Vertical and Lateral Distribution of Heavy Metals in the Euphrates River Sediments between Heet and Fallujah, Western Iraq

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Abstract

The present work aims to investigate the vertical and lateral distribution of heavy metals and to assess pollution level in the Euphrates River sediments along the area extended between Heet to Fallujah cities in October 2020. The sediment samples were collected from five sites and three depths range 0–30 cm, 30–60cm, and 60–90cm. The concentration of heavy metals sediment samples was compared with reference values. The results showed that the mean concentrations of heavy metals take the following ascending order: cadmium (1.8 mg/kg), lead (12.48 mg/kg), Cobalt (16 mg/kg), copper (24.88 mg/kg), chromium (46.07 mg/kg), zinc (58.75 mg/kg) and nickel (114.44 mg/kg). The results did not show a similar vertical distribution pattern of heavy metals among the five sampling sites. The cadmium showed high enrichment compared to the other metals that showed low to moderate enrichment according to reference values. The contamination factor and geoaccumulation index showed high contamination with Cd, moderate with Ni and low with other metals.

Keywords: Heavy metals; Pollution; Sediments; Euphrates

1. Introduction

In recent decades, significant attention has been paid to heavy metal contamination as a dangerous environmental problem because of the toxicity, abundance, persistence and bioaccumulation of these elements (Chen et al., 2016,) (Islam et al., 2017 and) (Jin et al., 2019). In general, the contamination state of the environment is evaluated by the total concentration of heavy metals (Kanda et al., 2018). The riverine sediments are considered as the main basins for various pollutants following the largely uncontrolled discharge of contamination resulting from human activities and geogenic processes (Liu et al., 2018). The sediment contamination is an important indicator of environmental variation as a result of anthropogenic influence (Gao et al., 2019). River sediments serve as not only a major sink and carrier of heavy metals but also as potential sources of secondary pollution, which can reflect their contamination level (Tang et al., 2014 and) (Hsu et al., 2016). The heavy metals are released into the aquatic environments from geogenic and anthropogenic sources. The geogenic sources include chemical leaching of bedrock, water drainage basins and runoff from banks (Raj et al., 2017). The anthropogenic sources of heavy metals pollution in aquatic systems include mining activities, industrial wastes

DOI:

disposal and pesticides use, (Chakravarty & Patgiri, 2009). The heavy metals pollution of sediments is an indicator of the aquatic systems water quality (Zhao et al., 2012). The heavy metal distribution in sediments and pollution levels supplies a base for consideration of sediments treatment methods and evaluation of the potential releasing of heavy metals into water and transport downstream (Nawrot et al., 2020). The vertical and spatial distribution of heavy metals and pollution levels have been evaluated in sediments of many world rivers, such as the Yinma River, China (Guan et al., 2018), the Voghji River, Armenia (Gabrielyan et al., 2018), the Barigui River, Brazil (Machado et al., 2017), the Harazdan River, Armenia (Petrosyan et al., 2019), the Thames River, the UK (Vane et al., 2020), the Yang River, China (Tang et al., 2020), and the Lu Lu River, China (Ye et al., 2020). The spatial distribution of heavy metals in sediments of the Euphrates River in Iraq has been investigated by many authors (Issa & Qanbar, 2016); (Al-Taher et al., 2020); and (Hussain & Al-Jaberi, 2020). The spatial variation of heavy metal concentrations in sediments of the Euphrates River along the studied area between Heet and Ramadi Cities has been studied by (Al-Bassam & Al-Mukhtar, 2008) and (Salah et al., 2012). The aim of the study is to investigate how heavy metals are laterally and vertically distributed in sediments of the given study area of Euphrates River between Heet and Fallujah Cities. This study represents the first attempt to investigate the vertical distribution of heavy metals in the Euphrates River sediments.

2. Materials and Methods

2.1. Sediment Sampling

The sediment samples were collected from five sites located between Heet and Fallujah Cities, (Table 1) and (Fig. 1). The sediments were sampled from three depth ranges (0-30, 30-60, and 60-90 cm) using an auger tube sampling device.

Table 1. Location of sampling sites in the study area

Sampling region	Sampling site	Latitude (N)	Longitude (E)
Heet	S1	33.649730°	42.817123°
Muhammadi	S2	33.565374°	42.900063°
Ramadi	S3	33. 441916°	43.315409°
Khalidiya	S4	33.397504°	43.523923°
Fallujah	S5	33.337579°	43.762461°

2.2. Sample Preparation and Analysis

The samples were dried and then inserted into the oven at a temperature of 70 degrees Celsius for a period of 24 hours. The sediment samples were crushed with ceramic mortar (Agate) and homogenized, then sieved with a 150 μm steel sieve for digestion (EPA Method 3050B, 2012) (Islam et al., 2015). A mixture of digesting sediments from concentrated acids (hydrochloric acid, and nitric acid) was prepared at a ratio of 3:1 respectively, (or Aqua Regia solution). Then, 1 gram of the dry sample was weighed and 10ml of the prepared lemon mixture was added to it and the sample was stirred. Next, the mixture was put in a microwave and digested for 15 minutes to near dry at a temperature of 190 0C, after which it was to cool down and washed with deionized water several times until the entire sample was removed. It was then filtered with Whiteman (42 Mm) filter paper and the volume were topped up with distilled water to make 50ml (Islam et al., 2015). The digested samples are analyzed for Zn, Cu, Fe, Cr, Co, Ni, pb and Cd using Atomic Absorption Flame Emission Spectrophotometer (ASC.7000) at Anbar Water Directorate, Central Water Laboratory.
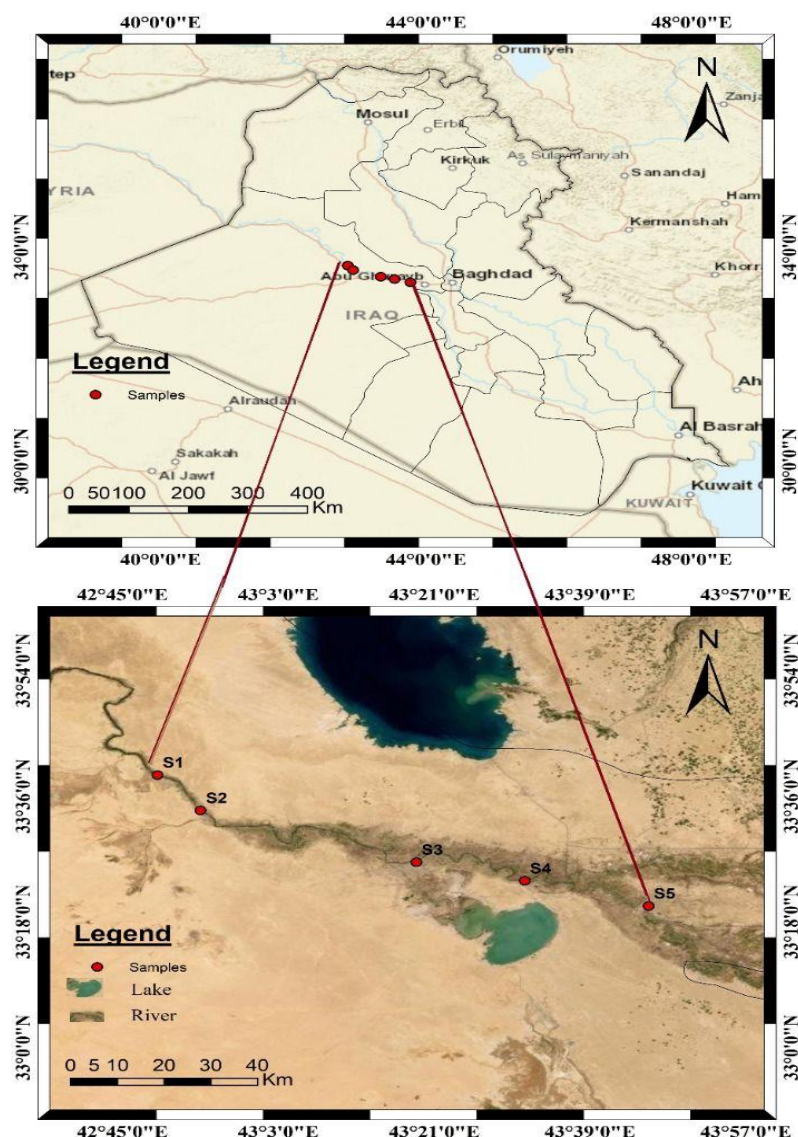
Fig.1.   Location map of the study area and sediment sampling sites

2.3. Pollution Level

The enrichment factor (EF), the geo-accumulation Index (Igeo), the contamination factor (CF), and the pollution load index (PLI) were used to evaluate level contamination by heavy metals in the studied stations. Specific to each of these measures, the values were calculated, as well as the specific levels for each of them. These levels were collected from many data sources related to pollution phenomena, and from these data the pollution limits were determined low, high, or severe. International reference values were used for the elements in our study (Martin & Meybeck, 1979)

Enrichment Factor (EF) = Cm/CFe sample)/(Cm/CFe) background (Sinex & Helz, 1981)   (1)

Cm = element concentration in sample

CFe= Concentration of Iron

Geo-accumulation Index (Igeo) = Log2 [Cm Sample/ (1.5× Cm Background)]   (2)

(I geo) = Ln {Cm/ 1.5 * CmB }(dos Santos et al., 2017)

Cm = element concentration in sample

CmB = the reference value of the element.

$$\text{Contamination factor (CF)} = Cm / Bn \text{ (Müller, 1969)} \qquad (3)$$

Bn = the reference value of the element in nature

Cm = element concentration in sample

$$PLI = (CF1 \times CF2 \times CF3 \times …...CFn) \ 1/n \text{ (Mmolawa et al., 2011)} \qquad (4)$$

n = number of elements

3. Results and Discussion

3.1. Heavy Metal Concentrations in Sediment

The results of analysis of heavy metals in sediment sample of the Euphrates River are listed in Table 2. The results showed that Pb concentration ranges from 8.79 and 17.14 with mean of 12.48 mg/kg. This is less than previous studies in the sediments of the Euphrates River in study area (Salah et al., 2012). As for Cd concentrations ranges from 1.129 and 3.255, mean of 1.8 mg/kg. The results obtained were lower than that's of the previous studies of the Euphrates River sediments (Salah et al., 2012; Al-bassam, 2011). The Fe concentration in sediment of the Euphrates River ranges from 15900 and 28692, mean of 24969 mg/kg. The results were higher than previous studies of the Euphrates River (Salah et al., 2012). In addition, Cu concentration in sediment of the Euphrates River stations ranges from 17.61 and 40.81, mean of 24.88 mg/kg. These results were comparable to previous studies of the Euphrates River (Al-Bassam & Al-Mukhtar, 2008), while the values of Cu concentrations in the current study were higher than in a previous study (Salah et al., 2012). While Zn concentrations ranges from 42.68 and 75.85, mean of 58.75 mg/kg. The results of Zn concentrations were lower in previous studies of the Euphrates River (Al-Bassam & Al-Mukhtar, 2008; Salah et al., 2012). As for the Cr concentration in sediment of the Euphrates River stations ranged between 30.48 and 59.88, mean of 46.07 mg/kg.

Table 2. Concentration of heavy metals(mg/kg) in sediment samples of Euphrates River

No	Station	Depth	Pb	Cd	Fe	Cu	Zn	Cr	Ni	Co
1		S.H.30	12.13	2.895	15900	18.326	42.68	30.480	62.064	11.12
2	S1	S.H.60	15.13	3.255	16741	18.929	75.85	33.671	65.507	11.23
3		S.H.90	14.80	3.075	18908	22.245	69.33	37.379	72.269	11.55
4		S.M.30	8.79	1.1820	26833	17.663	46.04	47.987	104.05	14.81
5	S2	S.M.60	10.79	1.9009	22135	17.603	62.92	47.125	85.485	13.21
6		S.M.90	10.45	1.4681	27612	20.195	47.48	50.143	95.628	14.93
7		S.R.30	9.78	1.1655	26097	19.532	49.25	46.262	117.76	16.35
8	S3	S.R.60	9.45	1.1582	27115	23.872	48.72	40.484	120.095	15.92
9		S.R.90	9.12	1.1291	26453	25.213	48.33	39.967	138.291	16.47
10		S.K.30	10.45	1.2107	28692	21.159	57.73	39.794	118.742	16.84
11	S4	S.K.60	12.46	1.5077	25984	25.500	57.44	52.558	131.713	17.95
12		S.K.90	14.80	1.6975	28218	28.394	62.91	50.661	137.369	18.62
13		S.F.30	17.14	1.9543	27880	35.989	72.48	57.301	146.959	19.91
14	S5	S.F.60	16.82	1.8446	28269	40.812	70.35	59.889	158.946	20.28
15		S.F.90	15.13	1.5713	27691	37.798	69.81	57.474	161.774	20.77

These results are comparable to previous studies in the Euphrates River (Al-Bassam & Al-Mukhtar, 2008), while they were less than in one other study (Salah et al., 2012). While, Co concentrations ranges from 11.12 and 20.77, mean of 16 mg/kg. The results that we obtained were lower due to an approach to previous studies of the Euphrates River (Al-Bassam & Al-Mukhtar, 2008; Salah et al.,

2012). As for The Ni concentration in sediment samples of the Euphrates River stations ranges from 62.06 and 161.77 mean of 114.44 mg/kg. The results of Ni obtained in our study were higher due to an approach to previous studies of the Euphrates River (Al-Bassam & Al-Mukhtar, 2008; Salah et al., 2012). Results of correlation matrix analysis for heavy metals in the sediment of Euphrates River are listed in Table 3. The correlation coefficient shows a significant positive relation between chromium with nickel, copper, and cobalt. A significant positive correlation between copper with zinc and lead, Also a positive correlation between nickel with iron and cobalt, addition to a positive correlation between cobalt and iron, at ($P < 0.05$) , (Table 3), which is evidence of a common source for these elements, the origin of the geological rock (Varol et al., 2020).

Table 3. Correlation matrix analysis of elements in sediments

Element	Cd	Cr	Cu	Ni	Zn	Pb	Co	Fe
Cd	1.00							
Cr	-0.45	1.00						
Cu	-0.12	**0.73**	1.00					
Ni	**-0.63**	**0.78**	**0.82**	1.00				
Zn	0.47	0.33	**0.54**	0.20	1.00			
Pb	**0.56**	0.38	**0.70**	0.24	**0.85**	1.00		
Co	**-0.60**	**0.84**	**0.83**	**0.98**	0.24	0.32	1.00	
Fe	**-0.86**	**0.73**	0.49	**0.84**	-0.08	-0.11	**0.85**	1.00

Marked correlations are significant at $p < 0.05$.

3.2. Horizontal and Vertical Distribution of Heavy Metals

3.2.1. Horizontal Distribution of Heavy Metals

Heavy metals are natural crystalline compounds that originate from igneous, metamorphic rocks, and sometimes sedimentary rocks, and are transported by rivers as a result of erosion and the erosion of the parent rocks. Heavy metals undergo a recycling phase as a result of the erosion of these rocks, which in turn become secondary source rocks for these minerals (Morton & Hallsworth, 1999). Thus, source rocks play a fundamental role in the quality of heavy metals in river sediments, in addition to the climatic conditions prevailing in the source areas playing an important role in the changes in heavy metals. The transportation distance may also play a similar role in this process (Pettijohn, 1975). Al-Bassam & Al-Mukhtar (2008) concluded that river torsions play a role in the concentration of some heavy metals as well as being important for the relationship of the grain size of minerals with their concentration ratios in modern river sediments. Human pollutants are directly related to the pollution of water and sediments with the advancement of mankind and practices of industry, commerce and agriculture (Al-Bassam & Al-Mukhtar, 2008). The distribution of Pb in the study stations showed the following ascending order: Fallujah >Heet > Khalidiya> Muhammadi> Ramadi due to the different sources of pollution from one station to another along the Euphrates River, (Fig. 2). The most important of these are the sewage and drainage water discharged from agricultural lands fertilized with phosphate fertilizers, which are directly thrown into riverbeds, leading to an increase in the Cd concentration in the riverine environment (Charkhabi et al., 2008). While the vertical distribution of Pb in each station was adopted differently and unevenly between the layers of each station, as the concentration increased in the upper layers (30cm), as in Fallujah and Ramadi, and decreased in the lower layers (90cm), (Fig.

2). This is due to the continuous increase in pollution over time at each station. Muhammadi and Heet stations show an increase in Pb concentration in the depth range (30- 60cm).
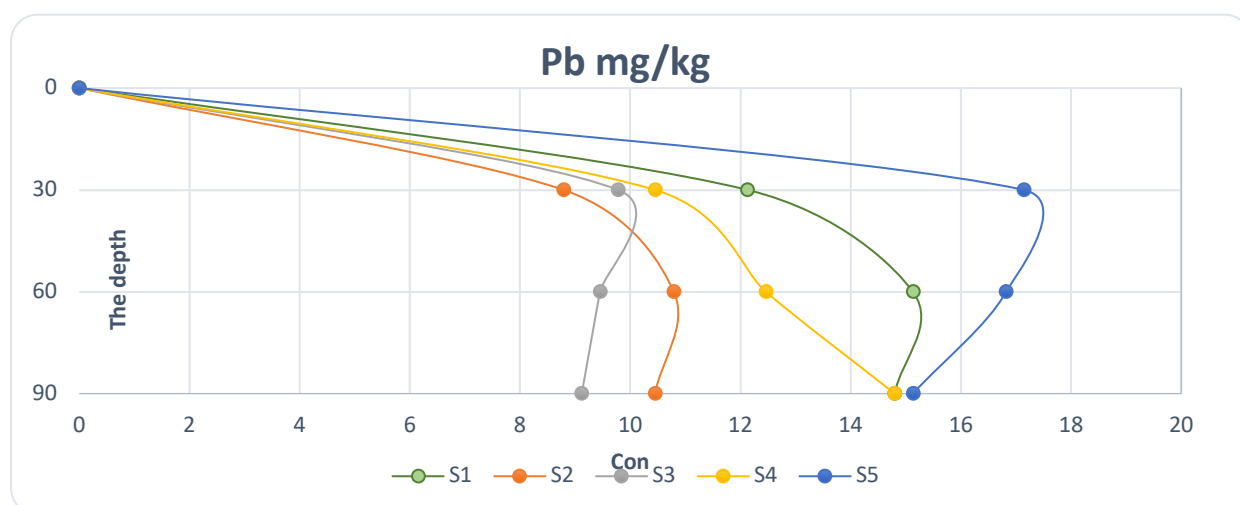

Fig. 2. Concentrations of lead (mg/Kg) in sediment of the Euphrates River

The distribution of the Cd in the sampling sites follows the descending order: Ramadi< Khaldiya< Muhammadi< Fallujah< Heet. This order is due to the different sources of pollution along the Euphrates River from one station to another (Fig. 3). The vertical distribution of cadmium in each site showed different distribution patterns between the layers, with an increased concentration in the middle layers (60cm), as in Heet and Muhammadi, and a decreased concentration in the other layers (Fig. 3). It increased in the upper layers (30cm) from the other layers, as in Fallujah and Ramadi, and it also increased in the bottom layer (90cm) from the other layers in the Khalidiya station.
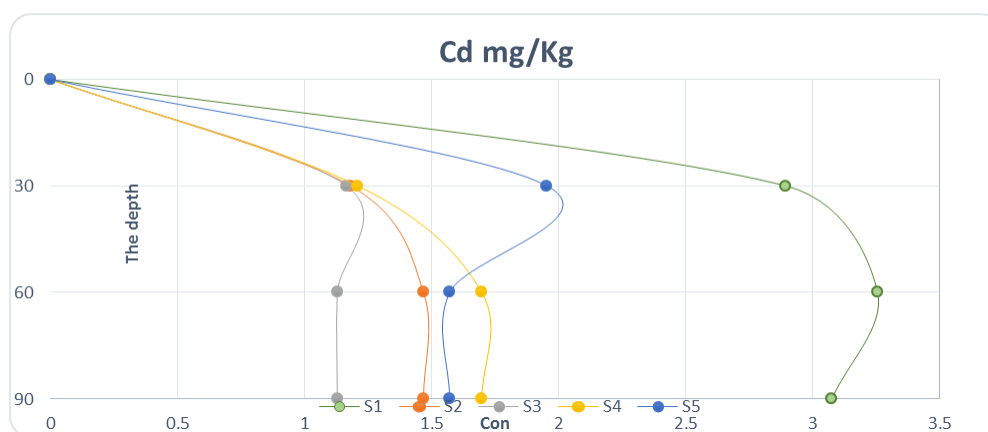

Fig. 3. Concentrations of cadmium (mg/Kg) in sediment of the Euphrates River

The results of the Fe distribution showed the following horizontal descending: Fallujah> Khaldiya> Ramadi> Muhammadi> Heet (Fig. 4). High iron concentrations are caused by the geological rock origin of the earth's crust and the accompanying processes of erosion of rocks and soil dredging that accompany torrents that enter the riverbed and lead to an increase in iron in river sediments. While the vertical distribution of iron in each station showed an upward distribution towards the lower layers in most of the stations. This is due to the natural origin of iron from the geological rocks of the earth's crust. In addition, the vertical distribution of copper in each station showed an ascending distribution towards the lower layers in all the studied stations, (Fig. 4).
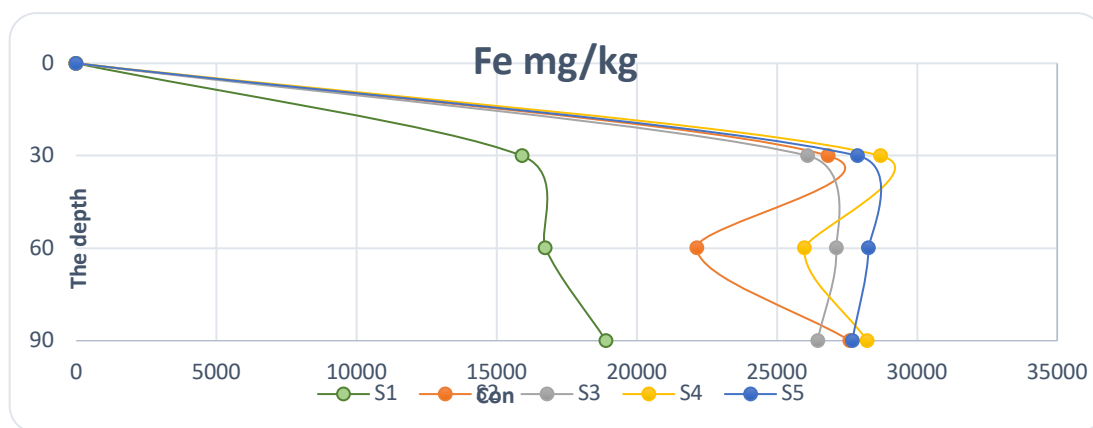
Fig. 4. Concentrations of iron(mg/Kg) in sediment of the Euphrates River

The results reveal the following horizontal distributions of Cu for the study stations: Muhammadi< Heet< Ramadi< Khaldiya< Fallujah (Fig. 5). The reason for this is the nature of the sediments of the crumbs and the torsional movement of the river's flow, erosion and weathering factors impacting sedimentary rocks, and human activity, which is represented by domestic and industrial wastewater such as thermal power stations and combustion emissions (Denton et al., 1997). In addition, the vertical distribution of copper in each station showed an ascending distribution towards the lower layers in all the studied stations (Fig. 5), the being the rocky origin of copper and the components of the earth's crust, considered a relatively abundant element in the lithosphere (Callender, 2000).
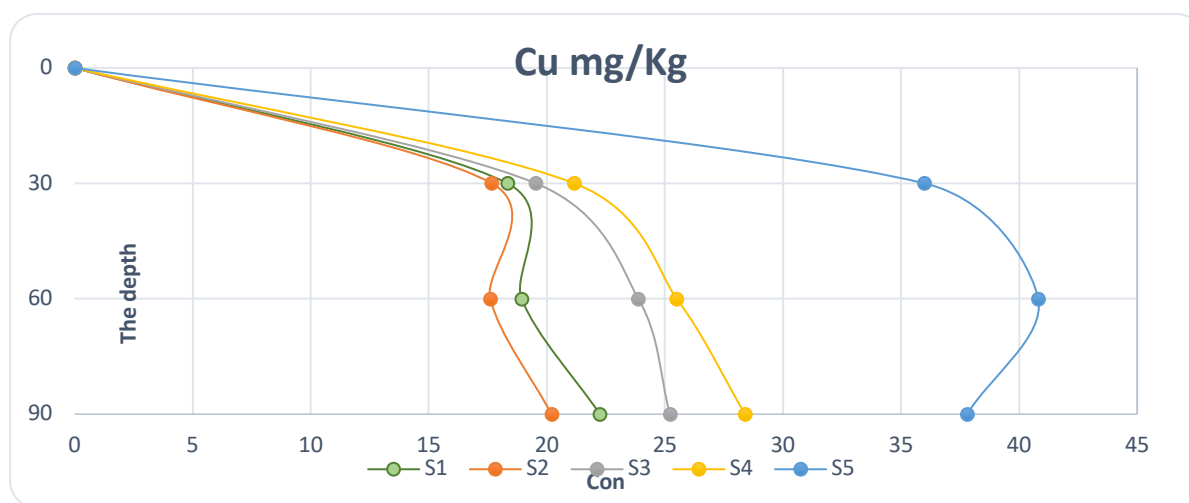

Fig. 5. Concentrations of copper(mg/Kg) in sediment of the Euphrates River

The Zn element distribution in the study was taken was as follows: Ramadi< Muhammadi< Al-Khalidiya< Heet< Fallujah (Fig. 6). The difference in concentration among the study stations, as well as the geological differences and the natural origin of the element according to the nature of the area, erosion factors and weather conditions, are due to the different sources of pollution along the Euphrates River, the most important of which are sewage and industrial water, mining operations and burning coal (Gautam et al., 2014). As for the vertical distribution of the zinc element in each station, it differed between the layers, and there were no significant differences for the zinc in the layers for each station, (Fig. 6.).
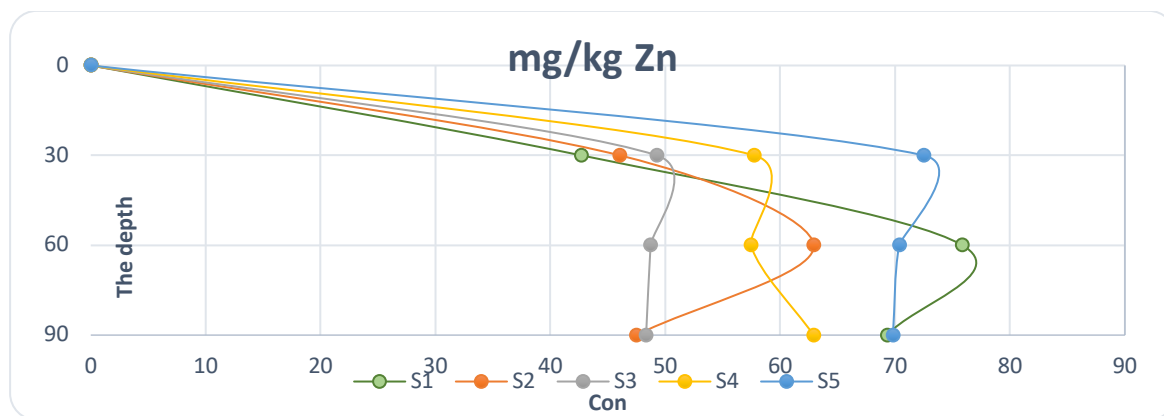
Fig. 6. Concentrations of Zn (mg/Kg) in sediment of the Euphrates River

The distribution of Cr for the study stations showed the following descending order: Fallujah> Muhammadi> Khalidiya > Ramadi> Heet (Fig. 7). The presence of chromium comes mainly from the rocky origin of the earth's crust and the impact of environmental and climatic factors such as erosion, weathering and redistribution according to river movements, in addition to human activity represented by water discharges, sanitation and industrial waste. While the vertical distribution of chromium in each station, it showed a different variation in each station, rising in the lower layers (90cm), as in Heet and Muhammadi, and decreasing in the other layers, as well as rising in the middle layers (60 cm), as in Khalidiya and Fallujah, and rising in the upper layer (30 cm) (Fig. 7). As in the Ramadi station, most of the elevation in the lower and middle layers is due to the rocky origin of the element and the impact of transport and river movements, in addition to the movement of aquatic organisms, leading to a redistribution between the layers (Sindern et al., 2016). In addition, the increase in the upper layers may be due to the increase in human pollutants excreted from industrial waste and sewage water.
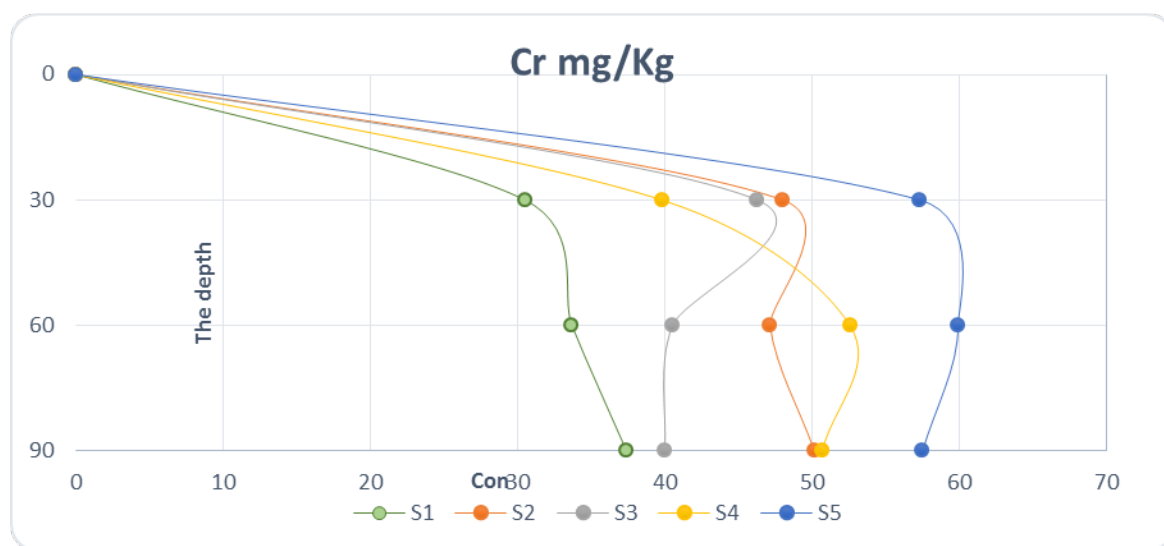

Fig. 7. Concentrations of Cr (mg/Kg) in sediment of the Euphrates River

The results for the distribution of Co for the study stations showed the following horizontal order: Fallujah > Khaldiyah> Ramadi> Muhammadi > Heet, (Fig. 8). The main reason for this successive difference or increase is the natural resources and the nature of the rocks and sediments that make up the area of each station, in addition to human activity factors represented by wastewater and industrial waste, (Rosenberg, 2015). While the vertical distribution of the cobalt element in each station also showed an upward distribution towards the lower layers in all stations due to the geological rock origin

and its high content in geological formations and the environmental and climatic conditions these formations suffer from that change the behavior of division in river sediments (Sindern et al., 2016).
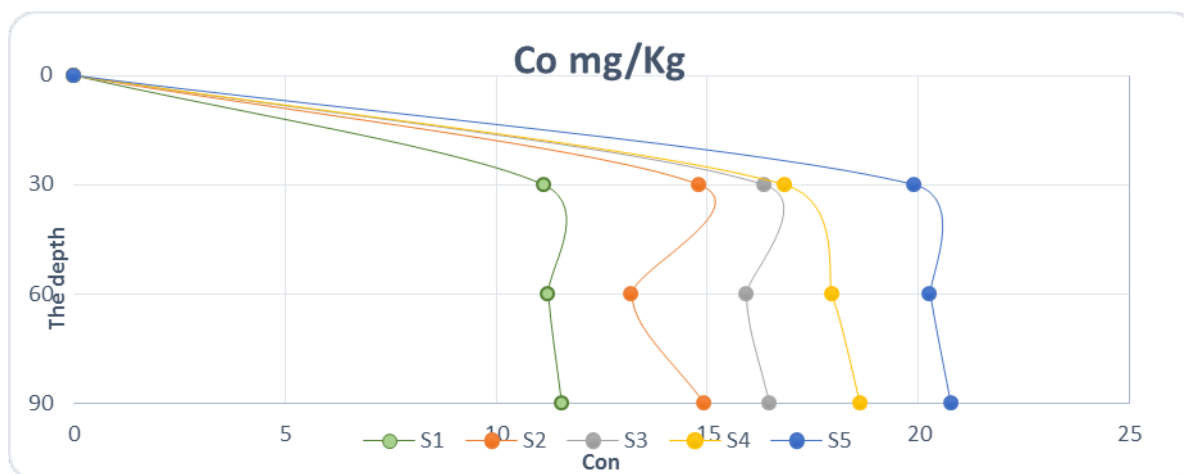

Fig. 8. Concentrations Co. (mg/Kg) in sediment of the Euphrates River

The distribution of Ni in the study stations showed the following ascending order: Heet< Muhammadi< Ramadi< Khalidiya< Fallujah (Fig. 9). Due to the geological rock origin and its high content in geological formations and environmental and climatic conditions, these formations suffer from changes in the behavior of division in river sediments (Sindern et al., 2016).
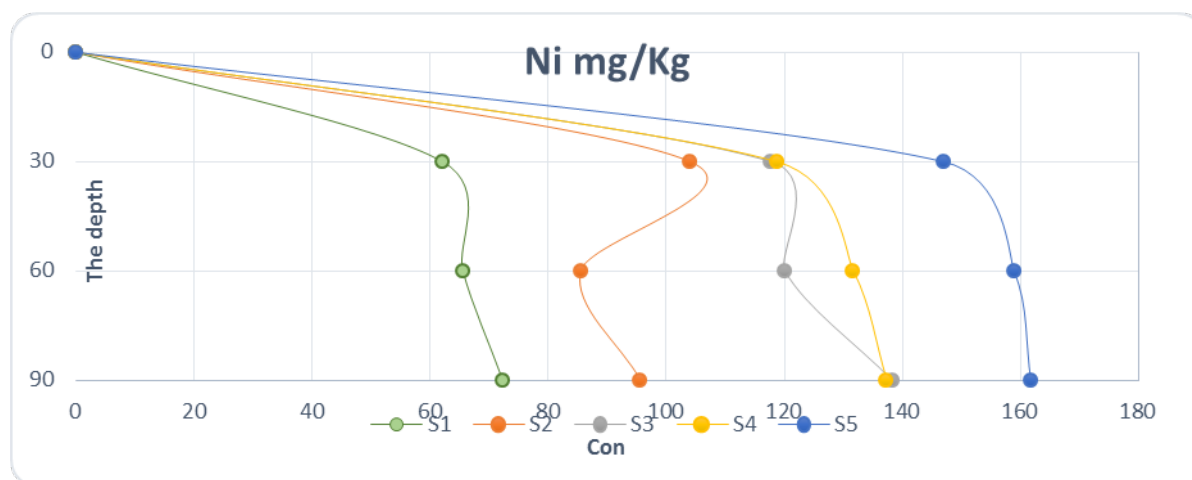

Fig.9. Concentrations of Ni (mg/Kg) in sediment of the Euphrates River

In general, the continuous or escalating increase in the concentrations of heavy elements from the city of Heet to the city of Fallujah are attributable to the movement of the river and the torsions that exist along the river and the elements carried by the river's water of elements that may be gradually deposited. This is in addition to the human waste that is thrown into the river without any prior treatment. In addition, the vertical distribution of nickel in each station also showed an upward distribution towards the lower layers in most stations, except for Muhammadi station, Fig. The reason for this is due to the natural origin of nickel in the rocks of the earth's crust as well as it being a natural component of the soil, plus the accompanying factors of erosion, weathering, torrential water and rain and what is carried from the elements that eventually drain into the river, leading to increased nickel in the sedimentary strata of the river (Rosenberg, 2015; Al-Bassam & Al-Mukhtar, 2008). In general, there were no significant differences between the layers in each station, the possible reason being the external

processes that result from the movement of the river and changes in the pH value of the water, which causes re-dissolution and sedimentation between the sedimentary layers,transfer operations as a result of the movement of aquatic organisms inside the bottom of the sediments, as well as the changing the state of sediments due to grazing, excavations, and the like (Khalid, 1980). Most of the increase in cadmium concentrations in river sediments, in addition to the geological origin of rocks, erosion factors, and air transport to industrial and agricultural human activity, as the release of heavy municipal water into rivers without treatment is one of the causes of increased pollution (Rauf et al., 2009).

3.2.1. Vertical Distribution of Heavy Metals

The distribution of the concentrations of the heavy metals for all depths in the sediment samples of Euphrates River was dependent on the following horizontal arrangement Ni< Zn< Cr < Cu < Co < Pb < Cd (Fig. 10). It was observed that there were slight differences in the concentrations of heavy elements between the three layers (0-30cm), (30-60 cm), and (60-90 cm), indicating that there were no sources of high pollution in that time period. In general, we notice an increase in the concentrations of the heavy metals towards the Fallujah station, that is, an increase from upstream to downstream in sediment samples of Euphrates River.
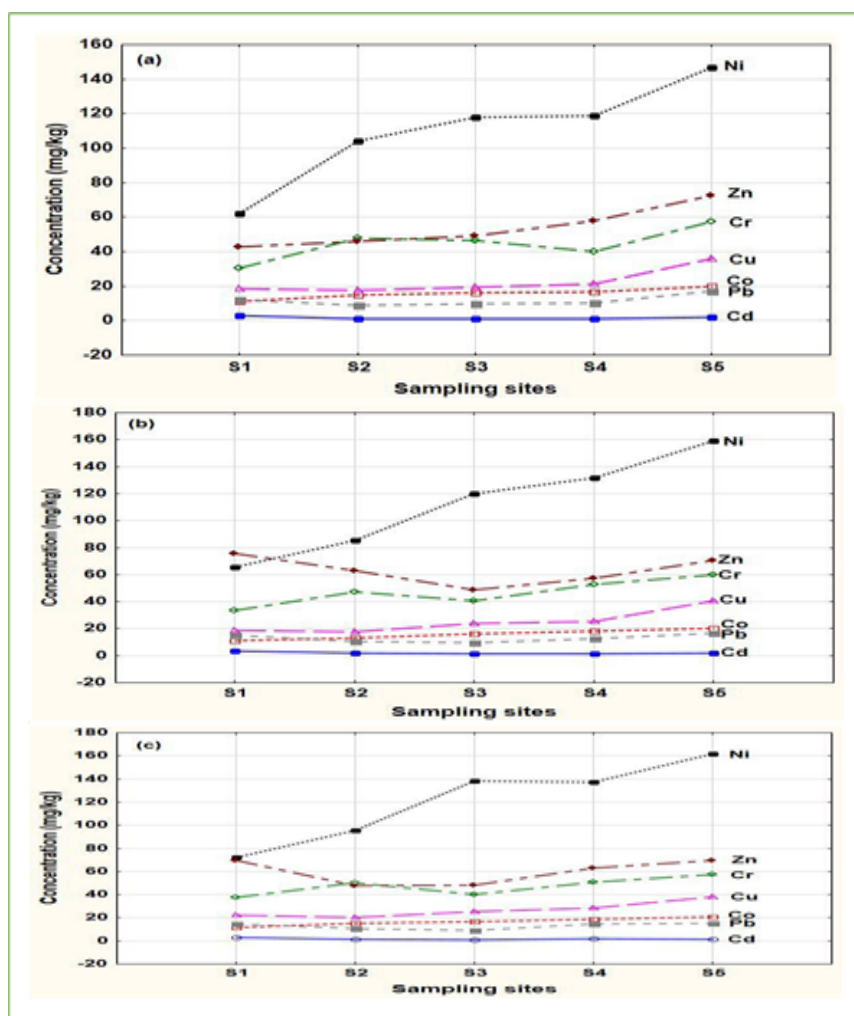

Fig. 10. A) The vertical distribution in sediment sample of Euphrates River in 30 layers; b) the vertical distribution in sediment sample of Euphrates River in 60 layers; c) the vertical distribution in sediment sample of Euphrates River in 90 layers

3.3. Pollution Levels for Sediment of the Euphrates River

An assessment of elemental contamination was carried out at the selected study stations to determine the state of the river. Expressions of concentration are insufficient for investigating the presence or absence of contamination. Therefore, pollution indicators were used to express the terms 'polluted' or 'non-polluted'. In general, the evaluation revealed the presence of moderate to low pollution in all the study stations. This pollution or increase in the concentration of some elements is due to the sedimentary layers and the pollutants carried by torrents that end up in the river, as well as, possibly, human activity, as in the increase in cadmium resulting from the use of phosphate fertilizers and the drainage of agricultural drainage water into the river water. The EF was used to distinguish between heavy metals caused by human activities and those resulting from natural formation (Fig. 11). The results indicate that all metals are deficient to minimally enriched in Euphrates River sediments. (EF < 2), with the exception of samples of cadmium, which showed a high level of enrichment (EF < 34) in all stations.   Ni showed a moderate level of enrichment ($2 \leq EF < 5$). The Igeo was used to evaluate the stages of pollution of heavy metals in sediment samples for the study area (Fig. 11). The results for the studied sediment samples of the Euphrates river showed the values of the averages of the geochemical accumulation factor for Cd (1.722), Cr (-0.852), Cu (-0.686), Ni (0.398), Pb (-0.66733), Co(-0.21), Zn (-1.182), and Fe(-0.79533).
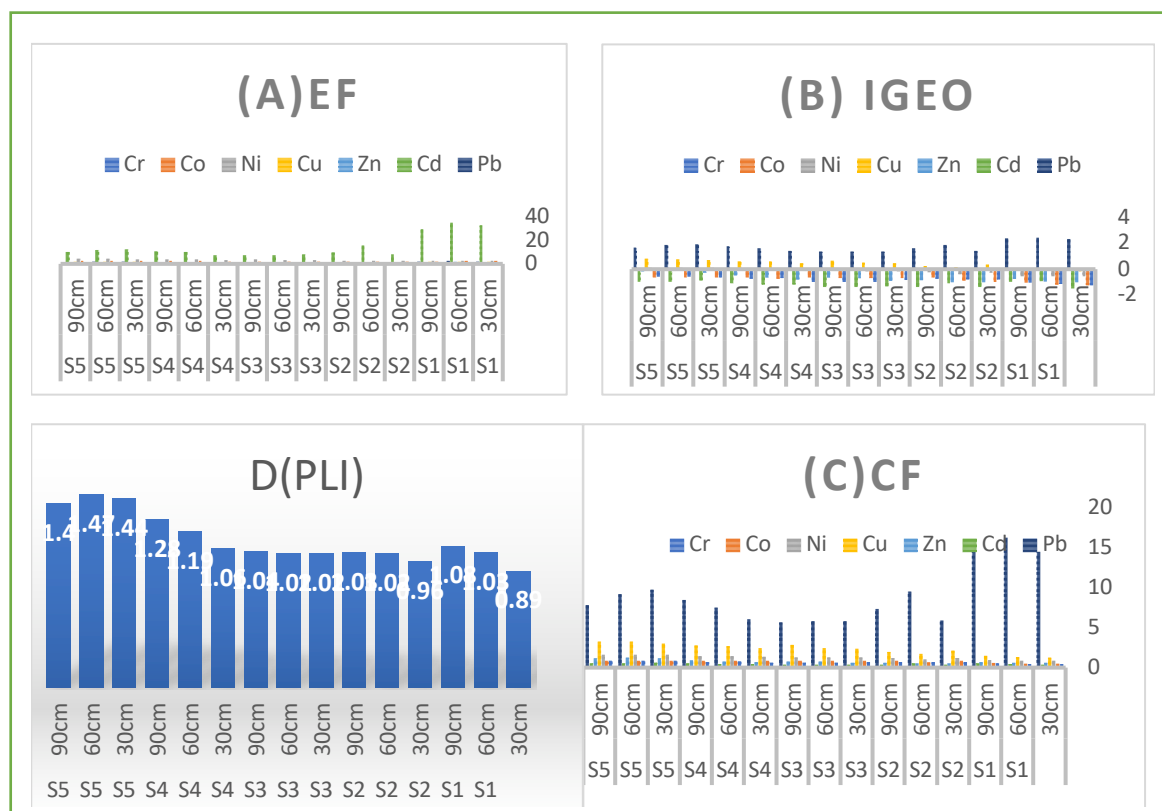

Fig.11. EF, Igeo, CF and PLI for sediment samples of the Euphrates river

The results indicate that the sediment samples for the Euphrates River are not contaminated with the following elements: Cr, Cu, Ni, and Zn, while lead contamination belongs to the unpolluted to moderately polluted category. In the case of Cd, Ni and Co, they belong to the heavily polluted category. The accumulation of heavy metals in sediment samples of the Euphrates River may be due to various human activities. The PLI evaluates the stage of total contamination in sediment samples for the study area. Fig.11 shows the results of the PLI for the eight metals studied in sediment samples, with a mean

value of 1.128667 and a range between 0.89 and 1.47. Based on the results presented, the degree of contamination by the eight metals in all sediment samples of the Euphrates River is classified as class 2 (deterioration on site quality), which indicates that for the studied sediment samples the Euphrates River is contaminated.

4. Conclusions

The increase of the concentrations of the studied elements in the sediments of the Euphrates River in the studied area is due to the rock origin of the elements in addition to the factors of erosion, weathering, and torrents. The continuous or escalating increase in the concentrations of heavy elements from the city of Heet to the city of Fallujah is attributable to the movement of the river and the torsions that exist along the river and the elements carried by the river's water of elements that may be gradually deposited. This is in addition to the human waste that is thrown into the river without any prior treatment. The results of the study showed that iron, nickel, cobalt, chromium and copper, which are natural in origin, increase vertically towards the lower layers. As for cadmium, lead and zinc, which are both natural and human sources, they increase vertically towards the upper layers, affected by human activities that cause pollution. Enrichment factor for assessing pollution showed that most of the stations have low to medium pollution. This indicator proves the absence of human activities in the industrial field in that time period.

Acknowledgements

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