

Polycyclic Aromatic Hydrocarbons in Some Fishes from the Iraqi Marine Waters

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Abstracts

This study represent the concentrations of 16 PAHs in 12 of commercial fish species (*Euryglossa orientalis* , *Acanthopagrus araticus* , *Epinephelus coioides* , *Chirocentrus dorab* , *Scomberoidescommersonnianus* , *Scomberoides lysan* , *Otolithes ruber* , *Alepes diedaba* , *Tenualosa ilisha* , *Parastromateus niger* , *Pampus argentatus* , *Ablennws hians*) . which were collected from the coastal area of the of Iraqi marine water during 2015. The analysis of Polycyclic Aromatic Hydrocarbons (PAHs) in fish muscles were determined using HPLC to identify the sources of PAHs, The total concentration of PAHs in fishes ranged between (0.432) ng/g dry weight in *P. niger* and (14.939) ng/g dry weight in *T. ilisha* muscles . When we calculated the ratio of (Flouranthene to Pyrene) and the ration of (Phenanthrene to Anthracene) and ratio of low molecular weight to high molecular weight of PAHs and the ration of Anthracene to (Anthracene + Phenanthrene) and the ration of Benzo[a]anthracene to (Benzo[a]anthracene+ Chrysene), it is showed that the PAHs origin in fishes were Pyrogenic and Petrogenic . *T. ilisha* shows high ability to accumulate PAHs compounds like Fluorene , Anthracene, Acenaphthene and Phenanthrene in muscles compared with other fishes , there was different ability of fish to accumulate the PAHs from surrounded environments .

Keywords: PAHs compounds, HPLC, fishes, Iraqi marine water.

1.Introduction

The Arabian Gulf region possesses some of the most endangered coastal areas in the world due to petroleum point and non point sources. Over half of the world oil supply is transported from Arabian Gulf and offshore oil exploitation in this region considered the most extensive in the world. In spite of the fact that the overall pollution in the Northern coasts of the Arabian Gulf is in medium range for many petroleum associated (Eghtesadi – Araghi & Farzadnia ,2011). Recently there has been a global growing concern on the release of harmful organics into the marine environment. So, the need for identification of organic and inorganic pollutants in the Arabian Gulf has become an important issue, too. That is because areas are considered among the oil producing regions, and among the busiest in ship traffic. The chronic oil pollution in such vital areas from industrial and other anthropogenic sources, which is described in some Literature as "acute oil pollution" (DouAbul *et al.*, 1998; Al-Saad ,*et al.*2006;AL-khion 2012).

Polycyclic aromatic hydrocarbons (PAHs: also called poly nuclear aromatic hydrocarbons), series of organic contaminants, are ubiquitous in the marine environment (Aghajanloo *et al.*, 2013; Bhuvaneshwari *et al.*, 2013; Garcia-Flores *et al.*, 2013).

PAHs generally possess high chemical stability and hydrophobic properties, which result in enhanced accumulation and a high capacity for distribution in the environment. A number of PAH compounds are considered as hazardous environmental chemicals (Adel-Razeq , 2012).

Fresh and marine Sediments often contain very high concentrations of PAHs compared to the surface water, so the particles transferred from the rivers, which contain oils enter the body of the fish through the skin or gills, In addition, contaminants such as tar balls in sediment entering the intestines through ingestion of water by Fish(AL-khion 2012). Fish greatly influenced by waves, tides and wind much where carried from place to place, a result likely that the samples do not reflect the degree of oil pollution at the sites (Ahmed, 2014).

Although human health had not been considered to be at risk from concentrations of petroleum hydrocarbons in fish, the possible consequences of bioaccumulation cannot be ignored especially in communities consuming large quantities of fish (Shriadah 2001).therefore have an increasing effect in the accumulation of PAHs Pollution by persistent chemicals is potentially harmful to the organisms at higher tropic levels in the food chain. The marine organisms like fish are able to accumulate several fold higher concentration of PAHs than the surrounding water (Law and Hellou, 1999; Johnson-Restrepo , *et al.*,2008). Fish is a major source of proteins and healthy lipids for people. In particular, the long-chain omega-3 fatty acids have been shown to have numerous beneficial roles in the human health, despite the human benefits of a fish diet, an issue of concern related to frequent fish consumption is the potential risk arising from exposure to toxic chemicals (Ismail, 2005).

The study of PAHs in coastal areas of Northern Arabian Gulf is of importance, since these areas are ecologically, and economically of important. These locations are known to face considerable pollutant input from variety of land-based sources via coastal discharges.

The Arabian Gulf is considered an extremely demanding shipping route for oil, with accidental spillage being almost unavoidable. Also, oil refinery effluents and lose, during operations have been identified as the major source of oil contamination in the waters of the Arabian Gulf . In combination , these source provide long term input of petroleum hydrocarbons into the Gulf ecosystem , where's major oil spills , either being unintentional or as an outcome of military activities , have added occasional dramatic plus of oil contamination to the long term background of oil pollution of this area (Al-Saad , *et al.*,2006).

Because of environmental pollution it has become clear in the last fifty years because of the population explosion and the throwing of industrial and agricultural waste and excessive use of energy resources, especially petroleum products, which leads to an imbalance in the balance of components of the atmosphere and the pollution of sources of life (Air, water and soil). The environmental importance to the region north-west of the Arabian Gulf for being a suitable place for the presence of feeding and reproduction of many organism and a good source for fishing and an important outlet for Iraqi regional waters, this study aimed to determine the concentration of polycyclic aromatic hydrocarbons compounds PAHs carcinogenic that cause serious effects in some fishes and identify the main sources of these compounds.

2.MATERIALS AND METHODS

Twelve fish species (*Euryglossa orientalis* , *Acanthopagrus araticus* , *Epinephelus coioides* , *Chirocentrus dorab* , *Scomberoides commersonnianus* , *Scomberoides lysan* , *Otolithes ruber* , *Alepes diedaba* , *Tenualosa ilisha* , *Parastromateus niger* , *Pampus argentatus* , *Ablennws hians*) were collected during 2015 from Iraqi marine water (Table 1) . The 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, grounded and sieved using a 63 μ m metal sieve then placed in clean a glass vial to become ready for analysis. The procedure of Grimalt and Oliver (1993) was used for the extraction of fish muscles. The concentrations of PAHs in muscles were determined by using High Pressure Liquid Chromatography (HPLC) Shimadzu CBM20A with UV-visible detector.

The fish samples were analyzed for a suite of 16 PAHs ; naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), Fluorene (FLU) , Phenanthrene (PHE) , anthracene (ANT), Fluoranthene(FLUE), pyrene (PYR) , Benzo[a]anthracene (BaA), chrysene (CHY), benzo(b)fluoranthene (BbF) , Benzo[k]fluoranthene (BkF) , benzo(a)pyrene (BaP), Dibenzo[a,h]anthracene (DahA) , benzo(ghi)perylene (BPY) and Indeno[1,2,3-cd]pyrene (InP) .

Some of ratios used to determine sources of aromatic compounds are represented as including the following : (Fl/Py), (Phen/Ant) , (LMW /HMW),(Ant / (Ant + Phe)) and (Bap / (Bap + Chr)) . The calculation of these concentrations can be an indicator of the source of the aromatic hydrocarbons found in the fish samples, ratio of (Fl/Py) Values greater than 1 have been used to indicate pyrogenic origins and values less than 1 are attributed to petrogenic source (Qiu *et al.*, 2009) . Ratio of (Phen/Ant) less than 1 indicates petrogenic (Zhu *et al.*,2004) . ratio of(LMW /HMW) values greater than 1 indicate petrogenic origins and values less than 1 are attributed to pyrogenic sources (Vrana *et al.*, 2001).

(Ant/(Ant+Phe)) ratio presumes that ratios less than 0.1 indicate PAHs source to be of petroleum origin while ratios larger than 0.1 indicate PAHs source to be of combustion origin and the ratio of (BaA /(BaA+CHR)) less than 0.2 implies petrogenic, from 0.2 to 0.35 indicates either petrogenic or pyrogenic origins , and larger than 0.35 implies pyrogenic sources (Guo *et al.*,2007) .

(Fl/Py) = fluoranthene to pyrene.

(Phen/Ant) = Phenanthrene to Anthracene .

Phenanthrene + Anthracene

LMW (low molecular weight) = Naphthalene + Acenaphthylene + Acenaphthene + Fluorene +

HMW(high molecular weight) = Fluoranthene + Pyrene + B a A+ Chrysene + Bbf+ BK f + BaP + DBA + B (ghi) perylene + indenol pyrene.

(Ant / (Ant + Phe)) = Anthracene to Anthracene + Phenanthrene .

(Bap / (Bap + Chr)) = benzo(a) anthracene / (benzo(a) anthracene + chrysene).

3.RESULTS AND DISCUSSION

The concentrations of the 16 compounds of PAH are presented in (Table 1). They ranged from 0.432 ng/g dry weight to14.939 ng/g dry weight. The maximum concentration of PAHs in marine fish species was

found in *T. ilisha* and the minimum concentration of PAHs was found in *P. niger*. The concentrations of PAHs determined in the muscle tissues of the fish from the Iraqi marine water are presented in (Table 2).

Table 1: The lipids content (%) along with length and body weight of selected fish species

Fish species	No. Of fish	Total weight (g)	Total length (cm)	Fat%
<i>Euryglossa orientalis</i>	32	480	30	2.66
<i>Acanthopagrus araticus</i>	40	410	23	4.1
<i>Epinephelus coioides</i>	10	1152	61	2.04
<i>Chirocentrus dorab</i>	15	819	75	2.84
<i>Scomberoides commersonnianus</i>	15	1214	80	2.25
<i>Scomberoides lysan</i>	20	408	25	9.1
<i>Otolithes ruber</i>	30	517	33	12.17
<i>Alepes diedaba</i>	25	219	22	3.22
<i>Tenualosa ilisha</i>	50	420	30	7.8
<i>Parastromateus niger</i>	20	333	23	11.1
<i>Pampus argentatus</i>	10	310	22	22.1
<i>Ablennws hians</i>	35	1350	79	2.51

Table 2: The concentrations and ratios of polycyclic aromatic hydrocarbons in the fishes tissues.

Name compound	<i>E. orientalis</i>	<i>Ac. raticus</i>	<i>Ep. coioides</i>	<i>C. dorab</i>	<i>S. commersonnianus</i>	<i>S. lysan</i>	<i>O. ruber</i>	<i>Al. diedaba</i>	<i>T. ilisha</i>	<i>P. niger</i>	<i>Pa. argentatus</i>	<i>A. hians</i>
Naphthalene	0.014	-	0.097	0.042	0.255	0.155	-	-	-	-	-	-
Acenaphthylene	0.512	0.892	-	1.011	2.606	0.338	6.953	2.162	-	-	-	-
Acenaphthene	0.085	-	0.555	-	0.139	0.554	-	0.156	1.028	-	-	-
Fluorene	-	0.254	-	0.345	0.526	0.997	-	0.388	9.64	-	4.324	4.642
Phenanthrene	0.006	-	0.022	0.014	-	-	-	0.007	0.832	-	0.762	-
Anthracene	0.977	0.368	-	-	-	-	-	1.284	2.434	-	-	1.174
Fluoranthene	-	-	0.016	-	-	0.101	-	-	-	-	-	-
Pyrene	0.001	0.017	0.024	0.065	0.070	-	-	-	0.281	-	0.280	-
Benzo[a]anthracene	0.021	-	0.079	0.013	0.011	-	0.054	0.032	0.278	0.259	0.129	-
Chrysene	0.002	0.008	-	0.023	0.011	-	-	0.002	0.127	-	-	0.054
Benzo[b]fluoranthene	0.016	-	0.009	0.022	0.067	-	-	-	-	0.060	0.008	-
Benzo[k]fluoranthene	0.028	-	0.001	-	-	-	-	0.114	0.158	-	0.012	0.023
Benzo[a]pyrene	-	-	0.005	-	-	-	0.584	-	0.157	-	0.005	0.029
Dibenzo[a,h]anthracene	-	-	-	1.527	-	-	2.867	0.764	-	-	-	-
Benzo[g,h,i]perylene	-	-	-	-	-	-	0.352	-	-	0.112	-	-
Indeno[1,2,3-cd] pyrene	-	-	-	0.071	-	-	-	-	-	-	-	-
Σ PAHs	1.666	1.541	0.813	3.140	3.688	2.148	10.811	4.913	14.939	0.432	5.522	5.923
fluoranthene/pyrene	-	-	0.688	-	-	-	-	-	-	-	-	-
Phen/Ant	0.006	-	-	-	-	-	-	0.006	0.342	-	-	-
LMW /HMW	22.663	60.030	5.728	0.819	21.964	-	1.802	4.378	13.885	-	11.672	54.364
Ant/(Ant+Phen)	0.993	-	-	-	-	-	-	0.993	0.745	-	-	-
BaA/(BaA+Chry)	0.892	-	-	0.368	0.482	-	-	0.923	0.686	-	-	-

The total PAHs concentration reported in fish samples of the present study appears to be higher than the concentration reported in edible fishes (0.207–3.365 ppm) of the Gomti river , Lucknow , India (Malik,*et al.* 2008) . The PAHs concentration detected in the fishes of present study is comparable with study reported in the muscles of the fish , from the Derna city coast (1.51 – 8.23 μ g/g dry wt.) (Hamad,*et al.* 2014) , and lower than the levels in the Red Sea Coast (57.90 ng/g dry wt.) (Al-Saad , *et al.* 2006) Table 3 .

Table 3: Comparison of PAHs levels in fishes samples with those in other studies in Iraq and the world.

References	Concentrations (ng.g-1) d.w	location
DouAbul, <i>et al.</i> 1987	14-106	North West of Arabian Gulf
Al-Saad &Al-Asadi, 1989	11.6-26.3	Shatt Al-Arab
Fowler, <i>et al.</i> 1993	21	Kuwait
Fowler, <i>et al.</i> 1993	7.6-239	Saudi Arabia
Fowler, <i>et al.</i> 1993	1.1-6.4	Bahrain
Fowler, <i>et al.</i> 1993	3.8-7.7	UAE
Fowler, <i>et al.</i> 1993	2.2-110	Oman
Fowler, <i>et al.</i> 1993	55.8-165.9	Qatar
Sreenivasa, and RAO 2001.	0.059- 0.183	India
Al-Saad, <i>et al.</i> 2006	23.90-57.90	Red sea coast of Yemen
Al-Saad, <i>et al.</i> 2006	6.78-23.83	North West of Arabian Gulf
Al-Khatib,2008	0.1 -42	Hor Al-Howaiza
Nasr, <i>et al.</i> 2012	0.062- 18.415	Egypt
Al-Khion ,2012	1.83-84.67	Iraqi coast regions
Hamad, <i>et al.</i> 2014	2.4093 - 5.5102	Libya
Olaji, <i>et al.</i> 2014	0.003- 0.038	Nigeria
Abdolahpur Monikh, <i>et al.</i> 2014	91.32 - 11 54.45	Arabian Gulf
Jazza, <i>et al.</i> 2015	1.095 -16.661	Al-Kahlai River / Missan
Persent study	0.432-14.939	Iraqi marine water

Among the various components of PAHs in present studies , Fluorene was the most frequently detected as reported in other studies (Hamad , *et al.* 2014 ; Liang , *et al.* 2007) , followed by Benzo[a]anthracene and acenaphthene. Indeno[1,2,3-cd] pyrene could not be detected in any fishes accept in *Ch.dorab* ,while the absence or rather low detection of certain PAHs in the fish samples may be attributed to their rapid depuration or biotransformation (Deb, *et al.* 2000). The accumulation and depuration of PAHs in fish can be influenced by various factors including route and duration of exposure, lipid content of tissues, environmental factors, differences in species, age, and sex, and exposure to other xenobiotics (Varanasi, *et al.* 1987).

In the muscle of *E. orientalis* , the most abundant PAHs are NAP , ACE ,ACY, PHE, ANT, FLUE, PYR, BaA, CHY, BaP, BbF and BkF (Fig. 1). In the muscle of *Ac. Araticus* the most abundant PAHs are ACE, FLU, ANT, PYR and CHY (Fig.2). In the muscle of *Ep. Coioides* NAP, ACE, PHE, FLUE, PYR, BaA, BbF, BkF, BaP and appear to be the most abundant PAHs(Fig.3). In the muscle of, *Ch.dorab* NAP, ACY, FLU, PHE, PYR, BaA, CHY, BbF, DahA and InP are the most abundant PAHs (Fig. 4). NAP, ACY, ACE, FLU, PYR, Bap, CHY and BbF are also abundant in the muscle of *S. commersonnianus* (Fig. 5). In the muscle of *S. lysan* , NAP, ACY, ACE , FLU and FLUE are the most abundant components of the PAHs(Fig. 6) . In the muscle of *O. ruber* , the most abundant PAHs are ACY, BaA, BaP, DahA and BPY (Fig. 7). In the muscle of *Al. dieddaba* the most abundant PAHs are ACY, ACE , FLU, PHE, ANT, BaA, CHY, BbF and DahA (Fig. 8). In the muscle of *T. ilisha* ACE, FLU, PHE, ANT, PYR, BaA, CHY, BkF and BaP appear to be the most abundant PAHs (Fig. 9). In the muscle of, *Pa. niger* BaA , BPY and BbF are the most abundant components of the PAHs (Fig. 10). FLU, PHE, BaA, BkF and BaP are also abundant in the muscle of *P. argentetus* (Fig. 11). In the muscle of *A. hians* , the most abundant PAHs are FLU, ANT, CHY, BaP and BkF (Fig. 12).

High amount of benzo[a]pyrene, the most carcinogenic polycyclic aromatic hydrocarbon, was found in the muscle of fish. Previous studies have also shown that benzo[a]pyrene, naphthalene and pyrene are the dominant compounds of PAHs in fish tissues (Hamad,*et al.* 2014; Vives,*et al.* 2004; Al-Saleh and Al-Doush, 2002).

The results showed that low molecular weight PAHs were accumulated much less in the fish tissues than high molecular weight PAHs. The concentrations of PAH compounds vary widely with molecular weight (Sverdrup *et al.*, 2002 ; Abdolahpur Monikh ,*et al.*2014). In general. Σ PAHs in this study were lower than that recorded for the Saudi Arabia coasts; 1.88-412 ng/l in Arabian Gulf (Awad , 1990),and comparable with that recorded for the Gulf of Libya Coast(Hamad,*et al.*,2014) .By comparing the present data with those reported by FAO/WHO for the guidelines of PAHs in food with value (0.001) for human consumer , it's clear that from the

present data it can be noticed that the levels of PAHs compounds in the muscles of these fishes were less than their levels in other fishes collected from other regions of Iraq and the world (Table 3).

Some ratios were calculated as attempt to identification the sources of the aromatic compounds, in the present study, the ratio of (FLU / PYR) < 1 (Tables 2) reflecting petrogenic origin. The HMW/LMW ratios values also were used to identification of the sources of the PAHs compounds , In this all the calculated values of the HMW/LMW ratio are >1 , and this is indicate the main source of the PAHs in the investigated area coming from petrogenic source (Tables 2) .

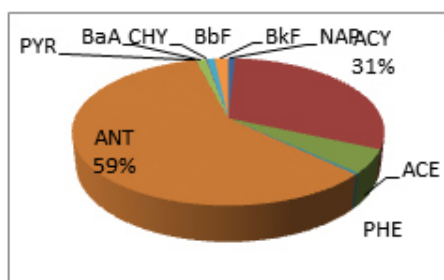


Fig.1 The percentage of the PAHs compound in *E. Orientalis*

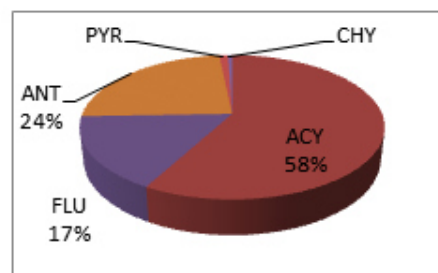


Fig.2 The percentage of the PAHs compound in *Ac. raticus*

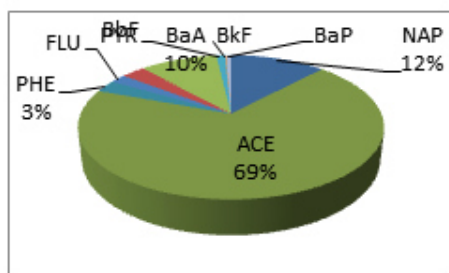


Fig.3 The percentage of the PAHs compound in *Ep. Coioides*

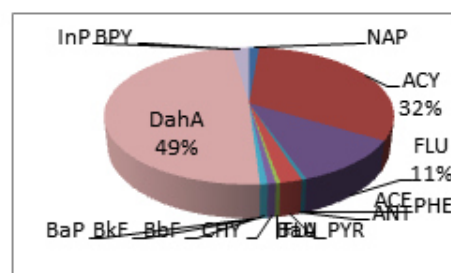


Fig.4 The percentage of the PAHs compound in *C. dorab*

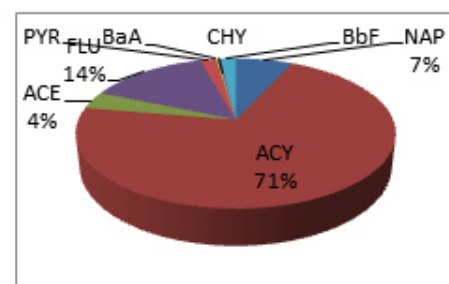


Fig.5 The percentage of the PAHs compound in *S. commersonianus*

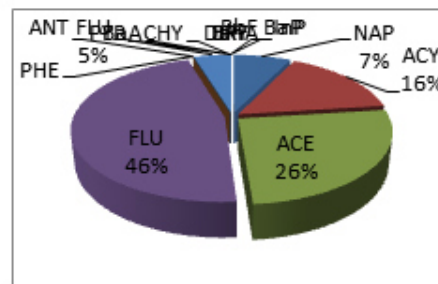


Fig.6 The percentage of the PAHs compound in *S. lysan*

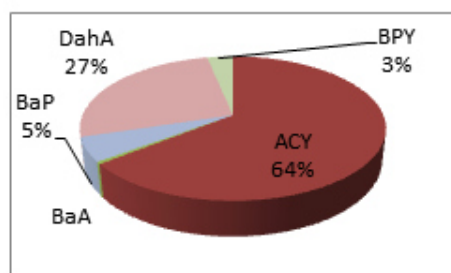


Fig.7 The percentage of the PAHs compound in *O. rube*

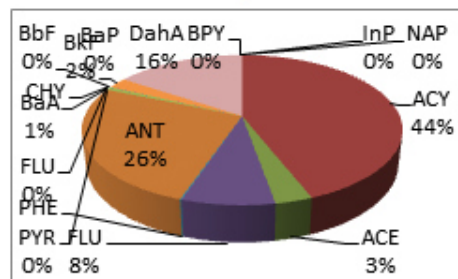


Fig.8 The percentage of the PAHs compound in *Al. diedaba*

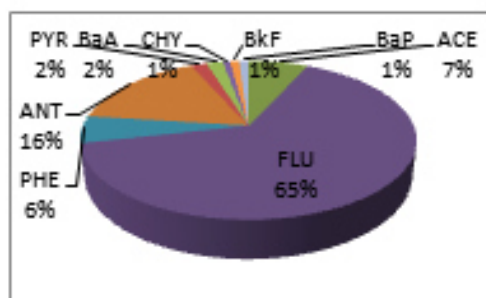


Fig.9 The percentage of the PAHs compound in *T. ilisha*

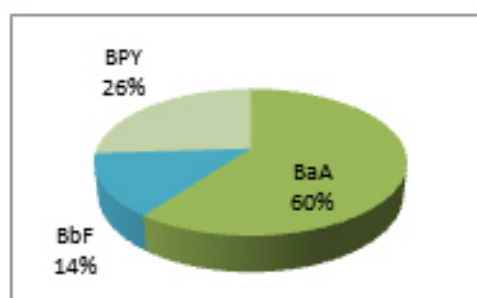


Fig.10 The percentage of the PAHs compound in *P. niger*

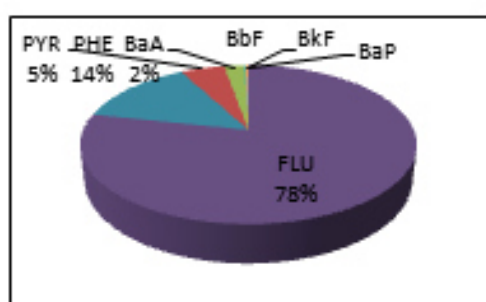


Fig.11 The percentage of the PAHs compound in *Pa. argentatus*

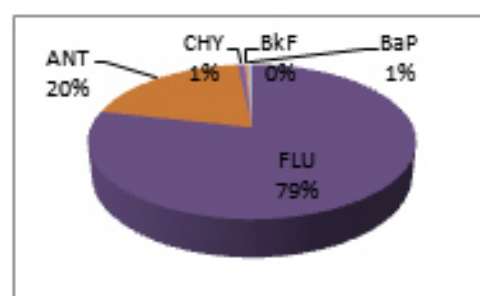


Fig.12 The percentage of the PAHs compound in *A. hians*

As a conclusion these fishes are contaminated with PAHs ,but these concentrations are not effects on human health, and the main sources of these compounds petrogenic or pyrogenic origins.

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