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Laboratory Evaluation of Low-Temperature Thermally Enhanced Soil Vapor Extraction

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ABSTRACT: The goal of this study is to understand the conditions in which heat collected from soil-borehole thermal energy storage (SBTES) systems can be used for thermally enhanced vapor extraction. Although thermal enhancement of soil-vapor extraction (SVE) has been investigated in several studies, they typically involved temperatures in the range of 50-400 °C. The soil in SBTES systems typically only reaches temperatures of 40-60 °C, which is not significant but is from a renewable source and can be applied for a long duration at low cost. The approach used in this study involves column tests in which a vacuum is used to draw air through an unsaturated soil layer that has a uniform initial diesel distribution. Three tests were performed in which the soil was heated to 20, 40 and 60 °C and a vacuum was imposed to cause upward air flow through the soil column. The amount of diesel removed from the soil increased with temperature. Although up to 90% of the pore water was removed in the 60 °C test, only a small fraction of the free-phase diesel was removed due to its low volatility. Nonetheless, the results indicate that combining SBTES systems with remediation strategies for volatile NAPLs may have a positive long-term impact on the reuse of contaminated sites.

INTRODUCTION

Many cities are encouraging reurbanization to stimulate economic revitalization in inner-city areas. Unfortunately, the subsurface in urban areas is often contaminated with non-aqueous phase liquids (NAPLs) from historic industrial activities or leaking underground storage tanks. Although redevelopment of such "brownfield" sites poses a significant economic incentive for investors, in many cases the contaminant concentrations are high enough that an active remediation approach is needed to eliminate threats to groundwater. This is the case for sites contaminated with relatively light, organic NAPLs that are found in gasoline and diesel fuel that are present in the vadose zone or atop the water table. For sites with a deep water table, the most common remediation approaches are bioremediation and soil-vapor extraction (SVE) (Rathfelder et al. 1995). Bioremediation is usually less expensive, but can take years to complete. Theoretically, both SVE process and bioremediation could be enhanced with a moderate increase in ground temperature to 40-60 °C. Unfortunately, thermal processes are often avoided due to prohibitive costs, and when they are used temperature magnitudes of several hundred °C are employed to facilitate vaporization and quick removal, in which case biological activity is eliminated. An opportunity to reduce the cost of supplying heat

to the subsurface may be to combine the soil remediation process with low temperature heat collected from Soil-Borehole Thermal Energy Storage (SBTES) systems.

Soil-borehole thermal energy storage (SBTES) systems are an approach to provide efficient renewable resource-based thermal energy to heat buildings (Sibbit et al. 2007; Pavlov and Oleson 2011; Chapuis and Bernier 2009; McCartney et al. 2013). They function in a similar way to conventional ground-source heat pump (GSHP) systems, where a fluid is circulated within a closed-loop pipe network installed in vertical boreholes to shed or absorb heat from the surrounding subsurface. Different from conventional GSHP systems, SBTES systems are configured to store thermal energy collected from solar thermal panels during the summer, and discharge the heat to buildings during the winter. The boreholes are spaced closer together in an SBTES system than in a conventional system, with spacings on the order of 1.5 to 2 m (Başer and McCartney 2015). The temperature of the ground within the borehole array increases from its ambient temperature (approximately 10-20 °C) to 40-60 °C during heat injection. SBTES systems are a convenient alternative to other energy storage systems as they are relatively inexpensive, involve storage of renewable energy (solar thermal energy), and are space efficient as they are underground. Most SBTES systems involve direct circulation of fluid through the closed-loop boreholes during heat injection and extraction, without the use of a heat pump.

A physical modeling approach is used in this study to characterize the coupled flow of heat, water and LNAPL in unsaturated soil layers, leading to a better understanding of their effects on contaminant removal in thermally enhanced soil vapor extraction. Specifically, an insulated, cylindrical column is filled with an unsaturated soil layer that has been uniformly mixed with diesel hydrocarbon before compaction, underlain by a vapor diffusion layer. After heating the soil column from the boundary, a vacuum is applied to the top of the soil column to induce soil vapor extraction at an approximately constant air flow rate. Dielectric sensors are used to measure the dielectric permittivity and temperature of the soil at different depths in the unsaturated soil layer, which can be used to characterize the flow processes for heat and water. Gas sampling needles inserted into the soil layer are used to measure changes in the vapor-phase LNAPL concentration as a function of space and time using gas chromatography. The vapor-phase LNAPL concentration in the effluent gas extracted from the top of the soil column is also tracked as a function of time. The instrumentation incorporated into the system is sufficient to characterize the flow process and the contaminant removal process under non isothermal conditions. Three tests were performed in which a soil column was heated to temperatures of 20, 40 and 60 °C and a vacuum was imposed to cause upward air flow through the soil column.

BACKGROUND

Soil vapor extraction (SVE) is a popular, effective and economical in-situ remediation method for vadose zone contamination (Marley and Hoag 1984). SVE creates an advective airflow through the contaminated porous media inducing volatilization and vapor transport of the contaminant. A SVE system consists of a vadose zone extraction well (or wells) connected to a vacuum pump at the surface. The pressure differential created by the vacuum pump induces horizontal airflow through the soil, inducing transfer of the contaminant to the air stream. The volatilized contaminant is then

extracted through the well and either released into the atmosphere or treated depending on local environmental regulations. The amount of contaminant transferred to the air stream is related to the contaminants vapor pressure at the given ground temperature.

The subsurface temperature at contaminated site is typically equal to the mean annual air temperature, ranging from 10-20 °C. Laboratory column tests have shown that performing SVE at elevated ground temperatures ranging from 50-400 °C increases contaminant removal from the ground (Oma and Buelt 1988; Lingineni and Dhir 1992; Mesbah-Ul Islam and Kaluarachchi 1995; Heron et al. 1998; Poppendieck et al. 1999a; Park et al. 2005). These observations have been confirmed in instrumented field tests (Poppendieck et al. 1999b; Park et al. 2005). The vapor pressures of LNAPLs increase with temperature (Wilhoit and Zwolinski 1971), and an increased vapor pressure signifies that more of the LNAPL will be in the vapor phase. Advective airflow through the soil generated by a soil vapor extraction system will then carry the water and vapor-phase LNAPL through the soil layer to be extracted at the surface. The saturated vapor pressure for LNAPL in air also increases with temperature, so the amount of LNAPL carried with the air also increases (Wilhoit and Zwolinski 1971). An increase in temperature may also lead to a decrease in the degree of water saturation and a corresponding increase in the air relative permeability. This will allow more air to flow through the soil, increasing the amount of LNAPL extracted. Some of the most common heating processes in the field are electrical resistance, radio frequency heating, and hot-air injection, all of which require a large amount of energy and high cost. Accordingly, the goal of this study is to quantify the efficiency of LNAPL removal for a moderate increase in ground temperature to only 40-60 °C encountered in more cost-efficient SBTES systems.

MATERIALS

Bonny silt was used in this experiment as the unsaturated soil layer. The specific gravity of the silt is 2.65. The liquid limit and plastic limit are 24 and 21. Based on these values and the fact that the soil has nearly 84% fines, the soil is classified ML according to the Unified Soil Classification Scheme (USCS). In this study, the Bonny silt was compacted to a void ratio of 0.89 (porosity of 0.47) and has an estimated saturated hydraulic conductivity to water of 1.3×10^{-6} m/s. This corresponds to an intrinsic permeability of 1.18×10^{-9} cm². The fluids used in this study were tap water and commercially-available diesel fuel. Diesel was chosen to demonstrate this remediation approach because it is made up of semi-volatile hydrocarbons so that only a small fraction of the contaminant would be lost while mixing the contaminant with the soil. However, higher volatility materials are likely more suitable for practical use in the field.

EXPERIMENTAL SETUP

The experiment was conducted in a cylindrical acrylic container having an inner diameter of 228.6 mm and a height of 609.6 mm. A cross-section elevation view of the soil container for a typical setup is shown in Figure 1. The cylinder is enclosed on the top

and bottom with large endcaps that are fitted to the cylinder with “O”-rings to ensure an airtight seal. The endcaps each have ports to which plumbing fittings can be connected. In the case of this experiment, the port in the bottom cap was used to supply air at atmospheric pressure to the bottom of the soil layer within the container, and the port on the top cap was used to supply vacuum to withdraw the contaminant from the soil using soil vapor extraction (SVE). Five dielectric sensors are placed throughout the center of soil column to record the variations of temperature and dielectric permittivity of the soil throughout the experiment. Due to a defective sensor, dielectric permittivity was only recorded in four locations. There are four vapor sampling ports. One of the sampling ports was connected to the effluent line at the top of the container, and three were placed into the side-wall of the container at heights of 60, 180, and 300 mm above the base of the soil. The vapor sampling port on the effluent line is used to measure the vapor phase total petroleum hydrocarbon (TPH) of the effluent gas and the three vapor sampling ports along the height of the soil column are used to measure the vapor-phase TPH concentration of the soil gas in the pore space throughout the experiment. The vapor-phase TPH concentration of the gas samples were determined using a Gas Chromatographer (GC) equipped with a flame ionization detector (FID). In the experiments involving elevated temperatures, heating tape wrapped around the outside of the cylinder was used to heat the soil to the desired temperature.

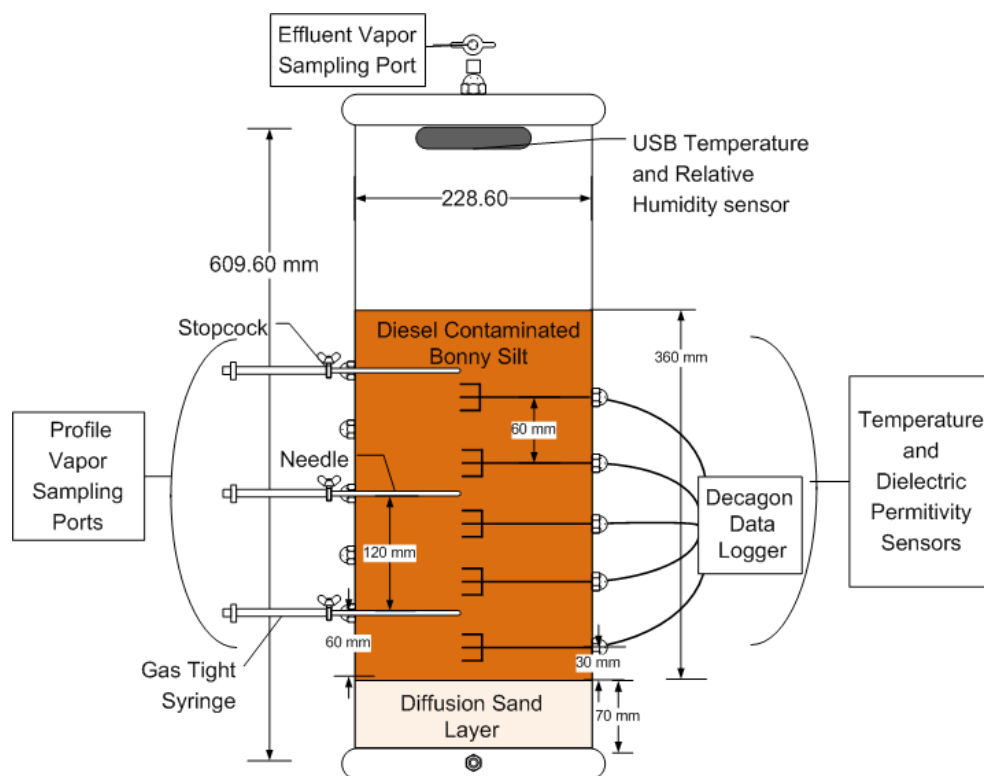


Figure 1: Schematic of the experimental setup showing instrumentation plan

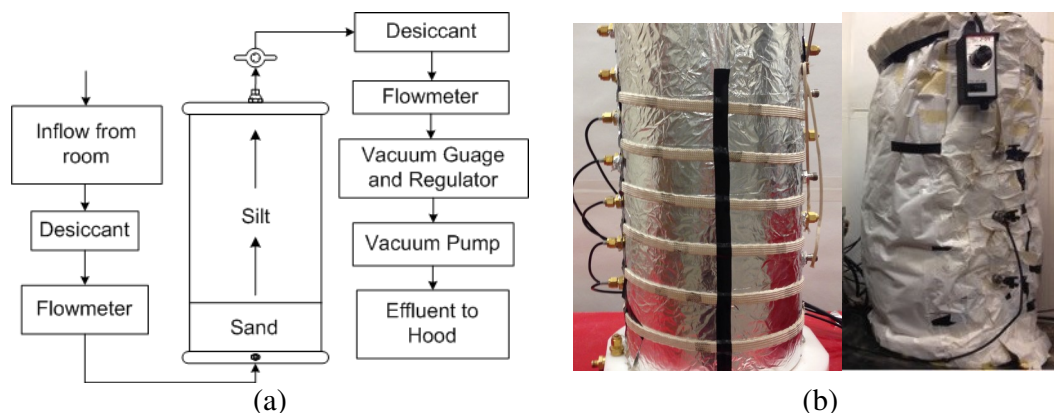


Figure 2: Overall experimental setup: (a) Schematic; (b) Picture

An overall schematic of the system is shown in Figure 2(a). The soil was heated using heating tape wrapped around the outside of the cylinder, as shown in Figure 2(b). Before placement of the tape, the cell was wrapped in aluminum foil. A temperature sensor was placed outside of the cell next to the heating tape to record the temperature of the heating tape. The whole cylinder was encased in fiberglass insulation to decrease the heat loss. The heating tape was manufactured by HTS/Amptek Company. The heating tape was connected to a variable voltage meter which was then plugged into an automatic timer. In all three experiments, a vacuum was applied to the top of the soil column in order to simulate the process of soil vapor extraction. The air pressure at the base of the soil column was vented to atmosphere, which leads to an upward flow of air through the soil column. Flow meters are connected to the port at the bottom of the soil column (inlet) and between the top of the column and the vacuum pump (outlet). The vacuum pump exhaust is vented to a hood. The flow meters used in this experiment were Riteflow flowmeters manufactured by Bel-Art. For air at 1 atm and a temperature of 20 °C, this flowmeter can measure air flow from 28 ml/min to 1249 ml/min. The flow tube for the inlet flow meter stopped working at the beginning of the third test so the inlet flow rate data is unavailable for that test.

Laboratory gas drying units filled with indicating Drierite (desiccant) were placed in line between the effluent vapor sampling port and the flowmeter to collect the moisture (diesel/water) that was removed from the column with the air flow. For the first test, the drying unit was not used because significant moisture removal was not expected under room temperature conditions. For the second test one drying unit was used. For the third test, two units were used in sequence due to the large quantity of water that was expected to be removed based on the observations from the second test. Unique to the third test, a drying unit was also placed at the air inlet port on the bottom cap. This was in an attempt to eliminate any water entering the column in the form of moisture in the laboratory air. As mentioned, there are four vapor sampling ports in the experiment. One effluent port installed immediately after the outlet in top endcap and three ports installed along the side of the cylinder. After the soil column has been compacted, a needle is inserted into the soil from the outside of the cylinder via an airtight fitting so that the tip of the needle is placed at the center of the soil column. The part of the needle that remains outside the cell is connected to a stopcock via a luerlock connection. The stopcock is closed when the port is not being used for sampling. Soil gas samples were extracted using 2.5 mL

gas-tight syringes and then manually injected into a Gas Chromatographer to determine vapor-phase TPH concentration.

The dielectric permittivity and temperature of the soil were measured using 5TM dielectric sensors manufactured by Decagon Devices of Pullman, WA. The sensors were placed at a uniform spacing of 60 mm at different depths along the centerline of the soil column. The sensors measure temperature by an onboard thermistor that is mounted within the probe body. The sensors measure the dielectric constant of the soil using capacitance/frequency domain technology. The dielectric permittivity data was corrected to account for the impact of temperature on the sensor reading by subtracting an error of 0.01 per °C from the measured dielectric permittivity (Iezzoni and McCartney 2016).

PROCEDURES

The silt was moisture conditioned to the targeted gravimetric water content, before the soil was placed into the cylinder. The target gravimetric water content was 16% for Test 1 but was 13.5% for Tests 2 and 3 to provide a lower initial degree of saturation. An increase in temperature causes an increase in water vapor in both the soil pore space gas and the effluent gas. Samples with a high water vapor content can create problems with the Gas Chromatographer (GC). The moisture conditioning was done by spreading the soil out in a thin layer on a flat surface, spraying a fine mist of water onto the soil using a pressurized spray bottle, then mixing it by hand. This was repeated until the required mass of water was added to reach the target value. The moisture-conditioned soil was placed loosely in an airtight container and allowed to sit for 24 hours allowing the water content to homogenize. Samples were obtained to determine the average initial gravimetric water content using oven drying. Immediately before compaction, the soil was mixed with diesel fuel to achieve a concentration of 20,000 mg/kg of dry soil. Using the gravimetric water content, diesel concentration and a target dry density of 1400 kg/m³, the total density was calculated as follows:

(1)

where M_s is the mass of the solids, M_w is the mass of the water, M_d is the mass of the diesel fuel, and V_T is the volume of the container.

As mentioned, three tests were performed at target soil temperatures of 23 °C (room temperature), 40 °C and 60 °C. For the first test no heating was required. For the second and third test the heating tape was used to heat the soil to the target soil temperature. First, the variable voltage meter, connected to the heating tape, was set to 50% while the soil was being increased from room temperature to the targeted testing temperature. Then the voltage meter was decreased to 10% for the rest of the experiment. To maintain the targeted soil temperature (± 5 °C) throughout the rest of the experiment, the automatic timer was set to turn the heating tape on and off in predetermined cycles, with a shorter cycle leading to higher temperatures. Although this leads to periodic fluctuations in temperature at the boundary of the specimen, the soil temperature within the column reaches a relatively stable value. Also, the length and frequency of these intervals changed as the experiment progressed because the soil was drying out and thus required less energy to keep at the targeted temperature. Preliminary tests indicate that the temperature difference across a horizontal cross section is within 2 °C after the system has reached a stable temperature.

After the soil layer was heated to the targeted temperature, the vacuum was turned on. Using the vacuum regulator, the vacuum was increased until targeted flow rate was achieved. The vacuum was then left on until the experiment was over. The vacuum gauge reading and inflow and effluent flow rates were recorded every few hours in the beginning of the experiment and at longer intervals as the experiment progressed. The total running vacuum time was different for each experiment. For Tests 1 and 3, the experiments were run until the vapor-phase total petroleum hydrocarbon (TPH) concentration in the effluent reached a steady state. Test 2 was stopped due to technical difficulties before steady state was reached.

RESULTS

The average temperature of the silt for each test with time is shown in Figure 3(a). Although there are slight temperature fluctuations due to the heating method, the target average temperature of soil (within an acceptable range) was achieved in both thermal tests. The airflow rates at the effluent side of the soil column are shown in Figure 3(b). Although a large amount of variability is observed in the data, the goal of the vacuum application was to maintain a constant airflow rate between 400 and 500 mL/min. It was easier to obtain a constant flow rate after a greater amount of water had been removed from the soil, which is why the airflow rate stabilizes over time in the two heated tests.

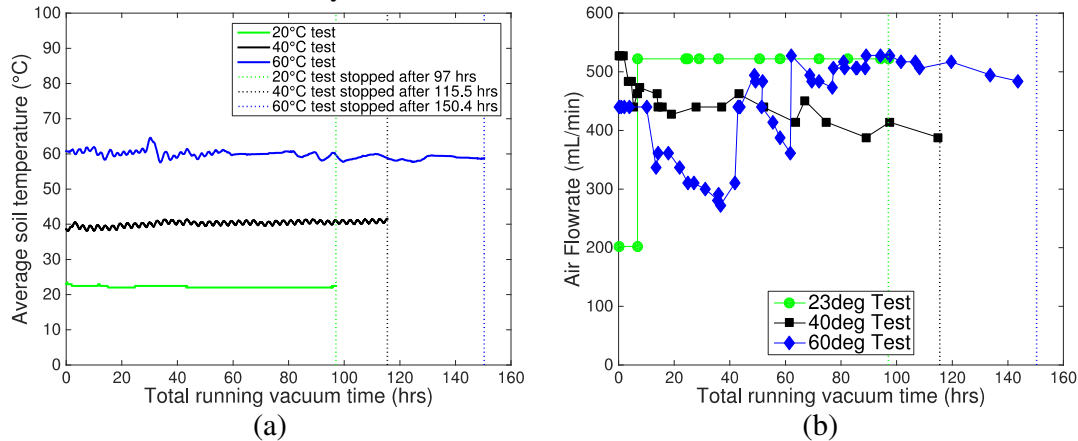


Figure 3: (a) Average temperature of the soil column during soil vapor extraction; (b) Effluent flow rate for all three tests

The change in dielectric permittivity at different locations in the column for both tests is shown in Figure 4. The timescale starts from when the silt was compacted to multiple hours after the vacuum was discontinued. The vertical dotted lines correspond with the ON/OFF vacuum interval. During the heating process, the dielectric permittivity of the soil showed an initial increase at the beginning, which corresponds with the upward flow of water and LNAPL past the location of each of the sensors. The dielectric permittivity then decreases throughout the tests. The amount of decrease is significantly greater in the higher temperature test, corresponding to a greater drying of the soil. An interesting observation is that the dielectric permittivity at all of the locations decreased at the same rate. This is possibly because the soil was initially relatively dry and the air permeability is uniform across the soil layer, leading to uniform drying. Some variability in the response of the sensors at different depths was observed, potentially due to the impact of preferential airflow paths through the soil layer.

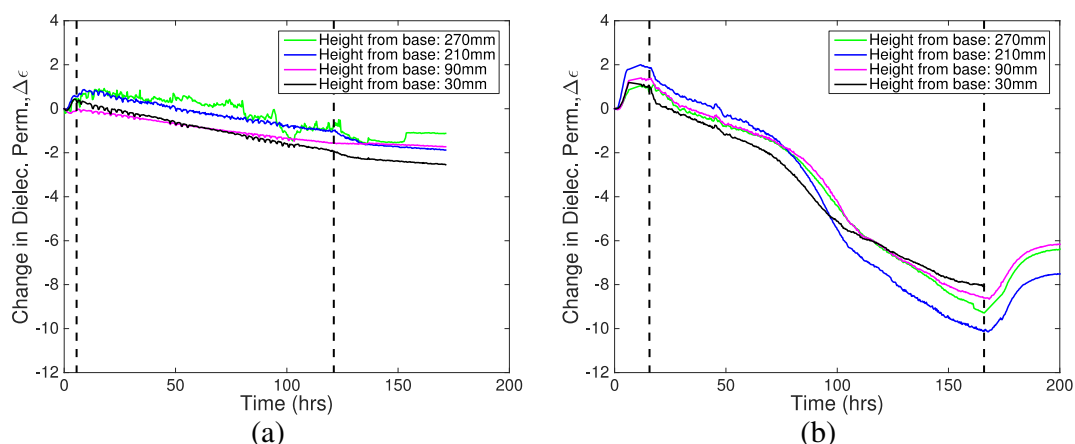
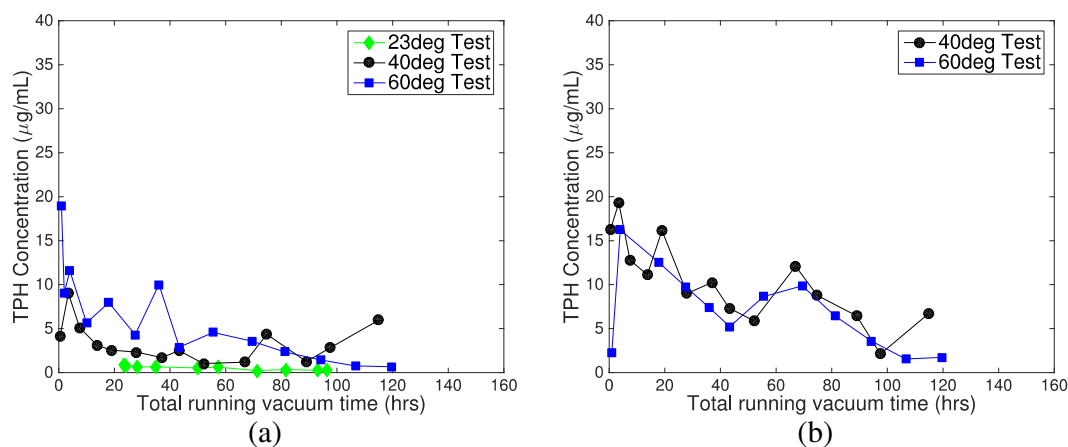
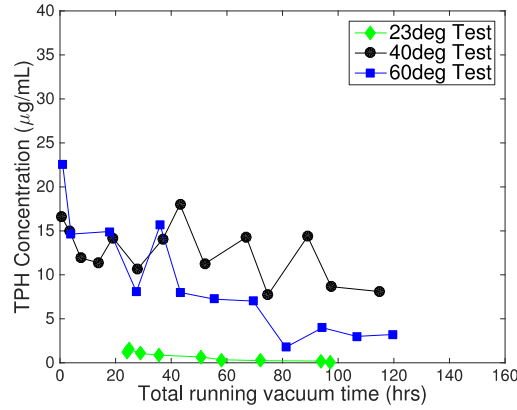


Figure 4: Change in dielectric permittivity with depth: (a) 40 °C test; (b) 60 °C test

The vapor-phase total petroleum hydrocarbon (TPH) concentrations for the effluent gas, and pore gas at two different heights in the soil column are shown in Figure 5. TPH concentrations for the pore space 300 mm above the base of the column is not available for the 23 °C test. All three tests show a decrease in TPH concentration with time. An initial sharp increase in effluent gas concentration followed by a sharp drop off is observed in Figure 5(a). The peak for the test at 60 °C is the highest, followed by the test at 40 °C. Although the data for the beginning of the test at 23 °C is missing, the beginning vapor-phase TPH concentrations for the test at 23 °C are likely lower than in the other two tests based on trends reported in the literature. The vapor-phase TPH concentration in the pore space 150-180 mm above the base of the column for the 60 °C test shown in Figure 5(c) decreases more rapidly and reaches a lower value than the 40 °C test. This implies that the rate of removal increases with temperature.

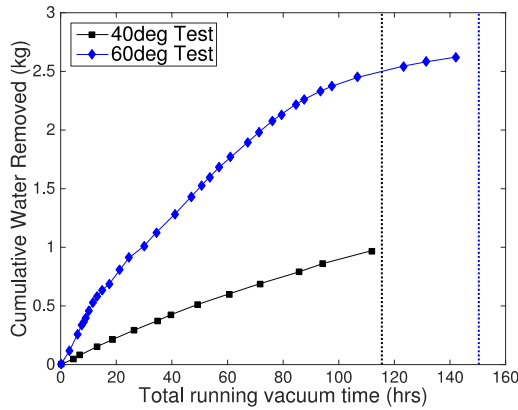




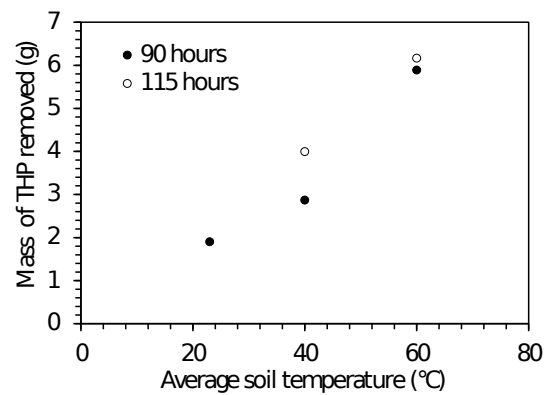
(c)

Figure 5: Results of the vapor phase TPH concentrations for each test: (a) TPH concentrations in the effluent; (b) TPH concentrations in the soil gas 300 mm above the base; (c) TPH concentrations in the soil gas 150-180 mm above the base

A summary of the cumulative amount of water removed during the tests at 40 and 60 °C is shown in Figure 6(a). A much larger amount of water was withdrawn from the column with the higher temperature. The mass of water removed after 90 hrs and 115.5 hours was estimated for all three tests based on water content measurements and is shown in Table 1. After 90 hours, approximately 8%, 31% and 88% of the water was removed from the soil in the 23, 40 and 60 °C tests, respectively. The relative humidity of the effluent gas for both of the heated tests reached 100% very quickly and maintained that value until the heat was turned off. This implies that the vapor pressure of water in the air leaving the column was fully saturated with water. The saturated vapor pressures of water in each test were 2.81, 7.37 and 19.92 kPa, respectively. Clearly an increased saturated vapor pressure at the higher temperature tests resulted in an increase of water removed.



(a)



(b)

Figure 6: (a) Cumulative mass of water removed; (b) Total mass of TPH removed after 90 and 115.5 hours of applied vacuum

Table 1: Water removed after 90 and 115.5 hours of applied vacuum

Test	Initial mass of water (kg) in soil	Mass of water (kg) removed		Percent removed	
		After 90 hrs	After 115.5 hrs	After 90 hrs	After 115.5 hrs
23 °C	2.62	0.21	NA	8.02 %	NA
40 °C	2.78	0.86	0.98	30.94 %	35.25 %
60 °C	2.80	2.47	2.50	88.21 %	89.29 %

The total mass of TPH removed from the soil after each test is shown in Figure 6(b). Mass removed was calculated assuming an average flow rate of 500 ml/min for each test. The average flow rate is multiplied by the vapor-phase TPH concentration and then integrated over time. Since the test at 23 °C was run for a shorter period of time, the total TPH removed for all three tests after 90 and 115.5 hours of applied vacuum are shown in Figure 6(b) for easier comparison. The total masses of TPH removed in the effluent for the 23 °C test (after 90 hrs), 40 °C test (after 115.5 hrs), and 60 °C test (after 115.5 hrs) was 0.15, 0.99, and 1.53 g, respectively. A clear increase in removal with temperature is observed. By comparing these cumulative TPH concentrations (7 g/mL) values to the original amount of TPH in the soil, the percent of TPH removed from the soil can be calculated. The percentage of the initial TPH in the soil removed was 0.55%, 0.97% and 1.49%, for the 23 °C test (after 90 hrs), 40 °C test (after 115.5 hrs), and 60 °C test (after 115.5 hrs), respectively. Although this clearly shows an increase of TPH removal with temperature, the overall removal percentage is still very low, even for the 60 °C test.

CONCLUSIONS

Although this study showed that TPH concentrations in the effluent increase with moderate increases in temperature, a significant change in the percent of initial diesel removed was not observed. This is likely due to the low volatility of diesel combined with the low air permeability of Bonny silt. This method may be more suited for use with more volatile LNAPLs, such as gasoline, where moderate increases in temperature may have a greater effect. Nonetheless, the heat collected from SBTES systems is from a renewable source and can be applied for a long duration for low cost. Overall, combining the heat storage aspects of SBTES systems with remediation strategies may have a net positive impact on the reuse of contaminated sites for moderate cost.

ACKNOWLEDGEMENTS

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