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Polycyclic Aromatic Hydrocarbons (PAHs) in water of Al-Kahlaa River in Missan Province, Iraq

Salih Hassan Jazza[1], Abdul-Hussain Y. Al-Adhub[2], Hamid T. Al-Saad[3]*

[1]*Department of Biology, College of Science, University of Missan, Missan, Iraq*
[2]*Department of Biology, College of Science, University of Basrah, Basrah, Iraq*
[3]*Department of Marine environmental Chemistry, Marine Science Center, University of Basrah, Basrah, Iraq*
Email: htalsaad@yahoo.com

Abstract

The present study was performed to have knowledge of Polycyclic Aromatic Hydrocarbons pollution status in water. The samples were collected during two seasons (winter and summer 2012 ,2013) from four different stations) Al-Magideh, Treatment unit, Al-Husaichi and Al-Zubair) distributed along Al-Kahlaa River in Missan province, in addition to reference station lies on the Tigris river before 25 Km from entering to Amara city. The concentrations of PAHs were determined in by using capillary Gas Chromatography. Results of the present study revealed that the total concentrations of PAHs in dissolved fraction ranged from 0.739 ng.l^{-1} in Reference station to 1.974 ng.l^{-1} in Treatment unit during winter, and from 0.300 ng.l^{-1} in Reference station to 1.125 ng.l^{-1} in Treatment unit during summer, while in the particulate fraction they varied from 0.79 ng.g^{-1} dry weight in Reference station to 24.42 ng.g^{-1} dry weight in Treatment unit during winter, and from 4.369 ng.g^{-1} dry weight in Reference station to 10.545 ng.g^{-1} dry weight in Al-Husaich during summer. It had been noticed that there were a predominance of high molecular weight PAHs on low molecular weight, while BaA/(BaA+Chr) ratio in water ranged from 0.218 to 0.804. InP/(InP+BghiP) ratio ranged between 0 and 0.578, where as Fl/Py ratio ranged between 0.150 and 2, this give an indication of the origin of PAHs compounds in water which were mainly pyrogenic and few from them petrogenic.

Keywords: *polycyclic aromatic hydrocarbons, PAHs, water, pollution*

Introduction

Pollution of the rivers water in Iraq is considered as important problem that must treated and controlled it, so not being complicated in future, such as what occurred in other industrial countries which have high population density (Al-Sayegh and Taka, 2002). Now a days, Petrochemical industries are considered as one of the most important basic industries all over the world. Although these industries have too many benefits for our life, the pollution caused by Petrochemical industries have been a cause for concern and major challenges to save the environment against their adverse impacts. These impacts not only affect the biological characters of the ecosystem but also can affect the water resource quality and threat human health (Esmaeli-Sari, 2002; Rooney, 2005).

Controlling such pollution problems in the aquatic environment is very complex because of the large number of input sources and their geographic dispersions. Contrary to popular views, evidences were accumulated to buttress the fact that these compounds mixes with water and penetrates to the under lying sediments (Patin, 1999). The chemical composition of each crude oil and petroleum products vary significantly, and can have diverse effects on different living organisms within the same ecosystem (Overton *et al*., 1994). These differences in toxic effects are attributed to qualitative compositional differences in the various products, in addition to concentration differences of the chemical constituents (Albert, 1995; Sverdrup, 2003).

Polycyclic Aromatic Hydrocarbons (PAHs) may reach aquatic environments from industrial and domestic sewage effluents, exhaust of gasoline and diesel combustion engines, surface run off from land, deposition of airborne particulates, and especially from spillage of petroleum and petroleum products into water bodies (Prahl *et al*., 1984). The majority of PAHs enter aquatic environments remain close to sites of deposition, suggesting that rivers near centers of human populations are the primary repositories of aquatic PAHs (Neff, 1979). In water, PAHs may either disperse into the water column or evaporate or become incorporated into bottom

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*) Corresponding author

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sediments or concentrate in aquatic biota or affected by biodegradation and chemical oxidation (Suess, 1976).

In the aquatic ecosystems, most PAHs are associated with the particulate fraction of PAHs due to their hydrophobic properties giving rise to the accumulation in the sediments (Perra *et al*., 2009; Qiu *et al*., 2009). This study is the first of its kind in Al-Kahlaa River which provides a necessary information on the pollution of this region by Polycyclic Aromatic Hydrocarbons (PAHs) and aim to determine the concentrations, seasonal and spatial variations in addition to the sources of these compounds in water.

Materials and Methods

Collection and extraction of dissolved and particulate fractions of these compounds in water samples

The samples were collected during two seasons (winter and summer 2012, 2013) from four different stations) Al -Magideh, Treatment unit, Al-Husaichi and Al-Zubair) distributed along Al-Kahlaa River in Missan province, in addition to reference station lies on the Tigris River before 25 Km from entering to Amara city as shown in (Fig. 1). The samples of water were suction filtered as soon as possible (within 5 h) through pre-weighted 0.45 µm pore size whatman glass fiber filter (GFF) pre-ignited at 450°C. Materials passing through the filter were considered as dissolved fraction and the retained as particulate fraction, then the CCl_4 was added to the dissolved fraction until the arrival to the lab for analysis, whereas particulate fraction was dried and wrapped in aluminum foil and transferred to the laboratory in ice packed and kept in freezer prior to analysis .

The hydrocarbons were extracted from dissolved fraction following the procedure of UNEP (1989). While from particulate fractions following a method of Goutex and Saliot (1980). The concentrations of PAHs in samples were determined by using capillary Gas Chromatography. Simple thermometer with range from 0 to 100 ^{0}C graduated at 0.2 ^{0}C was used to measure water temperature.

Evaluation indices of oil pollution

Many parameters have to be analyzed in order to evaluate the probable origin of Polycyclic Aromatic Hydrocarbons (pyrogenic or petrogenic). These are as follows :

A. Ratio of Low Molecular Weight (LMW-PAHs) to High Molecular Weight (HMW-PAHs). Values more than one indicate petrogenic sources from crude oil and their derivatives and values less than one are pyrogenic sources (Vrana *et al*., 2001).

B. Ratio of Benzo(a)Anthracene/(Benzo(a) Anthracene + Chrysene). BaA/(BaA+Chr). Ratio of BaA/(BaA+CHR) less than 0.2 implies petrogenic, from 0.2 to 0.35 indicates either petrogenic or pyrogenic origin sand larger than 0.35 implies pyrogenic sources (Guo *et al*., 2007).

C. Ratio of Indeno (1,2,3-cd)pyrene/(Indeno (1,2,3-cd)Pyrene + Benzo(ghi)Perylene). InP/(InP + BghiP). Values less than 0.2 implies petrogenic, from 0.2 to 0.5 indicates either pyrogenic or petrogenic origins and higher than 0.5 are implies pyrogenic origins (Guo *et al*., 2007).

D. Ratio of fluoranthene to pyrene (Fl/Py). Values more than one have been used to indicate pyrogenic sources and values less than one are attributed to petrogenic sources (Qiu *et al*., 2009).

Results and Discussion

Total PAHs concentrations as dissolved fraction ranged from 0.739 to 1.974 ng.l^{-1} and 0.300 to 1.125 ng.l^{-1}, where as in particulate fraction ranged from 5.796 to 24.42 ng.g^{-1} d.w and 4.639 to 10.545 ng.g^{-1} d.w during winter and summer, respectively (Tables 1,2,3 and 4). The highest and the lowest mean concentrations of PAHs in both dissolved and particulate fractions were observed in winter and summer, respectively. This may be attributed to the climatic condition that affect photo-oxidation, volatilization and high degradation during summer (Mohammed *et al*., 2009). PAHs dissolved in the water column will probably degrade rapidly through photo-oxidation and degrade most rapidly at elevated temperatures and dissolved oxygen and at higher incidences of solar radiation (Suess, 1976). Intensive solar radiation is a characteristics of weather feature in Iraq coupled with relatively high water temperature during summer (Table 5) leading to high evaporation rate and increase biodegradation processes by microorganisms (Al-Edanee *et al*., 1990). Photo degradation is an important degradation pathway in aquatic ecosystems for high molecular weight PAHs (Suess, 1976). Photo-oxidation can chemically transform PAHs, and the resulting products may be more toxic and carcinogenic than the parent compounds (NRCC, 1983).

Evaporation process plays an important role in the removal of low molecular weight PAHs from aquatic ecosystems. Also PAHs are subjected to biodegradation by various microorganisms such as fungi, bacteria, and certain algae that are suspended in the water column (Gibson, 1976). Microbial degradation of PAHs is one of the main processes responsible for removing these compounds from water column. Biodegradation of PAHs depends on such factors like type of ring fusion and the number of aromatic rings (Wild *et al*., 1991). Whereas the highest values during winter may be attributed to the lower rates of photo-oxidation and microbial degradation of PAHs during winter leading to relatively higher PAH concentrations compared with summer, in addition to increase hydrocarbon compound input to aquatic environment with run off and increase for use fuel in heating during winter (Al-Khatib, 2008).

Results of the present study revealed that the total concentrations of PAH compounds were in the particulate fraction more than dissolved fraction along the period of study, this due to the tendency of PAHs to adsorb on to solid materials inaquatic ecosystems and because of their hydrophobic nature and low water solubility (Sloof *et al*., 1989).

The association of PAHs with the solid objects depends on their molecular weight. In addition to that PAHs adsorbed to water suspended matters are more resistant to photo-degradation (McGinnes and Snoeyink,1974). Other researchers, however, found that PAHs attached to particulate matter are more susceptible to photolysis than PAHs in solution (Neff, 1979; Moore and Ramamoorthy, 1984).

In this study HMW-PAHs were more than LMW-PAHs in both dissolved and particulate fractions in all stations along the period of study, this may be attributed to the molecular weight. Differences in hydrophobicity and degradation processes in both low and high molecular weight PAHs. A wide array of microorganisms including fungi, algae and bacteria are known to degrade PAHs. However, bacteria play by far the most important role in completion of mineralization.

Lower molecular weight PAHs such as naphthalene and phenanthrene degrade rapidly, but higher molecular weight PAHs such as pyrene, fluoranthene, benzo(a) anthracene and

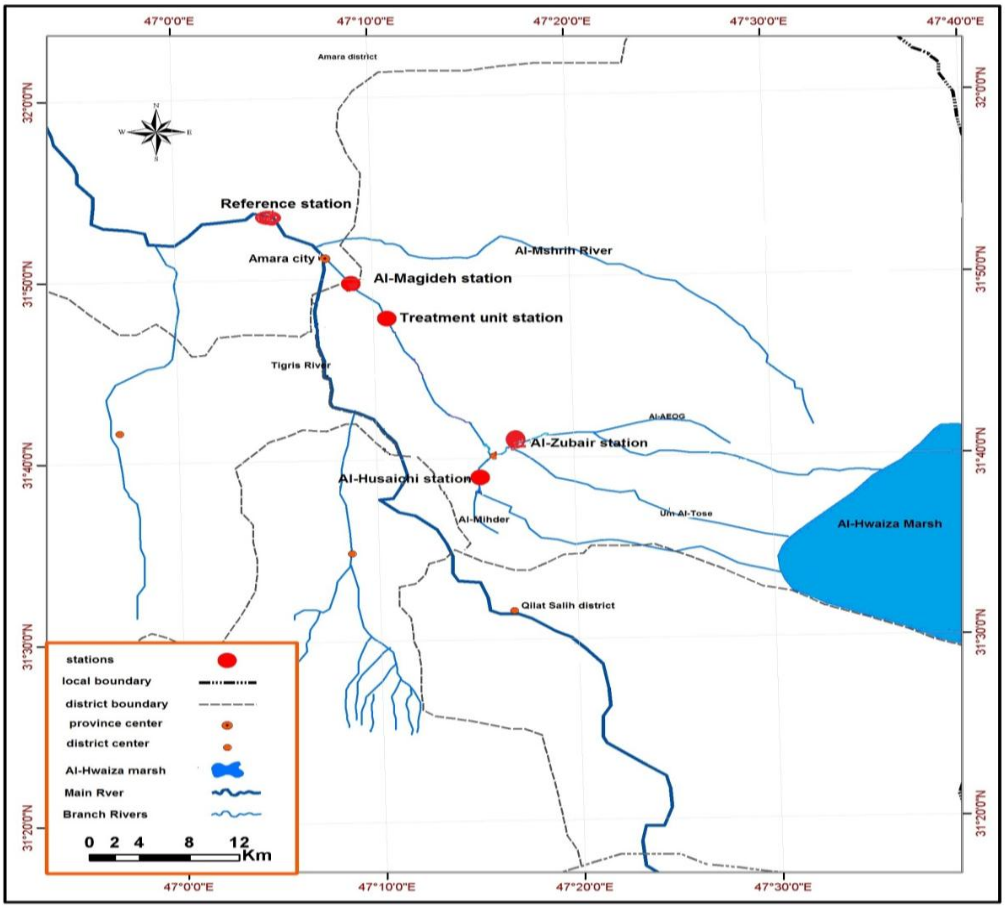

Figure 1. Map of Al-Kahlaa River showing the position of stations

Table 1. Levels of PAHs ($ng.l^{-1}$) in dissolved fraction during winter

Name compound	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Naphthylene	0.0185	0.007	nd	0.011	nd
Indol	nd	nd	nd	nd	nd
2-methyl naphthylene	0.031	0.051	nd	0.028	nd
1-methyl naphthylene	0.013	0.008	nd	0.022	nd
Biphenyl	0.008	0.008	nd	0.014	0.007
Acenaphthylene	0.009	0.014	0.014	0.011	0.010
Acenaphthene	0.018	0.017	0.011	0.033	0.009
Dibenzofuran +fluorene	0.020	0.024	0.015	0.014	0.011
Anthracene+phenthrathrene	0.009	0.014	0.029	0.013	0.018
Fluoranthene	0.012	0.024	0.020	0.013	0.013
Carbazole	0.129	0.090	0.027	0.112	0.068
Pyrene	0.023	0.02	0.072	0.015	0.033
Benzo(a)anthracene	0.019	0.063	0.025	0.048	0.017
Chrysene	0.016	0.226	0.058	0.063	0.017
Benzo(b+k)fluornanthene	0.017	0.037	0.027	0.151	0.094
Benzo(a) pyrene	0.448	1.365	0.644	0.574	0.326
Indeno (1,2,3-cd)pyrene	0.133	nd	nd	0.102	0.056
Dibenzo(a,h)anthracene	0.021	nd	nd	nd	nd
Benzo(ghi)perylene	0.092	nd	nd	0.078	0.053
Total	1.045	1.974	0.946	1.310	0.739
LPAHs	0.131	0.145	0.070	0.149	0.05
HPAHs	0.901	1.804	0.855	1.147	0.66
L/H	0.145	0.080	0.082	0.130	0.086
BaA/(BaA+Chr)	0.552	0.218	0.302	0.432	0.502
InP/(InP+BghiP)	0	0	0	0	0
fl/py	0.538	1.204	0.278	0.850	0.40

*nd :not detected

benzo(a) pyrene are more recalcitrant (Bakhtiari, 2009; Obayori and Salam, 2010). In addition to that the low molecular weight PAHs such as naphthalene has the highest vapor pressure of PAHs, therefore volatilization in aquatic ecosystems is probably the most important removal mechanism for these compounds (Southworth, 1979).

As an individual of PAHs in dissolved fraction Benzo(a)pyrene form the greater values ranged from 0.326 to 1.365 and from 0.123 to 1.125 during winter and summer, respectively while in particulate fraction benzo(a)pyrene also form the greater values 15.134 and 4.162 in Treatment unit and Al-Zubair stations, respectively. Acenaphthene and benzo(b+k)fluornanthene compound form the higher values 2.051 and 4.316 in Al-Magidehand Al-Husaichi stations during winter, respectively Alsobenzo(a)pyrene compound form greater values 1.606, 3.791 and 1.789 in Treatment unit, Al-Husaichi and Al-Zubair stations, respectively and Benzo(ghi)perylene form the higher values 1.390 in Al-Magideh station during summer. Benzo(a)pyrene levels and other individual of HMW- PAHs like total PAHs, were higher in winter than summer probably due to increase hydrocarbon compounds input to aquatic environment with run off because much of the PAHs released into the atmosphere and eventually reach the soil and these PAHs may be washed off by rain to reach the aquatic environment, in addition to that the increase in the use of fuel in heating during Winter (Lee and Grant, 1981; Al-Khatib,2008).

To determine the origins of PAHs as dissolved and particulate fractions according to the following ratios. The ratio of LMW-PAHs to HMW-PAHs (LMW/HMW) had been used to distinguish between petrogenic (>1 values) and pyrogenic (<1 values) origins.The ratios LMW-PAHs/HMW-PAHs were less than one in all stations during winter and summer except in Al-Magideh station was more than one during winter. Petrogenic and natural organic matter sources of PAHs were dominated by low molecular weight PAHs, while combustion derived PAHs are predominantly of high molecular weight (Neff, 1979). The results indicate that the origin of these compounds in dissolved fraction were pyrogenic except in Al-Magideh station was petrogenic. In addition to that BaA/(BaA+Chr) ratioranged from 0.218 to 0.552 and 0.243 to 0.368 in dissolved fraction and from 0.31 to 0.46

Table 2. Levels of PAHs (ng.l^{-1}) in dissolved fraction during summer

Name compound	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Naphthylene	nd	0.006	nd	nd	nd
Indol	nd	nd	nd	nd	nd
2-methyl naphthylene	0.008	0.008	0.005	nd	nd
1-methyl naphthylene	nd	0.0108	nd	nd	nd
Biphenyl	0.020	0.009	0.005	nd	nd
Acenaphthylene	0.023	0.013	nd	nd	0.008
Acenaphthene	0.017	0.012	0.006	nd	nd
Dibenzofuran +fluorene	0.022	0.014	0.007	0.011	0.009
Anthracene+phenthrathrene	0.024	0.008	0.012	0.012	0.006
Fluoranthene	0.013	0.013	0.023	0.009	0.018
Carbazole	0.049	0.034	0.065	0.017	0.053
Pyrene	0.059	0.031	0.021	0.020	0.026
Benzo(a)anthracene	0.012	0.030	0.007	0.024	0.015
Chrysene	0.023	0.052	0.014	0.075	0.011
Benzo(b+k)fluornanthene	0.013	0.044	0.018	0.016	0.019
Benzo(a) pyrene	0.149	0.757	0.211	0.123	0.131
Indeno (1,2,3-cd)pyrene	0.182	0.016	0.031	nd	nd
Dibenzo(a,h)anthracene	nd	0.037	nd	nd	nd
Benzo(ghi)perylene	nd	0.02	nd	0.017	nd
Total	0.620	1.125	0.430	0.328	0.300
LPAHs	0.116	0.084	0.037	0.023	0.025
HPAHs	0.490	1.027	0.369	0.295	0.256
L/H	0.237	0.082	0.101	0.07	0.097
BaA/(BaA+Chr)	0.354	0.368	0.322	0.243	0.580
InP/(InP+BghiP)	0	0	0	0	0
fl/py	0.23	0.419	1.112	0.435	0.683

Table 3. Levels of PAHs (ng.g^{-1}) in particulate fraction during winter

Name compound	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Naphthylene	nd	nd	nd	nd	nd
Indol	1.104	nd	0.197	nd	nd
2-methyl naphthylene	1.437	0.17	0.149	nd	0.141
1-methyl naphthylene	0.656	0.26	0.23	nd	nd
Biphenyl	0.608	0.13	0.119	0.141	0.183
Acenaphthylene	0.162	0.14	0.301	0.296	nd
Acenaphthene	2.051	0.39	0.32	0.314	0.525
Dibenzofuran +fluorene	1.135	0.38	0.83	0.177	0.23
Anthracene+phenthrathrene	0.192	0.42	0.468	0.614	0.169
Fluoranthene	0.412	0.49	0.606	0.219	0.409
Carbazole	1.696	0.60	1.217	1.712	0.53
Pyrene	0.225	0.32	0.800	1.462	0.378
Benzo(a)anthracene	0.305	1.15	1.949	0.284	1.03
Chrysene	0.352	1.51	1.082	0.608	0.607
Benzo(b+k)fluornanthene	0.699	1.42	4.316	1.054	0.407
Benzo(a) pyrene	0.204	15.134	2.299	4.162	1.173
Indeno (1,2,3-cd)pyrene	nd	0.397	3.064	1.759	nd
Dibenzo(a,h)anthracene	0.234	0.329	2.233	3.113	nd
Benzo(ghi)perylene	nd	1.137	0.336	0.314	nd
Total	11.480	24.42	20.535	16.23	5.796
LPAHs	7.348	1.913	2.628	1.544	1.253
HPAHs	3.719	22.023	17.30	14.47	4.134
L/H	1.975	0.08	0.151	0.106	0.303
BaA/(BaA+Chr)	0.464	0.431	0.643	0.318	0.629
InP/(InP+BghiP)	0	0.547	0.578	0.36	0
fl/py	1.834	1.506	0.75	0.150	1.080

Table 4. Levels of PAHs ($ng.g^{-1}$) in particulate fraction during summer

Name compound	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Naphthylene	nd	nd	0.103	nd	nd
Indol	nd	nd	0.158	nd	nd
2-methyl naphthylene	nd	0.153	0.214	0.147	0.138
1-methyl naphthylene	nd	nd	0.292	0.355	nd
Biphenyl	0.131	0.162	0.115	0.140	0.127
Acenaphthylene	0.123	0.226	0.103	nd	0.216
Acenaphthene	0.125	0.291	0.306	0.680	0.154
Dibenzofuran +fluorene	0.128	0.190	0.268	0.377	0.192
Anthracene+phenthrathrene	0.228	0.150	0.201	0.572	0.146
Fluoranthene	0.295	0.788	0.400	0.359	0.376
Carbazole	0.468	0.864	0.733	0.220	0.297
Pyrene	0.208	0.410	0.209	0.271	0.275
Benzo(a)anthracene	0.336	1.478	0.415	0.521	0.554
Chrysene	0.268	0.359	0.829	0.188	0.418
Benzo(b+k)fluornanthene	0.493	0.439	1.364	0.796	0.393
Benzo(a) pyrene	0.938	1.606	3.791	1.789	1.077
Indeno (1,2,3-cd)pyrene	nd	nd	0.207	nd	nd
Dibenzo(a,h)anthracene	nd	1.924	0.537	0.314	nd
Benzo(ghi)perylene	1.390	nd	0.291	nd	nd
Total	5.137	9.046	10.545	6.736	4.369
LPAHs	0.736	1.174	1.765	2.274	0.975
HPAHs	4.106	7.083	8.379	4.102	3.017
LPAHs/HPAHs	0.179	0.165	0.210	0.554	0.323
BaA/(BaA+Chr)	0.556	0.804	0.333	0.733	0.569
InP/(InP+BghiP)	0	0	0.278	0	0
fl/py	1.415	1.923	1.916	1.326	1.367

Table 5. Mean of water temperature(^{o}C) during winter and summer in all stations

Seasons	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Winter	19	18	19	19	19
Summer	30	30	30	31	31

and 0.33 to 0.80 in particulate fraction during winter and summer, respectively BaA/(BaA+Chr) ratio <0.20 indicates petroleum source, values between 0.20 and 0.35 either petroleum or combustion sources, and values >0.35 combustion source . Also InP/(InP+BghiP) ratio in particulate fraction was more than 0.20.InP/(InP+BghiP) ratio <0.20 indicates petroleum source, values between 0.20 and 0.50 either petroleum or combustion sources and values >0.50 combustion source (Guo *et al*., 2007). Fl/py ratio in dissolved fraction ranged from 0.23 to 1.204, while in particulate fraction from 0.150 to 1.923 during winter and summer respectively. The Fl/pyratio >1 indicates pyrogenic source and values <1 attributed to petrogenic source (Qiu *et al*., 2009). The results indicate that the origins of PAHs in dissolved and particulate fractions were pyrogenic from combustion sources and few of them petrogenic which come from petroleum contaminations by sewage effluents and industrial wastes which usually contained measurable levels of PAHs (Al-Khatib,2008).

Conclusion

Results of the present study revealed that the higher concentrations of PAHs were recorded in winter, where as the lower in summer. The total concentrations of PAHs were in the particulate fraction more than dissolved, also high molecular weight of PAHs were more than low molecular weight in both dissolved and particulate. According to the ratios of LMW-PAHs/HMW-PAHs, BaA/(BaA+Chr), InP/(InP+BghiP) and Fl/pythe sources of PAHs were mainly pyrogenic and few are petrogenicin these water.

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