Effects of Soil Heterogeneity on Airflow Patterns and Hydrocarbon Removal during In Situ Air Sparging

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ABSTRACT: This paper presents the results of a laboratory investigation performed to study the effect of soil heterogeneity on the removal of benzene from six different homogeneous and heterogeneous soil profiles using in situ air sparging. Air injected in homogeneous coarse sand profiles traveled in channels within a parabolic zone. Within fine gravel, injected air traveled in bubbles and was confined to a smaller zone of influence than within the sand profiles. Heterogeneous soil profiles were subjected to airflow patterns that were combinations of patterns observed in homogeneous soil profiles. When hydraulic conductivity differences between adjacent soil layers or inclusions were less than a factor of 10, air freely entered into the lower permeability soil regions. However, when differences were greater than a factor of 10, the injected air followed a path of least resistance and avoided regions of lower permeability. Regions that were subjected to high airflow were remediated rapidly as a result of efficient vapor-phase partitioning due to volatilization. Regions that were not subjected to high airflow, whether the result of locations outside of the zone of influence or airflow diversion due to permeability differences between adjacent soil layers/inclusions, did not experience rapid contaminant removal and required rate-limiting diffusion for contaminant removal. Overall, the importance of determining the degree of soil heterogeneity at a given site with a detailed site investigation prior to implementing air sparging is demonstrated.

INTRODUCTION

In situ air sparging is a technique that has proven to be efficient for the remediation of saturated soils and ground water contaminated with volatile organic compounds (Ardito and Billings 1990; Marley et al. 1992; Johnson et al. 1993; Leonard and Brown 1994; Reddy et al. 1995; Semer and Reddy 1997; Clayton 1998; Reddy and Adams 1998; Adams and Reddy 2000). During air sparging, a gas, usually air, is injected into the subsurface below the lowest known point of contamination. Due to effects of buoyancy, the injected air will rise through the saturated zone toward the surface. Through a variety of mass transfer processes, the contamination is either stripped into the migrating air or oxygen is transferred into the subsurface, effectively assisting in the degradation of the contamination. As the contaminant-laden air continues to rise, it encounters the unsaturated zone of soil, where it is extracted using a soil vapor extraction system. Once extracted, the contaminated air may be treated using standard methods such as activated carbon filters or combustion.

In situ air sparging is most successfully applied within a thick deposit of saturated, homogeneous granular soil. However, natural soils are often heterogeneous in nature, and, as a result, the design of air sparging systems using airflow and zone of influence assumptions based on homogeneous conditions can lead to inadequate performance. During air sparging, injected air migrates through the subsurface along a path of least resistance. As a result, injected air may completely bypass the regions of lower permeability when layers and lenses of low permeability are adjacent to regions of higher permeability. When bypassed, these regions are not directly impacted by the airflow and rely upon less efficient mechanisms for contaminant removal, including diffusive transport. This can lead to extensive tailing, incomplete contaminant removal, or recalcitrant contaminant conditions within substantial portions of the soil. Additionally, untreated contaminated soil inclusions can act as a source of contamination to surrounding remediated soils and ground water.

Very few studies have been performed to understand the effect of soil heterogeneity on injected airflow (Ji et al. 1993; McCray and Falta 1997; van Dijke and van der Zee 1998). Ji et al. (1993) performed a laboratory visualization study to investigate the effect of soil heterogeneity on flow patterns of injected air during air sparging. Glass beads with diameters ranging from 0.2 to 0.75 mm were used to simulate various soil matrices. McCray and Falta (1997) performed numerical modeling to study the behavior of airflow during air sparging in homogeneous and heterogeneous soils. van Dijke and van der Zee (1998) developed a model capable of incorporating steady-state conditions to study airflow within a high permeability soil overlain by a low permeability soil. Although these previous studies helped demonstrate the significance of soil heterogeneity, a systematic laboratory investigation to study how soil heterogeneity affects contaminant removal and, ultimately, air sparging performance has not been performed.

The objective of this study is to perform controlled laboratory physical model tests to determine how different heterogeneous soil profiles affect injected airflow and dissolved-phase benzene removal during the use of air sparging. To accomplish this objective, different combinations of natural soils are used to create both homogeneous and heterogeneous soil profiles contaminated with dissolved-phase benzene in a 2D aquifer simulation test setup. Air sparging system performance under homogeneous and heterogeneous soil conditions is assessed by carefully delineating airflow within the tested soil profiles as well as monitoring contaminant removal.

EXPERIMENTAL METHODOLOGY

Aquifer Simulation Apparatus

A 2D Plexiglas aquifer simulation test setup, shown in Fig. 1, was used for this study. The large dimensions of the aquifer simulation apparatus allow for the placement of a heterogeneous soil profile, including soil layers, lenses, and other localized variations. The simulator also offers additional flexibility by allowing control of contaminant placement and options related to ground-water flow.

The interior of the tank consists of three compartments; a soil chamber is centered within the tank and is flanked by two ground-water reservoirs. A detachable Plexiglas cover is used to seal the tank. To ensure a good seal with the cover, rubber

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weather stripping is attached to the rim of the tank. The cover is placed over the rim and is clamped into place with a machine screw/wing nut assembly.

On the face of the soil chamber, 20 sampling ports are arranged in four rows of five ports each protruding into the soil chamber. These sampling ports consist of a rubber septum held in place by a polyethylene spacer ring sandwiched between Teflon fittings. An inclined, slotted stainless steel sleeve extends from the sampling port on the face of the tank into the soil chamber, allowing for easy sampling of pore water. Small holes are located at the opposite end from the slots in order to allow for pore-water circulation as well as to prevent the existence of a dead-end pore condition that may exist within the tube.

Three soil removal ports were placed into the bottom of the soil chamber. The middle soil port cover is fitted with two entry ports, each with speed connectors. The first entry port leads to a Teflon filter. This port is used to inject contaminant solution into the test soil prior to a test. The second port leads to a slotted stainless steel tube fitted with a porous stone cover, which serves as a sparge point for air injection during testing.

The two water reservoirs on either side of the soil chamber are connected to constant-head, source reservoirs containing clean water, as shown in Fig. 1. The source reservoirs are adjustable in height, allowing for a specific head to be maintained in each reservoir. The tank reservoirs are separated from the soil chamber by a geotextile-lined perforated stainless steel screen. The screen allows water to freely enter or exit the soil chamber while preventing soil particles from entering the water reservoirs.

A panel is used to control the injection of air. The panel consists of a flow regulator, a pressure gauge, and a Cole-Parmer (Vernon Hills, Ill.) variable area flowmeter. Cole-Parmer C-Flex tubing is used to transfer air from the panel to the air injection port.

Three effluent gas exit ports were installed into the tank cover. These ports lead to three Cole-Parmer C-Flex effluent gas lines, which are fastened to Supelco (Belleville, Pa.) activated carbon tubes for contaminant vapor collection. The carbon tubes collect the contaminant vapors to allow for the performance of a mass balance investigation while preventing vapor emission into the fume hood.

**Materials**

Four different granular soils, a fine uniform gravel ($D_{50} = 4.5$ mm), a coarse uniform sand ($D_{50} = 0.52$ mm), a fine uniform sand ($D_{50} = 0.12$ mm), and a well-graded sand ($D_{50} = 0.66$ mm), were selected for this research. Two soils (coarse uniform sand and fine uniform sand) were obtained from the U.S. Silica Company (Ottawa, Ill.), while the other two soils were obtained from local suppliers. The grain size distribution curves of the four soils are shown in Fig. 2. Benzene (density $= 0.88$ g/cm$^3$, water solubility $= 1.780$ mg/L, vapor pressure $= 76$ mm Hg and Henry's constant $= 5.43 \times 10^{-3}$ atm-m$^3$/mol) was chosen as the representative hydrocarbon contamination because it is a carcinogenic constituent of gasoline and other petroleum products and is a significant contaminant of concern at petroleum release sites.

**Testing Program**

Two homogeneous soil profiles (a coarse uniform sand profile and a well-graded sand profile) were used to compare and contrast how soil grain size and grain-size distribution affect airflow patterns and contaminant removal. To simulate soil
heterogeneity, tests were performed using soil profiles consisting of different soils arranged in layers and lenses. Four heterogeneous soil profiles were used: (1) Well-graded sand between coarse uniform sand; (2) fine uniform gravel under coarse uniform sand; (3) fine uniform gravel between coarse uniform sand; and (4) fine uniform sand lenses embedded within coarse sand. All tests were performed with an air injection rate of 2,500 mL/min under a pressure of 4.5 kPa.

**Testing Procedure**

When preparing a test, two stainless steel sheets were placed into the soil chamber to create three subchambers. The chambers were created to help contain the placed contaminant before testing; the two outside subchambers were kept contaminant-free, while benzene solution was injected into the middle chamber. After the sheets were placed, all three soil chambers were filled by pouring dry soil through the top of the tank using a consistent drop height. When layers or lenses of different soil were used to create the soil profile, care was taken to assure accurate placement of the different soils into appropriate thickness. The mass and the volume of the placed soils were recorded to calculate density and porosity of soils.

A benzene solution with a concentration of 200 mg/L was mixed in 4-L-capacity borosilicate glass bottles. The bottle lid was fitted with an injection port consisting of a rubber septum held in place with polyvinyl chloride fittings. Benzene was directly injected through the injection port into the deionized water already sealed without headspace in the bottle. Since headspace in the bottle was eliminated, benzene loss through volatilization was minimized. The bottles of benzene solution were placed on stirring plates and allowed to mix for approximately 1 h.

Following mixing, the benzene solution was injected into the middle subchamber through the contaminant injection port located on the middle tank porthole cover. A peristaltic pump was used for contaminant injection at a rate selected to prevent fracturing of the soil profile. While the contaminant was placed, the two side subchambers were simultaneously saturated with clean tap water that infiltrated from the water reservoirs, preventing the development of a hydraulic gradient between the subchambers and reducing or eliminating unwanted migration out of the initial contaminant zone. Once the contaminant zone was established and saturation of the side subchambers was completed, the steel sheets were removed from the soil profile, the tank lid was fastened and sealed in place, and effluent air tubes filled with activated carbon filter tubes were fastened to the tank lid effluent ports. The initial benzene concentration distribution was determined through sampling and analysis of the pore water from the sampling ports, and then air injection commenced.

During the course of testing, air was continuously injected using a specified flow rate of 2,500 mL/min under an injection pressure of 4.5 kPa. The resulting airflow patterns were carefully sketched and photographed. Air injection was continued until a predetermined sampling time was reached. At this time, air injection was briefly interrupted to allow sampling of pore water and subsequent analysis using gas chromatography. The air injection was only stopped for the time needed to sample pore water from the ports (approximately 10 min). During the brief shutdown, the activated carbon tubes also were replaced. Once these tasks were completed, air injection resumed.

**Chemical Analysis**

To assess the remedial action during the experiments, chemical analysis of both the extracted pore water and the activated carbon tubes was performed. The chemical analysis was performed using gas chromatography. Two gas chromatographs (GCs) were used to perform analysis. The first GC was an SRI model 9300B equipped with a Por-a-pak column and a photodiode ionization detector used to monitor pore water within the test profile. Two microliters of pore water were directly injected into the gas chromatograph for analysis. Pore water was continually analyzed using this GC during the duration of a test to monitor remedial progress.

The second GC used was a Hewlett-Packard Series 6890 gas chromatograph. It was equipped with a Supelco 60/80 Carbopack packed column connected to a flame ionization detector. This GC was used to analyze the contaminant that had been adsorbed within the activated carbon tubes. The activated carbon used to collect effluent contaminant vapors for a given time period was placed into a 250 mL amber glass bottle, and 250 mL of acetone (Fisher Scientific A949-4 HPLC-grade)
was used as an extractant. The bottle was shaken for approximately 1 h, and an aliquot sample was drawn using a gas-tight syringe. The sample was then analyzed using the GC and the total contaminant adsorbed by the given activated carbon tube was determined.

Quality Control

Several precautions were taken to assure accurate, reproducible results. Due to cost and ease of construction considerations, Plexiglas was used for apparatus fabrication. To determine the extent of contaminant adsorption to Plexiglas, several batch tests were performed in which a Plexiglas column was filled with dissolved-phase benzene solutions. Over a range of test times and contaminant concentrations, adsorption of the contaminant to the Plexiglas was found to be negligible. Accuracy of the gas chromatography analysis was assured by (1) GC calibration prior to each test; (2) testing blank samples; and (3) analyzing duplicates to check the accuracy of this method of analysis. To verify the accuracy of the air sparging testing procedures, mass balance analysis was attempted using the effluent contaminant vapors collected during testing. Selected duplicate tests were also performed to assure that results of the testing were reproducible.

RESULTS AND ANALYSIS

Homogeneous Soil Profiles

Coarse Uniform Sand

Fig. 3 shows the homogeneous coarse uniform sand profile, including the soil profile height and width as well as the depth of the saturated zone. The figure also shows the arrangement of the sampling ports within the soil profile. Upon air injection into the saturated homogeneous coarse sand profile, air was observed to travel in the form of pore-scale channels, as shown in Fig. 3. The injected air formed a parabolic zone of influence emanating from the point of injection. As a result, the injected air did not traverse the lower left and lower right regions of the initial contaminant zone. As the injected air migrated vertically toward the surface, lateral migration also occurred, resulting in an increased radius of influence with increased height.

The initial contaminant distribution, as shown in Fig. 4(a), was relatively uniform throughout the initial contaminant zone, but a lower contaminant level was detected in the upper left region of this zone. The extent of the air channel pattern, as shown in Fig. 3, was confined to the initial contaminant zone, effectively intercepting all areas with the exception of the regions immediately to the left and right of the point of injection. Figs. 4(b–d) show the benzene distribution within the soil after an elapsed time of 135, 500, and 2,875 min, respectively. Because airflow was highest directly above the point of injection, this region experienced the greatest contaminant reduction during the initial stages of testing. Benzene removal occurred in a bottom to top pattern; the region immediately above the injection source experienced significant removal first with regions above experiencing removal in a consecutive manner. As the injected air migrated upward, substantial contaminant partitioned into the vapor phase. During migration, the benzene concentration within the air may have approached saturation, reducing the driving force for further volatilization and in turn resulting in less efficient removal at greater soil elevations (Hayden et al. 1994). Additionally, the contaminant-laden air may have caused repartitioning into the aqueous phase at greater soil elevations (Johnston et al. 1998). Figs. 4(b and c) show that as the middle regions became contaminant-free, the regions to the left and right also experienced significant benzene reductions, but at a slower rate than the middle regions. As stated earlier, these regions were subjected to substantial airflow but less than within the middle regions.

Volatilization resulting from the extensive airflow within these regions accounted for the efficient removal of benzene. The extensive effect of volatilization resulted from the high density of air channels carrying the injected air. The partitioning of the contaminant in the vicinity of air channels was rapid compared to the removal of contaminant from water filled regions outside of the channels (Johnson 1998). The flowing air also created mechanical dispersion within the ground water in these locations, aiding in further benzene movement toward the channels. Migration of contaminant as a result of mechanical dispersion is more effective than diffusion for removal (Johnson 1998).
Benzene removal due to volatilization in these regions occurred so quickly that the effects of other mass transfer/transport mechanisms were difficult to observe. However, benzene migration resulting from aqueous-phase advection was detected. This effect may have contributed to the higher benzene concentrations within the upper initial contaminant region compared to the middle regions, as shown in Fig. 4(b). These increases in benzene concentration, however, are slight, indicating that the benzene partitioned into the vapor phase before further migration occurred as a result of pore-water migration.

While the benzene in regions of substantial airflow was removed efficiently, benzene outside of such regions lingered at relatively high concentrations for a longer time. The lower left and lower right regions of the contaminant zone exhibited such behavior, as illustrated in Fig. 4(d). Because these regions were essentially free of airflow, dissolved-phase benzene did not partition into the vapor phase through volatilization. Instead, contaminant removal was controlled by the aqueous diffusion through the porous media to the air channels as a result of the induced concentration gradients. The diffusion of dissolved-phase volatile organic contaminants within the soil pore water toward the aqueous/vapor-phase interfacial mass transfer area is often the rate-limiting factor for contaminant removal (Hayden et al. 1994).

The rate of transport of aqueous-phase benzene was slow enough to require a substantial amount of time for diffusion to occur, regardless of the short transport path lengths. Eventually, the benzene was removed; however, complete benzene removal required nearly 4,000 min of air injection. Regions experiencing high airflow, however, were contaminant-free after approximately 500 min of air injection. This demonstrates that diffusion becomes the rate-limiting factor for contaminant removal. It should be noted that no spreading of contaminant (migration into previously clean areas) occurred, either as a result of diffusion or airflow-induced advection.

**Well-Graded Sand Profile**

Fig. 5 shows the height and width of the well-graded sand soil profile as well as depth of the saturated zone. Once air injection into the saturated well-graded sand profile began, the injected air traveled in pore-scale channel mode, but the extent of the zone of influence observed was much wider than in the previous test with the coarse sand, as depicted in Fig. 5. While the permeability of the well-graded sand (hydraulic conductivity = $1.30 \times 10^{-2}$ cm/s) was slightly lower than that of the homogeneous coarse sand (hydraulic conductivity = $4.64 \times 10^{-2}$ cm/s), the porosity of the well-graded sand (porosity = 0.35) was also lower than in the coarse sand (porosity = 0.45). The lower value of porosity led to increased tortuosity within
FIG. 5. Airflow Pattern in Well-Graded Sand Profile

FIG. 6. Benzene Distribution in Well-Graded Sand Profile: (a) $t = 0$; (b) $t = 60$ min; (c) $t = 225$ min; (d) $t = 520$ min

Note: All Concentrations at the Ports are Expressed in mg/l.
the well-graded sand. The greater tortuosity leads to greater lateral air migration and a wider zone of influence.

Fig. 6(a) shows the initial contaminant concentration distribution of this test. As shown, the initial contaminant distribution is more uneven than in the homogeneous coarse sand test. Notably, the initial concentrations in the upper left and upper right regions of the initial contaminant zone are lower than other regions. Figs. 6(b–d) show the contaminant distribution within the soil profile after an elapsed time of 60, 225, and 520 min, respectively. The middle regions showed the first significant benzene concentration reductions due to the substantial airflow within these regions, resulting in efficient benzene removal through volatilization. Additionally, the “bottom-up” removal behavior seen in the uniform coarse sand test occurred; the region surrounding the injection point was remediated first, followed by the middle and upper regions, respectively. The center left and middle right regions also experienced substantial benzene concentration reductions early in the remedial program, but not as fast as within the middle regions. The lower airflow within these regions led to a smaller mass transfer area, slowing the rate of volatilization.

Unlike the coarse uniform sand test, the lower left and lower right regions underwent substantial benzene reductions, as illustrated in Figs. 6(b–d). As stated before, the zone of influence of the injected air was much wider than in the previous test with the homogeneous coarse sand. A substantial volume of air migrated through these lower regions, expediting benzene removal through volatilization. As a result, necessary diffusion paths were greatly reduced, eliminating the lingering contaminant effect seen within the homogeneous coarse sand layer. While benzene was removed faster within the lower layers, Fig. 6(b) shows that benzene lingered in the upper left and upper right regions of the initial contaminant zone as a result of two phenomena. First, vertical benzene migration occurred as a result of advection due to the injected air. Second, since the zone of influence is wider than in the coarse sand but consists of the same volumetric airflow, the air channel density is reduced. This is especially true in the upper regions of the initial contaminant zone, where the flowing air has been subjected to substantial lateral expansion, greatly reducing air channel density within these regions.

Further, as shown in Figs. 6(c and d), the benzene is removed from the entire soil profile in an expeditious manner; dissolved-phase benzene is almost completely removed from the soil profile after 520 min. Even though the zone of influence is not as dense as in the homogeneous sand layer, the zone was more extensive, which led to greater coverage of the contaminant zone. Additionally, the lower permeability may have led to greater air saturation within the zone of influence, enhancing the effect of volatilization (Chen et al. 1996). As a result, the reliance upon diffusion to assist in contaminant removal is reduced.

While the wider zone of influence was able to efficiently treat a greater volume of soil than in the coarse uniform sand test, the wider zone of influence induced spreading of the contaminant into initially contaminant-free regions. The zone of influence of the airflow entered into regions that were previously free of contamination, resulting in lateral contaminant migration due advective-dispersive transport into these regions. Dissolved-phase benzene that infiltrated into these regions did not remain for an appreciable length of time; migrating air responsible for its advective-dispersive movement was also capable and responsible for its efficient removal through volatilization.

**Heterogeneous Soil Profiles**

**Well-Graded Sand between Coarse Uniform Sand**

To study a heterogeneous setting in which two adjacent soil layers have different permeability and grain-size distribution, a test was performed in which a layer of well-graded sand (hydraulic conductivity = $1.3 \times 10^{-2}$ cm/s) was underlain by a layer of coarse uniform sand (hydraulic conductivity = $4.64 \times 10^{-2}$ cm/s) as shown in Fig. 7.

The airflow pattern resulting from air injection is depicted in Fig. 7. This airflow pattern appeared to be a hybrid of two previous tests performed using a homogeneous coarse uniform sand layer and a homogeneous well-graded sand layer. The zone of influence maintained its characteristic parabolic shape as it migrated through the coarse uniform sand layer but expanded laterally when it encountered the well-graded sand.

![FIG. 7. Airflow Pattern in Well-Graded Sand between Coarse Uniform Sand Profile](image-url)
layer similar to what was previously observed. This behavior is consistent with that reported by van Dijke and van der Zee (1998) when a similar layered soil profile was mathematically modeled. They reported that near the interface of the two soils, capillary forces and gravity forces are of equal magnitude, creating a narrow zone at the coarse uniform sand/well-graded sand interface in which substantial injected air lateral migration occurs. Lateral expansion of the zone of influence results from this phenomenon. Since the same volume of injected air migrating through the coarse sand entered into and migrated through the well-graded sand, the resulting zone of influence within this layer led to a lower channel density within the well-graded sand layer than within the coarse sand layer.

Fig. 8(a) shows the initial contaminant distribution within this soil profile. The contaminant distributions following 60, 500, and 6,045 min of air injection are shown in Figs. 8(b–d), respectively. The contaminant removal behavior is consistent with the result of the injected airflow pattern. The middle region of the coarse sand layer is in the immediate vicinity of the air injection point and is subjected to substantial airflow; as a result, this region is the first to experience significant contaminant reduction through volatilization. As shown in Fig. 8(b), this region experienced significant reductions after 60 min. As before, the lower left and right initial contaminant regions within the sand require diffusive dissolved-phase transport for benzene removal.

The upper left region of the initial contaminant zone exhibits behavior worthy of further comment. The initial benzene concentration detected within this region is relatively lower than other regions, as demonstrated in Fig. 8(a). With increased remedial time, the benzene concentrations within this region increased greatly, as shown in Fig. 8(c). This increase may be a result of advective-dispersive transport of dissolved-phase benzene. Dissolved-phase benzene within lower regions migrated vertically into the well-graded sand layer. Contaminant spreading due to advective-dispersive transport must be considered during field application of air sparging.

**Fine Uniform Gravel under Coarse Uniform Sand**

To study the effect of adjacent layers with a significant difference in permeability, a test was performed in which a layer of coarse uniform sand (hydraulic conductivity = $4.64 \times 10^{-2}$ cm/s) was underlain by a layer of fine uniform gravel (hydraulic conductivity = 1.60 cm/s). Fig. 9 shows the test soil profile as well as the depth of the saturated zone.

Once air injection began, a very narrow band of airflow extended from the point of injection upward through the fine
gravel layer as depicted in Fig. 9. This air migrated in bubble form as opposed to the pore-scale channels observed in previously tested soils. Because a narrow zone of influence developed within this region consisting of the same volumetric flow as in previous tests, a high level of agitation due to the bubbling action of the airflow was noted. Once this air migrated vertically and reached the homogeneous coarse sand layer, the flow was diverted and migrated laterally along the sand-gravel interface. Very little flow into the coarse sand layer was observed. The diverted flow continued to migrate laterally, eventually exiting the soil chamber along the sand-gravel interface into the water reservoirs.

The injected air does not enter the coarse sand layer because of the large permeability difference between the two layers; the horizontal permeability within the gravel layer allowed the air to migrate laterally as opposed to continuing its vertical flow due to buoyancy. This difference in permeability between the adjacent layers (approximately a ratio of 1:35) is too great for the injected air to enter into the less permeable coarse sand layer. The injected air, traveling along a path of least resistance, travels laterally through the more permeable fine gravel, entirely avoiding the coarse sand layer.

Fig. 10(a) shows the initial contaminant distribution within the soil profile. The absence of airflow within the coarse sand layer is further illustrated by the contaminant concentration distribution; Figs. 10(b–d) show the contaminant distribution after an elapsed time of 220, 1,480, and 5,675 min, respectively. As shown in Fig. 10(b), the middle region of the fine gravel layer experiences significant contaminant reduction during the initial stages of air injection. This is a result of the substantial volumetric airflow traveling through this region. Additionally, because the air is migrating in bubble form as opposed to flow within pore-scale channels, partitioning of dissolved-phase benzene into the vapor phase occurs more efficiently due to the increased mass transfer interfacial area across which volatilization may occur.

With further air injection, the remainder of the fine gravel layer became free of dissolved-phase benzene. After 220 min of air injection, small concentrations of dissolved-phase benzene are detected within this layer, as shown in Fig. 10(b).

The injected air traveling along the interface between the coarse sand layer and the fine gravel layer is able to remove dissolved-phase benzene from the regions to the left and the right of the injection point. Even though the air does not directly traverse these regions, the high permeability of the fine gravel allows movement of dissolved-phase benzene toward the sand-gravel interface through advective-dispersive and diffusive transport. The injected air migrating along the sand-gravel interface removes dissolved-phase benzene in the immediate vicinity of the interface efficiently through volatilization. This creates a concentration gradient between regions near the interface and locations located away from these regions, inducing diffusional transport.

While advective-dispersive and diffusive transport within the gravel layer occurs at a rate fast enough to contribute to efficient removal, the rate at which diffusive transport occurs within the coarse sand layer prohibits efficient remediation. This observation is supported by Fig. 10(c); the fine gravel layer is virtually free of benzene contamination after 1,480 min, but substantial benzene concentrations remain throughout the coarse sand layer. The absence of airflow within the sand layer leads to notable diffusion behavior. The dissolved-phase benzene migrates from the upper regions of the coarse sand layer, but, in order to reach the mass transfer interface to partition into the vapor phase at the sand-gravel interface, it must first pass through the middle regions of the sand layer. As a result, there is actually an increase in benzene concentration in these regions. The changes in benzene concentrations observed in the upper regions of the coarse sand layer are seen in Fig. 10(d).

In addition to benzene migration along the sand-gravel interface, lateral benzene migration through the gravel layer occurred as a result of advective-dispersive transport. The airflow induced ground-water movement within this region. This is verified through detection of benzene within the gravel layer to the left of the initial contaminant zone. The highly permeable gravel overlain by coarse sand of lower permeability presents problems that may occur during field application. Since the injected air will only follow a path of least resistance, it will not vertically migrate through the soil unless it encounters
Note: All Concentrations at the Ports are Expressed in mg/L.

FIG. 10. Benzene Distribution in Fine Uniform Gravel under Coarse Uniform Sand Profile: (a) $t = 0$; (b) $t = 220$ min; (c) $t = 1,480$ min; (d) $t = 5,675$ min

FIG. 11. Airflow Pattern in Fine Uniform Gravel over Coarse Uniform Sand Profile
FIG. 12. Benzene Distribution in Fine Uniform Gravel between Coarse Uniform Sand Profile: (a) $t = 0$; (b) $t = 60$ min; (c) $t = 435$ min; (d) $t = 1,735$ min

Note: All Concentrations at the Ports are Expressed in mg/L.

a location in which movement is not resisted. Therefore, if the difference in permeability in two adjoining layers of soil is large enough, vertical migration toward the surface will not occur. The resulting lateral movement will result in unwanted off-site migration if vertical movement and migration is unattainable. Caution must be exercised in such a situation to assure that contaminant transport away from the treatment zone does not occur.

Fine Uniform Gravel between Coarse Uniform Sand

To study the effect of a stratified soil layer consisting of a higher permeability soil between a lower permeability soil, a test was performed in which fine uniform gravel was underlain and overlain by coarse uniform sand. The soil profile and the thickness of the saturated soil zone are shown in Fig. 11.

Fig. 11 shows the observed airflow pattern resulting from air injection. The air migration through the lowest layer of coarse sand resembled the flow observed within a homogeneous profile of coarse sand. As shown in Fig. 11, a parabolic zone of influence was formed; substantial airflow occurred within the middle region of the sand in close proximity of the injection point, but little airflow occurred within the lower left and lower right regions of the initial contaminant zone. Once the vertically migrating air reached the fine gravel layer, the injected air migrated in bubble mode, similar to what was observed in the previous test within the gravel layer. The boundaries of the zone of influence within the gravel layer fluctuated greatly; the majority of the airflow was confined to the center regions of the gravel. Substantial airflow created vigorous bubbling activity within this region. A transient boundary of the airflow, however, fluctuated laterally from the center of this region out toward the edges of the initial contaminated region. The bubbling activity of the air within these fringe regions was less vigorous than within the center regions of air flow.

As shown in Fig. 12(a), the initial concentration distribution is relatively uniform with the exception of the upper right region, where contaminant levels were slightly lower than within the rest of the region. The effect of the airflow pattern within the gravel is evident from the contaminant profiles that existed throughout the test. After approximately 60 min of air injection, a distinctive contaminant distribution exists as shown in Fig. 12(b); contours of benzene concentration are parallel to the vertical axis of the contaminant zone and thus also parallel to the flow of air through the gravel. The migrating air within the gravel effectively removed benzene that initially existed within the middle regions of the contaminated area. Additionally, the flowing air greatly reduced the benzene concentrations in the side regions within the gravel layer. These concentration levels continue to decrease until the regions are contaminant-
free after approximately 435 min of air injection, as shown in Fig. 12(c).

The high permeability of the fine gravel, however, allowed contaminant to spread away from the source. This migration may be caused by two possible mechanisms: (1) Diffusive transport resulting from concentration gradients; or (2) advective-dispersive transport resulting from pressure gradients induced by the flowing air. Due to the high rate of observed benzene migration, advective-dispersive transport is the most likely cause. Therefore, caution must be exercised when applying air sparging to soils of high permeability in order to account for and/or prevent unwanted off-site migration.

Airflow within the sand led to benzene removal similar to that observed within a homogeneous coarse sand layer. Due to its close proximity to the point of air injection, the middle region of the coarse sand layer experienced significant reductions in benzene concentrations during the initial stages of air injection, as shown in Fig. 12(b). As in previous tests, benzene removal from regions to the left and right of the middle region occurred through diffusive transport, requiring lengthy removal times. The effect of this transport is clearly seen in Figs. 12(c and d). After approximately 435 min of air injection, the middle contaminated region within the sand layer is essentially benzene-free, while the regions to the left and the right still contain significant concentrations of benzene. After 1,735 min of air injection, however, the benzene concentrations within the left and right regions in the sand layer decreased (the right region substantially), while the middle region experienced a noted increase in concentration. Benzene migrated from the two side regions into the middle region through diffusive transport. The increased concentration of benzene within the middle region is subsequently removed through volatilization. The contaminant within the side regions continues to migrate into the middle region through diffusive transport, eventually eliminating benzene contamination within these regions.

Coarse Uniform Sand with Fine Uniform Sand Lenses

To study the effects of the presence of lenses of low permeability soils within higher permeability soils on air sparging performance, a test was performed using coarse uniform sand containing two lenses of fine uniform sand. These lenses of lower permeability were located in the middle left and middle right regions of the initial contaminant zone. A 10-cm layer of coarse uniform sand was first placed into the test apparatus. Next, two regions were cordon off and filled with fine uniform sand, yielding two lenses measuring 10 cm in height and 15 cm in length. Regions between and outside of the fine sand lenses were filled with the coarse sand. Coarse sand was next placed to complete the soil profile. Fig. 13 shows the soil profile as well as the depth of the saturated soil zone.

Once air injection began, airflow patterns as depicted in Fig. 13 were observed. The airflow within the lower region (consisting of coarse sand) resembled the flow observed within a homogeneous coarse sand profile. After migrating vertically and encountering the fine sand lenses, the migrating air circumvented the lenses, either flowing within the space between or migrating around the lenses. The low permeability of the fine sand (hydraulic conductivity = 3.38 × 10⁻³ cm/s) as compared to the surrounding coarse sand (hydraulic conductivity = 4.64 × 10⁻² cm/s) forced the migrating air to circumvent the lenses. This demonstrates that the permeability ratio of approximately 10:1 between the two soils is significant enough to prevent air migration through the low permeability soil. It may be possible for injected air to enter beneath lenses and break through when sufficient pressure develops (Lundegard and Andersen 1996), but this effect was not observed during this test. Once the vertically migrating air passed around the fine sand lenses, a zone of influence characteristic of a coarse sand was again formed. While the fine sand lenses forced the lateral extent of the zone of influence to expand, their lower permeability prevented air migration within the lenses.

Fig. 14(a) shows the initial contaminant distribution within the soil profile. Benzene concentration changes because air injection reflected the observed airflow pattern. Since the majority of the flowing air migrated through the space between the fine lenses, these regions experienced the first substantial benzene reductions, as shown in Fig. 14(b). These middle regions were the only regions within the initial contaminant area that were in direct contact with the airflow; as a result, dissolved-phase benzene within this region efficiently partitioned into the vapor phase due to the substantial interfacial mass transfer area. Other regions within the initial contaminant

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area were reliant upon diffusive transport for benzene removal. The contaminated regions directly above the fine sand lenses were the next regions to experience significant benzene reductions. As mentioned before, once the air passed between the two fine sand lenses, it migrated laterally, forming the characteristic parabolic zone of influence previously observed in coarse sand. The migrating air partially intercepted these regions, shortening the diffusion paths required for benzene removal and increased removal rates. Diffusion was still the rate-limiting factor for removal in these regions as well as within the regions immediately below the fine sand lenses. The injected air did not migrate through the regions below the lenses. The diffusion paths for this migration were longer than those required in the regions above the fine sand lenses, leading to longer removal times.

The regions below the fine sand lenses are eventually remediated, leaving the fine sand lenses as the only contaminated regions within the soil profile, as shown in Fig. 14(c). Because the injected air completely avoided traveling into these lenses, the benzene needed to migrate outward through diffusive transport toward regions experiencing airflow. A substantial concentration of benzene remained within the lenses for an extended period of time, as shown in Fig. 14(d). Significant contaminant concentrations were detected within the lenses even after 5,760 min. Because the flowing air completely avoided the lenses, the contaminant within the lenses relied upon diffusion for removal. The long diffusion paths required for contaminant removal from the lenses led to the slow removal that was observed during the course of the test.

**CONCLUSIONS**

The effects of soil heterogeneity on the efficiency of air sparging was investigated using laboratory experiments. Simulated soil profiles using different combinations of layers and lenses as well as homogeneous soil layers were contaminated with dissolved-phase benzene and subjected to air injection. The following conclusions were drawn from this study:

- Within a coarse uniform sand soil profile, injected air led to the establishment of a parabolic zone of influence through which air flowed into channels with sizes on the scale of a few pore diameters. When a profile of well-graded sand was tested, the size of the zone of influence increased due to the lower air permeability and the increased effect of tortuosity resulting from the wider range of grain-size distribution. Within a larger zone, however, the air saturation (or channel density) is not as great as in a smaller zone of influence for a given air injection flow rate. When air was injected into fine uniform gravel, it
traveled in bubble mode. The resulting lateral dimensions of the zone of influence within the fine gravel was less than within the sand profiles due to the high vertical permeability and low airflow resistance within the fine gravel.

- When two adjoining layers of soil have small differences in permeability and tortuosity, injected air enters into the lower permeability layer. When the air enters into a layer with a low permeability and a greater tortuosity, the size of the zone of influence will increase, but the air channel density/degree of air saturation will decrease.

- When the permeability ratio between two adjoining layers or lenses is >10:1, such as coarse sand underlain by fine gravel or fine sand lenses embedded in coarse sand, migrating air bypasses the soil with lower permeability. As a result, large regions of soil may not be subjected to airflow, preventing efficient contaminant remediation.

- Volatilization is the controlling removal mechanism during the initial stages of air injection and is the means for vapor-phase contaminant partitioning and removal. For volatilization to occur, injected air must come into contact with contamination. In the case of lower permeable layers underlain by high permeability strata or low permeability lenses being surrounded by higher permeability soil, air is prevented from entering into the low permeability soils. Contaminant removal from the low permeability soils must occur through diffusion to concentration gradients induced by volatilization. Diffusion is the rate-limiting removal mechanism. If major portions of contamination require diffusion for removal, remedial times during the use of air sparging may be greatly increased.

- Contaminant spreading due to advective-dispersive transport was observed in well-graded sand. The airflow entered into regions initially free of contamination and led to slight benzene concentration increases. In more permeable fine gravel, however, advective dispersive transport leads to sizable benzene spreading. This phenomenon may become problematic in soils with relatively high permeability as off-site migration may contaminate regions initially free of contamination. This migration may take the form of either vapor migration or ground-water transport.

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APPENDIX. REFERENCES


