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Assessment of Heavy Metals Pollution In Euphrates River Water, Amiriyah Fallujah, Iraq.

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Abstract
The concentrations of heavy metals (Cd, Cr, Fe, Ni and Pb) in water samples for the Euphrates River in Amiriyah Fallujah, Iraq were evaluated to assess the pollution level. Ten sites were selected along the study area and sampled during December 2013 to March 2014. The decreasing trend of metals were observed in water as Pb > Fe > Cr > Cd > Ni. The concentration of the studied metals was higher than the recommended guidelines for drinking and aquatic life, indicated that the water is not safe for drinking and aquatic life uses. There were significant differences in the concentrations of heavy metal during the sampling period. Heavy pollution index (HPI) results showed that the water was seriously polluted with Cd, Cr and Pb. According to metal index (MI) results, the water is seriously threatened with metal pollution and unsafe for drinking, irrigation and aquatic life uses. Principal components analysis (PCA) suggests that the Cd, Cr and Pb are derived from anthropogenic sources.
Keywords: River pollution; Pollution index, Metal Index; Euphrates; Iraq

1. Introduction

Recently, metal contamination in the aquatic bodies has of great concern because of its toxicity for environment and human beings, non-degradable, persistence and ability to be accumulated in food chains (Sin *et al*. 2001; Armitage *et al*. 2007; Yuan *et al*. 2011; Wang *et al*. 2013; Sun *et al*. 2015). Some of metals such as Cu, Fe, Mn, Ni, and Zn are essential as micronutrients for the life process for animals and plants while many other metals such as Cd, Cr, Pb and Co have no known physiological consequences (Kar *et al*. 2008; Aktar *et al*. 2010). Sources of heavy metals in water are natural (weathering of rocks and soils) and anthropogenic (mining, industries, wastewater irrigation and agriculture activities (Ahmet *et al*. 2006; Chanpiwat *et al*. 2010; Muhammad *et al*. 2010). Large quantities of heavy metals have been released into rivers world due to global rapid population growth and anthropogenic activities (Srebotnjak *et al*. 2012; Su *et al*. 2013; Islam *et al*. 2014; Islam *et al*. 2015). When the heavy metals released into the river system by natural or anthropogenic sources, they are distributed during their transport between dissolved phase and bed sediments (Varol & Şen 2012). The analysis of dissolved metals in water is a useful tool for assessing the state of pollution in a particular ecosystem, reflecting its specific degree of contamination (Haloi &d Sarma 2012; Li *et al*. 2012; Su *et al*. 2013; Alves *et al*. 2014). The increasing military activities in Iraq since 1980 resulted in establishing many military factories along the Euphrates and Tigris rivers. These factories led to an increase in environmental problems including water contamination, air pollution and ecosystem degradation. In the study area, there is Al-Ikha General Company. The company is a large industrial complex with five factories. Therefore, the company produces a wide spectrum of products (e.g. Water purification units and its filters, Electricity power transmission towers production line, optics, cooking gas cylinders ……. etc.). Studies on heavy metals concentrations, distribution and contamination in water, sediments , aquatic fauna and flora of Euphrates river in Iraq have been conducted by several investigators (Kassim et al. 1997; Al-Khafaji 2001; Fahad 2006; Hassan *et al*. 2010 ; Al-Khafaji *et al*. 2011; Salman & Hussein 2012; Salah et al. 2012 ; Abdullah 2013; Hussein *et al*. 2014; Habeeb *et al*. 2015). The objectives of this study are to assess the concentrations of heavy metals and the contamination status of the Euphrates river water in Amiriyah Fallujah, Iraq. This study represent the first attempt to investigate the distribution of heavy metals and pollution level in Euphrates river water in the study area.

2. Materials and methods

2.1 Study area

The Euphrates River is one of the most important rivers in the world. Along with the Tigris River, it provided much of the water that supported the development of ancient Mesopotamian culture. Euphrates River rise in the highlands of Turkey and it is formed the Karasu and Murat tributary rivers. Euphrates enters Iraq at AlQaim city. During its passage through Iraq, the river crosses more than 1000 km. The water resources in Iraq are concentrated to the Euphrates and Tigris Rivers. Amiriyah Fallujah, is a city in the Al Anbar governorate, about 30 km south of Fallujah. Amiriyah Fallujah located at 33.16° N and 43.86° E, Figure 1. The total population of the city is 110 thousand people.

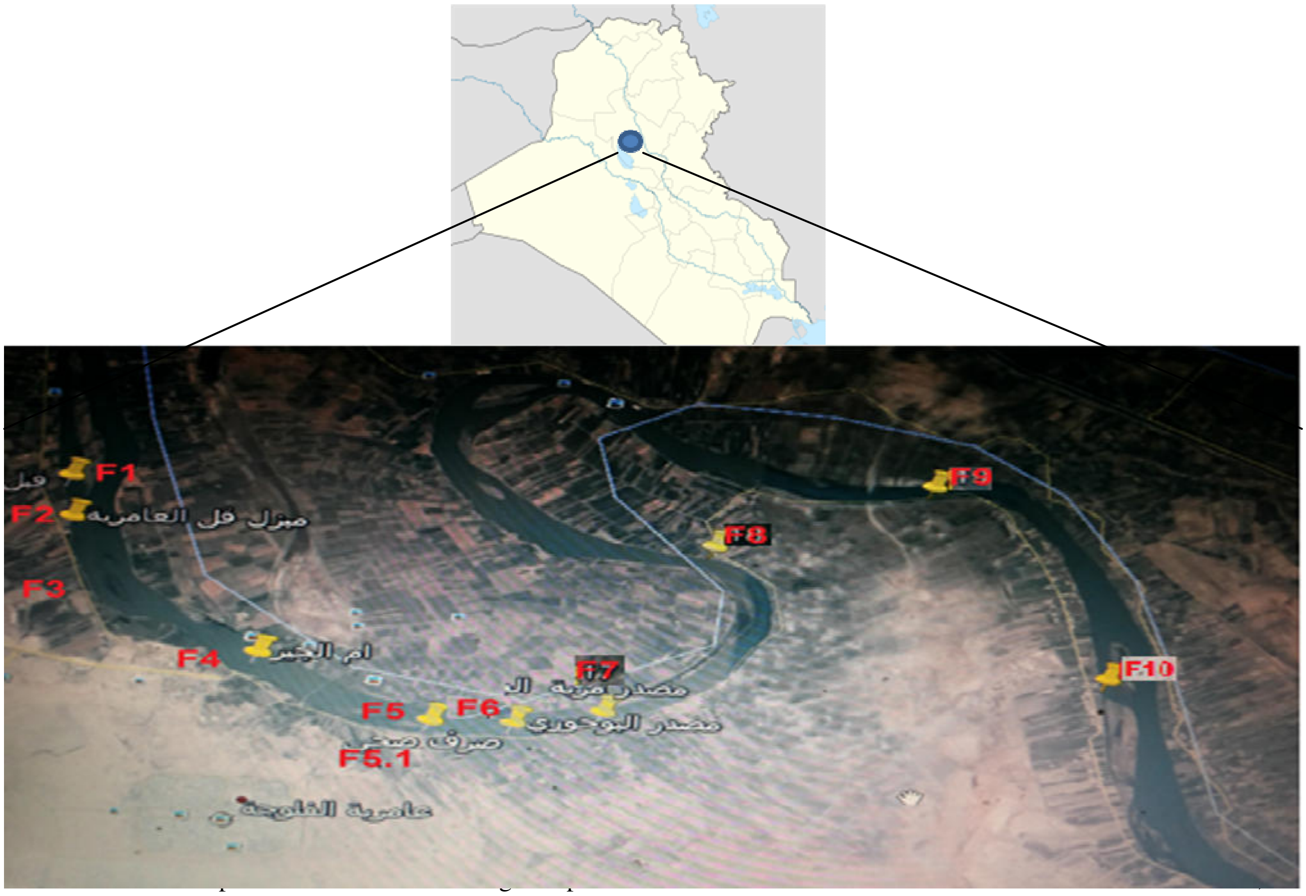

Figure 1and Table 1. The water samples were collected from a depth of 30 cm beneath the river surface. They have been done once a month and the sample were placed in clean poly-ethylene bottles and stored in ice containers. Concentrations of Cd, Cr, Fe, Ni and Pb were determined in water using flame atomic absorption spectrometry (Philips-England) according to the extraction procedures of (APHA 2005).

Table 1 Geographical coordinates of the sampling stations.

Stations	Latitude (N°)	Longitude (E°)
F1	33°12'3.63"	43°50'49.82"
F2	33°11'40.67"	43°50'54.18"
F3	33°11'16.44"	43°51'4.54"
F4	33°10'46.37"	43°51'51.46"
F5	33°10'25.95"	43°52'43.02"
F6	33°10'26.60"	43°53'8.08"
F7	33°10'28.02"	43°53'19.73"
F8	33°11'41.80"	43°54'1.38"
F9	33°12'12.44"	43°54'42.09"
F10	33°10'57.03"	43°56'0.74"

2.3 Metal quality Index

To assess the quality water of Euphrates river in terms of heavy metals, we employed two indices, the Heavy metal pollution index (HPI) proposed by Caerio *et al.*(2005) and the Metal index (MI) developed by Tamasi & Cini (2004). Heavy metal pollution index (HPI) is calculated using the following equation (Caerio *et al.* 2005):

$$HPI = \sqrt{[(C_i / S_i)^2_{max} + (C_i / S_i)^2_{min}] / 2} \qquad (1)$$

Where Ci is the reported concentration of each element and Si is the standard permissible value for the element. The Heavy metal pollution index (HPI) is categorized into six classes, Table 2. The metal index calculates the relative contamination of different metals separately and manifests the sum of generated components as a representative (Backman *et al.* 1998). The higher the concentration of a metal compared to its respective maximum allowable concentration (MAC) value, the worse the quality of the water. MI value > 1 is a threshold of warning (Bakun *et al.,*2010). Tamasi and Cini (2004) proposed the following equation to calculate the metal index (MI):

$$MI = \Sigma^{n}_{i=1} Ci / (MAC)_i \qquad (2)$$

Where Ci is the reported concentration of each element and MAC is the maximum allowable concentration.

Table 2 Categories of water pollution index.

Class	PI value	Effect
1	< 1	No effect
2	1-2	Slightly affected
2	2-3	Moderately affected
4	3-5	Strongly affected
5	> 5	Seriously affected

2.4 Principle Component Analysis (PCA)

This technique is the oldest multivariate technique. PCA analyzes a data table representing observations described by several dependent variables, which are, in general, inter-correlated. The goal of PCA is to extract the important information as a set of uncorrelated (i.e., orthogonal) variables. These variables are called principle components, factors eigenvectors, singular vectors, or loadings. Each unit is also assigned a set of scores which correspond to its projection on the components. The results of the analysis are often presented with graphs plotting the projections of the units onto the components, and the loading of the variables. The importance of each component is expressed by the variance (i.e., eigenvalue) of its projection or by the proportion of the variance explained (Abdi 2003).

2.5 Statistical analysis

The data were statistically analyzed using the STATISTICA software (Stat.Soft. Inc., 2007). The means , standard deviations, minimum, maximum and range were calculated of the metal concentrations in water calculated.. ANOVA was executed to detect any significant differences of metal contents temporally and spatially.

3. Results and discussion

1.3 Concentrations of heavy metals

The descriptive statistic results of concentrations of heavy metals in the water samples collected from Euphrates river in the study area are listed in the Table 3. The mean concentration of studied metals in water followed a decreasing order of Pb > Fe > Cr > Cd > Ni. The mean value of Pb (0.528 mg/l) was much higher than the World Health Organization (WHO) standard level for drinking water and Canadian Council of Ministers of the Environment (CCME) guideline for aquatic life and within the permissible limit of Food and Agriculture Organization (FAO) standards. The mean value of Fe (0.340 mg/l) was slight higher than the WHO and CCME standards for drinking water and aquatic life, respectively. It was within the permissible level of FAO guidelines. The mean value of Cr (0.124 mg/l) exceeded the guidelines of WHO, FAO and CCME for drinking, irrigation and aquatic life, respectively. The mean value of Cd (0.116 mg/l) was much higher than the WHO, FAO and CCME guidelines for drinking, irrigation and aquatic life. The mean value of Ni (0.038 mg/l) was with the permissible limit of WHO and FAO guidelines and higher than the CCME guidelines. According to the comparison between the mean values of the heavy metals and the WHO guidelines for drinking water mentioned above, the river was considered polluted by Pb, Cr and Cd. Due to the CCME guidelines, the river was unsafe for the aquatic life. The obtained results were compared with those reported in previous studies conducted in other parts of the Euphrates river and of rivers around the world (Tables 4 and 5). The results of comparison suggest that the metal levels are higher than that reported in the other regions of the Euphrates, with reference to these regions locate downstream relative to this study. This may be due to presence of local pollution sources in the study area and / or river discharge and the rainfall. Euphrates river was less pollution compared to the rivers around the world suggesting different geological, hydrological conditions and manmade activities. The temporal variations of concentration of heavy metals were given in Figure 2. ANOVA results show significant differences ($p < 0.05$) of heavy metals concentrations between the different sampling months. These differences may be attributed to the monthly differences of the river discharge and the rainfall and/ or the industrial and agricultural effluent discharge. Spatial variations of heavy metals concentrations of Euphrates River in the study area were given in Figure 3. ANOVA showed that there was insignificant difference in concentrations of heavy metals between the sampling sites. This result suggest that there are not local pollution sources in the study area.

Table 3 Descriptive statistic results of concentrations of heavy metals (mg/l) compared to guidelines.

Element	Mean	Min.	Max.	Range	Standard Deviation	Water guidelines		
						Drinking[a]	Irrigation[b]	Aquatic life[c]
Cr	0.124	0.040	0.210	0.170	0.041	0.050	0.10	0.001
Fe	0.340	0.142	0.560	0.420	0.103	0.300	5	0.3
Ni	0.038	0.000	0.130	0.130	0.038	0.070	0.2	0.025
Cd	0.116	0.076	0.190	0.110	0.020	0.003	0.010	0.001
Pb	0.528	0.010	1.510	1.500	0.498	0.010	5	0.007

[a] WHO (2011); [b] FAO (1994); [c] CCME (2007).

Table 4 Comparison of mean and range values of heavy metals (mg/l) of Euphrates river and its tributaries with the same in other regions .

Region	Cr	Fe	Ni	Cd	Pb	Reference
Amiriyah Fallujah, Anbar	0.124	0.340	0.038	0.116	0.528	This study
Shatt Al-arab estuary	-	0.389.2	-	0.00019	0.00018	Al-Khafaji (1996)
Qarmatt – Ali river – Iraq	-	0.6905	-	0.00013	0.00031	Al-Khafaji (2001)
Al-Garat river Nassiriya	-	-	-	0.02670	-	Fahad (2006)
Al-Hindiya-Al-Kufa	0.00007	0.10560	0.00007	0.00214	0.00010	Hassan et al. (2010)
Al-Nassiriya City	-	0.726	-	0.00015	0.00020	Al-Khafaji et al. (2011)
Shatt Al-Hilla River	-	0.040-8.110	0.080-13.000	-	0.030-16.090	Hussein et al. (2014)
Abo-Garak to south of Kifil city		0.1054		0.00661	0.00143	Habeeb et al.(2015)

Table 5 Average heavy metal concentrations of rivers around the world (mg/ l)

Rivers & Locations	Cr	Ni	Cd	Pb	Reference
Euphrates , Iraq	0.124	0.038	0.116	0.528	This study
Ganga, India	5.36	4.88	11.5	76.36	Pandey et al. (2010)
Challawa, Nigeria	0.47	-	-	0.44	Azumi and Bichi (2010)
Mghogha, Morocco	86.4	46.83	0.36	48.25	Rodríguez-Barroso et al. (2009)
Sava, Croatia	-	-	0.5	34	Halamić et al. (2003)
Rhine, Netherland	6.3	33.7	7.1	188.2	Middelkoop (2000)
Almendares, Cuba	90	-	2.5	93	Olivares-Rieumont et al. (2005)
Amazon, Brazil	65	26.7	-	83	Siqueira (2003)
Danube, Serbia and Montenegro	76.26	70.1	3.12	28.65	Milenkovic et al. (2005)
Siahroud, Iran	1.03		0.05	9.7	Charkhabi et al. (2008)
Nile, Egypt	-	0.010	0.0045	0.18	Goher et al. (2014)
Korotoa, Bangladesh	0.078	0.035	0085	0.031	Islam et al. (2015)
Haraz, Iran	-	0.0224	0.0026	0.0044	Nasrabadi (2015)
World's average of metals in river water	0.7	0.8	0.08	0.08	Gaillardet et al. (2003).

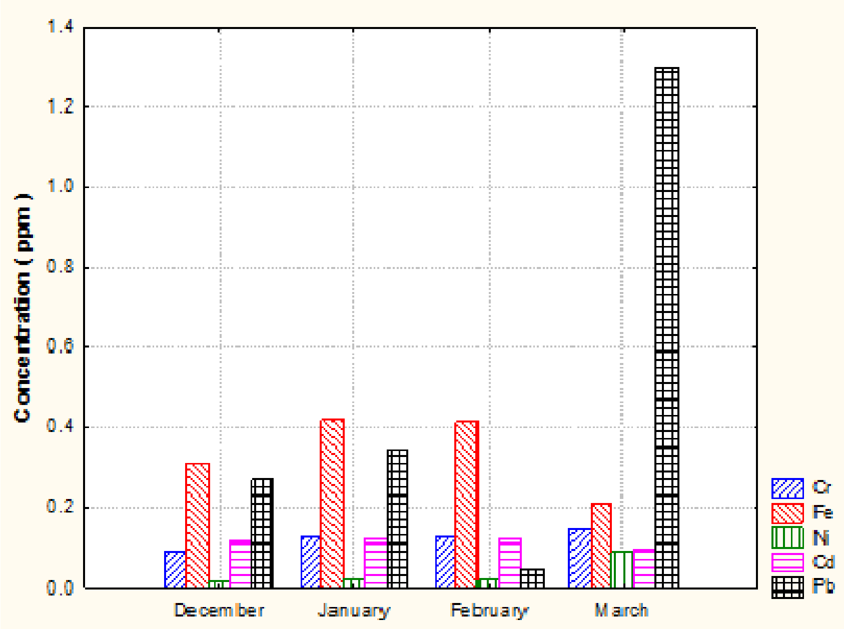

Figure 2. Temporal variation of heavy metals concentrations in Euphrates river.

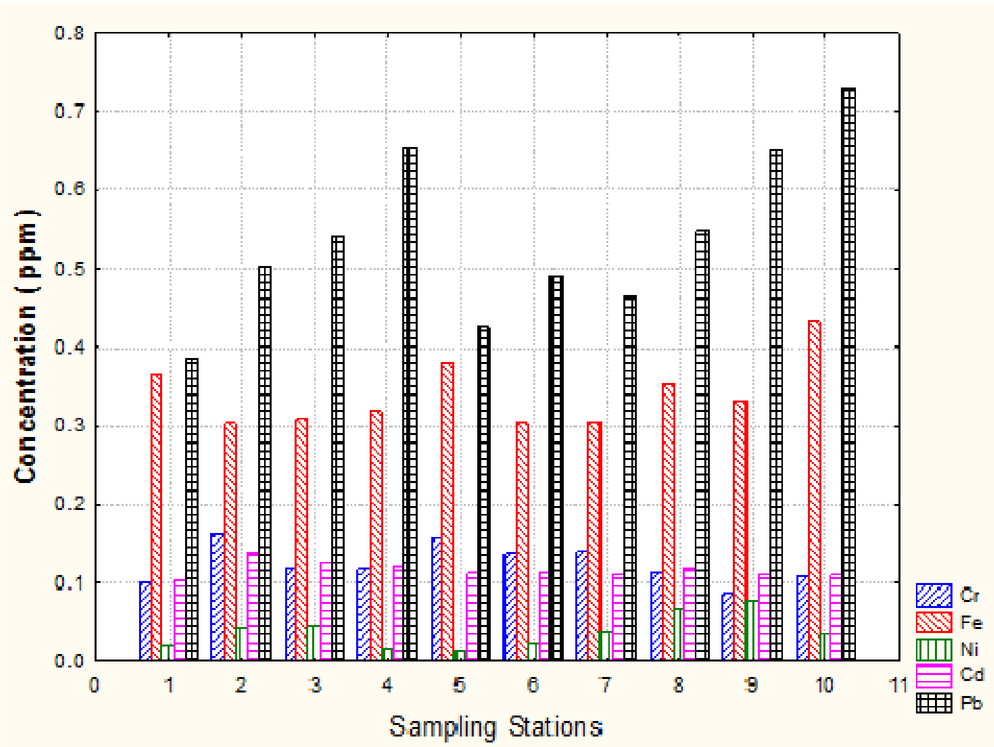

Figure 3. Spatial variation of heavy metals concentrations in Euphrates river.

2.3 Assessment metal pollution

To assess the (HPI) of Euphrates river in the study area, we selected five element (Cd, Cr, Fe, Ni and Pb). Results of calculation of HPI are listed in Table 6. Due to HPI, the metals show a different degrees of pollution in Euphrates river water for different uses. Chromium shows slight to moderate effect for drinking use, no to slight effect for irrigation use and serious effect of aquatic life use. Nickel exhibits slight to moderate effect on aquatic life and no effect for drinking and irrigation uses. The obtained results showed that Cd exhibits serious effect for drinking, irrigation and aquatic life uses. Pb shows serious pollution effect for drinking and aquatic life uses. The spatial variation of HPI for all water uses are given in Figure 4. Cd, Cr and Pb cause slight to serious pollution at all sampling stations for drinking uses, Figure 4a, and the highest values were reported at stations F2 and F10 for Cd and Pb, respectively. Figure 4b illustrates the spatial variation of HPI for irrigation uses. Cd was the serious polluter at all sampling stations along the Euphrates river in the study area. The maximum value of HPI for Cd was recorded at station F2. For aquatic life uses, Cd, Cr, and Pb show serious pollution at the all sampling stations along the Euphrates river, Figure 4c. Cd, Cr and Pb exhibit serious pollution at station F2 for all water uses. This station locates near to the agricultural drainage canal. Station F5 shows serious degree of pollution by Cr, Pb and Cd. This station locates near the municipal and industrial wastewater discharge activities. Only one study was carried out to assess the HPI and MI of Euphrates river in Iraq. Abdullah (2013) calculated the HPI and MI for Euphrates river before confluence with Tigris River at Qurmat-Ali. The mean value of HPI was found to be below the critical pollution index value of 100. Due to this result, Abdullah (2013) suggested that Euphrates River in the study area is unpolluted with heavy metal. Heavy metal index (HMI) is used to assess the metal pollution of Euphrates river water for different uses. Results of HMI estimation are presented in Table 7. These results suggest that the Euphrates river in the study area is seriously threatened with metal pollution (HMI > 1) for drinking, irrigation and aquatic life uses. The spatial variation of HMI is shown in Figure 5. According to the result of the MI, Abdullah (2013) found that Euphrates River is pure with respect to heavy metal pollution. The lower values of HPI and MI reported in study of Abdullah can be interpreted in terms of absence of the local anthropogenic sources and/ or transport of heavy metals from the dissolved phase to the bed sediments of the river.

Table 6 Heavy metal pollution index(HPI) in Euphrates river water according to guideline levels of drinking, irrigation and aquatic life water.

Element / Station	HPI		
	Drinking	**Irrigation**	**Aquatic life**
Cr			
F1	**1.56**	**0.78**	**78.26**
F2	**2.30**	**1.15**	**115.10**
F3	**1.58**	**0.79**	**79.05**
F4	**1.65**	**0.82**	**82.76**
F5	**2.46**	**1.23**	**123.49**
F6	**2.05**	**1.02**	**102.95**
F7	**1.87**	**0.93**	**93.94**
F8	**1.70**	**0.85**	**85.00**
F9	**1.55**	**0.77**	**77.62**
F10	**1.74**	**0.87**	**87.32**
Fe			
F1	**0.84**	**0.05**	**0.84**
F2	**0.70**	**0.04**	**0.70**
F3	**0.73**	**0.04**	**0.73**
F4	**0.76**	**0.04**	**0.76**
F5	**0.87**	**0.05**	**0.97**
F6	**0.70**	**0.04**	**0.70**
F7	**0.72**	**0.04**	**0.72**
F8	**0.80**	**0.05**	**0.80**
F9	**0.74**	**0.05**	**0.74**
F10	**0.97**	**0.05**	**0.97**
Ni			
F1	**0.65**	**0.22**	**1.82**
F2	**0.76**	**0.26**	**2.13**
F3	**0.57**	**0.20**	**1.60**
F4	**0.47**	**0.16**	**1.32**
F5	**0.37**	**0.13**	**1.04**
F6	**0.66**	**0.23**	**1.86**
F7	**0.67**	**0.23**	**1.88**
F8	**0.99**	**0.34**	**2.84**
F9	**0.96**	**0.33**	**2.69**
F10	**0.65**	**0.23**	**1.84**
Cd			
F1	**22.97**	**6.89**	**68.92**

F2	**36.48**	**10.94**	**109.44**
F3	**27.41**	**8.22**	**82.23**
F4	**27.24**	**8.17**	**81.72**
F5	**26.54**	**7.96**	**79.64**
F6	**25.56**	**7.67**	**76.70**
F7	**24.03**	**7.21**	**72.11**
F8	**26.25**	**7.87**	**78.77**
F9	**24.93**	**7.48**	**74.80**
F10	**24.90**	**7.47**	**74.70**
Pb			
F1	**51.84**	**0.10**	**74.06**
F2	**65.06**	**0.13**	**99.85**
F3	**58.50**	**0.11**	**83.58**
F4	**66.59**	**0.13**	**95.13**
F5	**64.65**	**0.12**	**92.36**
F6	**62.00**	**0.12**	**88.58**
F7	**61.50**	**0.12**	**87.86**
F8	**73.50**	**0.14**	**105.00**
F9	**74.17**	**0.15**	**106.00**
F10	**77.07**	**0.15**	**110.10**

3.3 Heavy metal pollution source identification

Because the heavy metals in the river water threaten the aquatic life and human health, it is important to identify the sources of pollution. The original heavy metal contents of rocks and parent materials, processes of soil formation, contamination by human activities, and other anthropogenic factors control the relative abundance of heavy metals (Li *et al*. 2008). Pearson correlation analysis and Principal components analysis (PCA) were performed to determine the most common pollution sources.

1.3.3 Correlation between heavy metals

Pearson correlation coefficient matrix was listed in Table 8. Generally, a correlation coefficient > 0.7 is interpreted as a strong relationship between two parameters, whereas values between 0.5 and 0.7 represents a moderate relationship. Significant correlations were found between Fe and Ni (r = - 0.660), Fe and Cd (r = 0.399), Fe and Pb (r = - 0.735), Ni and Cd (r = - 0.329), Ni and Pb (r = 0.841), and Cd and Pb (r = - 0.418) at the $p < 0.05$ level. Cr did not show significant correlations with these metals (Table 8). Correlation analysis provides an effective way to reveal the relationships between multiple variables and thus have been helpful for understanding the influencing factors as well as the sources of chemical components (Li *et al*. 2013).

Table 7 Heavy metal indices (HMI) of Euphrates river in the study area.

Station	HMI		
	Drinking	Irrigation	Aquatic Life
F1	76.35	11.56	260.09
F2	101.37	15.89	375.39
F3	99.63	14.04	321.88
F4	108.92	13.44	331.98
F5	84.42	12.99	331.50
F6	90.18	13.30	357.93
F7	87.16	12.63	317.91
F8	98.39	13.42	311.88
F9	105.96	12.55	295.08
F10	113.71	12.46	324.04

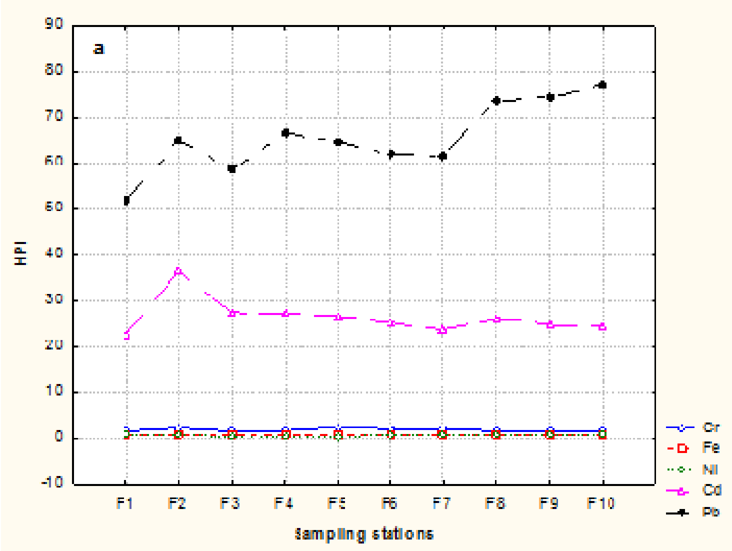

Figure 4 Spatial variation of HPI in Euphrates River for all water uses: (a) drinking (b) irrigation and (c) aquatic life.

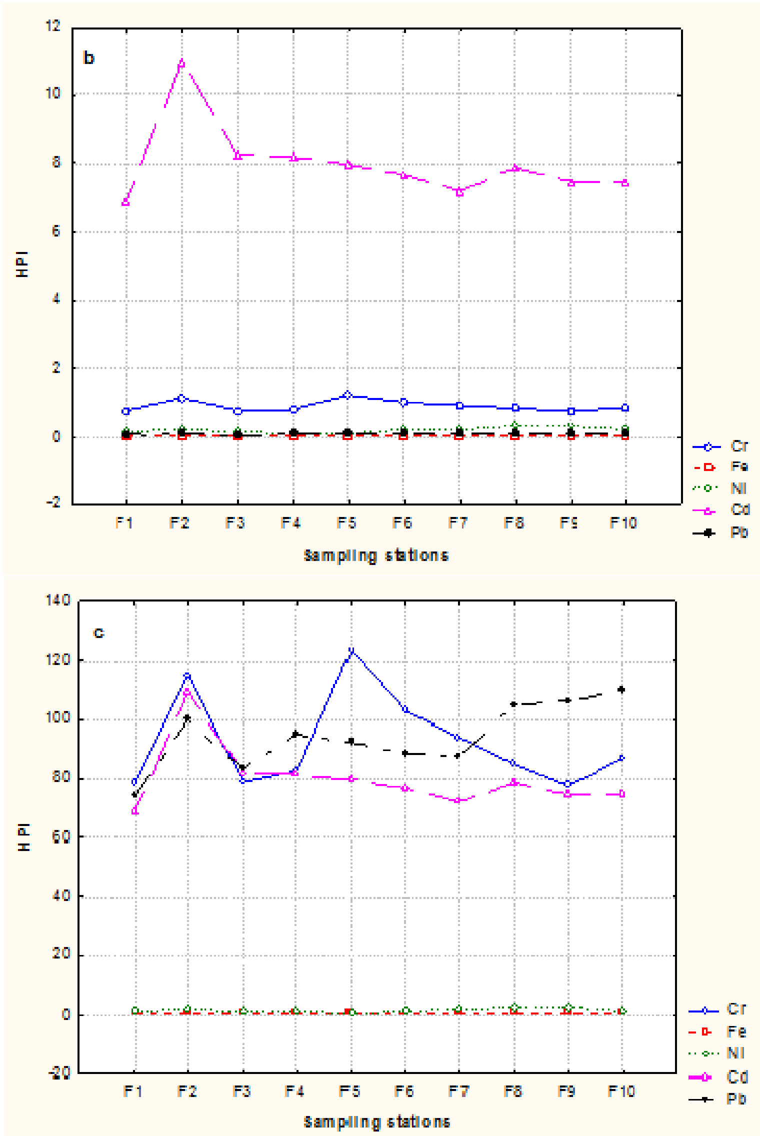

Figure 4 Continued.

The relationship between heavy metals can provide important information on heavy metal sources and pathways (Manta *et al*., 2002). High correlations between specific heavy metals in the water may reflect similar levels of pollution and/or release from the same sources of pollution. There was strong positive correlation between Ni and Pb suggesting anthropogenic sources of these heavy metals. The concentration of Cr showed a weak to very weak correlations with other metals suggesting that Cr was from different source than other metals.

2.3.3 Principle component analysis (PCA)

The results of PCA for heavy metal contents in water are listed in Tables 9 and 10. The first two principal components accounted for 71.493 % of total variance, and the variance of F1 and F2 was 41.107 and 30.386, respectively. Factor 1 had strong negative loadings on Cr, and positive loadings on Ni and Pb. The

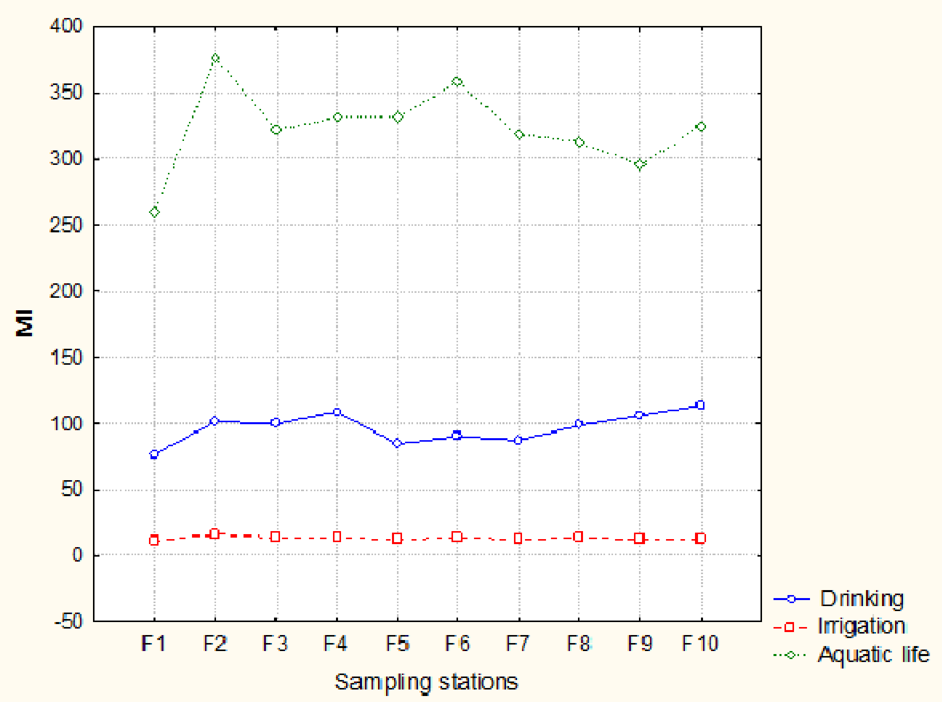

Figure 5 Spatial variation of MI in Euphrates river for all water uses.

Table 8 Pearson correlation matrix between different metals in the water of Euphrates River.

Element	Cr	Fe	Ni	Cd	Pb
Cr	1.000				
Fe	-0.161	1.000			
Ni	0.182	***-0.660***	1.000		
Cd	-0.109	***0.399***	***-0.329***	1.000	
Pb	0.311	***-0.735***	***0.841***	***-0.418***	1.000

Marked correlations are significant at $P < 0.05$.

association of elements Ni and Pb in F1 may be attributed to anthropogenic source and may originate from similar pollution sources. This result coincides with the conclusion of the correlation analysis. The source of these heavy metals in this area comes mainly from industrial activities , industrial wastewater, and domestic sewage. When the industrial activities is source of heavy metals, several metals show elevated level jointly. Factor 2 comprises Fe and Cd and had high positive loadings. This result is in good agreement with the conclusion of the correlation analysis.

4. Conclusions

In the present study, mean concentration of studied metals in water followed a decreasing order of Pb > Fe > Cr > Cd > Ni. The mean values of concentrations of heavy metals were higher than the safe recommended guidelines (WHO, CCME), which suggested that the Euphrates river is polluted by Cd, Cr and Pb and unsafe for drinking and aquatic life. Significant correlation relationships between heavy metals pairs were reported suggesting existence of a common source of these metals in the water and /or the same pollution level. ANOVA results show significant and insignificant difference ($p < 0.05$) of heavy metals concentrations between the different sampling months and sampling sites, respectively. The HPI values showed that Cd, Cr and Pb exhibit serious effect on aquatic life, and slight to serious effect on drinking water. The MI values suggest that the

Euphrates river in the study area is seriously threatened with metal pollution for drinking, irrigation and aquatic life uses. Principal component analysis (PCA) identified five heavy metals controlling the variability in water, which account for 41.107% (factor 1: Cr, Ni and Pb) and 30.386% (factor 2: Cd and Fe) of the total variance. F1 and F2 cover metals having origins in both natural and anthropogenic sources.

Table 9 Principal component analysis of Euphrates river water samples.

Component	Eigenvalue	%Total Variance	Cumulative Eigenvalue	Cumulative Variance %
1	2.055	41.107	2.055	41.107
2	1.519	30.386	3.574	71.493

Table 10 Factor loadings (Varimax rotation) on heavy metals in Euphrates river water samples.

Element	**Factor 1**	**Factor 2**
Cr	***-0.730***	-0.512
Fe	0.093	***0.801***
Ni	***0.814***	-0.254
Cd	0.063	***-0.884***
Pb	***0.774***	0.103
Eigen value	2.055	1.519
Variance explained %	41.107	30.386
Cumulative variance %	41.107	71.493

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