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Mohammed Orif, Amr El-Maradny*

Bio-accumulation of Polycyclic Aromatic Hydrocarbons in the Grey Mangrove (*Avicennia marina*) along Arabian Gulf, Saudi Coast

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Abstract: The Arabian Gulf is considered as one of the most important sources for the crude oil all over the world. Due to the vast oil exploration and exploitation, huge amounts of organic pollutants infiltrate to the gulf. An important class of organic pollutants is polycyclic aromatic hydrocarbons (PAHs). One of the marine habitats in Arabian Gulf area is the mangrove stands, that are undoubtedly impacted by all anthropogenic factors like oil industries and sewage discharge. In the monitoring framework for mangrove ecosystem along Saudi coasts, nine mangrove stands were examined for the accumulation of PAHs in the Arabian Gulf coast. PAHs were measured using Gas Chromatography-Mass Spectrometry. The mean values detected for total PAHs in mangrove sediments, roots and leaf were 105.39, 680.0 and 282.4 ng/g, respectively. The trend of total PAHs concentrations in all sites showed the descending order: roots > leaf > sediments. Despite the sandy nature and low organic carbon contents of the mangrove sediments, moderate values of PAHs were detected in the major sites. PAH bio-accumulation factors for roots are higher than that in leaf. The diagnostic ratios revealed that the sources of PAHs are mainly pyrogenic, except for Damam and Damam Port that were found to be petrogenic.

Keywords: Arabian Gulf; *Avicennia marina*; bio-accumulation; Mangrove; polycyclic aromatic hydrocarbons (PAHs).

***Corresponding author: Amr El-Maradny,** Faculty of Marine Sciences, King Abdulaziz University, P.O. Box 80207, Jeddah 21589, Saudi Arabia; National Institute of Oceanography and Fisheries, Qait Bay, Alexandria, Egypt, E-mail: amaradny@hotmail.com
Mohammed Orif: Faculty of Marine Sciences, King Abdulaziz University, P.O. Box 80207, Jeddah 21589, Saudi Arabia

1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous anthropogenic pollutants [1] characterized by high lipophilicity (accumulate in food chains) [2], high hydrophobicity (accumulate in sediments) [3]. Therefore, sediment could be reservoir and source for these pollutants [4]. PAHs also characterized by high toxicity, high stability and high resistance to microbial degradation [5]. Due to the probable cancer risk and mutagenic properties for PAHs; USEPA was listed 16 compounds of PAHs in the priority pollutants list [6]. Urban coastal areas often show elevated levels of PAHs [7,8]. In addition to PAHs biogenic origin, there are two main anthropogenic sources for PAHs which are: pyrogenic that generated from incomplete combustion of foil, and petrogenic that derived directly from crude oil [9].

The Arabian Gulf is a major production area for oil and gas, not only in the Middle East but also all over the world; where almost two third of the oil resources of the world is found in this Gulf [10]. In addition to tremendous stress exerted to the marine environment due to the production and transportation of oil in this area; during 1991, the Gulf was impacted by one of the largest oil spills in history (10.8 million barrels of crude oil were deliberately spilled in the Gulf) [11], synchronously with large amount of ash input to the Gulf due to blowouts of about 8 million barrels and fires in Kuwait oil fields [12]. The environmental recovery of such huge accidents may extend for decades. Other moderate and small oil spill accidents surely occurred; as in Deylam (Iran) (west northern of the Gulf) during 2012, where oil leakage covered 160 km^2 square area of surface water in the Gulf [13]. That is beside to urban sewage disposal, harbor activates, atmospheric deposition which are considered as source points for all kinds of organic pollutants include PAHs to the marine environment.

Mangroves are intertidal wetland plants usually found in tropical and subtropical coastal areas. The biological importance of mangroves is to provide foods for marine animals as the primary producers do, meanwhile several

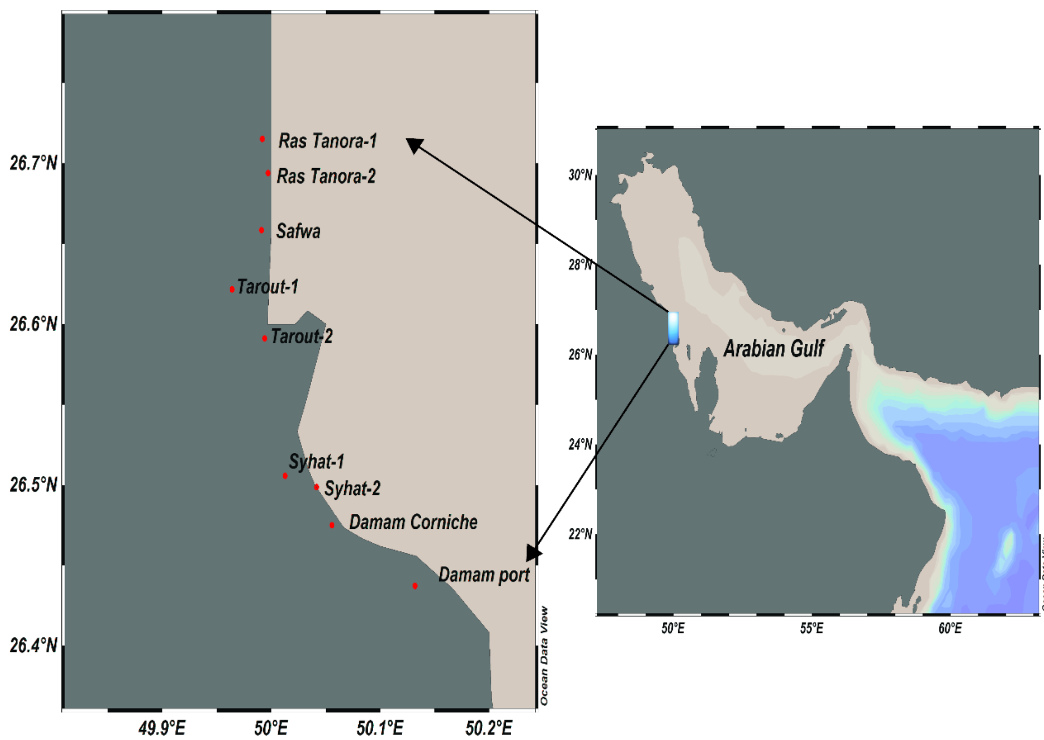

Figure 1: The map of the nine sampling locations of mangrove stands along Arabian Gulf.

kinds of organic pollutants like PAHs can accumulate in mangroves due to its unique characters such as high productivity and organic carbon content [14]. The levels of PAHs in contaminated mangrove swamps could exceed 10,000 ng/g [15]. Moreover, the levels of PAHs detected in the sediments of contaminated mangrove swamps were found to be higher than the levels in marine harbors, where it acts as security guard protecting the coastal line from organic pollutants [16]. PAHs could uptake from the sediment to leaf via mangrove roots. The accumulated PAHs in mangrove transfer to higher trophic levels through food web and could accumulate in animals and humane [17].

Despite the dangerous environmental activities in the Arabian Gulf, most of the studies were done to assess the PAHs levels in the Gulf sediments [18-21] and rarely extend to seawater [13]; with negligence for the marine habitats like mangrove stands that are definitely affected seriously by the accumulation of PAHs. The present work aims to study the concentration levels of the priority 16 PAHs and its methylated derivatives in the surface sediments, roots and leaf of mangrove stands for 9 selected sites in the Arabian Gulf-Saudi coast. The study will extend to evaluate the bio-accumulation of PAHs in mangrove plants. Moreover, the PAHs origin will be investigated based on two diagnostic ratios.

2 Material and Methods:

2.1 Study Area

The Arabian Gulf is located between Arabian Peninsula and Iran. The Saudi Arabian coastline of the Arabian Gulf extends for about 450 km. *Avicenna marina* is the only mangrove species existing in the Gulf. The present study cover mangrove stands in the Saudi coast of Arabia Gulf extended from 26° 42’ 54.616” N to 26° 26’ 14.69”N and from 50° 1’ 18.356” E to 50° 7’ 55.48”E. Some sites are located in remote areas far from direct anthropogenic effects (1, 5), while other sites directly affected by different anthropogenic sources like station 7 near sewage disposal area and sites 9 and 8 that are the Damam port and adjacent area which influenced by harbor activities. Mangrove samples mean heights were around 2.5-3.0 m and its age was estimated to be 25-30 years. The growth conditions of all swamps are comparable, therefore significant changes of PAHs contamination due to this factor is excluded. Figure 1 shows the map locations for the nine mangrove samples along the Arabian Gulf-Saudi coast.

2.2 Samples collection and pre-treatment:

Approximately the top 0-5 cm of sediments were collected from each studied stand and placed in aluminum bags, refrigerated, and transported to the laboratory within 8 hours after collection and kept frozen in -20°C. Plant samples were washed by tap water followed by deionized water before being freeze-dried. The dried plant samples were grounded into powder using a blender. The sediment samples were freeze-dried for 24 hours, grounded into fine powder using a pestle and mortar, and filtered through 80-mesh screen-sieve. All samples (sediments and plants) were transferred to glass bottles and kept in 4°C refrigerator for further analysis.

2.3 Organic carbon and grain size analysis

Total organic carbon in the sediments was analyzed by wet dichromate- sulfuric acid oxidation method [22]. Standard dry sieving technique was used for sediment grain size fractions, where classified as fine mud (< 0.063 mm), sand (0.063 – 2 mm) and gravel (> 2 mm) [23].

2.4 Extraction and purification of PAHs

Around 20 g of each sediment sample (5 g for leaf; 5 g for root) was subjected to a soxhlet extraction using 300 ml dichloromethane (DCM) for 24 h. Sulfur was removed by the addition of metallic copper. All samples were spiked with 250µL deuterated surrogate standard mixture (naphthalene-d8, phenanthrene-d10, and chrysene-d12) before extraction [24]. The crude extract was concentrated on a rotavapor at 35°C. Particulate impurities for plant samples were filtered. The extracts were concentrated to approximately 1 ml and transferred to silica column for cleanup and fractionation. The column was packed from top and bottom with pre-combusted anhydrous Na_2SO_4 at 450°C for 6 hours. Silica gel (230-400 mesh) was activated at 230°C for 12 hours then partially deactivated with 5% deionized water. The elution was done using n-hexane for aliphatic fraction followed by n-hexane/DCM (70:30 v/v) for PAH fraction [25]. PAH fractions were concentrated using gentle stream of pure N_2 gas to nearly 1 ml DCM. Deuterated internal standard mixture (acenaphthene-d10, flourene-d10 and perylene-d12) (100 µl, 5 ppm) was added just before injection to GC-MS.

2.5 Identification and Quantification of PAHs

GC-MS (Schimatzu 2010) with DB-5MS column (30 m* 0.25 µm, RTX) was used for PAHs analysis. The initial temperature programmed at 100°C with 1-minute hold, and then ramped at 6°C/minute to 300°C then held for 3 minutes. The electron energy of the mass spectrometer was 70 eV. Individual PAHs were identified based on both retention time and mass spectrum of selected ions with the external calibrated standards for the priority 16 PAH parent targets. Methylated PAH homologs quantified using the response factor of the corresponding non-methylated parent PAH [26].

2.6 Calculation of Bio-accumulation Factors:

The bio-accumulation factor (BAF) for roots and leaf was calculated by the formula: $BAF_{root} = C_{root}/CS$ and $BAF_{leaf} = C_{leaf}/CS$, where C_{root} and C_{leaf} are the concentration of total PAHs in mangrove roots and leaf, respectively, and CS = concentration of PAHs in sediment [27].

2.7 Quality control and quality assurance

Duplicate samples (10% of the analyzed samples) and procedural blanks (1 blank for each 5 samples) were performed at the same time with analysis. At the beginning of each working day, calibration standards were run before each sample batch to establish the calibration curves for PAHs. Before extraction, all samples were spiked with surrogate deuterated mixture for recovery calculations. Before GC-MS injection, deuterated internal standard mixture was added for all samples. The recoveries of samples were ranged between 69-102% with RSD% < 19%. The concentrations in the procedural blanks were no more than 3 times the method detection limit. Detection limits (DL) were calculated through five-point calibration curve for standers and extrapolated for determining the y axis intercepts [28]. All results were expressed as dry weight basis, and those samples with concentrations less than DL were reported as not detected (ND).

Ethical approval: The conducted research is not related to either human or animals use.

Table 1: Sediment characteristics for the studied mangrove stands along Arabian Gulf.

Site no.	Site name	Gravel%	Sand%	Mud%	$CaCO_3$%	TOC%	Textural Classification
1	Ras Tanora 1	3.8	91.1	5.2	24.7	1.71	Sand
2	Ras Tanora 2	6.2	92.7	1.1	35.2	2.86	Sand
3	Safwa	0.4	91.9	7.7	38.4	1.76	Sand
4	Tarout 1	0.6	93	6.5	31.2	2.69	Sand
5	Tarout 2	10.9	88.4	0.7	35.9	3.9	Gravel sand
6	Syhat 1	8.6	87.2	4.3	31.2	2.86	Slightly gravel sand
7	Syhate 2	0.8	90.5	8.8	30.9	4.55	Sand
8	Damam Corniche	0.4	91.9	7.7	34.5	2.31	Sand
9	Damam Port	8.9	88.6	2.4	22.9	2.01	Slightly gravel sand

3 Results and discussion

3.1 Levels of PAHs in mangrove stands:

Mangrove swamps protect coastal areas from erosion and storm waves. Meanwhile, mangrove trees act as an ocean safeguard from different pollutants [29]. It grows throughout the world, in the tropics and subtropics areas with low-oxygen soil, where slow-moving waters allow fine sediments to accumulate, and are adapted to life in harsh coastal systems. However, mangrove plants are able to grow in variable compositions of silt, clay and sand sediments contents [30]. Although several biogeochemical processes are influential in mangrove swamps to host organic pollutants [31], sediment particle size is principle factor affecting the levels of all organic pollutants [32]. Table 1 represents proportional ratios for gravel, sand, mud, $CaCO_3$, in addition to total organic carbon (TOC) ratios in the mangrove sediments for the nine studied sites. The textural classification of sediments in the present study were found mainly sandy (sand range: 87.2-93.0%) with small surface area that diminish adsorption of organic pollutants [33]. Although low values of total organic carbon were detected in the mangrove sediments (range: 1.71-4.55%) that were consistent with the sandy nature; gradual increase for TOC % was recognized though northern sites to reach maximum at site 7 (Syhate 2) that affected by sewage drainage.

Among the priority 16 inspected PAH congeners; 7 parent PAHs were detected either in sediments or in the mangrove plants which are: naphthalene, acenaphthene, phenanthrene, chrysene, fluoranthene, pyrene and benzo[k]fluorathene; while the rest of congeners remained below detection limit. Figure 2 represents the total PAHs concentrations in the nine studied mangrove stands (sediments, roots and leaf) along the Saudi coast of Arabian Gulf. The total PAHs concentration in sediment samples ranged from 4.39 to 533.40 ng/g with an average value of 105.39 ng/g.

These values were found comparable with the moderate values of PAHs detected in the mangrove sediments from the northern Arabian Gulf (Iran) (ranged from 75.24 to 581.94 ng/g) [34], Shantou mangrove wetlands (China) (ranged from 57 to 238 ng/ g) [35] and the Sunderban wetlands (Bay of Bengal, India) (ranged from 20 to 839 ng/g) [36].While PAH values in the mangrove sediments in the present study seem to be relatively lower than the extremely high PAHs levels detected in highly polluted mangrove sediments for two swamps in Hong Kong (average: 11098 ng/g; 3785 ng/g) [15, 37] and in Shenzhen, China (average: 4480 ng/g) [38]. On the other hand, the detected levels in the present study seem extremely higher than the levels detected in mangrove sediments in the regional area along eastern side of Red Sea coast-Saudi Arabia (ranged from 1.06 to 6.97 ng/g with an average value of 2.98 ng/g) [39].

Since the coastal areas of the Arabian Gulf are the largest source of crude oil and natural gases all over the world, and apart from mangrove stands; PAHs were studied in the sediments of different areas around Arabian Gulf extensively in the west northern part of the Gulf (Iran); recording very high values that ranged from 1,054 to 17,448 ng/g and 42.76 to 5596.49 ng/g in the industrial areas of Asaluyeh harbor [19] and Rural area [18] (Iran), respectively. While the moderate values recorded in Deylam, Busheher (Iran) (ranged between 15.3 to 759 ng/g) [13] and Qatar exclusive economic zone (ranged from 2.6 to 1025 ng/g, average: 117.3) [21] are comparable with the levels in the present study. It is worth to mention that the sediment core studied within Kuwait Bay for the depositional history of PAHs to the Arabian Gulf indicate low values fluctuated between 12 and 25 ng/g before 1970 [20].

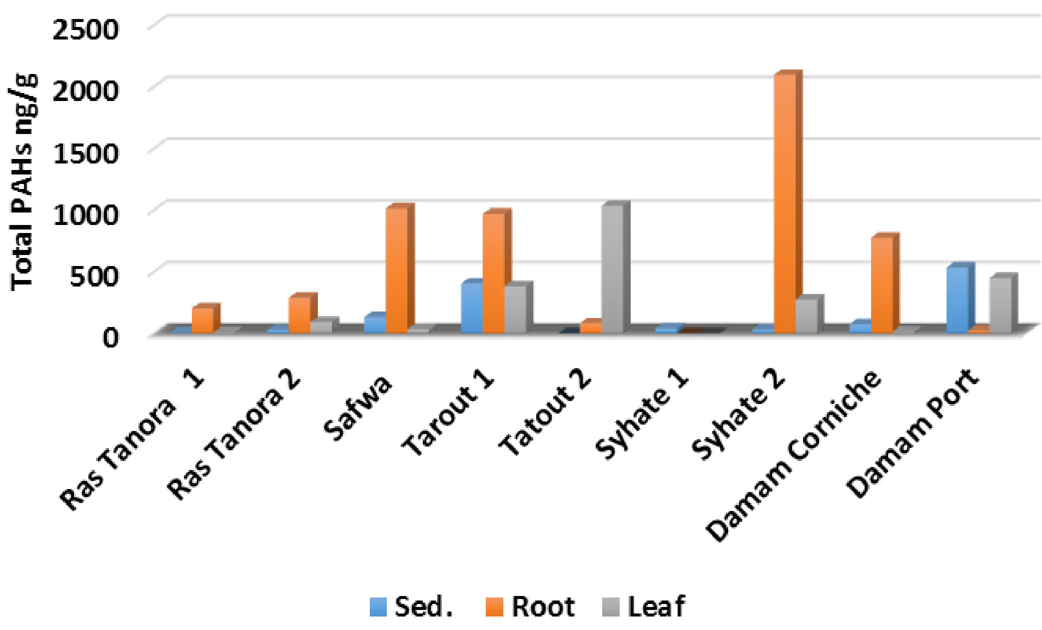

Figure 2: Total PAH concentrations (ng/g) for sediment, roots and leaf in the nine sites of the studied mangrove stands along Arabian Gulf.

Despite geochemical factors in the study area such as small particle size and low total organic carbon content, exclude hosting high concentrations of PAHs in the sediments; sites 3 and 4 showed moderate levels of total PAHs, in addition to site 9 (Damam port) that recorded the highest total PAH value (533.4 ng/g) which can be attributed to port activities like daily loading and washing of oil tankers, oil spills and leakages, and shipping wastes. Fluorancene appears as the highest detected individual parent congener (132.5 ng/g) in the sediment at site 9 (Damam port), while trimethyl phenanthrene recorded the highest concentration for methylated compound at site 3 (Safwa) (68.48 ng/g). PAH ring composition pattern covers 2, 3, 4 and 5 ring congeners. Average methylated congeners (mono-, di-, and tri-methyl) represented 22.5, 53.3, and 52.7% from total PAHs in the sediments, roots and leaf, respectively. Methylated PAHs ratios are distinctly increased in the mangrove roots and leaf; these compounds sometimes are more toxic than their parent PAHs, and tend to accumulate more than parent congeners [3,26]. According to sediment quality guidelines (SQG), individual and total PAHs detected in the present study were found below the effect range low (ERL) [40]; hence, adverse biological effects hardly to occur.

Except sites 5, 6 and 9, mangrove roots showed significant increase in the total PAHs levels (Figure 2), where roots values ranged from 22.7 to 2096.36 ng/g with an average of 680 ng/g; while leaf recording range between 7.3 to 1033.2 ng/g with an average of 282.40 ng/g. The trend of total PAHs concentrations for the major sites showed the descending order: roots > leaf > sediments. The observed accumulation trend of PAHs in this study is compatible with the mangrove swamps in Mumbai (India) [16]. In contrary, the highest levels of PAHs were detected in the mangrove leaf followed by roots then sediments in Shenzhen (Chania) [38], and eastern coast of Red Sea [39].

3.2 Bio-accumulation of PAHs in mangrove plant

The substantial mechanism for accumulation of PAHs in mangrove plant take place through sediment/root uptake [41,42]. Factors like sediment particle size around roots [43], the water solubility of PAHs and n-octanol-water partition coefficient (Kow) control the plant uptake of PAHs [44]. Another possible mechanism is leaf/atmospheric uptake, where PAHs horn to mangrove through the stomata of the large surface areas of mangrove leaf that usually covered with thick waxy layer, which can accumulate the lipophilic PAHs from atmosphere. The mean controlling factor in this mechanism is the volatility of PAH congeners and its partitioning between gaseous and practical forms [43]. It should be noted that both mechanisms are possible, that may lead to unsystematic correlations between PAHs in sediments and mangrove leaf and roots occasionally [45], as in the sites 5, 6 and 9 in the present study that showed

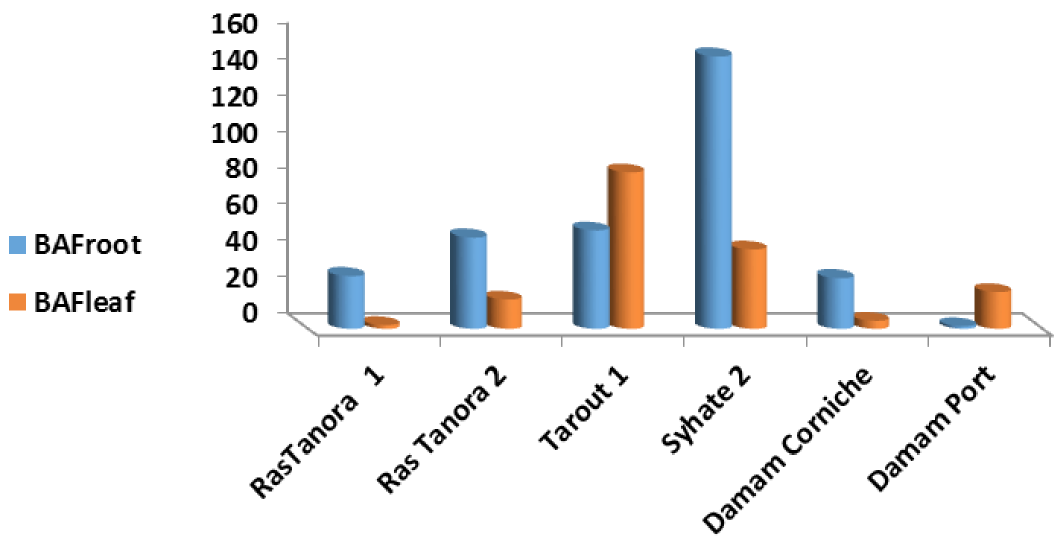

Figure 3: Bio-accumulation factors in roots and leaf for phenanthrene in mangrove plants in the study area.

different pattern from the major trend (Figures 2 and 3).

Bio-accumulation factors for roots (BAF_{root}) and leaf (BAF_{leaf}) were calculated for phenanthrene congener that was detected in most mangrove sediments, roots and leaf (Figure 3). Values of bio-accumulation factor (BAF)>1 mean that an organism absorbs PAH congener at a rate faster than that at which the substance is lost by catabolism and excretion. The results of the regression analysis for roots / sediments indicated p-values ≤ 0.05 that confirm a significant correlation for phenanthrene, harmonize with Sediment / root mechanism. BAF_{root} and BAF_{leaf} values elucidate high accumulation in the major sites (Figure 3) and the highest value for BAF_{root} (150.48) was recorded at site 7 (Syhate 2) which affected by sewage derange; while the highest value for BAF_{leaf} (86.50) was recorded at site 4 (Tarout 1) may support the presence of both mechanisms for phenanthrene accumulation in this site [42,46]. BAF_{root} and BAF_{leaf} values recorded in the present study for phenanthrene in mangrove roots and leaf are 60 and 12 times, respectively, higher than the accumulation factors recorded at the eastern coast of Red Sea [39].

It is worth to mention that, except for sites 3, 4 and 5; naphthalene and its methylated derivate were detected in the sediments of the rest sites, while they are absent in the corresponding roots and leaf. This phenomenon can be attributed to the extremely high temperatures that the Gulf area is exposed to all over the year. Despite the relative high hydrophilicity of these light congener, the high temperatures exceed solubility factor and allow these congener to evaporate and escape from sediments before absorption by roots. Hence, the harsh environmental conditions for the mangrove stands may prevent the bio-accumulation of some organic pollutants.

3.3 The probable origin of PAHs in the studied mangrove stands

The anthropogenic activities related to pyrogenic and petrogenic processes are the common sources for PAHs in the marine environment [47]. Discrimination between different origins of PAH mixture is highly difficult due to the complexity governing PAHs distribution in marine environment and co-exist of the compounds from both origins. Numerous studies presented at least 13 diagnostic ratios that were applied to distinguish between PAH origins [9,47,48]. Some diagnostic ratios are based on parent PAHs, others on the proportions of alkyl-substituted to non-substituted molecules. However, in the present study, based on the abandance of different congeners, two diagnostic ratios were selected to discriminate PAH origins (Table 2). Values higher than 2 for methylated phenanthrene/phenanthrene (MP/P) ratio indicate petrogenic origin, while it pyrogenic origin is indicated if the ratio less than one [48]. On the other hand, values higher than 0.22 indicate petrogenic origin if pyrogenic index is applied, while it indicates pyrogenic origin when the ratio less than 0.8 [9]. MP/P values for sites 8 (Damam) and 9 (Damam port) recorded 11.90 and 5.91 respectively, which are the only values which exceeded 2 among all sites, and pointed out to petrogenic origin, while this index ratio for all other sites showed pyrogenic origin. These results are expected, where sites 8 (Damam) and 9 (Damam port) are the closest sites to the oil wells, in addition to the other port activities like discharges of the balanced water of the ships and oil tankers that introduce considerable amount of PAHs originated from oil source. On the other hand, pyrogenic index values in the study area (ranged

Table 2: The selected molecular ratios for discriminate between pyroletic and petrogenic origins.

Diagnostic ratio	Pyrogenic origin	Petrogenic origin	This study	References
MP/P[a]	<1	>2	0.35-11.90	[48]
Pyrogenic index[b]	>0.8	<0.223	0.23-2.22	[9]

[a]MP/P: Ratio of (3-methylphenanthrene + 2-methylphenanthrene + 9- methylphenanthrene + 1-methylphenanthrene) to phenanthrene.
[b]Pyrogenic index: Ratio of the sum of the concentrations of EPA priority unsubstituted three- to six-ring PAHs to the sum of the concentrations of four targeted alkylated PAH homologues (naphthalenes, phenanthrenes, fluorenes, chrysenes).

between 0.23-2.22) are pointed out to pyrogenic sources for all sites, noting that, its value (0.23) at site 8 (Damam) is very close to the border line of petrogenic origin (0.22) which is consistence with MP/P index interpretation. Contribution percentages of pyrogenic and petrogenic origins were estimated at 78.8 and 22.2%, respectively, that are very close to the percentage recorded in the sediments northern of Arabian Gulf [34].

4 Conclusions

The ranges of total PAHs in the studied mangrove stands along Arabian Gulf–Saudi coast showed significant increase from sediment to leaf, and mangrove roots recorded the highest values. This trend support PAH uptake through sediment/root mechanism. Despite the sandy nature and low organic carbon content of sediments in the studied stands; moderate levels of PAHs were recorded either in sediments or mangrove plants that attributed to the presence of mangrove stands close to anthropogenic effects like sewage drainage, oil wells and port activates which increasing PAHs contamination probabilities. However, according to SQG adverse biological effects hardly to occur. Bio-accumulation in most sites showed high values reaching 150.50 for phenanthrene in the mangrove roots as the maximum recorded value for accumulation. PAH origin of the major mangrove stands in Arabia Gulf of Saudi coast was found mainly pyrogenic except two sites (Damam and Damam Port) that showed petrogenic origin.

Conflict of interest: Authors state no conflict of interest.

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