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# Evaluating the role of preferential pathways in exacerbating vapour intrusion risks



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# ABSTRACT

Vapour intrusion (VI) has been garnering more attention in the research field in the past few decades owing to its associated health risks. Many field studies have been conducted to understand the process of vapour migration from the source to indoor air and the factors affecting it. This research project primarily concentrated on the role of highly permeable backfill materials like sand and gravel used as bedding and embedment for utility lines. These can function as preferential pathways for vapour migration in the vadose zone. Extensive laboratory investigations were carried out over 13 days using two large two-dimensional (2-D) columns, one with preferential pathway and one without preferential pathway, to investigate the natural attenuation process and the role of preferential pathways in VI. The presence of preferential pathway has a role in determining soil gas vapour distribution in the vadose zone. A notable increase in vapour concentrations was observed in the top layer of the column with preferential pathway compared to the column with no preferential pathway. This was due to enhanced distribution of contaminant vapours along the column facilitated by the preferential pathway. An increase in concentration was observed in the column with preferential pathway throughout compared to the column with no preferential pathway, confirming that the presence of natural or man-made drainage features in the sun-surface can influence the distribution of contaminants throughout a site. This research project generates a better understanding of preferential pathways and recommends taking into account the role of highly permeable backfill materials when developing CSM and VI health risks assessments. In this way the strategic priorities for remediation projects will be successfully established.

#### 1. Introduction

The migration of volatile organic compounds (VOCs) from the subsurface source into buildings – *vapour intrusion (VI)* – has for decades been acknowledged as a major inhalation exposure pathway. Largescale production and use of organic solvents and hydrocarbon fuels have resulted in many VOC contaminated sites caused by leaking underground storage tanks, accidental spills, wastewater disposal lagoons, etc. (López et al., 2008). This has given rise to a significant number of hydrocarbon contaminated sites leading to potential VI impacts. Exposure to some chemical vapours that migrate through the soils and into a building can pose greater health risk than drinking chemicals found in tap water (Schuver, 2007). Most of these VOCs are potentially toxic and carcinogenic even in smaller concentrations (Karapanagioti et al., 2003). Due to these severe health impacts, much attention since the 1990s has been paid to VOC migration in vadose zone, its accumulation underneath foundation slabs of buildings and the subsequent VI into indoor air. Decades later, the focus is now on mitigating VI through research studies, mathematical modelling, field investigations and implementation of guidelines.

The major contaminants of concern in VI are petroleum hydrocarbons (PHCs), which can readily biodegrade under oxygenated soil conditions and chlorinated hydrocarbons (CHCs), which can resist aerobic biodegradation. Migration of PHCs can be limited in soil using aerobic biodegradation (Davis et al., 2009; DeVaull, 2007) producing carbon dioxide and water, whereas CHCs biodegrade much more slowly by anaerobic degradation. The latter process produces toxic by-products like dichloroethylene and vinyl chloride (McNab and Narasimhan, 1994). PHC vapours emanating from dissolved-phase sources are likely to be composed primarily of benzene, toluene,

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ethylbenzene, xylenes (BTEX) (Mitra and Roy, 2011) and other aromatic hydrocarbons. Benzene is estimated to be the primary constituent of potential concern for gasoline and diesel fuels based on a chronic (30year duration) human health residential exposure scenario (Bowers and Smith, 2014) because of its volatility, recognised toxicity and mass fraction in common fuels (gasoline and diesel).

In VI mitigation strategies, it is crucial to well understand the nature of contaminant vapours in the subsurface and to identify the settings where vapour transport leads to unacceptable safety and human risk (Roggemans et al., 2001). Research efforts such as field investigations and risk assessments have improved our understanding of the VI process, including the role of natural attenuations, soil heterogeneities, subsurface conditions, building characteristics and presence of preferential pathways. However, quantification of VI and vapour plume delineation have been a major challenge for concerned professionals who demand accurate site characterisations and evaluation of health risk assessment, primarily due to complexities involved in vapour transport in the vadose zone (Beck and Mann, 2010). The high spatial and temporal variability in soil vapour concentration makes it difficult to evaluate VI using conventional field investigation methods (McHugh et al., 2017a). Having recognised this issue, considerable efforts have been made by both researchers and regulators worldwide to evaluate and accurately predict VI exposure pathways (Bekele et al., 2018; Yao et al., 2017; Yao et al., 2016; Bozkurt et al., 2009). In the majority of vapour intrusion models (VIMs) and regulation documents, the conceptual site model (CSM) for VI primarily focusses on: i) VOCs partitioning from contaminated soil or groundwater into soil gas; ii) transport of soil gas through vadose zone mainly by diffusion; and iii) vapour entering buildings through cracks in foundations made possible by further diffusion or advection. Although the need to address preferential pathways is noted in guidance documents, information regarding their inclusion in the CSM or the prevalence of this pathway is very limited.

Preferential pathways play a significant role in delineating vapour plume accurately. The role of a preferential pathway in VI is poorly understood. As a result, these pathways are often not considered when developing conceptual site models and VI investigation or mitigation plans. Neglecting this pathway can complicate data interpretation, resulting in repeated and potentially redundant sampling. Preferential pathways are generally highly permeable natural or anthropogenic subsurface features that act as a path of least resistance for vapour transport. These pathways tend to transport contaminant vapours faster and farther than predicted by VIMs (McHugh et al., 2017a). Recent studies have identified the presence of sewers or utility tunnels in the subsurface, facilitating a preferential migration pathway in which the contaminant vapours can migrate into buildings with least resistance. In spite of this, there is currently limited regulatory guidance on how this will affect the plume size.

In many cases, the importance of the preferential pathway at the site was identified only after extensive site characterisation and VI testings (Beckley and McHugh, 2020; McHugh et al., 2017b). In addition, mitigation techniques like sub-slab depressurisation will not be effective where preferential VI pathways through sewer lines are dominant (Holton et al., 2015). Subsequently, identifying VI pathways and their significance is critical during VI mitigation system selection and design. Given the absence of sewer testing during most vapour intrusion investigations, it is likely there are additional sites where VOC transport through sewers and utility tunnels is important to learn but this has not yet been identified. This highlights the need for an improved understanding of VOC transport processes and the factors that make this transport significant at individual sites.

Currently, for VI site screening and human health risk predictive models, empirical values are derived from a large amount of data from subsurface samples of groundwater, exterior soil gas, sub-slab soil gas and building crawl spaces. These are often used in field investigations (USEPA, 2015; Friebel and Nadebaum, 2011; USEPA, 2013). As well, these values can serve to estimate the potential indoor air concentration based on which decisions to undertake response action for VI. Similarly, empirical attenuation values are employed for both vertical and horizontal exclusion of VI investigations relative to the distance of the source vapour plume. A buffer zone of 100 feet (30 m) vertically and laterally from the contaminant plume is recommended by US EPA (2002), beyond which the health risk of VI can be deemed insignificant in buildings at a distance greater than one house lot. However, these empirical values of exclusion distances have several assumptions attached to them, which include neglecting the role of preferential pathways in natural attenuation and application of exclusion distances. This research will investigate the limitations of using empirical values of exclusion distances in the presence of preferential pathways.

Several field studies conducted in the recent past confirm the role of preferential pathway in exacerbating VI, resulting in significant indoor air contaminant concentrations even in buildings outside the area of groundwater plume (Beckley and McHugh, 2021; Guo et al., 2020; Pennell et al., 2013; Vroblesky et al., 2011). Most of these studies dealt with the interception of compromised or deteriorated sewer systems or utility lines and their associated plumbing conduits, resulting in unhindered transport of VOCs into indoor air environments with little attenuation. However, the roles of highly permeable soil layers and backfill materials in VI have rarely been investigated. The presence of any highly permeable region in the vadose zone, either natural or anthropogenic, can function as a preferential pathway in contaminant vapour transport. These regions of high permeability can occur naturally as gravel layers or fractured rocks which facilitates higher contaminant flux owing to their higher porosity (USEPA, 2015). Additionally, granular fill materials laid down as bedding and embedment to utility lines can cause high contaminant flux laterally and vertically to the ground surface thereby acting as preferential pathways (Interstate Technology and Regulatory Council, 2014). Fig. 1 presents a typical CSM where a highly permeable utility line bedding/backfill acts as a preferential pathway.

The intention of this research was to: firstly, understand and evaluate the behaviour of benzene vapours under natural conditions; and secondly, conduct detailed evaluations of the role of highly permeable soil backfills as preferential pathways and natural attenuation in order to improve the accuracy of health risk assessment. This will help to save time, money and resources for all stakeholders. Laboratory column experiments using large 2D columns with and without preferential pathway were employed to achieve this objective. Data sets were obtained, analysed and compared for both preferential pathway (PP) and no-preferential pathway (NPP) scenarios to understand the role of preferential pathways in VI.

# 2. Research methodology

To understand the role of preferential pathways in VOC migration from subsurface sources to indoor air, a large laboratory-scale column study was conducted with information compiled from other published sources. The vapour transport mechanisms in the soil, design, fabrication and procedure of large soil column and laboratory analytical methods employed in this study are discussed in more detail below.

#### 2.1. Design and procedure of large lab-scale two-dimensional column study

This experiment was designed to understand the vapour migration pathway from source to building in the presence of a preferential pathway and to evaluate the factors contributing to it. The column set-up consisted of two large 2D columns with dimensions of 100 cm x 50 cm x 20 cm, one with preferential pathway (PP) and the other with no preferential pathway (NPP). These soil columns were custom-made with stainless steel and glass, comprising 15 sampling ports aligned vertically in three layers above the source injection port, at equal distance, to measure the vapour flux in both X and Y axis for both the columns as shown in Fig. 2. Laterally, the initial two sets of sampling ports were placed close to the source than the rest which were placed at a wider space





Fig. 2. Source and sampling ports layout of the 2-D soil column.

away from the source injection port. This was done in order to closely monitor the vapour behaviour close to the source. The PTFE-Silicon septum was used in the sampling ports for ease of sample extraction and prevent vapour escaping through the ports. The preferential pathway is introduced in one of the columns using a 20 mm pipe surrounded by highly permeable washed gravel laid down in a similar method of field installation. The soils were packed in the columns up except for the top 5 cm of each column. For this study, non-contaminated soil samples were sourced from Pilbara, Western Australia.

The soil mass was calculated by weighing the column before and after the soil was added. The experiment was conducted at a constant temperature of  $22\pm2$  °C by placing the column in a temperature regulated room. The fate and transport of the vapour as it migrated through the soil column, both in the presence and absence of preferential pathway, were monitored using the sampling ports. Soil vapour data from both columns were collected at frequent hourly, daily and weekly intervals to demonstrate the vapour migration process. This work focused on vadose zone vapour as the source to investigate the role of preferential pathways in VI from impacted soil or groundwater.

#### 2.1.1. Soil repacking

The soil columns were packed with non-contaminated soils from Pilbara, Western Australia to represent the field conditions. These soils Fig. 1. Conceptual site model for VI with a highly permeable backfill material acting as preferential pathway (Unnithan et al., 2021).

were highly mineralised and highly deficient in organic matter. The soils were air dried and passed through a 2 mm sieve and a detailed physicochemical characterisation was conducted. Processed soils were tested for baseline hydrocarbon contamination as well which was reported to be non-detectable. Soils were repacked in the soil column in 5 cm layers and tapped with a rubber hammer after each layer for the uniform repacking of the column. By packing air-dried soils in the column, the uniformity of the packing and higher bulk density was ensured, which helped to minimise macro-porosity. In total, 186 Kg and 172 Kg of soils were packed in 0.1125 m<sup>3</sup> vol in the PP and NPP columns, respectively, attaining bulk densities of 1680 Kg/m<sup>3</sup> and 1529 Kg/m<sup>3</sup> in PP and NPP, correspondingly.

To introduce a PP, washed gravel of size 4 mm was laid down around the 20 mm pipe for a total thickness of 50 mm, as depicted in Fig. 3, mimicking the pipe bedding and embedment of the utility lines as will occur in a real site scenario. Both ends of the pipe were sealed with airtight connections and screwed intact. This highly permeable gravel layer will act as the PP, intersecting the vapour migration route of the vapour and accelerating vapour migration away from the source. An airtight lid covers the top of the column, preventing ambient air contact.

#### 2.1.2. Introducing benzene vapour as the source

Pure analytical grade liquid phase benzene (> 99% purity) purchased from Sigma-Aldrich (Germany) served as the contaminant vapour source in the soil column. 8 mL of pure analytical grade liquid phase benzene was injected into the source port of both columns which then volatilised slowly at room temperature and migrated through the soil at room temperature via diffusion mechanism. The volume injected was chosen in order to avoid soil saturation limit calculated for a pure benzene chemical. The source port was injected periodically with benzene to prevent depletion before reaching the farthest sampling port.

#### 2.2. Laboratory analytical methods

#### 2.2.1. Soil characterisation

Air-dried soil passing through a 2 mm sieve was subjected to standard soil analysis to determine its physicochemical properties, namely



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Fig. 3. Soil repacking in 2-D PP column with gravel bedding.

moisture content, soil acidity (pH), electrical conductivity (EC), dissolved organic carbon (DOC), cation exchange capacity (CEC), water holding capacity (WHC), carbon (C) content, nitrogen (N) content and elemental composition. The moisture content was determined by ovendrying the soil to 105 °C. Soil pH and EC were measured in 1:5 (w:v) soil:water slurry after equilibrating in an end-over-end shaker for 1 hour (Rayment and Higginson, 1992). The DOC contents of the soil samples were determined using a TOC analyser (Shimadzu, China) in a 1:5 (w:v) soil:water slurry. The soil CEC was determined utilising the method described by (Gillman and Sumpter, 1986). Total Carbon (TC%) and Total Nitrogen (TN%) in soils were determined in a 0.25 g of oven-dried soil using the Trumac CNS analyser (LECO® Corporation, Australia). The total metal concentration of soils was analysed using US EPA 3051A, i.e. microwave assisted acid digestion procedure (USEPA, 2007). Soil organic content was measured by loss with the ignition method (Moebius-Clune, 2016).

# 2.2.2. Vapour sampling and analysis

A conventional gas chromatograph (Agilent 7980) equipped with auto headspace sampler and flame ionisation detector (FID) helped to analyse the concentrations of the VOCs. The detector was set to the highest sensitivity and the data were collected with a chromatography datahandling system. The vapour samples were drawn through the sampling ports of the column sealed with PTFE/silicon septa screw cap using a gas-tight syringe with stepping time from the time of injection. 1 mL of vapour samples was drawn from each sampling port, transferred to pre-treated 21 mL of GC vials and placed in the GC auto sampler for analysis. The sampling vials (21 mL) were prepared by purging them with pure lab-grade nitrogen gas using a gas dispenser for one minute and crimp capping with an aluminium cap with PTFE/silicon septum. The vials were then placed in a 60 °C oven and allowed to equilibrate for 15 min. Gas-tight syringes were washed with methanol, heat-treated between sampling tasks, and checked for contaminants before use.

Standards were prepared for the benzene vapours using pure analytical grade compounds injected into a pre-treated 121 mL crimpy top serum bottle and weight was measured using a high precision analytical scale. A range of gaseous volumes (e.g., 2, 5, 10, 20, 50, 75  $\mu$ L) were transferred from the standard bottle to the pre-treated 21 mL vials as per the calibration ranges required. The gaseous concentration in the crimp-capped bottle was calculated using Eq. (1):

$$S_{conc} = \frac{Benzene_{inj}}{V_{v}} \tag{1}$$

where:  $S_{conc}$  = Standard concentration in the vial (g/cm<sup>3</sup>); Benzene<sub>inj</sub> = weight of benzene injected (g); and V<sub>v</sub> = volume of the crimpy vial (cm<sup>3</sup>).

The experiment was not conducted until a steady state was reached but for a period of 13 days which was found to be the time taken for

Table 1	L
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Soil sample characterisation.

Parameter Measured		Data
	pH	$7.9 \pm 0.1$
	Ec (μS cm <sup>-1</sup> )	$2167 \pm 110$
	Moisture (%)	$3.4 \pm 0.4$
	CECb (cmol <sub>c</sub> /kg)	$3.58 \pm 2.9$
	Total N (mg/ Kg soil)	$465 \pm 92$
	Total P (mg/ Kg soil)	$559 \pm 21$
	TC (%)	$0.59 \pm 0.04$
	TN (%)	$0.03\pm0.01$
	Soil organic matter (%)	$1.1 \pm 0.3$
Particle distribution (%)	Sand	56
	Silt	33
	Clay	11
Elements	Al	$8555 \pm 341$
	As	$11 \pm 1$
	Ba	$34 \pm 1$
	Ca	$3426 \pm 266$
	Cr	$56 \pm 3$
	Fe	$179,436 \pm 2,8889$
	Mg	4684 ± 79
	Mn	$1120 \pm 80$
	Mo	$10 \pm 3$
	Na	$551 \pm 100$
	Zn	91 ± 4

the contaminant vapour to move from source port to the farthest port in the experimental setup (i.e. Port 1). During this time, the vapour concentration from each port was monitored and compared for both the setups. Before the commencement of the experiment, both the columns were purged with nitrogen to remove oxygen from the systems to eliminate any chance of vapour attenuation due to biodegradation. The soil columns were placed in the fume hood connected to the laboratory vacuum system to strip the vapour from the set-up after the experiment. Following verification of the sample from the sampling port for the stripping of vapour, the column was emptied and the soils were disposed of using licensed waste removal systems.

#### 3. Results and discussion

#### 3.1. Soil samples used in the column study

The physicochemical properties of the uncontaminated soil sample selected for the study are presented in Table 1:

The soil used in the study was highly mineralised with a slightly alkaline pH value of 7.9  $\pm$  0.1. The soil had a higher EC of 2167  $\mu$ S/cm and the moisture content was 3.4  $\pm$  0.4%. The CEC was 3.58  $\pm$  2.9 cmol<sub>c</sub>/Kg.



**Fig. 4.** Benzene vapour concentration plot as GC response area at all three layers (a) soil columns in the top, (b) middle and (c) bottom layers.

The soils had a lower TC (0.59  $\pm$  0.04%) and TN (0.03  $\pm$  0.1%) while the soil organic matter was 1.1  $\pm$  0.3%.

# 3.2. Vertical concentration profiles and attenuation of volatiles in the 2D column study

Rapid distribution of vapour was observed in the soil column, with detectable concentrations of vapour being measured at the furthest sampling port from the source. This indicates that the soil exhibited minimal vapour sorption and no biological activity.

The vapour concentrations from Port-13 and Port-1 were used to determine the rate of vapour transport vertically and laterally from the source, respectively. Vertical plot pairing aligned ports constitute the most practical way to measure the vertical migration of vapours and attenuation factor. Fig. 4 illustrates the contaminant vapour concentrations on Day-1 and Day-13 in both soil columns in the top, middle and bottom layers, respectively.

After the first day when the source was introduced, the vapour concentrations were higher at the source side sampling ports with the con-



Fig. 5. Benzene attenuation between bottom and top sampling ports.

centrations in the PP column being more pronounced. These then eventually decreased away from the source to virtually zero at the farthest end of all three layers for both columns. After Day-13, the vertical concentration for the column with PP revealed a higher vapour concentration at the top layer than NPP column and was more pronounced for columns farther away from the source. Based on this, it can be concluded that the PP resulted in an elevated concentration further away from the source at a rate greater than natural attenuation.

The rate of attenuation can be analysed by comparing the vapour concentrations from the bottom sampling port (15, 12, 9, 6 and 3) to the top sampling port (13, 10, 7, 4 and 1) aligned vertically and plotting it to understand the contribution of PP. An average rate of attenuation (average alpha) is computed by taking the ratio of the average vapour concentrations of the top and bottom sampling ports of each vertical layer for both PP and NPP soil columns as shown in Fig. 5.

Overall, the column with PP showed higher concentration in the top layer as compared to NPP as depicted in Fig. 4(a), which strongly suggests that the presence of preferential pathway has resulted in an elevated vapour concentration further away from the source at a rate greater than natural attenuation. The attenuation factor increased from Day-1 to Day-13 in the top layer compared to the bottom layer for both columns as the vapour tended to diffuse more upward with less pressure than laterally in natural soils. The concentration plot for column with PP shows that the VOCs which migrated laterally are quicker than the column with NPP. This can be extrapolated to a field scenario where the presence of preferential pathways will tend to enhance the migration of contaminant vapours from a higher source concentration, thus posing a more potential risk of VI.

#### 3.3. Soil vapour concentration 2D contour profiles

The interpolation of data was conducted using Kriging and the soil vapour concentration profile is generated using surface mapping software Surfer® developed by Golden software®. A significant difference in spatial variabilities was observed between columns with PP and NPP although both were applied identical sources as given in Fig. 6. Larger vapour concentrations can be observed for the PP 2-D column at top sampling ports when compared to the NPP 2-D column. The top layers of the PP column presents significant peak concentration as compared to NPP column, especially for Day 3 and Day 13 which indicates accelerated vapour transport occurring at the preferential pathway layer. This could be due to the enhanced distribution of the contaminant vapour facilitated by the preferential pathway acting as the path of least resistance in the PP 2-D column. It can be observed that the concentration on Day 3 for PP column peaks at ports vertically above the source port (Port 13 and Port 10) whereas on Day 13, the concentration peaks at the ports farthest from the source (Port 1 and Port 4). On both occasions, the NPP column shows significantly less concentrations than PP column.



Fig. 6. Day -3, -7 and -13 contour mappings of Benzene vapour concentration plot (in  $\mu$ g/L) for two scenarios investigated: a) with preferential pathway and b) with no preferential pathway.

This indicates that the PP layer is likely to accelerate vapour migration in both vertical and lateral directions.

In this study, it was observed that the vapour concentrations were higher at all monitoring ports in the top-layer of the PP column as compared to the NPP column. This difference was particularly noticeable during the sampling events on Day-13. The elevated concentrations could be explained by accelerated vapour migration as a result of the PP acting as a path of least resistance. This suggests that the presence of a highly permeable PP is likely to accelerate the vapour migration in the subsurface potentially exacerbating the risk of VI possibly to places beyond the exclusion distances suggested by US EPA (2002), for a high source concentration. Therefore, it would be advisable to take into account the existence of highly permeable layers beneath the surface when creating CSMs to ensure more precise health risk evaluations. The discussion in this research is focused on benzene as it is considered the primary risk driver amongst the BTEX compounds at petroleum hydrocarbon impacted sites.

# 4. Conclusion and recommendations

The study aimed at understanding how a highly permeable preferential pathway layer, in the form of embedment for utility lines and its influences on the vapour migration in the subsurface. The study was conducted using two-dimensional soil columns with and without preferential pathway. The contaminant mass flux at the top surface was observed to be higher in the column with PP than the NPP column, which suggests that natural or anthropogenic preferential pathways can influence the distribution of contaminants across the vadose zone. Day-13 sampling events revealed a higher concentration at all sampling ports for the top layer of PP 2-D column as compared to NPP column. This could be due to accelerated vapour migration along the preferential pathway acting as a conduit for the vapour to migrate.

The conceptual model scenario for the VI pathway presented in this study generates important insights into the factors involving VI pathway. At hydrocarbon-contaminated sites where sewers and utility connections cross the vapour plume, the mass flux chemical concentration can be substantially higher than the attenuation rate, depending on sitespecific conditions, location and depth of the source and sewer lines, and source vapour concentrations. This study has the limitation that it was conducted in a closed environment, with possibility of limiting downward oxygen diffusion. However, in actual site conditions, the ingress of atmospheric oxygen can be accelerated due to the presence of highly permeable preferential pathways and barometric changes, resulting in non-limiting biodegradation of petroleum hydrocarbon contaminants in the absence of an impermeable barrier, contrary to what was inferred from the study. This in turn reduces the VI risk and may complicate the decision making process. Therefore, this research project strongly recommends considering the role of preferential pathways when developing CSM and VI health risks assessments. This will enable professionals to save time and money at similar contaminated sites and put into motion strategic priorities for remediation projects based on knowledge of the potential risk from VI exposure pathways.

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#### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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