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A pilot study for the selection of a bioreactor for remediation of groundwater from a coal tar contaminated site

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Abstract

Coal tars in soil at a gasworks site in South Eastern Australia led to groundwater contamination with polycyclic aromatic hydrocarbons (PAHs), mono-aromatic compounds (BTEX) and phenols. The scope of the study included testwork in laboratory scale bioreactors and evaluation of available commercial groundwater treatment units. Two bioreactor configurations, a submerged fixed film reactor (SFFR) and a fluidized bed bioreactor (FBR) were effective, with high efficiencies of contaminant removal (typically >90%) over a range of hydraulic retention times (HRT) (3–29 h). Specifically, concentrations of total PAH, naphthalene, pyrene and total phenols in the feedstock and effluent of the SFFR were 123, 60, 51, 1.38 and 0.004, 0.001, 0.004, 0.1 mg/l, respectively. The FBR was only marginally less effective than the SFFR for the same groundwater contaminants. Discharge to sewer was the most appropriate end use for the effluent. SFFRs are regarded as being simpler in design and operation, and a commercially available unit has been identified which would be suitable for treating small volumes (<10 m^3 per day) of contaminated water collected at an interception trench at the site. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

1.1. Soil and groundwater contamination from gasworks sites

Former gasworks (or manufactured gas plant (MGP)) sites typically have soil and groundwater contaminated with various petroleum hydrocarbons [1–4]. Contaminants typically

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include polycyclic aromatic hydrocarbons (PAHs), phenols and various inorganic contaminants [5]. These are derived from coal tars and other residues that were often stored and/or disposed of at such facilities.

1.2. Groundwater treatment processes

Various bioreactor designs have been developed for treatment of groundwater contaminated with dissolved organic contaminants [6,7]. The most effective biological processes available for treating groundwater contaminated with organic compounds derived from coal tars use either submerged fixed film reactors (SFFRs) or fluidized bed bioreactors (FBRs).

Typically, SFFRs have been found to be more reliable than FBRs in the field. Process stoppages (such as those caused by power failures) often cause difficulties in re-establishing FBRs to normal and effective operation. However, fluidized beds and stirred tank bioreactors do allow for more effective aeration and mixing of bioreactor contents with the influent stream [8]. The increased mixing leads to increased contaminant removal efficiency. SFFRs are capable of operating in a number of different ways to treat liquid wastes [9]. SFFRs are also simpler in design than FBRs and require very little operator attention. The major advantage of SFFRs over the FBR design is that there is less vertical mixing, which enables a plug-flow regime to be established. This reduced mixing also means that SFFRs are suited to anaerobic degradation of contaminants.

In designing and operating a biological groundwater treatment process, a number of issues must be managed [7]. For example, bioreactors are typically inoculated with an acclimatized population of contaminant-degrading microorganisms prior to commissioning. Furthermore, both SFFRs and FBRs require the development of an active biofilm on a suitable support medium. The biofilm support material typically used in FBR treatment units is activated carbon. A number of different packing materials are used in SFFRs including kaolin, diatomaceous earth, polystyrene, wood chips, sand and ceramic saddles [7]. Activated carbon has been investigated for MGP applications because of its high capacity for retaining potentially toxic organic compounds (such as phenols), as well as a large surface area for development of the biofilm of microorganisms. These factors need to be considered in the selection of a process for treating groundwater contaminated with organic compounds such as coal tar contaminants. These factors are listed in Table 1 and have been considered in conducting the current study.

1.3. Aims and scope

Coal tars present in soil at a former gasworks site, contributed to groundwater contamination, underlying the site. This site is located in a major city in south eastern Australia, and is situated adjacent to a river, with the potential for discharge of contaminants to the river. The objective of the study was to assess the options for a field-scale water treatment process for the site. The principle aims of the study were to: determine the design parameters for a full-scale on-site treatment process for the contaminated groundwater to specified water quality standards (for discharge to sewer), and evaluate the principal bioreactor configurations and operating modes, to provide engineering designs and cost estimates for equipment procurement.

Table 1
Important parameters for establishing a bioreactor for treating contaminated groundwater[a]

Parameter	Description/importance
Temperature	As with all bioprocesses, treatment rates vary with temperature
HRT	Flexible handling of influent flow rates is required for effective control
Influent quality and flow direction	Variations in influent quality requires equalization or some other method of compensation; influent typically contains a mixture of contaminants, each with different degradation rates; it is important to minimize input of suspended solids and free or emulsified oily phases; selection of flow direction (a down-flow system requires backwashing of the bioreactor packing, whereas an up flow design will reduce clogging)
Mixing regime	Selection of mixing regime is important: a bioreactor may be set up as a completely mixed stirred tank or for plug-flow
Biomass	Potential problems arising in the attachment and detachment (sloughing) of biomass; biomass can build up and large oxygen demands associated with aerobic biotreatment processes may occur; potential problems with foaming (also dependent on types of chemicals in influent)
Nutrient requirements	Varies with contaminants
pH	pH control is critical and groundwater may not be an effective buffer

[a] Sources: [13–18].

The scope of the study included: test work in two laboratory scale bioreactors (i) a FBR and (ii) a submerged fixed film bioreactor (SFFR). These are considered the most appropriate reactor types for this type of groundwater contamination. An evaluation of available commercial groundwater treatment units, comparing with a purpose-built unit, on the basis of design, cost, availability and performance criteria, was also conducted. Consideration of recovery and/or remediation of tars present at the site is beyond the scope of this study.

2. Methodology

2.1. Groundwater sampling

A preliminary site assessment was conducted and 8 bores from around the site were analyzed for pH, phenols, BTEX, naphthalene, total petroleum hydrocarbon (TPH), PAH and ammonia. Previous analyses conducted on the groundwater from the site indicated that there were no inorganic compounds (such as cyanides) present in the groundwater. For the biotreatment design study, three 200 l drums were used to collect the groundwater and these were dispatched from the site. Drum 1 contained 200 l of interception trench water, Drum 2 contained 200 l of groundwater from 4 bores across the site, and Drum 3 contained approximately 150 l of trench water. Subsamples of these groundwaters were sent for chemical analyses. At the time of sampling, several bores contained viscous tarry liquid that was too thick to pump. Samples were not taken from these bores.

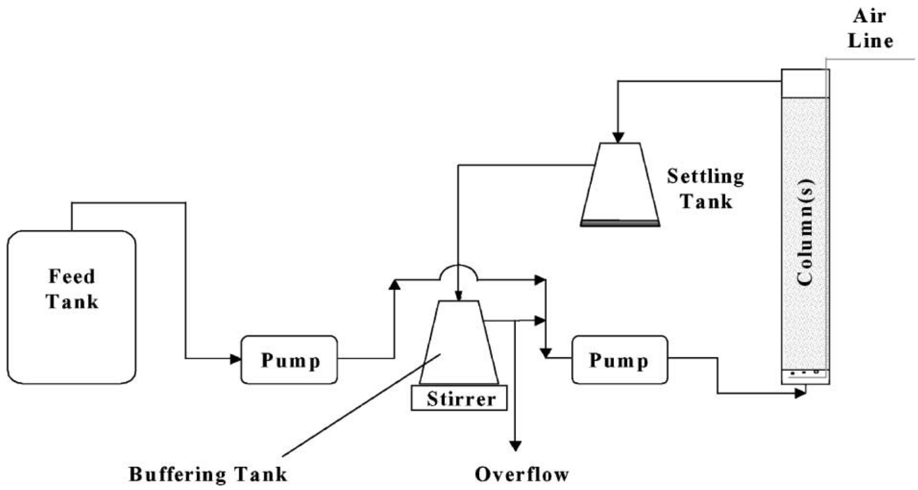

Fig. 1. Schematic of laboratory scale bioreactors.

2.2. Trial design

2.2.1. General

The following types of bioreactors were used: a SFFR (Column 1) and a FBR (Column 2). The bioreactors were constructed from columns of perspex tubing (80 mm diameter × 480 mm length). The experimental set-up is shown in Fig. 1. The FBR packing was wood-based carbon (CALGON BPL 4 × 10 mesh) and the SFFR packing was bituminous-based carbon (PICABIOL TE 1.2). The bioreactors were filled to capacity with the recirculating liquor. An air sparger (65 mm diameter ring of plastic tubing with 10 holes) was placed at the bottom of each of the bioreactors. Air was delivered into the sparger of the FBR to provide a flow rate of approximately 10 ml/min. A lower flow rate of 1–4 ml/min was delivered into the base of the SFFR. The columns were operated in upflow mode. Groundwater was fed into the base of each of the bioreactors using a peristaltic pump which was preset to deliver slow, medium or high flow rates, providing three different hydraulic retention times (HRT) (Table 2). The definition of the HRT is the time for which the influent (groundwater) resides

Table 2
Liquid flow rates and HRT

Trial number[a]	SFFR		FBR	
	Flow rate (ml/min)	HRT (h)	Flow rate (ml/min)	HRT (h)
1	2.4	32.6	3.0	26.1
2	6.1	12.8	7.0	11.2
3	22.5	3.5	23.5	3.3

[a] Trial 1 was conducted over a 6 days period, Trial 2 over 8 days and Trial 3, 5 days.

in the bioreactor. Quantitatively, this is related to the influent flow rate as follows:

$$\mathrm{HRT} = \frac{\text{net volume of bioreactor}}{\text{influent flow rate}}$$

The influent flow rates and corresponding HRT values are listed in Table 2 for each of the three trials. At the start of each of the trials, the flow rates of influent into both the bioreactors was adjusted to provide nominal HRTs of 24, 12, and 4 h. The actual HRT values achieved for each of HRT columns in the three trials were slightly different to these values (Table 2). The three trials were conducted at a temperature of 23°C.

2.2.2. Preparation of groundwater feed

Due to very low concentrations of contaminants present in the groundwater received from the site, a synthetic groundwater was prepared to contain kerosene (10 mg/l), phenol (20 mg/l), naphthalene (40 mg/l), pyrene (10 mg/l), and coal tar (0.1–0.25 g/l). Batches of 10 or 20 l were prepared as required.

2.2.3. Bioreactor operation and maintenance

Both bioreactors were aerated throughout the course of the trial. This provided oxygen to the attached aerobic microorganisms. The air also served to keep the activated carbon support fluidized during the trial. The air flow rate in the FBR was maintained at 8–10 ml/min and was regulated by a rotameter on a compressed air supply. The air flow rate in the SFFR was maintained at 1–4 ml/min using a small fish tank aerator. A solution of nitrogen and phosphate salts was dosed into each of the bioreactors to provide a C:N:P ratio of 100:10:1.

2.2.4. Microbial enrichment

Groundwater samples (5 ml) were obtained from the site. These were used to inoculate enrichment media containing phenol (0.05%), tar (0.5%), toluene (0.5%), and naphthalene (0.5%) and minerals. These cultures (100 ml) were shaken under aerobic conditions in 250 ml shake flasks for ∼2 weeks prior to being introduced to the bioreactors. Light microscopy was used to assess the development of the enrichment cultures prior to inoculation of the carbon packing.

Table 3
Methods used in the study

Parameter	Method source	Method number[a,b]
pH	USEPA	150.1
COD	APHA	5220
Phenols	USEPA	8321 (HPLC)
PAH	USEPA	8100 GC–MS
BTEX[c]	USEPA	5030B (Purge and Trap)
TSS[d]	APHA	2540D

[a] Recoveries for phenols, PAHs, BTEX from the groundwater samples varied between 90 and 100%.
[b] Methods are described in detail elsewhere [19,20].
[c] BTEX: benzene, toluene, ethyl benzene and xylenes.
[d] TSS: total suspended solids and is used as a measure of biomass.

2.2.5. Bioreactor sampling and analysis

Samples (10–50 ml) of influent and effluent were taken from each of the bioreactors at the start and completion ($t = 5$–8 days) of each of the three trials. Samples for head-space analyses (collected in 25 ml VOA vials) were stored at 4–6°C and samples for PAH and phenol analyses (collected in screw cap glass jars) were stored at −18°C until time of analysis. A summary of the analytical methods used in the trials is given in Table 3.

3. Findings

3.1. Site groundwater assessment

A preliminary investigation of soil and groundwater conditions at the gasworks indicated the presence of coal tar residues in soil. These residues were contributing to groundwater contamination. The presence of significant levels of ammonium is expected to aid biological degradation of the organic contaminants, since it will supply nitrogen, an essential nutrient for hydrocarbon degradation. The quality of groundwater under the gasworks site is summarized in Table 4. From the site investigations, it was estimated that the flow rates of contaminated groundwater, to be extracted from the site and treated, are ~1 m^3 per day.

Table 4
Key groundwater chemistry parameters at the gasworks site[a]

Bore	pH	Phenols (mg/l)	Benzene (mg/l)	Toluene (mg/l)	Naphthalene (mg/l)	Total PAHs (mg/l)	TPH (mg/l)	NH_4 (mg/l)
1	6.1	0.3	–	–	0.2	0.7	2	69
	5.8	61	0.3	0.1	1.2	1.7	141	84
2	7.2	90	–	–	60	182	620	321
	6.8	79	13	33	127	1864	13700	520
3	7.2	–	–	–	0.8	0.5	–	0.6
	–	–	–	–	–	0.5	–	–
4	6.6	170	–	–	25	55	170	141
	6.5	38	0.4	0.6	3.1	5.7	137	68
5	6.6	0.5	–	–	–	–	–	–
	6.5	ND	ND	0.01	0.17	0.3	2	3.2
6	7.4	730	–	–	4	4.7	4	1105
	7.1	480	5	0.2	5.8	7.0	786	680
7	7.0	1.2	–	–	0.07	0.1	–	8.6
	6.6	0.2	ND	0.03	0.18	0.3	2.9	7
8	6.7	170	–	–	0.4	1.5	2	22
	6.5	14	1.37	3.65	260	654	17300	60

[a] ND: not detected; no cyanide compounds were present from earlier reports (data not shown).

Table 5
Contaminant analyses of groundwater[a]

Contaminant	Drum 1[b]	Drum 2	Drum 3
Total BTEX (mg/l)	0.34	1.1	4.0
Total phenols (mg/l)	0.4	0.9	17.0
Total PAHs (mg/l)	0.23	1.4	8.1

[a] Drums 1 and 2 were from bores on the site.
[b] Drum 3 was representative of groundwater from the trench.

3.2. *Characterization of groundwater samples*

The results of the groundwater analyses are presented in Tables 5 and 6. These results indicate that there were only relatively low concentrations of monoaromatic hydrocarbons, benzene, toluene, ethyl benzene, xylene (BTEX), phenols, and PAHs present. These concentrations were relatively low compared with the data from the field analysis which included concentrations of up to 700 mg/l phenol and 1000 mg/l C_{10}–C_{14} *n*-alkane petroleum hydrocarbons in groundwater from selected bore holes from the site. The chemical oxygen demand (COD) of the groundwater samples was higher, up to 5700 mg/l (Table 5). This reflects the presence of a range of other organic compounds, presumably also deriving from the coal tar.

The total suspended solids (TSS) in the groundwater samples were low. Assuming the groundwater samples tested in the study are representative of the groundwater to be treated, than this finding suggests that only a small sedimentation or pretreatment tank would be required to remove solids prior to treatment in an on-site bioreactor. Note that the tank size would also need to be sufficient to provide for separation and collection of any free-phase oil or tar.

3.3. *Removal efficiencies of organic contaminants in bioreactor trials*

The bioreactor trials were aimed at determining the contaminant removal efficiencies in both the SFFR and FBR for three different loadings. The findings from the three trials in which both reactors were operated at different HRTs (see Table 2), are presented in Table 7. Total BTEX compounds in the feedstock were ∼0.7 mg/l (or 700 ppb) and the total phenols 1.38 mg/l (or 1380 ppb). The concentrations of total PAHs were relatively high at ∼120 mg/l

Table 6
Chemical and physico-chemical analyses

Parameter	Drum 1[a]	Drum 2	Drum 3[b]
COD (mg/l)	2200	2000	5700
pH	6.9	7.0	8.3
TSS (mg/l)	25	<10	460

[a] Drums 1 and 2 were from bores on the site.
[b] Drum 3 was representative of groundwater from the trench.

Table 7
Summary of bioreactor performance

Contaminant (mg/l)	Column 1: SFFR				Column 2: FBR		
	Feedstock	Initial[a] concentration (mg/l)	Final[b] concentration (mg/l)	Removal (%)	Initial concentration (mg/l)	Final concentration (mg/l)	Removalv (%)
Trial 1							
Total phenol	1.38	0.1	0.1	92.8	0.1	0.1	92.8
Naphthalene	60	0.001	0.001	100.0	0.004	0.016	100.0
Pyrene	51	0.001	0.004	100.0	0.004	0.001	100.0
Total PAHs	123	0.001	0.004	100.0	0.02	0.033	100.0
Benzene	0.2	0.15	0.001	99.5	0.16	0.001	99.5
Toluene	0.16	0.19	0.001	99.4	0.19	0.001	99.4
Ethylbenzene	0.042	0.015	0.001	97.6	0.018	0.001	97.6
Total xylenes	0.31	0.23	0.003	99.0	0.26	0.003	99.0
Trial 2							
Total phenol	1.38	0.1	0.1	92.8	0.1	0.1	92.8
Naphthalene	60	0.001	0.001	100.0	0.002	0.003	100.0
Pyrene	51	0.034	0.015	100.0	0.1	0.02	100.0
Total PAHs	123	0.034	0.016	100.0	0.12	0.024	100.0
Benzene	0.2	0.001	0.001	99.5	0.001	0.001	99.5
Toluene	0.16	0.001	0.001	99.4	0.001	0.001	99.4
Ethylbenzene	0.042	0.001	0.001	97.6	0.001	0.001	97.6
Total xylenes	0.31	0.003	0.003	99.0	0.003	0.003	99.0
Trial 3							
Total phenol	1.38	0.1	0.1	92.8	0.1	0.1	92.8
Naphthalene	60	0.001	0.002	100.0	0.008	0.02	100.0
Pyrene	51	0.035	0.13	99.7	0.1	0.18	99.6
Total PAHs	123	0.036	0.23	99.8	0.11	0.43	99.7
Benzene	0.2	0.001	0.001	99.5	0.001	0.005	97.5
Toluene	0.16	0.001	0.001	99.4	0.001	0.013	91.9
Ethylbenzene	0.042	0.001	0.001	97.6	0.001	0.001	97.6
Total xylenes	0.31	0.003	0.003	99.0	0.003	0.003	99.0

[a] Initial concentrations at inlet into bioreactor.
[b] Final concentrations on outlet of bioreactor.

(or 120,000 ppb). The concentrations of BTEX and phenols in the synthetic groundwater were somewhat lower than expected (based on the water solubilities of these compounds). This may have been due to laboratory artefacts such as losses due to volatilization or binding to the glass feedstock reservoir. At a HRT of ∼29 h, the efficiency of removal of all contaminants was >90% in both reactors. The results indicated that the SFFR was as effective as the FBR for removing phenol, BTEX, and PAHs from the groundwater.

At considerably lower HRTs of 3 and 12 h, the removal efficiencies after 5–8 days were also >90%. The results of the first set of analyses indicate removal efficiencies were, however, considerably lower and more variable at the start of the trials. The high removal efficiencies were obtained after a short period (5–8 days) of bioreactor operation. These efficiencies, together with microscopic observations of the recirculating liquor in the bioreactors, indicate that substantial biological activity occurred in both the bioreactors during the three trials conducted.

These results indicate that a SFFR is as efficient as a FBR in treating the organic contaminants in the groundwater and that high removal efficiencies can be obtained for all the organic contaminants tested over a range of HRTs.

3.4. Monitoring organic carbon and biomass production

Visual observations were made of samples collected throughout the trials. During Trial 1, no suspended solids (derived from biological growth) were observed in the effluent from either bioreactor at the beginning of the trial. However, at the completion of Trial 1, relatively large amounts of carbon fines were observed in the effluent from the FBR. During Trials 2 and 3, the amount of carbon fines in the effluent from both columns was negligible, indicating that attrition occurred in the first week of operation and only in a substantial way in the FBR. The amount of suspended solid due to biological growth had increased 2–4-fold in both reactors during Trials 2 and 3.

At the completion of all three trials, it was apparent that there had been a net loss of carbon packing from the FBR of ∼20%. There was considerably less loss of carbon packing from the SFFR (<5%).

As a result of attrition of the carbon packing in the bioreactors during the trial, carbon fines were present in all samples collected during the trial. The presence of carbon fines would interfere with COD and TSS analyses, so these were not conducted. Foam generation is a common characteristic of biotreatment systems, however, no foaming was observed to occur during the trials.

3.5. Selection and availability of a commercial groundwater treatment unit

The results of the laboratory study indicated that a SFFR could operate with a similar efficiency to a FBR on the same contaminated feedstock. Groundwater treatment processes, based on the SFFR concept, are commercially available. Engineering designs and specifications, cost estimates for construction, installation, commissioning and operation are all available from the manufacturers. This mini review highlights some of the potential problems associated with the groundwater treatment process. These particularly relate to

Table 8
Comparison of bioreactor costs[a]

Cost component	Supplier 1	Supplier 2
Reactor type	Fixed film, plug-flow	Fluid bed
Packing	PVC	GAC
Capacity[b]	10 gpm (3 m^3/h)	30 gpm (10 m^3/h)
Air supply	Blower, membrane diffuser	O_2 generator
Nutrient storage and delivery	Yes	Yes
Capital cost[c] (AUS$)	42000	132000
Operating cost (AUS$; AUS$ per m^3 treated)	0.05	0.30

[a] Cost in AUS$ = 0.55US$ (as at 1997).
[b] Estimates of flow rates of contaminated groundwater to be extracted from the site are ∼1 m^3 per day. Working volumes of both reactors are <1 m^3.
[c] This includes start up costs.

demonstration of the fate of influent contaminants (i.e. the proportion actually undergoing biodegradation as compared with other non-biological processes).

Two commercial bioreactors were evaluated (Table 8). The bioreactor module and associated equipment have a 2 m × 5 m base area, and both would be conveniently located in a container or small shed on the site. Preliminary cost estimates were obtained from both suppliers, which are summarized in Table 8. One of the commercially available technologies has previously been reviewed by the USEPA also for groundwater contaminated with PAHs [10].

3.6. Development of water quality criteria for bioreactor discharge

The river should not be used for discharge of treated effluent, due to the risk of contaminant release in the event of bioreactor failure. Discharge to sewer is likely to be more appropriate for the site. A nearby sewer connection exists at the site. Australia's recent national environmental protection measure (NEPM) lists specific acceptance guidelines for organic compounds [11]. The AWRC [12] guidelines include concentrations of organic compounds that inhibit activated sludge sewage treatment processes. The guideline concentrations for the relevant compounds are listed in Table 9. These guidelines do not necessarily reflect consideration of risks associated with the presence of organic compounds in the treated groundwater at the site. It is necessary for the site owners to confirm the effluent quality criteria that will need to be met. Nevertheless, the effluent quality achieved during this trial suggests that the AWRC [12] criteria, if imposed, will be easily met by the biotreatment process. All effluent from the trial bioreactors contained concentrations of benzene less than the guideline for discharge to sewer (Table 9). Only one value of 0.013 mg/l toluene was reported in the effluent. This was from Trial 3 in the FBR with the lowest HRT tested (∼3.5 h). However, the biotreatment process will not provide an effluent stream that is sufficiently clean for disposal back to groundwater or to a freshwater ecosystem.

The concentrations of total PAHs were less than 0.04 mg/l in all the effluent samples except in Trial 3 where the concentrations were up to 0.43 mg/l. Benzo(α)pyrene, a carcinogenic PAH, was below detection limits in the effluent of all the trials except Trial 3

Table 9
Applicable national water quality guidelines for treatment effluent

Parameter	Groundwater quality	Trade waste to sewer[a]
pH	No guideline[b]	6–10
Phenols (mg/l)	0.05[c]	100
Benzene (mg/l)	0.3[c]	125
Toluene (mg/l)	0.3[d]	35
Naphthalene (mg/l)	No guideline[c]	500
Total PAHs (mg/l)	0.003[c]	5
Petroleum hydrocarbons (TPH) (mg/l)	No guideline	30
NH_4	No guideline	No guideline

[a] Developed with the local regulatory authority (as is the case across Australia) and are not national guidelines.
[b] For marine and fresh water aquatic ecosystems [11].
[c] For freshwater aquatic ecosystems as published in the NEPM for protection of Australian groundwater [11].
[d] AWRC [12].

where the concentrations reached 0.005 and 0.025 mg/l in the SFFR and FBR, respectively. In all the trials, the total phenols were below the detection limits (0.1 mg/l).

4. Conclusions and recommendations

The bioreactor configurations established and trialed in this design study were effective in removing the hydrocarbons in the ‘synthetic’ groundwater feedstock. Both the FBR and SFFR demonstrated high efficiencies of contaminant removal (typically >90%) over the three HRT tested. At a HRT of approximately 29 h, it is apparent that the effluent to be discharged from either a SFFR or FBR process is likely to be acceptable for discharge to sewer.

Since SFFRs are considerably simpler in design and operation, it is advisable to consider procuring a treatment unit based on this process. Such a unit can be purchased ‘off-the-shelf’ and would require minimal adaptation for the site. A relatively simple and low cost unit is available from a supplier which would be suitable for treating the relatively low volumes of contaminated water collected in the interception trench on-site. The use of a non-carbon packing in this unit avoids problems with carbon fines generation. However, activated carbon packings act as a very effective buffer against toxicity of the influent towards the bound microorganisms. Thus, in the absence of carbon, operational precautions would need to be taken to avoid shock effects to the bound microorganisms.

SFFRs would be a preferable option for the treatment of the groundwater at the coal tar contaminated site described in this study. However, if contaminant concentrations were higher, then the higher aeration rates employed in the FBR could make the FBR design superior.

Discharge to sewer appears to be the most appropriate disposal for the effluent. More recent groundwater monitoring data (concentrations of the various contaminants) and groundwater generation rates will need to be collected and compared with previous estimates of concentrations and volumes. This will enable equipment vendors to provide a quotation for the supply of a treatment unit to treat the groundwater on-site.

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