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Research Article

Origins and distribution of polynuclear aromatic hydrocarbons (PAHs) in water and sediments of some rivers in Misan Province, Iraq

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Abstract: Polynuclear aromatic hydrocarbons (PAHs) are amongst the pollutants of main concern in the aquatic ecosystems. They are mostly characterized by mutagenic and carcinogenic effects. Origins and distribution of the 16 PAHs was studied in water and sediments samples which collected from Al-Musharah, Al-Musharah, Al-Batiruh and Al-Kahlaa rivers and three sites along Tigris River namely Ali Al-grbi, Maemal alzuyut and Qulat Salih, in Misan Province. The results revealed the sum concentrations of the 16 PAHs that in water ranging from 1667.15 to 5552.7ng/l in Al-Batiruh and Al-Mijer rivers, respectively. Low Molecular Weight PAHs (LPAHs) levels in the water column were lower than High Molecular Weight PAHs (HPAHs) in all stations except Al-Batiruh River. Also according to our findings, Σ16 PAHs in the sediments was varied from 10365.73 to 28876.2µg/gm.dry weight in Ali Al-grbi station and Al-Kahlaa River, respectively. The concentration of LPAHs in sediment samples was lower than HPAHs at all rivers. The mutagenic equivalent quotient *MEQ* and toxic equivalent quotient *TEQ* was generally low except Benzo[k]fluoranthene, Benzo[a]pyrene, Dibenzo[a,h]anthracene, Benzo[g,h,i]perylene and Indeno[1,2,3-c,d] pyrene that were found extremely higher in the sediments of the studied rivers indicating a possible cancer risk to those how perhaps exposed to the river sediments. According to diagnostic ratios of Inpy/(Inpy+BghiP) were more than 0.2 and LPAHs/HPAHs ratios were less than one. These results suggested that the origins of PAHs compounds were derived from pyrolytic origins in water and sediments of the studied rivers.

Keywords: PAHs, River, Tigris, Pollution.

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Introduction

Polynuclear aromatic hydrocarbons (PAHs) are unique types of organic harmful pollutants that are environmentally persistent, bio-accumulated in tissues of biota species having toxic impacts on living organisms. They are composed of two or more fused benzene rings which widely distributed in water, air and soil (Armstrong et al. 2004; Abdel-Shafy & Mansour 2016). Their physical and chemical properties are varying with both the pattern of ring linkage and the number of rings (Bayoumi 2009). PAHs are recognized as carcinogenic, mutagenic, endocrine disruption, oxidative stress and can cause potent immunological suppressants (CCME 2004; Onozata et al. 20016). The toxicity of PAHs to aquatic life is affected by the presence of ultraviolet light and metabolism which adverse effects on living organisms. Although un-metabolized PAHs can pose toxic effects, a main concern is the ability of the reactive metabolites like dihydrodiols and epoxides of some PAHs individuals to bind to DNA and cellular proteins that can cause cell damage occurrence lead to mutations, malformations, developmental, cancer and tumors (Honda & Suzuki 2020).

PAHs are lipophilic compounds readily available

absorbed from the digestive system then rapidly distributed in various lipid tissues like liver, muscles and gonads (Jazza 2015). There are two types from the major PAHs sources in the environment, including natural and anthropogenic, natural sources of PAHs are less dominant than anthropogenic source (Jazza et al. 2016a). Natural sources of PAHs are formed whenever organic materials are exposed to high temperatures under low oxygen or anaerobic conditions through pyrolysis process, whereas anthropogenic sources include residential heating, biomass burning, combustion engines and industrial activities (Abdel-Shafy & Mansour 2016; Hussain et al. 2018). Seventeen PAHs have been identified being of greatest concern with regard to potential exposure and adverse health effects on human; the health impacts of individual PAHs are not exactly analogical. The international agency for research on cancer IARC (2010) classified some individual PAHs into known, possibly or probably carcinogenic to humans such as are benzo[a]pyrene naphthalene, chrysene, benz[a]anthracene, benzo[k] fluoranthene and benzo[b]fluoranthene. Some PAHs are well-known as mutagens, carcinogens and teratogens and thus pose a serious threat to the human health (WHO 2003). The purpose of the present study is to determine the origins of the individual PAHs on the basis of their concentrations and to analyze their distributions in water and sediments of some rivers in Misan Province.

Material and Methods

Samples collection, preparation and extraction: Water samples were collected from Al-Musharah, Al-Musharah, Al-Batiruh and Al-Kahlaa rivers and also from three sites along the Tigris River (Ali Al-grbi, Maemal alzuyut and Qulat Salih) in Misan Province. Samples were taken from the surface and 50 to 100cm below the surface using cleaned glass bottles. PAHs in water sample were extracted according to (UNEP 1989). Five liters of water sample was mixed with 25ml CCl_4 for 30min using water mixer, the liquid fraction was drained, while the residual was transferred into separator funnel. The organic layer was carefully poured into a glass column containing 5g of anhydrous sodium sulfate Na_2SO_4, then collected and dried. The residual was dissolved with 50ml n-hexane and passed through a glass column filled with glass wool at the bottom, 10g deactivated silica gel 100-200 mesh, 10g deactivated alumina 100-200 mesh and 5g anhydrous sodium sulfate at the top. The aliphatic fractions were eluted from the column with n-hexane, whereas the aromatic fractions were eluted with benzene.

The sediment samples were collected from each station using a van veen grab sampler. Simultaneously sediment samples were taken at a depth of 5 to 10cm of sediment surface from the idem sites and placed in an ice box for transferring to the laboratory. The sediment samples were dried in an oven at 50°C for about three days then grinded and sieved through a 63μm mesh sieve and stored in glasses vials until analysis. 20g of sieved sediments were placed in cellulose thimble and soxhlet extracted according to Goutx & Saliot (1980) using soxhlet intermittent extraction with mixed solvents (methanol: benzene 1:1) for 36-48 hours at temperature does not exceed 40°C. Then the combined extracts were saponification for 2 hours through adding 20ml (4M) from methanol and KOH at the same temperature then left to be cooled at room temperature. The un-saponification layer was extracted with 50ml from n-hexan using separator funnel. The upper layer was passed through chromatographic column filled with glass wool at the bottom, 10g deactivated silica gel 100-200 mesh, 10g deactivated alumina 100-200 mesh and 5g anhydrous sodium sulfate at the top. The aliphatic fractions were eluted from the column with n-hexane, whereas the aromatic fractions were eluted with benzene. Aromatic fractions were dried and stored until detection with HPLC apparatus type Shimadzo in Basrah University (Marine Science Center).

Mutagenic and carcinogenic effects of PAHs in sediments: Mutagenic and carcinogenic toxicities of the HMWPAHs were detected in the sediments and

Table 1. Concentrations of PAHs (ng/l) in water samples.

Compound name	stations						
	Al-Musharah	Al-Mijer	Al-Batiruh	Al-Kahlaa	Tigris River		
					Ali Al-	Maemal	Qulat
Naphtalene	218.75	452.90	ND	483.82	23.90	81.94	ND
Acenaphtylene	352.64	ND	218.56	ND	92.64	ND	45.93
Acenaphthene	33.96	ND	69.81	581.43	ND	48.95	ND
Fluorene	11.75	73.81	27.45	13.10	26.44	ND	ND
Phenanthrene	353.44	39.94	ND	58.96	ND	436.89	54.93
Anthracene	978.48	ND	546.78	ND	786.34	ND	ND
Fluoranthene	ND	40.59	ND	27.21	101.68	45.61	ND
Pyrene	316.52	ND	59.94	49.80	ND	ND	453.84
Benzo[a]anthracene	ND	453.72	ND	ND	517.72	ND	ND
Chrysene	ND	ND	ND	81.50	ND	93.45	ND
Benzo[b]fluoranthene	531.78	602.19	73.90	ND	675.82	ND	52.84
Benzo[k]fluoranthene	79.31	ND	ND	88.49	ND	847.82	ND
Benzo[a]pyrene	291.49	ND	275.50	ND	639.71	ND	739.41
Dibenzo[a,h]anthracene	ND	129.71	68.55	772.83	222.49	183.72	ND
Benzo[g,h,i]perylene	492.36	607.28	25.89	274.61	649.11	89.27	17.93
Indeno[1,2,3-	523.16	3152.56	300.77	486.93	204.70	410.28	1875.67
∑16 PAHs	4183.64	5552.7	1667.15	2918.68	3940.55	2237.93	3240.55
LPAHs∑	1949.02	566.65	862.6	1137.31	929.32	567.78	100.86
HPAHs ∑	2234.62	4986.05	804.55	1781.37	3011.23	1670.15	3139.69

*ND = not detected

assessed relative to benzo[a]pyrene using Mutagenic Equivalent Quotient (MEQ) (referred as the ability of each congener to cause modification Deoxyribonucleic acid DNA of human) and Toxic Equivalent Quotient (TEQ) in equations of 1 and 2, respectively: (1) $MEQ=\sum C_n \cdot MEF_n$ and (2) $TEQ=\sum C_n \cdot TEF_n$, where C_n is concentration of each PAHs individual (n) in the mixture, MEF_n = mutagenic equivalent factor of each PAHs individual (n), TEF_n = toxic equivalence factor of each PAHs individual (n).

Results and Discussion

PAHs in water samples: The concentrations of PAHs in the water column are given in Table 1. ∑16 PAHs in the water samples was ranged from 1667.15 to 5552.7ng/l in Al-Batiruh and Al-Mijer rivers, respectively, the total concentrations of PAHs in water column were varied among the studied stations due to the difference in the discharged water sources because of the proximity to human activities (El-Nemr et al. 2013; Jazza et al. 2016b). Low Molecular Weight PAHs (LPAHs) was ranged between 100.86 and1949.02ng/l in Qulat Salih and Al-Musharah rivers, respectively, while High Molecular Weight PAHs (HPAHs) ranged from 804.55 to 4986.05ng/l in Al-Batiruh and Al-Mijer rivers, respectively. LPAHs levels in the water column were lower than HPAHs in all stations except Al-Batiruh River, this due to the higher volatility, solubility and biodegradability of LPAHs compared with HPAHs (Huanling et al. 2019). In addition, the temperature has a role in the evaporation processes of PAHs in the aquatic environment which can also eliminate LPAHs in the water surface (Jazza 2015; Jazza et al. 2016a). Alam et al. (2013) proved that the four -and six-ring PAHs found in the particle phase, whereas 2-3 ring PAHs occur in the vapor phase at ambient temperature (Alam et al. 2013). Also Sofuoglu et al. (2001) reported that temperature variation has more impact on vapor phase LPAHs dispersion than particle phase HPAHs. In addition, Photo-oxidation process plays an important role in degradation pathway of these compounds in water and can chemically transform them into other products may be pose more carcinogenic impacts and more toxic than the parent PAHs compounds (NRCC 1983). The presence of these compounds in the water indicates

Table 2. Concentrations of PAHs (ng/gm) in sediments samples.

Compound name	stations						
					Tigris River		
	Al-Musharah River	Al-Mijer River	Al- Batiruh River	Al-Kahlaa River	Ali Al-grbi	Maemal alzuyut	Qulat Salih
Naphtalene	ND	453.09	ND	ND	600.90	444.07	781.03
Acenaphtylene	376.69	ND	67.63	ND	802.50	ND	ND
Acenaphthene	ND	87.96	ND	721.06	ND	ND	377.17
Fluorene	449.03	ND	330.92	ND	226.91	ND	ND
Phenanthrene	480.40	488.92	498.91	ND	ND	501.61	ND
Anthracene	ND	710.53	ND	49.01	ND	105.83	ND
Fluoranthene	281.51	ND	ND	ND	169.39	ND	241.09
Pyrene	ND	502.29	550.72	ND	ND	47.32	ND
Benzo[a]anthracene	830.41	ND	ND	ND	ND	ND	ND
Chrysene	557.05	628.43	ND	7936.03	ND	64.71	5220.27
Benzo[b]fluoranthene	ND	ND	602.93	ND	77.81	ND	ND
Benzo[k]fluoranthene	8943.22	6398.37	ND	591.95	ND	4419.91	ND
Benzo[a]pyrene	586.60	3866.01	ND	7551.61	385.61	ND	151.89
Dibenzo[a,h]anthracene	729.83	ND	730.64	2838.57	1205.86	6601.49	5013.24
Benzo[g,h,i]perylene	401.16	135.78	3449.918	2494.51	798.38	228.97	2879.42
Indeno[1,2,3-	577.55	2103.17	6253.89	6693.46	6098.37	7643.564	9076.06
∑16 PAHs	14213.45	15374.55	12485.558	28876.2	10365.73	20057.47	23740.175
LPAHs∑	1306.12	1740.5	897.46	770.07	1630.31	1051.51	1158.2
HPAHs ∑	12907.33	13634.05	11588.098	28106.13	2637.05	11362.4	22581.97

Table 3. Mutagenic Equivalent Quotient (MEQ) of PAHs in sediments samples.

Compound name	Stations						
					Tigris River		
	Al-Musharah River	Al-Mijer River	Al-Batiruh River	Al-Kahlaa River	Ali Al-grbi	Maemal alzuyut	Qulat Salih
Benzo[a]anthracene	68.09362	ND	ND	ND	ND	ND	ND
Chrysene	9.46985	10.68331	ND	134.9125	ND	1.10007	88.74459
Benzo[b]fluoranthene	ND	ND	150.7325	ND	19.4525	ND	ND
Benzo[k]fluoranthene	983.7542	703.8207	ND	65.1145	ND	486.1901	ND
Benzo[a]pyrene	586.6	3866.01	ND	7551.61	385.61	ND	151.89
Dibenzo[a,h]anthracene	211.6507	ND	211.8856	823.1853	349.6994	1914.432	1453.84
Benzo[g,h,i]perylene	76.2204	25.7982	655.4844	473.9569	151.6922	43.5043	547.0898
Indeno[1,2,3-c,d]pyrene	179.0405	651.9827	1938.706	2074.973	1890.495	2369.505	2813.579

chronic or recent pollution in the studied rivers.

PAHs in sediments: The levels of PAHs composition in the sediments are summarized in Table 2. The sum concentrations of the 16 PAHs (Σ16 PAHs) varied from 10365.73 to 28876.2µg/gm.dry weight in Ali Al-grbi station and Al-Kahlaa River, respectively. The results showed that LPAHs was varied from 770.07 to 1740.5ng/gm.dry weight in Al-Kahlaa and Al-Mijer rivers, respectively, whereas HPAHs ranged from 2637.05 to 28106.13 in the Maemal alzuyut station in Tigris River and Al-Kahlaa River, respectively. According to the results, the concentration of LPAHs (2-3 ring) in sediment samples was lower than HPAHs (4-6 ring) at all stations (Table 2), this may be attributed to the climatic conditions and LPAHs may have undergone microbial degradation compared to HPAHs (Mohammed et al. 2009; Singare 2015). PAHs are subjected to microbial degradation by bacteria and fungi. Biodegradation is one of the most processes responsible for removing PAHs compounds which depending on the number of

Table 4. Toxic Equivalent Quotient (TEQ) of PAHs in sediments samples.

Compound name	Stations						
	Al-Musharah River	Al-Mijer River	Al- Batiruh River	Al-Kahlaa River	Tigris River		
					Ali Al-grbi	Maemal Alzuyut	Qulat Salih
Benzo[a]anthracene	83.041	ND	ND	ND	ND	ND	ND
Chrysene	5.57	6.284	ND	79.360	ND	0.647	52.20
Benzo[b]fluoranthene	ND	ND	60.293	ND	7.781	ND	ND
Benzo[k]fluoranthene	894.322	639.837	ND	59.195	ND	441.991	ND
Benzo[a]pyrene	586.60	3866.01	ND	7551.61	385.61	ND	151.89
Dibenzo[a,h]anthracene	729.83	ND	730.64	2838.57	1205.86	6601.49	5013.24
Benzo[g,h,i]perylene	4.011	1.357	34.499	24.945	7.983	2.289	28.794
Indeno[1,2,3-c,d]pyrene	57.75	210.317	625.389	669.346	609.837	764.356	907.606

Table 5. Toxic Equivalent Quotient (TEQ) of PAHs in sediments samples.

PAHs	Petrogenic origins	Pyrolytic origins	Stations						
			Al-Musharah River	Al-Mijer River	Al-Batiruh River	Al-Kahlaa River	Tigris river		
							Ali Al-grbi	Maemal alzuyut	Qulat Salih
Inpy/(Inpy+BghiP)	<0.2	>0.2	0.51	0.83	1.08	0.63	4.17	1.21	1.0
LMW/HMW	>1	<1	0.87	0.11	1.07	0.63	0.30	0.33	0.03

Table 6. Origins of PAHs in sediments samples.

PAHs	Petrogenic Origins	Pyrolytic origins	Stations						
			Al-Musharah River	Al-Mijer River	Al-Batiruh River	Al-Kahlaa River	Tigris River		
							Ali Al-grbi	Maemal alzuyut	Qulat Salih
Inpy/(Inpy+BghiP)	<0.2	>0.2	0.5	0.9	0.6	0.7	0.8	0.9	0.7
LMW/HMW	>1	<1	0.1	0.12	0.07	0.02	0. 6	0.09	0.05

aromatic rings, type of ring fusion and molecular weight of PAHs (Wild et al. 1991). LPAHs compounds are more degradable and soluble, but HPAHs compounds are more recalcitrant therefore more resistant to biodegradation (Obayori & Salam 2010). The higher levels of PAHs in the sediment compared to the surface water attributed to oxidation and volatilization processes can rabidly remove many PAHs in the water column. In addition to most of PAHs are associated with the suspended materials and this due to their hydrophobic characteristics giving rise to the increased accumulation them in sediments. Thus sediments represent important reservoir of PAHs in the aquatic ecosystem (Perra et al. 2009). These results suggest that the sediments could be cause adverse impacts to the aquatic organisms within these areas, so that PAHs will pose a risk of bioaccumulation in dwelling organisms which increases the of a toxic impact to humans (Singare 2015). The adverse impacts of PAHs on the aquatic ecosystem can be used as an early warning indicator of ecosystem health and potential effects on human health (Jazza 2015). Therefore, further studies are needed to observe the bioaccumulation of PAHs especially the HPAHs in the sediment and their effect on benthic living organisms.

Probable HMWPAHs in sediments: The MEF values for B[a]A, Chry, B[b]F, B[k]F, B[a]P, D[ah]A, B[ghi]P and Inpy stations were 0.082, 0.017, 0.25, 0.11, 1.0, 0.29 and 0.31, respectively, and their TEQ were 0.1, 0.01, 0.1, 0.1, 1.0, 10.0, 1.0 and 0.1, respectively (Benson et al. 2017; Zhao et al. 2017).

The results were revealed that MEQ and TEQ were low except B[k]F, B[a]P, D[ah]A, B[ghi]P and Inpy that were found extremely higher in the sediments of the studied rivers (Tables 3 and 4), indicating a possible cancer risk to those how perhaps exposed to the river sediments (Salem et al. 2014; Benson et al. 2017). In addition, there are another problems including lung disease, low intelligent quotient and birth defect (Hsu et al. 2014; Hussein et al. 2016).Origins of PAHs in water and sediments samples**:** In the present study, attributed to the limited data to determine diagnostic ratios, there are only one ratio of PAHs that available to use for origins identification Indeno[1,2,3-c,d]pyrene/(eno[1,2,3-c,d]pyrene+Benzo[g,h,i]perylene). Inpy/(Inpy+ BghiP), in addition to ratio between LMW-PAHs with HMW-PAHs (∑LPAHs / HPAHs) (Moyo et al. 2013; Adeniji et al. 2018, 2019; Ihunwo et al. 2019).

Diagnostic ratios of Inpy/(Inpy+BghiP) in water samples from all stations were more than 0.2 whereas ∑LMW/HMW ratios were less than 1at all stations except in Al-Batiruh River (Table 6). These results confirmed that the origins of PAHs in the water samples were derived from pyrolytic sources at all stations except in Al-Batiruh River were mixed sources (Jazza 2015; Adeniji et al. 2019; Ihunwo et al. 2019). Diagnostic ratios of Inpy/(Inpy+BghiP) in the sediments samples from all stations were more than 0.2 and ∑LPAHs/HPAHs ratios were less than 1at all stations (Table 6). These results suggested that the origins of PAHs compounds were derived from pyrolytic origins in the sediments of the studied rivers, this may be due to implying contributions from the burning of nonpetroleum substances such as biomass or waste as major pyrolytic origin (Jazza et al. 2016b; Zhao et al. 2017; Adeniji et al. 2019).

Conclusions

The higher levels of PAHs in the sediment compared to the water column attributed to weathering processes such as volatilization, photo oxidation and biodegradation etc. In water and sediments samples, there were mostly dominance of HPAHs on LPAHs at all rivers except Al-Batiruh River in water samples, this attributed to the higher volatility, solubility and biodegradability of LPAHs compared with HPAHs. Results of this study was revealed that the *MEQ* and *TEQ* was generally low except BkF, BaP, DahA, BghiP and Inpy that were found extremely higher in the sediments indicates a possible cancer risk to those how perhaps exposed to the river sediments. According to diagnostic ratios of Inpy/(Inpy+BghiP) and LPAHs/HPAHs, these findings suggested that the origins of PAHs compounds were derived from pyrolytic origins in water and sediments samples of studied rivers.

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