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SUBSURFACE TRANSPORT OF VOLATILE ORGANIC CONTAMINANTS INTO BUILDINGS

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Populations living near hazardous waste sites and landfills may be exposed to volatile organic compounds (VOCs) via several pathways including inhalation of contaminated outdoor air (1), ingestion of contaminated water (2), inhalation of contaminants which volatilize during residential water use (3, 4), and dermal sorption of contaminants while showering (5, 6). The subsurface transport of volatile contaminants into buildings located near contaminated sites has been considered as an additional route of exposure (7-14), but the overall impact of the pathway has not yet been placed in perspective. Indeed, there is reason to suspect (14) that subsurface soil-gas transport may be a dominant exposure pathway in some circumstances.

The flow of soil gas into buildings is driven by a pressure gradient that results from temperature differences between indoors and outdoors, wind loading on the building superstructure, and, in some instances, the operation of devices such as furnaces and exhaust fans. The small (several pascals) but persistent pressure difference that is established between the exterior and interior of the building can result in the infiltration of soil gas through penetrations in the building substructure. This advective transport of soil gas has been shown to be a dominant mechanism for radon entry into buildings (15). Contaminants present in soil gas may also diffuse into buildings through cracks or openings in the substructure, through permeable below-grade walls (9), or through other building materials.

Despite mounting evidence (reviewed in the following section) that transport of soil-gas into buildings is a potential pathway of exposure to VOCs for populations near contaminated sites and landfills, the significance of such exposures has not been evaluated quantitatively and our understanding of the pathway is rudimentary. The purpose of this paper is to provide order-of-magnitude estimates of the possible increase in indoor air pollutant concentrations in buildings near contaminated sites using current understanding of radon transport, existing measurements of VOCs in soil gas, an evaluation of some sources of subsurface contamination and simple transport models.

Experimental evidence

Available evidence suggests that transport of soil-gas contaminants into buildings can be an important pathway of exposure to VOCs. The results of several studies are summarized in this section.

Wood and Porter (7) conducted an investigation at the BKK landfill in Southern California. BKK is a Class I landfill which is permitted to accept hazardous waste. Methane was detected in enclosed spaces in nearby homes at concentrations approaching 1 percent (7,000 mg m^{-3}) and further detailed measurements suggested that chlorinated hydrocarbons had migrated into one of the houses located 180 m from the landfill. The migration of VOCs appeared to have resulted when a lowered water table opened a subsurface pathway through a permeable sandstone layer. Wood and Porter also conducted a survey of 20 Class II landfills. Despite the fact that these landfills are not permitted to accept hazardous wastes, they found landfill-gas concentrations of benzene and vinyl chloride in excess of 1ppm (~ 2 mg m^{-3}) in over half of the landfills surveyed with maximum concentrations of 100 mg m^{-3} benzene and 110 mg m^{-3} vinyl chloride.

In a study carried out in the Netherlands, Kliest et al. (8) found that 7 out of 77 homes located on contaminated soil had elevated concentrations of the same contaminants indoors and in the crawl space when compared to the average concentrations found in 200 reference homes. The average contaminant concentrations in the 'polluted' houses were increased by 250 to 8,000 times in the crawl space and by about 2 to 12 times in the living room relative to the reference homes. These relationships were found to be most influenced by the groundwater level, the crawl space ventilation rate and the type of floor (wood or concrete) present in the house.

Garbesi and Sextro (9) and Hodgson et al. (14) developed more specific evidence of soil-gas transport of VOCs in a pilot study conducted in a single, unoccupied house near a landfill. In that study, chlorinated hydrocarbons, freons and aromatic and aliphatic

hydrocarbons were found in the soil gases around the house at levels ranging up to about 1 mg m^{-3}. The same VOCs were found in cavities in the cement blocks of the basement walls at levels similar to those in the soil gases, and in the house at concentrations one to two orders of magnitude lower. When the basement of the house was artificially depressurized, an increase in the indoor concentrations of those VOCs present in the soil gas was observed. The indoor concentration of a tracer gas, which had been injected into the soil-gas on two sides of the house, also increased with increasing basement depressurization. The experiments were conducted in an empty house so that background levels of VOC were minimized. In this particular house, the contributions of contaminated soil gas would have accounted for more than three-quarters of the daily exposure to tetrachloroethylene (PCE), exceeding the combined exposures estimated for ingestion of water, inhalation in a shower and inhalation of outdoor air. It should be noted that subsurface ventilation pumps had been installed between the landfill and the house before the study began.

The California Air Resources Board (CARB) (10) reported sampling conducted by the South Coast Air Quality Management District (SCAQMD) in the vicinity of the BKK landfill in Southern California. A total of 500 air samples were taken at two outdoor sites and four indoor sites downwind of the landfill. All of the 120 samples that equaled or exceeded the state vinyl chloride standard of 10 ppb (0.025 mg m^{-3}) were taken inside the residences with a maximum indoor vinyl chloride concentration of 0.13 mg m^{-3}. During additional sampling at the OII landfill, the SCAQMD measured concentrations of vinyl chloride in some nearby homes from 0.02-0.3 mg m^{-3}. Since the maximum outdoor air concentration measured at the landfill was only 0.03 mg m^{-3}, atmospheric transport alone could not have accounted for the elevated indoor levels observed. In fact, grab samples of air taken from within water meter boxes at residences adjacent to the OII landfill revealed vinyl chloride concentrations ranging from 0.03-90 mg m^{-3}. The CARB report concluded

that for people residing near landfills, inhalation of indoor air may represent the most significant source of exposure to vinyl chloride.

Kullman and Hill (11) reported that gasoline contamination of indoor air at an office building was traced to three abandoned gasoline tanks buried beneath an adjacent building. Levels of benzene, toluene and xylene at 1,700, 3,800 and 7,500 mg m^{-3}, respectively, were found in the indoor air, with total gasoline hydrocarbons at 95,000 mg m^{-3}. Periodic gasoline odors had been reported over a period of several years with complaints of eye irritation, headache and nausea among some office workers. Even after the tanks had been removed, the contaminated soil beneath the building continued to be a source of occasional odors and complaints.

Finally, Moseley and Meyer (13) described the results of an air, soil-gas and groundwater monitoring survey that was undertaken following the discovery of explosive levels of gasoline vapors (the lower explosive limit (LEL) for gasoline is 1.3% by volume) in a house located approximately 50 m from a gasoline storage tank. Several cm of 'free product' found floating in nearby groundwater monitoring wells confirmed subsurface leakage from the tank. A contaminated groundwater plume was detected moving down the gradient in the water table accompanied by a plume of contaminated soil gas, lagging somewhat behind the groundwater plume. Four months after the initial discovery, the plumes reached a school located about 250 m from the tank, and some staff and students became ill from noxious indoor odors. At this point, the local fire department measured levels of airborne vapors in the school building of up to 40% of the LEL. The peak level of total hydrocarbons (THCs) measured in the soil-gas plume was 300 ppm (150 mg m^{-3} as C) with peak groundwater concentrations of benzene at 21 ppm (21,000 mg m^{-3}) and THCs at 300 ppm (300,000 mg m^{-3}). In further air sampling at the school, levels of THCs were measured at 1.3 mg m^{-3} as C in classrooms and 60 mg m^{-3} as C in the crawl space beneath the floor. Both residence and school were evacuated on discovery of the contamination.

Overview of contaminant sources and transport pathways

Figure 1 shows an idealized and schematic view of sources and transport pathways that may contribute to the contamination of indoor air with VOCs from the subsurface. Potential sources include volatile liquids such as gasoline or chlorinated solvents, contaminated landfill gas and contaminated groundwater. The VOCs introduced into the soil gas by these sources can be transported towards building substructures by means of advection under a pressure gradient or by diffusion if a concentration gradient exists. Other soil-gas transport mechanisms, such as flow induced by a gradient in buoyancy (16), appear to be of lesser importance and will not be considered here. On arrival at the zone of influence surrounding a building (see Figure 1), the VOCs may enter through cracks in the substructure either via advection or diffusion or a combination of these two mechanisms.

These potential sources and transport pathways are summarized schematically in Figure 2, and the parameters that are likely to have large influence over the transport mechanisms are indicated. A description of the symbols used to identify the parameters may be found in the glossary. Rectangular boxes represent the possible states that the VOCs may assume while arrows indicate transport pathways. The parameters of major significance for the various transport pathways are listed in ovals.

Evaluation of source strengths

Soil-gas surveys are increasingly used to locate the sources of subsurface contamination and to determine the extent of contamination. The results of four such surveys (17-20) are summarized in Table I. The studies reporting PCE and 1,1,1-trichloroethane (TCA) contamination were undertaken in residential/business areas while the measurements of benzene and toluene were made primarily at gasoline stations. These soil-gas concentrations may be compared with median levels of the same compounds reported for indoor air by Shah and Singh (21) and shown in Table II. The median indoor

air concentrations reported by Shah and Singh will be assumed to represent baseline indoor levels, and will serve as a point of reference for ascertaining whether indoor contamination arising from subsurface sources is significant. Both the average and the maximum soil-gas contaminant concentrations shown in Table I are many orders of magnitude higher than the baseline indoor air concentrations given in Table II, suggesting the potential for high indoor exposures should efficient transport pathways exist near contaminated sites.

The VOCs found in the field studies originated from spills or leaks of volatile organic liquids. However, as indicated in Figure 2, VOCs may also enter soil gas from other sources including landfills and contaminated groundwater. These potential sources are examined briefly in turn.

Chlorinated solvents. Liquid solvents such as PCE, TCA and trichloroethylene (TCE) that leak into permeable unsaturated soil will sink under the influence of gravity. Capillary (interfacial) forces will smear out the flow of liquid contaminant (22). Should a sufficient quantity of liquid reach a relatively impermeable and horizontal soil layer, it will form a pool. If the liquid is more dense than water, it will penetrate any unconfined groundwater aquifer it encounters. Wherever solvent is in contact with soil gas, it will vaporize, forming a boundary vapor concentration that drives diffusive transfer of the VOC into the soil gas. Therefore, the vapor pressure of a pure solvent determines the source concentration of contaminant in the soil gas (Table III).

Gasoline. Gasoline that passes into the unsaturated soil zone will behave in a similar fashion to the chlorinated solvents with the important distinction that gasoline is less dense than water and will spread out on the top of a water table (22). Gasoline is a complex mixture of VOCs and the vapor concentration at the liquid surface will depend on the liquid composition of the gasoline, which can vary widely. The more volatile components will escape more rapidly if the vapor is continually being removed, and the liquid composition will change with time. Therefore, predicting the vapor composition in equilibrium with a liquid gasoline spill is a more complex task than for pure solvents such

as PCE. However, an estimate of the vapor concentration in equilibrium with typical fresh gasoline (23) may be obtained by assuming that the partial pressure in the vapor phase equals the pure component vapor pressure multiplied by the liquid mole fraction (Raoult's law). Liquid gasoline concentrations of selected components and their corresponding vapor concentrations are given in Table IV.

Contaminated landfill gas. The major components of landfill gas are methane and carbon dioxide; both generated during anaerobic microbiological activity. The presence of VOCs in landfill gas may be due to one or more of the following processes: vaporization of organic liquids dumped in the landfill, volatilization from contaminated water, microbial action or chemical reaction. Average and maximum concentrations of VOCs in landfill gas taken from a survey of 20 class II landfills (24) are given in Table V. While depending on landfill construction and on the rate of gas generation, the pressure in a typical older municipal landfill is of the order of 1,500 Pa (25).

Contaminated groundwater. VOCs that are present in groundwater will volatilize into the soil gas. The gas-phase concentration at the groundwater surface that is in equilibrium with the water acts as a source and driving force for transport of VOCs through the soil gas. In a survey of 1,006 wells belonging to large water suppliers in California (26), 302 wells were found to contain detectable levels of TCA, TCE, PCE or carbon tetrachloride. The concentrations of the VOCs found in the contaminated wells are summarized in Table VI together with the concentrations which would be present in air immediately adjacent to the groundwater, assuming equilibrium predicted by Henry's law.

Transport models

A first-order estimate of the elevation in indoor VOC concentration resulting from subsurface contamination may be obtained by assuming that VOCs are transported into buildings in an analogous fashion to radon. However, the ratio of indoor concentration to source concentration is expected to be smaller for VOCs than for radon since radon

emanates from the soil immediately adjacent to the building while the VOCs will usually have to be transported over some distance before reaching the building. In addition, VOCs will be subject to sorption and desorption processes as they pass through the soil.

In this section the radon analogy will first be applied. Following that, a simple linear sorption isotherm will be described and then incorporated into transport models in the form of a retardation factor. The first transport model considers diffusion from a planar source located some distance from a building. Next, in a second diffusion based model, a building located in an extensive region of uniformly contaminated soil and soil gas is examined. Finally, a simple advective model is used to estimate the transport of VOCs from a landfill to a building. To simplify the three transport models, it is assumed that the VOCs will be swept into the building as fast as they arrive at the zone of influence (an assumption that is largely consistent with current understanding of radon entry) and that the concentration at the building perimeter is very much lower than the source concentration. Furthermore, the source concentration of the contaminant is assumed to be constant over time, and the soil medium is assumed to be isotropic and homogeneous.

Radon analogy. Using the mean indoor radon concentration found in the living space of single-family homes (27) and an estimate of the mean radon concentration in soil pores, a representative ratio of indoor radon concentration to soil-gas radon concentration is found to be $\alpha = C_{indoor}/C_{source} = 0.0016$ (see Appendix). This ratio varies widely across the housing stock. If it is assumed that VOCs behave in a similar fashion to radon, then the value of α (which can be thought of as an attenuation coefficient (12)) can be used as a first approximation of the likely indoor concentrations resulting from contaminated soil gas. Applying the radon ratio to the maximum measured soil-gas PCE and benzene concentrations in Table I gives indoor concentrations of about 20 mg m^{-3}. This value is over 2,000 times higher than the baseline indoor concentrations shown in Table II, justifying further attention to the problem.

Sorption models. The simplest model used to describe sorption between soil gas and soil assumes that the sorption equilibrium may be expressed as a linear isotherm:

$$q = K_d C \qquad (1)$$

where q is the mass of VOC sorbed per unit dry mass of soil, C is the gas-phase VOC concentration, and K_d is a partition coefficient. Ong and Lion (28) have shown that the sorption coefficient, K_d, for VOCs on soils that are close to field capacity in moisture content, is related to K_d^{sat}, the water saturated sorption coefficient, and H, the dimensionless Henry's law constant (concentration basis), by

$$K_d = \left(\frac{K_d^{sat}}{H}\right) + \left(\frac{\theta_m}{H}\right) \qquad (2)$$

where the activity coefficient of the VOC in water is assumed to be unity, and θ_m is the soil moisture content. The field capacity of soils is the moisture content that corresponds to their ability to retain moisture. Ong and Lion found that eq 2 successfully predicted K_d for TCE on Bandelier tuff for moisture contents greater than 4%. Note, however, that eq 2 substantially underpredicts K_d for oven dried soil or for soil dried in air at 68% relative humidity. In experiments with seven different soils, Ong and Lion found that K_d^{sat} for TCE varied between 0.06×10^{-3} and 3.3×10^{-3} m^3 kg^{-1}. The average value of 0.9×10^{-3} m^3 kg^{-1} will be used in this study as a representative sorption coefficient for TCE.

Diffusion models. The equation describing one-dimensional transient diffusion through sorbing unsaturated porous media is

$$\frac{\partial C}{\partial t} = \left(\frac{D_e}{\varepsilon_a R}\right)\frac{\partial^2 C}{\partial x^2} \qquad (3)$$

where t is time and x is distance. The effective diffusion coefficient D_e governs the rate of diffusion of VOCs through the soil gas and, according to Millington (29), is related to the diffusion coefficient of the VOC in air D_{air} and the soil porosity by

$$D_e = D_{air}\left(\frac{\varepsilon_a^{3.33}}{\varepsilon^2}\right) \qquad (4)$$

where $\varepsilon_a = \varepsilon - \theta_m \rho_b$ is the gas-filled porosity of the soil, ε is the total porosity, and ρ_b is the bulk density of the soil. Equation 4 has been verified experimentally by Karimi et al. (30) for diffusion of benzene through unsaturated soil. In eq 3, R is the retardation factor, or the ratio of the total amount of VOC to the amount present in the soil gas, and is defined as $R = 1 + \rho_b K_d/\varepsilon_a$. If no sorption occurs, $K_d = 0$, and R reduces to unity. The use of the retardation factor to account for sorption in the transport models assumes that the VOC in the soil gas is at all times in equilibrium with the sorbed fraction. This requires that the rate at which sorption equilibrium is established should be much faster than the rate at which contaminant is transported.

In the first diffusion model, a plane of contamination with gas-phase concentration C_{source} is introduced at a distance L from a building in an otherwise uncontaminated region of soil, as shown in Figure 3. This model is intended to approximate, for example, a layer of liquid solvent resting on an impermeable soil layer below a building, a layer of fresh gasoline on the water table, or contaminated groundwater itself. The areal extent of the source is assumed to be substantially greater than that of the building.

Once eq 3 has been solved for the appropriate initial and boundary conditions, the mass flux J entering the building is evaluated as

$$J = -D_e\left(\frac{\partial C}{\partial x}\Big|_{x=L}\right) \qquad (5)$$

where L represents the point of soil gas entry. The flux of VOC through the soil when multiplied by the appropriate cross-sectional area A gives the mass flow of VOC that enters the building. The resulting indoor concentration C_{indoor} may be estimated as the rate at which the mass of VOC enters the building divided by the volumetric flow rate of air through the building Q_b or

$$C_{indoor} = \frac{J\,A}{Q_b} \qquad (6)$$

A simple steady-state solution to eq 3 combined with eqs 5 and 6 results in

$$\alpha = \frac{C_{indoor}}{C_{source}} = \left(\frac{D_e A}{L Q_b}\right) \qquad (7)$$

The further the source is from the building, the longer it will take to establish steady-state conditions. A transient solution for the flux is therefore obtained by applying the boundary conditions $C(x=0, t > 0) = C_{source}$ and $C(x=L, t > 0) = 0$ and the initial condition $C(0 < x < L, t = 0) = 0$, to eq 3 to find (using eq 4.22 of ref 31)

$$J = \left(\frac{D_e C_{source}}{L}\right)(1 + 2 \sum_{n=1}^{\infty} (\cos(n\pi) \exp(-D_e n^2 \pi^2 t/R \varepsilon_a L^2)) \qquad (8)$$

Combining eqs 6 and 8 yields the ratio of indoor air concentration to source concentration as a function of time t and distance to source L, or

$$\alpha = \frac{C_{indoor}}{C_{source}} = \left(\frac{D_e A}{L Q_b}\right)(1 + 2 \sum_{n=1}^{\infty} (\cos(n\pi) \exp(-D_e n^2 \pi^2 t/R \varepsilon_a L^2)) \qquad (9)$$

At infinite time, the summation term in eq 9 reduces to zero yielding the steady-state solution of eq 7.

Typical transport and building parameters are listed in Table VII. As indicated, certain parameters are taken from Johnson and Ettinger (12) to facilitate comparison with that work. A detailed discussion of soil properties is presented in a recent review of radon transport from soil to air by Nazaroff (32). Eq 7 is used to calculate the steady-state value of α for source distances varying from 0.1 m to 100 m with results reported in Table VIII. The time taken to reach 90% of the steady-state value (τ_{ssc}) is calculated using eqs 7 and 9 and also reported in Table VIII. While the attenuation coefficient falls in a linear fashion from 0.03 to 0.00003 as the distance from the source increases, the time to steady state increases rapidly. For a retardation factor of 1, the time taken to reach steady-state is about

20 min at L = 0.1 m, but rises to 36 years for a source located 100 m from the building. Practically, this suggests that diffusive transport from subsurface sources at a distance of the order of 100 m or more from a building may not contribute to the contamination of indoor air. Sorption will delay the time taken to reach steady state by a factor equal to the retardation factor, which amounts to about 20 using the representative sorption coefficient for TCE. This shows that sorption can have a large impact on transport of VOCs through the unsaturated zone.

In the second diffusion model, a uniform source with gas-phase concentration C_{source} surrounds the building as shown in Figure 4. In this case, the model is intended to approximate a new house built in an extensive uniformly contaminated region. The VOC in the soil-gas is assumed to be in equilibrium with that sorbed on the soil. For simplicity, it is assumed that there is no transport of VOC from the ground surface to the surrounding ambient air. This model should therefore only be applied to situations which approximate these conditions such as might exist in paved environments. Once the house has been built, the VOCs in the zone of influence are swept into the building setting up a concentration gradient in the surrounding soil with a resulting flux of VOCs towards the building. The physical situation resembles diffusion into a region bounded internally by the sphere with radius $r = r_b$, where r_b is the equivalent radius of the zone of influence. Applying the boundary conditions $C(r<r_b, t > 0) = 0$ and $C(r\rightarrow\infty, t > 0) = C_{source}$ and initial condition $C(r_b < r < \infty, t = 0) = C_{source}$ to eq 3 modified for radial geometry (eq 6.1 in ref 31), a solution is obtained (using eq 6.60 in ref 31) as

$$\frac{C}{C_{source}} = 1 - \left(\frac{r_b}{r}\right) \mathrm{erfc}\left(\frac{r - r_b}{2\sqrt{(D_e\, t)/(R\, \varepsilon_a)}}\right) \qquad (10)$$

Combining the flux at $r=r_b$ obtained using eqs 5 and 10 with eq 6 yields the ratio of indoor air concentration to source concentration as

$$\alpha = \frac{C_{indoor}}{C_{source}} = \left(\frac{D_e}{r_b} + \sqrt{\frac{D_e\,\varepsilon_a\,R}{\pi\,t}}\right)\left(\frac{A}{Q_b}\right) \qquad (11)$$

The distance that the contaminant front has receeded from the building may be estimated as $r-r_b$, where r is the radius at which the concentration is equal to 0.9 C_{source}, and obtained from eq 10 by trial-and-error.

Using the transport and building related parameters in Table VII, and a value for r_b of 4.7m calculated as the radius of a hemisphere of surface area A, eq 11 gives the value of α as it changes with time with results reported in Table IX. The distance that the contaminant front has receeded as calculated from eq 10 is also given. The attenuation coefficient falls with time as the contaminant front slowly recedes from the building. In a situation where no sorption occurs, the attenuation coefficient varies from 0.004 after 1 day to 0.0007 after a period of 10 years. During the 10 year period, the contaminant front recedes by about 20 m. In contrast to the previous case, sorption results in higher levels of indoor contamination and a reduction in the rate at which the front recedes since the sorbed fraction acts as a reservoir of contaminant. These calculations illustrate how indoor air contamination which arises in such a case may persist for many years.

Advection model. The equation describing one-dimensional transient advection through sorbing unsaturated porous media is

$$\frac{\partial C}{\partial t} = -\left(\frac{v}{R\,\varepsilon_a}\right)\frac{\partial C}{\partial x} \qquad (12)$$

where all variables are as previously defined except v which is the Darcy velocity, given by

$$v = -\left(\frac{k}{\mu}\right)\left(\frac{dP}{dx}\right) \qquad (13)$$

where k is the soil permeability, μ is the dynamic viscosity of the soil gas and P is the pressure. Eq 13 applies only to viscous laminar flow in porous media.

For the advection model, a landfill with gas-phase concentration C_{source} is located a distance L from a building as shown in Figure 5. The ground surface between the building and the landfill is assumed to have a permeability which is much lower than the unsaturated zone. Practically, this could take the form of a thin surface clay layer or a paved surface. The landfill is at pressure P_s while the pressure at the building perimeter is P_b. The size of the landfill is assumed to be substantially greater than the width of the building as well as the distance to the building. The contaminant flux passing between landfill and building is $v\ C_{source}$ which, using eqs 6 and 13, gives the following steady-state solution

$$\alpha = \frac{C_{indoor}}{C_{source}} = \left(\frac{k}{\mu}\right)\left(\frac{P_s - P_b}{L}\right)\left(\frac{A}{Q_b}\right) \qquad (14)$$

Eq 14 is based on a crude estimate of the actual contaminant flux since the transport will not be entirely one-dimensional. The actual situation can be visualized by imagining the building to be a low pressure sphere and the landfill a high pressure plane at some distance from the sphere. The streamlines passing from the plane to the sphere will sweep out a broad path before converging near to the surface of the sphere. This implies that eq 14 underestimates the contaminant flux into the building. More accurate calculations are possible (see, for example, ref 33), but are not warranted for present purposes. The characteristic time taken for the pressure gradient between the landfill and the building to be established is (33, 34)

$$\tau_{ssp} \sim \left(\frac{\mu\ \varepsilon_a\ L^2}{k\ P_{atm}}\right) \qquad (15)$$

while the time to reach steady-state concentration is the time for the contaminant to travel from the landfill to the building, or

$$\tau_{ssc} \sim \left(\frac{L\ \varepsilon_a\ R}{v}\right) \qquad (16)$$

Under these idealized conditions, and providing that $\tau_{ssp} << \tau_{ssc}$, the attenuation coefficient should remain essentially zero until the steady-state conditions for concentration are met.

For the advection model, the attenuation coefficient is calculated using eq 14 and the appropriate parameters taken from Table VII. The landfill is assumed to be a distance of 100 m from the building, and the values of α for different soil permeabilities along with the times taken to reach steady state are reported in Table X. The attenuation coefficient falls as the permeability of the soil decreases and sorption delays the arrival of the contamination at the building. As k varies from 10^{-10} m^2 to 10^{-13} m^2, α ranges from 0.4 to 0.0004 with τ_{ssc} varying between 90 hrs and 10 yrs for R=1. The steady-state pressure gradient is established within about 1.5% of τ_{ssc} at R=1. Soils with permeability lower than 10^{-13} m^2 are unlikely to allow significant advective transport from a landfill at distances greater than about 100 m.

Johnson and Ettinger's model. The three transport models described above all use the assumption that the entire flux of VOC arriving at the zone of influence is swept into the building. To assess the impact of this assumption, consider the results of Johnson and Ettinger (12) who coupled steady-state diffusion from a planar source to the rate of infiltration into a building via both advection and diffusion. For a source located 10 m from the building, and for crack spaces in the building substructure amounting to 0.1% of the total subsurface area, the coupled model predicts upper and lower bounds for α of 0.0003 and 0.00002, respectively. Surprisingly, the range only spans an order of magnitude. The high value of 0.0003 corresponds to the condition for high advective soil-gas flow while the lower value of 0.00002 arises where the soil is so impermeable that there is no soil-gas flow and the VOCs enter the building via diffusion alone. The value of 0.0003 corresponds to, and is consistent with, the value for L = 10 m in Table VIII.

VOCs in indoor air from subsurface sources

Now, having evaluated soil-gas contaminant concentrations for the various sources and calculated a range of attenuation coefficients for different situations, the source concentrations can be multiplied by the attenuation coefficients to obtain order-of-

magnitude estimates of the likely ranges of indoor air contaminant concentration as shown in Table XI. The source strengths are taken from Tables III to VI. Ignoring the attenuation coefficients obtained for L = 0.1 m in Table VIII and at t = 1 day in Table IX, which are probably unrealistically high, the range of reasonable α values for the diffusion models is 0.00003 to 0.003. For the advection model, the likely range in α is taken as 0.0004 to 0.04, ignoring the value obtained in the case of highest permeability.

Table XI shows that the elevation in indoor air VOC concentrations attributable to subsurface contamination can be many orders of magnitude higher than typical baseline levels. Even if the models used to arrive at these estimates predict values that are too high by an order of magnitude, the results still give considerable cause for concern. For the representative conditions considered in this study, a liquid spill creates the greatest potential hazard for contaminant exposure through indoor air, while contaminated groundwater is of least concern. In addition, notice that for the gasoline, landfill and groundwater sources, only a single component has been considered, and the presence of other components will increase the overall contamination of the indoor air. Also, the landfill source has been considered at a distance of 100 m and in cases where a building is located directly on top of or adjacent to a landfill, the contamination could be one or two orders of magnitude higher than that shown in Table XI.

The health effects associated with living in houses nearby contaminated sites or landfills could be substantial. The upper limits of the estimated ranges of indoor concentrations for benzene and TCE are near or above the 8-hour threshold limit values for occupational exposures (35). Since the average person spends about 115 hours per week in the home (36), the exposure in houses close to strong sources could be almost three times greater than in occupational settings. Furthermore, more susceptible populations, such as children, the chronically ill and the elderly, are exposed in homes.

The cancer risks associated with indoor exposures to the benzene and TCE concentration ranges reported in Table XI are well above the generally accepted lifetime

risks of 10^{-6}, even under quite conservative exposure assumptions (36-38). For example, taking the lowest estimated indoor concentration of benzene (0.2 mg m^{-3}), the lifetime cancer risk for a person who spends 115 hours per week in a house over a period of 5 years amounts to 1×10^{-4}. Similarly, for the lowest estimated indoor concentration of TCE arising from a liquid spill (13 mg m^{-3}), the lifetime cancer risk for 5 years of exposure is estimated as 2.6×10^{-3}. For the higher predicted indoor concentrations and for longer exposure periods, the estimated risks increase in proportion both concentration and time.

Research directions

Contamination of indoor air by VOCs from the subsurface is of concern only in the relatively small fraction of houses located near contaminated sites or landfills. However, the high levels of contamination that have been measured in a few buildings, and that are predicted by the simple transport models, suggest that a clearer understanding of this transport pathway is urgently needed. Simple screening models for identifying buildings in which soil-gas infiltration may be a major pathway of contamination will be of considerable value. Such models could also allow estimates to be made of the size of the population subject to increased risks via this exposure pathway. The models presented here should provide a useful starting point in this regard. Caution is warranted when interpreting the results of these transport models as accepted models of radon entry into basements underpredict by almost an order of magnitude the observed radon entry rate (39).

Field studies at contaminated sites should be carried out to provide further evidence for the existence of this exposure pathway as well to improve the general understanding of the transport mechanisms involved. For example, the role of weather may be most rapidly elucidated via this research approach. Field studies can also provide a basis for testing the usefulness of various screening models, which in turn can suggest suitable methods to control the subsurface infiltration of VOCs into buildings. Existing techniques for the

mitigation and control of radon entry (40) are a useful starting point for research into the prevention of VOC entry.

Laboratory studies will be of most benefit in investigating specific aspects of the soil-gas transport pathway. For example, the transport of VOCs through unsaturated soil media needs to be more clearly understood. In particular, further research is required on multi-component sorption and transport, and the influence of soil moisture on soil sorption coefficients in the unsaturated zone at low moisture levels. An additional research requirement is to investigate the microbiological decay and/or generation of VOCs as they pass through the unsaturated zone. Depending on the rate of transport, these biotic mechanisms could have a large impact on the concentration of VOCs arriving at the building zone of influence.

Conclusions

This work has shown that subsurface transport of volatile contaminants into buildings near contaminated sites and landfills can result in levels of indoor air contamination that are many orders of magnitude higher than typical baseline levels. The estimated risks associated with the predicted indoor concentrations are in turn, orders of magnitude above acceptable levels. The transport models used are necessarily based on highly idealized representations of reality and are only intended to give first order estimates of indoor contamination levels. Indeed, these models should only be used with a clear understanding of the simplifying assumptions on which they are based. Nevertheless, the predicted risks are sufficiently high to justify further urgent attention to this exposure pathway.

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Appendix: Attenuation coefficient for radon entry from soil

The mean indoor radon concentration in the living space of single-family dwellings in the United States is approximately 55 Bq m^{-3} (27, 41). The dominant source of this radon is soil adjacent to the building substructure. The maximum or undepleted radon concentration in soil pores C_{source} is related to primary factors as follows

$$C_{source} = f \rho_s A_{Ra} \left(\frac{1 - \varepsilon}{\varepsilon} \right) \qquad \text{(A1)}$$

where the parameters can be estimated from data compiled by Nazaroff and co-workers (15, 32). Data on the radium content of surface soils A_{Ra} indicate a typical range of 10-100 Bq kg^{-1} with 40 Bq kg^{-1} constituting a reasonable estimate of the mean. The emanation coefficient f of ^{222}Rn has been observed to range from 0.05 to 0.7, and the geometric mean value of these limits, 0.2, can be taken as a baseline estimate. Typical values for the soil grain density ρ_s and total porosity ε are listed in Table VII. Combining these values yields C_{source} = 35,000 Bq m^{-3}, a number that is consistent with measured data. The resulting representative ratio C_{indoor}/C_{source} is therefore 55/35,000 = 0.0016.

The building substructure may be expected to influence the rate of soil-gas entry. Cohen (42) reported annual average indoor radon measurements in the living space of houses in the United States as 59, 48 and 47 Bq m^{-3} respectively, for basement, slab-on-grade and crawl space substructures. This suggests that the type of substructure has a relatively small influence on the rate of soil-gas entry.

Glossary

A	effective contaminant flux area (m^2)
A_{Ra}	radium activity concentration of soil (kg^{-1} s^{-1})
C	gas-phase concentration of contaminant (mg m^{-3})

$C_{baseline}$	baseline concentration of contaminant in indoor air (mg m^{-3})
C_{indoor}	concentration of contaminant in indoor air (mg m^{-3})
C_{source}	gas-phase contaminant source concentration (mg m^{-3})
D_{air}	diffusion coefficient for contaminant in air (m^2 s^{-1})
D_e	effective diffusion coefficient for contaminant in soil gas (m^2 s^{-1})
f	emanation coefficient of radon from soil (dimensionless)
H	Henry's law constant (dimensionless, concentration basis)
J	Mass flux rate (mg m^{-2} s^{-1})
k	permeability of soil to soil gas (m^2)
K_d	air to unsaturated soil partition coefficient (m^3 kg^{-1})
K_d^{sat}	air to water-saturated soil partition coefficient (m^3 kg^{-1})
L	distance from contaminant source to building (m)
P	dynamic gas pressure (kPa) (Pa ≡ kg m^{-1} s^{-2})
P_{atm}	ambient air pressure (kPa) (Pa ≡ kg m^{-1} s^{-2})
P_b	dynamic soil-gas pressure at building (kPa) (Pa ≡ kg m^{-1} s^{-2})
P_s	dynamic gas pressure in landfill (kPa) (Pa ≡ kg m^{-1} s^{-2})
P_{vap}	vapor pressure (kPa) (Pa ≡ kg m^{-1} s^{-2})
q	concentration of contaminant sorbed to soil (kg kg^{-1})
Q_b	volumetric air flow rate through building (m^3 s^{-1})
r	radial distance (m)
r_b	effective radius of zone of influence (m)
R	retardation factor
t	time (s)
T	temperature (^oC)
v	Darcy velocity (m s^{-1})
x	linear distance (m)
α	attenuation coefficient (dimensionless)

ε	total soil porosity (dimensionless)
ε_a	gas-filled soil porosity (dimensionless)
μ	dynamic viscosity of soil gas (kg m^{-1} s^{-1})
θ_m	soil moisture content (m^3 kg^{-1})
ρ_b	bulk density of soil (kg m^{-3})
ρ_s	density of soil grains (kg m^{-3})
τ_{ssc}	characteristic time to establish steady state concentration profile (s)
τ_{ssp}	characteristic time to establish steady state pressure profile (s)

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Table I. Measured concentrations of VOCs in soil-gas at selected contaminated sites.

VOC	Location (reference)	Number of samples	Average concentration (mg m^{-3})	Maximum concentration (mg m^{-3})
TCA	Vancouver, WA (19)	14	3.5	19†
PCE	Yakima, WA (18)	113	1.1	18
PCE	Vancouver, WA (19)	18	2.5	11†
PCE	Grand Island, NE (17)	28	460	11,000
Benzene	Bay Area, CA (20)	135	920	14,000
Toluene	Bay Area, CA (20)	147	2,900	42,000

†Capacity of sampling device was exceeded; actual value may be higher.

Table II. Median concentrations of VOCs in indoor air database (21).

VOC	Number of data points	Median concentration (mg m^{-3})
TCA	2,120	0.01
TCE	2,132	0.0007
PCE	2,195	0.005
Benzene	2,128	0.01

Table III. Vapor concentration in equilibrium with pure organic liquids (43).

VOC	Vapor pressure (20°C) (kPa)	Vapor concentration (mg m^{-3})
TCA	13	730,000
TCE	7.7	420,000
PCE	1.9	130,000

Table IV. Typical fresh gasoline liquid component concentrations (23) and estimated equilibrium vapor concentrations.

VOC	Mole fraction in liquid	Vapor pressure (20°C) (kPa)	Vapor concentration (mg m^{-3})
Benzene	0.022	9.8	7,000
Toluene	0.103	2.8	11,000
Xylenes†	0.078	0.79	2,700

†Vapor pressure taken as average value for all three isomers.

Table V. Measured concentrations of VOCs in Class II landfill gas (24).

VOC	Average concentration (mg m^{-3})	Maximum concentration (mg m^{-3})
Methane	350,000 (53% by vol)	390,000 (58% by vol)
Vinyl chloride	19	12
Methylene chloride	67	800
PCE	66	1,300
Benzene	10	170

Table VI. Measured concentrations of VOCs in groundwater supplies (26) and estimated equilibrium concentration in air (Henry's constants from ref 44).

VOC	Average groundwater concentration ($mg\ m^{-3}$)	Maximum groundwater concentration ($mg\ m^{-3}$)	Henry's constant (20°C)	Average air concentration ($mg\ m^{-3}$)	Maximum air concentration ($mg\ m^{-3}$)
CCl_4	5.3	29	0.88	4.7	26
TCA	9.1	100	0.57	5.2	57
PCE	6.9	170	0.54	3.7	92
TCE	23	540	0.32	7.4	170

Table VII. Typical transport and building parameters.

Parameter	Symbol	Value or Range	Reference
Diffusivity in air (TCE at 20°C)	D_{air}	$8.4 \times 10^{-6}\ m^2\ s^{-1}$	44
Henry's constant (TCE at 20°C)	H	0.32	44
Satd. partition coefficient (TCE at 20°C)	K_d^{sat}	$8.9 \times 10^{-4}\ m^3\ kg^{-1}$	28
Total soil porosity	ε	0.38	12
Soil moisture content	θ_m	$7 \times 10^{-5}\ m^3\ kg^{-1}$	12
Bulk density of soil	ρ_b	$1{,}700\ kg\ m^{-3}$	12
Permeability of soil to soil gas	k	10^{-10} to $10^{-13}\ m^2$	32
Dynamic viscosity of soil gas (air at 20°C)	μ	$1.8 \times 10^{-5}\ kg\ m^{-1}\ s^{-1}$	43
Ambient air pressure	P_{atm}	101 kPa	-
Dynamic landfill-gas pressure	P_s	1.5 kPa	25
Dynamic pressure at building perimeter	P_b	0	-
Ventilation rate of building	Q_b	$105\ m^3\ h^{-1}$	12
Effective contaminant flux area	A	$138\ m^2$	12
Effective radius of zone of influence	r_b	4.7 m	-

Table VIII. Attenuation coefficients and time to steady-state: diffusion from planar source.

	Distance from source (m)			
	0.1	1	10	100
α	0.03	0.003	0.0003	0.00003
τ_{ssc} (R=1)	20 min	32 h	140 d	36 y
τ_{ssc} (R=20)	7 h	27 d	7 y	720 y

Table IX. Attenuation coefficients and distance to front: diffusion from uniform source.

	Time			
	1 d	30 d	1 y	10 y
α (R=1)	0.004	0.001	0.0009	0.0007
Dist to front (m)	1	5	12	22
α (R=20)	0.02	0.004	0.002	0.001
Dist to front (m)	0.2	1	4	10

Table X. Attenuation coefficients and time to steady-state: advection from landfill (100m).

	Permeability (m^2)			
	10^{-10}	10^{-11}	10^{-12}	10^{-13}
α	0.4	0.04	0.004	0.0004
τ_{ssp}	1 h	10 h	5 d	50 d
τ_{ssc} (R=1)	90 h	40 d	1 y	10 y
τ_{ssc} (R=20)	70 d	2 y	20 y	200 y

Table XI. Potential ratio of indoor air VOC concentration at contaminated site relative to baseline levels (α = 0.00003 - 0.003).

Source	C_{source} (mg m^{-3})	C_{indoor}† (mg m^{-3})	$C_{baseline}$ (mg m^{-3})	$\frac{C_{indoor}}{C_{baseline}}$
Pure solvent (TCE)	420,000	13 - 1,300	0.0007	20,000 - 2,000,000
Landfill‡ (PCE)	1,300	0.5 - 50	0.005	100 - 10,000
Gasoline (Benzene)	7,000	0.2 - 20	0.01	20 - 2,000
Groundwater (TCE)	170	0.005 - 0.5	0.0007	7 - 700

†Calculated as α C_{source}

‡α = 0.0004 - 0.04 for a landfill at 100 m.

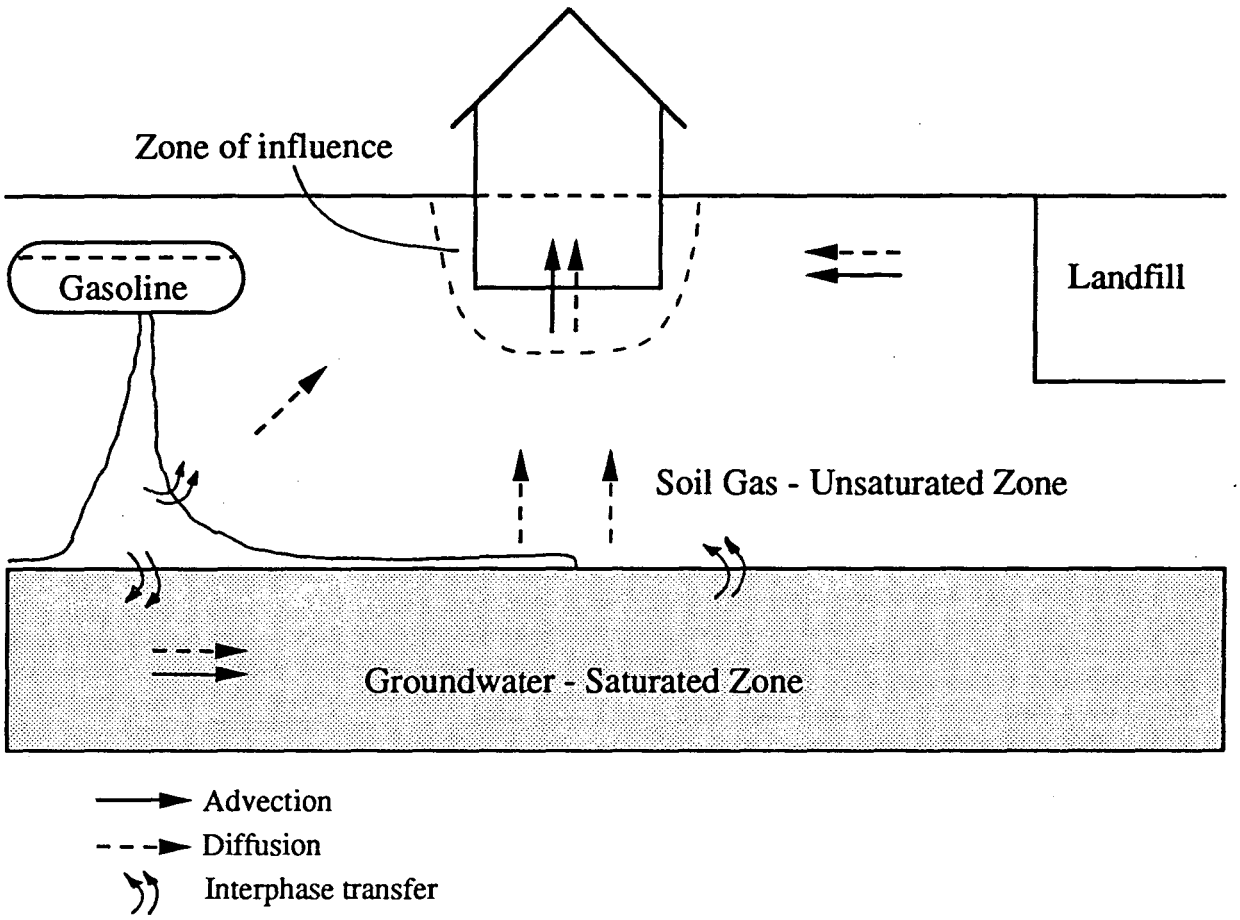

Figure 1. Idealized schematic representation of potential sources and transport pathways that could lead to contamination of indoor air with volatile organic compounds. The zone of influence surrounding the building is the region in which the building influences the movement of soil gas.

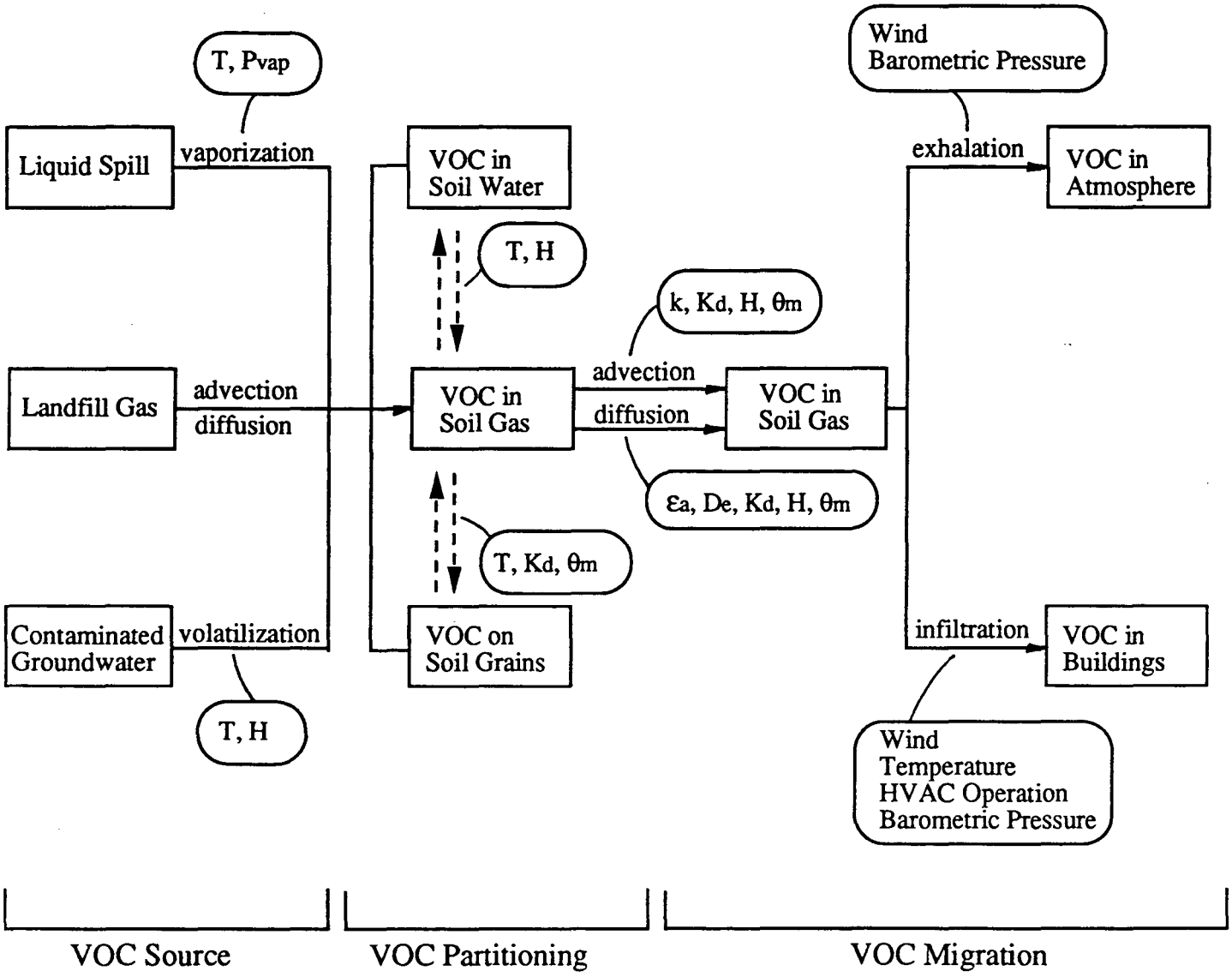

Figure 2. Schematic representation of VOC sources and transport pathways in the unsaturated soil zone. Parameters that influence source strength, partitioning and transport are shown in ovals and are defined in the glossary. HVAC refers to heating, ventilation and air-conditioning systems.

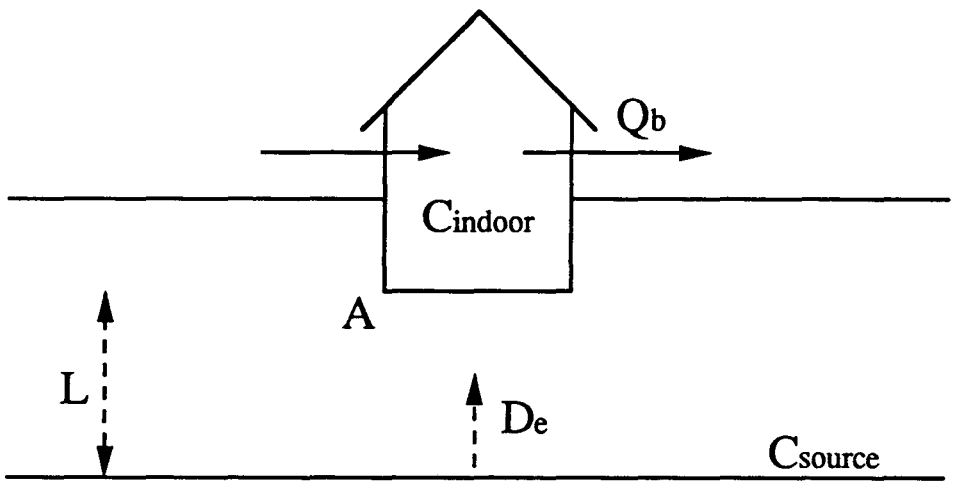

Figure 3. Schematic representation of the diffusion of VOCs from a planar source of contamination into a building.

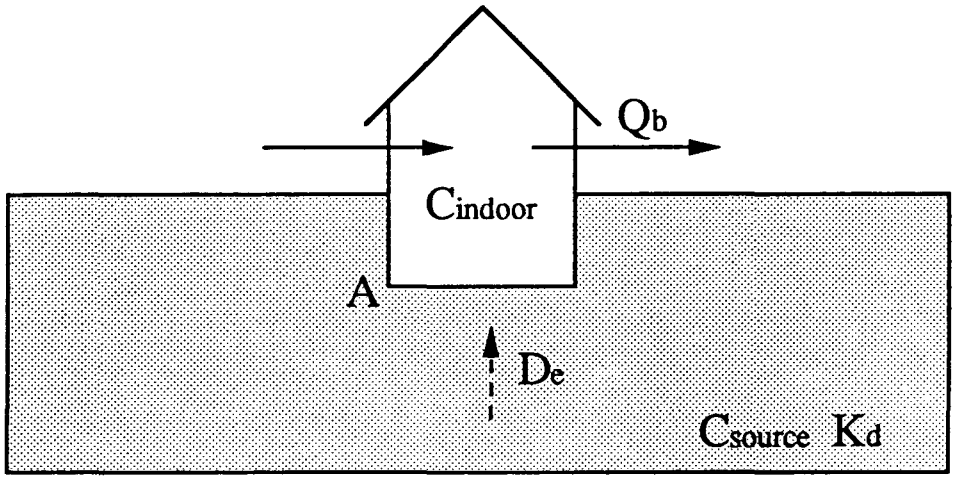

Figure 4. Schematic representation of the diffusion of VOCs from a uniform source of contamination into a building.

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