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DISTRIBUTION OF TOTAL PETROLEUM HYDROCARBONS AND POLYCYCLIC AROMATIC HYDROCARBONS IN MUSA BAY SEDIMENTS (NORTHWEST OF THE PERSIAN GULF)

Sediment samples were collected from the coastal area of the Petrochemical Special Economic Zone of Musa Bay. The samples were analyzed to determine the degree of hydrocarbon contamination and identify the sources of PAHs which were evaluated by employing the ratios of specific PAHs compounds and principal component analysis (PCA). The levels of TPH concentration in the studied area were relatively moderate compared to the world-wide chronically oil-contaminated locations. The concentration of total PAHs was lower than those in other parts of the Musa Bay but approached the NOAA sediment quality guideline value.

1. INTRODUCTION

The Petrochemical Special Economic Zone (PETZONE) was established in the southeast of Iran, in the vicinity of Musa Bay (northwest of the Persian Gulf) in 1997. PETZONE consists of the area of approximately 2600 ha and contains fifteen petrochemical companies and five sanitarium and effluent treatment stations. Since PETZONE was established, some parts (Zangee and Jafari creeks) of Musa Bay have become enclosed in PETZONE constructions and divided into two parts [1, 2]. The Musa Bay (Khowr-e Musa) is a basin with a 50 km long connection to the open ocean

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and it is not an estuary [3]. Moreover, several creeks are branched out from it, including the Zangee, Jafari, Moavi, Ghanam, and Marimus creeks [2, 4, 5]. It is located in a large cross-border area of intertidal mud flats and a common characteristic of almost all intertidal mud flats is the high productivity which makes them attractive for breeding, nesting, overwintering and migration stop-over grounds for birds and as nursery ground for many fish species [3, 6, 7].

Musa Bay is a semi-enclosed ecosystem, and several studies have shown that pollution and its adverse effects are stronger in semi-enclosed areas than in the open sea and based on the characteristics of these ecosystems, as the flow of water decreases, the concentrations of nutritious substances and biodiversity increase. The water circulation in Persian Gulf is anti-clockwise, and the water exchange rate from the narrow striate of Hormuz is 3 to 5 years, the water of Musa Bay is exchanged during this time frame. In addition, the semi-diurnal tide in the intertidal mud flat zone naturally dredges and transports sediments and associated pollutants [3, 6, 8].

According to the IMO declaration, the Gulf area is the most sensitive area in the world and Department of Environment has reported that, the Musa Bay is considered the most sensitive marine area in Iran and the Gulf area due to its unique ecosystem. Therefore, Musa Bay is important for the whole northwestern coast of the Persian Gulf [3]. Due to the sensitivity of the aquatic ecosystem of the Persian Gulf, several studies have been performed on various pollutants and primary sources. Based on the scientific reports, we learn that 49% of the world's oil production comes from the Gulf states and passes through this old waterway and it holds an estimated about 57–66% of the world oil reserves. Several studies showed that the Persian Gulf is the most oil-polluted marine area in the world, even before the Gulf war and the Gulf oil pollution is about 48 times that of any other similar area on the Earth [3, 9–11]. During the first Gulf war over 700 wells were destroyed and burned thus after the Gulf War, interest in the contamination of the Persian Gulf (especially oil pollution due to the presence of natural sources of oil and gas) and its adverse effects increased. In the past four to five decades, this area has been the major waterway for oil transport and it has suffered from oil spills into its marine environment thus several studies and regional sea programs were conducted to estimate the rate of damage to this valuable aquatic ecosystem [10, 12]. The risks to aquatic biota are difficult to assess due to the complex nature of the hydrocarbon mixtures. Therefore, during the last decades, several researches were done in this area due to its sensitivity and also several point source and nonpoint sources of pollutions. In 1996, studies showed that the Iranian coastal area (especially the southern region along the Imam Khomeini port) was contaminated with petroleum hydrocarbons but in a lesser scope than that of the most Arabian coasts [13]. In 2005, the concentrations and distributions of various petroleum hydrocarbons (PHs), chlorinated pesticides and PCBs were determined in biota and coastal sediments in seven countries (Bahrain, Iran, Kuwait, Oman, Qatar, Saudi Arabia and the United Arab Emirates) of the ROPME sea area. Results of this study showed that the

extensive marine contamination regarding to organochlorinated compounds and PHs was not observed [14]. Also during the last decades Iran's coastal problems have been increased which has been realized that related to the sectoral management [15].

Among the organic pollution of aquatic environments, aliphatic hydrocarbons and polycyclic aromatic hydrocarbons (PAHs) have received much attention due to their omnipresent distribution in marine sediments. Many researches showed that the lipophilic characteristics of polyaromatic hydrocarbons, their tendency to accumulate in sediments, their low degradation rate and high stability also their potential to manifest the ecotoxicological activity, the concentration of PAH in sediment is an appropriate index of the status of contaminates in marine environments [16–19]. Several studies have classified the sources of PAHs into two categories, pyrolytic and petrogenic. Pyrolytic PAHs are formed as a consequence of incomplete combustion although petrogenic PAHs are mainly derived from crude oil and its refined products. The use of ratios of PAH components of the same molecular weight is well founded as an index to interpret PAH composition and its possible sources, for example molecular indices such as fluoranthene/pyrene are are used to identify emission sources [20, 21]. Thus, in the present study, due to the vicinity of PETZONE, Bandar-e-Imam Khomeini petrochemical company and ports and other industries to Musa Bay, the distribution and sources of total petroleum hydrocarbons and polycyclic aromatic hydrocarbons in the sediment of Musa Bay close to the PETZONE (northwest of the Persian Gulf) was monitored.

2. MATERIALS AND METHODS

Sediment samples from Musa Bay were collected from 12 stations located in the coastal area of the Petrochemical Special Economic Zone (PETZONE) from June 2009 to June 2010 using an Ekman-Birge grab sampler (225 cm^2). The sampling sites are shown in Fig. 1.

Five (1–5) sampling stations were in the proximity of PETZONE wastewater discharge points and drown to make one sampling point 0.7 km apart on each transect. The sampling stations presented in Table 1.

The study area is covered with fine-grained sediments and the top 0–5 or 0–10 cm from each grab samples was obtained to provide an appropriate sample of surface sediment. Then, sediment samples were stored in aluminum foil and placed on ice after sampling, immediately transported to the laboratory and kept in the refrigerator at –20 °C before analysis. In the laboratory, 10–20 grams of freeze-dried sediment sample (freeze-drier model: Operon), ground and sieved at 125 µm, were placed in the glass tube of the reactor. The extraction was conducted using a microwave oven (at 115 °C for 20 min). A 15 g aliquot of freeze-dried sediment sample was placed in the glass tube of the reactor with 40 cm^3 of hexane:methylene chloride (1:1 v/v).

Fig. 1. Location of the study area and sampling stations in Musa Bay

Sulfur was removed using activated elemental copper in order to avoid potential interferences during gas chromatography, and the extracts were concentrated using a rotary evaporator. Sediment samples were fractionated on a silica gel–alumina col-

umn and the column of systems contained 10 cm^3 of silica powder, 10 cm^3 hydrated alumina, 1–2 g of sodium sulphate. After passing the extract through the system, the extract was placed in 5 cm^3 vials and then the extract was dried with anhydrous sodium sulfate and transferred in a graduated tube and concentrated down to 1–2 cm^3 using a flow of clean nitrogen. Individual PAHs were quantified based on the retention time with a reliable PAHs mixed standard (Sigma), and concentrations of each PAH were calibrated based on the standard calibration curve. Finally, the concentrations of the following PAHs were determined: six low molecular weight, two and three rings aromatics, naphthalene (Na), acenaphthylene (Acpy), acenaphthene (Acp), fluorene (Flur), phenanthrene (Phen), anthracene (Ant), and 10 high molecular weight, four, five and six aromatic rings, fluoranthene (Flu), pyrene (Py), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenzo(a,h)anthracene (DbahA), benzo(g,h,i)perylene (BghiP), indeno (1,2,3-cd)pyrene (IP) [22–24].

T a b l e 1

Sampling stations of Musa Bay study

Code	Station	E	N
1	Junction of Jafari and Zangi Creeks	49° 6′48.50″E	30°26′53.50″N
1-J	700 m from the junction of Jafari & Zangi Creeks	49° 7′29.49″E	30°26′36.12″N
2	BI-PC East Pond	49° 7′12.44″E	30°26′17.34″N
2-BI	700 m from BI-PC East Pond	49° 7′40.01″E	30°25′59.60″N
3	BI-PC South East	49° 7′0.47″E	30°25′50.09″N
3-BI	700 m from BI-PC South East	49° 7′28.11″E	30°25′28.77″N
4	BI-PC Aromatic	49° 6′26.87″E	30°25′37.39″N
4-BI	700 m from BI-PC Aromatic	49° 6′45.47″E	30°25′11.46″N
5	Razi	49° 6′2.06″E	30°25′29.16″N
5-R	700 m from Razi	49° 6′12.26″E	30°25′1.02″N
6	Musa1	49° 4′17.99″E	30°25′20.64″N
7	Musa2	49° 3′39.19″E	30°24′38.17″N

Initially, each sample was analysed for total petroleum hydrocarbons using UV fluorescence (UVF 2500 fixed excitation wavelength 310 nm; the emission wavelength 360 nm) and gas chromatography mass spectrometry (GC-MS; GC, Agilent, 6890N, MS: Agilent, 5973N), respectively[14, 25].

3. RESULTS AND DISCUSSION

3.1. TOTAL PETROLEUM HYDROCARBONS

The levels of total petroleum hydrocarbons (TPH) in sediments were relatively moderate. In 1996, experiments showed that the coastal area of Imam Khomeini port

was classified as slightly polluted; however, the concentrations of contaminants were close to the lower limit of moderately polluted areas. Nevertheless, the observed amount of contamination was lower than that of most Arab coasts and the TPH concentration in the Persian Gulf sediments was categorised into four levels (guideline): Unpolluted area/natural background level (10–15 μg/g), slightly polluted area/upper permissible limits (15–50 μg/g), moderately polluted area (50–200 μg/g), heavily polluted area (>200 μg/g). Thus the concentrations <15 μg/g, as chrysene equivalents are considered to represent natural background levels in this region [13].

In 2000, the Mangrove Afforestation Program was initiated in the coastal area surrounding the aforementioned creeks and Musa Bay, including PETZONE and the region surrounding Mahshahr oil export terminal to increase the amount of green space around industrial zones [26]. In 2004, the Research Institute of the National Petrochemical Industry of Iran showed that the most polluted areas in Musa Bay were located in the vicinity of petrochemical industries and Mahshahr oil export terminal. The Institute reported that the concentration of total oil and grease (TOG) in Musa Bay sediments was 9.15 mg/g; therefore, the Bay is more polluted than other Arabic coastal areas [27]. In 2009, Taatizadeh studied the impact of TPH on Musa Bay by evaluating regions located in the vicinity of Imam Khomeini port (near PETZONE). In the aforementioned study, the concentration of TPH ranged from 5.44 to 16.66 μg/g (average 10.13 μg/g) [28].

Table 2

Average concentrations of TPH
in the sampling stations of Musa Bay

Code	Concentration [μg/g]	Guideline
1	88.81	
1-J	75.65	moderately polluted
2	57.32	
2-BI	43.84	slightly polluted
3	32.73	
3-BI	97.15	moderately polluted
4	45.93	slightly polluted
4-BI	53.04	moderately polluted
5	17.51	
5-R	16.48	
6	40.05	slightly polluted
7	39.25	
Total	48.98	

The average concentrations of TPH obtained in the present investigation are shown in Table 2. The TPH concentrations of sediment samples presented an average

value of 48.98 µg/g and ranged from 16.48 to 97.15 µg/g and the highest TPH level was observed at 3-BI (97.15 µg/g). The TPH concentrations of the stations can be arranged as follows:

$$3\text{-BI} > 1 > 1\text{-J} > 2 > 4\text{-BI} > 4 > 6 > 7 > 3 > 2\text{-BI} > 5 > 5\text{-R}$$

At all of the stations, the concentrations of TPH were higher than the natural background value (unpolluted area/natural background level: 10–15 µg/g [13]). Moreover, the TPH concentrations were lower at stations 5 and 5-R due to high water depth at the site.

Fig. 2. Connective canals of Jafari and Zangi Creeks inside the PETZONE

As shown in Figure 2, high concentrations of TPH were observed at the first 3 stations near the main PETZONE outlets and also near the most important outlet of Bandar-e Imam Khomeini Petrochemical Company (3-BI). The concentration of TPH reached a maximum value at station 3-BI, while station 5 and 5-R presented the lowest concentrations. Lastly, the amount of TPH increased gradually at stations 6 and 7. The concentrations of TPH reached maximum values in the east, near the main effluent outlets of the PETZONE and Imam Khomeini petrochemical company east and southeast outlets, which could be related to the discharges of petroleum wastewaters. As a result, stations 1 and 1-J are polluted. During low tide, outlets of Jafari and Zangi Creeks located in the PETZONE interchange their water with Musa Bay (Fig. 2). Depth of the Bay decreased gradually until station 4 and was very low at station 4-BI. As a result, the sedimentation rate increased [29]. As the water depth decreased, the concentration of TPH increased and reached at a maximum at station 3-BI (97.15 µg/g).

Another reason may be related to the mangrove forest area which is located in the east part of the PETZONE. The presence of fine sediment along the mangrove line can increase the sedimentation rate. As the hydrodynamic energy time decreases, these types of forests can trap suspended solids; thus, enough time is available for the storage and re-deposition of fine particles. Due to the high total organic carbon content, aerobic conditions and rapid change of burial, the potential for the deposition of anthropogenic pollutants adsorbed by sediments in mangrove forests is high [31, 32].

Stations 5 and 5-R are located near the port of Razi and the outlet of Razi petrochemical company; however, the water depth is very high in this area. Therefore, this region is less polluted than the other stations and it is classified as slightly polluted; nevertheless, the concentration of contaminants was close to the natural background value [13].This variation is mainly related to differences in the locations (closeness to the main petrochemical effluents), depth of the sediment sampling stations and also there is no mangrove plants. Station 6 is located near the Imam Khomeini port, and station 7 is situated in the mouth of Marimus Creek. However, effluent outlets were not present alongside these stations. As a result, the stations were classified as slightly polluted (slightly polluted area/upper permissible limits:15–50 µg/g [13]). These sites were not as deep as at stations 5 and 5-R; thus, the observed contaminant concentration may be related to shipping activity and strong tidal currents in the mouth of Marimus Creek. On the other hand, due to another guideline, TPH concentrations higher than 500 µg/g are indicative of significant pollution and less than 10 µg/g are considered as unpolluted sediments [33], also according to some guidelines, concentrations lower than 15 µg/g as chrysene equivalents, are considered to represent natural background levels in the ROPME Sea area [14].

Moreover, the average cluster analysis results for TPH showed that this factor can be classified into four groups as follows: Group I: 6, 7, 4, 3; group II: 2, 4-BI; group III: 5, 5-R, 2-BI and group IV: 1, 3-BI, 1-J. In addition, high concentrations of TPH were observed at group IV (Fig. 3).
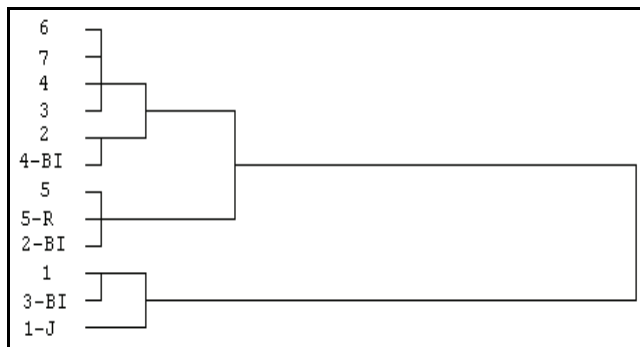

Fig. 3. A dendrogram representation of a hierarchical cluster analysis of TPH

In conclusion, the levels of TPH concentration in the study area relatively moderate compared to chronically oil-contaminated area such as, Gulf of Oman (0.05–779 µg/g) and also highly oil-contaminated coastline of BAPCO oil refinery in Bahrain (779 µg/g) [34], oil-contaminated coastline of Saudi Arabia after the Gulf war (11–6900 µg/g) [35], near-shoreline marine sediments of the United Arabian Emirates, in 1994 (0.4–212 µg/g), highly oil-impacted sediments of Hong Kong's Victoria Harbour (60–646 µg/g) [36] and New York Bight (35–2900 µg/g) [14].

3.2. POLYCYCLIC AROMATIC HYDROCARBONS

Concentrations of PAHs in sediment samples obtained in the present investigation are shown in Table 3.

Table 3

PAHs concentrations in the sampling sediments of the Musa Bay [ng/g dry wt.]

PAH	Station					
	1	1-J	2	2-BI	3	3-BI
Na	12.56	5.28	20.50307	23.38684	24.83834	217.8316
Acpy	57.42	51.9	89.64215	55.15262	130.5695	113.6414
Acp	3.27	2.38	17.05942	ND	34.74137	47.96575
Flu	26.33	18.78	104.1342	26.30694	138.8921	150.4455
Phen	153.36	100.76	520.505	97.07002	1003.686	582.7425
Ant	76.55	54.96	131.2831	94.85838	237.4756	196.9407
Flur	77.06	47.22	113.9081	83.32773	228.5946	127.9235
Py	92.32	63.55	151.2625	76.53803	317.9455	183.6341
BaA	72.87	56.23	62.49124	36.43397	246.8423	106.5354
Chr	53.8	41.83	73.14156	42.68393	235.2434	105.9908
BbF	ND	1.04	ND	ND	48.7879	12.39326
BkF	5.11	2.54	2.14402	ND	34.16614	11.51599
BaP	6.27	7.83	5.380938	2.14116	25.22545	13.42304
IP	3.91	ND	20.93167	ND	ND	2.668143
DbahA	ND	ND	ND	ND	ND	ND
BghiP	5.541056	ND	ND	ND	6.328635	1.5196
Σ PAHs	646.371056	454.3	1312.386968	537.89962	2713.336835	1875.171283
Station name	4	4-BI	5	5-R	6	7
Na	32.31006	39.31215	19.53378	17.15862	14.62426	ND
Acpy	109.7316	96.42878	67.37432	77.94457	25.06882	11.86068
Acp	68.82055	26.06835	4.159589	ND	10.09139	ND
Flu	580.3472	94.49087	51.57462	62.78317	8.22288	27.59102
Phen	2820.602	598.7857	220.8398	203.2178	95.24683	27.48902
Ant	668.3971	165.5599	121.2527	216.8339	50.25549	35.56072
Flur	2700.616	216.0918	133.6832	146.1367	220.4299	30.14026
Py	1772.815	230.4833	158.3324	162.3837	58.4439	38.58371
BaA	7079.156	308.8403	181.1581	184.9479	46.23929	15.69081

PAHs concentrations in the sampling sediments of the Musa Bay [ng/g dry wt.]

Chr	5510.745	242.9863	148.2293	202.9512	34.89384	11.93007
BbF	2898.684	54.08268	91.36013	330.4286	6.30478	ND
BkF	1327.789	45.64691	50.49311	170.4355	5.8342	1.97127
BaP	911.5719	41.64462	43.88357	111.4199	8.31854	5.829233
IP	83.42872	ND	61.35551	153.3058	21.75892	6.78132
DbahA	ND	ND	20.77299	20.91085	ND	ND
BghiP	94.05157	ND	36.4655	77.23912	7.39302	3.138818
Σ PAHs	26659.0657	2160.42166	1410.468619	2138.09733	613.12606	216.566931

ND – not detected.

Most values were exceptionally low, except for the sediments near the station 4, which is located near the aromatic outlet of Imam Khomeini Petrochemical Company (station 4: BI-PC aromatic) and its concentration level was higher than the NOAA sediment quality guideline value (ERL) of 4000 (ng/g dry weight) [37]. The cluster analysis of total PAH showed that this factor can be classified into two groups and classified the station 4 significantly different from other stations (Fig. 4).
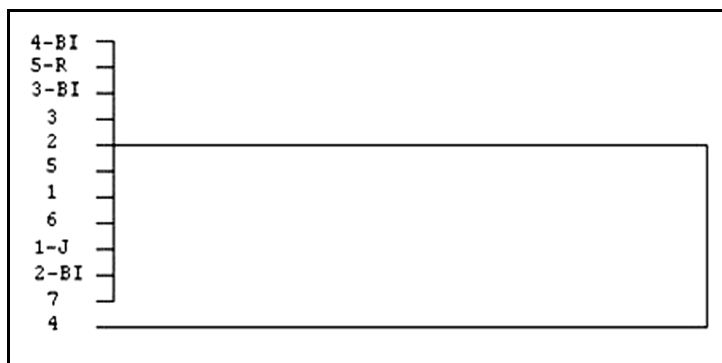

Fig. 4. A dendrogram representation of a hierarchical cluster analysis of the PAHs

PAHs in sampling sediments from the Musa Bay ranged from 216.57 to 26 6659.07 ng/g dry weight with the mean value of 3 394.76 ng/g (3.40 µg/g). The average concentration of PAHs in sampling stations of Musa Bay was lower than its concentration in the sediments of Musa Bay at Mahshahr oil export terminal (90 µg/g) and its concentration was very close to sediments of the Imam Khomeini port (0.19–3.52 µg/g), also concentration of PAHs was lower than the other part of the Ropme Sea area (RSA), near the BAPCO refinery in Bahrain (6.6 µg/g) [14, 34–38].

PAHs of molecular mass 178 and 202 are commonly used to recognize between combustion and petroleum sources of PAHs. According to the PAH isomer ratios

[20, 21, 23, 38], the following isomer ratios have been used to identify potential sources of PAHs: Ant/(An + Phe), BaA/(BaA + Chry), Flu/(Flu + Pyr) and IP/(IP + Bghi). The isomer pair ratios of PAHs were calculated. The Ant/(Ant + Phe) ratio showed that the main source of PAHs was combustion (Ant/(Ant Phe) > 0.1 were typical of combustion source). In contrast, the Flu/(Flu + Pyr) ratio showed that in the most of the sampling stations, the main source of pollution was combustion of petroleum (0.5 > Flu/(Flu + Pyr) > 0.4), except at stations 2-BI, 4 and 6, which received PAHs from different sources, (Flu/(Flu + Pyr) > 0.5) including were mainly from combustion of grass, wood and coal. The BaA/(BaA + Chry) ratio showed that the main source of PAHs at all of the sampling stations was combustion (BaA/(Chry + BaA) > 0.35). Finally, the ratio of IP/(IP + BP), at about half of the sampling stations were lower than 0.2 and it showed that the main source of PAHs was petroleum origin input, while in the rest of them it was greater than 0.5, indicating that the input of PAHs were mainly from coal, wood and grass combustion. As shown in Table 4, PC1 could explain 70.83% of the total variance in the data. This factor strongly affected the concentration of Flu, Ant, Flur, Py, BaA, Chr, BbF, BkF, BaP, Phen, Acp, and BghiP; thus, the main sources of contamination were incomplete combustion and fuel pyrolysis. PC2 was responsible for 15.60% of the total variance in the data and had a significant impact on the BaP and IP concentration. The third PC, which is responsible for 9.42% of the variance, was not associated with any of the PAHs; thus it is related to an unknown source.

Table 4

Rotated component loadings of the three principal
components of PAHs in Musa Bay sediments

PAH	PC1	PC2	PC3	PAH	PC1	PC2	PC3
Na	0.08	−0.55	0.67	Chr	0.98	0.00	−0.17
Acpy	0.49	−0.37	0.64	BbF	0.98	0.08	−0.14
Acp	0.80	−0.51	0.26	BkF	0.98	0.09	−0.13
Flu	0.98	−0.15	0.02	BaP	0.98	0.08	−0.13
Phen	0.96	−0.19	0.00	IP	0.44	0.80	0.34
Ant	0.98	−0.02	0.14	DbahA	−0.04	0.83	0.49
Flur	0.98	−0.00	−0.17	BghiP	0.77	0.60	0.17
Py	0.99	−0.04	−0.09	Variance %	70.83	15.60	9.42
BaA	0.98	0.00	−0.18	Cumulative %	70.83	86.44	95.86

In conclusion, the levels of TPH concentration in the studied area were relatively moderate compared to chronically oil-contaminated area in the RSA and to other highly oil-impacted sediments in other parts of the world. Compared to natural sources, hydrocarbon pollutants such as shipping activities, PETZONE and other industries significantly contributed to TPH contamination in Musa Bay.

The concentration of total PAHs in sampling sediments of the Musa Bay was lower than that other parts of the Musa Bay but its concentration level approached the NOAA sediment quality guideline value (ERL), also the average concentration of total PAHs was higher than the other parts of the RSA except the BAPCO refinery[34].

The results of source analysis showed that, in addition to petrogenic input as a major source because of the vast amount of oil and gas in the Persian Gulf, pyrolytic input are also a source for PAHs. Despite the high concentration of anthropogenic contaminates and other non-point source pollution, the results of the present study are unexpected in this semi-enclosed bay with a slow rate of water exchange. The unusual results may be related to the deposition of finer sediments along the Iranian eastern side and northwest area, which is associated with the counter-clockwise circulation from the Indian Ocean, deposition of eolian sediments and probably the effects of tidal currents [13]. According to the results of the current investigation, to assess ecological conditions, the status of the benthic community should be determined, ecological risk assessment should be estimated, the concentration of other contaminants should be evaluated and PETZONE effluents that are discharged into Musa Bay should be monitored annually.

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