Vadose Zone Profiling to Better Understand Vadose Zone Processes Related to Vapor Intrusion

Paper #

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ABSTRACT

This paper is focused on vadose zone processes and high resolution characterization as a means of refining site-specific conceptual models for vapor intrusion investigations. Much of the existing regulatory guidance is driven by the perception that vapor intrusion potential and indoor air quality are mainly a function of contemporaneous groundwater quality with limited recognition of vadose zone processes. In addition, regulatory policies founded on limited field experience are evolving toward not recognizing the use of exterior subsurface gas investigations as a component of vapor intrusion investigations. We believe that programs of vadose zone characterization including multilevel monitoring of gas phase can yield improved understanding of site conditions.

As shown in the first section of the body of this paper, data and observations from project work at soil and bedrock sites indicate that spatial and temporal variability often observed in vapor intrusion investigations can typically be explained by variations in subsurface conditions relevant to vapor migration, as well as seasonally variable infiltration and soil moisture conditions. Site data confirm that considerable volatile organic compound (VOC) mass may be partitioned into soil pore water and sorbed to soils in the vadose zone. This VOC mass can contribute to vapor intrusion potential and influence the longevity of this pathway even after substantial improvements in groundwater quality.

To aid in illustrating the combined influence of vadose zone processes a simple one-dimensional model of countercurrent vapor diffusion and infiltration was developed. The modeling was conducted using SESOIL, a model used historically to develop soil cleanup guidelines. The SESOIL modeling was completed with the goal of providing insight into the processes that lead to spatial and temporal variability in vapor concentrations and the potential longevity of VOC mass in the vadose zone. The results illustrate stylistically how mass partitioning among gas, aqueous, and sorbed phases within the vadose zone may influence vapor intrusion potential such that vapor conditions observed at a given place and time can be somewhat independent of groundwater quality. The term "stylistically" has been used by Parker[1] to reflect modeling that is not intended as an absolute simulation of specific site conditions but to indicate the style of transport. The modeling also illustrates the mechanisms behind seasonal variability in subsurface vapor concentrations.

INTRODUCTION

In the last ten years, vapor intrusion has become widely recognized in the U.S. and abroad as a potential pathway of human exposure. This recognition has led regulatory agencies, as well as industry and governmental consortiums, to publish guidance, policies, and rules to assess the vapor intrusion pathway. It has also led to the re-examination of sites that in some cases were first investigated more than two decades ago. The historical site characterization work has typically been focused on the saturated zone and, therefore, the available data is often insufficient to develop an adequate conceptual model, let alone to model or accurately predict vapor intrusion potential. Although there was substantial research[2,3,4] conducted in the late 1980s to the mid 1990s related to vadose zone processes and VOC vapor transport, there is not a scientific consensus on characterization techniques or the importance of the governing processes which are believed to be influenced by both subsurface conditions and building characteristics. It is generally accepted, however, that diffusion and advection are the primary mechanisms for transport of vapors in the subsurface.

The complex interactions between building conditions and the subsurface are recognized to result in a high degree of variability in indoor air quality despite similar groundwater source concentrations. In a 2008 presentation, the U.S. Environmental Protection Agency (USEPA)[5] concluded – based on empirical evidence from multiple granular soil sites – that for a given groundwater concentration, there may be variability in indoor air quality by as much as four orders of magnitude. We believe that spatial and temporal variability in vadose zone conditions relevant to vapor transport and VOC mass transfer among phases often explain some of this apparent variability.

Detailed characterization of the vadose zone, multilevel monitoring of vapor and modeling of vapor transport were summarized in a paper by Conant et al. [6] The field studies at the Canadian Forces Base Borden field site included measurement of soil texture and moisture and organic carbon content at vertical scales of centimeters, as well as multilevel soil gas monitoring at a sub-meter scale. The field data were compared against numerical model predictions.

On the basis of numerical modeling, the study concluded that sorption on organic carbon “has a very strong influence on relative migration rates of trichloroethene (TCE) vapor”. Sorbed and aqueous phase TCE was estimated to comprise about 50 to 70% and 15 to 26% of the total mass present in the vadose zone, respectively. Although the variations in soil moisture content were not found to have a strong influence on vapor migration in relatively permeable, higher porosity soils at Borden, it was concluded that “variations in moisture content could have a pronounced effect on vapor transport at other sites through its effect on gas phase permeability (k) and the effective diffusion coefficient (D_{eff})”.

Johnson[7] identified subsurface properties related to effectiveness of diffusion and advection – specifically D_{eff} and k – as some of the key primary input variables for modeling of vapor intrusion. Typically, these properties are non-linear functions of water- and gas-filled subsurface volumes, which may vary considerably in both space and time for a given soil type. Consequently, the properties may span several orders of magnitude across the range of moisture saturation. They cannot be readily measured and are typically estimated, assumed, or dictated by regulatory guidance.

The findings of two-dimensional, transient numerical modeling by Yu et al.[8] support the sensitivity of vapor transport by advection and diffusion to the infiltration rate and conclude that the “diffusive flux across the capillary fringe is the limiting process controlling indoor air impacts”.

Despite the influence of vadose zone processes on vapor intrusion potential, its characterization is materially addressed only in a small proportion of the available guidance. Most notably, New Jersey[9] and California[10,11] have allowed for development of a site-specific groundwater screening value on the basis of detailed characterization of soil texture. The text to follow presents a broad overview of vadose zone characterization work with examples of data and observations from field exploration and testing programs at vapor intrusion sites. This section is intended to introduce key concepts and the context for the stylistic[1] modeling. The details of the model application and results in relation to the field data and observations are outlined in the third major section of the document. The “Summary” section outlines our key conclusions.

FIELD OBSERVATIONS OF VADOSE ZONE CONDITIONS

Data and Observations from High Resolution Vadose Zone Profiling

In practice, characterization of the vadose zone involves sampling and testing for the physical properties of soil or rock materials and the development of site-specific estimates of parameters related to vapor transport. Analysis of soil and rock for VOCs has also proven valuable. When combined with multilevel gas monitoring, such analysis allows for quantitative evaluation of vapor transport by documenting concentration gradients that drive diffusion.

Applied with high resolution, vadose zone profiling would involve continuous sampling of vadose zone materials, preferably using methods that would retrieve minimally disturbed samples. Depending on lithology, the proper scale of sampling and laboratory analysis may be on the order of feet or less, informed by detailed field logging of texture, moisture, and fracturing. The focus is on identifying and quantifying conditions either conducive or limiting to vapor transport. As shown on Figure 1, wet soils near the water table within the capillary fringe and at capillary barriers that can be found at contacts between finer and coarser soil horizons[12] would be expected to limit vapor transport by both diffusion and advection. The position of “limiting layers” relative to foundation depth has been found to be important in assessing site conditions.

Figure 2 provides an example of a relatively low-resolution soil texture and moisture profile combined with vapor concentration data observed in nearly six years of multilevel vapor monitoring. The profile is comprised of well- and poorly-sorted granular soils with occasional interbeds of silt shown as “apparent limiting zones”. Laboratory analysis of less than a dozen soil samples indicates soil moisture saturation ranging from about 30 to 60% expressed as a fraction of soil porosity which ranges from about 0.2 to 0.35.

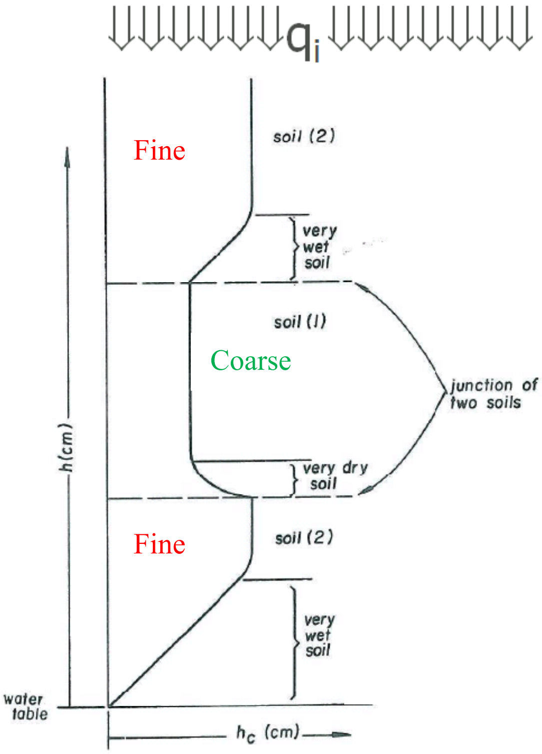
In this instance, the amount of water present in the vadose zone at the time of investigation was estimated to be the equivalent of about 40 inches of water or about two years of estimated infiltration.

Data recorded over 6 years of multilevel vapor monitoring indicate a decrease by about two orders of magnitude in vapor concentration from near water table depth to foundation depth. The concentrations observed near foundation depth on the order of 10,000 micrograms per cubic meter (µg/m^3) are about what would be expected from equilibrium partitioning with groundwater assuming a few tens of micrograms per liter (µg/L) of TCE. More typically, vapor concentrations are found to be one-half to two orders of magnitude or more below theoretical equilibrium conditions. Most of the concentration gradient occurs across high moisture content soils located between foundation depth and about 22 feet below ground. These conditions likely control the magnitude of diffusive flux and advection across this profile. The data support greater seasonal variability in concentration with distance from the water table.

Figure 1: Vadose Zone Concepts (Adapted by Sanborn Head from Corey[12])

Key Concepts

1. Transport occurs in pore space filled with liquid and gas.
2. Liquid and gas saturations are inversely proportional.
3. A high state of liquid saturation limits vapor transport by diffusion and advection.
    - As infiltration (q_i) approaches K_{sat}, the media approaches a water-saturated state.
    - Lithologic layering can result in capillary barriers that create a near-water saturated state even where q_i is less than K_{sat}.
    - Even thin zones can limit transport.



Phase Partitioning

The diagram included as Figure 3 depicts theoretical proportioning of mass among phases based on published partitioning coefficients for TCE.

Consistent with the findings of Conant et al.[6] the majority of the VOC mass is expected to be present in aqueous and sorbed phases across the range of soil moisture conditions observed in the example profile. In our experience, the observed vapor concentrations shown on Figures 2 and 4 imply the potential for soil concentrations ranging from hundreds to a few micrograms per kilogram on a dry weight basis (µg/kg). Changes in soil moisture conditions under seasonally variable recharge are believed to explain in part the apparent temporal variability in soil vapor concentrations that is typically observed at vapor intrusion sites. The example data set shown on Figure 4 is derived from foundation and water table depth. The data show over one-half order of magnitude seasonal fluctuations in TCE concentrations near foundation depth and more muted fluctuations near water-table depth. The fluctuations are repeatable and progressive trends.

Figure 2: Example of Soil Texture, Moisture and VOC Vapor Concentration Profile

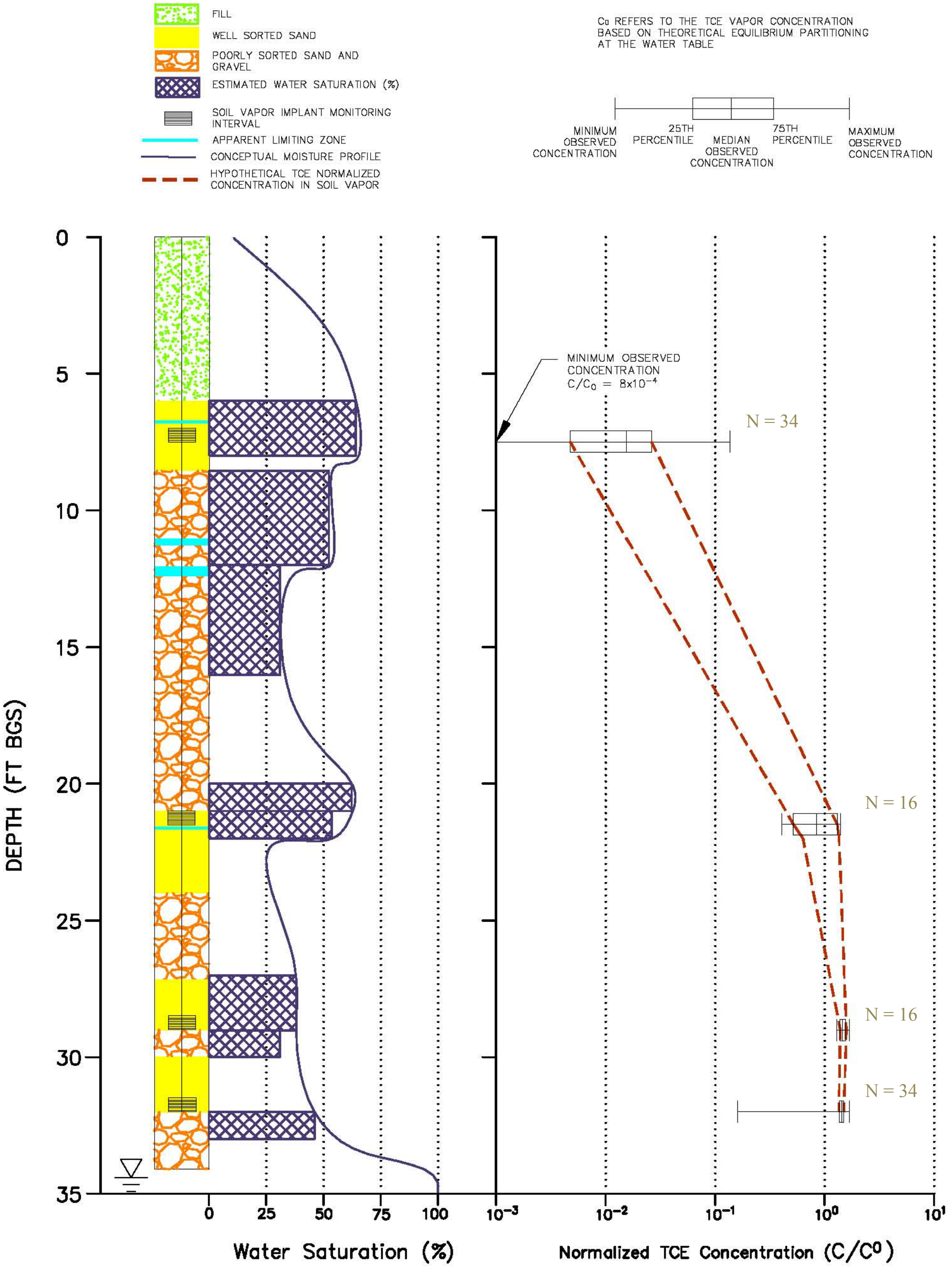


Figure 3: Example of Phase Partitioning

Mass Storage and Transfer:

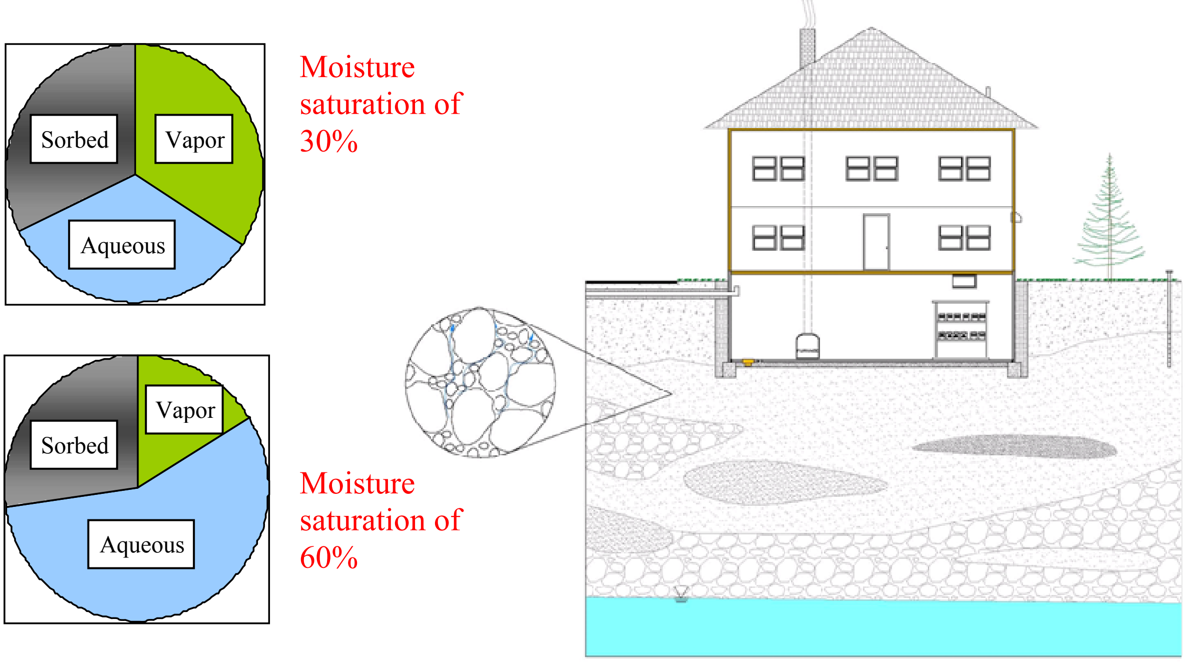
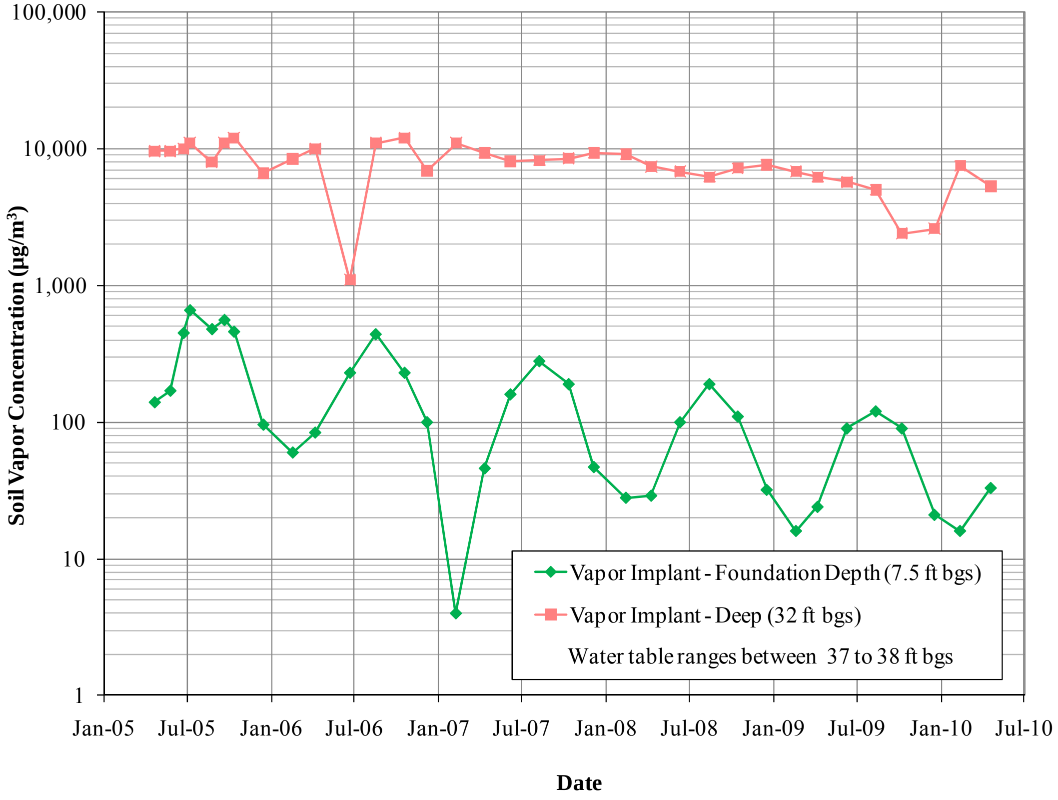


Figure 4: Seasonally Variable Soil Vapor Concentrations



MODELING OF VADOSE ZONE TRANSPORT

We developed a simplified application of the commercially available Seasonal Soil Compartment (SESOIL) model. SESOIL is a one-dimensional analytical model used to simulate vertical transport in the unsaturated soil zone. It was originally developed under contract with USEPA.[13] The model has routinely been used for estimating the potential impacts of surface spills on the water table and for development of site-specific soil cleanup standards. As illustrated on Figure 5, SESOIL accounts for water balance components of the hydrologic cycle (e.g., rainfall, surface runoff, soil moisture storage, evapotranspiration, and groundwater recharge), and contaminant fate processes including advection, vapor-phase diffusion, volatilization, linear/non-linear sorption, and chemical reactions. The model is capable of simulating time-varying contaminant loading, a heterogeneous initial contaminant distribution, and up to four soil layers with of varying properties.

The example simulation as detailed by Figure 5 was developed for tetrachloroethene (PCE) in a well-sorted sand profile. The initial condition is intended to reflect a simplified representation of a hypothetical mature vapor intrusion condition with PCE concentrations decreasing linearly from the water table to ground surface with partitioning into sorbed, aqueous, and vapor phases. The simulation was conducted to assess the potential style of transport assuming substantial cleanup of groundwater and, therefore, elimination of sourcing at the water table and allowing for desorption of VOC mass from soil and transport upward via diffusion and downward under seasonally variable infiltration.

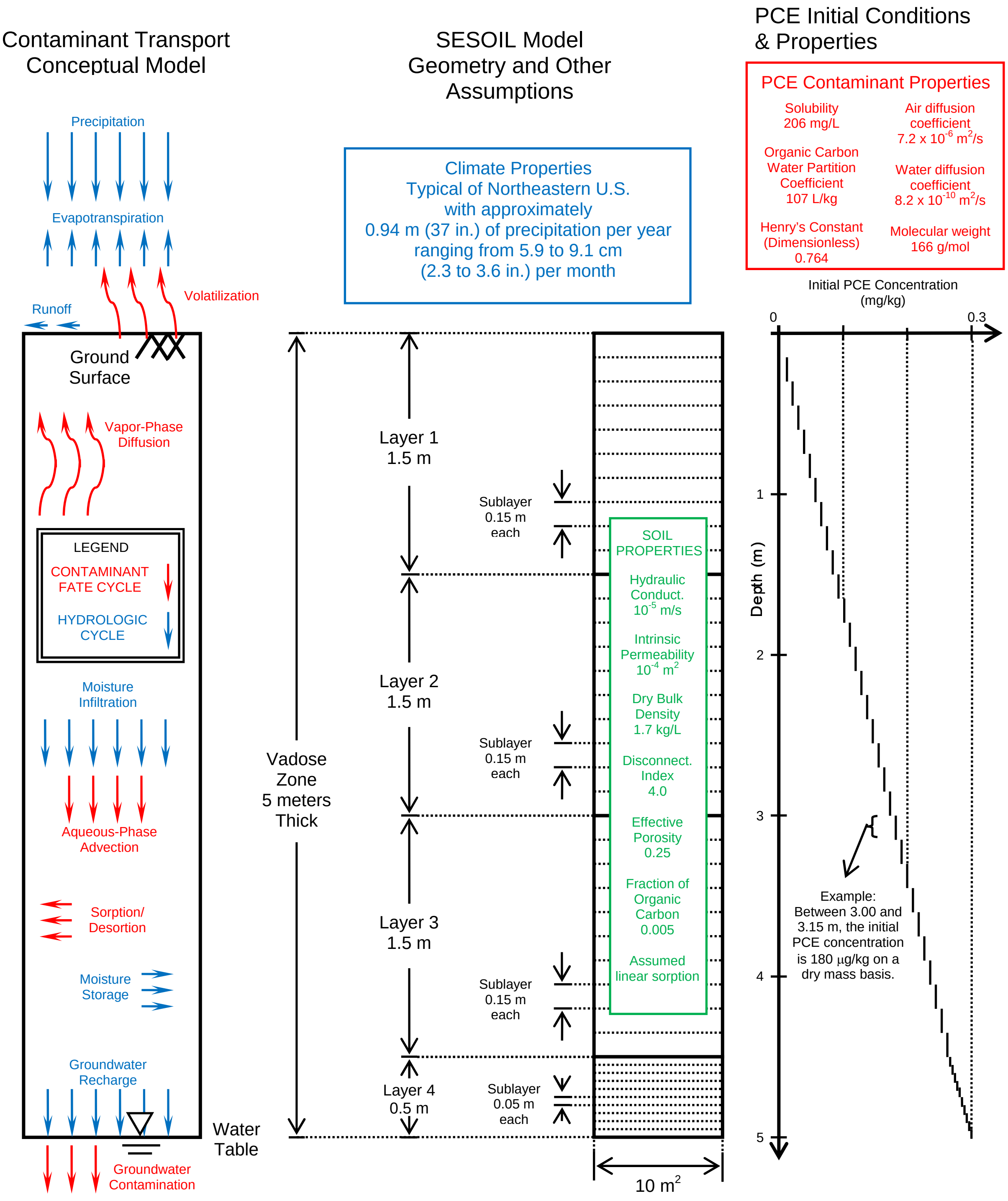
Model and Initial and Boundary Conditions

The model was developed for a 5-meter (m)-thick vadose zone (about 16 feet) divided into four layers, with the top three layers divided into equal thickness of 1.5 m, and a lower layer corresponding to the capillary fringe at a thickness of 0.5 m. For mass balance calculations each layer was subdivided into 10 sublayers of equal thickness as illustrated on Figure 5.

As detailed on Figure 5, the simulation was conducted using weather data believed generally representative of the humid northeastern U.S. As an additional simplification, the weather data was not varied year to year over the ten-year modeling period.

The initial linear PCE concentration distribution shown on Figure 5 reflects about 300 µg/kg (on a dry mass basis) near the bottom of the profile at the water table, declining to the uppermost sublayer from 0 to 0.15 m. Under fully water-saturated equilibrium conditions, 300 µg/kg would correspond to a groundwater concentration of about 400 µg/L, which would be considered a moderate to high source strength. At equilibrium, about 78% of the PCE mass would be sorbed to the soil solids with about 22% in the dissolved phase.

Figure 5: Modeled Profile, Assumed Hydrologic Properties, Initial Conditions and Loading

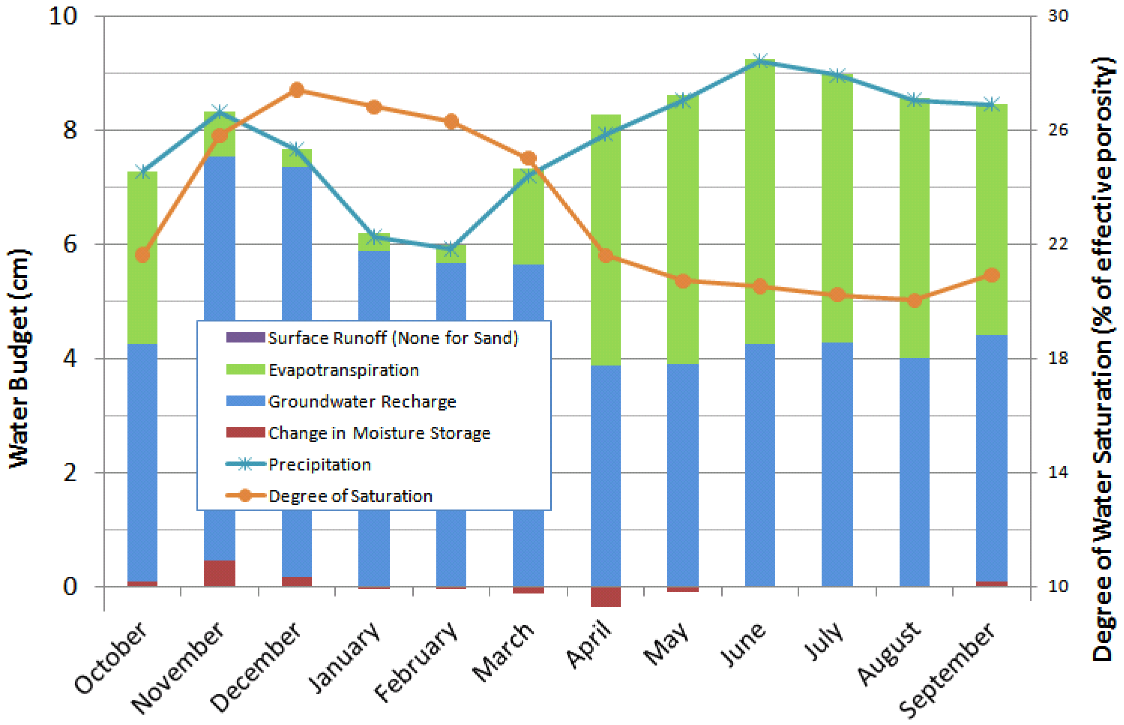


The properties related to soil conditions relevant to VOC transport are believed to be generally representative of a moderately dense, well-sorted, fine to medium sand soil with lower porosity and permeability compared to the Borden example, but with higher organic carbon content with the intention of providing an example of a soil column with moderate to high sorption. For comparison, the Borden vadose zone characterization[6] indicated soils with saturated hydraulic conductivity values ranging from about 3 x 10^{-5} to 2.4 x 10^{-4} meters per second (m/sec), with porosities ranging from about 0.41 to 0.56, expressed as a fraction of bulk soil volume, and a fraction of organic carbon f_{oc} averaging 0.0002 and ranging up to 0.022, netting partition coefficients (K_d) of about 0.03 and 2.8, respectively, which compare with a K_d of about 0.54 used in the modeling.

Model Water Budget and Soil Moisture Cycling

The water budget and soil moisture cycling as computed by the SESOIL model and summarized in Figure 6 shows that of the 94 cm of precipitation per year, about 34 cm (13 in.) or 36% is predicted to be lost to evapotranspiration which is predominant during the Spring and Summer growing season. About 60 cm (24 in.) or 64% is predicted to infiltrate across the vadose zone reaching the water table. Surface runoff – which may be equivalent to evapotranspiration for a fine-grained profile – is negligible for the relatively high-permeability sand.

Figure 6: Water Budget Components and Soil Moisture Cycling



The model-predicted change in soil moisture storage is small at -0.35 to +0.43 cm. The model-computed water saturation ranges from 20 to 27% of the effective soil pore space, and is about 23% on average. Given an effective porosity of 0.25, a 5-meter thick vadose zone would hold the equivalent of 29 cm water. At the model predicted annual groundwater recharge of about 60 cm, the soil moisture profile would be anticipated to turn over about twice a year instead of the about two years for the example field profile shown on Figure 2.

Model-Predicted Soil Vapor Concentration Profiles

Figure 7 shows PCE vapor concentration profiles as a function of depth (Figure 7.a) and time (Figure 7.b), as predicted by the SESOIL model. The PCE vapor concentration profiles on Figure 7.a reflect average of monthly concentrations for the given year while Figure 7.b shows the model-predicted vapor concentrations by month near ground surface (uppermost sublayer), near foundation depth (about 2.5 m or 8 ft), and near water table (5 m, lowermost sublayer).

The wavy patterns observed in Figure 7.b reflect seasonal cyclic patterns in concentration tied to changes in infiltration and soil moisture content similar to that observed in field monitoring programs. Since this simulation is for uniform, relatively permeable sand profile, the magnitude of those changes is logically smaller than observed for field sites with greater heterogeneity in soil texture and hence greater swings in moisture content (e.g., Figure 4). Overall, the profiles shown on Figure 7 indicate about one order of magnitude concentration gradient from water table to ground surface, with less than one-half an order of magnitude between water table and foundation depth.

Referring to Figure 7.b, the modeling suggests that after elimination of a groundwater source, it may take about seven years for vapor concentrations to drop below100 $\mu g/m^3$, a typical order of magnitude of soil gas screening threshold. It should be noted that this simulation was for a soil with low moisture saturation, moderate sorption, and relatively high permeability. Additional SESOIL simulations for lower permeability, silt-clay soil with higher moisture retention but similar sorption properties indicate that it could take substantially longer to reach this threshold.

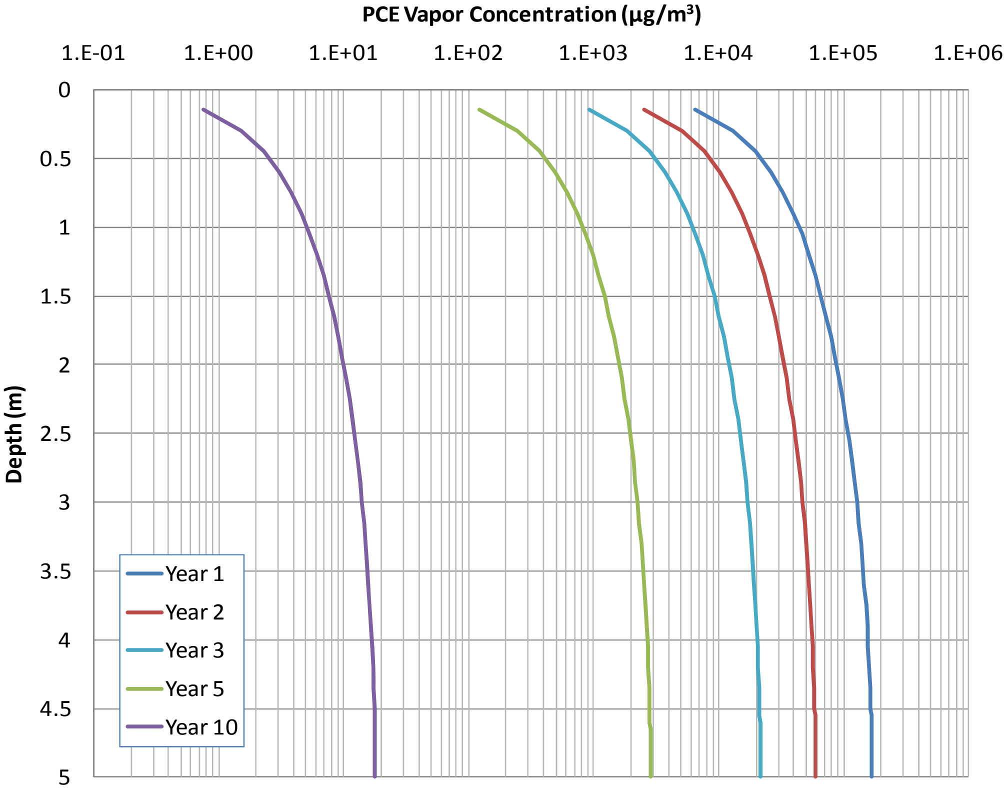
Model-Predicted Mass Flux

Figure 8 shows the PCE mass flux as a function of time over the ten-year simulation period near ground surface, near foundation depth, and near water table. Figure 8.a shows the daily mass flux, whereas Figure 8.b shows the cumulative mass transfer over the same period.

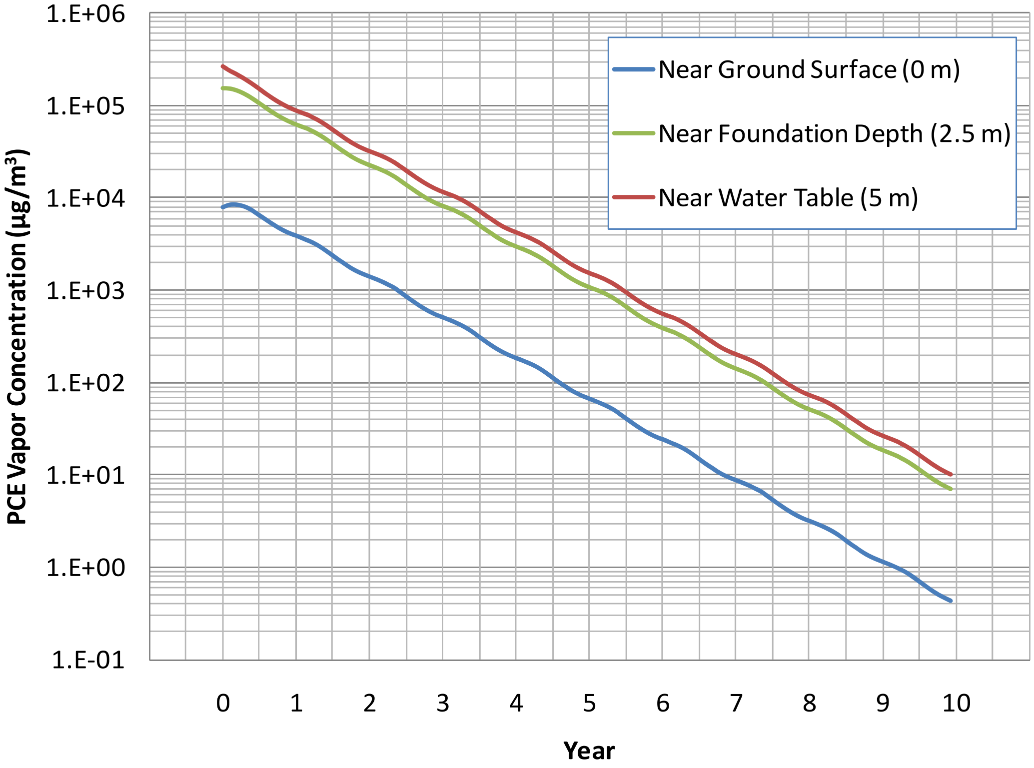
Figure 8.a shows that the upward diffusive flux is larger across the upper half of the vadose zone where PCE concentration gradients are steeper (see Figure 7.a). Near the water table, the downward mass flux to groundwater (advective transport via groundwater recharge) is about one order of magnitude larger than the upward diffusion flux. All of the mass fluxes decrease by one order of magnitude about every other year starting from hundreds to thousands of micrograms per square meter per day ($\mu g/m^2$-day).

For perspective, Table 1 provides a comparison of the model predicted flux against what would be inferred to achieve PCE indoor air concentrations observed at vapor intrusion sites from the USEPA's vapor intrusion database.[14] As summarized in Table 1, for PCE concentrations in indoor air to ranging between 0.6 $\mu g/m^3$ (25% percentile) and 52 $\mu g/m^3$ (95% percentile), the diffusion flux would be between about 9 and 750 $\mu g/m^2$-day under USEPA's default building assumptions for residences.[15] This range of flux back-calculated from indoor air statistics is within the range of model-derived flux rates estimated for foundation depth (Figure 8.a).

Figure 7: Model-Predicted PCE Concentrations as a Function of (a) Depth and (b) Time

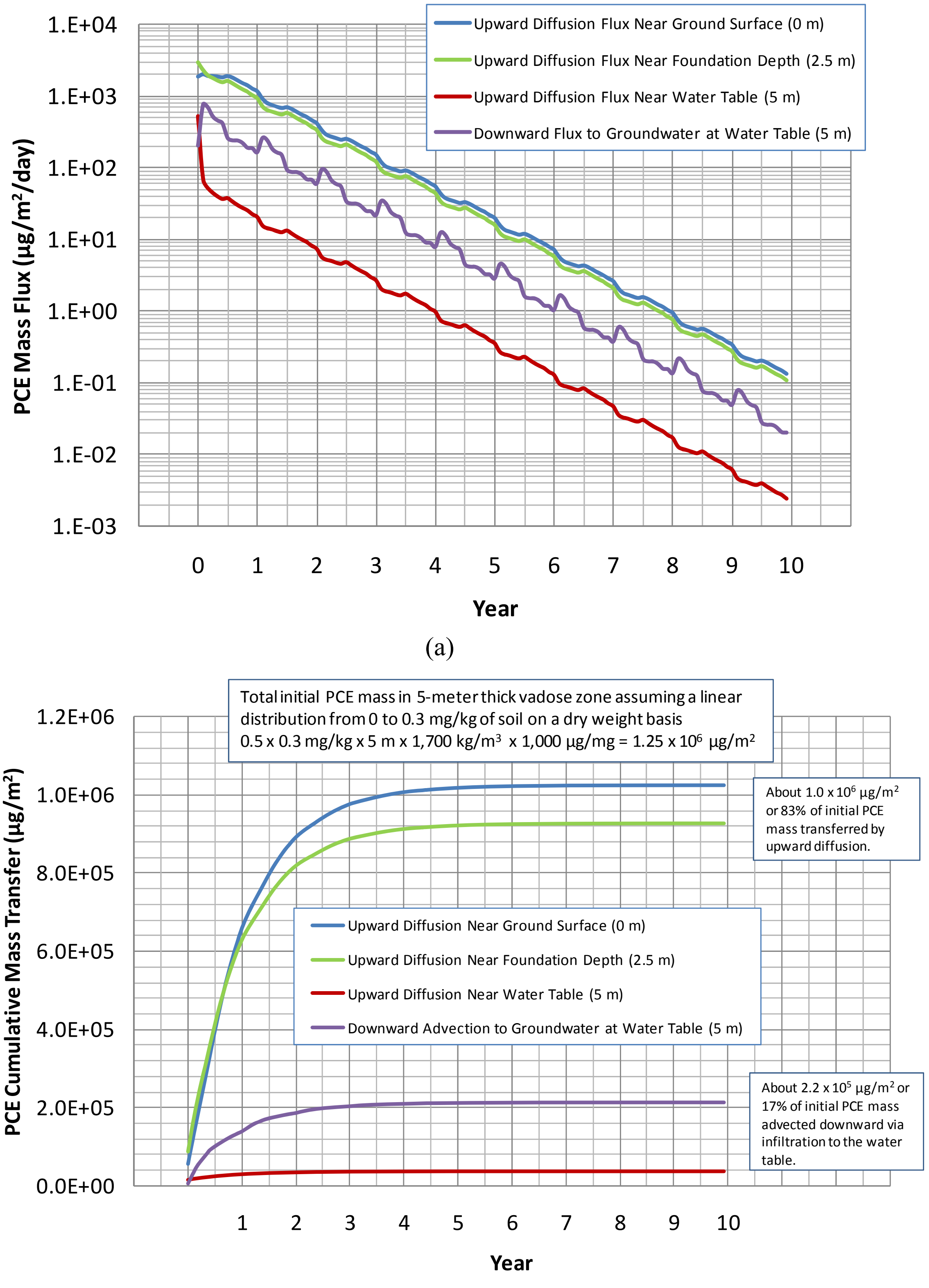


(a)



(b)

Figure 8: Model-Predicted Mass (a) Daily Flux and (b) Cumulative Mass Transfer



(a)

Total initial PCE mass in 5-meter thick vadose zone assuming a linear distribution from 0 to 0.3 mg/kg of soil on a dry weight basis
0.5 x 0.3 mg/kg x 5 m x 1,700 kg/m^3 x 1,000 µg/mg = 1.25 x 10^6 µg/m^2

About 1.0 x 10^6 µg/m^2 or 83% of initial PCE mass transferred by upward diffusion.

About 2.2 x 10^5 µg/m^2 or 17% of initial PCE mass advected downward via infiltration to the water table.

(b)

Table 1 - Indoor Air Statistics for PCE in Residences at Vapor Intrusion Sites and the Corresponding Probable Magnitude of Vertical Flux

PCE Indoor Air Statistics	Reporting Limit	25% Percentile	Median	75% Percentile	95% Percentile	Maximum
PCE Concentration in Residential Indoor Air from USEPA vapor intrusion database (μg/m^3)	0.2 to 2.7	0.6	1.5	5.6	52	1,896
PCE Upward Diffusion Flux* (μg/m^2-day)	2.9 to 39	9	20	80	750	27,000

* Flux calculated using USEPA's default building values, including building surface area *S* of 100 m^2 (1,080 ft^2), mixing height *H* of 2.4 m, and air exchange per hour (*AEH*) of 0.25 h^{-1}. The diffusion flux J can be calculated using a mass balance equation as:
J [μg/m^2-day] × S [m^2] = IA [μg/m^3]× S [m^2] × H [m]× ACH [h^{-1}] where *IA* is the indoor air concentration measured in the building.

The undulating patterns of seasonal increase and decrease in flux observed in Figure 8.a reflect seasonal variations in flux tied to infiltration and changing soil moisture conditions. At the beginning of the hydrologic cycle (fall and winter seasons) of the simulation year, the PCE mass flux to groundwater increase relative to the upward diffusion as a result of the increased groundwater recharge. During the second half of the cycle (spring and summer seasons), the magnitude of groundwater recharge decreases and diffusion increases.

Figure 8.b shows that under the modeled conditions, most of the PCE mass initially present in the vadose zone dissipates over the first five years through diffusive transport to the ground surface and to a lesser degree downward advection with groundwater recharge. Groundwater recharge accounts for about one fifth of the total PCE mass transfer, while the remaining four fifths are transferred via upward diffusion.

SUMMARY

The data and observations derived from field exploration and testing programs and the results of the limited modeling support and expand on what has been found by others referenced in the introduction. Specifically, we highlight the following important findings and conclusions.

- Progressive seasonal patterns in subsurface VOC vapor concentrations observed in long-term monitoring programs can be attributed to seasonal variability in infiltration and moisture transport across the vadose zone that influences gas phase diffusion and advection. VOC mass transfer among phases in the vadose zone can also contribute to the seasonal variability in vapor concentrations. The potential for seasonal and longer-term trends in vapor concentrations should be a consideration in designing and executing vapor intrusion investigation programs and in risk management approaches.

- The conventional model that indoor air quality and groundwater quality are directly linked in time and space is a flawed conceptual basis. VOC mass in soil moisture and sorbed onto the soil solids can contribute substantially to vapor intrusion potential and to the apparent spatial variability among groundwater and indoor air quality observed at vapor intrusion sites.

- VOC mass residing in the vadose zone may reflect past sourcing from historical groundwater quality conditions and, to a lesser degree, contemporaneous groundwater quality. In this light, VOC mass residing in the vadose zone may be an important factor limiting reduction in vapor intrusion potential following groundwater cleanup. The lag between groundwater quality improvements and reduced vapor intrusion potential may be considerable.

The findings support the value of vadose zone characterization as a component of a rigorous multiple lines of evidence approach to vapor intrusion investigations. In particular, collaborative data sets derived from characterization of lithology, soil texture and moisture, and other physical/chemical conditions along with multilevel soil gas monitoring, soil sampling and analysis for VOCs can greatly improve site conceptual models. Characterization at a scale of feet or less across the vadose zone may be required to identify conditions limiting to vapor transport.

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