



Original Research Article

## Estimation of Hydrocarbon Compounds in Drinking Water in Misan Governorate / Iraq

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Received: 20 July 2016

Revised: 03 August 2016

Accepted: 05 August 2016

### ABSTRACT

Levels, distribution and sources of hydrocarbon compounds were investigated in the drinking water of Misan governorate collected from fifteen selected stations during October to December, 2015. Results of this study revealed that the concentrations of Total Petroleum Hydrocarbons (TPHs) ranged between 0.838 to 11.23  $\mu\text{g.l}^{-1}$  in stations Al-Mailmen Quarter and Al-Hussein Quarter respectively, whereas total concentrations of Polycyclic Aromatic Hydrocarbons (PAHs) ranged between 8.460 to 3654.8  $\text{ng.l}^{-1}$  in stations Al-Meijer district and Awasha Quarter respectively. High Molecular Weight (HMW) of PAHs were more than Low Molecular Weight LMW-PAHs in all stations except in Al-Kahlaa district, Al-Fatimya Quarter, Kumat district, Awasha Quarter and Al-Meijer district. According to the ratios of LMW-PAHs/HMW-PAHs, Phenanthrene / Anthracene, Benzo[a]Anthracene / (Benzo [a] Anthracene + Chrysene) and fluoranthene / pyrene, they certainly reflected that the PAHs sources were pyrogenic as a main and few petrogenic in most stations. In all sampling stations, the concentrations of Benzo [a]Pyrene BaP in the drinking water samples was lower than the permissible limits of world health organization (WHO), Iranian national drinking water standards (ISIRI), US Environmental Protection Agency (USEPA) and European Union (EU) except in some stations which was above the permissible limits of European Union's drinking water standard. Sum of carcinogen PAHs Chrysene, Benzo [a]pyrene, Benzo [b] fluoranthene, Benzo[k]fluoranthene and Dibenzo [a,h]anthracene ranged between 1.47 and 3652.35  $\text{ng.l}^{-1}$  were above the permissible limit of USEPA in all stations except in Al-Kahlaa district, Kumet district, Al-Silam district and Al-Mailmen Quarter, while sum of carcinogen PAHs Benzo [b] fluoranthene, Benzo[k]fluoranthene, Benzo[g,h,i]perylene and Indeno [1,2,3-cd] pyrene were within the permissible limit of EU in all stations except in stations Al-Fatimya Q, Al-Hussein Q and Ali Al-Garbi district.

**Keyword:** Hydrocarbons; drinking water, Misan Governorate

## INTRODUCTION

Petroleum 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 these compounds have been a cause for concern and major challenges to save the environment against their adverse impacts all over the world and these can affect the water resource quality and threaten human health [1,2]. Polycyclic Aromatic Hydrocarbons (PAHs) are a class of diverse organic compounds containing two or more fused aromatic rings of carbon and hydrogen atoms. They are ubiquitous pollutants that can reach water bodies mainly through deposition of airborne particulates, road runoff, petroleum spills, industrial wastewater and fossil fuel combustion [3,4,5,6]. These compounds are always found as a mixture of individual compounds owing to their low solubility and high affinity for particulate matter. Their presence in drinking water is an indication of a source of pollution. PAHs are only slowly biodegradable under aerobic conditions and are stable to hydrolysis [7]. The main source of PAHs contamination in drinking water is usually not the raw water sources but the coating of the drinking water distribution pipes by using some coating material for water pipes to give effective protection against corrosion and after the passage of drinking water through those pipes or after repair work, significantly increased PAHs levels have been detected in the water [8,9]. PAHs compounds are teratogenic, mutagenic and carcinogenic such as lung, bladder, as well as skin cancer [10]. The main objective of the present study was to investigate the distribution of total petroleum hydrocarbons (TPHs) and sixteen PAHs in drinking water in Misan governorate as well as compare it with international standards for permissible levels of individual

polycyclic aromatic hydrocarbons for drinking purposes, especially carcinogenic PAHs.

## MATERIALS AND METHODS

Water samples were collected by using dark glass bottles (about 5 liter) from fifteen selected stations in Misan governorate. Hydrocarbons in water sample were extracted according to [11]. Analysis of Total Petroleum Hydrocarbons was determined by using Spectrofluorometer whereas Polycyclic aromatic hydrocarbons (PAHs) were determined by using HPLC instrument type Shimadzu at Department of Marine Environmental Chemistry, Marine Science Center, University of Basrah,

### Hydrocarbons pollution indices:

Many parameters have to be analyzed in order to evaluate the probable origin of PAHs in drinking water samples. These are as follows:

#### **a-Phenanthrene / Anthracene Ratio (Phe /Ant)**

The ratio values less than ten (10) show that the hydrocarbons are pyrogenic in origin. When the ratio values are higher than ten, the hydrocarbons are petrogenic in origin [12].

#### **b-Ratio of Low Molecular Weight to High Molecular Weight (LMW-PAHs/HMW-PAHs).**

Values more than one indicate petrogenic sources from crude oil and their derivatives and values less than one are pyrogenic sources [13].

#### **c-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 [14].

**d-Ratio of Indeno (1,2,3-cd)pyrene /(Indeno (1,2,3-cd)Pyrene + Benzo (ghi)Perylene). IcdP/(IcdP + 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 [14].

**e-Ratio of fluoranthene to pyrene (Flu/Pyr).**

Values more than one have been used to indicate pyrogenic sources and values less than one are attributed to petrogenic sources [15].

**RESULTS AND DISCUSSION**

Results of the present study indicated that the concentration of total petroleum hydrocarbons TPHs ( $\mu\text{g} \cdot \text{l}^{-1}$ ) in the drinking water samples of Misan province were 0.838, 1.123, 1.345, 1.736, 1.795, 2.103, 2.171, 2.602, 3.096, 3.796, 4.564, 4.898, 5.399, 10.348 and 11.23 in stations Al-Fatimya Quarter, Al-Azer district, Kumat district, Al-kadisea Quarter, Al-Meijer district, Ali Al-Garbi district, Al-Majidea Quarter, Al-Maymona district, Awasha Quarter, Al-Musharah district, Islamic Unit Quarter, Al-Kahlaa district, Al-Mailmen Quarter, Al-Silam district and Al-Husseini Quarter respectively (Figure 1). The main reason for TPHs presence in these stations may be from the pollution of water sources as well as leaching from pipelines [16,17]. In addition to that algae growth in water tanks in houses, which synthesized some of hydrocarbons compounds and released them to the water. [18] found that many species of biota have the ability to synthesize some of hydrocarbon compounds in their bodies and then release them to the water. The concentration of total PAHs in the drinking water shows variations in different stations, which ranged between 8.46 to 3654.8  $\text{ng} \cdot \text{l}^{-1}$ . The minimum PAHs concentrations was observed in Al-Meijer district, whereas the maximum PAHs concentrations in Awasha Quarter (Tables 1,2,3). Also PAHs contamination in drinking water may be attributed to the coating of the drinking water distribution pipes with mineral

materials which used to give effective protection against corrosion [16,17]. Water disinfection by chlorine and anaerobic condition remobilized PAHs from these materials [8,9]. Also during this study concentrations of high molecular weight-PAHs were more than concentrations of low molecular weight-PAHs in drinking water samples in all stations except in Al-Kahlaa district concentrations of LMW-PAHs were more than concentrations of HMW-PAHs (Table 1,2,3), whereas in stations Al-Fatimya Quarter, Awasha Quarter, Al-Meijer district and, Kumat district concentrations of LMW-PAHs were not detected (Tables 1,3). To determine the origins of PAHs in drinking water samples in Misan province according to the following ratios. The ratio of LMW-PAHs/HMW-PAHs 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 most stations except in Al-Kahlaa district the ratio of LMW-PAHs/HMW-PAHs were 12.86 (Tables 1,2,3), while in stations Al-Fatimya Quarter, Awasha Quarter, Al-Meijer district and, Kumat district this ratio were equal 0 (Tables 1,3). The results indicate that the origin of these compounds in drinking water samples were pyrogenic except in Al-Kahlaa district was petrogenic and this may be attributed to present some oil companies. [19] found that 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-PAHs. Phe/Ant ratio was recorded this only value (0.038) in Al-Majidea Quarter (Table 2). The results indicate that the sources of these compounds in drinking water of this station were pyrogenic [12]. Ratio of BaA/ (BaA+Chy) were 0.79, 0.915, 0.403 and 0.88 in stations Al-Fatimya Quarter, Al-kadisea Quarter, Al-Majidea Quarter and Al-Mailmen Quarter respectively. These ratios > 0.35 indicates combustion source (pyrogenic), whereas were

0.09 and 0.193 in stations Al-Silam district and Al-Hussein Quarter respectively (Tables 1,2,3). This ratio  $< 0.20$  indicates petroleum source (petrogenic) [13,14]. Flu/Pyr ratios were 1.08 and 1.62 in stations Al-Maymona district and Al-Mailmen Quarter respectively, while ratio of Flu/Pyris 0.59 in Islamic Unit Quarter. The ratio of Flu/Pyr  $> 1$  indicates pyrogenic source and values  $< 1$  attributed to petrogenic source which coming from petroleum contaminations by sewage effluents and industrial wastes which usually contained measurable levels of PAHs [15,20]. Also results of the present study when compared with international standards (Table 4) revealed that the concentration of BaP in the drinking water samples was lower than the permissible limits of world health organization (WHO) [16], Iranian National Drinking Water Standards [21] and [22], whereas the concentrations of BaP in all stations were within European Union's drinking water standard [23], which forces the concentration of BaP should not exceed  $10 \text{ ng} \cdot \text{l}^{-1}$  (Table 4) except stations

Al-Silam district, Al-Azer district (Table,1), Al-Hussein Quarter (Table ,2) and Kumat district (Table ,3). Sum of the carcinogenic PAHs concentration BaP, BbF, BkF, Chy and DahA ranged from 1.47 to  $3652.35 \text{ ng} \cdot \text{l}^{-1}$  in Al-Meijer district and Awasha Quarter respectively. The allowable level of carcinogen PAHs in [22] for drinking water standard is  $200 \text{ ng} \cdot \text{l}^{-1}$  which is higher than the guideline values proposed by [22] in all stations except Al-Kahlaa district, Kumat district, Al-Silam district and Al-Mailmen Quarter (table,4). While sum of the carcinogenic PAHs concentration BbF, BkF, BghiP and IcdP ranged from 0- $143.79 \text{ ng} \cdot \text{l}^{-1}$ . However, the concentrations of carcinogen PAHs BbF, BkF, BghiP and IcdP in drinking water samples were lower than [23] in all stations except Al-Fatimya Quarter, Al-Hussein Quarter and Ali Al-Garbi district, which forces that sum of the carcinogenic PAHs concentration BbF, BkF, BghiP and IcdP should not exceed  $100 \text{ ng} \cdot \text{l}^{-1}$  (Table ,4).

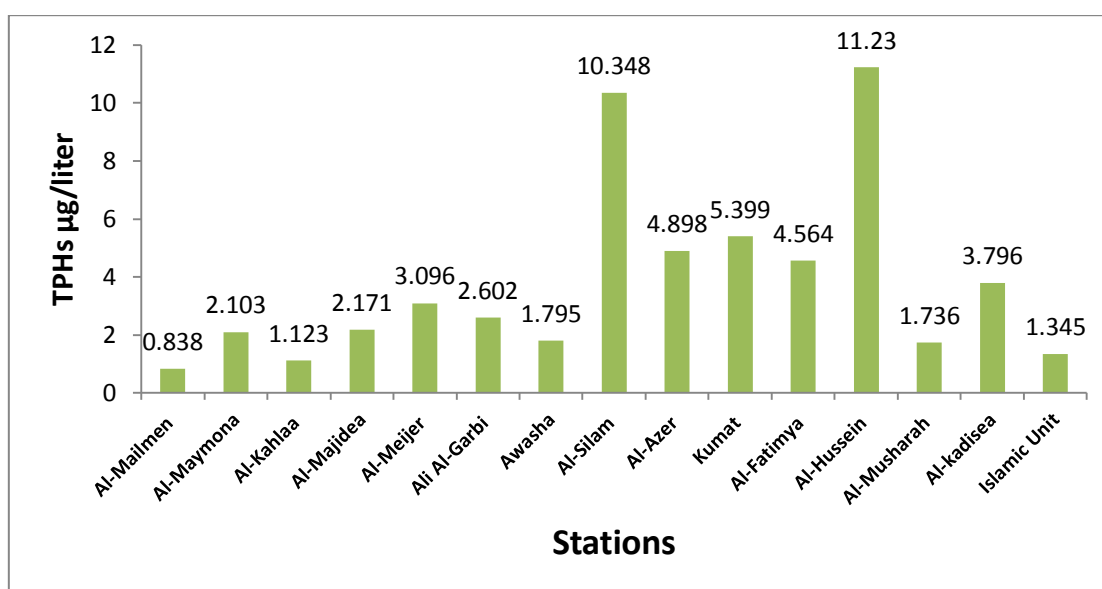


Fig. 1: TPHs concentration in different stations in drinking water samples ( $\mu\text{g} \cdot \text{l}^{-1}$ )

**Table 1: PAHs components concentration in stations Al-Silam , Al-Azer, Al-Fatimya , Al-kadisea and Al-Kahlaa (ng.l<sup>-1</sup>).**

Name compound	Al-Silam	Al-Azer	Al-Fatimya	Al-kadisea	Al-Kahlaa
Naphthalene Nap	ND	ND	ND	ND	ND
Acenaphthylene Ace	ND	ND	ND	ND	ND
Acenaphthene Acy	ND	34.06	ND	ND	248.22
Fluorene Fl	ND	ND	ND	ND	827.37
Phenanthrene Phe	ND	6.08	ND	2.65	ND
Anthracene Ant	50.65	ND	ND	ND	ND
Fluoranthene Flu	ND	ND	ND	1.82	ND
Pyrene Pyr	5.01	15.80	ND	ND	4.56
Benzo[a]anthracene BaA	3.49	ND	6.80	9.42	ND
Chrysene Chy	35.27	ND	1.78	0.84	3.43
Benzo[b]fluoranthene BbF	21.36	ND	3.68	3.04	8.02
Benzo[k]fluoranthene BkF	52.16	3.30	38.90	8.728	ND
Benzo[a]pyrene BaP	12.68	27.79	4.26	ND	3.18
Dibenzo[a,h]anthracene DahA	ND	211.35	213.31	244.57	64.38
Benzo[g,h,i,]perylene BghiP	ND	10.21	98.24	ND	ND
Indeno[1,2,3-cd] pyrene IcdP	ND	ND	ND	ND	ND
Σ16 PAHs	180.62	308.62	367.0	271.105	1159.211
Low PAHs	50.65	40.14	ND	4.47	1075.59
High PAHs	129.97	268.48	367	266.635	83.621
Low PAHs/ High PAHs	0.38	0.14	0	0.01	12.86
Phe /Ant	0	0	0	0	0
BaA/ (BaA+Chy).	0.09	0	0.79	0.918	0
IcdP /( IcdP + BghiP).	0	0	0	0	0
Flu/Pyr	0	0	0	0	0
ΣBaP, BbF, BkF, Chy and DahA	1.47	242.44	261.93	257.178	79.01
ΣBbF, BkF, BghiP and IcdP	64.52	13.51	140.82	11.768	8.02

**Table 2: PAHs components concentration in stations Al-Musharah, Al-Maymona , Al-Majidea , Al-Majidea and Islamic Unit (ng.l<sup>-1</sup>)**

Name compound	Al-Musharah	Al-Maymona	Al-Majidea	Al-Hussein	Islamic Unit
Naphthalene Nap	ND	ND	ND	ND	ND
Acenaphthylene Ace	ND	ND	ND	ND	ND
Acenaphthene Acy	ND	ND	ND	ND	ND
Fluorene Fl	22.33	ND	ND	ND	ND
Phenanthrene Phe	ND	2.30	2.44	29.59	ND
Anthracene Ant	70.79	ND	63.68	ND	ND
Fluoranthene Flu	2.85	1.48	ND	6.09	0.99
Pyrene Pyr	ND	1.37	2.98	ND	1.66
Benzo[a]anthracene BaA	11.71	ND	10.03	29.73	8.25
Chrysene Chy	ND	0.65	14.80	123.74	ND
Benzo[b]fluoranthene BbF	2.75	6.76	6.66	23.49	2.617
Benzo[k]fluoranthene BkF	ND	7.89	47.76	108.57	ND
Benzo[a]pyrene BaP	8.65	ND	ND	55.98	ND
Dibenzo[a,h]anthracene DahA	1241.93	406.39	2070.27	2778.6	789.52
Benzo[g,h,i,]perylene BghiP	ND	ND	ND	ND	ND
Indeno[1,2,3-cd] pyrene IcdP	ND	ND	ND	ND	ND

∑16 PAHs	1361.03	426.870	2218.674	3155.823	803.0687
Low PAHS	95.97	3.78	66.12	35.68	0.99
High PAHS	1265.06	402.61	2006.15	3119.92	802.07
Low PAHS/ High PAHS	0.07	0.009	0.03	0.01	0.001
Phe /Ant	0	0	0.038	0	0
BaA/ (BaA+Chy).	0	0	0.403	0.193	0
IcdP /( IcdP + BghiP).	0	0	0	0	0
Flu/Pyr	0	1.08	0	0	0.59
∑BaP, BbF, BkF, Chy and DahA	1253.33	421.69	2139.49	3090.38	792.137
∑BbF, BkF, BghiP and IcdP	2.75	14.65	54.42	132.06	2.617

**Table 3: PAHs components concentration in stations Al-Mailmen , Ali Al-Garbi , Awasha , Kumat and Al-Meijer (ng.l<sup>-1</sup>)**

Name compound	Al-Mailmen	Ali Al-Garbi	Awasha	Kumat	Al-Meijer
Naphthalene Nap	ND	ND	ND	ND	ND
Acenaphthylene Ace	ND	ND	ND	ND	ND
Acenaphthene Acy	ND	ND	ND	ND	ND
Fluorene Fl	ND	17.49	ND	ND	ND
Phenanthrene Phe	ND	ND	ND	ND	ND
Anthracene Ant	ND	ND	ND	ND	ND
Fluoranthene Flu	1.32	1.98	ND	ND	ND
Pyrene Pyr	0.81	ND	2.42	3.103	2.40
Benzo[a]anthracene BaA	5.97	7.27	ND	ND	ND
Chrysene Chy	0.75	56.16	11.10	ND	3.10
Benzo[b]fluoranthene BbF	1.73	19.30	12.77	9.08	ND
Benzo[k]fluoranthene BkF	ND	124.49	28.93	27.86	ND
Benzo[a]pyrene BaP	ND	6.40	7.79	17.42	2.95
Dibenzo[a,h]anthracene DahA	ND	ND	3591.76	98.77	ND
Benzo[g,h,l]perylene BghiP	ND	ND	ND	ND	ND
Indeno[1,2,3-cd] pyrene IcdP	ND	ND	ND	ND	ND
∑16 PAHs	10.60	233.136	3654.8	156.249	8.460
Low PAHS	1.32	19.47	0	0	0
High PAHS	9.28	213.66	3654.8	156.249	8.46
Low PAHS/ High PAHS	0.14	0.09	0	0	0
Phe /Ant	0	0	0	0	0
BaA/ (BaA+Chy).	0.88	0.114	0	0	0
IcdP /( IcdP + BghiP).	0	0	0	0	0
Flu/Pyr	1.62	0	0	0	0
∑BaP, BbF, BkF, Chy and DahA	2.48	206.35	3652.35	153.13	6.05
∑BbF, BkF, BghiP and IcdP	1.73	143.79	41.7	36.94	0



**Table 4: Comparing the results and PAHs standards in drinking water (ng.l<sup>-1</sup>)**

Components	USEPA ng.l <sup>-1</sup>	ISIR ng.l <sup>-1</sup>	EU ng.l <sup>-1</sup>	WHO ng.l <sup>-1</sup>	Results ng.l <sup>-1</sup>
BaP	200	700	10	700	ND – 55.89
∑BaP, BbF, BkF, Chy and ahA	200	--	--	--	1.47-3652.35
∑BbF, BkF, BghiP and IcdP	--	--	100	--	0-143.79

## CONCLUSIONS

Detection of TPHs and PAHs in the present study shows that water sources are contaminated which indicates the inefficiency of water resources management in Misan governorate in most sampling stations. Concentrations of HMW-PAHs were more than concentrations of LMW-PAHs in drinking water samples in most sampling points. According to the ratios of LMW-PAHs/HMW-PAHs, Phe /Ant, BaA / (BaA + Chy), IcdP / (IcdP + BghiP) and Flu/Pyr the sources of PAHs were pyrogenic as a main and few are petrogenic. In all sampling stations, the concentrations of BaP were less than the permissible limits of WHO, ISIRI, USEPA and EU except some stations which was above the permissible limits of European Union's drinking water standard.

Sum of the carcinogenic PAHs concentration BaP, BbF, BkF, Chy and DahA were higher than the guideline values proposed by USEPA for drinking water in all stations except in some stations, whereas sum of BbF, BkF, BghiP and IcdP were lower than the guideline values proposed by EU for drinking water in all stations except in some stations.

To protect drinking water sources as well as to prevent adverse effects on humans and biota, authors' recommendations to protection the water sources from different pollutants.

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**Cite this article as:**

Salih H. Jazaa, Hamid.T. Al-Saad, Salah M. Salih, Abd alhussein A. Khwadem. Estimation of Hydrocarbon Compounds in Drinking Water in Misan Governorate / Iraq. *J Pharm Chem Biol Sci* 2016; 4(2): 291-298.