Predicting hydrocarbon removal from thermally enhanced soil vapor extraction systems 2. Field study

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Abstract

The effectiveness of a thermally enhanced soil vapor extraction (SVE) system to remove semi-volatile organic chemicals (SVOCs) was investigated in a field study. The data allowed the calculations of SVOC removal rates at several temperatures. A previous laboratory study using the same field soils had developed a relationship between SVOC removal rate constants and inverse temperature. The laboratory and the field SVOC removal rate constants were compared and a linear log–log relationship between the laboratory and the field SVOC removal rate constants resulted. Subsequent analyses indicated that it was possible to use laboratory determined SVOC removal rate relationships to estimate SVOC removal from in situ field soil. The time dependence of SVOC concentration reduction using in situ thermally enhanced SVE systems was also shown. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Soil remediation; Thermally enhanced soil vapor extraction; Semi-volatile organic compound (SVOC); Vapor pressure; Temperature

1. Introduction

Higher vapor pressures allow thermally enhanced soil vapor extraction (SVE) to remove a broader range of compounds at a higher rate than an ambient temperature SVE system [1]. The various thermally enhanced SVE applications (radio frequency heating,

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electrical resistance heating, hot air injection, steam injection) elevate temperatures, and hence vapor pressures, to different levels. The design of thermally enhanced SVE systems is simplified if there is knowledge of the in situ soil temperature that is required to remove the chemicals of interest from the soil. However, there are few published data to predict the effect of elevated in situ soil temperatures on removal of semi-volatile organic compounds (SVOCs, e.g. diesel fuel).

Previous laboratory column studies conducted at elevated temperatures indicated that rate constants for the removal of *n*-alkane SVOCs (C_{13}–C_{19}) from soil could be correlated to the inverse of the soil temperature [2]. The goal of this evaluation was to use the rate constants determined in the laboratory column study to predict removal rates of SVOCs from in situ soil that was undergoing field-scale thermally enhanced SVE. Since the soil used to generate the previous laboratory data was obtained from the field site where thermally enhanced SVE was implemented, it was hypothesized that there should be a relationship between field-scale SVOC removal and the column study SVOC removal.

2. Material and methods

2.1. Field demonstration

A field demonstration of thermally enhanced SVE using radio frequency (RF) heating to remove SVOCs from in situ soil was conducted at Kirtland AFB in Albuquerque, NM. The RF-SVE demonstration site was a fire training pit abandoned in the late 1970s. The field demonstration was a cooperative investigation by the University of Texas at Austin, Rice University, KAI Technologies, and Brown & Root Environmental [3].

The objective of the field demonstration was to investigate the feasibility of in situ thermally enhanced removal of SVOCs using a RF-SVE system. The mass of soil targeted to be impacted by the RF-SVE system was approximately 3 to 6 m below ground surface elevation. The well configuration at the center of the demonstration site is shown in Fig. 1. Two RF applicator wells were located 2 m apart with the SVE well located between them in the center of the target volume of soil. Four monitoring wells were located 1.5 m from the SVE well. Each RF antenna casing contained a continuously monitored Luxtron® temperature probe. Temperatures in the monitoring wells were recorded daily with thermocouples. However, not all depths and wells were recorded each day. Hence, the number of temperature readings recorded on a given day ranged from 16 to 61. At the beginning and the end of the RF-SVE demonstration, soil borings were taken from the target area and analyzed to determine the change in SVOC concentration that occurred as a result of the RF-SVE demonstration.

Prior to the RF-SVE demonstration, SVE was operated alone at the field site to determine the chemical removal that could be achieved with ambient temperature SVE [3]. Gas chromatograph (GC) monitoring during this no-heat SVE phase revealed that the constituents in the off-gas were primarily gasoline range organic compounds ($< C_{12}$), e.g. the compounds removed were more volatile than the SVOCs of interest.
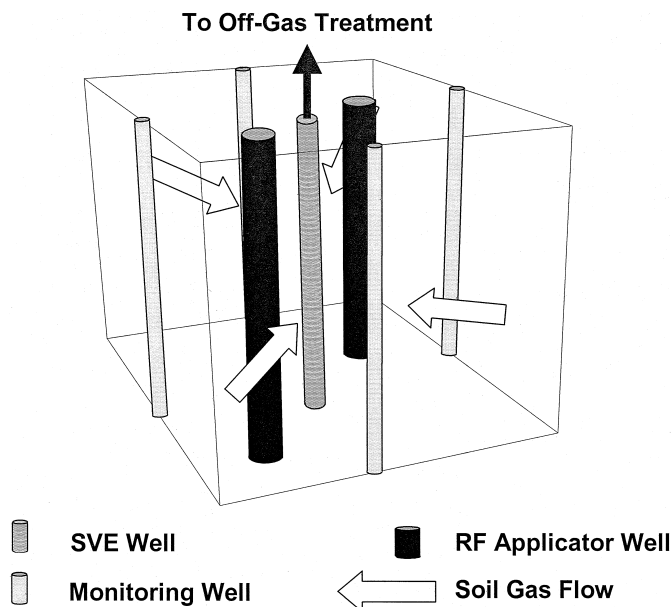
Fig. 1. Conceptual view of the RF-SVE system components.

Less than 0.01% of the total mass recovered during the demonstration of the alkanes of interest was removed during this ambient temperature SVE phase.

RF heating was conducted to enhance the removal of SVOCs beyond what could be achieved with SVE alone. During the RF-SVE demonstration, the RF system was powered for 87 days. Temperatures at the center SVE well reached a maximum of 139°C. The maximum temperatures of the four perimeter monitoring wells shown in Fig. 1 ranged from 100°C to 120°C. During these 87 days, the SVE extracted the soil gas at a varying rate of 37–538 l/min. The variation in flow rates was due to evaluation of various RF-SVE operating conditions.

As a measure of RF-SVE system performance, the non-aqueous phase liquid (NAPL) in the condensate knockout drums was analyzed. Chemical removal rate constants from the site soil were calculated using the cumulative NAPL mass collected in the knockout drums as a function of treatment time. In addition, RF-SVE performance was evaluated with respect to the soil temperatures achieved.

2.2. Soil analytical procedures

The volume of soil impacted by the RF-SVE system at the demonstration site was defined as the volume of soil that exceeded 94°C, the boiling point of water at the site (which was roughly 1500 m above sea level). Most of the NAPL in the condensate drums was recovered after soil near the RF antennas reached roughly 94°C. A kriging

analysis of the temperature data from the site monitoring wells was done with Neosys software (Fortner Research, Sterling, VA) to determine 70,300 kg of soil achieved temperatures greater than 94°C during the demonstration. A complete discussion of the temperature analysis used to determine this mass is presented in the field demonstration report [3]. The 70,300 kg of soil correlates to a volume that is roughly equivalent to the $3 \times 3 \times 3$ m^3 shown in Fig. 1.

Of the 21 soil borings drilled prior to the start of the RF-SVE demonstration, 14 borings were located in the defined impacted volume. The 3 m of each boring that were within this impacted zone were divided into five 0.6-m sections. Each 0.6-m boring section was composited and analyzed to determine SVOC concentrations. The SVOC concentrations of the 70 pre-treatment soil samples in the impacted zone were averaged to determine a single representative initial SVOC concentration within this impacted soil mass. Given the variability in the soil sampling, the averaging technique gave sufficiently accurate results so as not to warrant a more complicated weighting technique. The pre-treatment mass of each SVOC was then calculated by multiplying the SVOC concentration by the mass of the impacted soil. The post-treatment SVOC masses were determined in a similar manner using 30 soil samples from six post-treatment borings that were located in the impacted region.

The chemical analyses used to measure SVOC concentrations in the soil samples are outlined in Fig. 2. The moisture content of each soil sample was determined with a gravimetric analysis [4]. For chemical analyses, an 18-h methylene chloride soxhlet extraction of the soil samples was used. After removing the polar compounds from the extract with silica gel and concentrating the extract, a portion of each extract was analyzed by GC to determine the SVOC concentration in the soil samples.

The GC procedure followed EPA SW-846 method 8015B [5]. All GC analyses used methylene chloride as the solvent and a Restek® (Bellefonte, PA) 30-m, 0.32 mm i.d. 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 (C10-C25, Tennessee/Mississippi DRO Mix Standard) produced by Restek®.

2.3. Condensate analytical procedures

The condensate analysis began by sampling the NAPL that was floating on the water surface in each knockout drum of the RF-SVE off-gas treatment system. The NAPL phase was diluted with a known volume of methylene chloride. After a silica gel cleanup, the sample was analyzed using the GC methods outlined above. By multiplying the specific SVOC concentration in the NAPL by the volume of NAPL in each drum, the SVOC mass in each drum was determined.

The knockout drums were replaced every 1 to 2 days during the time period of interest. Analyses of the condensate water revealed that less than 1% of the total SVOC

Analysis   Methods

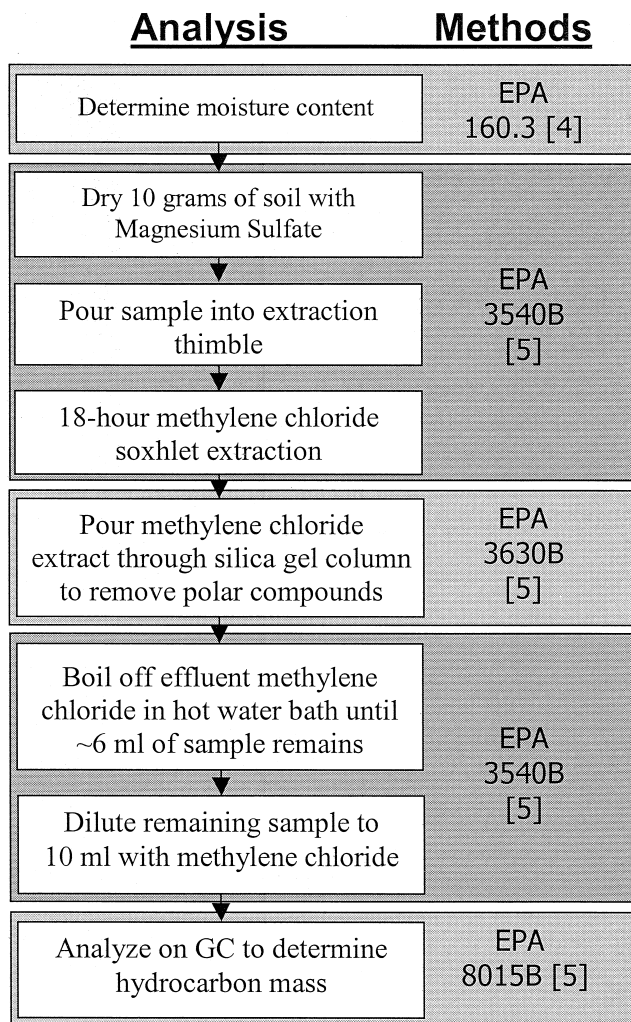| Determine moisture content	EPA 160.3 [4]
Dry 10 grams of soil with Magnesium Sulfate	EPA 3540B [5]
Pour sample into extraction thimble	
18-hour methylene chloride soxhlet extraction	
Pour methylene chloride extract through silica gel column to remove polar compounds	EPA 3630B [5]
Boil off effluent methylene chloride in hot water bath until ~6 ml of sample remains	EPA 3540B [5]
Dilute remaining sample to 10 ml with methylene chloride	
Analyze on GC to determine hydrocarbon mass	EPA 8015B [5]

Fig. 2. Chemical analysis process diagram.

mass in each knockout drum was dissolved in the aqueous phase. Thus, only the mass of the SVOC in the NAPL phase of each knockout drum was used in this evaluation.

3. Results

3.1. Condensate data

Fig. 3 shows the cumulative mass removed for pentadecane (C_{15}) found in the impacted region. The average monitoring well temperature noted in Fig. 3 is the average value of the four monitoring wells shown in Fig. 1.
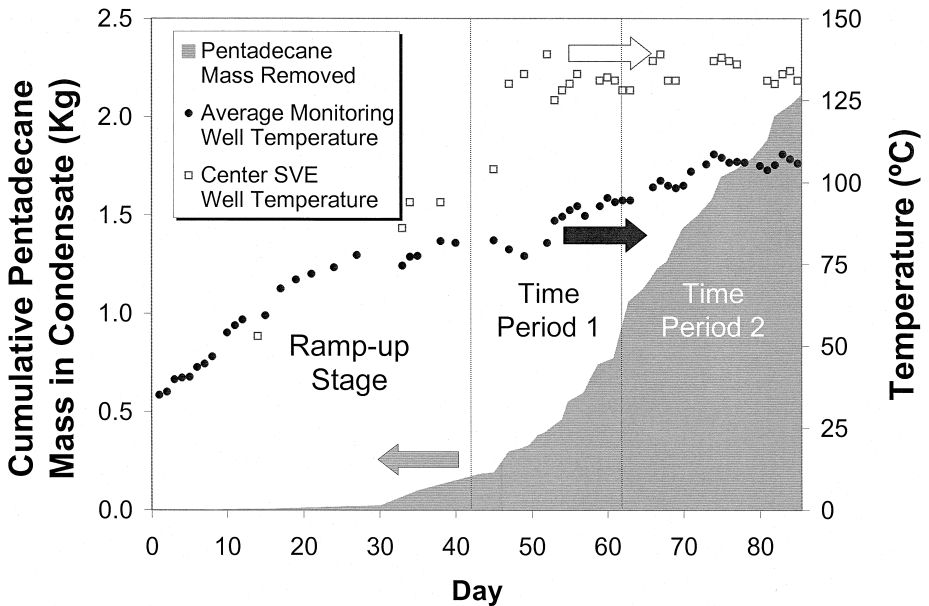
Fig. 3. Cumulative pentadecane (C_{15}) mass in the RF-SVE demonstration condensate.

Fig. 3 illustrates two of the three operational stages during the field investigation. The first operational stage, prior to day 45, was a ramp-up period where the soil was raised to the appropriate temperatures. The second operational stage encompassed the full power RF energy application. This operational stage is illustrated as Time Period 1 and Time Period 2 (described below) in Fig. 3. The final operation stage, the cool down period, began when the RF energy application was terminated. Condensate was collected during all the operational stages.

The objective of this evaluation was to determine if field SVOC removal rates during the full power RF energy application could be estimated from laboratory SVOC removal rates. However, the laboratory column studies were conducted at a constant temperature. Thus, for comparison to the column study removal rates, it was desirable to use rates from two periods where the soil temperatures were relatively stable. In this way, removal rate constants from the field could be compared to those from the constant-temperature laboratory column studies. In addition, the effect of temperature on field-scale SVOC removal could be investigated.

The RF-SVE system was operated at full RF power with relatively consistent SVE flow rates between day 45 and day 87. Thus, the condensate data for day 45 to day 87 were divided into two time periods of interest (Table 1). Between day 45 and day 64, the soil temperature hovered around the boiling point of water (94°C at the site). From day 64 to day 87 the soil temperature was significantly higher than the boiling point of water. The average temperature of the four monitoring wells and the SVE well (Fig. 1) was chosen to represent the in situ soil temperatures for the two time periods.

Table 1
RF-SVE demonstration time periods analyzed in this evaluation

Parameter	Time Period 1	Time Period 2
Average temperature (°C)	96	110
Duration (days)	19	23
Demonstration days	45–64	64–87
SVE flow rate (l/min)		
Average	385	272
Range	227–538	255–311

3.2. Soil data

Multiplying the average SVOC soil concentration by the mass of the impacted soil resulted in the SVOC mass in the soil. The pre-treatment and post-treatment masses of specific hydrocarbons in the volume of soil that reached 94°C are summarized in Table 2. The n-alkanes, C_{13}, C_{15}, C_{17}, and C_{19}, were chosen as representative compounds for the range of SVOCs affected by the RF-SVE at this site.

3.3. Removal rate constant calculation

The goal of this investigation was to determine if laboratory SVOC removal rate constants could be used to determine field SVOC removal rate constants. However, the laboratory column study removal rate constants were based on normalized data. Thus, for comparative purposes the field condensate data needed to be normalized by the SVOC mass in the impact volume of soil (Eq. (1)).

(Normalized SVOC mass remaining)

$$= 1 - \left(\frac{\text{SVOC mass removed (condensate drums)}}{\text{SVOC mass in impacted soil volume at start of time period}} \right) \quad (1)$$

Since the RF-SVE system had to be shut off and partially dismantled to obtain soil samples, SVOC masses in the soil could be only measured prior to and at the conclusion of the demonstration. Hence, the SVOC mass in the impacted soil volume at the start of the time period for evaluating temperatures of interest, $S_{\text{Start Day}}$, (denominator of Eq. (1)) was not measured and had to calculated. Fig. 4 illustrates how $S_{\text{Start Day}}$ was

Table 2
Mass of hydrocarbons in the field soil that reached 94°C

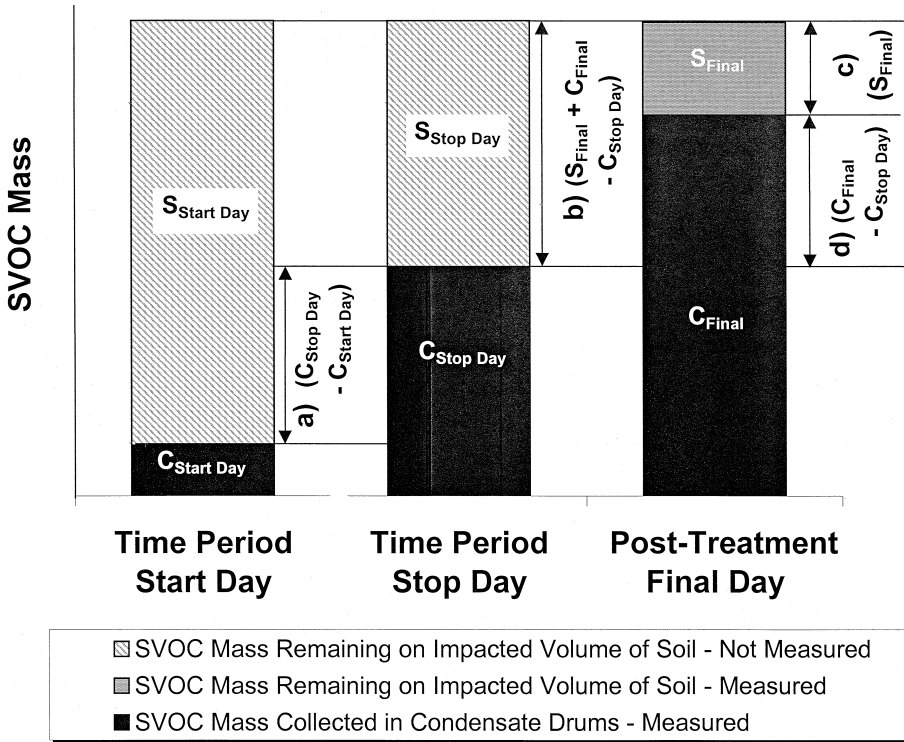| Compound	Pre-treatment mass (kg)	Pre-treatment mass standard deviation (kg)	Post-treatment mass (S_{Final}, kg)	Post-treatment mass standard deviation (kg)	Percent reduction
C_{13}	1.6	1.4	0.38	0.66	76%
C_{15}	4.4	3.7	1.4	1.9	68%
C_{17}	2.9	2.4	1.4	1.4	49%
C_{19}	1.2	1.0	0.86	0.69	26%

Fig. 4. Mass changes during operation of RF-SVE demonstration.

calculated as the sum of (a) the cumulative SVOC mass collected in the condensate knockout drums ($C_{\text{Stop Day}} - C_{\text{Start Day}}$) during the time period of interest plus (b) the SVOC mass remaining in the impacted volume of soil at the end of the time period, $S_{\text{Stop Day}}$. $S_{\text{Stop Day}}$ also could not be measured and had to be calculated as the sum of (c) the measured post-treatment mass in the impacted volume of soil (S_{Final}, Table 2) and (d) all SVOC mass collected in the condensate after the time period of interest ($C_{\text{Final}} - C_{\text{Stop Day}}$). The value, $C_{\text{Final}} - C_{\text{Stop Day}}$, includes all SOVC mass collected during the cool down period.

The SVOC mass removed at any time during a time period (numerator of Eq. (1)) was calculated by subtracting the cumulative hydrocarbon mass collected in condensate ($C_{(t)}$) from the total SVOC mass collected in the condensate drums prior to the first day of the time period ($C_{\text{Start Day}}$). In Fig. 3, $C_{(t)}$ for pentadecane is the data graphed using the left axis. It was assumed that all the SVOCs collected in the condensate originated in the defined impacted volume of soil. Eq. (2) shows the final normalized expression for the fraction of hydrocarbon compounds remaining in the soil over time.

Fraction remaining in soil (t)

$$= 1 - \left[\frac{C_{(t)} - C_{\text{Start Day}}}{\left(C_{\text{Stop Day}} - C_{\text{Start Day}} \right) + \left(S_{\text{Final}} + C_{\text{Final}} - C_{\text{Stop Day}} \right)} \right] \qquad (2)$$

The field SVOC fraction release data were fit using a first-order expression (Eq. (3)). In this first-order curve fit, k (1/h) is defined as the SVOC removal rate constant for the field demonstration.

$$\text{Fraction remaining in soil } (t) = \mathrm{e}^{-kt} \tag{3}$$

The field removal rate constants were used for comparisons with the column study removal rate constants.

3.4. Comparison of field and column study removal rate constants

Removal rate constants were determined for both the laboratory column studies and the field demonstration. However, different soil temperatures existed in the laboratory studies and the field demonstration. The field SVOC removal rate constants were calculated for the two time periods that represented average field soil temperatures of 96°C and 110°C. The column studies were conducted at 50°C, 100°C, 125°C and 150°C. A linear relationship between the column study removal rate constant and inverse temperature was determined [2]. This relationship was used to calculate column study removal rate constants at temperatures of 96°C and 110°C.

The calculated column study SVOC removal rate constants and the field-scale SVOC removal rate constants are compared in Fig. 5. There were a number of assumptions and estimations used to determine the field removal rate constants for the specific hydrocarbons. However, there was a linear log–log relationship between the field and column study removal rate constants. This relationship indicates that the column study SVOC removal rate constants can be correlated to field SVOC removal rates for the same soil.

The linear log–log relationship between the column study SVOC removal rate constants and the field scale SVOC removal rate constants is soil and site specific. In addition, it was derived using data obtained at the conclusion of the field study. The
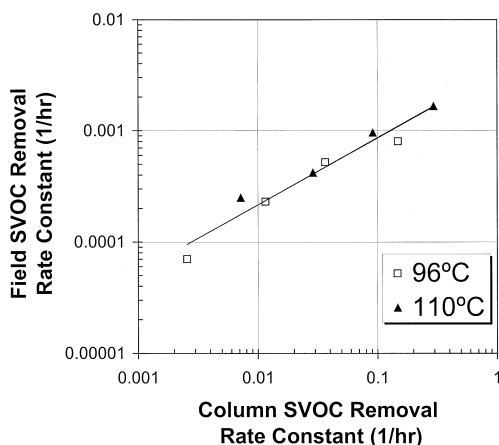

Fig. 5. Comparison of the field SVOC removal rate constants and the column study SVOC removal rate constants at equivalent temperatures.

column study removal rate constants are far more useful if they can be used to estimate field removal rate constants prior to full-scale field remediation operations.

3.5. Estimating field removal rate constants

To estimate the field SVOC removal rate constants from the column study rate constants it was assumed that the same physical and chemical processes controlled the SVOC release from the soil to the soil gas in both the field and laboratory. Previous work suggests that equilibrium partitioning from NAPL to soil gas controls a majority of the removal of alkanes from this soil at the temperatures of interest [2]. Hence, it was assumed that the SVOCs reached an instantaneous equilibrium between the soil gas and soil phases in both the laboratory and the field. This assumption means that the vapor pressures of the alkanes will strongly influence the estimated field removal rate constants of the alkanes. An increase in temperature will increase the alkanes vapor pressures and as result also increase the alkanes removal rate constant.

The above assumptions allowed the estimation of the field SVOC removal rate constants from the column study rate constants by comparing the time required to replace the soil gas volume in the two systems. Since the total gas flow rates were known for both the field and column study, only the volume of air in the soil, V_A (Eq. (4)), was needed to estimate the time to remove the soil gas.

$$V_A = \frac{M_S}{\rho_S}\left(\frac{n}{1-n}\right) - \frac{m_C M_S}{\rho_W} \tag{4}$$

The parameters and the values used to evaluate Eq. (4) are given in Table 3.

Each column study used 35 g of soil. In the field 70,300 kg of soil reached 94°C during the demonstration. The average density of the soil (ρ_S), soil porosity (n) and moisture content (m_C) determined during the RF-SVE field demonstration [3] were used in the field soil gas volume calculations. Since the column study used soil from the field site, the moisture content and soil density were assumed to be the same for both soil gas volume calculations. Changes in the density of water (ρ_W) with temperature were assumed to be negligible.

Table 3
Parameter values used to calculate soil gas volume

Parameter	Symbol	Value	Units
Mass of soil			
Column study	M_S	0.035	kg
Field demonstration		70,300	kg
Density of soil	ρ_S	2.70	kg/l
Porosity			
Column study	n	0.34–0.50	l pore volume/l total volume
Field demonstration		0.34	l pore volume/l total volume
Moisture content	m_C	0.07	g water/g dry soil
Density of water	ρ_W	1.0	kg/l

Table 4
Time required for soil gas removal

	Mass of soil (kg)	V_A (l)	Flow rate (l/min)	Time to replace V_A (min)
Column				
Porosity = 0.34	0.035	0.0042	0.014	0.30
Porosity = 0.50	0.035	0.011	0.014	0.75
Field				
Time Period 1	70,300	8500	390	22
Time Period 2	70,300	8500	270	31

Since the soil was loosely poured into the columns, it is unlikely that the porosity in the column study soil was equal to the field porosity. However, this parameter was not measured in the laboratory study. Thus, soil porosity values of 0.34 and 0.50 were used in the soil gas volume calculation for the column study to cover the possible range of soil porosities that may have existed in the column soils.

These values were then inserted into Eq. (4) and the time required for soil gas removal was calculated (Table 4). The field flow rates are from the RF-SVE demonstration report [3]. The time required to replace V_A by an equivalent volume of air moving through the soil was calculated by dividing V_A by the air flow rate.

The calculated column study removal rate constants (RRC) at the field temperatures were then used to estimate the field RRC using Eq. (5).

$$\text{Estimated field RRC} \left(\frac{1}{h} \right)$$
$$= \text{Column study RRC} \left(\frac{1}{h} \right) \left(\frac{\text{Column time to replace } V_A}{\text{Field time to replace } V_A} \right) \tag{5}$$

The actual field SVOC removal rate constants and the estimated field SVOC removal rate constants are compared in Figs. 6 and 7 at temperatures of 96°C and 110°C,
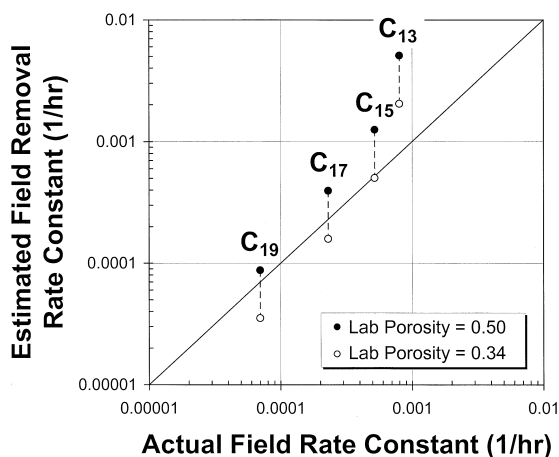

Fig. 6. Comparison of actual field and estimated field removal rate constants — 96°C.
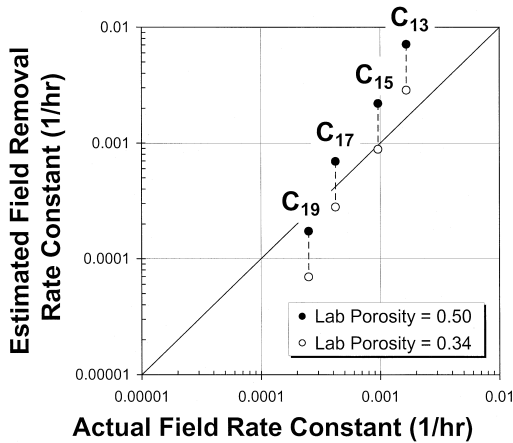
Fig. 7. Comparison of actual field and estimated field removal rate constants — 110°C.

respectively. The diagonal lines represent a perfect agreement between the estimated and actual removal rate constants. Each removal rate constant estimate is a range reflecting the range of estimated soil gas replacement times (Table 4). The solid black circles represent a column study porosity of 0.5. The open circles represent a column study porosity of 0.34.

Figs. 6 and 7 show that the estimated field removal rate constants were close to or within an order of magnitude of the actual field rate constants.

3.6. Estimation of mass removal

The estimated values for the removal rate constants were then used to predict the actual hydrocarbon fraction remaining in the soil at the conclusion of the RF-SVE demonstration. Since the estimated field removal rate constants were based solely on the column study removal rate constants and the parameters in Table 3, this fraction remaining could be estimated prior to the actual field implementation of a thermally enhanced SVE system.

The total hydrocarbon fraction remaining at the RF-SVE site at the conclusion of any given time period was the mass present at the beginning of the time period multiplied by the fraction removed during that time period. The fraction remaining at the beginning of the Time Period 2 of this evaluation was the fraction remaining at the conclusion of Time Period 1. Hence, the estimated total fraction remaining (at the conclusion of Time Period 2) was calculated by multiplying the fractions remaining in each of the two defined time periods (Eq. (6)). The fraction remaining was calculated using the estimated removal rate constants displayed in Figs. 6 and 7 (Table 5).

$$\left(\text{Estimated total fraction remaining}\right)$$

$$= e^{\left(-k_{96°C} \times 19\,\text{days} \times 24\,\frac{h}{\text{day}}\right)} e^{\left(-k_{110°C} \times 23\,\text{days} \times 24\,\frac{h}{\text{day}}\right)} \qquad (6)$$

Table 5
Comparison of actual field and estimated field fraction removed

Compound	Actual fraction remaining	Average estimated fraction remaining	Range of estimated fraction remaining
C_{13}	0.37	0.05	0.00–0.09
C_{15}	0.49	0.35	0.18–0.51
C_{17}	0.69	0.70	0.59–0.81
C_{19}	0.83	0.92	0.88–0.95

Despite the wide range in the estimated fraction removed, the average estimated fraction removed values were close to the actual fraction removed in the impacted volume of soil (Table 5).

Table 5 indicates that the relationship (Eq. (5)) over predicts the removal of the more volatile compounds (C_{13} and C_{15}). This reflects the fact that the estimated removal rate constants were higher than the actual field removal rate constant for C_{13} and C_{15} hydrocarbons. This means there was a greater C_{13} and C_{15} mass in the impacted volume of soil at the conclusion of the study than the column study rate constant predicted for these two SVOCs. Hydrocarbons migrating into the defined impacted soil volume from outside the impacted region may have been one source of the unexpected C_{13} and C_{15} residual mass in the impacted volume.

3.7. Estimating thermally enhanced remediation time

The value of using column studies to obtain field SVOC removal rate constants can be demonstrated by estimating treatment time prior to field scale implementation of a thermally enhanced SVE. Knowledge of treatment time allows the practitioner to better estimate thermally enhanced SVE system costs and benefits.

As the temperature of the soil is increased (increasing the cost of remediation), the vapor pressures of the alkanes will increase. Previous work [2] has shown that increasing both the alkane vapor pressure and soil temperature will increase the column study removal rate constant by a known amount. Since the estimated field removal rate constants are proportional to the column study removal rate constants, the time required to reduce field alkane concentrations decreases as both soil temperature and vapor pressure are increased.

By assuming that a thermally enhanced SVE system maintains a given temperature for a defined period, the time required to achieve a given field SVOC concentration reduction can be calculated (Eq. (7)). In Eq. (7), $k_{\text{Temperature}}$ (1/h) is the estimated field removal rate constant (Eq. (5)) at a specified temperature. This removal rate constant was derived solely from the parameters in Table 3 and the column study rate constant.

$$\text{Time required (h)} = \frac{\ln\left(\dfrac{\text{Desired soil concentration}}{\text{Initial soil concentration}}\right)}{-k_{\text{Temperature}}} \tag{7}$$
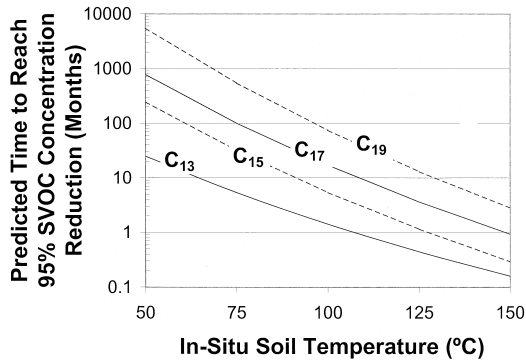
Fig. 8. Predicted time for a thermally enhanced SVE system to remove 95% of the specified hydrocarbon.

These times do not include initial temperature ramping nor cool down periods. At the RF-SVE demonstration site, less than 20% of the total mass removal occurred during the temperature ramping and cool down periods. In addition, this analysis does not include any chemical losses due to biodegradation. It was unlikely that there was biological activity at the temperatures achieved in the soil during the two temperature periods evaluated (average temperatures: 96°C and 110°C). However, biological degradation may have occurred during the temperature ramping period.

Fig. 8 shows the temperature dependence of the time required for a thermally enhanced SVE system to achieve a 95% concentration reduction of the specified SVOC for the soil used in this study. This figure demonstrates that increasing in situ soil temperatures from 100°C to 150°C will decrease the time required to achieve a 95% concentration reduction of nonadecane, C_{19}, using a thermally enhanced SVE, from 7 years to 3 months for the soil used in this study. For the data shown in Fig. 8, $k_{Temperature}$ was calculated using the relationships shown in previous work [2] and assuming that the ratio of field gas replacement time to the column gas replacement time was 62 (the average value for this study).

This study demonstrated that for a given soil, laboratory removal rate constants can be used to estimate field SVOC removals from a thermally enhanced SVE system. The laboratory study should be conducted using the field soil to be remediated at approximately field moisture and porosity values. Such estimated field removal constants can be used to estimate the time required for a field-scale thermally enhanced SVE system to reduce the concentration of individual compounds to a specified level.

Acknowledgements

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