



Comparison of the Polycyclic Aromatic Hydrocarbons and Heavy Metal Concentrations in Air for the July 2012 and Oct.2013 at Baiji District, Iraq

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ABSTRACT

The total suspended particles (TSP) were measured at selected locations in two periods July 2012 and Oct. 2013 at area near the Baiji Oil Refinery. The heavy metals (Co, Ni, Pb, Zn, Cr, and Cd) and polycyclic aromatic hydrocarbons (PAHs) concentrations in aerosol from air samples were determined. Aerosol samples were collected, total of 34 samples (17 samples for both sampling periods) were analyzed by GC-MS in order to determine 16 PAH compounds. The results of average concentration of suspended particles (TSP) are higher than the permissible limits of the Iraqi National and the world limits at the both periods, July 2012 (2714.1 $\mu\text{g}/\text{m}^3$) and October 2013 (1124.8 $\mu\text{g}/\text{m}^3$). Comparison of the averages of the heavy metals (Co, Ni, Pb, Zn, Cr, and Cd) with the national and world limits, noticed that they are lower than these limits. Except for the concentration of Cr and Cd at the two periods and Pb in July only, were higher than the world limits. Naphthalene was the most abundant PAH detected and there is strong correlation between total suspended particles (TSP) and total polycyclic aromatic hydrocarbons (TPAH) in all the sampling sites for both sampling periods. The variation between the two periods indicates the direction, speed and the intensity of the sand dust storms effect during the months of the research. Consequently, the total TSP, heavy metals and polycyclic aromatic hydrocarbons (PAHs) concentrations in aerosol concentrations for both sampling periods showed an increase at the direction away from the Baiji Oil Refinery mostly at the south and east directions depending on wind direction.

Keywords: TSP, Heavy Metals, PAH, Baiji Oil Refinery, Baiji, Iraq.

1. INTRODUCTION

The main source of air pollution is man-made sources that include automobiles, power generation and the industrial activities, especially oil industry activities using huge amount of consumable fuel like power plants and oil refineries; due the high rate emission of fume, solid particulates and toxic gases in quantity more than every other industry. These industries will be more hazardous upon its existence inside the limits of the cities, or its existence inside urban area (Masitah, et al., 2007). Suspended solids particle, which are air pollutants stuck in the air with a small volume ranges between (200-0.01) Micron, studying those particles is important because of their long survival periods in the atmosphere, unlike the other big particles which will settle down more rapidly. In addition to that smaller particles seem to interact with other air pollutants, leading to severe damages (WHO, 2000, Al-Dabbas , et al., 2015). Suspended solids are important pollutions which consist of suspended minerals and heavy metals. The sources of heavy metals in air may be natural (soils, and transported sediments by winds), or artificial include industrial sources that supply the heavy metals to the air and cause contamination of the atmosphere.

The trace elements (Pb, Zn, Cu, Ni, Cr, and Cd) are designated as priority pollutants by many researchers and are associated with a variety of health effects (Masitah, et al., 2007, Hashim , 2009). Limited surveys on air pollution have indicated in Iraq, particularly those on the air pollution from oil industrial activities (Al-Saadi, 2012). Particulate matter is a mixture of liquid droplets and small particles either organic (such as the polycyclic aromatic hydrocarbons (PAHs)) or inorganic substances. Polycyclic aromatic hydrocarbons (PAHs) belong to the group of persistent organic pollutants (POPs). These are organic contaminants that are resistant to degradation; they can remain in the environment for long periods, having the potential to cause adverse environmental effects. POPs are truly multimedia contaminants which occur in all parts of the environment atmosphere, inland and sea waters, sediments, soils and vegetation (Maliszewska-Kordybach, 1999).

The PAHs are predominantly formed as a result of the incomplete combustion of organic fuels such as the burning of petroleum and oil or emissions from automobiles and industrial processes (petrochemical industry and manufacturing of paints) . Many researchers had identified 16 different "priority pollutants" PAHs which have stronger toxicity than others and individual PAHs differ substantially in their physical and chemical properties (Shihua et al. 2001;

Bari et al., 2011). In ambient air, PAHs are generally present in both the gaseous phase and in association with particular matter (PM). They are partitioned in varying proportions in the two phases depending on a number of factors including the vapor pressure of the PAH, temperature, humidity, precipitation, the amount and nature of the PM present in the atmosphere (Rajput and Lakhani, 2010). The allowable PAHs in the atmospheric air is 0.2 mg/m^3 (WHO 2000; Husain 2003; Hashim2009; Al-Saadi 2012). Several researchers had carried out studies on environmental air pollution, but there are very limited studies particularly on the air pollution from oil industrial activities in Iraq (Ali, 2013, Shanshal, et al, 2014). The aim of this study is to analyze samples for total suspended solids concentrations (TSP) , heavy metals and PAHs in air for total of 34 samples (17 samples during each July 2012 and October 2013) , between latitudes ($34^{\circ} 56'' - 35^{\circ} 34''$) and longitude ($43^{\circ} 30'' - 43^{\circ} 34''$) (Fig.1).

2. MATERIALS AND METHODS

A- The TSP concentration in air at area near the Baiji Oil Refinery was determined in two periods July 2012 and Oct. 2013, by using Low volume air sampler (Sniffer), for total of 34 samples (17 samples at each sampling period) taking into consideration the prevailing wind direction that is an important factor in pollutants distribution (Husain, 2003, Christensen, et al, 2005, Ximei, et al, 2006).

B-Measuring the concentrations of some heavy metals (Co, Ni, Pb, Zn, Cr, and Cd), by using atomic absorption, for total of 34 samples, for two periods (17 samples at July 2012 and 17 samples at October 2013).

C- The chemical analysis of the air samples was done for measurements of the concentrations of the PAHs in the air, and their distribution in the studied area. As highly efficient separation tools, Gc-Ms and HPLC have been used for analyzing all kinds of samples containing complex components (Husain 2003; Gritti and Guiochon, 2010; Ali 2013). The used standards for polycyclic aromatic hydrocarbons analysis were of Sigma-Aldrich Company with high purity (not less than 99.5 %). A mixture of the 16 compound with a different concentrations for each standard material (naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, fluoranthene, chrysene, anthracene, benzo(a)anthracene, benzo(k)fluoranthene, benzo(b)fluoranthene, pyrene, dibenzo(a, h)anthracene, benzo(a)pyrene, benzo(g, h, i) perylene, indeno(1,2,3-cd) pyrene) were used (Rasdy, et al, 2008, Menezes and Cardeal, 2011).

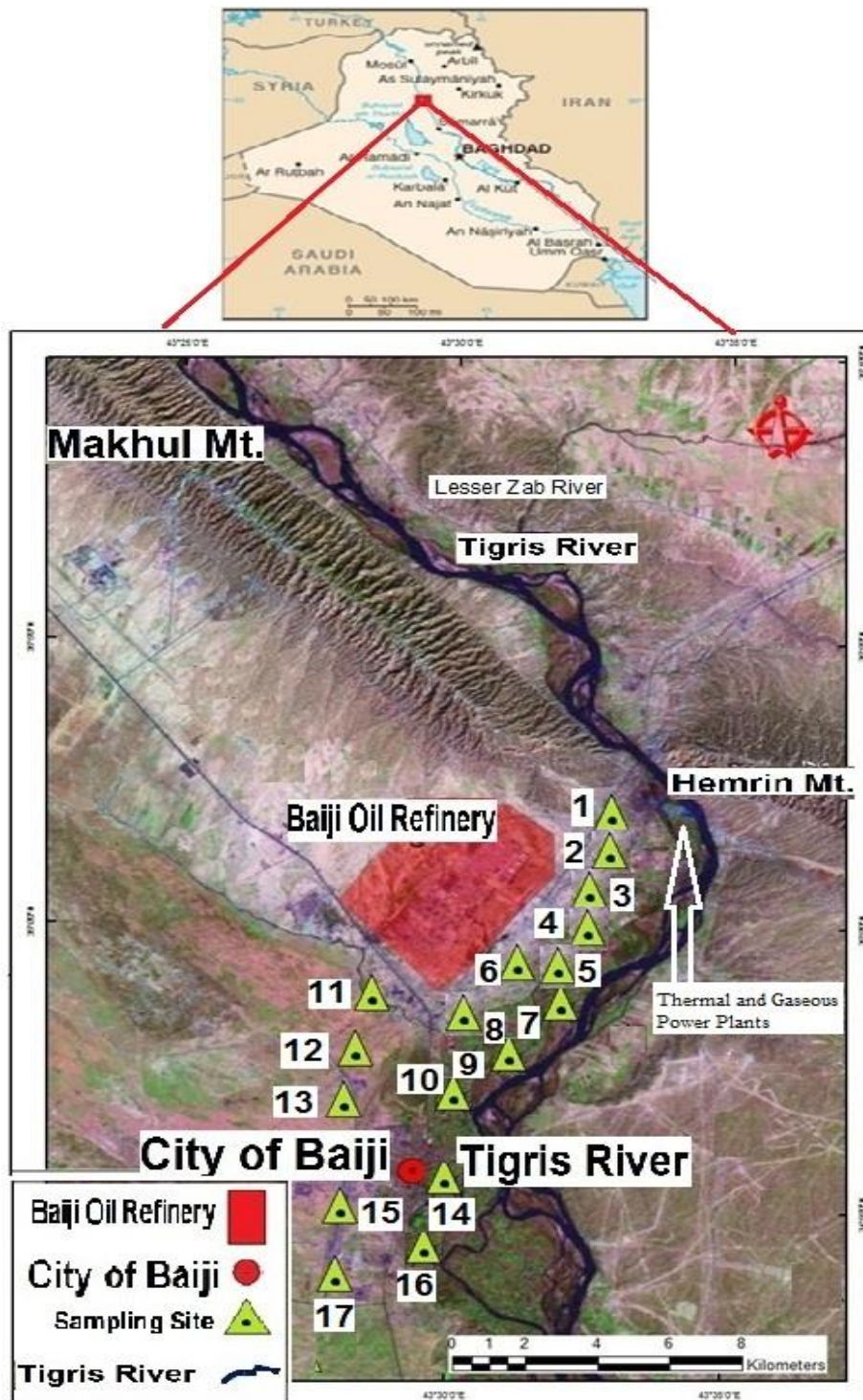


Figure 1: Location map of Baiji Oil Refinery (BOR) and the sampling sites.

3.RESULTS AND DISCUSSION

1-Total suspended particles (TSP):

The results of the total suspended particles concentration (TSP) analyses reflect that the average TSP concentrations ($\mu\text{g}/\text{m}^3$) in the air of the studied area in July 2012 was $2830 \mu\text{g}/\text{m}^3$ and October 2013 was $1090 \mu\text{g}/\text{m}^3$ (Table 1). The

results for TSP concentration show that Baiji Oil Refinery was not the only TSP source but also the neighboring sand dune area to the west and North West of the studied area which had an intensive effect in increasing the TSP in air

especially with increasing wind speed in July (Figures 2 and 3) (Kadhim, et al , 2012).

The analyzed climatic elements of Baiji meteorological station for the years 1980 - 2014, are shown in Table 2. The results reflect that the high value of mean monthly maximum, temperature minimum temperature and wind speed were (46.6) °C, (29.9) °C and (5.3) m/sec during July 2012, respectively. While the low value were (29.60) °C , (13.9) °C and (0.4) m/sec during Oct.2013, respectively. The high value of mean monthly Rainfall (mm) and Relative Humidity% were (32.80) mm and (63.0%) during Oct.2013, respectively, while the low values were (0.0) mm and (36.0%) during July 2012, respectively. Moreover, the prevailing mean monthly wind directions for Baiji meteorological station reflect mainly northwesterly winds direction (Figure 2). Hence, the variation between July and October month's effects of the direction, speed of the wind and the intensity of the dust storms are obvious (Figure 4). The variation between the two periods reflects the intensity of the dust storms and the direction, speed of the wind effect during both study periods (Table 2). It was also observed that the TSP concentrations distribute at

the direction away from the refinery that coincides with the prevailing wind direction during these months. More than 80 dust storms were captured by the MODIS sensor over Iraq during the period of 2003 to 2012. It is seen that dust storms could occur any time in the year in Iraq but most likely during the months of March to July (Al-Dabbas, et al., 2012). During dust storms, it is seen that distinct plumes were raised from many point sources in the Jezera area to the north west of the studied area. The dust plumes arise from discrete points in northwestern Iraq, and blow toward the southeast. The veil of dust is thick enough that the ground beneath was not visible (Alonso-Pérez, et al., 2013). Comparison of TSP averages with national and world limits reflect that the average concentration of suspended particles (TSP) were higher than the permissible allowable limits of the Iraqi National standards (350) µg/m³ and the worlds international allowable limits (60-90) µg/m³, (Table 1) for the both sampling periods. Although the TSP averages are within the same range of Ali, 2013 results of the TSP from Kirkuk oil refinery.

Table 1: TSP concentrations (µg/m³) in the air of the studied area in April and July 2012 with comparison TSP of national and world limits (WHO, 2000, Ministry of Environment, 2008, Ali, 2013).

No	Sample Location	TSP/Oct.2013	TSP/ July 2012
	Range	180 -1860	1400 - 4000
	Mean	1090	2830
	Iraqi National standards(MOE,2008)	350 µg/m ³	
	World allowable limits (WHO, 2000)	60-90 µg/ m ³	
	Ali, 2013	Range	230 - 3555.6
		Mean	956.8 µg/ m ³

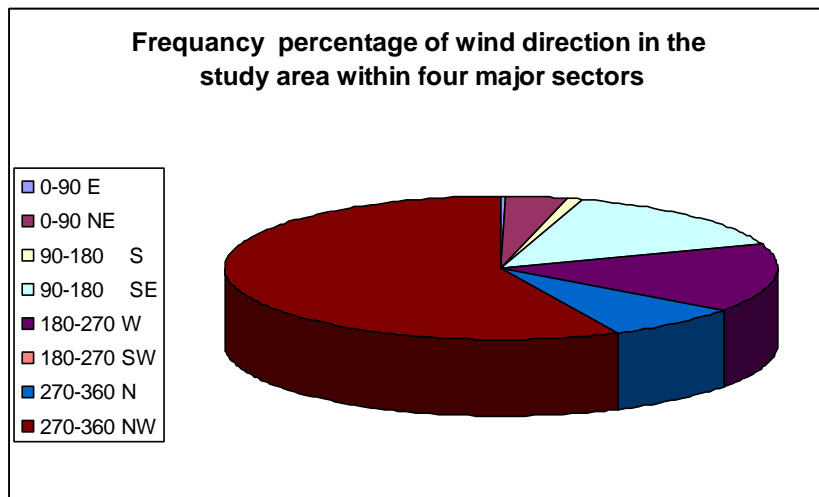


Figure 2: The frequency percentage of wind directions within different sectors (after Kadhim, et al , 2012).

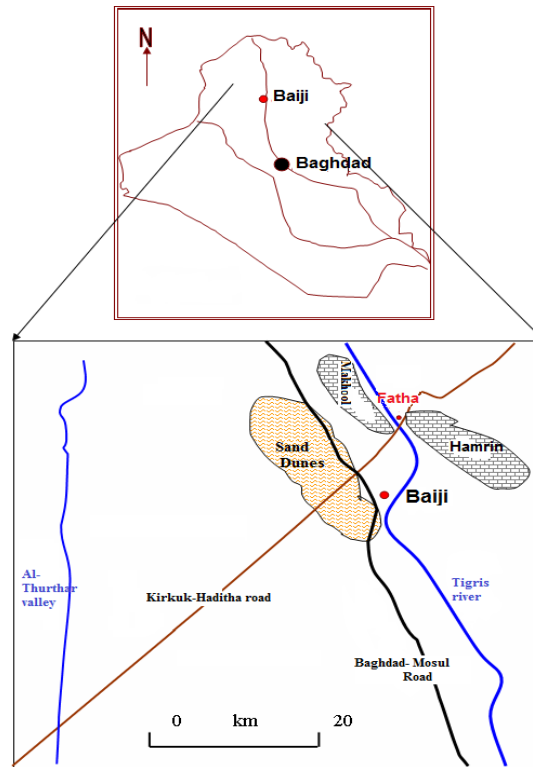


Figure 3: Location of sand dunes in NW of Baiji (after Kadhim, et al , 2012).

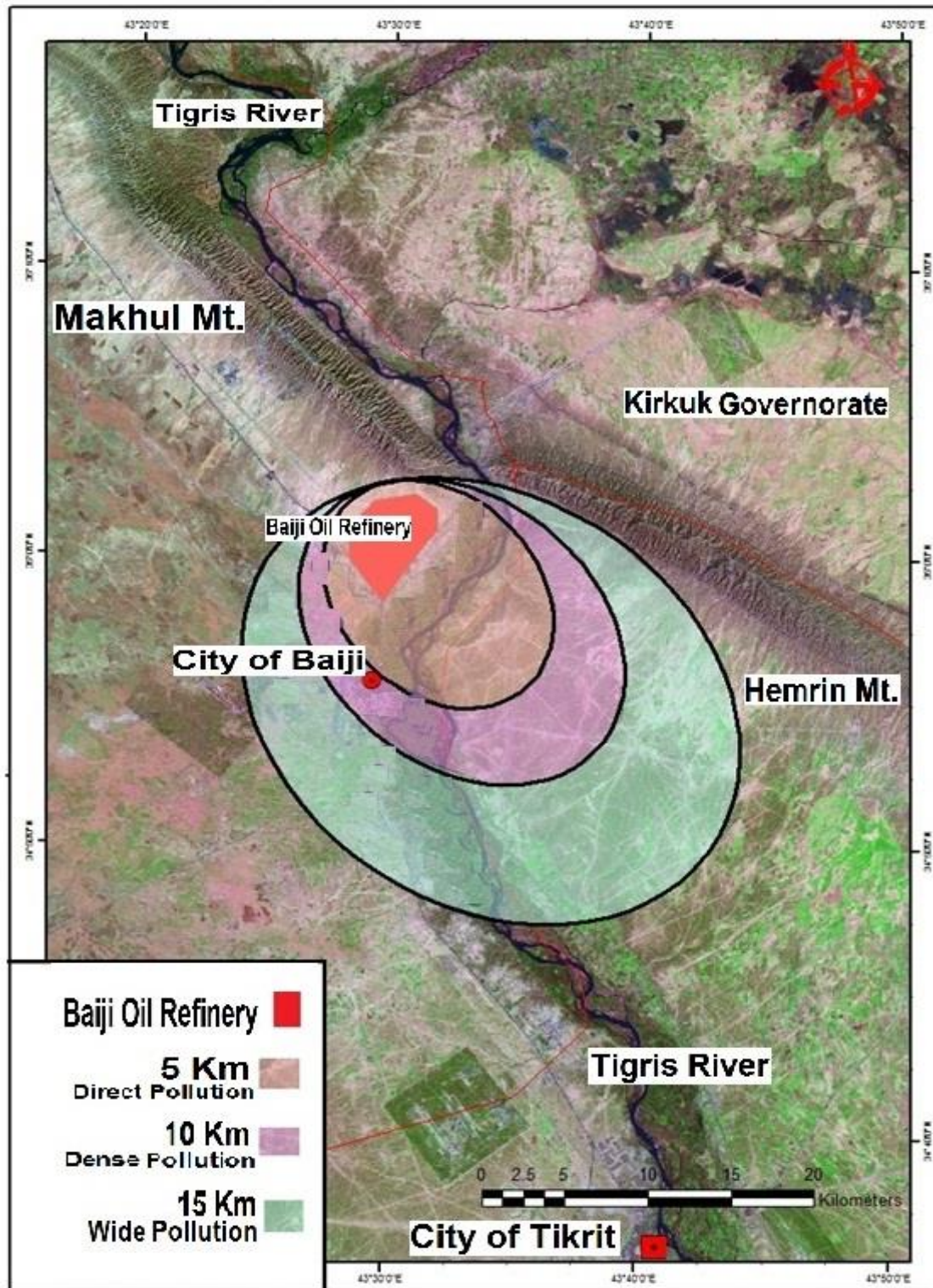


Figure 4: Concentrations of total suspended particles distribution in the air for study area.

Table 2: The average monthly rainfall (mm), Maximum, Minimum Temperatures ($^{\circ}\text{C}$), relative humidity% and wind speed m /sec of July 2012, Oct.2013 of Baiji meteorological station for years 1980 to 2014.

average monthly	Value	July 2012	Oct.2013
Rainfall (mm)	Min	0.00	0.00
	Max	0.00	32.80
	Mean	0.00	8.65
Maximum Temperature($^{\circ}\text{C}$)	Min	41.90	29.60
	Max	46.60	35.00
	Mean	43.73	32.56
Minimum Temperature($^{\circ}\text{C}$)	Min	24.10	13.90
	Max	29.90	20.20
	Mean	27.36	16.99
relative humidity%	Min	20.00	37.00
	Max	36.00	63.00
	Mean	26.63	45.83
wind speed (m /sec)	Min	0.80	0.40
	Max	5.30	2.90
	Mean	2.98	1.25

2. ANALYSES OF THE HEAVY METALS

The results of the heavy metals Co, Ni, Pb, Zn, Cr, and Cd concentration ($\mu\text{g}/\text{m}^3$) reflect that the average concentrations in July 2012 were 0.11, 0.23, 1.7, 0.03, 0.3 and $0.08 \mu\text{g}/\text{m}^3$ respectively, while in Oct.2013 were 0.08, 0.19, 0.13, 0.02, 0.05 and $0.07 \mu\text{g}/\text{m}^3$ respectively, Table 3.

Comparison of the averages of the heavy metals with the world limits, noticed that they are lower than these limits. Except for the concentration of Cr and Cd, at the two periods and Pb in July only,

were higher than the world limits $0.5 \mu\text{g}/\text{m}^3$, where the average was $1.7 \mu\text{g}/\text{m}^3$, although these average heavy metals values are within the same range or lower than that of Ali, 2013 results of the heavy metals for Kirkuk oil refinery, (WHO, 2000, Ministry of Environment, 2008, Ali, 2013), Table 3. This increasing in lead concentration in July can be referred to the increasing fuel combustion operations at the location such as the operations of the power plant which functions increasingly during the July month as well as the effect of meteorological factors on the air quality of the studied area at this period.

Table 3: Comparison of the concentrations of the studied heavy metals ($\mu\text{g}/\text{m}^3$) in the air with of WHO, 2000, Ali, 2013 results during both studied periods.

No	Sample Location	Cobalt (Co) ppm	Nickel (Ni) ppm	Lead (Pb) ppm	Zinc (Zn) ppm	Chromium (Cr) ppm	Cadmium (Cd)
July 2012	Range	0.02-0.35	0.02-0.65	0.09- 4.9	0.001-0.1	0.02-0.9	0.01-0.13
	Mean	0.11	0.23	1.7	0.03	0.3	0.08
Oct.2013	Range	0.01-0.2	0.01-0.6	0.03-0.8	0.001-0.09	0.01-0.08	0.01-0.1
	Mean	0.08	0.19	0.13	0.02	0.05	0.07
WHO,2000			0.2	0.5		0.04	0.05
Ali 2013	Range		0.5-0.9	0.9-9.1		0.01-2.5	0.07-0.2
	Mean		0.6	4.6		0.75	0.1

3. TPAH (ΣPAH) CONCENTRATIONS

The results of the total polycyclic aromatic hydrocarbons (PAHs) concentrations analysis in air show the existence of sixteen hydrocarbons in air, these are Naphthalene, Acenaphthene, Acenaphthylene, Fluorene, Phenanthrene, Fluoranthene, Chrysene, Anthracene, Benzo (a) anthracene, Benzo (k) fluoranthene, Benzo (b) Fluoranthene, Pyrene, Dibenzo (a, h) anthracene, Benzo (a) pyrene, Benzo (g, h, i) perylene, Indeno (1,2,3-cd) Pyrene Table 4.

During July 2012, most of the polycyclic aromatic hydrocarbons minimum concentrations were less than $3.0 \mu\text{g}/\text{m}^3$ except two of them had concentrations more than $3.0 \mu\text{g}/\text{m}^3$, these are Naphthalene and Benzo (g, h, i) perylene. While two of the studied polycyclic aromatic hydrocarbons

had less than $1.0 \mu\text{g}/\text{m}^3$ maximum values during July 2012, these are Anthracene and Chrysene. While, during Oct.2013, most of their minimum values were less than $1.0 \mu\text{g}/\text{m}^3$, except two of them indicate more than $1.0 \mu\text{g}/\text{m}^3$ (Fluoranthene and Indeno(1,2,3-cd)pyrene).

While most of their maximum values were more than $1.5 \mu\text{g}/\text{m}^3$ during Oct.2013, except for Phenanthrene, Anthracene, Chrysene and Benzo(b)fluoranthene which they had less than $1.5 \mu\text{g}/\text{m}^3$ concentrations (Table 4). The 16 EPAs priority PAHs detected in the studied area were not all found at all sites of measurements due to their physicochemical properties of these compounds. The concentration of PAHs in air of the studied area in July 2012 (ranging from 9.1 to $175.5 \mu\text{g}/\text{m}^3$ with average $40.3 \mu\text{g}/\text{m}^3$)

Table 4: Polycyclic aromatic hydrocarbons (PAHs) concentrations analysis in air of the studied area in July 2012 and Oct.2013.

No.	PAHs		July 2012			Oct.2013		
			Min	Max	Mean	Min	Max	Mean
1.	Naphthalene	NAP	76.3 ±12.6	114.2 ±30.5	96.3±13.9	0.6 ± 0.17	35.1 ± 8.6	11.2 ± 0.35
2.	Acenaphthylen e	ACY	1.7 ± 0.22	81.5 ± 13.5	30.5± 9.7	0.4 ± 0.16	4.8 ± 0.3	1.2 ± 0.22
3.	Acenaphthene	ACE	1.4 ± 0.15	3.7 ± 0.2	2.3 ± 0.2	0.9 ± 0.01	2.6 ± 0.2	1.3 ± 0.12
4.	Fluorine	FLU	0.8 ± 0.1	10.3 ± 0.4	3.6 ± 0.2	0.5 ± 0.1	3.1 ± 0.1	2.6 ± 0.13
5.	Phenanthrene	PHE	2.2 ± 0.13	31.4 ± 10.2	8.7 ± 0.17	0.2 ± 0.01	0.7 ± 0.1	0.4 ± 0.01
6.	Anthracene	ANT	0.3 ± 0.01	0.8 ± 0.1	0.5 ± 0.1	0.15± 0.01	0.19 ± 0.12	0.15 ± 0.1
7.	Fluoranthene	FLUA	0.3 ±0.01	19.2 ± 7.7	3.4 ± 0.17	1.5 ±0.1	4.1 ±0.2	2.0 ±0.1
8.	Pyrene	PYR	1.3 ± 0.1	27.6 ± 8.2	4.6 ± 0.2	1.0 ±0.1	2.0 ± 0.3	1.5 ±0.2
9.	Benzo(a)anthra cene	B(a)A	2.7 ± 0.22	52.6 ± 14.2	15.6 ± 11.3	0.2 ± 0.01	1.8 ± 0.2	0.6 ± 0.01
10.	Chrycene	CHR	0.3 ± 0.01	0.7 ± 0.12	0.4 ± 0.01	0.2 ± 0.01	0.5 ± 0.01	0.3 ± 0.01
11.	Benzo(b)fluora nthane	B(b)F	0.3 ±0.01	3.3 ±0.1	1.3 ±0.1	0.2 ± 0.01	1.2 ± 0.1	0.3 ± 0.01
12.	Benzo(k)fluora nthane	B(k)F	0.2 ± 0.01	12.4 ± 0.4	4.2 ± 0.2	ND	ND	ND
13.	Benzo(a)pyren e	B(a)P	0.5 ± 0.01	5.7 ± 0.18	2.3 ± 0.15	ND	ND	ND
14.	Dibenzo(ah)ant racene	Dib(ah) A	1.2 ±0.01	13.6 ±0.5	6.3 ±0.3	0.4 ±0.01	4.7 ± 0.2	2.4 ±0.1
15.	Benzo(ghi)pery lene	B(ghi)P	4.7±0.2	12.2±0.6	8.5±0.4	0.3 ± 0.01	7.0 ±0.2	2.7±0.1
16.	Indino(1,2,3- cd)pyrene	Ind P	2.5 ±0.1	5.3 ±0.2	3.4±0.2	1.3 ± 0.1	2.7 ± 0.1	1.5 ± 0.2
Total			9.1 ± 2.2	175.5 ± 16.4	40.3 ± 13.3	8.0± 1.8	80.0± 16.7	25.6 ± 6.5

was rather higher than that detected during Oct.2013 (ranging from 8.0 to 80.0 $\mu\text{g}/\text{m}^3$ with average 25.6 $\mu\text{g}/\text{m}^3$). This increment in PAHs concentrations at July 2012 period can be referred to the increase of dust storms with the increasing the wind speed, and air temperature with decreasing the relative humidity % (Table 2). The influence of meteorological parameters such as temperature, relative humidity and wind speed on the PAH emissions was analyzed by statistical package for social scientist (SPSS) program. The direct correlation between the total PAH concentration, values of temperature and the relative humidity% could be observed as listed in the Table 5. This could be due to a depositional effect on the particulate matter of PAHs in the gas phase as a consequence of environmental humidity. There are strong correlation between TSP and TPAH (0.912, 0.971) respectively for July 2012 and Oct.2013; due that the higher concentration of total suspended particulate leads to the adsorption more molecules of PAHs compounds (Bjorseth and Ramdahl, 1985, Dias, 1987). For the rate wind studied, no statistical significance was obtained.

Table 5- Calculation of Pearson Correlations of parameters in the different months.

Pearson Correlations –July 2012						
parameter s	Min. Temp . (°C)	Max. Temp . (°C)	RH %	Wind Speed (m/s)	Tsp (mg/m ³)	TPAHs (µg/m ³)
Tsp (mg/m ³)	0.573	0.692	0.521	0.623	1	
TPAHs (µg/m ³)	0.027	0.823	0.415	0.512	0.971	1
Pearson Correlations – Oct.2013						
parameter s	Min. Temp . (°C)	Max. Temp . (°C)	RH %	Wind Speed (m/s)	Tsp (mg/m ³)	TPAHs (µg/m ³)
Tsp (mg/m ³)	0.222	0.361	0.612	0.023	1	
TPAHs (µg/m ³)	0.196	0.291	0.831	0.431	0.912	1

4. CONCLUSIONS

1-The average concentration of total suspended particles (TSP) are higher than the permissible limits of the Iraqi National determinants of (350 µg/m³) and the world limits of (60-90 µg/m³) at the two periods: July 2012(2714.1 µg/m³) and Oct.2013 (1124.8 µg/m³). The difference between the two periods reflects the effect of the seasonal changes.

2- The average heavy metals concentrations in July 2012 were higher than in Oct.2013 and they are lower than international limits. Except for Cr and Cd, at the two periods and Pb in July only. This increasing in lead concentration in July can be referred to the increasing fuel combustion operations at the location.

3- The 16 EPAs priority PAHs detected in the studied area were not all found at all sites of measurements due to their physicochemical properties of these compounds.

4- The TPAH (ΣPAH) concentrations in July 2012 (ranging from 9.2 to 175.3 µg/m³ with average 40.2 µg/m³) was rather higher than during Oct.2013 (ranging from 8.1 to 79.7 µg/m³ with average 25.5 µg/m³). This increment can be referred to the increase of dust storms with the increasing the wind speed, and air temperature with decreasing the relative humidity %.

5- Positive correlation between TPAH and TSP with meteorological condition by the SPSS was found.

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