Published in final edited form as:

Ground Water Monit Remediat. 2010 ; 30(3): 57–64. doi:10.1111/j.1745-6592.2010.01286.x.

ANALYSIS OF SOIL VAPOR EXTRACTION DATA TO EVALUATE MASS-TRANSFER CONSTRAINTS AND ESTIMATE SOURCE-ZONE MASS FLUX

Mark L. Brusseau¹, Virginia Rohay², and Michael J. Truex³

¹School of Earth and Environmental Sciences, University of Arizona

²CH2M Hill Plateau Remediation Company

³Pacific Northwest National Laboratory

Abstract

Methods are developed to use data collected during cyclic operation of soil vapor extraction (SVE) systems to help characterize the magnitudes and timescales of mass flux associated with vadose zone contaminant sources. Operational data collected at the Department of Energy's Hanford site are used to illustrate the use of such data. An analysis was conducted of carbon tetrachloride vapor concentrations collected during and between SVE operations. The objective of the analysis was to evaluate changes in concentrations measured during periods of operation and non-operation of SVE, with a focus on quantifying temporal dynamics of the vadose zone contaminant mass flux, and associated source strength. Three mass-flux terms, representing mass flux during the initial period of a SVE cycle, during the asymptotic period of a cycle, and during the rebound period, were calculated and compared. It was shown that it is possible to use the data to estimate time frames for effective operation of an SVE system if a sufficient set of historical cyclic operational data exists. This information could then be used to help evaluate changes in SVE operations, including system closure. The mass-flux data would also be useful for risk assessments of the impact of vadose-zone sources on groundwater contamination or vapor intrusion.

Introduction

Management of chlorinated-solvent contaminated sites that have contaminant sources located in the vadose zone requires consideration of long-term vapor-phase mass flux. Mass flux, also referred to as mass discharge, mass flow rate, and source strength, is pertinent to assessing risk, establishing groundwater-based cleanup objectives, and for evaluating the performance of soil vapor extraction (SVE) operations. Evaluation of long-term vapor-phase mass flux at field sites is a complex endeavor, complicated by subsurface material-property heterogeneity, non-uniform contaminant distributions, and nonideal contaminant-transport processes.

Soil vapor extraction is a widely used method for remediation of sites with chlorinated-solvent contaminated vadose zones. It has been widely observed that the effectiveness of SVE operations typically decrease with time. This behavior is often attributed to the impact of rate-limited mass transfer on contaminant transport. Rate-limited mass-transfer processes may comprise dissolution of organic liquid trapped in regions of relatively high water content, diffusive mass transfer between lower-permeability and higher-permeability

domains, desorption, or some combination thereof. Characterization of mass-transfer constraints is critical to evaluating the long-term performance of SVE operations.

One approach for characterizing mass-transfer constraints is based on analysis of SVE operations data. For example, during the course of a SVE operation, the vapor concentrations for samples collected at the extraction well(s) are at some point typically observed to decline asymptotically. These temporal extraction-concentration profiles (i.e., elution tails) can be analyzed to evaluate rate-limited mass transfer (e.g., Digiulio et al., 1998; USACE, 2002). These data can also be used to estimate mass flux and to estimate time scales for meeting specified cleanup objectives. When using operational data such as these, it is critical to distinguish between mass-transfer related effects and other potential causes such as constraints associated with design or operation of the SVE system (e.g., dilution effects due to atmospheric exchange, far-source borehole locations, or stagnation points).

When available, SVE rebound data can serve as an alternate or additional source of information to help characterize mass-transfer constraints. Periodic operation of a fluid extraction system, such as pump and treat or SVE (flow interruption or pulsed pumping), has been used for some time to evaluate the occurrence of rate-limited mass-transfer processes (e.g. Brusseau et al., 1989; 2007; Harvey et al., 1994; USACE, 2002; Switzer et al., 2004). In addition, SVE rebound data have been analyzed to provide information on contaminant source location (e.g., Riley, 1998; Switzer et al., 2004; Switzer and Kosson, 2007). As we will demonstrate, SVE rebound data can also serve as an alternate or additional source of information to help characterize the magnitudes and timescales of mass flux associated with vadose zone contaminant sources. Furthermore, data collected for multiple rebound periods can be used to characterize temporal dynamics of mass flux, thus providing a means to forecast future SVE performance.

Operational data collected at the Department of Energy's Hanford site are used to illustrate the results that may be obtained from analysis of cyclic SVE data. An analysis was conducted of carbon tetrachloride vapor concentrations collected during and between SVE operations. The objective of the analysis was to evaluate changes in concentrations measured during periods of operation and non-operation of SVE, with a focus on quantifying temporal dynamics of the vadose zone contaminant concentrations, mass flux, and associated source strength.

Site Background

Carbon tetrachloride was found in the unconfined aquifer beneath the 200 West Area at the U.S. Department of Energy's Hanford Site in the mid-1980s. During this time, groundwater monitoring results indicated that the carbon tetrachloride plume was widespread and that concentrations were increasing. The source of the carbon tetrachloride contamination was determined to be waste liquids discharged to the vadose zone through engineered infiltration facilities. The carbon tetrachloride, which was contained in aqueous and organic-liquid wastes generated during plutonium-processing operations, was discharged primarily to the following infiltration facilities:

- 216-Z-9 Trench from 1955 to 1962 (referred to as the "Z9" site in this study)
- 216-Z-1A Tile Field from 1964 to 1969 and the adjacent 216-Z-18 Crib from 1969 to 1973 (together referred to as the "Z1" site in this study).

The vadose zone underlying the primary carbon tetrachloride disposal sites consists of approximately 70 m of relatively permeable sand and gravel within the Ringold Formation (lower portion) and Hanford Formation (upper portion). The Cold Creek Unit (CCU), a less-

permeable interval composed of 4 m of silt and sand and 3 m of carbonate-rich silt and sand, resides at a depth of approximately 38 to 45 m. A generalized cross section of the subsurface beneath the waste disposal sites is presented in Figure 1.

Soil vapor extraction (SVE) was initiated in February 1992 to remove carbon tetrachloride contamination from the vadose zone in the vicinity of these disposal sites. The objective of the remediation effort is to mitigate the threat to the environment caused by the migration of carbon tetrachloride vapors through the vadose zone and into the groundwater. Since February 1992, soil vapor extraction has been operated as an interim action taken pending final cleanup activities for these waste sites. The final cleanup activities will be determined as part of a Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA) remedial investigation/feasibility study (RI/FS) process.

The SVE system was operated in continuous mode between 1992 and 1996 (with downtime in 1993), using up to three SVE systems with design capacities of 14.2 m³/min, 28.3 m³/min, and 42.5 m³/min. The carbon tetrachloride mass recovery during the continuous operation is shown in Figure 2. In November 1996, a rebound study was initiated to determine if the carbon tetrachloride vapor concentrations would increase or rebound following temporary system shutdown. The operating strategy was modified based on the results of the rebound study and the declining rate of carbon tetrachloride removal during continuous extraction operations. The SVE system has been operated in a cyclic or periodic mode since 1997, with approximately 3 months of extraction at each location (Z9 and Z1) followed by an extended period of no extraction to allow time for carbon tetrachloride vapor concentrations to rebound.

Analysis of the data collected over the past 12 years shows that the vapor-phase concentrations of CT increase over time during the non-operation periods. Thus, concentrations at the beginning of the next SVE operation cycle are larger than at the end of the prior cycle. During the course of each operation cycle, the concentrations decline asymptotically. This behavior is illustrated in Figure 3, which shows the measured concentrations of CT in the extracted soil gas for two consecutive operational cycles. The observed behavior suggests that there is contaminant mass present within portions of the subsurface for which access and removal does not occur solely by gas-phase advective transport. The results of prior characterization efforts conducted at the site suggest that much of this poorly-accessible mass is likely associated with the lower-permeability CCU.

A total of 52 wells are available for use in the SVE operations. Thirteen of the 52 wells have two open intervals (i.e., screened or perforated casing), creating 65 intervals for vapor extraction. Thirteen of these 65 intervals are screened wholly or partially within the CCU. A subset of the intervals are used for the majority of flow. Screened interval lengths range from 1.5 to 22 m. Well diameters range from 2 to 20 cm (1 to 8 in.).

Data Analysis Methods

Mass Flux Analysis

For the mass flux analysis, the vapor-phase CT concentrations reported for the intake into the treatment system were used. These concentrations represent a composite or integrated concentration for the entire domain influenced by the SVE system under induced-gradient conditions. It is assumed herein that the SVE system encompasses the entire contamination zone, such that the observed temporal contaminant distributions are not influenced by capture of contamination from outside the SVE-influenced domain. The maximum mass flux (MF_m, in kg/d) was calculated as the product of the maximum CT concentration (C_m) and the mean SVE extraction flow rate at the beginning of the SVE operation cycle. The

asymptotic mass flux (MF_a) was calculated as the product of the asymptotic concentration (C_a) and the mean SVE extraction flow rate observed during the asymptotic phase of the preceding SVE operation cycle. Both of these quantities represent induced-gradient conditions. The equation to determine normalized mass-flux rebound (NR) is: $NR = (MF_m - MF_a)/T$. The rebound is normalized by the time (T) comprising the non-operation period that preceded the SVE cycle to account for the fact that the non-operation (or rebound) periods were not uniform. Simple linear normalization was selected for this analysis given the lack of information to support higher-order approaches. Regression analysis was conducted to evaluate the relationship between the normalized rebound and cycle (i.e., the temporal dynamics of the rebound).

A mass flux value (MF_r) was also calculated to estimate mass flux associated with diffusive mass transfer of contaminant from low-accessibility domains (e.g., the CCU) to the advective domain during the non-operation (rebound) period between each operation cycle. This value represents mass flux under natural-gradient conditions. This value was determined by first calculating the total mass of contaminant removed (M_{pv}) for the first gas-pore-volume extracted during SVE operation:

$$M_{pv} = \sum_{i=1}^n C Q T_s$$

where C is the contaminant concentration in the extracted soil gas (ML^{-3}), Q is the extraction flow rate (L^3T^{-1}), and T_s is the interval between sample times (T), and n is the sample where one pore volume of gas has been extracted. The pore volumes associated with the SVE systems (approximately 600,000 m^3) were obtained from prior characterization work conducted at the site (Rohay and McMahon 1996). The calculation of mass extracted is based on the assumption that the contaminant mass removed during the first pore volume equivalent of gas extraction represents primarily the mass present in the advective domain. It is further assumed that there is minimal CT mass present in the advective domain prior to the start of a rebound period and, therefore, all mass removed in the first pore volume of extraction represents mass that transferred from the low-accessibility domains during the preceding rebound period. This assumption is based on a qualitative analysis of spatial distributions of CT concentrations, which indicated that there is only a relatively small quantity of CT present in the advective domain prior to the start of a rebound period. M_{pv} was divided by the time of the non-operational period prior to the operational cycle to compute MF_r . It should be noted that the calculated mass-flux values may be conservatively low in that the use of a single pore volume assumes ideal removal (piston displacement). In addition, the length of the rebound periods may have been longer than the approximate equilibration time in some cases. In an attempt to partially account for this latter effect, the time required to attain maximum concentration, which was assumed to represent an approximate equilibration time for the mass-transfer process, was estimated using data collected at a well screened within and adjacent to the CCU for site Z9. This time was used in place of the total rebound time to calculate MF_r . Significant differences in equilibration time versus total rebound time were observed for only a small subset of the data.

Concentration Rebound Analysis

Concentration rebound data collected during the non-operation periods at selected individual monitoring wells (non-extraction wells) and extraction wells were analyzed. Four representative monitoring wells were selected, all located within the Z9 site, to illustrate the wide range of rebound behavior observed. The first is 299-W15-217, which is an extraction well screened within and adjacent to the CCU. The second is 299-W15-82, an extraction

well screened approximately 10 m above the CCU. The third is CPT-24, for which two individual point samplers were sampled, one emplaced within the CCU and one a few tens of meters above the CCU. The fourth is CPT-16, for which all of the point samplers are located more than 10 m from the CCU. The reported concentrations for the latter location represent a composite of all point-sampling locations. For this analysis, the maximum rebound concentration for each period was compared as a function of operation cycle.

Results

Characterizing Source-Zone Mass Flux

The mass flux values MF_m , MF_a , and MF_r are reported in Table 1. Concentration rebound is observed after each operational cycle of SVE, as illustrated in Figure 3. This rebound is attributed to mass transfer of contaminant residing within regions of the subsurface for which advective transport is limited (e.g., the CCU). Changes in pneumatic-gradient conditions, such as those resulting from periodic SVE operation, are expected to impact mass flux for systems with mass present in poorly-accessible domains. For a system wherein mass removal is primarily influenced by mass-transfer-limited conditions, the concentrations and associated mass flux observed during extraction of the first equivalent pore volume of gas in an SVE operation period represent primarily mass released from the poorly-accessible domains under natural-gradient conditions during the preceding non-operation period. This mass flux was estimated through calculation of the rebound mass flux (MF_r) quantity. Conversely, the concentrations and mass flux observed during the asymptotic phase of SVE operations (C_a , MF_a) represent primarily mass released from the poorly-accessible domains under induced-gradient conditions. The mass flux values calculated for the rebound and asymptotic phases can be compared to evaluate behavior under the two conditions. In conducting such a comparison, it is important to note that the values for MF_r generally have a greater degree of uncertainty due to the method of calculation, as noted above.

Inspection of Table 1 indicates that the mass flux values representing natural-gradient conditions (MF_r) are smaller than those representing induced-gradient conditions (MF_a). Interestingly, the MR_a values were 6–7 times larger during the initial phase of cyclic operation, whereas the two terms currently differ by approximately a factor of two. This reduction possibly reflects a decrease in the efficiency of mass removal obtained during SVE operation. Characterizing the relative magnitudes of MF_a and MF_r may serve as a means by which to evaluate the mass-removal effectiveness of a SVE operation. As the value of MF_a converges to that of MF_r , the SVE system in essence ceases to be an enhanced mass-removal operation and becomes a containment operation. At this point, the system may be considered ineffective based on mass-removal objectives. However, it still may be effective based on other factors influencing overall risk, which could be evaluated based on the actual values of MF_r .

The MF_r term represents mass flux under the natural-gradient conditions associated with the non-operation periods, and thus would be representative of mass flux after SVE is terminated. As such, this term may serve as contaminant-source input data for analyses of the impact of vadose-zone contamination on groundwater concentrations (or for vapor intrusion analyses), and therefore be of interest to regulators for evaluating termination of SVE operations. The use of SVE data to calculate MF_r serves as an alternative to existing approaches that may reduce uncertainty, particularly for heterogeneous systems.

Evaluating Effectiveness of Cyclic Operation

Both the maximum (MF_m) and asymptotic (MF_a) values have generally declined with cycle (Table 1). Interestingly, it appears that the magnitudes of the maximum and asymptotic

values are converging over time for both sites. This is reflected in the decline in normalized mass-flux rebound (NR) with cycle, which is observed for both sites (Figure 4). The normalized rebound is generally larger for the Z9 site than for the Z1 site, but the slopes of the regressions are statistically identical.

Convergence of MF_m and MF_a suggests that cyclic operation is becoming less effective. Under these conditions, in conjunction with evaluation of other factors that are important to this type of decision, transition to other SVE operational modes, transition to other remedial technologies, or termination of remediation should be considered. The analysis of normalized rebound establishes a trend that represents the likely progression of SVE removal efficiency under continued cyclic operational conditions. The regression equation obtained from the data analysis can be used to predict the number of cycles required to attain a condition wherein C_m and MF_m are not significantly different from C_a and MF_a , respectively, thus providing a means to estimate time frames of effective cyclic SVE operation.

Time-series Concentration Rebound Analysis

The maximum rebound concentration as a function of non-operation period for individual well locations are presented in Figure 5. The maximum rebound concentrations are much greater for the locations screened within and adjacent to the CCU, and are smaller for the locations screened away from the CCU. For extraction well 299-W15-217, the maximum rebound concentrations have decreased by two orders of magnitude over the study period. Conversely, they have decreased by approximately one order of magnitude for extraction well 299-W15-82 (data not shown), which is screened above the CCU. Similarly, the maximum rebound concentrations have decreased by approximately a factor of two for the point sampler of CPT-24 that is located within the CCU, whereas the maximum rebound concentrations have not changed over the entire time for the point sampler of CPT-24 that is located above the CCU (data not shown). There has also been no decrease in maximum rebound concentrations for CPT-16 (Figure 5). The disparity in rebound behavior reflects the proximity of each monitoring location to the CCU, and hence the relative impact of mass removal from this zone, which is considered to be the primary source of contaminant mass.

Assessing Rates of Mass Flux

The removal of mass from “non-advective” or low-accessibility domains would entail mass transfer to the gas-phase advective domain, which may be influenced by rate limitations. It is likely that contaminant mass removal associated with the current SVE operations at the site is controlled by this rate-limited mass transfer. There are established methods whereby data from a single rebound period can be used to characterize characteristic times of mass transfer (e.g., Brusseau et al., 1989; Harvey et al., 1994; USACE, 2002). The characteristic time for transfer of contaminant mass between the poorly-accessible and advective domains can be roughly estimated by conducting a simple Damkohler Number analysis incorporating the residence time associated with the SVE operation. The Damkohler Number represents a ratio of the residence time and the characteristic time of mass transfer. It has an operational range from approximately 0.01 to 100, with values of approximately 10 or greater indicating near-equilibrium conditions. The residence time can be estimated as the pore volume associated with the SVE system divided by the SVE flow rate, which produces a value of approximately 46 days for the Z9 site. Given that mass transfer is rate-limited during SVE operation (i.e., Damkohler Number much less than 10), as indicated by the rebound behavior, the characteristic time for mass transfer must be approximately similar to or greater than the operational residence time.

The characteristic time for transfer of contaminant mass between the non-advective and advective domains can also be roughly estimated by evaluating the time scales associated with contaminant rebound during the non-operation periods. These periods have ranged from approximately 6 months to 24 months. Analysis of individual-well data showed that the time required to reach the maximum concentration during the non-operation periods was approximately 8 months initially and approximately 5 months for the past few cycles for well 299-W15-217 located within and adjacent to the CCU. Treating mass transfer as a first-order process to simplify, and considering the time scale for 99% mass removal (7 half lives), results in an estimated characteristic time of approximately 1 month. This characteristic time translates to a rate coefficient of approximately 0.02 d^{-1} . In interpreting this value, and comparing to values obtained from other sites or from laboratory studies, it is important to note that the characteristic time and associated rate coefficient are composite values, representing all mass-transfer processes operative at the site. Mass-transfer coefficients could also be obtained by applying specific mathematical models to the rebound data (e.g., Brusseau et al., 1989; Harvey et al., 1994; USACE, 2002).

Conclusion

In prior work, SVE rebound data has been analyzed to provide information on the location of contaminant sources and associated mass-transfer constraints. As demonstrated herein, SVE rebound data can also serve to help characterize the magnitudes and timescales of mass flux associated with vadose zone contaminant sources. If a sufficient set of historical cyclic operational data exists, it may be possible to use the mass-flux data to evaluate the effectiveness of an SVE system. This information could then be used to help evaluate changes in SVE operations, including system closure. For example, comparison of MF_a and MF_r values, and of MF_m and MF_a values, respectively, were discussed. Convergence of the paired values may indicate that removal of remaining mass is influenced by mass-transfer processes whose characteristic times are much greater than the time scales associated with the SVE system operation, and/or that mass is being captured from outside of the effective operation domain. Under these conditions, modification of the SVE system or closure and transition to other remedies should be considered. Additionally, the natural-gradient mass flux (MF_r) can be used as an estimate of the contaminant flux that would exist if SVE is terminated. This value can be used as input for risk-based analyses of future contaminant flux to groundwater or land surface.

It is important to note that there are several limitations associated with the analyses reported herein. Non-uniformity in the SVE operation cycles with respect to the time of operation, the time of non-operation, and the number and sequence of wells used, as was the condition for the case study, imparts uncertainty to the calculated values. This uncertainty is of special significance for use of the data to provide predictions of future behavior. Thus, maintaining consistent operational conditions during cyclic operations would be preferred for applying this approach for trending analysis. Additionally, the analyses employ underlying assumptions that simplify contaminant mass-transfer processes. However, the method serves as a useful means of using available SVE operational data to provide insight into subsurface contaminant transport processes, estimate mass-flux quantities for use in remediation decisions, and evaluate trends in SVE performance during cyclic operations. It should be stressed that analysis of SVE operational data is one of many components of a complete assessment for evaluating SVE transition or closure.

Acknowledgments

This work was funded through the Department of Energy's Office of Environmental Management as a part of a project under Engineering and Technology, Groundwater and Soil Remediation, with additional support from the

NIEHS Superfund Research Program (ES04940). We thank Dave Becker (US Army Corps of Engineers) and the reviewers for their thoughtful and constructive comments.

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Figure 1. Generalized cross section of the subsurface beneath the waste disposal sites. bgs = below ground surface.

(a)

(b)

Figure 2.

Cumulative mass recovery of carbon tetrachloride during continuous SVE operations, 1992 through 1996 (a) and 1997 through 2008 (b). Data points correspond to the left axis.

Figure 3.
Carbon tetrachloride concentration in extracted soil gas for the 1998 and 1999 operational cycles of the Z9 site SVE system.

- A. Normalized mass flux rebound for the Z9 site. Input mass flux in units of kg/d. Rebound time in units of months. Regression equation: $\log NR = -0.18\text{Cycle} + 0.29$, $r^2 = 0.64$.

- B. Normalized mass flux rebound for the Z1 site. Input mass flux in units of kg/d. Rebound time in units of months. Regression equation: $\log NR = -0.13\text{Cycle} - 0.47$, $r^2 = 0.56$.

Figure 4.

Figure 5.

Maximum concentration rebound for three selected wells at the Z9 site. Regression equation (for 299-W15-217): $\log C = -0.2\text{Cycle} + 3.4$, $r^2 = 0.75$. The data for CPT-24 are from the point location placed within the CCU.

Table 1

Mass Flux Values (kg/day).

Cycle	MF _m	MF _a	MF _r
<i>Z9 Site</i>			
1	30.6	7.9	1.1
2	9.1	6.7	1.2
3	4.3	2.5	0.9
4	10.5	2.9	0.9
5	5.6	3.3	1.9
6	7.2	1.8	NA
7	5.8	2.3	1.4
8	2.4	1.7	0.4
9	1.3	1.0	0.7
10	1.3	1.2	0.6
<i>Z1 Site</i>			
1	7.6	3.2	NA
2	2.6	3.4	1.1
3	4.0	2.1	0.4
4	3.6	2.4	0.2
5	2.5	2.1	0.3
6	2.1	1.7	0.5
7	1.2	0.8	0.3
8	1.1	1.1	0.2
9	1.0	0.9	0.3
10	1.3	0.6	0.1
11	1.5	1.3	0.3

MF_m is mass flux during the initial period of SVE operation

MF_a is mass flux during the asymptotic period of SVE operation

MF_r is mass flux associated with the non-operation (rebound) period