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THE CHEMISTRY OF THE LEAVES OF PLANT *Eucalyptus camaldulensis* AS ENVIRONMENTAL CONTAMINATION INDICATOR OF SELECTED LOCATIONS AT KIRKUK - IRAQ

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ABSTRACT

The environmental contamination by the polycyclic aromatic hydrocarbons (PAHs) and heavy metals (Pb, Cu, Ni, Cr, and Cd) concentrations in the leaves of plant *Eucalyptus camaldulensis* were determined at the city of Kirkuk in 15 selected locations using GPS. The pickings up of samples were carried out in two periods October 2010 and March 2011. Compared with results of other studies, the concentration levels of determined heavy metals show values within these studies results. The average total concentration of polycyclic aromatic hydrocarbons (PAHs) in the leaves of Plant *Eucalyptus camaldulensis* indicated 37.1 ppb in October, while in March 165.2 ppb.

The model of cumulative effects of heavy metals and PAHs, which were determined by GIS for both sampling periods showed distribution of concentration towards wind directions and away from the site of Kirkuk Oil Refinery at the south east direction and that the refinery was not the only contamination source but there were other sources such as the Kirkuk main vehicle station on the edge of the urban areas.

Key words: Plant *Eucalyptus camaldulensis* leaves, contamination, Heavy Metals, PAHs, Kirkuk, Iraq.

INTRODUCTION

There is a high probability of contamination by the polycyclic aromatic hydrocarbons (PAHs) and heavy metals, due to the availability of sources of pollution. Plant’s leave 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, oil burning, waste incineration (Thorpe and Harrison, 2008). Heavy metals are that elements having specific gravity that is at least five times the specific gravity of water which is expressed as 1 at 4°C and refers to metallic elements with an atomic weight greater than iron (55.8 g/mol).

The composition and quantity of chemical matrix of road, highways dust and cement plants are indicators of environmental pollution with high concentration of Pb, Cu, Ni, Cr, and Cd (Kabata, and Pendias, 2001; Al-Sayegh and Al-Yazichi, 2001; Awadh, 2009 and 2014).

Determination of chemical composition of plants is one of the most frequently used methods of monitoring environmental pollution. Various plants have been used as bio-indicators to assess the impact of a pollution source on the vicinity which is due to high metal accumulation of plants (Lawal *et al*., 2011, Assadi *et al*., 2011). Generally, the climate of Kirkuk is semi-arid with maximum temperature up to 53ºC in July- August. 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. Besides, the used cars which are second hand remained in service. This exerts a heavy pressure on its urban environment. Exposure to heavy metals in road dust can occur by means of ingestion, inhalation include respiratory system disorders, nervous system interruptions, endocrine system malfunction, immune system suppression and the risk of cancer in later life (Ferreira-Baptista and Miguel, 2005). Because the Plants are collectors for all air pollutants, and their chemical composition may be a good indicator for contaminated-areas. *Eucalyptus camaldulensis* tree species were chosen. It has relatively high growth rate in Kirkuk and it attains heights between 25 and 35 m (Mohammed, 2009; Al-Edany, *et al*, 2012).

Heavy metals are non-biodegradable, hence are not readily detoxified and removed by metabolic activities once they are available in the environment. This may subsequently lead to their build up to toxic levels or bioaccumulation in ecosystem (Al-Hamadani 1987; Jan *et al*., 2000; Buszewski *et al*., 2000; Yongzhen *et al*., 2011).

The polycyclic aromatic hydrocarbons (PAHs) in the environment largely are a product of the incomplete combustion of petroleum, oil, and coal. Sources in the urban environment include industrial emissions and wastes, power plants, vehicles, mineral/crude oil extraction and petroleum refining processes and the majority of PAHs are suspensions (Masitah *et al*., 2007; Kluska 2013). Polycyclic aromatic hydrocarbons (PAHs) were ranked as the ninth most threatening compounds to human health. It is conceivable that faster deposition of exhaust aerosol droplets occurs close to the highway; while further spreading mediated by their adsorbed form on the dust particles that are distributed with wind, affects other media (e.g. air, water, soil and plants) (Samimi *et al*., 2009; Habib *et al*., 2012). Researchers had identified 16 different "priority pollutants" PAHs which have stronger toxicity than others and can be adsorbed by the leaves of plants (Venkataraman *et al*., 2002; Bari *et al*., 2011). Plants' leaves may accumulate an immense amount of pollutants, including heavy metals and PAHs. Most of these compounds are transported by air with particles of dust. (Bryselbout *et al*., 2000; Zitka *et al*., 2012). Many researchers who studied soil, water and plants in Kirkuk have detected high heavy metals concentration in different sampling media (Haqus and Hammed, 1986; Al-Khashman, 2007; Awadh, 2009; Ali, 2013). This work is expressed as geochemical study for further contribution of the local environmental pollution; it is going to discuss the concentration and distribution of the heavy metals and PAHs in Kirkuk plant *Eucalyptus camaldulensis* leaves in attempt for revealing the source of heavy metals.

The importance of this research lies in the risk pollutants and their impact on the public health. Therefore, it aims to measure the concentration of heavy metals (Pb, Cu, Ni, Cr, and Cd) and PAHs in the leaves of plant Eucalyptus for two periods October 2010 and March 2011, as well as the assessment of environmental impacts of the Kirkuk city.

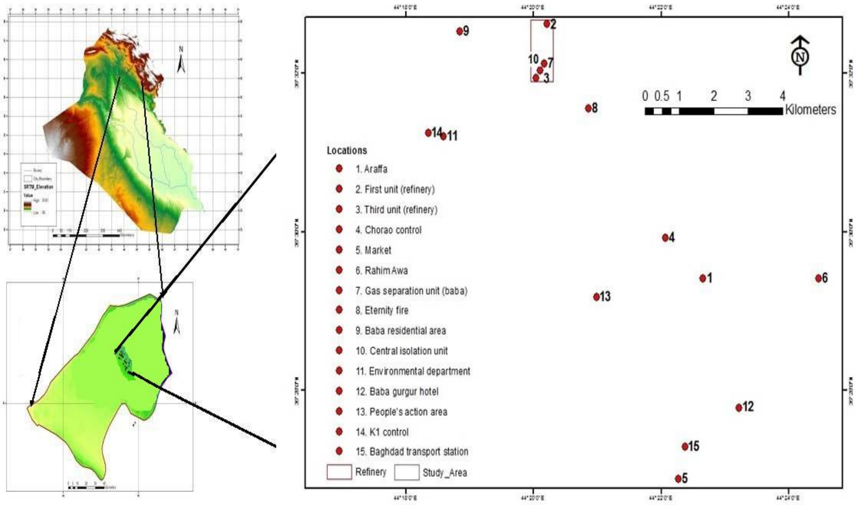

Fig. 1: The topographic map of Iraq showing the location of Kirkuk governorate and the sampling sites.

MATERIALS AND METHODS

A-The heavy metals in the leaves of plant *Eucalyptus camaldulensis* were analyzed. Fifteen different sampling sites inside and outside Kirkuk oil refinery have been selected between latitudes (35° 24" – 35° 29") to the north and longitude (44° 20" – 44° 26") to the east, (Fig.1). The site selection of the leaves of plant Eucalyptus sampling in Kirkuk Oil Refinery and around takes into consideration the prevailing wind direction that is an important factor in pollutants distribution, as well as the nearby populated sectors within Kirkuk city. The concentrations of the heavy metals (Pb, Cu, Ni, Cr, and Cd) were analyzed using Atomic Absorption Spectrometry equipment (Buszewski *et al*., 2000).

B-The polycyclic aromatic hydrocarbons (PAHs) in the leaves of plant (Eucalyptus) were determined by using HPLC (High Performance Liquid Chromatography) and GC-MS (Gas Chromatography Mass) of the polycyclic aromatic hydrocarbon compounds according to Henner *et al*., (1997) and Husain (2003) procedures. 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 materials, (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, (Henner *et al*., 1997; Ali, 2013).

C-The Geographic Information Systems (GIS) was applied. Arc GIS 10 modeling of measurements of heavy metals and the polycyclic aromatic hydrocarbons (PAHs) accumulated on the leaves of plant *Eucalyptus camaldulensis* was applied for the cumulative effects of both sampling periods October 2010 and March 2011.

RESULTS AND DISCUSSION

Heavy Metals Analyses in Plant

Obviously, the leaf plant tissues store the greater quantity of heavy metals. Nickel (Ni) can be found in high concentration around areas contained nickel-cadmium batteries and as product of diesel fuel. Cobalt (Co) can be found in diesel and gasoline fuel; in sludge, and commercial fertilizers that use, in addition to that many industrial sites occurring in Kirkuk as well as Kirkuk oil refining. The vehicle exhausts in heavy traffic are the main source of Pb and Cd (Mohammed, 2009). The anthropogenic sources of Cd compounds are production processes of zinc, copper and lead combustion processes of oil and incineration. Road dust receives varying inputs of heavy metals from diversity of mobile or stationary sources such as vehicular emission, industrial plants, power generation plants, oil burning, waste incineration, as well as re-suspension of surrounding contaminated soils (Ahmed and Ishiga, 2006; Al-Khashman, 2007).

Nickel compounds are mainly emitted by combustion (heavy residual oil burning units). Nickel compounds such as nickel sulfate, oxidic nickel, and nickel metal are the predominant species in stack fly ash from oil-fired combustion. The anthropogenic source of Pb is leaded gasoline in most as well as the other varied sources emitting from fuel combustion, and shops of radiator repairer.

The range and the mean concentrations of heavy metals in the leaves of plant *Eucalyptus camaldulensis* for October 2010 and March 2011 samples are shown in Table 1. The results during October 2010 reflect that the mean concentration of Pb, Cu, Ni, Cr, and Cd were 3.4, 4.1, 6.1, 54.2, and 12.0 ppm respectively, with the descending order of heavy elements in plant leaves as Cr > Cd > Ni > Cu > Pb. While during March 2011, the mean concentration of them was 1.4, 3.0, 37.7, 46.3, and 6.6 ppm respectively, with the descending order of heavy elements in plant leaves as Cr > Ni >Cd > Cu > Pb.

The results indicate that the studied heavy metals were relatively lower in March than in October, (Table 1). It is believed that such finding is obvious due to high wind speed and rainfall during the winter months (i.e. March) which wash and remove the pollutants from the leave surface.

Comparison of the studied heavy metals with other studies on other plants results (Khuwaidem, 2007; Habib, *et al*., 2012) referred that they are within or lower than their concentrations, Table 1.

Table 1: Heavy metals concentrations (ppm) in the leaves of plant *Eucalyptus camaldulensis* for October 2010, and in March 2011, compared with results of other studies.

Heavy Metals Conc.	Sample No.	Pb ppm	Cu ppm	Ni ppm	Cr ppm	Cd ppm
October 2010	Range	0.2-10.0	0.02-10.0	1.0-18.0	0.01-104.5	4.3-17.8
	Mean	3.4	4.1	6.1	54.2	12.0
March 2011	Range	0.01-4.0	0.01-10.3	16.3-54.8	20.7-88.4	3.6-10.5
	Mean	1.4	3.0	37.7	46.3	6.6
Khuwaidem , 2007	Range	4-42	10-35	5-14	4-12	7-13
	Mean	21.5	22	9	8.1	10
Habib, *et al*., 2012	Range	20-40		30-60		12-20
Dates leaves	Mean	30		45		16

Applying Arc GIS 10 model of heavy metals concentrations on the leaves of plant *Eucalyptus camaldulensis* for the cumulative effects of both sampling periods October 2010 and March 2011, shows that the concentrations of these pollutants distribute away from the refinery toward the wind direction and the refinery is not the only contamination sources, as in sampling site 4 and site 12 that represent the Chorao Control site and Baba gurgur hotel site respectively Figure 2. Moreover, the distribution of heavy metal concentration is displayed around Garage in which the gases from automobiles are continuously emitting; this led to suggest that the garages and crowded traffic area were a point and non point source respectively. These heavy metals could come from many different sources.

These sites indicate maximum recorded values due to traffic intensity as they represent the main bus and cars station sites, where the vehicle exhausts contain heavy metals concentrations and can be directly deposited on the leaves of *Eucalyptus camaldulensis*, Figures 1 and 2. It is clear that the pollutant affected by the prevailing wind direction (NW) was a controlling factor in transferring the pollutants. Pollutants (Pb, Cu, Ni, Cr, and Cd) are added to the road-side dust in quantities equivalent to the amount of gas emitted from automobiles, corrosion and wear of vehicle parts as well as atmospheric additives. The distribution pattern of the pollutants depends on the energy and direction of the wind.

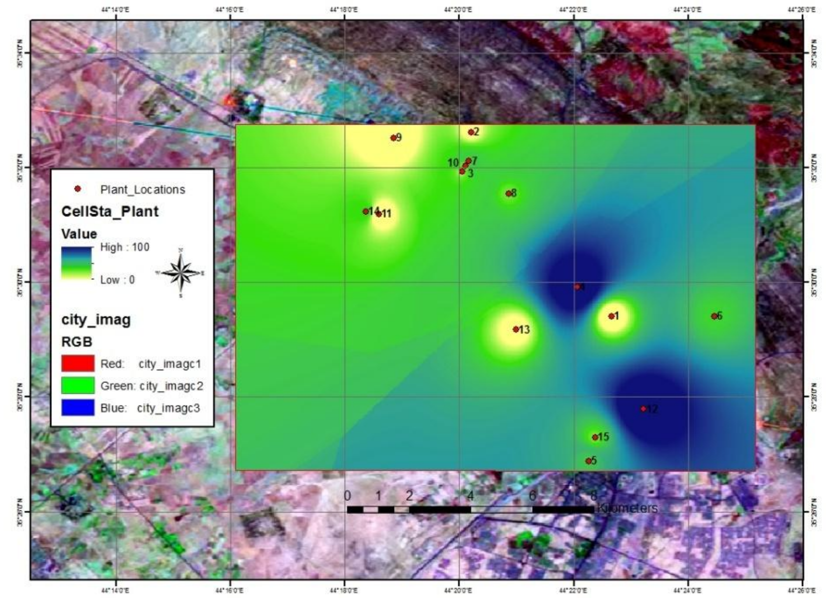

Fig. 2: Arc GIS cumulative model for total heavy metals in the leaves of plant *Eucalyptus camaldulensis* for both periods October 2010 and March 2011.

Total PAHs in Plant:

The results of the polycyclic aromatic hydrocarbons (PAHs) concentrations in the leaves of plant *Eucalyptus camaldulensis* show the existence of sixteen hydrocarbons, these are Naphthalene, Acenaphthene, Acenaphthylene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benzo (a) anthracene, Chrysene, Benzo (b) Fluoranthene, Benzo (k) fluoranthene, Benzo (a) pyrene, Dibenzo (a, h) anthracene, Benzo (g, h, i) perylene, Indeno (1,2,3-cd) Pyrene, Table 2. The 16 EPAs priority PAHs detected in the studied area were varied in concentration within the sites of measurements due to the physicochemical properties of these compounds.

Most of PAHs minimum values were less than 1.0 ppb during October 2010, except five Naphthalene 13.0 ppb, Acenaphthene 2.4 ppb, Phenanthrene 1.0 ppb, Benzo (a) pyrene 2.3 ppb and Dibenzo (a,h) antracene 2.3 ppb .While, most of their maximum values were less than 15.0 ppb except for six Naphthalene 67.7 ppb, Acenaphthene 75.0 ppb, Fluorine 33.2 ppb, Phenanthrene 15.3 ppb, Pyrene 27.4 ppb and Dibenzo (a,h) antracene 22.5 ppb, Table 2.

In March, 2011, most of PAHs minimum values were less than 6.0 ppb, except two Naphthalene 49.7 ppb, and Acenaphthene 18.4 ppb. While, most of their maximum values were less than 15.0 ppb except for six Naphthalene 63.7 ppb, Acenaphthene 129.4 ppb, Fluorine 187.5 ppb, Phenanthrene 28.9 ppb, Pyrene 45.2 ppb and Dibenzo (a,h) antracene 17.2 ppb, Table 2.

Table 2: Total polycyclic aromatic hydrocarbons (PAHs) concentrations (ppb) on the leaves of plant *Eucalyptus camaldulensis* for October 2010 and March 2011.

No.	PAHs		October 2010		March 2011	
			Min	Max	Min	Max
1.	Naphthalene	NAP	13.0	67.7	49.7	63.7
2.	Acenaphthylene	ACY	0.0	3.8	0.0	3.5
3.	Acenaphthene	ACE	2.4	75.0	18.4	129.4
4.	Fluorine	FLU	0.3	33.2	5.3	187.5
5.	Phenanthrene	PHE	1.0	15.3	2.3	28.9
6.	Anthracene	ANT	0.01	2.0	0.02	5.0
7.	Fluoranthene	FLUA	0.07	3.6	0.1	8.7
8.	Pyrene	PYR	0.7	27.4	0.2	45.2
9.	Benzo(a)anthracene	B(a)A	0.3	4.4	1.2	4.3
10.	Chrycene	CHR	0.3	3.1	0.5	4.0
11.	Benzo(b)fluoranthene	B(b)F	0.4	1.0	0.07	6.1
12.	Benzo(k)fluoranthene	B(k)F	0.6	6.2	0.02	1.0
13.	Benzo(a)pyrene	B(a)P	2.3	9.0	0.6	12.9
14.	Dibenzo(a,h)antracene	Dib(ah)A	2.3	22.5	3.8	17.2
15.	Benzo(g,h,i)perylene	B(ghi)P	0.5	7.0	2.2	7.3
16.	Indino(1,2,3-cd)pyrene	Ind P	0.0	0.03	0.0	1.4

The total accumulated PAHs in the leaves of plant *Eucalyptus camaldulensis* of each sampling sites are shown in Table 3. Most of total PAHs values were less than 20.0 ppb during October 2010, except for sites 4, 8, 10, 13 ,14 and 15 that their values reach 153.1, 67.8, 105.7, 59.2, 32.6 and 83.5 ppb respectively. In March, 2011, most of total PAHs values were less than 50.0 ppb, except for sites 1, 5, 6, 9, 12 and 15 were the PAHs values reach 50.6, 78.2, 99.4, 298.5, 221.0, and 51.3 ppb respectively, Table 3.

The maximum concentration of PAHs as a total accumulation in the leaves of plant *Eucalyptus camaldulensis* at October was 153.1 ppb at site No.4 (Chorao control site). While, the lower value of PAHs was 0.0 ppb at site No.2 (First unit at the refinery), with average 165.2 ppb, Table 3.

The maximum concentration of PAHs as a total accumulation in the leaves of plant *Eucalyptus camaldulensis* at March was 298.5 ppb at site No.9 (Baba residential area). While, the lower concentration of PAHs was 15.3 ppb at site No.13 (People's action area), with average 37.1 ppb, Table 3. The total concentrations of PAHs at March (165.2 ppb) was higher than at October (37.1 ppb) which could be due to the decreasing of the air temperatures winter, and evergreen plants scavenge the majority of emitted PAHs (Jan *et al*., 2000).

Comparison of the total PAHs on the leaves of plant *Eucalyptus camaldulensis* of each sampling site with other studies results indicates that the results were within the range or less than the other studies results ,Table 3, (Slaski *et al*., 2000; Ideriah *et al*., 2011).

Table. 3: Total PAHs concentrations (ppb) in the leaves of plant *Eucalyptus camaldulensis* for October 2010 and March 2011.

Sampling Site No.	Total PAHs conc. in Plant October 2010	Total PAHs Conc. In Plant March 2011
1.	4.0	50.6
2.	0.0	47.1
3.	15.8	27.1
4.	153.1	20.7
5.	17.9	78.2
6.	4.4	99.4
7.	4.4	15.7
8.	67.8	20.3
9.	14.2	298.5
10.	105.7	16.2
11.	2.3	16.2
12.	4.6	221.0
13.	59.2	15.3
14.	32.6	33.8
15.	83.5	51.3
Range	0.0-153.1	15.3-298.5
Average	37.1	165.2
Slaski, *et al*., 2000	Range	40-477
Ideriah, *et al*., 2011	Range	142.4-582.2

Moreover, Arc GIS 10 modeling of measurements of total accumulated PAHs in the leaves of plant *Eucalyptus camaldulensis* was applied for the total cumulative PAHs effects of both sampling periods October 2010 and March 2011, Figure 3. The result shows that the Kirkuk oil refinery was not the main source of the PAHs pollutants that distribute away from the refinery toward the south west direction but also ,it is noticed that PAHs increased in site No.4 (Chorao control) and site No.13 (people's action area) which reflect the PAHs high values due to traffic intensity as they represent the main bus and cars station sites, where, the vehicle exhausts contain PAHs concentrations and can be directly deposited on the leaves of Eucalyptus, Figure 3. Also, the meteorological condition (e.g. rainfall, wind direction and dust storms) plays an important role in distributing the total PAHs on the leaves of plant *Eucalyptus camaldulensis*.

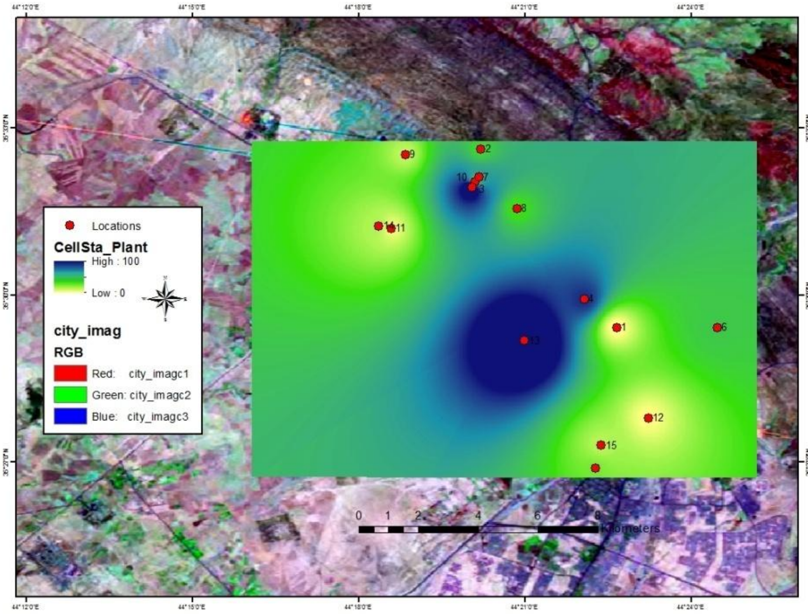

Fig. 3: Arc GIS model of total PAHs accumulation in the leaves of plant *Eucalyptus camaldulensis* at the studied area of both periods October 2010 and March 2011.

CONCLUSIONS

1-Comparison of the averages of the heavy metals Pb, Cu, Ni, Cr, and Cd with other studies referred that they are within or lower than the references values.

2-The results of PAHs indicate that in March 2011 (165.2 ppb) was higher than at October (37.1 ppb) due to the increase of fuel combustion operations accruing at the location such as the operations of the power plant that functions increasingly during the winter months and atmospheric contaminants (dust and smoke) are cumulated on the Plant surface. Comparison of the total PAHs in the leaves of plant *Eucalyptus camaldulensis* of each sampling site with other studies indicate that this study results were within the range or less than the other studies results.

3-The cumulative effects model of PAHs by using GIS for both sampling periods shown the distribution at the direction away from the refinery mostly at the south east direction and that the refinery was not the only contamination source but there were other sources such as the Kirkuk main vehicle station and electrical generators exhausts.

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كيميائية اوراق نباتات الكالبتوس كامالدولينسيس كدليل للتلوث البيئي لمواقع مختارة في مدينة كركوك — العراق

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**وزارة العلوم والتكنولوجيا — بغداد — العراق

الخلاصة

لقد تم تحديد التلوث البيئي لمعدل تراكيز العناصر الثقيلة (Pb,Cu,Ni,Cr,Cd) والهيدروكوربونات العطرية المتعددت الحلقات (PAHs) في نماذج اوراق نبات الكالبتوس كامالدولينسيس الماخوذ من ١٥ موقعاً مختاراً من مدينة كركوك باستخدام الـ GPS. ان جمع النماذج كان لفترتين هي تشرين الاول ٢٠١٠ وآذار ٢٠١١.

ان معدل تركيز الهيدروكوربونات العطرية المتعددة الحلقات (PAHs) في نماذج اوراق نبات الكالبتوس كامالدولينسيس كان ٣٧,١ جزء من البليون في تشرين ٢٠١٠ وفي آذار ٢٠١١ كان ١٦٥,٢ جزء من البليون.

اظهرت النتائج استخدام تاثير الهيدروكوربونات العطرية المتعددة الحلقات التراكمي حددت بتطبيق برنامج GIS لفترتي الدراسة بان تراكيزها تنتشر باتجاه الرياح السائدة الى جنوب الشرقي مبتعدة عن المصفى وان المصفى لم يكن المصدر الوحيد للتلوث حيث تلعب محطة وقوف السيارات والباصات العامة الواقعة على جانب المناطق السكنية دوراً كبيراً في زيادة التلوث.