



An application of permeable reactive barrier technology to petroleum hydrocarbon contaminated groundwater

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Abstract

A funnel and gate permeable reactive barrier was designed and built to treat groundwater contaminated with dissolved phase toluene, ethyl benzene, and xylene and *n*-alkanes in the C₆–C₃₆ fraction range. Removal efficiencies for the funnel and gate system varied from 63% to 96% for the monocyclic aromatic hydrocarbons. Average removal efficiencies for C₆–C₉, C₁₀–C₁₄, and C₁₅–C₂₈ fraction ranges were 69.2%, 77.6% and 79.5%, respectively. The lowest average removal efficiencies were 54% for the C₂₉–C₃₆ *n*-alkane fraction. The overall average removal efficiency for the funnel and gate system towards petroleum hydrocarbons present in the groundwater was 72% during the 10 month period over which the data were collected, and has allowed relevant water quality objectives to be met. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Treatment walls, or permeable reactive barriers, first reported by Mc Murthy and Elton [1], involve construction of permanent, semi-permanent, or replaceable units across the flow path of a dissolved phase contaminant plume. As the contaminated groundwater moves passively through the treatment wall, contaminants are removed by physical, chemical and/or biological processes, including precipitation, sorption, oxidation/reduction, fixation, or degradation. These barriers may contain agents that are placed either in the path of contaminant plumes to prevent further migration or immediately downgradient of the contaminant source to prevent plume formation. Several methods have been developed for the installation of permeable treatment

walls [2,3]. The funnel and gate system is one application of a permeable reactive barrier for in situ treatment of dissolved phase contamination [4,5]. Such systems consist of low hydraulic conductivity cut-off walls (e.g., 1×10^{-6} cm/s) with one or more gaps that contain permeable reaction zones. Cut-off walls (the funnel) modify flow patterns so that groundwater primarily flows through high conductivity gaps (the gates). The type of cut-off walls commonly used are slurry walls, sheet piles, or soil admixtures applied by soil mixing or jet grouting.

A white spirit petroleum hydrocarbon spill occurred at a factory facility in South Eastern Australia in December, 1997. A leaking underground storage tank (UST) caused white spirit (3000 L) to leak through the scoria fill material in which the UST was located, into the soil of the upper embankment at the rear of the facility. A quantity of the white spirit emerged at the base of the slope, discharging into a spoon drain, and then via a culvert, flowed into the nearby river. A proportion of the leaked white spirit was absorbed by

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the soil in the embankment and a quantity also entered the groundwater underlying the site. The aim of the remedial works conducted and reported in this study was to intercept the dissolved phase plume of petroleum hydrocarbons present in the shallow aquifer moving toward the river, using a funnel and gate permeable barrier, and to reduce these petroleum hydrocarbon concentrations to meet the relevant water quality objectives for the site.

2. Site description and methodology

2.1. Site layout and geology

The area between the rear of the facility and the nearby river consists of a relatively steep ($\sim 45^\circ$) embankment, which is bisected by a cycle path (2 m wide). A spoon drain runs parallel to the western side of the access track, terminating 20 m north of the northern boundary of the site in a culvert, which runs under the cycle path and discharges directly to the nearby river. The facility occupies relatively level ground, exhibiting a lesser incline towards the river. A tank farm is located in the northeastern corner of the premises, consisting of 4 USTs, used for white spirit storage. The geology of the site consists of basement siltstones and sandstones of the upper Silurian Dargile Formation, the upper profile of which has weathered to a tan-orange mottled sandy clay.

2.2. Site hydrogeology

A highly localised, shallow perched water table is present in the fill material which comprises the embankment between the factory and the nearby river. The regional water table is expected to be located at approximately the same depth as the level of the river. Residual clays from the weathered Silurian siltstones tend to be relatively impermeable, and as such perched water tends to be located at the fill–natural sandy clay interface.

2.3. Aquifer testing

Aquifer recovery tests were carried out on monitoring wells MW1, MW2, MW3, MW4, MW5, MW6, MW7, and MW8 (Fig. 1) on 20 November 1998. The procedure involved the removal of a known volume of groundwater from the well and monitoring the subsequent recovery with time. The hydraulic properties of the fill material were investigated (by slug test) to determine the hydraulic conductivity of this material. The Hvorslev method [6] was used to calculate the hydraulic conductivity in the immediate proximity to each well tested.

2.4. Development of the groundwater contamination model

Field scale implementation of funnel and gate systems, as with all treatment wall systems, requires careful design based on site specific hydrology and contaminant plume characteristics. In the current study, the modular finite difference groundwater flow model (MODFLOW) developed by the United States Geological Survey (USGS) was utilised. The model simulates three-dimensional flow of constant density groundwater through porous earth materials using the finite difference method, which provides an approximate solution to the partial-differential equation that describes the three-dimensional flow of groundwater. The finite difference method requires the modelled area to be divided into a grid of rectangular cells defined by a number of columns and rows. A model comprising 2835 active cells defined by 70 columns and 59 rows with equal cell dimensions of $0.5 \times 0.5 \text{ m}^2$ has been developed (Fig. 1).

Steady state calibration simulations of the flow model were conducted using the measured piezometric surface for eight monitoring bores on 7 October 1998. In addition, particle tracking and preliminary Modular Three-dimensional Transport Model (MT3D), simulations were used to provide a check on the flow calibration. MT3D is a computer model which simulates advection, dispersion, diffusion and some basic chemical reactions of contaminants such as sorption and first order decay [7]. MT3D was used for mass transport simulations of the contaminant plume. The model is designed for use in conjunction with a block-centred finite-difference flow model such as MODFLOW, and is based on the assumption that the changes in the concentration field will not measurably affect the flow field. For the particle tracking simulation, ten particles were placed along the western boundary of the model adjacent to the former underground tank farm.

The modular three-dimensional transport model, MT3D, was used for mass transport simulations of the contaminant plume. The linear isotherm, adopted in this model, uses a single distribution coefficient, K_d (L/kg or m^3/kg), to define the relation between the concentration in the dissolved phase and the concentration of sorbed material in the porous matrix. The distribution coefficient (K_d) approach has commonly been used to describe the sorption of hydrophobic organic compounds, or those compounds that are readily soluble in non-polar solvents, but are only sparingly soluble in water. Contaminant transport simulations incorporating physical transport processes and chemical adsorption reactions were conducted for ethylbenzene and total xylenes. Simulations utilised the measured and interpolated concentrations of these two constituents from 9 September 1998 as the initial starting condition for the model run. The calibrated steady state model was

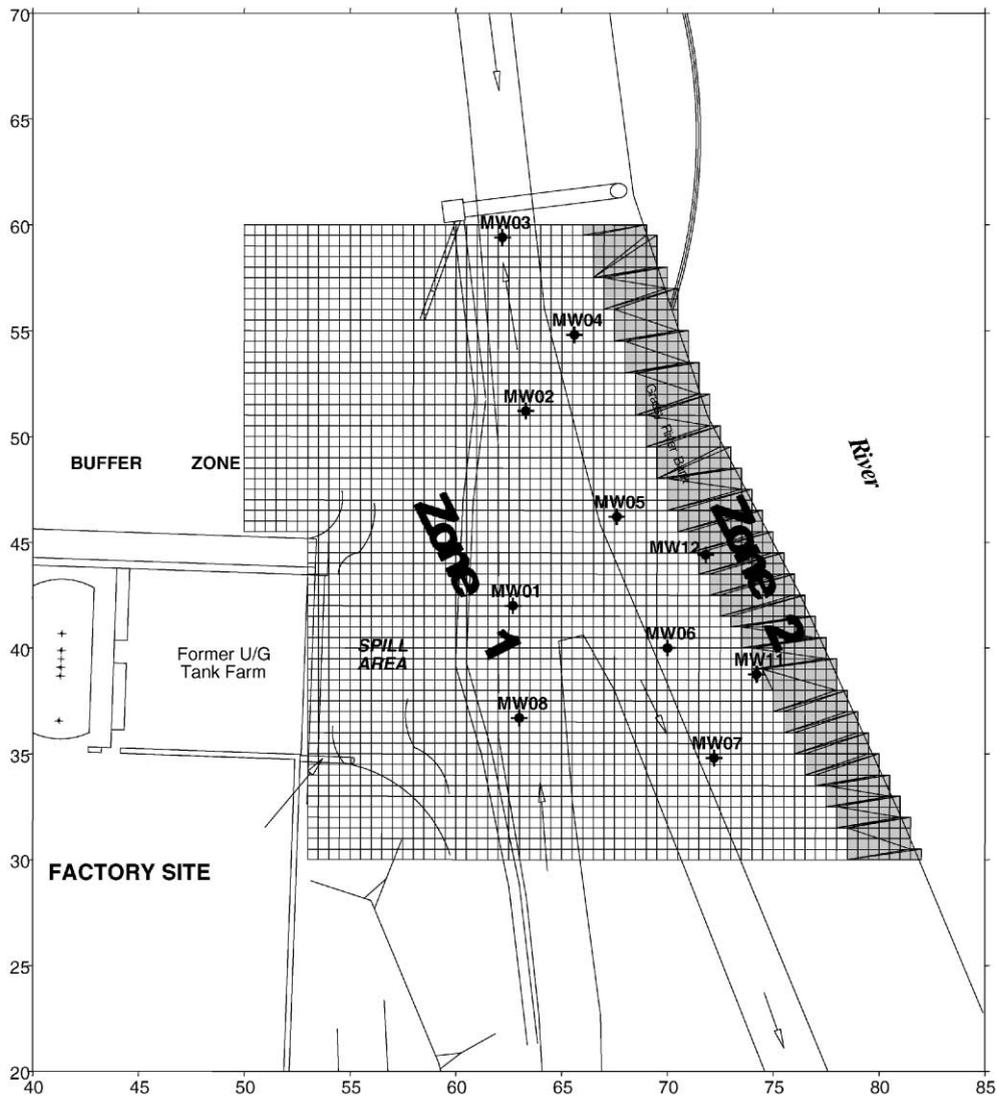


Fig. 1. The finite difference model grid and aquifer hydraulic property distribution (units in m).

modified to include the fabricated cut-off wall that was installed at the base of fill on 16 September 1998. In addition, drain cells were assigned to the upgradient side of the wall to simulate the cut-off trench by maintaining water levels at 1.2 m below ground level or approximately 2.6 m AHD. This facilitates removal of contaminants on the upgradient side of the wall.

The following parameters were used and assumptions were made in the modelling. The measured bulk density of the aquifer material was 1860 kg/m^3 . The average effective porosity was measured at 45%. The distribution coefficient (K_d) for linear adsorption reactions was taken as 1.76×10^{-3} and $3.74 \times 10^{-3} \text{ m}^3/\text{kg}$ for ethylbenzene and total xylenes, respectively. The model-

ling assumed no future spills. The volume of contaminated soil below the bicycle path, i.e. remaining in the soil (unexcavated), was calculated at 450 m^3 , with an average *n*-alkane petroleum hydrocarbon concentration of 2500 mg/kg and an average concentration of total xylenes at 18 mg/kg . A conservative estimate for the maximum amount of petroleum hydrocarbon entering the gate (desorbed from the contaminated soil remaining), was calculated to be 100 L (<4% of the original spill volume). Visual inspection of the contaminated water bailed out of all the wells showed no evidence of free phase NAPL. Approximately 100–200 L of petroleum hydrocarbon were recovered from the river and along the drain using booms at the time of the spill.

2.5. Construction of funnel and gate system

The funnel and gate system was comprised of an impervious barrier membrane (i.e. the funnel), directing groundwater into the treatment area (i.e. the gate). The gate consisted of a sparging unit upgradient of a blend of peat materials. The funnel component consisted of a 0.75 mm thick high-density polyethylene (HDPE) impervious barrier membrane positioned vertically in the cut-off trench to capture and redirect incident groundwater over the length of the spill area, parallel to the nearby river. The gate was composed of sequential treatment systems comprising a sparging unit emplaced in basaltic scoria, followed by blended peat materials. The funnel was designed to intercept groundwater flow from areas directly downgradient of the spill site, as well as adjacent areas in which lateral migration of the plume may be occurring. The dimensions of the funnel trench are 27 m long \times 5 m deep \times 0.6 m wide, excavated to a level of 0.5 m below the fill–natural siltstone interface.

2.5.1. Gate construction

Chain mesh fencing ($2 \times 15 \text{ m}^2$ long section) was laid flat along the river–embankment interface and fastened to the ground using 1.5 m long steel star pickets to aid long term stability. Sheet piling was inserted into the embankment parallel to the river bank at a distance of ~ 4.0 m from the river, dividing the areas to be excavated for both components of the gate system. The area designated for the peat (between the river bank and the sheet piling) was excavated to 0.5 m below the siltstone–fill interface, and infilled with peat to a depth of 0.5 m below surface. The remaining volume was filled with natural soil overlying a layer of A15 Type Bidim Geofabric to prevent fine soil particles mixing with and clogging the peat and scoria filtration media. Subsequent to the emplacement of the peat, soil adjacent to the opposite side of the sheet piling was excavated to a depth of 0.5 m below the fill–natural siltstone strata interface, and a horizontal sparging tube and scoria were emplaced. Thirty cubic metres of peat (on a wet volume basis of estimated 40% moisture) was used in the gate. The section of the gate that initially encounters influent groundwater consists of a volume ($\sim 5 \text{ m}^3$) of porous basaltic scoria. The scoria was emplaced over a submerged, perforated air sparging pipe, creating fine air bubbles, which permeate the water column within the gate, delivering oxygen to enhance biodegradation. Air was supplied to the system by a 12-cfm compressor, supplying air at $0.9 \text{ m}^3/\text{h}$. A transect diagram through the gate is given in Fig. 2.

2.5.2. Peat composition

After the groundwater was sparged (as it passed through the scoria as described above), the groundwater then passed through a peat mixture, immediately

downgradient of the sparging system. Previous studies indicate the high sorptive capacities of humic materials (such as peat) for petroleum hydrocarbons. Peats with low fibre contents and high lignin pyrolysis material and ash content have been shown to be the most effective peats to remove free and dissolved phase petroleum hydrocarbons from groundwater with removal efficiencies of 63–97% [8]. Using this information and conducting hydraulic conductivity tests, a locally available peat, referred to as “Biogreen Humic Reed Sedge”, with low fibre and high humic content, was selected for use. Hydraulic conductivity tests were performed on a variety of peats/sphagnum peat/cocoa fibre blends, in order that an optimal mixture, with respect to sorptive capacity, air circulation and hydraulic conductivity, is achieved. Tests were conducted using the “falling head test” method [9]. Equipment consisted of a column of packed, saturated peat. A blend of 70% biogreen humic reed sedge, 20% sphagnum peat and 10% cocoa fibre was selected for use based on the tests conducted in this study. Tests showed (data not shown) that this mix contained the maximum percentage of peat with the highest sorptive capacity (biogreen humic reed sedge), without reducing the hydraulic conductivity to less than that encountered in the surrounding fill materials. The hydraulic conductivity of the surrounding fill was 10^2 – 10^3 lower than the emplaced peat mixture.

2.6. Monitoring well installation, sampling and analysis

Testing of the funnel and gate system was primarily achieved via two groundwater monitoring wells, one emplaced immediately upgradient of the gate, and another downgradient of the gate area infilled with peat. A total of 14 monitoring wells were installed at the site. Monitoring wells MW 1 and MW 2 were installed in November 1997. Monitoring wells MW 3–MW 10 were installed in May 1998. Monitoring wells MW 11 and MW 12 were installed in December 1998. MW 13 and MW 14 were installed during March 1999. Bore wells were screened 0.5 m above the water table. All groundwater samples were analysed for *n*-alkanes (by GC-FID method, US EPA 8021B), and benzene, toluene, ethyl benzene and xylene (BTEX) using the purge and trap method (US EPA Method 5030) [10].

3. Results and discussion

3.1. Water table level and hydraulic conductivity tests

The water table configuration remained relatively constant throughout the monitoring period. The groundwater flow lines were observed to flow toward the east, approximately perpendicular to the bank of the river. Variations in the water table level were observed at

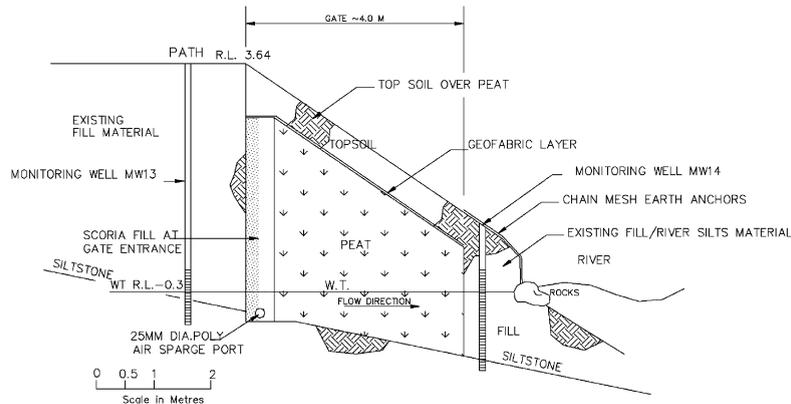


Fig. 2. Transect through the funnel and gate system.

Table 1
Measured hydraulic conductivity of subsurface

Monitoring well	Values (m/day)
MW01	0.18
MW02	1.63×10^{-8}
MW03	1.26×10^{-5}
MW04	0.0001
MW05	0.0046
MW06	0.0001
MW07	0.025
MW08	1.68

monitoring wells MW1, MW2 and MW3, illustrating similar temporal changes. These wells are furthest from the nearby river, located at the base of the slope. Closer to the river the water table variation is significantly less, indicating that the water table close to the river is being controlled by the water level in the river itself. Those wells located furthest from the river were expected to reflect rainfall (recharge) water table response relationships. As expected, the wells nearest the slope fluctuate the most in response to rainfall events, suggesting that the recharge is occurring from the slope to the west, most likely as interface drainage.

The results of the subsurface conductivity tests are presented in Table 1. As expected the material was found to have extremely variable hydraulic properties. Measurement of the conductivity of the fill material was in some instances erroneous as the well screens extended through two lithologies, the clay overlying the siltstone and the fill material itself. The large variability of the fill material suggests that the majority of groundwater and contaminant migration through this material will be occurring along preferential flow paths. If no treatment wall was installed, conditions were such that they were likely to result in faster migration times and a larger spread of the plume.

3.2. Modelling outcomes

The contamination distributions for ethylbenzene and total xylenes respectively, as measured on 9 September 1998, and with simulated transport and retardation at 6 months, 1, 2, 3 and 5 years following this date, indicated a general decline in concentration of groundwater contaminants and lateral movement to the river, as expected. Fig. 3 shows the predicted behaviour of ethyl benzene over these time periods. Given the initial concentrations, total xylenes appear to reside within the system for longer than benzene, which is consistent with the calculated retardation values. Based on the modelling results the concentrations of ethyl benzene and xylene in the groundwater being discharged to the river (ignoring mixing zones) will exceed the relevant criteria, $250 \mu\text{g/L}$, within ~ 6 months and will continue to exceed these criteria for a period in excess of 6 years.

The width of the discharge face (capture zone) was estimated at ~ 20 m, and this was effectively intercepted by the funnel.

The particle tracking simulations revealed that the longest travel time to the river estimated by the model is approximately 1000 days at the southern boundary, in comparison with travel times of approximately 475 days for the particle originating at the northern corner of the former UST farm (Fig. 4).

Fig. 5 illustrates the configuration of the water table on 7 October 1998.

3.3. Estimates of groundwater volumes and petroleum hydrocarbon mass entering funnel and gate system

The maximum concentrations of *n*-alkanes and the total of benzene, toluene, ethyl benzene and xylenes (BTEX) present in groundwater from bore wells across the site were 1000 and 25 mg/L , respectively. These values were reported between December 1997 and 1998.

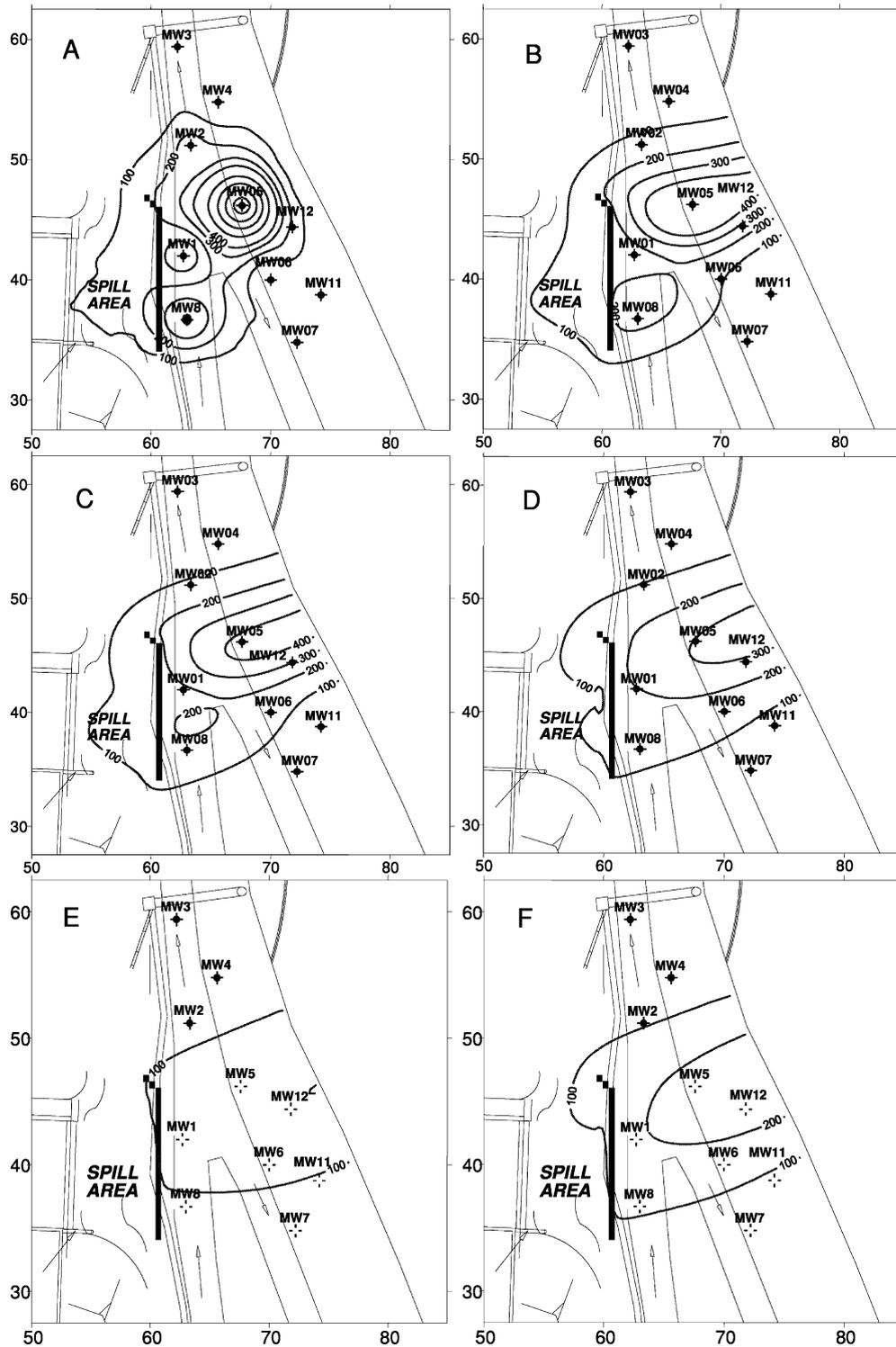


Fig. 3. Simulations of ethyl benzene distribution (μL) in groundwater: (A) 0 days, (B) 6 months, (C) 1 year, (D) 2 years, (E) 3 years, and (F) 5 years from 9 September 1998.

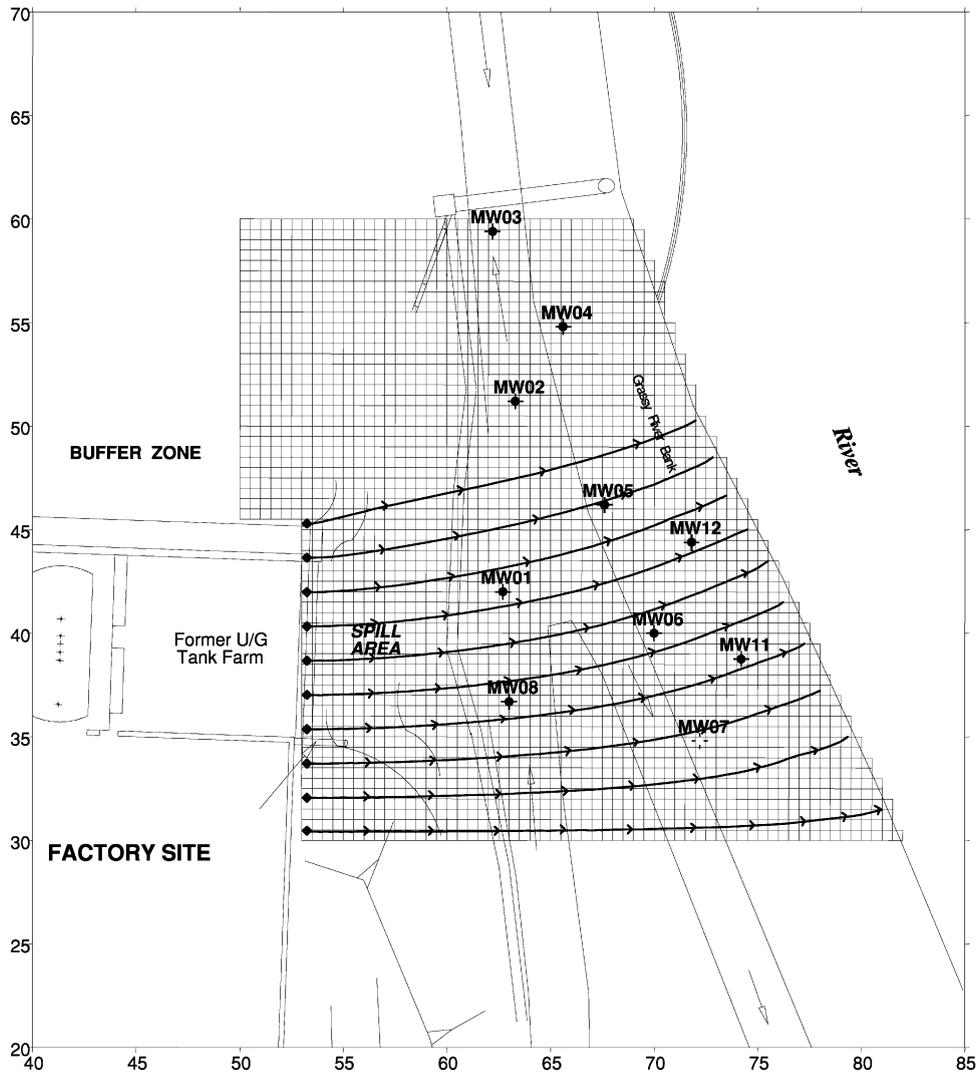


Fig. 4. Simulated particle tracking and flowpaths (units in m).

The average concentration of *n*-alkanes in groundwater over the period from December 1997 to August 1999, was 26 mg/L of *n*-alkanes, and the value for BTEX was considerably lower at <0.05 mg/L. Based on a median hydraulic conductivity of 0.4 m/day (estimated from the site data), the median volume of groundwater entering the gate was estimated to be $\sim 6 \text{ m}^3/\text{day}$. The maximum volume of water entering the gate, based on a conservative estimate, was expected to be $\sim 43 \text{ m}^3/\text{day}$. Assuming that the average concentration of petroleum hydrocarbons in groundwater was 26 mg/L, the maximum mass of petroleum hydrocarbons entering the funnel and gate was calculated to be 1.1 kg/day, with a median value of 160 g/day.

Assuming a median removal capacity of at least 20 kg/t peat (dry weight basis), as determined from previous studies with a range of peats tested for absorbing mono aromatic hydrocarbons [8], it was expected that the petroleum hydrocarbons in the groundwater entering into the funnel and gate (i.e. 1.1 kg/day), would be treated by the system for up to ~ 210 days before breakthrough of petroleum hydrocarbons. This period would be longer at 1448 days, assuming 160 g/day of petroleum hydrocarbons in the influent, entering the gate. These estimates, derived from [8], are conservative because they do not take into account a highly active microbial population which would be contributing to the apparent absorption (i.e. removal) process in the gate at the current site. Field measurements (using a portable GC-

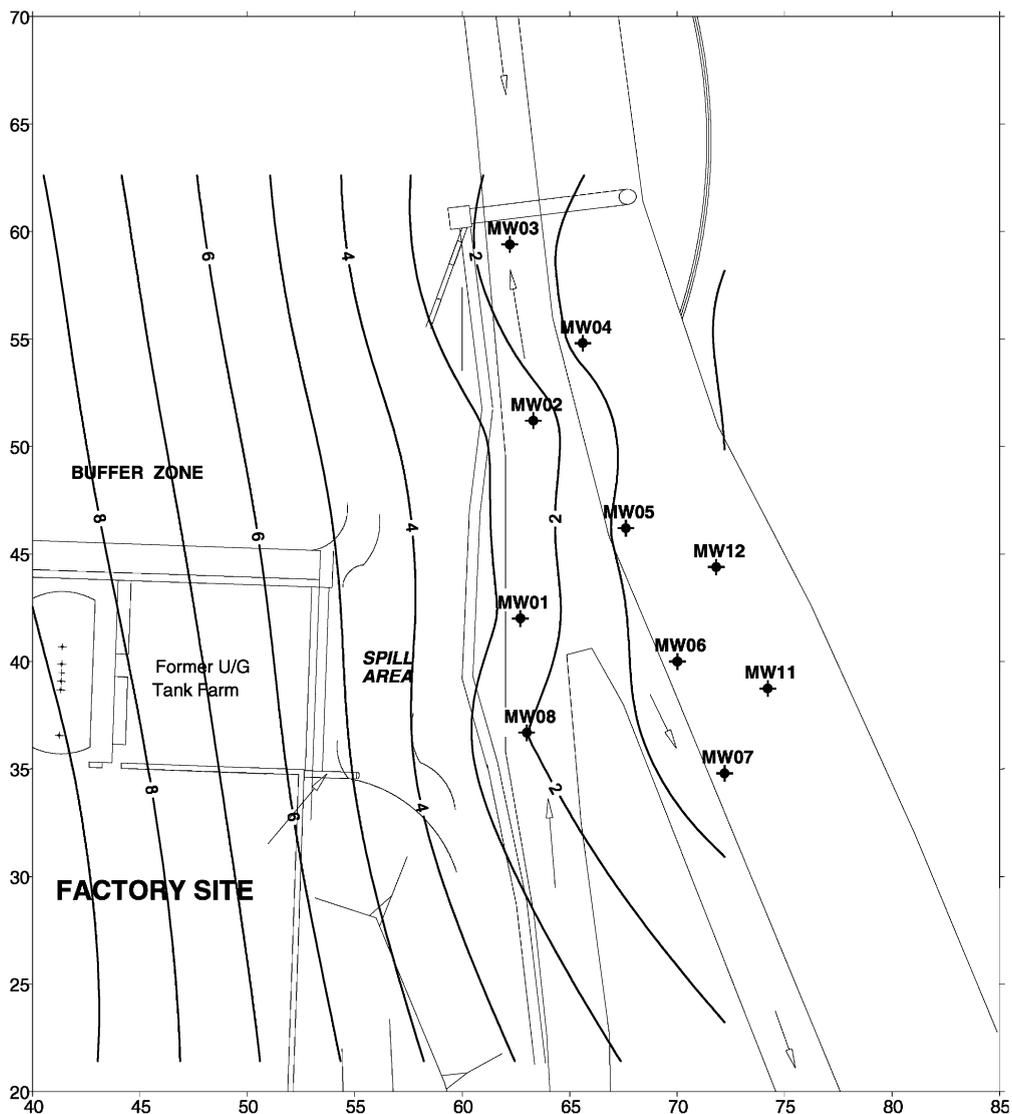


Fig. 5. Measured and interpolated piezometric surface on 7 October 1998 (units in m).

FID) indicated that there were no detectable volatilization of petroleum hydrocarbons from the sparging at the site. Therefore the petroleum hydrocarbon removal processes in the gate were primarily due to a combination of biodegradation and adsorption. At 306 days, the overall performance of the funnel and gate has been satisfactory.

3.4. Contaminant removal efficiencies

Concentrations of *n*-alkanes, toluene, ethyl benzene, and xylene, have substantially decreased in concentration between MW 13 (upgradient) and MW 14 (down-

gradient) of the funnel and gate. The removal efficiencies for the as-built funnel and gate system varied from 63% to 96% for the monoaromatic hydrocarbons, toluene, ethyl benzene, and xylene. Removal efficiencies for C₆–C₉, C₁₀–C₁₄, and C₁₅–C₂₈ *n*-alkane fractions were 69.7%, 81.1% and 67.2%, respectively. The lowest removal efficiencies were 54% for the C₂₉–C₃₆ *n*-alkane fraction. The overall efficiency for the funnel and gate system was 72% during the 10 month period over which the data were collected (Table 2). Removal efficiencies tended to decrease at 188 and 306 days with respect to toluene, ethyl benzene, and xylene and C₆–C₉ *n*-alkane fraction. Further sampling and analysis in the future will

Table 2
BTEX groundwater monitoring data from upgradient and downgradient of dissolved phase plume ($\mu\text{g/L}$)

Days	Toluene		Ethyl benzene		Xylene		C ₆ –C ₉		C ₁₀ –C ₁₄		C ₁₅ –C ₂₈		C ₂₉ –C ₃₆	
	UG ^a	DG ^b	UG	UG	DG	UG	DG	UG	DG	UG	DG	DG	UG	DG
0	1	1	6	280	250	990	1100	410	370	100	190	10	120	150
8	1	1	15	310	110	2600	50	2900	100	200	100	4	63	17
34	1	1	18	530	60	7400	2300	2200	950	1900	710	4	120	32
41	44	2	1	320	130	1900	560	780	440	100	100	1	170	62
48	310	3	1	170	80	3100	400	3500	100	320	100	2	151	26
58	40	2	1	780	110	3200	340	6600	630	50	80	3	9	1
153	15	1	1	230	60	2000	150	580	500	180	120	1	38	16
188	2	1	1	20	20	2400	780	750	480	100	100	2	2	16
215	17	1	1	270	30	2700	270	3500	410	140	0	1	<1	<1
245	1	1	110	940	270	2800	440	3000	510	150	120	16	840	250
274	1	1	1	340	30	1400	530	1700	730	150	0	1	75	11
306	1	1	5	100	80	2200	410	3100	740	100	100	6	10	18
Mean	36.2	1.3	13.4	358	110	2724	611	2418	497	308	143	4.3	145	54.5
Upper 95% CI	50.7	1.6	16	773	173	3642	1176	4397	823	416	164	5.5	388	65.3
Removal (%) ^c	Toluene		Ethyl benzene		Xylene		C ₆ –C ₉		C ₁₀ –C ₁₄		C ₁₅ –C ₂₈		C ₂₉ –C ₃₆	
Upper 95% CI	96.9		65.4		83.2		77.6		67.7		81.3		60.5	
Average	96.3		68.3		62.5		69.2		77.6		79.5		53.5	
Modal average	95.4		64.1		75.0		69.7		81.1		67.2		61.8	

^a Referring to upgradient bore (Bore MW 13).

^b Referring to down gradient (Bore MW 14).

^c Efficiency (%) of removal = [contaminant in groundwater in downgradient well] / [contaminant groundwater in upgradient well] \times 100.

ascertain whether this represents a decreasing trend in the performance of the funnel and gate. The apparent increase in concentrations of both the *n*-alkanes and toluene, ethyl benzene and xylenes in the upgradient well at day 245 correlates with the heavy rain in days before the samples were taken at that monitoring time. The removal efficiency data show that the model simulations were accurate in predicting the average removal efficiency of the contaminants in the dissolved phase.

Previous studies have shown that peats from various sources can remove 63–97% dissolved phase benzene, toluene, and xylenes from contaminated groundwater in laboratory studies and as these data were used in the selection of peat for the current study [8], the same range has been obtained in the current study [11], reporting efficiencies of BTEX removal from laboratory groundwater undergoing remediation with a briquet-peat barrier system, stated values of 86%, 71%, 43% and 28% for toluene, ethylbenzene, *m*-xylene and *o*-xylene, respectively. The removal efficiencies reported in the current field study were therefore considerably higher than those reported in previous laboratory studies. In another laboratory study, a peat column decreased petroleum hydrocarbon concentrations in an oil water mixture by 100-fold at 30,000–70,000 mg/L [12]. However, with a much lower dissolved phase concentration of 5 mg/L, the peat column was ineffective in removing

the petroleum hydrocarbons [12]. The literature values therefore vary considerably for petroleum hydrocarbon removal effectiveness. These previous studies reinforce the importance of testing local peat types prior to deploying peat at full-scale.

The downgradient monitoring well reported concentrations of petroleum hydrocarbons that were below the surface water guidelines, demonstrating that the funnel and gate system is also meeting the regulatory requirements. The surface water guidelines relevant to this site (Victorian State Environmental Protection Guidelines) were 250 $\mu\text{g/L}$ for both toluene and ethyl benzene and 10,000 $\mu\text{g/L}$ for the C₆–C₉ and C₁₀–C₁₄ *n*-alkanes. These guidelines were achieved with the constructed funnel and gate system.

4. Implications

The technical effectiveness of the funnel and gate, over the 10 month operating period in which the data were collected, demonstrates that the funnel and gate system has been effective in treating dissolved phase petroleum hydrocarbons in the shallow groundwater at the site. Ongoing measurements of the technical effectiveness of the funnel and gate system will be conducted over a longer term, by monitoring at 3 month

intervals to ensure continued effectiveness of the funnel and gate system. This is important as previous laboratory studies have shown that BTEX removal efficiencies decrease with time [8,12]. There are two key areas recommended for further research. These are (1) determining the mechanisms by which the petroleum hydrocarbons are removed from the groundwater and to monitor any deleterious effects on removal efficiencies from any potential increase in the biomass associated with any microbial growth in the peat within the gate, and (2) determining the effect of peat on the quality of the groundwater to ensure that these are not deleterious.

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