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Geo-environmental Approaches for the Remediation of Acid Sulphate Soil in Low-lying Floodplains

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ABSTRACT
Acidity generated from the oxidation of pyrite and other sulphidic compounds that exist at shallow depths in acid sulphate soils (ASS) presents a challenging environmental problem in coastal Australia. The generated acidic groundwater can adversely impact coastal ecosystems, aquaculture and agriculture. Groundwater manipulation using weirs and modified floodgates in creeks and flood mitigation drains in ASS-affected farmland, which has been practiced for over a decade for preventing pyrite oxidation, is not effective in low-lying floodplains due to the high risk of flooding. In this paper, the authors present an overview of their experience in coastal Australia, a critical evaluation of currently practiced geo-environmental remediation methods as well as a demonstration of a pilot permeable reactive barrier (PRB) to control acidic groundwater pollution. The selection of recycled concrete, a commonly available alkaline waste material, and the systematic investigation of its longevity are highlighted through a series of batch and column experiments. In addition, the improvement of the groundwater quality by a pilot PRB using recycled concrete in ASS terrain within the Shoalhaven region of NSW, Australia will be elucidated based on field data collected over the last 3.5 years.

Keywords: Acid sulphate soil (ASS), Longevity, Permeable reactive barrier (PRB), Recycled Concrete

INTRODUCTION

Acidic groundwater, generated from acid sulphate soils (ASS) is a major environmental problem in Australia. ASS occupies more than three million hectares of the coastal Australian landscape (White et al. 1997), which is estimated to contain 700 million tonnes of potentially environmentally hazardous sulphidic materials (commonly pyrite). If left undisturbed and submerged under the water table, this pyritic material is chemically inert. However, if exposed to complete atmospheric oxidation, the total amount of pyritic material in the Australian landscape would be equivalent to about 2.2 billion tonnes of sulphuric acid (Fitzpatrick 2003), which

would lead to a severe acidification of the coastal floodplains and estuaries. In fact, the use of large-scale artificial drains and one-way tidal restricting floodgates for reclaiming land has already enhanced pyrite oxidation and resulted in large volumes of *in-situ* acid production and storage in soils of low-lying floodplains of eastern coastal Australia (Indraratna et al. 1995; White et al. 1997). This acidic water can also mobilise potentially harmful metals (e.g. Al and Fe) from the soils (Dent 1992; White et al. 1997). In addition, natural changes in the hydrological system (e.g. drought) have exacerbated acid production by promoting pyrite oxidation in the shallow zone of ASS. Acidification of coastal waterways, massive kills of fish and oysters, sterilisation of land and groundwater for agriculture, and corrosion of concrete and steel infrastructures are major socio-environmental problems of ASS in coastal Australia. Although the causes of acidic water have been extensively investigated in Australia for over five decades (White et al. 1997), very limited studies have been conducted for their remediation.

In this paper, the authors will firstly present a critical evaluation of different geo-environmental approaches practiced in Australia for the prevention of pyrite oxidation in ASS floodplains. Secondly, this paper will demonstrate the application of Australia's first pilot subsurface PRB for the remediation of ASS groundwater in which: a) details of the screening process of the reactive media, b) evaluation of the efficiency of the reactive media, and c) evaluation of the performance of the PRB over the last 3.5 years in treating acidic groundwater generated from ASS will be presented.

PREVENTIVE APPROACH IN MANAGEMENT OF ASS

a) Water table manipulation using simple v-notch weirs

Groundwater manipulation techniques have been successfully practiced in acid rock drainage (Pedersen 1983) for decreasing the oxidation of tailings by complete inundation of acid producing materials. Similarly, Indraratna et al. (1995) suggested that simple v-notch weirs would decrease acid production in ASS terrain of coastal Australia by maintaining the water table above the pyritic zone. Blunden and Indraratna (2000) confirmed that such weirs could elevate the groundwater above the pyritic layer. Following extensive field monitoring and further hydrological modelling, Blunden and Indraratna (2001) developed a pyrite oxidation analytical model and demonstrated that the weirs could significantly decrease pyrite oxidation by allowing the phreatic surface to rise. Consequently, water manipulation by weirs has been practiced in coastal Australia as a low-cost management strategy for preventing further pyrite oxidation. These weirs work best in areas with good drainage and a water table not too close to the ground surface. However, the benefits occur in very localised regions near the weirs and conversely in low-lying areas with poor drainage, weirs may increase the risk of flooding and prevent tidal buffering from improving water quality.

b) Tidal buffering by modified two-way floodgates

The negative influence of tidal restricting structures including v-notch weirs regarding ecological effects on aquatic life (Pollard and Hannan 1994), wetland

development and productivity (Dick and Osunkoya 2000) and ASS (Indraratna et al. 2005) are now well recognised in Australia. In addition, the v-notch weirs prevent tidal ingress. Glamore and Indraratna (2004) designed two-way fully-automated modified floodgates (smart gates) as an alternative solution. Surface water quality measured continuously for three years (one year pre-modification and two years post-modification), upstream of a modified floodgate (Table 1) showed an increase in drain water pH above 6. Furthermore, Al and Fe also decreased significantly.

Table 1. Average drain water quality at different upstream locations of the floodgate pre- and post-floodgate modifications (modified after Golab and Indraratna 2009).

Parameters	Unit	Median	1m	15m	30m	60m
pH_{pre}		4.42	4.41	4.44	4.35	4.72
pH_{post}		6.22	6.29	6.21	6.20	6.24
Al^{3+}_{pre}	mg/L	14.61	16.64	15.43	13.79	4.15
Al^{3+}_{post}		1.82	2.80	1.96	1.69	1.21
% change		+85	+83	+87	+88	+71
Fe^{2+}_{pre}	mg/L	28.29	31.89	29.41	27.17	14.47
Fe^{2+}_{post}		3.79	5.75	3.85	3.74	3.46
% change		+84	+82	+87	+86	+77

Two-way floodgates allow entry of brackish creek water during high tide, improving the drain water quality by the buffering action of carbonate/bicarbonate (Glamore and Indraratna 2004; Indraratna et al. 2005). This technique also prevents the release of large slugs of acidic water into adjacent waterways at low tide. However, the amount of alkalinity observed during tidal ingress was in the range of 10-90 mg/L $CaCO_3$ and this amount was not high enough to buffer large volumes of acidic drain water, especially following large rainfall events, due to dilution of the buffering agents and an increase in total acidity flushed from the soil. Similar to v-notch weirs, two-way floodgates also elevate the risk of flooding for low-lying areas with poor drainage, are unable to neutralise the acidity already stored in the soil, and cannot prevent pyrite oxidation far from the drain. Nevertheless, due to its success in improving drain water quality before discharge into adjacent waterways (Indraratna et al. 2005), recently numerous local councils in Australia have installed smart gates elsewhere in coastal Australia.

REMEDIATION OF ACIDIC GROUNDWATER

Large quantities of acid generated over long periods are stored in ASS floodplains following the installation of flood mitigation drains. In addition, ongoing acid production continues due to pyrite oxidation in the soil either by hydrological changes or bacterial oxidation. This produces acid even under anaerobic and submerged conditions similar to acid drainage in some tailings dams under reducing conditions. Despite more than three decades of ASS research in Australia, a

comprehensive management strategy has not been developed to continuously neutralise acidic discharges because the water table manipulation approach did not remediate the previously stored acidity within the soil.

a) Active remediation approach in ASS

Soil liming to neutralise soil and groundwater acidity has been extensively practiced in Australia and can be effective but expensive for large areas. It also produces metal-rich sludge in the soil (Benner et al. 1999) from which metals may subsequently leach when mixed with freshly produced acid in the groundwater, thus being ineffective in the long-term (Pearson and McDonnell 1975; Webb and Sasowsky 1994). Pump-and-treat methods may be an alternative approach but with high operating costs. Injection of an alkaline slurry such as lime-fly ash near drains could control pyrite oxidation and improve groundwater quality close to the injected area (Golab and Indraratna 2009; Indraratna et al. 2006). However, its longevity is uncertain due to armouring and exhaustion of the reactive materials.

b) Passive Remediation Approach: Permeable Reactive Barrier

In low-lying floodplains, the application of PRBs for remediating acidic groundwater appears to be a cost-effective alternative to the conventional techniques practiced to date. Laboratory and field studies have established the efficiency of this remediation method for different organic and inorganic contaminants such as radionuclides, heavy metals and acid mine drainage (Blowes et al. 2000; Gu et al. 2002; Jurjovec et al. 2002). The pilot PRB appears to be the very first trial in Australia under reducing conditions for treating acidic water from ASS. Our currently demonstrated remediation strategy i.e. a PRB in the flow-path of acidic groundwater to neutralise the acidity and remove the toxic cations from solution has been in use for 3.5 years in ASS in southeast NSW, Australia. The PRB is briefly described in four stages covering: (i) site selection; (ii) selection of the reactive material; (iii) efficiency of the selected reactive material; and (iv) performance of the pilot PRB.

(i) Site Selection

Following site characterisation including: (a) physical and chemical properties analysis of the soil in ASS terrain, (b) monitoring of groundwater flow conditions and (c) groundwater quality analysis, a suitable site near Broughton Creek, southeast NSW, Australia was selected. The site has the following favourable properties for the installation of a PRB: (i) acidic groundwater (pH down to 3) with high Al (up to 40 mg/L) and Fe (up to 530 mg/L) levels; (ii) the site is low-lying (0-1 m AHD) thus weirs or floodgate modification are not suitable; (iii) a shallow ASS layer 0.3-1.5 m below the ground surface; (iv) a nearby drain for the treated groundwater to flow into; (v) 'all-weather' site access; (vi) a zone of preferential groundwater flow for passive interception.

(ii) Selection of reactive media

Selection of the suitable alkaline reactive material is of paramount importance for PRB design in ASS terrain because it determines the reactivity and metal removal capacity. Screening tests on 25 different alkaline materials, with an emphasis on

waste materials, including fresh and recycled concrete, limestone, calcite-bearing zeolitic, breccia and blast furnace slag (Figure 1) beginning with a series of batch tests (Golab et al. 2006) then short-term column tests (Golab et al. 2009; Golab and Indraratna 2009) were conducted with drain water collected from the field site. The six concretes, lime and air-cooled blast furnace slag, all achieved a pH consistent with the dissolution of portlandite/lime (pH 11 to 12). The limestone and zeolitic breccia achieved a pH consistent with the dissolution of calcite (pH ~7.4). The results show

that recycled concrete performed well by neutralising large volumes of acidity and removing Al and Fe from solution without leaching harmful ions into the groundwater. Therefore, it was selected for further investigation as the reactive medium of the PRB.

Figure 1. Change in pH with respect to time for selected reactive materials among the 25 alkaline materials in batch tests with acidic drain (after Golab et al. (2006))
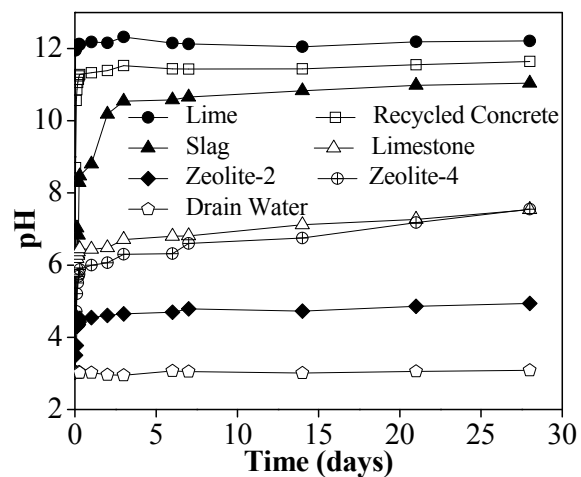
iii) Evaluation of the recycled concrete

The batch and short-term column tests could not confirm the longevity of the material since the material reactivity would decrease over time with the continuous flow of acidic groundwater. Consequently, following studies by Golab et al. (2006), and Golab et al. (2009), recycled concrete was selected for further investigation of its long-term efficiency to treat the acidic groundwater under the similar continuous flow conditions to the field situation. The recycled concrete used for this experiment was a waste material discarded after the demolition of old concrete structures. Calcium-bearing minerals present in the recycled concrete were identified as portlandite, anorthite and calcite by x-ray diffraction (XRD) and are considered as the most abundant alkalinity-generating compounds. The input solution for the column experiment was synthetic acidic water, with comparable characteristics to the average groundwater from the PRB field site (Regmi et al. 2009). A long-term column test was conducted in a 650 mm long and 50 mm inner diameter acrylic column at room temperature (23-25°C) with accelerated flow rate compared to the groundwater velocity at the field site. The effectiveness of the reactive material was assessed with respect to pH, bicarbonate alkalinity, effluent Al and Fe concentrations versus the number of pore volumes (PVs – defined here as the void volume of the column) of the acid passed through the column during the experiment.

A step-wise decreasing pH profile was observed in the column as the number of PVs passed through the column increased (Figure 2). The initial pH of the effluent was high (pH 11.2) due to the dissolution of portlandite, which was found consistent with the batch result of recycled concrete. The effluent pH decreased rapidly from 11.2 to 7.9 within 38 PVs due to the fast depletion of hydroxyl and carbonate alkalinity generated by very small amounts of portlandite present in the concrete.

Subsequently, three long plateaux (pH ~7.9 to 6.5 from 40 to 235 PVs, pH ~4 from 300 to 500 PVs and pH ~2.7 after 500 PVs; Figure 2) were observed where the pH dropped abruptly from one plateau to another over just a few PVs. These three plateaux of pH were attributed to three distinct pH-buffering reactions: the dissolution of carbonate/bicarbonate alkalinity from concrete at near neutral pH due to the dissolution of abundant calcium aluminium silicates (e.g. anorthite) and calcite present in the concrete, re-dissolution of aluminium hydroxide precipitates at pH ~4, and re-dissolution of ferric oxy/hydroxides minerals at pH < 3.

In addition, alkaline to near-neutral pH maintained by carbonate/bicarbonate buffering in the system favoured the precipitation of Al and Fe in different oxy/hydroxides forms. Thus, Al and Fe present in the influent acidic water were completely removed by mineral precipitation inside the column until maintenance of the first pH plateau at near neutral. Al and Fe in the effluent increased after 250 PV and 480 PV, respectively, corresponding to a fall in pH from near-neutral (pH 6.5) to acidic ($\leq$ pH 4.0), when the alkalinity was depleted.

The drop in pH from near-neutral to pH 4.0 occurred earlier (at 300 PV) than expected from the neutralisation capacity (520 PV), based on the Acid Neutralisation Capacity (ANC). This earlier drop in pH, along with observed white and orange precipitates of Al and Fe coating the reactive media from the bottom towards the top of the column shows that the efficiency of the reactive material was significantly decreased by armouring. However, negligible changes in pressure head showed that the hydraulic conductivity decreased by just ten fold confirming that chemical clogging would not be a major problem when larger particle sizes were used in the PRB compared to the column experiment. Nevertheless, the results from column experiment confirmed that recycled concrete could effectively neutralise acidic water and remove Fe and Al from the acidic groundwater even after the interaction of large volumes of acidic water in the system (Regmi et al. 2010).
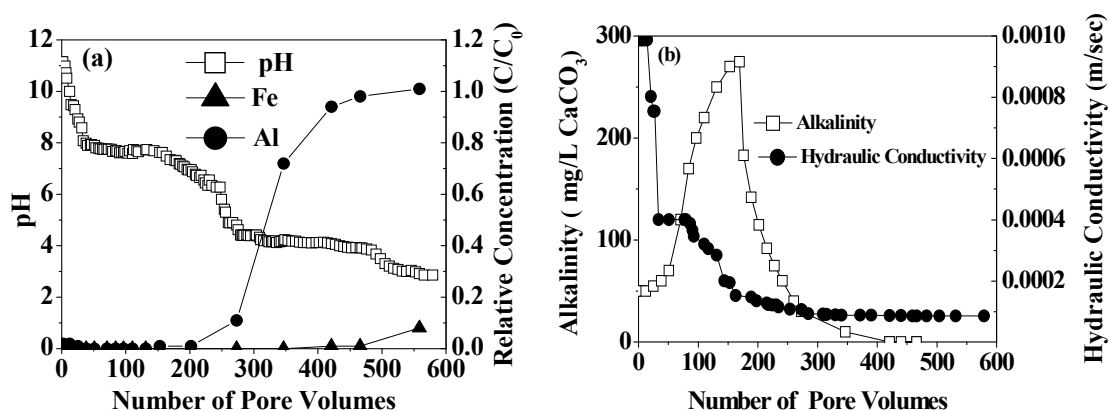

Figure 2. Results from column test (a) pH , Fe and Al vs. number of pore volumes of acid passed (b) alkalinity and hydraulic conductivity vs. number of pore volumes of acid passed.

iv) Performance of the pilot PRB

Based on the site characterisation and observed long-term performance of the recycled concrete in remediating acidic groundwater in the column test, a pilot PRB

(17.7 m × 1.2 m × 3.0 m) was installed, close to the flood mitigation drain (Figure 3A), intersecting the groundwater flow path (Indraratna et al. 2010). The PRB was filled with crushed 40 mm recycled concrete gravel and the trench was lined with geotextile fabric to protect the reactive media from physical clogging by soil and other fine particles. A monitoring network of 30 observation wells, 12 piezometers and 2 data loggers, as shown in

Figure 3A was installed up-gradient, inside, and down-gradient of the PRB to monitor its performance. The performance has been monitored continuously for nearly 3.5 years.

Figure 3B-D compares the groundwater quality parameters, up-gradient, inside and down-gradient of the PRB along transect a-a (Figure 3A). Throughout the monitoring period, the groundwater up-gradient of the PRB has been acidic (pH 3.2-4.5; average pH 3.7). Under acidic conditions, the groundwater contains high amounts of Al and Fe (Figure 3C and D). The groundwater pH inside the PRB has been consistently alkaline to neutral (pH 10.2 to 7.3). Similar to the column results, the PRB initially reached a high pH (~10.2), dropped down to a stable pH of ~8.0 after a few months followed by continual stability in the range of 8.0-7.0 to the latest stage. This pH plateau is consistent with the first plateau observed in the column tests. Furthermore, Fe and Al concentrations inside the PRB were negligible (0-2 mg/L) compared to high concentrations up-gradient (Figure 3C and D) due to mineral precipitation inside the barrier at high pH. Down-gradient of the PRB, pH is observed to rise up to ~6.8 in the monitoring wells at distances of 5 m and 10 m from the PRB. This is lower than the pH inside the barrier, but significantly higher than the pH up-gradient of the barrier. Fe and Al concentrations down-gradient were higher than those inside the PRB, but lower than those up-gradient of the PRB. The lower pH and higher metal concentrations down-gradient compared to those within the PRB are due to the (i) production and mixing of acid generated from pyrite oxidation in the soil in the down-gradient areas and the inability of the PRB to control acid generation; (ii) possibility of acid mixing (generated up-gradient of the PRB) from the sides of the barrier due to its small size; (iii) dilution of the effluent from the PRB. In addition, low pH at some observation wells down-gradient during some dry periods (e.g. Nov 2006 and Nov 2008) is possibly due to the flushing of large amounts of acidity stored within the soil by small rainfall events.

Despite the variable environmental conditions in the field, the current plateau of pH above 7.0, high generation of alkalinity inside the PRB (210-400 mg/L $CaCO_3$), and the removal of Fe and Al confirm that the pilot PRB can effectively neutralise acidic groundwater. These results are consistent with our previous column tests. However, the PRB's efficiency will ultimately decrease due to armouring by Fe and Al precipitates in addition to the exhaustion of the reactive material as demonstrated by the column test results. Exact calculation of the longevity of the PRB is difficult due to many factors such as groundwater flow rate, acidity generation rate and amount of the alkaline minerals present in the concrete and therefore, further performance monitoring is needed. The performance can be significantly improved if multiple small PRBs are installed in series in the area or if intermittent injection of alkaline fluid (preferably waste effluent for cost-effective management) into the PRB is adopted.
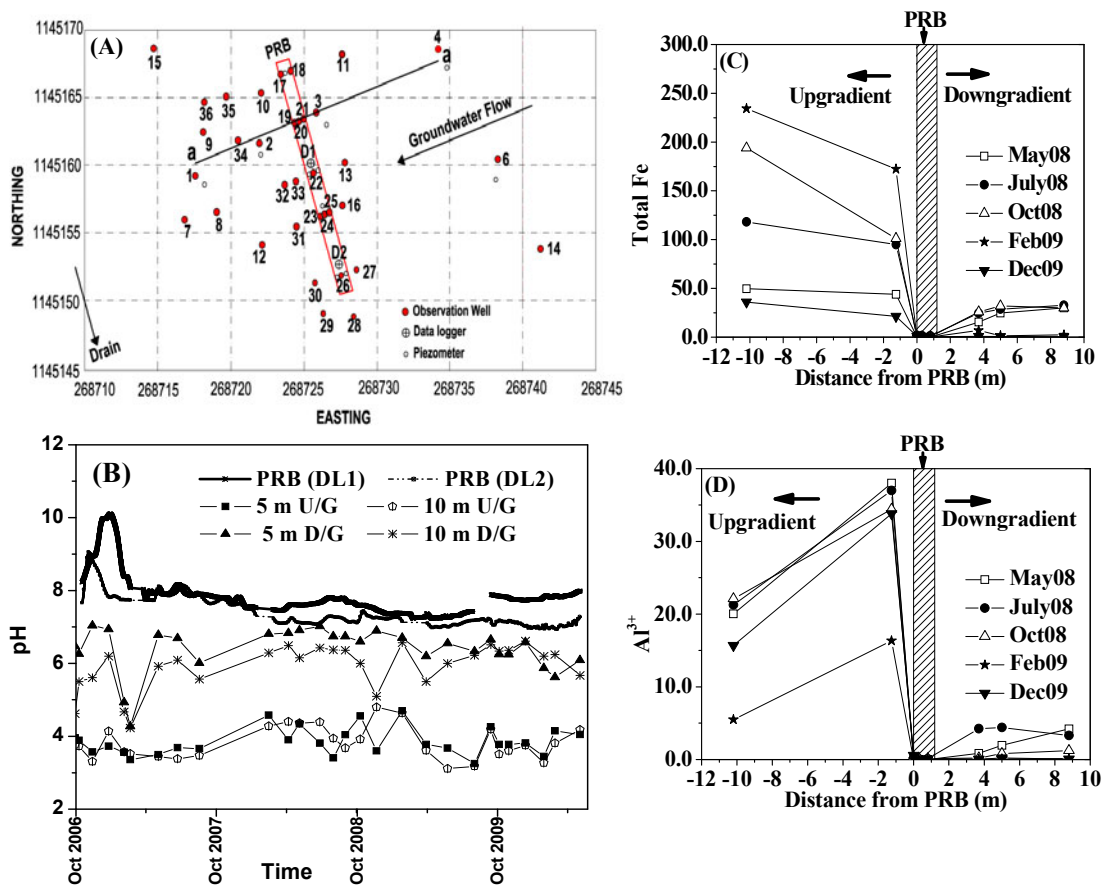
Figure 3. (A) Layout of the PRB and observation wells and transect a-a; Spatial and temporal performance evaluation of the PRB with respect to: (B) pH (C) Total Fe and (D) Al. U/G refers to up-gradient and D/G to down-gradient

CONCLUSIONS

This paper presents a critical evaluation of the currently practiced methods for the remediation of contaminated water from ASS in Australia and demonstrates the evaluation of the first pilot PRB in ASS terrain as an alternative. The preventive methods using modified floodgates and weirs currently practiced in Australia are not effective in low-lying floodplains as these techniques can neither remediate the acidity already present in the soil nor significantly prevent pyrite oxidation in areas far from nearby drains. Rather, these techniques increase the risk of flooding in coastal Australia. In addition, other active remediation methods are costly and not suitable for remediating large areas of ASS.

The findings of the first PRB using recycled concrete for spot treatment of acidic groundwater in ASS terrain show that PRBs using waste alkaline materials are a valid alternative to conventional methods. Column test results for maintaining near neutral pH and completely removing large amounts of dissolved Fe and Al for long

durations under accelerated flow confirm the suitability of recycled concrete. The buffering capacity is controlled by the dissolution of carbonate/bicarbonate alkalinity and the precipitation of Al and Fe as different oxy/hydroxide minerals. It has been observed that armouring effect by precipitated minerals reduces reduce the efficiency of the reactive material, further decreasing the PRB longevity. However, the threat by chemical clogging is minimised by the use of larger size reactive media.

The PRB with recycled concrete has effectively maintained neutral pH and removed almost all the acidic cations (Fe and Al) present in the groundwater in a manner similar to column experiment over the three and half year period following installation. However, it only marginally improved the chemistry of the groundwater down-gradient of the PRB. In addition, the longevity of the PRB is uncertain due to issues of pacification of the reactive media and long-term armouring, which might be a limiting factor. The longevity of the PRB and down-gradient water quality can be improved significantly if an alkaline effluent (preferably alkaline waste effluent for cost-effective management) is intermittently injected into the PRB, which is recommended for further work.

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