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Diffusion of volatile organic compounds (VOCs) through elastomeric bituminous geomembranes (BGMs)

A.M. Arif^a, F.B. Abdelaal^{b,*}^a GeoEngineering Centre at Queen's-RMC, Queen's University, Kingston ON, Canada, K7L 3N6^b Assistant Professor in Geotechnical and Geoenvironmental Engineering, GeoEngineering Centre at Queen's-RMC, Queen's University, Ellis Hall, Kingston ON, Canada K7L 3N6

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ABSTRACT

Diffusion of volatile organic compounds (VOCs) through a 4.1 mm elastomeric bituminous geomembrane (BGM) is investigated using a dilute aqueous solution of four aromatic hydrocarbons: benzene, toluene, ethylbenzene, and xylenes (BTEX). Due to the very different diffusion characteristics of the BGM components, double-compartment diffusion experiments are separately conducted on the bituminous and non-bituminous components of the BGM to assess their diffusion parameters. A two-layer computer model is developed to obtain the diffusion parameters of the BGM that correlates with the 890-day laboratory diffusion test data obtained for the multicomponent BGM and allows the modelling of transient diffusion. Using contaminant transport modelling, the BGMs performance as a diffusive barrier for different applications is evaluated and compared to different polymeric geomembranes (GMBs). It is shown that using BGM as part of the cover system for a hydrocarbon-contaminated soil landfill or as a vapour barrier below concrete building foundations can decrease the BTEX mass flux through the cover system and the peak concentration of contaminant in the indoor air compared to monolayer polymeric GMBs. It is also predicted to reduce the impact on the aquifer when modelled as a part of the composite liner without holes for a hypothetical solid waste landfill.

1. Introduction

In waste containment applications, geomembranes (GMBs) are used as part of the liner and cover systems of the waste to prevent the migration of contaminants to the surrounding environment. These membranes (typically polymeric) can also be used as vapour barriers to limit the contaminant intrusion into residential or commercial buildings or in redeveloping brownfield sites to limit the soil contaminant vapour intrusion into the indoor airspace (Jones and Rowe 2016). For a well-designed and well-constructed barrier system with a low hydraulic head in which advective transport is limited, or for an intact vapour barrier, diffusion becomes the main contaminant transport mechanism through the GMB (Rowe 1998, 2005; Rowe et al., 1995b, 2004).

Among the different contaminants that require controlling their diffusive migration through geosynthetic barrier systems are the volatile organic compounds (VOCs) (Haxo and Lahey 1988; Mueller et al., 1998; Park and Nibras 1993; Rowe et al., 1995a, 1996). VOCs are identified as hazardous air pollutants (USEPA 2003) and have the potential to contaminate groundwater (Edil 2003) whether existing in the vapour

phase, as in the landfill gas (Challa et al., 1997; USEPA 2005) or in the aqueous phase, as in the landfill leachate (McWatters and Rowe 2009; Rowe et al., 2004). They can also be found in brownfield sites with a history of chemical contamination (DiBattista and Rowe 2020; Jones and Rowe 2016; McWatters and Rowe 2015; Roghani 2018). Among these VOCs, benzene, toluene, ethylbenzene, and xylenes (BTEX) are the most common hydrocarbons that are typically used to examine their diffusion characteristics through different GMBs when used as covers or base liners for landfills or as vapour barrier in brownfield sites (DiBattista and Rowe 2020; Jones and Rowe 2016; Jones et al., 2023; McWatters and Rowe 2009, 2010; McWatters et al., 2016a).

The diffusion of BTEX through polymeric membrane materials such as high density polyethylene (HDPE), linear low density polyethylene (LLDPE), and polyvinyl chloride (PVC) and modified or co-extruded GMBs has been extensively studied by previous investigators (Aminabhavi and Naik 1998a, b, c, 1999a, b, c, d, e, f; Anjana et al., 2023; August and Tatzky, 1984; DiBattista and Rowe 2020; Edil 2003; Eun et al., 2014; Haxo 1990; Jones and Rowe 2016; Jones et al., 2023; Joo et al., 2004, 2005; McWatters and Rowe 2009, 2010, 2015, 2018;

* Corresponding author.

E-mail addresses: 18ama26@queensu.ca (A.M. Arif), fady.abdelaal@queensu.ca (F.B. Abdelaal).<https://doi.org/10.1016/j.geotexmem.2023.07.003>

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McWatters et al. 2016a, b, 2019; Nefso and Burns 2007; Park et al., 2012; Sangam and Rowe 2001, 2005; Touze-Foltz et al., 2013; Xiao et al., 1998; Xie et al., 2013; Yan et al., 2021). This enabled the assessment of the diffusive properties of these contaminants through different types of polymeric GMBs to ensure that their concentrations are within the acceptable limits in the surrounding environment (i.e., air, groundwater, soil). However, none of the previous studies examined bituminous geomembranes (BGMs; Fig. 1) to assess their effectiveness to resist the diffusive transport of contaminants such as VOCs for geo-environmental applications.

A survey conducted by the authors for the recent applications of BGMs in waste containment applications where migration of VOCs could be a valid concern showed their use in rehabilitating and capping old landfills for legacy and non-legacy waste such as the Hervey Range Landfill (Golder 2019; Keys 2021; Townsville 2020) and for municipal solid waste (MSW) such as Jensen Landfill (Golder 2022; Paten 2023; Townsville 2023) in Queensland, Australia. They were also used for capping engineered landfills to contain paper processing waste and inert waste in Petrie (ATCW 2021) and hydrocarbon and other types of waste in Ingham (Warren Hills Landfill; GHD 2020; Hinchinbrook 2023), also in Queensland, Australia. In Canada, they were used in Fort St. John, British Columbia to cap and close the Silverberry Landfill (Titan 2022) which accepted solid non-hazardous and selected solid hazardous wastes (oilfield, forestry and industrial wastes; IAAC 2016). They were also installed in the final cover of a MSW landfill at the Robin Hood Bay waste management facility in St. John's, Newfoundland and Labrador (Coletanche 2023; St. John's, 2023). In the US, they were used in the final cover for a MSW landfill adjacent to Suisun Bay, California (Hajd-Hamou and Breul, 2013). Apart from the cover system, BGMs were used for the extension of sideliner and piggy-back liner for an existing MSW landfill in McLaren Vale, South Australia (Golder 2018; Richardson and Wingrove 2021). However, the applications of BGMs associated with sources of VOCs have not been limited to waste containment. They were also used as a vapour and gas barrier in Canada (Shawinigan, Quebec and Toronto, Ontario), France (Gueugnon), and Ireland (Dublin) to prevent contaminant migration into the residential and commercial facilities developed near former oil fields, landfills, or industrial areas (Coletanche 2022). They were selected for the secondary containment liner system at a gas depot located in Portland, Oregon, to protect the soil and groundwater from contamination in the event of fuel leaks or spills (Daly et al., 2017). The selection of BGMs for all these applications can be attributed to their high mass per unit area, high puncture resistance, relatively high density, low coefficient of thermal expansion and good waterproofing characteristics (Breul et al., 2004, 2006, 2018; Cazzuffi et al., 2010; Daly and Breul 2017, 2021; Daly et al., 2018; Gautier et al., 2002; Giroud 1990; Heibaum et al., 2006; Mafra et al., 2008; Peggs 2008; Rowe and Jefferis 2022; Touze-Foltz et al., 2015). These are all favourable properties for a barrier system to prevent advective transport of contaminants, but not necessarily for preventing diffusive migration of contaminants.

Bitumen as a mixture of oligomeric hydrocarbons (Petersen 2000;

Redelius and Soenen 2015) can be expected to exhibit complex chemical properties towards the diffusion of different hydrocarbons. Solubility and diffusivity of hydrocarbons in bitumen have been studied due to their importance in the solvent-based recovery process in the petroleum industry and for research investigating the premature deterioration of asphalt pavements due to fuel spills (Badamchi-Zadeh et al., 2009; Das and Butler 1996; Diedro et al., 2015; Etminan et al., 2014; Fisher et al., 2000; Giuliani et al., 2009; James et al., 2012; Jamialahmadi et al., 2006; Maruffuzaman and Henni 2014, 2015; Mehrotra and Srcek 1982, 1985, 1988; Merusi et al., 2010, 2011; Richardson 2017; Sheikh et al., 2005, 2006; Schmidt et al., 1986; Tharanivasan et al., 2006; Upreti and Mehrotra 2002; Varet et al., 2013; Yang and Gu 2006, 2007; Yarranton et al., 2021; Zainal et al., 2011; Zhang et al., 2000). These studies conform to the classic "like dissolves like" theory where hydrocarbons such as methane, ethane, propane etc. work as a solvent for bitumen and vice versa. It was also concluded that the solubility and diffusivity of hydrocarbons in bitumen depend on the concentration of the solvent, type of bitumen, temperature, and pressure in the system. Based on these studies, it can be hypothesized that BTEX may be dissolved in the bitumen component of the BGM, and diffuse through it. However, due to the multicomponent structure of BGMs, the different types of bitumen used in their manufacturing, and the difference in the chemical of interest (light hydrocarbon gases or fuel vs BTEX), the results from these studies cannot be directly used to assess the diffusive transport of VOCs through barrier systems involving BGMs.

There is a paucity of research examining the diffusive transport of different contaminants through BGMs since previous research mainly focused on the BGMs chemical durability (Abdelaal and Samea 2023; Esford and Janssens 2014, Samea and Abdelaal 2023a, b; Touze-Foltz and Farcas 2017), physical response (Blond and Breul 2014; Clinton and Rowe 2017; Rogal et al., 2021), and hydraulic transfer by permeation of water only (Bannour et al., 2013; Coppinger et al., 2002; Eloy-Giorni et al., 1996; Lambert and Touze-Foltz 2000; Rocca-Serra et al., 1990). As such, the objectives of this paper are: (1) to experimentally establish the diffusion parameters of BTEX for an elastomeric BGM and (2) to use these diffusive parameters in a computer model to compare the performance of the BGM to polymeric GMBs in controlling the BTEX migration in different applications.

2. Experimental investigation

2.1. Materials

2.1.1. BGM

A 4.1 mm thick elastomeric BGM is investigated in the current study (Table 1). The BGM is reinforced using a polyester nonwoven geotextile (NW-GTX) and a glass fleece layer with mass per unit area of 235 and 50 g/m², respectively, that are fully impregnated with the bitumen. The bitumen used in coating and impregnating the reinforcement layers of this BGM is stabilized with Styrene-Butadiene-Styrene (SBS) copolymer. The top surface is roughened with sand to increase the interface friction of the liner system while the bottom surface retains an anti-root polyester (PET) film. The 0.012 mm thick PET film prevents the BGM from adhering to itself during storage in rolls and protects it from upward root penetration during service (Lazaro and Breul 2014; Samea and Abdelaal 2023a).

In addition to establishing the diffusion characteristics of the as-received multicomponent BGM, the bituminous component (i.e., BGM without the PET film; referred to herein as the modified BGM sample) and the thin PET film were separately examined in the diffusion tests. While the PET film was provided by the BGM manufacturer, the modified BGM sample that only included the bituminous component required manual removal of the PET film.

Different techniques were examined to remove the PET film such as peeling, slicing, and freezing the as-received BGM sample. However, melting the PET film with a torch flame was the only successful method

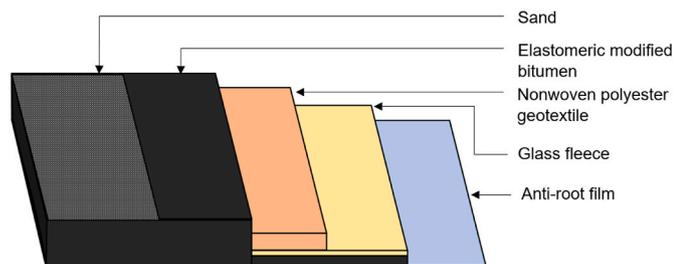


Fig. 1. Schematic of an elastomeric BGM structure (modified from Scheirs 2009); Note: schematic not to scale for enhancing the components of the geomembrane.

Table 1
Properties^a of BGM used for the experimental investigation.

Property	Method	Values	Unit
Designator	–	TERANAP 431 TP 4 M	–
Thickness	ASTM D5147	4.1	mm
Surface mass ^b	ASTM D5261	4.7	kg/m ²
Tensile strength at break (MD/ CMD) ^c	ASTM D7275	29/21	kN/m
Elongation at break (MD/CMD) ^c		55/60	%
Static puncture resistance	ASTM D4833	550	N
Tensile tear resistance (MD/ CMD) ^c	ASTM D4073	940/720	N
Tearing strength resistance (MD/CMD) ^c	ASTM D5884	130/105	N
Low temperature flexibility	ASTM D5147	–26	°C
Water permeability	ASTM E96	2×10^{-14}	m/s
Gas tightness	ASTM D1434	2.76×10^{-5}	m ³ /m ² / day

^a Based on technical datasheets provided by the manufacturer (Siplast 2022).

^b Measured by Samea and Abdelaal (2023b).

^c MD: Machine Direction, CMD: Cross Direction.

that completely removed the PET film. This involved placing the as-received BGM inside the fume hood with the PET film side facing up and cautiously hovering a propane flame from a height around 10 cm above the film which resulted in melting and crumbling of the film into tiny threads. The process started from the left edge of the sample and moved slowly towards its right edge accumulating the melted threads on the right edge. As the sample cooled down, the diffusion specimen was cut from the left portion after closely inspecting it to make sure that the PET film was completely removed without melting the bottom bitumen coat.

To confirm that the properties of the bitumen coat in the modified BGM sample remained the same as the as-received sample, the rheological properties of the modified BGM were examined using a Dynamic Shear Rheometer (DSR) after the removal of the PET film. The rheological indices such as the complex shear modulus (G^*) and the phase angle (δ) (Samea and Abdelaal 2023a) for the modified BGM samples were found to be similar to the as-received samples (Fig. 2). This implies that the melting process of the PET film did not affect the overall bitumen properties.

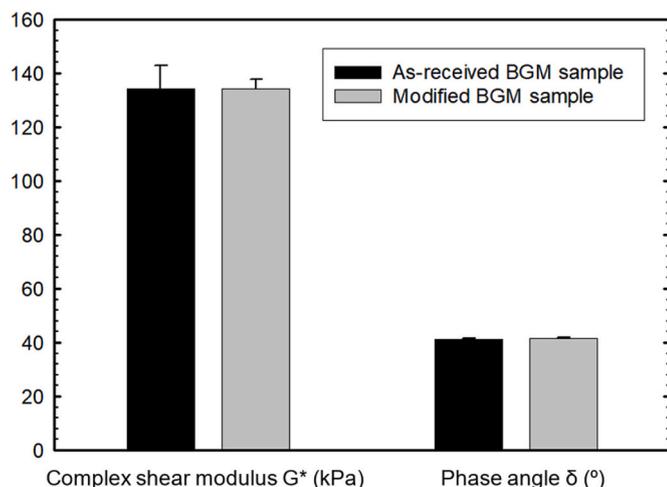


Fig. 2. Comparison of the rheological properties of the bitumen of the as-received and modified BGM samples.

2.1.2. BTEX

Four aromatic hydrocarbons (benzene, toluene, ethylbenzene, and xylenes) were examined in this study. Laboratory grade (99% purity) chemicals were purchased from Sigma-Aldrich, Mississauga, ON, Canada and Chromatographic Specialties Inc, Brockville, ON, Canada. Tests were conducted using dilute aqueous solutions prepared from high-concentration stock solutions. The essential properties of these chemical contaminants are presented in Table 2.

2.2. Test methods

2.2.1. Diffusion test

Diffusion tests (Fig. 3) were performed at room temperature (i.e., 24 ± 1 °C) in specially designed stainless steel double-compartment diffusion cells (e.g., DiBattista and Rowe 2020; Jones and Rowe 2016; McWatters and Rowe 2009, 2010, 2015; Rowe and Barakat 2021). The circular sample (from the as-received BGM, modified BGM or the PET film) was secured between the source and receptor compartments with PTFE (Polytetrafluoroethylene) gaskets and screws were tightened around its perimeter. The junction was then covered with a laboratory-grade silicone sealant. After the silicone seal was completely dry, the entire junction was covered with thick parafilm tapes. Both compartments of the diffusion cell were then filled with double deionized water (DDI) and the source was spiked with the BTEX solution of known concentration to initiate the diffusion process. For all diffusion tests (Table 3), the source compartment volume was two times larger than that of the receptor compartment (i.e., source to receptor fluid volume ratio 2:1) to accommodate the high partitioning of contaminant molecules onto the sample in the source compartment. During the test, samples from the source and receptor compartments were collected through the sampling ports and analyzed at different times.

Control tests were also conducted using the diffusion test setup to identify any significant mass loss either during the sampling events or due to the chemical/cell material interaction. A spiked control test and a blank control test were performed with the same sealing conditions as the diffusion tests described above. The spiked control test did not have any BGM sample, but it was spiked with the BTEX solution and sampled at a specific frequency to monitor the change in concentration. This test was performed to examine the effect of the system on the change in concentration (i.e., whether the BTEX was getting sorbed into the gasket or septa or if there was any mass loss due to leakage through the junction of the two compartments or the sampling ports). For the blank control test, the cell contained a BGM with only DDI water in both the source and receptor compartments. The compartment fluids on either side of the BGM sample were sampled at different times to identify any detectable BTEX concentration in DDI water. This test was performed to examine if the bitumen in the BGM was releasing any BTEX mass in the source or receptor fluids.

Table 2
Properties^a of organic contaminants tested (modified from Sangam and Rowe 2001).

Chemicals	Molar weight (g/mole)	Density (g/cm ³)	Molar volume ^b (cm ³)	Aqueous solubility ^c (mg/l)	Log K_{ow} ^d
Benzene	78.11	0.8765	89.11	1790	2.13
Toluene	92.14	0.8669	106.28	627	2.79
Ethylbenzene	106.17	0.8670	122.46	208	3.13
m-Xylene	106.17	0.8642	122.85	196	3.20
o-Xylene	106.17	0.8802	120.62	204	3.13
p-Xylene	106.17	0.8669	122.47	198	3.18

^a Data selected from Montgomery (2007).

^b Calculated based on chemical density and molar weight.

^c At 25 °C (shake flask-UV spectrophotometry).

^d n-octanol/water partition coefficient.

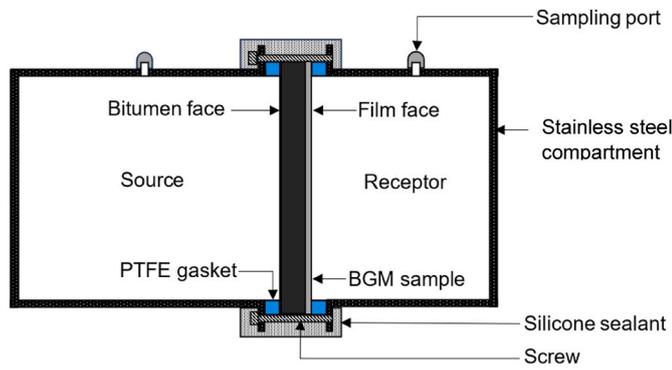


Fig. 3. Schematic of the typical diffusion cell used in the current study; Note: schematic not to scale for enhancing the features of the experimental setup.

2.2.2. Immersion test

Immersion tests were performed with BGM samples of known masses that were immersed in stainless steel cells filled with aqueous solutions of the contaminant. In these tests (Table 3), both surfaces of the BGM were in contact with the permeant with the same BTEX concentration so that the initial concentration of contaminants on either side of the BGM would be the same (i.e., there was no initial concentration gradient between the two faces of the BGM at the beginning of the test). Thus, chemical molecules in the solution could only partition into the BGM. Upon initiation of the tests, samples were collected through the sampling ports and contaminant concentrations in the immersion cells were monitored over time until no significant concentration change for successive samples was observed for 100 days within a percentage error of

±5.0 %.

2.3. Analytical methods

Chemical analysis was performed using solid-phase microextraction (SPME) gas chromatography/flame ionization detection (GC/FID) [Agilent (Santa Clara, California) 7890 B GC with a PAL 80 autosampler]. To minimize the effect of sampling while providing the 2 mL sample required for gas chromatography analyses, small volumes were collected from the test cells and diluted in a solution of methanol and water. Sample volume ranged from 10 to 100 µL, depending on the dilution factor for the sample and detection limit of the GC/FID. Samples were collected using laboratory grade glass syringes and then diluted in 4.0 mL nominal size glass vials sealed with open top cap equipped with Teflon lined septa. Contaminant concentrations were quantified based on a four-point calibration curve developed at the beginning of each set of analyses. This calibration curve was obtained through the analysis of certified standards of known concentrations. It was then verified using a calibration check standard or quality assurance/quality control (QA/QC) sample. The QA/QC samples contained the same analytes but were made from a different source. Blank samples were run for each set of analyses as well, with acceptable concentrations below the detection limit. An internal standard (10 mL of 2 mg/mL surrogate fluorobenzene) was added to each vial to check the consistency of analysis and to measure the percentage of recovery of the analyzed samples.

3. Theoretical modelling of the diffusion parameters for BGMs

The essential diffusion theories were discussed in detail in many

Table 3
Summary of the diffusion and immersion tests.

	GMB tested		Test duration, t (days)	Cell compartment volume (mL)	Contaminant concentration (ppm)						
	As-received	Mass (g)			Benzene	Toluene	Ethylbenzene	m&p-Xylenes	o-Xylene		
Diffusion test	As-received BGM	31	890	Source	241	t = 0	24	23	40	68	28
						t = t	1.8	0.62	0.38	0.58	0.29
	Modified BGM	30	150	Source	243	t = 0	0.46	0.14	0.06	0.09	0.07
						t = t	20	24	26	54	27
	PET film	0.13	200	Source	240	t = 0	3.5	1.5	0.52	0.98	0.60
						t = t	23	26	23	49	22
Immersion test	As-received BGM	29	120	SC	348	t = 0	41	48	52	112	54
						t = t	4.1	1.3	0.65	1.2	0.66
	As-received BGM	32	120	S-DC (bitumen side)	205	t = 0	41	27	34	77	38
						t = t	3.5	0.72	0.20	0.58	0.37
	As-received BGM	58	120	S-DC (film side)	211	t = 0	42	27	34	78	39
						t = t	3.9	0.76	0.29	0.64	0.43
As-received BGM	58	120	D-DC (left side)	207	t = 0	41	48	52	110	53	
					t = t	3.8	1.1	0.52	0.97	0.55	
As-received BGM	58	120	D-DC (right side)	213	t = 0	40	47	51	109	53	
					t = t	3.9	1.1	0.53	0.93	0.57	

previous studies (e.g., Rowe et al., 2004). This section briefly outlines the main concepts used in the current study to establish the diffusion parameters of BGMs.

Contaminant migration through an intact GMB by diffusion is a molecule-activated process and can be considered to occur in steps or stages (Sangam and Rowe 2001). For dilute aqueous solutions, diffusion through the GMB occurs in three fundamental steps, namely adsorption, diffusion, and desorption (Islam and Rowe 2009; Jones and Rowe 2016; McWatters and Rowe 2009, 2010, 2015; Rowe et al., 2004; Sangam and Rowe 2001; DiBattista and Rowe 2020). First the contaminant partitions between the source medium containing the contaminant and the surface of the GMB. When a GMB is in contact with the source medium for sufficient time to reach an equilibrium, there will be a relationship between the final equilibrium in concentration in the GMB, c_g (ML^{-3}) and the equilibrium concentration in the adjacent fluid, c_f (ML^{-3}) (Rowe 1998). This relationship can be described by Henry's law:

$$c_g = S_{gf} c_f \quad (\text{Eq 1})$$

where S_{gf} is the solubility, partitioning or Henry's coefficient (–). For the simplest case, where the permeant does not interact with the membrane material or at low concentrations of contaminants similar to those found in landfill leachate, S_{gf} is a constant for the given contaminant, fluid, GMB, and temperature (Rowe 1998; Sangam and Rowe 2001).

Once partitioned to the inner surface of the GMB, the contaminant then diffuses through the GMB due to the concentration gradient between the source and the receptor medium. The diffusion of contaminant molecules through an intact GMB can be expressed by Fick's first law:

$$f = -D_g \frac{dc_g}{dz} \quad (\text{Eq 2})$$

where f is the mass flux or rate of transfer per unit area ($\text{ML}^{-2}\text{T}^{-1}$), D_g is the diffusion coefficient specific to the contaminant and the GMB (L^2T^{-1}), c_g is the concentration of the contaminant in the GMB (ML^{-3}) and z is the distance parallel to the direction of diffusion (L).

In the transient state, the change in contaminant concentration in the GMB with time t , can be expressed by Fick's second law:

$$\frac{dc_g}{dt} = D_g \frac{d^2 c_g}{dz^2} \quad (\text{Eq 3})$$

In the last step, the contaminant partitions between the outer GMB surface and the receptor medium. This final step is similar to the first one with a reversed process, and can also be described by Henry's law:

$$c_g' = S_{gf}' c_f' \quad (\text{Eq 4})$$

where S_{gf}' is the contaminant partitioning coefficient between the receptor medium and the GMB (–). When the source and receptor fluids are the same, the partitioning coefficient into the GMB can be assumed to be equal to the partitioning coefficient out of the GMB ($S_{gf} = S_{gf}'$) (Jones and Rowe 2016; McWatters and Rowe 2009, 2010, 2015; Sangam and Rowe 2001).

Since the concentration of contaminant inside the GMB is very difficult to measure, the concentration in the fluid on either side of the GMB is analyzed and diffusion parameters are deduced based on measured mass transport from the source to the receptor.

$$f = -D_g \frac{dc_g}{dz} = -S_{gf} D_g \frac{dc_f}{dz} = -P_g \frac{dc_f}{dz} \quad (\text{Eq 5})$$

where P_g is the permeability or permeation coefficient (L^2T^{-1}), which represents the mass transfer across the GMB (Jones and Rowe 2016; McWatters and Rowe 2009; Sangam and Rowe 2001).

According to Rowe (1998), sorption of contaminants can take place within the GMB for GMBs with porous particles of high surface area such as carbon black, and silica gel, or thick polymeric GMB with voids or

other manufacturing defects. In these particular cases, some of the contaminant molecules, which are dissolved in the GMB, can be available to diffuse down the concentration gradient while the remaining molecules are confined to the adsorption sites or the voids within the GMB. This sorption within the GMB is unrelated to the solubility of the chemical contaminant in the GMB material and requires an additional term to Eq. (3) (Rowe 1998) viz:

$$\frac{dc_g}{dt} = D_g \frac{d^2 c_g}{dz^2} - \frac{dS_g}{dt} \quad (\text{Eq 6})$$

where S_g (ML^{-3}) is the mass sorbed per unit volume.

Based on the hydrocarbon-bitumen interaction literature discussed in Section 1, it can be hypothesized that the bitumen may behave as a solvent rather than an absorbent to the BTEX molecules. Additionally, most of the previous studies dealt with BGMs or bitumen as a non-porous material with solubility and diffusion coefficients (i.e., Eq. (3)) similar to hydrophobic GMBs such as HDPE and PVC (e.g., Coppinger et al., 2002; Eloy-Giorni et al., 1996; Rowe 1998). Thus, it is less likely that VOCs are confined to the bitumen adsorption sites and hence the term $\frac{dS_g}{dt}$ may not be applicable to modelling diffusion through BGMs. However, the validity of this assumption will be examined to identify the theoretical model (either Eq. (3) or (6)) that can accurately match and predict the experimental data.

Experimental data of the transient diffusion from the source to the receptor compartments collected during the diffusion experiments can be analyzed using the finite layer analysis program POLLUTE v7 (Rowe and Booker 2004). This software is widely used to infer the diffusion parameters for different materials and model the diffusion of various contaminants through GMBs. For a composite material such as BGM with an asymmetrical structure, a multilayer model may be needed to model the transit diffusion through the entire material (DiBattista and Rowe 2020; McWatters and Rowe 2015, 2018). This modelling method requires the assessment of a separate set of diffusion parameters for the different layers of the GMB to establish a theoretical model in POLLUTE v7 that can match the experimental data for the diffusion through the entire thickness of multilayered GMBs.

As shown in Fig. 1, the individual layers of the BGMs involve the modified bitumen coat, the bitumen-impregnated NW-GTX, the bitumen-impregnated glass fleece mat, and the PET film. Since it was not possible to reproduce or separate the bitumen-impregnated reinforcement layers from the bitumen coat of the BGM, the diffusion tests were conducted on the PET film only and on the modified BGM samples without the PET film (i.e., including the bitumen coat and the impregnated reinforcement layers only), representing the non-bituminous and the bituminous portions of the BGM, respectively. This is based on the assumption that the bitumen may govern the overall diffusion properties of the bituminous component of the BGM since the NW-GTX and the glass fleece layers are porous and completely impregnated with the bitumen.

4. Results and discussions

4.1. Diffusion and immersion tests results of the as-received BGM

While the diffusion tests were conducted using the different BTEX compounds, this section only deals with benzene to validate the theoretical and modelling methods used to assess the diffusion parameters for BGMs. The first attempt to examine the BGM using the diffusion tests involved the as-received BGM sample. This was done to explore the overall behaviour of the material before conducting the separate diffusion tests on the BGM components. The benzene concentration in the source and the receptor compartment of the diffusion test with the as-received BGM was initially monitored for 200 days (Fig. 4). With an actual initial source concentration (c_0) of 24 ppm, the benzene normalized concentration (c/c_0) decreased rapidly to reach 0.1 after 60

days from the initiation of the test and then stabilized at this value for the remaining 140 days. In the receptor, the normalized concentration was increasing but at a very slow rate to reach 0.015 by the end of the monitoring period. These results show an unusual behaviour in which there is a significant decrease in the source concentration while equilibrium in the diffusion test was not achieved for up to 200 days. Additionally, it was not possible to obtain a theoretical model with the assumptions for a single-layer composite material GMB in POLLUTE v7 (i.e., either using Eq. (3) or Eq. (6)) to accurately fit the experimental source and receptor concentrations and hence deduce the diffusion parameters for the as-received BGM.

The rapid decrease in source concentration in Fig. 4 was not attributed to the mass loss of the contaminant in the source compartment due to the cell material/VOCs interaction, or leakage through the junction of the compartments or sampling ports. This is because the control test conducted without the BGM samples showed the retention of the BTEX concentration in the diffusion cells for the entire test duration of two months. Furthermore, the experiments with the BGM (as-received samples) exhibited the same behaviour in replicate diffusion tests with different initial concentrations in the source (ranging from 2 ppm to 98 ppm), spiking the concentrations after the initiation of the tests, and different cell sizes (source to receptor compartment volume ratio 2:1 and 4:1). This also implies that the selected experimental configuration (i.e., initial concentration and cell size) did not affect the diffusion behaviour of the BGM. The potential mass loss due to biodegradation in the source solution over time was also considered. However, when a similar experimental setup was used by McWatters and Rowe (2015) for

long-term BTEX diffusion tests in double-compartment stainless steel diffusion cells for polymeric GMBs, the results did not show any significant mass loss due to biodegradation over approximately 6.5 years of testing. This excludes the possibility of mass loss due to biodegradation for the test result presented herein (Fig. 4) with a much shorter duration than the experiments reported by McWatters and Rowe (2015).

The aforementioned results suggest that benzene may be essentially retained in the bituminous component layer and not released to the receptor. This could be attributed to the multicomponent nature of the BGM and its asymmetric structure with the bituminous component facing the source and PET film facing the receptor side in the diffusion experiments. The other possible explanation for this behaviour is the absorption of the benzene molecules into the bituminous component of the BGM and hence the validity of Eq. (6) in modelling the diffusion through BGMs.

To investigate the effect of the multicomponent asymmetric structure of BGM, diffusion cells were used to perform immersion tests in three different setups (Fig. 5a). The first setup involves immersing the BGM sample with known mass inside a single-compartment (SC) immersion test spiked with a BTEX solution. The setup is similar to that used by Sangam and Rowe (2001) and McWatters et al. (2016a) for the sorption tests with polymeric GMBs and BTEX solution. The second setup (S-DC) involves a BGM sample placed between two compartments spiked with the same initial concentration of BTEX solution, to prevent any permeation through its 4.1 mm thick edges. For both SC and S-DC (bitumen side) the benzene concentration decreased rapidly to reach equilibrium after around 40 days (Fig. 5b). This shows that in the SC immersion test, the permeation through the edges of the BGM was negligible. However, for the S-DC PET film side, the decrease in concentration was much slower than the SC and the S-DC bitumen side, and equilibrium was not reached until 110 days (Fig. 5b).

Such a big difference in the partitioning of benzene to the different surfaces of the BGM was deduced in the third setup (D-DC) in which two BGM samples were welded on their PET film side so that both compartments face the bitumen coat of the double layer BGM sample. The decrease in concentration in the D-DC matched both the SC and the S-DC bitumen side and equilibrium was reached after 40 days. The immersion test results show the capacity of the PET film to obstruct the permeation of contaminant into the BGM. These results also highlight the role of the

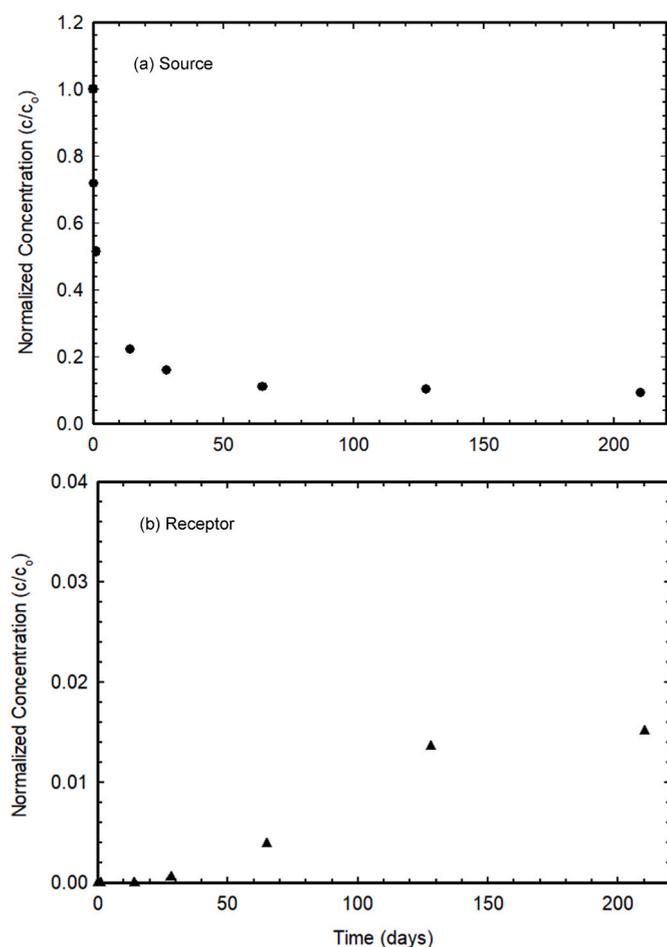


Fig. 4. Concentration changes for benzene during diffusion test at room temperature with as-received BGM in (a) source and (b) receptor; Note: the scale for normalized concentration (c/c_0) changes on the y-axis.

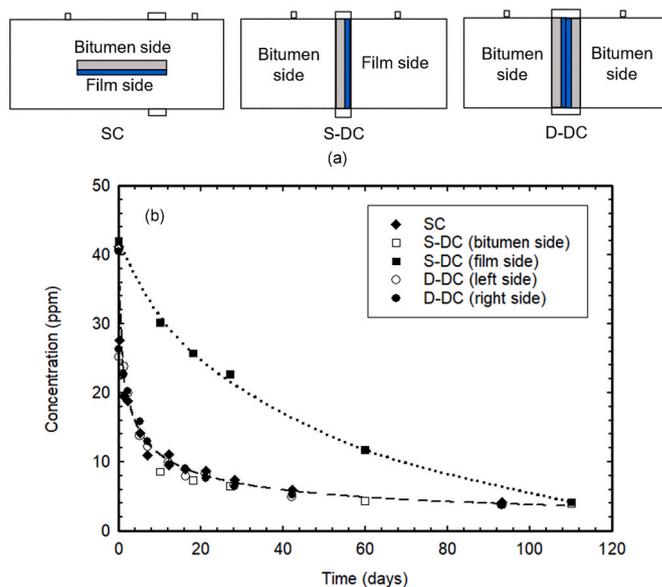


Fig. 5. Immersion tests conducted on the as-received BGM: (a) schematic of the immersion tests setup, single-compartment (SC), single layer-double compartment (S-DC), and double layer-double compartment (D-DC); (b) change in benzene concentration with time during the different immersion test setups.

multicomponent asymmetric structure in the contaminant permeation characteristic of BGM. However, with the data discussed so far, it is still not possible to conclude with absolute certainty whether there is any absorption of the benzene within the bituminous component of the BGM. This point will be further discussed in the next section.

4.2. Assessment of the diffusion parameters of multilayered BGMs

Due to the significant difference between the molecular contaminant migration through the BGM's different layers (bituminous and PET film), the diffusion parameters of the BGM cannot be inferred from modelling the changes in concentration in the source and the receptor of the diffusion cells using the as-received BGM in Fig. 4. Instead, a multilayer model was needed for the BGM to establish these diffusion parameters as discussed in Section 3. Thus, the diffusion tests were conducted on the PET film only and on the modified BGM sample without a PET film, representing the non-bituminous and the bituminous portion of the BGM, respectively.

For the diffusion test conducted using the modified BGM (Fig. 6), the change in concentration in the source showed similar behaviour to the immersion tests SC, D-DC and the bitumen side of S-DC. Although the source solution was in contact with the bituminous face of the BGM (i.e., similar to the diffusion test with the as-received BGM), equilibrium in the diffusion test of the modified BGM was reached within 100 days (Fig. 6). To check any potential mass loss in the system including the biodegradation of BTEX, the concentration of the contaminant in the source and receptor solution was monitored for additional 100 days. Both cell compartment fluids were able to maintain the equilibrium concentration during the additional monitoring period within a percentage error of $\pm 2.0\%$. Mass balance calculation was also performed on the modified BGM diffusion tests following the procedure presented by Jones and Rowe (2016), and the final mass measured in the system (m_f) was compared to the mass added to the system (m_i). A 99% mass balance (as a percentage, $m_f/m_i \times 100\%$) was achieved for benzene while the mass balance of the other BTEX molecules was within 92–99% which is typical for such analysis for BTEX diffusion performed in stainless steel diffusion cells (Jones 2016; Jones and Rowe 2016). These results imply that the contaminant molecules were partitioned to the modified BGM sample without any significant mass loss in the system and they were permeating the bituminous component (i.e., the bitumen coat and the bitumen impregnated NW-GTX and glass fleece layers) without being retained within the bitumen due to absorption. Hence, Fick's second law (Eq. (3)) can be used to model the molecular contaminant migration through the bituminous component of the BGM.

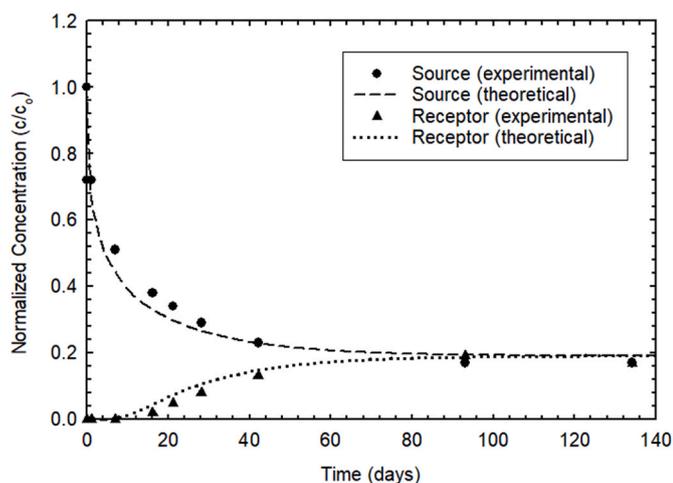


Fig. 6. Concentration changes for benzene in source and receptor during the diffusion test at room temperature with modified BGM (without PET film); Note: Source to receptor volume ratio = 2:1.

To infer the diffusion parameters of the bituminous component of the BGM (i.e., the modified BGM sample), modelling the transient diffusion from the source to the receptor compartments was performed using POLLUTE v7. Since equilibrium was reached in the diffusion tests, the partitioning coefficient (S_{gf}) can be also deduced from the mass balance (McWatters and Rowe 2015; Sangam and Rowe 2001) viz:

$$S_{gf} = \frac{[c_{fo} V_{fo} - c_{ff} V_{ff} - M_c]}{M_g c_{ff}} \times \rho_g \quad (\text{Eq. 7})$$

where c_{fo} and c_{ff} are the initial and final contaminant concentration in the solution (ML^{-3}); V_{fo} and V_{ff} are the initial and final solution volume (L^3); M_g is the initial mass of the BGM (M); M_c is the mass lost to the system as quantified from the control tests (M); ρ_g is the BGM density (ML^{-3}); and c_{ff} is the final concentration of the contaminant in the BGM at equilibrium (ML^{-3}).

The S_{gf} , D_g , and hence the P_g of the modified BGM inferred from modelling the change in concentration in the diffusion tests using POLLUTE v7 are presented in Table 4. The values of S_{gf} inferred from POLLUTE v7 modelling showed an agreement with the value of S_{gf} inferred from the mass balance at equilibrium (Eq. (7)).

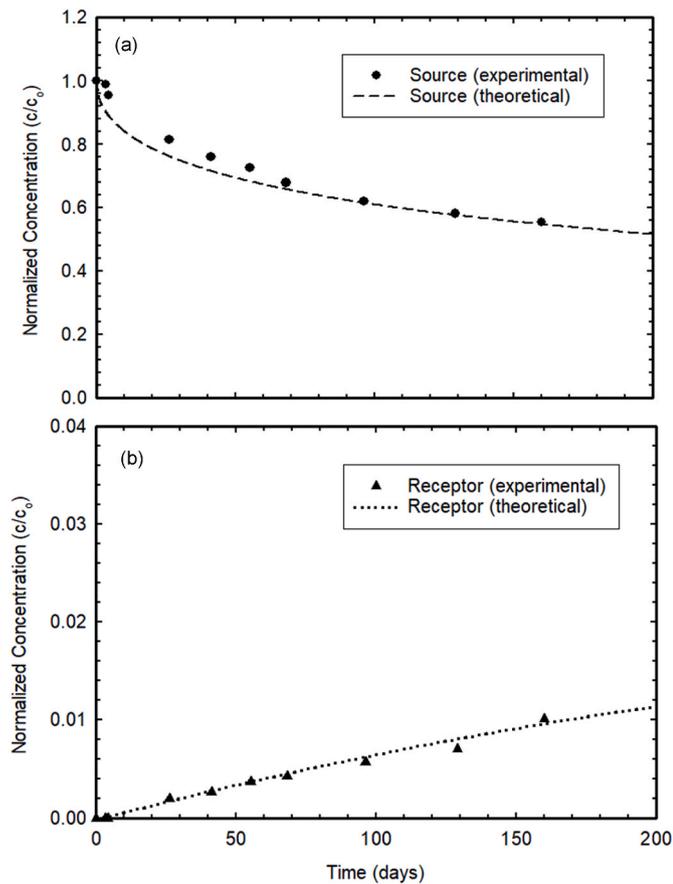
For the PET film (Fig. 7), the change in concentration was very slow in the source (similar to the PET side of S-DC; Fig. 5). In the receptor, the rate of increase in the concentration was also very slow and equilibrium was not reached by the end of the 200 days. This can be attributed to the relatively small D_g and P_g of the PET film (Table 4). Thus, although the source concentration was decreasing due to the relatively high S_{gf} , the contaminant molecules were moving very slowly through the PET film resulting in a very slow increase in receptor concentration. These results show that the slow increase in the receptor concentration of the as-received BGM (Fig. 4) was essentially due to the very low D_g and P_g of the PET film.

Based on the diffusion parameter inferred for the modified BGM and the PET film (Table 4), the D_g and P_g of the bituminous component of the BGM were four orders of magnitude higher than the PET film. For example, the diffusion coefficient, D_g of benzene was $150 \times 10^{-14} \text{ m}^2/\text{s}$ for the modified BGM, whereas for the PET film, it was only $0.003 \times 10^{-14} \text{ m}^2/\text{s}$. As such, it can be hypothesized that when the contaminant molecules enter the BGM matrix from the bitumen side, they may diffuse through the bitumen coat, then the bitumen impregnating the porous reinforcement layers without permeating the polyester fibres of GTX since this represents the path of the least resistance to diffusion. Overall, the plausible diffusion process involves the partitioning of BTEX molecules from the source solution into the BGM through the bituminous face due to the solubility of these contaminants in the bitumen, followed by the migration through the bitumen in the bituminous component layer due to the concentration gradient. As the molecules reach the interface between the bituminous component layer and the PET film, they partition into the film and then migrate at a much slower rate through the PET film layer due to the concentration gradient before getting released into the receptor. In this case, Fick's second law can separately apply to these two different component layers of the BGM (the bituminous component and PET film) with a different set of diffusion parameters.

To validate this interpretation, a two-layer model was established in POLLUTE v7 using the S_{gf} and D_g of the bituminous component and the S_{gf} and D_g of the PET film. This modelling approach was developed based on the method presented by McWatters and Rowe (2015) for multilayered co-extruded polymeric GMBs, where contaminant molecules permeate the different layers of the GMB (with different sets of diffusion parameters) in order. For a BGM, no special consideration was given to the interface between the bituminous component and the PET film since there were no tie-resin layers that were included in the co-extruded GMBs examined by McWatters and Rowe (2015). For this case, the source was modelled as a finite mass top boundary condition with an initial concentration that was similar to the initial concentration of the contaminant in the source solution (i.e., at time = zero) of the diffusion

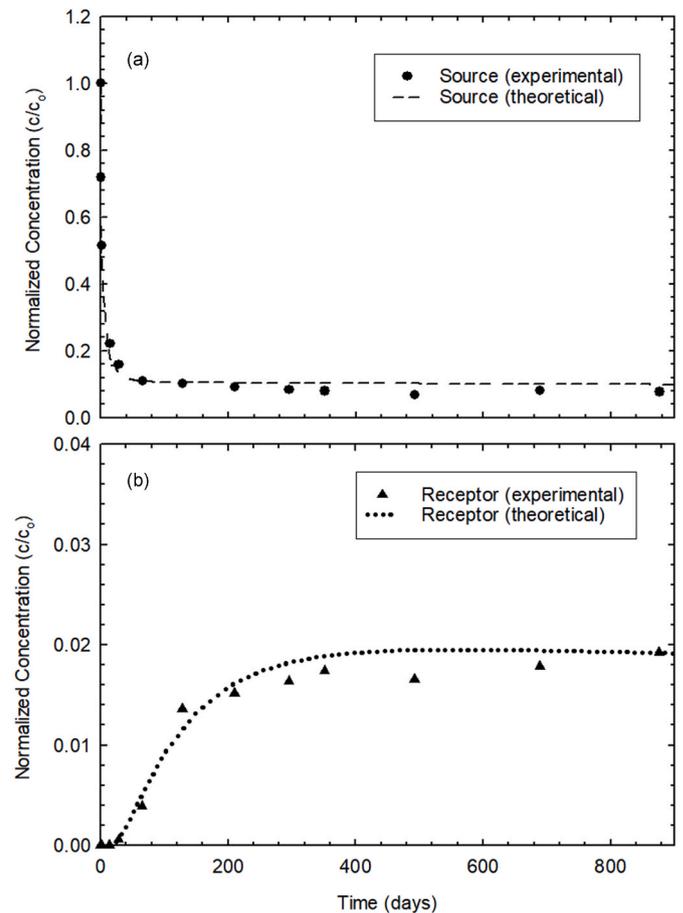
Table 4Partitioning (S_{gf}), Diffusion (D_g), and Permeation (P_g) Coefficients inferred for the BGM components at room temperature.

Contaminant	Modified BGM			PET film			HDPE ^a		
	S_{gf} (-)	D_g (10^{-14} m ² /s)	P_g (10^{-14} m ² /s)	S_{gf} (-)	D_g (10^{-14} m ² /s)	P_g (10^{-14} m ² /s)	S_{gf} (-)	D_g (10^{-14} m ² /s)	P_g (10^{-14} m ² /s)
Benzene	120	150	18,000	250	0.0030	0.75	30	35	1050
Toluene	380	120	45,600	700	0.0025	1.75	100	30	3000
Ethylbenzene	1100	75	82,500	2000	0.0015	3.00	285	18	5130
m&p-Xylenes	1300	73	94,900	2350	0.0013	3.06	347	17	5900
o-Xylenes	1050	71	74,550	1900	0.0011	2.09	240	15	3600

^a Values for a 2.0 mm HDPE reported by Sangam and Rowe (2001).**Fig. 7.** Concentration changes for benzene in (a) source and (b) receptor during the diffusion test at room temperature with PET film; Note: the scale for normalized concentration (c/c_0) changes on the y-axis; Source to receptor volume ratio = 2:1.

test. The reference height of the fluid was taken as the length of the source compartment. The receptor was modelled as a fixed outflow bottom boundary with a base outflow velocity of zero. The base thickness was equal to the length of the receptor compartment, and the base length and width were the diameter of the cell.

The theoretical data obtained from this two-layer model for BGM were compared to the experimental data from the diffusion test on the as-received BGM with all the components and showed that the two-layer model closely matched the experimental data for up to 890 days (Fig. 8). The good fit of the theoretical model to the long-term experimental data implies that the two-layer model captures the molecular contaminant migration through the multicomponent BGM. This also supports the assumptions made during establishing the theoretical modelling discussed in Section 3.

**Fig. 8.** Change in the normalized concentration versus time for benzene in the (a) source and (b) receptor compartments of the diffusion test with the as-received BGM. Fitting lines were obtained using the multilayer theoretical model output in POLLUTE v7; Note: the scale for normalized concentration (c/c_0) changes on the y-axis.

4.3. Comparison of the diffusion of the different aromatic hydrocarbons through BGM

The two-layer modelling discussed for benzene was used to establish the diffusion parameters for all other VOCs (Table 4). Similar to benzene, the source concentrations of toluene, ethylbenzene, m&p-xylenes, and o-xylene, decreased significantly during the diffusion test (Fig. 9). The rate of decrease in the source concentration was in the order of benzene < toluene < ethylbenzene < xylenes. This can be attributed to the increase in their hydrophobicity (Table 2; i.e., the n-octanol/water partition coefficient, $\log K_{ow}$) and hence the increase in their partitioning to BGM (Sangam and Rowe 2001). The trend observed in Fig. 9 agrees with the order of the estimated S_{gf} in Table 4 in which the benzene had the lowest S_{gf} while the m&p-xylenes had the highest values.

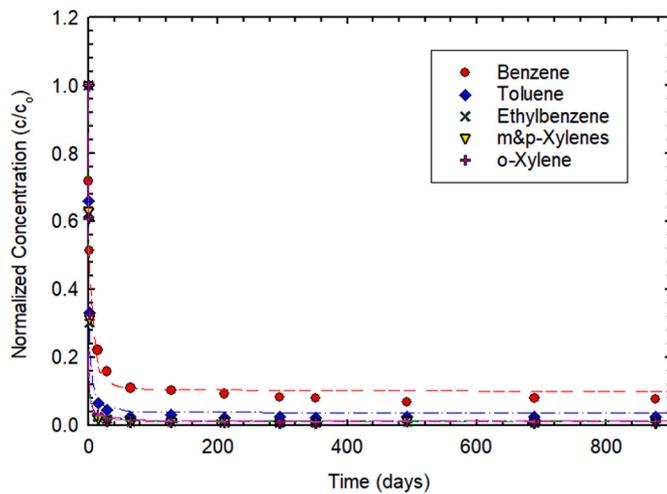


Fig. 9. Concentration changes for BTEX in the source during the diffusion test at room temperature. Fitting lines represent the theoretical model output obtained from POLLUTE v7 for the different contaminants.

For the receptor, the increase in the concentrations was in the order of benzene > toluene > ethylbenzene \approx xylenes (Fig. 10). This can be attributed to the lower molecular weight and molecular volume of benzene and toluene than the other VOCs (Table 2) since the diffusion of the permeant molecule increases with the decrease in permeant molecular weight and volume (Aminabhavi and Naik 1998b; Berens and Hopfenberg 1982; Saleem et al., 1989; Sangam and Rowe 2001). Additionally, due to the high partitioning of ethylbenzene and xylenes, the concentration gradient was lower than benzene and toluene which reduced their flux into the receptor. Thus, the estimated D_g was the highest for benzene, followed by toluene, and ethylbenzene and xylenes had the lowest value for both the bituminous component and the PET film of the BGM (Table 4).

5. Practical applications

Since the diffusion parameters of the BGM were assessed for its different components, these properties cannot be directly compared to other GMB materials (e.g., PE GMBs) established in the literature. Thus, to compare the efficiency of BGMs as a diffusive barrier to PE GMBs, three hypothetical cases of different containment applications were

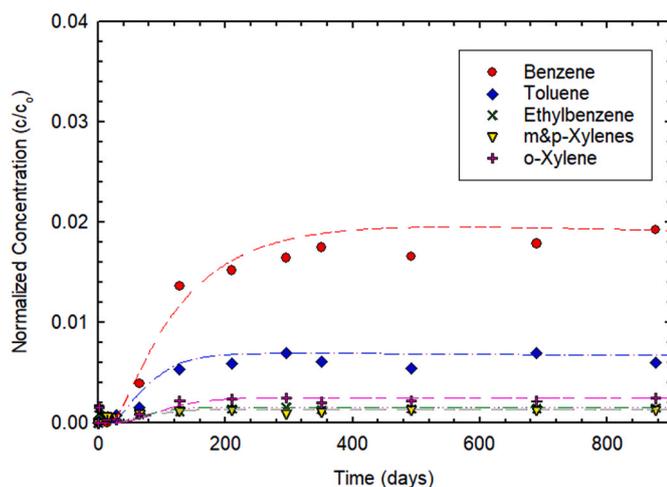


Fig. 10. Concentration changes for BTEX in the receptor during the diffusion test at room temperature. Fitting lines represent the theoretical model output obtained from POLLUTE v7 for the different contaminants.

analyzed based on existing guidelines.

5.1. Case 1: landfill base liner

A hypothetical MSW landfill (Table 5) lined with a single composite liner was examined for two general scenarios: (1) Generic Design Option I of O. Reg. 232/98 (MoE 1998) with a 1.5 mm-thick HDPE GMB (Case A) and (2) an alternative design with a 4.1 mm BGM (Case B). A 7 mm thick geosynthetic clay liner (GCL) was chosen to represent the low hydraulic conductivity layer of the composite liner. Benzene was selected as the main containment given its highest potential health impact among the BTEX. The reference height of the leachate was calculated based on the properties of the landfill and waste (finite mass boundary) provided in Table 4. The leachate collection system was assumed to be operational at all phases and capable of maintaining a constant leachate head above the liner system. The values of different parameters used in the barrier system model are presented in Fig. 11 and Table 6. For a more conservative scenario, the background concentration of contaminant in the groundwater of the receptor aquifer (fixed outflow boundary) was considered zero. The modelling also conservatively neglected any biological decay since it is an attenuation mechanism, and any natural soil with low nitrogen levels shall prevent biodegradation (DiBattista and Rowe 2020).

To evaluate the relative performance of BGM to HDPE GMB as a landfill baseliner with respect to the diffusive migration of BTEX, Cases A and B were modelled in POLLUTE v7 and the impact on the aquifer was assessed for comparison. In both cases, the peak concentration (c_{peak}) at the top of the aquifer (i.e., at the interface of the natural attenuation layer and the groundwater source) was compared to the maximum allowable concentration (c_m). The c_m was calculated as 1.25 $\mu\text{g/L}$ based on O. Reg. 232/98 (MoE 1998) for benzene with a zero-background concentration in the aquifer.

Without any holes or defects in the GMB, the mechanism for contaminant transport from a finite source waste of the landfill to the aquifer is pure diffusion (Rowe et al., 2004). For Case B, c_{peak} in the aquifer was 14% lower for benzene and arrived around 20 years later for the composite liner with a 4.1 mm BGM than that with a 1.5 mm HDPE GMB (Case A) (Fig. 12). While for both GMB materials, c_{peak} was below the c_m , all other conditions being similar, a composite liner with the BGM is expected to lower the impact on the aquifer than with the HDPE GMB considering only pure diffusion.

5.2. Case 2: vapour barrier

Although the current regulations specify low minimum thickness for vapour barriers below concrete building foundations (e.g., 0.15 mm (NYSDOH 2006) and 0.75 mm (USEPA 2008)), BGMs have been used as vapour and gas barriers as discussed in Section 1. Thus, a hypothetical warehouse (Fig. 13; adopted from Jones and Rowe 2016) was modelled in POLLUTE v7 using a 4.1 mm BGM to compare the effectiveness of BGMs as a vapour barrier to maintain the indoor air quality to the published data for other polymeric vapour barriers. For this case, a 100 \times 100 m and 5 m high building was required to be built over a

Table 5
Properties of the landfill.

Properties	Value	Units
Landfill length in direction of groundwater ^a	400	m
Landfill width ^a	100	m
Mass of waste per unit area ^b	250,000	t/ha
Proportion of Benzene in waste ^c	0.014	mg/kg
Initial concentration of Benzene in waste ^c	0.02	mg/L

^a Assumed; landfill length similar to the problem examined by Rowe and Barakat (2021).

^b Considered based on Keele Valley Landfill data (Rowe 1995).

^c Based on leachate characteristics given in OReg 232/98, MoE (1998).

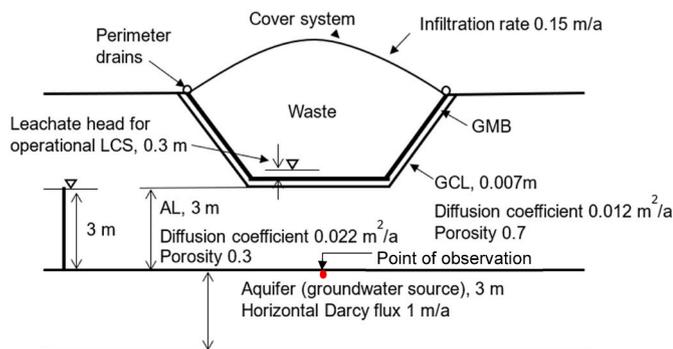


Fig. 11. Schematic diagram of the barrier system for the hypothetical MSW landfill; Note: schematic not to scale for enhancing the components of the barrier system.

Table 6

Different parameters used for the barrier system modelling in POLLUTE v7.

Input Parameters	Values	Justification/reference
Aquifer	Thickness = 3 m Porosity = 0.3 Horizontal Darcy flux = 1 m/a	Based on a conservative estimate by Rowe et al. (2004).
Natural attenuation layer (AL)	Porosity = 0.3 $D_g = 0.022 \text{ m}^2/\text{a}$	
GCL	Thickness = 0.007 m Porosity = 0.7 $D_g = 0.012 \text{ m}^2/\text{a}$ $K_d = 4.4 \text{ mL/g}$	Rowe et al. (2005)
HDPE GMB	Thickness = 0.0015 m $D_g = 1.1 \times 10^{-5} \text{ m}^2/\text{a}$ $S_{gf} = 30$	Sangam and Rowe (2001)
Leachate collection system (LCS)	Design maximum head on liner = 0.3 m; assumed to be functioning at all times	Rowe et al. (2004)
Cover	Infiltration rate = 0.15 m/a	The minimum value permitted by MoE (1998)

K_d = partitioning coefficient [$\text{L}^3 \text{ M}^{-1}$].

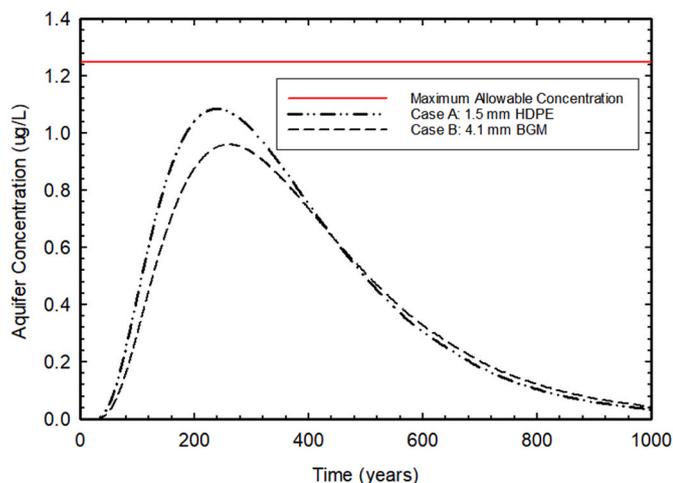


Fig. 12. Benzene concentration changes in aquifer with time for a composite liner with GCL and GMB with no holes or defects; Note: HDPE geomembrane modelled using parameters from Sangam and Rowe (2001), maximum allowable concentration (c_m) calculated based on O. Reg. 232/98 (MoE 1998).

hydrocarbon contaminated soil site (Fig. 13). Thus, a vapour barrier was placed between the subgrade (0.3 m thick) and the floor slab (0.3 m thick) to prevent contaminant migration from the soil to the indoor airspace. The assumed crack porosity of the concrete layer was 0.005

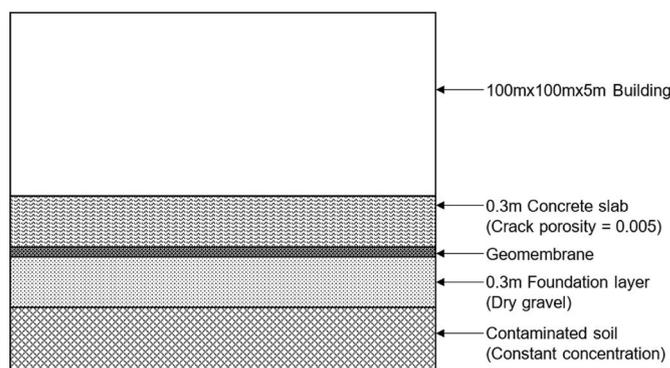


Fig. 13. Schematic diagram of the vapour barrier system for the hypothetical warehouse (based on Jones and Rowe 2016); Note: schematic not to scale for enhancing the components of the vapour barrier system.

and the porosity of the foundation gravel layer was 0.4. To be conservative, it was assumed that the source concentration would remain at the aqueous solubility limit for the specific contaminant as presented in Table 2. Benzene was selected as the critical contaminant for evaluating the worst-case scenario since it has the highest mobility among BTEX through GMBs (Jones and Rowe 2016; McWatters and Rowe 2010; McWatters et al., 2016a; Sangam and Rowe 2001). For the base case, an air exchange rate (AER) of 0.25 h^{-1} was assumed as an additional engineering exposure control measure without any passive ventilation system below the concrete floor. The peak indoor concentration (c_{peak}) was examined at zero air height (i.e., the interface of the concrete slab and the indoor air) since the concentration of the transported contaminant in the indoor air would be the highest at this level (DiBattista and Rowe 2020; Jones and Rowe 2016). The calculated and reported values for c_{peak} of benzene in the air for different types of vapour barriers were compared to the recommended exposure limits (Table 7).

A comparison of the POLLUTE v7 model results for the BGM with the different polymeric vapour barriers reported in the literature (Table 8) shows the effect of the material type (BGM, HDPE, LLDPE, and PVC) on c_{peak} of benzene for this particular case. Comparing the different types of traditional monolayer GMBs considered for this analysis to the BGM, the minimum c_{peak} (0.69 mg/m^3) was achieved for the 4.1 mm BGM. While the big difference in the thickness between the BGM and the other polymeric vapour barriers aided the better performance of the BGM, it still outperformed a 2.0 mm thick HDPE (Table 8). However, the BGM was not able to maintain the c_{peak} below the NIOSH recommended exposure limit (NIOSH REL) of 0.32 mg/m^3 as an 8-h time weighted average (TWA; Table 7). Although the BGM did not meet this criterion for an AER of 0.25 h^{-1} , it was able to maintain c_{peak} below the NIOSH REL STEL requirement (short-term exposure limit in any 15-min sampling period) which was the maximum acceptable value considered by

Table 7

Occupational exposure limits for benzene^a.

Guideline		Exposure limit (mg/m^3)
NIOSH ^b REL ^c (Ca) ^d	TWA ^e	0.32
	STEL ^f	3.2
ACGIH ^g TLV ^h (A1) ⁱ	TWA ^e	1.6
	STEL ^f	8

^a According to USEPA (2012) and CCOHS (2022).

^b NIOSH: National Institute of Occupational Safety and Health.

^c REL: Recommended Exposure Limit.

^d NIOSH classified as potential occupational carcinogen.

^e TWA: Time Weighted Average.

^f STEL: Short-term Exposure Limit.

^g ACGIH: American Conference of Governmental Industrial Hygienists.

^h TLV: Threshold Limit Value.

ⁱ ACGIH classified as confirmed human carcinogen.

Table 8

Peak indoor air concentration for benzene at the interface between the concrete floor and the building air for different vapour barriers at an AER of 0.25 h⁻¹.

Geomembrane	Thickness (mm)	c_{peak} (mg/m ³)	$c_{peak} (GMB) / c_{peak} (BGM)$
PVC	1.02	370 ^a	540
LLDPE	0.76	110 ^b	160
HDPE	0.79	22 ^a	32
HDPE	2.0	5.8 ^c	8.4
BGM	4.1	0.69	1
Co-extruded LLDPE/EVOH/LLDPE	0.53	0.19 ^d	0.28

^a Reported by Jones and Rowe (2016).

^b Modelled using diffusion parameters reported by McWatters and Rowe (2010).

^c Modelled using diffusion parameters reported by Sangam and Rowe (2001).

^d Modelled as a multilayer system based on the best estimate values provided by McWatters and Rowe (2015).

Jones and Rowe (2016). BGM also maintained c_{peak} below the ACGIH TLV-TWA and ACGIH TLV-STEL requirement (Table 7) which also matches the maximum exposure limits for benzene in many Canadian jurisdictions (CCOHS 2022). For the BGM to meet the NIOSH REL of 0.32 mg/m³ as an 8-h TWA for the simulated warehouse, an AER of 0.6 h⁻¹ or higher is required (Fig. 14).

The BGM was also compared to a 0.53 mm co-extruded LLDPE/ethylvinyl alcohol (EVOH)/LLDPE vapour barrier (McWatters and Rowe 2015). The LLDPE/EVOH/LLDPE was modelled as a multilayer system in POLLUTE v7 for the warehouse examined, based on the best-estimate diffusion parameters provided by McWatters and Rowe (2015). According to the modelling output, the c_{peak} for the LLDPE/EVOH/LLDPE vapour barrier was 28% of the c_{peak} obtained using the 4.1 mm BGM (Table 8). Additionally, it was the only vapour barrier that was able to maintain benzene concentrations at the interface below the NIOSH REL at an AER of 0.25 h⁻¹.

Based on the foregoing results, the 4.1 mm BGM reduced the c_{peak} significantly compared to the traditional monolayer HDPE, LLDPE and PVC GMBs. While the BGM was only outperformed by the 0.53 mm co-extruded LLDPE/EVOH/LLDPE vapour barrier, both materials are expected to perform as a very efficient vapour barrier for the simulated warehouse and can meet the different maximum exposure limits presented in Table 7.

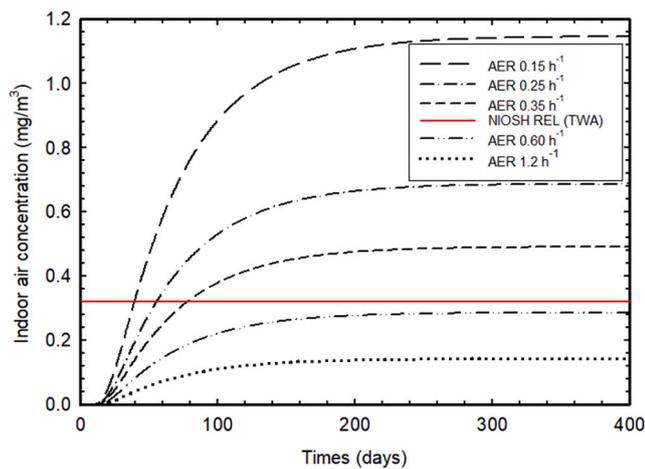


Fig. 14. Peak indoor air concentration for benzene at the interface between the concrete floor and the building air in a simulated warehouse with a 4.1 mm BGM installed as a vapour barrier.

5.3. Case 3: landfill cover system

A hypothetical landfill containing hydrocarbon-contaminated soil was examined to assess the relative performance of BGM as a cover liner material with respect to the diffusive migration of contaminants from the contaminated soil to the atmosphere. The problem examined is similar to the Arctic landfill modelled by McWatters et al. (2016a) using the same cover system of the Resolution Island landfill, Nunavut (ASU 2005, 2008, 2013, 2019) to contain contaminated soils. The hypothetical cover system (Fig. 15) comprises a GMB underlain by a 2 m soil cover. The porosity of the cover soil layer was assumed to be 0.4. For the analysis, the worst-case scenario with a dry cover was considered which implied that the soil layer was ineffective against the diffusive migration of the contaminant. Thus, the contaminant flux through the cover system would depend on the diffusion properties of the GMB and the concentration of contaminant in the contaminated soil in the landfill (McWatters et al., 2016a).

To quantify the effectiveness of the cover system, the steady-state (i.e., constant source and receptor concentration) benzene mass flux was calculated to assess the diffusive migration of VOCs through the intact liner system for different types of GMBs including a BGM. McWatters and Rowe (2009) suggested a typical landfill gas concentration of 74 g/m³ for benzene based on the data compiled from various landfill sites. This value was used as the initial concentration of the contaminant in the case examined herein.

According to McWatters and Rowe (2015), while multilayered modelling is required to assess the diffusion parameters of multilayered GMBs for the correct modelling of transient diffusion or time-dependent behaviour, for a simplified problem of a steady-state case, a single set of diffusion parameters can be calculated viz:

$$P_g^* = \frac{\sum_i t}{\sum_i \frac{t}{P_g}} \quad (\text{Eq 8})$$

$$S_{gf}^* = \frac{\sum_i S_{gf} t}{\sum_i t} \quad (\text{Eq 9})$$

$$D_g^* = \frac{P_g^*}{S_{gf}^*} \quad (\text{Eq 10})$$

where P_g^* is the harmonic mean of the permeability coefficient of the layers (L^2T^{-1}); S_{gf}^* is the weighted average of the partitioning coefficients of the layers (-); i is the number of layers and t is the layer thickness; D_g^* is a single-layer diffusion coefficient for the multilayer GMB (L^2T^{-1}) calculated from P_g^* and S_{gf}^* based on Eq. (10).

This method was used to calculate a single set of diffusion parameters for the multicomponent BGM for the landfill cover case. The mass flux of benzene through the cover system with BGM was calculated using Eq. (5) and was compared to different polymeric GMBs (Table 9). Among all the GMBs considered, the minimum flux ($1.4 \text{ g m}^{-2} \text{ a}^{-1}$) was achieved

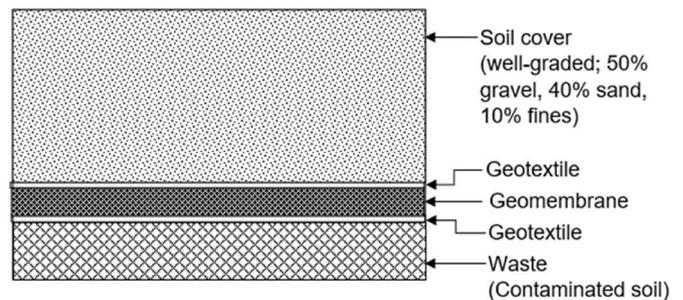


Fig. 15. Cross-section of the hypothetical cover system with a geomembrane and well-graded cover soil for a contaminated soil landfill; Note: schematic not to scale for enhancing all the components of the cover system.

Table 9

Calculated mass flux of benzene through the cover system in a hypothetical landfill containing hydrocarbon-contaminated soil.

Geomembrane	Thickness (mm)	flux (g.m ⁻² .a ⁻¹)	flux _(GMB) /flux _(BGM)
PVC	0.76	440 ^a	310
LLDPE	1.14	110 ^b	79
HDPE	1.5	20 ^a	14
BGM	4.1	1.4 ^c	1

^a Calculated using diffusion parameters reported by McWatters et al. (2016a).

^b Calculated using diffusion parameters reported by Jones and Rowe (2016).

^c Using single set of diffusion parameters.

for the BGM. This shows a 14-fold reduction in the benzene mass flux relative to the 1.5 mm HDPE, a 79-fold reduction relative to the 1.14 mm LLDPE and a 310-fold reduction relative to the 0.76 mm PVC GMBs. Based on these results for this specific case, and considering only diffusive transport, BGM can be expected to perform as a very good cover liner material for landfills with remediation purposes.

6. Conclusions

The diffusion properties of a 4.1 mm elastomeric BGM for BTEX migration were investigated to explore its performance as part of the barrier system to limit the diffusion of VOCs. Using the diffusion parameters deduced experimentally for the BGM, its performance as a diffusive barrier was compared to polymeric GMBs using POLLUTE v7 for three different hypothetical cases that only consider pure diffusion. For the test conditions and materials examined, the following conclusions were reached.

1. To assess the diffusion parameters for the multicomponent BGMs, it was necessary to examine the bituminous and non-bituminous (i.e., PET film) separately in the double-compartment diffusion tests. This is due to the significant difference in the BTEX migration through these different components of the BGMs. For instance, for benzene, the estimated values of the permeation coefficient, P_g for the bituminous and non-bituminous components were $18000 \times 10^{-14} \text{ m}^2/\text{s}$ and $0.75 \times 10^{-14} \text{ m}^2/\text{s}$, respectively.
2. A two-layer system in POLLUTE v7 was needed to model the transient diffusion through the as-received BGM. The two-layer model established using the S_{gf} and D_g of the bituminous component and the S_{gf} and D_g of the PET film captured the change in concentration in the receptor and source compartments of the diffusion experiment conducted using the as-received BGM for up to 890 days.
3. A comparison of the diffusion of the different aromatic hydrocarbons examined through BGM showed that the benzene had the lowest S_{gf} while the m&p-xylenes had the highest values. For the D_g , benzene had the highest values followed by toluene and ethylbenzene, and xylenes had the lowest values. This order was consistent with the findings of previous studies that examined BTEX diffusion through polymeric GMBs since the contaminant chemical properties such as their hydrophobicity, molecular weight and molecular volume play a critical role in contaminant migration through GMBs.
4. An alternative to the Generic Design Option I of O. Reg. 232/98, with a 4.1 mm BGM reduced the peak concentration of benzene in the aquifer by 14 % and increased the time of arrival of the peak concentration by about 20 years relative to a 1.5 mm HDPE GMB.
5. For a hypothetical case of a warehouse constructed on a hydrocarbon-contaminated site, the 4.1 mm BGM vapour barrier outperformed 2.0 mm HDPE, 0.76 mm LLDPE, and 1.02 mm PVC vapour barriers and was able to maintain the indoor air concentration below the NIOSH REL with an air exchange rate of 0.6 h^{-1} or higher. A 0.53 mm co-extruded LLDPE/EVOH/LLDPE was the only polymeric vapour barrier that outperformed the BGM and was able

to maintain the benzene concentrations at the interface below the NIOSH REL at an AER of 0.25 h^{-1} .

6. For a hypothetical steady-state mass flux case for a cover system in a contaminated-soil landfill, an approximate single set of diffusion parameters of multilayered BGM was calculated based on the simplified method proposed by McWatters and Rowe (2015). The 4.1 mm BGM showed a 14-fold reduction relative to the 1.5 mm HDPE GMB, a 79-fold reduction relative to the 1.14 mm LLDPE GMB, and a 310-fold reduction relative to the 0.76 mm PVC GMB, in the benzene steady-state mass flux through the cover system.

The results in this paper provided an evaluation of the diffusion parameters of BTEX through an intact elastomeric BGM. The results presented are related to the particular BGM and contaminants examined. Independent verification is needed before extrapolating the results to other types of BGMs or other contaminants. Furthermore, the chemical interaction between these contaminants and the BGM was not examined since the scope of this paper was only related to the diffusive transport of contaminants. Evaluation of the BGM performance as part of barrier systems requires examining many other critical factors affecting the GMB performance such as the chemical durability, physical response, and interface shear behaviour (Rowe et al., 2004) that were not considered in this study.

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. The authors declare the following financial interests/personal relationships which may be considered as potential competing interests.

Data availability

Data will be made available on request.

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