

Volatile organic compound diffusion and sorption coefficients for a needle-punched GCL

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ABSTRACT: Test apparatus designed to examine volatile organic compound (VOC) diffusion through geosynthetic clay liners (GCLs) are described, together with procedures for evaluating the relevant diffusion and sorption parameters. Test results show that the rate of contaminant migration proceeded through the hydrated GCL in the decreasing order of DCM and DCA > benzene > TCE and toluene. This was attributed to varying degrees of sorption of DCA, benzene, TCE and toluene to the geotextile component of the GCL as well as to the bentonite present in the GCL. Diffusion coefficients deduced from VOC diffusion testing conducted on the GCLs at confining pressures lower than approximately 10 kPa range from 2×10^{-10} m²/s to 3×10^{-10} m²/s. This is generally lower than those reported in the literature for compacted clay liner materials. Assessments of the environmental protection afforded by a landfill liner require that all underlying soil and geosynthetics components be considered in landfill contaminant migration assessments. The results in this paper provide some of the first published data for laboratory GCL diffusion and sorption coefficients, required to perform contaminant migration assessments for five VOC contaminants commonly found in municipal solid waste leachate.

KEYWORDS: Geosynthetics, GCL, Diffusion, Sorption, Contaminant transport, Volatile organic compounds

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1. INTRODUCTION

Leachate generated by municipal solid waste (MSW) landfills contains a variety of chemical compounds, including low concentrations of volatile organic compounds (VOCs). Even at low concentrations VOCs may pose a risk to groundwater quality and human health if allowed to migrate into the subsurface. Regulatory agencies such as the Ministry of the Environment of Ontario, Canada (MOE 1998), require that the design of any landfill liner system include an assessment of contaminant migration through the proposed liner system, and of potential impacts on groundwater near the landfill. For any contaminant this assessment requires, among other things, estimates of diffusion and sorption coefficients for the liner system of interest (Rowe 1998). Although there has been research investigating the contaminant migration of VOCs through various types of traditional liner system, such as compacted clay liners (Barone *et al.* 1992; Kim *et al.*

2001) and geomembranes (Park and Nibras 1993; Rowe *et al.* 1995a; Sangam and Rowe 2001), there is a lack of experimental data on the diffusion of VOCs through GCLs and the sorption that may occur. These data are also necessary to properly perform equivalency estimates of GCLs with compacted clay liners (CCLs) using numerical methods, as discussed by Rowe (1998) and Foose *et al.* (1999).

The objective of this paper is to examine the diffusion of several aromatic and chlorinated non-polar VOCs through a needle-punched nonwoven GCL. The methodology and procedures are developed for the range of low concentration levels of VOCs observed in MSW landfills. Estimates of diffusion coefficients for the VOCs examined are presented. As sorption onto the geotextile component of GCLs has not previously been examined, particular emphasis is placed on evaluating the influence of VOC sorption onto the geotextile with respect to the interpretation of the diffusion test results.

2. BACKGROUND

2.1. Volatile organic compounds (VOCs)

VOCs such as those examined in this study (dichloromethane (DCM), 1,2 dichloroethane (DCA), trichloroethene (trichloroethylene, TCE), benzene and toluene) are found in a variety of industrial and commercial applications as degreasers, as solvents, and in the manufacturing of other organic chemicals (Montgomery and Welkom 1990). Although most MSW facilities currently do not generally accept liquid waste that would contain large quantities of VOCs, many do accept contaminated soil and other waste that may possibly contain small amounts of VOCs (NSDEL 1994). Rowe (1995) examined five Ontario, Canada, MSW landfill leachates and found some or all of these VOCs present at concentrations generally less than 10 mg/l (see Table 1). The presence of VOCs in landfill leachate has also been reported by Farquhar (1989) and Alker *et al.* (1993).

However, even at below 10 mg/l, these VOC concentrations are often above drinking water guidelines set by regulatory agencies (see Table 1: MOE 2002; EPA 2002), and hence represent a possible risk to the environment if allowed to 'escape' unattenuated from a MSW landfill.

The physical properties of the VOCs investigated in this study vary, partly because of their chemical structure. Benzene and toluene are aromatic hydrocarbons with a six-member ring of carbon atoms in which there are alternating double and single bonds between the carbon atoms (Ebbing 1993). DCM, 1,2 DCA and TCE are aliphatic hydrocarbons in which two or three hydrogen atoms in the original structure have been replaced by chlorine atoms. The water solubilities of these compounds vary over an order of magnitude (Table 2), but all are generally considered non-polar. A summary of the various chemical properties of these VOCs is provided in Table 2.

Table 1. VOC landfill leachate concentrations ($\mu\text{g/l}$) for five Ontario landfills (Rowe 1995) compared with United States Environmental Protection Agency (EPA, 2002) and Ontario Ministry of Environment (MOE, 2002)

Compound	Regulatory guidelines		Landfill				
	EPA MCL ^(a) ($\mu\text{g/l}$)	ODWO ($\mu\text{g/l}$)	A	B	C	D	E
Dichloromethane (DCM, CH_2Cl_2)	5	50	<0.4–3700	ND ^(b)	0.3–2300	215–7100	ND-8300
1,2 Dichloroethane (1,2 DCA, $\text{C}_2\text{H}_4\text{Cl}_2$)	5	5	ND-230	NT ^(c)	11–2080	ND-900	NT
Trichloroethene (TCE, C_2HCl_3)	5	50	<1.9–79	ND	1–80	ND-<230	ND-110
Benzene (C_6H_6)	5	5	ND-57	ND-3.1	7–238	<0.1–25	1.8–590
Toluene (C_7H_8)	1000	N/A ^(d)	3.2–1600	120–600	2–7000	485–1821	350–5900

^(a)MCL: maximum permissible level of contaminant in water that is delivered to any user of a public water system (EPA 2002).

^(b)ND: below detection limit.

^(c)NT: not tested for.

^(d)N/A: not applicable.

All landfills given in Table 1 above commenced operation between 1972 and 1983 and are mainly MSW landfills (both active and closed), although some may have accepted hazardous waste in the past.

Table 2. Volatile organic compound properties

Compound	Specific density (g/ml)	Solubility in water (g/l)	Vapour pressure (kPa)	Octanol–water partition coefficient, $\log K_{ow}$	Dielectric constant	Aqueous diffusion coefficient at infinite dilution (m^2/s)
DCM	1.32 (20°C)	13.0–19.4	~60	1.15	8.9	1.26×10^{-9}
1,2 DCA	1.25	8.0–8.3	~9	1.47	10.4 (20°C)	1.08×10^{-9}
TCE	1.46 (20°C)	1.1–1.5	~10	2.42	3.3 (28°C)	0.99×10^{-9}
Benzene	0.87	1.7–1.8	~16	2.13	2.3 (20°C)	1.16×10^{-9}
Toluene	0.86	0.5–0.6	~4	2.69	2.4	0.97×10^{-9}

Data sources:

Specific density and solubility from Montgomery and Welkom (1990).

Vapour pressure and octanol–water partition coefficient from Schwarzenbach *et al.* (1993).

Dielectric constant from Lide (1995).

Aqueous diffusion coefficient at infinite dilution from Yaws (1995).

All properties are at 25°C unless otherwise indicated.

2.2. Contaminant transport

Modelling of contaminant transport through porous media has been generally described by several researchers over the years (Bear 1972; Freeze and Cherry 1979). Rowe and Booker (1984) developed a model for predicting one-dimensional contaminant transport through soils of finite thickness and to account for realistic landfill parameters such as surface boundary concentrations changing with time (as is the case for MSW landfills). Rowe *et al.* (2000) applied this model to predict one-dimensional contaminant transport for a single reactive solute (no degradation) through a saturated GCL, which utilises the following equation:

$$n_t \frac{\partial c}{\partial t} = \left(n_t D_t \frac{\partial^2 c}{\partial z^2} - n_t \bar{v} \frac{\partial c}{\partial z} \right) - \rho K_d \frac{\partial c}{\partial t} \quad (1)$$

where c is the concentration in the GCL at depth z and time t [ML^{-3}]; n_t is the total porosity of the GCL [-]; D_t is the diffusion coefficient deduced from the total porosity [L^2T^{-1}]; \bar{v} is the average linearised groundwater velocity [LT^{-1}]; ρ is the dry density [ML^{-3}]; and K_d is the partitioning coefficient [M^{-1}L^3]. Lake and Rowe (2000b) have shown that, for thin liner systems such as GCLs, it is the product $n_t D_t$ that controls the flux through the GCL, and hence by using the total porosity of the GCL (an easily measured parameter) the diffusion coefficient deduced from the total porosity will provide good fits to the experimental data from diffusion tests.

If the Darcy velocity, v_a ($= n_t \bar{v}$), is relatively low through a soil, as is often the case for MSW landfill liners, the diffusive migration of contaminants may be the dominant transport mechanism through the liner system. Diffusion, the migration of a contaminant from areas of high concentration to areas of low concentration, has been observed as a significant transport process through low hydraulic conductivity natural clay deposits (Goodall and Quigley 1977) as well as low hydraulic conductivity compacted clay liners (King *et al.* 1993). As shown by Shan and Daniel (1991), Ruhl and Daniel (1997), Petrov and Rowe (1997) and Jo *et al.* (2001), the hydraulic conductivity of a GCL can be similar to or lower than that of a compacted clay liner (provided similar testing conditions are employed). Thus the diffusive migration through GCLs will be as important as that for compacted clay liners. Therefore it is necessary to establish estimates of VOC diffusion coefficients for input into contaminant transport models to allow equivalency assessment of different landfill liner designs in relation to potential contaminant transport (Rowe 1998).

Volatile organic diffusion testing of clayey soil for use in landfills has been performed by numerous investigators, including Johnson *et al.* (1989), Barone *et al.* (1992), Myrand *et al.* (1992), Donahue *et al.* (1999) and Kim *et al.* (2001). Lo (1992) performed hydraulic conductivity testing with GCLs and monitored 1,4 dichlorobenzene (DCB) effluent concentrations during flexible wall hydraulic conductivity testing. The effective

diffusion coefficient for dichlorobenzene for a Claymax GCL was reported as $9.8 \times 10^{-11} \text{ m}^2/\text{s}$. Other DCB results are reported by Lo (1992) for GCLs containing organically modified bentonites (1.2 to $1.4 \times 10^{-10} \text{ m}^2/\text{s}$). However, apart from this, to the authors' knowledge no other research has been reported specifically relating to laboratory diffusion and sorption of VOCs through GCLs.

3. GCL VOC DIFFUSION TESTING

3.1. GCL VOC diffusion testing apparatus

Past experiences of others in relation to VOC diffusion testing with natural clayey soil (Barone *et al.* 1992; Donahue *et al.* 1999) and geomembranes (Rowe 1998; Sangam and Rowe 2001) had shown that the type of material chosen for a diffusion testing apparatus will depend *inter alia* on the chemical and physical properties of the VOC being tested. As a mixture of both aromatic and chlorinated compounds was to be used for GCL diffusion testing, glass was deemed to be the most appropriate choice of material to minimise the potential for sorption or reaction with the contaminants.

The specified volume GCL diffusion test concept discussed by Rowe *et al.* (2000) was adopted for VOC diffusion testing. In this type of diffusion test the GCL sample was restricted to hydrating to a specified volume (i.e. height). One challenge of designing a glass cell for specified volume diffusion testing is that the stresses applied to the glass cell by the hydration procedure must be less than that of any tensile resistance of connections or breaking stresses in the glass or fritted discs used in the diffusion cell. Preliminary work at swell pressures greater than approximately 25 kPa resulted in breakage of the glass apparatus during hydration of GCLs. For this reason, the VOC diffusion tests reported herein were performed at bulk void ratios of 4.1 and 4.6 and estimated swell pressures of less than about 10 kPa (from Lake and Rowe 2000a). Thus the results can be considered an upper bound to the diffusion coefficient at lower void ratios (relative to those expected in the field), with the effect of void ratio on diffusion coefficients having been discussed by Lake and Rowe (2000b). As shown in Figure 1, a variety of parts were utilised to construct the VOC diffusion cell. This cell essentially consisted of a 70 mm internal diameter glass cylinder (1) with two thick flanges (1a and 1b), which provided a contact surface onto which to glue the glass plates (2a and 2b). Glass supports (3a and 3b) were used in combination with glass fritted discs (4a and 4b) to restrict the hydrated volume of the GCL. The VOC diffusion cell shown in Figure 1 was assembled as described by Lake (2000).

GCL samples were hydrated in the diffusion cell by adding deionised distilled water (DDW) to the receptor reservoir (5a) of the diffusion cell through the receptor sampling port (6). This was accomplished by attaching flexible plastic tubing to the sampling port (6) and applying a hydrating head of approximately 200 mm to

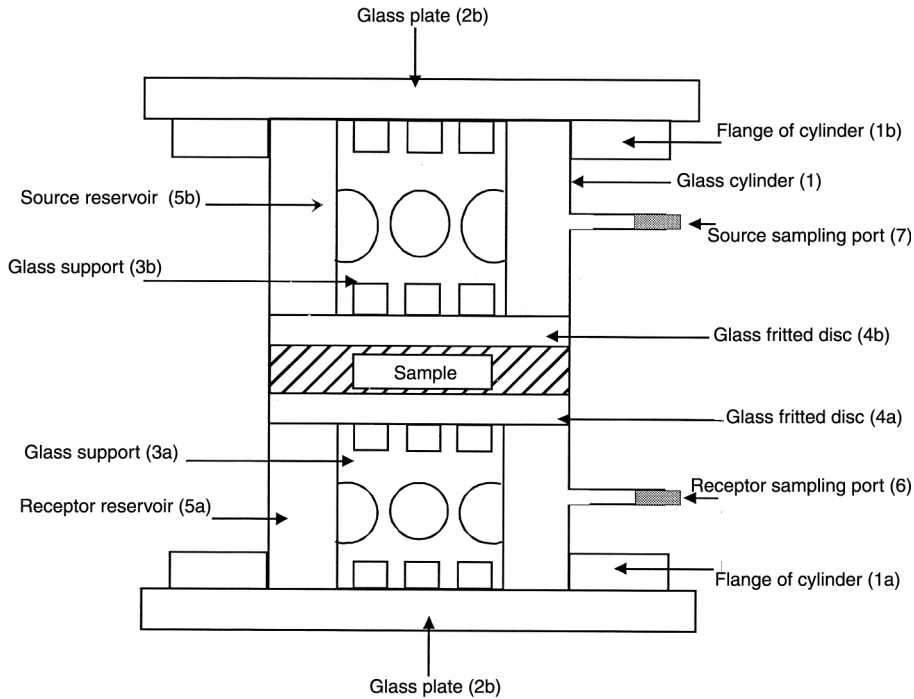


Figure 1. Schematic of VOC GCL diffusion apparatus (not to scale)

the receptor reservoir. After significant uptake of water by the GCL had ceased, 10 mm of DDW was placed above the sample, in the source reservoir. Based on the time required to reach chloride equilibrium for the inorganic GCL diffusion tests discussed by Rowe *et al.* (2000), the GCL was allowed to hydrate for at least 45 days prior to the diffusion test. After this time period, the 10 mm of water from the source reservoir (5b) was removed and replaced with the desired contaminant solution. The source and receptor sampling ports were sealed with a Teflon[®]-lined septum throughout the diffusion test. The test configuration ensured that there was no hydraulic gradient during testing.

Initial source solution concentrations for diffusion tests ranged from 3.0 to 3.2 mg/l for chlorinated VOCs and 0.9 to 1.1 mg/l for aromatic hydrocarbons. To commence diffusion testing, the source solution was transferred to the source reservoir as quickly as possible, with care taken not to induce aeration of the solution. Samples were immediately taken in the source solution and receptor solution to establish initial concentrations of each VOC contaminant. Daily sampling of both the source and receptor concentrations were performed by injecting a glass Gastight Hamilton Syringe (100 µl) into the source and receptor reservoir through the Teflon-lined septum inserted into the sampling ports (6 and 7).

Analysis of VOC concentrations in the source and receptor solutions was performed using a 3800 Varian gas chromatograph equipped with a Varian Saturn 2000 mass spectrometer detector. Complete details of the diffusion testing procedures and analytical methods used for analysis are given by Lake (2000).

3.2. GCL VOC diffusion coefficient estimation

VOC diffusion coefficients can be estimated by solving Equation 1 (see Rowe *et al.* 1995b) in combination with finite mass boundary conditions (see Rowe *et al.* 1995b) for the test set-up. Finite mass refers to the fact that the concentration is changing throughout time at both boundary conditions, owing to mass transfer through the GCL and any sampling of the laboratory tests. Mathematically, the concentration at any time in the source (contaminated) compartment, $c_t(t)$, is given by Rowe *et al.* (1995b) as

$$c_t(t) = c_0 - \frac{1}{H_r} \int_0^t f_t(t)dt - \frac{q_c}{H_r} \int_0^t c_t(t)dt \tag{2}$$

where c_0 is the initial concentration in the source solution [ML^{-3}]; H_r is the height of source fluid (volume of source fluid per unit area) [L]; $f_t(t)$ is the mass flux of contaminant into the soil at any time t [$ML^{-2}T^{-1}$]; and q_c is the fluid collected for sampling (replaced with DDW) per unit area, per unit time [LT^{-1}]. The concentration in the receptor compartment at any time, $c_b(t)$, can be expressed similarly as

$$c_b(t) = c_{b0} + \frac{1}{h_b} \int_0^t f_b(t)dt - \frac{q_c}{h_b} \int_0^t c_b(t)dt \tag{3}$$

where c_{b0} is the initial concentration in the receptor solution [ML^{-3}]; h_b is the height of the receptor (volume of receptor reservoir per unit area) [L]; $f_b(t)$ is the mass flux of contaminant into the receptor reservoir at any time t [$ML^{-2}T^{-1}$]; and q_c is the fluid collected for sampling (replaced with DDW) per unit area, per unit time [LT^{-1}].

The purpose of a laboratory diffusion test on a GCL (or any other liner material) is to obtain estimates of

Table 3. Summary of VOC GCL diffusion tests performed

Test no.	H_r (cm)	h_b (cm)	GCL thickness (mm)	e_B	n_t	Test duration (days)
NWNWT1	4.4	3.1	13.4	4.1	0.84	12
NWNWT2	3.5	3.5	13.8	4.6	0.86	12
NWNWT3	4.4	3.1	13.4	4.1	0.84	21
NWNWT4	3.5	3.5	13.8	4.6	0.86	21

diffusion and sorption parameters of the GCL. For GCL diffusion testing similar to that described herein, D_t , and often K_d , are unknown for the GCL, and hence the use of Equations 1, 2 and 3 requires values for D_t and K_d to produce the theoretical source and receptor reservoir curves using a contaminant transport program such as POLLUTE (Rowe and Booker 1999). Therefore an iterative technique is required to match theoretical source and receptor reservoir curves for a particular value of D_t and K_d for the GCL diffusion test with observed experimental data for a particular contaminant. The 'best fit' to the experimental data permits the selection of the D_t and K_d values for the GCL and contaminant being examined.

To fit theoretical POLLUTE curves to experimental data for a particular contaminant, a computer program, POLFIT[®] (Lake 2000), was developed. POLFIT has an algorithm that selects trial parameters (D_t , K_d), calls the contaminant transport program POLLUTE to calculate the theoretical source and receptor reservoir concentrations, and then compares these with the experimental source and receptor reservoir concentrations established in the tests. The square of the 'residual' or difference between the predicted POLLUTE concentration and experimental concentration is calculated at each point in time for which data are available. The sum of these squares (sum of squares error, SSE) is then calculated. This procedure is repeated for a range of D_t and K_d values until the sum of squares error (SSE) is a minimum for a particular D_t and K_d . These theoretical curves and hence D_t and K_d used to produce the curves are the best fit to the experimental data.

Best-fit curves obtained using POLFIT are given throughout this study. However, it should be noted that results obtained from a 'fit by eye' approach performed prior to the development of POLFIT provided very similar results, with diffusion coefficient obtained using the fit-by-eye approach varying by approximately $\pm 0.3 \times 10^{-10} \text{ m}^2/\text{s}$ (or less) from that obtained using POLFIT. It was also noted that variation between POLFIT and a fit-by-eye approach was random, indicating that there was very little bias in interpreting D_t and K_d 'by eye'.

3.3. GCL VOC diffusion test programme

GCLs used for diffusion testing were obtained from Naue Fasertechnik of Germany through the Bentofix distributor in Canada, Terrafix Geosynthetics of Toronto, Canada. The trade name of the GCL is a Bentofix NW GCL (described throughout the paper as NWNWT

(NonWoven carrier, NonWoven cover, Thermally treated needle-punched fibres) GCL), which consists of a nonwoven bottom geotextile (scrim reinforced with slit film woven polypropylene geotextile) and a nonwoven top polypropylene geotextile. The granular bentonite and the two geotextiles are attached by needle punching and thermal treatment of the punched fibres. A description of this GCL and the granular bentonite used in the NWNWT GCL was given by Lake and Rowe (2000b). The organic carbon content of the granular bentonite used in diffusion testing was 0.3%, as described in Lake (2000).

Two sets of GCL VOC diffusion tests were performed, each in duplicate, with the main difference between the two sets of tests being the dimensions of the glass cells used for each set of tests. Table 3 summarises the dimensions of the glass cells as well as the properties of the hydrated GCL used for testing. The duplicate tests were considered as one data set for modelling purposes to provide more experimental data to fit the theoretical data. As shown in Table 3, tests NWNWT1 and NWNWT3 are duplicates. Likewise, NWNWT2 and NWNWT4 are duplicates. When discussing the combined results of the duplicate tests, they are referred to as NWNWT1&3 and NWNWT2&4. The main difference between duplicate tests is that NWNWT1 and NWNWT2 were terminated after 12 days compared with 21 days for NWNWT3 and NWNWT4.

Geotextile sorption tests were also performed to independently investigate sorption of the contaminants to the geotextile component of the GCLs. Geotextile sorption tests consisted of the diffusion cells similar to that described above (see Figure 1) without the GCL. The diffusion cell was filled with a solution of contaminants similar in concentration to that used for diffusion testing, and monitored continuously with time.

4. DIFFUSION TEST RESULTS

4.1. General

Experimental diffusion test data for DCM, DCA, TCE, benzene and toluene are shown in Figures 2 to 6. An expanded scale of the y-axis is utilised for Figures 5 and 6. For convenience, data are presented as the median (symbol) and data range (vertical error bars) for each experimental point. The median value and data range were obtained from triplicate samples taken from the source and receptor each day. The curve fitting previously discussed in Section 3.2 was performed using individual data points and not the median and

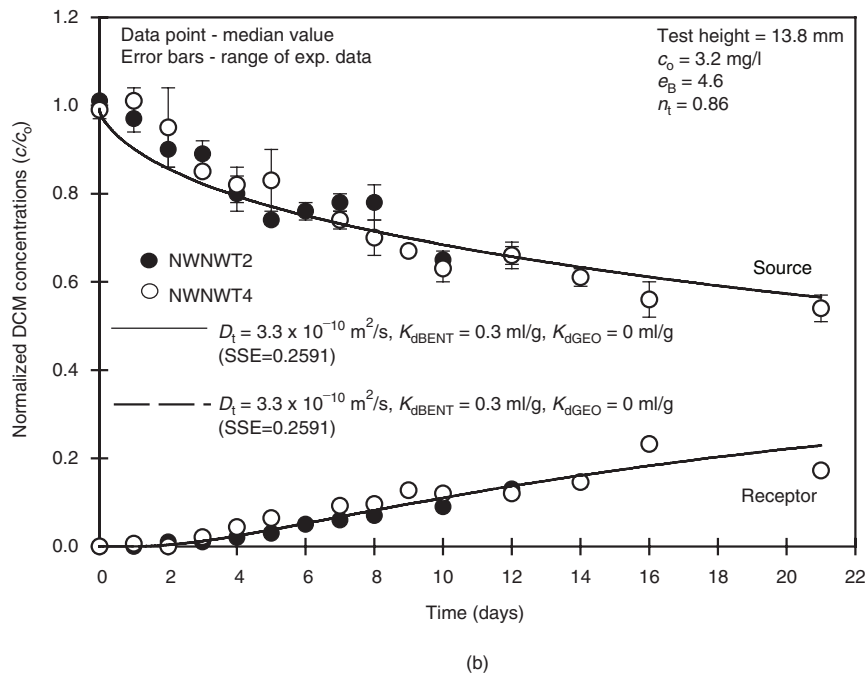
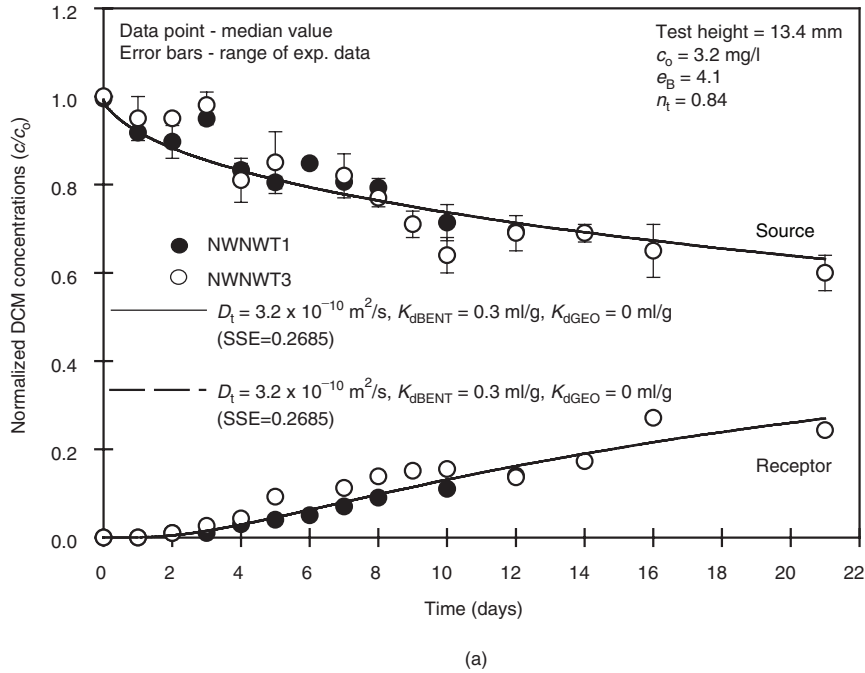
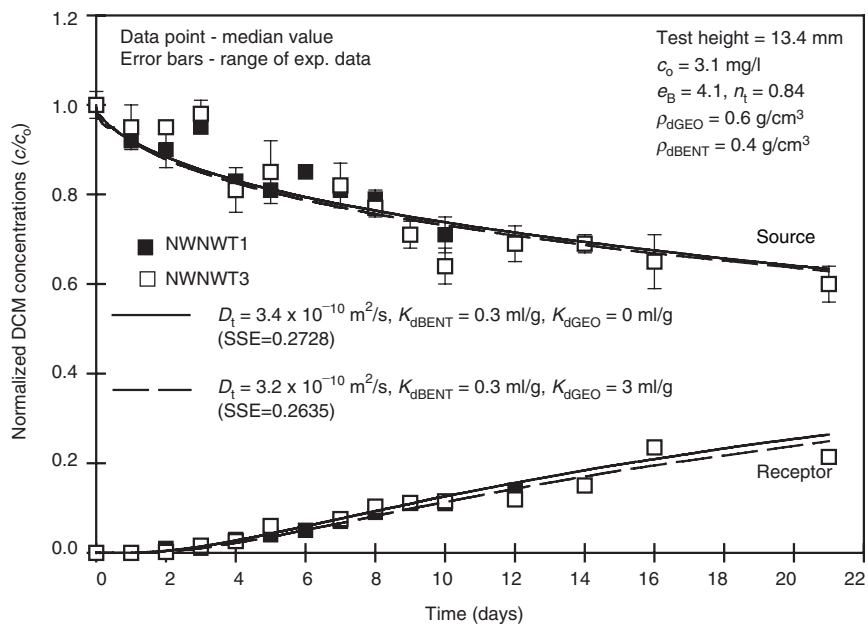


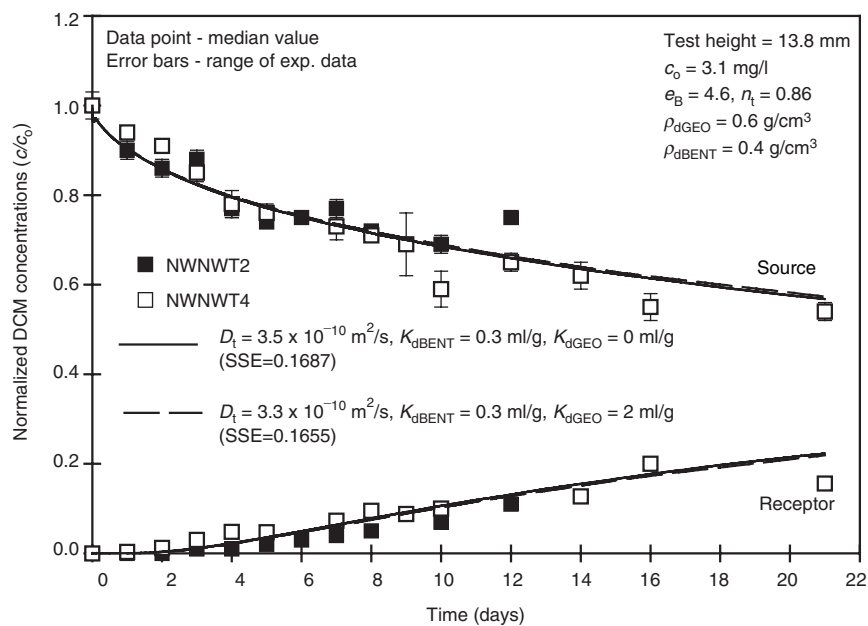
Figure 2. Theoretical POLLUTE curves (solid and dashed lines) fit to DCM experimental diffusion test data (solid and outlined circles). Solid lines represent optimum bentonite D_t (K_{dBENT} and K_{dGEO} constant), dashed lines represent optimum bentonite D_t and K_{dGEO} (K_{dBENT} constant): (a) NWNWT1&3 DCM diffusion; (b) NWNWT2&4 DCM diffusion

range values shown in Figures 2 to 6. For each plot, the solid symbols represent data for one of the duplicate tests (NWNWT1 or NWNWT2; test duration 12 days), and the open symbols represent data from the other (NWNWT3 or NWNWT4; test duration 21 days). The upper data points for each plot (starting at $c/c_0 = 1$) show the decrease in contaminant concentration with time in the source reservoir due to contaminant

migration into the GCL, and the lower data points (starting at $c/c_0 = 0$) represent the increase in contaminant concentration in the receptor reservoir with time. The solid and dashed lines on each plot represent the theoretical curves generated by POLLUTE modelling, as will be explained later. The final bulk GCL void ratio, e_B (as defined by Petrov and Rowe 1997), of each GCL tested is also shown in each figure.



(a)

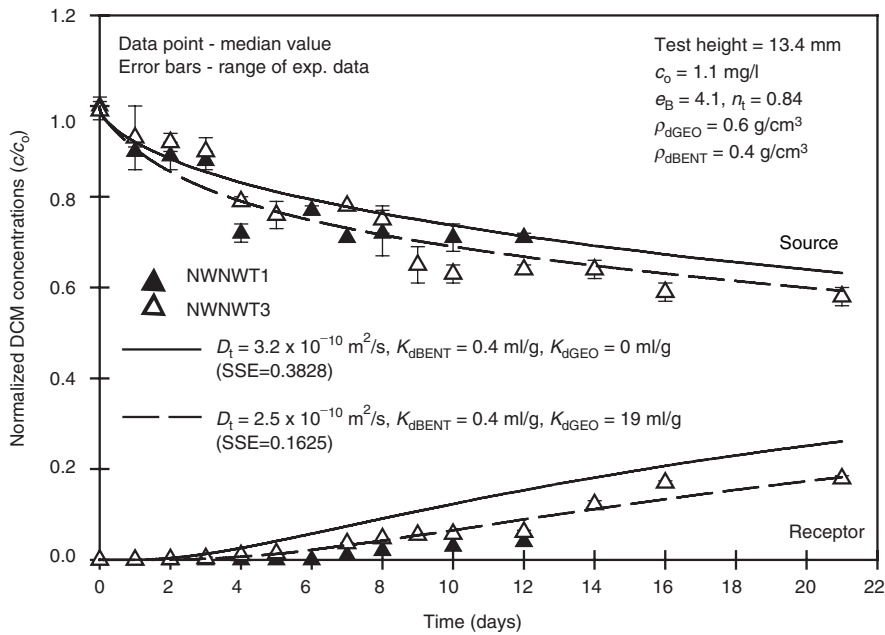


(b)

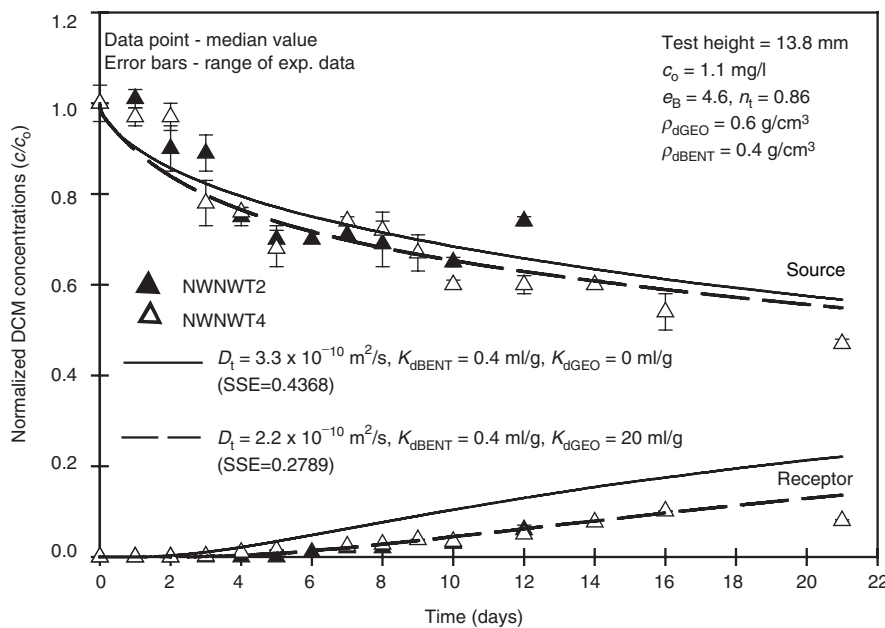
Figure 3. Theoretical POLLUTE curves (solid and dashed lines) fit to DCA experimental diffusion test data (solid and outlined squares). Solid lines represent optimum bentonite D_1 (K_{dBENT} and K_{dGEO} constant), dashed lines represent optimum bentonite D_1 and K_{dGEO} (K_{dBENT} constant): (a) NWNWT1&3 DCA diffusion; (b) NWNWT2&4 DCA diffusion

For the GCL samples and diffusion cell dimensions examined, all five contaminants exhibited a ‘break-through’ into the receptor reservoir at some point during the 21 days of testing; the actual time of breakthrough varied with each contaminant. When examining the experimental data for both sets of tests, DCM and DCA appear to be the contaminants with the quickest break-through times (2 to 3 days) relative to benzene (4 to 5 days), and TCE and toluene (5 to 6 days). The small

amount of attenuation of DCM and DCA results in concentration levels in the receptor reservoir of approximately 20% of the initial source concentration (i.e. $c/c_0 = 0.2$) after 21 days. Benzene exhibits final concentrations in the receptor reservoirs ranging from 0.1 to 0.2 of c_0 , and TCE and toluene final concentrations in the receptor reservoirs range from 0.04 to 0.08 of c_0 . Based on these initial observations, the level of retardation increases in the order DCM < DCA < benzene <



(a)



(b)

Figure 4. Theoretical POLLUTE curves (solid and dashed lines) fit to benzene experimental diffusion test data (solid and outlined triangles). Solid lines represent optimum bentonite D_t (K_{dBENT} and K_{dGEO} constant), dashed lines represent optimum bentonite D_t and K_{dGEO} (K_{dBENT} constant): (a) NWNWT1&3 benzene diffusion; (b) NWNWT2&4 benzene diffusion

toluene and TCE as the VOC contaminants migrate from the source reservoir, through the GCL and into the receptor reservoir because of the concentration gradient. As will be observed in subsequent sections, this difference in the level of retardation is the result of sorption to the geotextile.

The diffusion testing performed in this study lasted for 10 days or 21 days, depending on the test performed. As

described by Rowe *et al.* (1997a) this time frame is not long enough to initiate any significant biodegradation process of the VOCs in the test set-up. This is confirmed by the shapes of the source and receptor curves in Figures 2 to 6. If significant biodegradation resulted during testing, there would be a significant difference in concentration profiles in the source and receptor compartment, as demonstrated by Rowe *et al.* (1997a).

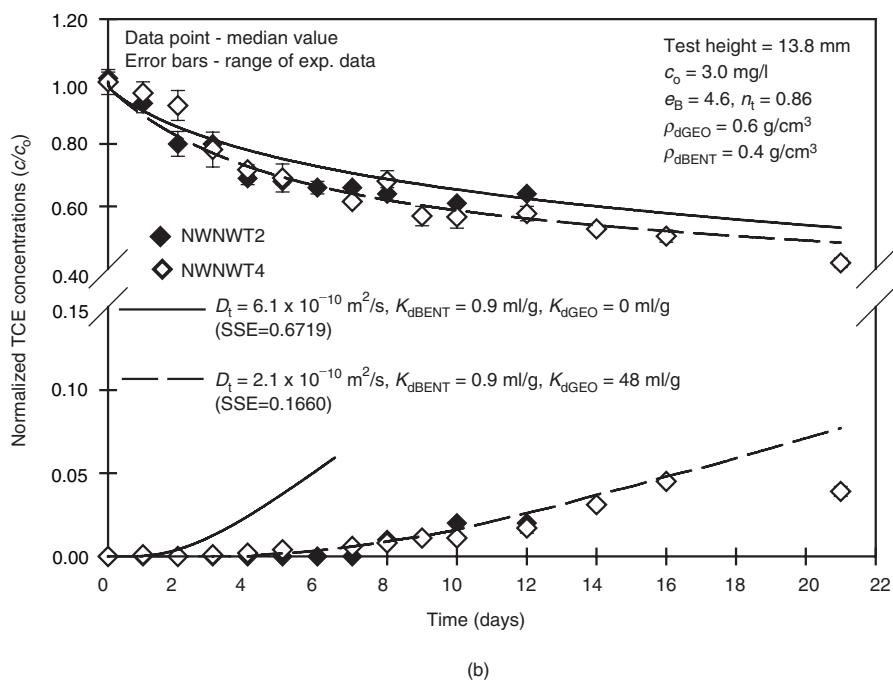
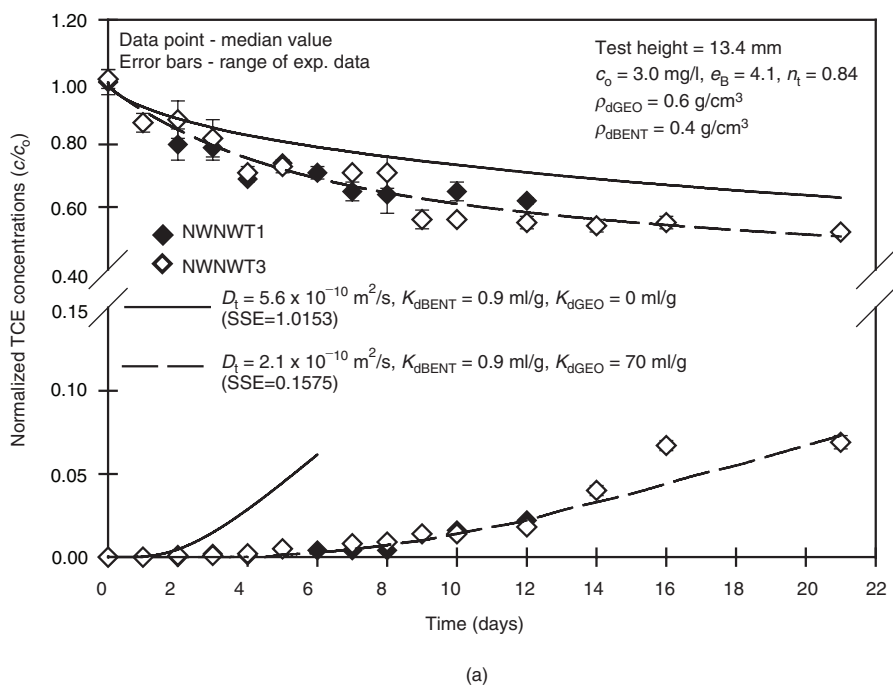


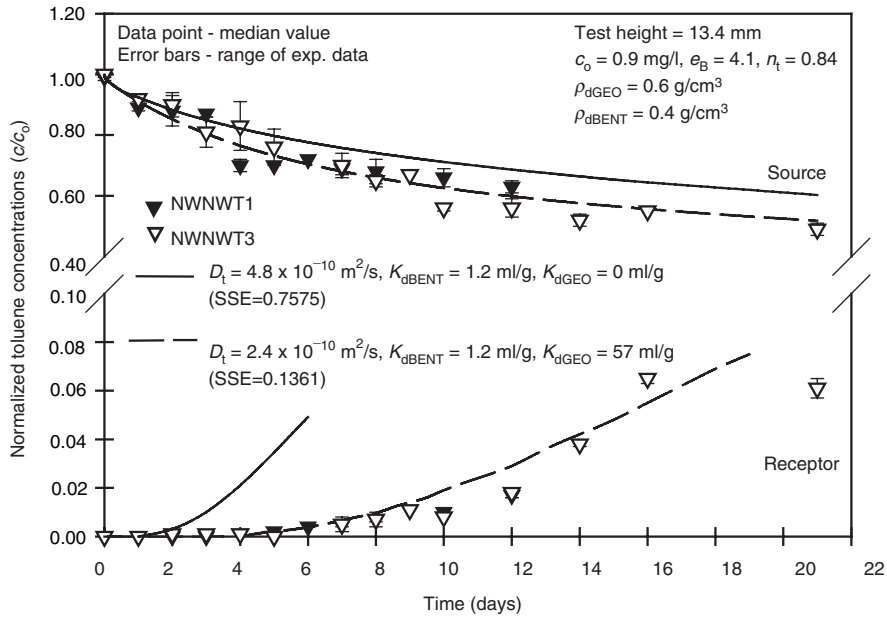
Figure 5. Theoretical POLLUTE curves (solid and dashed lines) fit to TCE experimental diffusion test data (solid and outlined diamonds). Solid lines represent optimum bentonite D_t (K_{dBENT} and K_{dGEO} constant), dashed lines represent optimum bentonite D_t and K_{dGEO} (K_{dBENT} constant): (a) NWNWT1&3 TCE diffusion; (b) NWNWT2&4 TCE diffusion

4.2. Diffusion coefficient (D_t) in the GCL neglecting sorption onto the geotextile

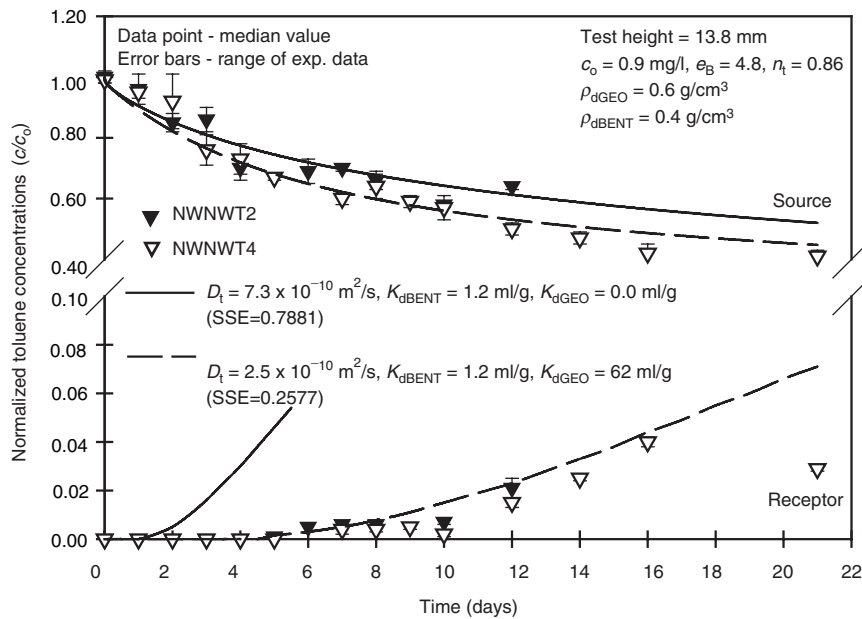
As an initial estimate of the sorption of the VOCs tested to the granular bentonite, batch tests were performed (see Lake 2000). Sorption of the VOCs to the bentonite was found to be relatively low, with K_d values of DCM 0.3 ml/g, DCA 0.3 ml/g, TCE 0.9 ml/g, benzene 0.4 ml/g, and toluene 1.2 ml/g. This low sorption of the non-polar

organic compounds tested in this study is due to the small percentage of organic carbon in the bentonite (0.3%). These low values are consistent with values obtained by Smith and Jaffe (1994) and Lo *et al.* (1997).

A first approach adopted for evaluating D_t and K_d for the NWNWT GCL was to assume that the sorption of the bentonite component of the GCL had a K_d value equal to that obtained from batch tests. These bentonite



(a)



(b)

Figure 6. Theoretical POLLUTE curves (solid and dashed lines) fit to toluene experimental diffusion test data (solid and outlined triangles). Solid lines represent optimum bentonite D_t (K_{dBENT} and K_{dGEO} constant), dashed lines represent optimum bentonite D_t and K_{dGEO} (K_{dBENT} constant): (a) NWNWT1&3 toluene diffusion; (b) NWNWT2&4 toluene diffusion

K_d values (hereafter referred to as K_{dBENT}) were taken as known, and hence were assumed to be constant throughout the iterative process in which D_t was optimised using the program POLFIT to find the best fit to the experimental data. It was also initially assumed that the geotextile component of the GCL had no sorptive capacity for the VOCs examined. As will be shown later, this is not the case for some of the VOCs examined in this study. However, it is of interest to

examine the theoretical fits to the data obtained assuming that the sorption corresponds only to the relatively low value for bentonite obtained from batch tests, and to examine the effect of not considering sorption of VOCs to the geotextile component of the GCL.

As shown in Figure 2, the theoretical fit to DCM experimental data (solid line) is quite good for the assumption of no sorption of DCM to the geotextiles.

Table 4. DCM, DCA, TCE, benzene, and toluene bentonite diffusion coefficients (D_t) obtained from experimental data assuming bentonite partitioning coefficient, K_d , of batch tests and no sorption of geotextiles. Sum of square error (SSE) values for curve fitting are in parentheses

Test no.	D_t (m ² /s)				
	DCM	DCA	TCE	Benzene	Toluene
NWNWT1&3	3.2×10^{-10} (0.2685)	3.4×10^{-10} (0.2728)	5.6×10^{-10} , NF (1.0153)	3.2×10^{-10} , PF (0.3828)	4.8×10^{-10} , NF (0.7575)
NWNWT2&4	3.3×10^{-10} (0.2591)	3.5×10^{-10} (0.1687)	6.1×10^{-10} , NF (0.6719)	3.3×10^{-10} , PF (0.4368)	7.3×10^{-10} , NF (0.7881)

NF, no fit; PF, poor fit.

The diffusion coefficient, D_t , obtained from both sets of diffusion data ranged from 3.2×10^{-10} m²/s to 3.3×10^{-10} m²/s as summarised in Table 4. DCA also provided a reasonable fit to the data, with D_t ranging from 3.4×10^{-10} m²/s to 3.5×10^{-10} m²/s. The sum of square error (SSE) for both DCM and DCA theoretical fits is also provided in parentheses in Table 4.

Benzene diffusion coefficients obtained from the fitting procedure ranged from 3.2×10^{-10} m²/s to 3.3×10^{-10} m²/s, although it can be seen from Figure 4 that these best-fit parameters tend to overestimate benzene source and receptor concentrations if sorption onto the geotextile component of the GCL is neglected. A comparison of the benzene SSE values with those for DCM and DCA also indicates a poor fit.

The theoretical curves for TCE and toluene, obtained using the batch test K_d value for the bentonite and no sorption onto the geotextile, did not fit the experimental data (Figures 5 and 6). This visually poor fit to the data is supported by the relatively large SSE values for TCE and toluene compared with DCM, DCA and benzene. This comparison shows that, when considering diffusion of benzene and toluene through a GCL, sorption on the geotextile component of the GCL must be considered, as discussed in the following section.

4.3. D_t in the GCL and K_d of geotextile (K_{dGEO})

In the previous section, D_t for the GCL was evaluated assuming that the only sorption for a particular contaminant was sorption to the bentonite component of the GCL (as obtained from batch testing). Although this approach produced reasonable theoretical fits to the experimental data for DCM and DCA, fits for the other contaminants examined were not good, suggesting an

additional mechanism of sorption of hydrocarbons to the polypropylene geotextile used in the manufacture of the NWNWT GCL. Polypropylene is a polymer formed from a series of repeating units or monomers chemically bonded together to form a high molecular weight compound (Cheremisinoff 1993). These relatively non-polar propylene units have the potential to provide a non-polar medium for the VOCs' sorption relative to the polar water in which they are dissolved. Thus it would not be unexpected that dissolved hydrocarbons would sorb onto the polypropylene geotextile, with the sorption being least for DCM, which has the lowest octanol-water partition coefficient (K_{ow}), and greatest for TCE and toluene, which are the most non-polar of the five VOCs examined and have the highest value of K_{ow} (Table 2).

To examine whether improved fits to the experimental data could be obtained by accounting for sorption of the VOCs to the geotextiles of the GCL, the iterative curve-fitting procedure described in the previous section was repeated. However, in addition to varying D_t , the K_d value of the geotextile (hereafter referred to as K_{dGEO}) was also varied. A 'dry density' of 600 kg/m³ was used for both geotextiles based on measurements of the mass per unit area of the geotextiles.

The dashed lines shown in Figures 2 to 6 indicate the best-fit theoretical curves obtained by varying D_t and K_{dGEO} , and a summary of results is given in Table 5. In the case of DCM (Figure 2), D_t ranging from 3.2×10^{-10} m²/s to 3.3×10^{-10} m²/s in combination with a K_{dGEO} value of zero provides the best fit to the data (the theoretical curve for K_{dGEO} plots on top of that produced for Section 4.2), indicating that sorption of DCM to the geotextiles is very low and is not influencing

Table 5. DCM, DCA, TCE, benzene and toluene bentonite diffusion coefficients and partitioning coefficients of geotextile (D_t and K_{dGEO}) obtained from experimental data assuming bentonite K_d of batch tests. Sum of square error (SSE) values for curve fitting are in parentheses

Test no.	D_t (m ² /s) and K_{dGEO} (ml/g)				
	DCM	DCA	TCE	Benzene	Toluene
NWNWT1&3	3.2×10^{-10} and 0 (0.2685)	3.2×10^{-10} and 3 (0.2635)	2.1×10^{-10} and 70 (0.1575)	2.5×10^{-10} and 19 (0.1625)	2.4×10^{-10} and 57 (0.1361)
NWNWT2&4	3.3×10^{-10} and 0 (0.2591)	3.3×10^{-10} and 0 (0.1655)	2.1×10^{-10} and 48 (0.1660)	2.2×10^{-10} and 20 (0.2789)	2.5×10^{-10} and 62 (0.2577)

Geotextile density of 600 kg/m³ (Lake and Rowe 2000b).

the value of the diffusion coefficient of DCM through the GCL.

The theoretical fit to the DCA experimental data (dashed line, Figure 3) obtained by considering sorption onto the geotextile (K_{dGEO}) is visually better than the case investigated in Section 4.2, which is confirmed by the lower value for the DCA SSE (compare values given in Table 5 with those in Table 4). However, the diffusion coefficient ($3.2 \times 10^{-10} \text{ m}^2/\text{s}$ and $3.3 \times 10^{-10} \text{ m}^2/\text{s}$) was only marginally affected relative to that obtained neglecting sorption ($3.4 \times 10^{-10} \text{ m}^2/\text{s}$ to $3.5 \times 10^{-10} \text{ m}^2/\text{s}$). The best-fit K_{dGEO} values varied from 2 ml/g to 3 ml/g. Thus the level of sorption was relatively low and had only a small effect of diffusion through the GCL.

For benzene, TCE and toluene good theoretical fits to the data were obtained when sorption onto the geotextile component of the GCL was considered (Figures 4, 5 and 6). This is also evident when comparing SSE for data fits without (Table 4) and with (Table 5) sorption onto the GCL. D_t found for benzene ranged from $2.2 \times 10^{-10} \text{ m}^2/\text{s}$ to $2.5 \times 10^{-10} \text{ m}^2/\text{s}$, with K_{dGEO} varying from 19 ml/g to 20 ml/g (Figure 4, dashed lines). TCE diffusion coefficients were $2.1 \times 10^{-10} \text{ m}^2/\text{s}$ with K_{dGEO} ranging from 48 ml/g to 70 ml/g (Figure 5, dashed lines). Toluene diffusion coefficients ranged from $2.4 \times 10^{-10} \text{ m}^2/\text{s}$ to $2.5 \times 10^{-10} \text{ m}^2/\text{s}$, with K_{dGEO} ranging from 57 ml/g to 62 ml/g (Figure 6, dashed lines). Overall, much better theoretical fits to the data were obtained by modelling GCL VOC diffusion tests considering sorption to the polypropylene geotextiles.

4.4. VOC sorption onto geotextile components of the GCL

To further investigate the influence of the polypropylene geotextile of the GCL on the sorption of the VOCs, the sorption tests described earlier were performed by placing the geotextiles from the GCL (bentonite removed) into an empty ('blank') cell. The results of the geotextile sorption tests are provided in Figure 7 for the chlorinated VOCs and in Figure 8 for the aromatic VOCs. Duplicate test results are shown in each figure as solid symbols (GEOBLK-1) and hollow symbols (GEOBLK-2). K_{dGEO} values were obtained as described in detail by Lake (2000). The deduced values of K_{dGEO} are summarised in Table 6.

Comparing the K_{dGEO} values given in Tables 5 and 6, it can be seen that the values for TCE, benzene and toluene are reasonably consistent compared with K_{dGEO} obtained from diffusion testing. For DCM and DCA the geotextile blank test gave higher values than obtained for the diffusion tests. The geotextile sorption tests are considered to provide an upper bound for sorption of VOCs to the geotextile during diffusion testing. The presence of bentonite in the GCL during diffusion testing also acts as a sorbent for the VOCs, and therefore the competitive sorption to the geotextile during diffusion testing appears to be slightly less than that obtained from blank tests, especially for DCM and DCA.

5. DISCUSSION OF GCL VOC DIFFUSION TESTS

Reasonable fits to the experimental diffusion test data were achieved for DCM and DCA by assuming no sorption of the VOCs to the geotextile component of the GCL, whereas poor fits were found for benzene, TCE and toluene. By optimising D_t and K_{dGEO} simultaneously, good fits to the experimental data were achieved for all VOCs examined. An independent examination of K_{dGEO} with geotextile sorption tests confirmed parameters obtained from diffusion tests.

The diffusion coefficients deduced from diffusion tests are similar for the contaminants examined (ranging from approximately $2 \times 10^{-10} \text{ m}^2/\text{s}$ to $3 \times 10^{-10} \text{ m}^2/\text{s}$). The variation in the D_t values obtained in duplicate tests for each of the VOC contaminants examined was small ($< 0.3 \times 10^{-10} \text{ m}^2/\text{s}$), and is probably due to small sampling errors and analytical reproducibility during the testing process. The factor that most influences the deduced diffusion coefficient is the choice of sorption parameters for the geotextile, K_{dGEO} . For DCM and DCA the effect of sorption is small, and the results were relatively insensitive to whether or not sorption was considered (Tables 4 and 5). For benzene, toluene and TCE, the consideration of sorption onto the geotextile (i.e. choice of K_{dGEO}) did have a significant effect on the deduced diffusion coefficient. The values given in Table 5 represent the combination of diffusion and sorption parameters that provides the best fit to the experimental data. In the case of toluene these best-fit values of K_{dGEO} are very close to the average value deduced from batch tests (Table 6). For benzene the best-fit values of K_{dGEO} (19–20 ml/g) are about 20% below the values deduced from the batch test (24 ml/g). Had the higher batch test values been taken as a given, a higher diffusion coefficient would have been required to match the concentrations in the receptor. For TCE the best-fit values of K_{dGEO} (48 and 70 ml/g) bracket the batch test value (62 ml/g). It can be concluded that these test results provide good values for DCM, DCA and toluene and reasonable values for benzene and TCE. It was assumed herein that sorption onto the carrier and cover geotextile were the same. This assumption warrants further investigation.

For field conditions, a manufacturer's installation specifications usually advise against allowing the GCL to hydrate under free swell conditions. Hence, for a landfill liner application, a cover soil would be immediately applied to the GCL, which would assist in attaining lower bulk GCL void ratios after hydration. Thus the results presented herein, which correspond to swell pressures under 10 kPa, represent conditions where the GCL hydrates under only 0.5 m of cover material. If the diffusion coefficient of the VOCs decreases with lower GCL bulk void ratios, as was shown for chloride and sodium in Lake and Rowe (2000a), then it would be reasonable to expect that the diffusion coefficients obtained in this study would provide an upper bound for GCL VOC diffusion coefficients for a range of stress

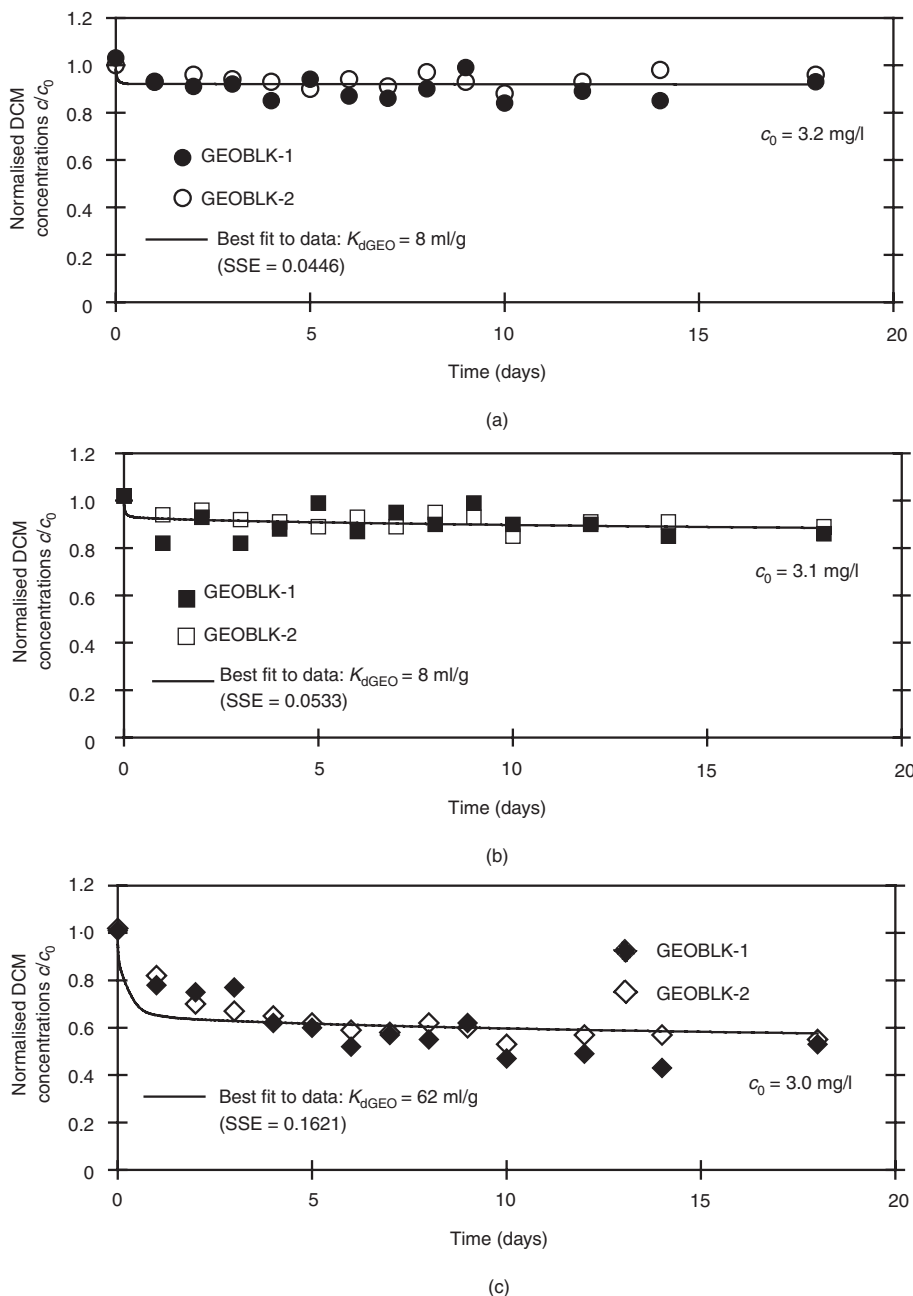


Figure 7. Geotextile batch test results and best theoretical fits to data for: (a) DCM; (b) DCA; (c) TCE

conditions expected in the field (for the five VOCs examined).

Diffusion test modelling as well as geotextile sorption test results suggest that there are varying amounts of VOC sorption to the polypropylene geotextiles present in the NWNWT GCL. This is not unexpected, owing to the similarities in chemical structure between the VOCs and polypropylene. It is apparent from the sorption tests that the polypropylene geotextile is responsible for sorbing the VOCs to varying degrees, and this should be considered both in the interpretation of diffusion test results and in the application of the results. Also, as the mass of geotextile available for sorption varies for different GCL products, the amount of sorption is also likely to vary from one product to another.

The values of diffusion coefficients reported in this paper are generally similar to or lower than those obtained by Rowe and Barone (1991), Myrand *et al.* (1992) and Kim *et al.* (2001) for VOC diffusion through CCLs. This is consistent with the results of Lake and Rowe (2000b) for inorganic contaminants. It is also consistent with lower hydraulic conductivity results of GCLs (Petrov and Rowe 1998) relative to CCLs (Quigley *et al.* 1987). Provided GCLs are hydrated with water, and subjected to confining stresses prior to exposure to MSWL, it has been shown that they can provide just as good, if not better hydraulic retention characteristics to CCLs. The same beneficial properties with respect to hydraulic conductivity also affect diffusive migration, namely the small surface area of

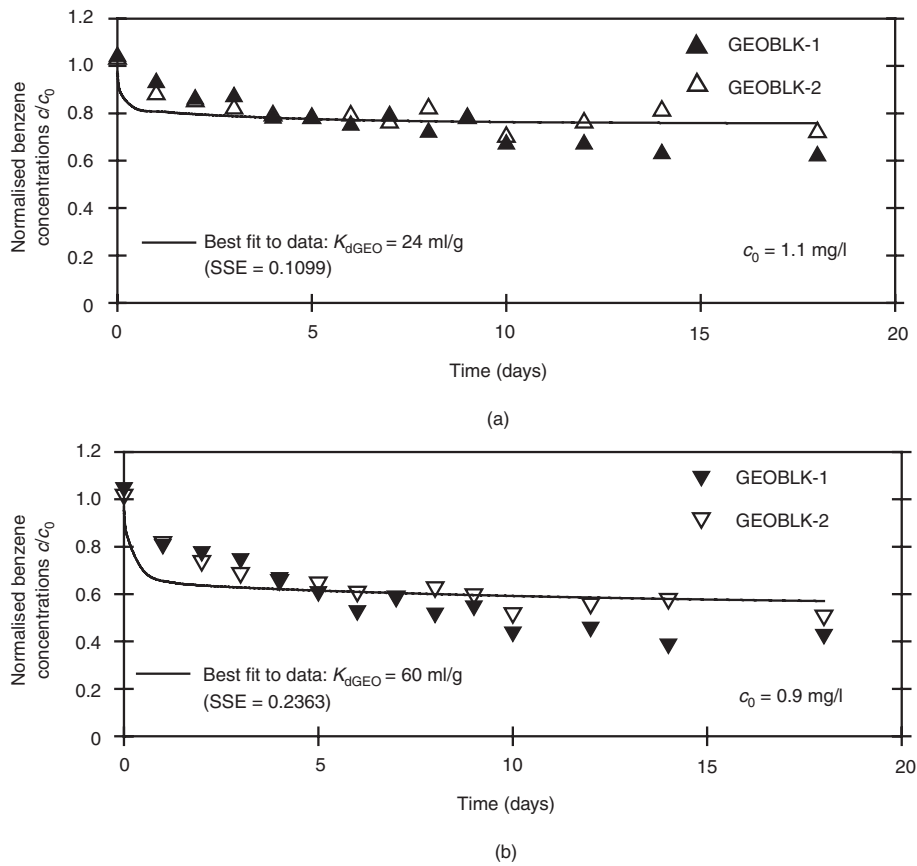


Figure 8. Geotextile batch test results and best theoretical fits to data for: (a) benzene; (b) toluene

Table 6. Geotextile sorption parameters obtained from geotextile sorption test

	DCM	DCA	TCE	Benzene	Toluene
K_{dGEO} (ml/g)	10	8	62	24	60
SSE	0.0446	0.0533	0.1621	0.1099	0.2363

the montmorillonite particles in the bentonite providing a tortuous path for contaminants to travel through. In addition, the influence of the large double layer in the hydrated bentonite reduces the effective pore space for contaminant migration.

The results in this paper provide the first information as to the diffusion of five common VOC contaminants through GCLs based on laboratory diffusion tests. However, the GCL represents only a portion of the landfill barrier liner system, and is often used in combination with a geomembrane (GM) to form an engineered composite liner system for a landfill that may overly native soil on the landfill construction site (attenuation/foundation soil). It is the engineered liner system (GM/GCL composite liner or GM/compacted clay composite liner) as well as this foundation soil that separates contaminants in the landfill from any underlying groundwater near the site. The mass of VOC contaminant actually sorbed by the GCL (per unit mass of GCL) will probably be much less than the thicker attenuation layer, or even a compacted clay liner. This is because, although the attenuation layer may have less

sorptive ability (per unit mass of attenuation layer), there is potentially 100 to 300 times the mass available for sorption (0.01 m thick GCL used over 1–3 m thick attenuation layer). A proper assessment of the feasibility of a landfill liner design requires that all these components be considered in contaminant transport modelling of the landfill liner (Rowe *et al.* 1997; Rowe 1998). The VOC results obtained in this study allow this process to be completed for a system involving advective-diffusive transport through a GCL.

6. SUMMARY AND CONCLUSIONS

This paper has provided details of the development of a glass diffusion test apparatus that can be used for obtaining GCL diffusion coefficients for VOCs. Methods of monitoring and interpreting test results were also described.

Test results showed that the rate of contaminant migration proceeded through the hydrated GCL in the decreasing order of DCM and DCA > benzene > TCE and toluene. This was attributed to varying degrees of

sorption of DCA, benzene, TCE and toluene to the geotextile component of the GCL as well as to the bentonite present in the GCL. Sorption tests performed with only the geotextiles assisted in examining the sorption of VOCs during diffusion testing. Diffusion coefficients deduced from VOC diffusion testing ranged from approximately $2 \times 10^{-10} \text{ m}^2/\text{s}$ to $3 \times 10^{-10} \text{ m}^2/\text{s}$. It was noted that VOC diffusion coefficients were obtained at relatively high bulk void ratios compared with what would be encountered if the GCL were placed as part of a base liner system in a landfill. For this reason, the diffusion coefficients reported in this study most likely fall near the upper range of VOC diffusion coefficients expected for the case of a GCL in a base liner situation.

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NOTATIONS

Basic SI units are given in parentheses.

D_t	diffusion coefficient deduced using the total porosity (m^2/s)
e_B	final bulk GCL void ratio (dimensionless)
$f_i(t)$	mass flux of contaminant into the soil at any time t ($\text{kg}/(\text{m}^2/\text{s})$)
h_b	volume of receptor reservoir per unit area (m)
H_r	reference height of leachate (volume of source fluid per unit area) (m)
K_d	linear distribution coefficient (m^3/kg)
$K_{d\text{GEO}}$	linear distribution coefficient of geotextiles in GCL (m^3/kg)
$K_{d\text{BENT}}$	linear distribution coefficient of bentonite in GCL (m^3/kg)
n_t	total porosity (dimensionless)
q_c	fluid collected for sampling per unit area, per unit time (m/s)
\bar{v}	average linearised groundwater velocity (m/s)
v_a	Darcy velocity (m/s)
ρ	dry density (kg/m^3)

ABBREVIATIONS

CCL	compacted clay liner
DCM	Dichloromethane
DCA	1,2 Dichloroethane
GCL	geosynthetic clay liner
GM	geomembrane

MSW	municipal solid waste
NWNWT	nonwoven, nonwoven, thermally treated GCL
TCE	Trichloroethene

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