ASSESSING EXPOSURE TO CHLORINATED SOLVENTS FROM THE SUBSURFACE TO INDOOR AIR PATHWAY

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ABSTRACT

JILL E. JOHNSTON: Assessing Exposure Of Chlorinated Solvents from the Subsurface to Indoor Air Pathway
(Under the direction of Jacqueline MacDonald Gibson)

The migration of chlorinated volatile organic compounds from groundwater to indoor air—known as vapor intrusion—is an important exposure pathway at sites with contaminated groundwater. However, monitoring indoor air quality in the hundreds or thousands of at-risk homes at each site is logistically and financially infeasible. Screening methods are needed to prioritize homes for monitoring and remediation. Current screening approaches do not adequately account for the substantial spatial and temporal variability in vapor intrusion risk, in part because the causes of this variability are not well understood. This work explores variability in vapor intrusion risk in a case-study community and then develops two different modeling approaches for screening at-risk homes.

We employed a community-based approach to collect indoor air samples and analyze vapor intrusion risk in 20 homes at a case-study site. Results demonstrate that indoor concentrations of tetrachloroethylene from vapor intrusion vary by an order of magnitude across space and time. We show that key factors affecting this variability include barometric pressure drop, humidity, wind speed, and season.

Using data collected from 370 homes in the National Database on Vapor Intrusion, we developed a multilevel regression model to predict vapor intrusion risks in

unmonitored homes. The resulting predictions decrease the rate of false negatives compared with the U.S. Environmental Protection Agency's (EPA) current screening approach, which assumes that indoor air concentration will not exceed 1/1,000 times the soil gas concentration just above the groundwater.

Finally, we demonstrate a second approach for improving the accuracy of screening by using Bayesian statistical techniques to integrate observational data into a mechanistic model describing the physical and chemical processes driving vapor intrusion. The resulting calibrated model also decreases the rate of false negatives in screening homes for vapor intrusion risks when compared with the current EPA approach.

The results suggest current policy may underestimate vapor intrusion exposures, and we demonstrate two approaches to improve exposure assessment. Future research should evaluate the potential for community-centered and real-time monitoring devices, the integration of localized and cumulative risk information into the framework, and assessment of the risks and benefits of a precautionary approach to mitigation.

Para los que luchan por un mundo mejor y la realización de la descontaminación y la salud de nuestro ambiente, nuestros cuerpos.

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To my daughter and my family, who navigated this process (and motherhood) with me, thank you for your unyielding support and encouragement. To my friends, I am grateful to each and every one of you.

PREFACE

This dissertation is organized in a nontraditional format, which includes three manuscripts. Chapter 1 provides an introduction to the dissertation and a description of the significance of the research. Chapters 2, 3, and 4 must stand alone as manuscripts to be submitted for publication and therefore have some redundancies with the earlier chapters. Chapter 5 presents a summary of the findings, policy implications, limitations of the studies, and directions for future research.

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LIST OF ABBREVIATIONS

AC	air conditioning
AIC	Akaike information criterion
AFB	Air Force base
ANOVA	analysis of variance
B	basement
BME	Bayesian maximum entropy
C	clay soil
CEJA	Committee for Environmental Justice Action
CL	clay loam soil
CS	crawl space foundation
CVOCs	chlorinated volatile organic compounds
DCE	dichloroethene
EPA	U.S. Environmental Protection Agency
F	fine-grained soil
GSD	geometric standard deviation
HAPSITE	Hazardous Air Pollutants on Site
HtA	Houston Black clay
JEM	Johnson-Ettinger model
LN	lognormal
LvA	Lewisville silty clay
MCMC	Markov Chain Monte Carlo
Med	median

OLS	ordinary least squares
PCE	tetrachloroethylene
RIOPA	relationship of indoor, outdoor, and personal air
RMSE	root mean squared error
S	summer
SC	sandy clay soil
SCL	sandy clay loam soil
Sd	standard deviation
SiC	silty clay soil
TCA	trichloroethane
TCE	tricholorethylene
TD-GC/MS	thermal desorption gas chromatograph/ mass spectrometer
USDA	U.S. Department of Agriculture
VC	very coarse-grained soil
VOCs	volatile organic compounds
W	winter

LIST OF SYMBOLS

A = building area, cm^2

A_b = area of enclosed space below grade, cm^2

α = alpha, vapor intrusion attenuation coefficient, unitless

C_{indoor} = contaminant concentration in indoor air (mass/volume)

C_{source} = contaminant source concentration (mass/volume)

D_{air} = chemical specific molecular diffusion coefficient in air, cm^2/s

D_{H_2O} = chemical specific molecular diffusion coefficient in water, cm^2/s

D^{eff} = effective diffusion coefficient, cm^2/s

D^{eff}_{crack} = effective diffusion coefficient through cracks, cm^2/s

$D^{eff}_{c,z}$ = effective diffusion coefficient across the capillary zone, cm^2/s

D^{eff}_{i} = effective diffusion coefficient across soil layer *i*, cm^2/s

D^{eff}_{total} = total overall effective diffusion coefficient, cm^2/s

ΔP = indoor-outdoor pressure difference, g/cm-s^2

E_b = air exchange rate (1/hr)

g = acceleration due to gravity, cm/s^2 (constant)

H_i = chemical specific Henry's law constant, unitless

k = soil permeability near foundation, cm^2/s

K_i = soil intrinsic permeability, cm^2

$K_{H,i}$ = chemical-specific Henry's constant, unitless

K_{rg} = relative air permeability, unitless (between 0 and 1)

K_s = soil saturated hydraulic conductivity, cm/s

L_{crack} = enclosed space foundation or slab thickness, cm

L_i = Thickness of soil layer *i*, cm

L_t = source-building separation, cm

M = van Genuchten shape parameter, unitless

MH = building mixing height, cm

μ = viscosity of air, g/cm-s

μ_w = dynamic viscosity of water, g/cm-s (constant)

η = fraction of foundation surface area with cracks, unitless

$Q_{building}$ = building ventilation rate, cm^3/s

Q_{soil} = volumetric flow rate of soil gas into the enclosed space, cm^3/s

R_{crack} = effective crack radius or width, cm

ρ_w = density of water, g/cm^3 (constant)

S_{te} = effective total fluid saturation, unitless

θ_m = volumetric moisture content, cm^3/cm^3

θ_r = residual soil water content, cm^3/cm^3

θ_T = total soil porosity, cm^3/cm^3

V_b = building volume, cm^3

X_{crack} = total length of cracks through which soil gas vapors are flowing (i.e. perimeter), cm

Z_{crack} = crack opening depth below grade, cm

CHAPTER 1

Introduction

1.1. Overview of this Research

Although a number of important pieces of environmental legislation have been enacted over the past 40 years, these regulations largely ignore the indoor sphere. No federal agency or law specifically regulates the quality of air in residential indoor environments, even though Americans spend 85-90% of their time indoors and the majority of exposure to air contaminants occurs there (Hodgson, Garbesi, Sextro, & Daisey, 1992; Klepeis et al., 2001; Spengler & Sexton, 1983). Compared to the consumption of drinking water, humans inhale 10,000 times more liters of air per day, an involuntary exposure that is very difficult to replace (Schuver, 2007). Over the past decade, vapor intrusion has been recognized as a possible significant health hazard to residents living near toxic sites and polluting facilities (Johnson & Ettinger, 1991; U.S. Environmental Protection Agency, 1992). Despite this evidence and growing interest in this exposure pathway, there has not been a systematic approach to exposure assessment or the development of appropriate, evidence-based policy to indoor air pollution due to vapor intrusion.

The vapor intrusion pathway is technically complex and incompletely understood. Detailed studies are few. Vapor intrusion involves both consideration of the pollutant source and its sink, the indoor air. It is an issue both of the commons (groundwater and/or soil) as well as private space (the interior of private buildings). At vapor intrusion sites, exposure is both inescapable and involuntary (Fitzgerald, 2009). Current regulatory guidance is limited in scope, and robust decision-making tools for managing vapor intrusion risks are lacking. The subsequent chapters of this dissertation evaluate the variability and uncertainty of the vapor intrusion pathway in order to inform the regulatory framework around the collection of measurements and the use of quantitative screening and modeling tools in the evaluation of exposure. The research focuses on an understudied region in the United States—the South—to examine the mechanisms and forces at work in a southern climate. This study compares data from measurements and modeling efforts and quantifies the uncertainty of measuring and predicting indoor air concentrations. The knowledge gained may be useful in creating and refining models to better predict exposure due to vapor intrusion and to support the development of quantitative decision-making tools useful in the assessment of contaminated sites. The research is structured around three objectives:

- **Objective 1:** Characterize spatial and temporal variability in the distribution of tetrachloroethylene (PCE) in indoor air in residences in a case study community that overlies groundwater contaminated with these chemicals. This objective has two components: (a) determine the concentrations of PCE in the air attributable to vapor intrusion in 20 homes, and (b) evaluate the factors that influence both temporal and spatial variability in the indoor concentrations of PCE.
- **Objective 2:** Evaluate the current U.S. Environmental Protection Agency (EPA) vapor intrusion screening-level approach with actual measurements, and develop

an alternative method based on a multivariate analysis of the vapor intrusion database.

- **Objective 3:** Demonstrate a novel approach to the integration of a mechanistic model with stochastic techniques in order to improve characterization of exposure due to vapor intrusion in a contaminated community.

1.2. Scope of Vapor Intrusion Problem and Potential Health Issues

When a subsurface release of volatile chemicals (those that easily transform to gas phase) occurs near buildings, contaminants can migrate upwards and result in vapor-phase contaminant intrusion into the indoor air. A particular class of volatile chemicals, chlorinated volatile organic compounds (CVOCs), includes commonly used solvents such as tetrachloroethylene (also called perchloroethylene, or PCE). CVOCs are among the most frequently detected groundwater contaminants at hazardous waste sites in the United States (Agency for Toxic Substances and Disease Registry, 2007; McCarty, 2010). They persist in the environment and are difficult to remediate (Simpkin & Norris, 2010; Travis & Doty, 1990). A commonly accepted practice in the remediation of CVOC plumes is “monitored natural attenuation”—that is, allowing natural physical processes to dilute and biological processes to degrade the contaminants to allowable levels, an approach that can take decades (U.S. EPA, 1999b). When contaminants remain in the subsurface, they may migrate indoors through the vapor intrusion pathway. The inhalation of vapors inside homes may be the most significant pathway by which communities are exposed to CVOCs from groundwater (Ferguson, Krylov, & McGrath, 1995; Fischer et al., 1996; Little, Daisey, & Nazaroff, 1992; Provoost et al., 2008).

Vapor intrusion exposures are real, direct, and chronic. The concentration of contaminants above recommended human health exposure levels in indoor air has been attributed to vapor intrusion from several sites (EerNisse, Steinmacher, Mehraban, Case, & Hanover, 2009; Folkes, Wertz, Kurtz, & Kuehster, 2009; McDonald & Wertz, 2007). Volatile organic compounds are reported at about half of known hazardous waste sites. Of these, approximately half may have conditions that favor intrusion of vapors into buildings, amounting to tens of thousands of sites nationwide (Schuver, 2007).

Levels of CVOCs in indoor air are typically five to 10 times those in ambient air (Steinemann, 2004; Wallace, 2001). Inhalation can lead to higher toxicities than exposures via oral routes (Pepelko & Withey, 1985). As a result, even low levels of exposure to indoor pollutants can present human health risks. Long-term exposure to CVOCs can cause both acute and chronic health effects, ranging from headaches and reproductive disorders to liver and kidney cancer (Buben & O'Flaherty, 1985; Chiu, Caldwell, Keshava, & Scott, 2006). While the epidemiological evidence of health impacts at vapor intrusion sites is limited, elevated disease rates—including elevated rates of liver, kidney and esophageal cancer— have been reported at sites with known vapor intrusion (Agency for Toxic Substances and Disease Registry, 2006; Colorado Department of Public Health and Environment, 2002). Elevated rates of adverse birth outcomes among newborns, including low birth weight, fetal growth restriction, and cardiac defects, have been associated with their mothers living in a community exposed to CVOCs via vapor intrusion (Forand, Lewis-Michl, & Gomez, 2011).

1.3. Vapor Intrusion Exposure Pathway: Monitoring Considerations

Determining when and where vapor intrusion is occurring—and subsequently remediating it—is challenging. Monitoring techniques must be able to measure minute concentrations of chemicals in air, in the realm of less than one part per billion (by volume), and chemicals of concern that volatilize from household products, including dry-cleaned clothes, must be identified and removed. Current practice requires the investigation assume a building-by-building analysis, and the evaluation techniques can be highly invasive, requiring entry by agency personnel and the placement of monitors inside private homes.

At the community scale, current knowledge of the vapor intrusion pathway derives from a few detailed case studies where indoor air concentrations were measured across space (Folkes et al., 2009; Kliest, 1989; McDonald & Wertz, 2007; Schreuder, 2006). The results have demonstrated significant spatial variability, and often the majority of the risk has been concentrated in a few homes. Further, recent research has demonstrated that concentrations attributed to vapor intrusion vary daily, weekly, and seasonally (Luo, Holton, Dahlen, & Johnson, 2011; McHugh, Nickles, & Brock, 2007; McHugh et al., 2012). Factors influencing temporal variability are a current area of investigation, with few published studies available.

In addition, indoor sampling is further complicated by the potential for confounding indoor sources (Dawson & McAlary, 2009; Gorder & Dettenmaier, 2011; Kurtz, Wolfe, Woodland, & Foster, 2010). Obtaining accurate readings of concentrations attributable to vapor intrusion requires the identification and removal of confounding sources. This process is inexact, time consuming, resource intensive, and intrusive.

1.4. Vapor Intrusion Exposure Pathway: Modeling Approaches

Determining if and when an exposure pathway exists is further limited by shortcomings in scientific understanding. The movement of vapors from the subsurface is dependent on multiple elements. The understanding of the transport mechanisms governing the migration of vapors from the subsurface to indoor air is still evolving (Folkes et al., 2009; McHugh et al., 2012; Yao & Suuberg, 2013). The potential for vapor intrusion to occur from a contaminant source is understood to be dependent on four interconnected processes as summarized in Figure 1.1: (a) the concentration of the contaminant in the groundwater; (b) the rate at which that contaminant can migrate through the soil toward the surface or building interface (soil properties); (c) the rate at which the contaminant is drawn into the building (foundation properties); and (d) the ability of the contaminant to accumulate indoors (building properties) (Johnson & Ettinger, 1991). Because of political, technical, and financial constraints on directly monitoring indoor air quality in private homes, it is typically more feasible to use a mathematical screening tool to identify at-risk areas. Vapor intrusion models attempt to simulate the transport of vapors from the source through the soil and into underlying buildings.

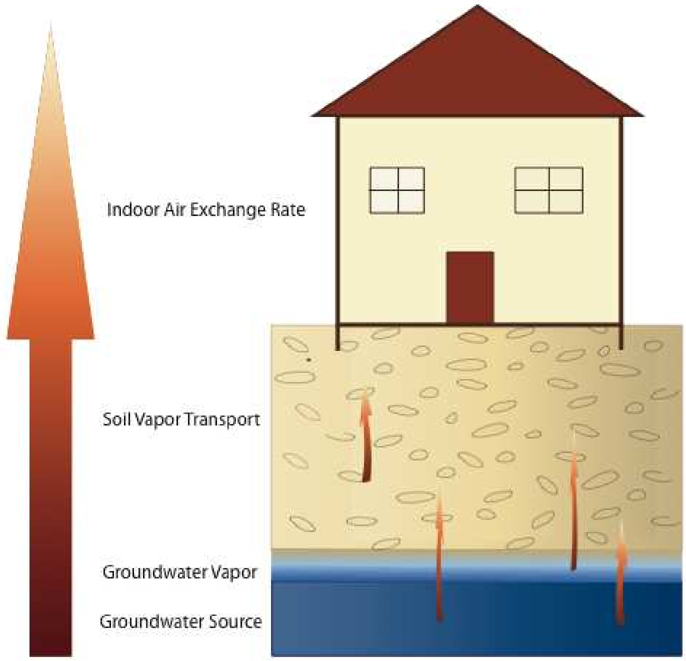

Figure 1.1. Summary of the physical processes that influence the vapor intrusion pathway.

There are two general categories of vapor intrusion models proposed: one dimensional (simplified) models and multidimensional numerical models. In general, one-dimensional models, like the Johnson-Ettinger model (JEM), are used in site evaluation to identify areas of potential highest risk and/or to determine whether further investigation of indoor air is warranted. The JEM (Figure 1.2) is widely used for regulatory guidance on vapor intrusion in the United States and estimates the vapor attenuation ratio, α, a unitless parameter that relates the indoor air concentration to the concentration in the vapor phase at equilibrium with the contaminated groundwater:

$$C_{indoor} = \alpha \times C_{source} \tag{1}$$

where α is the vapor attenuation ratio, C_{indoor} is the contaminant concentration in indoor air (mass/volume), and C_{source} is the contaminant vapor-source concentration (mass/volume).

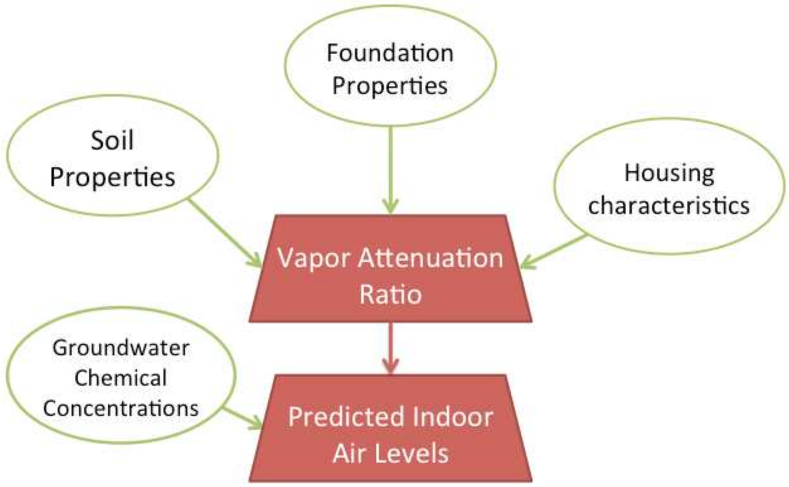

Figure 1.2. Conceptual framework of Johnson-Ettinger model.

The JEM couples one-dimensional steady-state diffusion of volatile compounds through porous media with diffusion and advection through the building foundation in the following equation to estimate α (Johnson & Ettinger, 1991):

$$\alpha = \frac{\left(\frac{D_{total}^{eff} A_b}{Q_{building} L_t}\right) \exp\left(\frac{Q_{soil} L_{crack}}{D_{crack}^{eff} \eta A_b}\right)}{\exp\left(\frac{Q_{soil} L_{crack}}{D_{crack}^{eff} \eta A_b}\right) + \left(\frac{D_{total}^{eff} A_b}{Q_{building} L_t}\right) + \left(\frac{D_{total}^{eff} A_b}{Q_{soil} L_t}\right)\left[\exp\left(\frac{Q_{soil} L_{crack}}{D_{crack}^{eff} \eta A_b}\right) - 1\right]} \qquad (2)$$

where D_{total}^{eff} is the total overall effective diffusion coefficient (cm^2/s), A_b is the area of enclosed space below grade (cm^2), $Q_{building}$ is the building ventilation rate (cm^3/s), L_t is the source-building separation (cm), Q_{soil} is the volumetric flow rate of soil gas into the enclosed space (cm^3/s), L_{crack} is the enclosed space foundation or slab thickness (cm), η is the fraction of foundation surface area with cracks (unitless), and D_{crack}^{eff} is the effective diffusion coefficient through the cracks (cm^2/s). However, many of these parameters are difficult to characterize.

The output of the JEM is intended to serve as an estimate of the influence of groundwater contamination on indoor air and to identify areas for further testing. Important parameters that influence vapor intrusion—and are included in the model—are soil characteristics (e.g., porosity, moisture content), building characteristics (air exchange rate, foundation type, and volume), and pressure differentials between the indoor and subsurface environments. Comparisons between modeled and measured α values indicate that with reasonable input parameters the JEM can predict within one order of magnitude the expected actual indoor air concentrations (Hers & Zapf-Gilje, 2003).

1.5. Vapor Intrusion Regulations and Policy Approaches

In November 2002, the EPA issued draft guidance titled "OSWER Draft Guidance for Evaluating the Vapor Intrusion to Indoor Air Pathway from Groundwater and Soils (Subsurface Vapor Intrusion Guidance)," which aimed to "provide a tool to help the 'user' conduct a screening evaluation as to whether or not the vapor intrusion exposure pathway is complete and, if so, whether it poses an unacceptable risk to human health" (U.S. EPA, 2002). The guidance proposes a risk-based approach for site management and the setting of remediation targets similar to the approach seen for the management of hazardous waste sites that requires site-by-site (and in this case even building-by-building) decisions at thousands of diverse sites (Daley, 2007; Sigman, 1998).

1.5.1. EPA Vapor Intrusion Assessment Tiers

The EPA guidance proposes a sequential order of assessment steps used to "screen in" a site for further investigation (U.S. EPA, 2002). As illustrated in Figure 1.3, the assessment process is divided into three tiers. The first tier considers whether volatile contaminants along with overlying structures are present at the site. The next tier employs a generic (not site-specific) screening process to estimate indoor air concentrations based upon the measured contaminant levels in the groundwater. Finally, the third tier uses site-specific modeling and data collection to assess human health risk. Failure to pass a specific step results in the conclusion that vapor intrusion either is not occurring or is not of adequate environmental or health concern, warranting no additional investigation. If any step is not sufficiently conservative or protective, sites can be falsely deemed "safe."

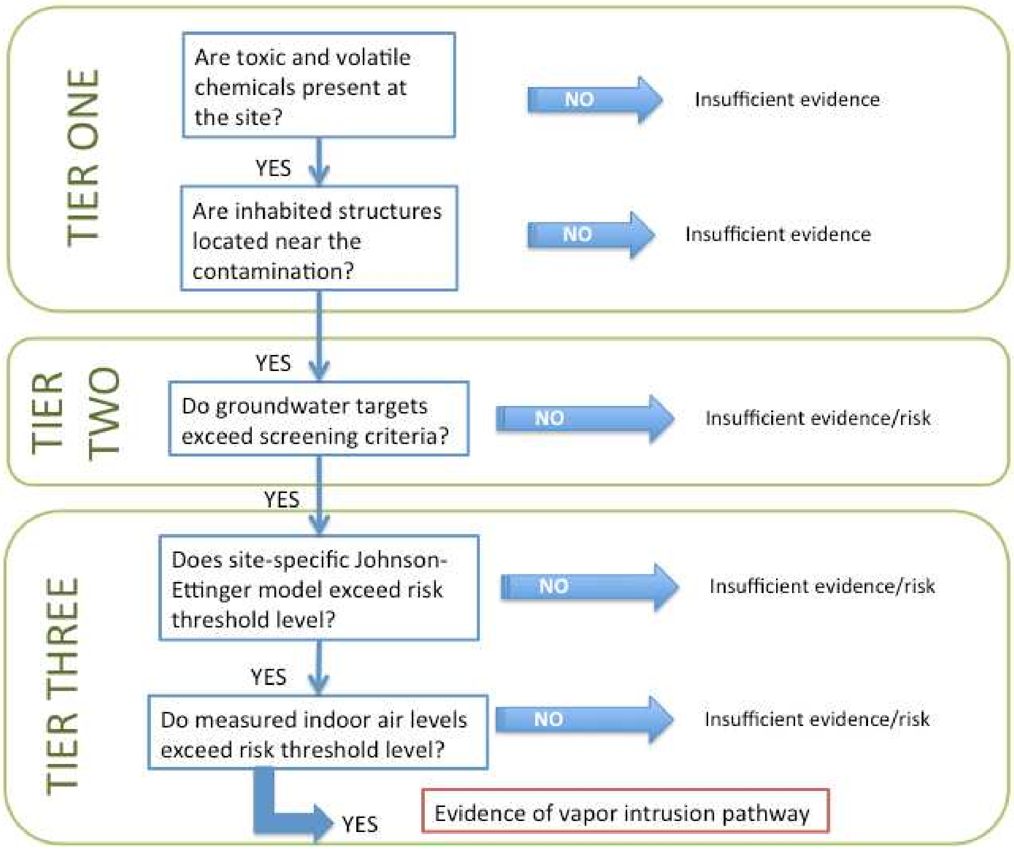

Figure 1.3. Schematic summary of EPA's approach to vapor intrusion sites, based on 2002 draft guidance.

Once a site passes the first tier, the second tier involves estimating the expected indoor air concentrations due to vapor intrusion. The 2002 guidance establishes groundwater targets by applying a generic attenuation factor, α, to screen vapor intrusion sites for study; EPA suggests that their assumptions represent the "worst-case conditions" (U.S. EPA, 2002). The attenuation factor is the ratio between the vapor-phase groundwater concentrations and indoor air concentrations of the chemicals of concern.

If sites screen into further analysis, the guidance document then proposes the use of the JEM to predict indoor air concentrations based on site-specific data such as groundwater depth and soil type (U.S. EPA, 2002). The guidance then outlines a method to use the indoor air concentration predictions to convert exposure into cancer risk estimates and recommend a course of action based on the risk calculation. There is no definitive action-level threshold; the EPA provides indoor air targets based on 10^{-4}, 10^{-5} and 10^{-6} risk levels. In addition, most state-level guidance provides an action-level threshold; across the various states, these values can vary by three orders of magnitude for the various CVOCs of concern (Eklund, Beckley, Yates, & McHugh, 2012).

Once sites have been selected for further screening, the next step is to take measurements of concentrations in indoor air. The collection of indoor air samples is suggested only if a home (or site) screens into the final stage based on previous modeling. The favored EPA method is a 24-hour active sample using a summa canister. This technique actively pumps air through the canister to capture a specified volume. This single sample is considered to be a representative concentration upon which to base an action decision.

1.5.2. Limitations of Current Regulatory Approach

The current approaches to estimating indoor air concentrations due to vapor intrusion are limited, may not be sufficiently protective, and fail to account for variability and uncertainty in the exposure pathway. EPA's proposed generic attenuation ratio of 1/1000 may underestimate exposure levels, as shown through the EPA's own analysis of vapor intrusion data (Figure 1.4) (Dawson, 2008a). The EPA's deterministic approach to modeling with the Johnson-Ettinger algorithm (i.e., not accounting for uncertainty and variability) can also underestimate actual risk and thus fail to provide adequate protection to affected households (Fitzgerald, 2009; Folkes et al., 2009; Schreuder, 2006; Tillman & Weaver, 2006). In a comparison with six other algorithms used in Europe, the JEM was found to produce the least conservative predictions (Provoost et al., 2009). The use of the JEM as a screening tool has been cautioned against because of the potential for false negatives and the frequency of underpredictions (Provoost et al., 2010). Complex three-dimensional models have been proposed and may be more accurate for an individual home, but these approaches are not scalable to a community level and require numerous detailed inputs (Bozkurt, Pennell, & Suuberg, 2009; Pennell, Bozkurt, & Suuberg, 2009; Yao, Shen, Pennell, & Suuberg, 2011; Yao & Suuberg, 2013). Few studies have compared modeling results to data, but in most cases the various models are unable to adequately explain the observations (Yao, Shen, Pennell, & Suuberg, 2013).

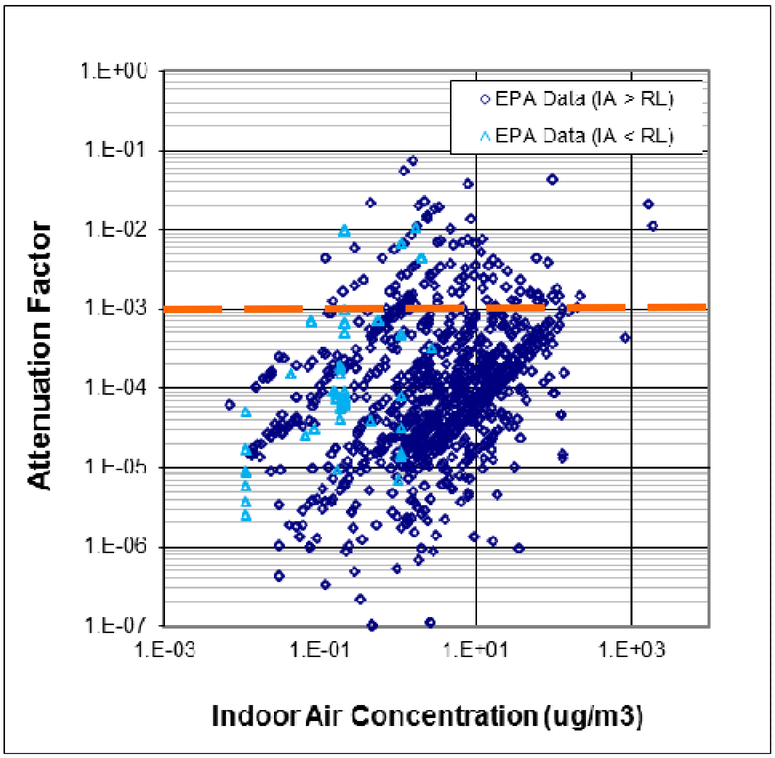

Figure 1.4. Measured groundwater vapor attenuation factors compared to the indoor air concentrations shown on a log scale for observations included in the EPA National Vapor Intrusion Database. The orange dashed line marks an attenuation factor of 1/1000. Dark blue diamonds represent observations where the indoor air (IA) concentrations exceeded the reporting limit (RL). Light blue triangles mark IA data less than the RL. Adapted from Dawson (2008a).

Finally, the method of only sampling from a single point in space and time is unlikely to reflect community-scale exposure. Due to the temporal variability of the process (as well as the potential for confounding indoor sources), a single sampling event is insufficient to provide definitive information about vapor intrusion risks (McHugh et al., 2012). Residents have expressed reservations about allowing monitoring using collection devices known as summa canisters, which are invasive and costly, prone to measurement error, and require batteries or electricity (Siegel, 2009; Wang & Austin, 2006). As a result of uncertainties, an accurate analysis requires robust data sets, which carry substantial costs.

1.5.3. Status of Current Federal Guidance

Since issuing the draft guidance, the EPA has yet not finalized the document. Separately, 29 states, stakeholder groups, and other federal agencies (including the Department of Defense, Department of Energy, and Department of Housing and Urban Development) have issued vapor intrusion guidance or other related technical documents, which vary widely in approach and scope (Fitzgerald, 2009; McAlary & Johnson, 2009; Simon, 2011). The Office of the Inspector General issued a report critical of the EPA's inadequate response and the incomplete scope of the 2002 guidance (U.S. EPA, 2009). The EPA has blamed the lack of progress toward finalizing guidance or developing regulations on both administrative and scientific barriers (U.S. EPA, 2009). The EPA was scheduled to release a final draft of the 2002 guidance in December 2012, but the deadline passed and a new draft is expected in the Spring of 2013. It is anticipated that the new guidance will increase emphasis on the collection and use of indoor air samples (McHugh et al., 2012). While the limitations of the guidance document are numerous, this research focuses on the tools proposed (or used) by environmental agencies to determine whether vapor intrusion is occurring and if a site requires monitoring or further remedial action.

1.6. Study Site

For much of this research, I used a case study site of a low-income neighborhood adjacent to the former Kelly Air Force Base (Figure 1.5), which operated in the southwest side of San Antonio, Texas, for nearly 85 years, serving as a logistic headquarters for the U.S. Air Force. Over that time period, the activities and practices at the base contaminated the shallow groundwater aquifer underneath it. Chlorinated solvents, including PCE, are the principal constituents of the contaminated plumes. These plumes extend five miles to the southeast of the base and occupy 12 square miles containing more than 30,000 homes in additional to farms, businesses, and schools. The shallow groundwater lies three to 40 feet below the homes, and PCE concentrations in the groundwater range from 5 μg/L (EPA drinking water maximum allowable limit) to almost 50,000 μg/L. Over 90% of residents living in the affected areas are Latino, and 75% live in poverty. San Antonio is situated in a warm, semi-desert climate with predominantly clay soil and an older single-story housing stock without basements.

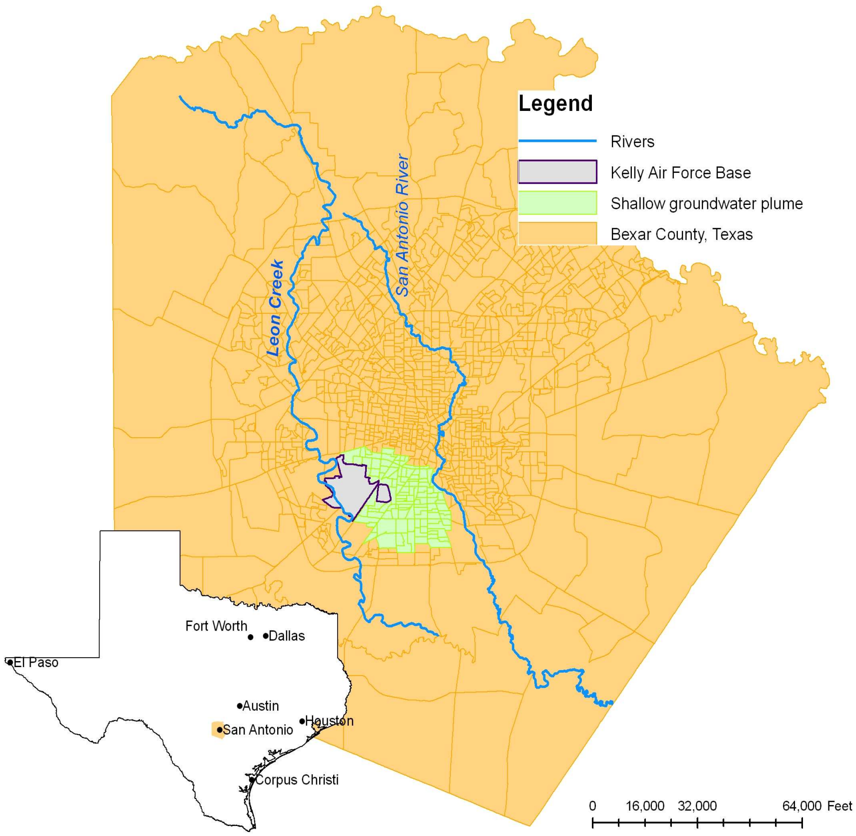

Figure 1.5. Map of Kelly Air Force Base and the adjacent contaminated groundwater plume.

In previous research, the JEM was used to estimate indoor air concentrations in approximately 31,100 homes in the community adjacent to Kelly Air Force Base, and the estimated concentrations were compared with measured values in approximately 20 homes tested by the EPA (Johnston & MacDonald Gibson, 2011). In this study, the JEM was employed using a probabilistic approach, in which the uncertainty and variability in model input variables were characterized, and Monte Carlo simulation was used to characterize the uncertainty in the resulting predicted indoor concentrations.

1.7. Motivation and Objectives

In summary, a key unresolved debate is what constitutes sufficient evidence of a complete vapor intrusion pathway and how to identify whether vapor intrusion exposure may be occurring in individual houses (U.S. EPA, 2009). Ultimately, the vapor intrusion pathway is house-specific and prone to temporal fluctuations. However, in most cases hundreds or even thousands of buildings are potentially impacted. While it is impractical to monitor every home, it is also problematic to use over-simplistic decision-making models to determine whether additional investigation is needed. Due to the known limitations of the proposed modeling-based tools, alternative approaches should be considered in order to improve exposure estimates and provide a better estimate of health risks and remediation needs. So far, neither an adequate tool to identify high-risk areas nor the ability to assess the exposure at a community-wide level exists.

The overarching aim of this dissertation is to investigate tools for predicting household indoor air contamination due to the migration of CVOCs from the subsurface and to assess decision-making tools used to make policy choices regarding vapor intrusion monitoring and remediation. This research represents the first community-wide study that examines vapor intrusion across space and time. The approach allows the quantification of relationships between weather conditions and household characteristics with vapor intrusion levels. The research improves upon current decision-making models to screen sites at risk of vapor intrusion and facilitates the allocation of resources to monitoring and/or remediation. Coupling modeling techniques with limited site-specific data can result in a process to effectively evaluate exposure at the community level, characterize the uncertainty and variability in the risks, and inform an alternative site-specific decision-making tool.

The remainder of this dissertation is organized into four chapters. Chapter 2 describes a method to quantify spatial and temporal variability in indoor concentrations of PCE in San Antonio, as well as integrate community participation. Chapter 3 examines the limitations of the current generic screening approach based on observations from the EPA vapor intrusion database and proposes a regression-based approach for screening potential vapor intrusion sites. Chapter 4 considers stochastic techniques to improve site-level exposure estimates based on the JEM when some site-specific data has been collected. Finally, Chapter 5 discusses key findings and implications from these analyses as well as future research needs.

CHAPTER 2

Spatiotemporal Variability of Tetrachloroethylene in Residential Indoor Air Due to Vapor Intrusion: A Longitudinal, Community-Based Study[1]

2.1. Introduction

Volatile organic compounds (VOCs) are often found at higher concentration indoors compared to the outdoor environment (Adgate et al., 2004; Dodson, Levy, Houseman, Spengler, & Bennett, 2009). VOCs are capable of migrating from contaminated groundwater through overlying soil and building foundations, resulting in vapor-phase contaminant intrusion into indoor air (Environmental Quality Management, 2004; Johnson & Ettinger, 1991). Tetrachloroethylene (PCE) is among the most frequently detected groundwater contaminants at hazardous waste sites in the United States (Agency for Toxic Substances and Disease Registry, 2007; McCarty, 2010). The inhalation of vapors inside homes is an understudied field, but prior research suggests it may be an important pathway by which communities at hazardous waste sites are exposed to chlorinated VOCs (CVOCs) in groundwater (Ferguson, Krylov, & McGrath, 1995; Fischer et al., 1996; Little, Daisey, & Nazaroff, 1992; Provoost et al., 2008). Long-term exposure to CVOCs has been linked to cancer, kidney and liver disease, and reproductive problems such as pregnancy loss, developmental abnormalities, and low-

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[1] *Johnston, J. E., & MacDonald Gibson, J. 2013, in press. Spatiotemporal variability of tetrachloroethylene in residential indoor air due to vapor intrusion: a longitudinal, community-based approach.* Journal of Exposure Sciences & Environmental Epidemiology

birth weights (Aschengrau et al., 2009; Agency for Toxic Substances and Disease Registry, 1997; Beliles, 2002; Doyle, Roman, Beral, & Brookes, 1997). Elevated rates of cancers, low birth weights, fetal growth restrictions, and cardiac defects have been reported at sites with CVOC vapor intrusion, although causality has not been established (Agency for Toxic Substances and Disease Registry, 2006; Colorado Department of Public Health and Environment, 2002; Forand, Lewis-Michl, & Gomez, 2011). Due to these potential health risks and the frequency of PCE detection in contaminated groundwater, the potential for PCE exposure via vapor intrusion is an important consideration when making decisions regarding groundwater remediation.

Spatial and temporal variability has been observed in subslab and indoor air concentrations of CVOCs above contaminated groundwater plumes (see, for example, Folkes, Wertz, Kurtz, & Kuehster, 2009; Luo, Holton, Dahlen, & Johnson, 2011; McDonald & Wertz, 2007; McHugh, Nickles, & Brock, 2007; Schreuder, 2006). Variability across space and time has also been observed in indoor radon concentrations, which also result from vapor intrusion (albeit from natural geologic sources rather than anthropogenic contamination) (Davies & Forward, 1970; Groves-Kirkby, Denman, Phillips, Crockett, & Sinclair, 2010; Steck, Capistrant, Dumm, & Patton, 2004). Hence, an indoor air sample from a single point in space and time is unlikely to reflect community-scale exposure to vapor intrusion risks. Furthermore, previous work suggests that groundwater concentrations are not adequate surrogates for measuring vapor intrusion exposure potential because variability in soil and household characteristics can lead to houses above relatively low groundwater PCE concentrations having higher PCE levels in indoor air than homes overlying higher concentrations and vice versa

(Fitzpatrick & Fitzgerald, 2002; Folkes et al., 2009). In site assessments, often only a single 24-hour indoor air sample is taken from a small number of homes in an affected community, although U.S. Environmental Protection Agency (EPA) and state guidance often recommend that multiple samples be collected from a single home following a multi-tiered approach to vapor intrusion investigations (Eklund, Beckley, Yates, & McHugh, 2012; U.S. EPA, 2002). For example, in the EPA's National Vapor Intrusion Database, the sampling frequency is as follows: a single-point-in-time sample in 84% of buildings, two samples collected in 10% of buildings, three to five samples in 5% of buildings, and more than five samples in 1% of cases. Collecting one or two samples, as is the current common practice, will not account for the potential spatial and temporal variability and may under- or overestimate the true exposure risk. An inaccurate characterization of exposure may result in inaccurate human health risk assessments.

Previous work has helped describe the mechanisms governing vapor intrusion and potential causes of variability. Pressure-driven flow is an important mechanism for gas entry into homes (Fitzpatrick & Fitzgerald, 2002; Nazaroff et al., 1985). Building underpressurization, changes in barometric pressure, wind, and diurnal fluctuations in temperature all can influence indoor-outdoor pressure differentials and hence vapor flow into homes (Adomait & Fugler, 1997; Garbesi & Sextro, 1989; McHugh et al., 2012). When these processes lead to negative building pressure (i.e., outdoor pressure greater than indoor pressure), the rate of vapor intrusion increases. However, the relationships among these factors are complex and the net effects on vapor intrusion difficult to predict. For example, in some cases, higher wind speeds have been associated with lower indoor radon concentrations, while in others no relationship between wind speed and indoor radon concentrations has been observed (Luo, 2009; Nazaroff & Doyle, 1985; Nazaroff et al., 1985; Turk, Prill, Grimsrud, Moed, & Sextro, 1990).

Further understanding of the spatiotemporal drivers of vapor intrusion is needed in order to inform decisions about the extent of indoor air monitoring necessary to adequately estimate exposure risks in communities overlying contaminated groundwater. Yet, indoor air monitoring is intrusive, and residents can be resistant to allowing researchers or government personnel into their homes (Siegel, 2009). Due in part to this challenge, other studies of temporal variability have focused on a single home rather than multiple homes, and studies of spatial variability have been able to collect only one or two 24-hour samples in each home.

This study addresses the need for community-wide assessment of spatiotemporal variability in vapor intrusion risks. The study, the first of its kind in the southern United States, integrated longitudinal and cross-sectional data collection at a contaminated site adjacent to the former Kelly Air Force Base in southwest San Antonio, Texas. We examined the effects of household characteristics and meteorological conditions on observed fluctuations in indoor air PCE concentrations to determine whether changes in (a) meteorological conditions, (b) soil type, (c) groundwater concentration, and (d) household characteristics significantly explain spatiotemporal variability in indoor PCE concentrations attributable to vapor intrusion. A better understanding of the drivers of temporal and spatial variability in vapor intrusion can inform decisions regarding monitoring and exposure assessment in affected communities.

The case study site is a low-income neighborhood overlying extensive plumes of CVOCs in groundwater emanating from the former Kelly Air Force Base. These plumes extended five miles to the southeast of the base and underlie approximately 30,000 homes. The shallow groundwater lies 1 to 12 m below the homes. PCE concentrations in the groundwater range from 1 μg/L to 200 μg/L in the residential areas. Off-base groundwater remediation began in 2004 and is ongoing.

The EPA evaluated a cohort of 24 houses for vapor intrusion in May 2008 and February 2009. During the 2008 sampling, the EPA collected one or two samples beneath each home's foundation, outdoor air samples in selected locations, and a single indoor sample in a subset of homes. The sampling protocol followed EPA method TO-15, in which 6-liter collection devices known as summa canisters (in this case with a PCE detection limit of 0.14 μg/m^3) are deployed to capture an air sample later analyzed in a laboratory (U.S. EPA, 1999a). For homes in which indoor air was tested, the EPA verified that all indoor sources had been removed by scanning each home with a real-time trace atmospheric gas analyzer. Figure 2.1 shows the results for PCE for the sampling events. The indoor air concentrations ranged from nondetectable to 1.83 μg/m^3. Ambient air sampled for PCE averaged 0.055 μg/m^3. The elevated subslab concentrations of PCE (4 to 600 μg/m^3), along with the very low outdoor PCE concentrations, provide one line of evidence suggesting that PCE vapors are migrating from the groundwater into homes.

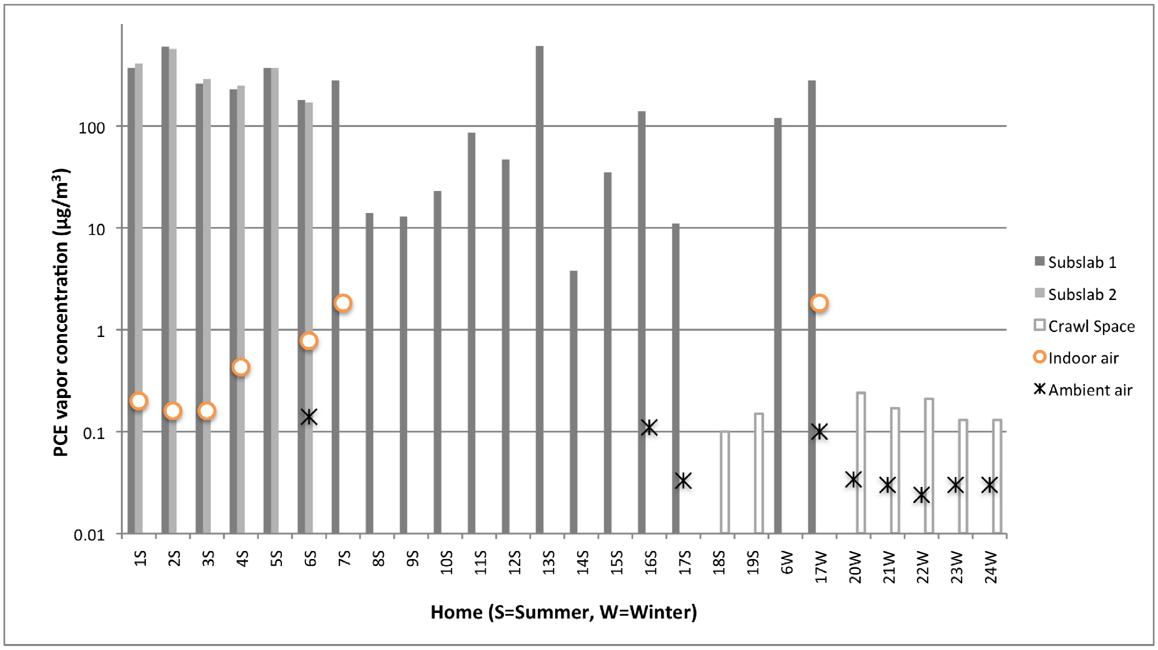

Figure 2.1. Summary of EPA's previous subslab, crawl space, and indoor air measurements for PCE.

Since this previous sampling was carried out only on a single day, the study design and results were not sufficient to evaluate the temporal variability in PCE concentration across the community. We previously modeled the scope of indoor air contamination in the community by employing a stochastic house-by-house approach based on the Johnson-Ettinger algorithm to account for variability and uncertainty in the parameters that influence vapor intrusion potential (Johnston & MacDonald Gibson, 2011). This modeling study estimated that PCE concentrations may exceed screening levels (0.41 $\mu g/m^3$ at the time of the analysis) in up to 72% of the homes, demonstrating potential vapor intrusion risk and highlighting specific neighborhoods that may be at higher risk. The present study was conducted to obtain field data to further explore the spatiotemporal variability suggested by our previous stochastic model.

2.2. Materials and Methods

We sampled indoor air for PCE in 20 homes over a 12-day period during summer (July-August) 2011 (Figure 2.2). We resampled nine of the homes over another 12-day period in winter (February-March) of 2012. For the winter period, we divided the homes into those with evidence of vapor intrusion (at least one detection above 0.25 μg/m^3) and those without. We randomly selected six homes from those that showed evidence of vapor intrusion and an additional three from the homes with no detectable PCE.

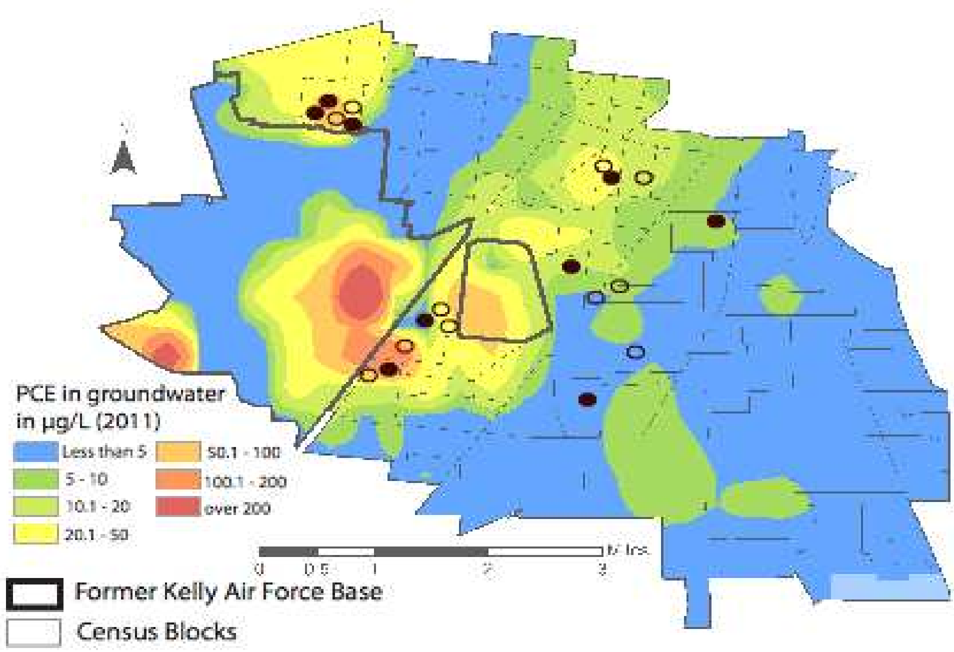

Figure 2.2. Map showing the approximate location of the 20 homes sampled (ovals, black ovals for homes sampled in winter) and the PCE concentrations in the underlying groundwater plume.

2.2.1. Indoor Air Sources Identification

PCE is a commonly used solvent that is contained in many common consumer products. A survey of indoor homes unaffected by vapor intrusion found a median PCE level of 0.9 μg/m^3 (Dawson & McAlary, 2009), and previous work has identified common consumer products containing PCE (Dettenmaier & Gorder, 2010). In order to identify and remove potential confounding indoor PCE sources, our study team sampled each home in real time with the Hazardous Air Pollutants on Site (HAPSITE) field portable gas chromatograph/mass spectrometer (Inficon, Syracuse, NY) prior to the deployment of the passive samplers. Previous work has used the HAPSITE as an effective tool to identify and remove indoor CVOC sources (Gorder & Dettenmaier, 2011). Prior to deploying the HAPSITE, we asked residents about the presence of common household products that could contain PCE and explained the importance of removing these sources for the duration of the study. We then asked to examine storage spaces for automotive, cleaning, and home repair supplies. We removed liquid/spray spot cleaners (15 homes), automotive lubricants (eight homes), and strong adhesives or shoe glue (four homes). No participant was known to use dry cleaning services or work in the dry cleaning industry. We also removed certain types of air fresheners because we found they interfered with the HAPSITE analysis. Next, within each home, we conducted an area-by-area investigation with the HAPSITE device to identify any additional potential vapor sources. After cleaning the concentrator, we collected a five-minute air sample that the HAPSITE automatically analyzed for PCE (detection limit 0.18 μg/m^3). Any detected household PCE sources were removed for the duration of the study period, and the air was resampled three hours after removal to assure that no confounding sources remained. Sampling began 24 hours later. In a randomly selected subset of 10 homes, the indoor air was reanalyzed with the HAPSITE on the fifth day of the study to evaluate whether confounding sources had been reintroduced into the home. We found no evidence of additional PCE sources during the mid-study resampling. In 18 of the homes, residents had a detached storage shed or garage, while two homes had no garage. These detached structures were neither evaluated nor included in the analysis.

2.2.2. Indoor Air Sampling

During the summer sampling event, we collected a total of eight duplicate samples (16 total samples) over a period of 12 days in each of the 20 study homes. For the second sampling period, four sample pairs per home were collected sequentially over a 12-day period in February and March. A total of 392 samples were collected (186 paired measurements). Passive monitoring devices were shipped to the field site, and duplicate field blanks were included in each sampling season. In each case, samplers were left in place for three days in order to ensure sufficient detection sensitivity. Duplicate samples were taken to help assure the quality of the collected data and avoid losing information if a device was mishandled. Figure 2.3 shows an example sampling schedule.

We deployed the indoor air monitoring devices on a simple, freestanding apparatus (constructed for this study) that enabled the sampling tubes to be hung in the breathing zone, 1.5 m (4.5 ft) above the floor. We located the samplers on the ground floor in an unused room, if available, or otherwise in a location where the monitors were less likely to be disturbed.

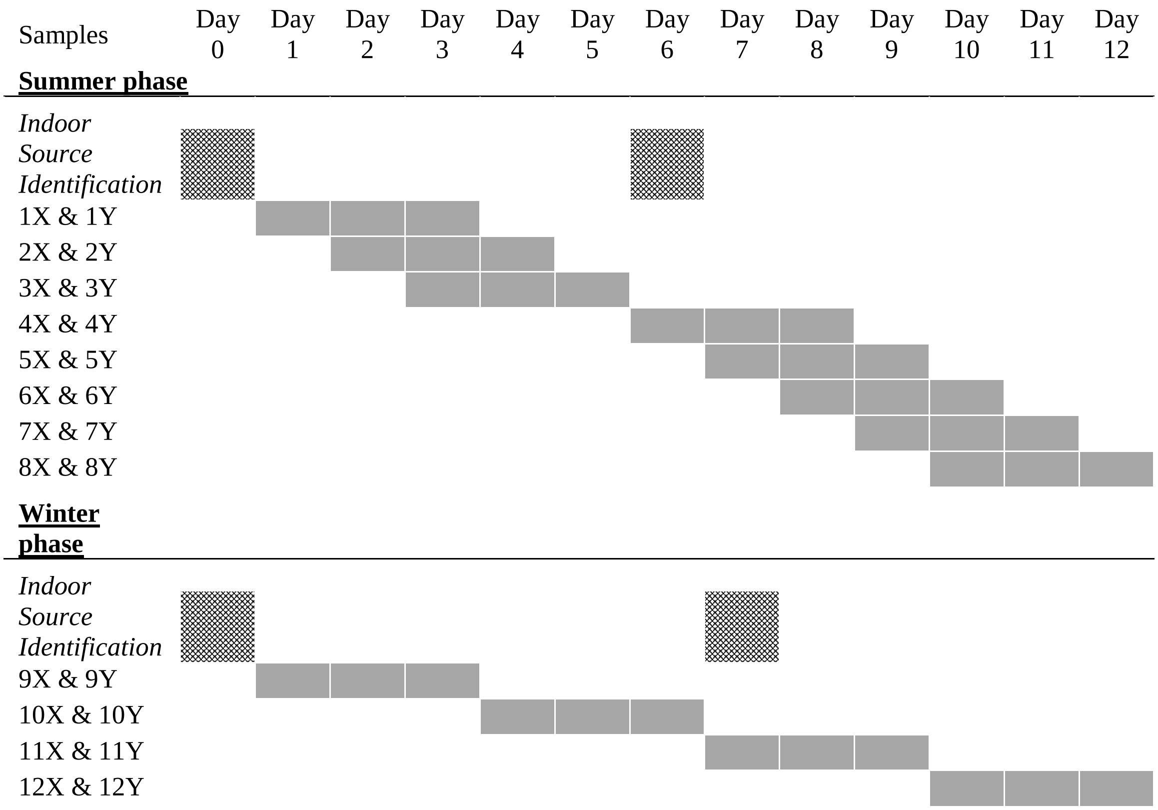

Figure 2.3. Example data collection schedule per house.

The sampling protocol followed ISO 16017-2:2003 (*Indoor, Ambient and Workplace Air-Sampling and Analysis of Volatile Organic Compounds by Sorbent Tube/Thermal Desorption/Capillary Gas Chromatography–Part 2: Diffusive Sampling*). Consistent with this protocol, the sampling devices were small (6.35 mm diameter × 89 mm height), stainless-steel tubes packed with an engineered adsorbent, Chromosorb-106, with a demonstrated affinity for chlorinated solvents. Beacon Environmental Services, Inc., (Bel Air, MD) thermally conditioned the samplers and shipped them to the study site. Previous studies have shown that these devices provide results comparable to those of summa canisters (Odencrantz, Thornley, & O'Neill, 2009).

Consistent with ISO 16017-2:2003 and also with U.S. EPA method TO-17 guidelines for sorbent samplers, Beacon Environmental analyzed the resulting samples with a Markes International thermal desorption system coupled with an Agilent 7890 gas chromatograph/ 5975 mass spectrometer (TD-GC/MS) (Woolfenden & McClenny, 1999). The concentration was calculated from the measured mass, exposure duration, and sorbent tube uptake rate for PCE (0.46 ml/min). The Beacon Environmental laboratory's reporting limit is 0.25 ng per sampling tube, yielding a detection limit of 0.13 $\mu g/m^3$. All field sample measurements were below the analytical system's upper calibration limit of 5.0 ng; therefore, no sample dilutions were required. The continuing calibration verification values for the system check compounds were all within ±20% of the true values. Laboratory method blanks were run with each sample batch to identify contamination present in the laboratory. In addition, laboratory control samples were included with each of the analytical batch samples and included the PCE compound. The average recovery rate of PCE for these samples was 95%.

In total, 392 total samplers were collected, with two lost due to leaks and sample handling errors. Five additional samplers were not used because the second internal standard was outside of the control limit, resulting in 385 observations. In these cases, we used only a single measurement to assign concentration. For all other cases, we averaged the two duplicate samples to estimate the measured concentration for a total of 186 distinct observations. On average, the relative percentage difference was 8.1% among duplicate samples that exceeded the detection limit.

2.2.3 Model Covariates

Meteorological data were acquired from the weather station at the former Kelly Air Force Base, which is within a 1.0 to 4.5 km radius of the study homes. Hourly data for temperature, wind speed, and humidity were averaged for the appropriate time period (based on the start and stop time for each sampler). Barometric pressure generally follows a diurnal cycle. The daily pressure drop was calculated as the difference between the crest and subsequent trough of the curve for each cycle (determined from hourly pressure measurements), with the first cycle commencing at the time of sampler deployment. These daily pressure drops were then averaged over the three-day exposure time for each sample tube. Information on chemical groundwater concentrations was acquired for April 2011 through the Kelly Air Force Base Semi-Annual Compliance Plans (1998-2011) from the Air Force Real Property Agency. Concentrations were interpolated from 900 monitoring wells using a Bayesian maximum entropy approach (see Christakos, Bogaert, & Serre, 2001; Johnston & MacDonald Gibson, 2011). All homes were located within 480 m of a monitoring well, with the majority of homes within 100 m of a well. While groundwater concentrations exhibit temporal changes, for this analysis the value was assumed to be constant for the study period. Groundwater depth was not included because the temporal resolution of the data was insufficient to allow such an analysis. The soil type beneath each home was determined from the Bexar County Soil Survey. For all homes, the identified soil type was either Houston Black clay or Lewisville silty clay (Taylor, Hailey, & Richmond, 1966).

Information was collected daily from participants about use of air conditioners, fans, and windows. These data were consolidated into a binary variable based on whether the home used air conditioning. In all homes that used air conditioning, the windows were kept closed. Since closed windows and cooling systems were strongly collinear, we only included the air conditioning variable in the model. Only one of the 20 homes used a dryer inside the home (others used dryers located in detached structures), so this variable was not considered in the model. Information on the age and square footage of each home was acquired from the Bexar County Appraisal District.

2.2.4. Community-Based Design

In partnership with a local community organization, the Committee for Environmental Justice Action (CEJA), we designed the research question, chose appropriate methods, recruited participants, and collected data. In this study, community cooperation was especially important because the sampling protocol necessitated access to participating homes on a daily basis over the sampling period and that participants adhere to the removal of products that could confound the results. To recruit participants, CEJA representatives circulated flyers describing the study. If a community member responded, CEJA arranged a meeting with a member of our study team, who then offered additional information about the data collection process. One participant in each household helped collect information on heating, cooling, and mechanical ventilation type for each home and completed an activity diary for each day of the study. The activity diary asked about use of products that might affect indoor air PCE concentrations and/or transport of vapors from the subsurface into the home (e.g., use of cleaning products, mechanical cooling devices, windows, and clothes dryers).

2.2.5. Statistical Analysis

We employed a longitudinal multivariate regression modeling approach to examine the temporal associations between the observed indoor PCE concentrations (dependent variable) in each home and barometric pressure drop, wind speed, and other meteorological characteristics. The form of the regression model was chosen to account for the detection limit (0.13 µg/m^3) of the sampling device as well as the longitudinal nature of the data collection. Typical techniques, such as exclusion of data, the assignment of half the detection limit to nondetects, or the substitution of a value randomly selected from an appropriate distribution, have been shown to bias parameter estimates and, in the case of the latter approach, bias the variance (Helsel, 1990; Lubin et al., 2004). To avoid such biases, we used the Tobit model, an extension of the probit analysis developed by Tobin (1958), which has been proven to provide an unbiased maximum likelihood approach for analyzing measurement data with detection limits (Slymen, de Peyster, & Donohoe, 1994; Tobin, 1958). Since the distribution of observed concentrations was right-skewed, we used a log-transformed dependent variable.

In this analysis, we employed clustered robust estimates of standard errors. In order to account for repeat and overlapping observations in each home and for the relatively small sample size, to estimate the standard error on each β coefficient we employed a method that is robust to serial autocorrelation and with good performance across a variety sample sizes (see Arellano, 1987; Hansen, 2007; Kezdi, 2003). Stata IC (Version 12) was used for statistical analyses, with an *a priori* significance level of 0.05.

To analyze the influence of changes in meteorological conditions on the within-home variation over time, a distinct intercept was modeled for each home in the regression, so that time-invariant characteristics would not bias the model. To investigate the variation between homes, we evaluated a pooled population average that examined both time-varying meteorological variables and time-invariant household characteristics.

2.3. Results

Table 2.1 shows the characteristics and minimum and maximum PCE concentrations measured in the 20 homes (all of which completed the study in its entirety). Figure 2.4 shows the detailed results for each house. PCE was detected in 12 of the 20 homes. The average PCE concentration across all samples above the detection limit was 0.28 $\mu g/m^3$ (Table 2.2); however, concentrations fluctuated as much as one order of magnitude (Figure 2.5).

In general, the measured concentrations were low, although about half exceeded the EPA Region 6 risk-based screening level for resident air of 0.33 $\mu g/m^3$ that was in place at the time of sampling. In April 2012, the EPA revised its PCE screening level to 9.4 $\mu g/m^3$, higher than all of the concentrations observed in this study. Nonetheless, the previous EPA analyses showing elevated subslab PCE concentrations and extremely low ambient concentrations (Figure 2.1) suggests that vapor intrusion may be an important source of the PCE observed in these homes, particularly because we removed indoor sources prior to sampling. (As an additional check on potential indoor sources, we also examined the correlations between measured PCE concentrations and self-reported days when cleaning products were used, but none of these correlations was significant.) Neither field blanks nor laboratory method blank samples had any measurable concentrations of PCE. Despite the low PCE concentrations observed in this study, the results nonetheless provide valuable new information on factors both within and between homes that influence variability in indoor PCE concentrations at sites affected by vapor intrusion.

Table 2.1. Characteristics of the 20 homes included in this study.

Year built[+]	Foundation	Cooling	Soil type[*]	Estimated groundwater PCE (µg/l)	Indoor PCE minimum (µg/m3)	Indoor PCE maximum (µg/m3)	Sampled in winter
1925	Crawl	Central AC	HtA	8	<0.13	0.14	Yes
1928	Crawl	Fans	HtA	12	<0.13	<0.13	No
1928	Crawl	Fans	HtA	21	<0.13	0.32	No
1934	Crawl	Fans	LvA	22	<0.13	<0.13	No
1940	Crawl	Fans	HtA	6	<0.13	<0.13	Yes
1945	Crawl	Window units	HtA	21	<0.13	0.16	Yes
1946	Crawl	Window units	HtA	15	<0.13	0.33	No
1949	Crawl	Fans	LvA	50	<0.13	<0.13	No
1950	Crawl	Central AC	LvA	21	<0.13	<0.13	No
1950	Crawl	Fans	HtA	20	<0.13	0.15	No
1951	Slab-on-grade	Central AC	LvA	4	<0.13	0.46	No
1953	Crawl	Window units	HtA	5	0.15	1.2	Yes
1955	Crawl	Window units	LvA	50	<0.13	0.46	Yes
1963	Slab-on-grade	Window units	LvA	8	<0.13	0.16	No
1965	Slab-on-grade	Central AC	LvA	98	<0.13	0.29	Yes
1965	Slab-on-grade	Central AC	LvA	98	0.15	0.67	Yes
1965	Slab-on-grade	Fans	LvA	98	0.16	.42	Yes
1972	Crawl	Window units	HtA	11	<0.13	1.50	Yes
1976	Slab-on-grade	Window units	LvA	98	0.14	0.75	No
1984	Crawl	Central AC	LvA	4	<0.13	<0.13	No

[+] All homes are single story. * HtA: Houston Black clay; LvA: Lewisville silty clay

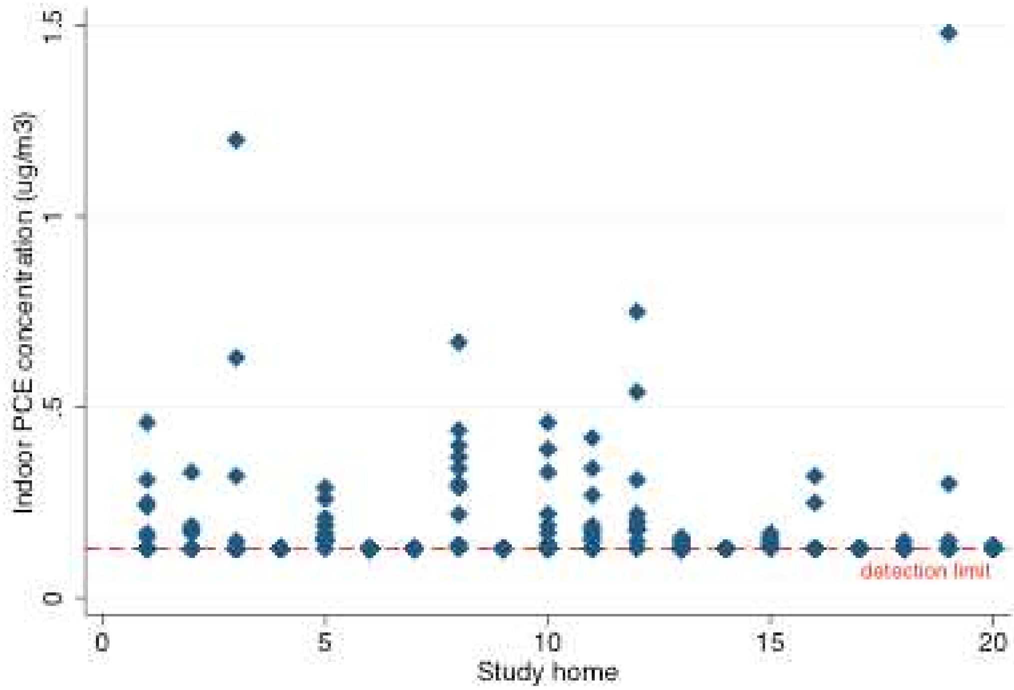

Figure 2.4. Indoor air concentration of PCE by study home.

Table 2.2. Summary statistics for the key continuous variables included in the regression model.

Time-variant variables	Above detection samples (n_s=90)	Below detection samples (n_s=106)
PCE concentration		
Indoor air (μg/m^3)	0.28 (0.22)+	≤0.13 --
Weather characteristics		
Barometric pressure drop (mm Hg)	6.05 (2.53)	5.91 (2.63)
Average wind speed (m/s)	4.03 (0.89)	4.32 (0.86)
Relative humidity (%)	62.64 (14.53)	58.2 (11.98)
Time Invariant Variables	Houses with any sample above the detection limit (n_h=12)	Houses with no samples above the detection limit (n_h=8)
PCE in the groundwater		
Groundwater concentration (μg/L)	43.6 (40.2)	32.8 (29.1)
Household characteristics		
House area (m^2)	124.6 (52.8)	111.2 (46.9)
House age (years)	58.9 (14.7)	61.1 (17.7)

+ Standard deviation is provided in parentheses.

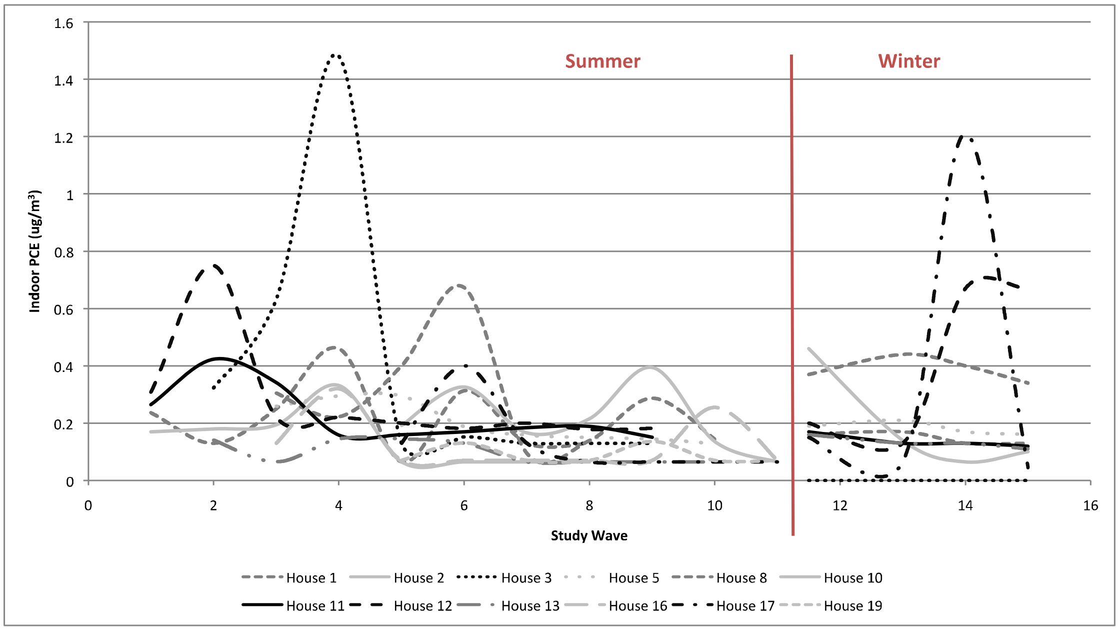

Figure 2.5. Temporal variation in indoor PCE concentrations in homes with at least one sample above the detection limit.

2.3.1. Within-Home Temporal Variability

Table 2.3 shows the results of the regression model examining the effects of weather variables on within-home temporal variability in PCE concentrations. As shown, barometric pressure drop, wind speed, relative humidity, and season all significantly predict the observed temporal variations. Specifically, indoor concentrations increase with magnitude of the pressure drop (p=0.048) and humidity (average marginal effect p<0.001), while concentrations decrease as wind speed increases (p<0.001) and during winter (p=0.001). As noted above, a similar relationship between wind speed and indoor concentration has been observed in previous studies of radon (Nazaroff & Doyle, 1985; W. Nazaroff et al., 1985; Turk et al., 1990). A recent, detailed study of a single house at a vapor intrusion site did not observe strong correlations between seasonal winds and vapor intrusion but suggested that wind may contribute to short-term vapor intrusion changes (Holton et al., 2012). In the model, humidity may be capturing some of the short-term effects of rainfall, such as groundwater rise. During rain events, humidity levels exceed 90%, while the average during the study period was 62%. In summary, for this community based on the regression analysis, it is expected that PCE concentrations in homes that are prone to vapor intrusion PCE concentrations will be higher during summer, during low-wind events, or when large barometric pressure drops occur.

Table 2.3. Average marginal effects for the within-home variability of natural log PCE indoor air concentration (ln-μg/m^3) due to vapor intrusion.

Covariate	Coefficient+
Weather characteristics	
Barometric pressure drop (mm Hg)	0.11*
	(0.051)
Average wind speed (m/s)	-0.26**
	(0.093)
Relative humidity (%)	0.29**
	(0.017)
Relative humidity squared (%)	-0.0017
	(0.00051)
Winter season	-2.50**
	(0.71)
Constant	-12.42**
	(2.48)

Note: There were 120 observations from 13 homes. Standard errors (in parentheses) were computed using clustered robust standard errors.

+McFadden's pseudo R^2: 0.2887. F-statistic: 3.98 ($p<0.0001$)
* Statistically significant at the 5% level.
** Statistically significant at the 1% level.

2.3.2. Spatial (Between-Home) Variability

Table 2.4 shows the regression model exploring the effects of household, environmental, and meteorological characteristics on between-home variability. The model results include all samples, including those from homes in which no indoor PCE was detected. As shown, indoor air PCE concentrations increase with groundwater concentration (expressed as a logarithmic term, p=0.030), a slab-on-grade foundation (p=0.028), magnitude of the barometric pressure drop (p=0.036) and humidity (expressed as a quadratic term, p=0.04). On the other hand, concentrations decrease in the absence of an air conditioning unit (presumably because windows are opened, p=0.015) and with wind speed (p=0.004). Although not statistically significant, larger homes tended to have higher indoor PCE concentrations, while lower PCE concentrations were measured in older (and presumably leakier) homes. Together, all of these included covariates are highly significant ($p<0.001$).

Table 2.4. Population-averaged effects of model covariates on between-home (spatial) variability of natural log PCE indoor air concentration (ln-µg/m^3) due to vapor intrusion.

Variable	Coefficient+
PCE concentration	
Log of groundwater concentration	0.16*
(log-µg/L)	(0.07)
Household characteristics	
Slab-on-grade foundation	0.83*
	(0.37)
No air conditioning units	-0.51*
	(0.20)
House area (m^2)	0.0017
	(0.0019)
Age of home (years)	-0.0060
	(0.0057)
Environmental characteristics	
Houston Black clay soil	-0.46*
	(0.23)
Weather characteristics	
Barometric pressure drop (mm Hg)	0.15*
	(0.07)
Average wind speed (m/s)	-0.36**
	(0.12)
Humidity (%)	0.051*
	(0.02)
Relative humidity squared (%)	-0.002**
	(0.0006)
Winter season	-2.84**
	(1.01)
Constant	-4.45**
	(1.13)

Note: There were 182 observations from 20 homes. Clustered robust standard errors in parenthesis.
+McFadden's pseudo R^2: 0.3389. F-statistic: 7.96 ($p<0.0001$)
* Statistically significant at the 5% level.
** Statistically significant at the 1% level.

2.4. Discussion

The indoor air concentrations observed in this study were similar to the results previously found in the EPA investigation (see Figure 2.1). The highest concentration measured by the EPA summa canister (1.83 $\mu g/m^3$) was on par with the highest observations in this study (1.50 $\mu g/m^3$). We do, however, observe a short-term temporal variability in indoor air concentrations that cannot be captured with a single-point-in-time sampling event. The relationship between meteorological conditions and indoor PCE observed here could help indicate the potential range of concentrations when only a single measurement is possible.

Several previous studies have found relationships between meteorological variables and vapor intrusion similar to those observed here. Radon studies have shown that atmospheric pressure drops contribute to the total radon entry rate into a building and can increase indoor concentrations by a factor of two over a daily time scale (Holford, Schery, Wilson, & Phillips, 1993; Robinson, Sextro, & Riley, 1997). Although atmospheric pressure fluctuations do not produce a net positive flow rate into homes over longer time intervals, they cause short-term changes in radon entry because of increases in the pressure differential between the subslab and indoor air (Robinson et al., 1997). That is, indoor air responds more quickly than subslab air to an ambient pressure change, leading to short temporal variations in subslab-indoor air pressure differentials that, in turn, affect advective flow of contaminant vapors into buildings. Similarly, a previous investigation of the intrusion of (unchlorinated) hydrocarbon vapors into a building in Australia found that semidiurnal decreases in barometric pressure caused a negative pressure differential between the building interior and subslab, increasing the rate of advective mass transfer of hydrocarbons into the indoor air (Patterson & Davis, 2009). Our analysis also suggests that an ambient pressure drop may increase the mass of PCE flowing into the home.

In studies in northern climates in homes with basements, higher concentrations of CVOCs have been observed in the winter compared with other seasons (Fitzpatrick & Fitzgerald, 2002; Holton et al., 2012). The inverse relationship observed here, in the hot and arid San Antonio climate, may be partly explained by the tighter sealing of homes during the summer months (to keep out the heat), higher subsurface and groundwater temperatures, desiccation of the shallow soils, or any combination of these. It should be noted that the temperatures in February and March 2012 were mild, ranging from 12 to 21°C. A detailed survey of homes in Houston, Texas, with characteristics similar to households in this study observed the lowest air exchange rates in the summer (Yamamoto, Shendell, Winer, & Zhang, 2009). Seasonal data on vapor intrusion in southern climates is limited; however, summertime increases in radon concentrations were observed in Alabama homes with crawl spaces and were attributed to preferential flow pathways associated with the area's karst geomorphology (Wilson, Gammage, Dudney, & Saultz, 1991). Using air conditioning has also been associated with higher indoor radon concentrations (Radford, 1985). This observation is worth further investigation because it suggests that seasonal effects on vapor intrusion in southern climates may differ from those in northern climates. However, the small sample size (necessitated by funding limitations) and selection criteria for resampled homes may have biased our observations.

The results of this study are limited by the small sample size and few homes studied compared with the size of the potentially affected population. Measurements were not taken during a rainstorm or during freezing conditions. The relationships identified here may not be generalizable to other sites, especially those with a different climate, hydrogeology, and housing stock. The homes included in the study were a convenience sample, not a random sample. Therefore, selection bias among the households that chose to participate in the study may have influenced the results of the analysis. The passive sampling devices showed relatively good precision, although in some cases the difference in the duplicates was high and the use of the average of the measurements may have biased the results. We did not collect summa canister samples (to compare the accuracy of the passive sampling devices), nor were ambient air samples collected. The sampling of the EPA and previous sampling by the local health department gave no indication of elevated ambient levels of PCE, but possible intrusion from outdoor sources may have affected our measurements.

Also worth noting are the potential advantages of the community-based research design used in this study. The study required daily access to each home and the motivation of residents to complete the entire protocol (including foregoing use of products that may contain PCE). Experience at other sites has suggested that access to homes is a barrier to data collection in vapor intrusion studies, particularly because at such sites animosity may exist between the community, those responsible for the potential pollution, and the involved government agencies (Siegel, 2009). In this case, participants appeared to adhere to the study protocol and participated for the duration of the study.

2.5. Conclusions

This study provides evidence of spatial variability as well as short-term and seasonal variability in PCE concentrations due to vapor intrusion. These results suggest that a single-point-in-time sample of indoor air in homes at risk of vapor intrusion is not adequate for characterizing the temporal variability in exposures. This study contributes to the body of evidence suggesting that vapor intrusion potential fluctuates on short and seasonal time scales and suggests that evaluating temporal variability is needed to adequately characterize the occurrence of vapor intrusion in a home. While PCE concentrations detected at this site did not exceed the new PCE risk-based standards, acknowledging spatial and temporal variability as well as understanding the drivers of these processes may be significant in designing and conducting vapor intrusion investigations at other sites, where concentrations may exceed EPA's standards.

CHAPTER 3

Screening Houses for Vapor Intrusion Risks: A Multiple Regression Analysis Approach[2]

3.1. Introduction

When groundwater or soil contamination occurs near buildings, volatile contaminants can migrate upwards and result in vapor-phase contaminant intrusion into the indoor air, a phenomenon called vapor intrusion. Chlorinated volatile organic compounds (CVOCs), which are among the most frequently detected groundwater contaminants at hazardous waste sites in the United States, persist in the environment, are difficult to remediate, and hence may pose long-term exposure risks (Agency for Toxic Substances and Disease Registration, 2007; Fischer et al., 1996; McCarty, 2010; Simpkin & Norris, 2010; Travis & Doty, 1990).

Policy debates concerning how to evaluate and minimize vapor intrusion risks are ongoing. Key questions include (Schuver, 2007; U.S. Environmental Protection Agency, 2009):

1) What constitutes sufficient evidence of a complete vapor intrusion pathway?
2) How can those responsible for contaminated sites identify which homes are at the highest risk for vapor intrusion?

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[2] *Johnston, J.E., & MacDonald Gibson, J. 2013, in press. Screening houses for vapor intrusion risks: A multiple regression analysis approach.* Environmental Science & Technology.

3) In which homes should vapor barriers, exhaust systems, or other measures be put in place in order to prevent exposure to vapors from subsurface contaminants?

At many contaminated sites, hundreds or even thousands of buildings may be affected by vapor intrusion. Due to political, technical, and financial constraints, monitoring indoor air directly in every potentially affected home is typically infeasible. Hence, decision-makers employ screening tools to categorize buildings according to the level of potential vapor intrusion risk. The current draft U.S. Environmental Protection Agency (EPA) guidance document on vapor intrusion, released in 2002, proposes a sequential order of assessment steps to “screen in” sites for further, and increasingly more site-specific, investigation (U.S. EPA, 2002). The suggested protocol begins with an examination of the source of vapors (contaminated groundwater or unsaturated soils), proceeds to monitoring soil gas in the unsaturated zone above the source, and, if there is evidence of vapor intrusion, continues upward to collect samples at the exposure point (e.g., indoor air or sub-foundation vapor). Buildings may be designated as not requiring further investigation at any of these steps.

As part of this sequential screening process, the EPA recommends the use of a generic screening-level model to determine whether site-specific data (such as indoor air measurements) should be collected. This initial assessment applies an “attenuation factor” to measured concentrations of contaminants in groundwater to predict the potential indoor air concentrations due to vapor intrusion. This attenuation factor is intended to represent the decrease in contaminant concentrations that occurs as the contaminant vapor migrates upward from the groundwater table, through the overlying soil, and into buildings; it is defined as (Johnson & Ettinger, 1991):

$$\alpha = \frac{C_{indoor}}{C_{source}} \tag{1}$$

where C_{indoor} is the contaminant concentration in indoor air (mass/volume) and C_{source} is the contaminant concentration in the soil gas just above the water table (mass/volume). Currently, the EPA recommends employing a generic attenuation factor of 1/1,000 to every building where the groundwater table is at least 5 feet from the ground surface, implying a contaminant concentration decrease of at least three orders of magnitude as the contaminant migrates upward into the building.

Previous reviews of empirical data at vapor intrusion sites have identified measured attenuation factors that range over several orders of magnitude. A comparison of attenuation factors found values as high as 0.1, with site averages ranging from 10^{-6} to 10^{-2} for chlorinated solvents (Johnston & MacDonald Gibson, 2011). Such results suggest that a factor of 1/1,000 could underpredict vapor intrusion risks (i.e., not be sufficiently conservative) in some cases. Others have suggested that the EPA generic attenuation factor is overly conservative—that, in practice, observed attenuation factors usually are significantly lower than 1/1,000 (Folkes, Kurtz, & Wannamaker, 2007; Johnson, Ettinger, Kurtz, Bryan, & Kester, 2009).

The transport of CVOCs from groundwater to indoor air is complex and incompletely understood. Differences in building construction, spatial variations in geology and soil type, and temporal and spatial variability in vadose zone transport processes and depth to ground water all may influence vapor migration, but existing models incorporating these factors still lack sufficient accuracy in predicting the substantial spatiotemporal variability observed empirically (Bozkurt, Pennell, & Suuberg, 2009; Hers, Zapf-Gilje, Evans, & Li, 2002; Hers & Zapf-Gilje, 2003; Johnson, 2005; McDonald & Wertz, 2007; Pennell, Bozkurt, & Suuberg, 2009; Tillman & Weaver, 2006).

In order to provide a data source for further studying factors that influence vapor intrusion, the EPA compiled the National Vapor Intrusion Database. This database represents the largest collection of vapor intrusion data in the United States (Dawson, 2008b; U.S. EPA, 2012), containing as of 2012 almost 2,400 indoor air observations collected during 1990-2007 in 913 buildings at 41 sites in 15 states. EPA personnel reviewed and quality-assured the data prior to inclusion in the database (U.S. EPA, 2012). The data represent a cross-sectional collection of vapor intrusion observations; most sites do not include multiple measurements in multiple buildings over time. A detailed description of the database, data sources, and included parameters is available from U.S. EPA's (2012) vapor intrusion database report.

This paper provides the results of the first systematic multivariate analysis of the EPA's vapor intrusion database. We employ a multivariate regression approach to evaluate the effects of contaminant properties, geologic conditions, groundwater depth, soil type, building foundation type, and season on the observed vapor attenuation factors. Our analysis focuses on chlorinated solvents, which are among the most common contaminants in groundwater and which are present in 98% of observations in the data set.

3.2. Methods

3.2.1. Dependent Variable

The dependent variable in this analysis is the attenuation factor (Equation 1). Hence, only observations for which paired data on groundwater and indoor air contamination were available were eligible for inclusion (~35% of the data). Using average groundwater temperature and adjusted chemical-specific Henry's constants, the measured groundwater concentrations were converted into groundwater-source vapor concentrations (μg/m^3), and then calculated the groundwater vapor intrusion attenuation factor as follows:

$$\alpha_i = \frac{C_{i,indoor}}{C_{i,gw} \times K_{H,i} \times \frac{1000L}{m^3}} \qquad (2)$$

where α_i is the vapor attenuation factor for chemical *i*, $C_{i,indoor}$ is the concentration of the chemical *i* indoors due to vapor intrusion (μg/m^3), C_{gw} is the concentration of the contaminant in the groundwater (μg/L), and $K_{H,i}$ is the chemical-specific Henry's constant for the average temperature of the groundwater (unitless). As Johnson et al. (2009) recommend, in order to avoid biasing the results we excluded samples with groundwater or indoor air concentrations below the detection limit.

CVOCs are often detected in indoor air even in areas not affected by contaminated soil or groundwater (Dawson & McAlary, 2009), due to the presence of indoor sources (e.g. cleaning products) and/or infiltration of contaminated outdoor air. The EPA marked all database entries (16.3%) they suspected of being confounded by non-vapor-intrusion sources (due, for example, to a lack of pre-screening for and removal of indoor sources) (Dawson & McAlary, 2009; Johnson et al., 2009; U.S. EPA, 2012). Our analysis excluded these observations.

3.2.2. Explanatory Covariates

Covariates in this analysis included geological, environmental, household, and chemical characteristics. We grouped contaminated sites into six geological groundwater regions, reflecting similarities in the composition, arrangement, and structure of subsurface formations as well as in broad water storage and transmission characteristics (Heath, 1984). Other environmental covariates were the depth to groundwater (m) and soil type. Soil type was classified as fine-grained (predominantly clay or silts), coarse (sandy soils), or very coarse (sand plus pebbles or rocks) generally based on the coarsest soil described in the vadose zone at the site.

Four types of foundations typified buildings in the database and hence also were considered as covariates: basement, slab-on-grade, crawl space, or partial basement. Buildings were further divided into residential, mixed-use, or commercial/institutional. In the final model, 96% of the observations were categorized as residential, so we restricted the analysis to residential buildings.

Seven CVOCs were included in the final analysis: 1,1-dichloroethane, 1,1-dichloroethylene (DCE), 1,1,1-trichloroethane, cis-1,2-dichloroethylene, tetrachloroethylene (PCE), trichloroethylene (TCE), and vinyl chloride. Since the chemical properties are important parameters in fate and transport models, we represented contaminants with their molecular weight (g/mol) and diffusion coefficient in air (cm^2/s), assuming standard temperature and pressure.

3.2.3. Statistical Analysis

Since the attenuation factors were right skewed, the regression analysis employed the logarithm of the observed attenuation factor as the dependent variable (see Appendix B, Figure B.1). The associations between the vapor intrusion attenuation factor and environmental parameters, household characteristics, and chemical properties were then explored using multivariate statistical techniques, as described in the following sections.

Multivariate Regression Model

Our first regression analysis fitted the following model to the pooled data:

$$\log y_i = \alpha + \mathbf{B}\mathbf{x}_i + \varepsilon_i \tag{4}$$

where y_i is the vapor intrusion attenuation factor for observation i, $\mathbf{x}_i$ is the vector of model covariates, $\mathbf{B}$ is the vector regression coefficient, and ε_i is the residual vector. The data exhibit positive spatial autocorrelation (the tendency for measurements in close spatial proximity to share similar attributes). Classical regression techniques applied to such mixed-level data often exaggerate levels of statistical significance of coefficient estimates (Moulton, 1990; Wooldridge, 2003). Our first multivariate pooled regression approach controlled for such site-level clustering by adjusting the standard error, replacing the independence-of-errors assumption with an independence-between-clusters assumptions, and employing a variant of Huber-White heteroskedasticity-consistent standard errors (Huber, 1972; Arellano, 1987; Graubard & Korn, 2006;). This approach allows the variance of the error term to vary by site, rather than remaining constant across all sites.

Multilevel Regression Model

Second, we implemented a multilevel linear regression model that views the data as arising from a hierarchical process in which individual buildings are nested within sites that share common characteristics and sites are, in turn, nested in regions with geologic similarities (Figure 3.1). Multilevel statistical techniques provide a technically robust analytical framework when the causal processes that affect the outcome are hypothesized to operate in such a nested fashion (Gelman & Hill, 2006; Subramanian, Jones, & Duncan, 2003). Exploratory analysis of the attenuation factor data suggests that a hierarchical framework may be appropriate (see Appendix B, Figure B.2). For hierarchical data, a pooled regression estimator of the effect of an observation-level predictor may be biased when using a flat regression approach such as in our first model (Steenbergen & Jones, 2002). The multilevel approach allowed us to examine the influence of building-specific characteristics on the attenuation factor as well as whether site-level and macro-scale geological-level contextual factors influence the attenuation factor when accounting for building-specific parameters.

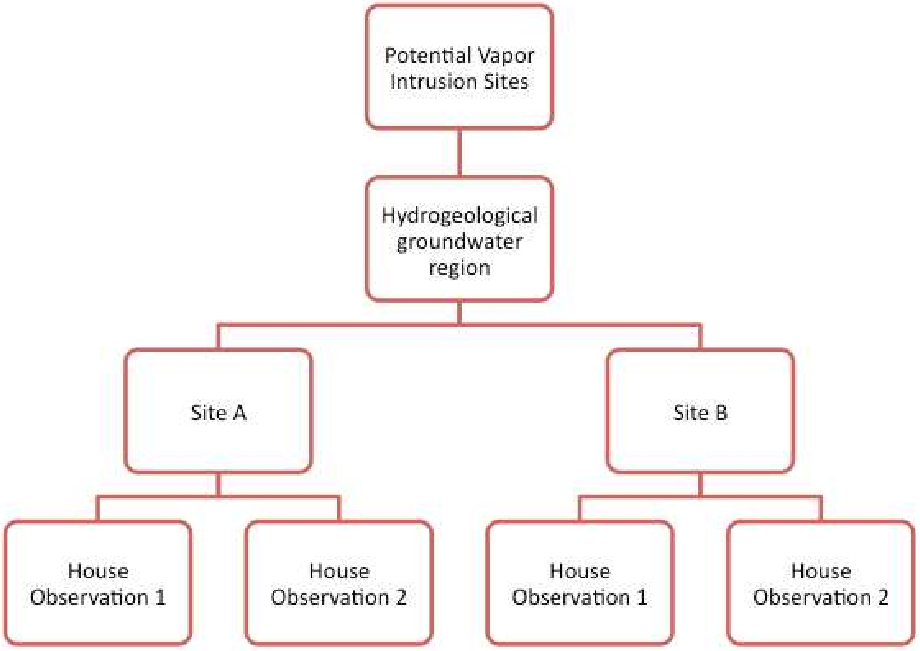

Figure 3.1. Schematic of nested multilevel model.

The multilevel approach models the intercept of each level as random, assuming that building i is nested within site j, which in turn is nested within geological region k:

$$\log y_{ijk} = \boldsymbol{\pi}_{0jk} + \sum_{p=1}^{P} \boldsymbol{\pi}_{pjk} x_{pjk} + \boldsymbol{\varepsilon}_{ijk} \tag{5}$$

where π_{0jk} is the intercept, π_{pjk} is the vector of regression coefficients and x_{pijk} is the vector of explanatory variables at the observation level. Assuming there are Q level-2 predictors and allowing z_{qjk} to be the qth predictor in site j influencing attenuation, then the model is further specified as:

$$\pi_{0jk} = \beta_{00k} + \sum_{q=1}^{Q} \beta_{0qk} z_{qjk} + \delta_{0jk} \tag{6}$$

where β_{0qk} and z_{qjk} are the vectors for the regression coefficients and explanatory variables, respectively. Finally, the level-3 intercept-only model is given by:

$$\beta_{00k} = \gamma_{000} + \upsilon_{00k} \tag{7}$$

The model further assumes that the components of the unobserved random effects, ε_{ijk}, δ_{0jk} and υ_{00k}, are independent and normally distributed with means of zero.

We used the empirical Bayes prediction method described by Rabe-Hesketh & Skrondal (2008) and the iterative generalized least squares maximum likelihood estimator as implemented in STATA IC 12 (StataCorp, College Station, Texas) to fit the model in Equations 5-7 to the data. Functional forms and interactions between variables were evaluated using the Akaike information criterion (AIC) and the likelihood ratio test. An *a priori* significance level of 0.05 was used.

3.3. Results and Discussion

After excluding observations with missing data for one or more variables, the study sample comprised 370 measurements from 21 sites, 84% of which were contaminated with DCE, PCE, and/or TCE. The majority of sites were located in the nonglaciated central groundwater region (n=235), but all six regions were included in the final models. Of the included data, the mean vapor intrusion attenuation value was 0.0008 (sd=0.005), but the observed values extended over five orders of magnitude. Among this sample, 11.6% of observations exceeded the suggested EPA screening level of 0.001. Table 3.1 shows the summary statistics for the subgroups of observations included in the analysis.

3.3.1. Regression Models

The results from the regression analyses reveal several statistically significant determinants of attenuation factors among residential buildings overlying chlorinated groundwater plumes. The final regression models used a logarithmic transformation of groundwater depth as the best-fit functional form. The coefficients of the two regression models follow similar patterns significance. Table 3.2 presents the results from both models. We use the multilevel model as the baseline for the discussion because it has the lowest AIC value, indicating the best fit. The models reveal several statistically significant determinants of attenuation factors among residential buildings overlying chlorinated groundwater plumes, as discussed in the following sections.

Table 3.1. Summary statistics for the key continuous variables included in the regression model.

Variable	Observations (n=370)	Observations if alpha>0.001 (n=44)
Environmental characteristics		
Vapor intrusion attenuation factor	0.0008 (0.005)[a]	0.0075 (0.016)
Groundwater depth (m)	4.35 (2.42)	2.70 (1.53)
Winter season	87%	91%
Soil type		
Fine-grained soil	34%	18%
Coarse-grained soil	60%	53%
Very coarse-grained soil	6%	29%
Foundation type		
Basement	49%	85%
Crawl space	26%	0
Slab-on-grade	16%	12%
Mixed foundation type	9%	3%
Chemical characteristics		
Molecular weight (g/mol)	128.4 (24.91)	137.6 (19.7)
Diffusivity in air (cm^2/s)	0.079 (0.0061)	0.077 (0.0032)
Groundwater hydrogeologic regions		
Alluvial basins	2.9%	12%
Atlantic & Gulf coastal plain	11%	11%
Colorado plateau & Wyoming basin	0.8%	0
Glaciated central region	16%	53%
Nonglaciated central region	63%	26%
Northeast & superior uplands	5.7%	9%

[a] Standard deviation is provided in parentheses for continuous variables.

Table 3.2. Effects of model covariates on variation in the log groundwater vapor intrusion attenuation factor.

Variable (n=370)	Model 1[a]	Model 2
	Clustered OLS	Multilevel
Chemical characteristics		
Molecular weight (g/mol)	0.002	0.0054**
	(0.002)	(0.0021)
Diffusivity in air (cm^2/s)	-10.93	-14.74
	(8.89)	(9.25)
Environmental characteristics		
Log-groundwater depth (m)	-0.92***	-0.79***
	(0.25)	(0.29)
Soil type (reference: coarse-grained)		
Fine-grained soil	-0.53	-0.58**
	(0.41)	(0.29)
Very coarse-grained soil	1.29***	0.66*
	(0.19)	(0.39)
Log-groundwater depth × fine-grained	0.75	0.55
	(0.88)	(0.53)
Log-groundwater depth × very coarse-grained	-1.70***	-1.84**
	(0.25)	(0.74)
Season (reference: summer)		
Winter season	0.45*	0.42***
	(0.23)	(0.15)
Foundation type (reference: basement)		
Crawl space	-0.37	-0.41*
	(0.40)	(0.22)
Slab-on-grade	0.78	0.67*
	(0.52)	(0.36)
Partial basement	1.47***	1.83***
	(0.24)	(0.67)
Crawl space × winter	-1.37***	-0.99***
	(0.45)	(0.38)
Slab-on-grade × winter	0.11	0.17
	(0.42)	(0.25)
Partial basement × winter	-1.12***	-1.55**
	(0.32)	(0.74)
Variance components		
Groundwater geological region υ_{00k}^2	--	0.24
		(0.16)
Site-level δ_{0jk}^2	--	0.26
		(0.10)
Observation-level ε_{ijk}^2	--	0.65
		(0.025)
Constant	-7.46***	-3.53***
	(1.86)	(0.92)
Akaike information criterion	846.58	793.02
Log likelihood	-478.15	-378.51

[a] Robust standard error reported in parentheses; * p<0.1, ** p<0.05, ***p<0.01

Environmental and Chemical Covariates

In both models, chemicals with higher molecular weights have slightly higher attenuation factors (statistically significant in Model 2, p=0.011), while those with high diffusivity in air have slightly but not significantly lower (p=0.11) attenuation factors. PCE and TCE have the highest molecular weights among the included contaminants and were also the most frequently observed in this dataset. The general solubility trend among chlorinated solvents is that as the number of chlorine atoms on a compound increases (which in turn increases the molecular weight), the aqueous solubility decreases and the octanol-water partition coefficient increases; these factors may influence contaminant partitioning from water into the vapor phase (Cwiertny & Scherer, 2010). The variance of air diffusivity between chemicals is relatively small (mean= 0.079, sd = 0.006). Our results concur with previous deterministic models of the vapor intrusion pathway concluding that small variations in diffusivity do not have a significant impact on the final predictions (Yao, Shen, Pennell, & Suuberg, 2011).

In the case of groundwater depth, we found a nonlinear relationship that is negative and statistically significant in both regression models (p=0.008 for Model 2). This finding is consistent with physical transport models and previous studies showing that as the distance between the structure and the source increases, more attenuation is expected (Johnson, 2005; Tillman & Weaver, 2006).

Compared to coarse-grained soil, fine-grained soil is associated with a significantly lower attenuation factor (p=0.046), while very coarse-grained soil is associated with a marginally higher attenuation factor (p=0.092). Soil type is a proxy for characteristics that influence the transport of vapor, namely hydraulic conductivity and porosity, and others have concluded that soil type is a strong indicator of vapor intrusion potential (Bozkurt et al., 2009; Hers & Zapf-Gilje, 2003; Pennell et al., 2009; Tillman & Weaver, 2006). A Wald's test of the interaction between soil type and groundwater depth shows high joint significance (p=0.004). This suggests that the relationship between groundwater depth and the attenuation factor is mediated by soil type. Shallow groundwater (less than 3 m below ground level) coupled with coarse or very coarse soil type puts a site at risk for higher-than-expected attenuation factors, that is, increases the possibility of exceeding the conservative screening estimate of 0.001.

Household and Seasonal Characteristics

Foundation type was the principal descriptor of household properties in this study. A Wald's test indicated that the categorical construct used to represent foundation type in this analysis was highly significant (p<0.001), even though, slab-on-grade and crawl-space foundations were only marginally significant (p=0.066 and p=0.064, respectively) when compared individually to basement foundations. In both models, homes with crawl space foundations were associated with a lower vapor intrusion factor, while those with slab-on-grade foundations were associated with higher intrusion factors when compared to homes with basements.

Season also significantly influenced the attenuation factor, as did the interaction between season and foundation type. On average, attenuation ratios in the winter were higher than in summer (p=0.007). However, for crawl-space homes, the seasonal effect was opposite, with higher intrusion factors in summer than in winter (p=0.010). Attenuation factors increased more in winter in slab-on-grade foundations than in homes with basements, but this difference was not statistically significant.

Figure 3.2 simulates the predicted relationships between soil type, foundation type, season and groundwater depth based on the multilevel model. In general, slab foundation and very coarse soil type are associated with the highest attenuation factors; at shallow groundwater levels, homes with these characteristics are predicted to experience much more vapor intrusion than the current generic attenuation factor (0.001) would indicate, as Figure 3.2 shows. We also observe that homes with crawl-space foundations are expected have attenuation factors below the screening level under almost all conditions, although these homes also show the widest range of predicted attenuation factors.

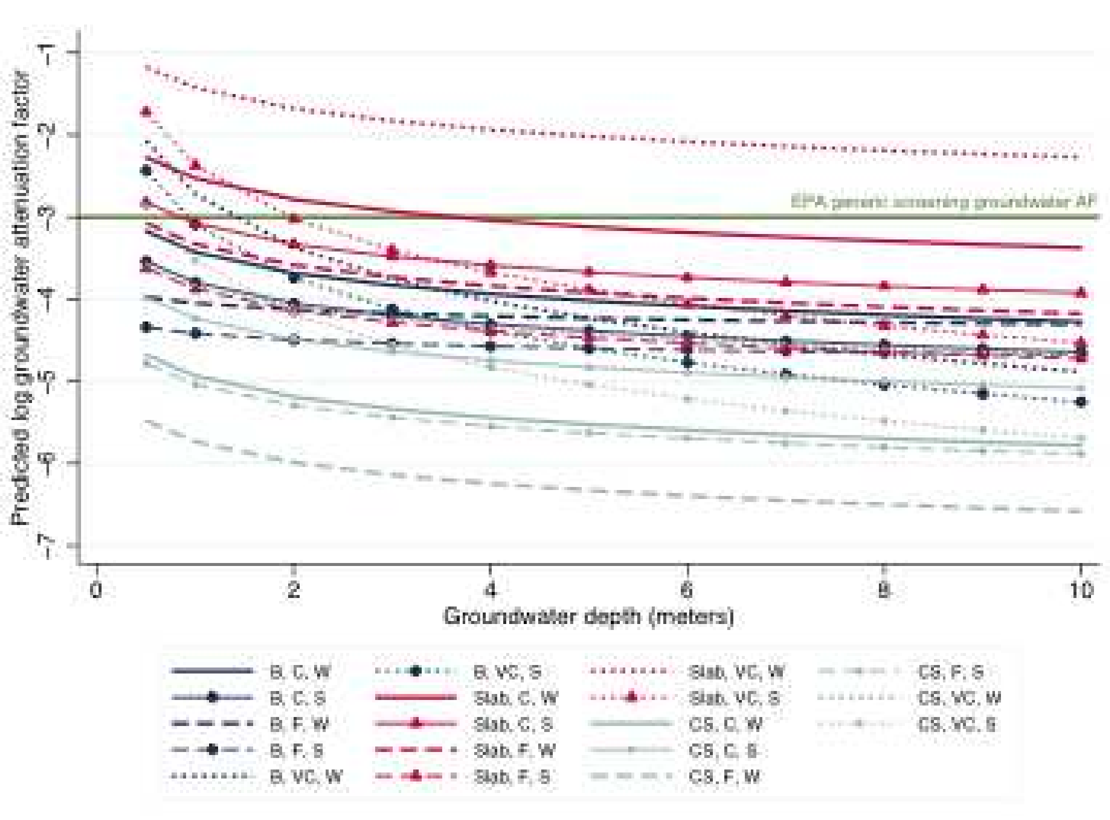

Figure 3.2. Predicted (log) vapor intrusion attenuation factor based on multilevel model for PCE, assuming mean groundwater region and site-level characteristics for various combinations of groundwater depth (m), soil type, foundation type, and season. B: basement; Slab: slab on grade; CS: crawl space; C: coarse-grained soil; F: fine-grained soil; VC: very coarse-grained soil; W: winter; S: summer.

Prior research on the physical mechanics of vapor intrusion suggests similar results to what we have observed: Differences in both construction styles and ventilation rates have been observed to contribute up to two orders of magnitude to the between-home variability in actual indoor air concentrations (Hers & Zapf-Gilje, 2003). Research at one well-studied site found statistically higher attenuation factors for slab-on-grade homes compared to basement or crawl-space homes (Folkes, Wannamaker, & Kuehster, 2004). Other studies have found only a weak relationship between attenuation factors and construction type, although these conclusions are based on modeling and simulations rather than field observations (Abreu & Johnson, 2005; Johnson & Ettinger, 1991). While vapor intrusion can occur in homes with any foundation type studies, our analysis suggests that foundation type significantly influences the magnitude of the attenuation factor.

Like the present study, other studies in northern climates in homes with basements have found higher concentrations of indoor CVOCs in the winter compared to other seasons, although the mechanisms underlying this phenomenon are not fully understood (Fitzpatrick & Fitzgerald, 2002; Holton et al., 2012; Luo, Holton, Dahlen, & Johnson, 2011; McHugh et al., 2007). One hypothesis posits that the differential pressure gradient across the foundation increases during the heating season as a result of indoor-outdoor temperature differences, increasing the vapor flow rate into the home (Nazaroff, Lewis, Doyle, Moed, & Nero, 1987).

The effects of foundation type and season, and the interaction between these two factors, that our regression analyses revealed for CVOCs is consistent with results of previous studies of the intrusion of radon vapors into homes. For example, a cross-sectional study of indoor radon concentrations in Danish homes found the highest mean radon concentrations in slab-on-grade homes and partial basement homes, followed by homes with crawl-space and full basements (Ulbak et al., 1988). A further review of radon in Danish homes found statistically significant seasonal differences observed in slab-on-grade and partial basement homes (Majborn, 1992). Among the data analyzed in the present study, foundation type influenced vapor attenuation, with seasonal effects the strongest for partial basement foundations and the highest mean concentrations predicted for slab-on-grade foundations (Figure 3.2). For crawl-space foundations, we observed the opposite seasonal effect, that is, higher concentrations, on average, in the summer compared to the winter. While field studies of vapor intrusion in crawl-space homes are limited, summertime increases in radon concentrations have been observed in Alabama homes with crawl spaces (Wilson, Gammage, Dudney, & Saultz, 1991).

Higher-Level Characteristics

Compared to the classical regression model, we find that including higher-level predictors in a hierarchical model offers a statistical improvement to an ordinary least squares (OLS) model, according to the likelihood ratio test (p=0.001). Using the flat data structure of OLS, but accounting for data clustering, we find coefficient estimates comparable to those in the multilevel model. However, in the OLS model, the post-estimation adjustment of standard errors may produce inefficiency in the estimates' error, masking statistically significant effects that become apparent in the multilevel model.

3.3.2. Predictions

We evaluated the ability of the models to serve as a predictive screening tool for the vapor attenuation factor (Figure 3.3). The correlation between measured and predicted attenuation factors is 41% for the multilevel model and 36% for the OLS model, indicating that these models have explanatory power but are not sufficient to accurately estimate vapor intrusion risk on a house-by-house basis. The correlation between the attenuation factors predicted by the two models is 95%, suggesting that the models yield very similar estimates.

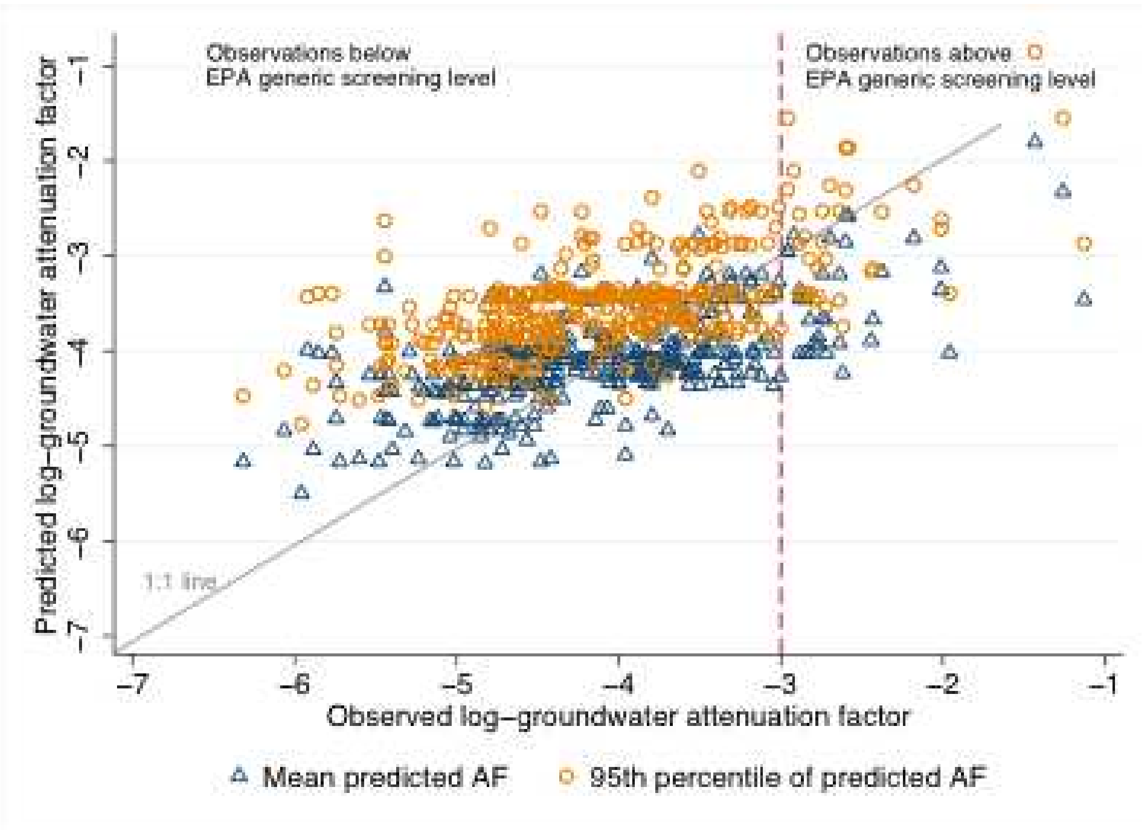

Figure 3.3. Comparison of the log mean predictions (blue) and 95th percentile predictions (orange) with the actual log of the attenuation factor. Dashed red lines represent the current EPA screening level.

In Table 3.3 we compare the performance of the models by dividing the measured attenuation factor into four groups and examining the frequency of over-/underprediction as well as the magnitude of the error (shown by the root mean squared error). In the case of the 95th percentile multilevel predictions, the model consistently overpredicts attenuation factors below 10^{-4}; however, the magnitude of these overpredictions is less, on average, than occurs when applying the generic EPA screening level. For example, for the observations with attenuation factors below 10^{-5}, the mean prediction for this cohort is 0.0001, an order of magnitude lower than the generic EPA screening level. For measured attenuation factors above 10^{-4}, the results are more mixed. Homes with higher attenuation factors are often the most at risk to high indoor air concentrations. Thus, to protect health, it is perhaps more important to be conservative in the estimations among this group of observations. The 95th percentile predictions are conservative (i.e., over-predict attenuation factors) in most cases (68%) and identified 72% of the homes with attenuation factors exceeding the current screening criterion of 0.001.

Table 3.3. Comparison of three methods for predicting groundwater attenuation factors to measured factors.

	Measured Attenuation Factor											
	$\alpha < 10^{-5}$			**$10^{-5} \geq \alpha < 10^{-4}$**			**$10^{-4} \geq \alpha < 10^{-3}$**			**$\alpha \geq 10^{-3}$**		
	High[+]	Low[+]	RMSE*	High	Low	RMSE	High	Low	RMSE	High	Low	RMSE
EPA generic screening factor	47	0	2.47	149	0	1.55	132	0	0.67	0	43	0.68
Multilevel model, mean	46	1	1.00	116	35	0.46	18	114	0.54	5	38	1.06
Multilevel model, 95th percentile	47	0	1.57	148	1	0.94	94	38	0.48	26	17	0.61

[+] High: model predictions exceed the observed groundwater attenuation factor (over-predictions); Low: model under-predict the observed attenuation.
*RMSE: root mean squared error (calculated based on log(α))

The predictions offered by the regression models developed in this analysis, coupled with an understanding of the relationships between various house and site-level conditions, could provide more nuanced screening-level information than a generic attenuation factor to support decision-making at potential vapor intrusion sites. In most cases, the 95th percentile predictions are slightly conservative, but allow for a range of screening values based on regional, site, and home characteristics.

This analysis is limited by the sample size and the lack of data on several key variables shown to be important in modeling vapor intrusion. While 21 different sites are represented in the final model, they are clustered in the northern half of the United States. Different weather, soil, and groundwater patterns in the southern United States may influence the relationships observed there (Johnston & MacDonald Gibson, 2013). While quality control measures were completed prior to inclusion of information into the database, the values are subject to measurement error as the protocols, classification systems, and sampling techniques varied by site. Soil type is used to approximate the ease with which vapors can flow through the soil but is a crude proxy for the true below-ground vapor flow rate, as the classification system fails to capture the presence of high-permeability flow paths (Garbesi & Sextro, 1989; Johnson, 2005). Further, previous research has shown that the attenuation factor is sensitive to air exchange rate, which regulates the ability of vapors to accumulate indoors (Johnston & MacDonald Gibson, 2011; Tillman & Weaver, 2006). The air exchange rate can be highly variable between houses depending upon heating/cooling systems, opening of windows, and the energy efficiency of a building. For future efforts, collected information on the foundation area and the age of the home may improve upon this model, as both sources of information are typically already available from other sources. While air exchange rate is not easily acquired, house age and foundation area have been shown to be a reasonable proxy for the leakiness of a structure (Chan, Nazaroff, Price, Sohn, & Gadgil, 2005).

3.4. Conclusions

This analysis provides insights into the effects of various household and environmental characteristics on the vapor intrusion attenuation factor, based on the largest currently available vapor intrusion dataset. The available data suggest that the relationship between vapor intrusion and groundwater concentration depends on soil type, groundwater depth, foundation, and season, with the effects of foundation varying by season. Our multivariate approach suggests that slab-on-grade foundations, shallow groundwater, and very coarse soil increase the risk of a home to vapor intrusion. In the majority of cases, the multilevel model resulting from our analysis was able to identify homes with vapor attenuation ratios above the current EPA screening level. This modeling approach may prove increasingly useful as more sites are identified as at-risk for vapor intrusion and may offer flexibility as a generally conservative screening tool (using the 95^{th} percentiles of regression parameters) that can be adapted to local conditions. The regression approach may help site-level initial screening estimates and identification of priority homes or neighborhoods for further investigation.

CHAPTER 4

Updating Exposure Models of Indoor Air Pollution Due to Vapor Intrusion: Bayesian Calibration of the Johnson-Ettinger Model[3]

4.1. Introduction

Chlorinated volatile organic chemicals (CVOCs) are capable of migrating from a subsurface plume upwards and cause vapor-phase contaminant intrusion in the overlying indoor air, an exposure pathway known as vapor intrusion. The challenges of collecting a robust data set inside private homes have complicated the characterization of vapor intrusion risks across potentially affected communities (Folkes, Wertz, Kurtz, & Kuehster, 2009; McDonald & Wertz, 2007; McHugh, Nickles, & Brock, 2007; Schreuder, 2006). Empirical evidence shows substantial spatial and temporal variability in contaminant exposures due to vapor intrusion, but the factors driving this variability are incompletely understood (Fitzpatrick & Fitzgerald, 2002; Folkes et al., 2009; P. Johnson et al., 2009). The scarcity of community-scale data, in turn, creates challenges to prioritizing homes for monitoring and assessing the need for remediation. The difficulty in assigning exposure at the individual or neighborhood level further restricts investigations of the potential association between vapor intrusion exposure and adverse health outcomes (Forand, Lewis-Michl, & Gomez, 2011).

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[3] *Johnston, J.E., MacDonald Gibson, J., & Sun, Q. 2013. In preparation for* Environmental Science and Technology

Because of political, technical, and monetary constraints on directly monitoring indoor air quality in private homes, mathematical screening tools typically are used to identify at-risk areas. While few studies have compared model predictions to measured indoor air pollutant concentrations, the existing literature reveals that the current models are, in general, inadequate to describe the observations (Yao, Shen, Pennell, & Suuberg, 2013). Previous research has identified the need for vapor intrusion models that more accurately reflect real-world conditions (Picone, Valstar, van Gaans, Grotenhuis, & Rijnaarts, 2012). The model most commonly employed during vapor intrusion site characterizations and recommended by the U.S. Environmental Protection Agency (EPA) draft vapor intrusion guidance is the Johnson-Ettinger model (JEM) (Eklund, Beckley, Yates, & McHugh, 2012; U.S. EPA, 2002). The JEM couples one-dimensional steady-state diffusion of volatile compounds through porous media with diffusion and advection through the building foundation (Johnson & Ettinger, 1991). In regulatory applications, deterministic values for variables that describe contaminant, environmental, and household properties serve as inputs to the JEM. The output is the vapor attenuation ratio, α, a unitless parameter that relates the indoor air concentration to the concentration in the subsurface water or soil (Johnson & Ettinger, 1991). The concentration of the contaminant indoors due to vapor intrusion is then estimated by:

$$C_{indoor} = \alpha * C_{source} \tag{1}$$

where C_{indoor} is the contaminant concentration in indoor air (mass/volume) and C_{source} is the contaminant vapor-source concentration (mass/volume), generally expressed as groundwater concentration multiplied by the appropriate Henry's constant.

In this research, we demonstrate a Bayesian approach for using empirically measured indoor air concentrations from a case study community to calibrate the JEM, in order to improve the accuracy and precision of its predictions of indoor air quality in the community. Systematic calibration procedures using site-level data have the potential to improve the accuracy of the JEM and hence to facilitate decision-making at vapor intrusion sites (Ellison, 1996; Larssen et al., 2006). The Bayesian method yields posterior calibrated distributions for the model input parameters of interest. The calibration approach used in this research has not been employed previously to update the JEM, although it has been applied successfully to a variety of ecological process-based and chemical fate-and-transport models (Arhonditsis et al., 2008; Larssen et al., 2006; Lehuger et al., 2009; Reinds, van Oijen, Heuvelink, & Kros, 2008; Saloranta et al., 2007; Van Oijen et al., 2011; Yeluripati et al., 2009). In these previous studies, the calibration procedure has narrowed the uncertainty of input parameters and improved overall model forecasting.

The Johnson-Ettinger algorithm is intended to estimate the influence of groundwater contamination on the overlying indoor air and to identify areas of concern for further evaluation (Environmental Quality Management, 2004). While the model contains numerous parameters, its application involves substantial uncertainty contributed by both model structure and parameter inputs (Fitzgerald, 2009). Since many sites contain hundreds or thousands of potentially impacted structures, the 2002 EPA guidance recommends the use of a combination of default and readily available site-specific-values as inputs to the JEM in order to evaluate whether the modeled indoor air concentrations exceed an established risk level. The EPA guidance provides default deterministic values for many of the inputs and suggests that these default values are "conservative," in other words, tend to over-estimate exposure risk.

Previous studies comparing modeled and measured α values have indicated that, with reasonable input parameters, the JEM can predict within one order of magnitude the actual indoor air concentrations of chlorinated volatile organic compounds (Hers & Zapf-Gilje, 2003). However, although purported to be conservative, in some past studies the JEM has underpredicted α and thus the indoor air concentration of the relevant pollutant (Fitzpatrick & Fitzgerald, 2002; Hers, Zapf-Gilje, Evans, & Li, 2002; Mills, Liu, Rigby, & Brenners, 2007; Provoost et al., 2010; Schreuder, 2006). More complex, three-dimensional models have been proposed and may be more accurate for an individual home, but these approaches require detailed local information, are not scaled to a community level, and have yet to be applied in a regulatory context (Bozkurt, Pennell, & Suuberg, 2009; Pennell, Bozkurt, & Suuberg, 2009; Yao & Suuberg, 2013).

The case study site employed to assess the potential for Bayesian calibration to improve the performance of the JEM is a neighborhood overlying extensive plumes of tetrachloroethylene (PCE) in groundwater emanating from the former Kelly Air Force Base in San Antonio, Texas. These plumes extend almost 8 km (5 miles) to the east and southeast of the base, affecting some 30,000 homes. The groundwater table is 1-10 m below the houses. Prior measurements of local monitoring wells have found PCE concentrations in the shallow groundwater ranging from 5 μg/L to almost 50,000 μg/L. In previous work we developed a method for incorporating parameter uncertainty into the JEM algorithm via Monte Carlo simulation, and we simulated indoor air concentrations of PCE in each of the case study community's 30,000 homes (Johnston & MacDonald

Gibson, 2011). While this previous Monte Carlo analysis offered a new approach to assessing the range of possible indoor pollutant concentrations in affected homes, the predictions still underestimated measured concentrations.

The goal of the research presented here is to further improve the JEM algorithm by calibrating it to measured indoor air pollutant concentrations, hence increasing both the accuracy and precision of the model. We compare the indoor air pollutant concentrations predicted by the calibrated stochastic model to the concentrations predicted by the deterministic JEM version currently used for policy and regulatory decisions. We compare both sets of predictions to measured indoor air pollutant concentrations, in order to evaluate whether the calibration technique can improve model vapor intrusion predictions. To our knowledge, such calibration of the JEM has not previously been attempted.

4.2. Methods

The mechanistic Johnson-Ettinger algorithm was combined with stochastic representations of input parameters and observed indoor air concentration data to update unknown model input parameters through Bayesian calibration using a Markov Chain Monte Carlo (MCMC) technique (Gilks & Roberts, 1996). The method involves specifying prior probability distributions for the input parameters and the likelihood function for the measured data (Figure 4.1). The approach iteratively computes posterior distributions for each uncertain input by comparing the predicted indoor air concentration at each new iteration with the measured concentration. The algorithm produces a sequence of updated input parameter values that comprise the posterior probability distribution functions for the inputs. This approach allows for the formal management of uncertainties in both the input parameters and the measured values and permits the explicit calculation of uncertainty in model results (Larssen et al., 2006).

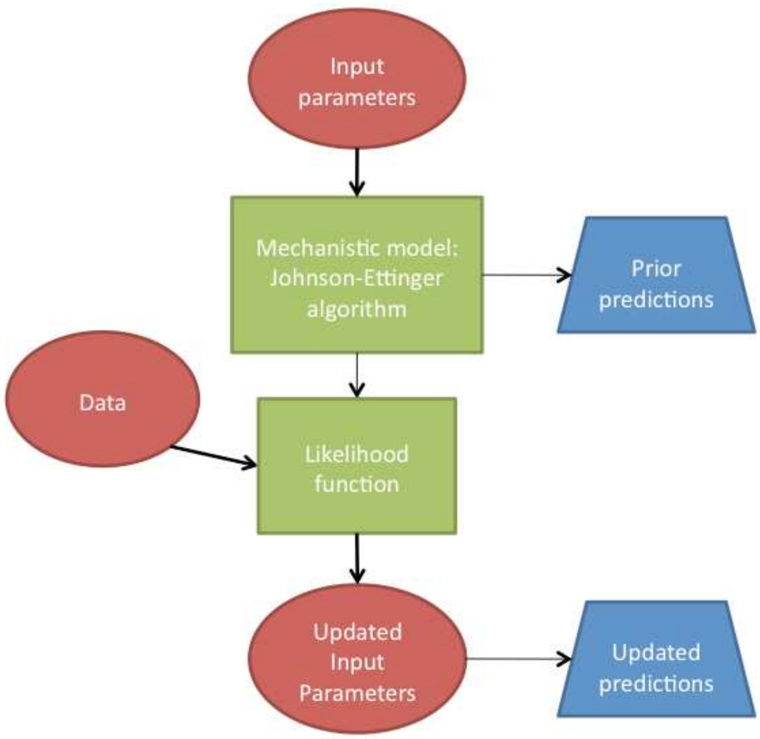

Figure 4.1. Sketch of model framework for the Bayesian calibration methodology.

<u>4.2.1. Johnson-Ettinger Model</u>

The JEM uses the following relationship to characterize the diffusion of pollutant vapors through the subsurface soil and across a building foundation and the subsequent accumulation of the pollutant inside the building (Johnson & Ettinger, 1991):

$$\alpha = \frac{\left(\frac{D_{total}^{eff} A_b}{Q_{building} L_t}\right) \exp\left(\frac{Q_{soil} L_{crack}}{D_{crack}^{eff} \eta A_b}\right)}{\exp\left(\frac{Q_{soil} L_{crack}}{D_{crack}^{eff} \eta A_b}\right) + \left(\frac{D_{total}^{eff} A_b}{Q_{building} L_t}\right) + \left(\frac{D_{total}^{eff} A_b}{Q_{soil} L_t}\right)\left[\exp\left(\frac{Q_{soil} L_{crack}}{D_{crack}^{eff} \eta A_b}\right) - 1\right]} \qquad (2)$$

where D_{total}^{eff} is total overall effective diffusion coefficient (cm^2/s), A_b is the area of enclosed space below grade (cm^2), $Q_{building}$ is the building ventilation rate (cm^3/s), L_t is the source-building separation (cm), Q_{soil} is the volumetric flow rate of soil gas into the enclosed space (cm^3/s), L_{crack} is the enclosed space foundation or slab thickness (cm), η is the fraction of foundation surface area with cracks (unitless), and D_{crack}^{eff} is the effective diffusion coefficient through the cracks (cm^2/s).

A majority of the eight inputs included in the JEM are not easily measured or characterized (Yao, Pennell, & Suuberg, 2012). Thus, in practice, parameter values are estimated from a series of secondary and tertiary equations that approximate the JEM inputs (see Table 4.1). As a result, the number of primary parameters considered in the model expands to 25. In practice, the EPA and/or state agencies suggest the use of deterministic values for each primary input parameter. However, in reality these parameters are both variable and uncertain. Hence, our stochastic modeling approach represents nine of the key input parameters (five of which are specific to the soil type) with lognormal probability distributions. The prior parameters for each distribution are informed by readily available site-specific data or, where such data are not available, extracted by national surveys or experimental studies available in the literature (Johnston & MacDonald Gibson, 2011). For this analysis, parameters describing soil properties were considered separately for each U.S. Department of Agriculture (USDA) classified soil type present at the sites of indoor air measurements (clay or silty clay). The distributions reflect the uncertainty in the environmental drivers as well as the variability in the parameter values. The remaining 14 inputs, the majority of which describe physical properties of PCE or measurable properties of a house (e.g., square footage), were represented as deterministic values. Collectively, the set of equations and parameter distributions constitutes the prior knowledge in the Bayesian framework to estimate the prior model-predicted indoor air concentration for each house.

Table 4.1. Complete set of equation needed to implement the Johnson-Ettinger model. The input parameters calibrated using Bayesian updating are marked with an asterisk (*).

Model equations	Variables:	
House/foundation calculations	V_b	= Building volume (cm^3)
$V_b = A \times MH$	A	= Building area (cm^2)
	MH	= **Building mixing height (cm)***
$Q_{building} = \frac{V_b E_b}{3600}$	$Q_{building}$	= Building ventilation rate (cm^3/s)
	E_b	= **Indoor air exchange rate (hr^{-1})***
$A_b = A + 2\sqrt{\frac{1}{2} \times A} \times L_{crack} + 4 \times \sqrt{\frac{1}{2} \times A} \times L_{crack}$	A_b	= Area of enclosed space below grade (cm^2)
	L_{crack}	= Foundation thickness (cm)
$R_{crack} = \eta \frac{A_b}{X_{crack}}$	R_{crack}	= Effective crack width (cm)
	η	= **Fraction of surface area with cracks***
	X_{crack}	= Floor-wall seam perimeter (cm)
Soil property calculations	S_{te}	= Effective total fluid saturation
$S_{te} = \frac{\theta_m - \theta_r}{\theta_T - \theta_r}$	θ_m	= **Volumetric moisture content (cm^3/cm^3)***
	θ_r	= **Residual soil water content (cm^3/cm^3)***
$\theta_v = \theta_T - \theta_m$	θ_T	= **Total porosity (m^3-voids/m^3-soil)***
	M	= **Van Genuchten curve shape parameter***
$K_{rg} = (1 - S_{te})^{0.5}(1 - (S_{te})^{1/M})^{2M}$	K_{rg}	= Relative air permeability
$K_i = \frac{K_s \mu_w}{\rho_w g}$	K_i	= Soil intrinsic permeability (cm^2)
	K_s	= **Soil saturated hydraulic conductivity (cm/s)***
$k = K_{rg} \times K_i$	μ_w	= Dynamic viscosity of water (g/cm-s)
	ρ_w	= Density of water (g/cm^3)
$Q_{soil} = \frac{2\pi k(\Delta P) X_{crack}}{\mu \ln(\frac{2 Z_{crack}}{R_{crack}})}$	g	= Acceleration due to gravity (cm/s^2)
	k	= Soil permeability near foundation (cm^2/s)
	ΔP	= **Indoor-outdoor pressure difference (g/cm-s^2)***
	μ	= Viscosity of air (g/cm-s)
	Z_{crack}	= Crack opening depth below grade (cm)
Diffusion calculations	D_i^{eff}	= Effective diffusion for soil layer, i (cm^2/s)
$D_i^{eff} = D_{air}(\frac{\theta_v^{3.33}}{\theta_T^2}) + \frac{D_{H_2O}}{H_i}(\frac{\theta_m^{3.33}}{\theta_T^2})$	D_{air}	= PCE diffusion coefficient in air (cm^2/s)
	D_{H_2O}	= PCE diffusion coefficient in water (cm^2/s)
$D_{total}^{eff} = \frac{L_t}{\sum_{i=1}^{n} \frac{L_i}{D_i^{eff}}}$	H_i	= PCE Henry's constant
	D_{total}^{eff}	= Total overall effective diffusion coefficient (cm^2/s)+
	L_t	= Distance between the source and the bottom of the enclosed space floor (cm)
	L_i	= Thickness of soil layer *i* (cm)

The groundwater depth (cm) and groundwater contaminant concentrations (μg/L) were interpolated for 2011 based on semiannual data for the 900 shallow groundwater monitoring wells located in and around the former Kelly Air Force Base. Monitoring well data are available through the Department of Defense Air Force Real Property Agency Semi-Annual Compliance Plans from 1998-2011. The 31-km^2 area was divided into 150-m^2 grid cells. A state-of-the-art spatio-temporal geostatistical interpolation method, Bayesian maximum entropy, was then used to describe the estimated mean concentration and variance for each grid cell using a technique described elsewhere (Christakos, Bogaert, & Serre, 2001; Serre, Carter, & Money, 2004). Every house in the grid cell was assigned the corresponding values (and uncertainty) for the groundwater depth (cm below surface) and PCE concentration (μg/L). Since these values are house-specific and based on a dense network of monitoring wells, these two parameters were not calibrated during the Bayesian updating. The resulting maps of PCE in the groundwater and the groundwater levels are shown in Figure C.1 and Figure C.2, respectively (Appendix C).

4.2.2. Indoor Air Concentration Measurements

From July to August 2011, we collected indoor air samples from 20 homes in the community (eight samples per home). The concentration of PCE indoors (μg/m^3) was measured with passive samplers taking sequential 72-hour integrated measurements. Potential confounding sources were removed from each residence before deploying the passive samplers. Further details about the sampling methods are available elsewhere (Johnston & MacDonald Gibson, 2013). The measured concentrations comprise the data values, y_{ij}, used to update the JEM parameters. The mean and variance of the measured concentration in each house were calculated, accounting for the detection limit (0.13 ug/m^3) and using the Kaplan Meier nonparametric technique (Antweiler & Taylor, 2008; Helsel, 2005). Table 4.2 presents indoor air concentration summary statistics and other relevant attributes for each sampled home.

Table 4.2. Basic properties of houses and measured concentration of PCE indoors. The mean and standard deviation is shown, assuming a lognormal distribution.

House	Soil Type+	Area (m^2)	PCE groundwater (μg/L)	PCE indoor air (μg/m^3)
1	SiC	119	24.9 (1.5)	0.18 (0.12)
2	C	116	7.6 (1.6)	0.15 (0.09)
3	C	97	2.3 (1.7)	0.25 (0.34)
4	SiC	118	11.2 (1.5)	0.08 (0.03)
5	SiC	109	49.2 (1.4)	0.18 (0.05)
6	C	74	2.8 (1.8)	0.08 (0.03)
7	C	100	5.9 (1.9)	0.08 (0.03)
8	SiC	122	49.2 (1.4)	0.31 (0.16)
9	SiC	74	2.1 (1.6)	0.08 (0.03)
10	SiC	272	1.9 (1.8)	0.20 (0.13)
11	SiC	136	49.2 (1.5)	0.20 (0.10)
12	SiC	109	49.2 (1.4)	0.28 (0.19)
13	C	145	10.5 (1.9)	0.11 (0.04)
14	SiC	93	104.3 (1.1)	0.08 (0.03)
15	SiC	207	4.2 (1.4)	0.15 (0.01)
16	C	82	10.5 (1.9)	0.14 (0.09)
17	SiC	74	24.9 (1.5)	0.08 (0.03)
18	C	109	10.0 (1.3)	0.08 (0.03)
19	C	178	5.3 (1.6)	0.22 (0.40)
20	C	40	3.8 (1.8)	0.09 (0.03)

+ SiC: silty clay; C: clay

4.2.3. Bayesian Approach for Updating JEM Parameters

Let y_{ij} denote the indoor air concentration of PCE for the jth measurement in the ith house, and let $f(\boldsymbol{\theta},\boldsymbol{\omega}, \mathbf{x}_i)$ denote the mean value of the indoor air concentration predicted in the ith house by the JEM using the uncertain parameter vector $\boldsymbol{\theta}$ (the nine parameters with asterisks in Table 4.1), the deterministic parameter vector $\boldsymbol{\omega}$, and the house-specific parameter vector $\mathbf{x}_i$ (house area, groundwater concentration and groundwater level). We can then use a stochastic model to investigate the relationship between the measured PCE concentration, $y_{ij,}$ and mean value of the concentration, $f(\boldsymbol{\theta},\boldsymbol{\omega}, \mathbf{x}_i)$, predicted by the JEM, as follows:

$$y_{ij} = f(\boldsymbol{\theta},\boldsymbol{\omega}, \mathbf{x}_i) + \varepsilon_{ij}\ , \quad i = 1, 2,\ldots,n;\ j=1,2,\ldots,j_i \tag{3}$$

where n is the number of houses, j_i is the total number of measurement within the ith house, and ε_{ij} is the difference between the predicted mean and the observed concentration (the error term). In accordance with previous work, the errors are assumed to be independent and identically distributed as normal random variables with mean of zero and variance of σ_i^2, i.e. ε_{ij}~N(0, σ_i^2) (Klemedtsson et al., 2008; Svensson et al., 2008; Van Oijen, Rougier, & Smith, 2005). The calibration process assumes there is an optimal set of input parameters that minimizes the error. These assumptions lead to the following likelihood function for the observed data:

$$p(\mathbf{y}\,|\,\boldsymbol{\theta})] = \prod_{i=1}^{n}\prod_{j=1}^{j_i}\frac{1}{\sqrt{2\pi\boldsymbol{\sigma}_i^2}}\exp\left(-\frac{\left(y_{ij}-f(\boldsymbol{\theta},\boldsymbol{\omega},x_i)\right)^2}{2\boldsymbol{\sigma}_i^2}\right) \tag{4}$$

The likelihood function evaluates how well the simulation model is able to reproduce the observed data y_{ij} at each value of $\boldsymbol{\theta}$. In the Bayesian paradigm, any prior information about the input parameters, $\boldsymbol{\theta}$, can be improved by incorporating the local data information (measured indoor PCE concentration) through the likelihood function, $p(\mathbf{y}|\boldsymbol{\theta})$, which further leads to the posterior distribution $p(\boldsymbol{\theta}|\mathbf{y})$, according to Bayes' Theorem:

$$p(\boldsymbol{\theta}\,|\,\mathbf{y}) = \frac{p(\mathbf{y}\,|\,\boldsymbol{\theta})\,p(\boldsymbol{\theta})}{p(\mathbf{y})} \tag{5}$$

4.2.4. Posterior Distribution

The posterior distribution of $\boldsymbol{\theta}$ cannot be specified analytically because of the complex relationships among the input parameters, the model output, and the data.

However, samples from the posterior distribution can be generated using the MCMC method using the Metropolis-Hastings algorithm (Kennedy & O'Hagan, 2001; Larssen et al., 2006; Lehuger et al., 2009; Metropolis, Rosenbluth, Rosenbluth, Teller, & Teller, 1953). The Metropolis-Hastings algorithm works by generating a sequence of sample values to estimate a posterior distribution for each parameter. These sample values are generated iteratively, with the distribution of the next sample being dependent only on the current sample value, denoted as $\boldsymbol{\theta}_t$. The candidate ($\boldsymbol{\theta}_{candidate}$) is either accepted, in which case the candidate value is used in the next iteration, or rejected, in which case the candidate value is discarded and the current value is reused for the next iteration. The probability of acceptance is determined by dividing the likelihood of the candidate parameter vector of model inputs ($\boldsymbol{\theta}_{candidate}$) by the likelihood of the current input vector ($\boldsymbol{\theta}_t$); the result is called the Metropolis ratio, r. If $r \geq 1$, the candidate vector is always accepted; if $r < 1$, the candidate vector is accepted with probability r.

In this paper, we implement Metropolis-Hastings algorithm using the following process (Gilks & Roberts, 1996):

1. Choose a starting value, $\boldsymbol{\theta}_{initial}$.
2. At iteration t, draw a proposed candidate, $\boldsymbol{\theta}_{candidate}$, based on the current state of the sample values, $\boldsymbol{\theta}_{t.}$
3. Compute the Metropolis ratio r:

$$r = \frac{p(\boldsymbol{\theta}_{candidate} \mid \mathbf{y}) q(\boldsymbol{\theta}_t \mid \boldsymbol{\theta}_{candidate})}{p(\boldsymbol{\theta}_t \mid \mathbf{y}) q(\boldsymbol{\theta}_{candidate} \mid \boldsymbol{\theta}_t)} \tag{6}$$

   where $q(\boldsymbol{\alpha}|\boldsymbol{\beta})$, is the proposed density of $\boldsymbol{\alpha}$ given $\boldsymbol{\beta}$.

4. Accept the candidate as the new value $\boldsymbol{\theta}_{t+1}$ with probability equal to min{r,1}.

If the candidate is accepted, then $\theta_{candidate} = \theta_{t+1}$; otherwise, $\theta_t = \theta_{t+1}$.

5. Draw a new candidate, and repeat steps 2-4 until the chains reaches a stationary distribution of p($\boldsymbol{\theta}$|**y**).

For each calibration, three parallel Markov chains were run from three different initial values for the parameter vector: the prior mean value and two randomly sampled points from the parameter distribution. We ran 20,000 iterations, disregarding the first 10,000 as the unrepresentative "burn-in" of the chain and adopting the final 10,000 iterations to characterize the posterior distribution of the parameter vector (Gilks & Roberts, 1996; Van Oijen et al., 2005). Chain convergence was assessed with Geweke's (1992) convergence for each single chain and with the Gelman-Rubin convergence statistic for multiple chains (Cowles & Carlin, 1996; Gelman, Carlin, Stern, & Rubin, 2003). All test statistics indicated that the resulting chains reached stationary distributions and therefore can be considered as a representative sample from the posterior probability distribution function. From this sample we calculated the mean, variance, and 90% confidence interval of the posterior estimate for each parameter based on the three chains. The generation and analysis of the Markov chains were carried out with the statistical package R, and diagnostics were conducted using the coda package for R (Plummer, Best, Cowles, & Vines, 2006).

4.2.5 Soil Parameters Inputs Based on Site Measurements

Previous research suggests that the Johnson-Ettinger model is sensitive to soil property inputs and that these inputs account for much of the uncertainty in the model output (Johnston & MacDonald Gibson, 2011; Tillman & Weaver, 2007). As a result, we collected site-specific soil samples in order to provide site-level prior estimates of volumetric moisture content (θ_m), hydraulic conductivity (K_s), and total porosity (θ_T) for each of the two soil types in the study area. Soil cores were collected at four of the 20 study homes, resulting in two cores for each soil type. These cores were evaluated at approximately 1.5 m and 5 m below the surface. Due to the limited sample size, we identified the minimum and maximum values and assigned a lognormal distribution for each parameter such that the minimum and maximum measured values equaled the lower and upper 95th percentile values of the calculated lognormal distribution. Steinberg, Reckhow, and Wolpert (1997) and Arhonditsis, Qian, Stow, Lamon, & Reckhow (2007) previously used a similar assumption with PCB fate and transport parameters and eutrophication model parameters, respectively. The extent to which the small number of soil cores collected represents soil properties across this study community is unknown. A second set of MCMC simulations was run to estimate a posterior probability distribution to evaluate whether these limited soil parameter measurements improve model calibration and posterior predictions, compared to using soil property estimates from available USDA databases.

4.2.6. Evaluation of Model Performance

The performance of the calibrated model was evaluated by: (a) comparing the 90% confidence intervals of the in-home PCE concentration measurements and the model predictions, (b) determining the pairwise correlations between the mean and 95th percentile values of the predictions and measurements, and (c) calculating the root mean squared error (RMSE), estimated as:

$$rmse = \sqrt{\frac{\sum_{i=1}^{n}\sum_{j=1}^{j_i}(y_{ij} - f(\boldsymbol{\theta}, \boldsymbol{\omega}, \mathbf{x_i})}{n}} \qquad (7)$$

4.3. Results and Discussion

4.3.1. JEM Deterministic Predictions

The indoor air concentrations of PCE in the 20 homes of interest were initially calculated using the standard adaptation of the JEM as recommended by the EPA vapor intrusion guidance document, incorporating the mean estimates for groundwater level and groundwater PCE concentrations, house-specific soil type and house area, and EPA default values for the other parameters (Environmental Quality Management, 2004). The JEM deterministic predictions for PCE ranged from 0.005 to 0.15 μg/m^3 for the 20 homes where measurements were collected. As shown in Figure 4.2, the deterministic method underpredicted the mean measured concentrations in all cases, and the predicted value was less than the 5th percentile of the observed concentrations in 11 out of the 20 study homes. Applied to the regulatory context, the use of the standard JEM approach would not accurately reflect the exposure levels occurring in the houses and would underestimate exposure and resulting risk.

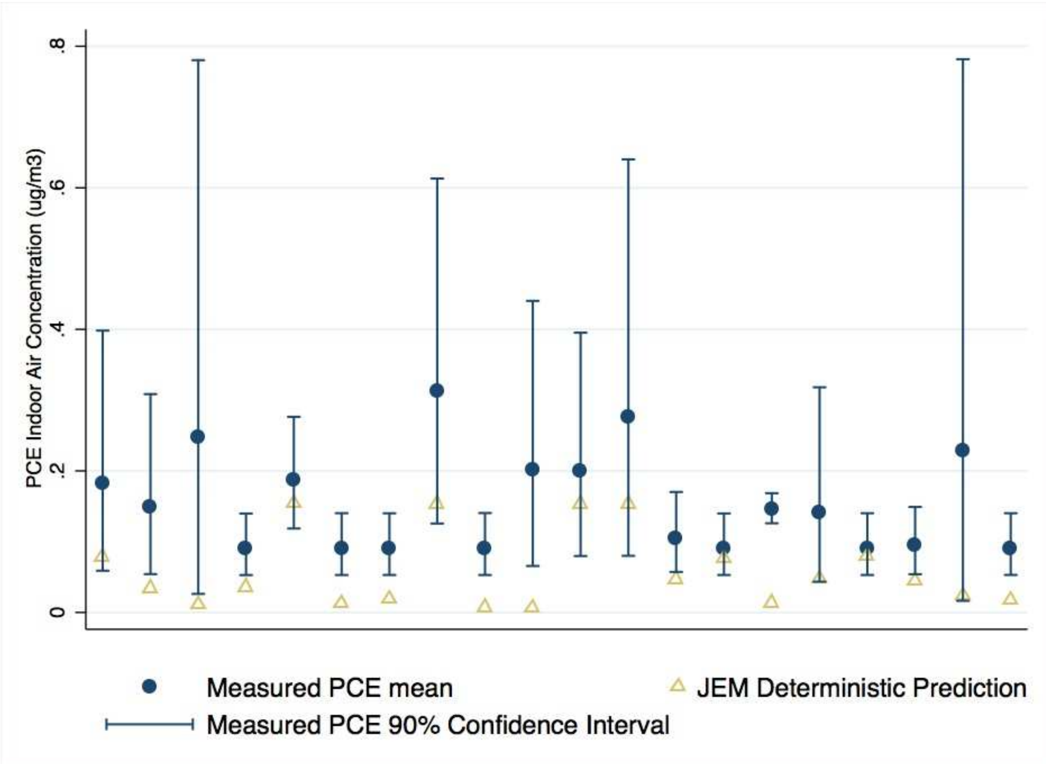

Figure 4.2. Comparison of the measured indoor air PCE concentrations due to vapor intrusion (Johnston & MacDonald Gibson, 2013) and EPA-recommended deterministic modeling based on the Johnson-Ettinger algorithm.

4.3.2. Simulation of Indoor Air Predictions

Monte Carlo techniques using the final 10,000 iterations produced prior probability estimates of PCE indoor air concentrations for each house based on the JEM. On average, the mean modeled predictions of indoor air concentrations for the prior probability distributions were within approximately one order of magnitude of the observed measurements, as generally expected for the JEM (see Appendix C, Figure C.3). In 19 out of the 20 cases, the modeled mean value is below the observed mean. However, it is also believed that the uncertainty of the prior distributions is generally overstated since the distributions do not incorporate all information available, that is, the data on the indoor air measurements (Van Oijen et al., 2005).

We calibrated the JEM against measured indoor air data using two different sets of prior distributions for soil parameters: (1) distributions based on publicly available USDA soil information for the study area (Model 1), and (2) distributions estimated from our local analysis of the four soil cores (Model 2). Figure C.4 (Appendix C) shows the MCMC trace plots for the nine uncertain model parameters. The irregular patterns of these sequences of iterations are characteristic of the Metropolis-Hastings MCMC algorithm, because the method aims to explore the range of potential values rather than only identify a single global optimum value (Gelman et al., 2003).

The means and standard deviations of the prior and posterior distributions of the nine updated model parameters are shown in Table 4.3, and corresponding box plots of the parameter distributions are compared in Figure 4.3. For the four house-related parameters, in both models the standard deviations of the posterior distributions were reduced as a result of the calibration. These results suggest that the Bayesian calibration technique was able to reduce the uncertainty in the model input variables and narrow the prediction interval. For example, the standard deviation of the air exchange rate (E_b) decreased to almost half of its prior value, and the mean also decreased (by about 20%) in both Model 1 and Model 2. We found a higher average differential pressure (ΔP) than initially chosen along with a decrease in the standard deviation of this parameter. Both of these changes suggest that our initial prior distributional assumptions were not sufficiently conservative, as a higher pressure differential and a lower air exchange rate increase the vapor attenuation factor and thus lead to higher indoor air concentrations.

Table 4.3. Prior and MCMC posterior estimates of the mean values and standard deviations of the model parameters**.**

	Input			EPA default	Prior distribution*	Model 1	Model 2	References for prior distribution assumptions
Household	MH	cm		300	270 (20)	268 (18.3)	269 (16.5)	(Johnson, 2005)
	E_b	hr^{-1}		0.25	0.66 (0.73)	0.55 (0.39)	0.55 (0.39)	(Meng et al., 2004; Yamamoto et al., 2009)
	η			0.004	0.004 (0.0015)	0.0029 (0.0011)	0.0029 (0.0011)	(Eaton & Scott, 1984; Johnson, 2005; Nazaroff, 1992)
	ΔP	$g/cm\text{-}s^2$		40	60 (35)	65.6 (28.5)	66.19 (24.96)	(Fischer & Uchrin, 1996; Nazaroff et al., 1987; Robinson, Sextro, & Riley, 1997)
Soil	θ_T	m^3- voids / m^3-soil	C	0.459	0.46 (0.09) M2: 0.35 (0.07)	0.46 (0.09)	0.36 (0.06)	(Carsel & Parrish, 1988; Environmental Quality Management, 2004)
			SiC	0.481	0.481 (0.07) M2: 0.35 (0.07)	0.48 (0.07)	0.36 (0.02)	
	θ_m	cm^3/cm^3	C	0.215	0.21 (0.08) M2: 0.12 (0.075)	0.21 (0.06)	0.13 (0.06)	(Environmental Quality Management, 2004)
			SiC	0.216	0.16 (0.04) M2: 0.14 (0.055)	0.16 (0.03)	0.16 (0.03)	
	θ_r	cm^3/cm^3	C	0.07	0.07 (0.03)	0.07 (0.03)	0.069 (0.03)	(Carsel & Parrish, 1988; Environmental Quality Management, 2004)
			SiC	0.09	0.10 (0.11)	0.09 (0.07)	0.16 (0.09)	
	N		C	1.25	1.25 (0.09)	1.26 (0.08)	1.25 (0.09)	(Carsel & Parrish, 1988; Environmental Quality Management, 2004)
			SiC	1.32	1.32 (0.05)	1.32 (0.05)	1.31(0.05)	
	K_s	cm/day	C	12.6	12.6 (3.55) M2: 0.058 (0.17)	16.6 (3.48)	0.16 (0.11)	(Carsel & Parrish, 1988; Environmental Quality Management, 2004)
			SiC	9.95	9.95 (3.93) M2: 1.3 (0.86)	9.95 (3.94)	1.41 (0.76)	

* Each distribution is assumed to be lognormal and defined by its mean with the standard deviation in parentheses.
+ C: clay; SiC: silty clay

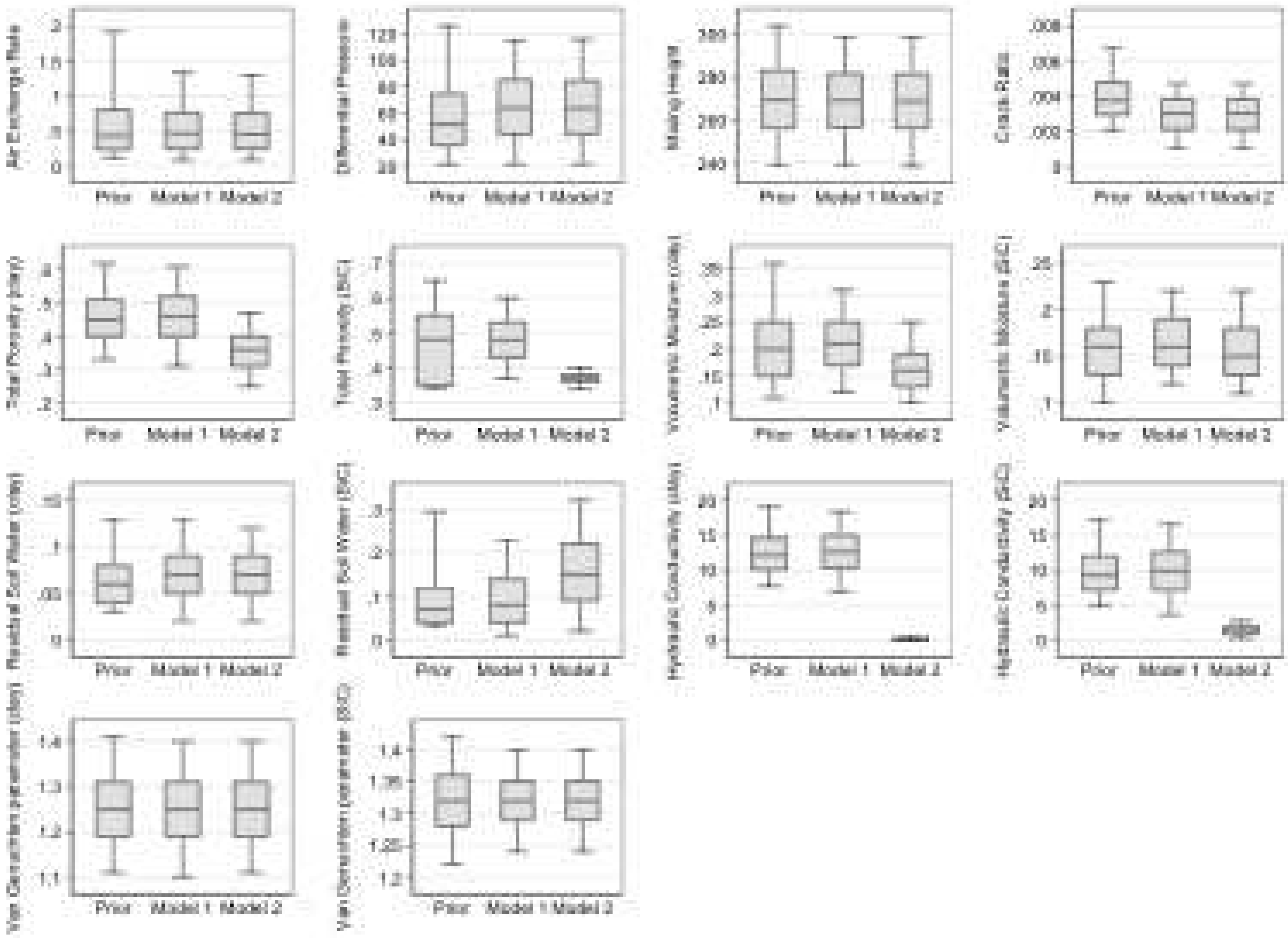

Figure 4.3. Box plots of the prior and posterior distributions (Model 1 and Model 2) for each of the four household parameters and five soil parameters (for clay and SiC: silty clay soil) calibrated during the Bayesian updating.

In the case of the soil parameters, the calibration did not result in noticeable reductions in the uncertainty or shifts to the mean. While these two moments (mean and standard deviation) of the distribution saw some change as a result of the calibration, there was a shrinking of the 90% confidence interval in Model 1 for four of the soil parameters (θ_T-SiC, θ_m-clay, θ_m-SiC and N-SiC), as shown in the Figure 4.3. For the second model (Model 2), we integrated site-level information on the total porosity, volumetric moisture content, and hydraulic conductivity of the soil into our prior knowledge. As a result, the calibration adjusted the soil parameters to some extent—although the posterior distribution is largely tied to the initial assumptions of the prior probability functions (from which the posterior candidate values were drawn). For Model 2, our prior distributions for these parameters were substantially different than the assumptions based on national USDA soil type data. Further, we assumed that the measurements taken were representative of the soil type across the neighborhood in spite of the small sample size and single sampling event. As a result of these assumptions, the average hydraulic conductivity converged on values significantly lower than the distributions estimated in the first model. However, this effect appears to be partially offset by the higher overall total porosity of the measured soil parameters.

4.3.3. Model Prediction Error

In general, the predictive ability of the Johnson-Ettinger model improved as a result of calibration with site-level data. The mean values of both Model 1 and Model 2 show better agreement with the measured mean values for PCE compared to both the deterministic and prior information model (Figure 4.4). In 18 of 20 the cases, the mean measured value is within the 90% confidence interval of the predicted concentration, and the confidence intervals are less than one order of magnitude in range. However, in cases of high indoor air concentrations (especially given homes above lower PCE groundwater concentrations), the calibrated model was still unable to adequately predict the upper ranges of the observed indoor air concentrations. In the two cases the model predictions were outside of the observed values, no detectable levels of PCE were measured in the home. In these cases, the precise performance of the model is difficult to assess. In general, Model 2 estimates are slightly higher (and have a somewhat larger uncertainty range) than Model 1, indicating a slightly more conservative (but less precise) estimate of the modeled exposure.

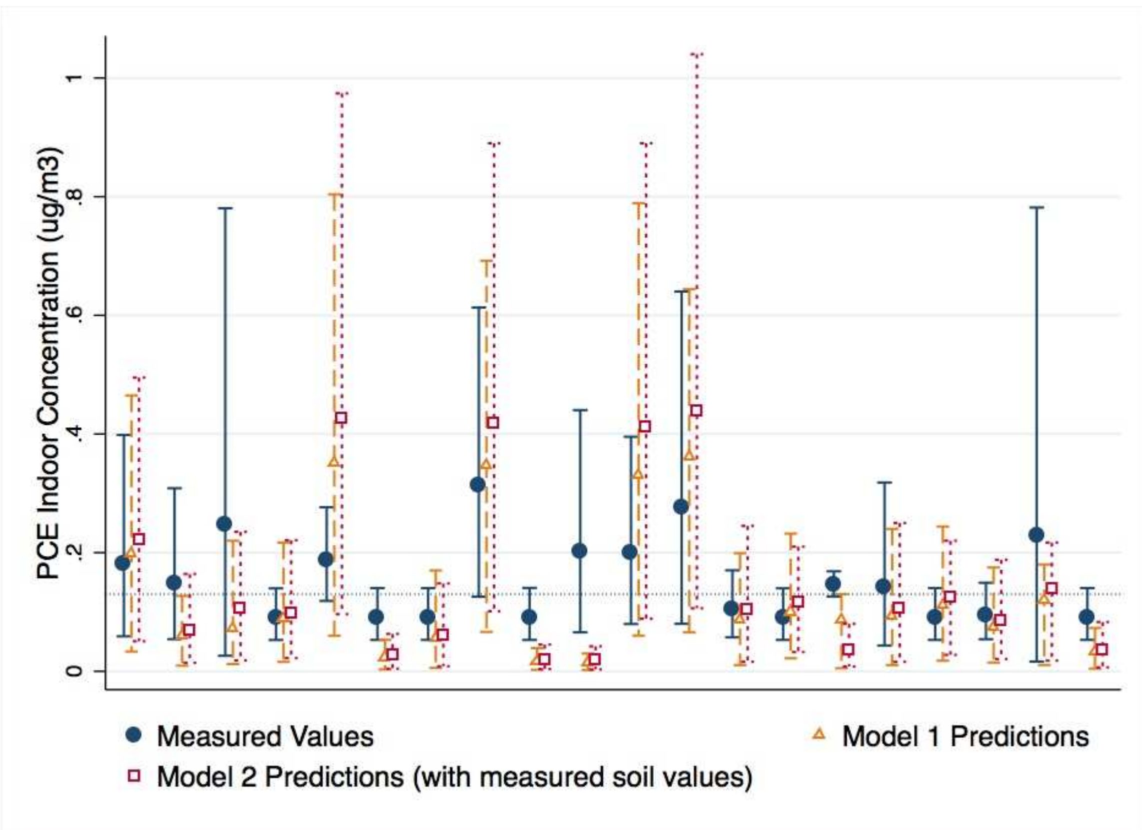

Figure 4.4. Measured predictions versus updated model predictions. The dotted gray horizontal line represents the PCE detection limit (0.13 μg/m^3).

As shown in Table 4.4, both of the calibrated models decreased the error between the measured and predicted values, as demonstrated by the decrease in the root mean squared error value (RSME). We compared the measured means with modeled means, as well as the measured 95th percentiles with the predicted 95th percentiles and found improvement in both cases. Compared to the deterministic approach, models 1 and 2 reduce the RMSE by almost 40% and 11%, respectively. In general, Model 1 indicates smaller residuals compared to Model 2, suggesting better agreement with observed values. The results of paired t-tests between measured and modeled values indicate that the mean differences between both of the calibrated models and the measured values is not significantly different than zero. In contrast, for both the prior model and the deterministic model estimates, the predicted means differed significantly from the measured means ($p<0.001$).

Table 4.4. Comparing model performance of the deterministic approach, the prior predictions and the two updated model predictions to the measured indoor air concentrations.

	Deterministic		**Prior**		**Model 1**		**Model 2**	
	Mean	95th	Mean	95th	Mean	95th	Mean	95th
Mean Difference (paired t-test)	0.097 (p<0.001)	--	0.082 (p<0.001)	0.087 (p=0.16)	0.024 (p=0.23)	0.035 (p=0.57)	0.0019 (p=0.94)	-0.14 (p=0.84)
RMSE	0.264	--	0.103	0.276	0.089	0.262	0.109	0.301

4.3.4. Sensitivity and Uncertainty Analysis

Previous work has found that, in order of significance, the predicted vapor intrusion attenuation ratio is most sensitive to the effective soil moisture content, air exchange rate, total porosity, and building mixing height (Johnson, 2005; Tillman & Weaver, 2006). A sensitivity analysis was conducted by setting each calibrated model input variable to the 5th and 95th percentile value of its posterior distribution, in order to determine the effects on the predicted indoor air concentration. Figure 4.5 shows the results for the household and silty clay soil parameters (the results for clay soil are very similar). As shown, increasing the air exchange rate or the mixing height decreases the predicted indoor air concentrations, while the opposite relationship is true of the other parameters. Among all of these input variables, the air exchange rate had the largest influence on the predicted indoor air concentrations. Air infiltration affects indoor air quality because insufficient air exchange with the outdoors can lead to higher exposure to pollutants of indoor or subsurface origin (Chan et al., 2005). Predicted concentrations also were sensitive to mixing height (used in the JEM to estimate the volume of the house) and the total soil porosity. Importantly, the calibration process decreased the uncertainty around each of these important input parameters, hence providing for narrower prediction intervals of indoor air concentration, compared to the prior model.

On the other hand, the uncertainty around some of the input parameters stayed the same, indicating that the calibration had little impact. This is particularly true for the first posterior model (Model 1), where only slight changes occurred in the probability distributions of the soil parameters. This may suggest either that data are insufficient to improve these parameters or that their optimal values were outside the prescribed range (Lehuger et al., 2009). The inability to narrow the uncertainty around the soil properties may also suggest that there is extensive heterogeneity among the soil properties influencing vapor diffusion even within a given soil type. This natural heterogeneity may make it impossible to reduce the standard deviation of these inputs below a certain range. Using the measured soil values of Model 2 increased the uncertainty of soil related parameters compared to assuming the values based on the USDA classification system (Figure 4.5c). Specifically, we observe greater uncertainty contributed by the hydraulic conductivity than seen in the other analysis, which further suggests that soil properties exhibit substantial, irreducible variability. The important influence of small-scale heterogeneity in soil on soil vapor transport has been noted in other studies (Carsel & Parrish, 1988; Garbesi, Robinson, Sextro, & Nazaroff, 1999). This observation may also be an artifact of the limited sample size used in the updating process.

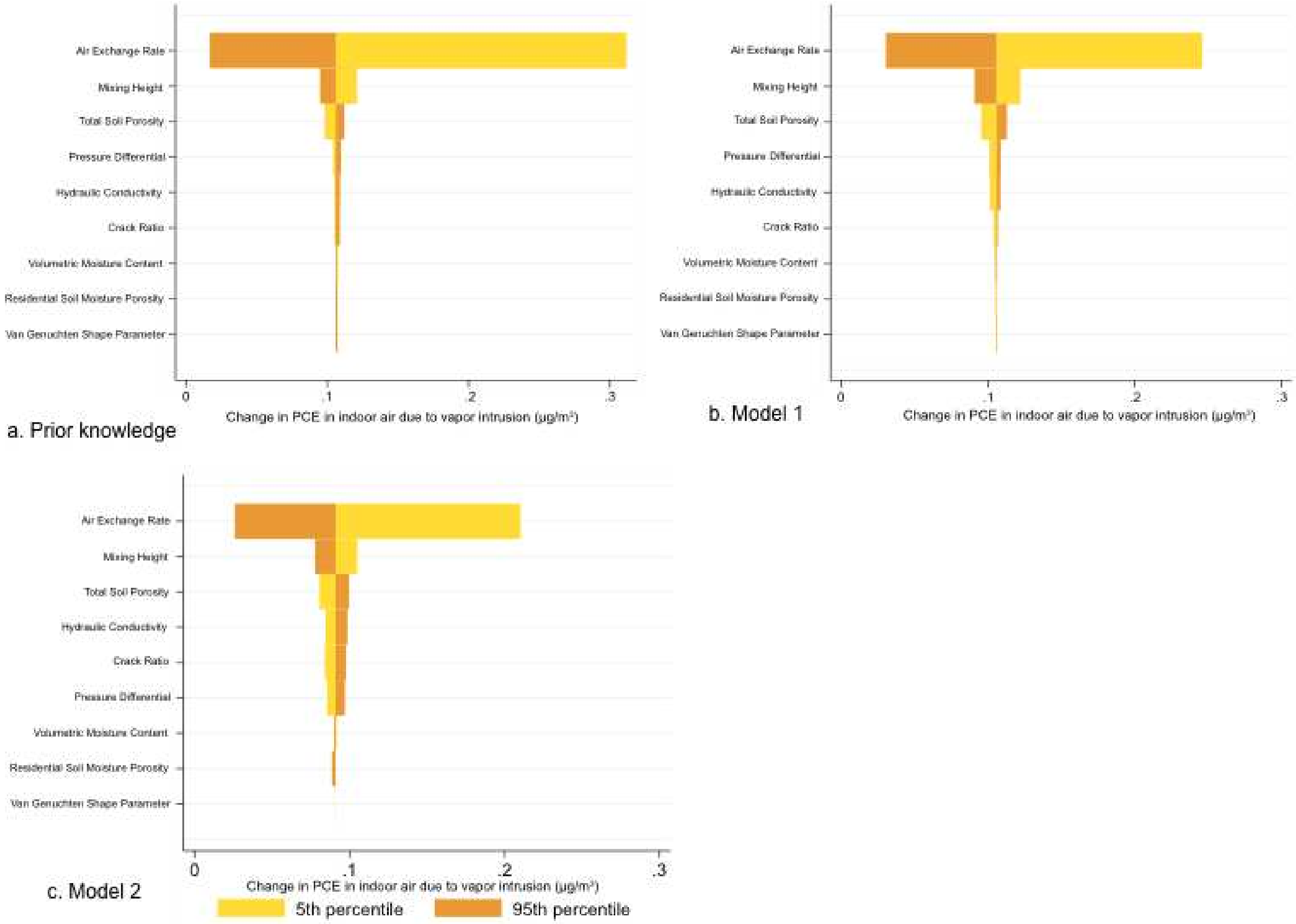

Figure 4.5. Sensitivity analysis of uncertain variables. The soil parameters displayed are for the silty clay (SiC) soil type.

4.3.5. Potential Suitability of Bayesian Calibration for Vapor Intrusion

Our primary goal in this study was to demonstrate the potential of a Bayesian calibration procedure to improve the parameterization of a vapor intrusion model, systematically quantify the uncertainty, and reduce the uncertainty in model output. Bayesian calibration with MCMC can handle a large number of parameters simultaneously, associate prior knowledge of parameter values with measurements of output variables, and reduce uncertainty when there is insufficient knowledge about the parameter distributions. In this study we chose to calibrate model parameters common across the community (rather than parameters, like groundwater depth, specific to each house). This approach searches for posterior distributions that simultaneously minimize error among all study homes at this site. In this study, indoor air measurements and model estimates do not exceed the revised EPA screening levels for PCE issued in April 2012, but the research nonetheless offers insights into a systematic approach to updating and improving the JEM exposure estimates based on limited data and evaluating the uncertainty of the predictions.

This technique could also be applied to multisite data, and in some ecological studies, to compare site-level and regional-level estimates (Lehuger et al., 2009). The calibration results can serve as the default values for spatial extrapolation in new houses where indoor air measurements have not been collected. However, Bayesian calibration cannot reduce the uncertainty of a parameter estimate or improve its accuracy without appropriate input data. While the calibrated models improve model predictions compared to a deterministic approach, there remains a large amount of uncertainty around each prediction. This is not entirely different from what is seen at vapor intrusion sites, where indoor air concentrations of the chemical of concern can fluctuate by more than an order of magnitude over days, weeks or months (Folkes et al., 2009; Johnson, Ettinger, Kurtz, Bryan, & Kester, 2009; McHugh et al., 2007). We did not see a substantial improvement in the predictions when including measured soil properties as the initial parameter assumptions, although this may not be the case if a large sample is available from which to establish the initial distributional properties. At least with this site, improving parameters of the soil properties proved challenging and possibly reflects that the spatial resolution scale of the soil classification scheme for assigning soil characteristics of houses may need to be increased to see improved efficiency of the parameter estimates.

4.4. Conclusions

We presented a statistical framework for model calibration and uncertainty estimation for complex deterministic vapor intrusion models. In this case, we combined a model with a mechanistic foundation with statistical methods; the former component can predict system behavior, while the latter allows for empirical parameter estimation and rigorous analyses of uncertainty. The inclusion of uncertainties in model predictions can strengthen the inferences drawn from model results in support of decision-making. The lack of quantifiable uncertainty often is used by scientists to justify their lack of involvement with the decision-making process and by some decision analysts as a vehicle to avoid using scientific information in the process (Morgan & Henrion, 1992; Underwood, 1995). However, uncertainty is fundamental to all scientific activities, and people regularly make decisions based on uncertain data (e.g., weather forecasts). The results need not be treated as final predictions, but rather can be viewed as new sources of information (new prior probability functions) for subsequent “experiments” that lead to modifications in management practices (new decisions) or improvement of exposure predictions.

For better model-based decision-making, the uncertainty in model projections should ideally be reduced, quantified, and reported in a way that can be used by decision makers. The Bayesian approach generates a posterior predictive distribution that accounts for both the uncertainty about the parameters and the uncertainty that remains when some parameters are known (Kennedy & O'Hagan, 2001), presenting a more realistic picture to support environmental management (Reichert & Omlin, 1997). In the case presented here, the currently regulatory framework for estimating exposure at sites potentially impacted by vapor intrusion could be improved through incorporation of uncertainty and variability of input parameters as well as the integration of site-level (even if limited) measurements to provide a more robust dataset from which to make decisions.

Chapter 5

Concluding Remarks

5.1. Overview of Policy Issues and Current Research Limitations

This dissertation has addressed several aspects of assessing vapor intrusion exposure risks. It demonstrated a method to quantify relationships between environmental, meteorological and household characteristics at the community scale (Chapter 2), to evaluate these relationships for sites across the country (Chapter 3), and to propose new methods for improving exposure predictions via the use of mechanistic models and site-level data (Chapter 4). At its core, the research presented in this dissertation explores the intersection of exposure assessment and quantitative environmental policy tools.

Exposure assessment is one of the key components of the regulatory decision-making approach regarding environmental and health risks and is frequently applied to hazardous waste sites. Accurate assessment of human exposures is a critical component of environmental health research and is paramount to the quality and utility of human health risk assessments (McKone, Ryan, & Özkaynak, 2008). As described in a National Research Council report (2009), exposure assessment first requires definition of the scope of the assessment in the context of the decision that needs to be made; that is, when designing an exposure assessment, the policy question should be posed first, and the assessment should then focus on gathering the information needed to answer the question (Abt, Rodricks, Levy, Zeise, & Burke, 2010). For example, screening-level information may be adequate to address some questions, targeted data may be useful for others, and extensive data may be needed in some circumstances, particularly for developing scientifically sound policies and regulations (Lioy & Smith, 2013). However, as noted in a recent report issued by the National Research Council (2012), policy and regulations have been slow to incorporate exposure science.

While it has been clear for some time that air pollution exposures are dominated by the indoor environment (the fraction of pollution inhaled from indoor sources is about 1,000 times greater than that from outdoor sources), policy innovations, monitoring programs, and other efforts to control indoor pollution have been limited (Bennett et al., 2002; Myers & Maynard, 2005; Wallace, 1991). This pattern extends to vapor intrusion. More than 20 years after the scientific, environmental, and regulatory community began to recognize this pathway, a comprehensive decision-making framework for addressing the potential exposure is still lacking. The full scope of the problem and the policy questions that need to be answered in order to make informed decisions regarding remediation and protecting public health are still unclear. More than 120,000 sites across the country known to have contaminated soil and groundwater have yet to be sufficiently remediated (Ehlers & Kavanaugh, 2013). It is unlikely, due to financial, political and technical limitations, that these tens of thousands of toxic groundwater plumes will be remediated anytime in the near future, and thus without interventions, exposures (such as from vapor intrusion) are likely to persist (Ehlers & Kavanaugh, 2013). Even hazardous waste sites that have been "closed," that is, considered sufficiently remediated, have been reopened specifically to investigate the vapor intrusion pathway (Rolph, Torres, & Everett, 2012).

Regulatory efforts to determine risks associated with vapor intrusion have been shaped by the results of only a small number of studies, which is problematic because broad generalizations about vapor intrusion may not be accurate. For example, early studies from Massachusetts found higher concentrations of chlorinated solvents in the winter compared with the summer, and recommendations to test in the winter were integrated into site assessment practices (Fitzpatrick & Fitzgerald, 2002). Outside of the research reported in this dissertation, this assumption has not otherwise been tested in warm southern or Mediterranean climates, where household behavior patterns and indoor/outdoor temperature and pressure differentials may differ from those in northern climates. Further, agencies have largely assumed a linear relationship between groundwater concentration and indoor air concentrations and thus prioritized (sometimes exclusively) collecting measurements in homes atop the highest plume concentration (Dawson, 2008a). The vapor intrusion monitoring program in Redfield, Colorado, showed that unhealthy indoor air concentrations are found in buildings overlying relatively low levels of contaminants and has helped dispel the belief that clay soil could sufficiently retard the migration of vapor into homes (Folkes et al., 2009; Renner, 2002). Furthermore, current U.S. Environmental Protection Agency (EPA) draft guidance on assessing vapor intrusion sites largely excludes homes with crawl spaces (U.S. EPA, 2002); this is likely to be a relic of early studies conducted in areas of the country where basements are typically found.

As noted in Chapter 1, the EPA has been examining vapor intrusion for decades and has yet to issue final guidance on assessing this pathway. Meanwhile, at least 29 states and several federal agencies have drafted their own vapor intrusion guidance or rules, in order to fill the void created by a lack of final guidance from EPA; however, the majority of these guidance documents are non-binding and, legal analysis suggests the documents may generate more confusion than clarity around the appropriate assessment and subsequent decision-making process (Eklund, Beckley, Yates, & McHugh, 2012; Rolph et al., 2012).

5.2. Key Findings and Implications

This dissertation aims to improve tools of exposure assessment—that is, tools to help forecast, prevent, and mitigate exposures that may lead to adverse human health effects and to help identify populations that face high exposures due to the vapor intrusion pathway. This work demonstrates methods to integrate evidence-based and quantitative tools into the analysis of the vapor intrusion pathway at the community level and proposes new approaches to evaluate exposure when equipped with only limited data and imperfect models. An additional outcome of this dissertation is the development of reproducible, quantitative techniques to incorporate and analyze both the uncertainty and variability among exposure predictions and measurements in order to improve exposure characterization.

5.2.1. Evaluating Current Guidance on Vapor Intrusion Exposure Assessment

Chapters 2-4 examined key assumptions behind the current EPA guidance for assessing the vapor intrusion pathway. The research finds that the scientific evidence and body of data, in general, do not substantiate the current approaches used by the EPA (and many state agencies) to assess exposure. Chapter 2 contributes to the small but growing body of evidence on the heterogeneity of exposures across space and time among residential homes in a contaminated community. Even over a few days, indoor air concentration can fluctuate by orders of magnitude, suggesting that a single sample or even a couple of samples in a home will not adequately characterize the exposure occurring in that space over time. Long-term averages of concentrations (over months or a year) also may mask short periods of high exposure. This work further shows that measurements taken in only one or a few homes in an affected community, as is often recommended by vapor intrusion guidance, are unlikely to be representative of the current exposure across the entire area. These results confirm other recent research that a robust sampling protocol is necessary for accurately characterizing exposure and perhaps challenges the feasibility of the current house-by-house approach to environmental management of vapor intrusion sites (Holton et al., 2013; Schumacher et al., 2013).

The current draft EPA guidance document for assessing vapor intrusion exposure risks outlines a process in which homes must be sequentially 'screened in' for further analysis based on increasingly site-specific and less conservative assumptions (Environmental Quality Management, 2004). For example, to remain on the list of candidates for further evaluation, a home must lie above an area of groundwater contamination for which the contaminant concentration in the soil gas just above the groundwater table is at least 1,000 times the indoor air concentration associated with potential health risks. Homes overlying aquifer sections with concentrations below this threshold are eliminated from the list of buildings that may require remediation. Chapter 3 shows that the use of the current generic vapor attenuation screening factor (1/1000) to screen in sites for further investigations may be insufficiently conservative and may underestimate indoor air concentrations in some cases. The analysis of the largest existing database of vapor intrusion site-level data demonstrates that the generic value is exceeded in 10% of the analyzed cases. The regression-based model developed using empirical data is a potential tool that can be used to improve screening of vapor intrusion sites, as it sharply decreases the percentage of underpredictions (compared to the EPA generic screening) while allowing a less conservative screening value under certain conditions. Understanding important factors affecting the vapor attenuation ratio can be refined (and perhaps improved) by including in the database easily acquired additional information, such as the area and age of the home and meteorological conditions during sampling. This work also supports the idea that regional differences in climate and geology may affect the observed relationships between groundwater chemical concentrations and indoor air chemical concentrations. The work demonstrates a preliminary screening method that is adaptable to local conditions and can serve as a generic method to identify high-risk areas to prioritize for further investigations.

As described in previous chapters, the Johnson-Ettinger model is a favored tool of regulators for predicting exposure from the vapor intrusion pathway. However, the current EPA protocol is to apply this model in its deterministic form, without considering the variability and uncertainty in model input parameters. This research showed that the deterministic approach systematically underestimates the observed concentrations in indoor PCE. Chapter 4 demonstrated a process for combining prior information with limited site-level data to represent uncertainty and variability in the indoor air concentrations predicted by the Johnson-Ettinger model. At least in the case study presented here, the JEM as applied by the EPA appears inadequate as a tool to estimate exposure, particularly if it is used to exclude sites from further analysis. These techniques demonstrate one way to improve the agreement between the JEM predictions and the measured indoor air concentrations, as well as represent the variability and uncertainty.

5.2.2. Quantifying Relationships Between Vapor Intrusion and Other Factors

This dissertation has demonstrated stochastic techniques for analyzing the relationships between indoor air concentrations and environmental, household, and meteorological variables. As the understanding of transport processes that govern vapor intrusion continues to evolve, the insights resulting from the stochastic analyses presented in this dissertation can contribute to the evolving body of knowledge about the physical and chemical processes affecting vapor intrusion. Using a panel study design that incorporates measurements across both space and time, as in this dissertation, allows for the quantification of relationships between indoor air concentrations and temporal and spatial variables. While these relationships are likely not applicable to sites nationwide, the research methods can be applied in other settings to better understand potential exposure patterns and high-risk scenarios for other communities. Estimating these relationships then allows for contextualizing the concentrations measured based on known (and in most cases easily acquired) meteorological, environmental, or household conditions. We have found that temporal changes in weather conditions (barometric pressure drop, wind speed, humidity, and season) can explain approximately a third of the observed variability in short-term fluctuations of indoor PCE concentrations. While this approach does not demonstrate the causal mechanisms behind these observed temporal changes, it does show statistically significant correlations among the variables. This approach can facilitate the understanding of the potential variability of concentrations in houses from a specific site. For future work, incorporating information about the indoor-outdoor temperature differential may refine this model, as it serves as an easily measured proxy of differences between ambient and indoor conditions.

The stochastic approach was extended to the multisite vapor intrusion database collected by the EPA (Chapter 3). Using multilevel statistical analysis of the vapor intrusion attenuation factor, we were able to examine patterns among attenuation and various other characteristics across many previously studied sites. As a result, we were able to evaluate correlations accounting for multiple factors, rather than just bivariate associations, in order to quantify the relationships between soil type, foundation, type contaminant properties, and season. However, few observations have been collected (or made available) from the southern United States and integrating this information may result in different relationships between the covariates and the vapor intrusion attenuation factor than reported in Chapter 3. In some cases similar patterns surfaced from both the local analysis (Chapter 2) and the national analysis (Chapter 3). For example, in both local and national analyses, the foundation type associated with the highest risk was cement slab-on-grade, and finer-grained soil was associated with lower risk. As mentioned, conflicting information also emerged, as summer was correlated with statistically higher indoor PCE concentrations in the San Antonio community, while the opposite effect was observed from the national database. In summary, regression-based approaches can support the understanding and quantification of such associations at vapor intrusion.

5.2.3. Integrating Multiple Sources of Information to Improve Exposure Predictions

A key unresolved issue is how to integrate site information into the process for deciding whether a home affected by vapor intrusion requires remediation. This research has developed tools to integrate site-specific information, empirical indoor air data, and a mechanistic model to improve the agreement between measured and predicted indoor air concentrations. While previous studies have attempted to improve the ability to model vapor intrusion at the individual building level, few studies have compared model results to measured data across multiple homes, and even fewer have used observations to influence model design (Yao & Suuberg, 2013). In this study, we integrated information about environmental factors, household properties, and contaminant characteristics to propose a model for the screening homes for further vapor intrusion analysis (Chapter 3) and we demonstrated a Bayesian approach to calibrating a probabilistic version of the Johnson-Ettinger model (Chapter 4). In face of the complexity and contentiousness around environmental problems, synthesizing information from multiple studies may be a good strategy for resolving environmental controversies (Biggs, Carpenter, & Brock, 2009). Scientists along with environmental managers frequently confront a diverse array of data relevant to a particular problem and need to combine information across different spatial or temporal scales (Wikle, 2003). Chapter 3 demonstrates the use of multilevel modeling to combine information about factors influencing the variability of the vapor attenuation factor both within and across vapor intrusion sites. This approach can easily be extended to include multiple samples over time if such data are available. In Chapter 4, the results show how combining mechanistic and stochastic tools along with observations can improve model performance as well as the utility of the model as a decision-making tool.

Further, these methods are not static, but rather facilitate the integration of new information, allowing an adaptability that is often seen as a desirable component of environmental policy tools. If new data are collected or a new understanding of important parameters or transport mechanisms emerges, previous analyses and data can be incorporated as prior knowledge through this method. Research has advocated for adaptive environmental management processes, in which analysis and decision-making are revisited in a continuous loop as conditions, information, and understanding of the complex system evolves (Polasky, Carpenter, Folke, & Keeler, 2011). The methods demonstrated in this research support this type of adaptive management approach.

5.2.4. Assessing Uncertainty

Recognition of the controversial nature of decision-making on complex environmental issues has spurred discussions about the role of uncertainty in environmental policy analysis. Assessing uncertainty is necessary for addressing the credibility of the scientific approaches used in a decision-making context (Ascough, Maier, Ravalico, & Strudley, 2008; Van der Sluijs, 2002). Inadequate articulation of uncertainties in environmental science for policy has contributed to inappropriate decisions and significant environmental and health damages (Maxim & van der Sluijs, 2011; Morgan & Henrion, 1992). Better understanding of uncertainty and how the level of uncertainty influences action is a prerequisite for better decision-making (Rowe, 1994; Walker et al., 2003). This research, along with emerging work from vapor intrusion sites, provides more conclusive evidence that the vapor intrusion pathway is highly complex, insufficiently described by current mechanistic models, and not well suited for application of simple decision-making tools. Regulatory approaches that support environmental management decision based on a single measured or predicted value are inadequate. Chapter 2 demonstrates that indoor air concentration is highly variable across space and time, and Chapters 3 and 4 show that EPA-recommended deterministic site evaluation models are insufficient as conservative prediction tools. Based on this knowledge, it is important that the uncertainties of both indoor air measurements and modeled values are both acknowledged and quantified. This research in turns supports a key objective of policy analysis as described by Morgan (1978), which is:

> to evaluate, order and structure incomplete knowledge so as to allow decisions to be made with as complete an understanding as possible of the current state of

knowledge, its limitations and its implications.

Because any site-level vapor intrusion assessment will be faced with imperfect information, and because modeling efforts are still advancing, the results of vapor intrusion exposure assessments need to be framed by confidence intervals. The case study research demonstrates the ability to use passive sampling techniques to facilitate repeat sampling of one home in order to collect multiple measurements from which to evaluate the potential range of concentrations. This temporal variability is described in a site-specific stochastic model that helps to assess the range of possible predictions using limited data. Chapter 3 demonstrates a method for predicting the generic attenuation factor, and using the 95th percentile of the prediction decreases the potential for false negatives. Finally, Chapter 4 shows how to explicitly quantify the uncertainty of Johnson-Ettinger model predictions. The analyses show how to compute uncertainty in modeled predictions, which is crucial for meaningful interpretation of model results (Warmink, Janssen, Booij, & Krol, 2010).

5.3. Future Research Needs

While a comprehensive study of the scope of potentially affected sites and populations has not been conducted, it is realistic that hundreds of thousands of buildings may be impacted by vapor intrusion and that this unregulated exposure pathway is likely to pose the greatest risks of exposure to chlorinated solvents for individuals living atop contaminated groundwater in and around hazardous waste or industrial sites (Ferguson, Krylov, & McGrath, 1995; Fischer et al., 1996; Little, Daisey, & Nazaroff, 1992; Provoost et al., 2008). While there is largely consensus from the scientific community that a single-point-in-time sample is insufficient to characterize the exposure occurring inside a single building, and certainly cannot represent an entire community, a framework for systematically evaluating the information in order to inform policy choices does not yet exist. In addition to collecting more data, future research needs include the improvement of tools and techniques to gather data, contextualize the data, and assess the value of such information in face of the uncertainty and potential health risks associated with vapor intrusion.

5.3.1. Decentralized Data Collection and Stakeholder Engagement

Recognized shortcomings of the conventional approaches to vapor intrusion monitoring include the logistical difficulty of placing and retrieving samples, the delay in obtaining results, and the high costs of analysis. Adding to policy demands are community demands for access to technologies that allow community members to work alongside scientists to generate their own exposure data, and more effectively participate in the environmental policy and regulatory processes (Brown et al., 2012). Evidence suggests that community-based research improves the relationship between scientists and residents and enhances both the quantity and quality of data collected (Altman, 1995; Viswanathan et al., 2004). Conducting exposure studies in the absence of community input or failing to maintain communication with affected communities may greatly diminish public confidence in exposure science and reinforce distrust of scientists engaged in this work (Stern & Fineberg, 1996; Wynne, 2006). Vapor intrusion is no exception, and the involuntary and unavoidable nature of the exposure likely fuels residents' desire to ensure agency's decisions are protective of the residents' health.

Innovations in science and technology provide opportunities to overcome data limitations, improve the transparency of exposure assessments, and support a framework to generate knowledge that is effective, timely, and relevant to current and emerging environmental health challenges. Engaging broader audiences may improve the responsiveness of exposure science and support problem formulation, collection of data, access to data, and development of decision-making tools (Lioy & Smith, 2013). In the case of vapor intrusion, the lack of field data, particularly from the southern part of the country, is striking. At the same time, community residents potentially affected by vapor intrusion are demanding more extensive monitoring efforts both pre- and post-mitigation, and the state of the science is showing that a robust monitoring network is necessary to adequately characterize exposure (Siegel, 2009). While work here demonstrates a passive monitoring technology that community members can deploy themselves (Chapter 2), a more comprehensive study could evaluate the comparative efficacy of allowing residents to collect relevant data themselves with current monitoring tools.

It has been suggested that scientific results derived from community and stakeholder engagement can empower individuals, communities, and agencies in preventing and reducing exposures, and in addressing environmental disparities (Boyer, 1996), but this approach has not be studied in relation to vapor intrusion. Increasingly, exposure monitoring equipment is being developed that is portable and can collect near-real-time data on contaminant levels. Efforts are under way to develop small portable sensors to measure ambient CVOCs that may be adaptable to vapor intrusion (Chen et al., 2012; Kim, Burris, Chang, Bryant-Genevier, & Zellers, 2012; Negi et al., 2010). Research into portable and inexpensive devices to collect vapor intrusion data has the potential to facilitate the collection of exposure data and to enable community residents to participate in the exposure assessment process. These devices can collect detailed information to characterize temporal variability, offer real-time readings to residents, and store data electronically.

Improving the characterization of exposure can facilitate further research on the health impacts of vapor intrusion. Evidence suggests acute exposures to CVOCs are linked to adverse pregnancy outcomes such as pregnancy loss, premature birth, developmental abnormalities, and low birth weights (Beliles, 2002; Chiu, Caldwell, Keshava, & Scott, 2006; Doyle, Roman, Beral, & Brookes, 1997). Elevated rates of low birth weight children, fetal growth restriction, and cardiac defects have been associated with exposure with CVOCs via vapor intrusion compared with unexposed populations (Forand, Lewis-Michl, & Gomez, 2011), but more studies are necessary to quantify this relationship further and improve environment health surveillance at vapor intrusion sites.

5.3.2. Vapor Intrusion in Context

Within the current regulatory approach to addressing hazardous waste (including vapor intrusion), it is not sufficient to show that contamination exists. It must also be shown that humans are exposed to that contamination and that this exposure increases the likelihood of an adverse health outcome. The framework for evaluating the vapor intrusion pathway follows the traditional chemical-by-chemical, pathway-by-pathway, one-disease-at-a-time approach widely used to make site-specific cleanup decisions at polluted sites in the United States (Montague, 2004; O'Brien, 2000). Nonetheless it is recognized that environmental health outcomes at the community level are a result of exposure to mixtures of chemicals, combined with social, economic, and psychological stressors that may increase vulnerability at the population level (Evans, Hubal, Kyle, Morello-Frosch, & Williams, 2007). To date, information and frameworks are lacking for evaluating vapor intrusion exposure in relation to cumulative exposures, and in relation to individual and population-level differences in vulnerability (Callahan & Sexton, 2007).

A cumulative exposure assessment framework is needed to incorporate community-level characteristics into vapor intrusion exposure assessments. For example, in the community used as a case study in this analysis, exposure to PCE alone may be occurring not only via vapor intrusion but also in drinking water or through inhalation and dermal exposure during bathing or wading in the local creek or in the workplace. Furthermore, PCE could interact with dozens of other chemicals identified in the community, such that exposure to multiple contaminants could exacerbate the risks posed by PCE. Future research should assess multiple pathways and chemical exposure occurring at vapor intrusion sites and explore how this information can be better integrated into environmental management.

Scientists have suggested that in spite of the increasingly sophisticated models and tools developed to address environmental problems, existing models are insufficient to represent the complex, dynamic nature of environmental systems (Biggs et al., 2009; Pahl-Wostl, 2007; Zellmer, Allen, & Kesseboehmer, 2006). Additional insights to complement existing scientific models and tools are possible through the consideration of community expertise (Corburn, 2002; Farrell, 2006). Therefore, also needed are processes for collecting and documenting community knowledge of vapor intrusion exposure and for integrating the results into the assessment process. The use of community-based exposure assessment techniques should be expanded; such techniques employ the community in defining the problems and the necessary data, supplying local knowledge, and interpreting the results in the context of the local reality (Barzyk et al., 2009; Corburn, 2007). Qualitative methods may offer insights into vapor intrusion research because of their ability to engage residents regarding local environmental health problems and to contribute to the understanding of population exposures by providing data on people's behaviors, their perceptions of risk, and the social, economic, cultural, and political considerations that influence personal exposure (Scammell, 2010). However, a method to combining quantitative and qualitative inputs from scientific studies of exposure, participatory community-based processes, and local knowledge is lacking (Lambert, Guyn, & Lane, 2006). Research should continue on systematic ways to incorporate into the assessment process the specialized knowledge of the community about the local environment, exposure and activity patterns, and lived experiences.

5.3.3. Acting Under Uncertainty

The need to make decisions regarding vapor intrusion is not going to wait until the scientific uncertainty has been resolved. It is clear that one or even a few indoor air samples will not tell the complete story of exposure, but rather adequately characterizing exposure requires long-term monitoring that collects samples on relatively fine temporal and spatial scales. One potential area of further research is on the use of mechanical techniques to create conditions of high exposure. Inducing negative pressure differentials can "turn on" vapor intrusion. Initial data suggest that controlling pressure differentials is technologically simple and reasonably reliable, and collecting samples during such negative pressure conditions may offer a better picture of the high potential indoor concentrations (McHugh et al., 2012). This approach could potentially reduce the potential for collecting false negative readings and decrease the uncertainty associated with a single sample, but further research is still necessary. Such an approach is still limited by the need for trained personnel and a house-by-house analysis to measure exposure, but it nonetheless offers a potential new tool to more quickly estimate potential indoor air concentrations at the household level.

In the face of a potential risk and the inability to fully quantify scientifically that risk, the regulatory framework surrounding vapor intrusion should consider alternative assessment approaches. The precautionary principle has been proposed as a tool to address decision-making in such circumstances (Deville & Harding, 1997; Sandin, 1999; Stewart, 2002). The principle has generally been defined as having two main components: preventive action in the face of uncertainty and reversing the burden of proof. The approach suggests that the decision-maker should anticipate harm before it occurs and provide for some measure of protection against this harm even if the probability cannot be determined accurately by existing science (Crawford-Brown & Crawford-Brown, 2011). This framework shifts away from the traditional paradigm that requires proof of some unacceptable level of risk. Future analysis of vapor intrusion guidance and regulation approaches should further consider: (a) who bears the burden of proof; (b) the evidence required to pass a confidence threshold; and (c) co-benefits , that is, others effects favorable to human welfare that are not directly related to the benefit of vapor intrusion mitigation.

Given the current state of evidence, the vapor intrusion community needs a thoughtful analysis of the costs, benefits, and health implications associated with an extended monitoring program compared to preventive remediation measures. The vapor intrusion pathway can usually be adequately controlled or eliminated in a cost-effective manner by some combination of sealing cracks, venting sumps, modifying HVAC operations, or installing subslab depressurization systems (Fitzgerald, 2009). These interventions may have significant public health co-benefits of reducing radon intrusion or decreasing moisture buildup, though no research on this topic has been published to our knowledge. This information could offer a framework from which to evaluate the trade-offs between collecting more information (additional monitoring) vs. taking action with imperfect information.

5.4. Conclusions

This dissertation examines several aspects of the migration of CVOCs from the subsurface to the indoor air, finding that current approaches to assessment of exposure are inconsistent with the current scientific evidence. While the understanding of the vapor intrusion pathway is still incomplete, the results demonstrate methods to better assess exposure at the community scale and integrate uncertainty analysis into both monitoring and modeling techniques. This research demonstrates quantitative techniques for integrating data into models in order to avoid such underestimates, one flaw in current policy design. Future research should evaluate the potential for community-centered and real-time monitoring devices, the integration of localized and cumulative risk information into the decision-making framework, and assessment of the risks and benefits of a precautionary approach to vapor mitigation.

APPENDIX A

Spatiotemporal Variability of Tetrachloroethylene in Residential Indoor Air Due to Vapor Intrusion: A Longtudinal, Community-Based Study – Supplementary Material

The supplemental materials contain the following information:

- Details on the equations behind the Tobit regression model,
- Analysis of covariance of the covariates considered for the model
- Picture of the sampling setup

Tobit Regression Model:

In the likelihood function the censored (below detection limit) data are represented as a probability of being less than the censoring threshold. In the likelihood function the censored (below detection limit) data are represented as a probability of being less than the censoring threshold. The resulting regression model is characterized by a latent regression equation:

$$y_i^* = \mathbf{x}_i^{\mathrm{T}}\beta + \varepsilon_i \tag{1}$$

where y_i^* is the latent dependent variable and the error term (ε_i) is assumed to be independent and normally distributed with a mean of 0 and a variance of σ^2. The observed dependent variable, y_i is defined as

$$y_i = y_i^* \text{ if } y_i^* > c \tag{2}$$

$$y_i = c \text{ otherwise.}$$

where c represents the detection limit. With a log-transformed dependent variable, the resulting likelihood function is:

$$L = \prod_{y_i = c} \Phi\left(\frac{\log c - \mathbf{x}_i^{\mathrm{T}}\beta}{\sigma}\right) \prod_{y_i > c} \phi\left(\frac{\log y_i - \mathbf{x}_i^{\mathrm{T}}\beta}{\sigma}\right) \tag{3}$$

Table A.1. Analysis of variance of covariates considered in the model.

	R^2	F-statistic	F-statistic significance
Age of home (years)	0.036	6.80	0.0099
Air conditioning (yes/no)	0.027	2.45	0.089
Area of home (m^2)	0.0082	1.48	0.22
Barometric pressure drop (mm)	0.038	0.61	0.092
Cleaning conducted (yes/no)	0.0035	0.61	0.44
Dryer used (yes/no)	0.0064	0.84	0.36
Humidity (%)	0.025	4.58	0.034
PCE groundwater concentration (µg/L)	0.0261	3.16	0.078
Season (summer/winter)	0.015	2.65	0.10
Soil (clay/silt)	0.013	1.61	0.11
Temperature (°C)	0.0057	1.03	0.3122
Wind (m/s)	0.033	6.14	0.014
Windows (opened/closed)	0.0093	1.69	0.19

Figure A.1. An example of the passive sampling set up used in residential homes in this study.

APPENDIX B

Screening Houses for Vapor Intrusion Risks:
A Multiple Regression Analysis Approach – Supplementary Material

<u>The supplemental materials contain the following information:</u>

- Details on the distribution of the vapor intrusion attenuation factors
- Figure showing the shows site-to-site and region-to-region variability of the attenuation factors.
- A univariate analysis of the vapor intrusion attenuation factors and the model covariates

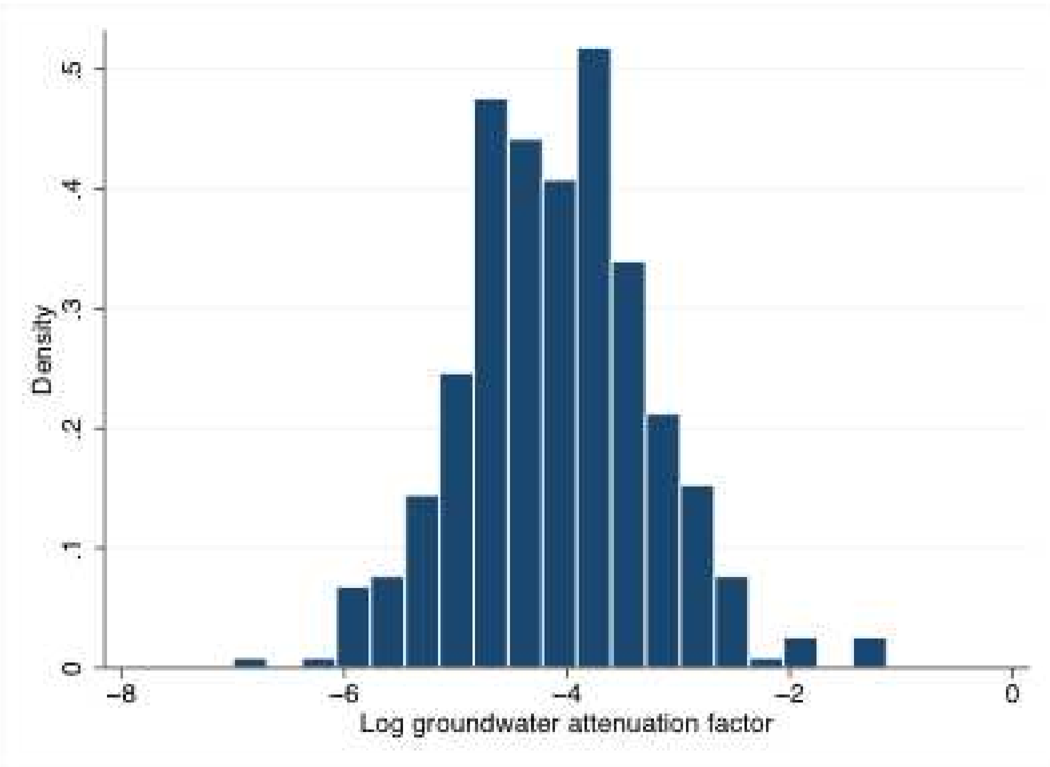

Figure B.1. Distribution of pooled vapor intrusion attenuation factors for all observations included in statistical analysis.

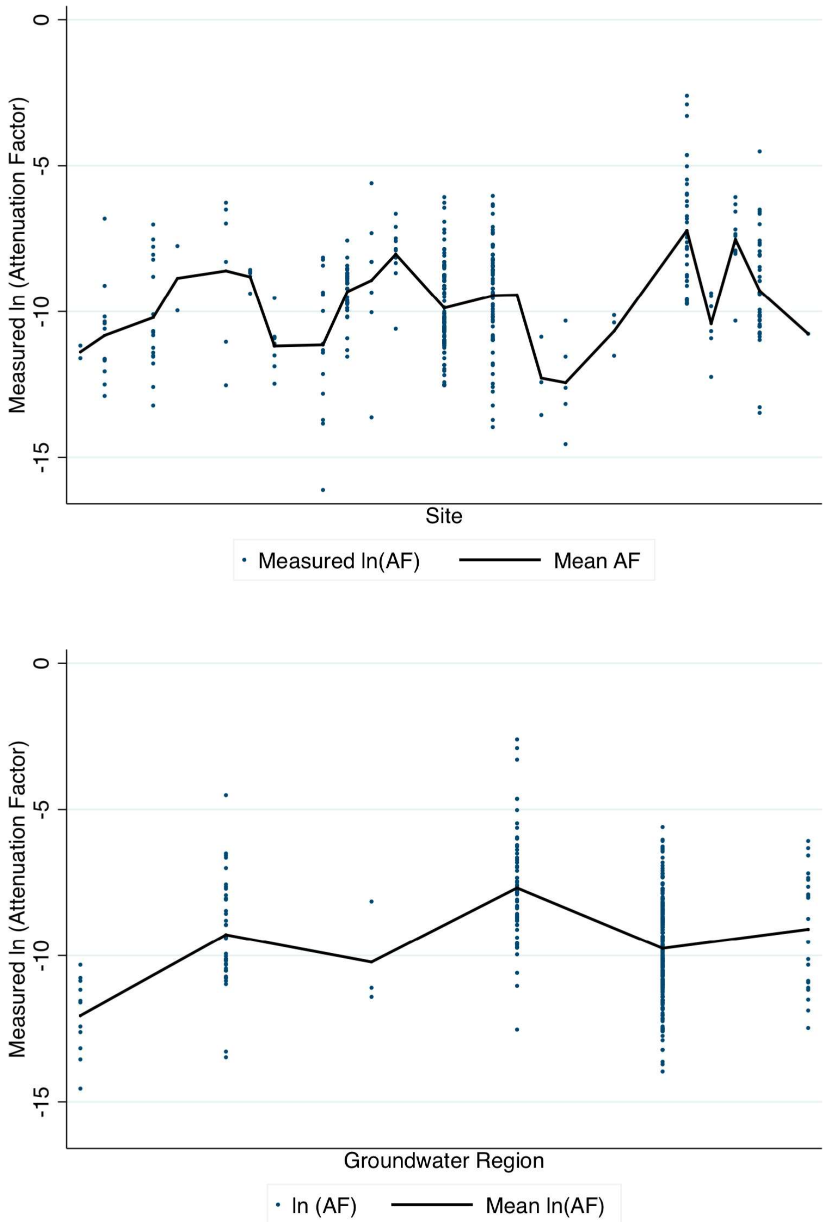

Figure B.2. Distribution of measured natural logarithm of attenuation factors for CVOCs across the sites and groundwater regions; black line tracks the mean attenuation factor at each site and region and shows site-to-site and region-to-region variability.

Univariate and Variance Component Analysis

Previous studies of radon have used the analysis of variance (ANOVA) approach as a technique to examine the proportion of variation in indoor radon contributions explained by various factors, such as geology, foundation type or building ventilation (Appleton & Miles, 2010; Burkhart & Huber, 1993; Louizi, Nikolopoulos, Lobotessi, & Proukakis, 2001). Similarly, we employed ANOVA as an initial approach to assessing the significance of the differences between means for pooled observations for the different factors potentially affecting vapor intrusion.

According to the one-way ANOVA, soil type followed by foundation type and groundwater depth explained the greatest proportion of variation in measurements among fixed effect covariates (Table B.1). All the explanatory variables were statistically significant ($p<0.05$), based on the *F*-test. The chemical properties explained the smallest portion of the variance in attenuation.

To assess the relative importance of the various levels of analysis prior to the inclusion of the additional covariates, we considered the ratio of each variance component—ε_{ijk}^2(representing variance between buildings), δ_{0jk}^2 (between-site variance), and υ_{00k}^2(between-region variation)—to the total variance in the attenuation factors. The random effects variance component at the observation level (ε_{ijk}^2) accounted for a major portion—60.9%—of the variance. Importantly, however, this implies that 39.1% of the variance is due to higher levels of analysis. Specifically, the site level (δ_{0jk}^2) accounts for 13.2% and the geologic region (υ_{00k}^2) for 25.9% of the variance. These results indicate that significant variation in the vapor intrusion attenuation factor occurs at all three levels of analysis, providing further evidence of the multilevel character of the vapor intrusion data.

Table B.1. Proportion of the variation of log vapor intrusion attenuation factor for residential homes explained by environmental, household and multilevel factors.

Covariates	Pooled observations	
	Number of categories	% variation explained
Diffusivity in Air	Continuous	2.5%
Chemical molecular weight	Continuous	2.9%
Season	2	3.9%
Groundwater depth (m)	Continuous	5.5%
Foundation	4	6.0%
Soil type	3	8.5%
Random effects variance components		
Groundwater region	6	25.9%
Site	1	13.2%
Observation	370	60.9%

APPENDIX C

Updating Exposure Models of Indoor Air Pollution Due to Vapor Intrusion: Bayesian Calibration of the Johnson-Ettinger Model – Supplementary Material

Table C.1. Additional model inputs, parameters, and references not in Table 4.3.

	Model Parameter	Description	Modeling Method*		Primary Reference
Groundwater	PCE concentration	Presence of TCE and PCE in groundwater aquifer, µg/L	Bayesian maximum entropy, lognormal (mean, standard deviation) for each grid cell		Air Force Real Property Agency
Groundwater	L_t	Aquifer distance from ground level surface, cm	Bayesian maximum entropy, normal (mean, standard deviation) for each grid cell		Air Force Real Property Agency
House & Foundation	L_{crack}	Foundation thickness, cm	15		(Environmental Quality Management, 2004; Johnson, 2005)
House & Foundation	Z_{crack}	Depth below ground surface to bottom of the foundation, cm	15		(Environmental Quality Management, 2004)
House & Foundation	X_{crack}	Total length of cracks through which soil gas vapors are flowing / floor-wall seam perimeter, cm	Sqrt(Area/2)*6		Author's judgment
House & Foundation	Area (A)	Area of enclosed space below grade, cm^2	Evaluated independently for each household (discrete)		Bexar County Appraisal District, 2009
Soil	$D^{eff}_{c,z}$	Effective diffusion through the capillary zone, cm^2/s	C SiC	.000016 .000026	(Carsel & Parrish, 1988; Environmental Quality Management, 2004)
Soil	$L_{c,z}$	Height of the capillary zone, cm	C SiC	81.5 192	(Carsel & Parrish, 1988; Environmental Quality Management, 2004)

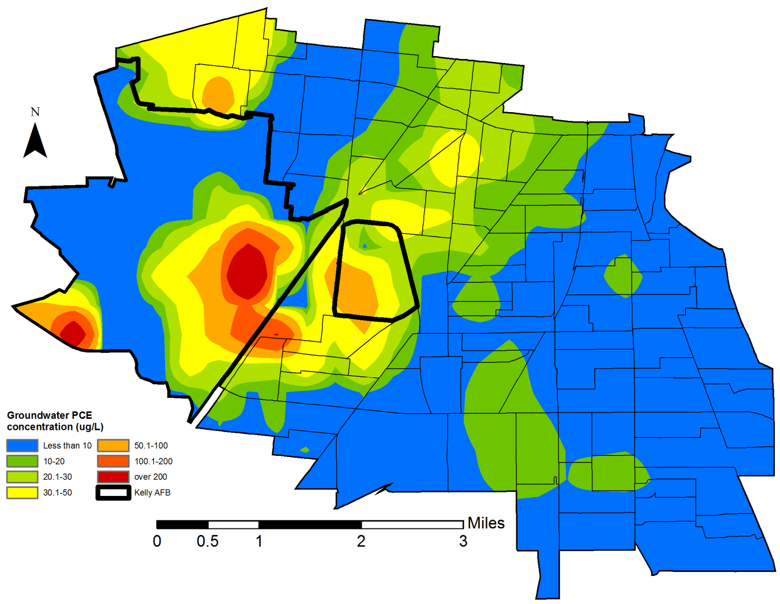

Figure C.1. Estimated PCE groundwater concentrations (μg/L) in 2011.

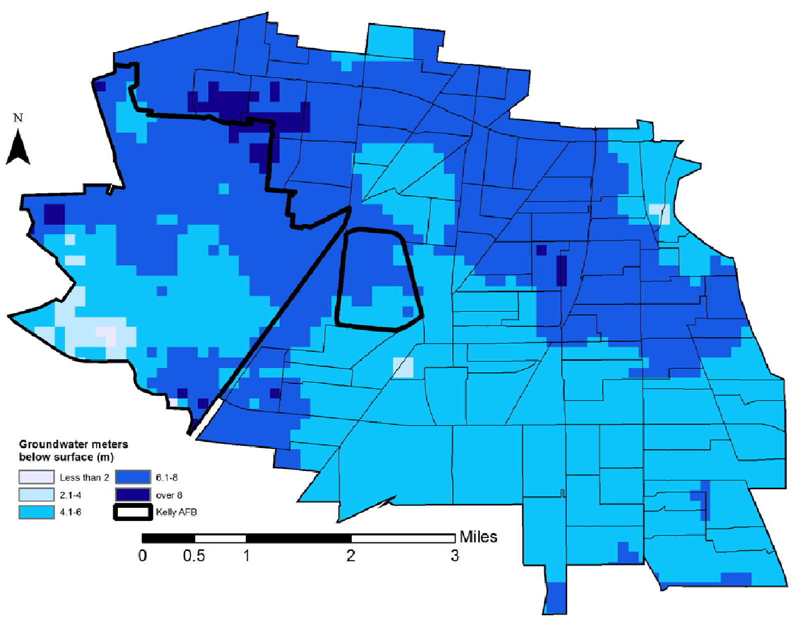

Figure C.2. Estimated groundwater levels (m below surface) in 2011.

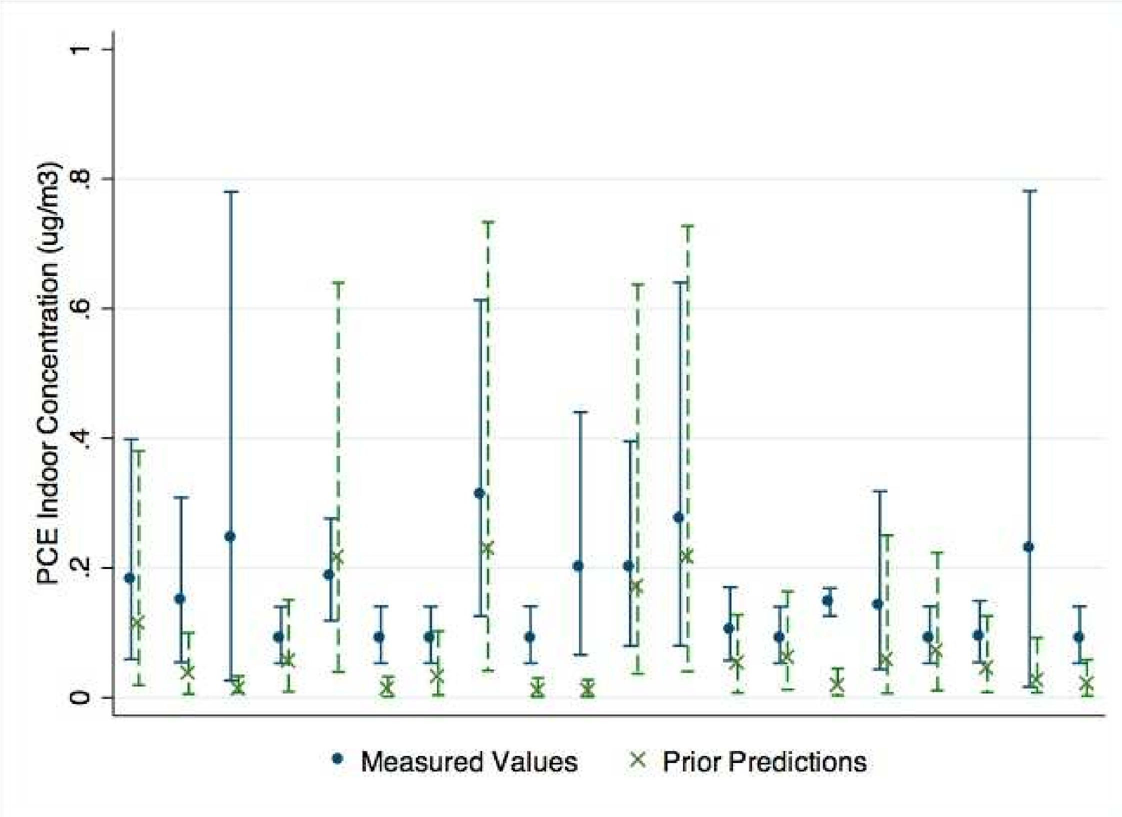

Figure C.3. Measured values (circle- mean with 90% confidence interval) compared to the prior probability predictions (x-mean with 90% confidence interval).

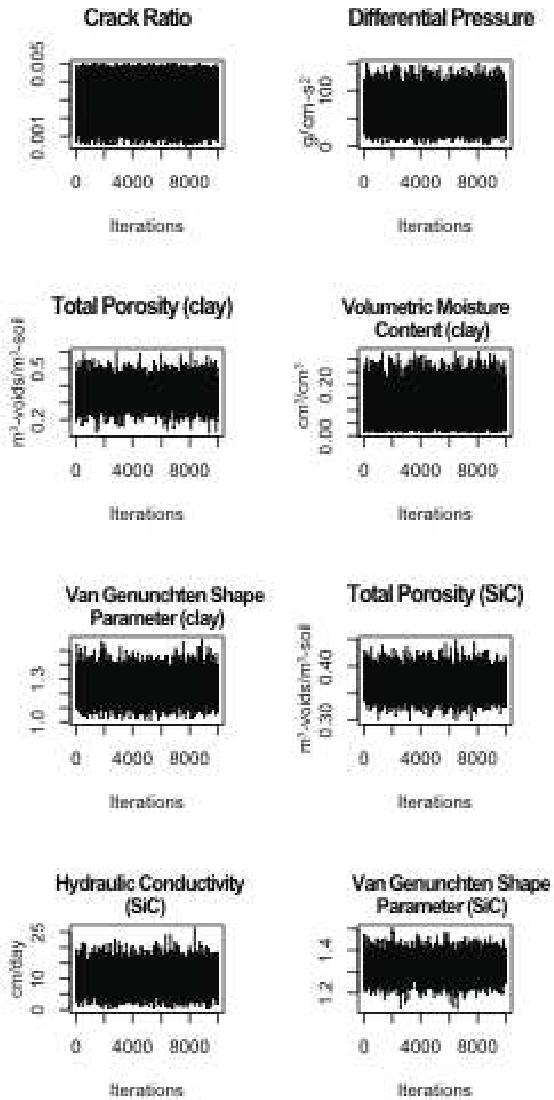

Figure C.4. Trace plots shown for one of the three parallel chains for Model 1.

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