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RESEARCH ARTICLE

Sources, Distribution and Ecological Risk Assessment of Polycyclic Aromatic Hydrocarbon in Sediment of Tigris River, Iraq

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ABSTRACT

Sediment samples were collected from five sites at Tigris River from Baghdad to Wasit Governorates River from December 2021 to November 2022. Sixteen polycyclic aromatic hydrocarbons (PAHs) in these sediment samples were analyzed. The results show that the total PAH concentrations ranged from 957.37 to 1805.05 μg/kg dry weight (dw) with mean values of 1368.034 μg/kg. Furthermore, the high molecular weight compounds (four, five, and six rings) of PAH in the analyzed sediment samples suggest that combustion sources such as fuel, gasoline, and vehicle emissions are the primary sources of PAHs in the environment. The sediment quality guidelines (SQG) based on the effects range low (ERL) and the effects range median (ERM) and risk quotient (RQ) were used to evaluate the ecological risks of polycyclic aromatic hydrocarbons (PAHs). The results showed that the risks were low to moderate, with potential biological effects of PAHs. The result of the current research work indicated that ongoing monitoring of PAH levels in sediments is needed.

Keywords: PAHs, Pollution, Risk quotient, Sediments, Tigris river

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are persistent organic contaminants often present in the environment and have two or more fused aromatic rings. [1–3] Because of their high persistence and moderate vapor pressure, the contaminants have the potential to travel long distances via the atmosphere and water, having a substantial impact on the ecosystem. By bioaccumulation and biomagnifying in food chains, the highly carcinogenic and mutagenic chemicals can also seriously threaten human health. [4,5]

The United States Environmental Protection Agency (USEPA) has classified sixteen PAHs as priority pollutants. It can be divided into two sets. Low molecular weight compounds (LMWC) are compounds with two to three rings, such as naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant) and fluoranthene (Fla). Second, the high molecular weight compounds (HMWC) with four or more rings, such as pyrene (Pyr), benzo(a)anthracene (BaA), crysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno[123-cd)pyrene (IcdP), dibenzo(a,h)anthracene (DahA) and benzo(g,h,i)perylene (BghiP). [6]

Most PAHs can be absorbed by particulate organic substances and eventually buried in the sediments, or they can be passed to biological organisms over an extended period of time due to their low aqueous solubility and high octanol-water partition coefficient (Kow). [7]

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The origins of PAHs might be anthropogenic emission sources and natural emissions. Organic matter decomposition and eruptions of volcanoes are illustrations of natural resources. In addition, some algae and plants can create PAHs in tiny amounts, and they are not considered sources of pollutio.[8]

Furthermore, the primary human sources of PAHs include fossil fuel burning, industrial processes, and vehicle exhaust.[9] They are generally insoluble in water and have slow migration, persistence and lipophilicity. As a result, they frequently build up and remain in the sediment for a long time. Compounds' molecular weight rises with an increase in benzene rings. This increases their toxicity while decreasing their volatility and biodegradability.[10,11] In sediment, PAHs are quickly accumulated, similar to other organic components in aquatic environments due to their hydrophobicity, low water solubility, and vapor pressures.[12,13] This implies that sediments are one of the most significant repositories of environmental contaminants. When PAHs reach the aquatic environment, they preferentially adsorb onto particles and accumulate in sediments due to their hydrophobic and persistent character.[14,15] Given that sediment serves as a major sink for most contaminants and influences the health of benthic species. This suggests that riverbed sediments are a valuable historical indicator of water pollution in a given river and its surrounding watershed throughout time.[16] Hence, the current study aims to look into (I) the concentration of PAHs, distribution, and levels of pollution in the Tigris River in the sediments, (II) pinpoint possible sources of these pollutants, and (III) determine the level of environmental hazards in these areas.

Materials and methods

Study area and sampling

Five sites were chosen on the Tigris River from Baghdad city (Al-Dura) to Al-Kut (Al-Suwaira, Al-Aziziyah, Al-Zubaidiya and Kut dam). The first site is located to the southeast of Baghdad. This site is influenced by many industrial activities on the river's bank, part of which belongs to the government sector and other parts to the private sector, like Al-Dura Power Station. The second site is located in eastern Iraq in the Wasit Governorate, about 35 km south of Baghdad, the capital of Iraq. It is located on the west bank of the Tigris. The third site is located on the left bank of the Tigris River and is part of Wasit Governorate. It is surrounded by several villages with rich land about 80 kilometers northwest of Kut. The fourth site is situated 75 km north of Kut, Wasit, and 93 km south of Baghdad;and the fifth site is located in Al-Kut city in Wasit Province. It is considered one of the longest dams in Iraq and is one of the important dams because of its impact on water storage and is sufficient to produce electricity.

Table 1. Geographical positioning system (GPS) of the selected sites.

	Site	Geographic coordinates		UTM coordinates	
		Longitude	Latitude	N	E
1	Al-Dura	44°25'19.16"	33°17'5.26"	3683008	446178
2	Al-Suwaira	44°47'34.67"	32°55'31.35"	3643033	480643
3	Al-Aziziyah	45° 3'49.86"	32°54'5.55"	3640373	505971
4	Al-Zubaidiya	45°10'32.42"	32°45'52.63"	3625206	516453
5	Kut dam	45°49'3.52"	32°29'55.24"	3596007	576811

The Global Positioning System (GPS) was used to determine the geographic coordinates (longitude and latitude) of the selected sites Table 1 and Fig. 1. Sediment samples were collected monthly from five sites on the Tigris River from December 2021 to November 2022. Using an Ekman Grab sampler, sediment samples were obtained, wrapped in aluminum foil, and stored at -20°C until analysis.[17] A total of (60) sediment samples were used in this study.

PAH extraction and analysis

PAHs were extracted from sediment samples using methods that have been described.[18] 3 g of dry and homogenized sediment sample was put into a clean centrifuge tube containing 20 mL of 5:5 (v/v) acetone/n-hexane. Blanks were prepared following the same procedure without adding sediment samples. All samples were vortexed for 1 min, and the mixture was subjected to ultrasonic treatment for 15 min for PAH extraction. The sample tubes were then centrifuged at 4,000 rpm for 10 min. After centrifuging, the organic layer containing the extracted compounds was siphoned out with a pipette, and the sediment was re-extracted twice with 2:2 (v/v) acetone/n-hexane (10 mL). The extraction of PAHs from sediment was analyzed using gas chromatography (Shimadzu, 2010, Japan), column oven (SE-30m) = 150 °C (hold 1 min.) -290 °C (10 °C /min) temperature of injector = 280 °C, temperature of detector (FID) = 310 °C, pressure100KPa, injection volume = 1u l) which was used to detect PAHs in the Ministry of Science and Technology laboratories. All chemicals used were of analytical grade and were used as received without any further purification;they were obtained from Sigma-Aldrich, Bio solve France and Supelco-German. All solutions were prepared with deionized water.

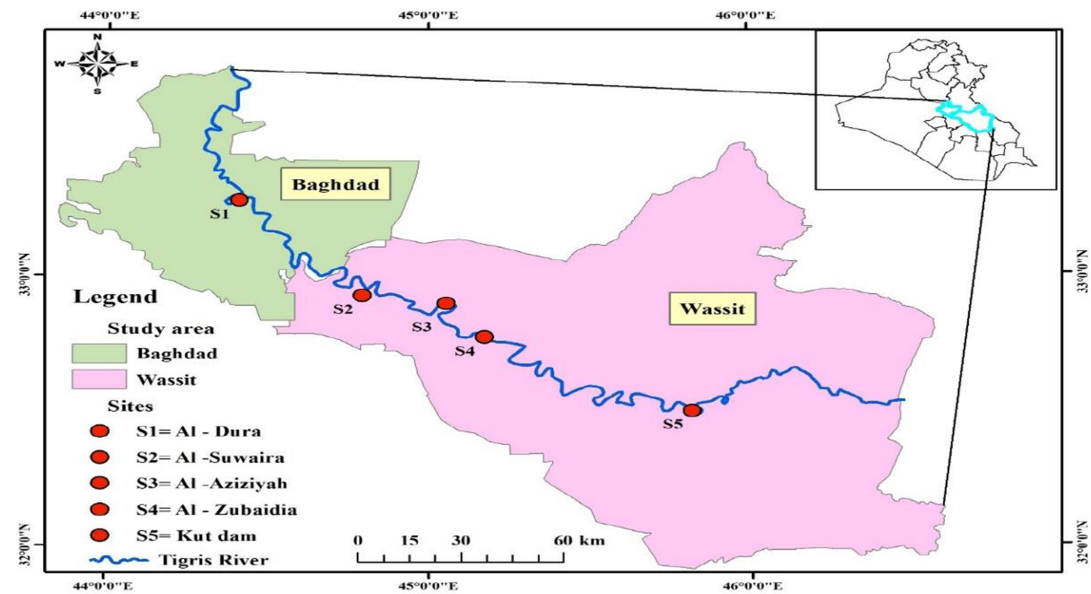

Fig. 1. Sampling sites along tigris river in Iraq.

Table 2. Diagnostic ratios for PAH compounds.[22,23]

Diagnostic ratios	Origin of PAH		
	Pyrogenic	petrogenic	Petrogenic or Pyrogenic
LMW/HMW	<1	>1	–
BaA/(BaA + Chr)	>0.35	<0.2	0.2–0.35
Flu/(Flu +Pyr)	>0.5	<0.4	0.4–0.5
Phe/Ant	<10	>10	–

Determining the PAH source

The primary sources of PAHs include incomplete combustion and industrial processes in various industrial operations, including producing iron and aluminum, cement, asphalt, dyes, waste incineration, vehicle emissions and other human-caused activities.[19] The ratios of specific PAH compounds can be used to determine the sources of PAHs, whether they originate from the burning of fuel pyrolytic or the contamination of crude oil (petrogenic), based on changes in PAH composition and distribution as a function of the emission source. Specific PAH diagnostic ratios were used to distinguish PAHs from petrogenic and pyrogenic sources Table 2. This study used LMW/HMW ratios, BaA/(BaA + Chr), Flu/(Flu+Pyr), and Phe/Ant to identify PAH sources.[20,21]

Ecological risk assessment of PAHs

Sediment quality guidelines (SQGs) were applied in this study to assess the potential negative impacts of PAHs on organisms.[22] The impacts of these pollutants on the aquatic environment are estimated by the SQGs using two criteria: effects range median (ERM) and effects range low (ERL).[23] Three chemical concentration categories found in the SQGs describe the varying degrees of detrimental chemical effects on biology: minimal effects with rare biological effects (<ERL), possible effects with occasionally biological effects (≥ERL and <ERM), and probable effects with frequent biological effects (≥ERM)[23] Table 3.

Risk quotient (RQ)

This study used risk quotients (RQs) to assess the ecological risk of PAHs in sediments. The quality values for negligible concentrations (NCs) and maximum permissible concentrations (MPCs) of PAHs in the media. Risk quotients (RQs) were used to quantify the risk levels posed by specific PAHs. RQs were calculated as follows[24,25]:

$$\mathrm{RQNCs}_{\mathrm{NCs}} = \frac{CPAHs}{CQV\,(NCs)}$$

$$\mathrm{RQ}_{\mathrm{MPCs}} = \frac{CPAHs}{CQV\,(MPCs)}$$

Where: cPAHs are the real concentration of a particular PAH in sediment, and CQV is the corresponding quality value of a certain PAH in the medium, Maximum permitted concentrations, or “MPCs,” are the quantities above which detrimental impacts on the ecosystem are undesirable, whereas negligible concentrations, or “NCs,” are the values below which

Table 3. Total PAH concentrations (μg/kg) in tigris river sediment samples, as well as the values of ERL and RRM for PAH_S (μg/kg dry weight) according to the sediment quality guidelines (SQG_S).

								SQGs	
PAH	No.of rings	Site 1	Site 2	Site 3	Site 4	Site 5	Average	ERL*	ERM*
Ace	3	143.5	83.35	86.9	123.0	131.0	113.55	16	500
Acy	3	115.77	56.75	60.11	97.12	102.19	86.388	44	640
Ant	3	127.17	79.55	71.85	104.80	118.25	100.324	85.3	
								1100	
BaA	4	118.19	52.05	55.43	86.21	96.35	81.646	261	
								1600	
BbF	5	131.90	72.16	72.53	114.90	122.20	102.738	320	
								1880	
BkF	5	152.97	91.63	93.59	124.40	140.02	120.522	280	
								1620	
BghiP	6	124.73	62.97	69.02	101.55	113.80	94.414	85	
								1600	
BaP	5	102.14	50.15	55.10	82.35	91.10	76.168	430	160
Chr	4	125.03	63.84	64.30	97.09	113.20	92.692	384	
								2800	
DahA	5	133.01	72.39	74.73	107.72	120.95	101.76	63.4	260
Fla	4	139.13	80.29	83.57	125.30	134.05	112.468	600	
								5100	
Flu	3	96.55	57.02	56.77	74.02	86.14	74.1	19	540
IcdP	6	76.72	41.95	44.05	64.80	70.89	59.682	240	—
Nap	2	83.47	37.98	42.25	62.65	74.79	60.228	160	
								2100	
Phe	3	77.17	29.85	34.52	48.85	67.58	51.594	240	
								1500	
Pyr	4	57.60	25.44	27.14	38.11	50.51	39.76	656	
								2600	
Σ_{16}PAHs		1805.05	957.37	991.86	1452.87	1633.02	1368.03		
ΣLMW		643.63	344.5	352.4	510.44	579.95			
ΣHMW		1161.42	612.87	639.46	942.43	1053.07			

*Effects range low (ERL), and effects range median (ERM).

unfavorable effects are minimal.[26] When RQ_{MPC} is greater than 1.0, it means that a PAH is associated with a serious risk, whereas its RQ_{NC} of less than 1.0 suggests that it can be ignored. The PAH of interest may provide moderate threats to the ecosystem, according to RQ_{NCs} greater than 1.0 and RQ_{MPCs} less than 1.0. While in a condition of $RQ_{\sum PAHs}$, values are interpreted as $RQ\sum_{NCs} \geq 800$ and $RQ\sum_{MPCs} \geq 1$ high risk, $1 \leq RQ\sum_{NCs} < 800$ and $RQ\sum_{MPCs} < 1$ low to moderate risk, and $RQ\sum_{NCs} < 1$ very low risk.[27,28]

Results and discussion

Distribution profiles and concentrations of PAHs in the sediment samples

PAHs were detected in all sediment samples in the study area. Concentrations of our target PAHs in sediment samples in this study are summarized in Table 3. The study showed that the concentration of Σ16PAHs in sediment samples ranged from 957.37 to 1805.05 μg/kg dry weight (dw) with an average of 1368.03 μg/kg. The highest concentration of Σ16PAHs in sediments was detected at S1, followed by S5, S4, S3 and S2 Fig. 2 and Table 3.

The average concentration of PAHs was in the order: BkF > Ace > Fla > BbF > DghA > Ant > BghiP > Chr > Acy > BaA > BaP > Flu > Nap > Inp > Phe > Pyr.

The results showed that BkF (5-ring) had the highest concentration in all sites, averaging 120.52 μg/kg. Because benzo(k)fluoranthene (B(K)F) has a high molecular weight (HMW) (5 ring), in which the HMW-PAHs are predominant with high concentrations constituted most of the sediment compounds, the high concentration of Bkf together with other HMW PAH species is indicative of recent, recurrent and widespread industrial pollution from pyrogenic sources. Benzo (k)fluoranthene is primarily present in the combustion byproducts of coal and oil, lubricating lubricants, old motor oils, coal tar, and cigarettes.[29] High molecular weight PAHs (HMW PAHs, four, five, and six-ring) constituted most of the sediments' compounds. Compared to low molecular weight (LMW) PAHs, they may have higher partitioning

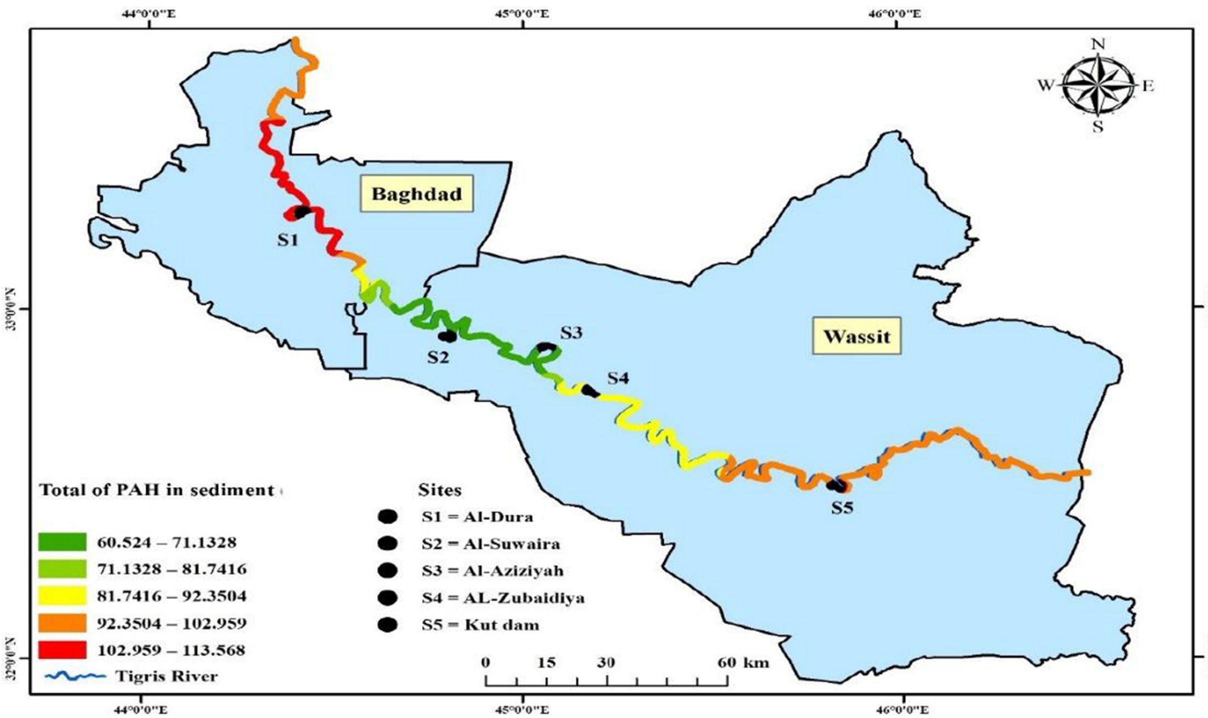

Fig. 2. Total PAHs (μg/kg) in the sediment of the current study.

coefficients in aqueous conditions, low solubility, and high hydrophobicity.[30] The average amounts of $\sum$LMW and $\sum$HMW PAH in the sediment were 579.95 and 1053.07 μg/kg, respectively Table 3.

The total concentration of PAHs in the current study is higher than the results of other studies Table 4. For example, total concentrations of PAHs in sediments from Kizilirmak River ranged from 43.15 to 386.115 μg/kg[31] and from Tigris River were between 24-1,222 μg/kg.[32] However, total PAH concentrations in this study were lower than the concentration of PAH in Delaware River, USA (3749 to 22,324 μg/kg).[33]

Emission sources of PAHs

In general, polycyclic aromatic hydrocarbons provide important information on where PAHs originate and how they affect the environment. The two primary pathways via which petroleum and refined products are generated, utilized, and transported are known as petrogenic pathways and pyrolytic pathways, which involve human-caused combustion of biomass and fossil fuels.[42] Comparing the ratios of LMW PAH compounds that contain 2-3 rings, such as Nap, Any, Ace, Flu, Phe, and Ant, to HMW of PAH compounds that contain 4-6 rings, such as Fla, Pyr,

Table 4. Comparing the levels of polycyclic aromatic hydrocarbons (PAHs) in sediments from around the world.

Study area	Concentration of ΣPAHs (μg/kg) dry weight	References
Delaware River, USA	3749 to 22,324	Kim et al.[33]
Suez Gulf, Egypt	1667.02–2671.27	Younis et al.[34]
Tigris river, Iraq	633.23–778.28	Kalaf et al.[35]
Euphrates River, Iraq	0.36–119.06	Hassan et al.[15]
Kizilirmak River ,Turkey	43.15–386.115	Şimşek & Bilgili.[31]
urban river(Turag, Bangladesh)	45.8–1901	Khan et al.[36]
River Conwy Estuary, Wales, UK	18–1578	Vane et al.[37]
Imiringi River, Nigeria	0.01–3,965.4	Aigberua& Seiyaboh.[38]
River Owan, Edo State, Nigeria	0.280–0.810	Akinnusotu et al.[39]
Suez Bay, Egypt	9.1–981.10	Soliman et al.[40]
Tigris River, Iraq	24–1,222	Abed et al.[32]
Ifiekporo Creek in Warri,Nigeria	850 to 3470	Berezi et al.[41]
Tigris River, Iraq	**957.37–1805.05**	**The current study**

Table 5. Diagnosis ratios of PAH compounds in sediment samples.

Index	S1	S2	S3	S4	S5
ΣLMW/ ΣHMW	0.554	0.562	0.551	0.541	0.550
Flu/(Flu + Pyr)	0.626	0.691	0.676	0.660	0.630
BaA/(BaA + Chr)	0.485	0.449	0.462	0.470	0.459
Phe/Ant	0.606	0.375	0.480	0.466	0.571

BaA, Chr, BbF, BkF, BaP, DahA, BghiP, and IcdP, may help identify the source of PAHs.[43]

In this work, the sources of PAHs in sediment samples are determined utilizing the ΣLMW/ΣHMW, Flu/(Flu + Pyr)/BaA/(BaA + Chr), and Phe/Ant ratios Table 5. The results of the current study showed that the values of ∑LMW/HMW were less than one (∑LMW/HMW < 1), the ratios of Flu/(Flu + Pyr) > 0.5, BaA/(BaA + Chr) > 0.35 and the ratio of Phe/Ant at all sites, indicating that the PAH in S1, S2, S3, S4 and S5 come from a pyrogenic source Table 5, which included vehicle emissions, the burning of fuel and gasoline combustion.[20] Because of this, these ratios are easy to utilize and have emerged as a crucial tool for locating the origins of PAHs, as shown by the numerous researchers that have used them.[44,45]

Assessment of ecological risk

Several assessment tools, such as sediment quality guidelines (SQGs), are commonly utilized for a preliminary investigation and evaluation of the ecological risk that aquatic habitats confront. ERL classifications are based on chemical concentrations below for which there is little chance of toxicity or other negative effects. However, ERMs indicate the potential for toxicity and unfavorable effects. Additionally, there are occasionally negative effects shown by the values between ERLs-ERMs.[46]

The data collected for this investigation indicated PAH concentrations below ERM levels;however, not all PAH concentrations were lower for ERL values.

The toxicity guidelines and concentrations of 16 PAHs are presented in Table 3. Based on comparing with SQGs, the concentrations of Ace, Acy, DahA, and Flu in the sediments at all sites were found to be within a possible-effects range, with values above ERL and below ERM. This requires continuous monitoring to avoid increasing concentrations above ERM.[23]

In all sites, the concentration of BaA, BbF, BkF, BaP, Chr, and Fla in the sediment was less than that of ERL and ERM. This indicates the minimal effects range with rare biological effects (< ERL). While BghiP and Ant concentration were less than that of ERL and ERM in the sediment of S2 & S3, which indicate rare biological effects while having radical biological effects at different sites S1, S4 and S5 (≥ERL and < ERM).[23]

This study utilized risk quotients (RQs) to yield more accurate results.

The majority of individual PAHs were found to have mean values of RQ (NCs) greater than 1, displayed in Table 6., indicating medium risk except for Cry (0.86), suggesting that these chemicals pose no ecological damage to river sediments, but values of RQ (MPCs) < 1 for all individual PAHs, indicating moderate threats to the ecosystem. According to the calculation of the sum of 16 PAHs RQ (NCs) and RQ (MPC) and show that the range of ΣRQ_{NCS} values was 326-619.22 (≥1 < 800) and the range of $\Sigma RQ_{(MPCS)}$ values was 3.2-6,3 (> 1), indicating low to moderate ecological risk.[28]

Table 6. Individual PAHs' NCs and MPCs along with their mean RQ (NCs) and RQ (MPCs) values in the tigris river sediment for all sites.

			RQNCs		RQMPCs	
PAHS	NCs μg/kg	MPCs μg/kg	Range	Mean	Range	Mean
Ace	1.2	120	(69.45–119.58)	94.62	(0.69–1.19)	0.94
Acy	1.2	120	(47.29–96.47)	71.98	(0.47–0.96)	0.71
Ant	1.2	120	(59.87–105.97)	83.6	(0.59–1.05)	0.83
BaA	3.6	360	(14.45–32.83)	22.67	(0.14–0.32)	0.22
BbF	3.6	360	(20.04–36.63)	28.53	(0.20–0.36)	0.28
BKF	24	2400	(3.81–6.37)	5.02	(0.03–0.06)	0.05
BghiP	75	7500	(0.83–1.66)	1.25	(0.008–0.01)	0.012
BaP	27	2700	(1.85–3.78)	1.01	(0.01–0.03)	0.02
Chr	107	10700	(0.59–1.16)	0.86	(0.005–0.011)	0.008
DahA	27	2700	(2.68–4.92)	3.76	(0.02–0.04)	0.037
Fla	26	2600	(3.08–5.35)	4.32	(0.03–0.05)	0.04
Flu	1.2	120	(47.30–80.45)	61.75	(0.47–0.80)	0.61
IcdP	59	5900	(0.71–1.3)	1.01	(0.007–0.013)	0.01
Nap	1.4	140	(27.12–59.62)	43.01	(0.27–0.59)	0.43
Phe	5.1	510	(5.85–15.13)	10.11	(0.05–0.15)	0.1
Pyr	1.2	120	(21.2–48)	33.13	(0.21–0.48)	0.33
RQΣPAH			(326–619.22)	466.63	(3.2–6.3)	4.63

Conclusion

Our results revealed that most of the PAHs measured in sediment samples were large molecules of PAHs with five or six rings in their structure. The Tigris River system has been shown to contain increased levels of PAHs, particularly carcinogenic chemicals, which may have ecological effects. According to the present results, PAH diagnosis ratio pyrogenic origins were the source of the PAHs found in the study area's sediments.

The primary sources of PAHs in the sediment were pyrogenic sources, including sources of combustion such as coal, fuel, and gasoline. According to the risk assessment, the concentration of numerous individual PAHs at several sites was above and below ERM, which occasionally negatively affected the ecosystem.

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Authors' declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- No animal studies are present in the manuscript.
- No human studies are present in the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee at University of Baghdad.

Authors' contribution statement

Sh. R. Z. handled the sample preparation, sample collection, chemical analysis, and authoring of the final manuscript. A. M.J. and M. H. m. the data interpretation and processing. Every author discussed the results and added them to the finished manuscript.

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مصادر وتوزيع وتقييم المخاطر البيئية للمركبات الهيدروكربونية الأروماتية متعددة الحلقات في رواسب نهر دجلة، العراق

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المستخلص

تم جمع عينات الرواسب من خمسة مواقع على نهر دجلة من بغداد إلى محافظة واسط للفترة من ديسمبر 2021 إلى نوفمبر 2022. تم تحليل ستة عشر هيدروكربون اروماتي متعدد الحلقات (PAHs) في عينات الرواسب. أظهرت النتائج أن التركيز ألأجمالي للهيدروكربونات الاروماتية متعددة الحلقات تراوح من 957.37 إلى 1805.05 ميكروغرام / كغم (وزن جاف) وبمتوسط حسابي مقداره 1368.034 ميكروغرام/ كغم (وزن جاف). علاوة على ذلك، أشارت نتائج الدراسة المتعلقة بالمركبات ذات الوزن الجزيئي العالي (أربع وخمس وست حلقات) من الهيدروكربونات الاروماتية متعددة الحلقات في عينات الرواسب إلى أن مصادر الاحتراق مثل الوقود والبنزين وانبعاثات المركبات هي المصادر الأساسية للهيدروكربونات الاروماتية. تم استخدام المبادىء التوجيهية لنوعية الرواسب (SQG) بالاعتماد على قيم RQ, ERL, ERM في تقييم المخاطر البيئية للمركبات الهيدروكاربونية الأروماتية، حيث اظهرت النتائج ان المخاطرالبيئية تراوحت بين منخفضة الى معتدلة مع تأثيرات بايولوجية خطره وهذا يتطلب الى استمرار في مراقبة مستويات المركبات الهيدروكاربونية الأروماتية في الرواسب.

الكلمات المفتاحية: الهيدروكربونات الاروماتية متعددة الحلقات، التلوث، حاصل الخطورة، الرواسب، نهر دجلة.