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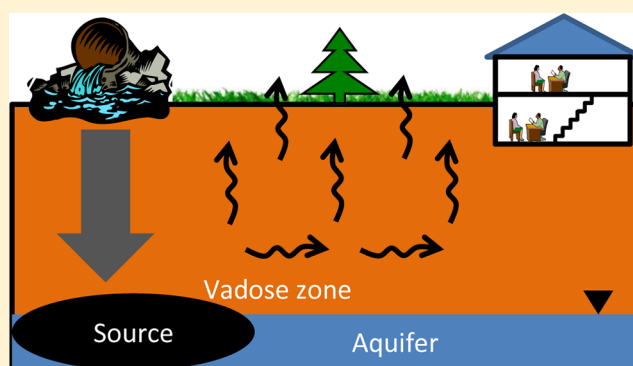
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Examination of the Influence of Environmental Factors on Contaminant Vapor Concentration Attenuation Factors Using the U.S. EPA's Vapor Intrusion Database

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ABSTRACT: Those charged with the responsibility of estimating the risk posed by vapor intrusion (VI) processes have often looked to information contained in the U.S. Environmental Protection Agency (EPA)'s VI database for insight. Indoor air concentration attenuation factors have always been a key focus of this database, but the roles of different environmental factors in these attenuation processes are still unclear. This study aims to examine the influences of these factors in the context of the information in the VI database. The database shows that the attenuation factors vary over many orders of magnitude and that no simple statistical fluctuation around any typical mean value exists. Thus far, no simple explanation of this phenomenon has been presented. This paper examines various possible contributing factors to the enormous range of observed values, looking at which ones can plausibly contribute to explaining them.



INTRODUCTION

The U.S. EPA's VI database¹ is a collection of measured data from VI sites throughout U.S.² The database includes all kinds of information involving building foundations types, contaminants, subsurface, and indoor air contaminant concentrations. The "attenuation factors" (AF) of indoor air concentration to subsurface soil vapor concentrations have always been a key focus of this database and related reports^{3–5} published by U.S. EPA's Office of Solid Waste and Emergency Response (OSWER). In both database and reports, statistical analyses and evaluation were made of different contaminant vapor concentration attenuation factors and data-screening criteria, and a conclusion was reported that VI conceptual models are basically reliable.⁶ However, except for background indoor air concentration,⁴ there has been little quantitative discussion about the role of different environmental factors based on results in this database. The possible role of both spatial and temporal variability, and even nonrepresentative samples was mentioned, but leaves practitioners with little quantitative guidance.

Figure 1 presents cumulative probability plots of the measured AF as reported in the EPA's VI database. These kinds of plots, also presented by U.S. EPA in its analysis of the database,^{1,3,5,6} present cumulative probability distributions for the indicated AF ratios, i.e., $(c_{in})/(c_{gw})$, $(c_{in})/(c_{ss})$, and $(c_{ss})/(c_{gw})$, where c_{in} , c_{ss} , and c_{gw} are contaminant vapor concentrations from indoor air, subsurface, and near groundwater surface, respectively. The value of c_{gw} in the VI database is normally based on groundwater sampling and vapor concentration is estimated from Henry's Law.¹

There are two types of contaminant source, polluted groundwater and contaminant vapor from nonaqueous phase vapor liquid (NAPL), in unsaturated media. In the EPA database,¹ polluted groundwater was considered as the major contaminant vapor source and is the focus of this study. The contaminant vapor concentration attenuation process from groundwater source to indoor air consists of two parts, the first involving transport from groundwater to subsurface and the second from subsurface to indoor air. Figure 1 shows that the distributions of these different measured AF values are similar. They all show a large variation (6–7 orders of magnitude) which is why they are plotted on a log scale. This range is independent of biodegradation, since cumulative probability plots are also similar for nonbiodegradable contaminants, such as PCE and TCE. There, however, is not an a priori reason to see this large a variation in AF, according to the governing equations of vapor transport used in intrusion modeling. The results of an examination of the influence of various factors on the variation seen in the database are presented below. We consider first the commonly accepted equations governing contaminant transport from source through soil to the receptor building.

Received: August 24, 2012

Revised: November 20, 2012

Accepted: December 19, 2012

Published: December 19, 2012

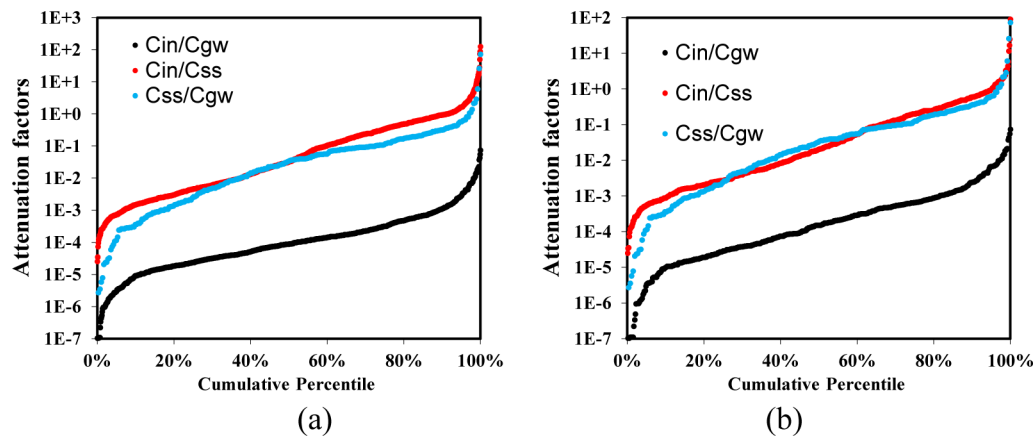


Figure 1. Cumulative probability plots for contaminant vapor concentration attenuation factors (a: all contaminants; b: PCE and TCE; c_{in} : indoor air contaminant vapor concentration; c_{ss} : subslab contaminant vapor concentration; c_{gw} : groundwater contaminant vapor concentration).¹

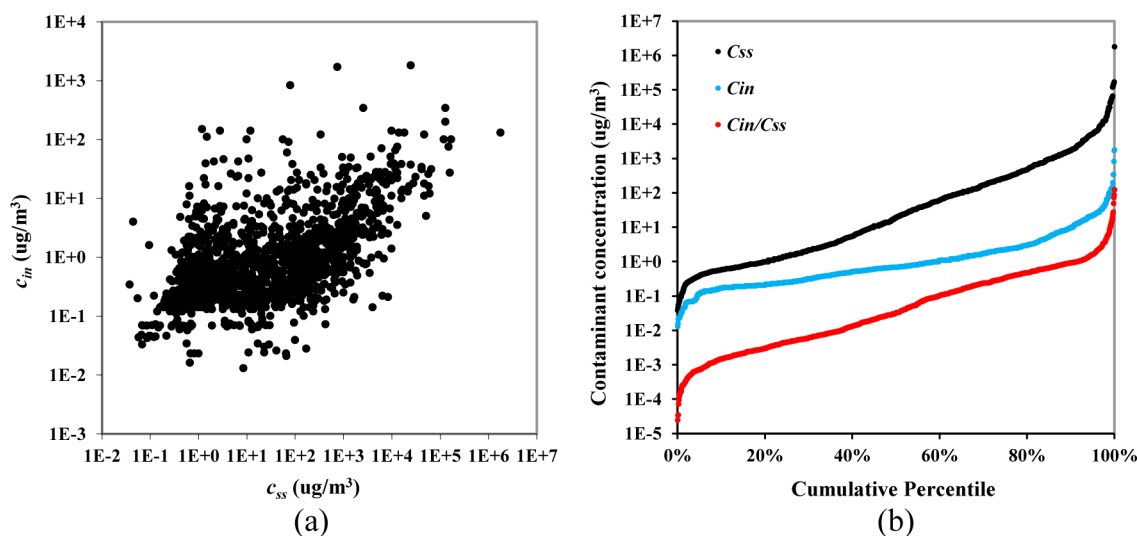


Figure 2. Indoor air and subslab contaminant vapor concentration data from the U.S. EPA's VI database.¹

GOVERNING EQUATIONS IN VI MODELS

Soil Vapor Transport. In steady state, the nondimensionalized equation governing nonbiodegradable contaminant vapor transport can be written as⁷

$$0 = -\nabla^* \cdot C^* - \nabla^* \cdot \left(\frac{C^* q_w}{H} \right) + \nabla^* \cdot (D^* \nabla C^*) \quad (1)$$

where ∇^* represents the nondimensional ∇ operator, nondimensionalized with respect to some characteristic length scale L , $C^* = (c_g)/(c_{gw})$, and $D^* = (D_L^*)/(q_g)$. c_g is the concentration of contaminant in the gas phase [M_i/L^3_{gas}], c_{gw} is the concentration of contaminant in the groundwater source vapor [M_i/L^3_{gas}], q_g is the soil gas flow per unit area [$L^3_{gas}/L^2_{soil}/T$], q_w is the water flux in the vadose zone if relevant [$L^3_{water}/L^2/T$], D is overall effective diffusion coefficient for transport of contaminant in porous media [L^2/T], and H is the contaminant Henry's Law constant [$(M_i/L^3_{gas})/(M_i/L^3_{water})$].

Note that this equation does not include terms with a logarithmic or exponential form involving concentration - it is a linear PDE.

Entry into the Building. The common way to estimate indoor air contaminant concentration c_{in} is to represent the

enclosed space of concern as continuous stirred tank(s) (CST)⁸

$$c_{in} = \frac{J_s + V_b A_e c_{atm}}{V_b A_e + Q_s} \approx \frac{J_s}{V_b A_e} \quad (2)$$

where J_s is the contaminant mass flow rate into the enclosed space [M/T], V_b is the volume of the enclosed space [L^3], c_{atm} is the contaminant concentration in atmosphere [M/L^3], Q_s is the volumetric flow rate of soil gas into the enclosed space [L^3/T], and A_e is the indoor air exchange rate [$1/T$].

In most cases, where depressurization of indoor air space determines the contaminant entry rate, the normalized indoor air concentration to subslab concentration is

$$\frac{c_{in}}{c_{ck}} \approx \frac{c_{in}}{c_{ss}} \approx \frac{Q_s}{V_b A_e} \quad (3)$$

where c_{ck} is the subslab crack contaminant soil vapor concentration [M/L^3].

Equation 3 suggests that c_{in} is linearly dependent on c_{ss} , and this conclusion also works for scenarios involving a permeable wall contaminant entry or any other case for which $Q_s = 0$.

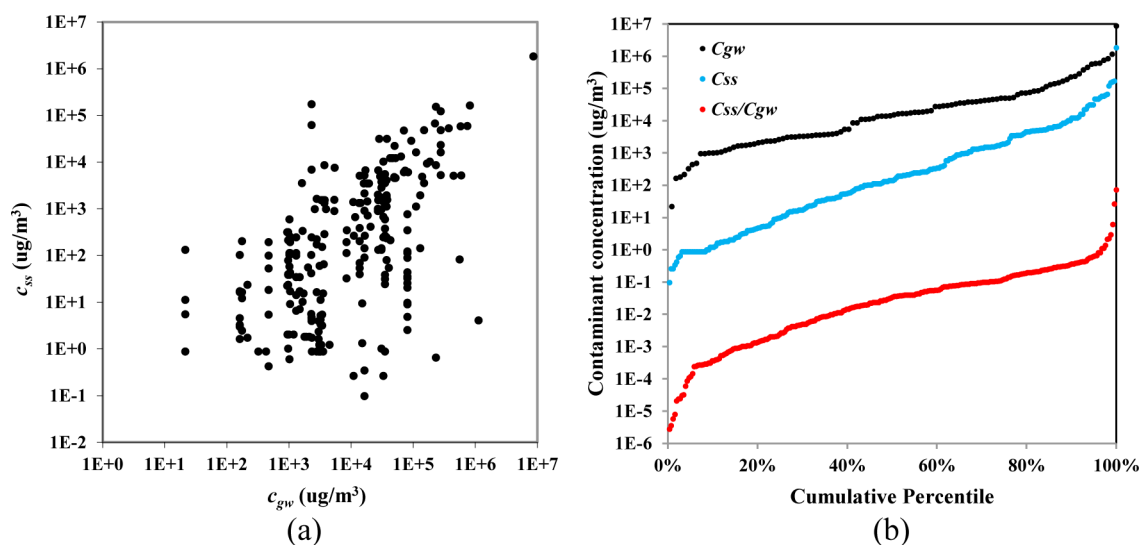


Figure 3. Groundwater and subslab PCE and TCE vapor concentration data from the U.S. EPA's VI database (duplicates of c_{gw} were removed in (b)).¹

Table 1. Summary of Influences of Environmental Factors in Normalized Subsurface Contaminant Soil Concentration Profile (oom Refers to Order(s) of Magnitude)^a

environmental factors		influence
building	building foundations (basement, crawl space, and slab-on-grade)	Contributes <1 oom for scenarios with uniform source and soil, supported by ^{9,11,17} $(c_{ss})/(c_{gw}) \approx ((d_f)/(d_s))^{1/2}$
	multiple buildings	Contributes <1 oom, supported by ^{9,15}
	indoor pressurization	Contributes <1 oom, supported by ^{9,11,17,37}
	indoor air exchange rate	Does not directly affect soil vapor transport
	impermeable surface cover	Contributes <1 oom for scenarios with uniform source and soil.
	indoor pressure distribution (wind load)	Contributes <1 oom, as indoor pressurization. ⁹
soil	soil permeability	Like indoor pressurization, it only changes the soil gas flow rate, which plays an insignificant role in soil gas concentration profile ^{9,11,17}
	effective diffusivity	Does not affect soil gas concentration profile for uniform soil.
	layered soil of different effective diffusivity or permeability (horizontal and vertical)	Can play a significant role ^{9,14} and depends on the distribution, i.e. the existence of capillary fringe may induce orders of magnitudes effect. ²⁹
	biodegradation rate (for petroleum hydrocarbon)	Can significantly affect soil gas concentration profile, depending on source concentration and biodegradation rate. ^{9,12,13}
contaminant source	sorption	Can induce significant change associated with transients. ⁹
	source concentration	When examines $(c_{ss})/(c_{gw})$, matters only for scenarios with biodegradation. ^{9,12,13}
	source depth	Contributes <1 oom for scenarios with uniform source and soil, supported by ^{9,11,17}
	source-building separation	For vertical separation, the influence is limited in a uniform soil. For lateral separation, the contaminant concentration decays exponentially with the horizontal transport distance. ^{9,17,33,34} $(c_{ss})/(c_{gw}) \approx ((d_f)/(d_s))^{1/2} e^{-(d_h)/(2d_s)\pi}$
	multiple sources	Complex, depends on source concentration and distance.

^aThe influences were only considered for nonbiodegradation cases, unless included in the explanation.

DISCUSSION

As of 2012, 2,929 paired measurements from 42 vapor intrusion sites across the country have been included in U.S. EPA's Vapor Intrusion database.¹ Of these measurements, "1,021 (35 percent) are paired groundwater and indoor air measurements, 235 (8 percent) are paired exterior soil gas and indoor air measurements, 1,582 (54 percent) are paired subslab soil gas and indoor air measurements, and 91 (3 percent) are paired crawlspace and indoor air measurements".⁵ In the discussion of this study, the entire database without any filters was used in the figures unless noted in the captions. As for possible spatial and temporal variability in the data in the EPA database, since this variability exists for every site and every structure, EPA

suggested a range of empirical attenuation factors and a statistical approach should be applied.⁵ Moreover, because the EPA VI database is such a large pool of data, the influences of any unknown factors, which might be responsible for temporal or spatial variation, are minimized, and the influence of the most important environmental factors should emerge through statistical approaches.

Subslab-to-Indoor Air Contaminant Vapor Concentration Attenuation Factor. Figure 2(a) is prepared from the EPA database and presents measured indoor air contaminant concentration c_{in} as a function of measured subslab contaminant vapor concentration c_{ss} . This figure shows that most c_{in} values fall in a small range of 0.1 to 10 $\mu\text{g}/\text{m}^3$, while there is a much more significant variation in c_{ss} - from 0.1 to 10⁵

$\mu\text{g}/\text{m}^3$. In a previous study,⁷ the lower limit of indoor air concentration is explained as typically being near the reporting limit or detection limit or background concentration for a contaminant, while the upper limit might be a result of indoor adsorption equilibria or background concentration.⁹ Whatever the reason, the indoor air concentration can be seen to be relatively tightly bounded when compared to subslab vapor concentration.

Figure 2(b) shows cumulative probability plots for actual reported values of c_{ss} , c_{in} , and $(c_{in})/(c_{ss})$, and suggests a relationship between the plots of $(c_{in})/(c_{ss})$ and c_{ss} . That is, the shape of the c_{in} curve is relatively “flat” in the range 0.1 to $10 \mu\text{g}/\text{m}^3$, so it must be the large variation in c_{ss} that gives the $(c_{in})/(c_{ss})$ curve its much broader range. At the same time, there should still be some kind of linear relationship between subslab and indoor air contaminant vapor concentration according to eqs 2 and 3. Consequently these data speak to the fact that there are physical restraints of some kind on indoor air data that will necessarily make use of eqs 1 and 2 to predict indoor air concentrations problematic.

Groundwater-to-Subslab Contaminant Vapor Concentration Attenuation Factor. Figure 3(a) presents the measured subslab contaminant vapor concentration c_{ss} as a function of estimated groundwater source contaminant vapor concentration c_{gw} . Again, what is normally measured is not c_{gw} , but groundwater contaminant concentration, from which c_{gw} is estimated. There appears to be a trend showing a relationship between c_{ss} and c_{gw} , which is in general agreement with eq 1. However, the same figure shows that the variance of groundwater-to-subslab contaminant vapor concentration attenuation factor $(c_{ss})/(c_{gw})$ can be as large as 5 orders of magnitude for same c_{gw} . There are many field results in the EPA database¹ suggesting that the subslab concentrations in a limited area (within a short distance of a monitoring well) can be significantly different from one another.

Figure 3(b) shows cumulative probability plots of c_{gw} , c_{ss} , and $(c_{ss})/(c_{gw})$. It suggests the variations in these three variables are similar (6–7 orders of magnitude). Based on this, it can be concluded that the distribution of observed c_{ss} results can be a consequence of both distribution of c_{gw} and transport process in soil. The nature of the groundwater contaminant source variation is a result of many factors and is discussed elsewhere.⁷ The focus now turns to the contaminant concentration attenuation during the soil vapor transport process or the role of environmental factors in determining $(c_{ss})/(c_{gw})$.

The Influences of Environmental Factors on VI. A list of environmental factors potentially influencing vapor intrusion was given in a technical report published by US EPA’s OSWER in 2012,¹⁰ and these are generally considered in models of the process.^{7,11–16} Values for all are not recorded in EPA’s database.

The environmental factors in the EPA report¹⁰ are summarized by main category⁷ in Table 1: building conditions (receptor), soil (pathway), and contaminant source (source). There are three types of building foundations described in the VI database, slab-on-grade, basement-partial, and basement. Virtually all models of VI show that subsurface concentration profiles beneath buildings with basements are not very different than profiles beneath buildings built slab-on-grade.^{12,15–18} Moreover, a significant variation in $(c_{ss})/(c_{gw})$ was observed even for the same foundation type, as shown in Figure 4.

Previous studies¹⁹ showed that for most cases contaminant soil gas intrudes overlying buildings by convection through slab

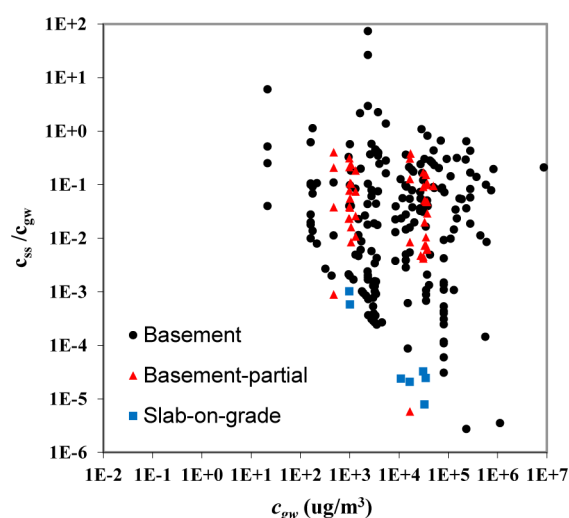


Figure 4. Groundwater-to-subslab PCE and TCE concentration attenuation factor for different building foundations.¹

cracks, and the level of indoor depressurization can significantly affect contaminant entry rate into a building⁸ but has little effect on overall subslab concentration profiles.^{12,18}

Figure 5 indicates that in a typical vapor intrusion scenario, there is little subslab crack concentration variation if the

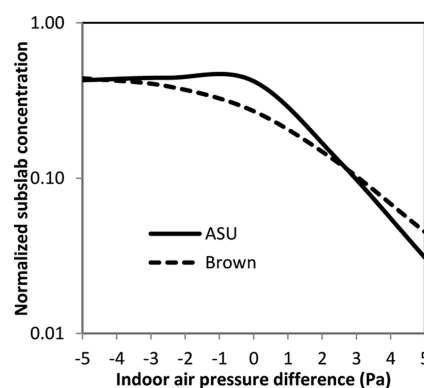


Figure 5. Simulated subslab crack concentration (normalized to source) change as a function of indoor air to ambient pressure difference.^{11,22}

building is operated under negative or atmospheric pressure conditions. When the indoor air pressure becomes positive, the subslab crack concentration will be diluted by the outflow of indoor air, which is normally relatively cleaner. This effect should be less significant if the subslab sampling location is away from the crack. Data showing the effect of indoor pressure on subslab concentration have recently been published.²⁰ This point is supported by a field study by McHugh et al.,²¹ which also suggested that in most cases the indoor pressurization change does not have a significant influence on subslab contaminant concentration.

Yao et al.¹⁸ suggested an estimate of subslab contaminant concentration for uniform soil effective diffusivity and source vapor concentration distribution:

$$\frac{c_{ss}}{c_{gw}} = \sqrt{\frac{d_f}{d_s}} \text{ or } \frac{c_{ss}}{c_{gw}} = \left(\frac{d_f}{d_s}\right)^{0.7} \quad (4)$$

The influence of building operational conditions (e.g., depressurization) on subslab contaminant concentration was not included in this estimate, because it was shown by accompanying numerical simulations that these have limited impact. So these operational factors do not explain orders of magnitude variation in subslab concentration. Likewise, variation in the foundation construction (including especially cracks and breaches) can significantly affect indoor air concentration but not subslab concentrations (except locally very near a breach).²⁰

The contaminant mass entry rate used in eq 2 can be obtained for entry through a crack, and its form, as is assumed in many common VI models,^{8,11–17,23} is

$$J_s = \frac{c_{ss} Q_s}{1 - \exp\left(-\frac{Q_s d_{ck}}{A_{ck} D^{ck}}\right)} \quad (5)$$

where A_{ck} is the area of the crack [L^2], d_{ck} is the thickness of the foundation slab [L], D^{ck} is the diffusivity in the crack [L^2/T^2], and $c_{in} \ll c_{ss}$ is typically assumed. Again, the entry rate is linear in c_{ss} . The effect of the area of the crack (A_{ck}) is mainly felt through the effect on Q_s , and of course larger areas of breach can lead to higher contaminant entry rates, but it is unlikely that orders of magnitude variations in c_{ss} are due to this factor.

Finally, the extent of impermeable ground cover (and with that, the presence of neighboring buildings) can influence subslab concentration levels to generally no more than an order of magnitude.^{10,16}

Hence none of these factors involving building (construction and operational conditions) can explain multiple orders of magnitude variation in subslab concentrations, especially when viewed in ratio to source concentration. It may, of course, be possible to take specific action to influence subslab concentrations (e.g., deliberate subslab venting), but to our understanding the EPA database does not include such samples.

This leaves factors that characterize the source and soil as the most likely origins of order of magnitude variation in $(c_{ss})/(c_{gw})$ observations.¹ Since a significant building-to-building variation in $(c_{ss})/(c_{gw})$ can be observed at the same central site over a short sampling time period, the factors involving long-term effects on time scales of months or years, e.g. sorption, would be ruled out as responsible.

In steady state, if diffusion is the dominant mode of contaminant transport in the soil, this attenuation factor $((c_{ss})/(c_{gw}))$ is mainly determined by diffusion direction, physical diffusion distance, and intervening soil effective diffusivity distribution. For most screening level VI tools,^{8,24–30} the contaminant is assumed to diffuse vertically from a uniform bottom source to the upper building receptor or open ground surface. In these models, the vertical distance from groundwater source to building foundation (or open surface) is believed to be the diffusion path length. 3-D simulations^{12,15} confirm that for scenarios with uniform soil diffusivity, the normalized contaminant concentration profile is roughly linear from groundwater table to open ground surface, as expected. Meanwhile values beneath buildings can approach source vapor concentrations. This by itself suggests that the relatively small range of vertical source-building separations (0.6–13 m) in the EPA database would not lead to a variation of 5–6 orders of magnitude in concentration or $(c_{ss})/(c_{gw})$, as confirmed in Figure 6, which shows no dependence of the attenuation factor on source depth.

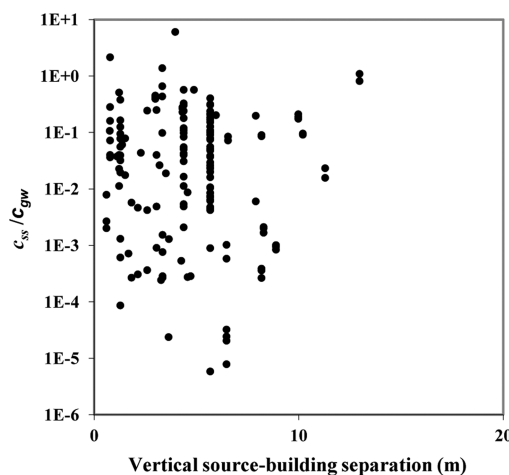


Figure 6. The influence of vertical source-building separation on groundwater-to-slab PCE and TCE concentration attenuation factors.¹

We turn now to the influence of soil diffusivity. It may be seen from typical estimates of gas diffusion in soils that the effective diffusivity variation in most dry soils is no more than an order of magnitude. What is more important than dry soil diffusivity is variation in air-filled voidage.³¹ If the soil is characterized by exceptionally low voidage, especially for soils containing water that fills most porosity, then soil diffusivity can vary by orders of magnitude from one place to another or one day to another. A capillary zone always exists above a watertable, and other saturated zones can be formed in soil after a heavy rain.³² The existence of such saturated low diffusivity zones can influence subslab concentration by orders of magnitude.^{15,33} The heterogeneity in soil effective diffusivity can thus help explain the orders of magnitude variation in $(c_{ss})/(c_{gw})$, as illustrated in Figure 7. What this means is that by interposing a very low diffusivity water-filled soil layer between a presumed source and receptor, there is created a zone of steep concentration gradient. Beyond this, if there is variability in the existence of such low diffusivity zones beneath sites, this is a plausible cause of large (orders of magnitude) variation in $(c_{ss})/(c_{gw})$ ratios.

In fact, there is an issue of characterizing a “source” right below the capillary zone, as noted in a previous paper.⁷ The correct groundwater concentration must be used with Henry’s law. There is an issue of characterizing this concentration. As shown in Figure 8, there can be a significant variation in measured c_{gw} that might simply have to do with variability of the contaminated groundwater source.^{33,35–37} A report suggested that only the first 10 cm below groundwater table should be used for risk assessment.³⁸

In most common VI screening tools,^{8,18,24–30} a uniform source is assumed such that vertical diffusion in the soil is presumed to dominate vapor transport. In practice there could be groundwater source contaminant concentration gradients in both vertical and horizontal directions.^{33,37} While the measured c_{gw} from one monitoring well has often been used as the groundwater source concentration for many buildings at different locations on a site,¹ Figure 8 illustrates the potential for problems arising from such a simplifying assumption. Again, since contaminant groundwater concentration is linearly related to estimates of c_{gw} , the uncertainty in groundwater concentration profiles will lead to comparable orders of magnitude

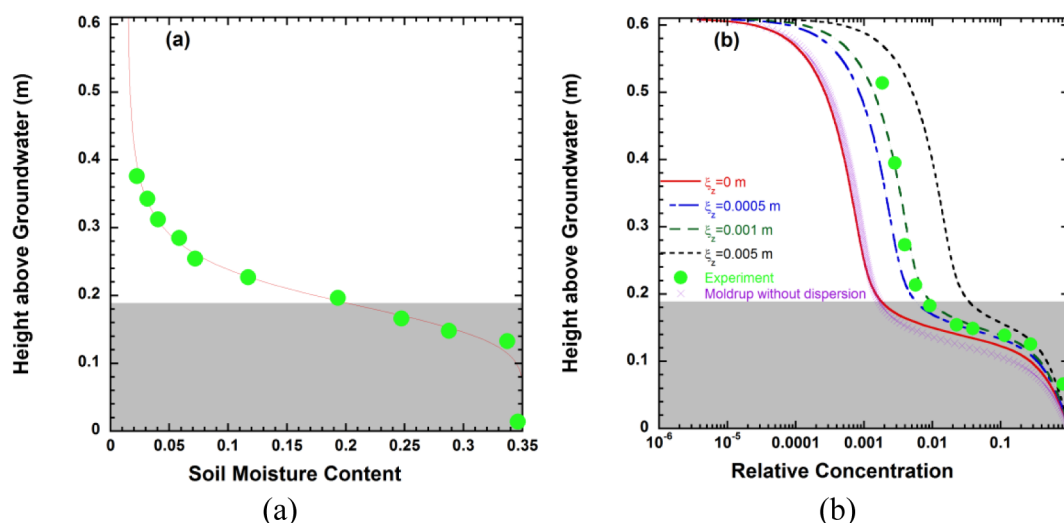


Figure 7. Simulated moisture content (a) and normalized contaminant concentration profile (b) caused by rainfall. A capillary fringe is shown as the shaded area.³⁴

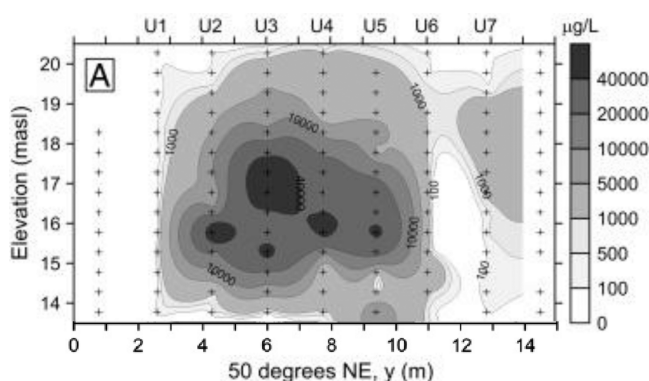


Figure 8. Sectional representations of the sum of measured chlorinated solvents (PCE, TCE, chloroform, and 1,1,1-TCA) in a typical groundwater source.³⁷ Reprinted from ref 37 with permission.

variation in c_{gw} . For example, while an aquifer underlying a particular set of structures might have a fairly uniform concentration at some depth, the main influence on a particular building might come from a portion of the aquifer located some distance from the building, but in which the contaminant concentration near the top of the aquifer is higher than it is right below the building.

For uniform soil diffusivity, if there is a building-source offset, the normalized subslab contaminant vapor concentration can be estimated as³⁹

$$\frac{c_{ss}}{c_{gw}} \approx \sqrt{\frac{d_f}{d_s}} e^{-\frac{d_h}{2d_s}\pi} \quad (6)$$

where d_f , d_s , and d_h are the building foundation depth, contaminant source depth, and horizontal building-effective source separation (lateral diffusion distance), respectively.

Equation 6 is consistent with 3-D simulation results as shown in Figure 9, where α is the indoor air concentration attenuation factor, and Q_s is the soil gas entry rate. In the scenario shown in Figure 9, $d_f = 2$ m, $d_s = 8$ m, and $d_h = 20$ m. Based on eq 6, $(c_{ss})/(c_{gw}) \approx 0.01$, which matches very well with the figure.

Recent studies^{40,41} have indicated that there has been overestimation of near-building soil gas concentration by

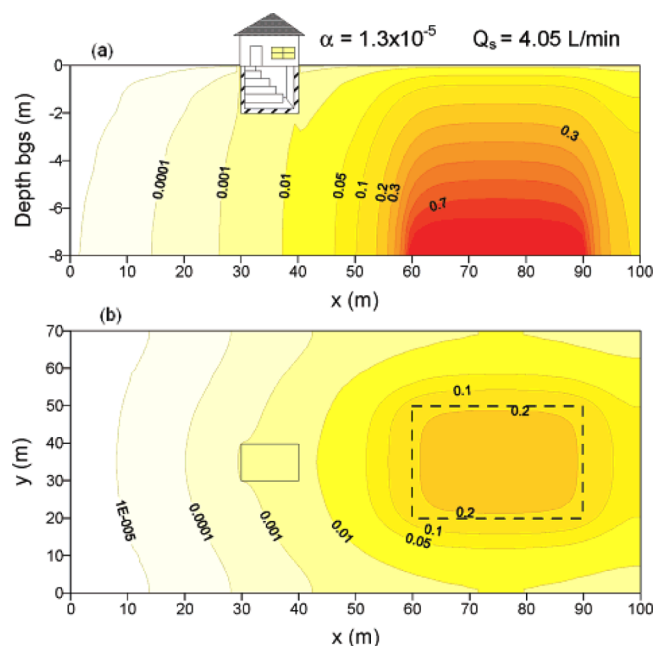


Figure 9. Simulated lateral diffusion effect on attenuation factor by Abreu and Johnson's 3-D model for nonbiodegradable contaminants.¹² Reprinted with permission from ref 12.

common vapor intrusion screening tools, and the failure to include lateral diffusion could be a possible explanation,³⁹ since both eq 6 and Figure 9 suggest that the contaminant vapor concentration decays exponentially with lateral diffusion distance (see also ref 42). This might also provide a further explanation for the orders of magnitude variation in groundwater-to-subslab contaminant concentration attenuation factor. For example, assume $(d_h)/(d_s)$ is uniformly distributed in a range of 0–4 for 90% of the measured $(c_{ss})/(c_{gw})$ values in the EPA database. For a $(d_f)/(d_s) = 0.25$ for basement and $(d_f)/(d_s) = 0.025$ for slab-on-grade, a distribution of AF can be obtained based on eq 6, and the results are compared with EPA database values, as shown in Figure 10.

Taken as a whole, the above results show that it is difficult to explain many order of magnitude variations in observed subslab

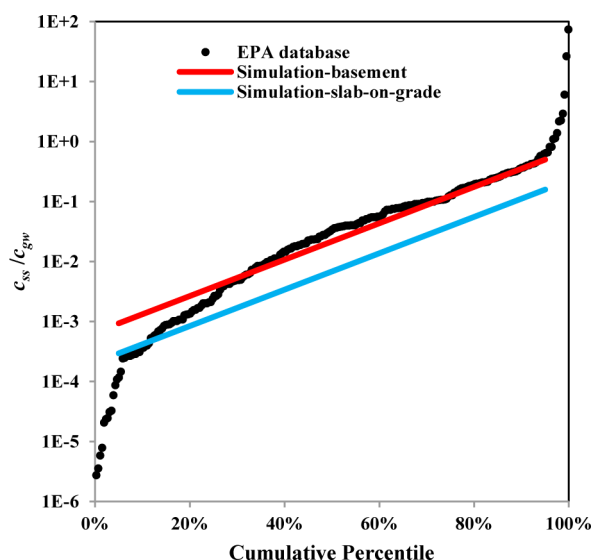


Figure 10. Cumulative probability plot of groundwater-to-subslab contaminant concentration attenuation factor based on simulated influence of lateral diffusion.¹

attenuation factors based upon most environmental factors, since these do not give rise to the sort of sensitivity needed to explain such large variation. It is not claimed that any one of the factors considered could not, in certain circumstances, be responsible for large variations. What the results do point to are factors, such as source displacement and inherent variability, as well as variations in capillary zone resistance as the factors that can make a significant, order of magnitude, difference in predictions.

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Notes

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The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This research was supported by Grant P42ES013660 from the National Institute of Environmental Health Sciences.

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