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Phytoremediation of organic pollutants in wastewater using native plants

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Abstract— Phytoremediation is a promising technology for the cleanup of petroleum contaminated soil. Total petroleum hydrocarbon TPH are elements of difficult management and they can cause carcinogenesis and toxicity in human. Different techniques have been used for the remediation of contaminated soils, but the phytoremediation is proposed as possible alternative, convenient and environmentally friendly than traditional physicochemical techniques. In this study two systems adopted; free surface flow (FSF) and sub-surface flow (SSF) to select which system is more suitable for plant to be applied in future study of phytotoxicity test. The preliminary test for two types of plants phragmites communis (The scientific name is phragmites australis) and Helianthus annuus was conducted in a greenhouse for 14 days. The diesel concentrations used in this experiment were (0,1,2,3,5)% V_{diesel}/V_{water} for both systems. Through this period the growth parameters measured; were wet weight, dry weight, stem length, root length and observation the withered for the two plants. From this work, SSF better than FSF and Phragmites communis is better in phytoremediation than Helianthus annuus.

Keywords— Phytoremediation, TPH, SSF, FSF, Phragmites communis, Helianthus annuus.

1. Introduction

Contamination of the aquatic environment has become an important and dangerous problem in many areas of the world with rivers and gulfs often affected in a serious way. Almost all marine coastal ecosystems have complicated structural and dynamic qualities that can be simply modified by human influence, estuarine and marine sediments are basins for different contaminants transported from other ecosystems. Total Petroleum Hydrocarbons (TPH's) assimilate one of the most common groups of persistent organic pollutants in the environment. They have been studied much more because they are toxic to many organisms and human health [31].

Oil-producing and industrialized countries of the world are considered as a threat to the environment and to human health. Phytoremediation as a green technology selection, is particularly useful in wetland environments because it uses plants and their associated microorganisms to heal water contaminated with hydrocarbons, and it is more environmentally supporting than the traditional mechanical clean-up methods [7, 29]. The basic mechanisms in the phytoremediation of organic contaminants are the immediate understand of

contaminants and their later metabolism in plant tissues, the transpiration of volatile organic hydrocarbons through the leaves, the launch of exudates that motivate microbial activity, and the improvement of mineralization at the root-soil interface, which is attributed to the microorganisms associated with the root surface [15][40]. Soil contamination petroleum hydrocarbons (PHC) is of concern according to potential adverse impacts on: (1) soil functions and services; (2) functional and structural variety of soil microorganisms, animals and plant communities and; (3) human health [8][25][26][49].

TPH is a mixture of many various compounds and everyone is exposed to TPH from many sources, including gasoline pumps, spilled oil on pavement, and chemicals used at home or work. Several of TPH compounds can affect your nervous system that leading to headaches and dizziness. TPH has been found in at least 23 of the 1,467 National Priorities List sites identified by the Environmental Protection Agency (EPA). Some chemicals that may be seen in TPH are hexane, jet fuels, mineral oils, benzene, toluene, xylenes, naphthalene, and fluorene, moreover other petroleum products and gasoline components [1]. Diesel is generally used fuel, basically in industry, and with the increase technological

development, it has become one of the most popular organic pollutants in the environment as well as it is toxic to a lot of organisms and detrimental to human health [31]. Phytoremediation for wastewater treatment uses plants species to break down, convert, digest, or removal toxic pollutants [13] and it is a natural mixing technology between plants and microbes to remediate contaminated site[22]. Recently, phytoremediation has been used to remediate industrial wastewater contaminated by organochlorines, including chlorobenzenes, chlorophenols, chlorinated hydrocarbons and chlorinated olefins [37]. The application of phytoremediation in recent years has changed the environment to the better using efficient and cheap in situ methods[15]. The aim of this study is to determine the maximum concentration of diesel which phragmites communis and Helianthus annuus plants that can survive and degrade the hydrocarbons.

2. Advantages and limitations of the phytoremediation technology

The advantages and limitations of the phytoremediation technology is:

Table 1: Advantages and limitations of phytoremediation [16].

Advantages	Limitations
Applicable to wide variety of inorganic and organic contaminants .	Limited by depth (roots) and solubility and availability of the contaminant .
Reduces the amount of waste going to landfills .	Although faster than natural attenuation , it requires long time periods (several years) .
Does not require expensive equipment or highly specialized personnel.	Restricted to sites with low contaminant concentration .
It can be applied in situ , Reduces soil disturbance and the spread of contaminants .	Plant biomass phytoextraction requires proper disposal as hazardous waste .
Early estimates of the costs indicate that phytoremediation is cheaper than conventional remediation methods .	Climate and season dependent . It can also lose its effectiveness when damage occurs to the vegetation from disease or pests
Easy to implement and maintain . Plants are a cheap and renewable resource . easily available .	Introduction of inappropriate or invasive plant species should be avoided (non-native species may affect biodiversity) .
Environmentally friendly . aesthetically pleasing . socially accepted . low-tech alternative .	Contaminants may be transferred to another medium , the environment . and or the food chain .

Less noisy than other remediation methods. Actually trees may reduce noise from industrial activities	Amendments and cultivation practices may have negative consequences on contaminant mobility .
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3. Mechanisms of phytoremediation

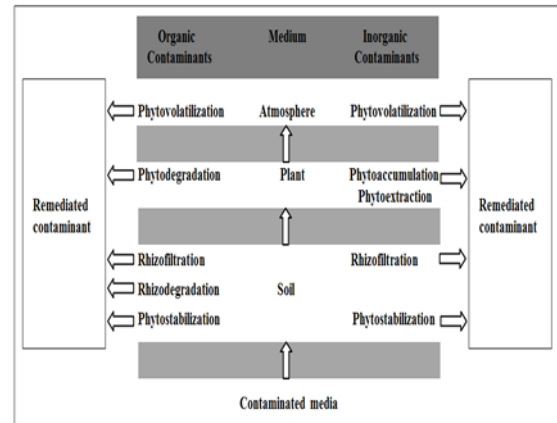


Figure 1: Mechanisms of phytoremediation [46].

3.1 Phytoextraction

Phytoextraction is a technique used in situ for the treatment of contaminated soils [2] [6] [24] [34] [49]. Contaminants are being absorbed by the roots, transported and accumulated in the shoots and leaves [30,44]. Phytoextraction is an eco-friendly technology and has many important advantages:

- It does not destroy/change the landscape.
- It protects the conservation and so the ecosystem.
- It is the major technique of phytoremediation to the removal of heavy metals from soil, sediment and water.
- It is also regarded as the most commercially hopeful technique because it is cheap [3].

3.2 Phytostabilization

Pollutants are immobilized in the root system by absorption of the roots or precipitation in the rhizosphere, so this process reduces the contaminant mobility, inhibiting migration into groundwater and reduce the bioavailability in the food chain [2,6,42,49].

The advantage of this technique consists:

In the changes of soil chemistry composition happened by existence of the plant itself and such these changes can facilitate the absorption or cause the precipitation of metals on the roots [51].

3.3 Phytovolatilization

In phytovolatilization process the pollutants are absorbed at root level and moved by the xylem and emitted into the atmosphere from the aerial parts of plant in less toxic forms because of metabolic modification. So, pollutants

are not only removed but they are being moved from one part to another [2, 6, 42, 49].

The most advantage of this technique is the probability that the contaminant can be changed into a less toxic substance, but, on the other hand, the weakness of this application is the probability that the modified substance, and still potentially toxic, can be emitted into atmosphere and then resettling in to the environment.

The phytovolatilization can be stratified to contaminants existence in soil, sediment or water, especially for organic contaminants like tetrachloroethane, trichloromethane and tetrachloromethane [39, 45, 52].

3.4 Phytodegradation

In Phytodegradation process, organic contaminants, after absorption by the root system, are degraded gratitude to the activity that completed by enzymes contributed in metabolism of the plant, or they will be incorporated into the plant tissues [2, 6, 42, 49]. Contaminants are stopped working after they have been started by the plant. As with phytoextraction and phytovolatilization, plant absorption generally happens only when the contaminants' solubility and hydrophobicity fall into a particular acceptable range. Phytodegradation has been noticed to remediate some organic contaminants, like chlorinated solvents, herbicides, and munitions, so it can treat contaminants in soil, sediments, or groundwater [48].

3.5 Rhizofiltration

Rhizofiltration allows the removal of organic and inorganic pollutants from groundwater, surface water as well as wastewater either adsorption or precipitation on the




roots, or for adsorption of contaminants around the root zone [51]. Several disadvantages of this technique consist in: adjust the pH, the importance of a first cultivation in a greenhouse, frequent harvests and subsequent get rid of the plants [6, 42].










3.6 Rhizodegradation






Rhizodegradation consists in the biodegradation of the organic contaminants at level of the radical apparatus of the plant, in a soil area called rhizosphere and this process happens gratitude to the activity of bacteria, fungi and yeasts (several studies have shown that the number of microorganisms in the rhizosphere is 100 times greater than the quantity present on the surface), that take out nutrients from the root exudates of the plant. Root exudates are a carbon and nitrogen source, and these root exudates are able to increase the efficiency of extraction and after that removal of contaminants via the plant [6, 12, 27, 28, 49]. Among the advantages and disadvantage [9][11][24][43]:

- It is a process, which happens in situ;
 - Translocation of the compounds to other parts of the plant or in the atmosphere is lesser than other technologies of phytoremediation.
 - It can get a perfect mineralization.
 - Installation and maintenance costs are low.
- The disadvantages is:
- It is a slow process and efficient only on surface of contamination (20-25 cm of depth).
 - The depth of the roots may be specified by the physical structure of soil.
 - Plants may demand fertilizer.

Table 2: Phytoremediation studies on the treatment of pollutants

Author	Image of plant	Species	Type of system	Concentration of pollutant	Methods (Period time, PH,T,Q,Temp.)	Removal efficiency
Israa Abdulwahab Al-Baldawi et al. 2013 [20]		Bulrush of scirpus grosses	Sub surface (horizontal) Emergent plant	Diesel (0,0.1,0.2,0.25)% V diesel/V water	T=72 days Temp=24-27c° pH=5.6-6.4 for 0 pH= 5.2-8.4 for all	eff.0.1=82% eff.0.2=71% eff.0.25=67%
Asmita V. Potil Jyoti P. Jedhav 2013 [5]		Tagetes patula L.	-----	Textile dye Reactive blue 160 (20-100) $\frac{mg}{l}$	T=3 weeks temp. = 25±2 pH=5.8	eff.at20 $\frac{mg}{l}$ = 90% eff.at40 $\frac{mg}{l}$ = 74% eff.at60 $\frac{mg}{l}$ = 67% eff.at80 $\frac{mg}{l}$ = 62% eff.at100 $\frac{mg}{l}$ = 57%
Nadya H.A.S et al. 2014 [33]		Eleocharis ochrostochys	Sub surface Free surface	Diesel (0,5,10,20,30,40) ml Diesel/L water	T = 15 days	bulrush resistant hydrocarbons , SSF system was batter compared FSF

Ray R. Hinchman et. al [35]		Hybrid polar (Populus sp.)	Sub surface horizontal and vertical	Heavy metal (zinc) 50-2000 ($\frac{\mu g}{g}$)	-----	For Hybrid polar eff.=99%
Ray R. Hinchman et. al [35]		Eastern Camagrarass (Tripsacum dactyloides)	Sub surface horizontal and vertical	Heavy metal (zinc) 50-2000 ($\frac{\mu g}{g}$)	-----	for Eastern eff.=(90-50)%
Israa Abdulwahab Al-Baldawi 2015 [18]		Typha angustifolia	Sub surface (horizontal)	COD TSS NH ₄ -N NO ₃ -N	T=95 days temp.=25-33 c° pH=4.5±0.1	eff.TSS = (73.3-95.5)% eff.COD = (35.2-86)% eff.NH ₄ = 33.9% 97.2% 18.9% eff.NO ₃ =79.6% 85.5% 71.8%
Kah Aik Jan et al. 2016 [23]		Eichhornia crassipes	free floating plant	dyes Methylene blue Methyl orange (5,100,150,200) $\frac{mg}{l}$	T=20 days temp. = 30±1 c° pH=4.4-10 Q=50 $\frac{mg}{l}$	eff.MB=98.42% eff.MO=66.8%
Israa Abdulwahab Al-Baldawi et al. 2013 [17]		scirpus grosses	Sub surface Free surface emergent plant	Diesel (1.2.3)% V diesel/V water	T=72 days temp _{SSF} =27.96 pH _{SSF} =5.27 temp _{FSF} =25.96 pH _{FSF} = 6.5	eff. _{SSF} =91.5% eff. _{FSF} =80.2%
Salmi Nur Ain Sanusi et al. 2012 [38]		paspalum vaginatum	Free surface	Diesel (5,10,15,20,25,30) $\frac{g}{kg}$	T=28 days	The plant could grow and has the potential in the phytoremediation of hydrocarbons range (0-30) $\frac{g}{kg}$
Israa Abdulwahab Al-Baldawi et al. 2015 [19]		scirpus grosses	Sub surface emergent plant	Diesel (0,1.0,1.0.175,0.25)% V diesel/V water	T=72 days temp. = 30±3 c°	eff.0.1=81.5% eff.0.175=71.4% eff.0.25=66.6%
Israa Abdulwahab Al-Baldawi et al. 2013 [21]		scirpus grosses	Sub surface Horizontal emergent plant	Diesel (0,8700,17400,26100) $\frac{mg}{l}$	T=72 days Temp.=24.28 c° pH=5.9-7.6 Q=7L	eff. for range (67.2-69.9)%
Nadali Alavi et al. [32]		Sorghum halepense (L.) pers.	Sub surface emergent plant	TPH (5,10,25,50,100) $\frac{g}{kg}$	temp. = 25±5 T=180 days	eff. Sorghum=86%

Nadali Alavi et al [32]		pers. Aeluropus littoralis (Guna) parl.	Sub surface emergent plant	TPH (5,10,25,50,100) $\frac{g}{kg}$	temp. = 25±5 T=180 days	eff. Aeluropus =43%
Eman A. Diab 2008 [10]		Vicia faba zea mays	Free surface	TPH (2.2-2.3)% oil V diesel/V water	T=60 days pH=7.6-7.8	eff.=62.4%
Eman A. Diab 2008 [10]		Triticum	Free surface	TPH (2.2-2.3)% oil V diesel/V water	T=60 days pH=7.6-7.8	eff.=19.9%
Eman A. Diab 2008 [10]		aestivuml	Free surface	TPH (2.2-2.3)% oil V diesel/V water	T=60 days pH=7.6-7.8	eff.=17.6%
Asia Fadhile Al-Mansoori Et al. 2015 [4]		Ludwigia Octovalvis (jacq.)	Free surface	TPH (1,2,3)% $\frac{g}{kg}$ V diesel/V water	T=72 days	eff.=79.81%

4. Methodology

a. Plant Propagation

Phragmites communis and Helianthus annuus were propagated for 6 weeks in the same number of selected plants of (phragmites communis , Helianthus annuus) having the same size, were used in each test pail for each species during propagation, the growth parameters measured were wet weight, dry weight, roots length and stem height.

b. Determination of Wet Weight

Two plants were combined at each sampling time and were rinsed with tap water then the water was absorbed onto tissue paper and the wet weight was recorded [41].

c. Determination of Dry Weight

Plants were dried in an oven at 70c° for 72h , after that dry weight was [41][47].

d. Measurement of Root Length and Stem Height

Root length was measured from the plant at ground level to the root tip, however plant height was determined starting from ground-height to the tip of the longest stems or leaves [14][36].

5. Preliminary tests

The preliminary test was tested in the same number of selected plants of (phragmites communis, Helianthus annuus) having the same size, were used in each test pail for each species. Synthetic wastewater was prepared by

mixing water with diesel in different percentages concentrations 1,2,3,5% (V_{diesel}/V_{water}). In this study, free surface flow (FSF) and sub-surface flow (SSF) systems were used to determine a flow system that enabled the plant to tolerate diesel.

Each pail was planted with plants of the plant in 3 kg of fine sand with 5 L of synthetic wastewater with 1, 2, 3, and 5% (V_{diesel}/V_{water}) diesel concentrations. Another pail without diesel acted as a control. The synthetic wastewater was kept at a level 5 cm above the sand to simulate a free-surface flow system. Physical observation was performed for two weeks, to determine the ability of plant to tolerate diesel contamination.

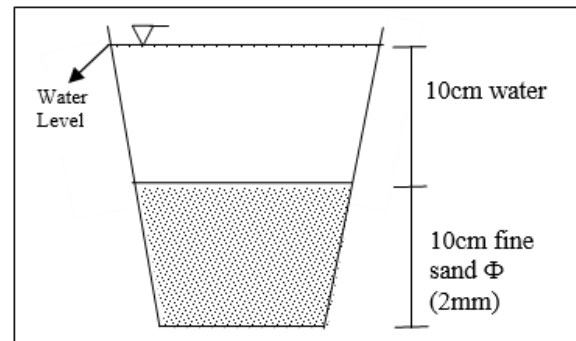


Figure 2: Free surface flow system.

For the sub-surface flow system, each pail was planted with plants of in 3 kg of fine sand with 2L of synthetic wastewater with 1, 2, 3, or 5% (V_{diesel}/V_{water}) diesel

concentrations. Another pail without standard diesel served as a control.

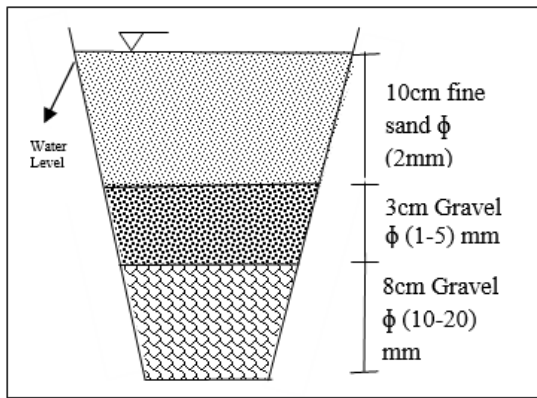


Figure 3: Sub surface flow system.

6. Results and discussion - propagation of phragmites australis and Helianthus annuus

1. Phragmites australis

The wet and dry plant weight was measured from the age of one day to 42 days to determine the plant biomass, the wet and dry weight increase from the second week.

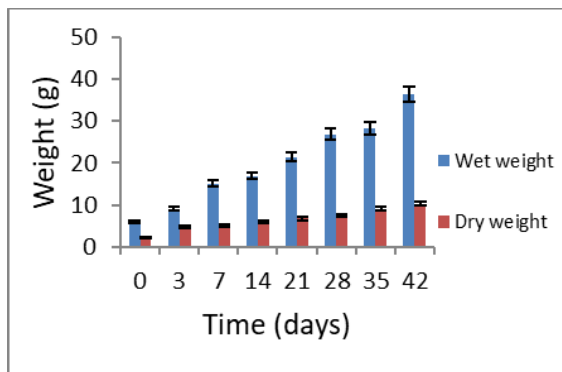


Figure 4: Wet weight and dry weight of phragmites communis during the propagation period.

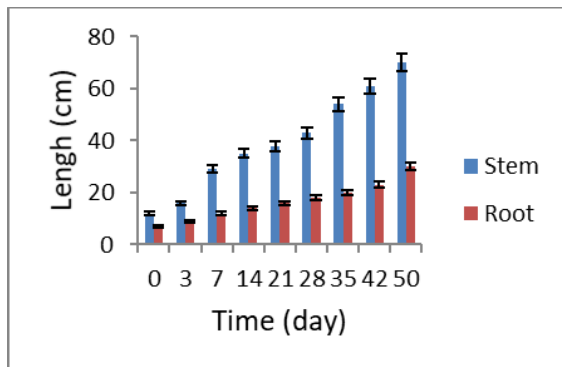


Figure 5: Stem length and root length of phragmites communis during plant propagation period.

The correlation formula was $y=3.919x-5.299$, with the correlation factor $R=0.945$ between wet and dry weight.

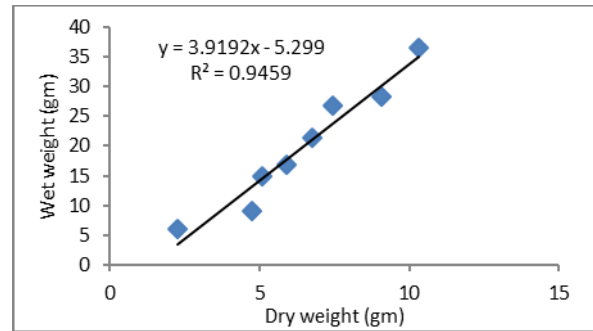


Figure 6: The correlation formula between wet and dry weight of phragmites communis during plant propagation.

2. Helianthus annuus

The wet and dry plant weight was measured from the age of 7 days to 50 days to determine the plant biomass, the wet and dry weight increase from the second week and decrease at 50 days.

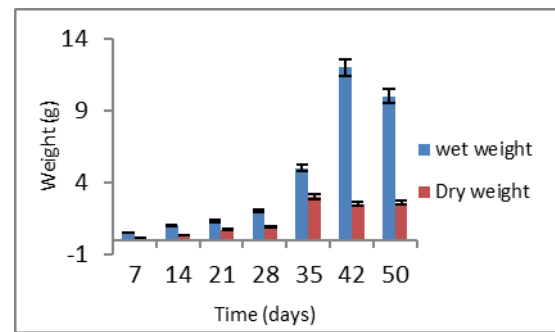


Figure 7: Wet weight and dry weight of Helianthus annuus during the propagation period.

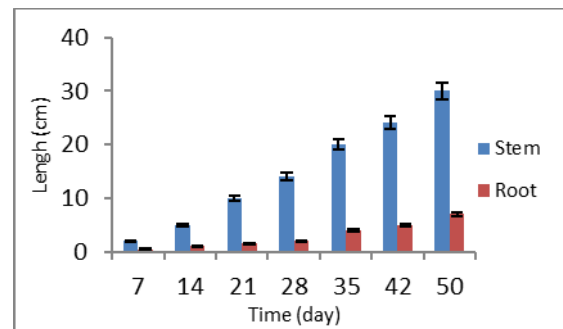


Figure 8: Stem length and root length of Helianthus annuus during plant propagation period.

The correlation formula was $y=3.188x-0.057$, with the correlation factor $R=0.683$ between wet and dry weight.

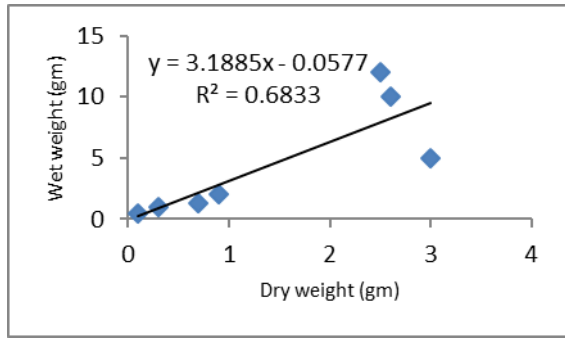


Figure 9: The correlation between wet and dry weight of Helianthus annuus during plant propagation.

7. Preliminary Test of Phytotoxicity

After two weeks of observation on both free- and sub-surface flow systems, the percentage of withered plants after exposure to diesel contaminants. The number of withered plants was recorded and the percentage of withered plants was determined using Equation:

$$\text{Withered plant \%} = \frac{\text{No.of withered plants}}{\text{No.of total plants}} \times 100 \quad (1)$$

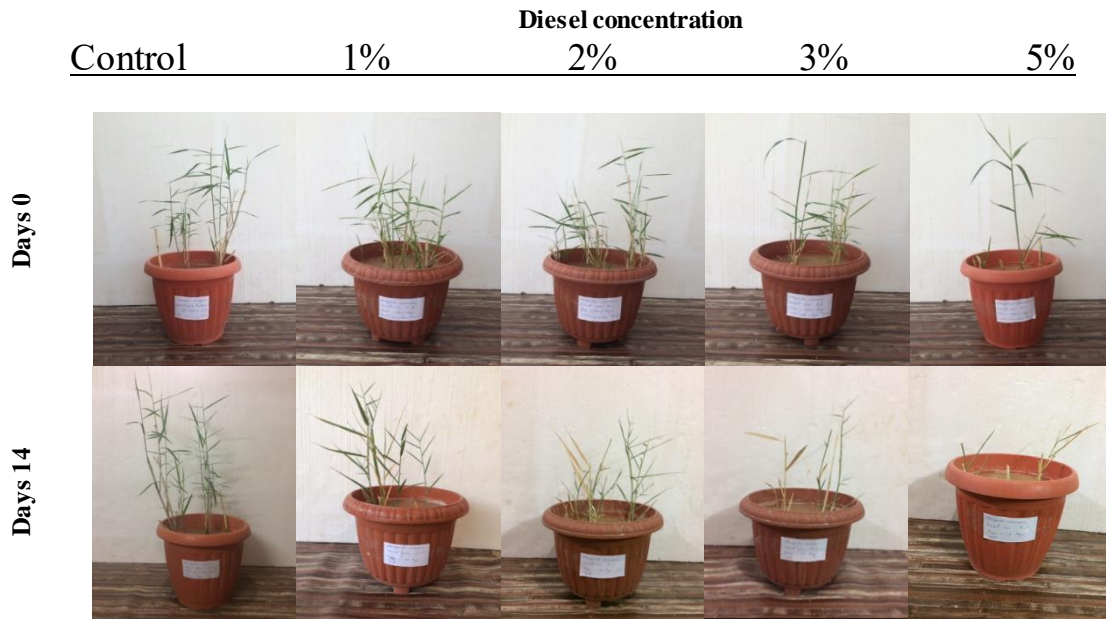


Figure 10: Withered plants in the sub-surface flow system after 14 days of observation in the preliminary test of phragmites communis.

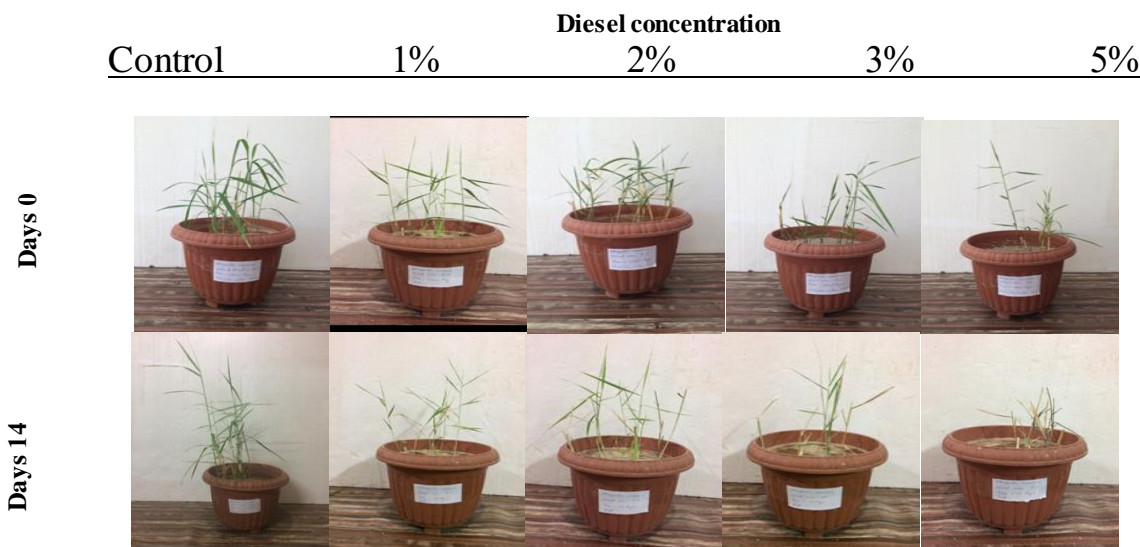


Figure 11: Withered plants in the free surface flow system after 14 days of observation in the preliminary test of phragmites communis.

Table 3: Metal foam properties.

No.	Conc (x) (V _{diesel} /V _{water})	Percentage of withered plants in SSF	Percentage of withered plants in FSF
1	0%	0	0
2	1%	10%	15%
3	2%	33%	40%
4	3%	43%	67%
5	5%	67%	100%

For the FSF and SSF, the minimum percentage of withered plants (15%) occurred at (1%) diesel concentration, it increase at (3%) in FSF at percentage (67%) and after two weeks of exposure to diesel at (5%) diesel concentration, (100%) plant is withered (no plant phragmites communis survived). In SSF the minimum percentage of withered plants, (10%) occurred at (1%) diesel concentration, it increase and at (5%) concentration diesel, withered plant percentage was (%67) .

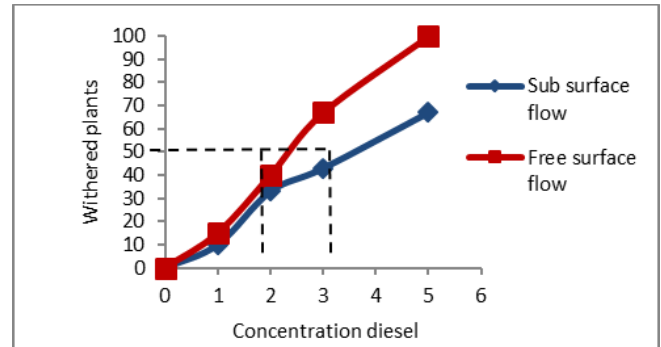


Figure 12: Percentage of withered plants of phragmites communis during the preliminary test

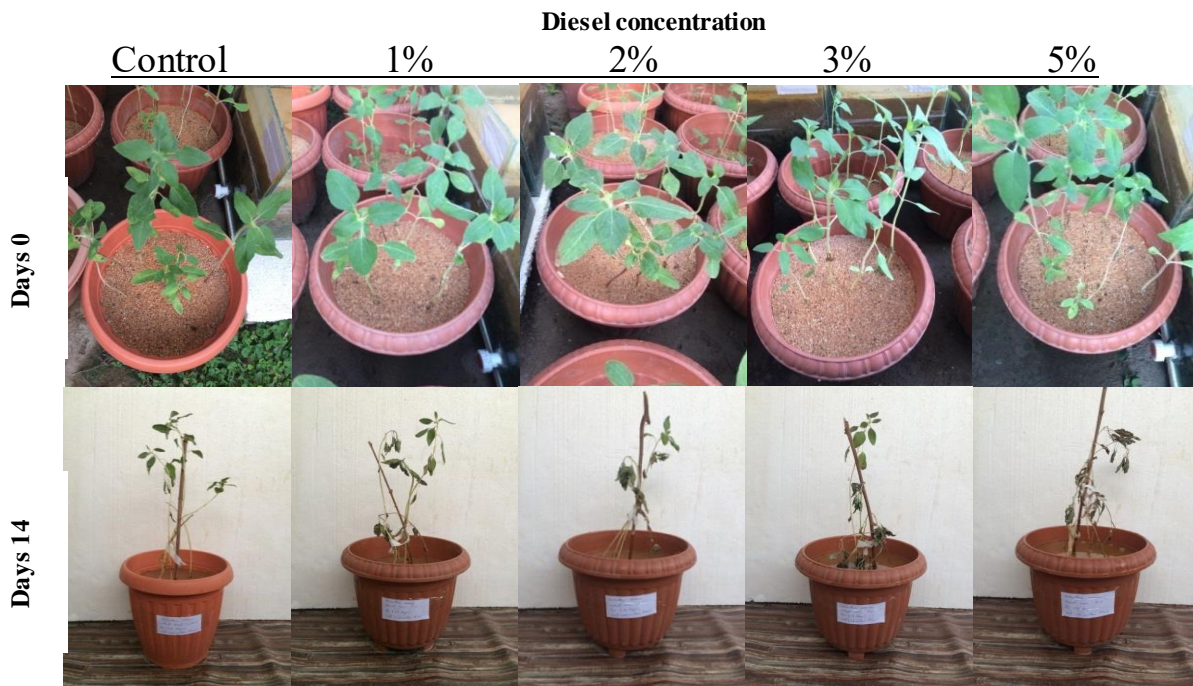


Figure 13: Withered plants in the sub-surface flow system after 14 days of observation in the preliminary test of Helianthus annuus.

Control 1% 2% 3% 5%



Figure 14: Withered plants in the free surface flow system after 14 days of observation in the preliminary test of *Helianthus annuus*.

Table 4: Preliminary tests of diesel exposure using *Helianthus annuus*.

No.	Conc (x) ($V_{\text{diesel}}/V_{\text{water}}$)	Percentage of withered plants in SSF	Percentage of withered plants in FSF
1	0%	0	0
2	1%	20%	33%
3	2%	70%	75%
4	3%	75%	100%
5	5%	100%	100%

For the free-surface flow system, the minimum percentage of withered plants (33%) occurred at 1% diesel concentration after two weeks and at the (3%) diesel concentration, no *Helianthus annuus* plants survived. After two weeks of exposure to diesel in the sub-surface flow system, the minimum percentage of withered plants was (20%) at the (1%) diesel concentration and it is increase, reached (100%) at the (5%) diesel concentration.

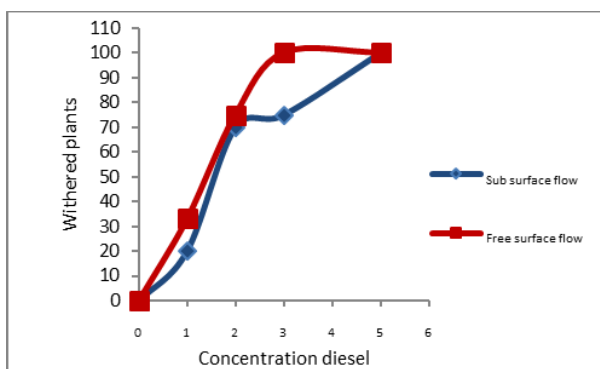


Figure 15: Percentage of withered plants of *Helianthus annuus* during the preliminary test.

8. Conclusion

The preliminary test conducted to assess the ability of *Phragmites communis* and *Helianthus annuus* survive with different diesel concentration in two systems of SSF and FSF. The results clearly show whenever concentration increase, the number of withered plant also increase. The plant in SSF resistant diesel compared FSF and *Phragmites communis* is better in phytoremediation than *Helianthus annuus*.

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المعالجة النباتية للملوثات العضوية في مياه الفضلات باستخدام نباتات محلية

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الخلاصة – تعد المعالجة النباتية تقنية واعده لتنظيف التربة من الملوثات النفطية الهيدروكربونية. ان الهيدروكربونات النفطية الكلية هي عناصر صعبة الادارة، ويمكن ان تسبب التسمم والامراض السرطانية في الانسان. وقد استخدمت تقنيات مختلفة لمعالجة التربة الملوثة ولكن تقترح المعالجة النباتية كبديل، مريح، صديق للبيئة ومن أفضل التقنيات الفيزيائية التقليدية. في هذه الدراسة اعتمد نظامين، التدفق الحر للسطح والتدفق تحت السطح لتحديد النظام الذي هو أكثر ملائمة للنبات ليتم تطبيقها في دراسة مستقبلية للسمية النباتية. تم اجراء الاختبار الاولي لنوعين من النبات (القيصوب الاسترالي وزهرة الشمس) اجريت في البيوت الزجاجية لمدة 14 يوماً. ان تراكيز الديزل المستخدمة في هذه التجارب (0,1,2,3,5 %Vdiesel/Vwater) لكلا النظامين. ومن خلال هذه الفترة قيست معاملات النمو وهي الوزن الرطب، الوزن الجاف، طول الساق، طول الجذر ومراقبة النباتات واعداد النباتات الذابلة. من هذا العمل، وجد ان نظام التدفق تحت السطح أفضل من نظام التدفق الحر للسطح وايضا ان نبات القيصوب أفضل في المعالجة النباتية من زهرة الشمس.

الكلمات الرئيسية – المعالجة النباتية، الهيدروكربونات النفطية الكلية، نظام التدفق تحت السطح، نظام التدفق الحر، نبات القيصوب الاسترالي، نبات زهرة الشمس.