



## Using Toxic Equivalent Quotients (TEQs) to Evaluate the Risk of Polycyclic Aromatic Hydrocarbons Compounds in Soil at Basrah Governorate, Iraq

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**Abstract:** The concentrations of PAHs compounds in this study were estimated in soil of different land uses at Basrah governorate including thirty stations distributed in eight locations (five residential areas, four oil areas, four agricultural areas, five public streets, four petrol stations, two power plants, two public parks and four near private electrical generators). The levels of PAHs compounds mean in all locations ranged from (34.2 - 382.5) ng g<sup>-1</sup> dry weight. The means of carcinogenic PAHs compounds in study area varied from (66.4 - 688.8) ng g<sup>-1</sup> dry weight and mean of non-carcinogenic PAHs compounds varied from (12.8 - 292.5) ng g<sup>-1</sup> dry weight. The carcinogenic PAHs compounds are more dominant than non-carcinogenic PAHs compounds. The human health risk was assessed depending on toxic equivalent quotients (TEQs). The results showed that TEQs ranged from (77.98 - 951.10) ng g<sup>-1</sup> dry weight. The TEQs of PAHs in soil samples of oil areas showed the highest level. In general all locations in Basrah governorate were much polluted with PAHs compounds due to urban and industrial development. The present study represents the first study to assess a human health risk due to exposure to PAHs compounds in soil along Basrah governorate.

**Keywords:** Carcinogenic compounds, Human risk, Iraq, PAHs.

### Introduction

Soil is a critical environment because different pollutants come from various human activities, such as agriculture, industry, mining and transports which can pollute the soil with many dangerous pollutants, one of them is polycyclic aromatic hydrocarbon compounds (PAHs) (Wang *et al.*, 2018). Polycyclic aromatic hydrocarbons are specific persistent organic pollutants that stay for long periods in the environment (Ukalska-Jaruga *et al.*, 2020).

PAHs compounds composed of two or more aromatic rings (Al-Imarah *et al.*, 2017). With increasing number of benzene rings, the molecular weight increases, hence decreases their volatility and biodegradability, whereas increases their toxicity (Al-Rudaini *et al.*, 2019; Abass *et al.*, 2019). Most of high molecular weight PAHs are carcinogenic and mutagenic, while the lowest molecular weight PAHs are severely toxic but non-carcinogenic to many

organisms (Laane *et al.* 2006; Karlsson & Viklander 2008).

Anthropogenic activities are the main source of PAHs that include incomplete combustion of petroleum products, wastes incineration, traffic emissions, house-heating, power plants and other industries (Jiao *et al.*, 2015; Ukalska-Jaruga *et al.*, 2020). Also these compounds come from natural processes such as volcanic activity, decaying organic matter. In addition to plants and algae which can create small amounts of PAHs (Wang *et al.*, 2018).

In general, PAHs occur in an air either bound with suspended particles or as gaseous phase, they may transfer in air for long distances and deposit on water, soil and plants by precipitation processes (Olgun & Doğan, 2020). These ubiquitous pollutants are distributed in environment and transport from soil to groundwater and plants causing human health risk (Cipa *et al.*, 2018).

PAHs compounds are very toxic and characterized for their potential influences of carcinogenicity, mutagenicity and teratogenicity, which are serious to human health due to their tendency to accumulate in the food chain (Cachada *et al.*, 2019). Exposure to these compounds can cause many problems to human including irritation (eyes, skin, and mucous membranes), reduces the function bone marrow, weakness in nervous system and many types of cancers (Moore *et al.*, 2015).

According to U.S. Environmental Protection Agency (US-EPA) sixteen PAHs compounds have been classified as priority pollutants and divided into two groups: carcinogenic and non-carcinogenic to human being. The carcinogenic compounds are seven, including (benzo (a)

anthracene, chrysene, benzo (b) fluoranthene, benzo (k) fluoranthene, benzo (a) pyrene, dibenzo (a) anthracene, and indeno (1,2,3-c,d) pyrene). The remaining nine compounds are non-carcinogenic, including (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene and benzo (g,h,i) perylene) (Kuppusamy *et al.*, 2020).

The route of human exposure to PAHs compounds are through breathing, ingestion and skin contact (Jiang *et al.*, 2016). Health risk estimation has the biggest effects to discover any potential exposure to PAHs compounds that is associated with harmful impacts either for short or long periods of exposure and the periods following the exposure (Kuppusamy *et al.*, 2020). Therefore the present studies focus on PAHs pollutants.

Because of there is a very little previous studies on PAHs compounds in soil at Basrah governorate, wherefore the aims of this study are to determine PAHs compounds in soil in different location at Basrah governorate and assess the human health risk due to exposure to these hazard compounds.

## Materials and Methods

### Study area

The study area is located at Basrah governorate, southern Iraq. Basrah covers an area of (19,070 km<sup>2</sup>) and has a population of (2.532 million people) (Al-Saad *et al.*, 2019). The studied stations are distributed from north to south at Basrah governorate, divided into thirty stations distributed in eight different land uses that included (five residential areas, four

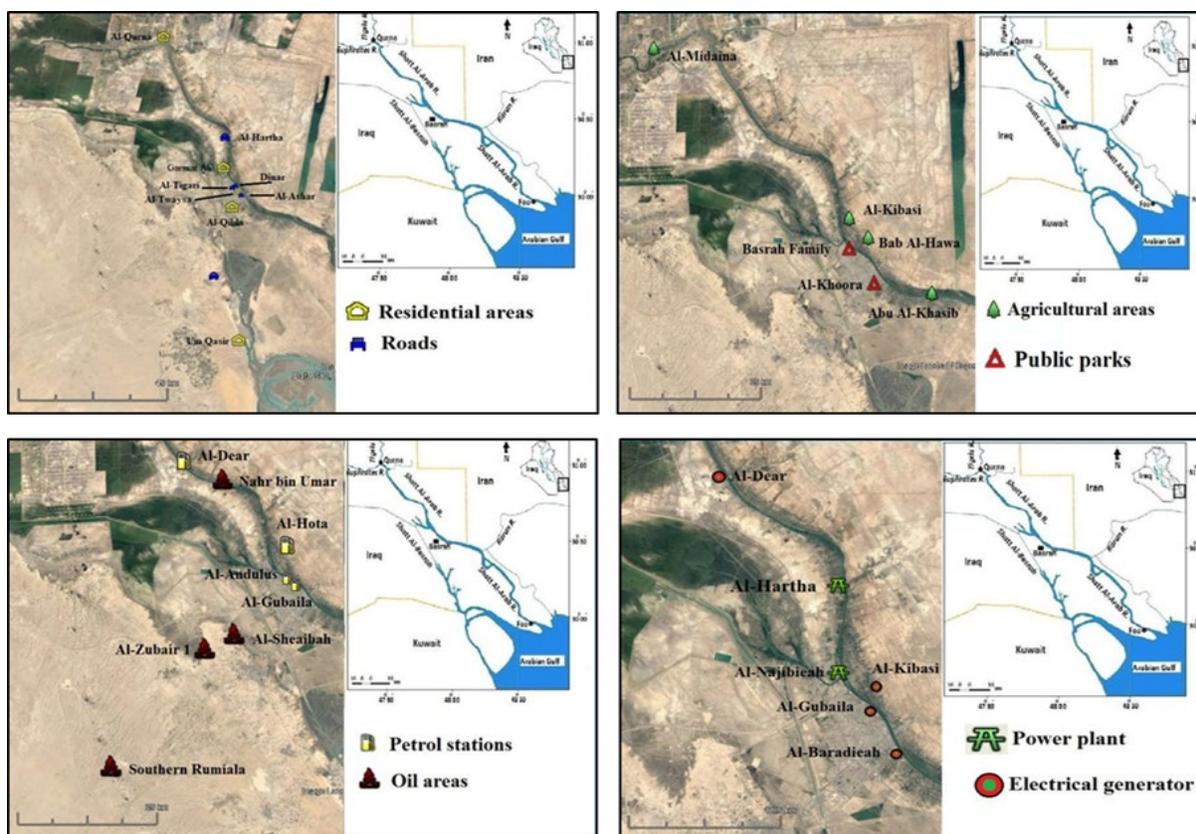


Fig. (1): Map of the study area.

oil areas, four agricultural areas, five public streets, four petrol stations, two power plants, two public parks and four near private electrical generators) as shown in fig. 1.

### Soil sampling and PAHs analysis

The composite soil samples were taken during two seasons the dry season (from July to October 2019) and wet season (from December 2019 to March 2020) from topsoil in depth 0-15 cm using stainless steel shovels. The soil samples taken from 3-5 random sites within each station and mix together as a complex sample and placed in aluminum foil wraps, then the soil samples air-dried at room temperature and sieved through 2 mm mesh sieve.

Dried soil samples were grinded well using a mechanical mortar, sieved through 63  $\mu$ m and stored in glasses vials until extracted PAHs

compounds according to (Goutx & Saliot, 1980). Twenty grams of soil were extracted in soxhlet intermittent extraction using thimble and mixed organic solvents (100 ml) methanol: benzene (1:1 v/v) in rounding flask for 48 hrs. at temperature below 40C°. The saponification process has been done on combined extracts using (15ml) 4M MeOH(KOH) for 2 hrs. at temperature which cannot be exceeded 40C°, then cooled at room temperature. The unsaponification part extracted in a separator funnel with n-hexan (50 ml). The upper unsaponification part (hydrocarbons) with hexane was passed through glass column with length 20 cm. (the bottom packed with glass wool, then about 10 g deactivated silica gel (100-200 mesh), 10 g deactivated alumina (100-200 mesh) , and 5g anhydrous sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) at the top ). The aliphatic parts

were eluted from the column with n- hexane (40 ml), while the aromatics were eluted with benzene (40 ml). The samples were air dried and stored until detection with a capillary Gas Chromatography and Gas Chromatography-Mass (GC-Mass) to measure PAHs compounds.

To estimate the quantities and qualities of PAHs compounds in extracted soil samples, standard polycyclic aromatic compounds were employed to be injected in Gas Chromatography instrument (GC) in Nahr bin Omar laboratories – Basrah Oil Company. GC was made in USA model (Agilent/ USA 7890A). Helium gas used as a carrier gas in Gas Chromatography with flow rate 1 ml /min using flame ionization detector (FID). The injector temperature was 300 C° with split mode ratio 50:1 and detector temperature 300 C°. Column model Agilent HP-1 methyl silicon with dimensions (30 m.\*320 µm \*0.25 µm) was used for aromatic compounds separation. Oven initial temperature was 120 C° was hold time 1 min, temperature rate graduated from 6 C° min<sup>-1</sup> to 300 C° hold time was 11 min.

### Health Risk Assessment

PAHs compounds are characterized by toxicity and carcinogenicity to humans, animals and

plants. To determine the carcinogenicity possibility associated with exposure to individual and total PAH compounds, the (TEQs) was calculated depending on the toxic equivalent factor (TEF) of each individual PAH. The (TEQs) was calculated using the following equation (Qiao *et al.*, 2006; Al-Saad *et al.*, 2019; Cao *et al.*, 2019; Li *et al.*, 2020):

$$\text{TEQs} = \Sigma(\text{TEFi} \times \text{CPAHi})$$

Where:

TEFi: toxic equivalent factor of each PAH compound as showed in table (1) according to (Qiao *et al.*, 2006; Al-Saad *et al.*, 2019).

CPAHi: concentration of individual PAH compound

### Statistical Analysis

Minitab ver.19 software program was used to analysis data through Analysis Of Variance (ANOVA) test. Also, Relative Least Significant Differences (RLSD) was calculated to identify the existence of significant variations between the mean concentrations of PAHs in soil samples.

**Table (1): Toxic equivalent factor (TEF) of each PAH compound.**

PAHs compounds	TEF	PAHs compounds	TEF
Naphthalene	0.001	Benzo(A)Anthrac	0.1
Acenaphthylene	0.001	Chrysene	0.01
Acenaphthnen	0.001	Benzo(B) Fluora	0.1
Fluorene	0.001	Benzo(K) Fluora	0.1
Phenanthrene	0.001	Benzo(A) Pyrene	1
Anthracene	0.01	Indeno(1,2,3-Cd)Pyrene	0.1
Fluoranthene	0.001	Benzo(G,H,I)Perylen	0.01
Pyrene	0.001		

**Table (2): Mean concentration (ng g<sup>-1</sup> dry weight) of PAHs compounds classified as carcinogenic and non-carcinogenic in soil at Basrah governorate.**

Location	Mean PAHs $\pm$ SD	Mean carcinogenic PAHs $\pm$ SD	Mean non-carcinogenic PAHs $\pm$ SD
Residential areas (n=5)	57.9 $\pm$ 65.0 <sup>b</sup>	79.2 $\pm$ 65.1 <sup>cd</sup>	43.7 $\pm$ 64.5 <sup>b</sup>
Oil areas (n=4)	382.5 $\pm$ 352.2 <sup>a</sup>	688.8 $\pm$ 318.1 <sup>a</sup>	178.3 $\pm$ 191.2 <sup>ab</sup>
Agricultural areas (n=4)	34.2 $\pm$ 42.5 <sup>b</sup>	66.4 $\pm$ 44.5 <sup>d</sup>	12.8 $\pm$ 25.2 <sup>b</sup>
Roads (n=5)	129.5 $\pm$ 155.0 <sup>b</sup>	235.5 $\pm$ 188.4 <sup>bcd</sup>	58.8 $\pm$ 76.1 <sup>b</sup>
Petrol stations (n=4)	148.4 $\pm$ 186.6 <sup>ab</sup>	244.5 $\pm$ 208.9 <sup>bcd</sup>	84.4 $\pm$ 148.8 <sup>b</sup>
Power plant (n=2)	221.8 $\pm$ 221.1 <sup>ab</sup>	330.8 $\pm$ 196.7 <sup>b</sup>	149.1 $\pm$ 215.7 <sup>ab</sup>
Public parks (n=2)	71.6 $\pm$ 113.6 <sup>b</sup>	149.2 $\pm$ 144.3 <sup>bcd</sup>	19.8 $\pm$ 45.2 <sup>b</sup>
Electrical generator (n=4)	292.0 $\pm$ 270.8 <sup>ab</sup>	291.3 $\pm$ 98.7 <sup>bc</sup>	292.5 $\pm$ 349.6 <sup>a</sup>
RLSD	237.6	218.74	178.85

The letters (a, b, c and d) according to RLSD value. The different letters refer to significant differences, while the same letters refer to non-significant differences.

## Results & Discussion

Statistical analysis (ANOVA - one way) showed significant differences ( $P < 0.01$ ) among mean concentrations of total PAHs compounds among locations at Basrah governorate table (2).

Oil area, electrical generator, power plant and petrol stations showed non-significant differences according to Relative Least Significant Differences (RLSD = 237.6,  $P = 0.05$ ). Also, non-significant differences were found among electrical generator, power plant, petrol stations, roads, public parks residential areas and agricultural areas, while there were significant differences between oil area and the remaining locations.

The lowest mean concentration was 34.2 ng g<sup>-1</sup> dry weight in agricultural areas, whereas the highest mean concentration was 382.5 ng g<sup>-1</sup> dry weight in oil areas. The sequences of mean PAHs compounds in the studied area followed this order: Oil areas > electrical generator >

power plant > petrol stations > roads > public parks > residential areas > agricultural areas.

Anthropogenic activities are the main source of PAHs in soil that includes pyrogenic sources from oil products and vehicle emissions (Liu *et al.*, 2019). The increase of oil products cause to soil structure disorder, Its lead to lack of organic matter contents, soil minerals, soil nutrients, soil fertility, and bad crop yield, It also lead to leaching and erosion of soil (Palese *et al.*, 2003; Nwaichi *et al.*, 2014).

The percentage of individual PAHs compounds at all locations at Basrah governorate, Fig. (2) showed that the most dominant PAHs in (residential areas, oil areas, agricultural areas, roads, petrol station, power plants, public parks and electrical generators) were (BghiP 20%, Chy 20%, BkF 29%, BbF 28 %, BkF 29%, Chy 19%, BbF 36 % and Pyr 23%) respectively.

The PAHs compounds are divided into groups depending on number of benzene rings (Cipa *et al.*, 2018), including two benzene rings

(Nap), three benzene rings (Acy, Ace, Flu, Phe, and Ant), four benzene rings (Fla Pyr, BaA, Chr), five benzene rings (BbF, BkF and BaP) and six rings ( InP, and Bghi).

The dominant PAHs compounds in all soil samples were 3–5 rings, this indicates that the PAHs sources originated from industrial activities and heavy traffic emission due to rapid industrial and economic growth (Kim *et al.*, 2019). Results showed that low molecular PAHs (two rings) have low concentrations due to their availability to biodegradability by microorganisms more than high molecular PAHs. Also low molecular PAHs were decreased gradually by dilution due to their high-water solubility (Cipa *et al.*, 2018), while The PAHs compounds with five or more rings have low solubility and volatility, so they are found as solid state and bond to soil particles (Han *et al.*, 2011). Therefore, they are less degradable by microorganisms and this causes that PAHs compounds are more persistent in the environment and stay for long periods (Kim *et al.*, 2019). The present results were in agreement with other reports. (Al-Saad *et al.*, 2017; Al-Saad *et al.*, 2019).

The mean concentration of carcinogenic PAHs compounds in the study area varied from (66.4) at agricultural areas to (688.8) ng g<sup>-1</sup> dry weight at oil areas. Statistical analysis (ANOVA one way) showed significant

differences (P<0.01) among locations of carcinogenic PAHs compounds.

Statistical analysis (RLSD) showed there were significant differences at (P<0.01) of carcinogenic PAHs compounds between oil areas and the remaining locations. The concentrations of carcinogenic PAHs compounds in the study area followed this order: Oil areas > power plant > electrical generator > petrol stations > roads > public parks > residential areas > agricultural areas.

The mean concentration of non-carcinogenic PAHs compounds varied from (12.8) ng g<sup>-1</sup> dry weight at agricultural areas to (292.5) ng g<sup>-1</sup> dry weight at electrical generators. Statistical analysis (ANOVA one way) showed there were significant differences at (P<0.05) among locations of non-carcinogenic PAHs compounds.

Statistical analysis (RLSD) showed there were no significant differences at (P<0.05) of non-carcinogenic PAHs compounds among electrical generators, power plants and oil areas and there were no significant differences at (P<0.05) of non-carcinogenic PAHs compounds among power plants, oil areas, agricultural areas, petrol stations, roads, residential areas, public parks; whereas electrical generators had significant differences with the remaining locations.

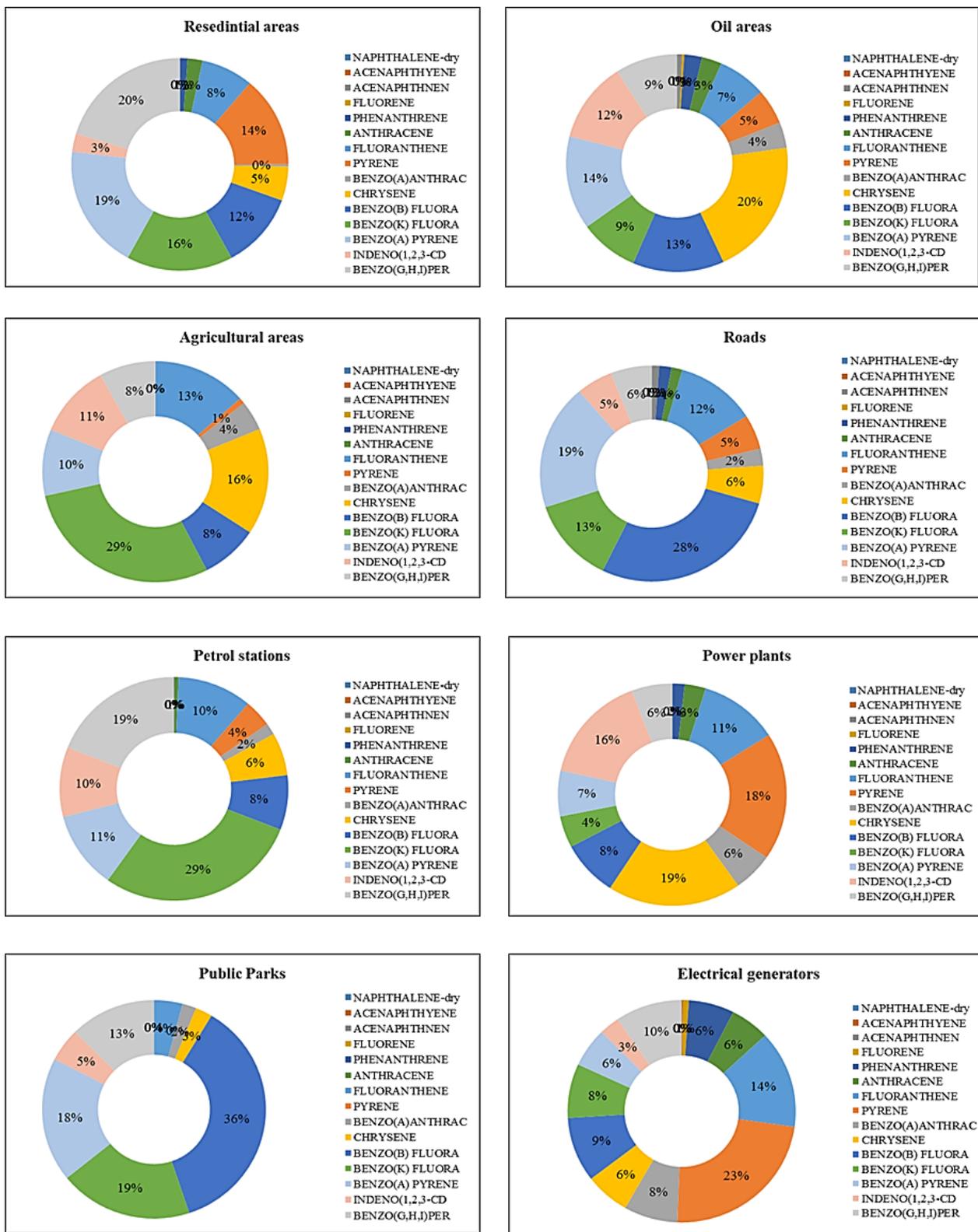


Fig. (2): The percentage of individual PAHs compounds in all locations at Basrah governorate during the study period.

The concentrations of non-carcinogenic PAHs compounds in the study area followed this order: Electrical generator > oil areas > power plant > petrol stations > roads > residential areas > public parks > agricultural areas. In general, all locations at Basrah governorate were very polluted with PAHs compounds and the concentration of these compounds was very high compared with previous studies except the study of (Khwedim, 2016), table (3). This is

due to oil refineries that emit PAHs, as well as combustion of gasoline in vehicles, power plant stations, petrol stations, private electrical generators and wastes incinerations. On the other hand, the burning of tyres in many roads, near oil companies and near Um Qasser port in Basrah governorate during demonstrations in October, 2019 revolution that added large quantities of hydrocarbons to soil.

**Table (3): Comparison the current concentrations (ng g<sup>-1</sup> dry weight) of surface soil PAHs compounds with previous studies at Basrah governorate.**

Study	Study area	PAHs compounds
Khwedim, 2016	Rumaila Oil Field	6600 - 4749300
Al-Saad <i>et al.</i> , 2019	Shatt Al-Arab River Delta	17.30 - 72.16
Kadhim <i>et al.</i> , 2019	West Qurna-1 Oil Field	1.335 - 3.592
Jalal, 2020	Basrah governorate	0.6 - 112.57
Current study	Basrah governorate	34.2 - 382.5

All these sources cause severe pollution with hydrocarbons which have harmful impacts on humans and environment. The carcinogenic PAHs compounds (BaA, Chy, BbF, BkF, BaP and InP) are more dominant than non-carcinogenic PAHs compounds (Nap, Acy, Ace, Flu, Phe, Ant, Fla. Pyr and BghiP) Fig. 2. The percentage of carcinogenic PAHs in

Basrah soils were very high compared with non-carcinogenic PAHs in all locations except the electrical generators in which the percentage of carcinogenic and non-

carcinogenic is equal, table (4). This indicates that the carcinogenic PAHs in the study area were the main contributor to total TEQ and this in agreement with Cao *et al.* (2019).

#### Health risk assessment

The results of TEQs in present study were ranged from 77.98 ng g<sup>-1</sup> dry weight in agricultural areas to 951.10 ng g<sup>-1</sup> in oil areas with mean 380.32 ng g<sup>-1</sup> dry weight, table (5).

**Table (4): The percentage of PAHs compounds as carcinogenic and non-carcinogenic in all locations at Basrah governorate**

Location	Non-carcinogenic PAHs	Carcinogenic PAHs
Residential areas	35 %	65 %
Oil areas	20 %	80 %
Agricultural areas	16 %	84 %
Roads	22 %	78 %
Petrol stations	26 %	74 %
Power plant	23 %	77 %
Public Parks	15 %	85 %
Electrical generator	50 %	50 %

**Table (5): Values of TEQs in all locations at Basrah governorate.**

Location	TEQs (ng g <sup>-1</sup> )
Residential areas	193.81
Oil areas	951.10
Agricultural areas	77.98
Roads	462.44
Petrol stations	362.17
Power plant	344.78
Public parks	266.16
Electrical generator	384.14
Mean	380.32
Min	77.98
Max	951.10
SD	260.05

**Table (6): Comparison the TEQs (ng g<sup>-1</sup>) in current study with previous studies in the world.**

Study	Study area	TEQs (ng g <sup>-1</sup> dry weight)
Jio <i>et al.</i> , 2015	Oilfield in China	13.3 - 4397
Moore <i>et al.</i> , 2015	Urban areas in Iran (Isfahan)	1- 900.53
Al-Saad <i>et al.</i> , 2019	Shatt Al-Arab delta –Basrah	0.523 - 5.834
Cao <i>et al.</i> , 2019	Coking plant in China	39.4 - 559.5
Cachada <i>et al.</i> , 2019	Urban areas in Lisbon (Portugal)	0.59 - 7653
Li <i>et al.</i> , 2020	petrol station in China	6.41 - 72.54
Current study	Basrah governorate	77.98 - 951.10

The TEQs of PAHs in soil samples of oil areas exceeded the safe level (600 ng g<sup>-1</sup>) according to the criterion risk of soil for human health protection (Wang *et al.*, 2018), whereas the

remain regions the TEQs were lower than 600 ng. g<sup>-1</sup>. This indicates that oil areas have high levels of carcinogenic dangerous compounds that harm workers and humans in surrounding

areas. In comparison the TEQs in current study with previous studies in the world (table 6). The concentration of TEQs in study area was very high compared with the study of Al-Saad *et al.* (2019), Cao *et al.* (2019) and Li *et al.* (2020). It was lower than the study of Jio *et al.* (2015) and Cachada *et al.* (2019), and it was near to the study of Moore *et al.* (2015).

## Conclusion

In this study the levels of PAHs compounds were very high compared with previous studies and the total of carcinogenic PAHs compounds was higher than non-carcinogenic PAHs compounds in all locations except the electrical generators. All studied stations were under the safe level of risk except oil areas were higher than the safe level. In general, Basrah governorate suffers from pollution with PAHs compounds due to increasing combustion processes and vehicle emissions. To confront the dangerous environmental issues, this study could present new views for the management of soil risk of different land uses in Basrah governorate contaminated with PAHs compounds or other organic pollutants.

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## Conflicts of interest

The authors declare that they have no conflict of interests.

## Contributions of Authors

**F.M.S.** : Collected the samples, samples preparation, chemical analysis and final draft writing.

**H.T.A.** : Laboratory work, data preparation and final draft editing submission.

**M.M.A.** : Field work sampling, samples preparation and statistical analysis.

All authors discussed the results and contributed to the final manuscript.

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## استخدام حاصل السمية المكافئة (TEQs) لتقييم مخاطر المركبات الهيدروكربونية الأروماتية المتعددة الحلقات في ترب محافظة البصرة، العراق

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**المستخلص:** المركبات الهيدروكربونية العطرية متعددة الحلقات (PAHs) هي ملوثات عضوية معقدة تأتي بشكل رئيسي من الأنشطة البشرية. في هذه الدراسة تم تقدير تراكيز مركبات الهيدروكربونات العطرية متعددة الحلقات في التربة من مناطق مختلفة الاستعمالات في محافظة البصرة شملت ثلاثين محطة موزعة على ثمانية مواقع (خمسة مناطق سكنية، أربع مناطق نفطية، أربع مناطق زراعية، خمسة شوارع عامة، أربع محطات وقود، محطتان لتوليد الطاقة الكهربائية، اثنتين من الحدائق العامة وأربعة مناطق بالقرب من المولدات الكهربائية الأهلية). تراوحت مستويات المركبات الهيدروكربونية العطرية متعددة الحلقات الكلية في جميع المواقع من (34.2-382.5) نانوغرام.غم<sup>-1</sup> وزن جاف. تراوحت المركبات الهيدروكربونية العطرية متعددة الحلقات المسرطنة في منطقة الدراسة من (66.4-688.8) نانوغرام.غم<sup>-1</sup> وزن جاف، والمركبات الهيدروكربونية العطرية متعددة الحلقات غير المسرطنة تتراوح بين (12.8-292.5) نانوغرام - 1 وزن جاف. المركبات الهيدروكربونية العطرية متعددة الحلقات المسرطنة (BaA و Chy و BbF و BkF و BaP و InP) هي الأكثر انتشارًا من المركبات الهيدروكربونية العطرية متعددة الحلقات غير المسرطنة (Acy و Nap و Ace و Flu و Phe و Ant و Fla. Pyr و BghiP). تم تقييم المخاطر على صحة الإنسان اعتمادًا على حاصل السمية المكافئة (TEQs). وأظهرت النتائج أن معادلات السمية المكافئة تراوحت بين (77.98 - 951.10) نانوغرام.غم<sup>-1</sup>. أظهرت معادلات السمية الشاملة للهيدروكربونات العطرية متعددة الحلقات في عينات التربة من مناطق النفط المستوى الأعلى. بشكل عام، كانت جميع المواقع في محافظة البصرة شديدة التلوث بمركبات الهيدروكربونات العطرية متعددة الحلقات بسبب التطور العمراني والصناعي. تمثل الدراسة الحالية أول دراسة لتقييم المخاطر على صحة الإنسان بسبب التعرض لمركبات الهيدروكربونات العطرية متعددة الحلقات في التربة على طول محافظة البصرة.

**الكلمات المفتاحية:** المركبات المسرطنة، الخطر على الإنسان، العراق، المركبات الأروماتية.