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POLYCYCLIC AROMATIC HYDROCARBONS(PAHs) IN SEDIMENT OF Al-KAHLAA RIVER IN MISSAN PROVINCE/IRAQ

Salih, H. jazza[1] ;Abdul -Hussain,Y. Al -Adhub[2] and Hamid, T. Al - Saad[3]
[1-]University of Missan /college of science .
[2-] University of Basrah /college of science.
[3-] University of Basrah/Marine science center(htalsaad@yahoo.com)

Abstract

The present study was performed to have knowledge of Polycyclic Aromatic Hydrocarbons pollution status in sediments along Al–Kahlaa river in Missan province .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),in addition to reference station lies on the Tigris river before 25 Km from entering to Missan city**.** The concentrations of Polycyclic Aromatic Hydrocarbons(PAHs) were determined using capillary Gas –Chromatography. Results of this study revealed that the total concentrations of PAHsin sediments ranged from 4.906 ng/g dry weight in Reference station to 35.479 ng/g dry weight in Treatment unit during Winter ,and from 2.391 ng/g dry weight in Reference station to 25.886 ng/g dry weight in Treatment unit during Summer.It had been noticed that there were a predominance of high molecular weight PAHs on low molecular weight in sediment samples ,while BaA/(BaA+Chr) ratio ranged from 0.520 to 0.66 , InP/(InP+BghiP) ratio ranged between 0.681 ,while Fl/Py ratio ranged between0.10 and 8.490 in sediments ,this give an indication of the source of PAHs compounds which were mainly pyrogenic and few of them petrogenic .

Keywords: Pollutioin,PAHs, Sediment, Al-Khalaa River,Missan Province

Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) may reach aquatic environments from domestic and industrial sewage effluents, exhaust of gasoline and diesel combustion engines, surface runoff from land, deposition of airborne particulates, and especially from spillage of petroleum and petroleum products into water bodies (Hoffman *et al*., 1984; 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 evaporate, disperse into the water column, become incorporated into bottom sediments, concentrate in aquatic biota, or affected by chemical oxidation and biodegradation (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 .Therefore, sediments represent the most important reservoir of PAHs in the aquatic environment (Qiu*et al* .,2009 **;**Perra*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 spatialvariations in addition tothe sources of these compounds in the sediments of this important area.

Materials and Methods

Collection of 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),in addition to reference station lies on the Tigris river before 25 Km from entering to Missan city as shown in Figure (1). The sediment samples were taken by using Van veen grab sampler and stored in aluminum foil then placed in ice-packed and transferred to the laboratory then kept in the refrigerator before analysis.

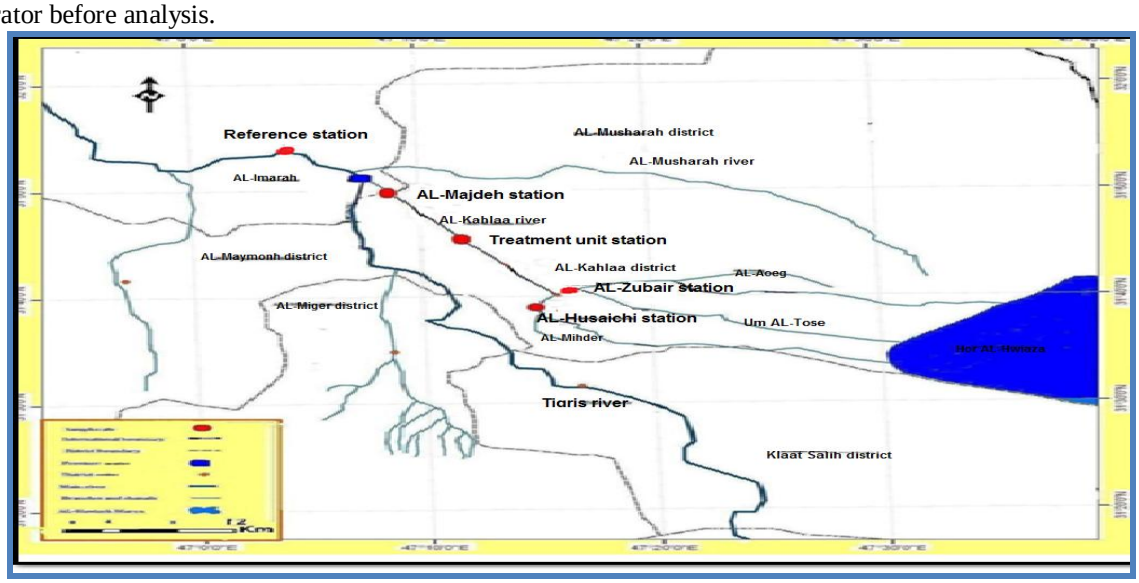

Figure (1) : Map of Al-Kahlaa River showing the position of stations.

Extraction of Hydrocarbon compounds from sediment samples.

50 gm dry sediments were placed in a pre-extracted cellulose thimble and soxhlet extracted with 150 ml methanol : benzene (1 : 1 ratio) for 24 -36 hours following a method of Goutex and Saliot(1980) .The concentrations of Polycyclic Aromatic Hydrocarbons(PAHs) were determined by using capillary Gas-Chromatography.

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) toHigh Molecular Weight (HMW-PAHs).

Values greater than one indicate petrogenic origins from crude oil and their derivatives and values less than one are attributed to 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 origins, and larger than 0.35 implies pyrogenic sources (Yunker *et al*. ,2002 ;Tolosa *et al* ., 2004; 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 petrogenic or pyrogenic origins and higher than 0.5 are implies pyrogenic origins (Yunker *et al*. ,2002 ;Tolosa *et al* ., 2004; Guo *et al*.,2007).

D-Ratio of fluoranthene to pyrene (Fl/Py) .

Values greater than 1 have been used to indicate pyrogenic origins and values less than 1 are attributed to petrogenic source (Qiu ,2009).

Results and Discussion

The total concentrations of PAHs ng/g (Tables 1 and 2) ranged from 4.667 to 22.047 , 25.886 to 35.479 , 11.591 to 16.166 , 11.216 to 11.72 and 2.391 to 4.906 in Al-Magideh , Treatment unit , Al-Husaichi , Al-Zubair and Reference stations respectively , the lowest concentrations recorded during summer , while the highest concentrations recorded during winter ,therefore results of statistical analysis (Table ,4) confirm that there are a significant differences ($p < 0.05$)between seasons. This may be attributed to the climatic condition effect photo-oxidation, volatilization and high degradation during the hot season . Temperature is generally higher in summer ,therefore increase the evaporation rate and also affect the biodegradation, which is higher than in other seasons (Boyd *et al*., 2001). Although hydrocarbons biodegradation can occur over a wide range of temperatures, the rate of biodegradation generally decreases with the decreasing temperature and vice versa ,so that the highest degradation rates generally occur in the range 20 to 30C^{o} in some freshwater environments(Bartha and Bossert ,1984). The amount of some PAH compounds in Hor Al- Howaiza , Euphrates River and coastal regions sediment in Iraq were measured by Al-Khatib (2008); Mohammed *et al*.(2009) and Al-Khion(2012) respectively and showed that the highest levels in winter, while the lowest in summer. Temperature also affects the solubility of hydrocarbons (Foght*et al* .,1996).Whereas elevated levels of them during winter may be attributed to their in precipitation which are significantly higher in winter than in summer because higher energy consumption for heating and increase hydrocarbon compound input to aquatic environment with run off during winter season. In addition to that temperature is lowest in the winter ,this will decrease evaporation rate, and causes lower rate of biodegradation(Van Noort and Wondergem , 1985; Al-Khatib ,2008). Also the higher concentrations of total PAHs were found in Treatment unit station compared with other stations ,therefore results of statistical analysis in table (4)confirm that there are some a significant differences ($p<0.05$)between stations especially between Treatment unit and Reference stations .This can be attributed to the fact that high amounts of industrial and domestic wastes that had been discharged to Al-Kahlaa river at this station (Jazza ,2009).

Benzo(ghi)perylene form the greater ratio of PAHs individual which was 4.934 in Al-Magideh station ,Benzo(a)anthracene compound 6.971 in Treatment unit station and Acenaphthene compound 3.116 in Al-Zubair station, where a Benzo(a) pyrene compound form 8.257 in Al-Husaichi station during winter (Table ,1) . While Pyrene compound form greater ratio of PAHs individual which was 0.615 in Al-Magideh station ,Benzo(a)anthracene compound 7.497 in Treatment unit station , Acenaphthene 2.762 in Al-Husaichi station and Dibenzofuran +fluorine 3.010 in Al-Zubair during summer(table,2).According to the present results, the concentration of low molecular weight (2-3 ring) polycyclic aromatic hydrocarbons (LMW-PAHs) in sediment samples of Al-Kahlaa river is lower than high molecular weight (4-6 ring) PAHs (HMW-PAHs)in all sampling stations during winter and summer except in Al-Zubair station during winter,(Tables 1 and 2) .This is due to that the lower molecular weight PAHs degraded rapidly in sediments, but higher molecular weight PAHs are more recalcitrant. These PAHs in the sediment are more resistant to degradation processes (Obayori and Salam, 2010) and may be due to differences in hydrophobicity between HMW - PAHs and LMW- PAHs . HMW -PAHs are more hydrophobic relative to LMW-PAHs and therefore will be more abundant in sediments (Luo *et al* ., 2006; Bakhtiari *et al* ., 2009) .

LMW-PAHs versus HMW-PAHs ratios ranged from 0.06 to 0.148 and from 0.043 to1.367during winter and summer respectively (Tables 1,2).These values were less than one at all station and the results indicate the predominance of pyrogenic inputs for PAHs in Al-Kahlaa river sediments at all stations during winter and summer and few of them were petrogenic specially in Al-Zubair station during winter. Higher concentrations of 1-methyl naphthylene , Acenaphthene and Dibenzofuran +fluorine in Al-Zubair station were present which indicate a Petrogenic origin supporting this conclusion(Vrana*et al*., 2001) . PAHs isomer pairs were calculated to infer the possible sources of PAHs

(Tables 1and 2) . Based on the PAH isomer pair ratio measurements compiled by Yunker*et al*. (2002) ,Tolosa*et al* .(2004)and Guo*et al*.(2007). BaA/(BaA+Chr) ratios for all stations were more than 0.35 (from 0.520 to 0.66 and from 0.492 to 0.773) during winter and summer respectively indicating that PAHs were mainly originated from pyrogenic sources. InP/(InP+BghiP) ratios for all stations were more than 0.2(from 0.33 to 0.840 and from 0.301 to 0.681) during winter and summer respectively these ratios indicate multiple PAHs sources (petrogenic and pyrogenic).Fl/py ratios for all stations ranged from 0.40 to 2.47 and 0.10 to 8.490 during winter and summer respectively, the results suggest that the PAHs in the sediments of this area had mixed sources with the predominance of pyrogenic inputs ,where pollutants come from multiple sources(Qiu ,2009) .The levels of PAHs compounds in sediment samples from Al-Kahlaa river were within the range in other regions of Iraq except in Shatt Al-Hilla (Al-Taee*et al* ., 2010) ,whereas these levels in other areas in the world were extremely low compared with the levels in this study (table ,3) , therefore the sediments analysis of this river considered as unpolluted (Al-Saad*et al* ., 1998 ; Al-Khatib ,2008 ; Al-Khion ,2012;Al-Taie ,2013) .

Table(1) :Concentrations of PAHs (ng/g) in sediments during winter.

Compounds	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Naphthylene	0.138	0.177	0.060	0.143	0.05
Indol	0.091	0.118	0.194	0	0
2-methyl naphthylene	0.148	0.404	0.125	0.114	0.079
1-methyl naphthylene	0.075	0.071	0.057	1.114	0
Biphenyl	0.114	0.123	0.063	0.077	0.083
Acenaphthylene	0.121	0.113	0.166	0.108	0.115
Acenaphthene	0.163	0.160	0.175	3.116	0.057
Dibenzofuran +fluorene	0.303	0.342	0.088	0.171	0.100
Anthracene+phenthrathrene	0.217	0.203	0.057	0.069	0.106
Fluoranthene	0.465	5.352	0.406	0.887	0.316
Carbazole	2.071	6.345	2.156	0.235	0.433
Pyrene	1.120	3.478	1.007	0.848	0.127
Benzo(a)anthracene	3.681	6.971	0.741	0.157	0.246
Chrysene	2.487	3.532	0.570	0.144	0.221
Benzo(b+k)fluornanthene	3.330	1.327	1.058	0.467	0.097
Benzo(a) pyrene	1.747	2.305	8.257	0.670	2.290
Indeno (1,2,3-cd)pyrene	0.702	1.386	0.266	0.316	0.163
Dibenzo(a,h)anthracene	0.132	0.261	0.378	0.637	0.155
Benzo(ghi)perylene	4.934	2.803	0.332	2.442	0.259
Total	22.047	35.479	16.166	11.72	4.906
LPAHs	1.374	1.7155	0.989	4.916	0.595
HPAHs	20.20	28.411	14.77	5.920	3.995
L/H	0.06	0.060	0.06	0.830	0.148
BaA/(BaA+Chr)	0.59	0.66	0.56	0.520	0.526
InP/(InP+BghiP)	0.841	0.840	0.41	0.33	0.51
fl/py	0.41	1.53	0.40	1.04	2.47

Table(2): Concentrations of PAHs (ng/g) in sediments during Summer.

Compounds	Stations				
	Al-Magideh	Treatment unit	Al-Husaichi	Al-Zubair	Reference
Naphthylene	Nd	0.053	0.242	0.052	nd
Indol	Nd	0.11	0.084	0.239	nd
2-methyl naphthylene	0.057	0.102	0.196	0.079	0.063
1-methyl naphthylene	0.306	0	0.849	0.103	nd
Biphenyl	0.099	0.111	0.054	0.131	0.056
Acenaphthylene	0.189	0.089	0.283	0.305	0.092
Acenaphthene	0.217	0.166	2.762	2.243	0.217
Dibenzofuran +fluorene	0.191	0.232	0.078	3.010	0.109
Anthracene+phenthrathrene	0.155	0.148	0.215	0.219	0.107
Fluoranthene	0.065	1.466	0.761	0.162	0.246
Carbazole	0.244	2.933	0.520	0.715	0.071
Pyrene	0.615	2.982	0.089	0.361	0.088
Benzo(a)anthracene	0.403	7.497	1.228	0.264	0.380
Chrysene	0.136	3.129	0.791	0.272	0.111
Benzo(b+k)fluornanthene	0.291	3.931	0.958	0.159	0.267
Benzo(a) pyrene	0.978	2.003	0.248	0.120	0.577
Indeno (1,2,3-cd)pyrene	0.164	0.422	0.603	0.388	nd
Dibenzo(a,h)anthracene	0.186	0.197	1.338	0.900	nd
Benzo(ghi)perylene	0.363	0.308	0.283	1.484	nd
Total	4.667	25.886	11.591	11.216	2.391
LPAHs	1.21	1.01	4.767	6.385	0.647
HPAHs	3.38	23.40	6.061	4.668	1.497
LPAHs/HPAHs	0.35	0.043	0.786	1.367	0.432
BaA/(BaA+Chr)	0.74	0.705	0.608	0.492	0.773
InP/(InP+BghiP)	0.46	0.681	0.310	0.301	0
fl/py	0.10	0.491	8.490	0.449	2.794

***nd =not detection**

Table (3): Comparison of concentrations of PAHs Compounds in sediments samples from Al-Kahlaa River with their concentrations in other areas of Iraq and the world.

References	Concentrations (ng/g)	Locations
Al-Saad , 1995	42.8 – 56.70	Shatt Al-Arab and NW Arabian Gulf
Al-Saad, 1987	0.2 – 76.25	Shatt Al-Arab and NW Arabian Gulf
Al-saad and Al-Timari, 1989	0.59 – 2.07	Hor Al-Hammar
Al-Hamdi, 1989	6.88-39.85	Khor Al-Zubair and N.W.Arabian Gulf
Al-Khatib ,2008	0.1- 145.8	Hor Al – Howaiza
Al-Taee*et al* ., 2010	26.668-900.042	Shatt Al-Hilla
Al-Khion ,2012	12.15-47.38	Iraqi coast regions
Al-Taie ,2013	0.252-10.363	Al-Azim Marshes
Hatzianestis*et al* .,1998	31-176.3	Aegean sea
Zakaria*et al*., 2002	4 -924	rivers in Malaysia
Zhou *et al*., 2000	247 - 480	Xiamen Harbor/ China
Doong and Lin,2004	8.0 - 356	Geo – ping river/china
Bakhtiari*et al* .,2009	306-7968	Lagant river /Malaysia
Kafilzadeh*et al* .,2011	167.4-530.3	The kor river /Iran
Present study	2.391-35.479	Al-Kahlaa river / Missan province

Table (4): A significant variations among stations and seasons.

Stations	Reference	Al-Zubair	Al-Husaichi	Treatment unit	Al-Magideh
	3.6485 c	11.468 Bc	13.8785 ab	30.6825 a	13.357 bc
Seasons	**Winter**		**Summer**		
	18.0636a		**11.1502b**		

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