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### Identification of polycyclic aromatic hydrocarbons in groundwater in the wells of Al-Zubair district near the Basra refinery, southern Iraq

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### Abstract

The current study was conducted to estimate the concentrations of polycyclic aromatic petroleum hydrocarbons with their dissolved and suspended particles in the groundwater of eight wells, the six wells from w1 to w6 close to the refinery area about half a kilometer to three kilometers and the two wells w7 and w8 are away from the site about 12.5 km (wells of control), in The area surrounding the South Refinery (Al-Shuaiba) Al-Zubayr district in Basra Governorate, southern Iraq. Sixteen compounds of PAHs in water were identified in both the dissolved and suspended parts in the spring of 2016 due to the high value of dissolved and suspended hydrocarbons and the transport of petroleum hydrocarbon pollutants through the rain that transport the particles and dissolved materials through the pores of soil to groundwater. The total concentrations of PAHs dissolved in water ranged between 7205 ng/l in well W1. In comparison, the lowest value was 39.1 ng / 1 in well W8. The highest concentration of Phenanthrene in well W1 was 3472 ng/l, and the lowest concentration of Benzo (a) Pyrene was 0.37 ng / 1 for well W8. Benzo (b) Fluoranthene was recorded predominantly in the dissolved part. The total concentrations of PAHs ranged from total suspended in water, between the highest value of 9334 ng/g dry weight in well W1. The lowest value of 370 ng/g dry weight in well W8 and the most abundant compound is Fluorene, as Dibenzo (a, h) anthracene recorded the highest concentration of 6689 ng/g dry weight in well W3 and the lowest concentration of 13 ng/g the compound Benzo (a) anthracene was in well W8. It was concluded that oil refinery has a significant impact in causing pollution of nearby water wells with some dangerous and carcinogenic polycyclic hydrocarbon pollutants .

Key words: polycyclic aromatic hydrocarbons - groundwater - Basrah refinerys.

### Introduction

people's Nowadays, society and livelihoods suffer due to disturbances in the sustainability of ecosystems, especially the exploitation of water resources. An impact on a component of our ecosystems can affect the resources available, economic growth, and social aspects. Mankind needs to be guided successfully by taking sustainable actions within the available environmental resources(Mahmoud .et al.2021)The problem of water pollution is a threat threatening human life and living

organisms. This problem has emerged as a result of industrial progress and population increase over the years. This problem has become one of the major global problems that resulted from poor planning and use of environmental programmed (Zietz et al., 2003). Water resources resources had the largest share of pollutants resulting from these sources, and this resulted in a significant deterioration in their quality at a time when the need for fresh resources increased, as well as a decrease in these resources due to the high temperatures and droughts that swept the globe (Fawell and Nieuwenhuijsen, 2003) and the degree of severity of many pollutants depends on the chemical nature of the pollutant, its concentration, the spread of pollutants and their distribution between water, air, land, and living things and biology, and its focus on the image of the pollutant was dissolved or stuck in the water or adhered to dust particles and because groundwater is a reserve reservoir for stored fresh water and represents the most important sources of drinking water in many countries the world, groundwater is exposed to many pollutants resulting from a variety of sources, including pollutants resulting from agricultural and urban activities, which have become today a global problem (Alia, et al, 2018). Surface or groundwater sources contain dissolved solids and dissolved gases as well as suspended materials. These components depend on the quantity and quality of geological and environmental factors. It always changes as a result of the interaction of water with the field associated with human activities, so chemical tests must be done before using water for domestic, agricultural, or industrial purposes. These tests are compared with the acceptable specifications for water use (Al-Sayed, 2005). The quality of groundwater is affected by the different stages of the hydrological cycle, such as rain and snow, which consists of water that lost its purity during its journey in the atmosphere and before its arrival to the earth, as pollutants in the atmosphere are the result of gases and dust in addition to the solids carried by the wind and gases resulting from human activities. When the rainwater reaches the soil, the water dissolves the minerals present in the soil and rocks (Al-Dabbas, et Oil pollution has occupied an al., 1989). important position among environmental pollution sources, and this importance comes due to the increase in global oil production rates (Elias, 1989). It leads to

damage to the water environment by changing its physical, chemical, and biological properties and damaging aquatic organisms, which leads to harm to humans, directly or indirectly. (Al-Sayegh and 2002) Petroleum hydrocarbon Tagah, compounds are among the most important pollutants in the water environment that affect water with dissolved and suspended particles and living organisms and sediments (Al-Saad et al., 1998; Al-Hamdi, 1989). The solubility of oil in water is usually low (GESAMP, 1993); it depends on the crude oil's chemical composition and temperature (Ehrhardt, et al., 1992). Alkanes are less soluble in water compared to aromatic hydrocarbons (Thomas, et al., 1995). The most common crude oil components dissolved in water are the aromatic compounds with low molecular weights, such as benzene, toluene and xylene (Zhu, et al., 2004). Polycyclic Aromatic Hydrocarbons PAHs have special environmental concern because they are carcinogenic or may turn into carcinogenic compounds that can cause serious health 2012: problems.( Douabul, et al., Otokunefor and Obiukwu ,2005) explained the effect of oil discards on the quality of the physical properties of those discards' received surfaces. The water environment is exposed to many sources of pollution, including industrial discards resulting from various industries such as the paper industry, fertilizers, spinning and weaving, rubber, oil refineries, and petrochemical industries, as the environmental impact resulting from polluted discards from industrial sources is one of the main problems in many countries of the world (Irshad, et al., 1997) hydrocarbons are a widespread pollutant that the environment is exposed to continuously. Polycyclic aromatic hydrocarbons (PAHs) are chemical pollutants formed from benzene cyclic structures of a hydrophobic nature. This property increases with increasing their molecular weight and are environmentally stable compounds (Juhasz and Naidu, 2000). Among the hundreds of aromatic hydrocarbon compounds 16 have been identified that are considered to be priority pollutants as being among the most harmful compounds for humans and other living organisms (Ravindra et al., 2008; Anyakora and Coker, 2006). PAHs (depending on their molecular weight) can be divided into two groups (CCME, 2008). **PAHs** are low molecular weight compounds and usually consist of two or three rings of benzene (such as naphthalene, fluorene, and acenaphthylene), and this type is usually accompanied by incomplete combustion of the fuel high molecular weight PAHs consist of four or more benzene rings, usually called Heavy PAHs, include Chrysene, Pyrene and anthracene pyrene. This type is associated with crude oil compounds spilled in bodies of water and soil, and hydrocarbon compounds that have the potential to cause cancer are Benzo (a) pyren Benzo (b) flouranthen, anthracene, Benzo (k) flouranthen (Hernandez, el at., 1995;. Stephanie, et al.1999). Benzo (a) pyren is one of the hydrocarbon compounds most capable of causing cancer in living populations and has dangerous and toxic effects (Guengerich, 1993). Petroleum hydrocarbons have a toxic effect on living organisms. Their toxicity depends on the type of organism, its life stages and the environmental conditions surrounding it (Linden, 1984). The lethal effect that occurs as a result of the chronic effect, which results from exposure of the organism to concentrations low of petroleum hydrocarbons for a long time, affects various vital activities such as growth, sexual maturity, metabolism rates. immunity, and enzyme activity (GESAMP,1977). The impact of refinery waste discharges as an oil region and its proximity to groundwater sources used as a main source for agriculture and animal watering, and due to the lack of studies on it. Contamination of groundwater with polycyclic aromatic petroleum

hydrocarbons determining the concentrations of polycyclic aromatic hydrocarbons in the groundwater in the wells and especially carcinogens.

### **Materials and Methods**

Water samples were collected during the spring of 2016 for the studied wells. Standard methods were adopted in collecting, transporting, and preserving samples for conducting the analyzes. The water samples for measuring hydrocarbons were collected in opaque glass bottles of 5-liter capacity and were field-fixed using carbontetrachloride.

### Description of the study area

Al-Zubayr district occupies the southwestern part of Basra governorate, extending over an area estimated at 11,618 km<sup>2</sup> of the governorate's area of 19,070  $km^2$  with a percentage estimated at 61.9% of its area. The district is characterized by high temperatures for most days of the year, offset by a rise in evaporation rates. Of sand and gravel, which is characterized by its high ability to store water. The injection process, has recently spread in the central and northern regions of the district due to the disruption of the quality control devices of these institutions, including the South refineriy in Al-Zubayr, (Shuaiba) as quantities of wastewater, along with petroleum hydrocarbons, are injected into newly dug wells, the depth of which reaches 18 Up to 20 meters to dispose of this waste as well as the operations of its disposal to the open, which forms a lake of waste with a width of 10 km<sup>2</sup> (Rahim 2008.). These wastes were previously dumped into the Shatt al-Arab canal, which caused an increase in contamination level (Hassan, et al., 2011). Eight groundwater wells were selected, with depth limits ranging between 24-22 meters, and the coordinates of each well were determined using GPS (Global Positioning System). Beside the refinery, a large lake with an area of 12 km<sup>2</sup> was formed from solid and liquid wastes that were discharged or leaked from the transport pipelines and tanks. Two wells have been identified far from pollution (control mains) in an area that is almost free of factories and industrial companies and is not surrounded by any waste dumped within the vicinity of Al-Zubair district.

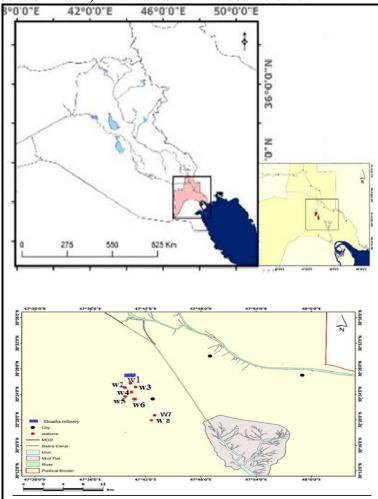


Figure (1) Map of Iraq and the city of Al-Zubayr showing the refinery in the south and the study

sites in the district of Al-Zubayr

Hydrocarbons were extracted from the water (suspended and dissolved), where 5 liters of water were filtered through a Millipore filter, the pore volume was 0.45  $\mu$ m, and the part that passed through the filter paper represented the dissolved part while what remained on the filter paper represented the suspended portion that was then dried weigh to measure.

# Extraction of petroleum hydrocarbons dissolved in water

According to the United Nations Environmental Protection Program, the dissolved hydrocarbons in the water were extracted (UNEP, 1989). Fifty ml of carbon tetrachloride was added for every 5 liters of the sample. The sample was shaken well using an electric mixer for 30 minutes, and the contents were transferred to a separating funnel and left until it stabilized. Then the lower layer representing the petroleum hydrocarbons was collected. With a beaker and evaporated using a rotary evaporator, then I passed a chromatographic separation column below it onto the glass wool layer, then 2 grams of alumina (Al<sub>2</sub>O<sub>3</sub>) and 2 grams of anhydrous sodium sulfate( Na<sub>2</sub>SO<sub>4</sub>) to remove the remaining water in the sampale , 30 ml of hexane is passed to obtain the aliphatic fraction. After that, 30 ml of benzene was added to obtain the aromatic fraction. The latter was evaporated to dryness, and then 5 ml of hexane were added to make the sample ready to measure the total concentrations of hydrocarbons petroleum using а Spectroflourometer. This device can determine the concentrations of aromatic hydrocarbons as an indicator of the total concentrations of hydrocarbons in the sample after comparing them with the fluorination of standard solutions prepared from Basrah regular crude oil in the same conditions.

## Extraction of suspended petroleum hydrocarbons in water

Petroleum hydrocarbons suspended in water were extracted according to the method mentioned by Goutx and Saliot (1980), and according to the following steps, the weighted suspended materials were taken on the filter paper, and the intermittent extraction process was performed Soxhlet Extraction. Noncontionuos using а mixture of methanol-benzene at a ratio of (1: 1) and conducted for it. The extraction process for 24-36 hours in the extraction device, after which the soaping process is carried out for two hours by adding 20 mL of the solution to the 4 standard KOH-Methanol potassium hydroxide solution obtained by dissolving 22.48 grams of KOH in 100 milliliters of methanol with continuous stirring traction of petroleum hydrocarbons suspended in water After the period is over. The extract is left to cool down, then 50 ml of n-hexane is added to the separating funnel and shaken well and then left to settle, forming the upper two layers, which are the layer containing hydrocarbons and the lower layer containing fatty acids, where they are disposed of. The layer containing the hydrocarbon compounds dissolved in hexane is taken and passed over a chromatography column that contains at the bottom a piece of glass wool topped by 2 grams of silica and 2 grams of anhydrous

sodium sulfate to absorb water if any and 2 grams of alumina to get rid of the fatty acid residue. Then 50 mL of hexane is added to separate the alpha fraction. Then 50 are added One milliliter of benzene to lower the aromatic compounds and evaporate to the point of dehydration so that the sample is ready for analysis by fluoridation device. The total concentration of aromatic hydrocarbons is measured using а fluoridation device, which is very sensitive low concentrations of aromatic to hydrocarbons compounds. Aromatic possess a high degree of resonant stability and have sufficient ability to fluoresce, especially compounds that have multiple aromatic nuclei, as they are among the most compounds for fluorescence. intense of Polycyclic Standards Aromatic Compounds provided by the American company Ultra Scientific, Fig. (2) Were used to determine the concentrations and specificities of the polycyclic aromatic compounds in the samples by injecting them into the Shimadzu High-Performance Liquid Chromatography (HPLC).

### **Statistical Analysis**

used the ready-made statistical program Special Package for Social Science (SPSS) in analyzing the data statistically and testing the Least Significant Difference (LSD) at a probability level of P < 0.05 and extracted the value of Standard Deviation.

### **Results and discussion**

Polycyclic aromatic hydrocarbons dissolved in water

Table (1) shows the concentration of polycyclic aromatic hydrocarbons for the eight wells during spring 2016 of water samples. The highest total PAHs concentrations reached 7205 ng/ L in well while the lowest total PAHs W1. concentrations reached 39.1 ng/ L in well w8 (Figure 3). The highest concentration of Phenanthrene was in well w1, with a value of 3472 ng/L, and the lowest concentration was 0.37 ng/ L of the compound Benzo (a)

pyrene in well w8 and the dominance of the compound Benzon (b) fluoranthene. followed by the two compounds Acenaphthlene and Acenaphthene. The results of the statistical analysis showed that there were significant differences at the likelihood level P < 0.05 among the study wells. Note that the lowest significant difference (LSD = 65). The current study of the water-soluble fraction showed that the highest total number of PAHs was recorded in well W1, while the lowest was recorded in well W8. This may be due to the proximity of well W1 to the source of pollution and the possibility of groundwater being affected by the flows of liquid and solid wastes that are discarded from the southern refineries, and this coincides with This is consistent with the study of( Mahmood and Al-Imarah,2001) .When there is a large increase in water, the torrents collect in the floods scattered in the study area and its vicinity, thus constituting a source of local nutrition, as in the study (Al-Kubaisi, 1996). In the study (Li, et *al.*,2017), it was shown that PAH

concentrations in the groundwater ranged from 8.51-402.84 ng / L this indicates pollution in the elevation stage The mean value of mixture ratio of the Yellow River water recharge to the groundwater was 65%, few anomalous sites can reach to 90% in light of the ongoing serious pollution, management practices for source control, improved control technologies, and the construction of a monitoring network to warn of increased risk are urgently needed. Consequently, these polycyclic hydrocarbons may be transported with the water of these rains and torrents to the groundwater. while the highest concentration of Phenanthrene was in the well W1. Contamination of the aquatic environment with polycyclic aromatics is mainly associated with human activities that are the main source of them in the aquatic environment compared to natural sources (Zakaria, et al., 2002; Yan, et al., 2012). These compounds are produced from oil and its derivatives and waste from factories and laboratories proposed for the environment (Mohammed, 2007).

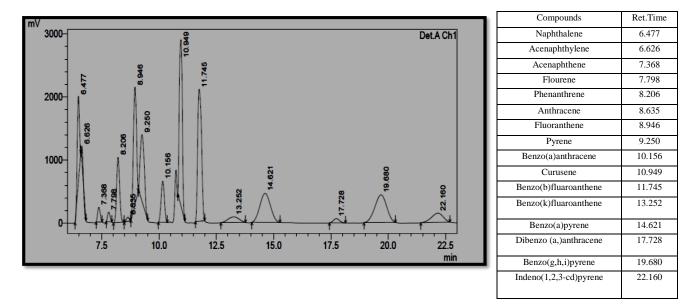


Figure (2) Liquid Chromatography (HPLC) of a standard sample of PAH

Table (1) the concentrations of polycyclic aromatic hydrocarbons ng/ L dissolved in water for
study wells during the spring 2016

PAHs	W1	W2	W3	W4	W5	W6	W7	W8	Total
Naphthalene	-	-	-	-	1.066	0.446	-	-	1.51
Acenaphthylene	-	395.3	26.87	191.3	139.0	50.25	88.01	-	1132
Acenaphthene	676.4	117.7	48.47	100.3	93.99	53.73	-	-	1090
Fluorene		175.8	414.2	-	-	-	-	-	590
Phenanthrene	3472	-	-	-	20.12	13.72	-	-	3505
Anthracene	-	-	-	54.43	-	-	2.663	-	57
Fluoranthrene	-	-	3.599	-	-	-	-	-	3.59
Pyrene	-	-	-	10.89	7.964	4.430	-	-	23.28
Benzo(a)anthracene	-	10.66	19.41	-	58.08	10.48	2.510	-	101.1
Chysene	51.01	9.822	24.83	-	0.698	-	-	4.826	91.1
Benzo(b)fluoranthene	223.2	96.60	39.29	1.046	2.878	0.351	3.470		366.8
Benzo(k) fluoranthene	117.7	-	-	-	2.878	4.001	3.577	-	128.7
Benzo(a)pyrene	1371	-	-	-	5.503	55.82	4.403	0.375	1437
Dibenzo(a,h)anthracene	-	1337	-	16.71	-	-	-	5.340	1359
Benzo(a,h,i)peryene	-	-	-	-	-	-	-	20.32	20.32
Indeno(1,2.3-cd)pyrene	1294	180.7	-	-	-	-	-	8.258	1482
Total	7205	2323	818.5	374.7	332.6	193	104.6	39.1	11390

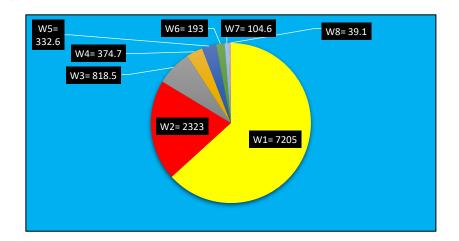


Figure (3) Total of PAHs dissolved in water for study wells during Spring 2016

## Polycyclic aromatic hydrocarbons in the suspension

Table (2) shows the PAHs in the suspended wells in the selected wells. PAHs recorded the highest value of 9334 ng/g dry weight in well W1, while their lowest value reached 370 ng/g dry weight in well W8. most abundant compounds The are composite Fluorene As well as the two compounds Phenathrene and Benzo (b) fluoranthene during Spring 2016, the highest concentration of Dibenzo (a, h) anthracene were represented as its value reached 6689 ng/g dry weight in well W3 While the compound Benzo (a) anthracene was the least concentrated, as it recorded 13 ng / grams of dry weight in well W8, the results of the statistical analysis showed that there were significant differences at a probability level P <0.05 between the study wells, noting that the lowest significant difference LSD = 23.5. The results showed that the total number of PAHs in the suspended part of the study wells is higher than in the soluble part because they are poor soluble compounds in water because they do not dissolve with polar solvents (Muhammad and Al-Saiti, 2008) Therefore, it may adsorb or surround the particles of plankton with rainwater, and this explains its greater percentage than that dissolved in it. The diffusion of organic matter and particulate matter in the water column and the volumetric distribution of the organic particles helps to adsorb the PAHs compounds particles' on the surface (Elkhon, 2012). Hydraulic cracking and cracks between the geological layers, exploration work, surface spills. leaching, and industrial subterranean wastewater may lead to groundwater damage, which may explain the contamination of nearby wells with high concentrations of polycyclic aromatic compounds compared to control wells that are far from the pollution area.

# Table (2) Concentrations of PAHs (ng/g) by dry weight of suspended wells in studied wells during spring

PAHs	W1	W2	W3	W4	W5	W6	W7	W8	Total
Naphthalene	-	-	-	-	-	15		-	15
Acenaphthylene	1769	4320	-	-	1950	61	-	-	8100
Acenaphthene	-	1358	-	369	209	-	317	-	2253
Fluorene	2057	-	124	265	347	1135	163	-	4091
Phenathrene	-	1792	51	26	2105	-	53	-	4030
Anthracene	2640	-	-	159	115	3332	-	1	6246
Fluoranthrene	-	68	-	25	-	-	23	-	116
Pyrene	-	-	106	-	189	14	-	1	309
Benzo(a)anthracene	-	-	-	-	21	28	-	13	62
Chysene	-	-	-	242	12	215	75	-	544
Benzo(b)fluoranthene	2868	-	261	-	357	529	-	356	4372
Benzo(k) fluoranthene	-	-	-	4913	-	-	203	-	5116
Benzo(a)pyrene	-	-	-	-	-	-	619	-	619
Dibenzo(a,h)anthracene	-	-	6689	-	-	-	-	-	6689
Benzo(a,h,i)peryene	-	-	-	I	-	-	-	-	-
Indeno(1,2.3-cd)pyrene	-	-	-	-	-	-	-	-	-

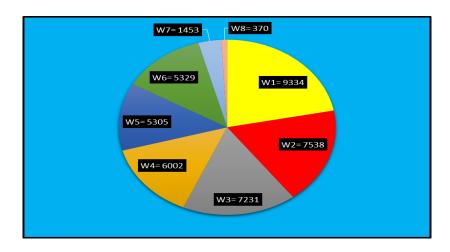


Figure (4) Total of PAHs in the suspended for study wells during spring 2016

### Conclusions

From the quantitative and qualitative analysis of the dissolved and suspended fraction of water in this study, we concluded that the transfer of toxic and carcinogenic polycyclic aromatic compounds to groundwater is a dangerous matter, especially for agricultural lands that use groundwater for irrigation ,concluded that the concentrations of the polycyclic aromatic compounds in the suspended part of the study wells are higher than the soluble The wells close to the pollution fraction. source have more concentrations of hydrocarbons than those distant and control wells. The water of the studied wells carcinogenic hydrocarbon contains compounds, including Phenanthrene and Dibenzo (a, h) anthracene, which recorded the highest concentrations and contained the most important carcinogenic compounds, Benzo (a) pyrene in Some wells Therefore, when constructing a well, it must be protected from pollutants by not randomly throwing and injecting oil residues, treating leaks from pipelines that transport crude oil, and treating spent flows, and the responsible authorities in the governorate must monitor oil spills near the wells.

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# تشخيص الهيدروكربونات الأروماتية متعددة الحلقات في المياه الجوفية لأبار قضاء الزبير القريبة من مشخيص الهيدروكربونات الأروماتية من مصفى نفط البصرة جنوب العراق

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#### المستخلص

أجريت الدر اسة الحالية لتقدير تراكيز الهيدروكربونات النفطية الأروماتية متعددة الحلقات بجزيئها الذائب والعالق في المياه الجوفية لثمانية أبار , الابار السنة من W1 اليW6 قريبه من منطقة المصافي حوالي نصف كيلو متر الي ثلاثة كيلو مترات والبئرين W7 وW8 بعيدان عن المنطقة حوالي 12.5 كم (ابار سيطرة) في المنطقة المحيطة لمصافى الجنوب (الشعيبة) قضاء الزبير في محافظة البصـرة جنوب العراق. تم تحديد 16 مركب من مركبات الهيدروكربونات الأروماتية مُتعددة الْحلقات في الماء في كل من الجزء الذائب والعالق في ربيع عام خلال ربيع 2016 وذلك لارتفاع قيمه الهيدر وكربونات الذائبة والعالقة خلال الربيع بسبب انتقال الملوثات الهيدر وكربونية النفطية عن طريق الأمطار التي تنقل الدقائق والمواد الذائبة عبر مسامات التربة الى المياه الجوفية ولكون الامطار كانت غزيرة في فصل ربيع 2016 اختير هذا الفصل. وتراوحت مجموع تراكيز PAHs الكلية الذائبة بالماء بين اعلى قيمة 7205 نانوغرام /لتر في البئر W1 بينما بلغت اقل قيمة 39.1 نانوغرام /لتر في البئر W8 وكان اعلى تركيز للمركب Phenanthrene في البئر W1 3472 نانوغرام / لتر واقل تركيز كان للمركب 0.37 Benzo(a)Pyrene نانوغرام / لترفي البئر W8 ســجل المركب Benzo (b)Fluoranthene سيادة في الجزء الذائب . تراوحت مجموع تراكيز PAHs الكلية العالقة في الماء بين أعلى قيمة 9334 نانوغرام/ غم وزنا جافا في البئر W1 واقل قيمة 370 نانوغرام/ غم وزنا جافا في البئر W8 وأكثر المركبات تواجدا هو مركب Fluorene, اذ ســجل Dibenzo(a,h)anthracene اعلى تركيز 6689 نانوغرام/غم في البئر W3 واقل تركيز 13 نانوغرام/ غم كان للمركب Benzo(a)anthracene في البئر W8. استنتج من الدراسة ان مصــافي النفط لها اثر كبير في احداث تلوث ابار المياه القريبة ببعض الملوثات الهيدر وكربونية متعددة الحلقات الخطرة والمسر طنة.

الكلمات المفتاحية: هيدروكربونات الأروماتية متعددة الحلقات -المياه الجوفية – مصفى البصرة.