Analysis of Petroleum-Contaminated Soils, Treatment Using Selective Ornamental Plants

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Abstract:

Pot-culture trials were completed to survey the phytoremediation capability of 14 elaborate plants in weathered petroleum-debased soil, which was gathered in the Oil Field, one of the greatest oil fields in Iraq, by looking at their effect on the corruption capability of aggregate petroleum hydrocarbons (TPHs) and its arrangement. Comes about indicated Gaillardia aristata, Echinacea purpurea, Fawn (Festuca arundinacea Schreb), Fire Phoenix (a consolidated F. arundinacea), and Medicago sativa L. could viably lessen TPHs and its creation in 10,000 mg kg−1 TPH-sullied soil. Following a 30-day pot-culture analyze, the expulsion rates were 37.16%, 46.74%, 49.42%, 41.00%, and 37.93%, separately, essentially higher than that in the control (just 12.93%). Expulsion rates of TPH sythesis including immersed hydrocarbon, fragrant hydrocarbon, asphaltene, and polar compound achieved 39.41%, 38.47%, 45.11%, 42.92%, and 37.52%, individually, additionally higher than that in the control (just 6.90%). Advance, the aggregate biomass did not essentially diminish for all plants tried in 10,000 mg kg−1 TPH-debased soil. Fourier change infrared spectroscopy affirmed the nearness of oil in the plant tissues. These outcomes proposed that the run of the mill fancy species including G. aristata, E. purpurea, Fawn, Fire Phoenix, and M. sativa can be embraced in phytoremediation of oil-contaminated soil.

Keywords: petroleum-contaminated soil, phytoremediation, total petroleum hydrocarbons (TPHs), Ornamental Plants.

Introduction: With quick improvement of industry, vehicles, and planes, the interest for petroleum is progressively extended. Be that as it may, a lot of petroleum was impregnated to soil amid the investigation, translocation, and preparing, and it brought about critical ecological contamination. As per pertinent reports, the centralization of aggregate petroleum hydrocarbons (TPHs) around the

Liaohe Oil Field might be more than 10,000 mg kg−1, preferably higher than the hazard based cleanup levels (500 mg kg−1) in mechanical soils for TPHs. The levels of TPHs around the very debased destinations were 30%–50% in the surface soil (0–20 cm). Subsequently, the harmful impacts of TPHs have been broadly reported; the exploration on petroleum-debased soil has been given careful consideration. Commonplace medicines for petroleum-debased soil include in exhuming the dirt and expelling it for treatment utilizing physical or compound strategies. These medicines, however powerful, are expensive and include in broad site unsettling influence. To discover all the more fiscally satisfactory choices, natural strategies have been explored, for example, phytoremediation, that is, utilizing living green plants as a part of situ to "tidy up" defiled grounds. Phytoremediation is a low-input approach contingent upon regular constriction by biodegradation and physiochemical systems that abatement the toxin focus wherein sowing plants might be the main intercession [1].In the previous decade, it has delivered a broad group of research on the phytoremediation of both natural and inorganic contaminants. Most hydrocarbon debasement is accepted to happen through a rhizosphere impact; plants radiate natural mixes through their underlying foundations, which increment the thickness, assorted qualities, and movement of particular microorganisms in the encompassing rhizosphere, which thusly corrupt hydrocarbons. Few concentrates just by Zhou and associates, be that as it may, have inspected decorative plants for phytoremediation of petroleum-debased soils. This has their favorable circumstances in revegetation cover and embellishing encompassing environment. Phytoremediation utilizing decorative plants can abstain from entering evolved ways of life and successfully lessen the contamination, than utilizing crops. The point of this work was to screen out decorative plants with high viability for treating petroleum-debased soil, by analyzing the expulsion rate of TPHs and its creation following a 30-day pot-culture try in TPH-tainted soil [2].

SOIL POLLUTION:

Geosphere, or physical layer, is that some piece of the earth on which the individuals live and extricate the most extreme of its assets. Past it was trusted that the earth had boundless ability to assimilate the effects of mankind. At present, the geosphere is viewed as extremely delicate and powerless against wounds beginning from anthropogenic exercises. As indicated by Manahan the meaning of contamination can be depicted as the increment in the centralization of a sure substance to more elevated amounts than that they happen actually, emerging from an outer source, by and large identified with the human movement. There is awesome trouble in anticipating the conduct of a xenobiotic in the dirt, since its arrangement is absolutely unpredictable and heterogeneous. Along these lines, the information of the physico-substance qualities of the contaminant mixes and the earth is basic to anticipate its element. It ought to be noticed that few soils have the ability to absorb and kill such toxins, since compound and biochemical marvels are fit for lessening the destructive way of the poisons. These marvels incorporate procedures of oxi-decrease, hydrolysis, corrosive base responses, precipitation, adsorption and biochemical debasement [3].

A few perilous natural concoction items can be corrupted to harmless items on the dirt and the overwhelming metals can be sorbed, immobilized or mineralized. By and large, a great deal of consideration ought to be taken in the buildups' disposal, rejects and other conceivably dangerous materials to the dirt, especially where there is the likelihood of sullying the current water. At the point when the contaminant achieves the dirt, either intentionally or inadvertently, it endures the activity of geochemical and natural wonders and is appropriated by the subsurface in the vaporized, remaining or adsorbed stages, free stage and broke down stage. The dispersion of such stages will rely on upon their physico-concoction qualities furthermore on the dirt's sort. Along these lines, the contaminants' portability and, hence, their lethality are straightforwardly identified with the dirt's limit in keeping up them held in their strong stage, making them inaccessible to be consumed by plants, disintegrated and/or leachate. Among the components that focus the coupling of contaminants to the dirt there is the accessible surface range of the particles (m^2/g). In addition, the electrical charges of the dirt's particles lattice likewise impact in the contaminants' adsorption to the earth. It is essential that in connection to their physico-concoction properties the contaminants are delegated Dense Non-Aqueous Phase Liquid (DNAPL), when the substance is more thick than the water and Light Non-Aqueous Phase Liquid (LNAPL), when it is less thick.

The primary procedures of association between the natural mixes or metals and the earth are the maintenance by adsorption, retention or precipitation; biotic and abiotic changes and transport by volatilization, filtering or spillover. There are mixes exceptionally impervious to debasement that can communicate unequivocally in a reversible or irreversible route with the colloidal segments of the dirt. This procedure is called sorption, both for adsorption and ingestion. Adsorption is described as an interfacial procedure while assimilation varies for including the compound's entrance in the dirt's particles and can be gathered inside the safeguard framework. By and large, the contaminants' element in the dirt can be displayed by three instruments of mass transference, specifically: shift in weather conditions, scattering and weakening. Shift in weather conditions – it comprises in the component where the contaminants circumstantially take after the stream vectors and keep an immediate association with the rate of permeation in the dirt. It is the instrument in charge of the arrangement and activation of the free period of hydrocarbons [4].

Scattering – Consists in the instrument in charge of the lessening in the contaminants' centralization in the liquid permeation and that can happen by two procedures: hydrodynamic scattering and atomic dissemination. Hydrodynamic scattering happens by the stream limitation in the dirt's pores that creates the lessening in the permeation speed of the more gooey segments while the atomic dispersion is, naturally, a marvel of weakening of the more solvent mixes, and is the principle development procedure of the disintegrated stage, in charge of the more noteworthy portability of the contaminants. On account of emulsions, for example, hydrocarbons, the scattering can happen in a more perplexing system, because of the wonders of hysteresis (deferral) of the contaminants' entrainment, particularly in the immersion fronts and slim periphery. This procedure is related to the development of the adsorbed stage furthermore by the generation of a small amount of emulsions that can compose the dissolved phase.

Lessening – Consists in the decrease of contaminants transported by shift in weather conditions or weakening by compound or physico-synthetic responses. Concoction weakening is the more exceptional in soils with higher cation trade limit and acts lessening mixes in the free and adsorbed stage. Additionally in the rundown of responses there are the bioconversion responses, in which a hydrocarbons' piece is changed or completely oxidized in natural acids. Concoction weakening is more extraordinary in the area with higher accessibility of oxygen. Physico-concoction constriction is in charge of the development of the adsorbed stage and comprises in the contaminants' detainment that hold fast to the dirt's grains, particularly to the grumes of mud with higher action. In any case, connected with the systems of synthetic lessening, it is in charge of the development of the broke down stage (encouraged by the diminishment of pH).

EXPERIMENTAL

Sampling and tested materials

Weathered petroleum-contaminated soil was collected (sampled to a depth of 250 mm) from the Shengli Oil Field in Iraq. The contaminated soil had been classed as a drained brown soil with pH 7.66, and carbon (C), phosphorus (P), nitrogen (N), and available P concentrations were 45.77, 0.65, 0.73, and 0.002 g kg^{-1}, respectively. Uncontaminated reference soil samples were collected. The average concentration of TPHs in contaminated soil collected was 28,000 mg kg^{-1} and its composition of TPHs was 40.76% of saturated hydrocarbon fraction, 27.02% of aromatic hydrocarbon fraction, and 30.82% of asphaltene and polar fraction. Collected soil was sieved through a 4.00-mm sieve to ensure homogeneity. According to the pretest results, all plants tested could not grow in the weathered petroleum-contaminated soil directly. Through the addition of uncontaminated reference soil, contaminated soil collected was diluted to 10,000 mg·TPHs kg^{-1} (W_{TPHS}/W_{soil}) according to the experimental design [5].

Experimental design

The tested plants and their basic botanical characteristics are summarized. Soil tested (2.5 kg) was added to 20-cm-diameter pots. A disc of filter paper was placed in the bottom of each pot to prevent the dry soil escaping out from the drainage holes and pots were placed on saucers. To each pot, single plant species treatments (n=6) were transformed, and germination of each seed took place in 15 days.

Then, each ornamental plant studied in three replicates in contaminated soil (TPHs=10,000 mg kg^{-1}) was harvested at the end of the experiment. Three replicates of the control (no plants and only soil) were also simultaneously maintained with the same contaminated soil. The control soils were identically processed at the time of watering plants and all treatments were processed during 30 days. Plants were sown in a grown chamber with a 16 h/25°C day and 8 h/15°C night cycle. Pots were watered every second day to maintain approximately 25% gravimetric water content. Roots were shaken to dislodge loose soil and then the attached rhizosphere soil was archived at −20°C for hydrocarbon analyses. Following

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analysis, washed roots and detached shoots were dried at room temperature for 1 week and then weighed.

Table 1: Tested Ornamental Plants and Their Basic Characteristics

Tested number	***General name***	***Scientific name***	***Family and genera***	***Basic characteristic***
1	Cornflower	*Centaurea cyanus* L.	Asteraceae, *C. cyanus*	An annual plant growing 16–35 inches tall, with grey-green branched stems.
2	Snapdragon	*Antirrhinum majus* L.	Plantaginaceae /Veronicaceae, *A. majus* L.	A herbaceous perennial plant growing 0.5–1 m tall, rarely up to 2 m. The leaves are spirally arranged and broadly lanceolate.
3	Green false hellebore	*Adonis aestivalis* L.	Ranunculaceae , *A. aestivalis* L.	A perennial flowering plant; the flowers appear in springtime and are up to 80 mm in diameter, with up to 20 bright yellow petals.
4	Annual chrysanthemum	*Chrysanthemum carinatum*	Compositae, *C. carinatum*	An annual plant, growing 30–70 cm tall with branched stems.
5	Barberton daisy	*Gerbera jamesonii* Bolus	Asteraceae, *G. jamesonii* Bolus	A perennial flowering plant, growing 20–30 cm tall, with hairy stem; it is a tuft-forming plant.
6	Blanketflower	*Gaillardia aristata*	Asteraceae, *G. aristata*	A perennial herb reaching maximum heights of anywhere between 20 and 70 cm. It has lance-shaped leaves near the base and several erect, naked stems holding the flowers.
7	Purple coneflower	*Echinacea purpurea* (L.) Moench	Asteraceae, *E. purpurea*	A perennial flowering plant that is 1.2 m tall and 0.5 m wide at maturity.
8	Aster Callistephu	*Callistephus chinensis* (L.)	Asteraceae, *C. chinensis*	An annual plant, growing 20–80 cm tall with branched stems. The leaves are

	s	Nees		alternate, 4–8 cm long, ovate, and coarsely toothed.
9	Black or common nightshade	*Solanum nigrum* L.	Solanaceae	Annual; 15–60 cm tall, with a suffrut escent base; branches and shoots subglabrous or pubescent to glandular villous; hairs appressed or patent.
10	Sandland No. 1		Gramineae	A kind of combined pasture. Evergreen, creeping perennial grass; deeply rooted specimen grass.
11	Fawn	*Festuca arundinacea* Schreb	Poaceae, *F. arundinacea*	Evergreen, tuft-forming grass, with a deep-root system.
12	Fire Phoenix		Poaceae, *F. arundinacea*	A kind of combined *F. arundinacea*. Evergreen, tuft-forming grass; deeply rooted specimen plants.
13	Cold-Tolerant No. 1		Gramineae	A kind of combined cold pasture. Herbaceous, evergreen, tuft-forming, deeply rooted specimen plants.
14	Alfalfa	*Medicago sativa* Linn.	Leguminosae	A cool season perennial legume, with height up to 1 m and a deep root system sometimes stretching to more than 15 m.

TPH analysis

TPHs in soil were examined. At first, TPHs were extracted from 5.0 g petroleum-contaminated soil, which had been previously sieved through a 4-mm sieve, and transferred to a 40-mL glass centrifuge tube, 25 mL chloroform was added, and the tube was closed to cover. Then they were ultrasonically extracted for 1 h. During the extraction, some cold water was added to keep the bath temperature below 40°C. After extraction, the samples were centrifuged for 10 min under 3000 rpm, and the extracts were transferred into an Erlenmeyer flask, dried to a constant weight, and bathed under 65°C to evaporate volatile chloroform. After evaporation of the solvent, the amount of residual TPHs was gravimetrically determined [6].

The removal rate of TPHs was calculated using the following expression:

$R = (M_2-M_1)/M2x100$

where M_2 is the concentration (mg kg^{-1}) of TPHs in soil before remediation, M_1 is the concentration (mg kg^{-1}) of TPHs in rhizosphere soil after remediation, and R is the removal rate of TPHs.

TPH composition and soil pH

The determination of saturated hydrocarbon, aromatic hydrocarbon, asphaltene, and polar compound in petroleum-contaminated soil was performed by separation using aluminum oxides. First, the sample was confected, after TPHs were examined, into ethane with 0.05 g mL^{-1}, saturated hydrocarbons were cleaned out using ethane with 50 mL after rendering the sample, and the effusing was received with a bottle and dried to a constant weight; afterward, aromatic hydrocarbons were cleaned using ethane and methylene dichloride (v/v=1:1) with 50 mL and the effusing was received with another bottle and dried to a constant weight; and then, asphaltene and polar compound were cleaned using methanol with 50 mL and the effusing was received with the third bottle and weighed after drying. Put the three bottles above in draught cupboard, until solvent in the bottles had been evaporated to a constant weight; then the saturated hydrocarbon, aromatic hydrocarbon, asphaltene, and polar compound were gravimetrically calculated, respectively. Soil pH was determined by the pH meter (pHs-3B) [7].

Statistical methods

Statistical analysis was carried out using the Excel XP, SPSS 17.0. Sampling and chemical analyses were examined in triplicate in order to decrease the experimental errors and to increase the experimental reproducibility. The confidence of data generated in the present investigations has been analyzed by standard statistical methods to determine the mean values and standard deviation (SD). The values in figures were expressed as mean±SD of the three replicates. Differences among treatments were analyzed by one-way analysis of variance (least significant difference test) [8].

RESULTS

Growth of plants and their biomass

The describes a variation in plant biomass among plant species in the study. After a 30-day culture, root and shoot biomass ranged from 1.55 to 5.76 g dry weight in TPH-contaminated soils. The tested plant species including #1, #3, #4, #5, and #8 grown in the contaminated soil yielded significantly (p<0.05) less dry weight than that in the control, particularly in #3 (*Adonis aestivalis* L.), and had decreased to about 50% (p<0.01) of that in the control. The biomass of tested plant species decreased in the petroleum-contaminated soil. A 77% reduction of ryegrass biomass after 30-day growth in soil contaminated with 25 g petroleum hydrocarbons kg^{-1}. The existence of 5 g diesel kg^{-1} soil led to a biomass reduction of 82%. In this work, the biomass reduction was less than the previous studies.

This may be attributed to the use of freshly spiked soil, which was reported to be more toxic to plants and microorganisms than aged contaminations, where pollutants are in general adsorbed to a higher degree on soil particles and therefore less available for growing. After a 30-day culture, the biomass of the species including #2 (*Antirrhinum majus*), #6 (*Gaillardia aristata*), and #14 (*Medicago sativa* Linn.) increased significantly (p<0.01) in contaminated soil than that in the control, whereas the biomass of #14 (*M. sativa* Linn.) in contaminated soil rose by 67.93% than that in the control. It may be because carbon in TPH-contaminated soil can be absorbed as a nutrition substance by #14 (*M. sativa* Linn.) and also because of its ability to fix atmospheric nitrogen as legumes. There was no significant correlation between the concentration of TPHs in contaminated soil and biomass of the tested plant species including 7, 9, 10, 11, 12, and 13 (p>0.05). However, 12 (Fire Phoenix) and 13 (Cold-Tolerant No. 1) were species whose yields increased a little in contaminated soil than that in the control. The differences may be attributed to the different organic compounds exuded by plants through their roots, which affect the density, diversity, and activity of specific microorganisms in surrounding rhizosphere [9].

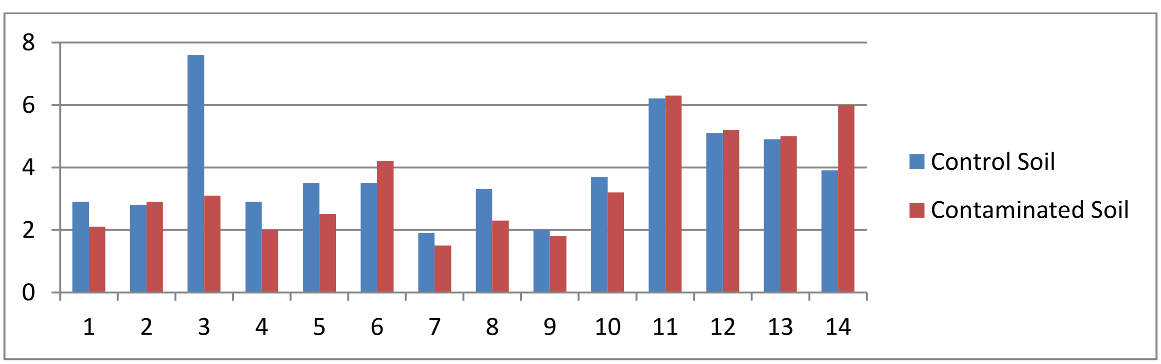

Figure 1: Growth of plant species tested and their biomass in control and rhizosphere soil at 30 days following transformation.

Treatment effectiveness of TPHs

After treatment by the specific plants, reduction in TPHs was observed during this study. After a 30-day culture, the removal rate of TPHs ranged from 19.54% to 49.42% in contaminated soil after remediation, and that was 12.93% in the control. The species #1 (*Centaurea cyanus*) (19.54%) and #2 (*A. majus*) (20.69%) had the lowest removal rate, whereas #7 (*Echinacea purpurea*) (46.74%) and #11 (Fawn) (49.42%) had the largest overall TPH removal rate among 14 species tested. The concentration of TPHs after remediation by #11 (Fawn) after a 30-day culture could reach the Canada industrial levels (5000 mg·kg^{-1}) for surface soil. The removal rate of TPHs may reach more than 35% for the tested plant species including #4, #5, #6, #10, #12, and #14 after a 30-day culture. The enhanced degradation of TPHs was observed in petroleum-contaminated soil compared with that in the control, by sowing plant species tested. This was in agreement with the results, who also observed a degradation of 64%–72% in 4700 mg TPHs kg^{-1} soil using Tall Fescue. It may be brought about by a combination of plant and soil interactions such as improvement of physical and chemical properties of a contaminated soil, increase in soil microbial activity, and increase in contact between rhizosphere microbes and TPHs in a contaminated soil. Further, the degradation mediated by plant-secreted enzymes in the rhizosphere could also cause the enhancement of TPH. The mechanisms of plant and soil interactions will be studied in the future. Integrating the results of biomass and the removal rate, we believe that the tested plant species including may have better ability of remedying petroleum-contaminated soils for further study [10].

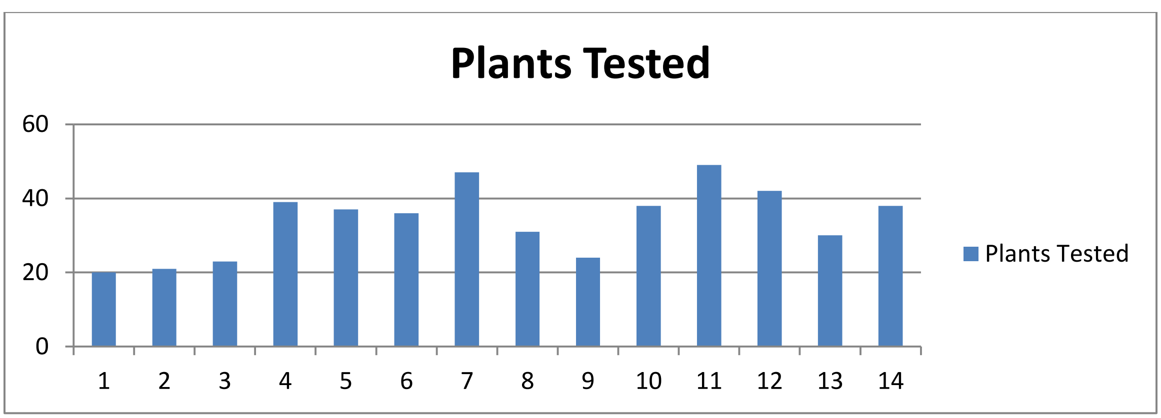

Figure 2: Removal rate of TPHs for plant species tested in control and rhizosphere soil at 30 days following transformation. TPHs, total petroleum hydrocarbons.

Changes in TPH composition

The total removal rates of various TPH composition including saturated hydrocarbon, aromatic hydrocarbon, asphaltene, and polar compound are depicted. There were great differences in the removal rate among 14 plant species tested. The removal rate ranged from 16.79% to 45.11% in the contaminated soil, higher than that in the control (only 6.9%). The degradation of saturated hydrocarbon fraction was mostly greater than that of the other two fractions, which was in line with other reports. The removal rate was higher for saturated hydrocarbons, for example, 20.81% by #7 (*E. purpurea*), 19.61% by #10 (Sandland No. 1), and 19.64% by #11 (Fawn) [11].

The fraction of asphaltene and polar compound had a lower degradation generally during phytoremediation. It might be because oxidation of TPH composition in soil depends on enzyme types, which are mostly related to the type of oil-degrading bacteria. More extensive degradation of saturated hydrocarbon than aromatic hydrocarbon, asphaltene, and polar compound in the rhizosphere of plants might be due to changes in the bacterial community structure, with special enzymes to oxidize these compounds. Further, TPHs with composition including aromatic hydrocarbon, asphaltene, and polar compound are hydrophobic solids and consequently are difficult to degrade because of their low water solubility and high aliphatic solubility. However, #11 (Fawn) (14.20%) and #12 (Fire Phoenix) (14.91%) contained significantly higher removal rate of asphaltene and polar compound after a 30-day culture than all other plant species tested, compared with that in the control (only 1.47%). It may be because more root biomass and root surface area, such as grass, which increased secretion of microbial enhancing metabolites such as water-soluble phenols, could also stimulate microbial activity in soil. It was also beneficial to the degradation of TPH composition [12].

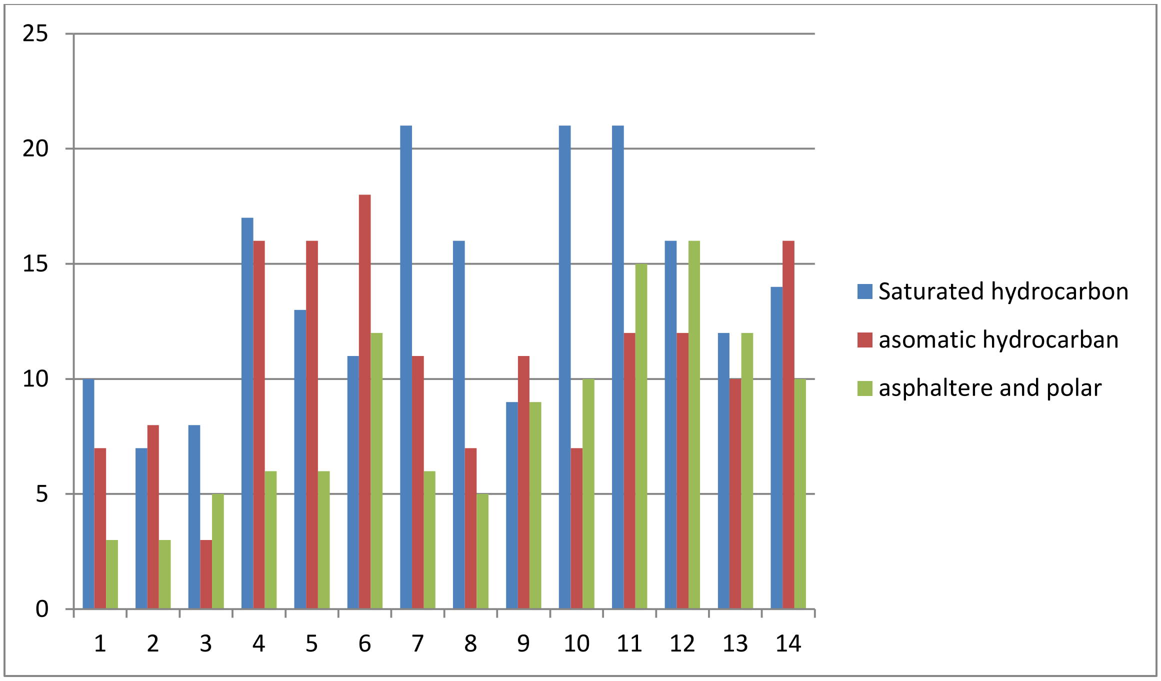

Figure 3: Removal rate of TPH composition for plant species tested in control and rhizosphere soil at 30 days following transformation

Changes in soil pH

A change of pH in planted soils after a 30-day culture. The little significant difference was observed among 14 species; it was in accordance with the reference. There was a little increase in soil pH, except 11 (Fawn) and 14 (*M. sativa* Linn.). This may be attributed to the difference in properties of rhizosphere secretion by sowing different plant species. pH values decreased after growing 11 (Fawn) and 14 (*M. sativa* Linn.), perhaps because organic acid occurred in process of growing plants and carbonic acid which was sacrificed, in the process that carbon dioxide formed from biorespiration was dissolved in water. The plants had a certain effect on special rhizosphere secretions (i.e., organic acid and citric acid) and rhizosphere microbial communities in plant treatment [13].

Fourier transform infrared analysis

The infrared (IR) spectra of the ornamental plants in noncontaminated soil as well as those planted in oil-contaminated soil were obtained using a Fourier transform infrared spectrometer (FTIR RX 1; Perkin-Elmer) [14]. For the FTIR study, 30 mg of finely ground biomass of whole ornamental plants (including roots and stems) was encapsulated in 300 mg of KBr (Sigma) in order to prepare translucent sample disks. The results clearly showed that the IR spectra of plants sown in contaminated soil and that of noncontaminated soil were showing different absorption patterns in the IR region. Particularly, the shifting of bands in IR spectra was observed in the hydrocarbon-contaminated plants around 1000, 1400, 1600, and 3400 cm^{-1} because of interaction between hydrocarbons of the oil and various functional groups present in these plants. Many of these bands shift toward the higher or lower frequency, clearly indicating the impact of oil contamination. For instance, the band around 1640 cm^{-1} in the IR spectra of ornamental plants may be attributed to the C=O functional groups present in the structure, which shift toward the lower wave number (3–15 cm^{-1}) in case of plants sown in petroleum-contaminated soil [15].

Conclusion

Considering these results for biomass, TPHs, TPH composition, and pH change, 6 (*G. aristata*), 7 (*E. purpurea*), 11 (Fawn), 12 (Fire Phoenix), and 14 (*M. sativa* Linn.) are plant species that have a larger potential for removing TPHs and its composition in petroleum-contaminated soil. This study has shown that phytoremediation using special ornamental species is one of the treatment methods in terms of effectiveness of TPH degradation in petroleum-contaminated soil.

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