Transient Variability In Vapor Intrusion And The Factors That Influence It

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2 Abstract

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Temporal variability in indoor air contaminant concentrations at vapor intrusion (VI) sites has been a concern for some time. We consider the source of the variability at VI sites located near Hill Air Force Base and Naval Air Station North Island using statistical analysis methods and three-dimensional subsurface computational fluid dynamics modeling. The results suggest that an order of magnitude variation in indoor air contaminant concentrations may be expected at "normal" VI sites where preferential pathways do not play a role, whereas three or more orders of magnitude can be observed at sites characterized by preferential pathways. A CFD modeling sensitivity analysis reveals that it is not only the presence of contaminant vapor in a preferential pathway that is required to see large observed variations, but there must also exist a permeable region beneath the structure. Large temporal fluctuations in indoor air contaminant concentrations may be observed where no preferential pathways exist, but this requires a particular combination of large fluctuations in pressure driving force and high soil permeability.

17 Introduction

Long term vapor intrusion (VI) studies in both residential and larger commercial structures have raised concerns regarding significant observed transient behavior in indoor air contaminant concentrations. ¹⁻⁶ VI involves the migration of volatilizing contaminants from soil, groundwater or other subsurface sources into overlying structures. VI has been a recognized problem for some time, but many aspects remain poorly understood, particularly with respect to the causes of large temporal transients in indoor air concentrations. There is uncertainty within the VI community regarding how to best develop sampling strategies to address this problem. ^{1,3,7}

Results from a house operated by Arizona State University (ASU) near Hill AFB in

Utah, an EPA experimental house in Indianapolis, IN and a large warehouse at the Naval
Air Station (NAS) North Island, CA have all shown significant transient variations in indoor
air contaminant concentrations. All were outfitted with sampling and monitoring equipment
that allowed tracking temporal variation in indoor air contaminant concentrations on time
scales of hours. All have shown that these concentrations varied significantly with time orders of magnitude on the timescale of a day or days. ^{5,8,9}

In one instance the source of the variation was clearly established during the study; at
the ASU house a field drain pipe (or "land drain"), which connected to a sewer system, was
discovered beneath the house, and careful isolation of this source led to a clear conclusion that
this preferential pathway significantly contributed to observed indoor air contaminant levels
and their fluctuations. ^{9,10} While in this case the issue of a contribution from a preferential
pathway was clearly resolved, what it left open was a question of whether existence of such
a preferential pathway to an area beneath a structure would always be expected to lead to
large fluctuations in indoor air contaminant concentrations.

Similarly, a sewer pipe has recently been suggested to be a source of the contaminants found in the EPA Indianapolis house. ¹¹ That site was also characterized by large indoor air contaminant concentration fluctuations. Sewer lines have been generally implicated as VI

source at several sites to date. 11-14 A Danish study estimates that roughly 20% of all VI sites in central Denmark involve significant sewer VI pathways. Thus while the consideration of a role of possible sewer or other preferential pathways is now part of normal good practice in VI site investigation, it is still not known whether the existence of such pathways automatically 47 means that large temporal fluctuations are necessarily to be expected. In some of these cases. 12,14 the sewer provided a pathway for direct entry of contaminant into the living space. While potentially important in many cases, this scenario is not further considered here, where the focus is on pathways that deliver contaminant to the soil beneath a structure. 51 It is, however, now known that even absent a preferential pathway, there may be sig-52 nificant transient variation in indoor air contaminant concentrations at VI sites. ^{2,4,15} One 53 example is a site at NAS North Island at which no preferential pathways have been identified. 54 Instead, a building at this site is characterized by significant variations in indoor-outdoor pressure differential.⁵ It is believed that this is the origin of the fluctuations at that site. This paper investigates the sources of the temporal variation in indoor air contaminant 57 concentrations in both the presence and absence of in soil preferential pathways. In this work, the latter scenarios are referred to as "normal" VI scenarios, in which there is typically a groundwater source of the contaminant. Specifically, we pose the question of just how much variation in indoor air contaminant concentration may be expected at such normal VI sites vs. those characterized by preferential pathways. The conditions required for preferential pathways to be significant contributors to temporal variations in indoor air con-63 taminant concentrations are also explored, and the consequences for sampling strategies are

also discussed.

66 Methods

67 Statistical Analysis Of Field Data

To begin to characterize transient behavior in indoor air contaminant concentrations, actual datasets are analyzed to establish common levels of variability at VI sites. For this purpose, the datasets from the ASU house in Utah, the EPA Indianapolis site and North Island NAS were chosen for analysis. This paper relies on statistical analysis of published field data, and 71 readers are referred to the original works for details regarding data acquisition. 3,5,8,9,16 The ASU house and data were obtained over a period of a few years. During part of 73 this time, controlled pressure method (CPM) tests were being conducted, in which the house was underpressurized to an extent greater than that characterizing "normal" operation. This 75 caused greater than normal advective flow from the subsurface into the house, thus increasing 76 VI potential. ^{6,8,17} This period of CPM testing is considered separately from the otherwise 77 "natural" VI conditions in the analysis. Likewise, the existence of a preferential pathway at the ASU house needs to be considered in examining the dataset, noting that during some of 79 the testing, this pathway was deliberately cut off, resulting in what we have termed "normal" VI conditions in which the main source of contaminant was believed to be groundwater. The NAS North Island dataset has not (as far as is known) been influenced by a prefer-82 ential pathway, but the structure there was subject to large internal pressure fluctuations, much more extensive than those typically recorded at the ASU house during normal operations. Additionally, the underlying soil at NAS North Island is sandy and more permeable than that at the ASU site, which, as will be shown, contributes to the indoor air contaminant concentrations being more sensitive to pressure fluctuations.⁵ Likewise, the Indianapolis site investigation spanned over a number of years and period-88 ically included the testing of a sub-slab depressurziation system (SSD). The goal of which

is to mitigate the VI risk by drastically depressurizing the sub-slab area underneath the

house, preventing the contaminants from entering the structure above, and therefore only

the period before the installation of this system was considered in the analysis. It is likely a sewer line beneath the structure acted as a preferential pathway, ¹¹ however at no point was this PP removed, making it difficult to assess how significant the PP was at this site, regardless it is of interest to analyze this site due to its wealth of data.

The typical variation in indoor air contaminant concentrations with time will first be 96 considered below in the case of the ASU house during "natural", (i.e. non-CPM conditions), 97 in the case of the NAS North Island site over the entire available dataset, and for the Indianapolis case we consider the variations before the installation of the SSD system. The deviations from the mean TCE (Chloroform and PCE at the Indianapolis site) in indoor 100 air concentration, as well as the indoor-outdoor pressure differential associated with these 101 concentration fluctuations, were examined, and both univariate and bivariate kernel density 102 estimations (KDE) were constructed. KDE is a technique that estimates the probability 103 distribution of a random variable(s) by using multiple kernels, or weighting functions, and 104 in this case, Gaussian kernels are used to create the KDEs. This means that it is presumed 105 that the variables of interest (i.e., indoor air contaminant concentrations and indoor-outdoor 106 pressure differentials, as sampled) are normally distributed around mean values (that there 107 is statistical fluctuation associated with each sampling event). In this instance, the scipy statistical package was used to construct the KDEs, assuming a bandwidth parameter determined by Scott's rule. The distributions of the individual parameters and the relationship between them will be examined using the KDE method. 111

112 Modeling Work

In addition to examining the actual field data, a previously described three -dimensional computational fluid dynamics model of a generic VI impacted house was used to elucidate certain aspects of the processes. This model was implemented in a finite element solver, COMSOL Multiphysics. In the present work, there has been an addition of a preferential pathway to the "standard" model that has been described before in publications by this

group. 18-20 In this modeling work, only the vadose zone soil domain is directly modeled.

The modeled structure is assumed to have a 10x10 m foundation footprint, with the 119 bottom of the foundation slab lying 1 m below ground surface (bgs), simulating a house 120 with a basement. The indoor air space is modeled as a continuously stirred tank (CST)¹ 121 and all of the contaminant entering the house is assumed to enter with soil gas through a 1 122 cm wide crack located between the foundation walls and the foundation slab that spans the 123 perimeter of the house. All of the contaminant leaving the indoor air space is assumed does 124 so via air exchange with the ambient. The indoor control volume is assumed to consist of only 125 of the basement, assumed as having a total volume of 300 m³. Clearly different assumptions 126 could be made regarding the structural features and the size of the crack entry route, but for 127 present purposes, this is unimportant as the intent is only to show for "typical" values what 128 the influence of certain other features can be. The modeled surrounding soil domain extends 129 5 meters from the perimeter of the house, and is assumed to consist of sandy clay (except 130 as noted). Directly beneath the foundation slab, there is assumed to be a 30 cm (one foot) 131 thick gravel layer, except in certain cases where this sub-base material is assumed to be the 132 same as the surrounding soil (termed a "uniform" soil scenario). The preferential pathway 133 is modeled as a 10 cm (4") pipe that exits into the gravel sub-base beneath the structure. The air in the pipe is assumed to be contaminated with TCE at a vapor concentration equal to the vapor in equilibrium with the groundwater contaminant concentration below 136 the structure, modified by a scaling factor χ , which is allows the contaminant concentration 137 in the pipe to be parameterized. The groundwater beneath the structure is assumed to be 138 homogeneously contaminated with trichloroethylene (TCE) as a prototypical contaminant. 139 The groundwater itself is not modeled, as the bottom of the model domain is defined by the 140 top of the water table. The ground surface and the pipe are both assumed to be sources of 141 air to the soil domain. Both are assumed to be at reference atmospheric pressure. Vapor 142 transport in the soil is governed by Richard's equation, a modified version of Darcy's Law, 143 taking the variability of soil moisture in the vadose zone into account. 21 The van Genuchten equations are used to predict the soil moisture content and thus the effective permeability of the soil.²² The effective diffusivity of contaminant in soil is calculated using the Millington-Quirk model.²³ The transport of vapor contaminant in the soil is assumed to be governed by the advection-diffusion equation, in which either advection or diffusion may dominate depending upon position and particular circumstances. The equations and the boundary conditions are given in Table 1.

Drivers For Indoor Air Contaminant Variability

The Role Of Building Depressurization/Pressurization

Out of the factors that influence IACC in VI, pressure is one of the most dynamic ones, 153 and the relationship between changes in indoor/outdoor pressure difference and IACC are examined in Figure 1. Three well-studied VI sites are considered, the "ASU house" near Hill 155 AFB in Utah, a building at North Island NAS in California and a duplex in Indianapolis. The absolute IACC at these sites vary significantly, therefore some means of comparing 157 them all to each other is necessary. Additionally, the focus in this section is the driver for 158 variations in IACC, and representation of the variations between the different sites must 159 also be achieved, specifically variations on the scale of order of magnitudes is of the greatest 160 interest, as these are the largest hinderance to proper site IACC characterization. To achieve 161 these goals, the log-10 deviation from the mean IACC (μ) within each dataset is calculated, 162 e.g. 10μ indicates the IACC is an order of magnitude above the mean IACC for the dataset. 163 On the y-margin in figures 1a and 1b, the univariate KDE distribution of deviation from the 164 mean may be seen. 165

The indoor/outdoor pressure differences are simply taken as it, and their univariate KDE distributions are shown on the x-margin. A negative pressure difference indicate that the building is underpressurized relative to outdoor.

The relationship between the deviation from mean IACC, and indoor/outdoor pressure difference may be seen in the central portion of each figure, in the form of a bivariate KDE

Table 1: Governing equations, boundary conditions & model input parameters. (See below for table of nomenclature).

(a) Governing equations

Unsteady-CSTR	$V_{\frac{du}{dt}} = \int_{A_{ck}} j_{ck} dA - u A_e V$
Richard's equation	$\nabla \cdot \rho \Big(- \frac{\kappa_s}{\mu} k_r \nabla p \Big) = 0$
Millington-Quirk	$D_{ ext{eff}} = D_{ ext{air}} rac{ heta_g^{10/3}}{ heta_t^2} + rac{D_{ ext{water}}}{K_H} rac{ heta_w^{10/3}}{ heta_t^2}$
Advection-diffusion equation	$\frac{\partial}{\partial t} \Big(\theta_w c_w + \theta_g c \Big) = \nabla (D_{\text{eff}} \cdot \nabla c) - \vec{u} \cdot \nabla c$
van Genuchten equations	$Se = \frac{\theta_w - \theta_r}{\theta_t - \theta_r} = [1 + \alpha z ^n]^{-m}$ $\theta_g = \theta_t - \theta_w$ $k_r = (1 - Se)^l [1 - (Se^{-m})^m]^2$ $m = 1 - 1/n$

(b) Boundary conditions

Boundary	Richard's equation	Advection-diffusion equation
At foundation crack	$p = p_{\rm in/out}$ (Pa)	$j_{\mathrm{ck}} = \frac{uc}{1 - \exp\left(uL_{\mathrm{slab}}/D_{\mathrm{air}}\right)}$
At groundwater source	N/A	$c = c_{\rm gw} K_H \; (\mu \rm g/m^3)$
At ground surface	p = 0 (Pa)	$c = 0 \; (\mu g/m^3)$
Exit of preferential pathway	p = 0 (Pa)	$c = c_{\rm gw} K_H \chi \; (\mu g/m^3)$

(c) Soil & gravel properties $^{24-26}$

Soil	Permeability (m ²)	Density (kg/m ³)	θ_s	θ_r	$\alpha (1/m)$	n
Gravel	$1.3 \cdot 10^{-9}$	1680	0.42	0.005	100	3.1
Sand	$9.9 \cdot 10^{-12}$	1430	0.38	0.053	3.5	3.2
Sandy Clay	$1.7 \cdot 10^{-14}$	1470	0.39	0.12	3.3	1.2

(d) Trichloroethylene (diluted in air) properties 25,26

$D_{\rm air}~({ m m}^2/{ m h})$	$D_{\rm water}~({\rm m}^2/{\rm h})$	Density (kg/m^3)	Viscosity ($Pa \cdot s$)	K_H	M (g/mol)
$2.47 \cdot 10^{-2}$	$3.67 \cdot 10^{-6}$	1.614	$1.86 \cdot 10^{-5}$	0.403	131.39

(e) Building properties

$V_{\text{base }}(\text{m}^3)$	$L_{\rm slab}$ (cm)	$A_e (1/\mathrm{hr})$
300	15	0.5

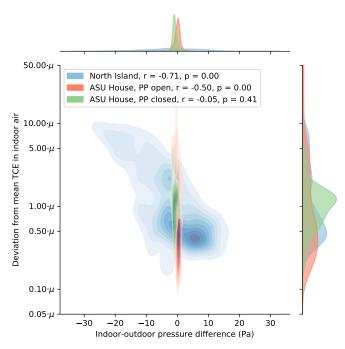
distribution. The p-value and Pearson's r-value for each bivariate distribution is shown in the legend.

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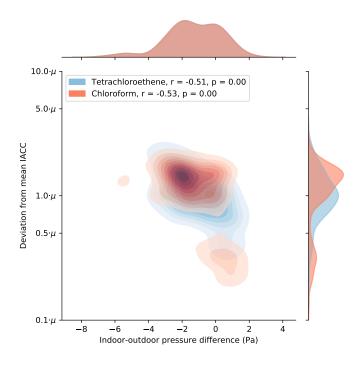
In Figure 1a the North Island and ASU house datasets are plotted, with blue representing

Figure 1: KDE analysis of IACC dependence on indoor/outdoor pressure difference at the ASU house and North Island site (1a) and the Indianapolis site (1b). p-values and Pearson's r-values shown for each dataset.

(a) Period before and after the PP was shut at the ASU house considered separately; North Island dataset considered in its entirety.



(b) Chloroform and PCE considered separeately at Indianapolis duplex 422.



the North Island site, and with red and green representing the ASU house. The ASU house dataset is split into two different parts, the first (red) is data from the time period before the PP was discovered, the second (green) is from after the PP was sealed off. At both of these sites, TCE was the only contaminant considered. The IACC of Chloroform and PCE in the 422 part of the Indianapolis duplex are considered in Figure 1b.

From Figure 1a, it is apparent that the three datasets differ significantly from each other.

The North Island site exhibit the greatest variation in both pressure difference and IACC, in
fact, looking at the univariate pressure distribution - it is almost entirely flat and spanning
from extremly high and low values. A likely explation for this is the poor condition of the
structure, rendering it highly susceptible to envivornmental influnces.

To accompany this, there is significant variation in IACC, that has a relatively strong 184 link with pressure difference (r = -0.71), although there is clearly a distinct peak (around 185 (0.5μ) that is primally associated with the smaller negative and positive pressure differences 186 ($-4 \le \Delta p \le 8$). This demonstrates that at this particular site, the very large variations 187 (an order of magnitude or more) are primarily influenced by the unusually large pressure 188 difference fluctations. Likely, this relationship is amplified by the fact that the soil sur-189 rounding the structure is sandy and therefore relatively permeable - increasing the influence 190 of pressure differences. However, despite all this, pressure differences can only partly explain 191 the variations observed at this site. 192

The two ASU datasets are not only significantly different from the North Island dataset, but also different from each other. Consider the dataset before the PP was discovered (red) and one can see that the IACC varies significantly (from 0.05μ to more than 50μ , albeit rarely.) This is more variation than was observed at North Island, and yet the distribution of pressure differences is narrow, mostly varying by a few Pa around zero (± 2 Pa), and with r = -0.50, suggests that pressure difference is not insignificant but still a weak predictor for the variations in IACC.

The picture changes completely after the PP was closed off, where a similar distribution of

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pressure differences is observed, but with a signficant reduction in IACC variance to around $\pm 0.5\mu$, resembling something of a log-normal distribution. This clearly shows just how much 202 variance the PP at this site contributed with, and how critical it is to assess whether a PP 203 is present at a particular site in general, lest potentially face unacceptable uncertainty in 204 determining the relevant contaminant exposure. It is also clear that in the absence of a 205 PP, the pressure difference becomes even less important, with the p- and r-value indicating 206 that pressure differences are here insignficant in determining variation in IACC. This also 207 suggests that the PP at the ASU house acted not only as a PP for contaminant vapors, but 208 for air in general - allowing the impermeable soil surrounding the house to be circumvented. 209 PPs, and the factors that influnce them will be explored later in this article. 210

The Indianapolis duplex also featured a PP, but despite this, there is significantly less variance in IACC of both Chloroform and PCE. The exact reason for this is not known, but considering that the PP at the Indianapolis site was a sanitary sewer, and at ASU the PP was a foundation drain system, it is not inconceivable that their dynamics are quite different. Nevertheless, both PCE and Chloroform are more or less log-normally distributed, with (mostly) less than an order of magnitude variance around the mean, indicating that the varation in IACC are species independent.

The pressure difference distribution exhibit some bimodal characteristics, with a larger variation than was observed at the ASU site. This may explain why the r-values for both PCE and Chloroform, r = -0.51 and r = -0.53 respectively, are so similar to the ASU site before the PP there was closed; the larger pressure differences, which occured more often than ASU, increase contaminant entry rates to the extent that a PP was needed to "achieve" at the ASU site.

All of these data clearly suggest that there is significant variation in IACC that cannot be easily explained by variations in indoor/outdoor pressure difference - even with a PP. It is also clear that PP may contribute with unacceptable levels of variations, and screening for PPs should be part of any site investiation. It may also be prudent, even in the absence of PP, to add around a half to one order of magnitude margin of error to early screening
measurements of indoor air. E.g. if a single sample is less than an order of magnitude
from some target limit, it may be justified to perform subsequent samples to better assess
the relevant exposure. These data also call the role of advection in VI into question, as
"normal" pressure difference ranges are such a poor predictor of IACC variation, which is
the opposite of what one would expect if advection was a dominant transport mechanism.

The Role Of Air Exchange Rate

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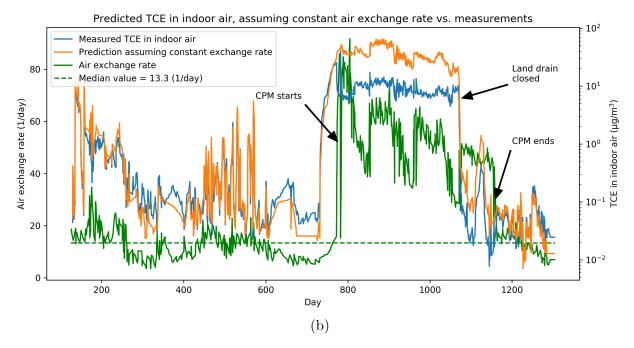
Air exchange rate, which measures how often the interior air in a given building is exchanged 235 with exterior air, is the main parameter characterizing contaminant expulsion, and another 236 highly dynamic process. Exploring the impact of fluctuations in air exchange rate is thus also 237 important for the development of sampling strategies. At the ASU house, the contaminant 238 entry rate into the structure (called emission rate in that study) as well as the building air 239 exchange rate were recorded across time. This allows for exploration of the impact that fluctuating air exchange rate has on the final IACC by solving the CST equation, using the recorded contaminant entry rate as one input, and assuming a constant air exchange rate. The rationale is to show the extent of variability in IACC due to factors other than fluctuations in air exchange rates. We have taken this factor out of consideration by assuming a single constant exchange rate, characteristic of the whole period. 245

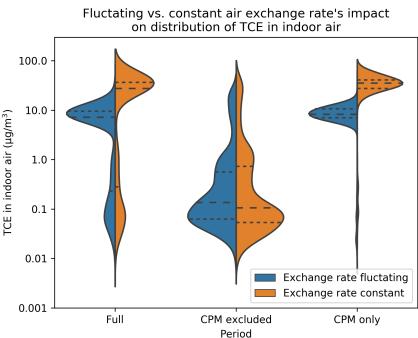
In Figure 2a, the measured IACC and air exchange rate across time are shown, with
the predicted IACC assuming a constant air exchange rate. The median air exchange rate
value for the pre-CPM period was chosen as this seemed to be the most representative value
for this time period. From Figure 2a it may be seen quite clearly that for the non-CPM
periods, the calculated IACC assuming constant and actual fluctuating exchange rates are
quite similar. In other words, in this instance it is unlikely that the variations in IACC were
driven by fluctuations in air exchange rate.

This is more clearly shown in Figure 2b, where KDEs of the IACC are constructed

Figure 2: Comparison between the recorded and the calculated TCE in indoor air at the ASU house, assuming constant air exchange rate. 2a shows the TCE in indoor air across time as well as the exchange rate. 2b shows the distribution of these values for three periods.

(a)





assuming the constant and fluctuating air exchange rate cases. This figure shows the relative probability of being at a particular TCE in indoor air concentration at the ASU house. These

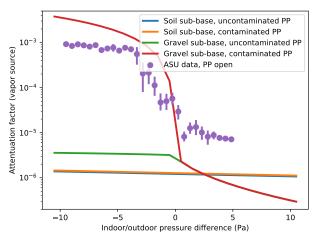
are compared to each other for 1. the full measurement period, 2. the non-CPM periods and
3. the CPM period. Looking at the full period, it does not appear as though the two cases of
steady and fluctuating air exchange rate are comparable, the distribution functions for IACC
are different and offset. The curves to the different sides of the vertical line for the "full"
measurement period represent the probability distributions for the indicated values of IACC.
It appears as though assuming a constant air exchange rate shifts the IACC probability
distribution to higher values. The reason for this are more apparent when considering the
CPM and non-CPM periods separately.

Considering only the CPM period, assuming a constant air exchange rate does not in-264 fluence the shape of the IACC distribution, but assuming constant exchange rate shifts the 265 IACC to a higher values. This is because the actual air exchange rate for this CPM period is 266 significantly higher than the assumed median value, and much more contaminant is actually 267 exchanged with the exterior. The shape of the IACC distribution is not very different from 268 that which takes the air exchange fluctuations into account however, indicating that fluctu-269 ating exchange rate is not a major contributor to observed IACC variation for this period 270 (the distinction being drawn between variation and absolute levels of IACC). 271

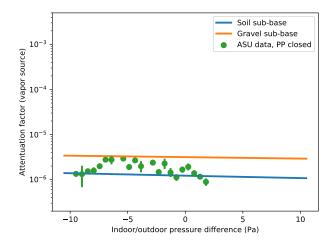
The non-CPM period is the condition under which field practitioners would normally collect samples at a VI site. It is clear here that there is only a very minor difference made in assuming a constant vs. real fluctating air exchange rate, again indicating that account for the minor fluctuations of air exchange rate's impact on variations in IACC is not important. Rackes and Waring ²⁷ drew similar conclusions regarding the relationship between IACC and air exchange rate.

Figure 3: Sensitivity analysis of IACC dependence on indoor/outdoor pressure difference for a model featuring a PP (3a) and without a PP (3b). Results compared with field data from ASU house.

(a) PP present. Sensitivity to the presence of a gravel sub-base and contamination in the PP considered.



(b) No PP present. Sensitivity to the presence of a gravel sub-base considered.



²⁷⁸ The Influence of Preferential Pathways

279 Conclusions From Steady-State Modeling

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Table 2: Abbreviations & symbols

Abbreviation or symbol	Explanation
VI	Vapor intrusion
PP	Preferential pathway
$L_{ m slab}$	Thickness of the foundation concrete slab
p	Pressure
u	TCE in indoor air concentration
c	TCE in soil gas concentration
$D_{ m air}$	Diffusion of TCE in air
$D_{ m water}$	Diffusion of TCE in water
$D_{ m eff}$	Effective Diffusion of TCE in soil
K_H	Henry's Law constant
$ heta_g, heta_w \ \& \ heta_s$	Vapor, water filled & saturated soil porosity
$ heta_r$	Residual moisture porosity
$\alpha, l, n \& m$	van Genuchten parameters
Se	Soil moisture saturation
k_r	Relative permeability
$lpha_{ m gw}$	Attenuation factor

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