

Air flow optimization and surfactant enhancement to remediate toluene-contaminated saturated soils using air sparging

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Abstract

This paper presents the results of laboratory experiments that investigate the removal of volatile organic compounds from saturated soils through the use of air sparging. Three series of experiments were performed in a column test apparatus using two different soils to represent actual field conditions, namely, a fine gravel and a medium to fine Ottawa sand (both obtained from sources near Chicago, Illinois, USA) contaminated with toluene, a major constituent of petroleum products. The results showed that toluene was removed from gravel very efficiently using air sparging; complete removal was achieved using a variety of air flow rates. However the toluene removal rates in tests using sand were significantly less. Even at the highest air flow rate used during testing, complete toluene removal took eight times longer than in comparable tests using gravel. With low air flow rates this was not achieved even after 17 hours of testing. It was further found that the injection of foams generated with surfactants, SDS and witconol SN70, at low air flow rates during the use of air sparging was found to accelerate the bulk removal of toluene in sand, but the use of surfactants did not facilitate the removal of residual levels of contamination.

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Introduction

Subsurface contamination caused by the infiltration of hydrocarbons, including petroleum products, poses serious environmental problems. Hydrocarbon releases often occur initially within the vadose zone and then migrate downward through the soil, eventually reaching the water table. Light non-aqueous phase liquid (LNAPL) constituents of petroleum products tend to float on top of the water table and as a result may be transported vertically and horizontally due to water table fluctuations. Such fluctuations may be the result of seasonal variations, well pumping, or groundwater flow conditions. These LNAPLs will persist for decades within the subsurface unless remedial action is taken.

A number of remedial technologies, including pump-and-treat, bioremediation and soil washing/flushing have been used to treat contaminated groundwater. However, in many cases, these methods have proven to be ineffective, expensive, unreliable, and often require extended time periods for adequate clean-up (Ardito and Billings, 1990; Leonard and Brown, 1992; NRC, 1994; Reddy *et al.*, 1995). The soil vapor extraction (SVE) method has proven to be an effective and less expensive technique for the remediation of volatile organic compounds within the vadose zone; however, it cannot be used to treat saturated soil zone contamination. Therefore, a different remediation technique must be used for saturated soils and groundwater.

In-situ air sparging is becoming an increasingly popular option within the USA for the remediation of saturated soils and groundwater contaminated with volatile organic compounds (VOCs). Previous field investigations have shown air sparging to be an efficient and effective method when applied to such contaminant conditions (Johnson *et al.*, 1993; Reddy *et al.*, 1995). Air sparging is most effective when applied to contaminants with Henry's Law constants greater than 10^{-5} atm m^3/mol and vapor pressures greater than

5mm Hg (Leonard and Brown, 1992). It is a simple method to apply; field implementation is performed using readily available equipment. Since no special equipment is required, air sparging has proven to be a cost-effective remedial method. *In situ* air sparging is performed by injecting air below the groundwater contamination plume, as shown in Figure 1. The injected air then travels towards the ground surface, creating air passageways. The contaminants are partitioned into the vapor phase through the action of different mechanisms, including volatilization, dissolution, and desorption, which result from the action of the injected air. The contaminant-laden air continues to travel upward through the air passageways, eventually entering the vadose zone. The vapors are then collected using a soil vapor extraction system, as shown in Figure 1. Additionally, the injected air increases subsurface oxygen levels, aiding in the action of aerobic biodegradation. A detailed review of the mechanics of the air sparging process is provided by Reddy *et al.* (1995).

Current air sparging design methods are heavily based on empirical approaches and limited field experience rather than a rational design basis because very little is understood regarding the mass transfer/transport/transformation mechanisms that occur during the use of air sparging. Additionally, little is known about the effects of control variables, including injection air flow rate and pressure. To provide a better understanding of these mechanisms and variables, a comprehensive air sparging research program has been underway at the University of Illinois at Chicago (UIC) since 1993. The research includes the performance of extensive one-dimensional and two-dimensional laboratory experiments. Knowledge gained from the controlled laboratory experiments will be used to develop a conceptual model of the air sparging process as well as a comprehensive mathematical model that can serve as a predictive tool for the optimal design of air

sparging field systems based on site-specific data (Reddy and Zhou, 1996). Additionally, data from air sparging field systems currently in operation will be used to validate and improve the model.

During the use of air sparging, contamination either in contact with or in the vicinity of an air passageway is volatilized and removed while contamination distant from the passing air moves toward the air channels through diffusion. Diffusion is often a rate-limiting process; remediation of contaminant trapped in soil pores or in the interstices of clay occurs very slowly. While the majority of the contaminant mass is removed relatively quickly through volatilization, asymptotic levels are reached when residual contaminant removal becomes diffusion-dependent. The removal of residual contamination often takes significantly longer than the removal of the initial bulk contamination. Increased dissolution of the residual organic compound would accelerate the remediation process considerably by mobilizing the contaminant and allowing it to interact with the flowing air more quickly. The use of surfactant foams as an air sparging enhancement technique

may significantly increase the mobility of residual contaminants, allowing for more efficient removal.

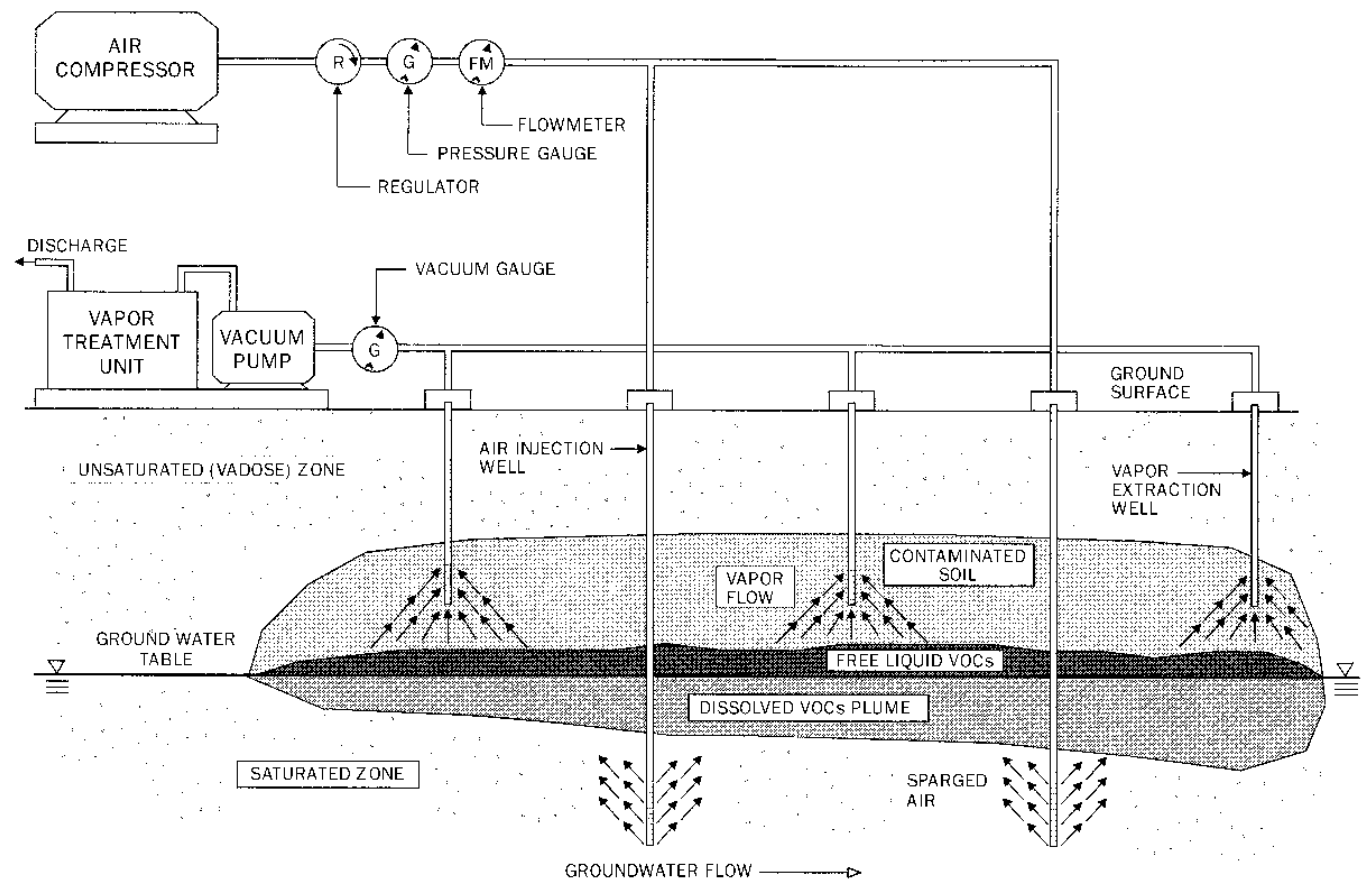
This paper presents the results of laboratory experiments which investigated the effectiveness of air sparging in removing toluene from different types of saturated soils. One-dimensional column experiments were conducted using two different types of soil, a fine gravel and a medium to fine sand, under different injected air flow conditions. The effect of surfactant foam injection coupled with air sparging on contaminant removal efficiency has also been evaluated in this paper.

Experimental program

Materials

Two different soil types were used in this research: a fine gravel, and a medium to fine Ottawa sand. Laboratory tests were first performed on these soils to determine their index and engineering properties. These tests included grain size analysis, specific gravity, minimum and maximum densities, and hydraulic conductivity. The test results for

Figure 1
Schematic of *In Situ* Air Sparging System



these two soils are summarized in Table I. The grain size distribution for the two soils is shown in Figure 2. Toluene, a major constituent of gasoline and other petroleum products, was selected as the representative VOC for this investigation and is routinely used as an indicator of organic contamination in groundwater at field sites. The properties of toluene are shown in Table II.

Two different surfactants were used to generate foams in this study, sodium dodecyl sulfate (SDS) and witconol SN70. Each of these

Table I
 Characteristics of soils used in experiments

Property	Sand	Gravel
Unified soil classification, ASTM D 2487	SP	GP
Effective particle size, D_{10} , ASTM D 422	0.18	2.35
Coefficient of uniformity, C_c^* , ASTM D 422	2.1	2.0
Coefficient of curvature, C_u^* , ASTM D 422	1.0	1.1
Specific gravity, G_s , ASTM D 854	2.67	-
Minimum dry density (g/cm^3), ASTM D 4254	1.56	1.62
Maximum dry density (g/cm^3), ASTM D 4253	1.85	1.90
Hydraulic conductivity (cm/s), ASTM D 2434	1.97×10^{-2}	1.60

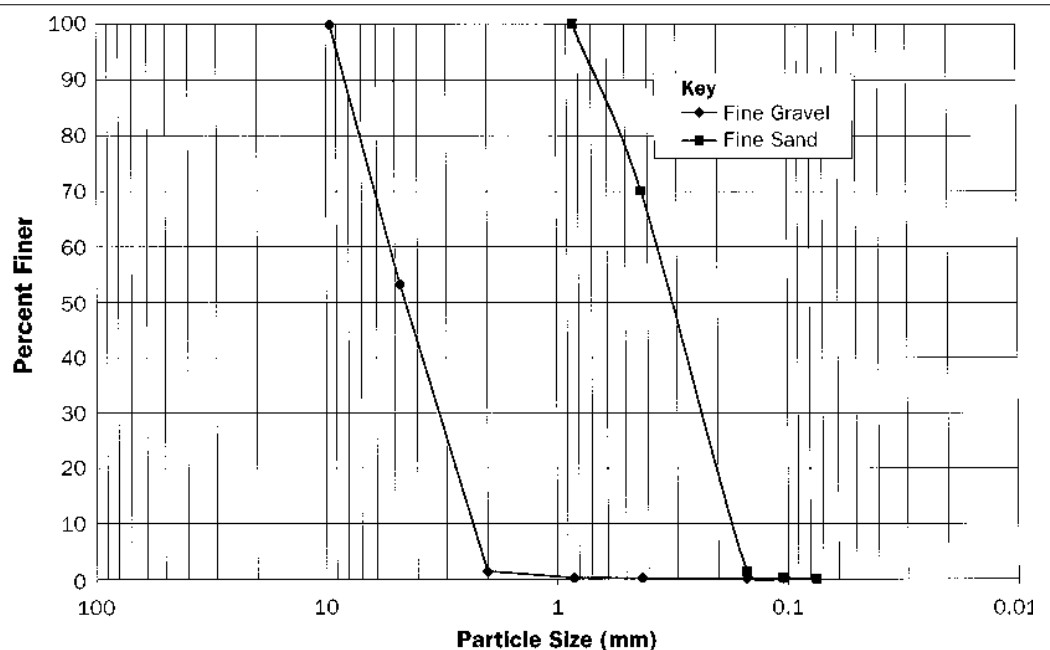
Note:
 $C_c^* = D_{60}/D_{10}$ and $C_u^* = D_{30}^2/(D_{10}D_{60})$

Table II
 Physical and chemical parameters for toluene (Clarke *et al.*, 1993)

Molecular Formula	C_7H_8
Aromaticity	Yes
Molecular weight	92.13
Color/form	Colorless liquid
Boiling point, $^{\circ}C$	111
Melting point, $^{\circ}C$	-95
Density, g/cm^3	0.8661
Aqueous solubility, mg/L	515
Vapor pressure, mm Hg	40 (at $31.8^{\circ}C$)
Vapor density (air=1)	3.14
Log K_{ow} (Octonol water coefficient)	1.83 - 2.94
Log K_{oc} (Soil/water distribution coefficient referenced to soil organic content)	1.12 - 3.28
K_H , $Pa \cdot m^3/mol$ (Henry's Law Coefficient)	680

surfactants is relatively inexpensive and is commercially available. These surfactants were selected because they possess low surface tension, low specific gravity, good detergency and solubilization, and low soil dispersion. Additionally, both surfactants are biodegradable. The properties of these surfactants are summarized in Table III. While SDS is anionic, witconol SN70 is non-ionic; therefore, their use in the study allows for the investigation of the suitability of two different groups of surfactants. Cationic surfactants were not used in this study because such surfactants have been shown to retard contaminant removal (Abdul, 1990; Clarke *et al.*, 1993).

Figure 2
 Grain size distribution of soils used in this study



Experimental set-up

A one-dimensional air sparging column apparatus was utilized during the investigation. A schematic of the test apparatus is shown in Figure 3. The plexiglas column, measuring 93 cm in height with an inside diameter of 8.7 cm, was used to contain the test soil. Seven sampling ports were located at distances of 9cm, 19cm, 29cm, 39cm, 49cm, 45cm, and 59cm, respectively, from the base of the column. These locations are designated as ports 1 through 7, respectively. The ports consisted of a septa and brass sleeves held in place with Teflon fittings. The top and bottom of the column were sealed with plexiglas covers. Each cover featured a groove holding a rubber O-ring in place, providing an air-tight seal between the cover and the column.

Six machine screws were used to connect the covers to the column.

The bottom cover is provided with two one-way inlet quick connectors. The first inlet was used for air injection into the soil column. The inlet led to a star-shaped grid of holes that assured a uniform distribution of injected air. The second inlet served two purposes. First, it was used to inject contaminant solution into the soil column. After this was accomplished, the contaminant source was disconnected, and a feed line leading to a clean water reservoir was connected in its place. The reservoir was used to maintain a constant head within the column during testing.

Both the clean water reservoir and a contaminant reservoir were placed so as to maintain the appropriate hydraulic head in the column. The clean water reservoir contained a level regulator and was connected to a tap. This system allowed a desired, constant water level to be maintained in the reservoir throughout the testing. The water reservoir was connected to the column bottom cover using Teflon tubing. A check valve was provided to assure unidirectional flow from the reservoir to the column. Another valve for flow shut-off was provided. The contaminant reservoir, which had a two-liter capacity, was also connected using Teflon tubing to the column bottom cover.

The controlled injection of clean air into the column was accomplished through the use of an air source (compressed air), a regulator, a pressure gauge and a flow meter as shown in

Table III

Properties of surfactants used

Surfactant	SDS	Witconol SN70
Chemical Name	sodium lauryl sulfate	ethoxylated alcohol
Ave. molecular weight	288	392
Form	powder	liquid
Specific gravity	0.40	0.98
Solubility in water	10 % in water	moderately at 25°C
Vapor pressure (mm Hg at 25°C)	negligible	<1
pH of 1 per cent solution	8-10	6-7
Critical micelle concentration	0.231 wt% (2.31 g/l)	0.500wt% (5 g/l)
Biodegradable	yes	yes
Net charge	anionic	nonionic

Figure 3

Schematic of one-dimensional experimental air sparging set-up

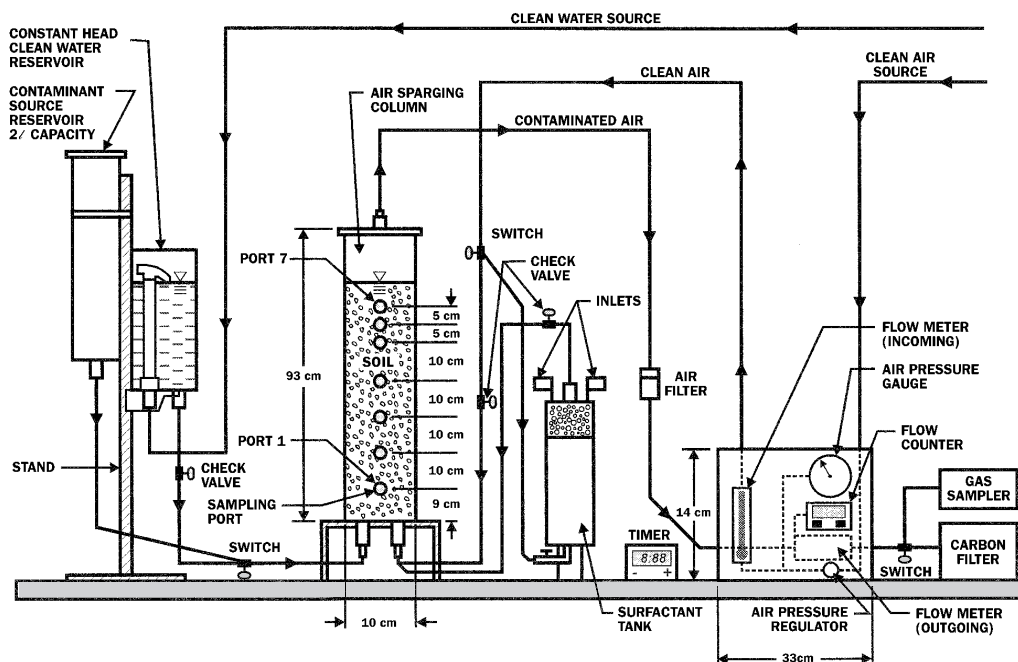


Figure 3. The injected air pressure and flow rate were set to the desired levels and were kept constant throughout the testing. The top cover of the column was provided with an outlet for the contaminated air to exit during the testing. The outlet was connected to a filter, a flow meter, an air sampler and a carbon filter using Teflon tubing as shown in Figure 3. Both the rate of outflow and cumulative outflow were recorded using an electronic flow meter with a flow counter. An air sampler allowed for the intermittent sampling of the outflow air to determine contaminant concentration levels. A carbon filter was used to remove contaminant vapors from the effluent air before emission into the atmosphere.

A timer was used to accurately record the sampling times and test duration. For testing with surfactant foams, a reservoir containing surfactant solution was included in the air flow injection system as shown in Figure 3. When surfactants were used, the injected air entered through the bottom of the surfactant reservoir. The surfactant foams were then forced through tubing at the top of the reservoir along with the air. A gas chromatograph (GC) equipped with a photo-ionization detector (PID) and a Por-a-pack column was used to measure the toluene concentrations both in the air and liquid samples that were collected during the testing.

Experimental procedure

When performing an air sparging test, a Whatman #42 filter paper was first placed over the bottom cover to prevent clogging of the injection grid. The cover was attached to the column. Next, the test soil was placed into the column through the top opening by dropping it from a desired height. Both the mass and volume of the placed soil were recorded. After soil placement, the top cover was fastened into place, sealing the column. All other necessary equipment was connected to the column, as depicted in Figure 3.

The contaminant solution was prepared by mixing predetermined amounts of toluene and deionized water in a sealed glass container. The mixture was thoroughly stirred using a magnetic stirrer for over 12 hours. The contaminated solution was transferred into the contaminant reservoir and allowed to infiltrate the soil column through an inlet on the bottom cover. Once the test soil was saturated to the desired level (approximately 2.5 cm above port 7), contaminant infiltration ended. The volume of contaminant solution injected into the soil was recorded; the amount was approximately equal to the estimated pore volume of the soil column. The initial contaminant concentration profile within the soil was measured by extracting

two microliters of pore water from each sampling port and analyzing using gas chromatography (GC). The syringes used for pore water extractions were decontaminated through repeated washings with deionized water and acetone following each use.

Following contaminant placement, the contaminant reservoir was disconnected from the column, and the clean water reservoir was connected in its place. By opening the valve along its pathway, water was introduced into the column, helping to stabilize the water level. Once the water level stabilized, air injection began. The injected air flow rate and injection pressure were controlled using a flow meter and regulator, respectively, which were built into the column control panel. If desired, the surfactant reservoir was attached in-line with the injected air feed line, allowing for surfactant introduction. The air entering into the column was observed, and the electronic timer was switched on to begin the test.

Contaminant concentrations within the soil profile were monitored by extracting pore water from the sampling ports and analyzing in the same manner as before. The contaminant concentrations within the extracted pore water were then recorded. Additionally, an air sampler was used to collect effluent gas from the column intermittently. The collected gas was also analyzed using gas chromatography. The flow rates of effluent gas as well as cumulative flow were recorded at different time intervals. Testing and data collection continued until insignificant contaminant concentrations remained within the soil or time constraints prevented continued testing.

Testing program

Three series of experiments were conducted during this study. The first series consisted of three tests performed using gravel as the test soil; the second series consisted of two tests utilizing sand as the test soil; and the third series consisted of two tests performed using sand subjected to the introduction of two different surfactants: sodium dodecyl sulfate (SDS) and witconol SN70. A summary of the test conditions is provided in Table IV.

Results and discussion

Experiments on gravel

The results of the three experiments conducted with gravel contaminated with an initial toluene concentration of approximately 250mg/l and subjected to injection air flow rates of 380, 960 and 2,225 ml/min are shown in Figures 4(a) through 4(c), respectively. The air entry pressure for all three experiments was 6.9kPa. The measured

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Environmental Management and Health
 10/1 [1999] 52-63

Table IV

Summary of one-dimensional air sparging test conditions

Test	Soil	Initial density (g/cm ³)	Initial concentration (ppm)	Injected air flow rate (ml/min)	Injected air entry pressure (kPa)	Surfactant
1	Gravel	1.62	250	380	6.9	NA
2	Gravel	1.62	250	960	6.9	NA
3	Gravel	1.62	250	2,225	6.9	NA
4	Sand	1.62	250	2,225	6.9	NA
5	Sand	1.58	50	960	6.9	NA
6	Sand	1.58	50	380	9.7-12.4	SDS
7	Sand	1.58	50	380	8.3-9.7	Witconol SN70

Notes:

NA = Not applicable

toluene concentrations in each port were normalized with the initial toluene concentration detected at that port and plotted. Through visual observation, it was determined that air traveled predominantly in the form of bubbles distributed evenly throughout the soil cross-section. With increasing air flow rate, the activity of these bubbles significantly increased.

The normalized concentration profiles in Figures 4(a) through 4(c) show that complete toluene removal was achieved in all three tests. Complete removal was accomplished in 390 minutes using an air flow rate of 380 ml/min; in 140 minutes at an air flow rate of 960ml/min; and in 80 minutes at an air flow

rate of 2,225 ml/min. These results show that increasing air flow rates led to faster contaminant removal. The concentration profiles shown in Figures 4(a) through 4(c) were used to calculate the percent toluene remaining in the soil at different times; these values were plotted as shown in Figure 5. The results in Figure 5 clearly demonstrate the effect of injected air flow rate on the contaminant removal. Rapid initial contaminant removal was followed by slower removal rates during the use of the lowest flow rate. At higher air flow rates, rapid initial contaminant removal also occurred, but the remaining contaminant was removed slightly faster than during lowest air flow rate usage. However, the

Figure 4a

Air sparging test on gravel with air flow rate 380ml/min

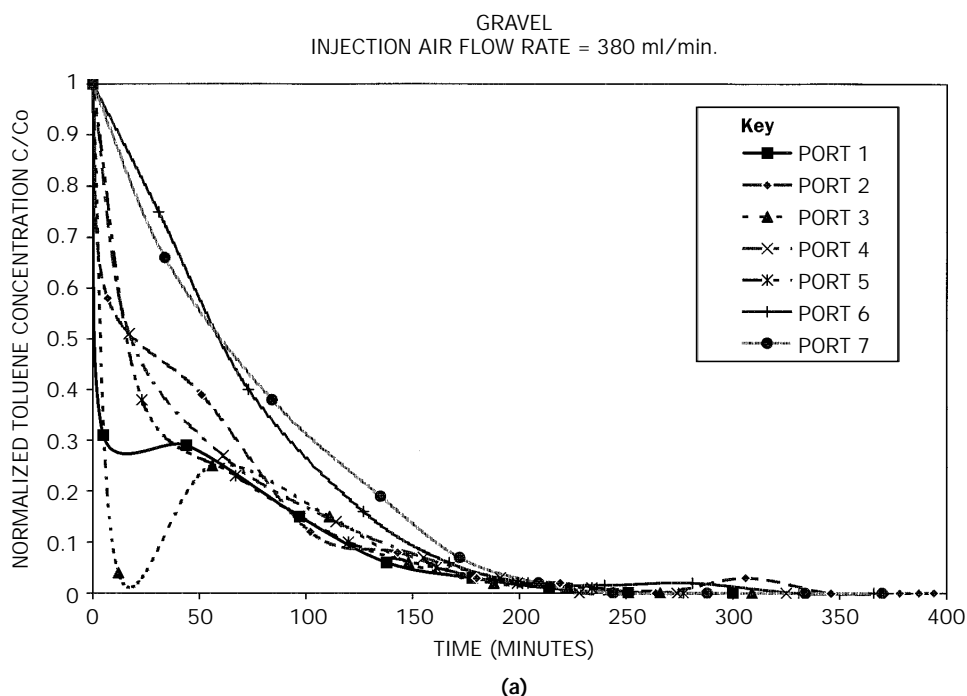


Figure 4b
 Air sparging test on gravel with air flow rate = 960ml/min

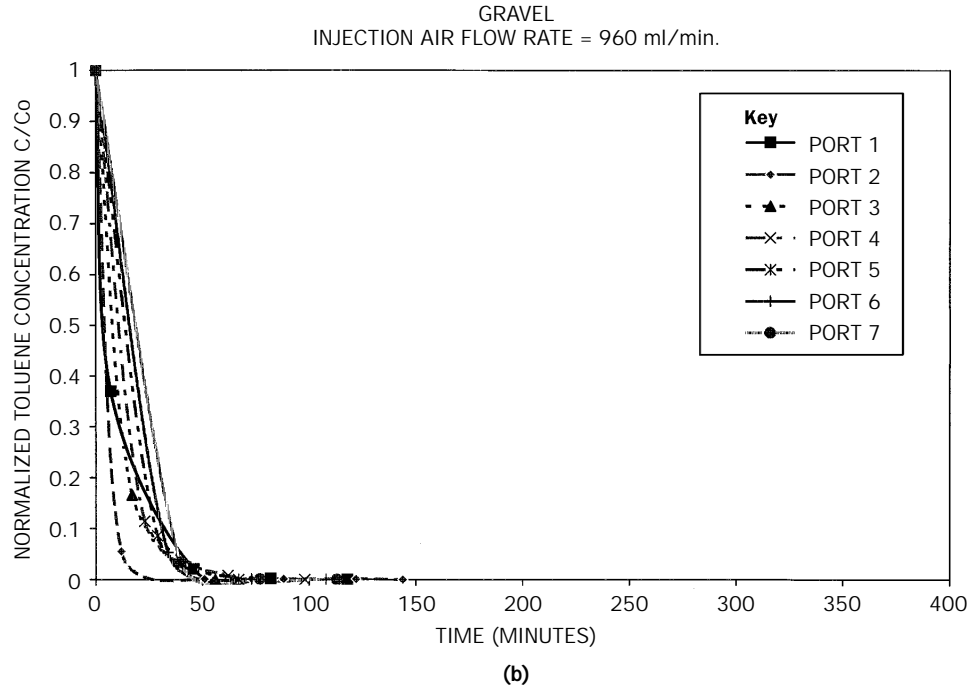
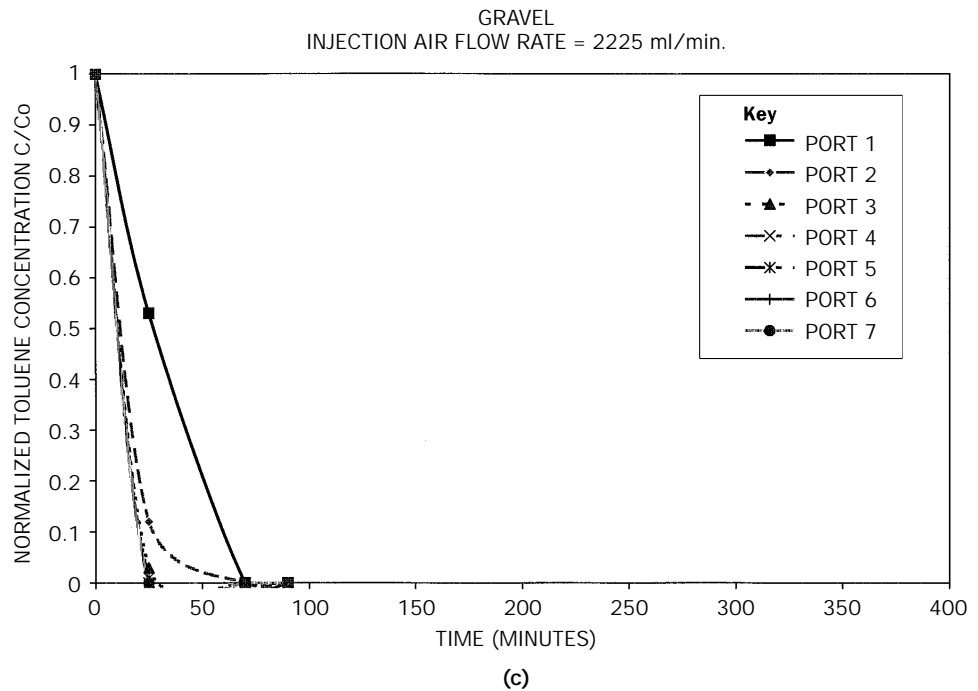


Figure 4c
 Air sparging test on gravel with air flow rate 2,225ml/min

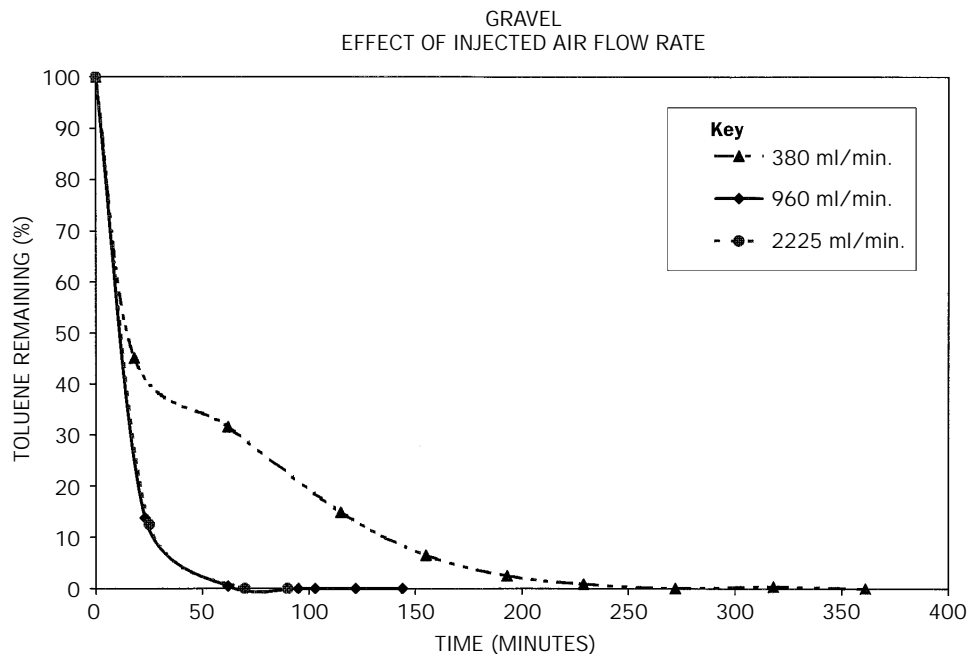


difference in the rate of removal between the flow rates of 960ml/min and 2,225ml/min was not significant, indicating that an optimal air flow rate had been reached, beyond which an increase in flow rate does not significantly increase the rate of contaminant removal.

Experiments on sand

Two experiments were conducted using sand to evaluate the effect of different initial contaminant concentrations and varying injected air flow rates. The first experiment was performed with an initial toluene concentration

Figure 5
 Effect of injected air flow rate on contaminant removal



of 250mg/l and injected air flow rate of 2,225 ml/min. The results of this test, in the form of normalized concentration versus time, are shown in Figure 6. From these results, it can be seen that complete toluene removal was accomplished within 660 minutes. The visual observations made during testing revealed that air flow within the sand occurred within a channel network. Because of the high air flow

rate, a high density of channels were formed, allowing the flowing air to interact with a greater volume of contaminated soil.

The second air sparging test with sand was performed using a lower initial toluene concentration (50mg/l) and a lower flow rate (960 ml/min). These test results, shown in Figure 7, indicate that the toluene was not fully removed even after 1,000 minutes. The visual

Figure 6
 Air sparging test on sand with air flow rate = 2,225ml/min

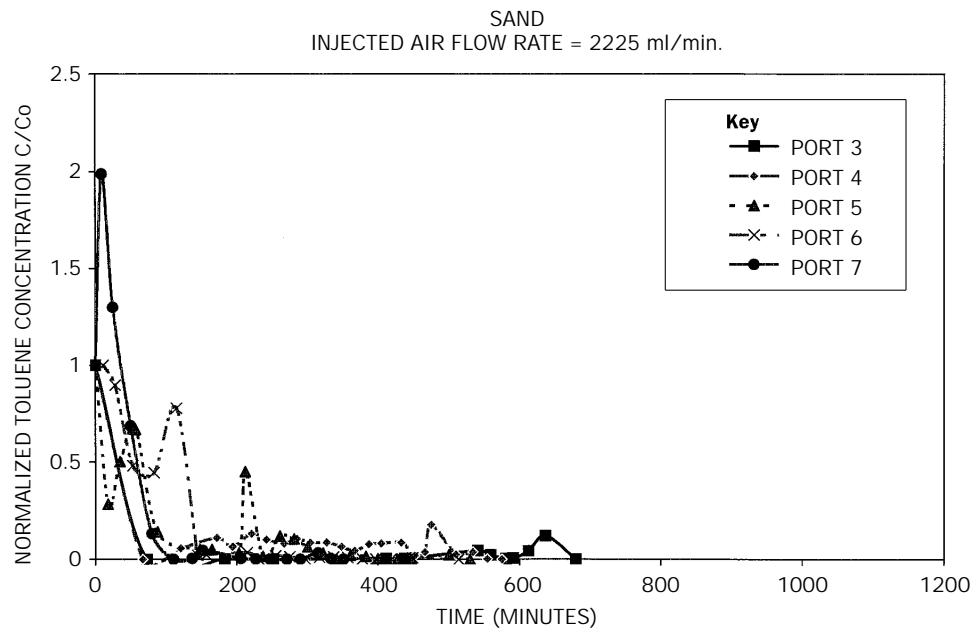
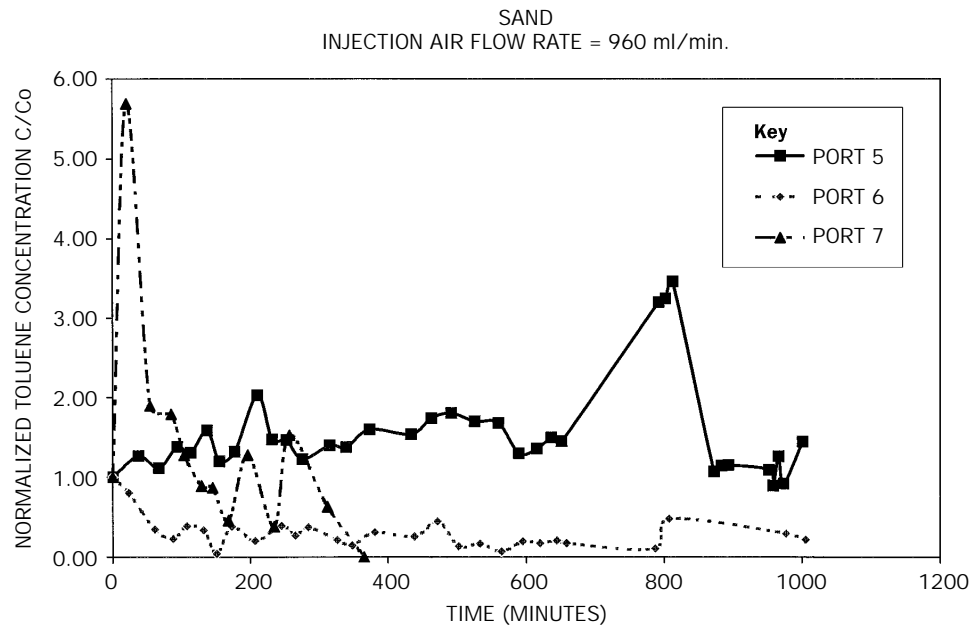


Figure 7

Air sparging test on sand with air flow rate = 960ml/min



observations made during testing revealed that because of the low flow rate, the injected air traveled through a few discrete channels within the soil. Because less air channels were created, a greater portion of contaminated soil was left untreated. Contaminant in the immediate vicinity of the air channels was removed effectively, but contaminant located away from the air channels did not interact with the air and, consequently, remained in the soil. These results demonstrate that it may be difficult to achieve adequate removal efficiencies with low air flow rates in sandy soils.

The effect of soil type on the contaminant removal efficiency is clearly evident from the comparison of the results for gravel and sand shown in Figures 4(c) and 6, respectively. Both tests used the same initial maximum toluene concentration of 250mg/l and the same injected air flow rate of 2,225ml/min. Using the concentration profiles, the contaminant remaining in these soils at different times was calculated and compared in Figure 8. In both tests, complete removal of toluene was achieved; however, the time required for complete toluene removal varied significantly between tests. While toluene removal was accomplished within 80 minutes in the gravel, a removal time of 660 minutes was necessary within the sand. Therefore, it took approximately eight times longer to completely remove the toluene from the sand than required for the test with gravel. This difference in removal efficiency is attributed to the air flow patterns observed within the two

soils. The air flow in gravel occurred in the form of evenly distributed bubbles, but within the sand, the air flow occurred through a channel network. The contaminant in the gravel was completely in contact with the air, leading to quick removal. Within the sand, however, the contaminant located in regions not traversed by channels had to diffuse toward the air channels in order to be removed, significantly increasing the removal time.

Experiments on sand using surfactant foams

As discussed in the previous section, high contaminant removal efficiencies in sands are difficult to achieve using low air flow rates. One option that can enhance the contaminant removal in sand is the use of high air flow rates. However, the injection of air at high flow rates requires the use of higher capacity equipment, resulting in additional energy expenses. Instead of employing high air flow rates, the feasibility of using surfactant foams at very low air flow rates to enhance contaminant removal was investigated in this study.

Two tests were conducted by injecting surfactant foams along with air into sand contaminated with toluene at an initial concentration of approximately 50mg/l. Foams generated using two different types of surfactants, SDS and witconol SN70, were tested. Both surfactant solutions were used at concentrations of 1 per cent solution by weight, which exceeded the critical micelle concentration of each surfactant as shown in

Figure 8

Comparison of contaminant removal in gravel and sand

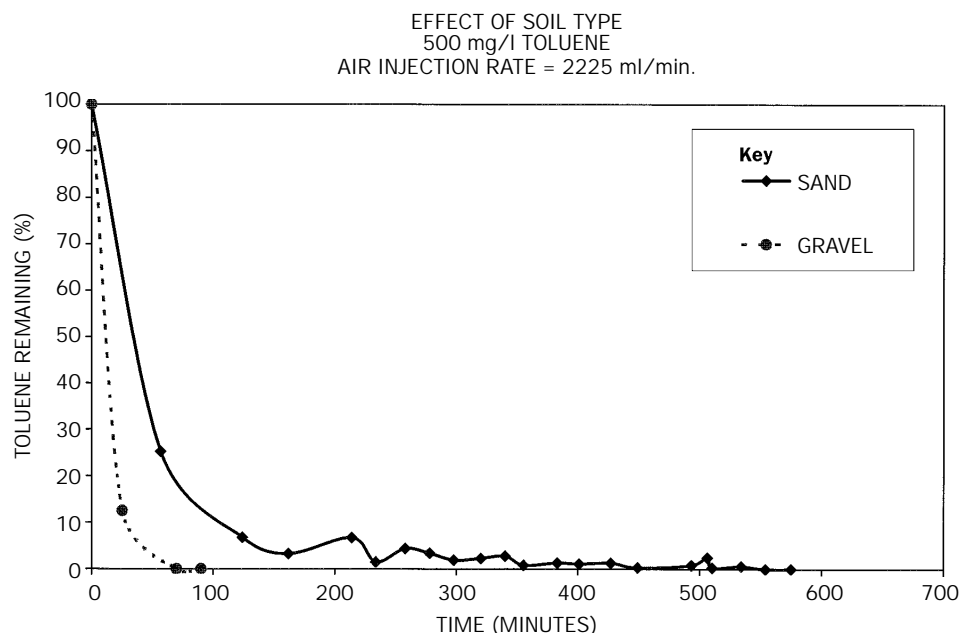


Table III. In each test, air was injected at a flow rate of 380ml/min. The measured air entry pressure for SDS varied from 9.7kPa to 12.4kPa, while the air entry pressure for witconol SN70 varied from 8.3kPa to 9.7kPa. The air entry pressure represents the minimum pressure needed to overcome the resistance to air flow caused by the surfactant reservoir solution and the hydrostatic pressure within the soil column. The test results for SDS and witconol SN70 are shown in Figures 9 and 10, respectively.

From Figures 9 and 10, it can be seen that significant toluene removal was achieved in approximately 500 minutes with the use of surfactant foams. However, residual contamination was present even after 1,000 minutes for both surfactants. The performance of SDS foams was better than witconol foams since the final contaminant concentrations were lower and the residual contamination was less in the test performed using SDS. Both surfactants had a positive effect when compared to the results of the tests performed on sand subjected to an air flow rate of 960 ml/min and free of surfactant (Figure 7). The higher removal with the surfactant foams is attributed to the lowering of the interfacial tension between the water and the toluene, resulting in increased dissolution of the toluene into the water.

During testing, the surfactant foams were observed at the bottom of the column and, to a much lesser degree, in the effluent at the top of the column. Air channels that developed along the walls of the column allowed

visualization of surfactant foams traveling through the soil. The surfactant foams may have followed the air passageways created by the air flow channels, restricting the migration of the surfactant into the interior of the sand. Thus, residual contamination persisted within the soil although the bulk of the contaminant was removed faster with surfactant treatment. If the surfactant foams could be injected in such a way as to penetrate the soil more evenly, the mobility of the contaminant could be significantly enhanced.

Conclusions

The following conclusions can be drawn based on the results of this study:

- 1 During application to gravel, air sparging completely and efficiently removed toluene contamination. Increased air flow rate led to faster contaminant removal; however, at high flow rates, the increase in the rate of removal was not significant.
- 2 For sand, toluene removal was more efficient using high air flow rates. At low air flow rates, complete toluene removal was not achieved. The bulk of toluene contamination was removed relatively quickly, but when low flow rates were used, residual levels of contamination lingered within the soil.
- 3 In addition to dependence on contaminant properties, including the Henry's Law constant and vapor pressure, contaminant

Figure 9
 Air sparging test on sand with SDS foams and air flow rate = 380ml/min

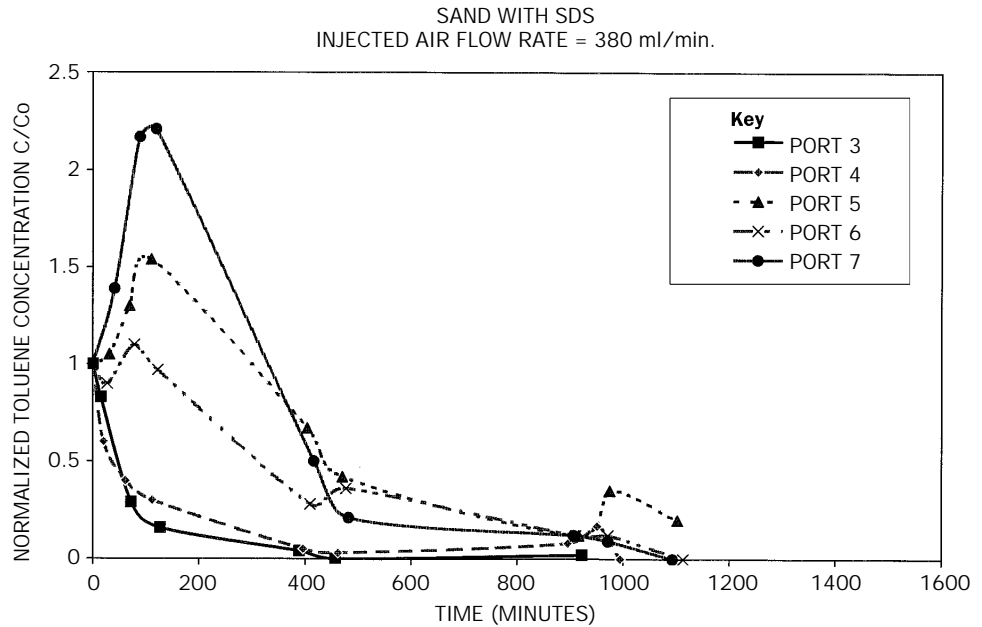
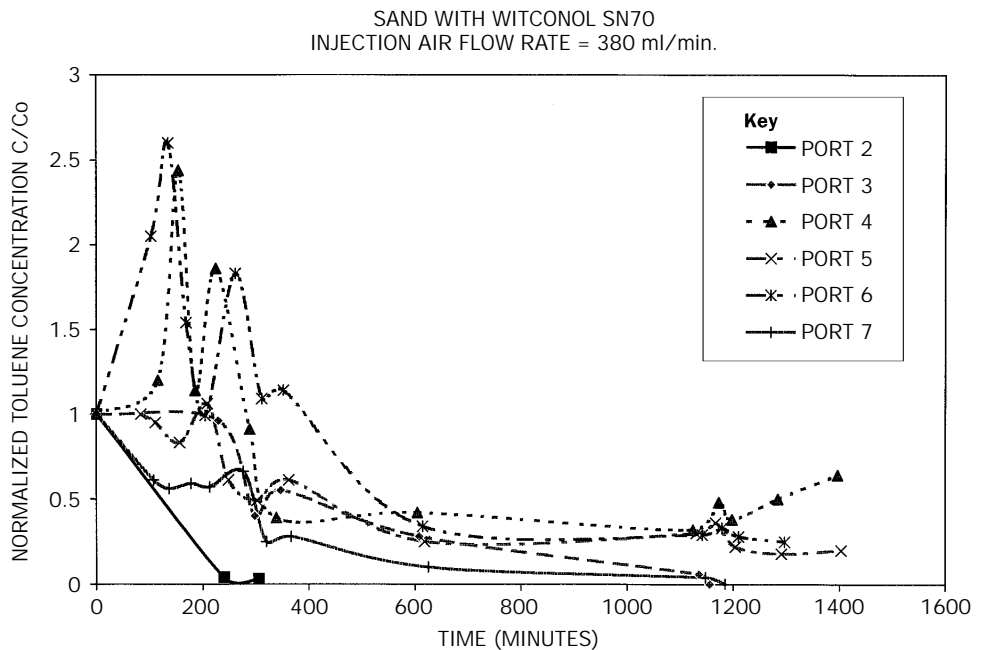


Figure 10
 Air sparging test on sand with witconol SN70 foams and air flow rate = 380ml/min



removal depends significantly on the soil type. The differences observed in contaminant removal rates between gravel and sand were attributed to the mode of injected air travel. In general, the injected air traveled through the gravel in the form of bubbles distributed evenly throughout the gravel. Because of this distribution, more thorough interaction between the contaminant and the injected air occurred,

leading to complete and efficient removal. In sand, the injected air traveled through a channel network. The number and distribution of these channels was found to be a function of the injected air flow rate. At high air flow rates, a large number of channels were formed which caused greater interaction between the air and the contaminant and, consequently, higher contaminant removal. At low air flow rates, a lower

number of channels were formed. The contaminant that was present in the vicinity of these channels was removed quickly; however, the contaminant in regions untouched by channels had to diffuse toward the channels in order to be removed. This diffusion-controlled removal allowed residual contamination to persist with removal occurring very slowly.

- 4 Surfactant foam injection at very low air flow rates was found to accelerate the bulk contaminant removal; however, the total removal of the contaminant was difficult to achieve using surfactant foams. SDS was found to be a more effective surfactant than witconol SN70. Surfactant enhancement may be a viable option for obtaining significant removal efficiencies when high flow rates are not possible due to site geology or other site-specific conditions.
- 5 Overall, based on the experiments of this study, air sparging has shown to be efficient in removing VOCs from saturated soils. High rates of removal can be achieved from highly permeable soils such as gravels with low flow rates; however, in soils with relatively lower permeability such as sands, higher flow rates are required to remove contamination. In sandy soils, the use of low air flow rates may result in lingering residual contamination, even with the use of surfactants.

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