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Levels of polycyclic aromatic hydrocarbons in some marine and freshwater fish in Basrah City

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Abstract

This study represents the concentrations of 16 PAHs in six commercial fish species (*Cyprinus carpio* , *Oreochromis aureus* , *Planiliza abu* , *Pampus argenteus* , *Epinephelus coioides* , *Otolithes ruber*) , which were collected from Shatt Al-Arab river and Iraqi marine water from August 2018 to May 2019. The analysis of Polycyclic Aromatic Hydrocarbons (PAHs) in fish muscles was determined using Gas chromatography (GC). The total concentration of PAHs in fish ranged from (2.791) ng/g dry weight in *E.coioides* during summer to (42.953) ng/g dry weight in *C.carpio* muscles during autumn. The results were showed that the origin of PAHs in fish is Pyrogenic and a little bit of it is Petrogenic. *C. carpio* showed a high ability to accumulate PAHs compounds in muscles compared with other fish. There was a different ability of fish to accumulate PAHs compounds from the surrounded environments.

Key Words: PAHs, Marine and Freshwater fish, GC, Shatt Al-Arab River, NW Arabian Gulf.

Introduction

Aromatic hydrocarbons can be defined as a large group of cyclic organic compounds consisting of hydrogen and carbon arranged in two or more fused benzene rings and can also be called polynuclear aromatics (PNAs) or Polycyclic Organic Matter (POM) (ATSDR, 2012). Physical and chemical properties of PAHs vary with molecular weight, for example, are more resistant to oxidation, reduction, and volatilization with increased molecular weight, while their solubility decreases with the increase of molecular weight. PAHs differ in their behavior and distribution in the environment and their effects on biological systems (Nagpal, 1993). Depending on molecular weight, PAHs can be divided into two groups (Columbus, 2014; Stogiannidis and Laane, 2015):

1. Low molecular weight PAHs usually consist of two or three rings, such as acenaphthalene, fluorine, and naphthalene, which have acute toxicity to aquatic organisms.
2. High molecular weight PAHs consist of four or more benzene rings, including Chrysene, Pyrene, Benzo (a) Pyrene and, Benzo (a) anthracene, known to cause cancer.

In general, PAHs are environmentally stable and lipophilic compounds that have a high solubility potential and water-threatening nature. This property increases by increasing its molecular weight (Juhasz and Naidu, 2000). Among the hundreds of aromatic hydrocarbons, 16 PAHs compounds were identified as priority contaminants as the most harmful compounds for humans and other organisms (Ravindra *et al*., 2008).

Aromatic hydrocarbons reach to the aquatic environment from different sources, either directly or indirectly, which causing contamination of the aquatic environment. (USEPA, 2000). There are two sources of PAHs: natural and human processes (Morillo *et al*., 2008). Natural sources include natural oil spills from the bottom, erosion of old deposits, volcanoes and, forest burning, as well as the ability of algae, bacteria and, some plants to manufacture a few concentrations of these (Farid, 2007).

The major sources of human emissions of PAHs in the environment are industrial emissions, which are the main sources of emissions of PAHs in the environment (Zakaria *et al*., 2002; Tsymblyuk *et al*., 2011 and Abdullatif, 2015), formed when processing organic raw materials at high temperature as well as household waste, fossil fuel combustion, oil spillage, waste disposal, cigarette smoke, aviation, mining and, oil transportation (Fadzil *et al*., 2008; Tsymblyuk *et al*., 2011). Smoke from vehicles and transport is a source of pollution from PAHs that are made up of four to six benzene rings (Wong and wang, 2001). More than 150 PAHs were found in car exhausts (Abdullatif, 2015).

PAHs reach the aquatic environments by domestic and industrial effluents, diesel and diesel engine exhausts, the deposition of airborne particulates, the spill of oil and petroleum products into water bodies and, the majority of aromatic contaminants entering aquatic environments remain close to sedimentation sites, suggesting that rivers close to human centers are the primary reservoirs of PAHs (Jazza, 2015).

It is important to note that incomplete combustion originating from a natural or human source contributes significantly to the addition of PAHs to the environment (Abdel-Shafy and Mansour, 2016). The oil contaminants are transferred to different fish tissues through the circulation system, which increases their absorption and accumulation within their bodies, where hydrocarbons enter the fish's body through the gills or the minutes of food or drinking water (Al Saad *et al*., 1997). These dangerous compounds accumulate in the brain, glands and, genitals and accumulate in them at a higher concentration than those in the tissues of muscles and liver due to the high-fat ratio in those organs (Deb *et al*., 2000).

Because of the high toxicity of PAHs compounds, it is necessary to study these compounds in marine and freshwater fish because of their economic importance and large consuming of fish in Basrah. Therefore, the aim of the present study is to evaluate and determine 16 compounds of PAHs residues in six marine and freshwater commercial fish species from North West Arabian Gulf and Shatt Al-Arab river.

Materials and Methods

Six fish species (*Cyprinus carpio* , *Oreochromis aureus* , *Planiliza abu* , *Pampus argenteus* , *Epinephelus coioides* , *Otolithes ruber*) were collected from August 2018 to May 2019 from Shatt Al-Arab river and Iraqi marine water by local fishermen, Figure (1). Fish samples were placed in plastic boxes, immediately transferred to the laboratory before analysis, the muscles were cut to small parts and dried in freeze-drier, grind and, placed in clean a glass vial to become ready for analysis. The procedure of Grimalt and Oliver (1993) was used for the extraction of hydrocarbons from fish muscles.

The concentrations of PAHs in muscles were determined by using gas chromatography, which is equipped with a Flame Ionization Detector (FID). The device has a type separation column (Agilent 125-103 KHP-5) with dimensions (30 m * 320 µm. * 0.25 µm.). Fish samples were analyzed for a suite of 16 PAHs compounds.

Figure (1). Location map showing fishing sites: (1) National Iraqi waters, and (2) Shatt Al-Arab River.

Results and Discussion

Table (1)shows the percentage of fat, lengths and, weights of the fish studied during the different study seasons. The concentrations of 16 PAH compounds are ranged from 2.791 ng/g dry weight to 42.953 ng/g dry weight. The maximum concentration of PAHs was found in *C.carpio* during autumn while the minimum concentration of PAHs in *E.coioides* during summer. The concentrations of PAHs determined in the muscle tissues of the fish from Iraqi marine water and Shatt Al-Arab River are presented in Tables (2-5). The results of the statistical analysis showed no significant differences between fish species (LSD = 0.629 $P > 0.05$). There were significant differences between the seasons (LSD = 0.759 $P \leq 0.05$), and significant differences between polycyclic aromatic compounds (LSD = 1.235 $P \leq 0.05$).

The results of the study showed the presence of high molecular weight PAHs is higher than percentages of low molecular weight PAHs Figure (2), due to their environmentally stability and resistance to microbial degradation (Anyakora *et al.*, 2005; Anyakora and Coker, 2007), as well as the ability of fish to metabolize lower molecular weight PAHs higher than those with high molecular weights (Dasilva *et al.*, 2006; Ramalhosa *et al.*, 2012). PAHs can be eliminated outside the body through rapid and effective metabolism through an effective Mixed Function Oxygenase (MFO) system in fish (Hylland, 2006).

The variation in total PAHs concentrations in the studied species during four seasons is due to different species, feeding system, abundance of food, living space, fat content and, environmental conditions (Al-Saad, 1995; Al-Saad *et al*., 1997; Al-Khion, 2012). The difference is also due to the ability of these compounds to bind and move through the surface of the cell membranes of fish or so-called life availability as well as the location of the living organism from the water column (Neff, 2002), as well as the biological availability of chemicals in food, water and chemical measurements of the aquatic environment (Al-Saleh and Al-Doush, 2002).

Table (1) Lipids content (%) along with lengths and body weights of selected fish species .

Fish	Season	Total weight	Total length	Fat%
P. argenteus	**summer**	**451.96**	**27.4**	**4.87**
	Autumn	**202.02**	**25.7**	**3.80**
	Winter	**00.302**	**26.3**	**3.55**
	Spring	**128.37**	**20.5**	**5.06**
E.coioides	**summer**	**901.10**	**38.8**	**2.33**
	Autumn	**425.84**	**31.8**	**5.33**
	Winter	**950.38**	**41.8**	**4.11**
	Spring	**600.35**	**35.8**	**5.13**
O.ruber	**summer**	**351.96**	**35.7**	**2.31**
	Autumn	**600.53**	**38.1**	**7.58**
	Winter	**475.41**	**36.8**	**3.03**
	Spring	**443.07**	**34.9**	**5.03**
C.carpio	**summer**	**803.31**	**38.9**	**5.45**
	Autumn	**1426.95**	**45.8**	**12.45**
	Winter	**1025.33**	**43.7**	**6.02**
	Spring	**225.03**	**23.0**	**5.07**
O.aureus	**summer**	**246.67**	**21.4**	**1.78**
	Autumn	**243.64**	**22.0**	**4.24**
	Winter	**76.48**	**17.5**	**4.01**
	Spring	**172.49**	**20.7**	**4.21**
P.abu	**summer**	**19.23**	**11.7**	**4.38**
	Autumn	**15.49**	**10.6**	**9.94**
	Winter	**37.48**	**14.7**	**6.90**
	Spring	**22.58**	**12.7**	**7.67**

Seasonal variations in concentrations may also relate to specific factors such as fish biology, mainly their dietary habits and sexual cycle (Hantoush *et al*., 2001). Hydrocarbons are readily available to fish in the aquatic system through the food chain and by water and sediment (Visciano *et al*., 2008).

The low and high molecular weight of PAHs was calculated to determine the source of PAHs within fish muscle tissue. The value was smaller than one for all fish samples and in all seasons indicating that the source of PAHs was Pyrogenic (Doong and Lin, 2004).

The ratio of Phenanthrene to anthracene is less than 10, indicating that it is derived from Pyrogenic (Zakaria *et al*., 2002).

The ratio of Fluoranthene to Pyrene ranged 0.117-1.974 in *P.argenteus* , 0.651-9.785 in

E.coioides , 1.028- 2,913 in *O.ruber*, 0.027-4.293 in *C.carpio*, 2,285-13.818 in *O. aureus* and 0.331-4.384 in *P.abu*, indicating that the source of PAHs in the studied fish is pyrogenic and a small part of it petrogenic (Zakaria *et al*., 2002).

The highest concentrations of PAHs were found in *C.carpio* during autumn and the lowest was found in the *E.coioides* fish during summer. This result, which indicates the susceptibility of *C.carpio* to accumulate pollutants with high concentrations compared to other species may be due to the high proportion of fat in these species or maybe the reason that this type of freshwater fish has a greater ability to accumulate aromatic hydrocarbons in their tissues through water and food. The dangerous results showed high concentration of Benzo (a) pyrene in all samples. This compound is not manufactured by the organism and is produced from incomplete combustion of fuel (Ramesh *et al*., 2004). Previous studies have also shown that B (a) P the most common of all polycyclic aromatic hydrocarbons predominance of PAHs in fish tissues (Al-Saleh and Al-Doush, 2002; Vives, *et al*., 2004; Hamad, *et al*., 2014).

Table (2) Concentrations of PAHs in fish muscles studied during summer in ng/g dry weight.

PAHs Compounds	**Types of Fish**					
	P. argenteus	***E. coioides***	***O. ruber***	***C. carpio***	***O. aureus***	***P.abu***
Naphthalene	**ND**	**ND**	**ND**	**ND**	**ND**	**ND**
2-methyl naphthalene	**ND**	**ND**	**ND**	**ND**	**ND**	**ND**
1-methyl naphthalene	**ND**	**ND**	**ND**	**ND**	**ND**	**ND**
Acenaphthlene	**0.016**	**ND**	**ND**	**ND**	**ND**	**ND**
Acenaphthene	**0.149**	**ND**	**ND**	**ND**	**ND**	**ND**
Fluorene	**0.065**	**0.101**	**0.049**	**0.026**	**0.011**	**0.012**
Phenanthrene	**0.071**	**0.040**	**0.032**	**0.041**	**0.031**	**0.013**
Anthracene	**0.131**	**0.020**	**0.014**	**0.018**	**ND**	**ND**
Fluoranthene	**0.067**	**0.432**	**0.041**	**0.019**	**0.032**	**0.039**
Pyrene	**0.062**	**0.663**	**0.174**	**0.032**	**0.014**	**0.038**
Chrysene	**0.058**	**0.016**	**0.040**	**0.145**	**0.089**	**0.064**
Benzo(a)fluoranthene	**0.207**	**0.041**	**0.281**	**0.068**	**0.019**	**0.036**
Benzo(k)fluoranthene	**1.241**	**0.061**	**1.169**	**0.584**	**0.057**	**0.431**
Benzo(b)fluoranthene	**0.548**	**0.081**	**0.310**	**0.324**	**0.208**	**0.149**
Benzo(a) pyrene	**9.167**	**1.241**	**3.994**	**7.463**	**3.611**	**1.862**
Indeno(1,2,3-cd)pyrene+Dibenz(a,h)anthracene	**9.414**	**0.041**	**2.167**	**1.434**	**1.668**	**0.548**
Benzo(g,h,i)perylene	**0.825**	**0.054**	**0.335**	**0.308**	**0.305**	**0.199**
Total	**22.021**	**2.791**	**8.606**	**10.462**	**6.045**	**3.391**
LPAHs	**0.432**	**0.161**	**0.095**	**0.085**	**0.042**	**0.025**
HPAHs	**21.589**	**2.630**	**8.511**	**10.377**	**6.003**	**3.366**
LPAHs /HPAHs	**0.020**	**0.061**	**0.011**	**0.008**	**0.006**	**0.007**
Phenanthrene /Anthracene	**0.541**	**2.000**	**2.285**	**2.277**		
Fluoranthene/ Pyrene	**1.080**	**0.651**	**0.235**	**0.593**	**2.285**	**1.026**

LPAHs /HPAHs

LPAHs : (Low Polycyclic Aromatic Hydrocarbons)
HPAHs : (High Polycyclic Aromatic Hydrocarbons)
ND: Not Detected

Table (3) Concentrations of PAHs in fish muscles studied during autumn in ng/g dry weight.

PAHs Compounds	Types of Fish					
	P. argenteus	*E. coioides*	*O. ruber*	*C. carpio*	*O. aureus*	*P.abu*
Naphthalene	ND	ND	ND	ND	ND	ND
2-methyl naphthalene	ND	ND	ND	ND	ND	ND
1-methyl naphthalene	ND	ND	ND	ND	ND	ND
Acenaphthlene	ND	0.010	0.048	ND	ND	0.024
Acenaphthene	0.036	0.026	0.041	0.028	0.013	0.138
Fluorene	0.096	0.141	0.018	0.015	0.091	0.061
Phenanthrene	0.021	0.053	0.054	0.111	0.017	0.046
Anthracene	0.049	0.039	0.081	0.103	0.051	0.114
Fluoranthene	0.065	0.081	0.067	0.817	0.066	0.171
Pyrene	0.359	0.011	0.023	29.789	0.021	0.039
Chrysene	0.028	0.021	0.038	0.588	0.028	0.0799
Benzo(a)fluoranthene	0.253	0.087	0.217	0.392	0.133	0.163
Benzo(k)fluoranthene	1.234	0.715	2.779	0.108	0.844	1.831
Benzo(b)fluoranthene	0.637	0.321	0.513	0.616	0.502	0.494
Benzo(a) pyrene	12.855	6.844	12.88	6.775	7.230	11.736
Indeno(1,2,3-cd)pyrene+Dibenz(a,h)anthracene	2.307	0.885	4.245	2.903	1.292	9.539
Benzo(g,h,i)perylene	0.869	0.549	0.799	0.708	0.527	0.818
Total	18.809	9.783	21.803	42.953	10.815	25.253
LPAHs	0.202	0.269	0.242	0.257	0.172	0.383
HPAHs	18.607	9.514	21.561	42.696	10.643	24.870
LPAHs /HPAHs	0.0108	0.028	0.011	0.006	0.016	0.015
Phenanthrene /Anthracene	0.428	1.358	0.666	1.077	0.333	0.403
Fluoranthene/ Pyrene	0.181	7.363	2.913	0.027	3.142	4.384

LPAHs /HPAHs
LPAHs : (Low Polycyclic Aromatic Hydrocarbons)
HPAHs : (High Polycyclic Aromatic Hydrocarbons)
ND: Not Detected

Table (4) Concentrations of PAHs in fish muscles studied during winter in ng/g dry weight.

PAHs Compounds	Types of Fish					
	P. argenteus	*E. coioides*	*O. ruber*	*C. carpio*	*O. aureus*	*P.abu*
Naphthalene	ND	ND	ND	ND	ND	ND
2-methyl naphthalene	ND	ND	ND	ND	ND	ND
1-methyl naphthalene	ND	ND	ND	ND	ND	ND
Acenaphthlene	ND	ND	ND	0.014	0.011	0.024
Acenaphthene	0.025	ND	0.029	0.069	0.029	0.138
Fluorene	0.064	0.029	0.061	0.020	0.120	0.061
Phenanthrene	0.016	0.030	0.034	0.087	0.046	0.046
Anthracene	0.027	ND	0.017	0.075	0.023	0.114
Fluoranthene	0.018	0.047	0.013	0.022	0.039	0.171
Pyrene	0.153	0.034	0.457	0.017	0.016	0.039
Chrysene	0.035	0.048	0.016	0.019	0.025	0.079
Benzo(a)fluoranthene	0.147	0.249	0.101	0.071	0.106	0.163
Benzo(k)fluoranthene	1.710	0.359	0.953	1.280	0.086	1.831
Benzo(b)fluoranthene	0.409	0.171	0.294	0.421	0.302	0.494
Benzo(a) pyrene	6.005	3.976	7.817	9.180	4.932	11.736
Indeno(1,2,3-cd)pyrene+Dibenz(a,h)anthracene	8.485	0.466	3.263	1.381	3.542	9.539
Benzo(g,h,i)perylene	0.503	0.258	0.432	0.596	0.460	0.818
Total	17.597	5.667	13.487	13.252	9.737	25.253
LPAHs	0.132	0.059	0.141	0.265	0.229	0.383
HPAHs	17.465	5.608	13.346	12.987	9.508	24.870
LPAHs /HPAHs	0.007	0.011	0.011	0.020	0.024	0.015
Phenanthrene /Anthracene	0.592		2.000	1.160	2.000	0.403
Fluoranthene/ Pyrene	0.117	1.382	0.028	1.294	2.437	4.384

LPAHs /HPAHs
LPAHs : (Low Polycyclic Aromatic Hydrocarbons)
HPAHs : (High Polycyclic Aromatic Hydrocarbons)
ND: Not Detected

Table (5) Concentrations of PAHs in fish muscles studied during spring in ng/g dry weight.

PAHs Compounds	Types of Fish					
	P. argenteus	*E. coioides*	*O. ruber*	*C. carpio*	*O. aureus*	*P.abu*
Naphthalene	**ND**	**ND**	**ND**	**ND**	**ND**	**ND**
2-methyl naphthalene	**ND**	**ND**	**0.004**	**ND**	**ND**	**0.007**
1-methyl naphthalene	**ND**	**ND**	**0.002**	**ND**	**ND**	**ND**
Acenaphthlene	**ND**	**ND**	**0.002**	**0.003**	**ND**	**0.017**
Acenaphthene	**0.030**	**0.032**	**0.080**	**0.002**	**0.036**	**0.031**
Fluorene	**0.006**	**0.009**	**0.066**	**0.007**	**0.013**	**0.005**
Phenanthrene	**0.045**	**0.047**	**0.127**	**0.054**	**0.081**	**0.143**
Anthracene	**0.035**	**0.015**	**0.064**	**0.218**	**0.025**	**0.114**
Fluoranthene	**0.077**	**0.137**	**0.164**	**0.395**	**0.152**	**0.095**
Pyrene	**0.039**	**0.014**	**0.115**	**0.092**	**0.011**	**0.172**
Benzo(a)fluoranthene	**0.025**	**0.023**	**0.066**	**0.076**	**0.031**	**0.082**
Chrysene	**0.115**	**0.084**	**0.797**	**0.349**	**0.204**	**0.151**
Benzo(b)fluoranthene+ Benzo(k)fluoranthene	**0.987**	**0.593**	**2.410**	**1.104**	**0.704**	**0.610**
Benzo(a) pyrene	**0.610**	**0.128**	**0.530**	**0.268**	**0.250**	**0.122**
Indeno(1,2,3-cd)pyrene +Dibenz(a,h)anthracene	**3.896**	**2.236**	**13.102**	**4.094**	**2.398**	**2.511**
Benzo(g,h,i)perylene	**0.273**	**0.017**	**0.226**	**0.073**	**0.091**	**0.051**
Total	**6.138**	**3.335**	**17.755**	**6.735**	**3.996**	**4.111**
LPAHs	**0.116**	**0.103**	**0.345**	**0.284**	**0.155**	**0.317**
HPAHs	**6.022**	**3.232**	**17.410**	**6.451**	**3.841**	**3.794**
LPAHs /HPAHs	**0.019**	**0.031**	**0.019**	**0.044**	**0.040**	**0.083**
Phenanthrene /Anthracene	**1.285**	**3.133**	**1.984**	**0.247**	**3.240**	**1.254**
Fluoranthene/ Pyrene	**1.974**	**9.785**	**1.426**	**4.293**	**13.818**	**0.552**

LPAHs /HPAHs
LPAHs : (Low Polycyclic Aromatic Hydrocarbons)
HPAHs : (High Polycyclic Aromatic Hydrocarbons)
ND: Not Detected

When comparing the results of the current study with those in the region and the world, they are located within the recorded concentrations and contain contaminants table (6).

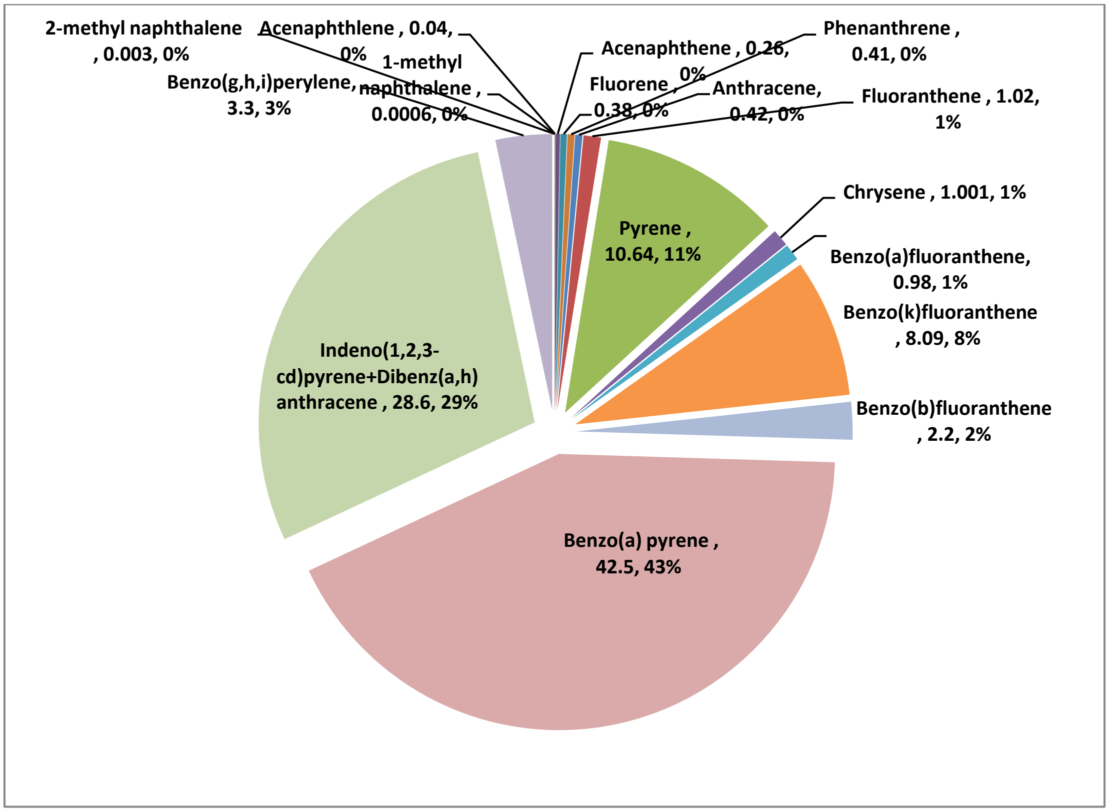

Figure (2) Percentage of PAHs in fish studied during four seasons .

Table (6) Comparison of PAHs levels in fishes samples with those in other studies in Iraq and the world.

References	Location	Concentrations (ng.g-1) d.w
Al-Saad *et al*. (2006)	**Northwest Arabian Gulf**	**6.78-23.83**
Al-Saad,*et al*. (2006)	**Red sea coast of Yemen**	**23.90-57.90**
Al-Khatib (2008)	**Hor Al-Howaiza**	**0.1– 92.7**
Beg *et al*. (2009)	**Kuwait Bay**	**1.079-15.022**
Al-khion (2012)	**Iraqi coast regions**	**12.19-86.48**
Mirza *et al*. (2012)	**Iranian coast**	**126.0-226.1**
Hamad *et al*. (2014)	**Libya**	**2.409-5.510**
Jazza (2015)	**Al-Kahlaa River / Missan**	**1.095 -16.661**
Al-Imarah *et al*. (2017)	**Shatt Al-Arab and N.W. Arabian Gulf**	**23.046-407.835**
Sun *et al*. (2018)	**China**	**86.70-256.0**
The Present study	**Shatt Al-Arab and N.W. Arabian Gulf**	**2.791-42.953**

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مستويات الهيدروكاربونات الاروماتية متعددة الحلقات في بعض الأسماك البحرية والنهرية في مدينة البصرة

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الخلاصة

شملت هذه الدراسة قياس الهيدروكاربونات الاروماتية متعددة الحلقات في ستة أنواع من الأسماك التجارية في محافظة البصرة وهي و *Cyprinus carpio* الكارب الاعتيادي و*Oreochromis aureus* البلطي و *Planiliza abu* الخشني و *Pampus argenteus* الزبيدي و *Epinephelus coioides* الهامور و *Otolithes ruber* النويبي والتي جمعت من شط العرب و شمال غرب الخليج العربي للمدة من أغسطس 2018 الى أيار 2019 . تم تحديد الهيدروكاربونات الاروماتية متعددة الانوية باستخدام جهاز الغاز كروماتوغرافي (GC) . تراوح المجموع الكلي لتراكيز ال PAHs من 2.791 نانوغرام/غرام في اسماك الهامور خلال فصل الصيف الى 42.953 نانوغرام/غرام في اسماك الكارب الاعتيادي خلال فصل الخريف. اظهرت النتائج ان مصدر ال PAHs في عضلات الأسماك هو Pyrogenic وجزء قليل منه Petrogenic ، كما أظهرت النتائج ان اسماك الكارب الاعتيادي لها قدرة عالية على تراكم مركبات ال PAHs في عضلاتها مقارنة مع الأسماك الأخرى ، وكان هنالك قدرات مختلفة للأسماك لتراكم هذه المركبات من البيئات المحيطة .