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"Gheorghe Asachi" Technical University of Iasi, Romania

MASS TRANSFER MODEL OF SOIL VAPOR EXTRACTION UNDER THERMAL TREATMENT FOR REMOVING A VOLATILE CONTAMINANT

Peng Li[1,2], Xiaoyong Liao[2*], Xiulan Yan[2], Dong Ma[2], Xiaoyong Cui[1], Huan Tao[2]**

[1]University of Chinese Academy of Sciences, Beijing 100049, China
[2]Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Science, Beijing 100101, China

Abstract

A series of column experiments were conducted to study volatile contaminant (benzene) removal and process principles using soil vapor extraction (SVE) under different thermal conditions. Compared with the normal SVE method, two thermal methods-steam injection and electrical resistance heating effectively increased the benzene vapor diffusion and removal rates. The removal efficiency of benzene with the electrical resistance heating method was 96.5%, which was significantly higher than that with the steam injection method (90.5%, $p=0.002$) or with the normal SVE method (85.2%, $p=0.001$). In addition, SVE with electrical resistance heating showed a more stable heating effect than SVE with steam injection in homogeneous sandy soil. Based on existing equilibrium SVE models, we established a one-dimensional non-equilibrium mass transfer model for estimating volatile contaminant removal under thermal enhancement. The improved model can accurately reflect benzene content variations using SVE with electrical resistance heating and provides a theoretical basis for preliminary site remediation assessments using SVE under continuous thermal treatment.

Keywords: mass transfer model, soil vapor extraction, thermal treatment, volatile contaminant

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1. Introduction

Soil vapor extraction (SVE), also known as soil venting or vacuum extraction, has proven to be an effective method for remediating unsaturated soils that have been contaminated with volatile organic compounds (VOCs) (DiGiulio and Varadhan, 2001; Lee et al., 2016). In recent years, SVE has been the most commonly used technique for remediating unsaturated soils contaminated with volatile and semivolatile organics. From 1982-2005 in the United States, SVE technology was used in 248 of 977 Superfund sites and accounted for 26% of the total number of remediation sites (USEPA, 2007). As early as 1995, Frank and Barkley (1995) attempted to clear contamination from low permeability subsurface formations using SVE technology and 60% leakage contamination was successfully removed. At the Utah Air Force Base, an aviation engine fuel leak caused 0.4 ha and 15 m depth of soil contamination, with a peak oil concentration of 5000 mg/kg that had decreased to 410 mg/kg using SVE technology (Hinchee et al., 1991). Campagnolo and Akgerman (1995) designed a ventilation system to remove gasoline from saturated soils based on a simplified mechanism analysis and the removal efficiency of this SVE system reached 70%. Mohammad et al. (2014) evaluated the effects of soil water content and air flow rate on SVE and investigated the transition regime between the integration of bioventing and SVE for toluene removal from sandy soils site. In current site

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[*] Author to whom all correspondence should be addressed: e-mail: liaoxy@igsnrr.ac.cn; Phone: +86 10 64889848; Fax: +86 10 64888162
[**] Co-corresponding author: e-mail: cuixy@ucas.ac.cn

remediation, there are limiting factors for efficient removal using SVE, including the slow desorption of contaminants, slow diffusion of contaminants from less permeable layers into areas where vapor exchange is possible, limited volatilities of contaminants at in-situ temperatures and vapor-phase retardation because of adsorption (Heron et al., 1998). Moreover, conventional soil vapor extraction systems have a limited effectiveness for removing semi-volatile chemicals and contaminants from low permeable soil layers (Lenhard et al., 2004). These factors greatly limit the development of SVE. Improving the efficiency of SVE technology using enhancing methods has become an important research focus in environmental remediation.

These enhancing methods include thermal enhancement, microbial enhancement and air sparging. Thermal enhancement, also known as in-situ heating technology, is considered to be an excellent potential SVE enhanced technology (Poppendieck et al., 1999). These technologies include radio frequency heating, hot air injection, steam injection and electrical resistance heating, which significantly shorten remediation period and enable the removal of low- or semi-VOCs that cannot be removed by conventional SVE. Marley (1985) conducted a one-dimensional sand-column ventilation experiment, and found that the removal rate of gasoline vapor was mainly controlled by air flow and saturated vapor pressure at the gasoline–air interface. Poppendieck et al. (1999) observed elevated removal rate and the range of compounds that could be removed from the soil when soil temperature increases to 50-150°C. Park et al. (2005) performed TE-SVE by hot-air injection for removing total hydrocarbons, and found that the 30 d removal rate of total hydrocarbons reached 95%. However, there is little published about process principles and mathematical models of *in-situ* soil heating. Therefore, a series of column experiments were conducted to study contaminant (benzene) removal and process principles using different thermal treatments. The major objectives of this study were to prove the enhanced thermal treatment removal efficiency, to explore the strengthening mechanism of thermal treatment, to establish a one-dimensional non-equilibrium mass transfer model under the state of thermal treatment and verify the accuracy of this model and to explore the possibility of predicting the actual possibility of site remediation using the model.

2. Experimental

2.1. Experimental setup

A plexiglass column (70 cm height; 12 cm diameter) with a resistant coating of organic corrosion was used for the experiments. A schematic diagram of the apparatus is shown in Fig. 1. At the bottom of the column, there was a dense copper wire net to keep the soil at the bottom of the column and distribute the inlet gas uniformly. Under the net, there was an injection port to inject contaminants. At the top lip, there was an extraction port and a port for installing an electrical resistor or a stainless steel pipe with some holes which can inject steam. Four monitoring ports, which were named port 1, port 2, port 3 and port 4, were located at distances of 5, 25, 45 and 65 cm from the bottom of the column, respectively. Each monitoring port included a sampling port and a temperature detection port. WT-1 probe-type digital thermometers (Hongtai Alloy Electric Apparatus Co., LTD, Yancheng, China) were installed in the temperature detection ports. Sampling ports were sealed using silicone mats. Temperature detection ports were sealed using silicone rubber. The fifth sampling port, a valve and adjustable gas flow meter were located in the primary extraction pipeline. A vacuum pump was connected to the pipeline to produce negative pressure and air flow in the soil. The exhaust gas was absorbed using dichloromethane. Two thermal treatment methods were chosen for this experiment: steam injection and electrical resistance heating.

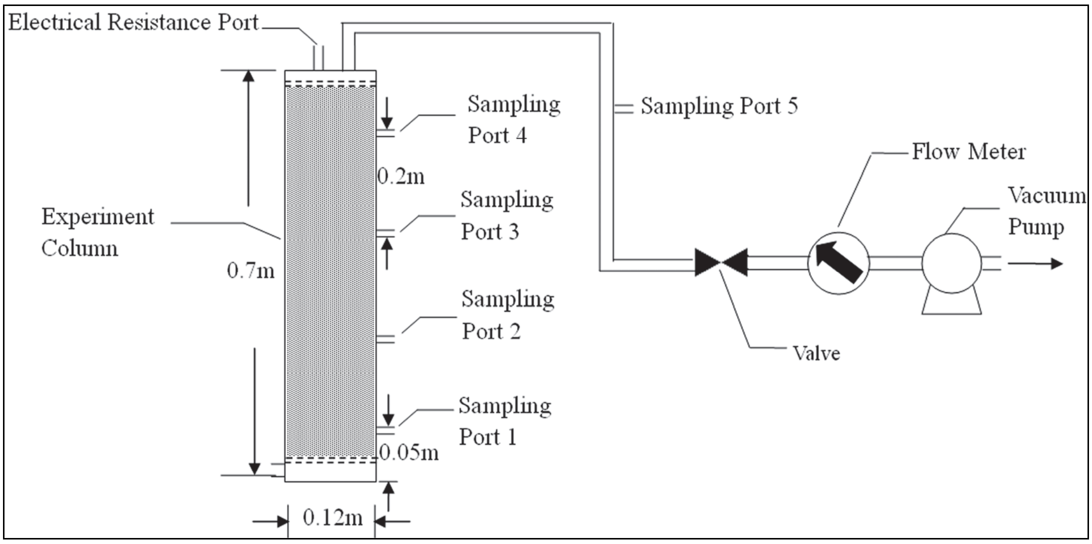

Fig. 1. Experimental setup

Table 1. Major physicochemical properties of tested soil

Soil type	***Sand (%)***	***Silt (%)***	***Clay (%)***	***Porosity (%)***	***Organic matter content (%)***
Sandy soil	80.3	10.5	9.2	33	1.5

Steam was produced using an LYT003 household steam generator (Juneng Energy Technology Co., LTD, Yuyao, China) and was injected into the stainless steel pipe with some holes which was buried in the soil before the experiment.

Electrical resistance heating was achieved by burying the electrical resistor when the soil was carefully and quickly placed into the column. The electrical resistor was an industrial straight heating pipe (50 cm length) that had 0.5 kW of power. A manual temperature control device was installed that could control the temperature of the electrical resistor from 20°C to 120°C.

2.2. Experimental procedure

Benzene, an important raw material in the chemical industry and a pesticide intermediate, was used as the target contaminant. It was analytically pure and was supplied by the Beijing Chemical Plant, China. The SVE experiments were conducted using a soil-column ventilation method. The major physicochemical properties of tested soil are shown in Table 1.

After the top lid was removed, 10 kg of unpolluted tested soil, which was dried and passed through a 40-mesh sieve, was quickly placed into the column. Electrical resistors and a steam injection humidifier were immediately buried in the soil to guarantee uniform heating. Then the top lid was secured to the column with screws to prevent any VOCs leakage to the atmosphere. An initial benzene volume of 100 mL was injected into the bottom of the column and the contact proceeded for 12 h. Then, a soil sample from port 1 and gas samples from ports 1-5 were collected for contaminant analysis. Three treatments were used: (1) normal SVE, referred to as CK; (2) SVE with steam injection, referred to as H-S; and (3) SVE with electrical resistance heating, referred to as H-R. All other experimental conditions were kept the same. The temperature of heating was 50°C and the extraction flow was 3 L/min. The extraction time was 180 h. Gas samples from ports 1-5 were collected, and the temperatures at ports 1-4 were recorded every half hour in the first 12 hours. In the remaining experimental time, gas samples from ports 1-5 were collected and temperatures from ports 1-4 were recorded every 4 hours. The soil from port 1 was collected after the experiment.

2.3. Chromatography

The gas samples were directly injected using a manual syringe (SGE Analytical Science). The soil samples were freeze-dried, and a portion of the samples (2g) were weighed and mixed with dichloromethane (10mL). The mixture was ultrasonicated (1h) and centrifuged. The supernatant (2mL) was collected and added to a silica gel extraction column (10g of 200-300 mesh silica, with anhydrous sodium sulfate placed on top of the silica) and eluted with n-hexane/dichloromethane (1:1 v/v). The purified extract was condensed to approximately 0.5mL using a rotary evaporator (RE-52AA, Shanghai) and diluted to 1mL with n-hexane for analysis. Benzene was analyzed by gas chromatography (Agilent 7890A) with an HP-5MS capillary column (30m×0.25mm) under the following conditions: an injector temperature of 260°C, detector temperature of 230°C, helium as the carrier gas, an inlet pressure of 0.03MPa, a linear velocity of 37cm/s, an inlet temperature of 50°C, a start temperature of 40°C, which was increased to 45°C at a rate of 2°C/min and then increased to 50°C at a rate of 4°C/min, which was maintained for 1 min until the elution of benzene was complete.

2.4. Mathematical model development

During the SVE process, it is assumed that soil vapor is the key focus. Based on the law of conservation of mass, the mass-transport equation for soil vapor can be described by (Fischer et al., 1996) (Eq. 1):

$$\frac{\partial}{\partial x_i}\left(\theta_g D_{ij}\frac{\partial C_g}{\partial x_j}\right)-\upsilon_g\frac{\partial\left(\theta_g C_g\right)}{\partial x_i}=\theta_g\frac{\partial C_g}{\partial t}+\theta_l\frac{\partial C_l}{\partial t}+\rho_s\frac{\partial C_s}{\partial t} \tag{1}$$

in which: D: diffusion coefficient (m^2/s); C_g: concentration of pollutants in the gas phase (kg/m^3); C_l: concentration of pollutants in the liquid phase (kg/m^3); C_s: concentration of pollutants in the solid phase (kg/m^3); θ_g: gas volume fraction; θ_l: volumetric moisture content; ρ_s: density (kg/m^3); υ_g: flow rate (m/s).

Because the diameter of column is small, the soil vapor movement can approximately be considered as a one-dimensional steady-state flow. So Eq. (1) can be described as Eq. (2):

$$\theta_g D_e\frac{\partial^2 C_g}{\partial x^2}-\theta_g\upsilon_g\frac{\partial C_g}{\partial x}=\theta_g\frac{\partial C_g}{\partial t}+\theta_l\frac{\partial C_l}{\partial t}+\rho_s\frac{\partial C_s}{\partial t} \tag{2}$$

In the process of normal SVE, existing models can be classified into two types: equilibrium and non-equilibrium. Equilibrium models assume that the mass transfer processes of contaminants between gas, liquid and solid states are balanced. Contaminants present in the gas, liquid and solid states are considered as gaseous, dissolved and adsorbed, respectively. We assumed that there is a continuous liquid film along the interface of soil particles.

Therefore, the mass transport between gas and solid states should go through the liquid state (Zaidel and Zazovsky, 1999). The total mass transport processes can then be divided into gas-liquid and liquid-solid mass transfers.

Non-equilibrium models are based on the assumption that when the gas and liquid phases are transferred by a liquid boundary, the power of transfer is the concentration gradient between the average concentration of the liquid phase and the equilibrium concentration of the gas-liquid interface. Mass-transfer processes between gases and liquids are approximated by a first-order kinetic equation and mass-transfer processes between solids and liquids act according to equilibrium-transfer. So the mass balance equation for a liquid can be described by Ho (1998) (Eq. 3):

$$\theta_l \frac{\partial C_l}{\partial t} = \theta_g \lambda_{gl}\left(C_g - HC_l\right) - \theta_l \lambda_{gl}\left(C_l - \frac{C_s}{K_d}\right) \tag{3}$$

in which: H: Henry coefficient; K_d: adsorption equilibrium constant (m^3/kg); λ_{gl}: gas-liquid mass transfer coefficient (s^{-1}).

The corresponding mass balance equation for solids can be described as Eq. (4):

$$\rho_s \frac{\partial C_s}{\partial t} = \theta_l \lambda_{ls}\left(C_l - \frac{C_s}{K_d}\right) \tag{4}$$

where λ_{ls} is the liquid-solid mass transfer coefficient (s^{-1}).

We can obtain the model (Eqs. 5 and 6), which is based on the concentration of gas contaminant by substituting Eqs. (3 and 4) into Eq. (2). This model can simulate the process of normal SVE, which is an extraction process under constant temperature.

$$L\left(C_g\right) = \theta_g D_e \frac{\partial^2 C_g}{\partial x^2} - \theta_g \upsilon_g \frac{\partial C_g}{\partial x} \tag{5}$$

$$\theta_l R \frac{\partial C_l}{\partial t} - \theta_g \lambda_{gl}\left(C_g - HC_l\right) = 0 \tag{6}$$

where R is the retardation factor (Eq. 7):

$$R = 1 + \frac{\theta_l}{\theta_g H} + \frac{\rho_s K_d}{\theta_g H} \tag{7}$$

If the temperature changes, the Henry coefficient will change simultaneously (García-Herruzo et al., 2000), (Eq. 8):

$$H = H_{ci} RT / V \tag{8}$$

where H_{ci} represents the gas-liquid distribution coefficient.

Thus, Eq. (5) can be transformed into Eq. (9):

$$\begin{aligned} L\left(C_g\right) &= \theta_g \frac{\partial C_g}{\partial t} + \theta_l R \frac{\partial C_l}{\partial t} \\ &= \theta_g \frac{\partial C_g}{\partial t} + \theta_l \frac{\partial C_l}{\partial t}\left[1 + \frac{V\left(\theta_l + \rho_s K_d\right)}{\theta_g H_{ci} RT}\right] \end{aligned} \tag{9}$$

We attempted to simulate the SVE process under thermal treatment, which is an extraction process under varying temperatures using Eq. (9).

3. Results and discussion

3.1. Soil temperature variations

Fig. 2 presents temperature variation curves over time (12h) at the different sampling port heights. Under normal SVE, soil temperatures at different heights changed with room temperature. When using electrical resistance heating, soil temperatures at different heights stabilized at approximately 50°C, which was the temperature of the resistor. Temperature variations were small. The temperature of lowest position (end of electrical resistance) was lower than the highest position (top of electrical resistance). When using steam injection, soil temperatures at different heights all decreased slowly. At final, the temperature had decreased to approximately 45°C. The heating created instability. Because the steam was injected at the bottom of the column, soil at higher positions had lower temperatures.

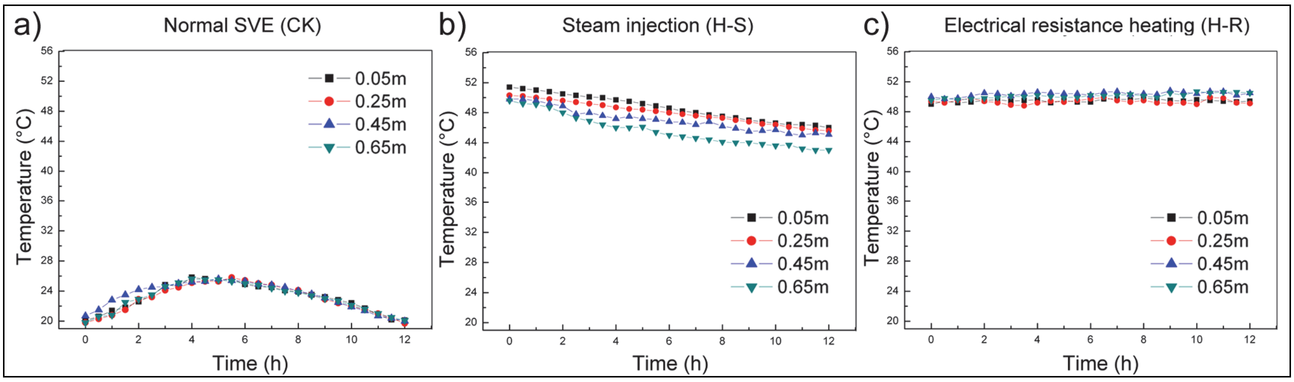

Fig. 2. Soil temperature variation curves under experimental conditions a), b), and c)

Poppendieck et al. (1999) conducted experiments on thermal enhancement SVE using electrical resistance heating and four different setting temperatures (from 50°C to 150°C) were consistent with the measured temperatures. This indicated that the effect of electrical resistance heating was stable. And their result is consistent with our study. Di et al. (2002) developed an empirical model to simulate the removal of VOCs from soils via steam injection and in their experiment, to ensure the temperature stability, the volume of steam injection was increased. It also indicated that the soil temperature will decrease using steam injection.

It is likely that because of the extraction, the steam's temperature will decrease gradually after being injected into the column. Steam does not thermally exchange well with soil, creating temperature instability. From this result, it is apparent that although electrical resistance heating and steam injection can raise soil temperatures, electrical resistance heating has a more stable heating effect than steam injection.

3.2. Benzene diffusion

Fig. 3 presents gaseous benzene content variation curves over time (180 h) under different experimental conditions. It is apparent that trends in benzene content variations at different sample ports are similar under all SVE conditions, but the actual benzene contents are significantly different. During the first 12h of the normal SVE experiment, the benzene content at port 1 decreased dramatically, but the benzene content at ports 2–4 increased early and then declined. Because benzene was injected at the bottom of the column, the benzene content at port 1 decreased constantly. This also shows that the removal of benzene by SVE is a dynamic process. During the process, the benzene constantly volatilizes and forms a vapor group with high benzene concentrations. Then, the vapor group is forced by extraneous air from the bottom to the top of the column. From diffusion of the vapor group, the concentrations at ports 2–4 peaked and then decreased constantly as the vapor group moved up the column.

When using the two thermal treatments, the benzene content at ports 1 and 2, which were at the bottom of the column, decreased dramatically during the first 12h of the experiment. The decrease was largest when using electrical resistance heating. When using electrical resistance heating, the benzene concentrations at ports 3 and 4 also decreased during the first 12h of the experiment. When using steam injection and normal SVE, the benzene concentrations at port 3 decreased slightly, but increased at port 4. During the remainder of the experiments, the concentrations at all sample ports decreased. When using electrical resistance heating, the benzene concentrations at each port were lowest compared with steam injection and normal SVE, which had the highest residual benzene concentrations.

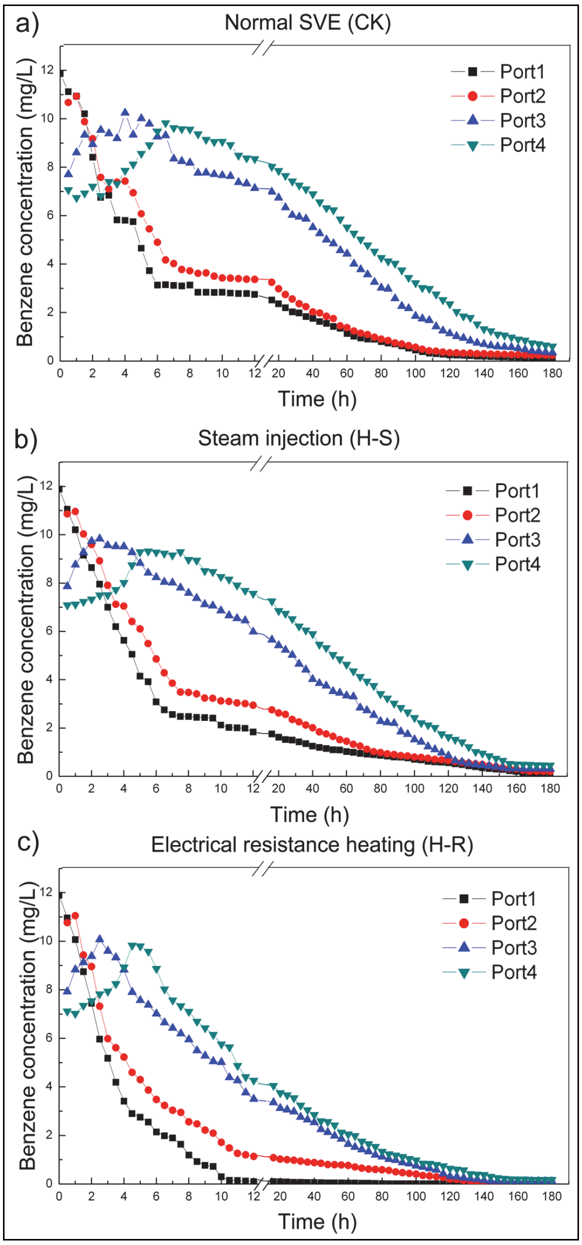

Fig. 3. Gaseous benzene content variation curves under experimental conditions a), b), and c)

Comparing the height (determined based on port location) of peak benzene concentrations per hour under the three conditions, it is evident that when using the two thermal treatments, peak concentrations reached each subsequent port of the column more quickly than under normal SVE. When using electrical resistance heating, the peak concentration reached a height of 0.25m (port 2) after 1h. At a height of 0.65m (port 4), peak benzene concentrations were reached after h. Under normal SVE, the height of 0.25m was reached after 2h and the height of 0.65m was reached after 6h. It is clear that using thermal treatments increased the diffusion rate of benzene.

3.3. Benzene variations in extracted gas and soil

Fig. 4 presents benzene content variation curves in the extracted gas from port 5 over time (180h) under different experimental conditions. When using electrical resistance heating, the concentration of benzene in the extracted gas was higher than under normal SVE during the first 120h. Steam injection produced higher benzene concentrations than normal SVE for the first 84h. Compared with normal SVE, SVE under thermal treatment can remove more volatile contaminants during the early stages of the experiments. At the final of the experiments, the benzene concentration in the extracted gas from normal SVE was higher than those from thermal enhancement SVE. This is because, using thermal treatment, most contaminants in the column were removed in the early stages of the experiments. In these three experiments, the amount of benzene injected was the same (87.86g).

Using an integration method, we have determined the benzene removal efficiency under different conditions. Under electrical resistance heating, 84.5g of benzene was removed and the removal efficiency was 96.5%, much higher than under the other two conditions. Under steam injection, 78.38g was removed and removal efficiency was 89.21%, while normal SVE removed 75.42g resulting in an efficiency of 85.85%. SVE under thermal treatment can increase the removal efficiency of volatile contaminants from soil.

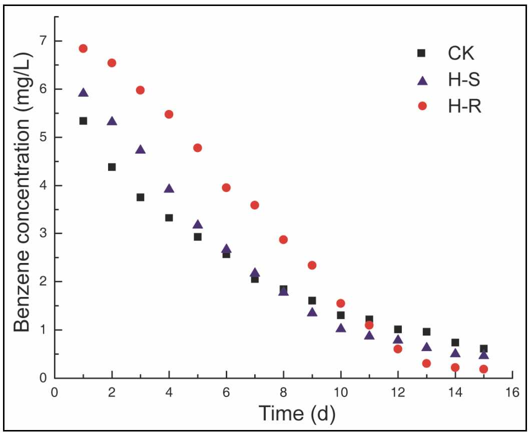

Fig. 4. Gaseous benzene content variation curves under different experimental conditions

Fig. 5 presents benzene content variations in soil (port 1) under different experimental conditions. SVE under thermal treatment increased the removal of volatile contaminants in soil compared with normal SVE. Under electrical resistance heating, the removal efficiency was 96.5%, which was significantly higher compared with the efficiencies achieved under steam injection or normal SVE (removal efficiencies 90.4 and 85.2%, respectively). The removal efficiencies calculated based on concentration variations in soils were similar to the values calculated using concentration variations in the extracted gas.

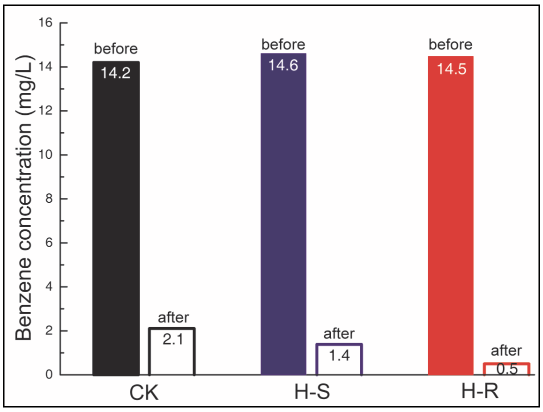

Fig. 5. Benzene content in soil before and after extraction under different experimental conditions

Poppendieck et al. (1999) conducted experiments on SVE under electrical resistance heating for removing C_{13} hydrocarbons. Their study indicated that when the temperature was 100°C, the C_{13} removal efficiency was 90% over 24h. Van Zutphen et al. (1998) conducted experiments on the removal of trichloroethylene using electrical resistance heating and the trichloroethylene removal efficiency was 99.8% after 45 days when the temperature of the soil was 33°C. Mohamed et al. (2007) designed an experiment for removing benzene using SVE under steam injection and the removal efficiency increased by 9% compared with normal SVE.

Based on previous study, this paper compares the enhanced efficiency of different thermal enhancement methods (electrical resistance heating and stream injecting) under the same experimental condition. It proves that thermal treatment achieve the purpose of enhancement for SVE and electrical resistance heating is more stable and has better enhanced efficiency than stream injecting. Electrical resistance heating improves the efficiency of SVE by increasing the stability of heating effect and reducing the disturbance to soil. In the normal SVE, however, steam may increase the moisture and reduce the porosity of the soil, which thereby affects the SVE efficiency. Indeed, Albergaria et al. (2008) proved that the moisture of soil in one-dimensional column is higher, the time of removing cyclohexylamine by SVE is longer and the efficiency is lower.

Although our results showed that the electrical resistance method had better efficiency than the normal SVE and steam method, we have to keep in mind that our data was based on homogeneous sand as the tested soil. For more heterogeneous conditions the electrical resistance method or the steam method may suffer from obstruction in heat transfer, whereas the normal SVE has been proved more disadvantageous for heterogeneous soil (USEPA, 2007).

3.4. Removal of contaminants and mathematical model

Some information on mathematical and mass transfer models of normal SVE exists. Campagnolo and Akgerman (1995) developed a mathematical model to simulate the removal of VOCs using normal SVE. More recently, Kneafsey and Hunt (2004) established a mass transfer model to simulate the removal of the non-aqueous phase during normal SVE. A numerical model which was incorporated with the first-order mass transfer kinetics developed for the design and operation of air sparging and soil vapor extractions systems was used to simulate two field case studies (Rahbeh and Mohtar, 2007). During the process of normal SVE, existing models can be classified into two types: equilibrium and non-equilibrium. We attempted to simulate the process of SVE under thermal treatment using Eq. (8) based on existing models and to verify the accuracy of the model. The parameters used are presented in Table 2. Using these parameters, the concentration of benzene in the gas phase could be calculated using Eq. (8).

Fig. 6 presents calculated and actual values of benzene content in gas samples from port 1 over time. The calculated values were determined using Eq. 5 and 8 and the actual values were determined under different experimental conditions. Comparing the calculated and actual values under normal SVE indicates that the non-equilibrium mass transfer model (Eq. 5) can accurately reflect the benzene content variation under normal SVE. Comparing the calculated and actual values under thermal treatment indicated that deviations between the calculated and actual values were large under steam injection. The improved model (Eq. 8) could not accurately reflect benzene content variations under these conditions. However, when using electrical resistance heating, the calculated value was in good agreement with the actual value during most of the experiment, particularly late in the experiment. The improved model (Eq. 8) can accurately reflect the benzene content variation using electrical resistance heating, but cannot be used to model steam injection. Two main reasons for this are that the soil temperature cannot be stabilized at a set value and the moisture content is variable during steam injection, but Eq. 8 does not fully consider the effect of soil moisture on temperature. For SVE under electrical resistance heating, this model demonstrates the relationship between enhancement and repair effects and provides a theoretical basis for the preliminary assessment of site remediation using SVE under thermal treatment.

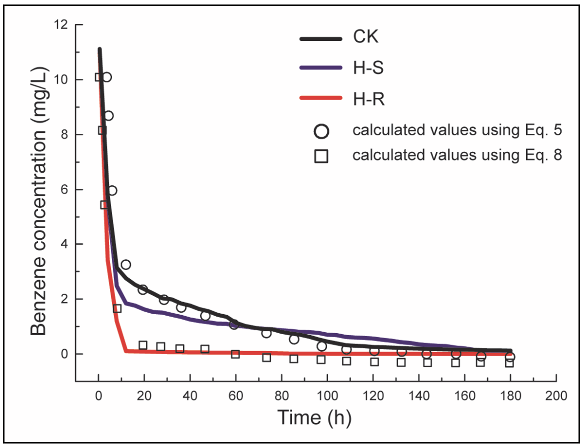

Fig. 6. Calculated and actual values of benzene content under different experimental conditions

It is noted that the numerical model we established in this study is also valid for other aqueous contaminants that have similar physicochemical properties as benzene and other soil types, but with the limitation of temperature which has to be under the boiling point of contaminant. Althought whether this model can be applied under a larger scale needs further validation, we believe that this model can facilitate not only SVE-related studies, but also studies on site assessment and remediation technology screening.

4. Conclusions

SVE with electrical resistance heating achieves higher benzene vapor diffusion and removal rates than the normal SVE or SVE with steam injection. In addition, electrical resistance heating shows a more stable heating effect compared with steam injection in homogeneous sandy soil.

Table 2. Model parameters

Soil properties		
Gas volume fraction θ_g	0.2148	Measured in this study
Average volumetric moisture content θ_l	0.1843 (CK) 0.33454 (H-S) 0.1534 (H-R)	Measured in this study
Benzene properties		
Molar mass M	78.11 g/mol	(Høier et al., 2007)
Density ρ_s	0.8786 kg/m^3	(Li et al., 2008)
Henry coefficient H	0.0018	(Li et al., 2008)
Adsorption equilibrium constant K_d	7.9672×10^{-4} m^3/kg	(Marley, 1985)
Effective absorption coefficient D_e	0.3568×10^{-4} m^2/s	(Marley, 1985)
Gas-liquid mass transfer coefficient λ_{gl}	1.3×10^{-4} s^{-1}	(Halmemies et al., 2003)
Gas-liquid distribution coefficient H_{ci}	4.25	(Halmemies et al., 2003)

A one-dimensional non-equilibrium mass transfer model was proposed, which is capable of accurately reflecting benzene content variations under the electrical resistance heating condition. However, this improved model cannot be applied to the steam injection condition due to the instable temperature and the variable moisture content of soil.

Acknowledgments

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