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ASSESSMENT OF POLYCYCLIC AROMATIC HYDROCARBONS OF BOTTLED WATER AVAILABLE IN IRAQI LOCAL MARKETS

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ABSTRACT

The present study is intended to assess the type and content of 16 specific polycyclic aromatic hydrocarbons in bottled drinking water available in Iraqi local markets (The study included eight local brands, one tap water sample, and three imported brands). The evaluation is conducted using gas chromatography, and the results are compared against the standards set by US Environmental Protection Agency and World Health Organization. The results indicate that most of the samples of bottled drinking water in Iraqi local markets contained polycyclic aromatic hydrocarbons (PAHs) compounds, sample 1 contained ∑PAHs High (2.21 µg/L) and highest content of Acenaphthylene and Anthracene of concentrations 0.445 and 0.325 µg/L respectively , sample 5 contained ∑PAHs Low (0.99 µg/L), the presence of the naphthalene compound (2 rings) was observed in most of the study samples, and sample 1 contained the highest concentration (0.275 µg/L), the content of tricyclic compounds (3 rings) prevailing in most samples 31.25%, quaternary compounds (4 rings) 18.75%, five-cyclic compounds (5 rings) 12.5%, hexacyclic (6.25%). The absence of compounds such as Dibenzo (A, H) anthracene, Chrysene, Benzo (A) Pyrene, Benzo (G, H) Perylene and Benzo (K) Fluoranthenen in all the samples is a positive finding. While the compounds Anthracene, Phenanthrene (3 rings), and Benzo (A) Anthracene (5 rings), were present and did not conform to the standard specifications of US Environmental Protection Agency and World Health Organization in more than 50% of studied samples.

INTRODUCTION

Drinking Water is defined as any liquid that does not hurt people or is not significantly contaminated, water is considered polluted when it exceeds the standard specifications. No living thing can exist without water; hence, it is a crucial and required component of all industries and processes. It is required for all bodily interactions. Neighborhoods (Abbas, 2018). Access to water is one of the most basic human needs that cannot be overlooked. It must meet particular physical, chemical, and biological specifications and is a generally acknowledged human right (WHO, 2022). One of the most common food and beverage industries is bottled water. Drinking water consumption is continually increasing due to consumer demand and water pollution caused by chlorination difficulties. The stability of ecosystems is generally seriously threatened by water pollution, which can result from any factor that modifies the properties or character of water. As a result of the bottled water industry's rapid growth over the past ten years, Iraqi consumers have largely come to rely on water packaged in plastic bottles of various sizes, some of which are used only once and others repeatedly (Ibrahim and Kashmola 2011; Alobaidy and Almahdawi, 2016). Currently, Iraq has 54.5% of all the various food industry factories in the country. The number of licensed industrial projects that produce bottled water in Baghdad reached 15 factories, compared to 234 unlicensed factories distributed among the different governorates (Razuki and Al-Rawi, 2010). PAHs are natural chemicals consisting of multiple connected rings. These rings are composed of carbon atoms and are connected in a cyclic structure. PAHs are characterized by the absence of heteroatoms (atoms other than carbon and hydrogen) or substituents within their ring structures (Chen *et al*., 2018). Nowadays, a number of organic pollutants can be detected in different water sources because of many man-made and natural disasters. Organic pollutants that contaminate water are frequently hazardous and carcinogenic, raising concerns across the globe (Shallal and Ubaid 2007; Gorji *et al*., 2016). Many sources and activities can contribute to the presence of organic pollutants, including polycyclic aromatic hydrocarbons (PAHs), in the environment. Such as: Fossil Fuels: Oil, gasoline, coal, Wood and Biomass Burning, Industrial Operations: such as foundries, steel, aluminum, and iron manufacturing, Waste Incineration, and tobacco smoke (Shariatifar *et al*., 2021). Polycyclic aromatic hydrocarbons (PAHs) are classified as persistent organic pollutants (POPs). PAHs are considered xenobiotics, which refers to compounds that are foreign to an organism or ecosystem. The specific PAHs that have been classified as pollutants by the United States Environmental Protection Agency (USEPA) (USEPA, 2011). USEPA, (2018) has indeed identified certain PAHs as priority pollutants due to their potential toxicity and adverse effects on human health and the environment. The 16 PAHs are: [(indeno[1,2,3-cd]pyrene (IP), dibenzo [a,h] anthracene (DhA), benzo[g,h,i]perylene (BgP), anthracene (A), fluorene (F), fluoranthene (Fl), acenaphthene (Ac), acenaphthylene (Ace), benzo(a)anthracene (BaA), phenanthrene (Pa), pyrene (P), naphthalene (NA), chrysene (Ch), benzo(k)fluoranthene (BkF), benzo[a]pyrene (BaP), and benzo[b]fluoranthene (BbF), are commonly found in various environmental matrices and have been studied for their toxic properties (Purcaro *et al*., 2013). Human exposure to polycyclic aromatic hydrocarbons (PAHs) has been associated with an increased risk of various malignancies and other health conditions. PAHs are known or suspected carcinogens, meaning they have the potential to cause cancer in humans. Long-term exposure to PAHs, particularly through inhalation or ingestion, has been linked to an increased incidence of several types of cancer, including bladder, lung, stomach, and oral cancers. Polycyclic aromatic hydrocarbons (PAHs) have been associated with various effects on the immune system and are also suspected to be endocrine-disrupting chemicals (EDCs) (Gorji *et al*., 2016; Sharifiarab *et al*., 2022). PAHs are found in water like groundwater, saltwater, and river water because people are doing more things that create them and not managing industrial waste. Many lakes and rivers around the world are not safe to drink from because they have too many PAHs in them (Sharifiarab *et al*., 2022). Drinking water from bottles is liked by a lot of people around the world. People from all around the world are increasingly choosing to drink bottled water. From 1994 to 2002, the amount of bottled water sold went up from 58 billion liters to 144 billion liters, for example, per capita consumption of bottled water is about 47 gallons in the United States per year in 2022 (PCCBWUS, 2024). Indeed, with the significant increase in the production and consumption of bottled water, it is crucial to continuously monitor water quality to ensure its safety and adherence to regulatory standards. Monitoring water quality in bottled water involves various aspects, including source water quality, production processes, and finished product testing (Vega *et al*., 2011). There are no previous studies have been conducted on the presence of PAHs in different bottled waters around the world, with specific focus on Iraq. As a result, conducting a study to evaluate the polycyclic aromatic hydrocarbon (PAH) content in bottled water in Iraqi local markets according to USEPA and WHO standards is a valuable initiative.

MATERIALS AND METHODS

The Collecting of Samples

Seventy-eight samples total,11 brands,1 tap water, 7 samples from each brand were collected randomly from different Iraqi markets, taking into account production date convergence and same storage conditions (at room temperature), Then the samples were taken to the lab and kept in the refrigerator at 4°c. The information of bottled water is showed in Table (1), (according to the bottled water label) (Clesceri,1998 and Al-Emam,2012).

Table (1): Bottled water Information.

S. Type	S. ID	St. type	Water source	Cont.	Size (L)	P.C.
Local	1	O	MW	P	0.5	Iraq
	2	---	MW	P	0.5	Iraq
	3	O	SW	P	0.5	Iraq
	4	O	SW	P	0.5	Iraq
	5	O,UV	---	P	0.33	Iraq
	6	---	---	P	0.5	Iraq
	7	O,UV	---	P	0.5	Iraq
	8	O	TR	P	0.5	Iraq
	9*	Cl	TR	---	---	Iraq
Imported	10	---	MW	P	0.5	Italy
	11	---	SW	P	0.5	Uk
	12	---	MW	P	0.33	Turkey

MW= Mineral water, SW= Spring water, *= Tap water, TR= Tigris River, O= Ozone, UV= Ultraviolet, Cl= Chlorine P=Plastic, S= Brand Sample, St.= Sterilization, Cont.= Container, Producing country= P.C.

Sample Preparation for the Determination of PAHs by Gas Chromatograph (GS)

Extraction

50 ml of the homogenized sample were put into clean glass tubes, and 15 mL of acetone/n-hexane (1:1) were then added. The tubes with the sample were mixed for 1 minute using a vortex and then treated with ultrasonic waves for 15 minutes to

extract the samples. A centrifugation at 3000 rpm for 10 minutes was employed to separate the mixed sample and organic phase. The organic layer containing the extracted compounds, including PAHs, was collected into another clean glass tube using a Pasteur pipette. Furthermore, the residual sample was re-extracted twice with a mixture of acetone and n-hexane in a 1:1 volume ratio (v/v) using 5 mL of the solvent mixture. After collecting all the extracts containing the extracted compounds, including PAHs, they were combined. Then, activated copper was added to the extract for desulfurization. The extract was further processed by drying over anhydrous sodium sulfate, concentrating to 0.5 mL using a gentle stream of nitrogen, and finally, transferred to the refrigerator for storage until analysis is performed (Chen *et al*., 2019).

GC condition

The analysis of polyaromatic hydrocarbons (PAHs) was conducted at the Food Research Center, Ministry of Science and Technology of Iraq, using gas chromatography (GC) technology. Specifically, a Shimadzu GC-2010 model from Japan was employed for the analysis. The separation column used was an HP-5MS capillary column with the following specifications: length of 30 meters and an internal diameter of 0.25 mm. The following temperature program was utilized: Initial temperature: 40 °C (held for 1 minute), temperature ramp rate: 35 °C/min, temperature 1: 120 °C, temperature ramp rate: 10 °C/min, temperature 2: 160 °C temperature ramp rate: 5 °C/min, and temperature 3: 300 °C (held for 10 minutes), the Carrier gas helium was 5 mL/min (Chen *et al*., 2019).

RESULTS AND DISCUSSION

Table (2) and Figures (1,2,3,4,5,6,7,8,9,10,11 and 12) show the content of PAHs in bottled water samples in Iraqi local markets. The highest sample that contained total PAHs ($\sum PAHs_{High}$) is sample 1 (2.21 µg/L), and the lowest sample that contained total PAHs ($\sum PAHs_{Low}$) is sample 5 (0.99 µg/L) These results agree with what was found by Güler, (2007) when studying the PAHs in bottled water from mineral sources, springs and drinking water and you don't agree with what he said (Vega *et al.,* 2011*)*. Fortunately, all samples were free of the following compounds: Benzo (K) Fluoranthenen, Benzo (G, H) Perylene, Benzo (A) Pyrene and Chrysene, it matches what found (Aygun & Bagcevan, 2019) with his study of drinking water sources in Turkey, while Anthracene, Phenanthrene (3 rings) of medium molecular weight and Benzo (A) Anthracene with a high molecular weight (5 rings) are present in 50% or more of the samples, and this is compatible with (Ambade *et al*., 2021) in the fact that these compounds are dominant, but they are not compatible with it because they do not conform to the standard specifications when studying the risks of compounds PAHs in India's drinking water.

Sample 1 contained the highest content of Acenaphthylene and Anthracene, where the concentrations 0.445 and 0.325 µg/L respectively, which is not in accordance with the specifications of the WHO and USEPA (maximum level of 0.2 µg/L), As well as compounds such as Acenaphthene, Benzo(A) Anthracene, Naphthalene, and Phenanthrene, which are not in compliance with the WHO and USEPA specifications And correspond to (Güler, 2007) .

In sample 6, The content of compounds such as Acenaphthene, Acenaphthylene, Anthracene , and benzo (A) Anthracene where the concentrations 0.26,0.22,0.31 and 0.24 µg/L respectively, did not comply with the standard specifications (Sharifiarab *et al*., 2022). Samples 3, 4, 7, 9, and 10 contained three compounds; samples 2, 11, and 12 contained two compounds; and samples 5 and 8 contained one compound, all of which contained these compounds that did not conform to standard specifications. These results do not agree with what was observed by (Awaz *et al*., 2010) and agree with (Güler, 2007; Sharifiarab *et al*., 2022) when they studied PAHs in drinking water and bottled water, respectively.

The presence of the naphthalene compound (2 rings) was observed in most of the study samples, and sample 1 contained the highest concentration (0.275 µg/L) and was also not in conformity with the standard specifications, and this is consistent with what was found Guart *et al.,* (2014) when studying it to evaluate bottled water in 77 samples collected from 27 countries from all over the world to identify 69 organic pollutants It contained thirteen compounds of PAHs. The samples were analyzed using GS-MS technology. It was noted that the samples did not contain PAHs except for naphthalene, as its content ranged between 0.005 – 0.202 µg/L. High levels of contaminants in water can arise from both primary pollution sources (including industrial discharges, agricultural runoff, sewage discharge, oil spills, and chemical waste disposal) and secondary contamination (including water treatment, storage, distribution, and consumer use).

Table (2): Value and types of PAHs compounds in bottled water samples in Iraqi local markets

S. ID/ PAHs (µg/L)	1	2	3	4	5	6	7	8	9	10	11	12
Ac	0.29	Nd	Nd	Nd	0.16	0.26	0.12	Nd	Nd	Nd	Nd	0.06
Ace	0.44	0.11	Nd	0.17	0.07	0.22	0.22	0.16	0.18	0.20	0.12	0.16
A	0.32	0.16	Nd	0.11	0.22	0.31	0.16	0.26	0.22	0.27	0.17	0.12
BaA	0.22	0.26	Nd	0.22	0.12	0.24	0.24	0.10	0.22	0.13	0.11	0.20
BbF	Nd	0.20	Nd	Nd	Nd	0.17	Nd	0.16	Nd	Nd	Nd	Nd
BkF	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
BgP	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
BaP	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
Ch	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
DhA	Nd	Nd	0.16	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
Fl	Nd	Nd	0.33	0.27	Nd	Nd	Nd	Nd	Nd	Nd	Nd	Nd
F	Nd	0.16	0.18	Nd	0.11	0.13	0.16	0.11	Nd	Nd	Nd	Nd
IP	0.18	0.12	0.20	0.22	Nd	0.12	Nd	Nd	0.15	0.20	0.22	Nd
Na	0.27	0.11	0.12	0.16	Nd	Nd	0.12	0.17	0.17	Nd	0.11	0.18
Pa	0.34	0.05	0.22	Nd	0.12	0.16	0.21	0.12	0.20	0.16	0.22	0.22
P	0.12	Nd	0.17	Nd	0.17	0.08	0.16	Nd	Nd	0.12	0.12	0.16

Nd= not detected

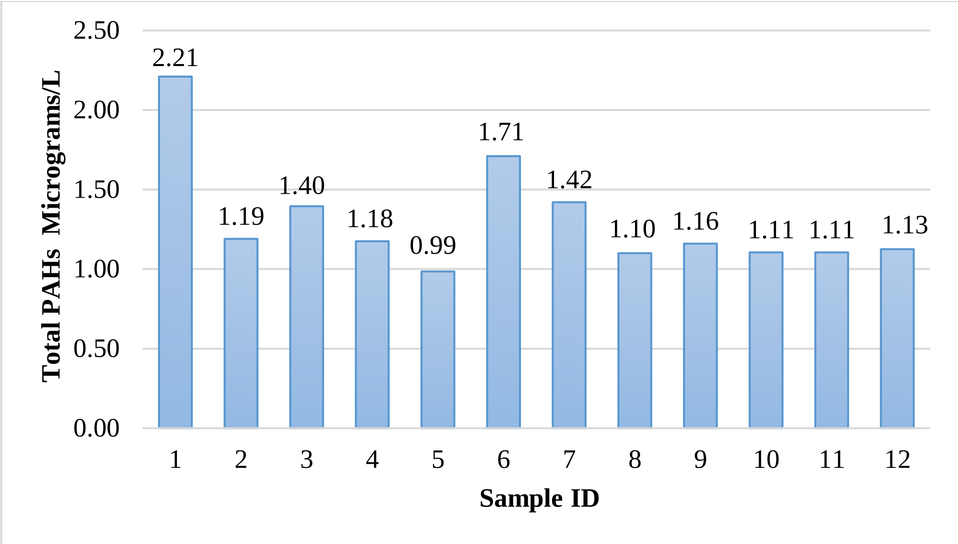

Figure (1): Total content of PAHs in bottled water samples from Iraqi local markets

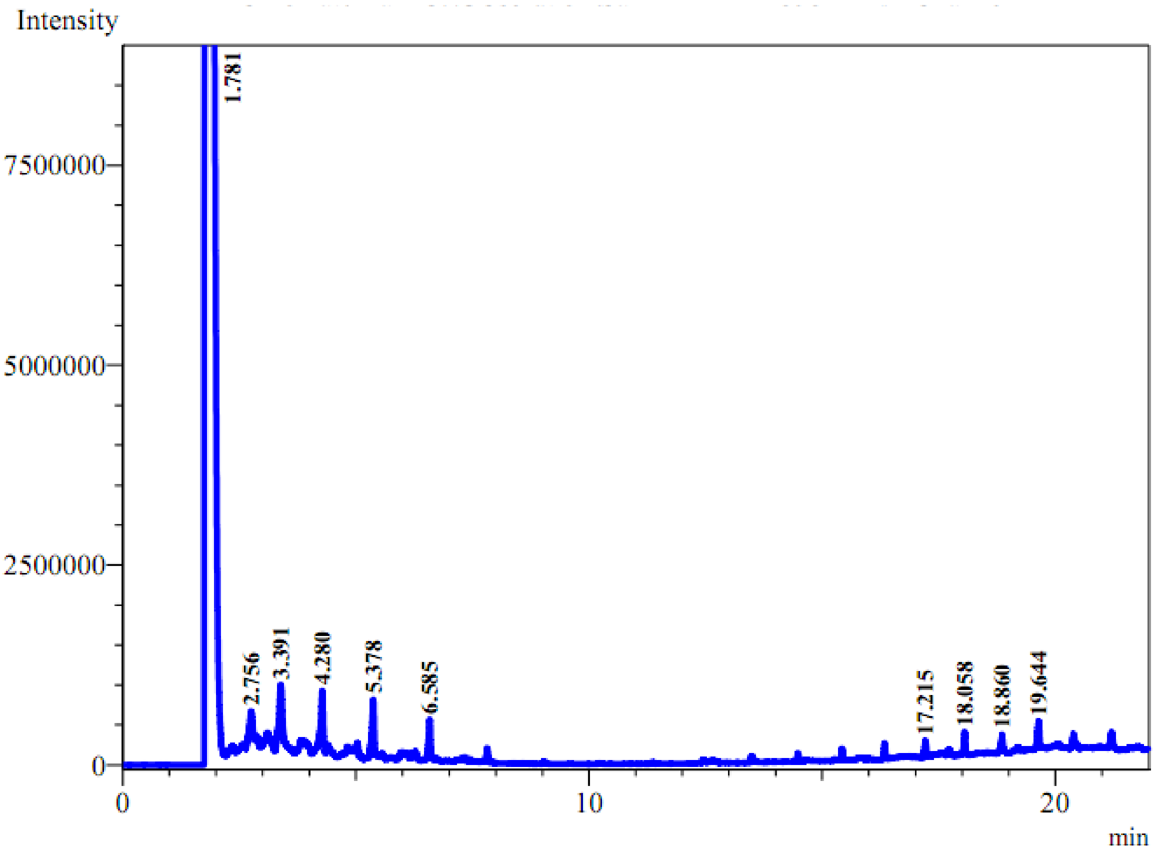

Figure (2): Chromatogram of PAH compounds for sample 1

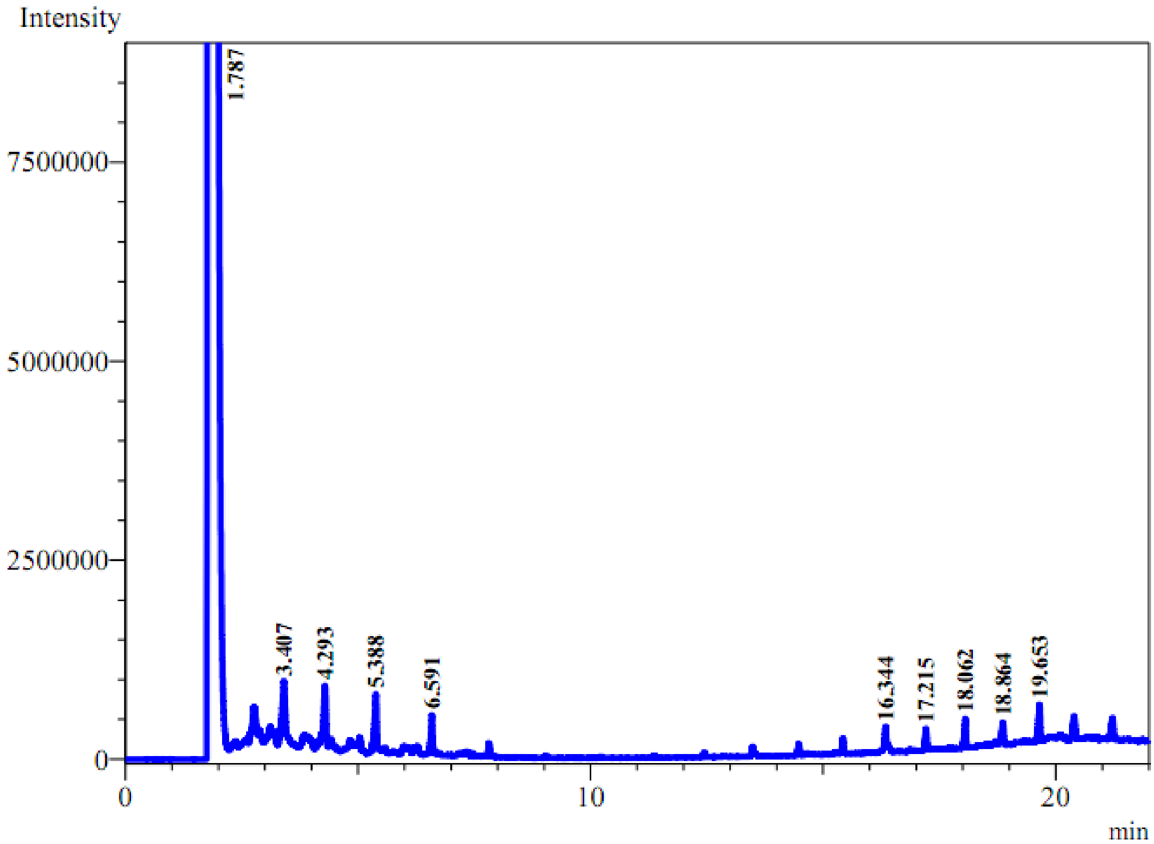

Figure (3): Chromatogram of PAH compounds for sample 2

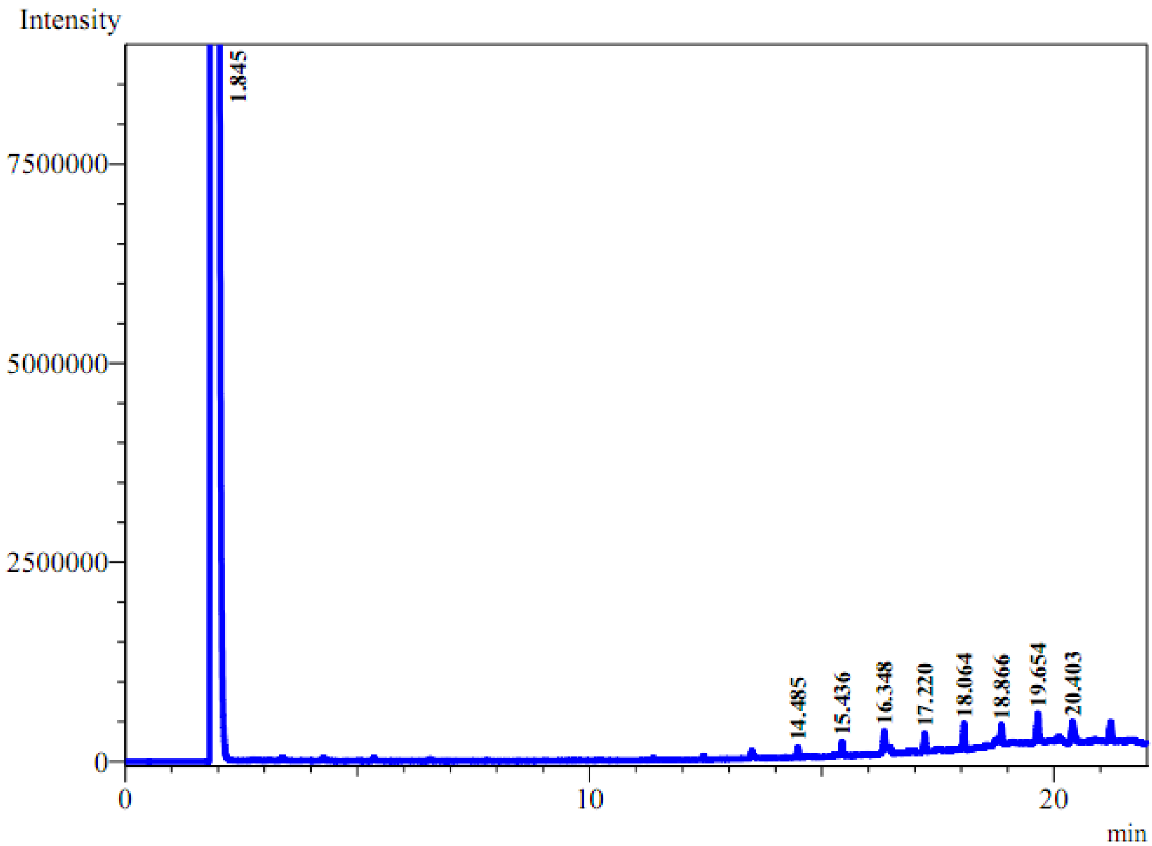

Figure (4): Chromatogram of PAH compounds for sample 3

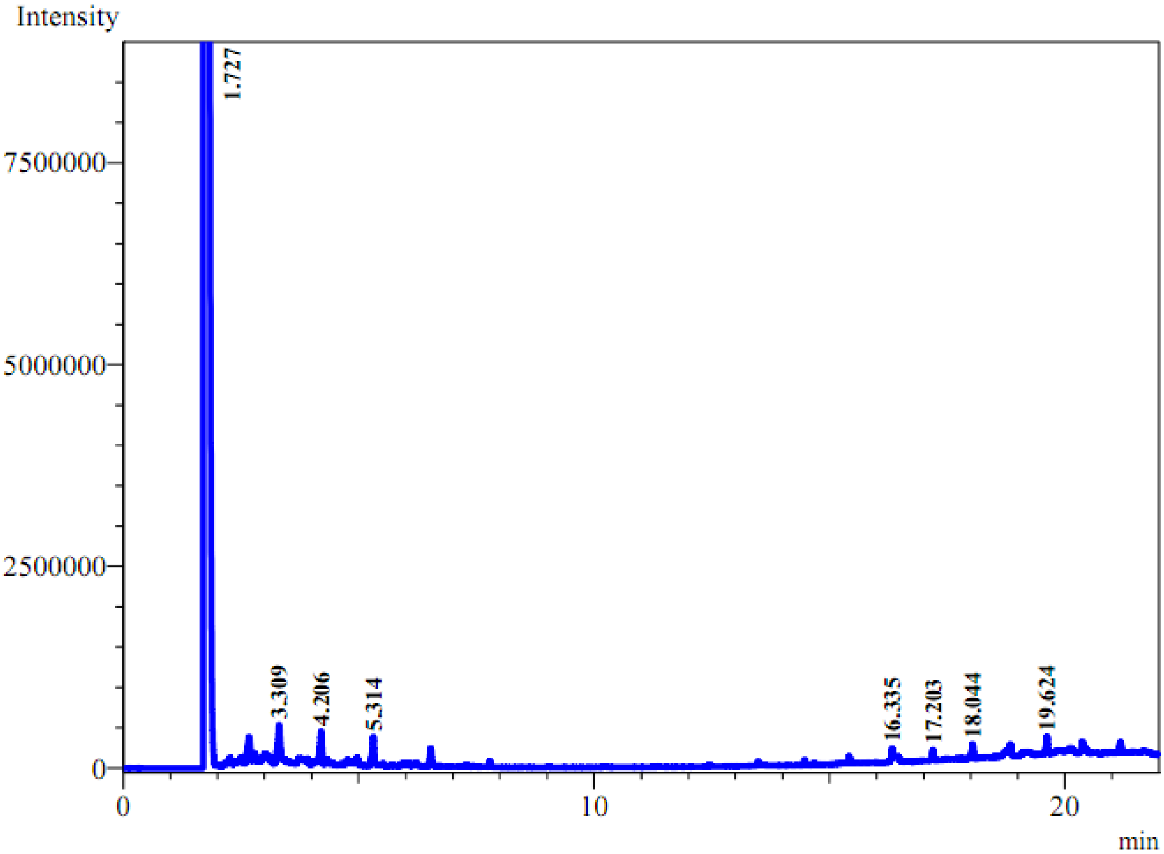

Figure (5): Chromatogram of PAH compounds for sample 4

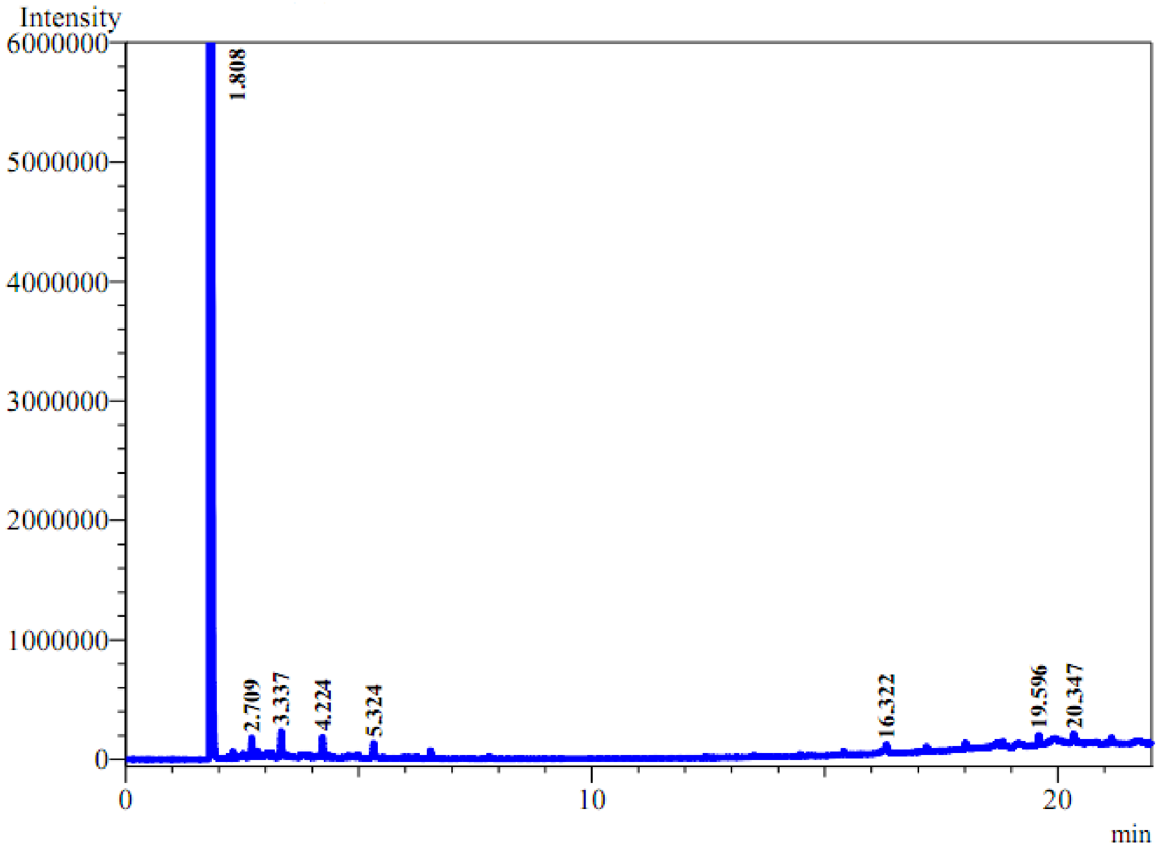

Figure (6): Chromatogram of PAH compounds for sample 5

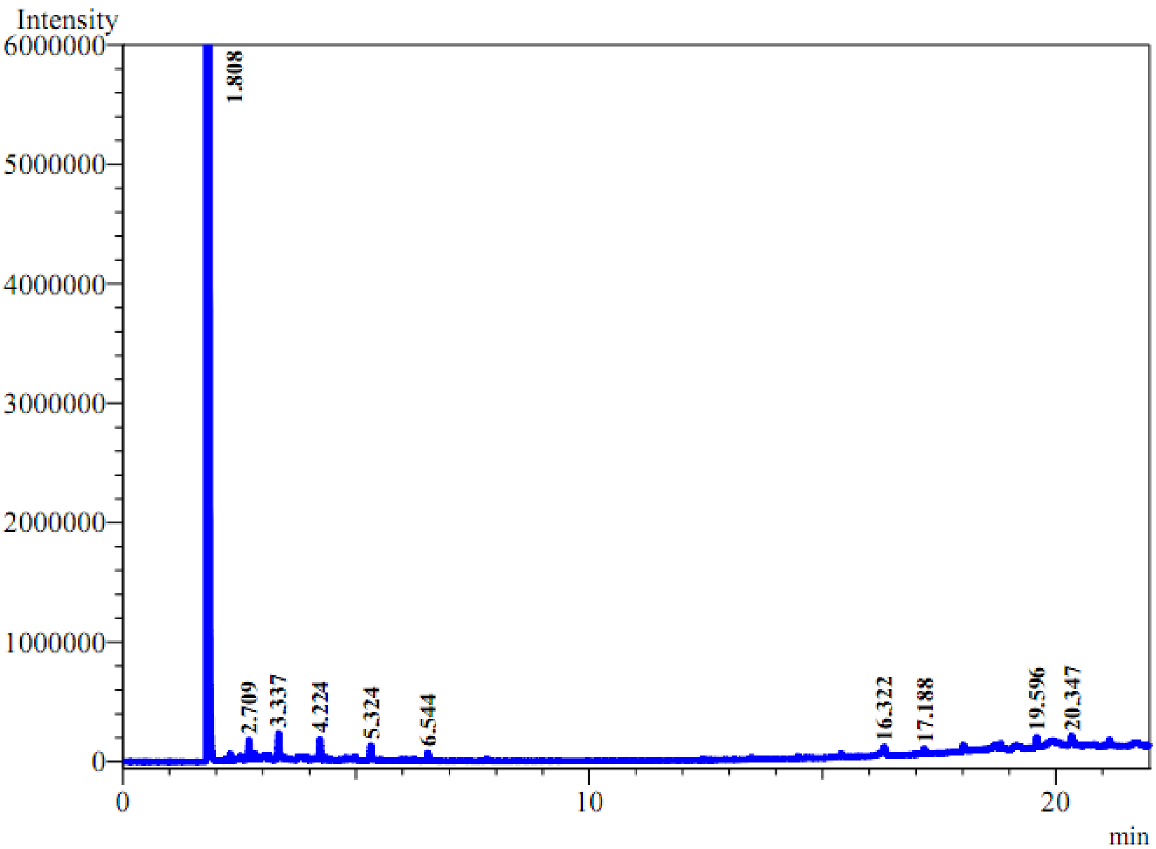

Figure (7): Chromatogram of PAH compounds for sample 6

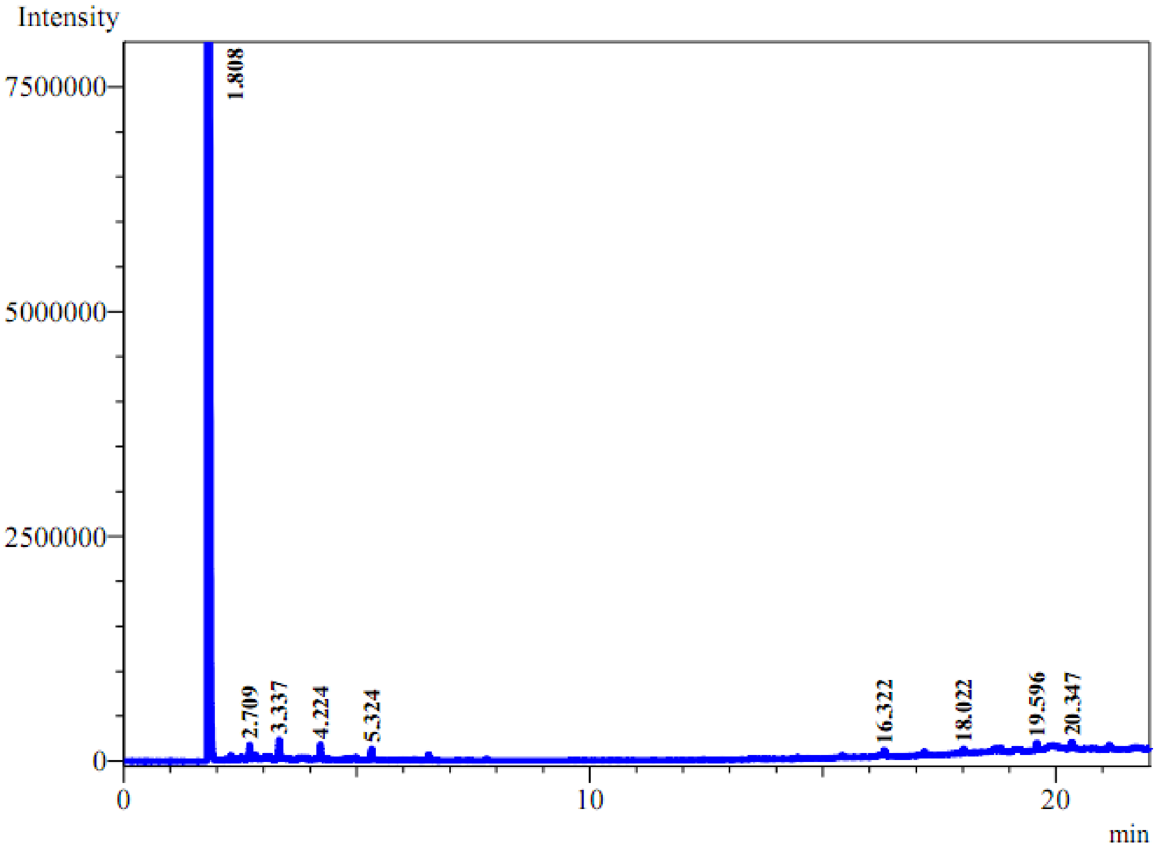

Figure (8): Chromatogram of PAH compounds for sample 7

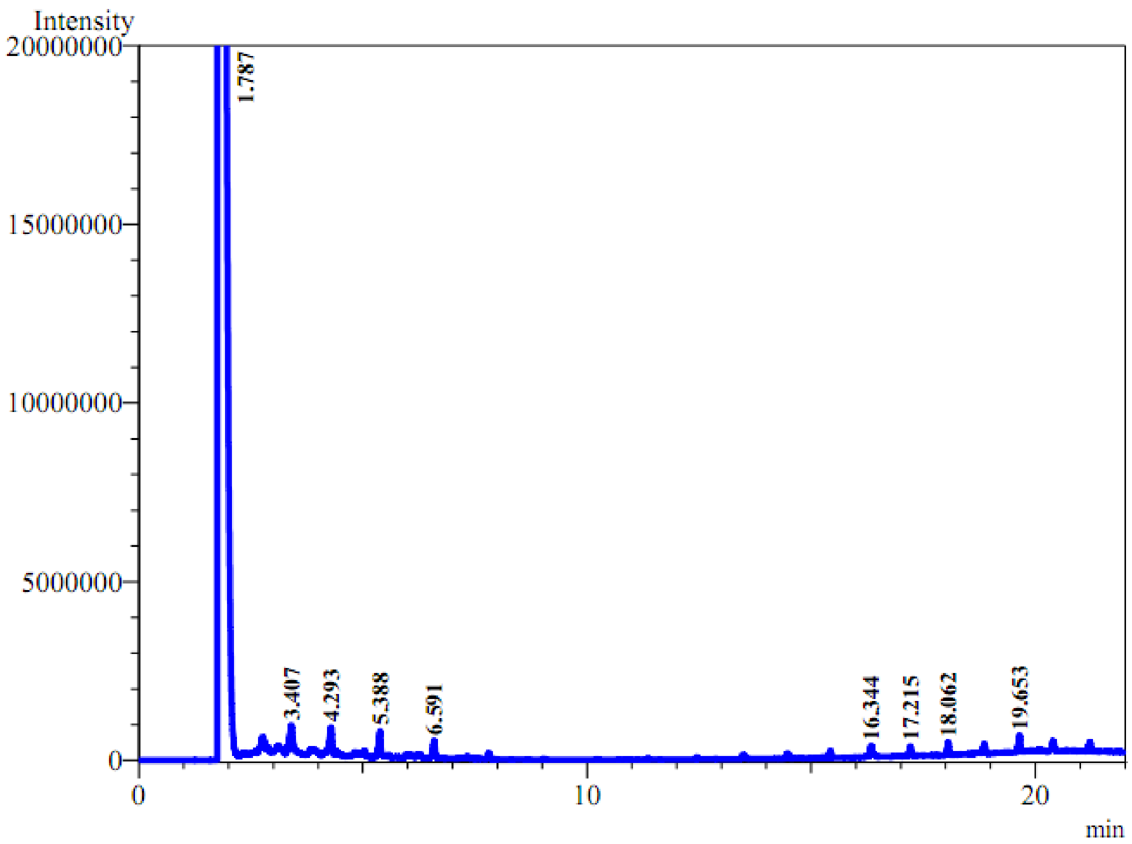

Figure (9): Chromatogram of PAH compounds for sample 8

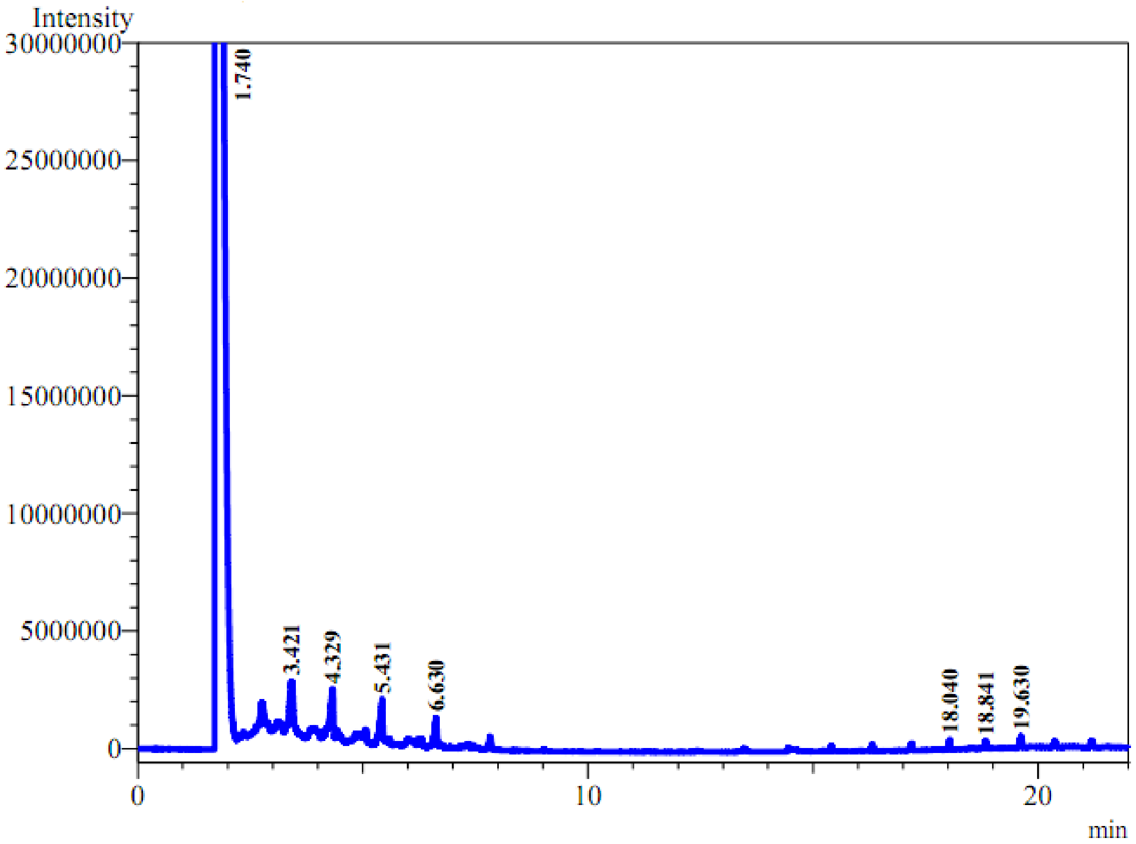

Figure (10): Chromatogram of PAH compounds for sample 9

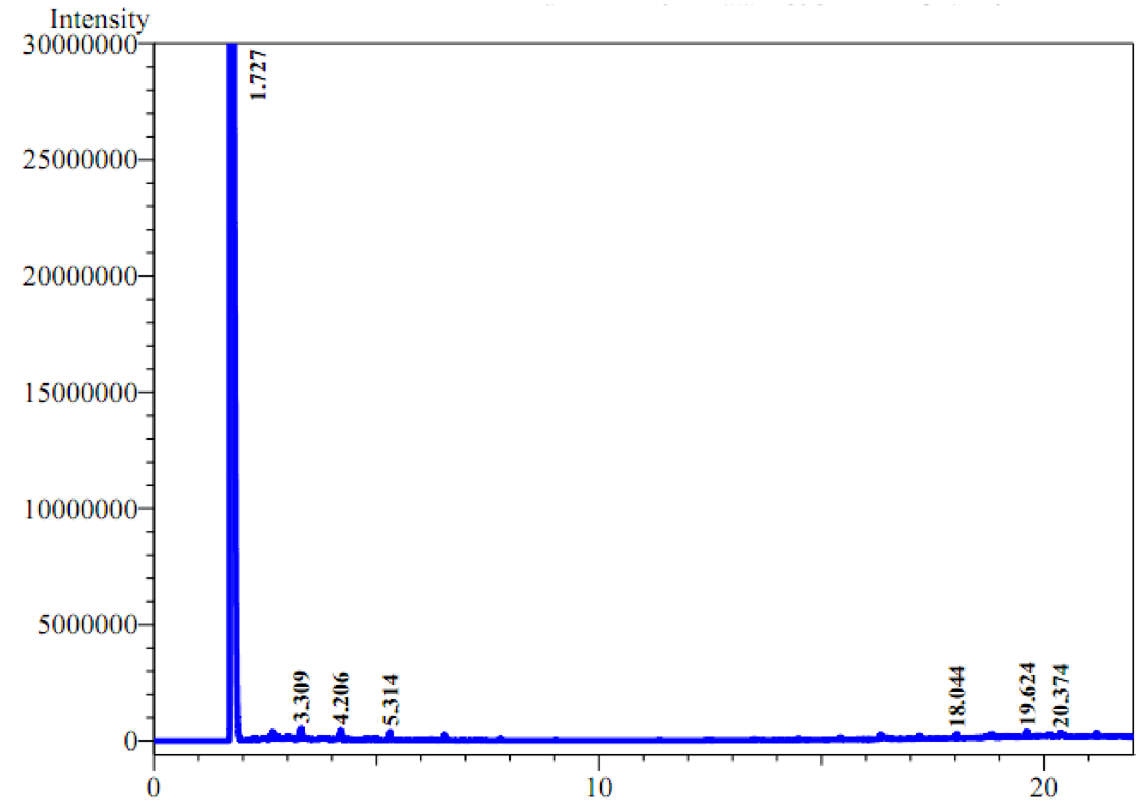

Figure (11): Chromatogram of PAH compounds for sample 10

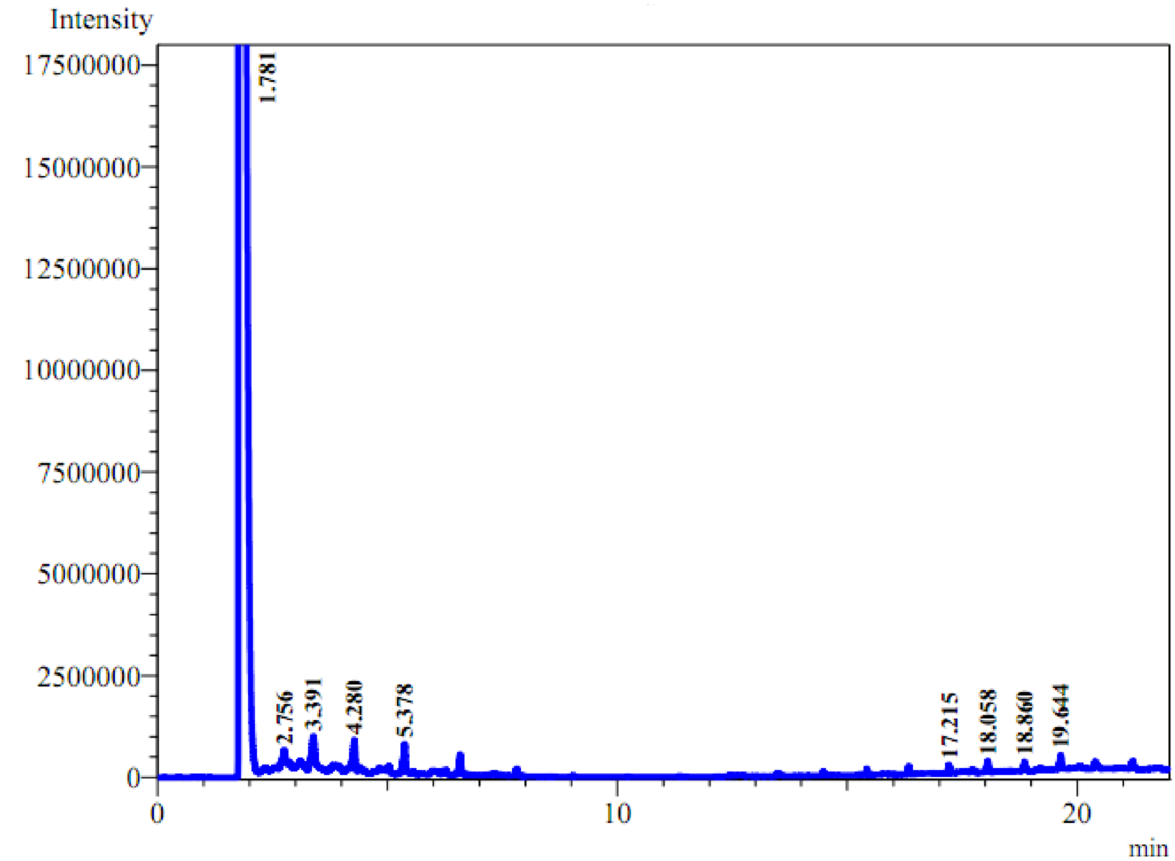

Figure (12): Chromatogram of PAH compounds for sample 11

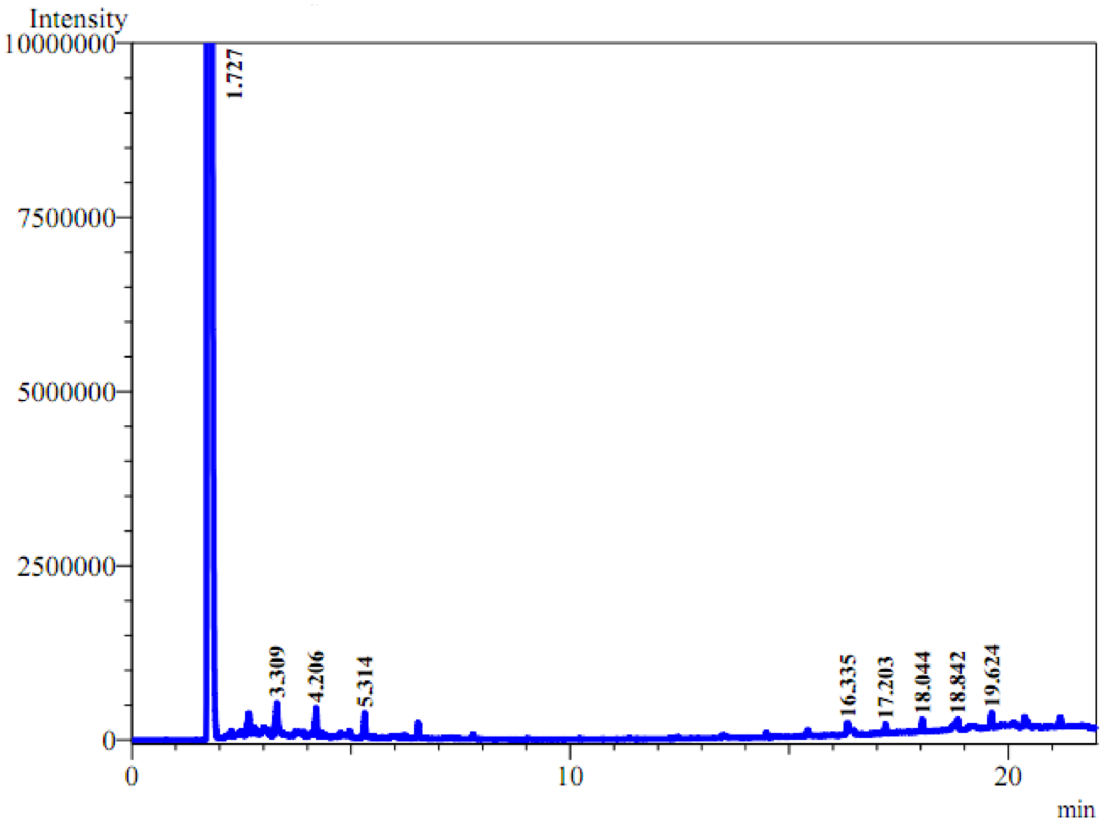

Figure (13): Chromatogram of PAH compounds for sample 12

CONCLUSIONS

We conclude that PAHs present in most samples; sample 1 contained ∑PAHs $_{High}$ the high amount of hydrocarbon pollutants can cause various cancers because they are considered priority pollutants, and sample 5 contained ∑PAHs $_{Low}$ it is the best of brands studied due to its low content of PAHs compounds. The content of tricyclic compounds (3 rings) prevailing in most samples was 31.25%; quaternary compounds (4 rings) was 18.75%; five-cyclic compounds (5 rings) was 12.5%; and hexacyclic compounds were 6.25%. Fortunately, all samples were free of the following compounds: Benzo [G, H] Perylene, Benzo[K] Fluoranthenen, Benzo[a]Pyrene and Chrysene While the compounds Anthracene, Phenanthrene (3 rings), and Benzo (A) Anthracene (5 rings) were present and did not conform to the standard specifications in 50% or more of the samples, the presence of the naphthalene compound (2 rings) was observed in most of the study samples. High levels of pollutants can be due to primary contamination of water (source), and secondary pollution such as contamination of surfaces, tools, bottles during filling, etc.

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CONFLICT OF INTEREST

The authors stated that there are no conflicts of interest with the publication of this work.

تقدير الهايدروكاربونات العطرية متعددة الحلقات للمياه المعبأة المتوفرة في الاسواق المحلية العراقية

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الخلاصة

هدفت الدراسة إلى تقييم نوع ومحتوى 16 مركب من الهيدروكربونات العطرية متعددة الحلقات (PAHs)الملوثة للمياه المعبأة (8 علامات تجارية محلية ، عينة مياه اسالة، 3 علامات تجارية مستوردة) في الأسواق المحلية العراقية باستخدام كروماتوكرافي الغاز وفق معايير وكالة حماية البيئة الأمريكي ومنظمة الصحة العالمية، تم الكشف عن وجود مركبات الهيدروكربونات العطرية متعددة الحلقات في معظم عينات الدراسة، اذ احتوت العينة (1) على أعلى تركيز (∑PAHs) 2.21 ميكروغرام/لتر وأعلى محتوى من الأسينافثيلين والأنثراسين بالتركيزين 0.445 و0.325 ميكروغرام/لتر على التوالي، في حين احتوت العينة (5) على أقل تركيز (∑PAHs) 0.99 ميكروغرام/لتر، كما لوحظ وجود مركب النفثالين (ذو الحلقتين) في معظم عينات الدراسة، الا ان التركيز الأعلى من المركب المذكور لوحظ في العينة (1) واحتوت العلامة اذ بلغ (0.275 ميكروغرام/لتر)، ومحتوى المركبات ثلاثية الحلقات سائد في معظم عينات الدراسة بنسبة 31.25% و المركبات الرباعية الحلقات 18.75% و المركبات الخماسية الحلقات 12.5% و السداسية الحلقات 6.25%. كانت جميع العينات خالية من المركبات التالية: بنزو [K] فلورانثينين و بنزو [G,H] بيريلين و بنزو [a] بيرين و كريسين و ثنائي بنزو [A,H] أنثراسين. بينما كانت مركبات الأنثراسين والفينانثرين (ثلاثية الحلقات) والبنزو [A,H] أنثراسين (خماسية الحلقات) موجودة وغير مطابقة للمواصفات القياسية لوكالة حماية البيئة الأمريكية ومنظمة الصحة العالمية في أكثر من 50% من العينات المدروسة.

الكلمات المفتاحية: ماء الشرب، كروماتوغرافيا الغاز، الزجاجات البلاستيكية، الملوثات.

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