original article

Soil remediation via bioventing, vapor extraction and transition regime between vapor extraction and bioventing

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ABSTRACT

Aims: The main objectives of this study were evaluation of the efficiencies of bioventing (BV), soil vapor extraction (SVE) and transition regime between BV and SVE (air injection bioventing [AIBV]) for benzene and toluene removal from polluted sandy soils.

Materials and Methods: Laboratory-scale set-up consisted of three cylindrical units (with 29 cm in length with a 7.29 cm i.d.) was conducted to study the removal efficiency of three *in-situ* remediation technologies.

Results: The results showed that, after 48-h air injection with constant air flow rate of 250 mL/min, benzene (initial concentration of 1 mg/g of soil) removal efficiency in BV, SVE and AIBV reactors were almost 84, 98 and >99.5%, respectively. Also results indicated that, toluene with a similar concentration was successfully (>99.5%) reduced via AIBV technology, after 72-h continuous air injection.

Conclusion: Comparison of the BV, SVE and AIBV technologies indicated that all of those technologies are efficient for remediation of unsaturated zone, but after specific remediation time frames, only AIBV able to support guide line values and protect ground waters.

Key words: Benzene, bioventing, soil remediation, toluene, vapor extraction

INTRODUCTION

Industrial activities, municipal and industrial wastes disposal or environmental accidents can cause soil contamination. In many cases, the little amount of an organic compound introduce in the soil is sufficient to pollute great volumes of soil and groundwater.[1] The contamination of the groundwater, surface and subsurface soils with organic chemicals is one of the major problems has occurred during the last decades. Aromatic hydrocarbons are accounted to be more hazardous than of aliphatic types because of their higher mobility in the subsurface and their various adverse effects.[2]

Benzene and toluene are flammable and volatile monoaromatic hydrocarbons that have used in different solvents and find in many fuels including petroleum and gasoline,[3] and which are the main water-soluble compounds in the fuels.[4] These chemicals have detrimental effects on the environment and human health. Cancer, irritation of mucosal membranes, hematological changes, destruction of the central nervous

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system, liver, and kidney interruption are the main adverse health effects of these compounds on human.[5] United States Environmental Protection Agency (USEPA) categorizes these compounds as environmental priority pollutants that their removal from the environment is indispensable.[6] The bioremediation can be classified into *in-situ* and *ex-situ* techniques, in general.[7] Contrary to *ex-situ* technologies, *in-situ* techniques treat the contaminants in place avoiding excavation and transport of them.[8]

Bioventing (BV) is one of the *in-situ* standard technologies for vadose zone remediation.[9] This reliable technique has many advantages such as: Relative simplicity, high efficiency, lower cost than other technologies,[10] minimize off-gassing[11] and minimal site disturbance,[12] but the most important disadvantage of BV is extended remediation time often required.[10,13] Furthermore, BV needs oxygen and nutrient to motivate microbial activity.[7] Soil vapor extraction (SVE) as another *in-situ* standard aeration-based technique can combine with BV and improve biodegradation by an oxygen supply and reduce its treating time as a final point.[13]

Magalhães *et al*. indicated that toluene with initial concentrations of 2 and 14 mg/g of natural soil, were treated successfully (99% removal efficiency) using a transition regime between BV and SVE (air injection bioventing [AIBV]) techniques, at a constant air flow rate of 130 mL/min during 5 days.[13]

Österreicher-Cunha *et al*. studied the AIBV of gasoline–ethanol contaminated undisturbed residual soil with applying a 2 psi constant airflow. They observed that gasoline with concentration of 117 mg/g of soil was reduced by 95%within 60 days.[14]

However, mentioned studies[13,14] found that BV can be integrated with SVE and dramatically remediate a contaminated vadose zone.

This paper is focused on the investigation of three *in-situ* technologies to remove benzene and toluene from contaminated sandy soil, and on soil moisture and microbial population variations under laboratory-scale conditions.

MATERIALS AND METHODS

Chemicals and reagent

The composition of the solution was per liter of sterilized tap water: $Na_2HPO_4.H_2O$ (0.134 g/L; Merck, Germany), KH_2PO_4 (0.03 g/L; Merck, Germany), NaCl (0.5 g/L; Merck, Germany), NH_4Cl (3.982 g/L; Scharlau, Spain), $MgSO_4.7H_2O$ (2.47 g/100 mL; Merck Germany); 1 mL salt/100 mL medium, $CaCl_2$ (111 mg/100 mL; Merck Germany); 1 mL salt/100 mL medium.

Benzene (0.5 g/L; with ≥ 99% purity, Merck, Germany) and toluene (0.5 g/L; purity of 99.5%, Merck, Germany) were added to the medium as the sole carbon sources and agar-agar (Merck, Germany) has been used as a thickening agent only in agar-plate technique.

Inoculum preparation

Soil sample for microbial isolation were collected from aged contaminated soil by petroleum products located in south-western of Iran. Adaptation and enrichment procedures of toluene and benzene degrading bacteria was conducted using batch methods as described in Wolica *et al.*,[15] and were used as inoculum for BV and AIBV reactors.

Microbial cell count

Two gram each of moist sandy soil was taken from two ports on the BV and AIBV reactors. Eighteen milliliter of sterilized water solution containing 0.9% NaCl was added to each soil sample. This solution was vortex robustly at maximum speed for 5 min and serially diluted with sterilized water and then inoculated into nutrient agar-agar media. The plates were incubated at least 5 days at 25°C, before counting the colonies. The concentration of bacteria was reported as number of colony-forming units per gram dry weight of sandy soil (colony forming unit [CFU]/g d.w of soil).

Extraction method

Benzene and toluene were extracted from the soil samples with carbon disulfide and diethylether as low boiling solvents, respectively. One gram of soil sample were added and mixed with 5 ml of solvent in tightly stoppered glass tubes (Schot, Germany), was blended in maximum speed and centrifuged at 6000 rpm for 1and 5 min, respectively. The recoveries of the extraction methods are 74.5% and 93.8% for benzene and toluene, respectively.

Analytical method

The extracted benzene and toluene were analyzed by a gas chromatograph (Agilent GC, 7890A, Netherlands) equipped with flame ionization detector Gas Chromatography-Flame Ionization Detector (GC-FID).

The characteristic of GC column was Agilent 19091S-433: 30 m × 250 μm × 0.25 μm. Helium with flow rate of 1.11 mL/min was used as the carrier gas. The temperatures of the oven, injector, and detector were fixed at 150, 210, and 250°C, respectively.

Soil pH level was determined by adding 10 g of sandy soil to 20 mL of calcium chloride solution (0.01 M). The pH value of the suspension was measured using a pH meter (Eutech, UK).

The porosity of sandy soils was measured using method as described in Qin *et al.*,[16] and the size distribution and bulk density were tested accordance with ASTM C-136 using standard sieve sizes and ASTM D-854,[17] respectively.

Characterization of sandy soil

The sandy soil was collected from the 2 m under surface of a coastline area of Assalouyeh, Iran. After repeated washing to get lucid water, the soil was dried, first at room temperature during 5 days and after that in oven at 110°C for 24 h. The size distribution of the soil as mm is: 61.4% (0.3-0.425), 30.2% (0.425-0.6), 7.3% (0.6-0.85), 0.1% (0.85-1.18), 1% (1.18-2). The bulk density, porosity and pH of the soil were 992 kg/m^3, 36%, and 7.8, respectively.

Experimental set up

A schematic of the experimental set-up is shown in Figure 1. Three columns made of stainless steel having an internal diameter of 7.29 cm and 29 cm in length were used for experiments in this study. A perforated stainless steel plate at the height of 5 cm of columns was placed to support the sandy soil and distribution of the inlet gas uniformly. Two sampling ports, at the height of 10 and 15 cm of columns were provided. One kilogram of polluted soil with initial concentration of 1mg/g was rapidly placed into the column for each experimental run. Lastly, three activated carbon columns were used for the collection of off-gases.

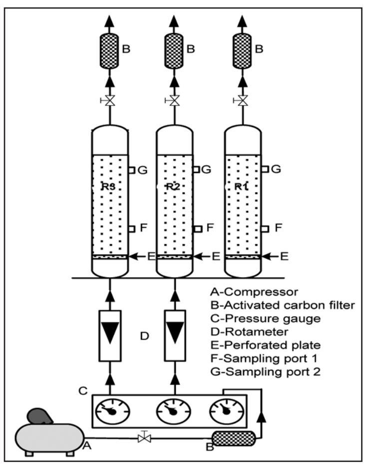

Figure 1: Schematic of the experimental setup for bioventing, soil vapor extraction and air injection bioventing (Reactor 1 [R1]: Sandy soil + inoculum/MM, reactor 2 [R2]: Sandy soil + inoculum/MM + air injection, reactor 3 [R3]: Sandy soil + sterilized water + air injection)

RESULTS

Performance comparison of BV, SVE and AIBV for benzene and toluene removal from sandy soils is shown in Figures 2 and 3, respectively. As can be seen in the figures, all of the three above technologies are useful for vadose zone remediation, but at the same time AIBV is more efficient compared to other technologies.

Enumeration of CFU of benzene and toluene-degrading bacteria are shown in Figurers 4 and 5, respectively. In both figures, microbial count is presented as CFU per gram dry weight of sandy soil (CFU/g d.w. of soil). As can be seen, the number of colonies in AIBV test was higher than the number of colonies formed in BV experiment at the same time. Variation of soil water content as percentage by weight is shown in Figure 6. As it can be observed, the humidity of polluted sandy soil in AIBV reactor was decreased by the

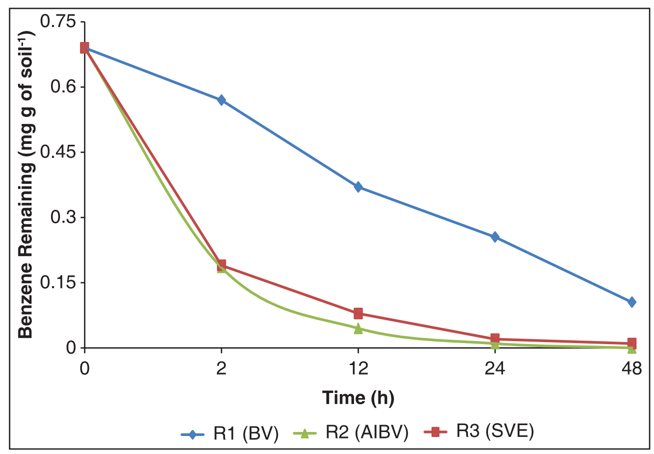

Figure 2: Efficiency of bioventing, soil vapor extraction and air injection bioventing technologies for benzene removal

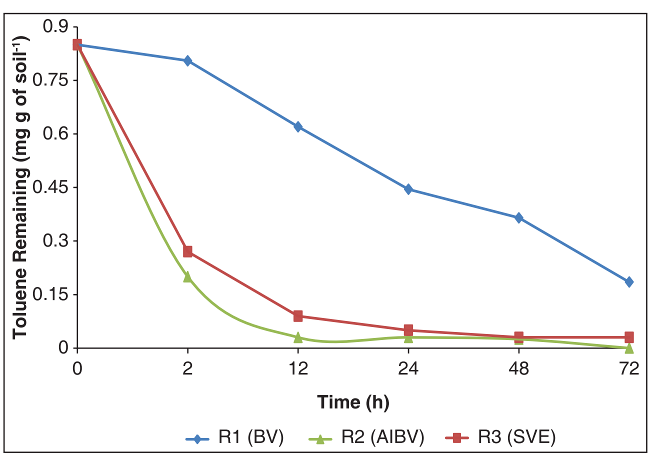

Figure 3: Efficiency of bioventing, soil vapor extraction and air injection bioventing technologies for toluene removal

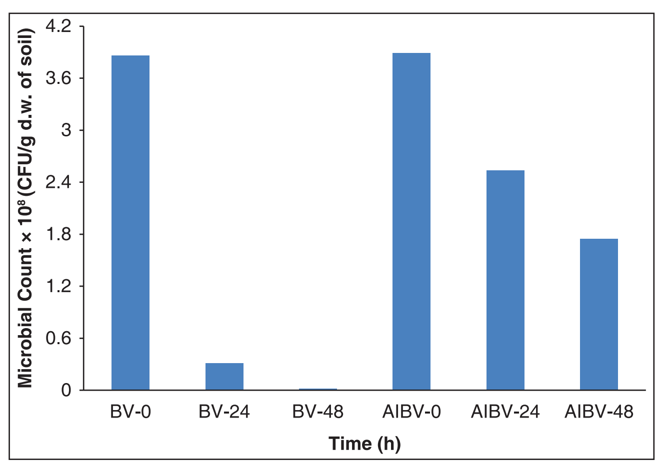

Figure 4: Benzene degrading microbial count during hours of operation

elapse of time, while variation of soil water content in BV reactor is negligible.

Benzene and tolene mass balances on BV, SVE and AIBV reactors are shown in Figure 7. As can be seen in this figure maximum volatilization was occurred in R2 reactors and maximum adsorption of contaminants to sandy soil particles was detected on BV reactors.

It should be noted that all of the presented results are mean values that obtained from two sampling ports (ports 1 and 2).

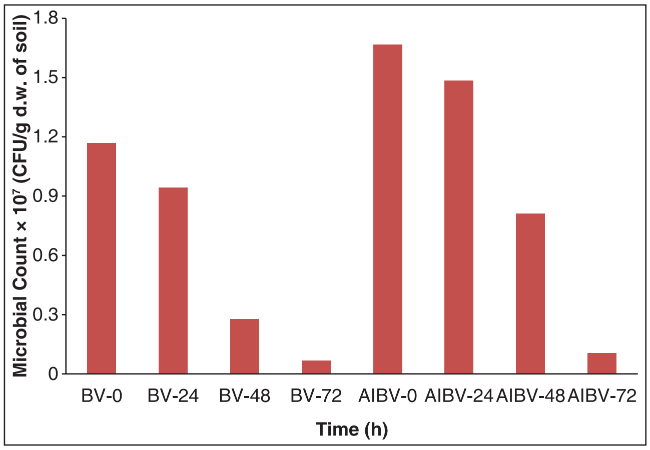

Figure 5: Toluene degrading microbial count during hours of operation

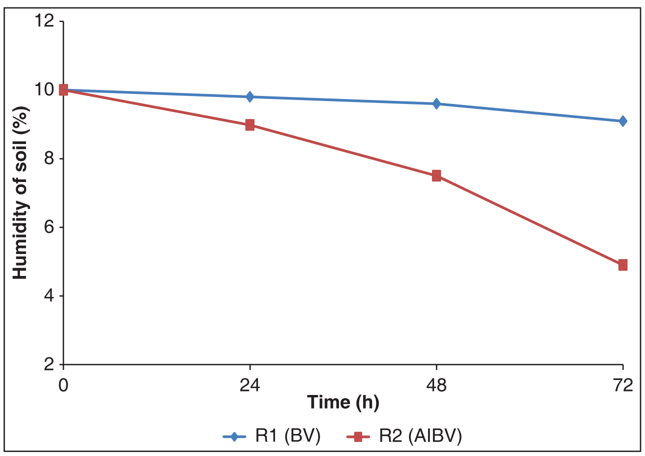

Figure 6: Variation of soil water content during bioventing and air injection bioventing operation

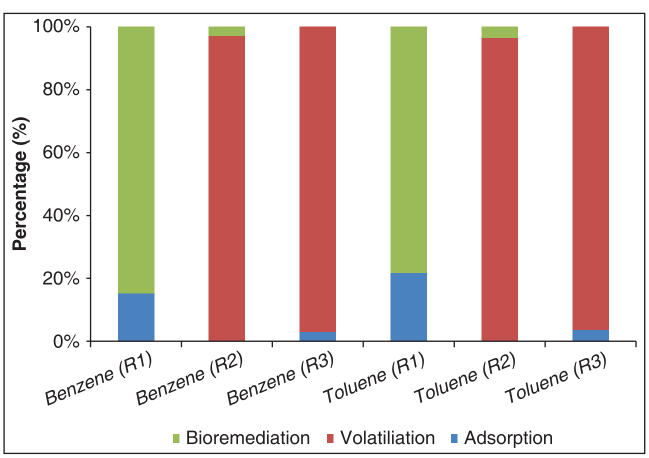

Figure 7: Benzene and toluene mass balances on bioventing, soil vapor extraction and air injection bioventing reactors

DISCUSSION

Efficiency of three remediation technologies

In the present study, three columns in parallel were used as BV (R1), SVE (R3) and AIBV (R2) reactors [Figure 1]. The constant air flow rate of 250 mL/min was injected in R3 and R2 columns for oxygen supply. R1 reactor was used as control for biodegradation without air injection and it can be said that, R3 column was conducted as control for volatilization and has not been inoculated. Figure 2 shows the efficiency of three technologies for benzene removal from contaminated sandy soil. As seen in Figure 2, after a period of 48-h air injection, in BV and SVE reactors, approximately 105 and 10 mg of benzene per kg of soil was remained, respectively. Figure 3 shows that after a 72-h air injection period, 185 and 30 mg of toluene per kg of sandy soil were remained in BV and SVE reactors, respectively.

As can be seen in these figures, benzene and toluene were not detected in AIBV reactors after the distinguished remediation timeframes of these experiments.

These results of toluene removal can be compared to results that presented in Magalhães *et al.*[13] They concluded that approximately 99% of toluene (with initial concentrations of 2 and 14 mg/g of soil) was removed in AIBV reactors after 5 days, but in present study the removal efficiency of toluene via AIBV reactor was >99.5%. The main differences between studies that mentioned above are the soil porosity and flow rate of air injection. So that, remediation in higher flow rates is much more than lower flow rates, and consequently lower time is required for the remediation.[16]

The remediation of soils combining SVE and bioremediation has been studied by Soares *et al.*[1] Their experiments indicated that 170 mg benzene/kg of soil was reduced to 92 mg/kg of soil after 4.2-h air injection with a flow rate of 18 L/h, and then diminished to 37 mg/kg of soil after 646-h bioremediation process. They reported that high level of organic matter (24%) led to incomplete remediation via SVE. It should be noted that in present study the content of organic matter of sandy soil was less than 0.25%.

In other study, remediation efficiency of vapor extraction of sandy soils contaminated with cyclohexane and with an emphasis on influencing factors such as air flow rate, water and natural organic matter content was conducted by Albergaria *et al.*[18] They indicated that increase of soil water content and natural organic matter resulted in negative impacts on remediation process.

The US EPA guideline values for benzene and toluene in vadose zone to protect ground water are 0.03 and 12 mg/kg of soil.[19] In accordance with these guidelines, within a similar timeframe, only AIBV can efficiently protect groundwater.

Monitoring of microbial count

Presence and distribution of active microbial population are responsible for *in situ* bioremediation.[20] More successful *in situ* remediation have a microbial population ranging from 10^7 to 10^8 CFU/g d.w. of soil, and at least 10^5 CFU should be present in the 1 g of soil for BV to be feasible.[21] Figures 4 and 5 shows that at the beginning of the BV experiments, benzene and toluene-degrading microbial counts were 3.86×10^8 and 1.17×10^7 CFU/g d.w. of the soil, respectively. The AIBV experiments were also started in healthy and satisfied condition, because 3.89×10^8 and 1.67×10^7 microbial colony were formed for 1 g of benzene and toluene polluted soil, respectively. The reductions of the enumerated colonies were appeared in next days, especially in BV rectors. It seems lack of venting or oxygen supply led to diminish microbial population.

Furthermore, reductions in cell growth were observed in AIBV as ventilated reactors. However comparison between microbial count in BV and AIBV reactors indicated that reduction of colonies number is not due to lack of oxygen only. Hellekson reported that many factors such as soil moisture, nature of the contaminant, soil structure, soil particle size, and soil permeability can influence on the efficiency of BV system.[12]

Monitoring of soil moisture

As can be observed in Figure 6, variation of soil moisture content in BV reactor is negligible, but in AIBV reactor, soil moisture content reduced to 49% of the initial value after 72-h air injection. The moisture content of the soil plays a very important role in BV experiments.[22] Therefore, it seems that reduction in soil moisture may be led to mass mortality and diminish microbial populations in AIBV [Figures 4 and 5]. Bezerra *et al.* reported that optimum soil water content for BV is 18%wt, or approximately 50% of the soil's field capacity.[23] In the study conducted by Magalhães *et al.*, the initial soil moisture was considered 10% wt,[13] same as present study. According to Bezerra *et al*.[23] although the soil water content is less than18% wt is not optimum for microbial growth, but 1.4×10^6 CFU/g d.w. of the soil was obtained, after 72-h air injection at 4.9% soil moisture [Figure 5].

Benzene and toluene mass balance in three remediation reactors

Mass balance is based on the principle of conservation of contaminants mass on each reactor. As the generation of benzene and toluene in reactors are equal to zero, Equation (1) can be applies to material balance of a pollutant.

$$m_i = m_{vol} + m_{biodeg} + m_{ads} \qquad (1)$$

where, m_i is initial pollutant mass detected into the soil before beginning an experiment, m_{vol} is the pollutant mass removed by volatilization, m_{biodeg} is the pollutant mass removed by bioremediation and, m_{ads} is the pollutant mass that adsorbed to sandy soil particles.

The result shows that almost 97% of benzene and 96.5% of toluene was volatilized after 72 and 48-h air injection during AIBV processes, respectively. In the other hand, maximum mineralization for benzene and toluene was obtained 3 and 3.5%, respectively. These results can be compared to results that presented in Magalhães *et al*.[13] They concluded that after 120-h air injection at a constant air flow rate of 130 mL/min, 92% of toluene (initial concentration of 2 and mg/g of soil) was volatilized and about 8% of toluene with noted initial mass was biodegraded. However, rate of mineralization in our study was lesser than mentioned study,[13] that its can be due to higher air flow rate that used in present study.

CONCLUSION

The remediation tests of BV, SVE and AIBV were performed in sandy soils contaminated with benzene and toluene with similar initial concentration of 1 mg/g of sandy soil. This study highlights that AIBV is very efficient technique than other *in-situ* technologies such as BV and SVE. Furthermore, we conclude that injected air passed through contaminated soil diminish water content of the soil and reduce the microbial population.

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