Concentration And Distribution Of Total Petroleum Hydrocarbons In Two Emerged Aquatic Plants From The River Euphrates Near Al-Nassiriya City -South Of Iraq

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Abstract

Concentration and distribution of Total Petroleum hydrocarbons (TPHs) were estimated in water, surface sediment and two emerged aquatic plants (*Phragmites australis*) and (*Typha domengensis*) .They were collected from three selected stations in the river Euphrates near A1-Nassiriya ,a city south of Iraq, during the period between February and July/2006. Concentration of TPHs in the sample solutions, prepared after extraction, were analyzed by using spectrofluorometer (shimadzu RF. 540). Total organic carbon (TOC) content in sediment was measured , air and water temperature, dissolved oxygen (DO), Biochemical oxygen demand (BOD_5) and suspended particulate matter(SPM) were estimated. Monthly variations were recorded in air and water temperature. The lowest value of DO (4.8 mg/l) in July was recorded in station 3, and the highest (8.9 mg/l) in February was recorded in station 1. The highest value of BOD_5 (8.51 mg/l) in July and the lowest (2.33 mg/l) in May were recorded in stations 3 and 1 respectively. The high concentration of TPHs found in sediment was (23.4 l μg/g) dry weight in April, while it were (10.43 μg/l) in February and (6.30 μg/g) in April in (dissolved and particulate) phases of water samples respectively. Different concentrations of TPHs were found in the aquatic plant samples. *P. australis* recorded higher concentration of TPHs than *T. domingensis* in (shoot and root parts), whereas TPHs concentration in root part were higher than its concentration in shoot part in both plants. Posative correlation was recorded between TPHs concentration and fat content in (root and shoot) parts for both plants. Furthermore, posative correlations were observed between TPHs concentration and TOC content in sediment for all study stations. In conclusion both plants are capable of accumulation TPHs and we can use them as bioindicators for study and monitoring the pollution by this type of pollutants in aquatic environment.

Introduction

Pollutants are substances that adversely affect physical, chemical and biological properties of the environment. Some pollutants are generated by industrial plants, oil refineries, motor vehicles and many other sources within the watershad (GESAMP, 1993). Water pollutants may come from point and nonpoint sources. Point sources involuve factories, sewage treatment units and oil tankers, whereas nonpoint sources are runoff water containing pesticides and fertilizers from acres of agricultural lands. This type of sources is much more difficult to control (Morgan *et al*., 1993). Many studies were conducted to determine TPHs in Iraqi water body. DouAbul *et al*. (1984) for example,studied distribution of petroleum hydrocarbons in surficial sediment from shatt Al-Arab. Al-Saad (1995) studied distribution and source of hydrocarbons in Shatt Al-Arab estuary and northwest the Arabian Gulf. Also, many studies estimated TPHs in plants such as that by Aziz (1998) which studied the concentration of hydrocarbons in the fruit and leaves of date palm from Basrah. Aziz (2004) studied concentration of petroleum hydrocarbons in Shatt Al-Basrah Canal.From his study in shatt Al-Arab Al-khafaji (2006) has indicated that the different species of fish concentrated different levels of aromatic hydrocarbons in their muscles. In the river Euphrates near Al-Nassiriya city, few studies such as that by Ali (2005)were done on petrolum hydrocarbon concentrations.The major objective of the present study is to provide informations on the concentration and distribution of TPHs in water ,sediment and two emerged aquatic plants *Phragmites australis* and *Typha domengensis* collected from the river Euphrates near Al-Nassiriya city.The data may be of use to other researchers who require a baseline for comparsion of TPHs distribution.

Study Area:

Three sampling stations were chosen to represent different sectors of the Euphrates near Al-Nassiriya city, north before the river enters the city station 1, middle station 2 was near Al-Nassiriya Electric Power Station (EPS) and station 3 was in southern part of the river near the treatment unit of the municipal effluents Fig. 1.

Materials and Methods:

Samples were collected monthly from the above mentioned stations during the period between February and July 2006. Air and water temperature were measured in the field of the time of sample collection, while DO and BOD_5 were estimated in laboratory according to (AOAH, 1984) procedure . Water (20)L was collected from each station every month. Water samples were filtered through preweighted 0.45μm por size filters. Material passed through the filter were considered as dissolved, while that retained as particulate. Dissolved hydrocarbons were solvent extracted following UNESCO (1976) procedure, in which carbon tetrachloride (CCl_4) was used for extraction. Ultra violet fluorescence was used for analysis. The extraction procedure of hydrocarbon in particulate matter was based upon that of Goutx and Saliot (1980). The filter with particulate and Soxhlet extracted 150 ml methanol benzene 1:1 mixture for 24 hours. The final extract was analyzed by ultraviolet fluorescence.

Sediments were collected by means of van Veen grab sampler. Triplicate samples were taken. In lab sediments samples, dried in oven with

80^{o}C for 48 hrs, ground finely in agate mortar and sieved through a 63μm mesh metal sieve. The extraction procedure of petroleum hydrocarbons residues in sediment employed was based upon that of Goutx and Saliot (1980). Total Organic Carbon (TOC) content in the sediment was determined according to El-Wakeel and Riely (1957)procedure, by using exothermic heating and oxidation 1gm grind dry sample with chromic acid. Plant samples *Phragmites ustralis* (Cav) extende (common red) and *Typha domengensis* (cattais) are the most widespread plants species world wide with a wide ecological amplitude (Al-Essa, 2004). The above mentioned plants were monthly collected from the margin of the river in each study station. Exactly 10gm of dried part of plant were extracted employed procedure of Goutx and Saliot (1980). Fat content in plant tissues were extracted by solvent (petroleum ether & aceton) and determined according to AOAC (1984) procedure, the value was determined as percentage. Concentration of TPHs in all samples solutions prepared after extraction was analyzed by using spectrofluorometer (Shimadzu-Rf-540). SPSS computer program was used for the analysis of the study data.

***Results and Discussion*:**

Mean values of air and water temperature ^{o}C, (SPM)g/L and (DO, BOD_5)mg/L were represented in (Table 1). Mean concentration of TPHs in water (dissolved & particulate) phases and average monthly variation of these concentrations in water (expressed as Iraqi crud oil equivalents) were listed in (Table 2), while their regional average values were shown in fig. 2.

Concentration of TPHs in water (dissolved phase) was found to vary from ND in July in station 1 to 10.43 μg/L in February in station 3 while its concentrations in particulate phase varied from ND in July in station 1 to 6.30 μg/gm in April in station 3. The higher concentration of TPHs in dissolved and particulate phase specially in station 3, in February and March, due to the city sewage effluents,which contain petroleum hydrocarbons. There are many sources released liquid hydrocarbons in the aquatic environment such as wastewater discharged as domestic sewage effluents. Oil refinery, in addition to natural hydrocarbon was released by aquatic biota Aziz (2004).DouAbul and Al-Saad (1985) have considered that sewage discharge and urban run-off were the most significant sources of oil entering the river of Shatt Al-Arab. Inverse relationship was found between DO and TPHs concentration in particulat phase. This is due to the higher microbial activities in hydrocarbons degradation. BOD_5 is a process that needs dissolved oxygen. Negative relationship was observed between DO and BOD_5. This means that the wastewater contains a wide variety of organic compounds which are measured as biochemical oxygen demand (BOD5). It is the measurement of the oxygen which is required by the microorganisms to oxidize the organic matter. In July TPHs concentration in both water phases are less than its concentration in other months. This is due to the higher temperature in the mentioned month (Table 1). Air and water temperature played un important role in TPHs evaporation and increased the activition of microbile degradation to TPHs in aquatic environment. This is promoted with higher BOD_5 in this month in station 3,(Table 1).

Atlas and Bartha (1972) have indicated that temperature accelerates the degradation of TPHs by microorganisms.The concentration of TPHs obtained in water (dissolved phase) was in general agreement with those reported eleswhere (Table 3).

The highest concentration of TPHs in sediment was 23.41 µg/gm dry weight in April in station 3, while the lowest concentration was 2.11 µg/gm dry weight in July in station 1, (Table 4). The lowest value of TOC in the sediment was 0.49% in July in station 1, while the highest value was 1.97% in March in station 3, (Table 4).The quantity of organic carbon in the sediment is due in part,to sedimentation rate and reactions within the sediment Al-Khafaji,(2005). Positive correlation was recorded between mean values of TPHs and TOC% in the sediment for all stations .In the sediments higher concentration of TPHs and TOC were always observed in station 3, while the lowest concentrations were in station 1 for all study months, (Fig. 3 and 4). The sediment acted as archive for many pollutants. So it accumulated the contaminants above that water, because the high capacity of the sediment for adsorption hydrocarbons, Zerba *et al.*(1985). Al-Saad (1995) has indicated that petroleum hydrocarbons were essential components of the organic materials in sediment. The higher and lower concentrations of TPHs in the sediments were in stations (2 and 3) in Winter and Summer months respectively, (Table 4).This is due to several factors such as the fluctuation in discharged effluents from domestic sewage treatment unit and the effluents of electric power station which are situated near the above mentioned stations respectively. In addition higher temperature in Summer months (Table1)increases microbial activity for hydrocarbons degradation, so TPHs concentration was reduced in the mentioned months and this case was reversed in Winter months. Microbiles activity increases with increasing of water temperature and organic matter to obtain energy and carbon for their growth (Fahad,2006).

A comparison between the present study values of TPHs in sediment and those reported for some other rivers and estuaries elsewhere(Table5), indicated that the present study was of little exposure to sources of this type of pollutants. Concentrations of TPHs in *P. australis* and *T. domegensis* were showed in (Tables 6 and 7) respectively. TPHs are one of the lipophilic pollutants. So analysis of fat in both studied plants gives indices on the level of TPHs in plant tissues. Fat contents% were found in *P. australis* plant as being higher than its content in *T. domengensis* for both studied parts(Table 8).In *P. australis* the highest concentration of TPHs in shoot part was 4.22 µg/gm dry weight in February in station 3, while the lowest concentration was 0.19 µg/gm dry weight in July in station 1. *T. domengensis* recorded highest concentration 5.22 µg/gm dry weight of TPHs in root part in February in station 3, while the lowest concentration was 0.13 µg/gm dry weight in July in station 1, whereas TPHs concentration in shoot part was ranging around (ND-2.73) µg/gm dry weight (Table 7). Positive correlations were recorded between fat content and TPHs concentration in (root and shoot) parts in both plants. The higher concentration of TPHs in root part was more than in shoot part for both plants due to the higher concentration of this pollutants in the sediment found more than that in water. It enters the plants by the absorption process. The higher concentration of TPHs in both plants in station 3. Is due to the richness of this situation with domestic sewage effluents, Ali (2005) indicated that the Euphrates waters in Al-Nassiriya city near municipal effluents treatment unite were rich in petroleum hydrocarbons.

In conclusion,both aquatic plants revealed its' ability to accumulate TPHs. Concentration of TPHs in root part was more than that in shoot part for both plants, while *P.australis* accumulates TPHs more than that in *T. domengensis*. The concentration of TPHs in the sediment was more than that in water. The highest concentration of TPHs in all samples was always found in stations 2 and 3, while the lowest was found in station 1. This indicates that TPHs are possibly originated from sewage discharge, urban run-off and electricity generation station.These may be considered as the most significant sources of oil entering the river Euphrates. According to other values in the world,TPHs concentration in the study area was in an acceptable range. Both plants in the present study can be used as bioindicator for TPHs pollution.Additional studies are needed to know the ability of both plants as phytotechnology to remove TPHs from wetland water.

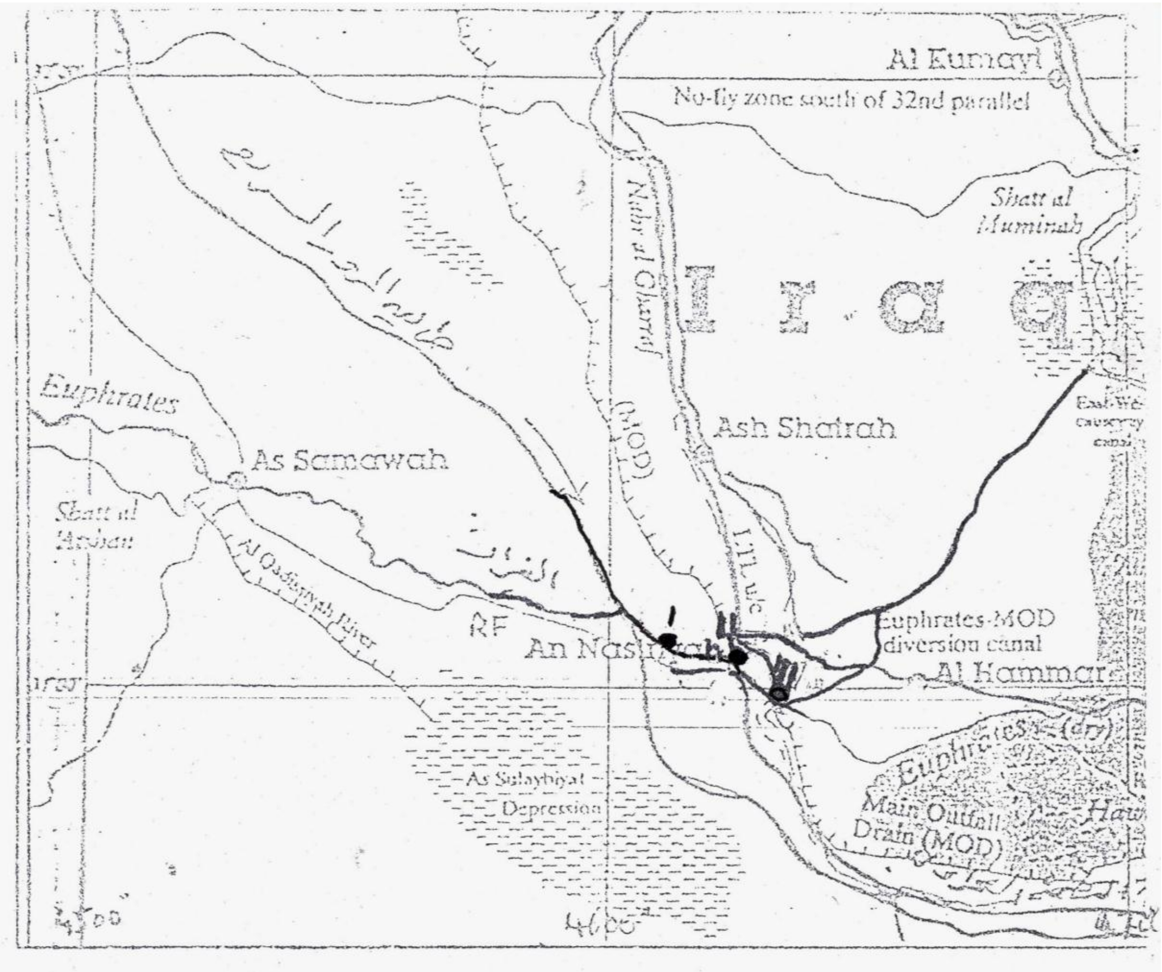

Fig. 1: **Sample Locations**

***Table 1*: Mean values of selected physical and chemical factors in the study area during the study period**

Month	Station	Air Temp. ^oC	Water Temp. ^oC	S.P.M. gm/l	DO mg/l	BOD_5 mg/l
Feb.	1	19	13	0.29	8.9	2.66
	2	19	13	0.62	7.2	3.50
	3	20	14	0.84	7.8	5.71
Mar.	1	21	17	0.33	7.0	2.91
	2	22	18	0.71	6.7	5.82
	3	22	18	0.68	7.3	6.92
April	1	25	21	0.31	7.5	3.81
	2	25	21	1.33	7.0	4.92
	3	26	22	1.20	6.8	6.71
May	1	28	23	0.42	8.0	2.33
	2	28	23	1.02	6.2	5.23
	3	29	24	1.33	7.2	5.78
June	1	33	28	0.88	8.1	2.50
	2	34	28	1.61	7.0	5.11
	3	34	29	1.43	5.5	7.11
July	1	35	31	0.81	7.73	2.41
	2	35	32	1.03	6.20	6.24
	3	36	33	1.07	4.8	8.51

* S.P.M. : **Suspended Particulate Matter**.

Table 2: **Mean concentration of TPHs in water sample (Dissolved and Particulate) phases during the study period**

Month	Station	Concentration	
		Dissolved μg/l	Particulate μg/gm dry wt.
Feb.	1	N.D	N.D
	2	6.71	2.51
	3	10.43	3.63
Mar.	1	1.61	1.11
	2	3.40	2.21
	3	8.0	4.23
April	1	1.12	1.21
	2	3.13	2.64
	3	6.82	6.30
May	1	1.33	1.31
	2	3.21	3.11
	3	4.62	3.61
June	1	1.52	1.2
	2	3.12	3.42
	3	5.69	4.61
July	1	N.D	N.D
	2	4.22	2.6
	3	5.33	2.1

N.D. : **Not Detected.**

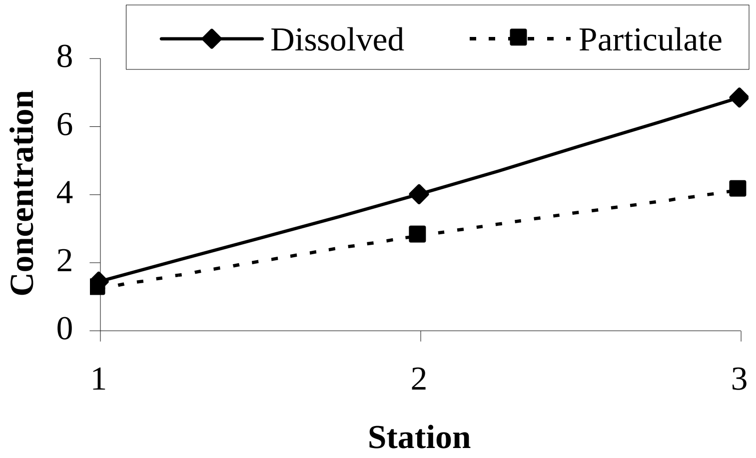

***Fig. 2*: Mean regional concentration of TPHs in water (Dissolved µg/L and Particulate µg/gm dry wt.) during the study period**

***Table 3:* Comparsion of mean concentration (µg/l) of TPHs in water (dissolved phase) in different locations.**

Location	Conc. (µg/l)	Reference
Shatt Al-Arab & Nw. Arabian Gulf	**2.7- 86.7**	**DouAbul (1984)**
Shatt Al-Arab & Nw. Arabian Gulf	**3.25-25.23**	**Al-Saad (1995)**
Euphrates river	**2-15.6**	**Ali (2005)**
Shatt Al-Basrah canal	**25-75**	**Azize (2005)**
Shatt Al-Arab river	**2.7-75.2**	**Al-Khafaji (2006)**
Euphrates river	**1.2-10.43**	**Present study**

Table 4: **Mean concentration of TPHs (µg/gm)dry weight and TOC% in the sediment**

Month	Station	Concentration in sediment	TOC%
Feb.	1	3.40	0.6
	2	16.6	0.96
	3	21.7	1.36
Mar.	1	3.11	0.63
	2	18.5	0.86
	3	22.59	1.97
April	1	2.88	0.75
	2	19.50	0.86
	3	23.41	1.68
May	1	2.56	0.67
	2	19.70	0.81
	3	22.51	1.37
June	1	2.25	0.65
	2	16.5	0.86
	3	20.62	1.43
July	1	2.11	0.49
	2	15.6	0.76
	3	20.04	1.33

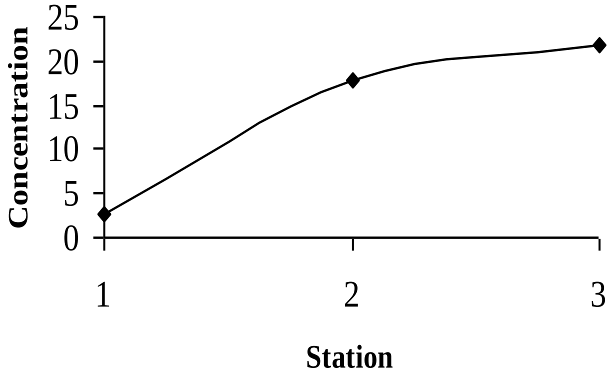

Fig. 3: **Regional concentration (µg/gm) dry weight of TPHs in sediment during the study period**

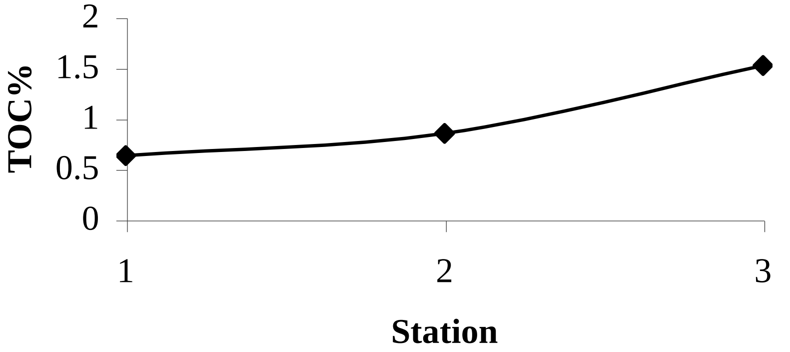

***Fig. 4*: Regional mean values of total organic carbon TOC% content in sediment during the study period**

***Table 5*: Comparison of mean concentration (µg/gm) dry weight of TPHs in the sediment in different locations.**

Location	Conc. (µg/gm) dry weight	Reference
Kuwait	1-291	Zerba *et al*.,(1985)
UAE	10-12	Flower *et al*., (1993)
Shatt Al-Arab estuary	8.92-16.89	Al-Saad & Timari (1993)
Saudia Arabia	0.5-1.03	Ehrhardt & Burns (1993)
Shatt Al-Arab	6.97-55.67	Al-Saad (1995)
Shatt Al-Basrah	2.19-64.98	Aziz (2004)
Shatt Al-Arab/ vicinity of oil processing regions	2.7-93	Al-Khafaji (2006)
Euphrates river	2.11-23.41	Present study

Table 6: **Mean concentration (μg/gm) dry weight of TPHs in parts of *P. australis* during the study period**

Month	Station	Concentration(μg/gm) dry weight	
		Root part	*Shoot part*
Feb.	1 2 3	0.95 5.36 8.11	0.22 2.33 4.22
Mar.	1 2 3	0.36 5.03 7.88	0.25 2.12 4.20
April	1 2 3	0.88 6.11 7.08	0.23 3.05 4.21
May	1 2 3	0.86 4.71 6.17	0.25 2.11 3.88
June	1 2 3	0.48 3.80 6.60	0.21 1.93 3.77
July	1 2 3	0.44 3.60 5.18	0.19 1.89 3.11

Table 7: **Mean concentration (µg/gm) dry weight of TPHs in parts of *T. domengensis* during the study period**

Month	Station	Concentration (µg/gm) dry weight	
		Root part	*Shoot part*
Feb.	1	0.22	N.D
	2	3.11	1.26
	3	5.22	1.92
Mar.	1	0.19	N.D
	2	3.16	1.07
	3	5.03	2.73
April	1	0.19	N.D
	2	2.88	1.11
	3	4.88	2.50
May	1	0.18	N.D
	2	2.78	1.23
	3	4.79	2.10
June	1	0.16	N.D
	2	2.65	1.08
	3	4.26	2.11
July	1	0.13	N.D
	2	2.30	1.09
	3	4.22	2.05

Table 8: **Fat content in different samples of *P. australis* and *T. Demengensis***

Month	*P. australis* Fat content %		*T. demengensis* Fat content %	
	Root part	*Shoot part*	*Root part*	*Shoot part*
Feb.	0.28±0.01	0.15±0.02	0.16±0.02	0.13±0.01
Mar.	0.25±0.03	0.17±0.02	0.18±0.02	0.15±0.02
April	0.27±0.02	0.18±0.01	0.20±0.03	0.17±0.01
May	0.24±0.03	0.19±0.01	0.18±0.02	0.16±0.01
June	0.23±0.03	0.18±0.02	0.16±0.02	0.13±0.01
July	0.23±0.02	0.15±0.02	0.17±0.01	0.13±0.02

الخلاصة :

تضمنت الدراسة الحالية تحديد تركيز وتوزيع الهيدروكاربونات النفطية الكلية في الماء والرواسب السطحية ونوعين من النباتات المائية البارزة ، القصب *Phragmites australis* والبردي *Typha domingensis* جمعت عينات هذه الدراسة من ثلاث محطات أختيرت في نهر الفرات قرب مدينة الناصرية، خلال الفترة من شباط ولغاية تموز/٢٠٠٦.

تم قياس الهيدروكاربونات النفطية في عينات المحاليل بعد استخلاصها بوساطة جهاز التفلور نوع (Shimadzu-RF-540) ، كذلك تم قياس محتوى الكاربون العضوي الكلي في الرواسب، كما قيست درجة حرارة الهواء والماء والاوكسجين المذاب والمتطلب البايوكيميائي للأوكسجين وكمية المادة العالقة في الماء ولجميع المحطات.

ظهرت تغيرات شهرية في درجة حرارة الهواء والماء كما سجلت أدنى قيمة للأوكسجين المذاب (٤.٨ ملغم/لتر) خلال شهر تموز في المحطة ٣ وأعلى قيمة له (٨.٩ ملغم/لتر) سجلت خلال شهر شباط في المحطة ١، بلغت أعلى قيمة للمتطلب البايوكيميائي للأوكسجين (٨.٥١ ملغم/لتر) في شهر تموز وأدنى قيمة له (٢.٣٣ ملغم/لتر) خلال شهر مايس في المحطتين
٣، ١ على التوالي.

ولوحظ أن أعلى تركيز (٢٣.٤١ ميكغم/غرام) وزن جاف للهيدروكاربونات النفطية في الرواسب سجل خلال نيسان، في حين بلغت تراكيزها (١٠.٤٣ ميكغم/لتر) خلال شهر شباط و(٦.٣٠ ميكغم/غرام) وزن جاف خلال شهر نيسان في الجزئين الذائب والعالق للماء على التوالي.

ولوحظت أيضاً تراكيز مختلفة للهيدروكاربونات النفطية الكلية في عينات النباتات المدروسة حيث سجل نبات القصب *P. australis* تراكيز اعلى مما هي عليه في نبات البردي *T. domingensis* وفي كلا الجزئين(الخضري والجذري)، في حين كانت تراكيزها في الجزء الجذري أعلى مما هي عليه في الجزء الخضري ولكلا النوعين. ولوحظ وجود ارتباط موجب بين تركيز الهيدروكاربونات النفطية الكلية والمحتوى الدهني في الجزئين الخضري والجذري لكلا النباتين، كذلك ظهر ارتباط موجب بين تركيز الهيدروكاربونات وكمية الكاربون العضوي الكلي في الرواسب، كما سجل ارتباط سالب بين المتطلب البايوكيميائي للأوكسجين والاوكسجين المذاب في جميع محطات الدراسة. استنتج من هذه الدراسة ان كلا النباتين لهما القدرة على تراكم الهيدروكاربونات النفطية ويمكن استخدامهما كدلائل احيائية لمراقبة التلوث بهذا النوع من الملوثات في البيئة المائية.

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