were digested in an open system with a mixture of concentrated HNO_3/ $HClO_4$/ HF (3:2:1) according to Oregioni and Aston (1984). The determination of the some metals in the sediment samples were performed with a SHIMADZU AA6800 atomic absorption spectrophotometer equipped with a deuterium background corrector. An atomizer with an air/acetylene burner was used for determining all the investigated elements. All instrumental settings were those recommended in the manufacturer's manual book. Suitable internal chemical standards (Merck Chemicals, Germany) were used to calibrate the instrument.

To remove any contamination, all glassware and plastic vials were washed with 10% nitric acid solution and rinsed thoroughly with Milli-Q water and dried. All reagents were Merck analytical grade or super pure quality. In order to check for the quality of the method applied for the
analysis of heavy metals, the accuracy of the analytical method was estimated by analyzing sediment Standard Reference Material (IAEA-405): estuarine sediment, International Atomic Energy Agency, Vienna, Austria). The recovery of the selected metals ranged from 90 to 104% and the measurements of precision was under 10% RSD.

Statistical analysis :

SPSS 16 software were used in the present study for the calculation of Pearson Correlation coefficient matrix, Factor analysis and constructing a dendrogram for the Hierarchical Cluster analysis.

RESULTS AND DISCUSSION

Characteristics of sediments

Table 1 show that the studied sediments contain high sand fractions (95.7 %) according to result in table (1). This indicates that the sand is the dominant component in the collected samples. Additionally, the sum of silt and clay fractions fluctuates between 4.3 and 8.7 %. Organic matter (OM) content was low, ranging from 0.86 % at St4 to 1.48% at St5.

The results showed differences in the distribution of $CaCO_3$%. The Station 1,3,4,5 was characterized by relatively medium $CaCO_3$ while the Station 2 was characterized by high percentage of $CaCO_3$.

These results are in agreement with the results of Hamouda and Wilson (1989).

Heavy metals distribution :

The range and mean some heavy metal concentrations in surface sediments of Study area coast are summarized in Table 2. The mean value of Some heavy metals concentrations in the study area follows a descending order as: Fe>Pb>Mn> Zn> Cu> Cd. A comparative study between metal concentrations in sediments of with those of other regions along the Mediterranean .

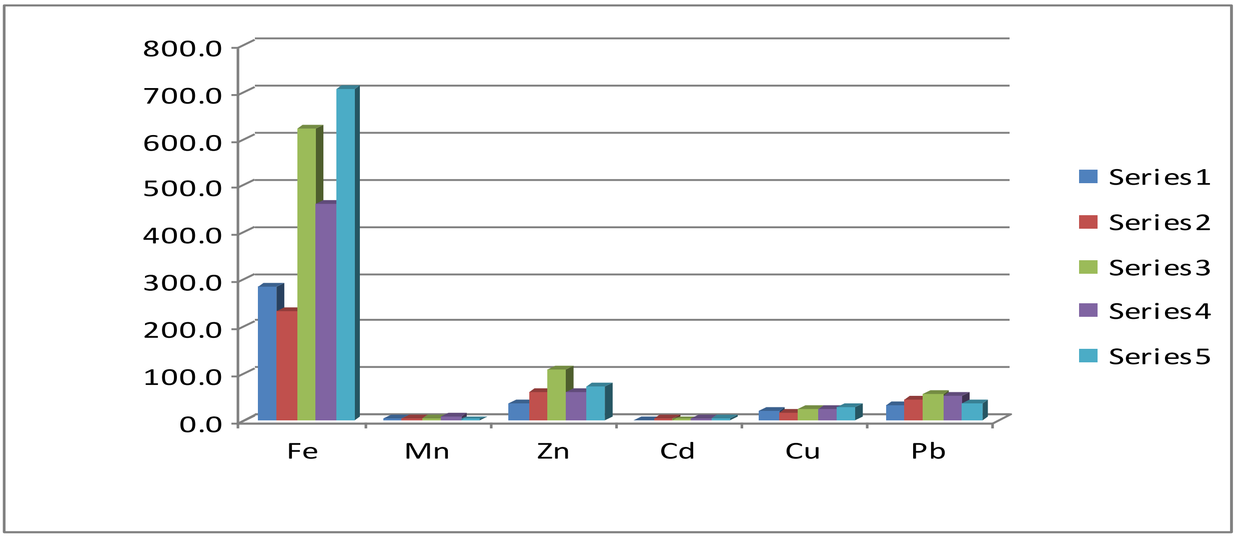
Table 3 revealed to have higher concentrations of Fe, Zn,Pb while, , Mn,Cu and Cd have lower concentrations than those of other regions , Concentrations of Pb were lower than those measured in Sardina, Italyj , , Coast of Safax, Egyption Med. Coastb and higher than those reported The eastern Libyan coasta .(Hasan and Islam, 2010, Okbah et al., 2014, Sabhi et al., 2000, Caredda et al., 1999).

Table 1: the Physicochemical characteristics of Mediterranean Sea sediments at Jerpoly coast Libya.

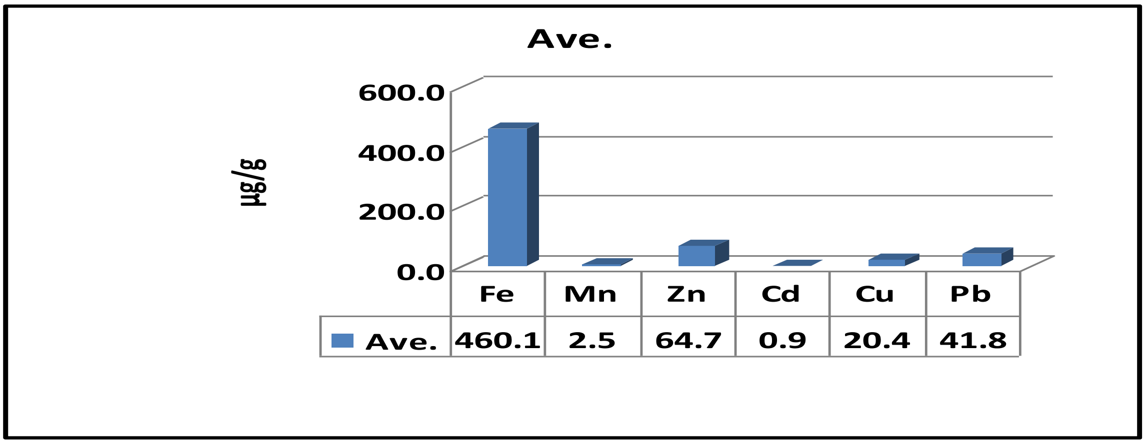
Sites	O.M. %	$CaCO_3$ %	Grain size analysis (%)	
			Sand	Silt+Clay
St1	1.23	44.38	93.6	6.4
St2	1.38	74.85	91.3	8.7
St3	1.23	53.7	95.7	4.3
St4	0.86	55.68	92.4	7.6
St5	1.48	47.67	95.6	4.4

Table 2:- the Distribution of Some heavy metal concentration (µg/g) in Mediterranean Sea sediments at Jerpoly coast Libya.

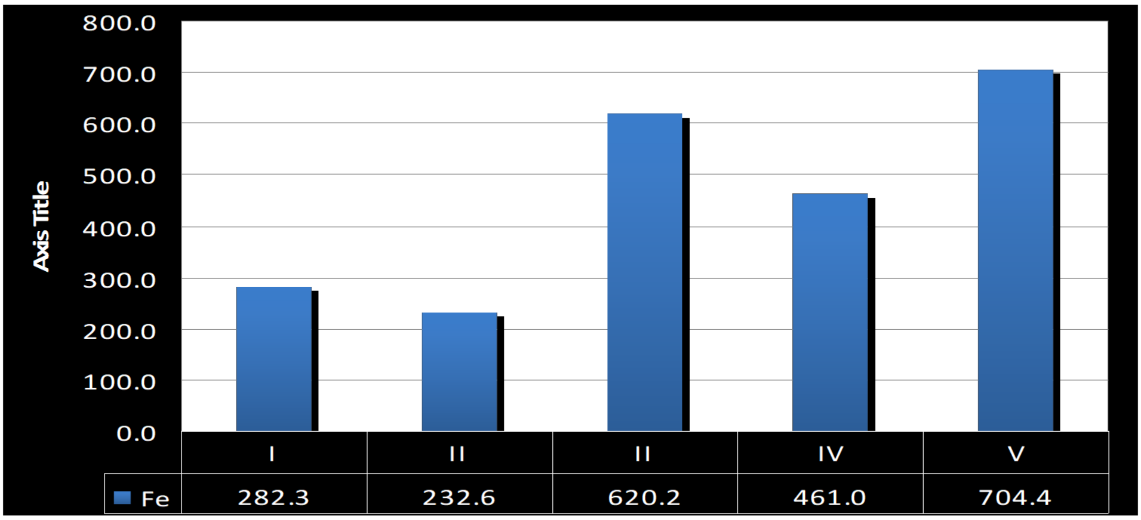
Stations	Fe	Mn	Zn	Cd	Cu	Pb
I	282.3	3.2	33.3	0.0	18.5	30.0
II	232.6	0.6	57.0	1.1	14.1	41.2
II	620.2	0.9	105.7	0.5	21.3	53.6
IV	461.0	7.3	58.4	1.2	22.5	49.4
V	704.4	0.6	69.1	1.5	25.5	34.8
Ave.	460.1	2.5	64.7	0.9	20.4	41.8



"Fig. 2,"**: the Distribution of some heavy metal concentration (µg/g) for Mediterranean Sea sediments at Jerpoly coast Libya**



"Fig. 3,":- **the Distribution average of some heavy metal in Mediterranean Sea sediments at Jerpoly coast Libya**



"Fig. 4,": the **Distribution concentration Fe in Mediterranean Sea sediments at Jerpoly coast Libya.**

Table 3: Some Heavy metal concentrations Mediterranean Sea sediments at Jerpoly coast Libya and other selected regions along the Mediterranean Sea (µg/g) .

Location	Fe	Zn	Pb	Cd	Mn	Cu
This study	282.3	33.3	30.0	0.0	3.2	18.5
Sardina, Italyj	NA	198-3239	74-772	0.21-13	NA	2.77-51
The eastern Libyan coasta	NA	2.3-27.3	NA	ND-1.73	37-76.7	8.7-42
Egyptian Med. Coastb	243.5-38045.1	2.05-62	3.34-53	0.04-0.47	17.3-1086	0.46-26
Moroccan Med. Coastl	NA	9.2-37	0.17-0.25	0.03-0.25	NA	1.2-6
Libya Coast	141.8-1056.8	11.6-30.5	18.9-56.9	5-10.5	14.3-49.4	9.1-22.7

NA; Not available

(Hasan and Islam, 2010, bOkbah et al., 2014, Sabhi et al., 2000, Caredda et al., 1999).

However, Cu showed values higher than those measured in the Mediterranean coast of Moroccan Med. Coastl , Finally, Cd showed lower concentrations than those reported in The Libyan Coast .

Assessment of sediment contamination

A number of methods have been put forward for quantifying the degree of metal enrichment in sediments. Various authors **(Tomlinson et al., 1980; Muller, 1969; Hakanson, 1980**) have proposed pollution impact ranges to convert the calculated numerical results into broad descriptive bands of pollution ranging from low to high intensity **(Abrahim and Parker, 2008**). In this study, three different indices were used to assessment the degree of some heavy metal contamination in **Mediterranean Sea sediments at Jerpoly coast Libya.**

The geo accumulation index (*Igeo*):

The geo accumulation index (Igeo) originally introduced by **Müller (1969),** determine and define metal contamination in sediments. The geoaccumulation index (Igeo) is defined by the following "equation (1),"

$I^{geo} = Log2\ (C_n / 1.5 x B_n)$

where Cn is the measured concentration of the examined metal (n) in the sediment and Bn is the geochemical background concentration of the metal (n). The factor 1.5 is the background matrix correction factor due to lithological variability. The background values of the metals here are the same as those recorded in the enrichment factor calculation. Similar to metal enrichment factor, geo accumulation index can be used as a reference to estimate the extent of metal pollution **(Zhang et al., 2009).** the geo accumulation index from Class 0 (Igeo=0) to Class (Igeo>5).

The Igeo is associated with a qualitative scale of pollution intensity, samples may be classified as unpolluted (Igeo≤0), unpolluted to moderately polluted (0≤Igeo≤1), moderately polluted (1≤Igeo≤2), moderate to strongly polluted (2≤Igeo≤3), strongly polluted (3≤Igeo≤4), strongly to extremely polluted (4≤Igeo≤5), and extremely polluted (Igeo≥5). The geo accumulation indexes calculated for the sediments are summarized in Table 4 in the form of a Box Whiskers plot. According to Muller's scale, the mean geo-accumulation indexes of Cu (6), Fe (9), Mn (8),Pb(6.8), and Zn (8) are higers than zero (Igeo>0),and Cd less than zero (Igeo<0), suggesting that Study area has been polluted overall by these metals. The Igeo class of Pb was higer from 1 in the sediments of all sites which usually had moderately polluted" class. Among the studied elements, Cd had the less mean Igeo values (-1), suggesting the sediments has unpolluted with this metal.

(**Table 4).**

Table 4:- Metal contamination factors (Igeo) in Mediterranean Sea sediments at Jerpoly coast

Igeo	Cu	Fe	Zn	Cd	Mn	Pb
I	6	9	8	0	8	6.8
II	6	7	8	0	6	7.1
III	6	8	9	-1	6	7.4
IV	7	8	8	0	8	7.3
V	7	8	8	0	6	7.0

Contamination factors (CF) and Degree of contamination (Dc)

The CF is the ratio obtained by dividing the concentration of each metal in the sediment by baseline of background value. "equation (2),"

CF =M x/Mb

CF value were interpreted as suggested by **Hakanson (1980)**, where: CF < 1 indicates low contamination; 1<CF<3 is moderate contamination; 3<CF<6 is considerable contamination; and CF>6 is very high contamination. The results of contamination factors (Table 5) indicated that in this study Zn, Cu, Mn and possess the lowest CFs reflecting lowest contaminated sediments (CF<1). Both Pb and Cd showed low to moderate contamination level of contamination. On the other hand, Sediments of are highly contaminated (CF>6) with Fe. (**Table 5).**

Table 5: Metal contamination factors (CF) and pollution load indices (PLI) for Mediterranean Sea sediments at Jerpoly coast Libya.

CF	Cu	Fe	Zn	Cd	Mn	Pb
I	0.41	6.05	**0.35**	0.00	0.00	0.79
II	0.31	4.98	**0.60**	1.20	0.00	1.08
III	0.47	13.28	**1.11**	0.56	0.00	1.41
IV	0.50	9.87	**0.62**	1.25	0.01	1.30
V	0.57	15.08	**0.73**	1.63	0.00	0.92

Pollution Load index (PLI).

For the entire sampling sites, PLI has been determined as the nth root of the product of the"equation (3)," CF: PLI = (CF1 x CF2 x CF3 x ….x CFn) 1/n This empirical index provides a simple, comparative means for assessing the level of heavy metal pollution. When PLI >1, it means that a pollution exists; other wise, if PLI<1, there is no metal pollution **(Tomlinson et al., 1980).** The pollution load index (PLI) ranged from 0.0 to 0.04. According to the mean PLI value (0.04), **Mediterranean Sea sediments at Jerpoly coast Libya.** was unpolluted.(Table 6).

Table 6:- Metal contamination factors (MPI, PLI) IN study area.

MPI	PLI
0.0	0.000
13.3	0.001
18.0	0.005
25.2	0.040
18.4	0.006

Table 7:- Metal contamination factors (EF) IN study area.

EF	Cu	Fe	Zn	Cd	Mn	Pb
I	0.1	6.0	0.1	0.0	0.0	0.1
II	0.1	5.0	0.1	0.2	0.0	0.2
III	0.0	13.3	0.1	0.0	0.0	0.1
IV	0.1	9.9	0.1	0.1	0.0	0.1
V	0.0	15.1	0.0	0.1	0.0	0.1

3.5. Multivariate Statistical analysis

Multivariate analysis (Principal component analysis; PCA and Cluster analysis; CA) has been proved to be an effective tool for providing suggestive information regarding heavy metal sources and pathways **(Hu et al., 2013).**

Correlation matrix :

In order to establish relationships among metals and determine the common source of metals in Study area a correlation matrix was calculated for some heavy metals in the sediments. According to the values of Pearson correlation coefficients (Table 8), a significant positive correlation existed among the metals studied. In this study, fe was significantly correlated with Cu, Zn and sand . It indicated strong association of these metals in the sediments where they might be share. The significantly positive correlation of Cu (r=0.8, p<0.1) and Pb (r=0.7, p< 0.1) suggested that these metals were redistributed in the sediments by the same physico-chemical processes or had a similar source **(Bai et al., 2011).** The major role of carbonate as metal carrier is reflected by the positive correlations between Cu, fe with sand . On the other hand, the positive correlation of OM with Cu suggested that OM has an important role in the binding of these elements. The negative correlation of sand with Pb (r = -0.174) indicate that these elements can be easily released by ion exchange processes due to the electrostatic interaction of trace metals as they are weakly bound and is bioavailable to the liquid phase **(Fostner and Wittman, 1979; Morillo et al., 2004).**

Table 8 . Correlations Matrix of some heavy metal concentrations grain content in Mediterranean Sea sediments at Jerpoly coast Libya (N = 14)

	Fe	Mn	Zn	Cd	Cu	Pb	O. M	CaCO3	Sand	Silt+Clay
Fe	1									
Mn	-0.18	1								
Zn	0.69	-0.03	1							
Cd	0.41	-0.05	0.19	1						
Cu	0.89	0.19	0.34	0.35	1					
Pb	0.30	0.21	0.75	0.15	0.10	1				
O.M	0.13	-0.93	0.09	0.16	-0.11	-0.50	1			
Caco3	-0.47	-0.18	0.07	0.35	-0.69	0.33	0.07	1		
Sand	0.84	-0.38	0.58	-0.07	0.71	0.03	0.32	-0.69	1	
Silt+Clay	-0.84	0.38	-0.58	0.07	-0.71	-0.03	-0.32	0.69	-1.00	1

Table 9. Risk assessment Code (RAC)

Risk	Criteria (%)	Present study %					
		Fe	Mn	Cu	Zn	Cd	Pb
No risk	< 1	10.11-30.6	4.7-57.9	13.8-25	10.3-32.7	12.0-35.2	14.3-25.6
Low	1 - 10						
Medium	11 - 30						
High	31 - 50						
Very high	> 50						

Criteria(%),(Perin et al.,1985; Singh et al., 2005).

Principle component analysis

Principle component analysis has been applied to determine the degree of pollution by metals from lithogenic and anthropogenic sources. The results of PCA for heavy metal contents are listed in Four principal components were extracted, which covered 88.417% of the total variance. Apparently the result of PCA corresponds well with the correlation coefficients **(Hu et al., 2013).**

Cluster analysis :-

Cluster analysis was performed on the same data as PCA to understand the similarities among them. Fig. 6 depicts a dendrogram with single linkage Euclidean and correlation coefficient distance. The cluster analysis results indicate four clusters: (1) Cd- Cu-Ni-Zn-Mn-CaCO3-OM; (2) Sand (3) Fe (4) –Pb in terms of similarities.

This indicates that Cd, Cu, Ni, Zn and Mn appear to have originated mainly from same sources. Cluster 2 further shows that sand has no association with these elements. Cluster 3 further shows that Fe has no association with these elements. In addition, Cluster 4 shows that Pb seem to drive partly from the same sources. This is consistent with our PCA results.

CONCLUSION

This work provides the first comprehensive analysis of some metal status in surface **Mediterranean Sea sediments at Jerpoly coast Libya**. In this study, three different indices have been employed for the evaluation of some heavy metal contamination status.

The results showed that total heavy metal concentrations in the sediments samples followed the order: Fe>Pb>Mn> Zn> Cu> Cd. According to Muller's scale, the mean geo-accumulation indexes of Cu, Fe, Mn, and Zn are less than zero (Igeo<0), suggesting that the Libyan.

Mediterranean Sea has not been polluted overall by these metals. On the other hand, the sediment has been strongly to very strongly polluted with pb (7≤Igeo≤6), Cu (7≤Igeo≤6), Mn (8≤Igeo≤6), fe (9≤Igeo≤7), This was supported by CF and Ef (>6) for,fe. According to the mean MPI value (25.2), present study in the Libyan Mediterranean coast was polluted. The heavy metal concentrations in assessed sediment samples were compared with (TEL-PEL) values. The results indicate that fe and Zn would rarely be expected to cause adverse effects on biota. 79% percent of sediments would be expected to occasionally be associated with the toxic adverse effects on aquatic organisms because of Pb. While, Fe exceeded the PEL value at 100% of the sediment samples.

Multivariate analysis (PCA/CA) and correlation matrix were used in this study. A significant positive correlation is observed among , Cu, Fe, and Zn, indicating that these metals were derived from similar sources. Iron has association with other elements, suggesting that Fe has another different sources or pathways. The study point out that although there were slight variations in the results of the three indices, the combination of the three indices gave us a comprehensive understanding of heavy metal risks in the surface **Mediterranean Sea sediments at Jerpoly coast Libya.**

RECOMMENDATION

A continuous monitoring program is highly recommended to follow up if more improvement in the environmental condition of the sea water with time is still going.

Further modeling studies must be carried out in the future to predict improvement or detoriation of the environmental condition of the sea water.

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