Study of hydrocarbon compounds levels in water, sediments and some aquatic biota in Al-Mushrah river in Misan province/ Iraq

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Abstract :

In this study levels and sources of total petroleum hydrocarbon compounds (TPHs) and Polycyclic aromatic hydrocarbons (PAHs) were estimated in water , sediments and some aquatic biota which used as a bio-indictor to assess the pollution status of the water in Al-Mushrah river in Misan province.TPHs concentrations in water samples ranged from 0.759 to 2.476 µg/l ,whereas in sediment ranged from 1.55 to 3.408 µg/gm dry weight in station 1 and station 2 respectively .TPHs levels in plant species ranged from 4.222 to 13.985 µg/gm d.w in *Phragmits australis* and *Ceratophyllum demersum* respectively and in fish muscles ranged between 1.765 µg/gm d.w and 92.592 µg/gm d.w in *Liza abu and Barbus luteus* respectively ,while in tissues of *Unio tigridis* were 13.18 µg/gm d.w. Concentrations of total PAHs in water samples ranged between 18148.43 and 155416.327 ng/l ,whereas in sediments ranged between 10866.47 and 44417.53 ng/gm in St1 and St2 respectively .The levels of low molecular weight PAHs(LPAHs) in water and sediment samples were less than high molecular weight PAHs (HPAHs). Sources of PAHs in water samples were pyrogenic origin, whereas in sediments samples were petrogenic. Total PAHs recorded in plant species ranged between 18532.11984 ng/gm and 123631.6954 ng/gm in *P. australis* and *C. demersu*m ,sources of PAHs in plants were petrogenic and in fish samples total PAHs ranged from 51161.66 to 358080.1 ng/gm in *L. abu* and *B. luteus* respectively, the fish samples in study area were very highly contaminated with PAHs according to Baumard *et al* (1998) classification ,sources of PAHs were in *B. luteu*s were pyrogenic ,whereas in *L. abu* and *Cyprinus carpio* were petrogenic , while total PAHs in *U. tigridis* were 152462.93 ng/gm and sources of PAHs in *U. tigridis* were pyrogenic. High levels of TPHs and PAHs in studied aquatic biota observed in this study suggests that these organisms can be used as a tool for bio-monitoring of hydrocarbon pollution in study area .

Introduction:

The petroleum hydrocarbon compounds (TPHs) are environmental contaminants that are released into the aquatic environment through different routes .They are absorbed and Bio-accumulated by aquatic organisms via food ,water and sediments (Neff ,1985 ; Mashinchian *et al* .,2012). The control on oil pollution problem in the aquatic ecosystem is very difficult because of the large number of input sources (Patin , 1999) . The sources of hydrocarbon compounds in the aquatic ecosystem are either as anthropogenic or as biogenic. Major sources of these chemical compounds enter into water by many routes of different activities like oil refineries, transportation and tanker accident , off shore oil production ,urban runoff and municipal wastes(Maktoof *et al* ., 2014 ; Al-Saad *et al* ., 2000). PAHs are pose high environmental stability, high toxicity, high hydrophobicity and high affinity to fatty tissues of organisms and cause the acute lethal and sub-lethal toxic effect in freshwater organisms so that accumulation through the trophic chain with final destination the human body. (Salman *et al* ., 2014 ; Warren *et al* .,2003).The organisms exposure to Petroleum compounds through sediments ,respiration , dermal exposure or consumption of contaminated prey such as Mollusca (Varanasi *et al.*, 1989). Okpashi *et al* .(2017)and many studies showed that aquatic organisms can accumulate PAHs compounds from ambient. The present study aims to assess the TPHs and PAHs in the water, sediments and some aquatic biota as a bio-indicator for oil pollution of Al-Musharah river in Misan province and this data would serve as baseline information on the extent of contamination for management purpose to diluting the detrimental inputs of hydrocarbon compounds to the study area.

Materials and methods:

Collection and preparation of samples

Water and sediment samples were collected during December 2016 from two selected stations. Water samples were collected by using dark glass bottles ,whereas the sediment samples were taken by using Van veen grab sampler and stored in aluminum foil then placed in ice-packed and transferred to the laboratory then kept in the refrigerator before analysis.

The plant species *Phragmits australis* and *Ceratophyllum demersum* (Al-Saadi and Al-Mayah,1983) were collected by hand, washed in the river water and rinsed thoroughly with distilled water in the lab then were cut to small parts then dried, grounded and sieved by using a 63 µm metal sieve then placed in clean glass vial to become ready for analysis. Fish species *Liza abu ,Barbus luteus and Cyprinus carpio* (Coad ,2010)and bivalve molluscs species *Unio tigridis* (Ahmed ,1975)were collected by using the trawl net, washed thoroughly in the river water and were placed in aluminum foils before being transferred to the lab ice-packed. The samples were rinsed thoroughly with distilled water in the laboratory the muscles were cut to small parts and dried ,grounded and sieved then placed in clean glass

vial to become ready for analysis (Talal ,2008). The molluscs tissues were removed from the shell with the help of a clean ,sterile steel pin then dried. They were then grounded and sieved and placed in clean glass vials to prepare for analysis (Farid ,2007).

Extraction of hydrocarbon compounds from samples

The procedure of UNEP(1989)was used for the extraction of hydrocarbon compounds from water samples **.**The procedure of Goutex and Saliot(1980) was used for the extraction of hydrocarbon compounds from sediments , whereas the procedure of Grimalt and Oliver (1993) was used for the extraction of hydrocarbon compounds from aquatic biota species.

PAHs sources

The PAHs sources was assessed according to the following ratios :

a-Ratio of Low Molecular Weight to High Molecular Weight (LPAHs /HPAHs).

Values less than one are pyrogenic sources , while values more than one indicates petrogenic sources from crude oil and their derivatives (Vrana *et al.*, 2001) .

b-Phenanthrene / Anthracene ratio (Phe /Ant)

The ratio values higher than one the hydrocarbons are petrogenic in origin, whereas the ratio values less than one show that the hydrocarbons are pyrogenic in origin (Beg *et al*., 2009).

c- Fluoranthene to Pyrene ratio (Flu/Pyr) .

The ratio less than one are attributed to petrogenic sources and the ratio more than one have been used to indicate pyrogenic sources (Qiu *et al* .,2009).

Analysis of samples

TPHs concentrations in samples was determined by using Spectrofluorometer , whereas PAHs concentrations in samples were determined by using HPLC instrument type shimadzo at department of marine environmental chemistry, Marine Science Center, Basrah University .

Results and discussion :

1-TPHs in water and sediments

The concentrations of TPHs in water samples in figure 1 which ranged from 0.759 to 2.476 µg/l in station 1 and station 2 respectively. While in sediment samples in figure 2 ranged between 1.55 and 3.408 µg/gm dry weight in St1 and St2 respectively. The levels of hydrocarbon compounds in sediments were more than water in all stations ,this due to the tendency of these compounds to adsorb on the suspended particulates in water then accumulated in sediments from anthropogenic and natural emissions . Also the death of aquatic plants and phytoplankton which lead to increase hydrocarbons levels in sediments (Qiu *et al* .,2009 ; Jazza, 2015) .The present study revealed that there were a spatial variations of TPHs in both water and sediments ,the highest levels of TPHs were

recorded in St2 may be attributed to the reception of high amounts of agricultural wastes and domestic wastes which contain on high amount of organic matters that come from Al-Musharah city because the river flow through agriculture and urban industrial area carry a great amount and variety of pollutants as well as fishing boats (Al-Khatib ,2008 ; Maktoof *et al* .,2014 and Jazza ,2015) .

2- TPHs in aquatic biota species .

a- TPHs in plants

TPHs concentrations in plant species ranged between 4.222 and 13.985 µg /gm .dw in *P.australis* and *C.demersum* respectively (figure,3) ,from results of the present study it could be noticed that there was a difference in the levels of TPHs in different species of aquatic plants ,these variations perhaps attributed to eliminate certain pollutants to the environment or the different abilities of plants to accumulate , the accumulation processes of pollutants may depend on many chemical and physical properties such as concentration of nutrients , dissolved oxygen , salinity, pH and temperature (Al-Saad ,1994 ; Jazza ,2015). In addition to that the variation concentrations of these pollutants between plants species may be return to their ability to absorb the pollutants from water and sediment (Hasssan *et al* ., 2016).

b- TPHs in fishes

Results of this study revealed that TPHs in *B.luteus* had the highest levels 92.592 µg/gm d.w ,followed by *C. carpio* 3.934 µg/gm d.w and *L.abu* 1.765 µg/gm .d.w (figure,4) , this variation in concentrations of TPHs among different species was attributed to genetic makeup, age, exposure way to contaminants, health of the fish ,physiology factors, nutritional status, tissue size and lipid content (Okpashi *et al* ., 2016) . Fish may take up TPHs contaminants from water through diffusion across their gills then accumulate these compounds in their tissues (Meador *et al.*,1995).

c- TPHs in bivalve molluscs

The levels of TPHs in *U. tigridis* were 13.18 µg/gm d.w which illustrated (figure,5) ,this concentrations may be attributed to the content of lipids as many studies conducted with mollusca species in different regions indicated that the accumulation of hydrocarbons in their tissues increased on the increase of their lipid content (Allen *et al* ., 2002 ; Farid ,2007 and Jazza ,2015). Biotransformation of hydrocarbon compounds in mollusca and other aquatic organisms occurs to varying degrees depending on many factors such as the rate of uptake, physical condition like temperature , metabolic capability, age and feeding habits (Varanasi *et al.*, 1989) in addition to that generally all mollusca have lower hydrocarbon petroleum compounds metabolizing capability compared to aquatic species such as crustaceans, fish and polychaetes (Meador *et al.*,1995).

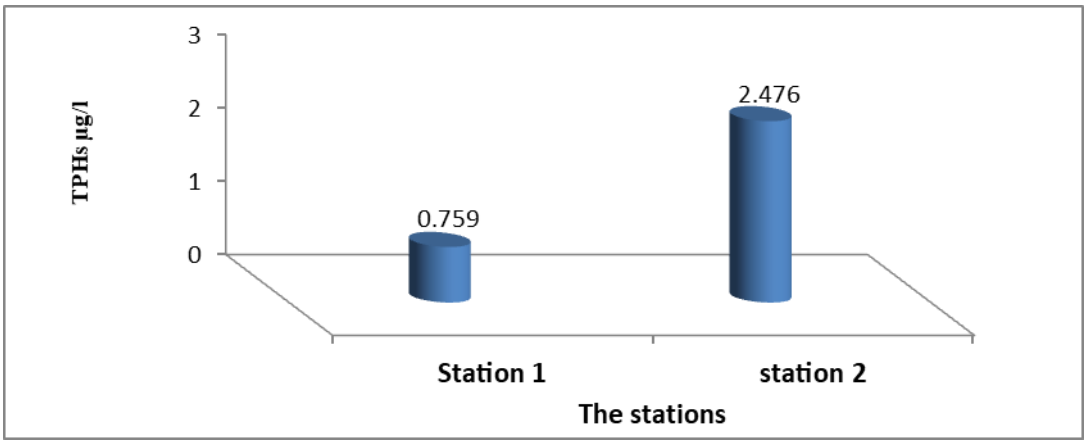

Figure (1) : concentrations of TPHs (µg /l) in water samples .

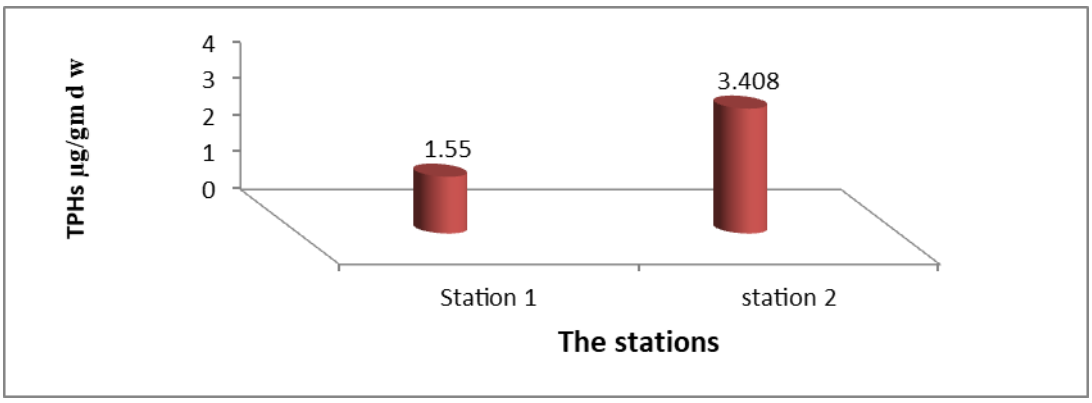

Figure (2) : concentrations of TPHs (µg /gm .d w) in sediments samples .

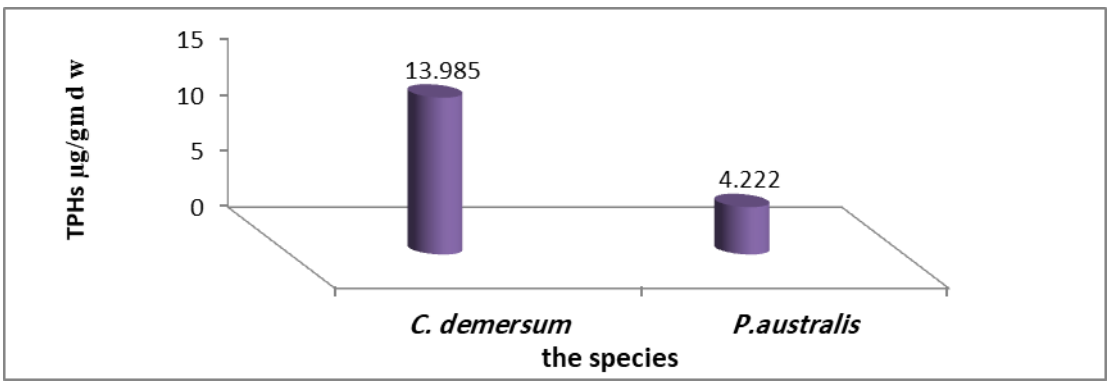

Figure (3) : concentrations of TPHs (mg /gm .d w) in plant species.

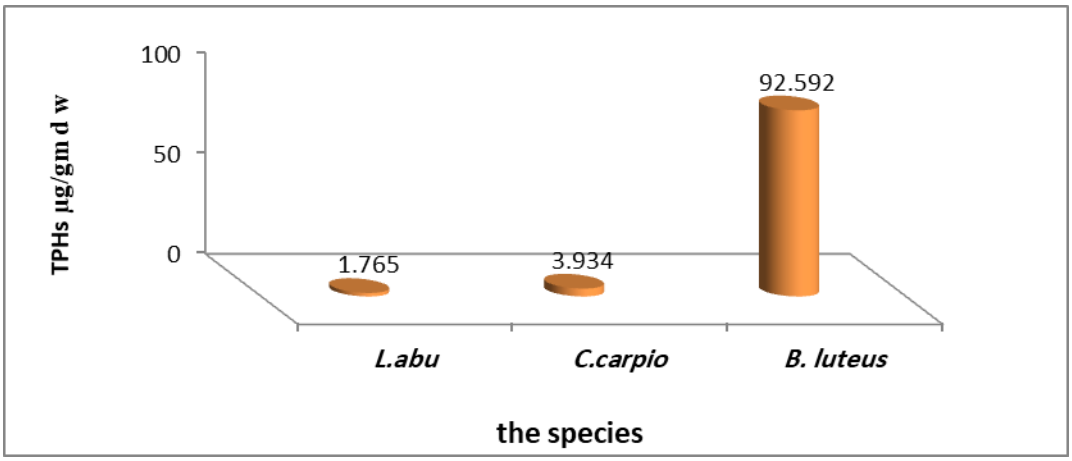

Figure (4) : concentrations of TPHs (µg /gm .d w) in fish species .

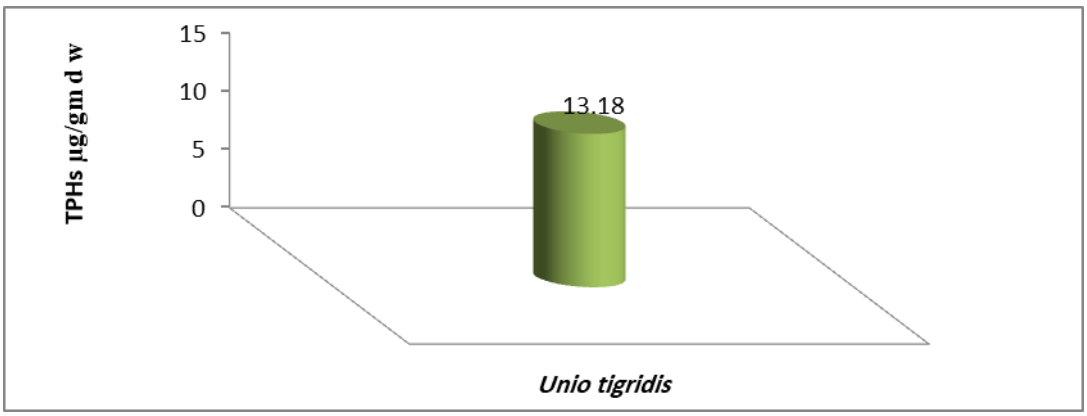

Figure (5) : concentrations of TPHs (µg /gm .d w) in *U. tigridis*.

3- PAHs in water and sediments.

The concentrations of total PAHs in water samples ranged from 18148.43 to 155416.327 ng/l in St1 and St2 respectively (table,1) ,whereas their concentrations in sediments samples ranged between 10866.47 and 44417.53 ng/gm in St1 and St2 respectively (table,2). The higher concentrations of total PAHs were recorded in water and sediments samples collected from St2 ,whereas lower concentrations are detected in St1 for both samples ,this variation in PAHs content may be due to the different sources of discharged waters from natural and human activities. These results may be attributed to large amounts of waste water contained PAHs was discharged into the river which caused the higher concentrations of total PAHs in station 2. However, according to Baumard *et al.* (1998) classification(contamination level of PAHs in sediments was very high when more than 5000 ng/gm) sediment samples in both station studied were very high contaminated with PAHs compounds. According to the results of the present study , the levels of low molecular weight PAHs(LPAHs) in water samples of the river

were lower than high molecular weight PAHs (HPAHs) in both stations (table, 2), concentration of LPAHs in water samples ranged between 6701.28 and 5793.232 in St1 and St2 respectively ,while HPAHs ranged between 11447.13 and 149623.1 in St1 and St2 respectively. LPAHs in sediments ranged from 10866.47 to 29483.171 in St1 and St2 respectively ,whereas HPAHs ranged between o and 14934.36 in St1 and St2 respectively (table, 2). HPAHs are often in surface water , whereas in sediment samples were dominated by LPAHs . The variations between PAH pattern in water and sediments perhaps due to molecular weight and bacterial degradation by microorganisms such as bacteria, fungi and algae especially bacteria play important role in complete mineralization (Mohammed *et al.*, 2009 ; Nasr *et al.*,2010). LPAHs were more degradable and soluble, whereas HPAHs are more recalcitrant (Obayori and Salam, 2010).To determined origin of PAHs according to LPAHs/HPAHs ratios were less than 1 in water samples for both stations which indicate that they derived from pyrogenic origin ,whereas in sediments LPAHs/HPAHs ratios were more than 1 in St2 (table,2) ,which indicate that they derived from petrogenic origins. The Flu/Pyr ratios in sediments samples were less than 1 in St2 (table,2) ,this indicated that source of PAHs in sediment is from petrogenic origins (Li *et al.*, 2006).

Table 1 :Concentrations of PAHs compounds in water and sediments samples.

Name of compound	Sample name			
	Water station 1 (ng/l)	Water station 2 (ng/l)	Sediment station 1 (ng/gm)	Sediment station 2 (ng/gm)
Naphtalene	ND	ND	ND	ND
Acenaphtylene	5599.90	2978.348	ND	25578.35
Acenaphthene	ND	2127.034	ND	ND
Fluorene	ND	ND	ND	1912.741
Phenanthrene	1101.38	687.85	10866.47	260.8061
Anthracene	ND	ND	ND	ND
Fluoranthene	ND	ND	ND	1731.272
Pyrene	ND	ND	ND	3114.801
Benzo[a]anthracene	ND	ND	ND	11819.56
Chrysene	ND	ND	ND	ND
Benzo[b]fluoranth	ND	ND	ND	ND

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Benzo[k]fluoranthene	706.70	796.594	ND	ND
Benzo[a]pyrene	ND	ND	ND	ND
Dibenzo[a,h]anthracene	10740.43	148826.499	ND	ND
Benzo[g,h,i]perylene	ND	ND	ND	ND
Indeno[1,2,3-c,d]pyrene	ND	ND	ND	ND
16 PAHs∑	18148.43	155416.327	10866.47	44417.53

ND: not detected

Table 2 : Sources of PAHs in water and sediment samples according to ratios of individuals PAHs.

The equation	Sample name			
	Water st 1	Water st2	Sediment st 1	Sediment st 2
Low PAHS ∑	6701.28	5793.232	10866.47	29483.17
∑High PAHs	11447.13	149623.1	0	14934.36
LPAHS/ HPAHs	0.585411	0.038719	0	1.974184
Phe /Ant	0	0	0	0
Flu/Pyr	0	0	0	0.555821

4- PAHs in aquatic biota species .

a- PAHs in plants

In the plant samples ∑PAHs shown variations in different species ,the highest concentrations recorded in *C. demersu*m (123631.6954 ng/gm whereas the lowest in *P. australis* (18532.11984 ng/gm) in table 3. These variation between plant species may be attributed to the different their ability to absorb the pollutants from sediment and water (Hassan *et al* ., 2016) or eliminate certain pollutants to the environment ,in addition to that the accumulation processes of these pollutants may depend on some physical and chemical factors such as temperature, pH, nutrients

and salinity (Al-Saad ,1994 ; Jazza ,2015) .Also this variations may be return to the contents of lipids in living cells which caused an increase of hydrophobic hydrocarbons in their tissues (Fisher ,1995).The LPAH concentration were dominated by HPAHs (table,4)in both studied species. To determined origin of PAHs according to LPAHs / HPAHs ratios in table 4 were more than 1in *P. australis* (1.986201) and in *C. demersu*m (10.00487) which indicated that they derived from petrogenic source (Vrana *et al.*, 2001) .

b- PAHs in fishes

In fish samples ∑PAHs shown variations in different species ,the highest concentrations recorded in *B. luteus* (358080.1 ng/gm), followed by *L. abu* (51161.66 ng/gm) and *C. carpio* (13892.94 ng/gm).This variations in accumulation of hydrocarbon compounds among different fish species were may be due to many factors may include age ,sex, health ,genetic makeup of the fish, amount contaminants, uptake rate, metabolic capability ,nutrition habit of the fish, lipid content and tissue size(Okpashi *et al* .,2017).According to Baumard *et al*. (1998) contamination of fish with PAHs could be classified based on measured concentrations into low contaminated (0-100), moderately (100-1000), highly (1000-5000) and very highly contaminated if greater than 5000 ng/gm ,thus based on this classification the fish sampled from Al-Musharah river could be said to be very highly contaminated (13892.94 -358080.1 ng/gm) with PAHs. HPAHs were less than LPAHs (table,4) in fish species *L. abu and C. carpi*o, whereas vice versa in *B. luteus* , the dominance of L PAHs in fish muscles had been due to the low breakdown of LPAHs and thus longer retention in fish muscles compared to HPAHs that were enzymatically metabolized (Okpashi *et al*.,2017) .The ratio of LPAHs /HPAHs in table 4 were less than 1 in *B. luteu*s (0.028898) which indicated that they derived from pyrogenic origin , whereas the ratio of LPAHs /HPAHs in *L. abu* (8.092585) and *C. carpio* (9.092448) was more than one and Phe /Ant ratio in *L. abu* (0.211) and *C. carpio* (0.047) in table 4 was less than one , which was indication of pollution origin from petroleum (Vrana *et al.*, 2001 ; Beg *et al*., 2009).

c- PAHs in bivalve molluscs

Concentrations of total PAHs in molluscs species *U. tigridis* were 152462.93 ng/gm (table,3).PAHs levels in *U. tigridis* more than in sediment because this animal is sediment dwelling and filter feeding. Thus, *U. tigridis* was more susceptible to contamination with PAHs compounds , in addition to that filter large volumes of water and have a low metabolic capacity for PAHs and then accumulate in their tissues (Baumard *et al* ., 1998). Also may be attributed to lipid content because many investigators found that there were a direct relationship between concentrations of PAHs and lipid content in the mollusca (Gold-Bouchot *et al*.,1995 ;Farid ,2007; Al-Saad *et a* .,2009 and Jazza ,2015).The continuous

release of PAHs to the river environment, can result in elevated concentrations in bivalves which living in the study area. Results of the present study showed that HPAHs were dominated by LPAHs (table 4),this was may be return to HPAHs which were more resistant to degradation processes compared with LPAHs ,in addition to that the mollusca were lack the Mixed Function Oxidase (MFO) system, therefore they unable to efficiently metabolize PAHs (Hylland ,2006).To determined sources of PAHs according to LPAHs/HPAHs ratios in table 4 were less than 1(0.433238) which indicated that they derived from pyrogenic source .

Tab 3 :Concentrations of PAHs compounds in species biota .

Name of compound	Sample name					
	P. australis	*C. demersum*	*B. luteus*	*L. abu*	*C. carpio*	*U. tigridis*
Naphtalene	**1604.590565**	**1177.75**	**395.9587**	**2684.135**	**ND**	**ND**
Acenaphtylene	**ND**	**103780.90**	**ND**	**ND**	**ND**	**46086.34**
Acenaphthene	**ND**	**ND**	**ND**	**14394.02**	**1730.783**	**ND**
Fluorene	**9987.790071**	**5200.11**	**9661.115**	**ND**	**ND**	**ND**
Phenanthrene	**733.8213007**	**1002.32**	**ND**	**4696.201**	**493.6913**	**ND**
Anthracene	**ND**	**ND**	**ND**	**22246.08**	**10291.9**	**ND**
Fluoranthene	**ND**	**1236.30**	**ND**	**1514.477**	**ND**	**ND**
Pyrene	**6205.917902**	**ND**	**ND**	**ND**	**1222.624**	**ND**
Benzo[a]anthracene	**ND**	**11234.27**	**ND**	**ND**	**ND**	**ND**
Chrysene	**ND**	**ND**	**ND**	**1815.442**	**ND**	**ND**
Benzo[b]fluoranthene	**ND**	**ND**	**ND**	**3811.303**	**153.9441**	**ND**
Benzo[k]fluoranthene	**ND**	**ND**	**3276.532**	**ND**	**ND**	**ND**
Benzo[a]pyrene	**ND**	**ND**	**1244.742**	**ND**	**ND**	**ND**

Dibenzo[a,h]anthracene	**ND**	**ND**	**343501.7**	**ND**	**ND**	**101179.60**
Benzo[g,h,i]perylene	**ND**	**ND**	**ND**	**ND**	**ND**	**5196.98**
Indeno[1,2,3-c,d]pyrene	**ND**	**ND**	**ND**	**ND**	**ND**	**ND**
16 PAHs∑	**18532.11984**	**123631.6954**	**358080.1**	**51161.66**	**13892.94**	**152462.93**

Tab 4 : Sources of PAHs in species biota according to ratios of individuals PAHs.

The equation	**Sample name**					
	P. australis	***C. demersum***	***B. luteus***	***L. abu***	***C. carpio***	***U. tigridis***
Low PAHS ∑	**12326.2**	**112397.4**	**10057.07**	**45534.91**	**12516.37**	**46086.34**
∑High PAHs	**6205.918**	**11234.27**	**348023**	**5626.745**	**1376.568**	**106376.6**
LPAHS/ HPAHs	**1.986201**	**10.00487**	**0.028898**	**8.092585**	**9.092448**	**0.433238**
Phe /Ant	**0**	**0**	**0**	**0.211**	**0.047**	**0**
Flu/Pyr	**0**	**0**	**0**	**0**	**0**	**0**

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دراسة مستويات المركبات الهيدروكربونية في المياه والرواسب وبعض الاحياء المائية في نهر المشرح في محافظة ميسان / العراق

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الخلاصة :

في هذه الدراسة قدرت مستويات ومصادر المركبات الهيدروكربونية النفطية الكلية (TPHs) والمركبات الهيدروكربونية الاروماتية المتعددة الحلقات(PAHs) في المياه والرواسب وبعض الانواع المائية التي استخدمت كدلائل حياتية لتقييم واقع التلوث في مياه نهر المشرح في محافظة ميسان. تراكيز مركبات TPHs في عينات المياه تراوحت من 0.759 الى 2.476 مايكروغرام / لتر ، بينما في الرواسب تراوحت من 1.55الى 3.408 مايكروغرام / غرام وزن جاف في المحطة الاولى والثانية على التوالي .مستويات مركبات TPHs في الانواع النباتية تراوحت من4.222 الى 13.985 مايكروغرام / غرام وزن جاف في *Phragmits australis* و *Ceratophyllum demersum* على التوالي وفي عضلات الاسماك تراوحت تراكيزها بين 1.765 و 92.592 مايكروغرام / غرام وزن جاف في *Liza abu* و *Barbus luteus* على التوالي ،بينما في انسجة المحار *Unio tigridis* كانت 13.18 مايكروغرام / غرام وزن جاف .

التراكيز الكلية للمركبات PAHs في عينات المياه تراوحت بين 18148.43 و155416.327 مايكروغرام / لتر ، بينما في الرواسب تراوحت بين 10866.47 و 44417.53 في المحطة الاولى والثانية على التوالي . مستويات مركبات PAHs ذات الوزن الجزيئي الواطئ كانت اقل من المركبات ذات الوزن الجزيئي العالي في عينات المياه والرواسب ومصادر مركبات PAHs في عينات المياه هي pyrogenic (طبيعية) ، بينما مصادرها في الرواسب هي petrogenic (من الفعاليات البشرية).التراكيز الكلية لمركبات PAHs في الانواع النباتية تراوحت بين 18532.11984 و123631.6954 نانوغرام/غرام في *P. australis* و *C. demersum* ، ومصادرها كانت petrogenic وفي الانواع السمكية تراكيزها الكلية تراوحت من 51161.66 الى 358080.1 نانوغرام/غرام في *Liza abu* و *Barbus luteus* على التوالي ،وكانت العينات السمكية المدروسة ملوثة جدا طبقا الى تصنيف Baumard *et al.* (1998). مصادر هذه

المركبات في *B. luteus* كانت pyrogenic ،بينما في *L. abu* و *Cyprinus* كانت petrogenic. تراكيزها الكلية في المحار كانت 152462.93 نانوغرام/ غرام، ومصادرها كانت pyrogenic. المستويات العالية من المركبات الهيدروكربونية النفطية الكلية والمركبات الهيدروكربونية الاروماتية المتعددة الحلقات المسجلة في الانواع المائية المدروسة تقترح بان هذه الكائنات نستطيع استخدامها كأدوات لتقييم التلوث بالمركبات الهيدروكربونية في هذه المنطقة .