

## **Polycyclic Aromatic Hydrocarbons (PAHs) and Aliphatic Hydrocarbons (AHs) in Edible Fish from the Arabian Gulf**

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Petroleum deposits in the Arabian Gulf have fuelled the region's economy for over half a century. The Gulf's environment and its resources are influenced by natural processes and pressures arising from human activities (Price 1993). The 1990 - 1991 Iraqi invasion of Kuwait caused much concern, following the conflagration of over 700 oil wells and the intentional discharge of 6-10 million barrels of crude oil into the marine environment (Price 1998). Major national, regional and international research programmes have been initiated in an attempt to determine the nature, magnitude and environmental significance of the oil spill. Despite constraints and uncertainties associated with such assessments, a number of predictions have been made concerning the recovery of the region's natural ecosystem (McGlade and Price 1993).

Polycyclic aromatic hydrocarbons (PAHs) and aliphatic hydrocarbons (AHs) are important components of crude oil, which can produce significant contamination of the marine environment and adversely affect marine life. Some of the PAHs, which are commonly found in the environment, can become acutely toxic to DNA when exposed to solar radiation (Newsted and Giesy 1987). Thus breaks in single DNA strand resulting from PAH contamination of fish have been used as biomarkers of genotoxicity in fish and other aquatic species (Al-Hassan 2000a). The need for such biomarkers has been implemented in relation to carcinogenicity, reproductive problems and other adverse effects of pollution (Mitchelmore and Chipman 1998). Similarly, PAHs are found to have deleterious effect on the vitellogenesis of fish that varies with species and population and can even differ between individuals (Nicolas 1999).

During metabolism of petroleum-derived hydrocarbons, the ability of an organism to process PAHs may be altered by the presence of some polar components. Reports of high prevalence of liver neoplasm in bottom dwelling fish from urban waterways have heightened awareness of the potential problems concerning the quality of fish habitats, as well as food-chain transfer of contaminants from fish and shellfish to humans (Al-Hassan 2000a). The present study was undertaken to evaluate the trends of PAH and AH contamination in edible fish from the North Western part of the Gulf, in order to assess their potential health impact on consumers.

## MATERIALS AND METHODS

Fish were caught at various locations in the Arabian Gulf (Table 1) and others were purchased from the local fish market. Liver, gills and muscle samples were each placed in aluminium foil and kept in clean dark glass bottles, labelled and stored at  $-80^{\circ}\text{C}$  until analysed. PAHs and AHs were extracted from fish as described previously (Al-Hassan et al 2000a ,2000b and 2001). All sample extractions were carried out in triplicate and the hydrocarbon concentrations as shown in tables are the average of the triplicate experiments. Statistical analysis was carried out using analysis of variance (ANOVA).

## RESULTS AND DISCUSSION

Twelve species of edible fish were collected and analysed for PAHs and AHs (Tables 2 and 3). Our earlier analysis attempts were focused on gills and liver of fish, as these organs could indicate the degree of hydrocarbon pollution in the marine environment. Since hydrocarbons were detected in appreciable amounts in both of these organs, muscle analysis was also included for some fish in the present study. A high concentration of total PAHs ( $13.52\text{ }\mu\text{g/g}$ ) was found in Silvery pomfret (Zubaidi) with the most potent carcinogen benzo [a] pyrene at a concentration of  $0.22\text{ }\mu\text{g/g}$  accumulated in its gills and  $1.78\text{ }\mu\text{g/g}$  of PAHs in its muscle. In this fish species, the concentration of total AHs was found to be  $37.74\mu\text{g/g}$ , which was the highest of all the fishes included in this study. Its liver contained  $23.33\text{ }\mu\text{g/g}$  of n- $\text{C}_{26}$  alkane. Although benzo [a] pyrene could not be detected in the muscle of this fish, a high level of this mutagenic PAH in its gills indicated the prevalence and persistence of this carcinogen in the Gulf waters . An elevated level of benzo [a] pyrene in gills and dibenzo [ah] anthracene, the second most potent carcinogen at  $1.26\text{ }\mu\text{g/g}$  in its liver may have adverse health effects for its consumers. Seabream (She'im), another popular fish among the Gulf inhabitants, showed benzo [a] pyrene at a level of  $0.72\text{ }\mu\text{g/g}$  in the muscle, while its total PAHs concentration in gills, muscle and liver was found to be  $11.85\text{ }\mu\text{g/g}$ . This was one of the highest amongst all the samples analysed in this study. Muscle of this fish had a high accumulation of PAHs at  $4.11\text{ }\mu\text{g/g}$ , indicating the vulnerability of this fish to PAHs burden and the health hazard it represents to its consumers. A total of  $17.50\text{ }\mu\text{g/g}$  of AHs in the liver, gills and muscle of this fish, was also among the highest level of aliphatic hydrocarbons of all the fishes included in this study. The second highest concentration of benzo [a] anthracene  $0.78\text{ }\mu\text{g/g}$  with total PAHs ( $11.65\text{ }\mu\text{g/g}$ ) was recorded for Silvery Grunt (Nagroor). The major portion of this carcinogenic PAH was concentrated in the liver of the fish. A ratio of PAHs/AHs has been used as a marker for toxicity (Pena-Mendez et al, 1999). A ratio of 3.8 PAHs/AHs indicated that this fish was probably worst affected by PAH carcinogens in the Gulf. The high ratio of PAHs/AHs reflected that the fish was exposed to an area where concentration of PAHs was greater than AHs leading to its higher accumulation of PAHs. It could also be possible that this species may not be metabolising PAHs efficiently, thus resulting in their accumulation. Mullet (Maid) showed the second highest ratio of PAHs/AHs of 2.3

**Table 1.** Sample collection locations.

Name of Fish	Scientific Name	Location of Sample	Date of Col	Latitude N	Longitude E
1 Red Snapper	<i>Lutjanus coccineus</i> (Cuvier & Valenciennes)	Garaoh	30 05 1997	280 52.92'	480 49.82'
2 Emperor Rubber Lip (Shaary)	<i>Lethrinus nebulosus</i> (Forsk.)	Garaoh	30 05 1997	280 52.92'	480 49.82'
3 Grouper (cat)	<i>Epinephelus areolatus</i> (Forsk.)	Garaoh	17 10 1997	280 52.92'	480 49.82'
4 Needlefish	<i>Tylosurus leiurus</i> (bleeker)	Al-Durrar oil field	12 06 1997	280 57.51'	490 08.45'
5 Silvery Croaker (Nuwaibi)	<i>Otolithes argenteus</i> (Cuvier & Valenciennes)	Yacht Club Salmiya	16 10 1997	290 21.41'	480 04.77'
6 Brown Spotted Grouper	<i>Epinephelus tauvina</i> (Forsk.)	Kuwait fish Market	07 12 1997	NA	NA
7 Silvery Pomfret (Zubaidi)	<i>Pampus argenteus</i> (Euphrasen)	Kuwait fish Market	07 12 1997	NA	NA
8 Silvery Grunt (Nagroor)	<i>Pomadasy Kaakan</i> (Forsk.)	Kuwait fish Market	07 12 1997	NA	NA
9 Yellow Finned Black Seabream (She'im)	<i>Acanthopagrus latus</i> (Houttuyn)	Kuwait fish Market	07 12 1997	NA	NA
10 River Shad (Soboor)	<i>Hilsa ilisha</i> (Hamilton-Buchanan)	Kuwait fish Market	07 12 1997	NA	NA
11 Mullet (Maid)	<i>Liza subviridis</i> (Valenciennes)	Kuwait fish Market	07 12 1997	NA	NA
12 Grooved Tiger Prawn	<i>Penaeus semisulcatus</i> (De Haan)	Kuwait fish Market	07 12 1997	NA	NA

NA = Not Applicable

**Table 2.** Levels of PAHs in edible fish ( $\mu\text{g/g}$ ).

Local Name	Organ	Naph	Flu	Phe	Anth	Fluor	Pyr	B[a]	Chry	B[b]F	B[k]F	B[a]P	Ind	Dibe	Benz	Total
1 Brown Spotted Grouper (Hammoor)	Liver	ND	ND	0.47	0.89	ND	0.38	ND	ND	ND	ND	ND	ND	ND	ND	1.74
	Muscle	1.06	0.33	0.29	0.62	0.12	0.21	ND	ND	ND	ND	ND	ND	ND	ND	2.63
	Gills	1.02	ND	0.54	0.89	0.38	1.44	ND	ND	ND	ND	ND	ND	ND	ND	4.27
	Total	2.08	0.33	1.3	2.4	0.5	2.02	ND	ND	ND	ND	ND	ND	ND	ND	8.63
2 Yellow Finned Black Seabream (She'im)	Liver	2.23	ND	0.27	0.54	0.1	0.16	0.2	0.21	ND	ND	ND	ND	ND	ND	3.71
	Muscle	0.64	ND	0.25	0.52	0.1	0.2	0.31	0.36	0.5	0.51	0.72	ND	ND	ND	4.11
	Gills	3.06	ND	0.34	ND	0.38	0.25	ND	ND	ND	ND	ND	ND	ND	ND	4.03
	Total	5.93	ND	0.86	1.05	0.57	0.61	0.51	0.56	0.5	0.51	0.72	ND	ND	ND	11.82
3 River Shad (Soboor)	Liver	1.54	0.35	0.41	0.75	0.15	0.27	ND	ND	ND	ND	ND	ND	ND	ND	3.47
	Muscle	ND	ND	0.34	0.66	0.11	0.22	ND	ND	ND	ND	ND	ND	ND	ND	1.33
	Gills	1.41	ND	0.32	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.73
	Total	2.95	0.35	1.07	1.41	0.26	0.49	ND	ND	ND	ND	ND	ND	ND	ND	6.53
4 Silvery Grunt (Nagroor)	Liver	1.12	ND	0.6	0.99	3.46	0.55	0.78	ND	ND	ND	ND	ND	ND	ND	7.5
	Muscle	0.73	0.24	0.18	0.69	0.17	0.24	ND	ND	ND	ND	ND	ND	ND	ND	2.25
	Gills	0.67	0.23	0.17	0.4	0.16	0.28	ND	ND	ND	ND	ND	ND	ND	ND	1.91
	Total	2.52	0.47	0.94	2.08	3.79	1.07	0.78	ND	ND	ND	ND	ND	ND	ND	11.65
5 Silvery Pomfret (Zubaidi)	Liver	0.81	2.1	0.69	1.06	0.59	0.59	0.39	0.51	ND	ND	ND	2.79	1.26	ND	10.79
	Muscle	ND	0.71	0.34	0.58	0.15	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.78
	Gills	ND	ND	0.29	0.2	0.24	ND	ND	ND	ND	ND	0.22	ND	ND	ND	0.95
	Total	0.81	2.82	1.31	1.84	0.98	0.59	0.39	0.51	ND	ND	0.22	2.79	1.26	ND	13.52

**Table 2.** (Cont.) Levels of PAHs in edible fish (ug/g).

Local Name	Organ	Naph	Flu	Phe	Anth	Fluor	Pyr	B[a]	Chry	B[b]	F	B[k]	F	B[a]	P	Ind	Dibe	Benz	Total
6 Mullet (Maid)	Muscle	1.1	0.25	0.19	0.47	0.15	0.27	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	2.43
	Gills	ND	ND	0.36	0.51	0.18	0.52	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.57
	Total	1.1	0.25	0.55	0.98	0.33	0.79	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	4
7 Shrimps	Muscle	1.08	ND	0.28	ND	0.18	0.23	ND	2.76	ND	2.87	ND	ND	ND	ND	ND	ND	ND	1.77
8 Grouper (Gato)	Muscle	ND	0.96	0.89	1.11	0.98	1.31	ND	2.76	ND	2.87	ND	ND	ND	ND	ND	ND	7.17	18.06
	Gills	ND	1.05	0.88	ND	ND	1.44	ND	2.76	3.72	ND	ND	ND	ND	ND	ND	ND	7.84	14.93
	Total	ND	2.01	1.77	1.11	0.98	2.75	ND	2.76	3.72	2.87	ND	ND	ND	ND	ND	ND	15.02	32.99
9 Red	Liver	ND	0.44	0.83	ND	1.17	ND	1.19	0.95	0.33	0.29	0.44	0.72	0.91	0.78	8.05			
Snapper (Hamra)	Gills	0.08	ND	0.62	ND	ND	1.18	0.77	0.73	ND	0.23	ND	0.61	0.67	0.59	5.48			
	Total	0.08	0.44	1.45	ND	1.17	1.18	1.96	1.68	0.33	0.52	0.44	1.33	1.58	1.37	13.53			
	Liver	0.59	0.85	1.97	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	3.41
Croaker (Nuwaibi)	Muscle	ND	ND	0.74	ND	1.11	1.21	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	3.06
	Gills	0.09	ND	0.68	ND	0.97	ND	0.8	0.78	0.29	0.24	ND	ND	ND	ND	ND	ND	ND	3.85
	Total	0.68	0.85	3.39	ND	2.08	1.21	0.8	0.78	0.29	0.24	ND	ND	ND	ND	ND	ND	ND	10.32
11 Needlefish (Hagool)	Liver	ND	0.37	0.7	ND	1.07	1.19	0.86	0.79	0.29	0.24	ND	0.58	0.65	ND	6.74			
	Gills	0.38	ND	0.66	ND	0.88	1.11	ND	0.69	0.26	0.22	ND	0.51	ND	0.59	5.3			
	Total	0.38	0.37	1.36	ND	1.95	2.3	0.86	1.49	0.56	0.47	ND	1.09	0.65	0.59	12.07			
12 Emperor	Liver	0.13	ND	1.09	ND	ND	ND	ND	ND	ND	0.47	ND	ND	ND	ND	1.69			
Rubber Lip (Shaary)	Gills	0.19	0.46	1.01	ND	1.42	ND	ND	1.08	ND	0.43	ND	ND	ND	ND	4.59			
	Total	0.32	0.46	2.1	ND	1.42	ND	ND	1.08	ND	0.9	ND	ND	ND	ND	6.28			

Gato; Hamra; Silvery Croaker (Nuwaibi); Hagool; Shaary are wet sample extractions while others are freeze dried samples  
Naph=Naphthalene; Flu=Fluorene; Phe=Phenanthrene; Anth=Anthracene; Fluor=Fluoranthene; Pyr=Pyrene; B[a]=Benzo  
[a] anthracene; Chry=Chrysene; B[b]=benzo[b]fluoranthene; Benzo [k] fluoranthene; B[a]P=Benzo[a]pyrene; Ind=Indeno[123-cd] pyrene;  
Diben=Dibenzo[an]anthracene; Benz=Benzo[ghi]perylene; ND = Not detected

**Table 3.** Concentration of alkanes in edible fish (µg/g).

Local Name	Org	C14	C15	C16	C17	C18	C19	C20	C21	C22	C23	C24	C25	C26	C27	C28	C29	C30	C31	C32	C33	Tot
1 Brown	Liv	0.17	0.13	0.1	0.23	0.08	0.08	0.3	0.21	0.34	0.27	0.37	0.27	0.82	0.7	0.21	0.31	0.28	0.18	ND	ND	5.06
Spotted	Mus	0.05	0.07	0.08	0.12	0.08	0.05	0.18	0.12	0.19	0.74	0.21	0.14	ND	ND	0.02	ND	ND	ND	0.12	ND	1.51
Groupers (Hammoor)	Gil	0.04	0.07	0.08	0.12	0.06	0.15	0.21	ND	1.3	0.05	0.09	ND	0.1	ND	ND	ND	0.05	ND	ND	ND	2.28
Tot		0.26	0.27	0.26	0.47	0.22	0.27	0.69	0.33	1.8	0.39	0.68	0.41	0.92	0.7	0.23	0.31	0.33	0.18	0.12	ND	8.85
2 Black	Liv	0.00	0.02	0.03	0.11	0.08	0.08	0.16	0.19	0.14	0.07	0.14	0.15	0.33	0.25	0.04	0.14	0.09	0.01	0.06	0.53	2.62
Seabream	Mus	0.1	0.17	0.14	0.22	0.1	0.1	0.17	ND	0.22	0.08	0.28	0.23	0.83	0.12	ND	0.13	0.11	0.12	ND	ND	3.14
(She'im)	Gil	0.29	0.28	0.19	0.2	5.26	ND	ND	ND	ND	ND	0.5	ND	2.08	1.58	0.53	0.32	0.26	0.18	0.08	ND	11.7
Tot		0.39	0.47	0.36	0.53	5.44	0.18	0.33	0.19	0.37	0.15	0.92	0.39	3.24	1.95	0.57	0.59	0.45	0.3	0.15	0.53	17.5
3 Silvery	Liv	0.04	0.08	0.11	0.26	0.1	0.06	0.14	0.08	0.12	0.02	0.04	0.1	ND	ND	ND	0.19	0.05	ND	ND	ND	1.28
Grunt	Mus	0.01	0.04	0.06	0.11	0.05	0.02	0.08	0.03	0.04	0.01	0.01	0.01	ND	ND	ND	ND	ND	ND	ND	ND	0.46
(Nagroor)	Gil	0.08	0.08	0.06	0.1	0.05	0.08	0.18	ND	0.12	0.08	0.14	0.09	0.12	0.05	ND	ND	0.09	ND	ND	ND	1.33
Tot		0.13	0.2	0.22	0.46	0.2	0.15	0.4	0.11	0.28	0.11	0.19	0.1	0.12	0.05	ND	0.19	0.14	ND	ND	ND	3.07
4 River	Liv	1.2	0.71	0.43	0.94	0.29	2.07	0.36	0.33	0.13	0.31	0.47	0.57	1.21	1.02	0.18	0.18	0.13	0.22	ND	ND	10.8
Shad	Mus	0.04	0.13	0.2	0.87	0.24	1.96	0.16	0.05	0.04	0.03	0.01	0.12	0.26	0.13	ND	ND	ND	ND	ND	ND	4.25
(Soboor)	Gil	0.54	0.6	0.48	2.36	0.47	3.96	0.18	0.07	0.04	0.06	0.15	0.31	0.46	0.52	ND	ND	ND	ND	ND	ND	10.2
Tot		1.78	1.45	1.1	4.18	1	7.99	0.7	0.45	0.21	0.4	0.63	1.01	1.93	1.67	0.18	0.18	0.13	0.22	ND	ND	25.2
5 Mullet	Mus	0.2	0.01	0.01	0.01	0.05	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.00	ND	ND	ND	ND	ND	ND	ND	0.12
(Maid)	Gil	0.01	0.03	0.07	0.25	0.08	0.11	0.1	0.06	0.02	ND	0.03	0.1	0.21	0.21	ND	0.09	0.12	0.08	0.05	ND	1.62
Tot		0.02	0.04	0.08	0.26	0.13	0.12	0.1	0.07	0.03	0.00	0.03	0.1	0.21	0.21	ND	0.09	0.12	0.08	0.05	ND	1.74
6 Shrimp	Mus	0.28	0.33	0.22	0.26	0.09	0.03	0.08	ND	0.04	0.01	0.02	ND	0.13	0.03	ND	ND	ND	ND	ND	ND	1.52
7 (Zubaidi)	Liv	0.85	0.91	0.99	0.69	0.09	0.64	0.38	0.87	0.45	0.39	1.47	1.52	23.3	2.68	2.48	ND	ND	ND	ND	ND	37.7

ND = Not detected; Org = Organ; Liv = Liver; Mus = Muscle; Gil = Gills; Tot = Total

with a total PAHs concentrations of 4.0 µg/g, with a potential health hazard for its consumers due to its high level of PAHs, PAHs/AHs ratio and total pyrene, which is the fourth most potent PAH recorded in the literature (Saeed et al. 1995). Among all the fish samples included in this study, Reef Grouper (Gato) showed the highest total concentration of PAHs at 32.99 µg/g, while Red Snapper (Hamra), showed the highest level of dibenzo [ah] anthracene.

In many of the analysed samples, two, three and four ring compounds were more common, compared to higher member ring compounds which is in agreement with a published report (Baumard et al. 1999). The most prevalent PAHs found in these samples were naphthalene, phenanthrene, anthracene, fluoranthene and pyrene. Krahn et al (1984) have reported that routine analyses for parent PAHs in fish, captured in polluted areas, often show only traces of PAHs, even when the sediments contained high concentration of these compounds. The values obtained in our study may reflect only part of the true concentration of the total PAHs, because of their ready metabolism by fish (Malins & Hodgins 1981). In most of the samples, the identified alkanes were homologues of C<sub>14</sub> to C<sub>30</sub> and a dominance of C<sub>18</sub> to C<sub>24</sub> n-alkanes suggested a relatively recent oil input. (Al-Hassan 2000a).

There are reports to show that the concentrations of total petroleum hydrocarbons in the Gulf coastal and offshore sediments and fish are increasing with time (Al-Omran and Rao, 1997; Al-Yaqoob et al. 1993). Our results undoubtedly support this contention and indicate that fish are acutely exposed to petroleum hydrocarbons in the Gulf. Our studies which involved pelagic fish (Zubaidi, Soboor, Maid and Needlefish), demersal fish (She'im and Sharks) (Al-Hassan et al. 2000a; 2000b) and shrimp and other bottom dwellers (Gato, Hamra, Nuwaibi, Nagroor, catfish) (Al-Hassan et al. 2001) have indicated that these marine organisms carry a noticeable concentration of PAHs and AHs, which can be hazardous to fish consumers. According to the results of this study, public awareness concerning risk to human health may be aroused in order to avoid adverse health effect to consumers of the Gulf seafood.

A highly significant correlation for accumulated PAHs in muscle, liver and gills was found ( $p = 0.0005$ ). In addition, results from fish samples were compared between themselves, in order to establish the significance of accumulated PAHs. A highly significant correlation in the accumulated PAHs could be established for gills ( $p = 0.004$ ) and muscle ( $p = 0.006$ ), while liver showed a significant relationship ( $p = 0.05$ ). This statistical analysis showed that these parts of fish accumulated PAHs.

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