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Laboratory Studies of Air Stripping of VOC-Contaminated Soils

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ABSTRACT: Bench-scale laboratory experiments were conducted to evaluate the effectiveness of air stripping for *in situ* remediation of benzene-contaminated soils. Several parametric studies were performed to evaluate the effect of soil grain size, air injection flow rate, and air inlet temperature on the benzene recovery efficiency.

An increase in soil grain size produced a significant increase in benzene recovery efficiency especially during the early stages of air injection. After 2 h of treatment, an increase in soil grain size from $D_{50} = 0.31$ mm to $D_{50} = 1.20$ mm resulted in an increase in benzene recovery efficiency from 49 to 65%. When the air-flow rate was increased from 5 l/min to 10 l/min, the benzene recovery efficiency increased from 56 to 70% after 4 h of venting operation. Maximum recovery of benzene was reached after approximately 37 h of soil venting at a flow rate of 5 l/min and after approximately 24 h at a flow rate of 10 l/min. Preheating the air to 45°C at the inlet resulted in an increase in recovery efficiency from 70 to 90% after 5 h of air stripping.

KEY WORDS: soil vapor extraction, soil remediation, contaminated soils, air stripping.

I. INTRODUCTION

Soil contamination by volatile organic chemicals (VOCs) has become a major environmental problem in many industrialized countries. These contaminants are introduced into the subsurface as a result of accidental surface spills, leakage from underground storage tanks, or waste disposal. In general, VOCs are highly toxic and water soluble, and their presence in soils poses a serious threat to groundwater. In recent years, several *in situ* remediation techniques have been developed for soils contaminated by VOCs, including biological degradation, vapor extraction, steam stripping, vitrification, supercritical solvent extraction, low temperature thermal desorption, and radio frequency heating. Among these techniques, soil vapor extraction (also called air stripping or soil venting) has become one of the fastest growing *in situ* remediation techniques because of its relatively low cost and simple design and operation. Air stripping has been studied by several investigators and tested extensively in the field, showing its effectiveness as an *in situ* remediation technique. A soil vapor extraction operation consists of pumping air through one or more wells within the unsaturated zone. The induced air flow causes *in situ* evaporation of VOCs, volatilization of chemicals dissolved in residual pore water, and desorption of chemicals from soil surfaces. Mobilized organic vapors and air move along the induced flow path to the withdrawal well, where they are removed.

Experimental work reported so far on vapor extraction is confined to a few laboratory studies performed under controlled conditions. Among recently reported investigations, an experimental study of air stripping in homogeneous columns of gasoline-contaminated sand was conducted by Baehr *et al*. (1989), who showed that the gasoline removal rate was proportional to the air-flow rate and that essentially all of the gasoline in the soil column was eventually removed. They observed that the contaminant concentration in the effluent air declined with increasing time of treatment due to a shift in equilibrium that was caused by the selective removal of more volatile hydrocarbons. Ho and Udell (1992) performed two-dimensional experiments in homogeneous and heterogeneous porous media, and displayed a visual observation of the evolving geometry of the liquid contaminant pool under various conditions. They also investigated the mechanisms of multicomponent evaporation and transport during soil venting. Lingineni and Dhir (1992) conducted an experimental study of air stripping to determine the evaporation rates of multicomponent miscible contaminants in the soil. Results from their experiments indicated lowering of soil temperatures during venting, and they found that preheating the inlet air significantly improved the removal rates of heavier fractions. One-dimensional column experiments were also conducted by Katsumata and Dhir (1992) to examine the effects of preheating the air in a soil-venting system. Their results showed that the gasoline components were being removed according to their volatility such that the higher-volatility components were being removed first, and preheating significantly increased the removal rate of each component. More recently, Ho and Udell (1993) conducted one-dimensional experiments to study two limiting conditions of air flow in heterogeneous geologic settings: (1) ideal through flow through contaminated regions and (2) a bypass flow around the contaminated region. They examined in detail the mechanisms of evaporation and transport for each case. A recent literature survey revealed the lack of detailed parametric studies of the efficiency of soil vapor extraction for *in situ* remediation of VOC-contaminated soils.

The main objective of this study was to conduct several laboratory experiments and correlate the effects of three important parameters on VOC removal efficiency using air stripping: (1) soil grain size, (2) air injection flow rate, and (3) air injection temperature.

II. APPROACH

Laboratory experiments were conducted in an instrumented one-dimensional soil column as shown in Figure 1. The column was 61 cm long and 5.1 cm in diameter and was packed uniformly with the selected soil sample. The pressure gradient across the soil column was measured using two high-accuracy pressure gauges placed at each end of the column. A differential manometer was used to measure the small pressure gradients that occurred in soils with coarser grain size. Steady air flow was provided to the soil column using an air compressor. The air-flow rate was modulated by an adjusting valve on the compressor and measured by a high-precision rotameter. At the exit from the column, the vapors were routed through a vapor treatment unit before they were discharged into the atmosphere through a fume hood. Vapor samples were collected in a 250-ml gas sampling glass tube that was fitted with inlet and outlet valves and a septum port. To determine the benzene concentration in the air, the gas samples were analyzed using a packed column gas chromatograph instrumented with a flame ionization detector (Varian 3700 GC/FID). With this measurement system, the detection limit of benzene in the effluent gases was 0.1 mg/l (Lingineni and Dhir, 1992), with a random error of approximately 5%. Copper-constantan thermocouples were placed at regular intervals along the column centerline. The thermocouples were connected to an automatic signal scanner and a digital thermometer that displayed the temperature readings.

The selected soils were characterized by their grain-size distribution. The soil samples were prepared and packed into the column as described by Hadim *et al.* (1993). The permeability was determined using Darcy's law and measured values of air-flow rate and pressure drop.

Initially, the soil column was mounted in the vertical position and saturated with water, which was allowed to drain under the influence of gravity until a field

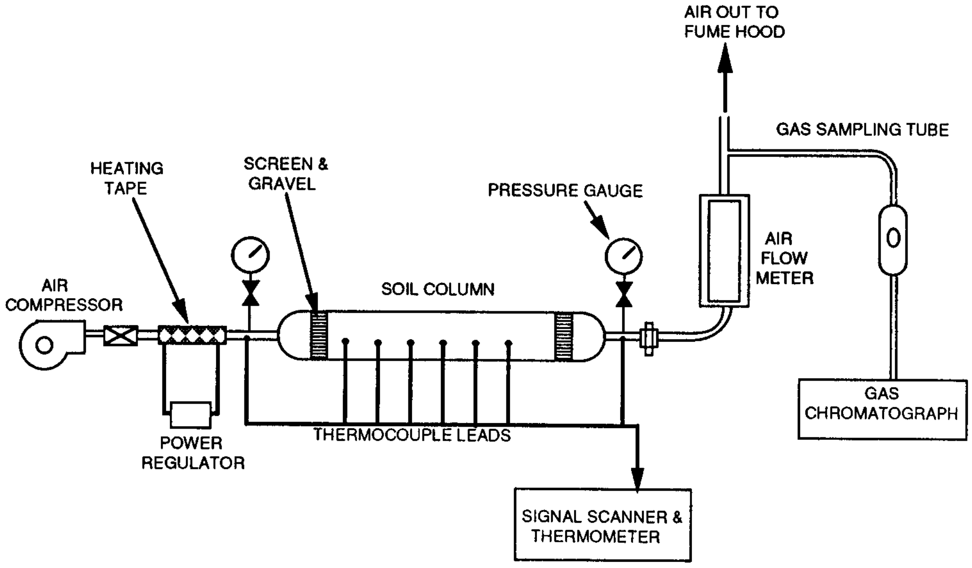


FIGURE 1. Schematic diagram of the experimental setup.

capacity condition was simulated. A measured quantity of benzene was then added at the top of the soil column and allowed to drain from the column until equilibrium was reached. This procedure simulated the movement of benzene in the vadoze zone. The residual saturation of benzene in the soil column was determined from mass balance.

The contaminated soil column was then connected to the set-up shown in Figure 1. Atmospheric air was injected into the column at a prescribed flow rate. Using the gas-sampling tubes, air samples were collected from the column at regular intervals. Samples of 20 μl were extracted from the gas-sampling tubes using a syringe and then injected into the gas chromatograph for analysis. The benzene removal rate was calculated as a product of the steady air-flow rate and the concentration of the contaminant in the effluent air (Johnson *et al.*, 1990). The concentration was estimated by numerically integrating the variation of the benzene concentration over a selected time interval. The experiments were run until the benzene concentration in the air samples was found to be below the detection limit of the gas chromatograph. In addition, soil samples were taken from the inlet, center, and exit of the soil column, and their analysis by gas chromatography confirmed the absence of benzene in the soil after treatment.

III. MECHANISMS INVOLVED DURING SOIL VENTING

The mechanisms affecting vapor movement and the removal of volatile contaminants by vapor extraction include gas advection, gas diffusion, gas-organic-water-soil chemical equilibrium and mass transfer, liquid diffusion, and water and organic phase advective and dispersive transport. All these mechanisms may be important, but it is not possible to distinguish individually their relative impact on the effectiveness of vapor extraction. In most cases, removal of the organic phase is dominated by advection, but the other mechanisms are also important within the dissolved and soil phases.

The efficiency of a soil vapor extraction system depends on the properties of the soil and contaminant and on the process conditions. The contaminant properties affect the distribution of the contaminant among four states: vapor, dissolved in pore water, sorbed to soil particles, and as a separate nonaqueous-phase liquid (NAPL). This distribution of the compound is affected by its degree of partitioning into the vapor phase, its ability to adsorb to soil particles, and various site-specific conditions. The rate and degree of partitioning of the residual organic liquid in the soil into the vapor phase and dissolved phase depend on the type of soil and the contaminant chemical composition. Initially, when a contaminant is at a residual saturation condition in a soil, an equilibrium is established among all three phases (gas, liquid, and solid). During soil venting, removal of contaminated air causes an equilibrium shift as fresh air replaces the contaminated vapors. The contaminant remaining as residual liquid volatilizes into the fresh air, seeking to establish equilibrium. As the process continues, more and more contaminant in the residual liquid is “mobilized” into the vapor phase and removed by the soil-venting system.

A contaminant’s volatility is directly associated with its degree of partitioning into the vapor phase. Highly volatile compounds such as gasoline and some chlorinated solvents have a greater tendency to exist in a vapor phase than less volatile products such as heating oil. The volatility of the product is the most important property affecting the efficiency of soil vapor extraction. A compound’s degree of partitioning is also affected by its vapor pressure, its solubility in water, and the soil sorption coefficient.

The air permeability of the soil incorporates the effects of several soil characteristics, including grain size distribution, stratigraphy, air-filled porosity, water content, and contaminant residual saturation.

In general, during a vapor extraction operation, the contaminant concentration in the extracted vapor declines over time according to three performance phases (Anderson, 1993). During the initial phase (“flushing” phase), contaminant vapor concentrations are relatively high, reflecting the long-term soil-gas equilibria condition. During the second phase (evaporation phase), the contaminant is rapidly removed from the more permeable layers through which extracted vapor readily flows. The third phase (diffusion phase) occurs when contaminants are removed more slowly by diffusion from the less permeable regions of the formation to the more permeable and cleaner zones.

IV. RESULTS

Several experiments were performed in order to correlate the effects of soil grain size distribution, air injection flow rate, and air inlet temperature on the VOC recovery efficiency. In all the experiments, benzene was selected as the contaminant. The air-flow rate was fixed at 10 l/min for all experiments except when the effects of air-flow rate were studied. Except for the case when the air was preheated to 45°C, all the other experiments were conducted by injecting air at the ambient temperature of 22°C. The properties of the selected soils are listed in Table 1. Other relevant experimental data are listed in Table 2.

TABLE 1
Physical Properties of the Selected Soil Samples

U.S std. sieve #	Mean grain size (D_{50}, mm)	Porosity	Hydraulic conductivity (cm/s)	Air conductivity (cm/s)	Permeability (m^2)
10–20	1.20	0.39	7.64×10^{-1}	5.56×10^{-2}	8.6×10^{-10}
20–40	0.61	0.36	1.14×10^{-1}	7.76×10^{-3}	1.2×10^{-10}
40–60	0.31	0.33	6.65×10^{-2}	4.16×10^{-3}	6.7×10^{-11}
60–100	0.22	0.31	1.66×10^{-2}	1.29×10^{-3}	2.0×10^{-11}

TABLE 2
Experimental Data

Mean grain size (mm)	Flow rate (l/min)	Initial concentration of benzene in air[a] (mg/l)	Air inlet temperature (°C)	Treatment time (min)	Maximum recovery efficiency %
1.20	10.0	457.0	22.0	1255	97.1
0.61	10.0	353.0	22.0	1402	97.6
0.31	10.0	340.0	22.0	1870	97.8
0.61	10.0	356.3	45.0	1220	100.0
0.61	5.0	342.4	22.0	2200	99.8

[a] After 30 s of air stripping.

A. Effect of Soil Grain Size

Experiments were conducted with three types of soils having a uniform grain size and contaminated with benzene. Contamination of the soil column by gravity drainage yielded a residual saturation of benzene that varied from 11% in the coarse-grained soil (D_{50} = 1.2 mm) to 17% in the fine-grained soil (D_{50} = 0.31 mm). Due to higher capillary forces in finer soils, the benzene residual saturation increased with decreasing particle diameter.

At the beginning of air injection, the concentration of benzene in the effluent air was higher in coarser soils (Table 2). In fact, experiments with the soil having the larger grain size (D_{50} = 1.2 mm) revealed that the benzene concentration in the effluent air at the beginning (after 30 s) of air injection was 457 mg/l and was higher than the saturation limit of benzene in air (320 mg/l, 20°C). This is due to the presence of droplets of liquid benzene and water in suspension in the exiting air during the early stages of soil venting. A similar observation was made by Zaman (1989) and Marley (1985).

In the present experiments, the variation of benzene concentration with time of treatment showed trends similar to the general ones discussed in Section III. During air injection, clean air replaced contaminated air within the column and the liquid contaminant in the pores volatilized into the clean air, seeking to establish equilibrium. The variation of benzene concentration with time of treatment showed a similar trend for all three types of soils (Figure 2). After the beginning of air injection, the benzene concentration decreased rapidly with increasing time of treatment (evaporation phase) and approached a very low asymptotic value during the final stage of air injection (diffusion phase). During the beginning of air injection, the benzene removal rate was higher in the coarser soil, as explained earlier. Beyond the initial stages of air injection, the benzene removal rate was higher in finer soils because the grain surface area is relatively higher in finer soils

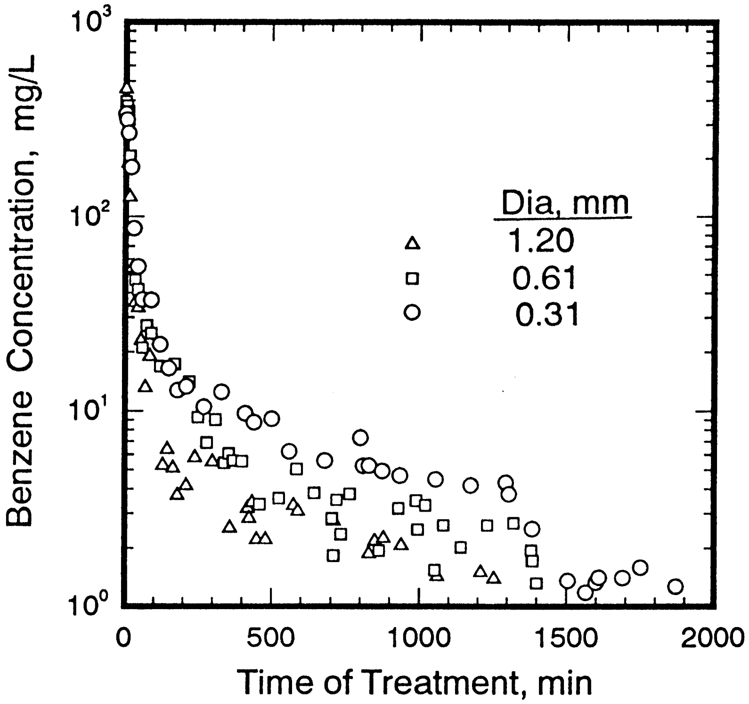


FIGURE 2. Effect of soil grain size on benzene concentration in the effluent air.

than in coarser soils, such that the volatilization of benzene is increased due to increased contact with the flowing air. As the time of treatment increased, the benzene concentration in the effluent air maintained a higher value in finer soils than in coarser soils (Figure 2).

For the range of soil grain sizes considered, the final benzene concentration was below the detection limit of the gas chromatograph and treatment was completed when no benzene was detected in the effluent air sample. As the soil grain size decreased, the time of treatment increased.

The results presented so far are illustrated in Figure 3, which shows the variation of recovery efficiency with time of treatment for the three selected soils. The removal efficiency increased exponentially with time of treatment, and it approached asymptotically a maximum value at the end of treatment. It is shown that maximum recovery was achieved faster in coarser soils due to the relatively large amount of contaminant that was recovered at the beginning of air injection, as discussed earlier. After 2 h of treatment, approximately 65% of the benzene was removed in the soil with grain size D_{50} = 1.2 mm, while within the same time span, the removal efficiency was approximately 49% in the finer soil (D_{50} = 0.31 mm).

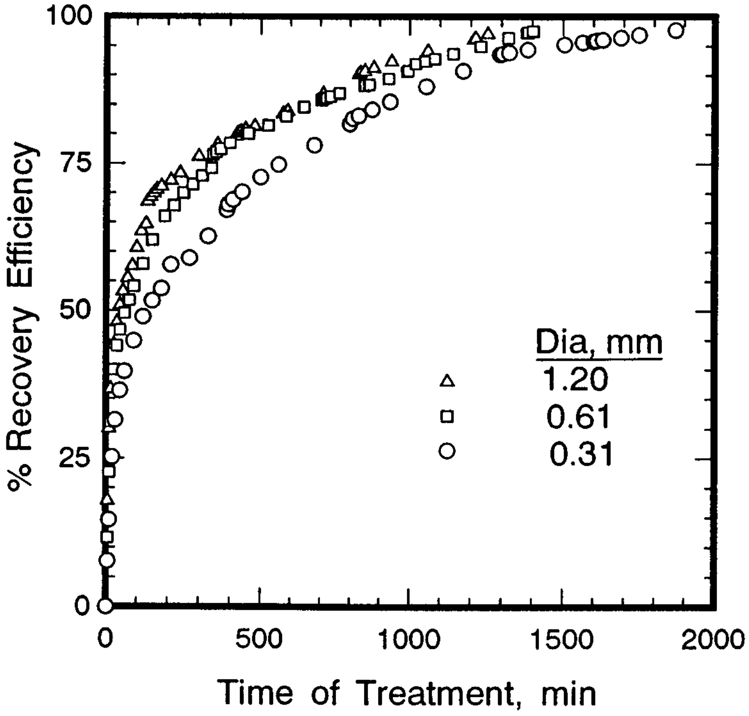


FIGURE 3. Effect of soil grain size on benzene removal efficiency.

B. **Effect of Flow Rate**

In order to investigate the effect of air-flow rate on the mass transfer rate of benzene from the immiscible and water phases to the air phase, experiments were conducted with air-flow rates of 10 and 5 l/min. During this phase of the study, all tests were performed using soil samples having a grain size of D_{50} = 0.61 mm and contaminated with benzene.

At the beginning of air injection, the concentration of benzene in the effluent air was higher at the higher flow rate because more liquid droplets of benzene and water were transported with the air to the exit of the soil column, as indicated earlier. Beyond the initial stages of air injection, the benzene concentration in the effluent air sustained a higher value at the lower flow rate, as shown in Figure 4. At the lower flow rate, the interstitial velocity of the air was lower such that the contact time of the air with the soil was higher, which resulted in a higher mass transfer rate of benzene from the liquid to the gas phase, as discussed in Section III. After 2 h of venting, the benzene concentration in the effluent air increased from 17 mg/l at the flow rate of 10 l/min to 35 mg/l at a flow rate of 5 l/min.

Because the removal rate is proportional to the air-flow rate (Crow *et al.*, 1987; Baehr *et al.*, 1989) and a relatively larger amount of contaminant was recovered

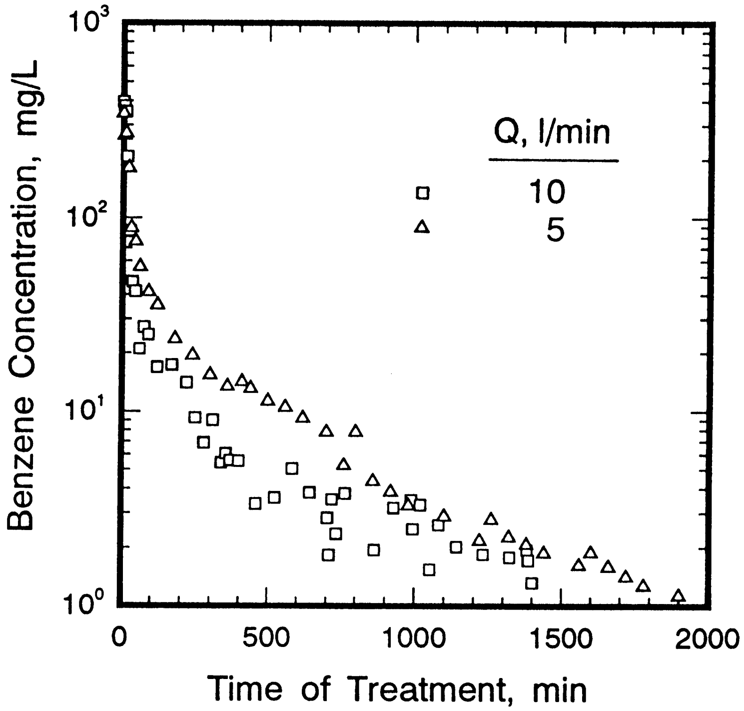


FIGURE 4. Effect of air injection rate on benzene concentration in the effluent air.

at the higher flow rate during the beginning of air injection, a higher recovery efficiency was obtained at the higher flow rate of 10 l/min (Figure 5). Maximum recovery efficiency was reached after approximately 37 h of venting at a flow rate of 5 l/min and after approximately 24 h at a flow rate of 10 l/min. Therefore, as the flow rate was increased, the time of treatment of the contaminated soil was reduced.

One experiment was conducted in a pulsed venting mode such that the air compressor was shut off at regular intervals. As shown in Figure 6, the benzene concentration in the effluent air peaked sharply during each temporary shutdown of the compressor, suggesting that a higher amount of contaminant was removed. During shutdown of the compressor, the contaminant that remained adsorbed to the soil surface diffused slowly into the air within the pores (as discussed in Section III) before it was recovered when the compressor was started again. When air stripping was resumed, the benzene concentration in the effluent air decreased rapidly to a concentration that was less than the value before the shutdown because the time required to reduce the amount of hydrocarbon vapors in the soil was much less than the time required for the vapors to reestablish their original concentration levels in the soil (Crow *et al.*, 1987). For this experiment, the variation of benzene

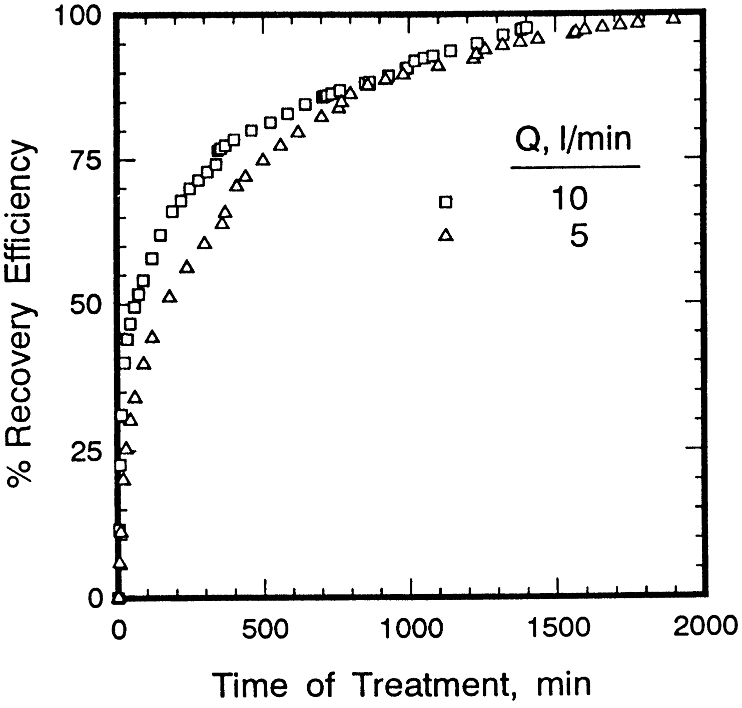


FIGURE 5. Effect of air injection rate on removal efficiency.

concentration with time of treatment was similar to what occurred during continuous operation except for the sharp peaks that occurred during compressor shutdowns. The resulting recovery efficiency during pulsed venting was nearly the same as during continuous operation at the same flow rate, but the time of treatment was reduced. This suggests that pulsed venting may be a cost-effective technique due to its lower energy requirements.

C. **Effect of Air Inlet Temperature**

It is well known that soil vapor extraction becomes less effective for compounds with a vapor pressure less than 0.1 mmHg at ambient temperatures (Anderson, 1993). However, the range of applicability of this technique can be extended by heating the subsurface or by injecting hot air because the vapor pressure of a VOC increases with increasing temperature, leading to a higher volatilization rate. The vapor pressure is related to the driving force required for the movement of the vapors from the liquid to the gaseous phase. For most organic compounds having an intermediate molecular weight (e.g., benzene), vapor pressure increases three to four times for each 10°C rise in temperature (Jury and Ghodrati, 1989).

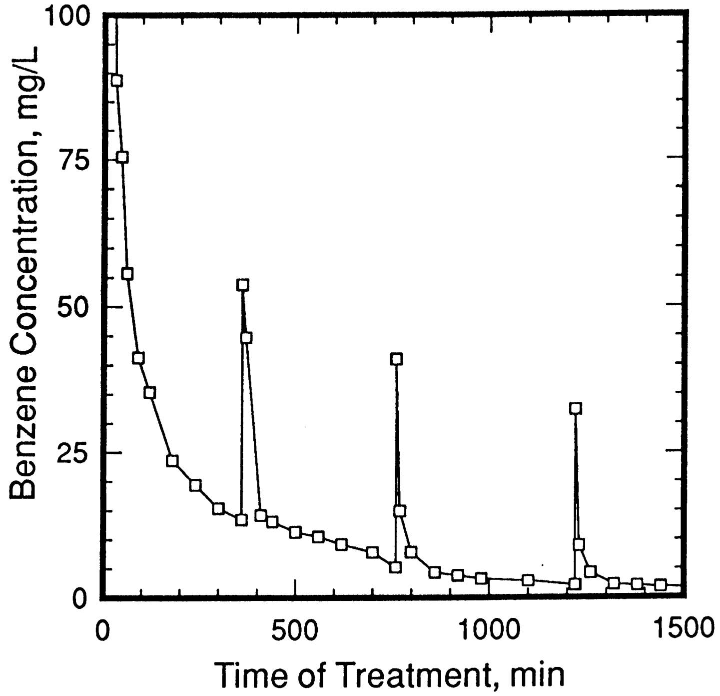


FIGURE 6. Variation of benzene concentration in the effluent air with time of treatment during pulsed venting.

One experiment was carried out to examine the effect of preheating the air on the efficiency of air stripping. Medium-grained soil (D_{50} = 0.61 mm) contaminated with benzene was used in this experiment, and the air was preheated to 45°C at the inlet. Due to the low thermal diffusivity of the soil, heat propagation through the soil column by conduction was very slow. Hence, heating of the soil downstream of the inlet region was mainly due to convective heat transfer from the upstream hot air.

In Figure 7, the variation of benzene concentration with time of treatment is displayed for unheated air and preheated air. At the beginning of the venting process, the difference between the two cases was very small because heating of the air had not yet had a chance to affect the initially high removal rates (Katsumata and Dhir, 1992). As air stripping continued, the benzene concentration in the effluent air maintained a significantly higher value in the case with preheated air. The removal rate with preheated air was significantly higher than with unheated air (Figure 8). After 2 h of air stripping, it was found that approximately 76% of the contaminant was removed with the preheated air, while only 56% of the benzene was stripped from the soil with unheated air. As shown in Figure 7, the remediation time was substantially reduced when the air was preheated.

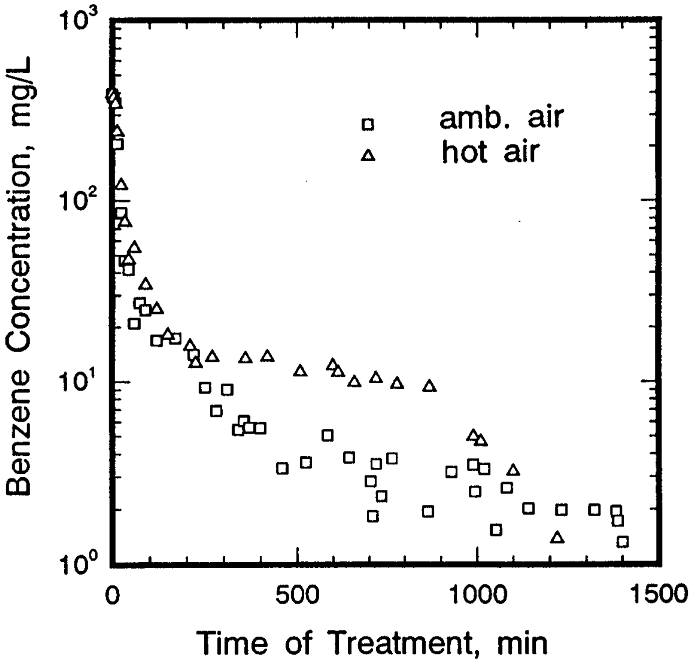


FIGURE 7. Effect of air inlet temperature on benzene concentration in the effluent air.

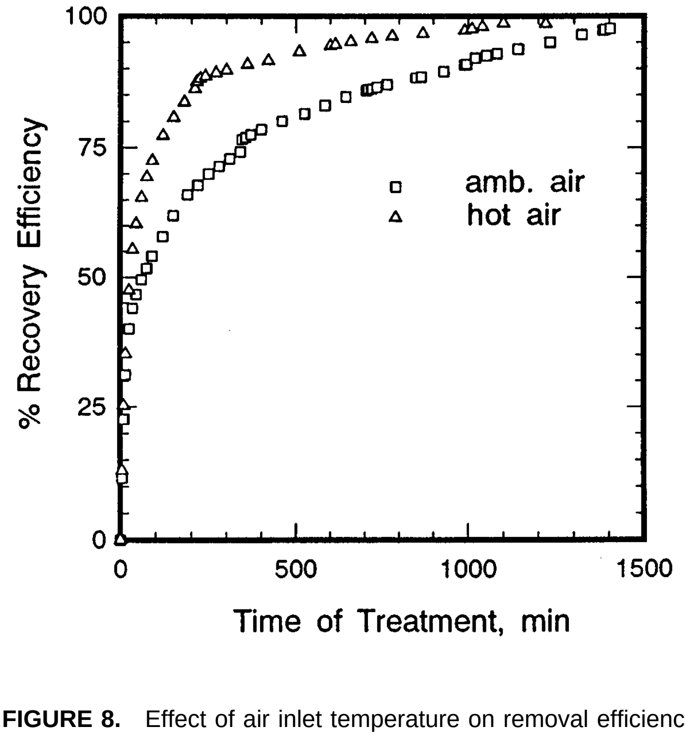
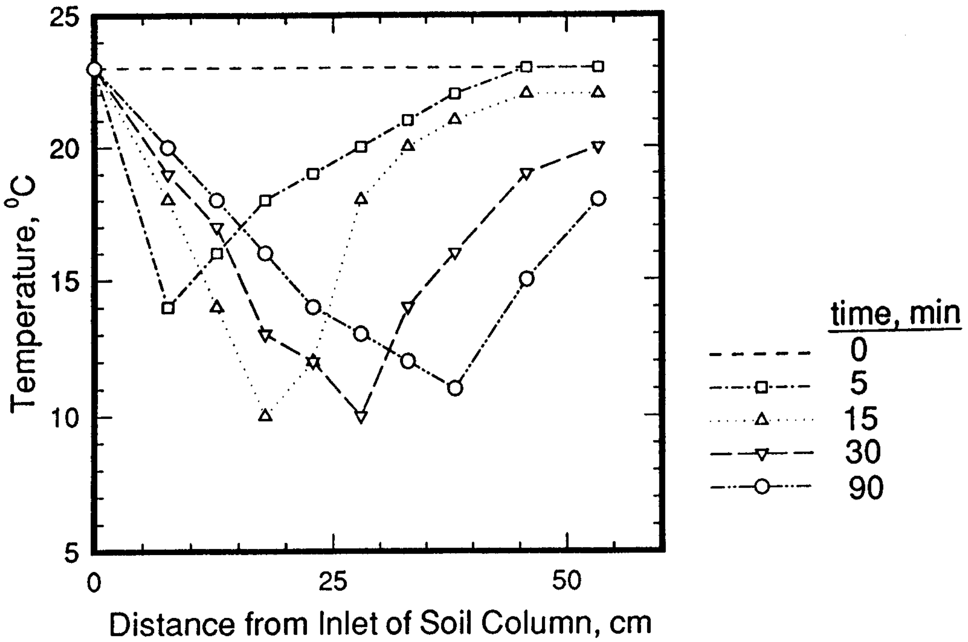


FIGURE 8. Effect of air inlet temperature on removal efficiency.

D. **Temperature Profiles**

The temperature profiles in the soil column were obtained for three experiments. In case 1, unheated air was injected at the rate of 10 l/min. In case 2, unheated air was injected at the rate of 5 l/min, and in case 3, the air was preheated to 45°C at the inlet and injected at a rate of 10 l/min. These experiments were conducted with benzene-contaminated soil samples having a mean grain size of 0.61 mm.

The column centerline temperature distribution is displayed in Figure 9 for all three cases. As air passes through the test section, evaporation of the liquid contaminant and water occurs at the air-liquid interface. Due to the high surface area per unit volume of packed soil, the air becomes saturated with contaminant vapor within a narrow region. The saturation vapor concentration in the air depends on the local soil temperature and the composition of the contaminant. The latent heat of vaporization of the liquid is partly provided by the incoming air, while the rest of it is absorbed from the soil. This leads to a decrease in soil temperature below ambient until an equilibrium condition is established between the heat transfer from the incoming air to the soil and the latent heat absorption or until all



A

FIGURE 9. Temperature distribution along the column centerline at various times. (A) Unheated air at 10 l/min; (B) unheated air at 5 l/min; (C) preheated air at 45°C and 10 l/min.

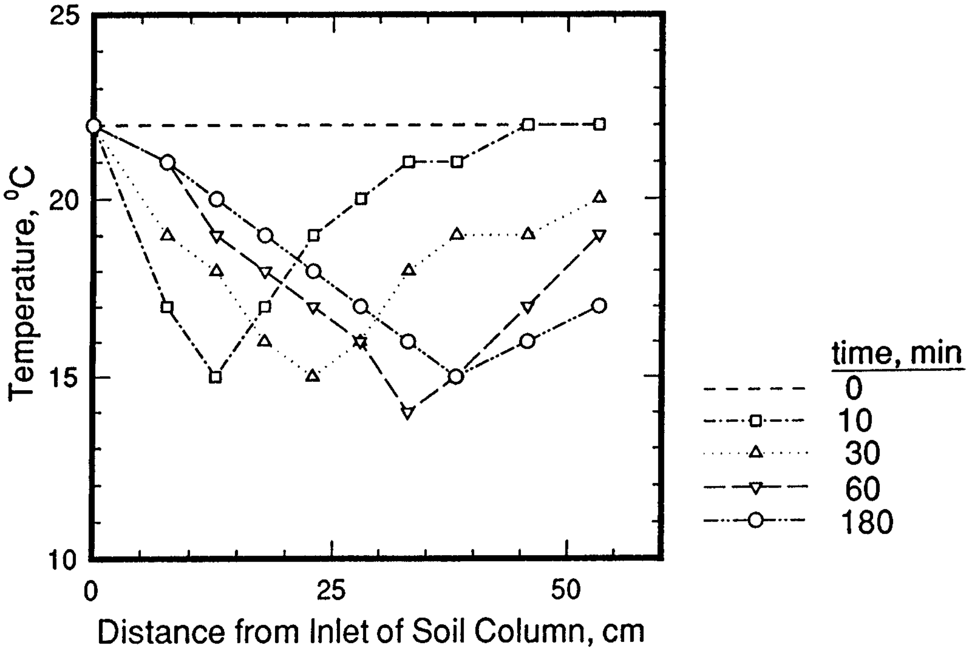


FIGURE 9B

of the contaminant (liquid) at that location is completely vaporized. After complete evaporation of the contaminant at a given location, the local soil temperature increases due to heat transfer from the incoming air to the lower-temperature soil. This trend is shown clearly in Figures 9A and B (cases 1 and 2). The location of the minimum temperature represents that of the evaporation front, which moves from the inlet to the exit with the passage of time. As the evaporation front moves toward the exit, vapor concentration decreases in the exiting air.

In case 1 (Figure 9A), the difference between the inlet temperature and minimum temperature in the column was 14°C. In case 2 (Figure 9B), the difference was only 8°C because the rates of evaporation of liquid benzene and water were less than that of case 1, as explained earlier, and the cooling effect was reduced. In case 3 (Figure 9C), as the air was preheated to 45°C at the inlet, the temperature at the evaporation front was higher than in both cases with unheated air. In this case, a minimum temperature of 18°C was obtained compared with 10°C in case 1 with unheated air at the same flow rate.

The rate of propagation of the thermal front depends primarily upon the ratio of the energy contents of the air and the soil, and on the air-flow rate. By increasing the heat flux at the inlet, the temperature of the incoming air increases, but the rate of thermal front movement does not change significantly. On the other hand, the

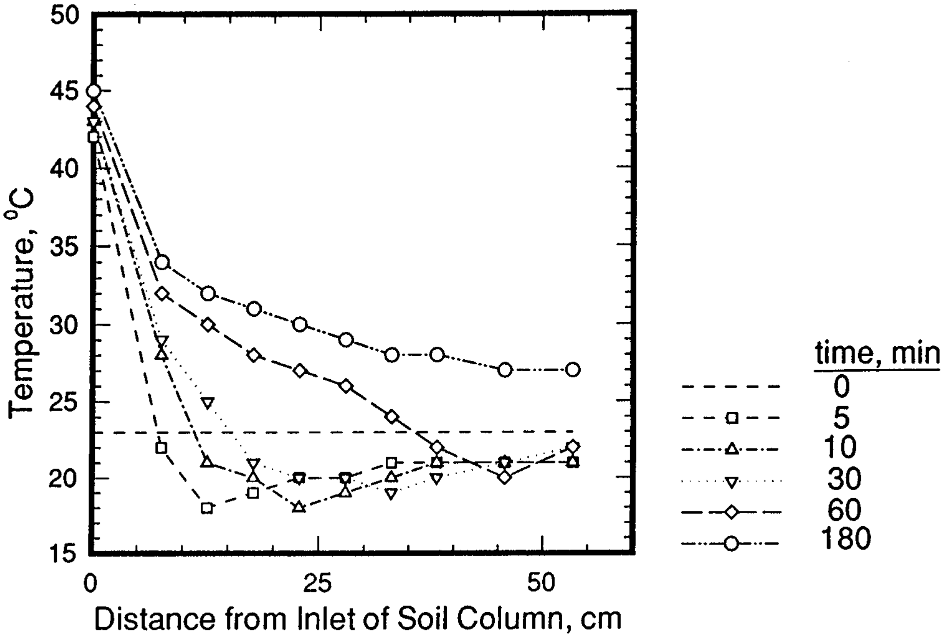


FIGURE 9C

movement of the evaporation front depends upon the air-flow rate, the benzene residual saturation, and the volatility of benzene. Hence, in the case of a highly volatile contaminant such as benzene, heating the incoming air enhances the evaporation process over a very short region, which results in a sharp decrease in temperature (Figure 9C). For contaminants with lower volatility, the rate of evaporation is slower and the temperature change occurs over a wider region of the soil column, as reported by Katsumata and Dhir (1992). Ideally, as time increases, the temperature should reach a constant value throughout the column, equal to the temperature of the incoming air, but due to heat losses to the surroundings, a temperature gradient was sustained within the column (Katsumata and Dhir, 1992).

VI. SUMMARY AND CONCLUSIONS

An experimental study was performed to evaluate the effectiveness of air stripping for remediation of VOC-contaminated soils. Experiments were conducted in a one-dimensional soil column contaminated with benzene, and parametric studies were performed to evaluate the effect of soil grain size, air injection flow rate, and air inlet temperature on benzene recovery efficiency. Important results are summarized as follows:

1. The benzene concentration in the effluent air decreased with time of treatment, mostly during the early stages of air injection, indicating that a relatively large amount of contaminant was removed at the beginning.
2. Due to higher removal rates at the beginning of air injection, the benzene recovery efficiency was higher in coarser soils than in finer soils.
3. A higher air injection flow rate yielded maximum benzene recovery efficiency in significantly less time than the lower flow rate.
4. Pulsed venting operation resulted in the same recovery efficiency as continuous venting at the same flow rate but required less time, suggesting that it may be more cost effective than continuous treatment.
5. The benzene removal rate increased significantly when preheated air was injected into the soil column, and less time was required to reach maximum recovery efficiency than with unheated air.
6. Lowering of the temperature in the soil column was observed, and the minimum temperature was measured at the evaporation front, which moved progressively toward the column exit.

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