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Research article

Recent advances in vapor intrusion site investigations

Thomas McHugh [a, *], Per Loll [b], Bart Eklund [c]

[a] GSI Environmental Inc., Houston, TX, United States
[b] DMR A/S, Jerslev, Denmark
[c] AECOM, Austin, TX, United States

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ABSTRACT

Our understanding of vapor intrusion has evolved rapidly since the discovery of the first high profile vapor intrusion sites in the late 1990s and early 2000s. Research efforts and field investigations have improved our understanding of vapor intrusion processes including the role of preferential pathways and natural barriers to vapor intrusion. This review paper addresses recent developments in the regulatory framework and conceptual model for vapor intrusion. In addition, a number of innovative investigation methods are discussed.

1. Introduction

Vapor intrusion is the vapor phase migration of volatile contaminants from a subsurface source into overlying buildings or other structures. Vapor intrusion has been recognized as a potential exposure pathway at contaminated sites for decades, however, before the year 2000, few regulatory guidance documents provided comprehensive recommendations for field investigation of this pathway. Following the discovery of vapor intrusion problems at a small number of sites in the late 1990s and early 2000s, this exposure pathway has received more attention in regulatory guidance and among the regulated community. As a result, our understanding of the vapor intrusion pathway has evolved rapidly since 2000.

Previously published guidance documents provide a general review of vapor intrusion and investigation methods (e.g., ITRC, 2007; USEPA, 2015a). This paper focuses on recent developments in vapor intrusion with a specific focus on recent developments to i) regulatory framework, ii) conceptual model, and iii) investigation approaches. This paper focuses on chlorinated volatile organic compounds (VOCs). There is an extensive literature related to potential vapor intrusion of petroleum hydrocarbons, methane, and radon that is largely outside the scope of this paper. Until recently, vapor intrusion for chlorinated VOCs and petroleum VOCs were addressed in a similar manner. Recently, separate guidance has been developed for petroleum VOCs (USEPA, 2015b; ITRC, 2014) because they often rapidly biodegrade in the vadose zone greatly reducing the vapor intrusion risk (McHugh et al., 2010; USEPA, 2012a).

2. Regulatory framework

Previous reviews of vapor intrusion guidance outside the United States found that where the vapor intrusion pathway was being addressed, the usual approach relied upon numerical modeling and risk assessment (Ferguson, 1999; Eklund, 2007). This largely continues to be the case. Field investigations to evaluate vapor intrusion are most common in only a few countries: Australia, Canada, Denmark, the United Kingdom, and the United States. As shown in Supplemental Material, Table S1, these countries have all issued guidance documents in the last decade to take into account recent developments in site characterization methods, data evaluation techniques, and site decision-making.

The United States Environmental Protection Agency (USEPA) issued a draft vapor intrusion guidance in 2002 with the intent to update and finalize the guidance within a few years. However, the

* Corresponding author. GSI Environmental Inc., 2211 Norfolk, Suite 1000, Houston, TX, United States.
E-mail address: temchugh@gsi-net.com (T. McHugh).

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USEPA did not issue final guidance for addressing vapor intrusion at non-petroleum (USEPA, 2015a) and petroleum (USEPA, 2015b) sites until 2015. For non-petroleum sites, the USEPA guidance recommends very conservative screening criteria and intensive sampling for sites where VOC concentrations exceed these criteria. Recommended sampling includes groundwater, soil gas and indoor air with multiple rounds of sampling recommended to characterize temporal variability. The guidance recommends a multiple-lines-of evidence approach for evaluating the investigation results and sets a high bar for concluding an absence of vapor intrusion concern. Numerous state governments in the United States issued their own vapor intrusion screening levels and guidance. These guidance documents have been previously summarized (Eklund et al., 2007; Eklund et al., 2012). Compared to the USEPA guidance, the state guidance documents typically provide more detail regarding the specific procedures to be used for collection and analysis of soil gas and indoor air samples. In Canada, there is guidance at both the Federal and Provincial level. In addition to the Federal guidance listed in Table S1, there also are guidance documents for specific provinces (e.g., Alberta, Ontario, British Columbia, Atlantic provinces). In Europe (outside of the United Kingdom and Denmark), vapor intrusion is primarily addressed through modeling and risk assessment (NICOLE, 2004). In most of these countries, there is little guidance for how to proceed at sites that fail this screening and show a potentially unacceptable risk from vapor intrusion. In addition to establishing regulatory frameworks, researchers in Australia, Canada, Denmark, and the United States have been active in developing investigation methods and elucidating vapor intrusion processes.

In other countries outside of North America, Europe, and Australia, there is little or no regulatory guidance on vapor intrusion. Vapor intrusion is addressed in New Zealand on a case-by-case basis, but no new guidance has been issued in the last 15 years. NICOLE Brasil, an industry-led non-governmental organization, issued vapor intrusion guidance that draws heavily upon practices used in the United States. The guide presents a conceptual model for vapor intrusion and covers investigation methods and modeling but does not recommend default screening values or attenuation factors. Malaysia has issued guidance for addressing vapor intrusion that provides screening levels for residential and industrial indoor air, guidance for soil vapor measurement, and a tiered risk assessment process. However, the guide does not recommend follow-up actions for sites where the risk assessment process indicates a potential vapor intrusion concern. In South Africa, the industry-led NICOLA group has a Working Group that is developing vapor intrusion guidance.

A number of multi-national companies have company-specific risk management policies that include evaluation of the vapor intrusion pathway even in countries without established guidance. In addition, government agencies such as the U.S. Department of Defense have evaluated vapor intrusion at some overseas installations. These parties commonly utilize vapor intrusion guidance and investigation practices from their home countries. In many of these cases, these investigations require shipment of equipment and return of samples to the United States or Europe for analysis.

3. Vapor intrusion conceptual model

The standard conceptual model for vapor intrusion consists of i) partitioning from groundwater or soil into soil gas, ii) diffusion through the vadose zone from the source area to the immediate vicinity of the building and iii) advection and/or diffusion through the building foundation (USEPA, 2015a, Fig. 1). Once vapors have

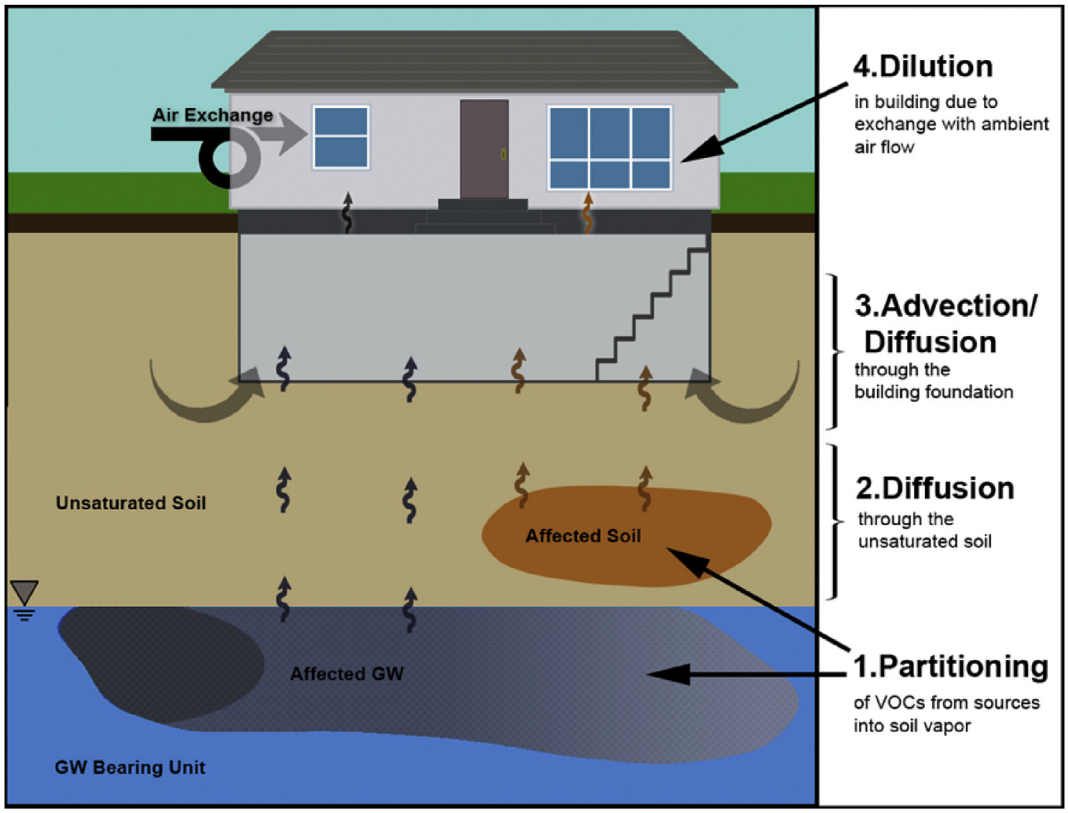

Fig. 1. Standard conceptual model for vapor intrusion.

entered a building, the indoor air containing these vapors is usually assumed to be reasonably well-mixed for a given floor or level of a building (USEPA, 2015a). This standard conceptual model can be used to explain vapor intrusion observed at many sites where the locations of the impacted buildings correlate well with the observed groundwater plume. Two well-studied examples from the United States are the Raymark site in Connecticut (USEPA, 2006) and the Redfield Rifle site in Colorado (Folkes et al., 2009). However, at many sites, the conceptual site model (CSM) needs to incorporate various site-specific features. If preferential pathways or physical barriers to vapor intrusion are present, the CSM needs to incorporate these features to explain the presence or absence of vapor intrusion.

3.1. Preferential pathways

A preferential pathway is typically defined as a high permeability conduit that can serve as a high-capacity transport pathway for VOC vapors from the source area to or into a building. For example, a sewer line can serve as a preferential pathway connecting an area of contaminated groundwater to a building. Most buildings contain some penetrations through the building foundation such as cracks, expansion joints, and plumbing penetrations that can serve as conduits for air flow through the foundation. Because these features are common to most buildings and they do not extend beyond the building foundation, they are considered to be potential entry points for vapors but not "preferential pathways" for vapor intrusion (ITRC, 2007). In other words, except for sources located directly adjacent to the building foundation, penetrations through the building foundation do not provide a continuous high capacity connection from the source area to the interior of the building. Nonetheless, high permeability building features can have an important impact on the entry into and/or the distribution of VOCs within the building. For example, VOCs may migrate within wall cavities, elevator shafts, stairwells, or open attic spaces. Such transport pathways can result in an unexpected distribution of VOCs within the building such as high VOC concentrations in building rooms or floors not directly adjacent to the VOC source. This type of transport is more likely to occur in older multi-level buildings.

Exterior preferential pathways for vapor intrusion must connect to a building and extend some distance away from the building towards the source. For example, dry wells or cisterns can act as vertical preferential pathways enhancing the transport of VOC vapors into a building from a groundwater source located below the building. In addition, sewers or utility tunnels can serve as preferential pathways for vertical and lateral migration of VOC vapors (Fig. 2). Sewers or utility tunnels are of greatest concern when they pass directly through contaminated groundwater or vadose zone non-aqueous phase liquid (NAPL) sources. When sewers pass through contaminated groundwater, this groundwater may infiltrate into the sewer allowing partitioning into the gas phase to occur within the sewer line. In these cases, vapor intrusion impacts may occur in buildings laterally offset from the groundwater plume but connected to the sewer line running through the plume (Fig. 2). Examples of sites with sewer preferential pathways are provided in Table S2. In Denmark, it is estimated that, at approximately 20% of vapor intrusion sites, sewer lines are either the primary pathway or a significant contributing pathway for VOC transport to buildings (Hvidberg, 2016). Sewer lines may facilitate migration of VOC vapors directly into the building or may result in the release of vapors below the building foundation (Fig. 2). In cases where the sewer line is facilitating transport directly into the building, sub-slab depressurization may not be an effective mitigation method.

Preferential pathways are problematic when they are not recognized and/or cannot be addressed by standard site investigation and mitigation measures. At most of the well documented sites with sewer line preferential pathways, the importance of the sewer line was identified only after obtaining investigation results that could not be explained by the standard vapor intrusion conceptual model (e.g., Guo et al., 2015) or, in some cases, after standard mitigation measures failed to control the vapor intrusion.

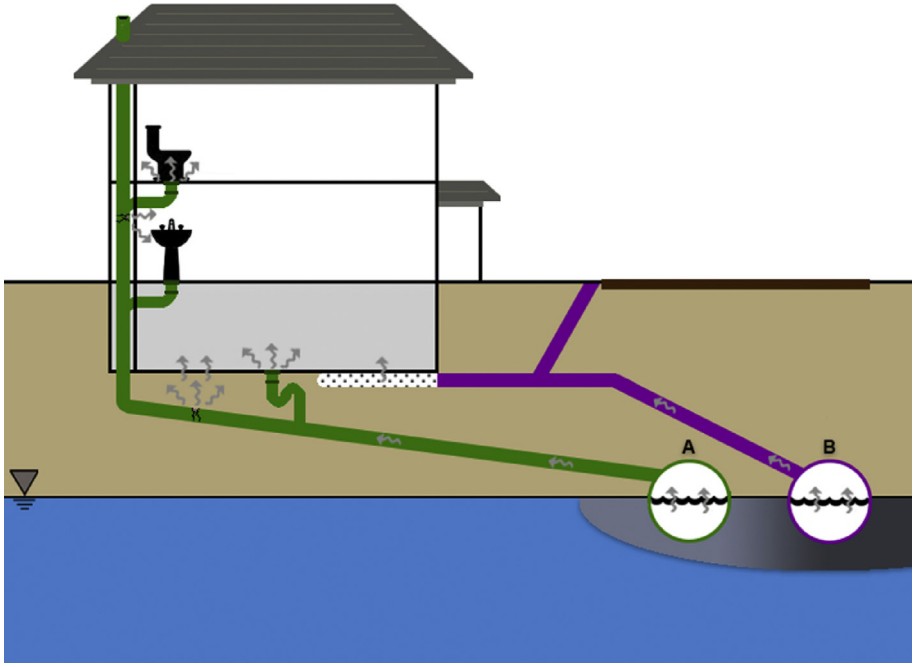

Fig. 2. Simplified Conceptual Model for Sewer Preferential Pathway Vapor Intrusion: A) Sanitary sewer line, B) Storm sewer or land drain system connected to building foundation drain (not applicable for some buildings). In some older sewer systems, sanitary and storm water flow through a combined sewer system.

Some guidance documents suggest that naturally occurring high permeability zones in soil may act as preferential pathways for vapor intrusion (e.g., gravel layers or vertically fractured rock; ITRC, 2007; USEPA, 2015a). Similarly, some guidance documents suggest the potential for preferential vapor migration within permeable backfill around utility lines (e.g., NJDEP, 2016; CalEPA, 2011). However, we are aware of few examples where migration of vapors through backfill or naturally-occurring high permeability zones has been documented to be important. This suggests either that i) this type of transport is rare or ii) when this pathway occurs, it is adequately addressed by standard vapor intrusion investigation and mitigation measures.

3.2. Barriers to vapor intrusion

Vapor intrusion impacts have not been observed at some sites with VOC concentrations in groundwater far above pathway-specific screening values (e.g., Pennell et al., 2016). At these sites, specific barriers to vapor intrusion may be preventing migration of VOCs into the building. These barriers can include: a clean water lens at the top of the water-bearing unit, a saturated clay or silt layer above the VOC source (McHugh et al., 2013), biodegradation or other transformations within the vadose zone, an impermeable layer or ventilated air gap in the building foundation, or positive building pressure (Fig. 3). Aerobic biodegradation in the vadose zone is well recognized to limit the potential for vapor intrusion of petroleum VOCs (USEPA, 2012a). Aerobic biodegradation can also limit the potential for vapor intrusion for some of the less chlorinated VOCs such as vinyl chloride (Patterson et al., 2013) and, at some sites, cis-1,2-dichloroethene (Schmidt et al., 2010).

4. Innovative investigation approaches

Most vapor intrusion guidance documents recommend a multiple-line-of-evidence approach for evaluating the vapor intrusion pathway (ITRC, 2007; USEPA, 2015a). Using this approach, no single investigation result is considered definitive; rather all available results are evaluated to determine whether the pathway is likely to be complete or not. Although a wide variety of investigation methods may be included in the multiple-lines-of-evidence approach, VOC concentrations in groundwater, soil gas, and indoor air typically constitute the primary lines of evidence considered (ITRC, 2007). When VOCs are detected in one or more of these media at concentrations exceeding screening levels, the key challenges to evaluating the pathway include: spatial variability in VOC concentrations, temporal variability in VOC concentrations, and other sources of VOCs (i.e., indoor and ambient sources). In recent years, a number of innovative investigation approaches have been developed or improved in order to address these challenges. The more promising or more widely-used approaches are discussed below.

4.1. Passive samplers

Although evacuated stainless-steel canisters are most commonly used for the collection of soil gas and indoor air samples in the United States and Canada, sorbent samplers are widely used in other parts of the world. Attainment of accurate results using passive samplers requires selection of a proper sampler and sorbent combination to avoid problems of starvation, poor retention, and poor recovery. However, for sampling of indoor and ambient air, these issues and the solutions are generally well understood

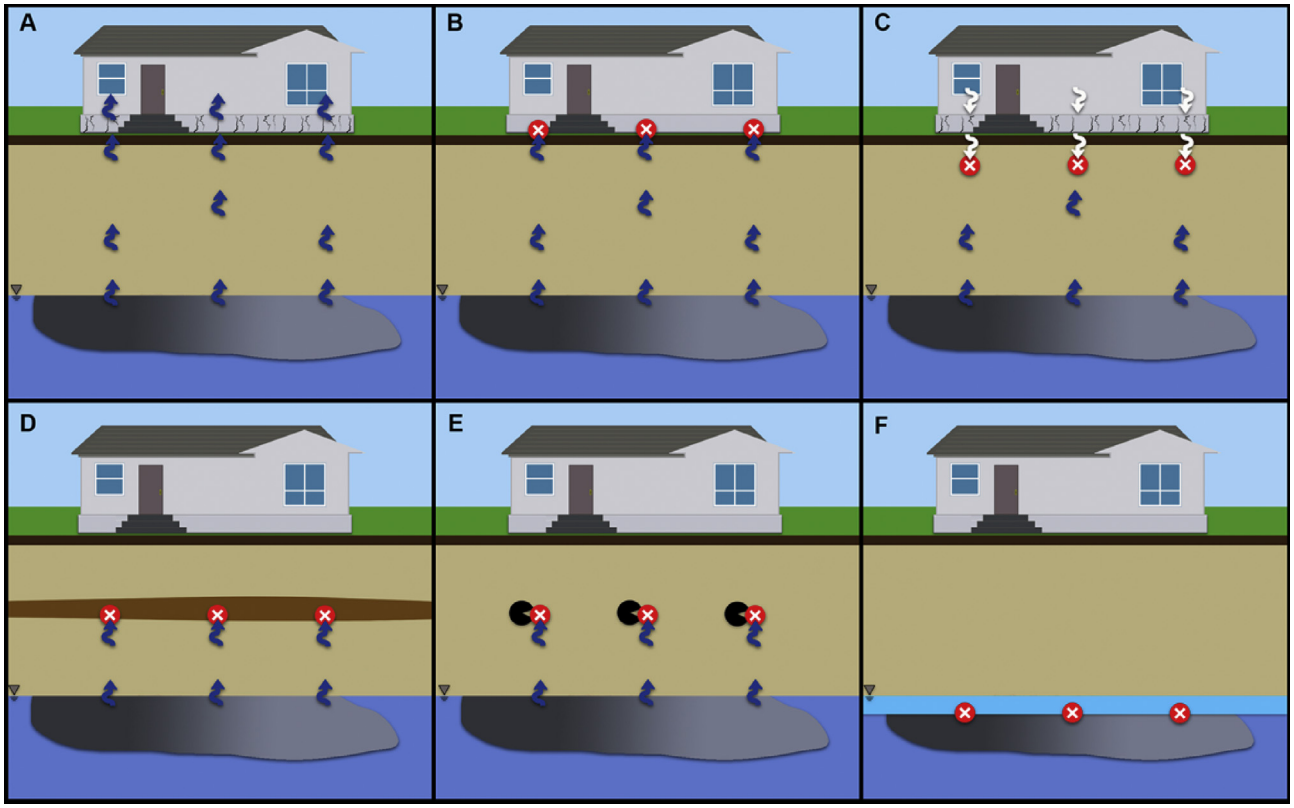

Fig. 3. Potential Barriers to Vapor Intrusion: A) No barriers, B) Impermeable building foundation, C) Positive building differential pressure, D) Saturated clay/silt layer between vapor source and building, E) Biodegradation of vapors in the vadose zone, F) Clean water lens at the top of the water table.

(USEPA, 2014). Active and passive sorbent-based methods are commonly employed in Europe where evacuated canisters are typically not available locally. In Denmark, for example, passive sorbent sampling has been the default method to measure VOCs in indoor air since the late 1990s. A mix of passive sorbent and whole air sampling approaches have been used in Australia and New Zealand, with a trend towards developing in-country analytical options in recent years for canister samples.

Passive sorbent samplers have long been used in industrial settings to monitor worker exposure, but they have less commonly been used to measure VOC concentrations for vapor intrusion investigations in the United States. Compared to evacuated stainless-steel canisters, passive sorbent samplers are considerably smaller, can be handled more easily, and can easily be adapted to sample collection times of greater than 24 h. Longer sample collection times reduce the effects of short-term temporal variability by yielding a time-integrated average VOC concentration. The standard deployment period for passive sorbent samplers in Denmark is two weeks.

In recent years, studies have focused on validation of passive sorbent samplers for measurement of time-averaged VOC concentrations in air over exposure periods of weeks to months. With passive sorbent samplers, the performance of the sampler depends on several factors including: the physical design of the sampler, the sorbent material, and the target VOCs. Specifically, the sorbent material must be compatible with the target VOCs so that the VOCs are retained on the sorbent for the full duration of the sample period (i.e., no back diffusion) but can be fully desorbed for laboratory analysis (i.e., no irreversible sorption). ISO 16017-2 provides guidance for use of passive samplers for deployment periods of up to four weeks (ISO, 2003). As part of a USEPA-funded study, researchers evaluated the performance of the Radiello passive sampler for sampling periods of up to one year (USEPA, 2012b). For longer sample durations, they found low bias for some VOCs with the maximum acceptable sample duration ranging from two weeks for chloroform to one year for tetrachloroethylene (PCE).

Utilizing passive sorbent samplers for measuring VOC concentrations in soil gas can result in data that are biased low due to sampler starvation. Starvation occurs when the uptake rate for the sampler is greater than the rate of VOC flux into the sample point. Because the effect of starvation on sample results is variable and difficult to quantify, passive samplers are most commonly used to obtain qualitative or semi-quantitative information on the distribution of VOCs in soil gas. McAlary et al., 2014a showed that the potential for starvation can be reduced by using a passive sampler with a lower uptake rate. However, very low uptake rates <0.1 mL/min are required to avoid significant starvation effects in high moisture soils (McAlary et al., 2014b). In addition, uptake rates are sampler and chemical specific. For most passive samplers, the uptake rates are considered proprietary by the manufacturer and are not publically available.

4.2. High purge volume sub-slab samples

At the scale of individual buildings, spatial variability in VOC concentrations is much higher for sub-slab soil gas samples compared to indoor air samples (McHugh et al., 2007). As a result, a larger number sub-slab sample points are required to characterize the average VOC concentration with a reasonable degree of accuracy or confidence. In order to reduce the number of sample points required to characterize the VOC concentration below a building foundation, McAlary et al., 2010 have developed a procedure for sub-slab testing of large buildings using high purge sampling from a small number of sub-slab sample points. Using this procedure, a high-flow vacuum blower is used to purge at a high flow rate (e.g., 10s to 100s L/min). Under these purge conditions, an integrated sample can be collected from the purge stream for laboratory analysis providing an integrated measure of VOC concentrations below the slab in the general vicinity of the sample point. Alternatively, a field instrument can be used to measure temporal changes in VOC concentrations in the purge stream providing some information concerning the spatial distribution of VOCs below the foundation (Fig. 4; Eklund, 2010). While this approach can provide an improved understanding of spatial variability in sub-slab soil gas at buildings with relatively large footprints, it may be difficult, in some cases, to accurately account for the dilution of the sample by indoor air drawn down through the slab or ambient air drawn from the edge of the foundation. As a result, regulatory acceptance of this sampling method has varied.

4.3. Real-time on-site analysis

On-site analysis of environmental samples has the potential to stream-line an investigation by allowing the investigator to choose the location for subsequent samples based on the contaminant concentrations results from prior samples. This rapid iteration of sample collection and results interpretation greatly improves the ability to locate the source(s) of VOCs detected in indoor air. For vapor intrusion, mobile analytical laboratories, such as the USEPA TAGA unit, have been deployed to field sites to perform on-site analysis for many years. More recently, Gorder and Dettenmaier, 2011, demonstrated the use of a 20 kg field portable GC/MS to address the challenge of indoor VOC sources in residences. They developed a procedure for area-by-area screening of a building followed by more focused sampling in the areas with higher VOC concentrations. By using the analytical results from each sample to move progressively to the area of highest VOC concentration, they were able to identify specific indoor sources and/or vapor intrusion

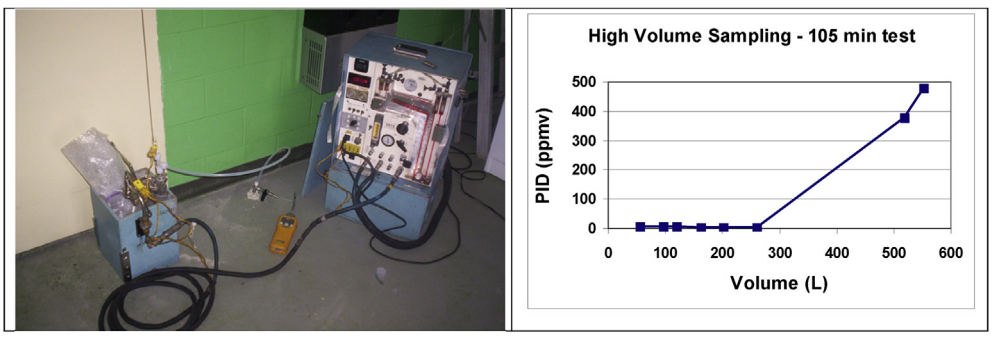

Fig. 4. Equipment used for high-volume sub-slab sampling and example of field data showing "hot spot" some distance from probe.

entry points in all of the buildings they evaluated. They also developed procedures for isolation and testing of potential indoor sources to estimate the VOC emission rate and thus estimate the contribution of the source to VOC concentrations measured in the building. Using these procedures, they found that common consumer products and other indoor VOC sources were responsible for most of the exceedances of indoor air screening levels observed in residences near Hill Air Force Base in Utah (Gorder and Dettenmaier, 2011). Beckley et al., 2014 validated this investigation method in large industrial buildings. They found that this investigation procedure is effective in industrial buildings even if they contain large open indoor spaces. Even within large rooms, the differences in VOC concentrations between sample locations was sufficient to identify locations with indoor sources or vapor intrusion entry points.

4.4. Measurement and control of building pressure

A small pressure difference (i.e., less than 5 Pa) between the building and the subsurface is sufficient to control the direction of air flow through the building foundation. A small positive pressure is sufficient to suppress (i.e., "turn off") vapor intrusion while a small negative pressure is sufficient to "turn on" vapor intrusion. Thus, measuring building differential pressure during indoor air sampling events provides important information concerning the likely sources of any detected VOCs. When building differential pressure is positive, VOCs detected in indoor air are unlikely to have originated from the subsurface and, therefore, indoor or ambient sources are likely.

In most buildings, the building differential pressure can be manipulated between positive and negative pressure using a fan, blower, or the existing heating, ventilation, and air conditioning (HVAC) system. As a result, measuring VOC concentrations in indoor air under controlled positive or negative building differential pressure can provide an improved understanding of vapor intrusion conditions (McHugh et al., 2012). When baseline building differential pressure is neutral or negative, induction of a positive building differential pressure can be used to evaluate the presence of indoor sources of VOCs. The detection of VOCs in indoor air under controlled positive differential pressure conditions provides evidence for indoor or ambient sources. If vapor intrusion and indoor sources are both contributing VOCs to indoor air, the change in VOC flux between baseline and controlled positive differential pressure conditions can be used to estimate the relative contribution of these different sources (McHugh et al., 2012). When baseline building differential pressure is neutral or positive, induction of a negative differential pressure can be used to evaluate the potential for vapor intrusion to occur during the winter heating season or under other conditions when natural negative building differential pressure may occur. The building pressure control method for evaluation of vapor intrusion has been validated through the USEPA Environmental Technology Verification (ETV) program (MacGregor et al., 2011).

Holton et al., 2015 evaluated the controlled building differential pressure method at Sun Devil Manor, the vapor intrusion research house near Hill AFB used for a multi-year study of vapor intrusion processes. Under natural building conditions, VOC concentrations in indoor air attributable to vapor intrusion varied by more than two orders of magnitude with the highest VOC concentrations occurring on only a few days of the year. Under a sustained induced negative building differential pressure condition, they found that i) VOC concentrations in indoor air attributable to vapor intrusion exhibited much lower temporal variability compared to baseline conditions, ii) false negative results were not obtained (i.e., vapor intrusion was always detectable under controlled negative building differential pressure conditions), and iii) VOC concentrations in indoor air were similar to the maximum concentrations observed under natural building conditions. These results suggest that measuring VOC concentrations in indoor air under controlled negative differential pressure conditions provides a relatively simple way to evaluate potential worst case or near worst case vapor intrusion for that building. Short-term building differential pressure control studies can be conducted in occupied buildings, although the air flow used to control building pressure may have some effect on building temperature. The controlled building pressure method is cited in Interstate Technology & Regulatory Council (ITRC) and USEPA guidance as a method to distinguish between vapor intrusion and background sources of VOCs (ITRC, 2014; USEPA, 2015a).

4.5. Stable isotope analysis

Biodegradation of chlorinated VOCs in the subsurface typically results in enrichment in heavy isotopes for the remaining undegraded parent compound (USEPA, 2008). For sites where biodegradation is occurring, this isotope enrichment effect can result in clear differences in the isotope signature between the chlorinated VOCs in the subsurface and indoor sources of the same VOCs (McHugh et al., 2011; Beckley et al., 2016). Un-degraded manufactured sources of chlorinated VOCs exhibit a relatively narrow range of carbon and chlorine isotope ratios (USEPA, 2008; McHugh et al., 2011); thus the enrichment in heavy isotopes caused by biodegradation in the subsurface commonly results in isotope ratios for the subsurface source that are distinct from the range observed for manufactured sources found in consumer products (McHugh et al., 2011). As a result, the compound-specific stable isotope analyses of a small number of subsurface source and indoor air samples from a site are often sufficient to classify the VOCs detected in indoor air as originating from a subsurface source verses an indoor source (Beckley et al., 2016).

4.6. Tracer gases

Measurements of tracer gases can be used to obtain a better understanding of specific components of the vapor intrusion pathway at a site. Tracer compounds may be natural or anthropogenic compounds already present at a site or may be compounds released at specific locations as part of the pathway investigation.

Radon is a naturally-occurring compound that has been widely used as a tracer for vapor intrusion. Although the concentration varies with soil type, radon gas is present at some level in almost all soils. Because radon is ubiquitous in soil gas, it can be used as a general tracer for soil gas entry into a building. Radon measurements are generally inexpensive compared to VOC concentration measurements and, for most buildings, indoor sources of radon are not a concern. As such, radon measurements can be used to estimate soil gas entry rates and the sub-slab to indoor air attenuation factor (McHugh et al., 2008). However, because distribution of radon below a building foundation may be different from the distribution of site VOCs, the attenuation factors for radon and for VOCs may differ. As a result, radon measurements are best used as a measure of bulk soil gas entry into a building and a qualitative or semi-quantitative measure of sub-slab to indoor air attenuation for VOCs. Despite this limitation, radon monitoring can be a cost-effective method to evaluate temporal variability in soil gas entry into buildings (Schuver and Steck, 2015). In addition, radon can be used to verify that induced positive building differential pressure has suppressed soil gas entry into a building (McHugh et al., 2012).

A variety of compounds can be introduced into the environment in order to provide a better understanding of gas movement at

vapor intrusion sites. Tracer compounds such as sulfur hexafluoride (SF_6) and helium are used to measure air exchange in buildings for a variety of purposes unrelated to vapor intrusion such as evaluating building air exchange rates to understand energy efficiency (ASTM, 2011). These compounds have also been released below buildings in order to evaluate the movement of soil gas into buildings. For example, Olson and Corsi (2001) introduced an SF_6 tracer into soil gas at two test houses in New Jersey. At a former drycleaner site, Eklund and Simon (2007) introduced SF_6 in the subsurface to evaluate soil gas entry and released helium indoors to evaluate building ventilation rates. Note, however, that the use of SF_6 is restricted in some countries because it is a greenhouse gas. Lundegard et al., 2008 used nitrogen gas to displace the soil gas below a residence overlying deeper petroleum contamination and then monitored oxygen rebound to obtain an understanding of the movement of atmospheric air below the building. In general, the use of introduced tracer gases to understand the movement of contaminants in the vadose zone is limited by the ability of the tracer release to mimic the distribution of the contaminant vapors. As a result, the use of introduced tracers in soil gas typically has been limited to research sites.

Perfluorinated tracers (PFTs) are anthropogenic compounds that can be used in designed experiments to investigate vapor migration pathways in a qualitative or quantitative way. Most commonly applied are the two compounds perfluoromethylecyclopentane (PCMP) and perfluoromethylecyclohexane (PCMH) manufactured by Brookhaven National Laboratory although several other PFTs also are available. PFTs can be detected at part per trillion concentrations and thus can be released into the environment at very low rates over extended periods of time allowing the assessment of gas movement over periods of days to weeks. PFTs are released at a constant rate from emitters and are sampled by passive samplers – typically over a period of approximately 2 weeks. One passive sampler can be used to measure the concentration of multiple PFTs. With a controlled release of tracers on the source side of a structure and measurements on both the source and receptor side, a well-designed tracer study can be used to estimate attenuation factors for that building. When PFTs are released at different locations, it is possible to estimate the contribution of different VOC sources or transport pathways. Successive tracer studies can be applied to improve the site-specific CSM (Loll et al., 2016). PFT study results are most useful when the PFT distribution at the release point is comparable to the distribution of the VOCs. As a result, the method tends to perform better when the PFT is released into an open space such as a crawl space, basement, wall cavity, or sewer line. Results from a point release in the vadose zone are more difficult to interpret. The PFT method has been applied to quantitatively assess air exchange between dry cleaners and adjacent apartments (Mortensen and Glensvig, 2003), to qualitatively assess vapor intrusion through sewer systems (Riis et al., 2010), and to estimate attenuation factors for crawl spaces, cavity walls and industrial settings (motorcycle repair shop and dry cleaning) (Loll et al., 2016).

4.7. Mass flux/mass discharge analysis

Mass flux is the rate (mass per time) of contaminant movement per unit area. Mass discharge is the rate (mass per time) of contaminant movement through a defined structure or area. Mass flux and mass discharge analyses have been proposed as an alternative to concentration-based analyses for the evaluation of risk associated with various exposure pathways at contaminated sites. For example, in 2001, Einarson and MacKay proposed the use of mass discharge analysis to evaluate the potential for a contaminant plume in groundwater to impact a pumping well at a concentration above a regulatory standard. Although some tools exist to directly measure mass flux, mass flux or mass discharge within a groundwater plume as a whole is most commonly estimated using contaminant concentration and groundwater flow data. Several researchers have recently suggested the use of a similar approach to evaluate mass flux and mass discharge along the vapor intrusion pathway using VOC concentration and other site data.

McHugh and Nickels, 2008 proposed using mass flux and mass discharge analyses to evaluate VOC transport along the vapor intrusion pathway from a groundwater source, through the vadose zone, and then into the building. They found that high uncertainty in the estimated mass flux values made it difficult to evaluate the consistency in mass flux along the vapor intrusion pathway. However, McHugh et al., 2012 found that the evaluation of mass discharge through a building under baseline conditions and controlled positive and negative differential pressure conditions was useful for distinguishing between subsurface and indoor or ambient sources of VOCs in indoor air.

More recently, Guo et al., 2015 found that mass discharge analyses were useful for identification of preferential pathways for vapor intrusion. At their research house (Sun Devil Manor), they found that the mass discharge of TCE through the residence under controlled negative pressure conditions was approximately three orders of magnitude higher than would be expected based on the mass discharge through the vadose zone beneath the residence. This large discrepancy in mass discharge values provided strong evidence of preferential mass transport through a land drain sewer line connected to the building foundation. When the land drain system was disconnected from the residence, the mass discharge of TCE through the residence decreased dramatically and more closely matched the estimated mass discharge through the vadose zone (Guo et al., 2015).

Mass discharge also may be useful for determining when vapor intrusion mitigation systems are no longer necessary for a given building. The exhaust piping from a sub-slab venting system allows measurement of mass emission rate using standard stack testing methodology. This mass discharge through the vent stack represents the maximum potential vapor intrusion rate in the absence of mitigation. As a result, monitoring of decrease in mass discharge through the vent stack during site remediation is likely to indicate when the mitigation system is no longer needed.

4.8. Identification of vapor entry points

For some buildings, vapor entry occurs primarily through a small number of foundation features such as cracks, plumbing penetrations, or floor drains. The identification of these features can confirm the occurrence of vapor intrusion and can also provide an opportunity for at least partial mitigation of the vapor intrusion (i.e., through sealing of these specific features). A number of methods have been developed to identify vapor intrusion entry points. All of these methods require negative differential pressure (relative to the sub-slab) to provide advective flow through the potential entry point during testing.

- **ppb-Level Screening**: VOC-screening tools, like the ppbRAE® or similar instruments with ppb-level detection capabilities, can, in some instances, be applied for identification of vapor entry points. This screening instrument provides a real-time semi-quantitative measurement of the total VOC concentration in air being sampled by the instrument. Fuglsang, 2004 used a ppb-level detector with an attachment that served to isolate the potential entry point being tested from the indoor environment. This allowed for testing vapors moving through the entry point without interference from VOCs in indoor air. Lauridsen and Overgaard, 2006 documented the use of this method at three

former dry cleaning sites. The success of the method depended on a significant signal contrast between the source (i.e., VOC concentration at the entry point) and the background VOC levels in the building. Because the instrument measures total VOC concentrations, any background VOC (not just the specific VOCs associated with the subsurface source) can cause interference. In addition, humidity can cause negative interference with the photo-ionization detector (PID) resulting in a low bias for readings in humid soil gas. Background total VOC levels in buildings vary widely and are difficult to predict. As a result, VOC concentrations of 100 ppb or higher relative to background may be required at the VOC entry point before they can be reliably distinguished from indoor background. One advantage of this screening method is that only actual VOC entry points are identified. Some of the other entry point screening methods discussed below will identify soil gas entry points regardless of whether VOCs are present at the entry point.

- **Thermography**: Sebastian, 2009 suggested using thermography together with an applied negative differential building differential pressure of 50 Pa, relative to the sub-slab, to test for soil gas entry points. Thermography uses a hand-held sensor to identify temperature differences on surfaces. When soil temperatures are lower than indoor temperatures, soil gas entry can result in lower temperatures at the soil gas entry point. Sebastian, 2009 found that this method was effective at the test site but also pointed out limitations with regard to applying the method: i) under conditions of a low temperature difference between the sub-slab soil and building environment (i.e., <5 °C) where soil gas entry may not result in measureable temperature differences, and ii) at sites with wet building materials where observed temperature differences may be attributable to evaporative cooling. Langeland et al., 2014a tested thermography at four sites with a range of negative building differential pressures (2–60 Pa) in a comparative study of three methods for locating vapor intrusion points. They concluded that the thermography method was inferior to both the hydrogen tracer gas method and ppb-level screening method, and was basically only able to identify entry points that were visible to the naked eye and/or could be felt by hand due to high air flow. Langeland et al., 2014b concluded that thermography is not well suited for finding potential soil gas entry points due to a high risk of false negative results.
- **Hydrogen tracer gas**: Loll et al., 2008 and Loll et al., 2010a suggested using hydrogen tracer gas to identify soil gas entry points. This method consists of: i) a controlled release of a 5% H_2/95% N_2 tracer gas on the source side of a structure, ii) verification of the sub slab saturation, and iii) screening of potential entry points with a fast response hand held hydrogen detector with both audible and visual semi-quantitative measurement of the hydrogen level. This type of hydrogen detector is commonly used to detect leaks in natural gas pipelines and other equipment. The method requires little training, uses inexpensive equipment, and yields rapid results. Loll et al., 2010b documented applications of this method at six different sites. Riis et al., 2010 applied the method for investigation of sewer preferential pathways. In recent years, the method has more frequently been used in Denmark for documenting the functionality of mitigation systems that include a barrier/membrane (e.g., Larsen et al., 2013). Langeland et al., 2014a and Langeland et al., 2014b compared the hydrogen tracer investigations to other methods at two sites and concluded that the method is well suited for identification of soil gas entry points, but pointed out that there are limitations at sites with water logged soils below the structure to be investigated.
- **Thoron/Radon-measurements**: Thoron (^{220}Rn) is a naturally occurring short-lived isotope of Radon (^{222}Rn) with a half-life of only 56 s. Petersen et al., 2012 utilized the short-lived nature of Thoron to locate advective soil gas vapor entry points at 28 sites (including five sites with VOC vapor intrusion concerns). Two of these sites were also investigated using ppb-level screening, thermography, and hydrogen tracer tests for comparison. Petersen et al., 2012 concluded that concurrent measurements of Thoron, Radon, and differential pressure across the concrete slab is a suitable method for locating potential advective vapor entry points. Langeland et al., 2014b found that the method was effective and had a slightly lower cost than ppb-level screening and hydrogen tracer methods. However, the authors point out that each measurement takes 5–10 min compared to the near instantaneous reading provided by the ppb-level VOC detectors and the hydrogen detectors.

4.9. Vapor intrusion models

USEPA vapor intrusion guidance documents have consistently recommended a limited role for mathematical models to evaluate the vapor intrusion pathway (USEPA, 2002; USEPA, 2015a) compared to the stand-alone model-based evaluations commonly conducted prior to the early 2000s. The USEPA has recommend that models can be used as line of evidence in conjunction with site investigation data as part of a multiple-lines-of-evidence approach. European researchers and regulatory agencies have been active in the development and evaluation of vapor intrusion models (Provoost et al., 2008; Provoost, 2013; CityChlor, 2013; Hulot et al., 2010; Traverse et al. [Fluxobat], 2013). Similar work has been undertaken in Australia (Turczynowicz et al., 2012; www.crccare.com/publications). Further, Yao et al., 2013 conducted a comprehensive review of available vapor intrusion models including simple 1-D screening models and more complex 2-D and 3-D models. They concluded that, although a number of new vapor intrusion models have been developed in recent years, these models lack formal validation limiting the confidence that can be placed in the model predictions.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2017.02.015.

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