The feasibility of a permeable reactive barrier to treat acidic sulphate- and nitrate-contaminated groundwater

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Abstract

Manufacturing of fertilisers at a site in Johannesburg, South Africa, has resulted in the localised contamination of the shallow groundwater. The latter migrates through the weathered granite zone and emerges as base flow to the natural drainage system that ultimately exits the site. The water quality is acidic and is contaminated with respect to nitrate, sulphate, aluminium, fluoride and manganese. Treatment of this water is therefore necessary before discharge.

In situ passive rehabilitation of groundwater using a permeable reactive barrier offers a potentially cheaper alternative to conventional treatment technologies for acidic groundwater. Feasibility criteria used in this study were based on the suitability of the site for the installation of a permeable reactive barrier (PRB) and on the amenability of contaminants for treatment using locally available reactive media. The suitability of the reactive media was assessed from laboratory leach tests on reactive materials (mushroom compost, dolomite and limestone). These tests were followed by batch testing of various composites of the mushroom compost and the neutralisation gravel media (dolomite and limestone for comparison).

The results of the study indicated that the site conditions were potentially suitable for the installation of a PRB but that the mushroom compost is not suitable as a carbon source. This was indicated by the limited success achieved in reducing sulphate concentration and the poor quality of the leachate with respect to sodium, chloride, ammonia and potassium. The neutralisation capacity of the dolomite aggregate was similar to that of the limestone for the batch tests. However, it can be expected that the long-term performance would be inhibited by clogging and armouring by aluminium and manganese precipitates.

Keywords: permeable reactive barrier, nitrate, groundwater, dolomite

Introduction

A large fertiliser manufacturing facility near Johannesburg, South Africa, has been in operation since 1896. Various activities have been undertaken on the site, from the manufacture of explosives for the military to the production of fertilisers and associated chemicals. Site assessments of the soil, surface and groundwater quality show that the seepage of contaminated water from the historical fertiliser factory area has resulted in the localised contamination of the shallow groundwater (SRK, 2002).

The shallow groundwater migrates through the highly weathered granite zone and emerges as base flow to the stream flowing through the contaminated area. The stream flows to the southwest where it joins an east-west tributary of the main river flowing through the site (Fig. 2). The flow of 3.1 m^3/h (flow varies from 1.3 m^3/h to 20 m^3/h) is acidic with concentrations of nitrate, sulphate, aluminium, fluoride and manganese in excess of the recommended receiving water quality objectives (Table 1).

The current treatment strategy for wastewater and effluent is the addition of lime to neutralise the water to a pH of 7. The water is gravitated to an evaporation dam and excess water is sprayed onto grasslands for nitrogen uptake. Effluent minimisation and alternative treatment strategies are being considered to reduce the impact on groundwater and to improve the quality in streams to within the compliance levels stipulated in the effluent site permit.

The initial proposal for rehabilitation of the site's contaminated groundwater was for the installation of a seepage cut-off trench immediately down-gradient of the factory area where the majority of the contamination emanated (SRK, 2001). Subsequent investigations indicated that the depth to bedrock is extremely variable (0.5 m to >12 m) making the installation of a cut-off trench along the proposed trench line neither practical nor effective for intercepting groundwater emerging into the stream further down-gradient (SRK, 2003).

A more practical option would be to intercept the groundwater where it is laterally constrained within the channelled stream approximately 1km down-gradient of the factory and where the depth to bedrock is shallower than that typically associated with the deep weathering profile on the slopes of the granite outcrop higher up the hill (SRK, 2003). *In situ* passive rehabilitation of groundwater using a permeable reactive barrier at this point could offer a potentially cheaper, low maintenance and self-sustaining alternative to pumping this water up the hill to the effluent treatment plant.

A permeable reactive barrier (PRB) is defined as 'an engineered treatment zone of reactive media that is placed in the subsurface and designed to intercept a contaminant plume, provide a flow path through the reactive media and transform the contaminants into an environmentally acceptable form to attain remediation concentration goals down-gradient of the barrier' (Carey et al., 2002).

An anoxic limestone drain (ALD) comprising a buried bed of limestone is an example of a PRB. The acidic water reacts with the limestone aggregate and results in the addition of alkalinity to the water. Reducing conditions are induced to limit the potential for armouring by the precipitation of iron or manganese oxides on the aggregate. A carbon source may be mixed with the limestone to enhance reducing conditions. A typical

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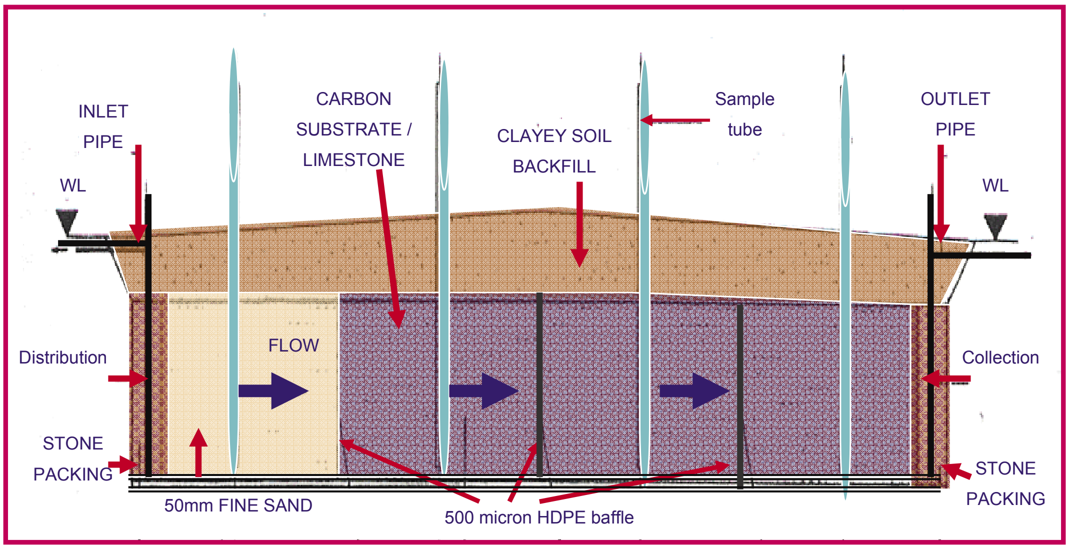

Figure 1
Diagrammatic representation of a PRB

cross-section of horizontal flow in an anoxic limestone drain is illustrated in Fig. 2 (Pulles et al., 2001).

Passive remediation strategies for inorganic species aim to utilise natural transformation mechanisms to reduce, retard, immobilise or transform potentially harmful inorganic species dissolved in the water into immobile species. This would be achieved by, for example, sorption, precipitation or biological transformation of the dissolved species to less polluting species within the barrier (Carey et al., 2002). Denitrification and sulphate reduction only occur in oxygen-deficient systems. Similarly, once nitrate is no longer available, conditions are optimised for sulphate-reducing bacteria to utilise the oxygen in sulphate as an electron acceptor, provided ferric iron is absent. This would typically result in increased reducing conditions along the groundwater flow path.

In order to comply with the site's effluent permit concentrations (otherwise termed the receiving water quality objectives) it would be necessary to neutralise the water from a pH of 3 to around 7 and decrease the concentrations of nitrate (520 to 12 mg/ℓ as N), sulphate (1 700 to 200 mg/ℓ), aluminium (120 to 20 mg/ℓ), manganese (64 to 0.4 mg/ℓ) and fluoride (1.5 to 2 mg/ℓ).

Optimal conditions for denitrification include a neutral pH, temperatures of around 25°C, anoxic conditions (Eh around 0mV), and a readily available carbon source. (Shrimali and Singh, 2001; Robertson and Cherry, 1995). Similarly, optimal conditions for sulphate reduction include a pH of between 5 and 8, anaerobic conditions (Eh<100 mV), sulphate concentrations of greater than 100 mg/ℓ and a readily available single-chain carbon source. (Hedin et al., 1994)

The reactive media that are used in the PRB must be sufficiently reactive to treat water for several years and should reduce contaminants of concern without leaching additional contaminants to the environment. The hydraulic permeability should be high enough to avoid clogging but sufficient to allow for the **residence time** required to allow the bacteria to reduce contamination to the required levels. The reactive media required for biologically mediated transformations must also support colonisation by the required bacteria.

Feasibility and laboratory tests

The feasibility of a using a PRB to treat the sulphate- and nitrate-contaminated acidic groundwater at this site was assessed using screening methods outlined for EPA sites by the USA National Groundwater and Contaminated Land Centre (Carey et al., 2002). The feasibility assessment was based on a site investigation and conceptual site model, used to determine the practical constraints to installing the barrier, and preliminary laboratory tests to assess the feasibility of using locally available reactive media in the PRB. The reactive media include mushroom compost, to be used as a carbon source for the biological transformation of nitrate and sulphate, and dolomite gravel for the dual purposes of neutralising acidity and creating permeability in the PRB.

Laboratory tests

The suitability of reactive media for the PRB was determined using laboratory leach tests on dolomite and mushroom compost, to determine the constituents that could potentially leach into the solution. Small-scale batch tests of various composites of the mushroom compost and gravel media were to assess the amenability of contaminants for treatment. The batch tests were used to establish the proportion of mushroom compost to neutralisation media that would be optimal for the PRB. Other laboratory tests included analysis of the seepage water and of the composition of the mushroom compost, dolomite and limestone. Small-scale batch testing of various composites of the mushroom compost and gravel media was conducted to determine the conditions that would occur in the PRB and to establish the optimal proportion of mushroom compost to neutralisation media.

Due to cost constraints, only mushroom compost was considered for the reactive media in the batch tests. The compost, derived from chicken manure and sterilised for sale to the public, was obtained from a nursery in Midrand. The dolomite aggregate was obtained from Lyttelton Quarry in Midrand and grades of <19 mm and <37 mm were selected for use in the batch tests (Taylor et al., 2001*).* Limestone was supplied by PPC cement at a similar grade of <20 mm.

Results and discussion

Seepage water analyses and composition

The detailed analysis of the water in the seepage sample is compared with the receiving water quality objectives required by

TABLE 1
Seepage analysis and receiving water quality objectives

Parameter	Receiving water quality objective	Seepage analysis
pH	5.5-8*	3.34
Eh (mV)		425
COD (mgO/*l*)	50*	44
Conductivity (mS/m)	120*	729
Nitrate as N (mg/ℓ)	10*	520
Ammonia as N (mg/ℓ)	1*	<0.2
Total N (mg/ℓ)		520
Total P (mg/ℓ)	1*	19
Sulphate (mg/ℓ)	200* (600)	1720
Aluminium (mg/ℓ)	2 (2)	120
Fluoride (mg/ℓ)	2*	82
Calcium (mg/ℓ)	80(3)	640
Magnesium (mg/ℓ)	100(3)	310
Manganese (mg/ℓ)	0.4* (5)	64
Iron (mg/ℓ)	2(2)	0.32
Strontium (mg/ℓ)	1(3)	5.4
Sodium (mg/ℓ)	180* (600)	140
Chloride (mg/ℓ)	100 (600)(3)	470
Potassium (mg/ℓ)	50 (400)(3)	120

Notes:* = Receiving water quality guidelines specified by DWAF for the site effluent permit
(1) = Drever, 1997
(2) = Concentrations must be <2 mg/ℓ to reduce the potential for armouring (Laine and Jarvis, 2003; Younger et al., 2003)
(3) = Domestic water quality guidelines; concentrations generally do not constitute a health risk. Concentrations in excess of concentration in brackets represent health risks (DWAF, 1996)

TABLE 2
Summary of analyses of limestone, dolomite and compost by ICP and of analyses from the de-ionised water leach tests

Parameter (mg/ℓ)	ICP analysis			De-ionised water leach	
	Limestone	Dolomite	Compost	Dolomite	Compost
PH				10.01	6.63
NO$_3$				<1.0	540
SO$_4$				<1.0	1710
PO$_4$				<2.0	130
Cl				9	510
F				0.18	<0.10
Al	70	130	1970	<0.08	0.2
As	18	35	47	<0.34	<0.34
Ba	7.3	7.7	150	<0.02	0.09
Ca	32 900	19 500	4 980	5.7	402
Cu	<10	<10	33	<0.02	0.03
Fe	170	610	910	<0.02	0.54
K	40	70	2 250	0.44	870
Li	<10	<10	<10	<0.02	<0.02
Mg	650	11200	500	13	95
Mn	420	490	40	<0.01	0.6
Mo	<10	<10	<10	<0.05	0.05
Na	10	20	620	1.6	170
Zn	<10	<10	150	<0.17	<0.17

TABLE 3
Paste pH and neutralising potential (NP)

Parameter	Dolomite	Limestone
Paste pH	9.34	8.75
pH at titration	1.62	1.67
Neutralisation potential CaCO$_3$ (kg/t)	1048	1018

the site effluent permit as stipulated by the Department of Water Affairs and Forestry (DWAF) in Table 1.

Composition of reactive media

An ICP metals scan, de-ionised water leach test and paste pH were undertaken for the mushroom compost, dolomite and limestone. The results are summarised in Table 2. The ICP analysis indicates that dolomite is relatively impure in comparison to the limestone and comprises higher concentrations of silica, manganese, iron and aluminium.

It should be noted that concentrations of aluminium and barium were unexpectedly high in ICP solids analyses for the compost sample. The reason for this is unknown. The deionised leach tests indicate, however, that the potential for concentrations of aluminium and barium to leach from the compost is negligible. The deionised water leach tests on the dolomite and compost indicate that calcium, magnesium, potassium and sodium will leach from the dolomite and sodium, phosphate, potassium, calcium, magnesium, nitrate, sulphate and chloride from the compost.

The saturated paste pH and available neutralising potential (NP) of the limestone and dolomite are presented in Table 3. The high positive NP indicates that both the limestone and dolomite will generate alkalinity.

Small-scale batch tests

Although batch tests do not reproduce the solid-to-liquid ratios likely to be found within the PRB (Carey et al., 2002), they can be used as a screening tool to provide initial confirmation that the water is amenable to treatment. Batch reactor vessel tests following a methodology similar to the sequential batch leachate test (modelled after the ASTM test method D4793) were therefore used to evaluate the effectiveness of each blend of material treating the seepage from the site (Elliot, 2003). A composite of 50 ℓ of shallow groundwater was obtained from auger holes at the site for use in the test.

Five duplicates were made for each test so that each cell could be kept closed and airtight from the initial addition of the seepage solution until analysis. Samples were collected from the batch reactor vessels after: 1, 7, 14, 29 and 43 d.

Amenability to treatment

A summary of the results for the limestone, dolomite and compost mixes of the batch tests at 1 d and after 48 d is compared to the seepage analyses and permit concentrations in Table 5.

The results from the batch tests indicate that:

TABLE 4
Cell contents for batch tests (Elliot, 2003)

Cell ID	Dolomite (g) <19 mm	Dolomite (g) < 37 mm	Limestone <20 mm	Compost (g)	Weight % compost
D1-25	125	-	-	375	75
D1-50	250	-	-	250	50
D1-75	375	-	-	125	25
D-100	1 000	1 000	-	0	0
D2-25	-	125	-	375	75
D2-50	-	250	-	250	50
D2-75	-	375	-	125	25
L-25	-	-	375	125	75
L-75	-	-	125	375	25
L-100	-	-	2 000	0	0

- Both the dolomite and limestone aggregates successfully increased the pH from 3.3 to between 6 and 7 within 24 h.
- In separate batch tests, the compost blend with both dolomite and limestone resulted in a decrease in Eh (anoxic conditions), and concentrations of nitrate, fluoride, phosphate, aluminium, manganese, copper, nickel and lead decreased over the period of 43 d.
- Concentrations of sulphate, chloride, sodium, COD and ammonia increased over the 43 d period.

Field tests

Auger holes were drilled to refusal along and across the valley channel from the granite outcrop 300 m up-gradient of the confluence with the downstream tributary to about 1 km from the old factory site. The position of the auger holes and study area is presented in Fig. 2. The study indicated that subsurface seepage is along a broad front and is controlled by the depth to bedrock. The depth to bedrock varies from 6.5 m up-slope becoming shallower down-slope at 1.2 m and outcrops before the confluence. The study showed that the groundwater appears to be naturally funnelled in the valley area, constraining lateral flow. A cross-section of the area is presented in Fig. 3. Auger holes were used to study subsurface seepage, soil profiles, bedrock topography and permeability.

Groundwater flow conditions

Soil types were found to vary from fine silty sands to localised areas of clay residual from granite. Ferricrete concretions and mottling characteristic of ferruginisation and a shallow water table were evident in some of the soil profiles, especially adjacent to the channel. Water is intercepted in a narrow zone adjacent to the stream at a depth of about 2 m (upslope) to 1 m (down-slope).

Permeability tests

Percolation permeability tests were completed in two of the holes, one up gradient within the clayey horizons and one down gradient in the silty sand. The permeability varied from 0.93 m/d (1.07 x 10^{-5} m/s) in the clayey sand formation to 24 m/d or 2.8 x 10^{-4} m/s in the silty sand, correlating well with literature values for silty sand (Freeze and Cherry, 1979).

Feasibility assessment

The feasibility of using a PRB down-gradient of the fertiliser manufacturing site is summarised in Table 6.

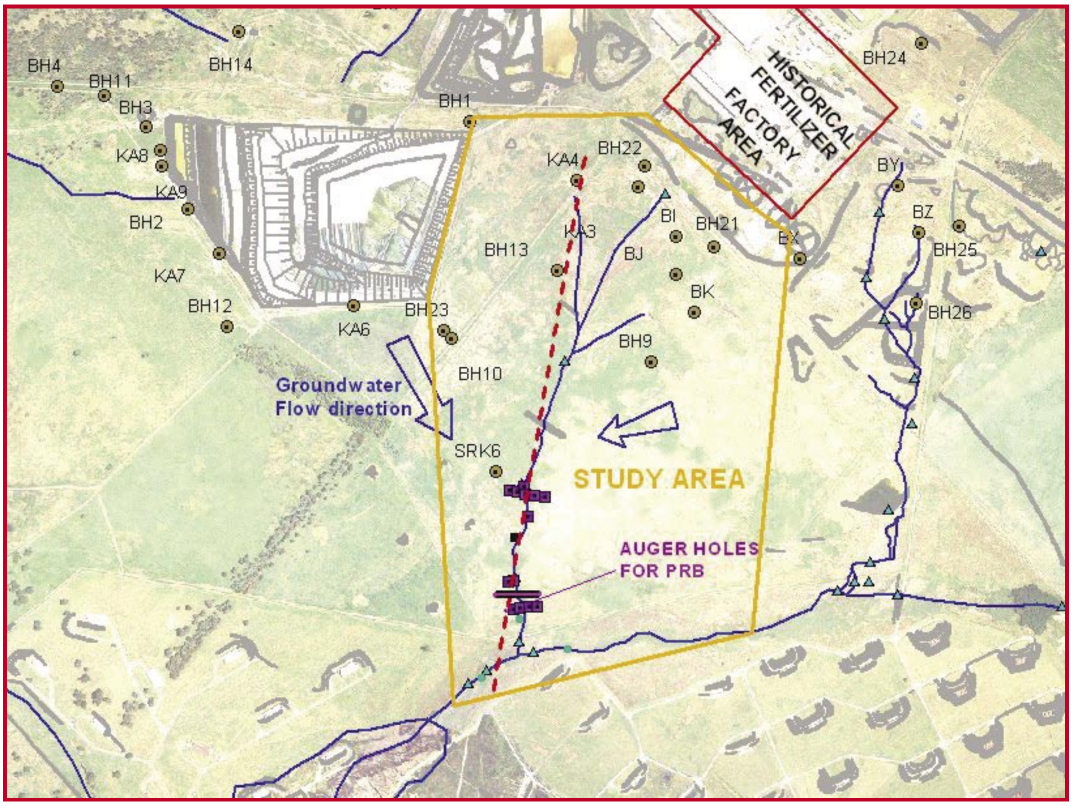

Figure 2
Position of auger holes in the study area (not to scale)

TABLE 5
Summary of analytical results

Parameter	Seepage water	RWQO	Concentration at Day 1		Concentration at Day 48	
pH	3.34	5.5-8*	Dolomite	5.7	Dolomite	6.1
			Limestone	6.4	Limestone	6.8
			Compost mixes	5.9-7.3	Compost mixes	7.1-7.4
Eh (mV)	425	0 mV for NO_3; -100 mV for SO_4 (1)	Dolomite	190	Dolomite	115
			Limestone	182	Limestone	150
			Compost mixes	132-227	Compost mixes	9 to -326
COD (mgO/ℓ)	44	50*	Dolomite	16	Dolomite	40
			Limestone	50	Limestone	37
			Compost mixes	570-970	Compost mixes	1 270 -3 460
Conductivity (mS/m)	729	120*	Dolomite	732	Dolomite	765
			Limestone	768	Limestone	766
			Compost mixes	1 077-1 341	Compost mixes	1 376-1 604
Nitrate as N (mg/ℓ)	520	10*	Dolomite	499	Dolomite	569
			Limestone	522	Limestone	589
			Compost mixes	449-512	Compost mixes	0.1-1.8
Ammonia as N (mg/ℓ)	<0,2	1*	Dolomite	0.16	Dolomite	0.16
			Limestone	0.16	Limestone	0.16
			Compost mixes	194 -341	Compost mixes	217-411
Total N (mg/ℓ)	520		Dolomite	500	Dolomite	569
			Limestone	522	Limestone	589
			Compost mixes	695-852	Compost mixes	219-411
Total P (mg/ℓ)	19	1*	Dolomite		Dolomite	
			Limestone		Limestone	
			Compost mixes		Compost mixes	
Sulphate (mg/ℓ)	1720	200* (600)	Dolomite	1 610	Dolomite	1 810
			Limestone	1 660	Limestone	1 780
			Compost mixes	1 920-3 340	Compost mixes	3 100-5 440
Iron (mg/ℓ)	0,32	2(2)	Dolomite	0.67	Dolomite	0.41
			Limestone	0.3	Limestone	0.4
			Compost mixes	0.32-0.77	Compost mixes	1-3.2
Aluminium (mg/ℓ)	120	2 (2)	Dolomite	32	Dolomite	7.1
			Limestone	0.3	Limestone	0.44
			Compost mixes	1.2-14	Compost mixes	0.46-2
Fluoride (mg/ℓ)	82	2*	Dolomite	40	Dolomite	24
			Limestone	16	Limestone	9.4
			Compost mixes	13-29	Compost mixes	6.5-13
Calcium (mg/ℓ)	640	80(3)	Dolomite	710	Dolomite	790
			Limestone	940	Limestone	1 100
			Compost mixes	430-700	Compost mixes	490-750
Magnesium (mg/ℓ)	310	100(3)	Dolomite	360	Dolomite	410
			Limestone	330	Limestone	300
			Compost mixes	280-370	Compost mixes	310-390
Manganese (mg/ℓ)	64	0.4* (5)	Dolomite	61	Dolomite	61
			Limestone	55	Limestone	35
			Compost mixes	13-38	Compost mixes	4.1-11
Iron (mg/ℓ)	0.32	2(2)	Dolomite	0.67	Dolomite	0.41
			Limestone	0.3	Limestone	0.4
			Compost mixes	0.32-0.77	Compost mixes	1-3.2
Sodium (mg/ℓ)	140	180* (600)	Dolomite	140	Dolomite	150
			Limestone	150	Limestone	150
			Compost mixes	320-450	Compost mixes	500-590
Chloride (mg/ℓ)	470	100 (600)(3)	Dolomite	470	Dolomite	500
			Limestone	490	Limestone	450
			Compost mixes	490-1 830	Compost mixes	1 720-2 350
Potassium (mg/ℓ)	120	50 (400)(3)	Dolomite	100	Dolomite	130
			Limestone	130	Limestone	110
			Compost mixes	1 060-1 800	Compost mixes	2 000-2 500

Notes: * = Receiving water quality guidelines specified by DWAF for the site effluent permit
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TABLE 6
Feasibility of utilising a PRB

Criteria	Site	Limitations/ further studies required
Source-pathway-receptor	The source is assumed to be mainly historical. The main impact is to the aquatic environment but there is a marginal impact on the deeper aquifer.	Impact to the deeper aquifer was assumed to be marginal and was therefore not fully quantified. Monitoring is required to confirm that there is, in fact, a marginal effect. The impact to the aquatic environment and the east-west stream must still be quantified in terms of the dilution effects and assimilative capacity of this stream. A water balance across this area is also recommended to prioritise the need for protection of the water source.
Groundwater flow	Groundwater flow is estimated from literature and initial permeability tests	Seasonal variations to be confirmed and additional geochemical work required. Long-term column tests are required to determine contaminant flux and retention times
Technical feasibility of PRB • Amenability of treatment	Acidity and nitrate removal was successful but there was only limited success achieved for sulphate	Long-term column tests are required to determine the long term performance of the barrier
• By-products from treatment process do not present an unacceptable risk to the environment	Ammonia, sodium, chloride and potassium is leached from compost and would constitute a risk to the environment	The potential for leaching of organic acids was not addressed
• The PRB will capture the contaminant plume	The site investigations indicate that the shallow groundwater contamination can be intercepted in this area	The deeper groundwater contamination can not be treated by the PRB. The impact on the deeper aquifer was assumed to be marginal
• The residence time, as a function of the rate of treatment relative to travel time, within the reactive media is adequate	Batch tests are not sufficient for residence times to be calculated. However, sulphate was not removed after 43d in the batch tests and is therefore unlikely to have sufficient time to be treated in PRB where groundwater flow is 5.8 m^3/d	Column tests required
• Successful achievement of remedial objectives	Treatment of sulphate not successful. Nitrate was removed by reaction with mushroom compost, but ammonium concentrations increased.	Alternative media should be investigated
• Long-term effectiveness of barrier	Long-term effectiveness cannot be confirmed before column tests are done but results of batch tests indicate that concentrations of aluminium and manganese may result in armouring of the dolomite aggregate. The precipitation of aluminium hydroxide in the barrier may also result in clogging. The clay and silt of the residual granite may also result in clogging of the barrier over time.	Column testing required to assess long-term effectiveness of the barrier.
How PRB will affect groundwater flow and geochemical regime	To be addressed in the detailed design stage	
Prioritisation of action to protect water source	Still to be confirmed by a mass and salt water balance of load contributions for each stream	
Characterisation of the site	A detailed geotechnical investigation should be done to confirm initial study	
Is contamination plume and geometry amenable to PRB application	Yes	
Cost-effectiveness	The cost of dolomite and mushroom compost is much cheaper than that of limestone aggregate primarily because of the shorter transport distances from suppliers.	
Access	Open land area. Access is good.	
Regulatory	Design and construction of a PRB would require application to be made for a water use licence.	

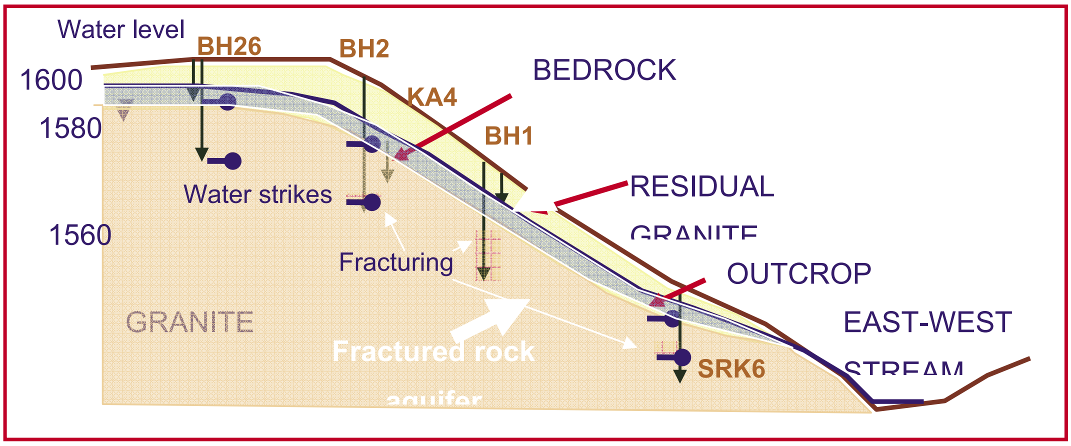

Figure 3
Schematic cross-section through the study area

Conclusions

Feasibility of dolomite as an alternative to limestone

Based on the initial tests, dolomite, while not as pure as limestone, is a feasible alternative for use as neutralising gravel. Both dolomite and limestone added alkalinity to the water. The increase in pH, for the pure limestone, dolomite, and limestone/dolomite mixed with compost cells, in turn reduced concentrations of fluoride (by precipitation as CaF_2) and aluminium in the water.

The main limitation, for both dolomite and limestone, would be the long-term effect of armouring and permeability. Armouring with aluminium hydroxide or manganese oxides would decrease the reactive surface area and may cause clogging of the pore spaces, restricting the flow of groundwater through the PRB. The long-term efficiency of the dolomite as a neutralising agent can, however, only be confirmed by long-term column testing.

Feasibility of mushroom compost as a carbon source

Limited success was achieved in the compost mix cells for the treatment of nitrate, sulphate, fluoride and metals (manganese and aluminium). This was demonstrated by improved water quality with respect to fluoride, aluminium, manganese and nitrate. In contrast, water leaching from the compost composite was of poorer quality water with respect to sulphate, chloride, sodium, potassium, COD and ammonia. This resulted in a net increase in salt concentrations represented by the increased electrical conductivity.

Suitability of site conditions for a PRB

The site conditions, as defined by the shallow depth of granitic bedrock, low permeability (10^{-5} m/s) and natural funnelling of seepage indicate that it would be practical to intercept groundwater within the proposed area.

Overall conclusion

The overall results indicate that groundwater interception and the installation of a PRB at the site is practicable but that mushroom compost is not suitable as a carbon source. Alternative options should therefore be considered.

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