Predicting hydrocarbon removal from thermally enhanced soil vapor extraction systems
1. Laboratory studies

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Abstract

Conventional soil vapor extraction (SVE) systems have a limited effectiveness at removing semi-volatile chemicals from soil. Raising chemical vapor pressures by heating soil in-situ can decrease remediation time and help remove semi-volatile chemicals that otherwise would not be removed by conventional SVE. The increased compound removal rate that results from use of thermally enhanced SVE was investigated in laboratory studies. Increased soil temperatures (50–150°C) increased both the rate of removal of the compounds studied and the range of compounds that were removed in column studies. The column studies indicated that if soil temperatures are raised enough to elevate the vapor pressure of a compound above 70 Pa, SVE will remove most of the compound from the soil. Thermally enhanced column study hydrocarbon removal rate constants were shown to have a definable relationship with vapor pressure. The relative removal rate constants also demonstrated an Arrhenius relationship with temperature. Laboratory studies can be used to develop these relationships and the results can be extrapolated within certain temperature ranges and compound types for a given soil. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Soil vapor extraction (SVE) can be used to remove relatively volatile compounds (e.g. gasoline) from unsaturated soil. Heating the soil allows thermally enhanced SVE to

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accelerate the removal of volatile chemicals and increases the range of compounds that can be removed from a soil using SVE.

Thermally enhanced SVE technologies (radio frequency (RF) heating, electrical resistance heating, hot air injection, steam injection) exploit the higher chemical vapor pressures that result when soil temperatures increase. The vapor pressures of organic chemicals double to triple with a 20°C rise in temperature [1]. Thus, an SVE system operating in soil at elevated temperature will remove compounds at a higher rate than an ambient temperature SVE system [2].

However, there is little published data for predicting the effect of in-situ soil heating on compound removal. Field experience at conventional SVE sites indicates that compounds with vapor pressures greater than approximately 70 Pa at 25°C are candidates for SVE removal [1,3,4]. Using Antoine's equation (Eq. (1)), one can predict that straight-chain hydrocarbons up to C_{20} will have vapor pressures above 70 Pa at 150°C [5].

$$\ln(P_0) = -\frac{B}{T+C} + A \tag{1}$$

where A, B, and C are constants valid over a defined range of temperatures; T is the temperature; and P_0 is the vapor pressure.

Most compounds in diesel fuel have vapor pressures between those of C_{12} and C_{20}. Thus, when heat raises the vapor pressures of the diesel range organic (DRO) compounds above 70 Pa, one can expect such a thermally enhanced SVE system to remove a considerable fraction of such compounds. However, due to lack of published data, the benefit of the extra cost to heat the soil at a given site is not quantifiable prior to a pilot scale study.

The goal of this research was to determine, using a laboratory column study, the rates of removal of the *n*-alkane (C_{13}–C_{19}) compounds from a field soil at varying temperatures. The resultant hydrocarbon removal rate constants were related to the physical properties of the chemicals.

This laboratory effort was conducted in conjunction with a field demonstration of enhanced SVE using RF heating to remove diesel-range compounds from subsurface soil at Kirtland AFB in Albuquerque, NM. The RF-SVE demonstration site was a fire training pit abandoned in the late 1970s. The objectives of the field demonstration were to prove the feasibility of RF-SVE technology by removing significant quantities of semi-volatile hydrocarbons using a RF-SVE system. The field demonstration was a cooperative investigation by The University of Texas at Austin, Rice University, KAI Technologies, and Brown and Root Environmental [6].

2. Materials and methods

2.1. Column apparatus

The conceptual approach to the laboratory column study was to thermally release and then strip the compounds from the field demonstration soil. This two step contaminant removal process will be referred to as simply removal.

Fig. 1 shows the laboratory column study apparatus used. An instrument-grade compressed air cylinder provided clean sweep air for the column apparatus. A pressure regulator and a dial valve limited flow through the system to 13–15 ml/min. All piping between apparatus components was 0.3 cm (0.1 in.) outer-diameter stainless steel tubing.

The air flowed into a variable setting oven in which temperatures were measured with a 0–200°C thermometer placed in a sand-filled beaker. The ovens temperature-control vent was removed to allow flow lines and the sample tubes to pass in and out of the oven. After looping to the bottom of the oven to warm the inflow, the air flowed into 1.7-cm inner diameter, 30-cm long, vertical, stainless steel, soil sample pipe.

All soil samples came from a well-mixed composite of unremediated demonstration site soil. Table 1 summarizes the physical properties of the field soil [6]. The composite was made from site borings taken from the middle of the RF-SVE demonstration site. The samples for the composite were taken from the same depths as the target depths (3–6 m) for the field RF-SVE system. Thus, the column studies were conducted using the same soil, chemicals and concentrations that were present in the field site soil. To help provide soil homogeneity, rocks and debris were removed with a 2-mm opening sieve (US Standard Mesh Number 10).

Roughly 35 g of the soil were loosely poured into the bottom 20-cm of the 30-cm sample pipe atop a glass wool plug. A second glass wool plug was pushed down from the top to hold the sample in place. The sample pipe was contained entirely inside the oven during each run, with the soil-filled section approximately centered in the oven.

Chemicals in the sample column off-gas were collected on a sorbent that was periodically analyzed. Chemicals were extracted from the sorbent with methylene chloride and the extract was analyzed on a gas chromatograph (GC). Carbotrap C® was chosen as the column sorbent based largely on the supplier recommendation and reusability. In this paper, Carbotrap C® is referred to as Carbotrap.
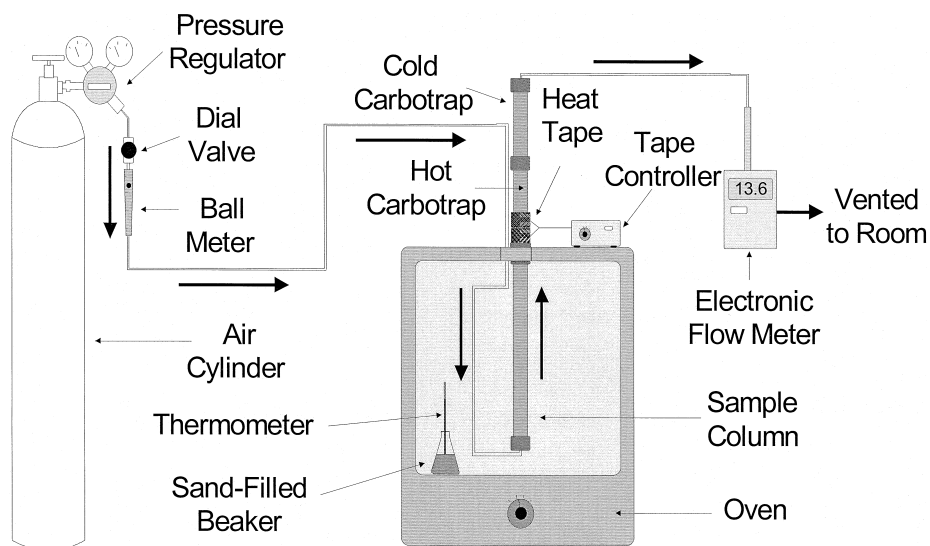

Fig. 1. Column study schematic.

Table 1
In-situ demonstration site soil properties [7]

Grain size	
Sand	60–75%
Silt	5–15%
Clay	0–15%
Chemicals present	
DRO (C_{12}–C_{20})	∼ 1000 mg/kg dry weight
Total petroleum hydrocarbons	∼ 7500 mg/kg dry weight
Tridecane, C_{13}	∼ 40 mg/kg dry weight
Pentadecane, C_{15}	∼ 80 mg/kg dry weight
Heptadecane, C_{17}	∼ 60 mg/kg dry weight
Nonadecane, C_{19}	∼ 30 mg/kg dry weight
Organic carbon content	3.0%
Moisture content (dry weight basis)	
Range	1–10%
Mean	7%
Porosity	0.34
Dry density	17.5 kN/m^3
Air permeability	0.1–100 Darcys

Sorbent containing traps were placed immediately outside the oven vent. The sample Carbotrap tubes were prepared as follows. A tightly packed glass wool plug was placed at the bottom of a 1.7-cm inner diameter, 7.6-cm long, stainless steel sample tube. Roughly 4 g of Carbotrap were poured into the tube prior to pushing in another tightly packed glass wool plug to hold the Carbotrap in place.

A two-trap system was used. A hot Carbotrap tube, at oven temperature, collected the semi-volatile, heavy hydrocarbons. A cold Carbotrap tube, at ambient temperature, trapped the more volatile hydrocarbons. To maintain the hot Carbotrap at the oven temperature, the bottom half of the hot trap was wrapped in heat tape. Although the hot Carbotrap tube was placed just outside the oven (to allow trap changes without affecting the oven temperature), it maintained a temperature slightly above the oven temperature. The cold Carbotrap tube sat roughly 13 cm above the oven exit. For all oven temperatures, the cold trap was at ambient room temperature (roughly 22°C). During the 150°C column study, no hydrocarbon mass was detected on additional Carbotrap tubes placed beyond the cold trap.

The hot and cold Carbotrap tubes were replaced with a fresh set of hot and cold Carbotrap tubes 10–20 times during each experiment. The Carbotrap tubes were removed from the column apparatus after time periods long enough for the traps to acquire detectable amounts of hydrocarbons for GC analysis. However, the sample periods were short enough to prevent the compounds from saturating and breaking through the Carbotrap. Thus, at the beginning of each column study, both Carbotrap tubes were replaced every hour. Near the end of each study, the Carbotrap tubes were replaced every 12–24 h. The Carbotrap from the hot and cold tubes were combined into one sample and extracted on a daily basis. By analyzing the amount of chemical in each set of combined Carbotrap tubes, the chemical removal from the soil was related to the time elapsed since the start of the column study.

An electronic flowmeter was connected to the system after the traps. The average flow (13–15 ml/min) varied by 10% during experiments. The flow rate was not varied on purpose. Rather the flow variation was a result of the experimental setup. The carbotrap tubes had a varying resistance to airflow. Each glass wool plug was packed slightly differently. Carbotrap tubes with densely packed glass wool had a higher resistance to airflow. In addition, as the soil was heated from ambient temperatures to the desired temperatures, water was driven out of the soil and onto the Carbotrap tubes. Varying moisture contents in the soil and carbotrap led to varying airflow resistance during each column study. Thus, the carbotrap tubes used earlier in a study had a higher resistance to airflow than did the tubes used later in a study.

Analysis　　　Methods

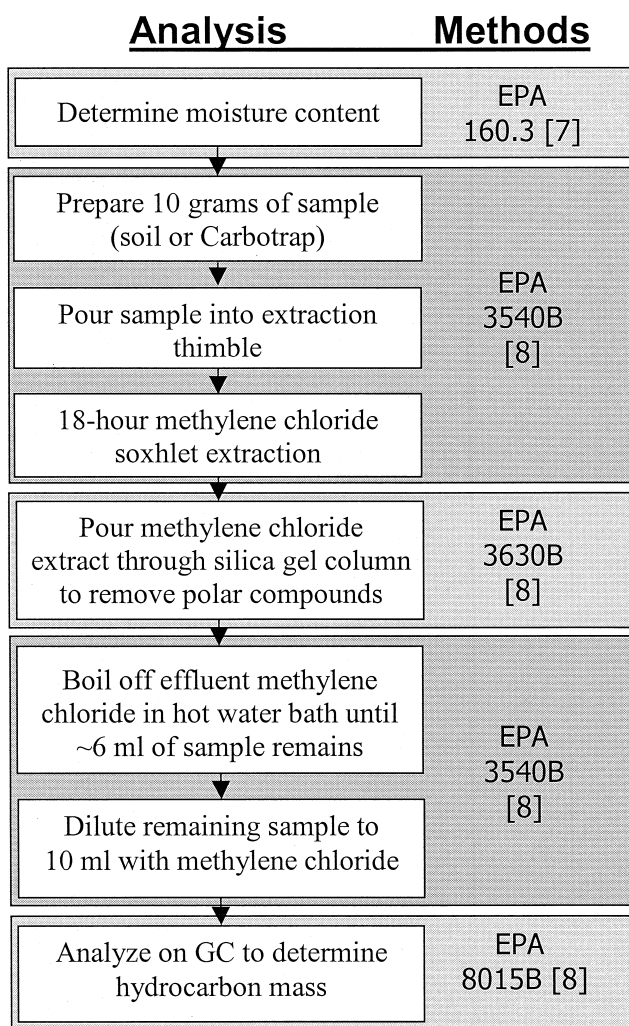|	
Determine moisture content	EPA 160.3 [7]
Prepare 10 grams of sample (soil or Carbotrap)	
Pour sample into extraction thimble	EPA 3540B [8]
18-hour methylene chloride soxhlet extraction	
Pour methylene chloride extract through silica gel column to remove polar compounds	EPA 3630B [8]
Boil off effluent methylene chloride in hot water bath until ~6 ml of sample remains	EPA 3540B [8]
Dilute remaining sample to 10 ml with methylene chloride	
Analyze on GC to determine hydrocarbon mass	EPA 8015B [8]

Fig. 2. Chemical analysis process diagram.

2.2. Analytical procedures

The chemical analyses used to measure hydrocarbon masses are outlined in Fig. 2. The moisture content of each soil sample was determined with a gravimetric analysis. For chemical analysis, an 18-h methylene chloride soxhlet extraction removed chemicals from the soil and the Carbotrap samples. After removing the polar compounds with silica gel and concentrating the extract, a portion of each extract was injected into the GC to determine the hydrocarbon concentration. In turn, the concentration data were used to determine the hydrocarbon mass removed from the soil and collected on the carbotrap.

The GC procedure followed EPA SW-846 method 8015B [8]. All GC work used methylene chloride as the solvent and a Restek® (Bellefonte, PA) 30-m long, 0.32 mm ID, 0.25 µm *df* (film density), Rtx-5 column. Samples were analyzed using a HP 5890 Series II Plus GC with a HP 6890 Autosampler and a flame ionization detector set at 300°C. For each sample, the GC temperature was held at 37°C for 1 min, ramped 19°C/min to 280°C, held at 280°C for 5 min, ramped 20°C/min to 310°C before being held for 30 min. Samples were compared to standards made from a straight chain hydrocarbon standard (C_{10}–C_{25}, Tennessee/Mississippi DRO Mix Standard) produced by Restek®.

The masses of C_{13}–C_{19} hydrocarbons were determined for all carbotrap and soil samples. Tracking individual C_{13}–C_{19} hydrocarbon masses collected in the off-gas over time revealed the fraction of the compound removed, the relative rate of the compound removal and the effect of temperature on the compound removal rate from the soil. At the conclusion of the column study, the soil in the column was extracted to determine the hydrocarbon mass remaining on the soil after the column study. Data for C_{13}, C_{15}, C_{17}, and C_{19} hydrocarbons are presented in this paper to show the general trends affecting release of diesel compounds at the temperatures investigated.

3. Results

Four column studies were conducted to determine the effect of temperature on the relative rate of compound removal from the soil. The target temperatures of the four studies were 50, 100, 125 and 150°C. However, due to oven variations, the actual average temperatures of the soil columns varied slightly (Table 2). All calculations in

Table 2
Oven temperature variations for column study experiments

Target temperature (°C)	Actual average temperature (°C)	Standard deviation (°C)	Number of measurements	Low (°C)	High (°C)
50	47	1.0	18	46	49
100	102	3.4	15	95	109
125	128	2.9	20	120	132
150	157	4.4	10	154	166

this paper were done using the actual column average temperatures. However, in this paper, the studies are referred to by the target temperatures.

Temperature affected both the rate of compound removal from the soil and the fraction of the original compound mass remaining in the soil at the conclusion of the column study. The rate at which the individual compounds were removed from the soil was determined from the cumulative mass collected in the Carbotrap tubes. Mass below detection limits (0.03 mg alkane/kg dry weight) was assumed to be zero.

The removal rate constant of hydrocarbon mass from the soil was determined as follows. The final hydrocarbon mass remaining on the soil was determined from analyzing the soil in the column at the conclusion of each study. The initial hydrocarbon mass, used for mass balance determinations, was determined from a subset of soil separated from the column study soil prior to each experiment.

The cumulative mass collected data were normalized to enable the comparison of the removal rates at different temperatures. This was required because each column study had somewhat different initial hydrocarbon masses. Normalizing also allowed comparisons among different hydrocarbons with different initial masses in a single column study.

Normalization was done by dividing the cumulative Carbotrap hydrocarbon mass at specific points in time (G_t) by the total mass of the hydrocarbon of interest in the system. The total hydrocarbon mass in the system was defined as the final cumulative mass collected on all the Carbotraps (G_F) plus the hydrocarbon mass remaining on the soil at the conclusion of the column study (S_F). Eq. (2) shows the final normalized expression for the fraction of hydrocarbon compounds remaining on the soil.

$$\text{Fraction Remaining in Soil at Time } t = 1 - \left[\frac{G_t}{G_F + S_F} \right] \qquad (2)$$

To illustrate the hydrocarbon removal patterns that resulted, Fig. 3 displays the fraction of tridecane (C_{13}) remaining in the soil over time for all column studies. The times for each data point in Fig. 3 denote when the Carbotrap tubes were replaced with
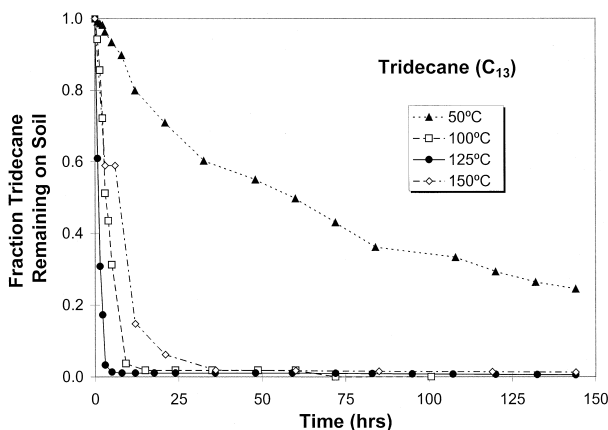
Fig. 3. Fraction of tridecane (C_{13}) remaining on soil as a function of time and temperature.

fresh Carbotrap tubes. Similar patterns resulted for the other hydrocarbon compounds evaluated. Faster hydrocarbon removal occurred in the higher temperature column studies than in the lower temperature studies. In addition, in the time frame of the experiments, a greater total fraction of the initial mass in the soil was removed in the higher temperature studies. Anomalous data are discussed in greater detail below.

Figs. 3 and 4 suggest that the mechanisms controlling the release of alkanes from the soil change over time. Conceptually, the data can be divided into three zones (Fig. 4; 125°C data). Zone 1 incorporates the initial linear alkane removal from the soil. An analysis not presented in this paper suggests that this initial alkane removal is limited by equilibrium partitioning from non-aqueous phase liquids to the gas phase. Zone 2, the transition zone, describes the bend in the data. This is the result of the mole fractions of the alkanes decreasing both the partial pressures of the alkanes and the rate of alkane removal. Zone 3 describes the data points at the end of the experiment where minimal alkane removal is occurring. In this region, removal is likely dominated by non-equilibrium mechanisms, such as kinetic film transfer limitations and or diffusion of alkanes from organic matter. Unfortunately, all the mechanistic based mathematical models that were examined were unable to describe the entire set of data. Hence, the data was analyzed in the following manner.

A first-order model did not provide a reasonable fit for the entire range of curves generated in the column experiments. Thus, a two-region, first-order equation was applied to the column data. The equation has been used to model the release of compounds in water-saturated soils [9,10]. This model assumes that a certain fraction of the compound is removed at the fast rate and the remainder is removed at some slower rate (Eq. (3)).

$$F_{RS}(t) = F_{fast}(e^{-k_{fast}t}) + (1 - F_{fast})(e^{-k_{slow}t}) \qquad (3)$$

where $F_{RS}(t)$ = fraction remaining on the soil at time t; F_{fast} = fast fraction released from the soil; k_{fast} = removal rate constant for fast fraction (1/h); k_{slow} = removal rate constant for slow fraction (1/h).
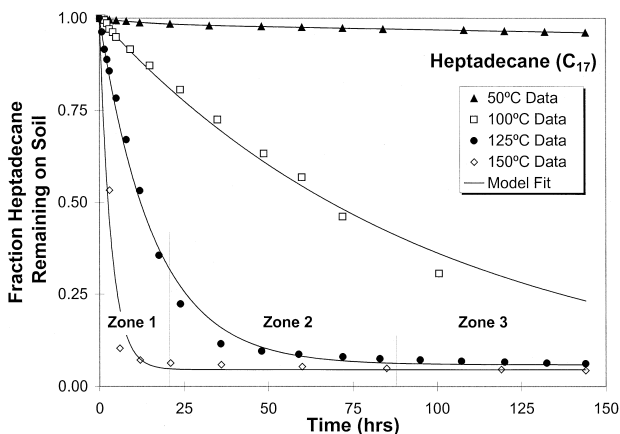

Fig. 4. Heptadecane (C_{17}) comparison of two-region first-order curve fit with experimental data.

For both the 125 and 150°C column studies, the k_{slow} rate constant was zero. For those column studies, the zero k_{slow} value effectively modifies Eq. (3) to a first-order model with a non-releasing residual.

Fig. 4 illustrates the fit of Eq. (3) for heptadecane (C_{17}) in each of the column studies. With different model parameters, Eq. (3) was able to reproduce both the steep reductions in the 150°C column study data and the slow removal in the 50°C experimental data. Similar calculations were performed to obtain best-fit parameters for the other hydrocarbons evaluated.

The objective of the column studies was to predict at a given temperature the relative rate of hydrocarbon removal (described by Eq. (3)) from the demonstration site soil. The conceptual model presented above suggests that the removal of alkanes from the soil is related to vapor pressure and temperature.

The vapor pressures presented in this paper were calculated using the OLI$^®$ Environmental Simulation Program software (Version 5.2; Morris Plains, NJ). OLI$^®$ is a private company that is developing computer software to model environmental systems. The Environmental Simulation Program provides a database of physical properties for a broad range of chemicals. The software used a refined Antoine equation to predict chemical vapor pressure from experimental data. Since relevant vapor pressure data for the temperatures of interest could not be obtained from other references, the vapor pressure data used in Figs. 5 and 6 were determined using the OLI$^®$ software.

As shown in Fig. 5, vapor pressure had a non-random relationship with the fast fraction released from the soil, F_{fast}. Fig. 5 displays only data from the 50, 125 and 150°C studies. In the 100°C study, k_{fast} equaled k_{slow}. Thus, for the 100°C study, Eq. (3) reduces to a first-order model for which there is no F_{fast} term.

The results presented in Fig. 5 indicate that if soil temperatures are raised enough to elevate the vapor pressure of a compound above 70 Pa, SVE will remove most of the compound from the soil. This coincides with field experience at conventional SVE sites that indicates that chemicals with vapor pressures of 70 Pa or greater are candidates for
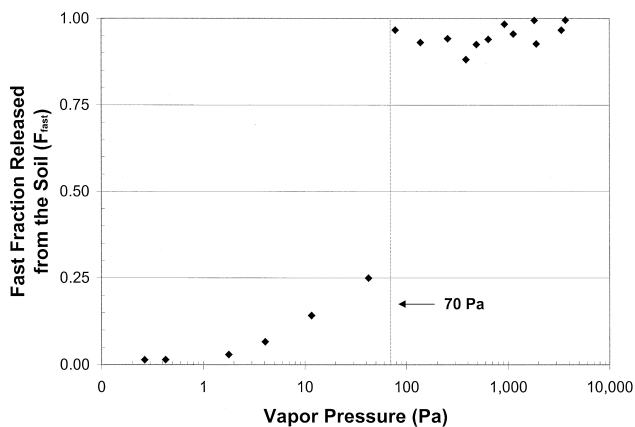

Fig. 5. Fast fraction released from soil as a function of vapor pressure.
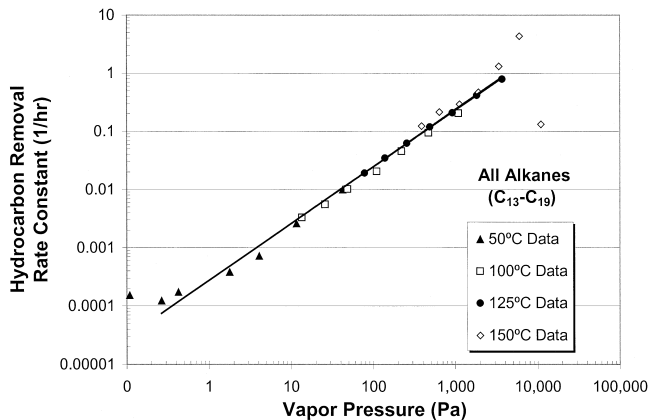
Fig. 6. Hydrocarbon removal rate constant as a function of vapor pressure.

SVE removal [1,3,4]. Compounds with vapor pressures below 70 Pa will still be removed, albeit at a slower rate and possibly to a lesser extent.

As Fig. 5 illustrates, a majority of the F_{fast} values were either above 0.9 or below 0.1. This indicates that a majority of the fraction of the hydrocarbon remaining (F_{RS}) was dominated by one part of Eq. (3). For instance, when F_{fast} was greater than 0.9, k_{fast} represented a majority of the chemical removal from the soil. In addition, when F_{fast} was less than 0.1, k_{slow} represented a majority of the chemical removal from the soil. Thus, correlation to physical properties of the compounds could be done with a single rate constant representing a majority of the chemical removal.

In 125 and 150°C experiments, F_{fast} was always greater than 0.93. In other words, in these two studies, k_{fast} represented at least 93% of the curve. In addition, for these two studies, a majority of the compound mass was removed early (i.e., fast) in the experiment. Thus, k_{fast} represented the important part of the curve for these two column studies.

As mentioned above, the more gradual curves in the 100°C column study resulted in rate constants for the fast and slow fractions that were identical. Thus, one value was used to represent chemical loss for the 100°C column study. Except for C_{13}, F_{fast} in the 50°C experiments was always below 0.13. Thus, for these experiments, k_{slow} represented at least 87% of the curve and the majority of the compounds in the 50°C study were dominated by a slow removal. Thus, k_{slow} was the rate constant most relevant for the 50°C column study.

Table 3 summarizes the removal rate constants that were used in the subsequent predictive evaluation. As discussed above, these rate constants correspond to either k_{fast} or k_{slow} of Eq. (3).

Fig. 6 shows the linear relationship between the log of the vapor pressure and the log of the removal rate constants presented in Table 3. Fig. 6 indicates that vapor pressure has a strong influence on the hydrocarbon release rate constant.

The two points on the far right of the graph represent C_{13} and C_{14} hydrocarbons in the 150°C study. In this study, there were anomalies in the raw Carbotrap data for these

Table 3
Summary of column study chemical removal rate constants (1/h) used in subsequent analysis

Compound	Column study			
	150°C	125°C	100°C	50°C
C_{13}	0.13	0.79	0.21	0.0099
C_{14}	4.3	0.41	0.094	0.0026
C_{15}	1.3	0.21	0.045	0.00072
C_{16}	0.47	0.12	0.020	0.00038
C_{17}	0.29	0.062	0.010	0.00017
C_{18}	0.21	0.035	0.0055	0.00012
C_{19}	0.12	0.019	0.0033	0.00015

hydrocarbons. At any temperature, the C_{13} and C_{14} hydrocarbons have higher vapor pressures than the C_{15}–C_{19} hydrocarbons. Given this vapor pressure data, one would expect that the C_{13} and C_{14} hydrocarbons would reach 100% chemical loss faster than the other compounds. This trend is shown by the column studies at other temperatures. However, the 150°C data did not show this trend. In fact, no detectable mass of C_{13} was collected between 3 and 6 h (Fig. 3). Significant quantities of the heavier hydrocarbons were collected during the same time period. Therefore, the C_{13} and C_{14} hydrocarbon rate constants at 150°C appear to be based on anomalous data. A thorough examination of the data was done but no explanation for the anomalies was found.

The C_{19} hydrocarbon rate constant from the 50°C column study resulted from data that were barely above the GC detection limits. For these reasons, the linear interpolation shown in Fig. 6 does not include the rate constants for the C_{19} hydrocarbon from the 50°C column study and the C_{13}–C_{14} hydrocarbons from the 150°C column study.

Most vapor pressure models predict the log of the vapor pressure using an inverse temperature relationship. Both the Antoine equation (Eq. (1)) and the thermodynami-cally derived Clausius–Clapeyron equation have this inverse temperature relationship with the log of the vapor pressure [5]. Since vapor pressure could be correlated with removal rate constants, a relationship between the removal rate constants and inverse temperature was also expected.
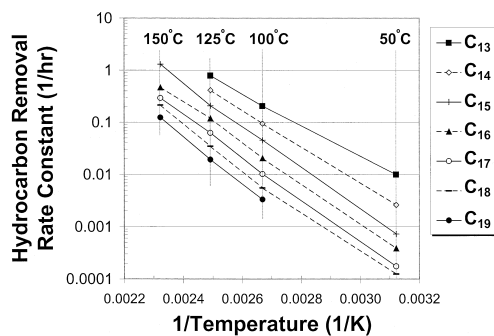

Fig. 7. Hydrocarbon removal rate constant as a function of inverse temperature.

Fig. 7 shows the relationships between the inverse temperature and the log of the previously derived hydrocarbon removal rate constants (Table 3). The temperatures are plotted as inverse Kelvin.

Because of the reasons outlined earlier, the rate constants for the C_{13} and C_{14} hydrocarbons in the 150°C study and the rate constant for C_{19} hydrocarbon in the 50°C study were not shown in Fig. 7.

4. Discussion

The results of this study quantified the extent that an increase in soil temperature increases the range of compounds removed from a soil. Increased soil temperatures also increase the rate of removal of the compounds studied. The relative removal rate constants can be related to temperature and vapor pressure. These experiments indicate that laboratory studies can be used to develop such relationships and it appears that the results can be extrapolated within certain temperature in the following manner (equations for linear interpolation on Figs. 6 and 7).

$$\text{Chemical Removal Rate Constant} \left(\frac{1}{h} \right) = \beta \left[\text{Vapor Pressure (Pa)} \right]^{\alpha} \tag{4}$$

$$\text{Chemical Removal Rate Constant} \left(\frac{1}{h} \right) = \delta * 10^{\left[\frac{\gamma}{\text{Temperature (K)}} \right]} \tag{5}$$

The exact parameter values presented are compound and soil specific. The removal rate constants presented in Eqs. (4) and (5) are also dependent on the airflow characteristics of the system and thus cannot be directly used to predict remediation times for field thermally enhanced SVE systems.

However, chemical and soil specific parameter values for Eq. (4) can be obtained by following the methods outlined. Only one column study using soil from a site of interest is necessary to evaluate the α and β constants in Eq. (4). By examining just two compounds present in the soil that encompass the range of vapor pressures that are of interest, hydrocarbon removal rate constants for all physically similar compounds in the soil can be estimated.

Thus, appropriate laboratory studies will help define, prior to implementation, the applicability of thermally enhanced SVE at a given site.

References

[1] K. Rathfelder, J.R. Lang, L.M. Abriola, Rev. Geophys. Suppl. (1995) 1067.

[2] T. Bergsman, B. Trowbridge, Soil and Groundwater Cleanup, July 6, 1997.

[3] N.L. Hutzler, B.E. Murphy, J.S. Gierke, US Environmental Protection Agency, Risk Reduction Engineering Laboratory, OH, January 1990, EPA 600/S2-89/024.

[4] A.S. Goldfarb, G.A. Vogel, D.E. Lundquist, Waste Manage. 14 (1994) 153.

[5] R.P. Schwarzenbach, P.M. Gschwend, D.M. Imboden, Environmental Organic Chemistry, Chap. 4, Wiley, New York, 1993.

[6] M.T. Webster, R.C. Loehr, A field demonstration of enhanced soil vapor extraction with radio frequency heating, Final Report to Advanced Applied Technology Demonstration Facility, Rice University, Houston, TX, 1998.

[7] Methods for Chemical Analysis of Water and Wastes, 3rd edn. US Environmental Protection Agency, Washington DC, 1983, EPA 600/4-79-020.

[8] Test Methods for Evaluating Solid Waste, 3rd edn. US Environmental Protection Agency, Washington DC, 1995, EPA 530/SW-846.

[9] G. Cornelissen, P.C.M. van Noort, H.A. Govers, J. Environ. Toxicol. Chem. 16 (1997) 1351.

[10] J.J. Pignatello, J. Environ. Toxicol. Chem. 9 (1990) 1107.