

PAHs in Seawater of the Semi-Closed Areas along the Alexandria Coast of Egyptian Mediterranean Sea

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ABSTRACT

The distributions of 15 out of 16 polycyclic aromatic hydrocarbons (PAHs) were determined in surface and bottom water of Alexandria coasts (Egyptian Mediterranean Sea). Total average PAH concentrations were ranged from 9.89 to 677.25 ng/l, and from 2.21 to 345.29 ng/l in surface and bottom water, respectively. PAH concentrations were at relatively low levels compared to those reported for other estuaries and marine systems around the world. The Σ COMB and Σ TF-PAHs suggest the origin of hydrocarbons. The higher concentrations of total pyrolytic hydrocarbons (Σ COMB) than that of total fossil hydrocarbons (Σ TFPAH) declared that atmospheric fall-out from traffic exhausts was the significant source of PAHs to marine environment of the semi-closed areas of Alexandria. Interferences of rather petrogenic and pyrolytic PAH contaminations were noticed in the harbours, especially marine area due to petroleum products deliveries and fuel combustion emissions from the ships awaiting along side the quays.

Keywords: Surface Seawater; Bottom Seawater; PAHs; Alexandria; Egypt; GC-MS

1. Introduction

Water pollution is a serious problem in the global context. Marine environments are considered to be the ultimate pool of many organic pollutants [1-3] introduced directly or indirectly through anthropogenic and/or natural sources. The study of trace organic contaminants in coastal marine environment is of great importance, since these areas are biologically productive and receive considerable pollutant inputs from land-based sources via point and non-point sources [4-7]. The quality of the aquatic ecosystem is of great interest to the entire world. Scientific interest in the quality of marine ecosystems is quite recent and has especially increased in the past 15 years in relation to application of the European Union (EU)'s Water Framework Directive (WFD) [8]. One of the main objectives of the WFD is achievement and preservation of "Good Chemical Status" of surface waters of the EU member states. Of all marine pollutants, hydrocarbons have received the greatest attention due primarily to the highly lethal effects of most hydrocarbons on marine organisms, espe-

cially aromatics class. The occurrence of PAHs in marine environment has attracted the attention of the scientific community, as these compounds are frequently detected in seawater and sediments at increasing levels and have adverse health effects on marine organisms and humans. Several PAHs are potential human carcinogens and are included in the priority list of the European Union Water Framework Directive (2000/60/EC). Sixteen PAHs, identified by the US Environmental Protection Agency (EPA) as priority pollutants [9-14], are a class of organic pollutants which have an important impact on marine ecosystems because they include the largest known class of chemical carcinogens and mutagens [15-17]. This study aims to assess the distributions and characteristics of selected parent PAHs in aqueous phase along Alexandria coast and the correlation of the physicochemical parameters on their distribution. Conversely, in the Eastern Mediterranean few data have been published on the presence of PAHs in coastal water close to point sources (municipal and river discharges, etc.). In addition, hydrocarbon budgets are available for the Western part of Mediterranean Sea [18-20], but there is a tremendous lack of informa-

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tion regarding the Southern Mediterranean [21-28]. This is the first systematic study to be undertaken along the semi-closed areas of Alexandria (Egyptian Mediterranean Sea coast) as a comparative study between four sectors of different identity. The available data of hydrocarbon levels in the Egyptian marine environment are scarce in the last ten years [23-30]. Along the Mediterranean coast of Alexandria city, there are many areas with high activities of shipping and pleasure boating activities, incorporating numerous Harbours and marina [28-39].

2. Materials and Methods

2.1. Sampling Plan

Water samples were collected from 49 sampling stations; (49 surface water & 43 bottom water samples), along the semi-closed areas of Alexandria coasts, Egypt as shown in (Figure 1).

Surface and bottom water samples were collected from 49 stations distributed along the semi-closed areas of Alexandria (Egyptian Mediterranean Sea coast), namely Abu-Qir Bay (AQ), Eastern Harbour (EH), Western Harbour (WH), and El-Mex Bay (Max) during 2010 on local motorboat using Niskin bottle. The location of each sampling station was determined using a GPS (Trimble Navigation, Japan) to pinpoint sampling locations. Sampling sites were selected taking into consideration the expected polluted areas due to industrial and human impacts. Temperature and pH values were obtained at each site by deploying a multi-variable sensor. A set of water samples were fixed to determine dissolved oxygen (DO), also oxidizable organic matter (OOM) was determined in all samples. Samples were subsequently kept in a refrigerator at -4°C prior to analysis in the laboratory.

2.2. Extraction

2.2.1. Physicochemical Characteristics of Water Samples

Salinity (‰) was measured in situ using a Salinometer equipment (Thermo Electron Corporation 088749, USA). pH was measured in the field by the use of a pH meter equipment (JENWAY 3505, UK), DO was fixed immediately and determined according to the classical Winkler method (Strickland and Parsons) [40]. OOM was done as described by FAO [41].

2.2.2. Determination of Petroleum Hydrocarbons in Water

One liter of seawater was poured into glass separatory funnel, extracted twice with 2×40 ml of methylene chloride. The extract was stored in dark at low temperature ($\sim 5^{\circ}\text{C}$). Before analysis, the stored extracts were evaporated to dryness in a rotary evaporator at 30°C under reduced pressure. The residue was re-dissolved in 1 ml of n-hexane before fractionation with column chromatography [42]. The extracted volumes were passed through the silica column prepared by slurry packing 20 mls (10 g) of silica (activated at 185°C overnight and deactivated with 12.5% water afterwards), followed by 10 mls (10 g) of alumina (90 neutral, 70 - 230 mesh previously activated at 185°C overnight and deactivated with 12.5% water afterwards) and finally 1 g of anhydrous sodium sulphate. Elution was performed using 40 mls of hexane (aliphatic fractions), then 40 mls of hexane/dichloromethane (90:10) followed by 20 mls of hexane/dichloromethane (50:50) (which combined contain PAHs). Finally, the eluted samples were concentrated under a gentle stream of N_2 to about 0.2 ml. Activated copper was added for desulfurization [43].

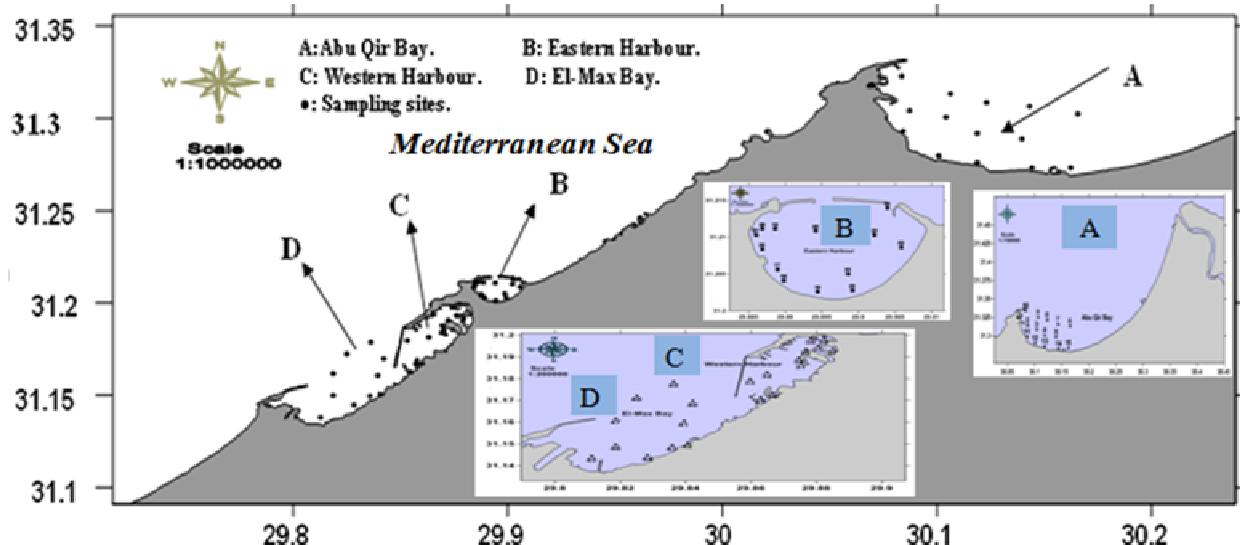


Figure 1. Study area along Alexandria Coasts of the Egyptian Mediterranean Sea.

1) Blank preparation

According to Parson *et al.*, [42] eighty mls of methylene chloride were poured into a rotary evaporator flask and evaporated to dryness at 30°C. The residue was dissolved in 1 ml of n-hexane before fractionation.

2) Identification of PAHs using Gas chromatography-Mass spectrometry (GC-MS)

Blanks of 1000 fold concentration were analyzed by Gas Chromatograph-Mass Spectrometer; Trace-Ultra coupled to DSQ-II MS (Thermo Electron SPA, UK) equipped with auto injector. The mass spectrometer was operated in full scan mode (50 - 650) Daltons per second. The ionization source was supplied with voltage at 70 eV. Three μ l volume of each sample was injected in the splitless mode. The 15 out of 16 EPA-PAH compounds were naphthalene (Naph, m/z 128), acenaphthylene (Acthy, m/z 152), acenaphthene (Ace, m/z 154), phenanthrene (Phe, m/z 178), anthracene (Ant, m/z 178), fluoranthene (Flu, m/z 202), pyrene (Pyr, m/z 202), benzo(a)anthracene (BaA, m/z 228), chrysene (Chr, m/z 228), benzo (b)fluoranthene (BbF, m/z 252), benzo(k)fluoranthene (BkF, m/z 252), benzo(a)pyrene (BaP, m/z 252), benzo (ghi)perylene (BghiP, m/z 276), indeno (1,2,3-cd)pyrene (InP, m/z 276), and dibenz(a,h) anthracene (DBA, m/z 278). GC/MS is equipped with splitless injector and a fused silica capillary column; Thermo TR-35 MS (30m, 0.25 mm, 0.25 μ m) with 35% phenyl polysilphenylene-siloxane. Helium was used as carrier gas at 1.5 $\text{ml}\cdot\text{min}^{-1}$. The oven temperature program was 60°C (1 min) ramped at 8°C/min to 100°C, hold for 1 minute then ramped at 5°C/min to 300°C maintaining about 20°C. The injection and the transfer line temperature were maintained at 280°C and 300°C, respectively.

The response factor of individual PAH compounds to the internal standard was measured and calculated at least three times at the beginning, in the middle and at the end for each batch of GC injections (15 samples). Identification and quantification of 15 out of 16 EPA-PAH compounds were based on matching their retention times with a mixture of PAH standard.

3) Analytical quality control

To control the analytical reliability and assure recovery efficiency and accuracy of the results, 6 analyses were conducted on PAH compound reference materials, HS-5 (sediments) provided by NRC-IMB of Canada and SRM-2974 (Freeze-dried mussels tissue) (Mytilus edulis) provided by NIST of USA as well as artificial seawater samples of known PAH levels spiked with a mixture consisting of 2 μ g each of PAHs were analyzed as above to validate the analytical method used in this study. The recovery efficiency ranged from 92% to 111% for HS-5, 88% to 96% for SRM-2974 and 93% to 105% for the spiked samples. The mean recovery for PAHs were as followed: Naph 90.1%, Acthy 92.3%, Ace 105.2%, Flu

95.4%, Pyr 92.5%, BaA 89.7%, Chr 107.1%, BaP 94.6%, BbF 90.5%, DBA 101.8%, BghiP 93.7% and InP 97.5%.

3. Results and Discussion

3.1. PAHs in Water Samples

Tables 1(a)-(d) showed the total concentrations of the PAHs in water. The concentrations below their limits of detection were given a value of zero for the calculation. (Σ PAHs) concentrations in water varied significantly among the studied locations. The values ranged from 2.2 to 677.25 ng/l with an average value of 192.29 ng/l. The highest concentration of total PAHs was measured in water collected from St. 6S (AQ), while lower concentrations were detected in sample of St. 2B (AQ). In this study, the order of decreasing PAHs concentrations for surface and bottom waters is illustrated in Figure 2. The average concentration of PAHs in the present study was moderate in comparison to those in most areas of urbanized estuaries in the other countries represented in Table 2.

3.2. PAHs and OOM

No correlation between hydrocarbons and organic matter in water column where; ($R = -0.2368$, $n = 92$). This may attribute to the weak ability of hydrocarbons to adsorb onto organic matter in water phase at low concentrations of PAHs (Figure 3).

3.3. Comparison of Different PAHs Fractions in Water Samples of the Four Areas

Figures 4 and 5 showed the percentages of the average combustion, carcinogenic and fossil PAHs of the four sectors to each other. There is no significant difference in percentages of combustion PAHs in AQ, EH and WH in surface and bottom water and they are larger than that calculated for El-Max Bay. In contrast, Max possesses the highest percentage of fossil PAHs may be interpreted that the anthropogenic sources are loaded mostly with LMW PAHs (molecular weight < 178) which have ability to dissolve in water than HMW PAHs (molecular weight > 178), so the percent of saturation of fossil PAHs becomes 45% or more greater than that belong to combustion PAHs < 30%. This may give indication that the origin of PAHs in AQ, EH, and WH are generally pyrogenic; while it is petrogenic in max.

3.4. Physicochemical Parameters in Water Samples

Tables 3 and 4 showed some physico-chemical parameters; Salinity %, pH, OOM (mg/l), DO (mgO₂/l), Σ PAHs (ng/l), Σ COMP (ng/l), Σ CARC (ng/l), and Σ TFPAH (ng/l) in

Table 1. (a) Concentrations (ng/l) of PAHs in water samples collected from Abou Qir (AQ) during 2010; (b) Concentrations (ng/l) of PAHs in water samples collected from Easter Harbor (EH) during 2010; (c) Concentrations (ng/l) of PAHs in water samples collected from Western Harbor (WH) during 2010; (d) Concentrations (ng/l) of PAHs in water samples collected from El-Max (Max) during 2010.

(a)															
Parameter Site	Naph	Acthy	Ace	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkF	BaP	InP	B(ghi)P	DBA
1S	10.56	9.6	4.8	9.6	7.2	23.76	2.4	17.6	67.2	99.92	52.8	21.6	21.08	10.32	6.88
2S	17.68	ND	6.73	2.6	10.4	62.4	6.98	26	28.6	83.2	13	39.78	109.08	101.14	67.43
2B	ND	ND	<DL	<DL	<DL	<DL	ND	ND	<DL	<DL	<DL	ND	ND	<DL	
3S	7.56	<DL	3.6	9.89	3.6	45.97	1.8	13.8	7.2	52.2	14.4	97.2	17.83	78.48	52.32
3B	ND	ND	ND	<DL	<DL	<DL	ND	7.93	ND	5.77	12.8	3.98	17.95	78.2	52.13
4S	8.36	ND	3.8	18.68	5.3	41.8	<DL	13.3	9.5	24.7	17.1	76	13.25	156.37	104.25
4B	ND	ND	6.4	<DL	10.98	3.7	ND	5.3	3.15	18.29	11.09	3.92	26.18	57.2	38.13
5S	19.32	<DL	5.67	4.6	11.5	46.59	2.3	7.93	16.1	43.7	85.1	27.6	7.73	38.64	25.76
5B	6.72	ND	ND	8.71	<DL	2.4	<DL	8	12.88	16.8	24	7.2	27.95	49.44	32.96
6S	6.24	<DL	5.7	3.5	5.2	11.45	2.6	11.27	7.8	13.28	109.2	49.4	10.24	28.5	19
6B	ND	ND	ND	<DL	2.3	16.96	<DL	5.37	4.6	10.26	20.7	4.83	7.75	24.61	16.41
7S	11	<DL	10	2.5	12.5	50.98	9.84	12.5	17.5	115	192.5	112.5	18.01	49	32.67
7B	ND	ND	<DL	2.6	4.7	ND	ND	ND	ND	13.64	15.23	19.45	3.26	15.08	10.05
8S	17.48	<DL	<DL	<DL	3.8	5.7	1.9	46.87	22.8	106.4	32.3	47.5	52.98	47.31	31.54
8B	ND	ND	<DL	<DL	1.8	3.65	<DL	9.6	1.8	7.2	13.76	10.8	8.24	51.12	34.08
9S	7.22	ND	1.9	13.6	7.4	7.6	<DL	9.5	3.8	11.4	43.7	70.3	19.95	48.83	32.55
9B	7.04	1.6	<DL	8.3	3.2	6.4	1.6	31.47	3.2	14.4	6.4	16	17.53	3.26	2.18
10S	10.08	1.8	3.6	5.4	4.3	7.2	1.8	13.87	12.6	73.8	12.6	34.2	27.79	124.02	82.68
10B	7.22	ND	ND	<DL	1.9	1.9	<DL	ND	ND	3.8	3.8	5.7	10.4	23.56	15.71
11S	19.72	1.7	12.79	<DL	3.4	6.8	<DL	4.53	5.1	61.2	6.8	39.1	16.41	45.9	30.6
12S	8.16	2.4	2.4	7.2	4.8	21.6	2.4	37.6	7.2	9.6	12.56	43.2	26.32	157.44	104.96
13S	9.86	23.65	1.7	5.1	3.4	15.3	1.7	12.47	11.9	35.7	18.7	27.2	55.46	17.51	11.67
14S	10.92	7.9	13.45	23.01	10.98	28.6	2.6	30.33	13	36.4	12.98	23.4	9.59	17.32	11.54
15S	7.6	15.65	10.76	3.8	10.45	22.8	<DL	16.47	3.8	13.3	7.6	41.8	17.73	26.98	17.99

Naph: Naphthalene, Acthy: Acenaphthylene, Ace: Acenaphthene, Phe: Phenanthrene, An: Anthracene, Flu: Fluoranthene, Py: Pyrene, BaA: Benzo(a)anthracene, Chr: Chrysene, BkF: Benzo(k)flouranthene, BaP: Benzo(a)pyrene, BbF: Benzo(b)fluoranthene, InP: Indenopyrene, B(ghi)P: Benzo(g,h,i)perylene, DBA: Di-benz(a,h)anthracene, ND: Not detected, <DL: Lower detection limit, S: Surface, and B: Bottom.

(b)															
Parameter Site	Naph	Acthy	Ace	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkF	BaP	InP	B(ghi)P	DBA
15S	7.6	15.65	10.76	3.8	10.45	22.8	<DL	16.47	3.8	13.3	7.6	41.8	17.73	26.98	17.99
16S	6.05	2.5	8.2	14.23	3.7	2.5	9.8	16.67	19.8	35	13.89	11.87	10.48	78.5	52.33
16B	25.48	10.4	23.76	<DL	5.2	15.82	47.18	28.6	5.2	12.65	15.87	10.18	7.61	13.78	9.19
17S	19.36	35.2	12.26	<DL	2.2	7.56	<DL	16.13	6.6	16.92	11.29	8.39	40.38	75.9	50.6
17B	15.2	3.8	12.56	19.76	9.4	13.7	1.9	14.57	12.5	13.15	16.4	1.9	8.54	26.22	17.48
18S	11.02	30.6	10.8	6.7	3.6	<DL	<DL	22.2	12.76	10.8	18	7.2	20.99	13.68	9.12
18B	15.04	12.8	4.87	1.6	<DL	4.87	<DL	22.4	13.87	6.4	5.21	3.2	5.33	15.04	10.03
19S	29.4	30	13.45	<DL	12.5	<DL	34.21	18	17.13	5.98	23.98	2.94	26.75	41.4	27.6
19B	13.64	15.4	2.2	13.53	8.5	6.6	12.87	8.8	15.4	21.87	<DL	2.2	6.96	31.9	21.27
20S	73.44	4.8	12.34	5.4	4.8	13.87	3.7	8	7.2	13.29	21.26	3.84	20.5	103.92	69.28
20B	11.84	13.87	1.6	<DL	13.72	3.2	<DL	8	15.43	1.6	10.65	13.98	5.11	11.36	7.57
21S	6.56	35.7	6.8	1.7	21.5	1.7	<DL	14.17	15.3	13.6	5.93	9.63	20.82	56.27	37.51
21B	13.5	19.5	3	<DL	3.8	12.7	10.98	14	9	3.6	2.7	5.6	2.62	20.1	13.4
22S	8.64	9.6	17.6	8.5	3.2	<DL	<DL	11.2	8	5.11	12.6	3.29	10.1	39.84	26.56
22B	14.06	5.7	3.8	<DL	1.9	3.5	1.9	19.63	7.6	3.8	3.8	6.56	10.56	14.06	9.37
23S	10	2.5	13.23	<DL	3.65	3.98	<DL	15.83	ND	4.76	<DL	2.5	3.35	19.25	12.83
23B	34.08	2.4	6.47	ND	5.47	<DL	<DL	16	7.2	9.6	2.4	15.82	3.19	35.52	23.68
24S	13.64	13.2	4.4	11.74	3.3	<DL	12.67	8.8	13.23	12.85	17.27	2.2	10.8	17.16	11.44
24B	ND	7.6	10.89	<DL	13.25	18.54	<DL	29.13	9.5	7.6	1.9	9.5	8.68	5.89	3.93
25S	6.03	4.4	4.4	<DL	2.2	<DL	2.2	12.47	4.4	12.76	16.23	13.28	9.91	57.2	38.13
25B	8.64	4.8	13.29	<DL	<DL	7.2	<DL	22.35	16.8	32.84	12.95	2.4	20.2	39.12	26.08
26S	14.76	5.4	12.35	<DL	<DL	<DL	1.8	14.28	15.93	1.8	12.36	11.29	6.17	24.12	16.08
26B	5.12	<DL	11.94	<DL	25.14	6.2	1.6	23.47	34.73	3.2	12.93	4.8	4.61	6.88	4.59
27S	3.61	9.5	3.8	1.9	5.5	5.3	6.7	14.57	3.8	5.7	1.9	5.55	8.64	36.29	25.59
27B	9.86	7.54	3.21	<DL	<DL	<DL	6.8	5.1	7.95	5.84	3.92	3.06	9.52	6.35	
28S	4.94	5.7	11.4	<DL	7.98	3.8	<DL	14.57	5.7	3.8	6.94	11.4	20.84	47.31	31.54
28B	4.7	26.4	16.8	3.76	3.2	11.72	2.4	12.8	7.2	4.8	9.48	5.28	29.98	27.36	18.24

Naph: Naphthalene, Acthy: Acenaphthylene, Ace: Acenaphthene, Phe: Phenanthrene, An: Anthracene, Flu: Fluoranthene, Py: Pyrene, BaA: Benzo(a)anthracene, Chr: Chrysene, BkF: Benzo(k)flouranthene, BaP: Benzo(a)pyrene, BbF: Benzo(b)fluoranthene, InP: Indenopyrene, B(ghi)P: Benzo(g,h,i)perylene, DBA: Di-benz(a,h)anthracene, ND: Not detected, <DL: Lower detection limit, S: Surface, and B: Bottom.

(c)

Parameter Site	Naph	Acthy	Ace	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkF	BaP	InP	B(ghi)P	DBA
29S	15.64	18.4	21.94	<DL	2.3	ND	<DL	15.33	<DL	<DL	<DL	2.3	5.62	ND	<DL
29B	27	21.6	1.8	22.13	32.95	8.4	1.8	9.6	5.4	35.71	3.1	54.37	37.23	21.78	14.52
30S	8.68	13.87	4.2	6.56	4.2	7.6	1.4	12.13	5.89	1.4	16.51	<DL	3.01	10.36	6.91
30B	8.1	7.5	1.5	2.54	2.89	3.79	7.6	5	5.8	4.88	5.89	3.78	2.84	ND	<DL
31S	14.44	17.1	10.82	<DL	8.24	2.98	1.9	8.23	3.8	7.92	3.09	1.87	1.96	11.78	7.85
31B	34.84	5.2	12.09	12.34	23.75	<DL	9.5	15.6	5.2	12.34	10.64	<DL	5.17	ND	ND
32S	12.48	11.22	13.72	<DL	<DL	3.87	10.4	5.2	13.98	12.6	2.6	7.09	ND	ND	<DL
32B	48	7.2	4.8	<DL	4.86	6.78	3.96	10.4	7.2	10.92	11.28	3.87	42.44	48.96	32.64
33S	11.5	25	2.5	<DL	6.8	5	<DL	3.67	16.89	5.32	7.16	2.5	4.04	ND	ND
33B	17.02	18.4	23.76	2.3	11.92	<DL	<DL	5.37	11.82	2.3	<DL	6.15	3.93	42.09	28.06
34S	ND	ND	<DL	<DL	<DL	2.9	<DL	6.77	22.93	3.53	<DL	5.98	3.65	11.02	7.35
34B	4.41	5.7	5.34	9.87	3.87	7.98	<DL	8.23	1.9	7.95	<DL	<DL	2.09	6.84	4.56
35S	40.12	8.5	1.7	<DL	<DL	2.19	<DL	4.53	3.4	1.7	<DL	<DL	4.96	10.2	6.8
35B	30.6	12	3.33	4.27	5.29	3.7	5.18	13	7.5	1.5	2.17	3	101.66	12.6	8.4
36S	4.42	3.4	<DL	ND	ND	<DL	ND	ND	ND	12.67	<DL	<DL	1.29	ND	<DL
36B	75.36	24	23.14	<DL	3.76	<DL	<DL	3.91	12.67	8.98	4.99	2.4	68.72	11.02	7.34
37S	ND	5.5	7.8	9.6	8.5	2.4	<DL	16.2	9.6	2.4	<DL	<DL	ND	9.12	6.08
37B	4.75	17.5	10	12.95	3.65	2.5	<DL	6.67	9.37	8.64	13.24	2.5	181.85	22.25	14.83
38S	16.2	5.4	8.1	2.7	11.9	4.5	2.7	7.2	35.1	5.4	2.7	<DL	ND	ND	<DL
38B	3.78	ND	<DL	2.7	<DL	<DL	<DL	<DL	<DL	2.83	12.86	3.98	20.33	28.35	18.9
39S	28.5	7.5	7.5	25	2.5	<DL	7.98	39.17	7.12	2.5	14.5	7.5	4.33	24.75	16.5
39B	36.48	7.2	5.94	3.79	<DL	2.4	ND	5.6	4.8	7.68	4.8	<DL	22.8	14.4	9.6

Naph: Naphthalene, Acthy: Acenaphthylene, Ace: Acenaphthene, Phe: Phenanthrene, An: Anthracene, Flu: Fluoranthene, Py: Pyrene, BaA: Benzo(a)anthracene, Chr: Chrysene, BkF: Benzo(k)flouranthene, BaP: Benzo(a)pyrene, BbF: Benzo(b)fluoranthene, InP: Indenopyrene, B(ghi)P: Benzo(g,h,i)perylene, DBA: Di-benz(a,h)anthracene, ND: Not detected, <DL: Lower detection limit, S: Surface, and B: Bottom.

(d)

Parameter Site	Naph	Acthy	Ace	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkF	BaP	InP	B(ghi)P	DBA
40S	5.02	8.8	2.2	11	3.8	7.93	8.3	6.6	3.98	17.6	13.2	12.67	6.61	20.9	13.93
40B	15.84	11.98	23.45	13.2	12.34	11	11.43	22	5.67	6.6	12.78	23.89	5.64	4.4	2.93
41S	23.56	3.98	2.98	5.7	21.67	3.6	3.29	8.87	4.67	3.86	1.97	6.98	5.75	1.33	0.89
41B	21.08	5.98	4.67	6.8	3.4	8.5	3.87	29.47	3.4	18.7	11.45	7.65	9.22	24.82	16.55
42S	6.8	5.1	3.4	3.4	3.4	3.4	4.67	5.67	3.98	6.8	0	2.97	4.22	15.64	10.43
42B	11.78	7.6	3.8	6.41	5.7	38	2.68	5.07	1.9	7.6	3.8	9.5	5.4	9.31	6.21
43S	24.82	8.5	7.93	5.1	2.86	8.2	9.57	9.63	8.5	10.2	12.29	6.94	5.04	11.05	7.37
43B	9.5	5.7	5.7	7.6	5.7	7.6	3.8	10.77	5.7	11.4	5.7	20.9	4.38	32.49	21.66
44S	23.52	3.29	7.95	3.76	2.4	6.3	2.4	10.4	4.8	4.8	7.98	3.18	8.45	24.72	16.48
44B	35.36	7.94	10.4	4.57	2.6	13	7.8	13	10.4	26	15.6	23.4	14.17	25.74	17.16
45S	11	5.9	12.5	23.7	3.9	1.92	5.3	2.78	5.94	8.8	3.94	2.2	3.72	10.78	7.19
45B	27.14	4.6	6.9	7.94	4.6	13.8	<DL	9.2	23	4.6	13.8	9.2	14.55	92.23	61.49
46S	4.37	3.99	10.4	<DL	11.3	2.6	8.91	7.8	18.2	7.8	11.84	7.94	7.75	26.26	17.51
46B	5.2	3.29	23.94	7.5	4.68	6.88	6.48	10.83	7.96	17.5	10.19	4.96	9.21	23.5	15.67
47S	24.48	6.93	8.74	9.39	4.8	8.49	3.92	4.38	2.4	9.88	8.97	4.83	5.98	37.44	24.96
47B	7.92	6.59	10.8	3.02	3.6	10.8	7.86	21	5.4	14.4	18.75	13.56	10.78	28.44	18.96
48S	12.6	8.95	3.6	21.83	9.48	7.49	5.48	9.6	4.94	12.39	6.59	5.4	6.44	27	18
48B	24.24	10.8	4.8	26.4	5.94	7.2	8.75	13.6	3.6	10.29	8.4	13.2	3.02	15.48	10.32
49S	25.22	9.87	3.55	6.5	2.6	3.9	5.9	8.67	8.96	9.1	8.77	4.39	4.62	26.65	17.77
49B	11	4.76	8.65	12.5	2.2	13.2	6.95	11.73	9.76	15.4	17.3	6.6	13.02	37.62	25.08

Naph: Naphthalene, Acthy: Acenaphthylene, Ace: Acenaphthene, Phe: Phenanthrene, An: Anthracene, Flu: Fluoranthene, Py: Pyrene, BaA: Benzo(a)anthracene, Chr: Chrysene, BkF: Benzo(k)flouranthene, BaP: Benzo(a)pyrene, BbF: Benzo(b)fluoranthene, InP: Indenopyrene, B(ghi)P: Benzo(g,h,i)perylene, DBA: Di-benz(a,h)anthracene, ND: Not detected, <DL: Lower detection limit, S: Surface, and B: Bottom.

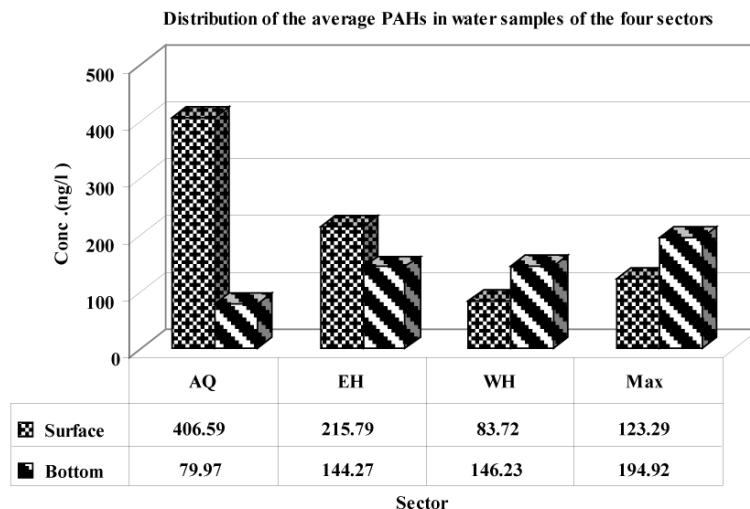


Figure 2. Variation of average PAHs concentrations (ng/l) in surface and bottom water samples of the four areas.

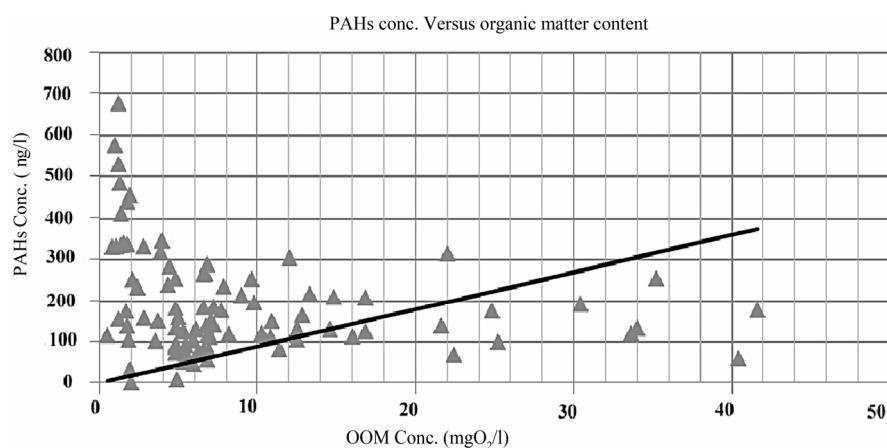


Figure 3. Relationship between total PAH concentrations (ng/l) and OOM (mgO₂/l) of the studied area.

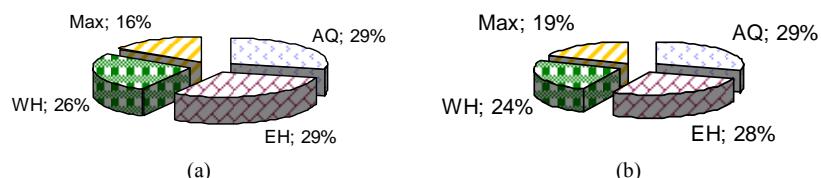


Figure 4. A comparison of the average combustion% of PAHs in: (a) surface and (b) bottom waters in the four areas.

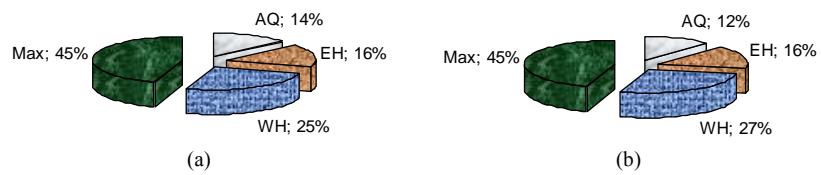


Figure 5. A comparison of the average fossil% of PAHs in: (a) surface and (b) bottom water in the four areas.

water samples collected from the studied area during 2010. The average values of water salinity at different areas revealed that the salinity decreased in the order: WH > EH > AQ > Max, the average pH values are decreasing in the order; AQ > WH > EH > Max. Concentration of DO was decreasing in the order; EH > AQ > W > Max, in case of OOM; it was noticed that the general trend showed that AQ exhibited the lowest content of OOM compared to other areas and the order was decreasing from: EH > Max > WH > AQ.

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Table 2. Comparison of PAHs concentrations (ng/l) in water recorded in the present study with areas of the surroundings.

Name of Site	Range (ng·l ⁻¹)	Reference
Abu-Qir Bay, Alexandria, Egypt	127.17 - 456.19	Present study
Eastern Harbour, Alexandria, Egypt	75.10 - 286.02	
Western Harbour, Alexandria, Egypt	29.31 - 292.72	
El-Max Bay, Alexandria, Egypt	107.95 - 209.0	
Deep Bay, South China	69.4 - 24.7	[3]
Tonghui River of Beijing, China	192.5 - 2651	[9]
Red Sea Coast, Egypt	400 - 96,450	[25]
Alexandria Coast, Egypt	13 - 120	[26]
Alexandria Coast, Egypt	117.5 - 564	[27]
Brighton Marina, UK	<2 and 11,400	[40]
Jiulong River Estuary and Western Xiamen Sea, China	6960 - 26,900	[43]
Aquatic Environment, El-Menofiya Governorate, Egypt	226.9 - 1492.2	[44]
Creek, Niger Delta region	24,390 - 283,600	[45]
Bahía Blanca Estuary, Argentina	ND - 4000	[46]
Gerlache Inlet Sea, Antarctica	5.27 - 9.43	[47]

Table 3. (a) Salinity %, pH, OOM (mgO₂/l), DO (mg/l), ΣPAHs (ng/l), ΣCOMP (ng/l), ΣCARC (ngng/l), and ΣTFPAH (ng/l) in water samples collected From Abou Qir Bay (AQ) during 2010; (b) Salinity %, pH, OOM (mgO₂/l), DO (mg/l), ΣPAHs (ng/l), ΣCOMP (ng/l), ΣCARC (ngng/l), and ΣTFPAH (ng/l) in water samples collected From Eastern Harbour during 2010; (c) Salinity %, pH, OOM (mgO₂/l), DO (mg/l), ΣPAHs (ng/l), ΣCOMP (ng/l), ΣCARC (ngng/l), and ΣTFPAH (ng/l) in water samples collected From Western Harbour during 2010; (d) Salinity %, pH, OOM (mgO₂/l), DO (mg/l), ΣPAHs (ng/l), ΣCOMP (ng/l), ΣCARC (ngng/l), and ΣTFPAH (ng/l) in water samples collected From El-Max Bay during 2010.

Parameter	(a)															
	Salinity %		pH		OOM (mgO ₂ /l)		DO (mg/l)		ΣPAHs (ng/l)		ΣCOMP (ng/l)		ΣCARC (ngng/l)		ΣTFPAH (ng/l)	
Site	S	B	S	B	S	B	S	B	S	B	S	B	S	B	S	B
1	38.7	NR	8.48	NR	1.35	NR	2.93	NR	411.17	NR	327.17	NR	206.57	NR	67.2	NR
2	38.5	38.5	8.65	8.71	1.19	1.98	4.55	5.53	531.16	2.21	427.16	2.21	323.42	2.21	91	NC
3	8.5	38.4	8.22	8.57	1.9	1.19	6.18	4.23	456.14	156.6	409.34	156.6	280.46	78.4	41.4	NC
4	35.1	38.7	8.9	8.5	1.27	1.66	5.85	3.87	484.98	175.56	431.78	171.16	224.11	109.56	45.6	2.2
5	34.7	38.6	9.08	8.51	1.74	2.37	6.94	4.52	440.5	232.54	325.5	196.54	210.96	142.3	98.9	33.6
6	36.6	38.6	9.01	8.75	1.19	1.82	7.1	5.16	677.25	106.81	635.65	104.51	285.69	70.7	33.8	NC
7	38	38.5	8.87	8.73	0.95	1.9	5.97	3.55	578.03	34.61	498.03	29.41	410.03	14.33	65	NC
8	38.4	38.6	8.72	8.69	0.79	1.74	6.45	4.52	330.15	139.59	238.95	137.79	161.24	83.07	87.4	NC
9	38.4	38.5	8.75	8.74	1.35	2.37	4.52	3.23	333.83	235.12	292.03	193.52	231.8	179.06	38	36.8
10	38.4	38.4	8.81	8.81	1.03	0.47	4.52	6.45	330.79	116.57	265.99	78.57	231.98	53.11	55.8	36.1
11	38.9	NR	8.98	NR	2.77	NR	3.23	NR	330.33	NR	224.93	NR	167.13	NR	102	NR
12	28.7	NR	8.98	NR	1.5	NR	5.48	NR	339.27	NR	286.47	NR	239.83	NR	45.6	NR
13	33.8	NR	8.72	NR	2.06	NR	4.19	NR	252.14	NR	190.94	NR	144.53	NR	52.7	NR
14	35	NR	9.07	NR	1.74	NR	3.55	NR	335.5	NR	265.3	NR	203.79	NR	62.4	NR
15	38	NR	8.68	NR	6.65	NR	3.87	NR	267.63	NR	218.23	NR	164.65	NR	41.8	NR

OOM: Oxidizable organic matter, DO: Dissolved oxygen, Av: average, Max: maximum, Min: minimum, NR: Not collected, NC: Not calculated, PAHs: Sum of aromatic hydrocarbons, ΣCARC: Sum of BaA + BbF + BkF + BaP + DBA + InP, ΣTFPAH: Sum Acenaphthylene + Acenaphthene, ΣCOMP: Sum of all PAHs except those calculated for ΣTFPAH.

Parameter	(b)															
	Salinity %		pH		OOM (mgO ₂ /l)		DO (mg/l)		ΣPAHs (ng/l)		ΣCOMP (ng/l)		ΣCARC (ngng/l)		ΣTFPAH (ng/l)	
Site	S	B	S	B	S	B	S	B	S	B	S	B	S	B	S	B
16	39.1	39.7	7.76	7.78	3.84	24.8	6.13	5.24	317.2	176.93	274.45	143.39	173.45	119.21	35.25	28.34
17	38.9	39.4	7.63	7.56	22	33.6	3.15	4.6	313.74	121.2	262.26	104.1	177.56	72.18	49.28	13.3
18	39.1	39.2	7.5	7.56	6.8	21.6	3.23	3.79	288.94	140.42	187.06	113.7	166.18	93.86	96.48	25.12
19	39.1	39.4	7.57	7.65	9.6	34	3.63	5.16	253.59	134.05	202.89	105.23	155.49	49.13	47.7	24.42
20	39.1	39.3	7.64	7.71	12	22.4	4.84	4.76	304.03	70.7	250.51	58.38	132.19	42.22	43.92	10.72
21	39.2	39.6	7.75	7.71	4.4	16.8	5.81	4.19	283.71	126.59	205	95.84	131.73	63.74	75.31	29.25
22	39.3	39.5	7.72	7.78	2.8	4.8	5.32	4.68	158.77	135.32	122.45	116.89	74.61	91.43	31.52	16.53
23	39.3	39.4	7.82	7.86	25.2	7.2	6.05	5.81	100.95	142.94	88.45	118.7	64.2	75.98	10	21.84
24	39.3	39.3	7.74	7.81	10.8	16.8	6.13	4.76	113.78	209.46	84.96	198.06	61.2	180.77	24.42	9.5
25	39.5	39.7	7.75	7.8	30.4	7.2	5.24	5.97	192.96	187.03	151.82	175.51	88.02	112.39	38.94	11.52
26	39.5	39.7	7.59	7.69	4.8	16	5.89	5.08	88.83	114.66	72.45	107.3	42.93	92.42	16.38	5.76
27	39.5	39.6	7.68	7.71	41.6	40.4	4.68	5.65	178.2	61.37	143.05	53.04	97.07	38.42	31.35	8.33
28	39.4	39.7	7.69	7.62	14.8	35.2	4.68	3.39	210.6	254.83	166.9	183.31	110.09	131.95	41.8	66.72

OOM: Oxidizable organic matter, DO: Dissolved oxygen, Av: average, Max: maximum, Min: minimum, NR: Not collected, NC: Not calculated, PAHs: Sum of aromatic hydrocarbons, ΣCARC: Sum of BaA + BbF + BkF + BaP + DBA + InP, ΣTFPAH: Sum Acenaphthylene + Acenaphthene, ΣCOMP: Sum of all PAHs except those calculated for ΣTFPAH.

(c)

Parameter	Salinity %		pH		OOM (mgO ₂ /l)		DO (mg/l)		ΣPAHs (ng/l)		ΣCOMP (ng/l)		ΣCARC (ng/l)		ΣTFPAH (ng/l)	
	Site	S	B	S	B	S	B	S	B	S	B	S	B	S	B	S
29	40.8	42.2	7.77	7.75	4.8	6.56	3.99	3.72	94.97	185.29	64.15	144.79	64.15	114.01	28.52	36.9
30	39.1	40.3	7.72	7.77	4.8	5.92	3.99	3.87	75.32	48.72	59.78	32.67	45.22	28.17	9.94	13.05
31	40.1	40.5	7.71	7.86	6.88	3.52	4.55	4.19	81.81	103.17	53.69	72.75	34.31	64.95	26.22	25.22
32	40.5	40.7	7.72	7.83	6.8	4.8	4.03	4.11	58.24	253.84	41.6	215.44	26	152.08	16.64	36
33	41	41.6	7.77	7.78	6.4	6.96	3.91	4.11	73.83	113.67	38.08	75.26	30.58	28.57	33.25	31.51
34	40.8	40.7	7.75	7.76	5.28	5.44	4.47	5.69	54.23	74.86	54.23	41.42	37.41	30.78	NC	29.64
35	40.4	40.1	7.78	7.76	6.16	6.88	5.93	4.94	72.47	123.23	42.21	89.93	26.91	65.33	30.26	28.8
36	41	41.3	7.76	7.78	4.88	3.92	4.43	4.19	9.89	345.29	4.28	278.81	4.28	163.85	5.61	64.08
37	40.5	41.2	7.8	7.79	5.36	3.68	4.74	6.33	54.72	152.37	45.12	96.12	24	68.87	4.8	51.25
38	40.8	41.2	7.81	7.81	5.92	6.56	6.05	5.53	105.3	81.7	78.3	77.11	37.8	48.76	21.6	1.89
39	40.5	40.6	7.77	7.74	4.32	5.28	5.06	4.19	240.15	126.42	183.4	91.38	151.15	69.78	29.25	32.64

OOM: Oxidizable organic matter, DO: Dissolved oxygen, Av: average, Max: maximum, Min: minimum, NR: Not collected, PAHs: Sum of aromatic hydrocarbons, ΣCARC: Sum of BaA + BbF + BkF + BaP + DBA + InP, ΣTFPAH: Sum Acenaphthylene + Acenaphthene, ΣCOMP: Sum of all PAHs except those calculated for ΣTFPAH.

(d)

Parameter	Salinity %		pH		OOM (mgO ₂ /l)		DO (mg/l)		ΣPAHs (ng/l)		ΣCOMP (ng/l)		ΣCARC (ng/l)		ΣTFPAH (ng/l)	
	Site	S	B	S	B	S	B	S	B	S	B	S	B	S	B	S
40	36.2	40	7.27	7.3	10.88	7.68	3.87	3.64	151.6	177.61	102.32	141.09	74.82	114.69	36.08	18.92
41	23.6	38.5	7.04	7.33	11.36	8.96	3.56	4.03	83.51	213.61	60.33	182.67	51.4	144.25	15.58	20.74
42	9.7	38	6.83	7.13	5.44	12.48	1.11	4.27	93.77	132.39	75.07	107.5	50.93	56.39	11.9	17.29
43	25.8	38.5	6.94	7.14	8.16	12.8	2.93	3.87	120.24	165.7	90.83	136.25	67.88	86.66	22.61	16.15
44	36.3	38.6	7.17	7.29	6.72	7.84	4.03	4.19	139.78	235.56	104.02	199.68	69.7	142.74	26.16	30.68
45	35.7	38.7	7.12	7.3	12.48	6.56	2.21	4.19	107.32	266	84.22	229.43	64.64	100.4	14.3	25.07
46	28.8	38.8	7.1	7.29	7.04	4.8	2.69	5.14	152	182.93	114.56	131.93	64.9	90.93	34.84	41
47	27.4	38.6	7.07	7.27	14.56	13.28	3.16	5.38	131.23	217.76	106.99	186.8	62.35	140.36	17.04	18.36
48	30.8	38.9	7.08	7.27	10.24	4.96	3.8	5.69	121.79	160.84	99.29	104.32	61.49	75.64	11.7	27.72
49	34.9	38.9	7.2	7.3	6.08	9.76	4.66	5.53	131.7	196.77	94.39	171.47	58.64	103.05	28.21	20.9

OOM: Oxidizable organic matter, DO: Dissolved oxygen, Av: average, Max: maximum, Min: minimum, NR: Not collected, NC: Not calculated, PAHs: Sum of aromatic hydrocarbons, ΣCARC: Sum of BaA + BbF + BkF + BaP + DBA + InP, ΣTFPAH: Sum Acenaphthylene + Acenaphthene, ΣCOMP: Sum of all PAHs except those calculated for ΣTFPAH.

Table 4. Correlation coefficient matrix for water individual PAHs and some physic-chemical parameters (n = 92).

Cpd	Naph	Acthy	Ace	Phe	Ant	Flu	Pyr	BaA	Chr	BbF	BkF	BaP	InP	B(ghi) P	DBA	OOM	pH	DO	PAH
Naph	1.000																		
Acthy	0.212	1.000																	
Ace	0.274	0.312	1.000																
Phe	0.034	-0.017	0.001	1.000															
Ant	0.068	0.188	0.144	0.205	1.000														
Flu	-0.032	-0.197	-0.013	0.147	0.155	1.000													
Pyr	0.183	0.158	0.300	0.073	0.098	0.067	1.000												
BaA	0.012	0.024	0.163	0.140	0.118	0.212	0.212	1.000											
Chr	0.020	0.059	0.064	-0.011	0.228	0.244	0.055	0.227	1.000										
BbF	-0.013	-0.188	-0.070	0.065	0.113	0.568 ^{**}	0.034	0.288	0.482 ^{**}	1.000									
BkF	-0.037	-0.187	-0.006	0.004	0.102	0.472 ^{**}	0.104	0.069	0.242	0.554 ^{**}	1.000								
BaP	-0.095	-0.221	-0.112	0.167	0.194	0.652 ^{***}	-0.028	0.209	0.101	0.641 ^{***}	0.606	1.000							
InP	0.168	0.200	0.025	0.011	0.013	0.163	-0.059	0.106	0.136	0.254	0.044	0.119	1.000						
B(ghi)P	0.040	-0.187	-0.089	0.072	-0.060	0.387 [*]	-0.073	0.190	0.053	0.322 [*]	0.151	0.455 ^{**}	0.211	1.000					
DBA	0.040	-0.187	-0.090	0.071	-0.060	0.387 [*]	-0.073	0.190	0.053	0.321 [*]	0.151	0.454 ^{**}	0.211	1.000 ^{***}	1.000				
OOM	-0.023	0.240	0.144	-0.069	0.012	-0.133	0.195	0.029	-0.088	-0.256	-0.202	-0.279	-0.158	-0.138	-0.135	1.000			
pH	-0.229	-0.298	-0.261	-0.116	-0.026	0.359 [*]	-0.223	0.143	0.088	0.458 ^{**}	0.396 ^{**}	0.516 ^{**}	0.177	0.355 [*]	0.354 [*]	-0.400 ^{**}	1.000		
DO	-0.062	-0.267	-0.101	-0.064	-0.083	0.151	-0.046	0.101	0.028	0.106	0.281	0.209	0.102	0.195	0.195	-0.053	0.325 [*]	1.000	
PAH	0.127	-0.050	0.097	0.131	0.109	0.650 ^{***}	0.038	0.362 [*]	0.313 [*]	0.640	0.642	0.721	0.299	0.576	0.575 ^{***}	-0.237	0.494 ^{**}	0.205	1.000

Correlations are significant at ^{*}p < 0.05 (low significant), ^{**}p < 0.01 (moderate significant) and ^{***}p < 0.001 (high significant).

4. Conclusion

The present comprehensive study represents the detailed study of the distribution and correlation between physicochemical parameters and PAHs in 92 water samples collected from four semi-closed areas along the Alexandria coast of Egypt. The studied samples were less contaminated by petroleum hydrocarbons. The average concentration of PAHs in eastern-southern Mediterranean Sea waters are shown to be substantially lower in the present study to those in most areas of urbanized estuaries and coastal areas in the other countries. The maximum concentration of PAHs of 677.3 ng/l was recorded in the most contaminated site (station 6; AQ). This study indicated that the origin of contamination was pyrogenic in all sectors except Max was petrogenic. The present study declared that HMW PAHs are correlated significantly with most of the physicochemical parameters; while no correlations were found between LMW PAHs and most physicochemical parameters at low concentrations of PAHs.

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