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Concentration of Total petroleum Hydrocarbons and Selected Heavy Metals In Soil And plant nearby Al-Nassiriya oil Refinery South of Iraq

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Abstract:-

The present study deal with measure concentration of Total petroleum hydrocarbons (TPHs) and selected heavy metals (HMs) (Cd , Pb , Co , V, and Ni) in soil and plant *phragmits austriles* (cav.) nearby AL-Nassiriya oil refinery during the period extended from winter up to summer 2018 . Some environmental factors such as , pH, salinity‰ , Electrical conductivity (Ec.) and soil temperature were measured , also total organic carbon (Toc) % content in soil was determined, and soil texture was analyzed and both of them expressed as percentage . The result observed that the range of the following factors were : soil temp. ranged from (26-36) ◦c , pH (7.3-7.9) , Ec. (14.3-25.3) µs/cm and salinity (5.1-7.3)‰ , while the value of Toc ranged from (0.7-8.55)% , soil texture was silty soil in control station , but they were sandy soil at polluted stations . The mean concentration of TPHs in soil , St.1 , St.2 , St.3 and St.4 were (16.47 ,14.78 ,12.65 and 1.36) µg /gm dry weight respectively , while the mean concentration of HMs in the soil for both season were Cd (0.78), Pb (26.85), Co (26.83), Ni (227.17) and V (95.67) µg /gm dry weight . In plant (shoot and root) part , mean concentration of TPHs were (4.86 and 8 .44) µg /gm dry weight respectively ,whereas the mean concentration of heavy metals in both parts for both seasons were Cd (0.49 and 0.65) , Pb (30.36 and 49.25) , Co (3.08 and 5.36 , Ni (8.01and 20 .38) and V (1.10 and 1.94) µg /gm dry weight respectively . It concluded from the present study that the oil refinery waste discharge affected upon the soil and plant of the study area through the increase in concentration of TPHs and HMs in its, also significant correlation observed between both pollutants and Toc content.

Keywords:- Total hydrocarbons ,Heavy Metals , Soil , *Phragmites austriles* , Oil refinery Thi-qar Iraq.

Introduction:-

Soil is important part of land ecosystem, It is exposure to many types of pollutants among which are petroleum hydrocarbons and heavy metals (AL-saad, 2016) . The release of crude oil in to the environment by oil spill is receiving worldwide attention . Crude oil products represent the one of the most common environmental pollutants and most oil pollution sources are anthropogenic such as motor fuel stations, underground storage tanks, home and commercial heating oil storage tanks, fuel distribution centers, crude oil production sites, accidental spill and refineries (Marineseu, *et al*., 2011). The accumulation of oil hydrocarbons in the soil of terrestrial ecosystem, also cause health risks for human and animals as well (Prakash and Irfan, 2011). Petroleum pollution is changing the physical, chemical and biological properties of the soil, thus changing nutrient values which sustain plant growth (Agbogidi, *et al*.,2006). Heavy metals are elements having atomic number more than twenty, atomic weight between 63.46 and 261 and specific gravity greater than 5 gm/cm^3 (Al-Doghachi, 2008). Heavy metal pollution it is a global problem, although severity and level of pollution differ from place to place In a small quantities, certain heavy metals are nutritionally essential for health life, but large amount of any of them may cause acute or chronic toxicity (poisoning) (Al-Awady, 2012). Concentration of heavy metals in soil are associated with biological and geochemical cycles, they are influenced by anthropogenic activities such as, transport ,waste disposal, industrialization social and agricultural activities have an effect on environmental

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pollution and the global ecosystem (Oyem and lowrence ,2013). Soil is exposure to petroleum pollution specially that which situated nearby the oil establishments. Al-Nassiriya refinery was established since (1980) and from this time till now its waste discharge on the soil close to it (personal communication with director manager of Al - Nassiriya refinery) . There are few studies about soil pollution by oil in AL-Nassiriya city, so the present study deal with effect of waste discharge from oil refinery in Al - Nassiriya city upon the soil properties close to the refinery. The major objective of this study was to provide information about distribution and concentration of heavy metals (Cd , Co, Ni , V and Pb), and total petroleum hydrocarbons in soil close to Al – Nassiriya oil refinery, these data may be of use to other researchers who require a baseline for comparison of petroleum hydrocarbons and heavy metals concentration and distribution.

Study area

The study area included 4 station on the soil close to Al –Nasiriya oil refinery by using GPS.72 Griming as follow:

St_1 :	N 30° 59 22.5	St_3 : N 30°	59 21.5
	E 46° 13 24.4	E 46°	13 28.3
St_2 :	N 30° 59 21.1	St_4 : N 30°	59 25.3
	E 46° 13 21.9	E 46°	13 29.1

The distance in between the stations (1,2,3) was about 20m , while fourth station act as control station , it was 500 m far from the mentioned stations (Fig.1).

Fig.(1): Map of the study area showing the study stations. (by Google)

Material and Methods:-

1. Samples collection

Soil Samples were collected in triplicate by mean of steel scope from about 10 cm depth from soil surface from each station during the period extended from winter and summer 2018, while plant *P. austriles* (cav.) was collected randomly from the study area in the same time.

Methodology:-

1- EC. pH and salinity of soil were measured according to (APHA , 2003) .
2- Total organic carbon (TOC) content in soil was determined according to (Ball,1964) as percentage by use two grams of soils dried and sieved by sieve with <63 µm mesh size , were put in pre –weight crucible and burned by furnace at 550˚c for 48 hrs , the differences in mass of crucible and soil sample before and after burning was calculated as Toc content .
3- Grain size analysis : grain size analysis of soil was carried out using the combined dry sieve and pipet techniques according to Folk (1974) method . The grain size (sand ,silt and clay) of soil determined as percentage .
4- Extraction and determine of TPHs:

In lab. soil samples dried in oven at 80 ◦c for 48 hrs , ground finely in agate morter and sieved throw 63 µm metal sieve . The extraction procedure of petroleum hydrocarbons residues in soil employed was based upon that of (Goutx and saliot , 1980) , while extraction of TPHs from the plant *P. austrillus* ,exactly 10 gm of dried parts of plant (shoot and root) were extracted employed procedure of (Goutx and saliot ,1980). Concentration of TPHs in all samples (soil and plant parts) solution prepared after extraction , Were measuring by shimadzu - RF 540 .

5- Extraction and determine of HMs :

Heavy Metals extraction from soil station were performed on the <63 µm fraction of the soil , which had been separated by sieving after dried by 80 °c in oven and grinding . The extraction of HMs from soil were done following the procedure described by (Sturgeon, *et al*., 1982) , while the extraction of HMs from the plants was employed the procedure of (Barman, *et al*., 2000) . 1 gm of specimen (shoot and root) part , dried , ground and sieved by plastic sieve with 40 µm mesh size , acids mixture (HNO_3 and $HClO_4$) 4:1 were used to digestion the plant tissues HMs (Cd, Pb , Ni , Co and V) were extracted in triplicates from the two parts of the plants. The mentioned HMs were determined in soil and plant tissues by using air / acetelyene Atomic abosorption spectrophotometry (Flam AAS.) Model SP9.pye-unicam.Blank values negligible for all studied metals, acid used were ultrapure and water was deionized distilled water (DDW) . ANOVA test were done to know the significant differences between parameters by using Minitab program.

Results and discussion:-

Soil parameters value were presented in Table (1). High temperature values (26.4 and 36.8) °c were observed at st.4. in winter and summer season respectively. The mention values due to the time of measure, st.4 is the last station in measure , also the polluted stations recording high temperature values specially at st.3, this due to the existance of oil hydrocarbons and their reactions in the soil . pH values were in alkalin side for all stations and the high value 7.9 was recorded at st.4 in the summer, also st.4 and 3 in winter and summer respectively recorded high values . Crude oil spill on the soil of Thi-Qar refinery influenced the pH values in the soil, but there is no significant differences in the pH values in polluted soil and control . Oil pollution may have had some direct impact in lowering in pH, it is also possible that microbial actions through metabolic processes contributed to changes in pH by producing organic acids (Manahan,1994) . The E.C. values were higher in the polluted stations more than its value in control station (st.4). The higher values were (23.4 and 25.3) µs/cm recorded at st.2 for both season respectively, while in control station (st.4) (non - polluted) the values of E.C. were (14.3 and 15.5) µs/cm in control station (st.4) for both season respectively . The higher values of E.C. in polluted station refer to high presence of changed ions (cations and anions) in the soil . The values electrical conductivity represent the ratio of soil salinity , so the key to determine soil salinity is to obtain electrical conductivity (Oyem and Lawrence ,2013) . Salinity was recorded high value 7.3‰ at st.1 in the summer ,whereas the low value 5.1% was recorded at st.4 in winter . The higher values of salinity were recorded at polluted station for both season . The flactuation in salinity values among the study station refer to disappearance of plants in polluted station , is not due to the decrease or increase of salinity level (sonon *et al*.,2015) .oil pollution has deleterious effect on soil by changing the physical and chemical properties and hence a significant effect on the growth of the plants (Abogidi , *et al*., 2007).

Table (1) mean values ±SD. of soil parameters in the study station during the study period.

parameter / station	Temp. °C	pH	E.C. µs/cm	salinity %	
Winter season					
st.1	26±0.02a	7.4±0.02a	20.3±0.5a	6.5±0.3a	
st.2	26±0.01a	7.2±0.01a	23.4±0.3b	6.2±0.05b	
st.3	26.2±0.03a	7.7±0.03a	17.5±0.3c	5.3±0.2c	
st.4	26.4±0.01a	7.8±0.01a	14.3±0.01d	5.1±0.1c	
Summer season					
st.1	30.1±0.02a	7.8±0.03a	25.0±0.5a	7.3±0.51a	
st.2	32.5±0.4b	7.7±0.03a	25.3±0.3a	6.5±0.8b	
st.3	36±0.22c	7.8±0.05a	20.56±0.5b	7. 2±0.71a	
st.4	36.8±0.8c	7.9±0.2a	15.5±0.5c	6.4±0.51c	

- Each value represent mean ± standard deviation
- The similar letters of each parameter refer to non-significant differences P<0.05.

Mean concentration of total petroleum hydrocarbons (TPHs) in soil and plant *P. austriles* (Root and Shoot) part , were observed in Table (2). TPHs concentration in soil found to vary from 0.79 at st.4 to 17.6 Mg/gm dry weight at st.1 in winter season, while, it's concentration in summer season vary from 0.79 Mg/gm dry weight at st.4 to 15.33 Mg/gm dry wt. at st.1. it showed higher level of TPHs in soil were recorded in winter more than the summer season, this due to the weathering process, such as light and temperature which led to vapour of hydrocarbons in the summer (AL-Khafaji,2007). High content of TOC in the study stations Fig (2) play an important role to increase the

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soil absorbed capacity to TPHs, statistical analysis showed good correlation r=0.880 P ≤ 0.05 between TPHs concentration and Toc in the soil, this refer that TOC affected upon TPHs concentration in the soil (zebral, *et al*.,2013). Soil textures for all station were send with acceptation of st.4 control station was muddy (Table,3) . This due to that the management of the refinery using sand soil time to time to cover the spill for treat and hidden its (Refinery manager personal communication). Low concentration of TPHs and Toc % content were recorded at st.4, while soil texture was muddy at this station, this due to the situation of the mention station was far from the effect of refinery spill and it's gases emission, because it's site was against of the prevailing wind direction. Plant *P. austriles* concentrated TPHs in their parts (Root and Shoot), higher concentration of TPHs was recoded in root part, while the lower concentration was observed in shoot part for both season , but concentration of TPHs in the studied parts of the plant was higher in summer and lower in winter, this due to the effect of many weathering process such as, temperature also some process which done by plant such as, transpir ation and transformation by metabolic activaties. In this study concentration of TPHs in the soil exceed the permissible level of these compounds in the soil as per the relevant soil quality standards (Trench, *et al*.,1994).

Table (2) mean concentration ± S.D. µg/gm dry weight of Total petroleum hydrocarbons (TPHs) in soil and plant *P. australis* (Root and shoot part) in winter (A) and summer (B) at the study stations

(A)			
structure / Station	soil	*P. austriles*	
		Root	Shoot
St.1.	17.6±0.35a	9.2±0.5a	6.4±0.05a
St.2.	15.32±1.22b	6.3±0.22b	4.35±0.31b
St.3.	13.0±2.31c	8.11±0.31a	3.16±0.05c
St.4.	1.93±0.02d	0.68±0.01c	0.78±0.02d
(B)			
St.1	15.33±0.5a	10.3 ±1.31 a	6.2±0.03 a
St.2	14.23±1.21 b	7.4±0.51 b	5.41±0.02 b
St.3	12.3±0.66 c	9.3±0.70 c	3.66±0.01 c
St.4	079±0.02 d	0.88±0.01 d	0.65±0.03 d

*The similar letters of each parameter refer to non- significant differences p<0.05.

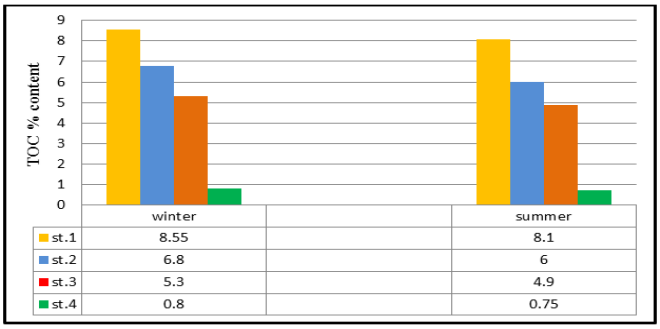

Fig (2) Total organic carbon (Toc %) content in the soil of the study area during the study seasons

Table(3) Soil texture % and its description in the study stations during the study period.

	Winter season				Summer season			
Texture / station	Send	Silt	clay	description	Send	Silt	clay	description
St.1	40.4	30.6	29.0	Sandy	40.0	31.2	28.8	Sandy
St.2	44.3	28.2	27.5	Sandy	43.3	28.2	28.2	Sandy
St.3	45.0	27.9	27.1	Sandy	44.2	28.5	27.3	Sandy
St.4	20.3	39.1	45.6	Muddy	23.5	38.1	44.2	Muddy

Concentration of heavy metals in soil and plant *P. austriles* in both (shoot and Root) µg/gm dry weight during winter and summer season, are presented in Table (4 and 5) respectively. Soil recorded high concentration of HMs in both season , because soil is the source and origin of thes types of pollutants (Al-Khafaji, 2017) . Higher concentration of studied Metals were observed in root part, the order of HMs concentration in the root part were Cd<Pb<Co<Ni<V, high mean concentration was 232.2 µg/mg dry weight recorded by Ni for both seasons, while the lower mean concentration was 0.78 µg/mg dry weight recorded by Cd for both seasons, crude oil content many substances among which heavy Metals, specially Co, Ni, V, Pb, and Cd . Many studies specially in aquatic environment used Ni and V as indicators for oil pollution (AL-Khafaji,2007). Heavy Metals have negative effect start by adsorbed by soil and absorption by plants and less to lesser extent by ground water contamination through leaching (Dawaki, *et al*.,2013) .All Heavy Metals in soil of present study recorded high concentration at st.1,2, and 3, whereas record low concentration at st.4 (control st.), this indicate that oil refinery effected upon the soil nearby it's. and provide it's with heavy metals through the waste discharge. High concentration in the study

stations (st.1,2, and 3) due to the presence of industries specially oil refinery, as well as contribution of heavy traffic of motor vehicles such as oil tankers, which exist every time to bring crude oil to the refinery. Maldonado, *et al.*, (2008) indicated that the higher concentration of Cr, Zn, Pb, Ni, Cd and Co in the soil of some cities under the study due to presence of industries and domestic sewage . Concentration of most heavy metals under the study passed the maximal permissible metals concentration according to Dutch ecologists (crommentuijn, *et al.*, 1997). Significant correlation $r = 0.653$, $r = 0.088$, $r = 0.85$, $r = 0.79$ and $r = 0.80$ $p \leq 0.05$ were observed between TOC and Metals concentration (Cd , Pb , Co , Ni and V) in the soil . Bioconcentration is often a good integratine indicator of the chemical exposures of organisms (among which are plants) in polluted ecosystem (phillips and Rainbow ,1994) . Pollutants entering soil interact with its active phase (clay minerals ,oxides and hydroxides of iron and manganese and organic substance and change own activity either increasing or decreasing their hazard (Fairs, *et al.*,2018) .*P. australis* showed high concentration to heavy metals in both parts , this mean that the mention plant has high ability to accumulate these type of pollutants . Root part showed high concentration of heavy metals more than their concentration in shoot part ,this due to that the root part contact directly with the soil , whereas shoot part correlated with soil through the root organ . The trend of metal concentration in both parts was Cd ≤ Co ≤ Ni ≤ V ≤ Pb . High mean concentration (30.36 and 49.23) µg/gm dry weight of Pb recorded in shoot and root part of plant respectively , this mean that the source of this metal is the soil , and it transport through the root and accumulated in the shoot part of the plant ,this indicate that the plant can transport the pollutants among which heavy metals to another organism through the food chain , because plants are the first trophic level in the food chain , this mean also that the plant can transfer the risk or hazard to another organism specially consumers.

Table (4) Mean concentration µg/gm ±SD. of heavy metals in soil of the study area during the study period .winter (A) summer (B)

A					
Element / Station	Cd	Pb	Co	Ni	V
St.1	0.98±001 a	28.6 ±1.2 a	27.2±0.8 a	246±10.5 a	110.5±2.7 a
St.2	0.72 ±0.03 b	29.5 ±2.3 a	25.7±1.4 a	214±12.5 b	98.1±2.2 b
St.3	0.79 ±0.01b	25 ±0.5 b	22.4±2.5 b	225±11.6 b	92.4 ± 5.3 b
St.4	0.09 ±0.01 c	10.5 ±0.16 c	5.66±0.01 c	1.6 ±0.02 c	1.16±0.02 c
B					
St.1	0.82 ± 0.03 a	25.5±0.8 a	29.7 ± 1.2 a	234±6.7 a	113± 6.3 a
St.2	0.65 ±0.01 b	27.4±0.5 a	23.5±1.33 b	255± 10.2 a	78± 1.2 b
St.3	0.70±0.03 b	25±0.21 a	20.5± 1.51 b	189±7.6 b	82±2.6 b
St.4	0.05±0.00	11.6±0.62 b	3.54±0.02b	1.7±0.05 c	0.55±0.05 c

*Means following by same letter in the same row are not significantly different (p≤0.05).

Table (5) Mean concentration Mg/gm ±SD of heavy metals in plant *P. austrials* parts (Root and shoot) during winter (A) and summer (B)

A						
Element / Station	Part	Cd	Pb	Co	Ni	V
St.1	Shoot	0.38±0.05 a	29.5±0.31 a	3.2±0.01a	8.55±1.05 a	0.88±0.01 a
	Root	0.55±0.01 a	58.4±1.4	4.8±0.03a	20.5±2.03 a	1.33±0.05 a
St.2	Shoot	0.26±0.03b	26.1±2.1	2.65±0.01b	8.0±0.05 a	0.58±0.01 b
	Root	0.43±0.02b	50.4±3.6	3.95±0.02a	22.0±0.01b	1.20±0.07 a
St.3	Shoot	0.28±0.02b	29.5±2.2	3.77±0.05a	6.9±0.22 b	0.90±0.01 a
	Root	0.45±0.01c	54.2±2.6	5.86±0.03a	19.8±1.05b	2.1±0.01 b
St.4	Shoot	0.11±0.01c	10.4±0.2	1.22±0.05c	0.78±0.01c	0.55±0.03c
	Root	0.15± d	16.5±0.61	2.30±0.02c	0.85±0.02c	0.95±0.04c
B						
St.1	Shoot	0.64±0.01a	33.9±1.6a	4.5±0.02 a	9.5±0.05 a	1.55±0.03a
	Root	0.78±0.03a	45.2±3.7	6.2±0.03a	21.2±0.32a	2.11±0.01a
St.2	Shoot	0.61±0.01b	33.1±1.1	2.16±0.01b	8.5±0.26 b	1.21±0.03a
	Root	0.88±0.01a	41.4±2.3	4.55±0.05b	19.9±0.03b	2.2±0.01a
St.3	Shoot	0.65±0.03b	30.1±0.9	2.2±0.03c	6.6±0.01 c	1.5±0.06a
	Root	0.78±0.05b	46.2±2.2	3.9±0.01c	18.9±0.03b	2.7±0.05 a
St.4	Shoot	0.09±0.01c	11.5±1.1	0.81±0.02d	2.3±0.02d	0.61±0.01b
	Root	0.11±0.02c	25.4±2.5	0.95±0.01d	3.9±0.01c	0.9±0.03b

It concluded from the present study the following .

1-Discharge waste from AL-Nassiriya oil refinery affected upon the nearby soil through accumulated TPHs and HMs in its.

2-Total organic carbon content (TOC %) support the heavy metals concentration.

3- *P. austriles* is a good bioindictator for HMs and TPHs pollution.

4- Root part of *P. austriles* concentrated both types of pollutants more than shoot part .

5- study area is polluted by HMs and TPHs according to the standard values seted by Dutch ecologesit (Crommentuijn, *et.al*.1997).

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