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Research Article

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Aromatic Hydrocarbons as Implication from Fingerprints and Their Impact to The Oil Pollution in The Suez Gulf, Egypt

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Abstract

Chemical fingerprinting is an aspect of environmental forensic investigation which involves chemical analysis of contaminants and associated chemicals to provide source-specific information. In this research, ten fish species from the Suez Gulf region were subjected to the total concentration of 16-PAHs ranging from 81.499 to 5895.608 ng/g wet ng/g. wet wt., and chemical fingerprinting employing some of the quantitative diagnostic ratios with the aim of ascertaining the precise nature and source of the contaminants. PAHs fingerprinting involves the determination of a number of quantitative diagnostic ratios of source-specific marker PAH compounds. These quantitative diagnostic ratios may be used to distinguish petrogenic PAHs including phenanthrene/anthracene; benz (a) anthracene/chrysene; fluoranthene/pyrene; phenanthrene/(phenanthrene +anthracene) and indeno (1,2,3-cd) pyrene/indeno (1,2,3-cd) pyrene + benzo (ghi) perylene from other sources. It was found that the PAHs contamination in the Suez Gulf is not only emanating from petrogenic sources but other multiple sources contribute significantly.

Keywords: Aromatic Hydrocarbons Fingerprints, Oil Pollution, Suez Gulf, Egypt

Introduction

In recent years, oil pollution has become a global environmental issue in that oceanic ecosystems and inland aquatic breeding ecosystems are threatened greatly. The evaluation and prediction of the effects of oil pollution on the water environment have become a very urgent and important issue. It has been estimated that approximately 5 million tons of crude oil enter the marine environment each year from different sources [1]. The Suez Canal alone is crossed by some 20,000 vessels, annually, that carry about 14% of the world's trade water and the biological life within it. Oil on the water surface reduces the exchange of gases contributing to reduced levels of dissolved oxygen and promoting the absorption of solar energy that increases water temperature and reduces the solubility of oxygen. Some of the components of the oil are soluble in water, such as phenyls, and are directly toxic to marine life. These chemicals may cause physiological damage to fish and lead to premature mortality. Even at low concentrations, the fish may absorb hydrocarbons from the water so that the meat becomes 'tainted' and unfit for human consumption. The major impacts of oil pollution on fisheries and aquaculture are the smearing of nets and fish cages and the tainting of fish and shellfish, rending them unfit for marketing. As compared to the adult, which can avoid contaminated areas, the early developmental stages such as eggs, larvae, and juvenile fish, to oil in surface waters, is at higher risk [2].

PAHs comprise the largest class of chemical compounds known to be cancer-causing agents. Some, while not carcinogenic, may act as synergists [3]. They are classified among the Semi-Volatile Organic Compounds (SVOC) having boiling points greater than 200°C. There are several hundred different PAHs, which always occur as complex mixtures of different PAHs. Some PAHs are considered as a priority because they are supposed to be more harmful than the other PAHs, more information is available on them and there is a greater possibility of people being exposed to them. PAHs are highly stable and have a multiplicity of which could be broadly classified as diagenetic in origin, pyrogenic in origin, or petrogenic in origin [4]. Also, environmental awareness has grown tremendously, hence appropriate regulatory bodies both at national and international levels have continued to promulgate and implement stricter environmental regulations in this regard. This attribute is a very important tool in determining source-specific information for PAHs. Even though, molecular distribution of PAHs cannot be said to be 100% accurate, since factors can affect the molecular distribution over time, such as; evaporation, dissolution, photo-oxidation, and biodegradation which can lead to compound specific degradation [5]. In this research work, some of these diagnostic ratios to environmental samples from the Suez Gulf, Egypt were applied with the aim of ascertaining the source of PAHs contamination. Even though petroleum production and crude exportation are the primary activities done in the area, our findings show that there may be multiple sources of PAH contamination in the area.

Material and Methods

1. Ten aquatic species were collected from the Suez Gulf seashore and purchased from fishermen operating small crafts within a 15 km radius in these areas which extend from AL-Nasr Oil Company (NPC) up to Beach of oil pipeline region and the morphometry measurements were taken immediately (Table 1). Aquatic species samples were stored in ice wrapped in aluminum foil, frozen, and stored until analysis.
2. 20 g of fish sample was treated with 100 ml methanol and 3 g. KOH. The methanolic phase was extracted twice with n-hexane, dried over un-hydrous sodium sulfate. The extract was air-dried to constant weight. The oil content was calculated as ng/g = (A-B) × 10(9)/ weight of gram of sample, where: A & B are the weight of bottle after and before action.
3. High-Performance Liquid Chromatography Analysis (PAHs) identification and quantification in the extracted oil by using HPLC apparatus. Water HPLC 600, AutoSampler 616 Plus, Dual Absorbance Detector 2487, attached to the computerized system with Millennium 32; Software; HPLC Conditions: Sample Volume: (100µl), Column: Supelcosill LC-PAHs 15cm. 4.6 mm ID, 4µm particles, Mobile phase: Acetonitrile: water 60:40 and changed gradually to 100% acetonitrile within 20 min, Flow rate: 1.2 ml/min and Detector: u.v.254 mm.

Table 1: The ecological characteristics and recorded morphometric measures of examined Aquatic Species along Suez Gulf, Egypt

Code	Stations	Scientific name	English name	Feeding habits	Biotype complex	No.	Length (cm)	Body weight (g)
1	AL- Nasr Oil Company (NPC)	Sauridaundo Squamis	Brushtooth lizard fish	Carnivore (small fish)	Demersal, (benthic)	7	14	400
2	Outlet of Suez Oil Petroleum company (SOPC)	Euthynnus Affinis	Kawakawa	Feeds on small fish, squids, and sometimes zooplankton.	found in open waters but always close to the shoreline	1	40	350
3	Old Al-Kabanon	Rhabdosargus Haffara	Haffarasea bream	Feeds on benthic invertebrates. Consumed fresh.	Inhabits shallow waters, mainly around coral reefs, and over sandy or mud-sandy bottoms	4	13.5	420
4	New Al-Kabanon	Argyrops Spinifer	Porgies	Feeds on benthic invertebrates, mainly mollusks. Important food fish.	Inhabits a wide range of bottoms. Young fish occur in very shallow waters of sheltered bays; larger individuals in deeper water	4	15	480
5	Inlet of Suez Oil Petroleum Company (SOPC)	Nemipterus Japonicus	Japanese threadfin bream	Carnivore (small fish, invertebrates polychates)	Demersal	5	18	425
6	Atakah Harbor	Oreochromis Niloticus	Nile Tilapia	Herbivorous (feed on phytoplankton)	benthic and pelagic due to air bladder	3	14.5	389
7	Adabiya Harbor	Trachurus Indicus	Horse Mackerel	Carnivore (invertebrates and fish)	Pelagic	4	24	431

8	Suez Beach	Peneus Japonicas	Red mullets	Prey of small fish and crustaceans	inhabit the inshore area and coral reefs, can be found on a range of sea beds including sand, mud and coarse gravel	8	11.5	450
9	El- Sukhna of Loloha Beach	Scomber Japonicus	Chub mackerel, Pacific mackerel or blue mackerel	feed on copepods and other crustacean, fishes and squids	A coastal pelagic species, to a lesser extent epipelagic to mesopelagic over the continental slope	4	21	470
10	Beach of oil pipeline	Pomadasys Stridens	Striped piggy	Feeding on a variety of crustaceans, mollusks and small juvenile fishes, called a predator.	Living in the reef environment and sandy	3	13.5	495

Results and Discussion

Aromatic Hydrocarbons

Results for PAHs analysis in the ten fish species are summarized in Table 2. The distribution of PAHs studied shows different patterns for ten species studied. The total concentration of the PAHs varies in the range 81.499 to 5895.608 ng/g wet weight. The fish species *Oreochromis Niloticus* showed the lowest concentration 81.499 ng/g wet weight, while the highest (5895.608 ng/g wet weight) was found in *Argyrops Spinifer* species. The percent distribution of PAHs based on a number of aromatic rings indicated the predominance of 85.365 % for 2-3 rings PAHs in all studied species except with *Oreochromis Niloticus* species where the four-membered rings prevailed 52.712 % (Table 3, Fig. 1). Among the different components of PAHs, Acenaphthene (Ace) was the most abundant one with the highest value found in *Argyrops Spinifer* (2084.697 ng/g wet weight) and lowest value found in both of *Oreochromis Niloticus; Trachurus Indicus; Peneus Japonicas; and Scomber Japonicus species* where under limit of detection Table 2, with stark contrast found in species *Sauridaundo squamous, Rhabdosargus Ghaffar, Argyrops Spinifer,* and *Trachurus Indicus*, where Naphthalene (Nap) was the highest PAHs with concentrations 1493.244; 1045.162; 2060.756 and 1796.83 ng/g wet weight respectively. Generally, the LPAHs were highly prevailing compared with HPAHs. This finding is consistent with that reported by some authors [6,7].

Table 2: Concentration of individual PAHs (ng/g) in the oil extracted from the Aquatic Species

		Aquatic Species									
Concentration Compounds		1	2	3	4	5	6	7	8	9	10
2Ring	Nap.	1493.244	N.D	1045.162	2060.756	10.555	N.D	1127.310	N.D	N.D	N.D
3Ring	A.	401.046	279.120	175.435	431.046	352.039	N.D	N.D	N.D	N.D	N.D
	Ace.	1589.435	1752.503	2032.565	2084.697	527.6998	N.D	N.D	N.D	N.D	961.293
	Phe.	59.443	56.250	43.513	42.777	12.666	13.314	N.D	N.D	11.456	N.D
	F.	49.052	3.800	39.631	N.D	60.497	1.701	92.626	3.710	9.632	15.184
	Ant.	44.038	16.517	7.586	16.453	93.708	N.D	N.D	N.D	N.D	N.D
4Ring	Flu.	63.697	77.332	218.595	677.086	49.636	N.D	N.D	N.D	N.D	N.D
	Pyr.	53.081	670.942	137.369	111.297	45.192	38.198	102.970	6.039	30.447	N.D
	BaA	8.001	14.186	0.229	62.425	4.153	4.762	9.287	2.626	1.194	15.478
	Chr.	28.902	6.834	9.238	32.229	4.933	N.D	4.758	N.D	N.D	N.D
5Ring	Bbf	9.191	20.441	53.459	13.709	25.591	N.D	14.515	N.D	N.D	3.541
	Bkf	1843.138	N.D	N.D	N.D	N.D	N.D	977.388	N.D	N.D	425.290
	Bap	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D
	DahA	N.D	167.467	87.261	56.497	44.031	N.D	N.D	437.512	419.088	N.D
6Ring	Bp	22.430	108.721	25.067	262.979	129.266	N.D	N.D	51.484	N.D	N.D
	Ip	101.624	37.485	42.046	43.657	51.055	23.524	17.476	39.762	44.095	103.900
	Total PAHs	**5766.322**	**3211.599**	**3917.156**	**5895.608**	**1411.019**	**81.499**	**2346.330**	**541.134**	**515.910**	**1524.686**

Table 3: Percent of PAHs collected Aquatic Species based on the number of aromatic rings

S.no	Aquatic Species	2-3 ring %	4-ring%	5-ring %	6-ring%
1	*Sauridaundo squamis*	63.060	2.665	32.123	2.151
2	*Euthynnus affinis*	65..643	23.954	5.851	4.552
3	*Rhabdosargus haffar*	85.365	9.329	3.592	1.173
4	*Argyrops Spinifer*	78.630	14.978	1.191	5.201
5	*Nemipterus Japonicus*	74.922	7.364	4.936	12.779
6	*Oreochromis Niloticus*	18.424	52.712	0.0	28.864
7	*Trachurus Indicus*	51.993	4.988	42.275	0.745
8	*Peneus Japonicas*	0.686	1.932	80.851	16.862
9	*Scomber Japonicus*	4.087	6.135	81.233	8.547
10	*Pomadasys Stridens.*	64.044	1.015	28.126	6.815

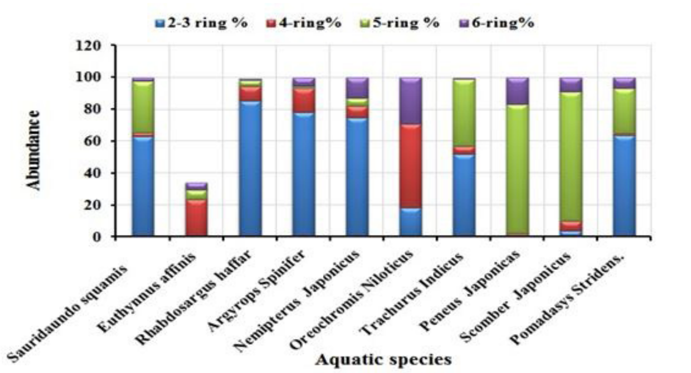

Figure 1: Percent of PAHs collected Aquatic Species based on the number of aromatic rings

Chemical Fingerprinting of Aromatic Hydrocarbons

Chemical fingerprinting is very important to effectively determine and identify the sources and origin of PAH contamination, making use of different diagnostic ratios. But for the reliability of the fingerprinting results, quantization of PAHs was focused on sixteen priority PAHs and in the same way the quantitative diagnostic ratios used in this study for the determination of source-specific information of PAHs were restricted to those sixteen. For the identification of the PAHs origin in Aquatic Species, the ratios of low molecular weight PAHs to high molecular weight PAHs (LPAHs/HPAHs), phenanthrene to anthracene (Phe/Ant), fluoranthene to pyrene (Flu/Pyr), fluoranthene to fluoranthene plus pyrene (Flu/(Flu+Pyr)) and anthracene to anthracene plus phenanthrene (Ant/(Ant+Phe)) were examined by several authors [8-12].

PAH ratios of Aquatic Species are presented in Table 4. The LPAHs/HPAHs ratio <1 and Flu/ (Flu+Pyr) ratio >0.4 calculated for the ten Aquatic Species suggest petrogenic sources for T*rachurus Indicus* and *Pomadasys Stridens* while other Species was near the boundary between pyrogenic and petrogenic sources. In contrast the Flu/Pyr ratio < 1, which indicates petrogenic origin, was recorded at *Nemipterus Japonicus, Oreochromis Niloticus; Trachurus Indicus; Peneus Japonicas; Scomber Japonicas';* and *Pomadasys Stridens*, while Flu/Pyr ratio > 1 suggesting pyrogenic origin was recorded at Species *Sauridaundo squamous; Euthynnus affinis; Rhabdosargus haffara* and *Argyrops Spinifer.* In addition, Phe/Ant ratio <10 and Ant/(Ant+Phe) ratio >1 indicating petrogenic sources was found at all Species except at *Sauridaundo squamis; Euthynnus affinis and Nemipterus Japonicus*, suggest mixed pyrogenic and petrogenic sources. For better determination and demonstration LPAHs/HPAHs ratio against Flu/Pyr ratio, Phe/Ant ratio against Flu/Pyr ratio, Ant/ (Ant+Phe) against Flu/Pyr and Ant/ (Ant+Phe) against Flu/ (Flu+Pyr) were plotted. Different PAH cross plot ratios of Aquatic Species are presented in Table 5. The PAH cross plots of the ratios of LPAHs/HPAHs against Flu/Pyr indicate different PAH origins (Figure 2a). Aquatic Species *Nemipterus Japonicus, Trachurus Indicus* and *Pomadasys Stridens* suggest ptrogenic origin while, LPAHs/HPAHs ratio<1 and Flu/Py ratio >1 suggest pyrogenic origin, while LPAHs/HPAHs ratio <1 and Flu/Py ratio <1 suggest mixed origin (pyrogenic and petrogenic) with the rest of Aquatic Species.

The origin of PAHs in Aquatic Species was also examined by plotting Phe/Ant ratio against Flu/Py ratio (Figure 2b). The Phe/Ant ratio<10 and Flu/Py ratio >1 found in *Nemipterus Japonicus, Oreochromis Niloticus; Trachurus Indicus; Peneus Japonicas; Scomber Japonicus;* and *Pomadasys Stridens*; suggest contamination from combustion processes, confirmed by a cross plot of LPAHs/HPAHs against Flu/Py, while Phe/Ant ratio > 10 and Flu/Py ratio < 1 found *Sauridaundo squamous; Euthynnus affinis; Rhabdosargus haftarah* and *Argyrops Spinifer*; confirmed mixed origin beside, both of *Argyrops Spinifer* and *Rhabdosargus haftarah* indicates the predominance of mixed origin.

In order to determine PAH sources in Aquatic Species, where different ratios and plots show mixed origin for *Euthynnus affinis; Rhabdosargus haffara* and *Argyrops Spinifer*; the ratios Ant/(Ant+Phe) against Flu/(Flu+Py) were plotted (Figure 2c). The cross plot of the Ant/(Ant+Phe) versus Flu/(Flu/Py) ratio reveal that S*auridaundo squamis* and *Nemipterus Japonicus*, indicate mainly pyrogenic origin where, *Oreochromis Niloticus; Trachurus Indicus; Peneus Japonicas; Scomber Japonicus;* and *Pomadasys Stridens* suggest petrogenic origin.

Table 4: PAHs parameters ((ng/g wet weight) used to detect the sources of PAHs in the Aquatic Species

S.no	Aquatic Species	Phe/Ant	Flu/Pyr	Flu /(Flu + Pyr)	Ant/(Ant + Phe)	BaA/(BaA + Chr)	IP/(IP-+BP)	6LPAHs	10HPAHs	Σ6LPAHs/ Σ10HPAHs
1	Sauridaundo squamis	1.350	1.446	0.545	0.409	0.217	0.819	2130.053	3636.259	1.707
2	Euthynnus affinis	3.406	4.682	0.103	0.176	0.675	0.256	1103.405	2108.191	1.911
3	Rhabdosargus haffar	5.736	28.816	2.102	0.034	0.024	0.626	573.264	3343.892	5.833
4	Argyrops Spinifer	2.600	41.148	0.859	0.024	0.660	0. 142	1259.879	4635.729	3.680
5	Nemipterus Japonicus	0.135	0.530	0.523	0.654	0.457	0.283	353.856	1057.163	2.988
6	Oreochromis Niloticus	ND	ND	ND	ND	1.0	1.0	66.484	15.015	0.225
7	Trachurus Indicus	ND	ND	ND	ND	0.661	1.0	1126.394	1219.936	1.083
8	Peneus Japonicas	ND	ND	ND	ND	1.0	0.436	537.424	3.710	0.007
9	Scomber Japonicus	ND	ND	ND	ND	1.0	1.0	494.823	21.087	0.0436
10	Pomadasys Stridens.	ND	ND	ND	ND	1.0	1.0	548.212	976.474	1.781

ND: under the limit of detection, concentration ng/L, LPAHs/ HPAHs >1 petrogenic, <1pyrogenic, Flu/ Flu+pyr <0.4 petrogenic >0.4 pyrogenic, Ant/ Ant+phe <0.1 petrogenic, >0.1 pyrogenic, Flu/pyr <1 petrogenic, >1 pyrogenic, Phe/Ant <10 petrogenic,>10 pyrogenic, BaA/BaA+Chr <0.2 petrogenic, >0.35 pyrogenic, Flu/Flu+pyr 0.4-0.5 petrogenic,>0.5 pyrogenic, Ipy/Ipy+Bp <0.2 petrogenic,>0.2pyrogenic, Ip/ Ipy+BP 0.2-0.5 petrogenic,>0.5 pyrogenic.Ant: Anthracene,Phe: Phenanthrene, BaA: Benzo[a] anthracene, Chr: Chrysene, Flu: Fluoranthene, BP: Benzo [g,h,i] perylene; BaP Benzo [a] pyrene, LPAHs: Low molecular weight PAHs, HPAHs: High molecular weight PAHs; IP: Indo [1, 2, 3-cd] perlyene; Pyr: Pyrene [24,25].

Table 5: Determination of PAH pollution sources of in aquatic species

		Ratio		
S. no	**Aquatic Species**	**LPAHs/ HPAHs vs Flu/pyr**	**Phe/Ant vs. Flu/pyr**	**Ant/Ant+phe vs. Flu / Flu+pyr**
1	Sauridaundo squamis	Mix.	Mix.	Pyro
2	Euthynnus affinis	Mix.	Mix.	Mix.
3	Rhabdosargus haffara	Mix.	Mix.	Mix.
4	Argyrops Spinifer	Mix.	Mix.	Mix.
5	Nemipterus Japonicus	Petro	Petro	Pyro
6	Oreochromis Niloticus	Mix.	Petro	Petro
7	Trachurus Indicus	Petro	Petro	Petro
8	Peneus Japonicas	Mix.	Petro	Petro
9	Scomber Japonicus	Mix.	Petro	Petro
10	Pomadasys Stridens	Petro	Petro	Petro

vs.: versus; **Pyro:** Pyrogenic; **Mix:** Mixed; **Petro:** Petrogenic.

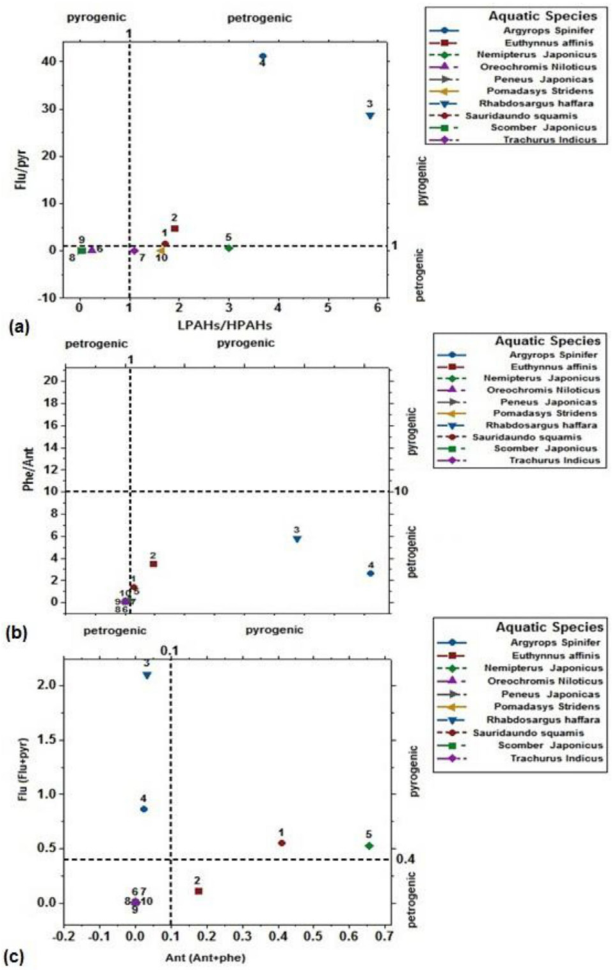

Figure 2: Cross plot of the Flu/Pyr versus LPAHs/HPAHs ratio (a), Phe/Ant versus Flu/Pyr ratio (b) and Ant/ (Ant+Phe) versus Flu/ (Flu/Pyr ratio, c)

Conclusions

In this study, 16 polycyclic aromatic hydrocarbons (PAHs) were tested in ten fish species collected from Suez Gulf indicated that:

1. Total average levels in fish species ranged from 81.499 to 5895.608 ng/g wet. The compositions of PAHs determined in all samples were measured in order to use them as chemical fingerprinting for identifying different sources of PAHs pollutants in the studied region.
2. The sources of PAH contamination could contribute to different degrees is either petrogenic, biogenic or mixed petrogenic and biogenic, hence opening up a new argument on how to determine sources of PAH contamination when there are multiple sources and how to apportion the contribution of each source.
3. This study provides useful information about the extent and source of PAHs pollutants in different fish species from the Gulf of Suez, which will be helpful for future comparison and recommended that a continuous monitoring program for the Suez Gulf should be formulated and conducted to ensure that the concentrations of petroleum hydrocarbons are within the Baseline levels.

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