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PHYTOREMEDIATION WITH AN AIR INJECTION SYSTEM

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Abstract : Phytoremediation is a plant-based technique with potential for enhancing the remediation of land contaminated by various pollutants. The use of deep-rooted plants for phytoremediation of contaminated soils has attracted a great deal of interest as an innovative alternative to other methodologies. As an innovative technique, in this study, air injected phytoremediation technique was introduced to enhance the remediation efficiency or to apply to the former soil vapor extraction or bioventing sites.

To select the plant species, plant-screening test was conducted by comparing growth rates that were affected by four levels of diesel addition. Alfalfa showed the strongest resistance against the contaminant and its growth rate was not inhibited by diesel additions as corns and barnyard grasses were. Effects of air injection, alfalfa treatment, and air injection with alfalfa treatments on the removal of hydrocarbon were investigated by column studies to simulate the field situation. Polyvinyl chloride (PVC) columns were packed with diesel-contaminated soils. Both the removal efficiency of diesel and the microbial activity were the highest in air-injected alfalfa-planted column soils. It was suggested that increased microorganisms activity stimulated by plant root exudates enhanced biodegradation of hydrocarbon compounds. Air injection has contributed to the promotion of the microbial activity. Air injected phytoremediation may be useful as a further approach to remediate diesel contaminated sites.

Key Words : Phytoremediation, air injection, alfalfa, diesel, remediation

INTRODUCTION

Phytoremediation is a promising plant-based technique for enhancing the remediation of land contaminated by various pollutants[1-3]. The use of deep-rooted plants for phytoremediation of contaminated soils has attracted a great deal of interest as an innovative alternative to conventional methodologies[4-7]. Although phytoremediation can be applied to large volume of slightly contaminated soils at a lower cost than the other techniques, plant-based remediation might have some restrictions because it uses solely naturally driven energy and mechanisms. Therefore, the combination of other techniques can be introduced to overcome these disadvantages and increase the remediation efficiency. Phytoremediation can be used as a supplementary technique or final treatment to the sites where other techniques have already been applied. Among other remediation techniques, bioventing and vapor extraction can be used with phytoremediation. Air injection may promote the microbial activity particular in anaerobic conditions. Air injection can enhance the physicochemical properties of the medium and contaminants and increase the bioavailability i.e., the plant and microbial accessibility to the contaminants. This technique can be introduced to enhance the remediation efficiency or to apply to the former soil vapor extraction or venting sites.

Experiments were conducted to investigate the

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effects of artificial air injection to the conventional phytoremediation and potential application as a supplementary technique or final treatment. Effects of air injection, alfalfa treatment, and air injection with alfalfa treatments on the removal of hydrocarbon were investigated by column studies to simulate the field situation

MATERIALS AND METHODS

Plant Selection and Germination Test

To select the plant species, plant-screening test was conducted by comparing growth rates that were affected by four levels of diesel addition. Alfalfa (Medicago sativa), corn (*Zea mays*), and barnyard grass (*Echinochloa crus-galli var. crus-galli*) were grown for 10 weeks in 0, 500, 1000, and 3000 mg TPH kg^{-1} of initially contaminated soils. Inorganic nutrients such as nitrogen and phosphorous, that can be limiting factors in bioremediation of organic contaminant[8] were provided. After germination, the shoot length of each treatment was measured every week to calculate growth rates.

Separate alfalfa germination tests at high diesel concentrations were conducted to evaluate the acceptable diesel concentration levels in phytoremediation applications. Alfalfa seeds were planted at a rate of 50 seeds per pot with five different concentrations; 0, 5000, 10000, 20000, and 30000 ppm. Germination rate were observed 7 days after seeding.

Column Preparation and Experimental Design

Multiple PVC pipes (0.24 m in diameter and 1.0 m in height) were installed in the greenhouse. Twelve holes (2 mm in diameter) from each depth of 0.25m, 0.5m, and 0.75m were made to collect samples. To inject air into columns, small PVC pipe (16 mm in diameter and 1.0 m in height) with twelve 3.2 mm holes, was inserted at the center of the columns. The coarse sand soils were filled up to 15 cm of the bottom of the columns (See Figure 1). Topsoil that was excavated from a neighboring hillock,

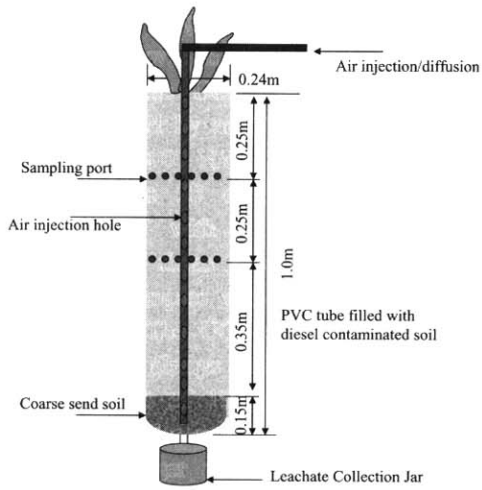


Figure 1. Schematic representation of the experimental column.

passed through a 2-mm sieve, initially contaminated with 782 ± 41 mg TPH kg^{-1} of soil was packed into the rest of the columns. Soil texture is classified as a sandy loam with organic matter content of 9.14%. Soil pH was adjusted to 7.0 using $CaCO_3$. Alfalfa was seeded 4 days after the column set up.

Four types of experiment were conducted to evaluate effectiveness of air injected phytoremediation for 100 days; treatment 1: unplanted columns; treatment 2: planted columns with no air treatment; treatment 3: columns unplanted but with an air treatment; and treatment 4: planted columns with an active air injection. Air from the compressor was supplied for 10 minutes in every 4 hour at 100 mL/min so that removal effect of TPH by air injection was minimized. The experimental units were watered weekly with deionized water to simulate 100 cm rainfall yr^{-1}. Moisture content in the soil column was maintained to 20-40% gravimetrically. Watering of the experiments did not allow water loss as leachate and the washing effect by water was not considered in calculation of TPH removal.

Soil samples were collected after 64 and 100 days of operation and PVC columns were cut off at day 100. The roots were carefully removed from the soil and the root length was measured manually.

Gas Chromatographic Analysis

5g of soil sample was obtained from each soil sample and 15g of Na_2SO_4 added to remove the soil moisture. Total petroleum hydrocarbons (TPH) was extracted from the dried soil samples with 10mL MtBE (Methyl tert-Butyl Ether) for 30 min using sonication extraction method. The supernatant was collected and filtered with membrane filter and stored at a temperature of 4℃. All extracted samples were analyzed on a Hewlett-Packard 5890 Gas Chromatograph equipped with a flame ionization detector.

Soil Microbial Activity

Soil microbial activity was also analyzed using INT (iodonitrotetrazolium) assay method.[9] 0.25% of INT solution (Sigma Chemical Co.) was added to 1 g of sample soil, incubated for 24 hours at 38℃. 10 ml of methanol was added, and mixed for 1 min using vortex mixer, then the supernatant was extracted and filtered through Whatman GF/C filter. Then, the sample was analyzed using U.V spectrometer at 480 nm.

Root Distribution

The soil was carefully removed from the roots by gentle washing in a water current and root length was measured. Then, the root distribution model developed by Sung et al.[10] was applied to simulate total root length and root distribution in both treatments using the experimental data. The root distribution in terms of rooting density can be written as

$$L_d = \frac{L_T f}{A\left(1-e^{-fz_m}\right)} e^{-fz} \tag{1}$$

where L_d is the rooting density, defined as the length of roots per unit volume of soil (cm cm^{-3}), L_T is the total root length (cm), f is a constant over soil depth at a given time period for plant growth (cm^{-1}), A is the cross-sectional area normal to the z-axis (cm^2), z_m is the rooting depth (cm) at time t, and z is the depth (cm), respectively. The rooting depth under favorable environmental conditions for root growth can be described as a sigmoidal curve.[11]

$$z_m = z_T S_{nf} \tag{2}$$

$$S_{nf} = [0.5 + 0.5\sin[3.03(t/t_T) - 1.47]] \tag{3}$$

where z_T is the maximum rooting depth to be achieved at t = t_T , i.e. time to maturity, under given conditions. The S_{nf} term is a sine function derived by Borg and Grimes by analyzing 135 reported field observations [1986] and demonstrated good performance at predicting root distribution in various soil systems.[12] Because total root length and rooting depth are related to plant growth, total root length (L_T) can also be described in the same way using a rooting depth expression with the assumption that those variables also followed a sigmoidal function.

$$L_T = L_{TT} S_{nf} \tag{4}$$

where L_{TT} is the maximum value of total root length (cm).

RESULTS AND DISCUSSION

The inhibition effect on plant growth of TPHs was estimated by

$$Inhibition\ (\%) = \frac{\left(\frac{\Delta L_0}{\Delta t} - \frac{\Delta L_C}{\Delta t}\right)}{\left(\frac{\Delta L_0}{\Delta t}\right)} \times 100 \tag{5}$$

where $\Delta L_0/\Delta t$ and $\Delta L_c/\Delta t$ are the growth rates of the plant in control (uncontaminated) and contaminated soils, respectively. Alfalfa showed the strongest resistance against the contaminant and its growth rate was not inhibited by diesel additions as corns and barnyard grasses were. At 3,000 ppm, growth rates of alfalfa, barnyard grass, and corn were decreased to 10.1, 30.1 and 31.8%, respectively, compared with

Table 1. Growth inhibition(%) of plants grown at diesel-contaminated soil

Concentration (ppm)	Inhibition (%)		
	Alfalfa	Barnyard Grass	Corn
500	-44.3*	0.0	-0.3
1000	-25.3*	20.8	15.5
3000	10.1	30.1	31.8

*Minus sign means the increased growth rate

Table 2. Germination rate of alfalfa in diesel contaminated soil

Diesel Concentration (ppm)	Germination Rate (%)
0	72
5000	82
10000	60
20000	6
30000	2

plants grown in uncontaminated soil (Table 1). On the contrary, the growth rate of alfalfa increased up to 44% and 25% at lower concentration (500 and 1,000 ppm). Alfalfa germination test showed that alfalfa also could be applied up to 5,000-10,000 ppm of diesel-contaminated soils (Table 2) although the germination rate decreased rapidly above 10,000 ppm. The above results suggest that alfalfa can be an appropriate plant to remediate soils contaminated by diesel.

The rooting density profiles of alfalfa and alfalfa with air treatment at each depth were

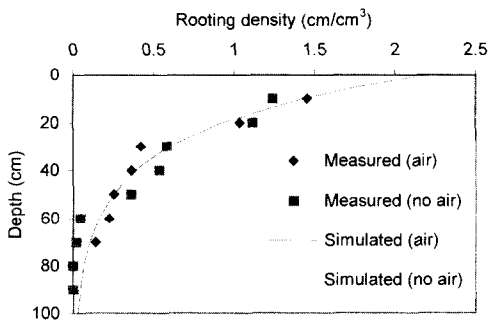


Figure 2. Comparison between measured and simulated spatial variation of rooting density of (a) alfalfa-planted column and (b) alfalfa planted with air treatment column at 100 day after seeding.

Table 3. Parameters used in root distribution model simulation

Parameters	Notation	Values
Maximum rooting depth	Z_T	100cm
Plant maturity time	t_T	100day
Cross-sectional area normal to the z-axis	A	24cm^2
Maximum value of total root length (Alfalfa + air injection)	L_{TT}	1200cm
Maximum value of total root length (Alfalfa only)	L_{TT}	950cm
Constant over soil depth for plant growth (Alfalfa + air injection)	f	30cm^{-1}
Constant over soil depth for plant growth (Alfalfa only)	f	25cm^{-1}

shown in Figure 2. There was no significant difference of rooting density between the treatments in the upper soil. However, measured rooting density of alfalfa with air treatment was higher in the lower section of the column than alfalfa without air treatment. Root distribution model[10] was applied to simulate total root length and root distribution in both treatments using the experimental data. The results showed that total root length was also higher in the air treated column (1,200 cm) than just alfalfa planted column (950 cm). The parameter values used in the model were listed in Table 3. Those results suggested that soil aerobic conditions caused by air injection might be helpful for root growth in depth below 50 cm where O_2-limiting conditions might be present.

The soil microbial activity in each treatment was presented in Figure 3. Alfalfa planted soil with air treatment showed the highest microbial activity in the upper soils of column as well as at 50 cm depth. The microbial activity declined in the order of alfalfa with air treatment, alfalfa without air treatment, air treatment soil, and control soil at 25 cm depth of columns and alfalfa with air treatment, air treatment soil, alfalfa without air treatment, and control soil at 50 cm depth. Results suggest that air injection could provide sufficient opportunity for promoting the microbial activity in the lower soil. The root and microbial activity data also

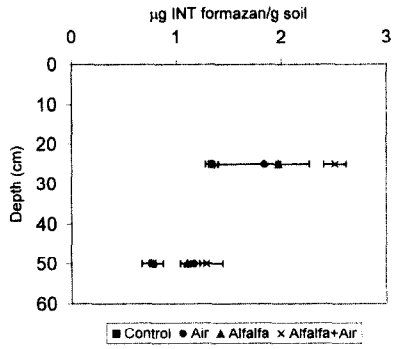


Figure 3. Dehydrogenase activity (DHA) profiles in each treatment at 100 day after seeding.

suggested that increased microorganisms activity stimulated by plant root exudates and air provision may enhance biodegradation of hydrocarbon compounds.

It is observed from the results that the THP concentrations in soil were affected by microbial degradation and transport to the lower sections with time (Figure 4). In the top soil of the column (0 to 25 cm) there was a constant TPH dissipation rate throughout the entire experiment period (Figure 4(a)). In the lower sections (25 to 50 cm), however, different dissipation rates were applied with time. Until day 45, the TPH concentration of each treatment were almost the same or even higher than initial concentrations, then slow dissipation until day 64, and then fast dissipation during the end of the experiment were occurred (Figure 4(b)). The results suggested that contaminant migration with water flow could be a main contribution of TPH dissipation in the top soil until day 45 although the slow microbial degradation occurred as well. The results explained that the TPH concentrations in the lower sections showed the same or even higher than initial concentration. TPH dissipation due to microbial degradation increased with time and almost the same dissipation rates with topsoil showed until day 100.

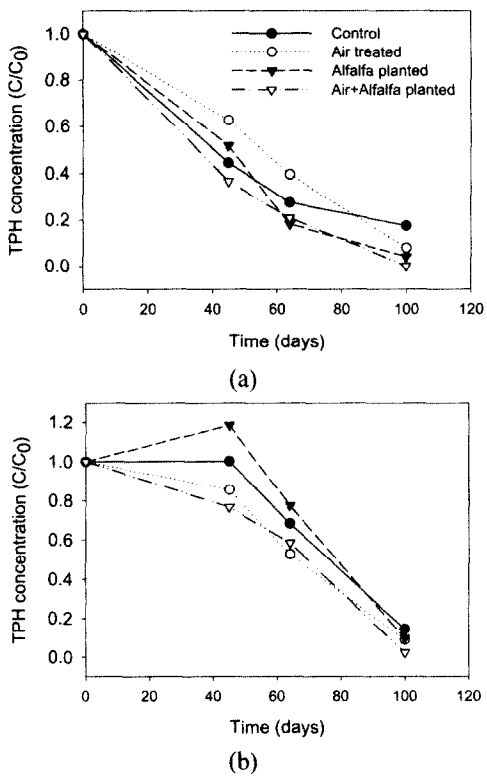
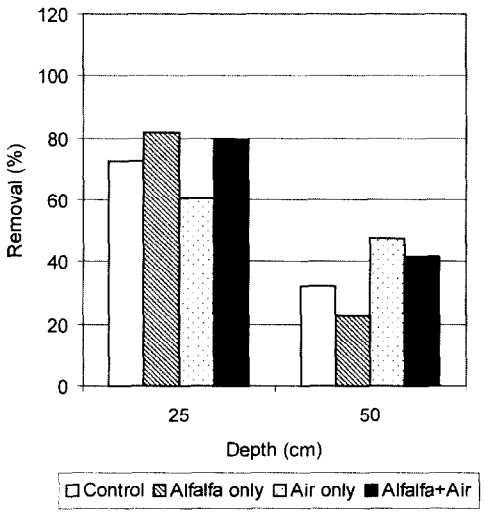


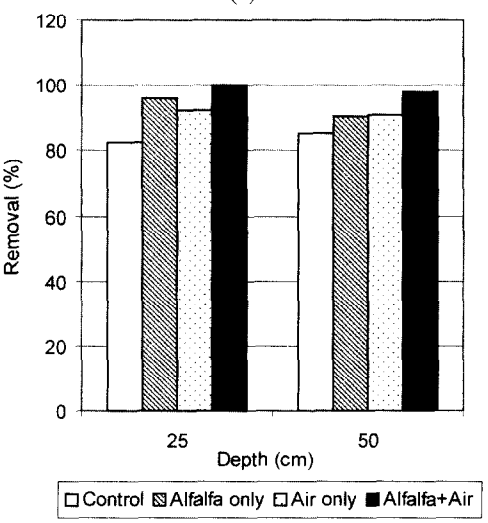
Figure 4. Relative concentration (C/C0) of TPH change with time at depth of (a) 0 to 25 cm and (b)25 to 50 cm.

In the topsoil, there were no distinct effects from plants or air injection on TPH dissipation in soils. Those are because the plant roots were not fully grown to provide sufficient substrates for microbial activity and proper air could also be provided from the ambient air in the topsoil when there was no artificial air treatment. The results also supported that the main dissipation mechanism could be the migration with water flow not microbial degradation as mentioned earlier. However, it is noticeable that there was more dissipation of TPH at air treated columns at lower sections at day 45 and day 65 (Figure 4(b)). The results suggested that the artificial air treatment can enhance the remediation efficiency in the deep soil, especially in the early period, when the roots are not fully developed. Until day 100 after alfalfa seeding, there was clear distinction among the treatments and alfalfa planted with air treatment column showed the lowest concentration and the results showed that both plant and artificial air could provide favorable environment for enhanced dissipation of TPH in phytoremediation.

TPH removal rates at 64 days and 100 days after alfalfa seeding were shown in Figures 5(a) and (b), respectively. At depth 25cm, alfalfa and alfalfa with air treatment showed higher reduction of TPH than treatments without plants. Because roots did not develop fully to lower column until day 64 and the alfalfa root mainly present in the upper soil, the dissipation of TPH by plant root mainly occurred at upper soil in the alfalfa planted column. At depth 50 cm, however, columns with air treatment showed higher TPH reduction than columns without air treatment. Alfalfa with the air injected column showed high removal efficiency in both depths and the results suggested that the artificial air treatment could enhance the remediation efficiency in the early period when the roots are not fully developed.



(a)



(b)

Figure 5. TPH removal rate of control, alfalfa planted, air injected, alfalfa with air injected treatments (a) at 64 days after seeding and (b) at 100 days after seeding.

In upper soil, alfalfa with air treatment and alfalfa without air treated columns showed high removal efficiency (100 and 96 %, respectively) than air treated or control columns (92 and 82%, respectively). The results showed that the plant effect was greater in the upper soil where the rooting density was high, and phytoremediation can be an effective remediation method for diesel contaminated soil in this case. However, the TPH reduction in the alfalfa with air treatment was greater than the other treatments at 50 cm depth where O_2-limiting conditions were present, with reduction of 98%, compared with 80% in control, 91% in air treatment only, and 90% in alfalfa planted soils. That is due to the air injection effects on the root growth and microbial activity where anaerobic conditions were present. The results suggested that the phytoremediation with air treatment could enhance the conventional phytoremediation methods by provide additional opportunity for promoting the microbial activity. Increased microbial activity due to air injection in conventional phytoremediation sites can reduce plant contamination.

It is suggested from the results that air injected phytoremediation system can be used as a supplementary technique at conventional phytoremediation sites. It can also be applied at the sites where bioventing or soil vapor extraction methods had been applied as a final or a complementary treatment.

CONCLUSIONS

The following conclusions were drawn from this study.

- Alfalfa showed the strongest resistance against the contaminant and its growth rate was not inhibited at diesel contaminated soils. On the contrary, the growth rate of alfalfa increased up to 44% and 25% at lower concentration (500 and 1000 ppm). Alfalfa could also germinate at soil highly contaminated by diesel (5000- 10000 ppm).
- Rooting density of alfalfa with air treatment was higher in the lower section of the column than alfalfa without air treatment. Air provision could be favorable for root growth at contaminated soil.
- Both the removal efficiency of diesel and the microbial activity were the highest in air-injected with alfalfa planted column soils. This study showed that the air injected phytoremediation technique can be introduced to overcome disadvantages of conventional phytoremediation method and increase the remediation efficiency.

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