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THE ENVIRONMENTAL IMPACT OF KIRKUK REFINERY ON SOIL QUALITY AT KIRKUK GOVERNORATE, N IRAQ

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ABSTRACT

The average concentrations of heavy metals (Pb, Cu, Ni, Cr, and Cd) with the polycyclic aromatic hydrocarbons (PAHs) in the soil of Kirkuk Oil Refinery and the surroundings were determined in 15 selected locations. The sample collection was carried out in two periods: October 2010 and March 2011. In comparison with other studies, the concentration levels of heavy metals show that the values are within or lower than those of other studies, except for the concentration of Cd in the two periods, which are higher. The average concentrations of polycyclic aromatic hydrocarbons (PAHs) in soil indicates 3.5 ppb in October, while in March it is 14.2 ppb.

The model of cumulative effect of PAHs, determined by GIS program for both sampling periods, shows an increase of concentration in wind directions away from the site of Kirkuk Oil Refinery.

التاثير البيئي لمصفى كركوك على نوعية ترب مدينة كركوك، شمال العراق

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المستخلص

ان معدل تركيز العناصر الثقيلة (Pb, Cu, Ni, Cr, Cd) والهيدروكربونات العطرية المتعددة الحلقات (PAHs) في نماذج التربة الماخوذة من مصفى كركوك والمناطق المحيطة بة قد تم في 15 موقعا مختارا. ان جمع النماذج كان لفترتين هي تشرين اول 2010 وآذار 2011.

أوضحت مقارنة نتائج البحث مع نتائج دراسات اخرى بان قيم العناصر الثقيلة وقعت ضمن او اقل من مدى نتائج الدراسات العراقية والعالمية ماعدا عنصر الكادميوم الذي كان اعلى من تلك المعدلات وفي فترتي الدراسة.

ان معدل تركيز الهيدروكربونات العطرية المتعددة الحلقات (PAHs) في التربة كان 3,5 جزء من البليون في تشرين 2010 وفي اذار 2011 كان 14,2 جزء من البليون.

أظهرت نتائج استخدام نموذج تاثير الهيدروكربونات العطرية المتعددة الحلقات التراكمي الذي حدد بتطبيق برنامج (GIS) لفترتي الدراسة بان تراكيزها تنتشر باتجاه الرياح السائدة مبتعدة عن المصفى.

INTRODUCTION

The source of the polycyclic aromatic hydrocarbons (PAHs) and heavy metals can be divided into natural and artificial sources. Soil receives pollutants from a variety of sources, including automobile exhaust gases, and emissions of factory chimneys, household electric power generators and dust storm (Habib *et al*., 2012). Also, tire friction adds some metals, particularly cadmium, the motor oils consumed contain heavy metals in addition to oil

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burning and waste incineration (Thorpe and Harrison, 2008). Heavy metals are those elements having specific gravity at least five times that of water (which is 1 at 4 °C) and are also referred to as metallic elements with an atomic weight greater than iron (55.8 g/mol). The composition and quantity of chemical matrix of roads, highways dust and cement plants are indicators of environmental pollution with high concentration of Pb, Cu, Ni, Cr, and Cd (Awadh, 2009 and 2014). The sources of heavy metals in soil are mainly natural, including geologic sources such as rock formations, soils, and wind transported sediments, while the artificial sources include industrial sources that supply the heavy metals to the air and soil causing pollution of the environment.

Henner *et al*. (1997), Hussain (2003), and Al-Saadi (2012) identified 16 different "priority pollutants" PAHs which have stronger toxicity than others. Several researchers, such as Al-Maliki (2005) and Li *et al*. (2010), carried out studies on environmental soil pollution, but there are very limited studies particularly on the soil pollution from oil industrial activities in Iraq. Kirkuk has experienced rapid growth in population and urbanization over the last few decades. It is estimated that between 2003 and 2012 a huge number of vehicles were registered in Kirkuk, in addition to the second hand used cars which are still in service. This exerts a heavy pressure on the urban environment.

The climatic parameters have an important effect on the concentration of pollutants in the soil and play a key role in controlling the spread of various soil pollutants (Awadh, 2014). The wind direction plays an important role in the distribution of pollutants in air, moving the pollutants with the general direction of the prevailing winds (Ahrens, 2005). Many researchers who studied soil, water and plants have detected high concentrations of heavy metals in different sampling media (Awadh, 2009 and Ali, 2013). The present work is a further contribution to the geochemical studies of local environmental pollution, discussing the concentration and distribution of some heavy metals and PAHs in Kirkuk soil in an attempt to reveal their source.

This work is based on analyzing the soil quality in and around Kirkuk Refinery and to identify location and nature of pollution. The study area is located around the oil refinery. The climate of the area is continental being hot in summer, cool wet in winter, and the wind prevails in a northwestern direction (Ali, 2013). The generally dry and warm to hot climate dominates most of Iraq and has its influence on the type of Quaternary sediments and the soil developed, being generally immature, without profile zonnation and generally with minor mineralogical and chemical alterations compared to source rocks. The main changes have been the size degradation of rocks into gravel, sand, silt and mud by weather factors and mechanical transportation, rather than by chemical weathering processes. It must be stressed that the climatic conditions were different in Iraq in the Pleistocene, where more pluvial periods and temperate climate existed. The present day climate is an extension of the Holocene climatic conditions, which have dominated the region for the past 6000 years or so (Al-Bassam and Yousif, 2014). Kirkuk surface rocks consist of recent sediments and sedimentary rocks. The rock formation outcrops are wide spread ranging in age from Miocene (Fatha Formation) to Quaternary. The common lithology is limestone and clastics with the presence of gypsum. These formations, from the oldest to the recent are as follows: Fatha Formation, M. Miocene, Injana Formation, U. Miocene, Mukdadiyah Formation, U. Miocene – Pliocene, Bai Hassan Formation, Pliocene, Quaternary deposits and recent sediments (Sissakan, 1993, and Majeed, 2004).

The area is divided geomorphologically into two parts. The first, where the study area lies, is generally flat with some hills and undulations, whereas the second part is mountainous and located in the north and northeast of the province.

This study aims to measure the concentration of the heavy metals (Pb, Cu, Ni, Cr, and Cd) and PAHs in soil as well as assessing the environmental impacts of Kirkuk Oil Refinery and surroundings, (Fig.1).

MATERIALS AND METHODS

Fifteen sampling sites inside and outside Kirkuk Oil Refinery have been selected between (35' 24" – 35' 29") N and (44' 20" – 44' 26") E (Fig.1). The site selection of the soil samples takes into consideration the prevailing wind direction, pollutants distribution, as well as the population. The concentrations of the heavy metals (Pb, Cu, Ni, Cr, and Cd) were analyzed using Atomic Absorption Spectrometry.

The polycyclic aromatic hydrocarbons (PAHs) in soil were determined using HPLC (High Performance Liquid Chromatography) and GC-MS (Gas Chromatography Mass) of the polycyclic aromatic hydrocarbon compounds (with less than 1 ppb detection limits) according to procedures of Henner *et al*. (1997) and Husain (2003) in the laboratories of the Ministry of Science and Technology. 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 compounds 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(ah)anthracene, Benzo(a)pyrene, Benzo(ghi)perylene, Indeno(1,2,3-cd)Pyrene were used, (Henner *et al*., 1997 and Ali, 2013).

HEAVY METALS IN SOIL

The results of analyzing heavy metals in soil are shown in Table 1 for October 2010 and March 2011 samples. Arc GIS model for Nickel and for Chromium distributions in soil (ppm) for October 2010 and March 2011, as representative for heavy metal distribution in Kirkuk soil, are shown in Figure 2. The results indicate that the concentrations are relatively higher in winter (March period) due to combustion of gas and petrol products and atmospheric contaminants (dust and smoke) which accumulate in the soil surface after rain (Figure 2). The meteorological conditions play an important role in the heavy metals distribution in air especially the factor of wind direction (Ali, 2013). Therefore the effects of seasonal changes are obviously noticed on the model made, as shown in Figures (2a-2e). The results show that the concentrations of these pollutants are distributed away from the refinery, toward the southeast part of the studied area. Furthermore, the results show that the refinery may not be the only contaminating source.

Comparison of the mean concentration of the heavy metals (Pb, Ni, Cr, and Cu) with the results of other studies shows that they are within or lower than these values except for the concentrations of Cd which are higher in the two sampling periods, (Table 1).

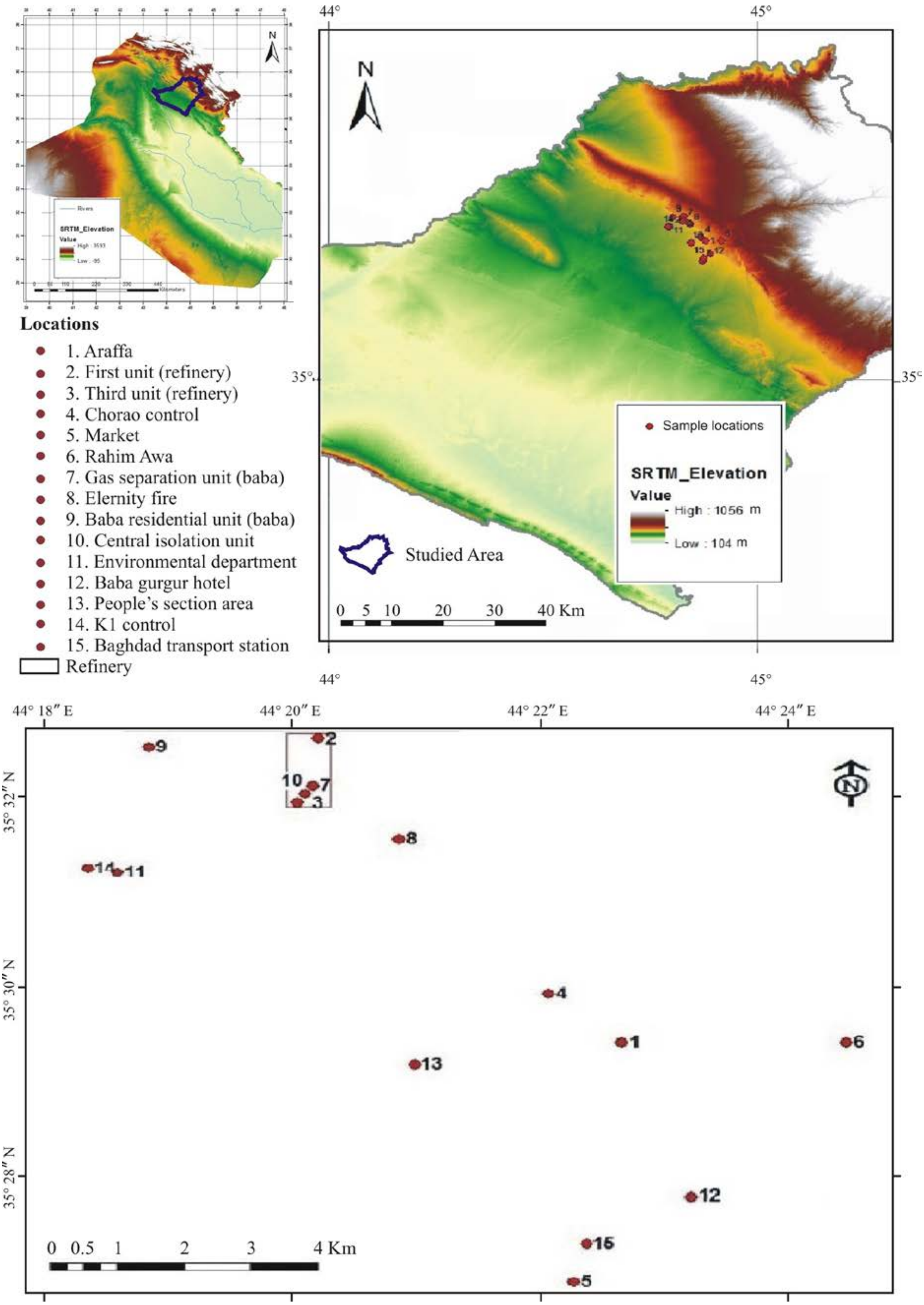

Fig.1: Topographic map of Iraq showing location of Kirkuk Governorate and sampling sites

Table 1: Heavy metal concentrations in the soil of the studied area, compared with results of other studies

Heavy Metals Concentration		**Pb ppm**	**Cu ppm**	**Ni ppm**	**Cr ppm**	**Cd ppm**
October, 2010	Range	0.5 – 15.0	3.0 – 15.0	11.0 – 62.0	31.0 – 81.0	3.8 – 19.8
	Mean	5.7	8.1	32.4	56.6	12.6
March, 2011	Range	0.23 – 49.9	0.1 – 28.9	0.03 – 77.3	25.0 – 90.0	3.6 – 11.9
	Mean	14.3	5.3	44.0	66.0	7.4
Salman, 2007	Mean	39.4	16.9	20.9	161.9	5.5
Hussain, 2014	Mean	50.6	–	59.0	52.0	2.6
Al-Bassam and Yousif, 2014	Mean	6.0	16.0	91.0	238.0	–
Burt *et al.*, 2003	Mean	10.1	17.3	18.3	24.1	0.2
Deabreu *et al.*, 2005	Mean	50.0	53.0	30.0	100.0	3.0
Peter, 2004	Mean	18.0	12.0	25.0	30.0	–
Lindsay, 1979	Mean	10.0	30.0	40.0	100.0	0.1

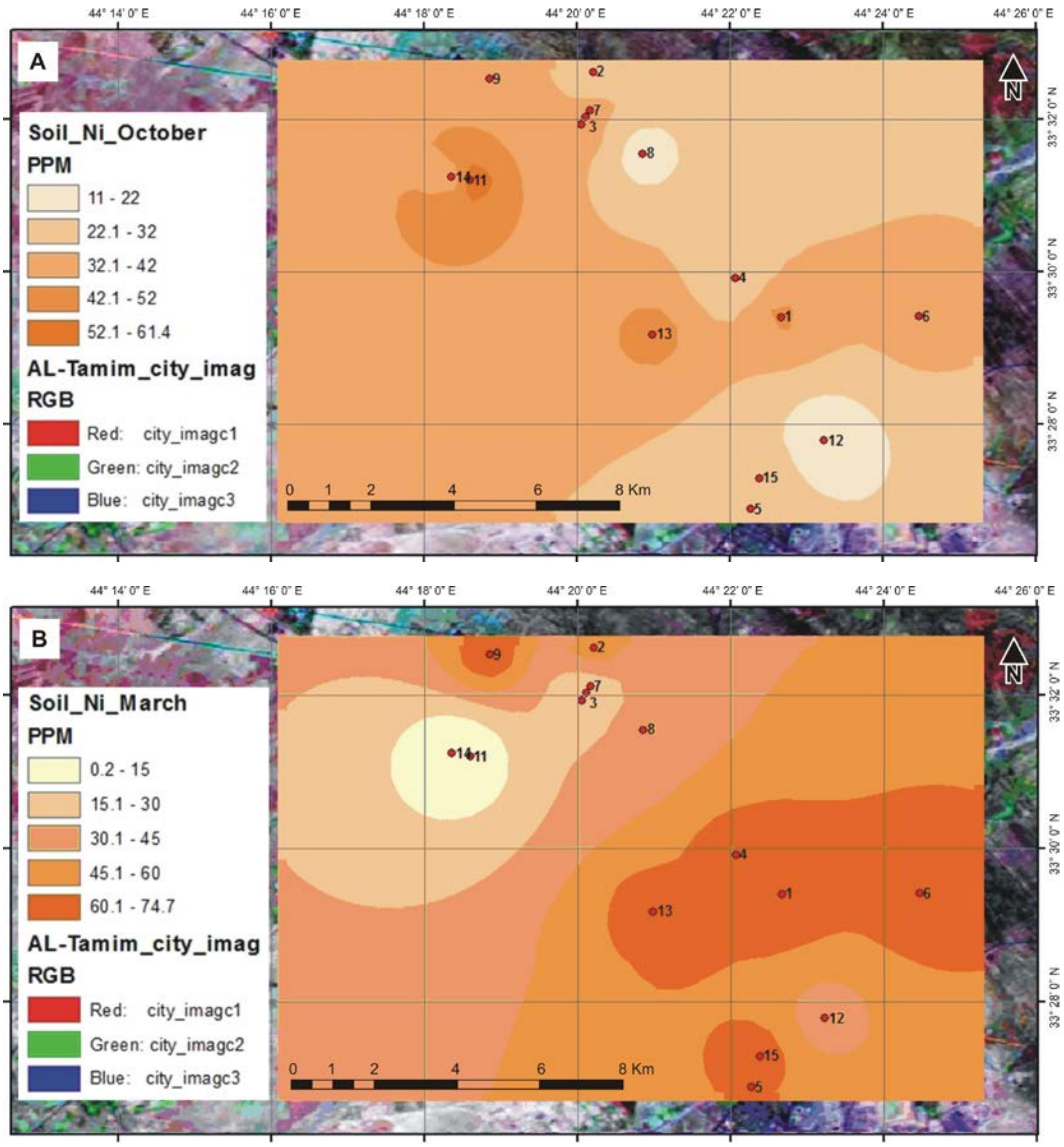

Fig.2a: Arc GIS model for nickel distribution in soil (ppm): **A)** for October, 2010 and **B)** for March, 2011

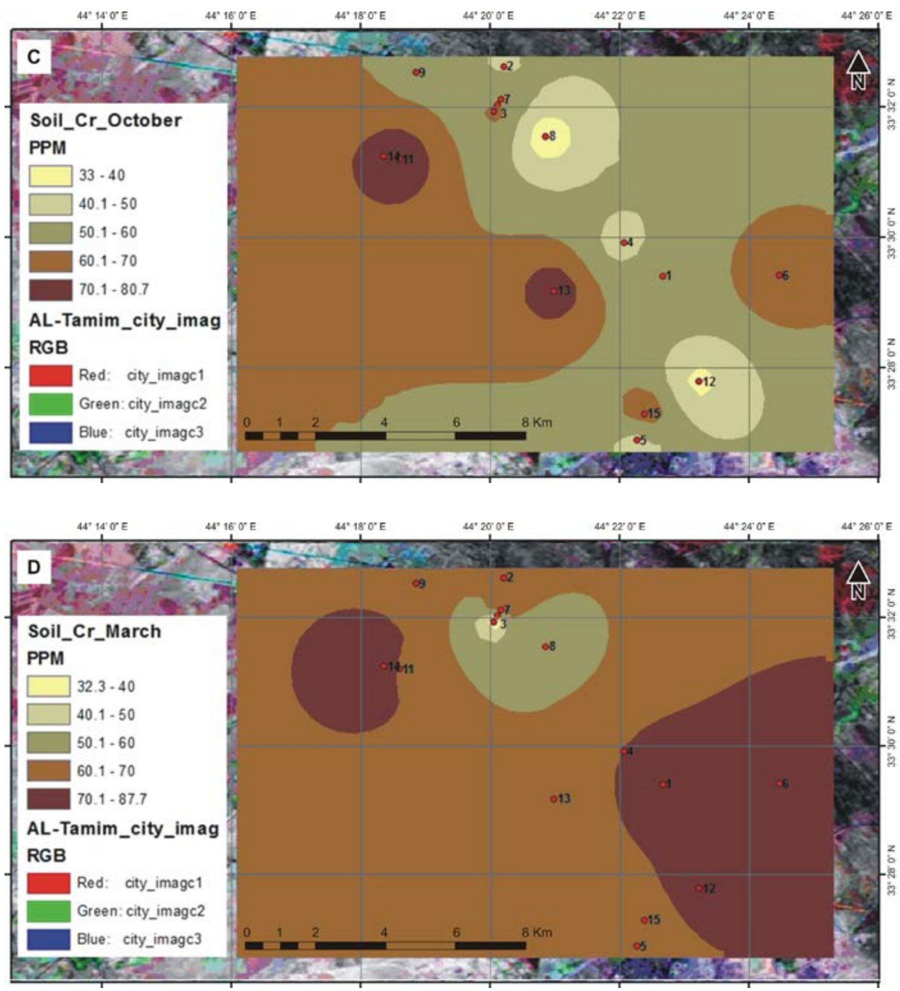

Fig.2b: Chromium distributions in soil (ppm): **C)** for October, 2010 and **D)** for March, 2011

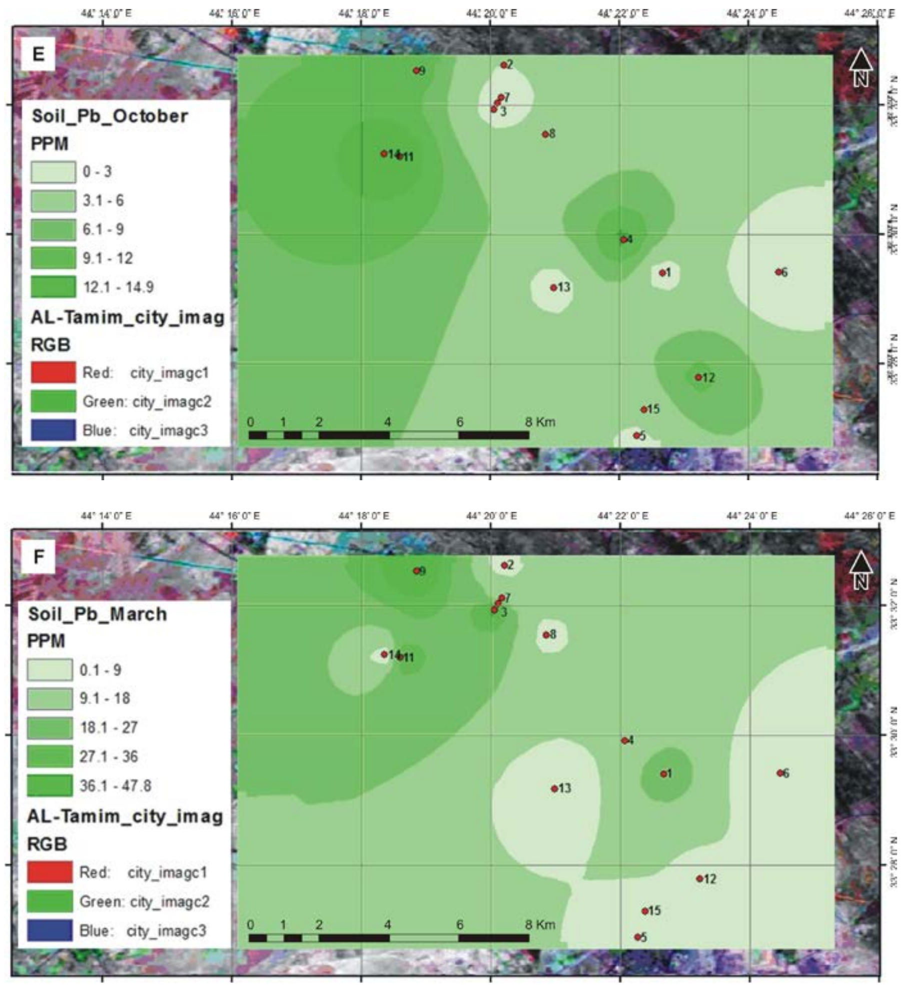

Fig.2c: Lead distribution in soil (ppm): **E)** for October, 2010 and **F)** for March, 2011

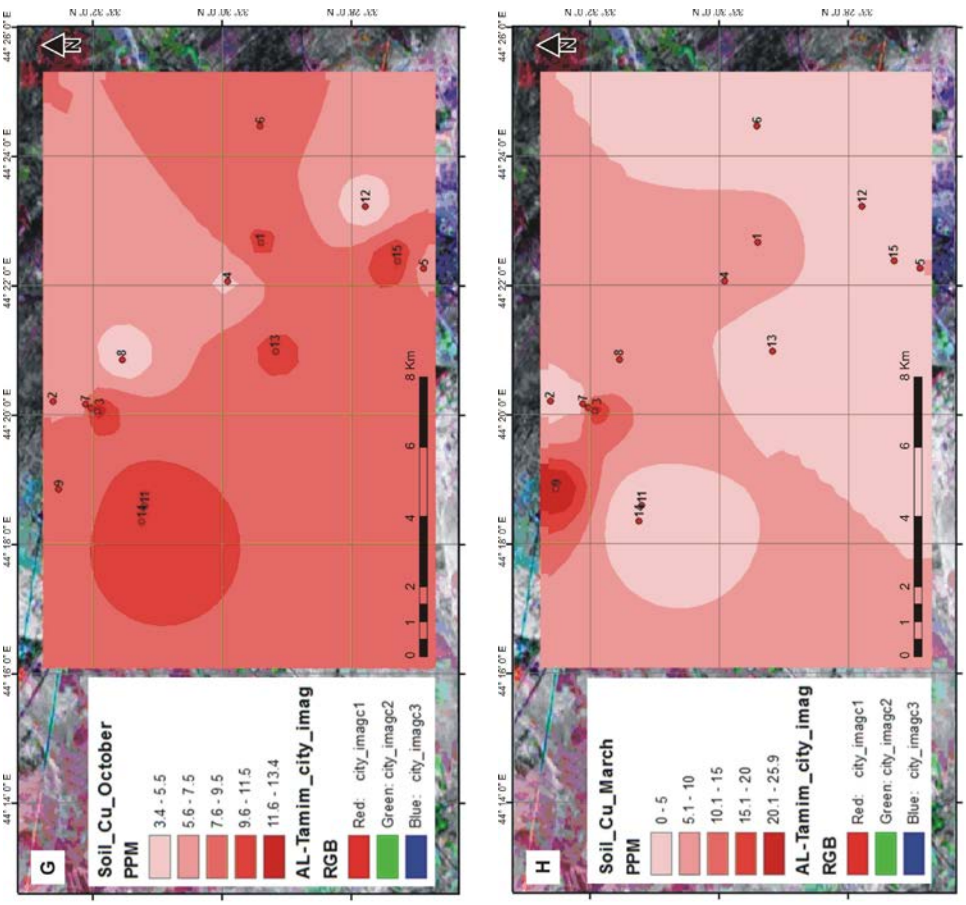

Fig.2d: Copper distribution in soil (ppm): **G)** for October, 2010 and **H)** for March, 2011

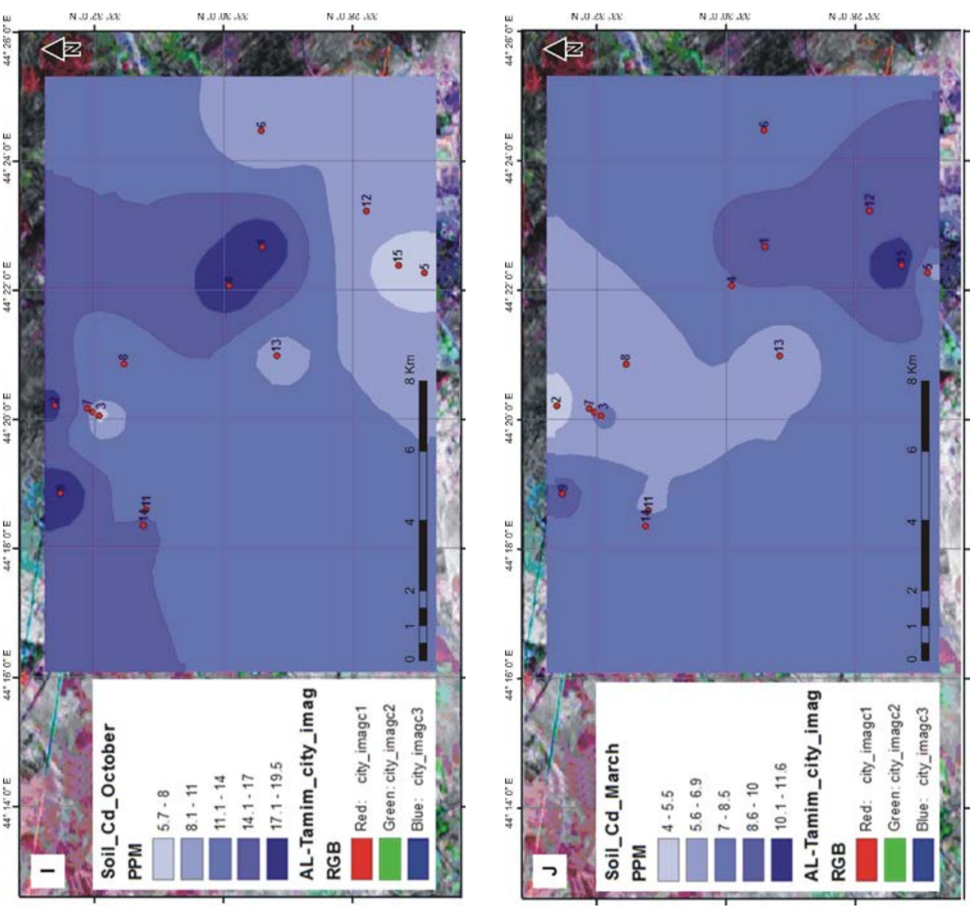

Fig.2e: Cadmium distribution in soil (ppm): **I)** for October, 2010 and **J)** for March, 2011

PAHs IN SOIL

The results of the total polycyclic aromatic hydrocarbons (PAHs) concentrations in soil show the existence of sixteen hydrocarbons These are Naphthalene, Acenaphthene, Acenaphthylene, Fluorene, Phenanthrene, Fluoranthene, Chrysene, Anthracene, Benzo(a) anthracene, Benzo(k)fluoranthene, Benzo(b)Fluoranthene, Pyrene, Dibenzo(ah)anthracene, Benzo(a)pyrene, Benzo(ghi)perylene, Indeno(1,2,3-cd)Pyrene, Table 2. The 16 EPAs priority PAHs detected in the studied area were not all found in all sites of measurements due to their physicochemical properties, so that Naphthalene was detected in the refinery and in the sites nearby due to the low molecular weight found in its gas phase compared with the compounds of high molecular weight found in the particular phase detected in the far away sites such as Benzo(b)flouranthen, Benzo(k)flouranthen, Benzo(a)pyrene, etc.

Most of PAHs minimum values were not detected (ND) during October 2010, except two Benzo(a)pyrene 0.3 ppb and Indino(1,2,3-cd)pyrene 0.5 ppb. On the other hand, most of their maximum values exceed 10.0 ppb except for Fluorine 10.1 ppb, Phenanthrene 27.6 ppb, and Naphthalene 35.3 ppb, (Table 2). In March, 2011, most of PAHs minimum values were less than 1.0 ppb, except two Acenaphthene 1.1 ppb and Dibenzo(ah)antracene 1.1 ppb, while, most of their maximum values were less than 10.0 ppb except for Dibenzo(ah)antracene 12.8 ppb, Fluorine 13.8 ppb, Phenanthrene 32.0 ppb, and Naphthalene 71.1 ppb, (Table 2).

Table 2: Total polycyclic aromatic hydrocarbons (PAHs) concentrations (ppb) in soil of the study area

No.	PAHs		October 2010		March 2011	
			Min	Max	Min	Max
1	Naphthalene	NAP	0.0	35.3	0.24	71.1
2	Acenaphthylene	ACY	ND	1.43	0.3	5.8
3	Acenaphthene	ACE	ND	8.3	1.1	6.5
4	Fluorine	FLU	ND	10.1	0.2	13.8
5	Phenanthrene	PHE	ND	27.6	0.1	32.0
6	Anthracene	ANT	ND	0.14	ND	0.3
7	Fluoranthene	FLUA	ND	7.7	0.1	8.7
8	Pyrene	PYR	ND	2.1	ND	3.9
9	Benzo(a)anthracene	B(a)A	ND	0.2	ND	2.0
10	Chrycene	CHR	ND	0.26	0.3	0.9
11	Benzo(b)fluoranthene	B(b)F	ND	0.25	ND	0.4
12	Benzo(k)fluoranthene	B(k)F	ND	1.0	ND	0.7
13	Benzo(a)pyrene	B(a)P	0.3	0.4	ND	0.6
14	Dibenzo(ah)antracene	Dib(ah)A	0.0	2.2	1.1	12.8
15	Benzo(ghi)perylene	B(ghi)P	0.0	ND	ND	5.3
16	Indino(1,2,3-cd)pyrene	Ind P	0.5	2.0	0.3	7.3

ND: not detected.

The total PAHs in soil of each sampling site is shown in Table 3. Most of total PAHs values are less than 10.0 ppb during October 2010, except for sites 14 and 11 where the PAHs values reach 11.7 and 12.7 ppb respectively. In March, 2011, most of total PAHs values are less than 23.0 ppb, except for sites 9, 6 and 8 where the PAHs values reach 31.1, 32.0 and 39.33 ppb respectively, (Table 3).

Comparison of the total PAHs in soil of each sampling site with others indicates that the results of this study are higher, within or lower than the reference ranges Table 3. (Raymond, and Guiochon, 1974, Husain 2003, Wang *et al*., 2010, Shihab-Aldin and Aziz, 2013, and Jiao *et al*., 2013).

The mean of total concentrations of PAHs in March of (14.2 ppb) is higher than that of October (3.5 ppb). This increment in PAHs concentrations in March, 2011 can be attributed to the increase in fuel combustion operations occurring at the location, such as the operations of the power plant, in addition to the effect of atmospheric contaminants and meteorological conditions such as temperature, rain and prevailing wind that accumulate the PAHs on the soil surface after the winter rain, (Table 3).

Moreover, Arc GIS 10 modeling of measurements of total PAHs in soil was applied for both sampling periods. The cumulative effects of both periods are shown in Figure 3 that represents the cumulative soil model of total PAHs in soil of both periods. The result shows that the concentrations of these pollutants spread away from the refinery. Such finding indicates that the refinery is a most effective source of these pollutants as well as the effects of the meteorological conditions that play an important role in distributing the total PAHs from the air to the soil especially the wind direction and rainfall, (Figure 3).

Table 3: Total polycyclic aromatic hydrocarbons (PAHs) concentrations (ppb) in soil of the study area

Sampling Site No.	Total PAHs concentrations in soil October 2010	Total PAHs concentrations in soil March 2011
1	0.85	7.88
2	0.02	0.14
3	0.84	1.69
4	2.0	2.11
5	9.4	20.5
6	0.38	32.0
7	0.48	20.16
8	2.11	39.33
9	2.5	31.1
10	7.6	8.7
11	12.7	11.3
12	0.1	3.4
13	0.50	2.22
14	11.7	22.8
15	1.65	9.25
Range	0.02 – 12.7	0.14 – 39.33
Mean	3.5	14.2
Wang *et al*., 2010	Range 0.27 – 1.30	
Lonneman *et al*., 1974	Range 3.9 – 12.7 ppb	
Raymond, and Guiochon, 1974	Range 3.8 – 11.2 $\mu g/m^3$	
Husain, 2003	Range 0.3 to 5.17 $\mu g/m^3$.	
Shihab-Aldin and Aziz, 2013	Range 0.3 to 7.0 $\mu g/m^3$	

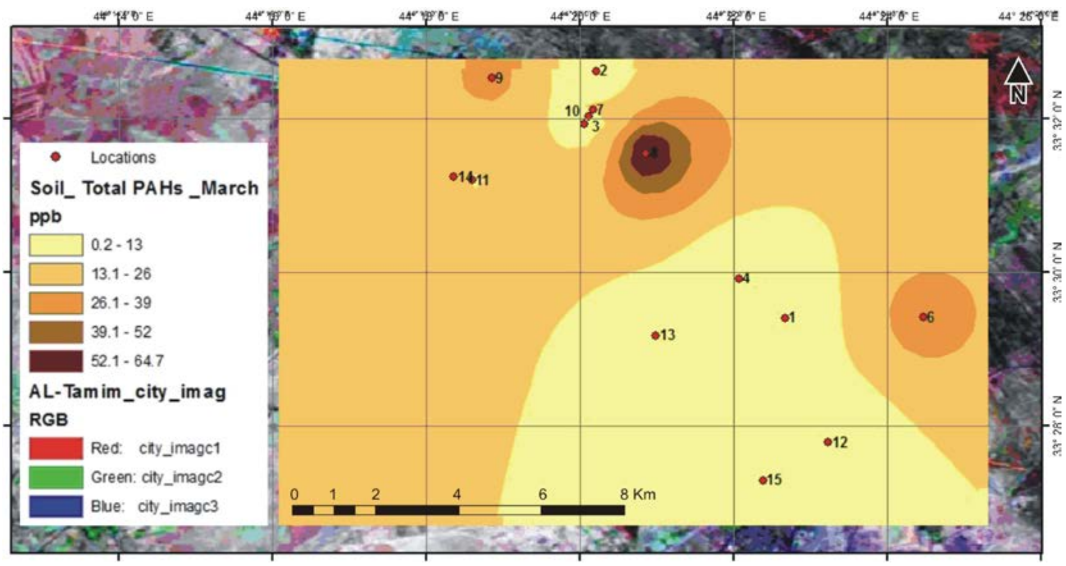

Fig.3: Arc GIS model of total PAHs pollutants of cumulative soil model of PAHs of both periods (total) October 2010 and March 2011

CONCLUSIONS

- Comparison of the average concentration of the heavy metals Pb, Ni, Cr, and Cu with results of other studies shows that they are within or lower than these limits, except for the concentration of Cd which is higher at the two periods.
- The results of PAHs indicate that in March 2011 they are higher due to the increase of fuel combustion operations occurring at the location, such as the operations of the power plant that functions increasingly during the winter months and due to atmospheric contaminants (dust and smoke) enriching the soil surface. This dependence can be explained by the relative high amounts of rain in March.
- The cumulative effects model of PAHs using GIS for both sampling periods show an increase in the direction away from the refinery indicating that the wind direction and that the refinery was not the only contaminating source but there are other sources such as the Kirkuk main vehicle station and electrical generators exhausts.

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