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Installation of a lime injection barrier for the remediation of acid sulphate soil problems.

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Abstract

Oxidation of naturally occurring pyrite (FeS_2) in certain low-lying clayey soils generates sulphuric acid, hence the term acid sulphate soils. A horizontal alkaline barrier was installed by radial grouting, for the purpose of remediating leachate from acid sulphate soils and preventing further oxidation. The current research relates to a large-scale field trial of this technique and the effect on the groundwater composition. In coastal Australia, a pyritic layer commonly exists in the soil at shallow depth that is at risk of oxidation, hence the main objective was to inject the barrier above the pyritic layer to (a) stop infiltration of oxygen to the pyritic layer and (b) neutralise any acidity stored in the soil. Two fine-grained alkaline materials, lime and fly ash, were assessed in this study. Lime was selected for its neutralising capacity, while the fly ash was selected to accompany the lime to enhance the pozzolanic reactions. The optimum mix ratio of lime, fly ash and water to form an ideal slurry and the optimum depth and pressure of injection were experimentally determined. For the large-scale field trial, the slurry was injected into a systematic grid of 22 holes to form the reactive barrier. The groundwater composition was monitored in a network of observation holes across the study site to determine the effectiveness of the barrier. The average groundwater pH was 3.25 prior to installation of the barrier, and it rose to 4.6 after the barrier was installed. The influence of the barrier on the groundwater pH was greater in observation holes close to the barrier than those further away. The concentrations of aluminium and iron decreased in the groundwater after the installation of the alkaline barrier. The ratio of Cl/SO_4 in the groundwater increased after the barrier was installed which confirmed that the barrier had successfully controlled the subsequent pyrite oxidation in the soil.

1. Introduction

Acidic groundwater is a common problem in coastal Australia. Sulphuric acid is produced by the oxidation of sulphide minerals (e.g. pyrite) contained in soils on coastal floodplains, commonly known as acid sulphate soils (ASS). In a study site near the low-lying town of Berry (34°S, 150°E), the effectiveness of using a sub-surface, horizontal reactive barrier to decrease oxidation of a pyritic soil layer and to improve the quality of groundwater and surface water was investigated. In this area, especially during drought conditions, the groundwater and surface water are usually acidic and contain high concentrations of dissolved aluminium, total iron and sulphate, as a result of lowered water tables exacerbated by deep flood mitigation drains (Glamore and Indraratna, 2004). Although initially recognised in Australia in the 1970's (Walker, 1972), serious research on ASS was not conducted until major fish kills occurred in coastal rivers in the 1980's. Since that time, techniques that either prevent pyrite oxidation or remediate the resultant acidic groundwater have been studied, and Indraratna *et al.* (2005) provide a critical and comparative review of those techniques. The focus of the current research was to develop a remediation technique suitable for low-lying areas where most other techniques are not feasible. The use of a sub-surface, horizontal reactive barrier is a novel technique that has not been implemented anywhere else in the world for remediating ASS problems.

1.1. Barrier Description

This study aimed to test the hypothesis that a sub-surface alkaline barrier will effectively remediate acidic groundwater in low-lying areas. The barrier consisted of a slurry of experimentally determined reactive materials injected into the soil at a level above the pyrite layer. The slurry would create an impermeable barrier to both atmospheric oxygen and surface water infiltrating down from the surface, and to the groundwater travelling up towards the surface. In this way, the barrier would serve a dual purpose. Firstly gaseous oxygen from the unsaturated zone is prevented from coming into contact with the pyrite layer, thus preventing further formation of acid. The second function of the barrier is to act as an aquitard and prevent surface water containing entrained oxygen from percolating through the pyritic zone. Moreover, when acidic water comes into contact with the alkaline barrier, the acid would be neutralised by the lime and fly ash mix. In this manner, the barrier would effectively remediate acidic groundwater and reduce the amounts of aluminium and iron that reach the drain, because, the solubility of both cations is pH and Eh dependent.

In acidic groundwater with relatively high organic content (>5%), the bacteria *Thiobaccillus ferrooxidans* can directly oxidise pyrite under saturated conditions (Ritsema *et al.*, 2000). Column tests were conducted in the laboratory to determine if a subsurface lime barrier would affect the population of *T. ferrooxidans* and the soil pH. The lime barrier not only reduced the number of oxidising bacteria (from 18400 to 37 cells per gram of soil) but also neutralised the acidity of the soil (from 4.48 to 9.45), thereby reducing the efficiency of biotic oxidation of pyrite in the soil (Rudens, 2001). Pilot studies conducted in the laboratory and the field suggest that lateral injection of a lime-fly ash slurry to create an alkaline barrier above the pyrite layer can reduce bacterial activity, while simultaneously neutralising the acid already produced.

2. Barrier Properties

The slurry injection process was based on the fundamental principles of grouting. Grouting may be broadly defined as the injection of grouting fluids through boreholes to fill and seal voids, cracks, seams, fractures or other cavities in soils or rock strata (Bowen, 1981). The grouting fluid solidifies over time by physico-chemical processes and interaction with pores, thereby increasing the strength and decreasing the permeability of the grouted mass (Shroff and Shah, 1993). A number of authors have demonstrated the importance and many specific applications of grouting (Bowen, 1981; Nonveiller, 1989; Fell *et al.*, 1992; Karol, 2003). The factors that require consideration prior to the selection and application of grouting are: type of grout and its physical and chemical properties, location of boreholes, depth and inclination angles of penetration, and the in-situ soil characteristics.

2.1. Grout composition

Indraratna (1996) demonstrated the importance of hydrated lime injection into acidic soils (Figure 1). A marked increase in pH was noted when even small quantities of hydrated lime were added to acidic soil. If hydrated lime is added to wet acidic soil in the presence of a source of silica, for example fly ash, the following chemical interactions occur:

(i) acidity is neutralised;

(ii) base-exchange whereby the high pH of the lime alters the nature of the adsorbed water layers of the soil particles (Hausmann, 1990); and

(iii) pozzolanic/cementing action whereby the lime reacts with available silica and alumina to form ‘natural cements’ composed of calcium silicate hydrate

(Hausmann, 1990) and calcium aluminate hydrate gels (Rogers and Glendinning, 1997), as represented by Eqn. (1).

$$3Ca(OH)_2 + 2SiO_2 \rightarrow 3CaO.2SiO_2.3H_2O \quad (1)$$

$$CaO + CO_2 \rightarrow CaCO_3 \quad (2)$$

The silicate gels fill the soil pores and facilitate the formation of an intact alkaline barrier (Figure 2). Some of the injected CaO will react with CO_2 available in the soil pores and groundwater and form $CaCO_3$ (Eqn. 2) forming a relatively weak cementing layer, hence increasing the shear strength of the barrier.

Prior to selecting grouting materials or a grouting technique, it is essential to perform preliminary test injections. From these tests, a number of characteristics can be determined including the boring possibilities in the soil, the stratification and heterogeneities present in the soil, the in-situ permeabilities and the grouting pressures that yield the optimum ground improvement results (van Impe, 1989). The use of fly ash and lime to stabilise and strengthen silty soils and clays have been tested, and it has been found that both additives increase the shear strength of the tested soil (Indraratna *et al.*, 1991; Indraratna *et al.*, 1995). Studies have also been conducted into the use of fly ash in conjunction with lime for grouting, producing promising characteristics in the treated soil (Akbulut and Saglamer, 2003). Also, chemical injections of lime and/or fly ash slurries have previously been conducted to stabilise landfills prior to construction (Blacklock *et al.*, 1984).

2.2. Radial Flow of Grout

The method for void sealing used in this project is mainly permeation grouting, which is the replacement of water/air in voids between soil particles with a chemical slurry that is injected under appropriate pressure. The theoretical principles of radial grouting have been discussed by Indraratna (1983), Cambefort (1987), and Naudts and van Impe (2000). The optimum injection pressure is related to the in-situ stresses, initial grout properties (density and viscosity), soil properties (permeability, void ratio etc.) and the anticipated grouting rate. Excessive grout injection pressures can lead to heaving of the overburden soil as well as hydraulic fracturing in the lateral plane. The relevant equations relating the injection pressures to the above parameters have been well documented and some well-known closed form solutions are given by Hausmann (1990), Naudts and van Impe (2000) and Karol (2003). The viscosity and setting time of the grout must be controlled such that sufficient time is available

for the grout to permeate the required lateral extent within the soil stratum. This essentially dictates the determination of injection hole spacing.

3. Site Description and Characterisation

A study site was chosen that is low-lying and unsuitable for manipulation of the water table (Figure 3). This site is underlain by acid sulphate soils and is easily accessed by grouting equipment. It has a relatively shallow pyrite layer, i.e. within 1.2 m of the surface, and has an artificially lowered water table (due to deep flood mitigation drains), thus an acid environment has been created as a result of pyrite oxidation. In some other sites where deep flood mitigation drains are scarce, the submergence of the pyrite layer by the water table minimises pyrite oxidation.

A Digital Elevation Model (DEM) of the site was produced from high-resolution airborne laser surfacing and the site was surveyed at 1:100 scale. A grid of 31 observation wells and five piezometers were installed to monitor the level of the phreatic surface, hydraulic gradient, hydraulic conductivity, pore water pressures, and the acidity and chemical composition of the groundwater (Figure 4). The groundwater composition was monitored for more than 400 days, including at least nine months of monitoring prior to the barrier installation. The groundwater was routinely sampled and analysed in the field for pH, electrical conductivity (EC), temperature and water table elevation and in the laboratory for Al^{3+}, Fe^{2+}, Ca^{2+}, Mg^{2+}, by atomic absorption spectroscopy (AAS), SO_4^{2-} by inductively coupled plasma-optical emission spectrometry (ICP-OES) and Cl^- by APHA 4500-Cl^- (APHA/AWWA/WEF, 1998). These factors are critical for deciding the thickness of the barrier. The volume of water that comes into contact with the barrier per unit time and the acidity of that water determine the amount of neutralising material that is required.

Soil cores were also collected to determine the depth to the pyrite layer, because the injection depth is critical as it needs to be above the pyrite layer throughout the treated zone. Samples were acquired by pushing a 60 mm diameter steel tube into the soil to a depth of 1.6 m. The core was then sectioned and the soil was analysed for pH, EC, total actual acidity, reduced inorganic sulphur content, and dissolved Cl^- and SO_4^{2-}. The acidity of the soil profile indicates the depth to ASS (large peak in Figure 5), and the acidity decreases below that as the potential acid sulphate soil (PASS) layer is reached.

3.1. Field Trials

As discussed earlier, lime and fly ash were chosen as the grout components due to their neutralising and pozzolanic characteristics respectively. A number of properties and requirements of lime-fly ash slurries have an impact on the injection process including fluidity, strength, minimum shrinkage, minimum viscosity and the optimum injection pressure. A lower viscosity of the grout fluid allows easier penetration into the ground. Different lime-fly ash slurry ratios were tested to choose the most appropriate viscosity and the mix ratio. The final mix ratio of water: lime: fly ash was 2:2:1. A volume of 314 L of lime-fly ash/water slurry was calculated for injection into each hole, to provide minimum barrier thickness of 100 mm and a radius of influence of at least 1 m.

The injection pressure was experimentally determined as 60-80 kPa to prevent hydraulic fracturing or uplift of the soil, based on the existing mathematical models (Hausmann, 1990; Naudts and van Impe, 2000). The equipment used in the injection process consisted of a grout pump and a 150 L mixing tank with an air powered motor. The injection pipe consisted of one hollow pipe inside another with radial slits at the base for injection and packers to seal the hole during injection, using a simple design as described by Cambefort (1987).

Two preliminary test injections were carried out to a depth of 1.1 m in a small area close to the main site. The depth of injection was determined based on the depth to the top of the PASS layer. The depth of injection was calculated as 0.1 m above the PASS layer but initial attempts showed that the soil at this level was too soft to create an adequate seal between the injection pipe and the surrounding soil. For instance, during the initial injections, the rubber packers on the injection pipe did not create a tight seal and some of the slurry emerged from the top of the hole. Consequently, an additional set of packers had to be placed on the pipe to achieve a seal sufficient to prevent vertical flow of grout along the pipe-soil boundary. Also, the slits at the base of the pipe were blocked during the preliminary injection due to the small size of the slits and the relatively high viscosity of the mix, but once the slits were slightly widened, this problem was overcome.

After the preliminary injection testing was completed, boreholes were drilled in the selected area in a triangular grid pattern. The depth of injection was raised to 0.7 m given the shallower depth of the PASS layer in this site. After the injection of the slurry into several boreholes, a few random test pits were dug to examine the treated ground. The radius of

influence of each borehole was found to be over 1 m and an excellent overlap of the influence zones was observed to give a continuous and structurally sound barrier. A complete coverage based on the injection pattern was then executed as shown in Figure 4.

In summary, the lime-fly ash slurry was injected into 22 holes, to form the impermeable reactive barrier. The volume of slurry that was injected into each hole varied from 209 L to 314 L depending on the lateral permeability of the soil surrounding the borehole. Two weeks after the barrier was injected, further random coring of the treated area confirmed that the barrier was continuous and sufficiently hardened. The grout thickness of the barrier varied from 100-130 mm.

4. Results and Discussion

The average pH of the groundwater across the study site increased from 3.25 immediately before barrier installation to 4.6 after the barrier was installed (Figure 6). The peak in the pH around the 125th day was due to dilution caused by heavy rain plus a burst water main that flooded the site with freshwater. The peak on day 251 is also due to a significant rain event (111.8 mm). The subsequent dip in pH is caused by the accumulation of acidic products in the groundwater. Following the installation of the barrier on the 313th day, the pH of the groundwater steadily rose to 4.6. Normally the pH dropped after a rain event, but during the post-barrier period a rain event occurred on the 384th day and the pH continued to rise, which indicates that the barrier was effective in decreasing the groundwater acidity.

The groundwater pH is affected by several climatic and hydrological factors, including rainfall, making the assessment of the barrier effectiveness somewhat complex. However, as shown in Figure 7, when a comparison is made of pH at observation wells OH2, 3, 6, and 28 (see Figure 4 for location of wells), the groundwater pH certainly showed a steady improvement during the post-barrier period. Groundwater flows towards the drain in a northwesterly direction and hence OH28 is a control point. The groundwater pH in OH28 decreases with the rainfall while the groundwater affected by the barrier increases due to the influence of the barrier. The observation wells on the down-flow side of the barrier (e.g. OH2) show the greatest improvement during the final monitoring phase. This is clear evidence of a positive impact from the barrier. Similarly, Figure 8 shows the groundwater pH measured in observation wells OH2 (1 m) and OH1 (2 m) from the barrier. As expected, the

influence of the barrier on the groundwater pH was greater in the observation hole which is 1 m from the barrier than the hole 2 m from the barrier.

Prior to installation of the barrier, the EC of the groundwater varied greatly (Figure 9). A significant peak occurred during the period of low rainfall (i.e. days 56-99) and low water table. The peak is probably due to the accumulation of acidic products in the groundwater caused by the oxidation of exposed pyrite. Following the completion of the barrier (beyond day 313), the groundwater EC was relatively stable despite the low rainfall and low water table. Normally, the EC would be expected to climb rapidly during a dry period due to pyrite oxidation, but since the barrier installation, this did not occur during the dry period. The steady, low level of EC in the groundwater indicates that a significant amount of pyrite oxidation has not occurred. This is a testament to the effectiveness of the impermeable barrier in preventing the ingress of atmospheric oxygen to the pyritic layer.

The concentrations of aluminium (Al) and iron (Fe) in the groundwater (Figure 10) varied greatly during the first 100 days of the study but since the barrier installation, the levels of each have stabilised. The high concentration of Al and Fe at the beginning of the study period is due to the accumulation of acidic products during the dry period that preceded the study. The two significant rain events (125th and 251st days) have flushed the Al and Fe from the soil and diluted the groundwater. After the installation of the barrier, the Al and Fe levels remained relatively low despite the low rainfall during that period. This may indicate that either a large amount of pyrite oxidation did not occur despite the lowered water table or the pH was high enough to cause removal of the Al and Fe from the groundwater. The low Al and Fe concentrations coupled with the low EC in the groundwater suggest that the impermeable barrier decreases the amount of pyrite oxidation. The sustained low Al and Fe levels also imply that the alkaline barrier may cause some precipitation of these ions out of the groundwater.

If the ratio of chloride to sulphate (Cl/SO_4) is below two, it is an indicator of ASS conditions (Mulvey, 1993). Chloride is a conservative ion but sulphate is released into the groundwater during oxidation of pyrite (sulphate minerals such as gypsum are not common in ASS). Therefore, if the Cl/SO_4 ratio decreases over time, it indicates that oxidation of pyrite is occurring. The average Cl/SO_4 of groundwater at the study site was below one on all but one occasion (Figure 11). The large peak in Cl/SO_4 on day 99 also occurred in the EC (Figure 9),

indicating an elevated level of ions in the groundwater. After the large rain event on the 125th day, the ratio sharply decreased reflecting the dilution of the groundwater and flushing of chloride from the soil. The Cl/SO_4 in the groundwater increased after the barrier was installed (Figure 11), which is surprising considering the low rainfall and water-levels during that period. This again indicates that the barrier is effective in decreasing the amount of pyrite oxidation that occurred.

The quality of the water in the drain adjacent to the barrier was not expected to improve significantly due to the lime-fly ash barrier because of the large influence of upland flows into the drain from ASS-affected areas. An influence was noticed however in the aluminium content of the drain water (Figure 12). Downstream of the barrier, the Al content decreased from 37 mg/L at the beginning of the study to 9 mg/L after the barrier was installed. The decrease in Al content from the beginning to the end of the study is clear evidence that the barrier had removed the Al from the groundwater prior to release into the drain. The peak in the concentration of Al on the 70th day (75.3 mg/L) occurred after light rain that followed a dry period. This is attributed to the rain flushing the accumulated Al out of the soil and into the drain. The levels dropped (14.2 mg/L) then rose again (day 125, 51.1 mg/L) following 162 mm of rain that had flushed more Al from the soil. The level of Al then plummeted and on the 140th day (0.4 mg/L) the level was very low due to the large rain event and the bursting of a fresh water main that fully diluted the drain water. In the post-barrier period, a rain event followed a dry period and this would normally increase the level of Al in the drain, however, the level of Al continued to stay low. In contrast in the drain water upstream of the barrier (Figure 12), the Al content increased after the post-barrier rain event. This is convincing evidence that the barrier reduced the amount of Al that was released into the drain.

5. Conclusions

The installation of the sub-surface lime-fly ash barrier at the study site was successful in relation to improving the groundwater quality. The groundwater quality data showed that pyrite oxidation products were generated in the pre-barrier period as a result of falling groundwater tables and biotic oxidation. After the installation of the barrier, substantial improvements in groundwater quality occurred due to decreased pyrite oxidation, as indicated convincingly by:

1) a rise in pH after installation of the barrier;
2) a decrease in EC (4.60 to 1.47 mS/cm);

3) decreases in levels of Al (65.5 to 20.3 mg/L) and Fe (161 to 42 mg/L); and
4) an increase in Cl/SO_4 ratio from 0.23 to 0.80.

The investigation of the durability and structural integrity of the alkaline barrier in the long term was not within the scope of this study. However the current field observations indicate without any doubt a structurally sound barrier with a high level of performance in the short-to-medium term. Numerical modelling is currently ongoing to study the long-term performance of the barrier in terms of chemical decomposition and potential instability due to soil movement and these results will be reported at a later stage.

The implications of the research presented here extend beyond the scope of remediation of ASS terrains. Impermeable, alkaline horizontal barriers should have similar potential for the prevention of oxidation of sulphidic tailings and other buried acidic waste materials. In this context, the observations of this study may be considered as encouraging evidence to promote the implementation of reactive barriers for such circumstances as well.

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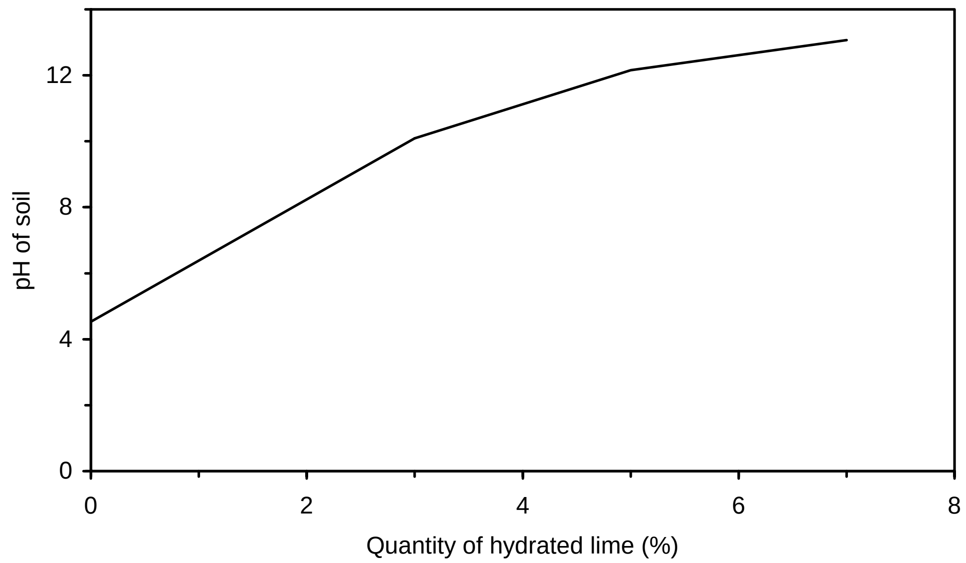

Figure 1. Relationship between percentage of hydrated lime added (compared to mass of the soil) and pH of acid soil (adapted from Indraratna, 1996)

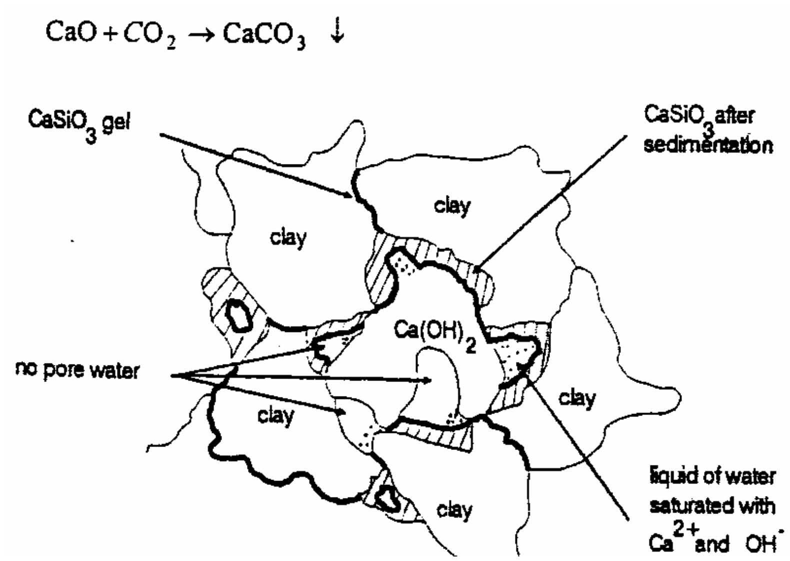

Figure 2. Formation of calcium silicate around soil particles (van Impe, 1989).

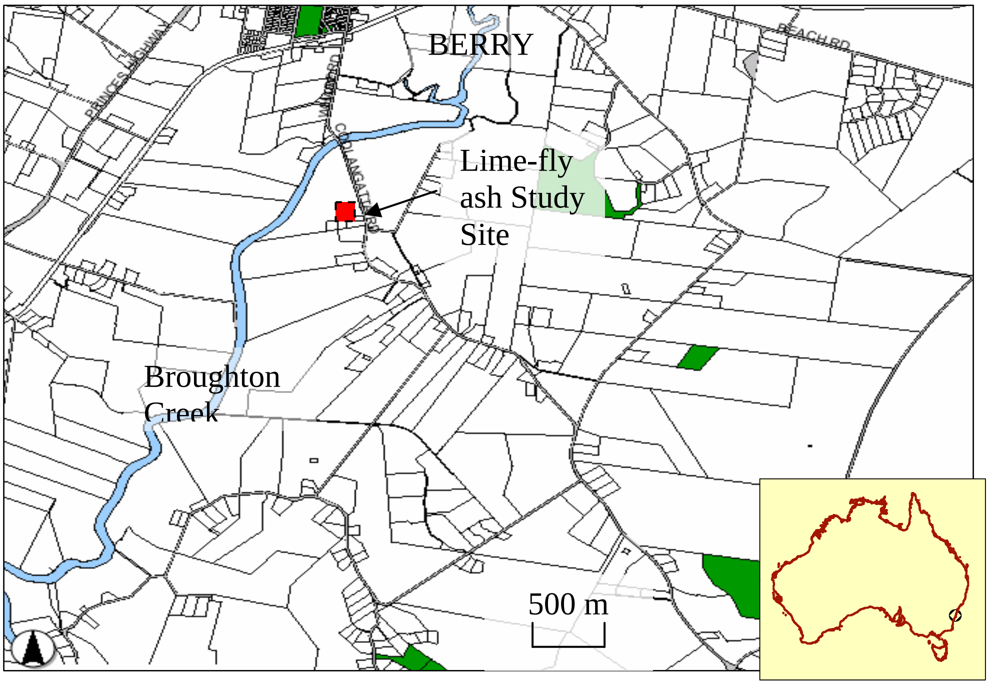

Figure 3. Location of the study site.

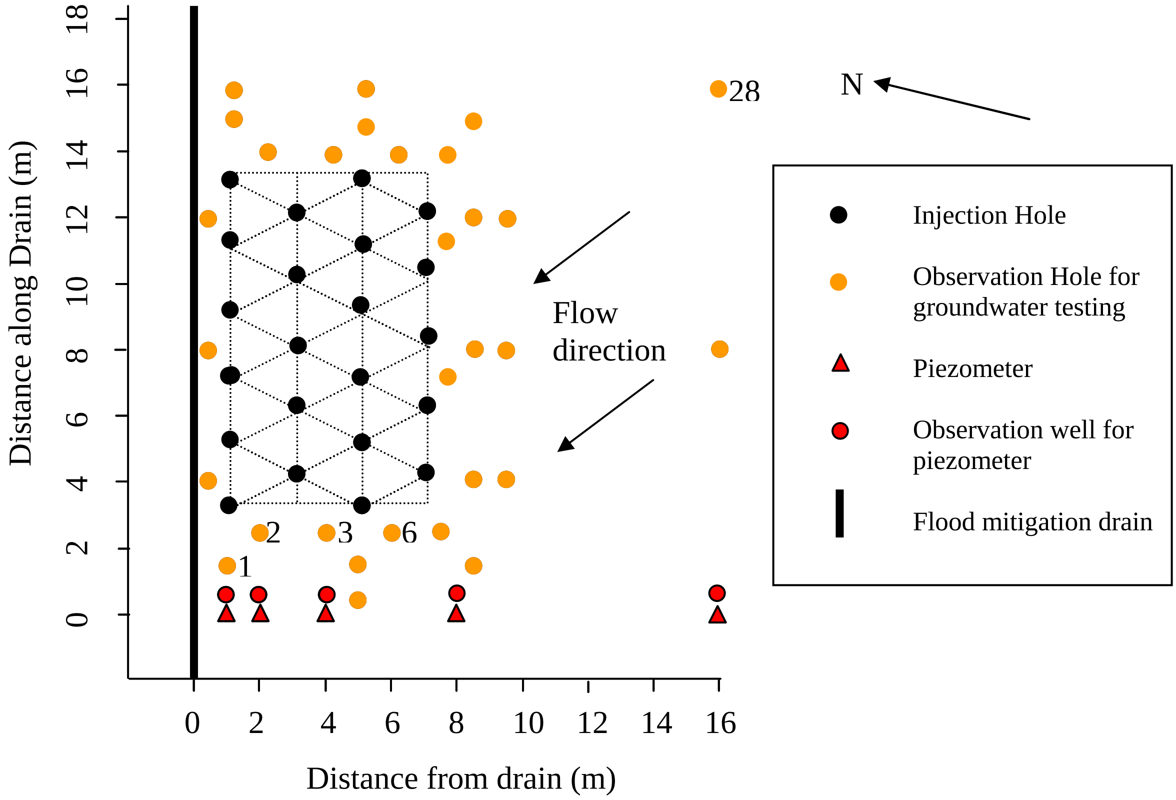

Figure 4. Layout of study site showing the location of observation wells and piezometers.

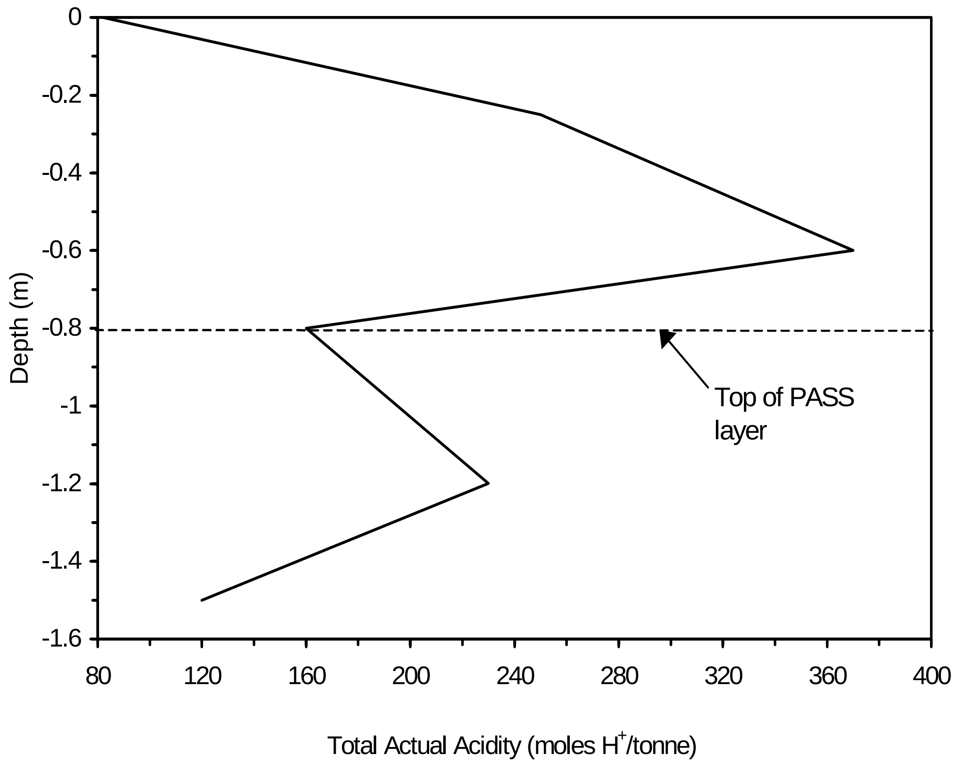

Figure 5. Change in soil total actual acidity with depth below ground surface.

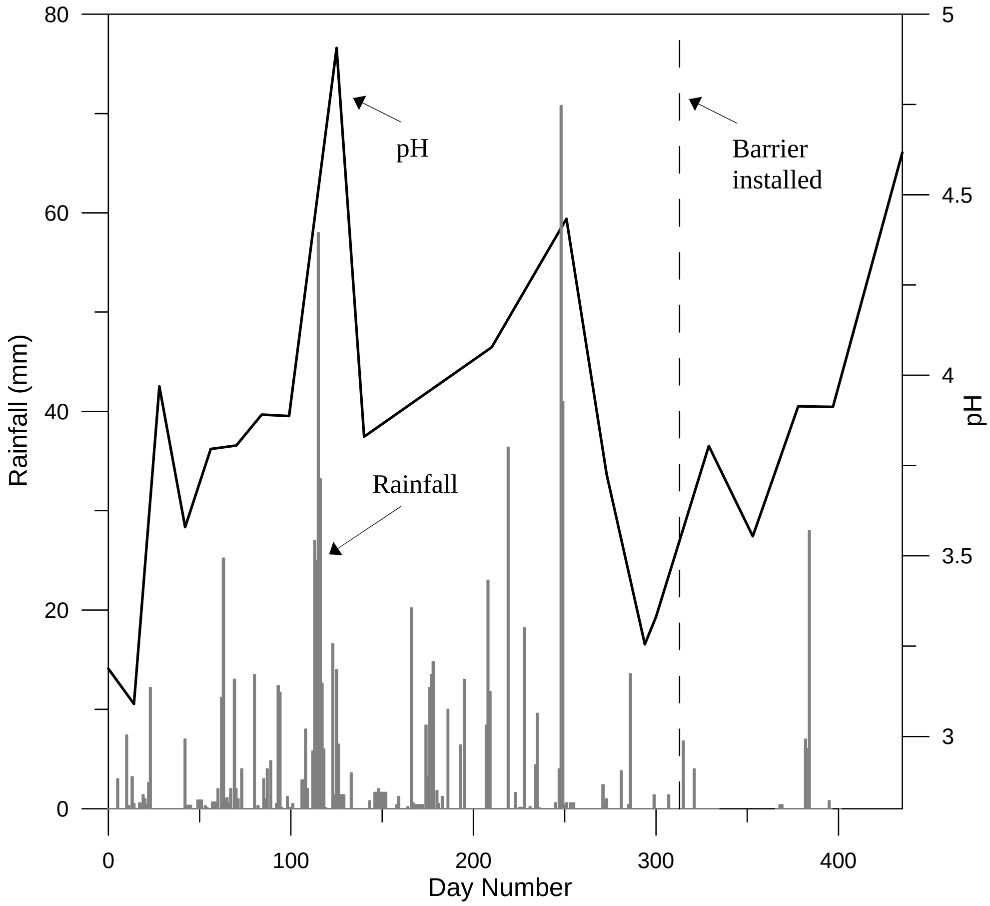

Figure 6. Groundwater pH and rainfall during the study period, averaged from 31 observation wells.

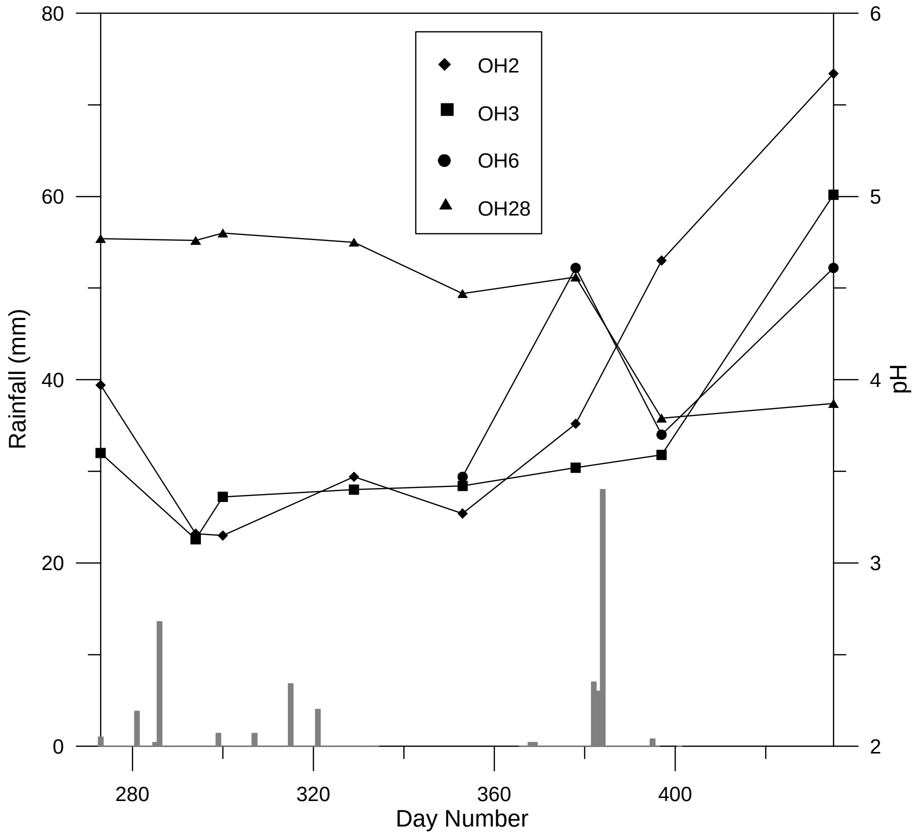

Figure 7. Groundwater pH measured at observation wells OH2, 3, 6 and 28 and rainfall after the 273rd day. The barrier was installed on the 313th day.

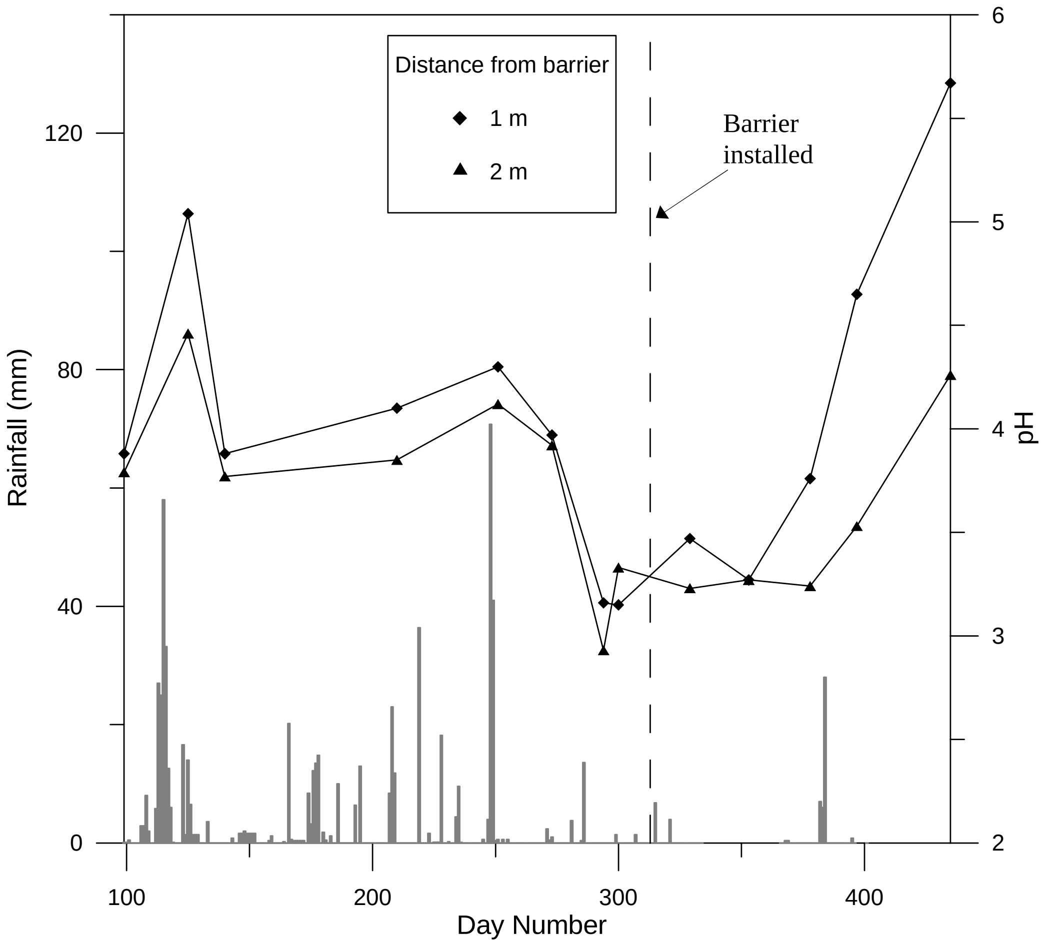

Figure 8. Average groundwater pH measured at 1 and 2 m from the barrier and rainfall.

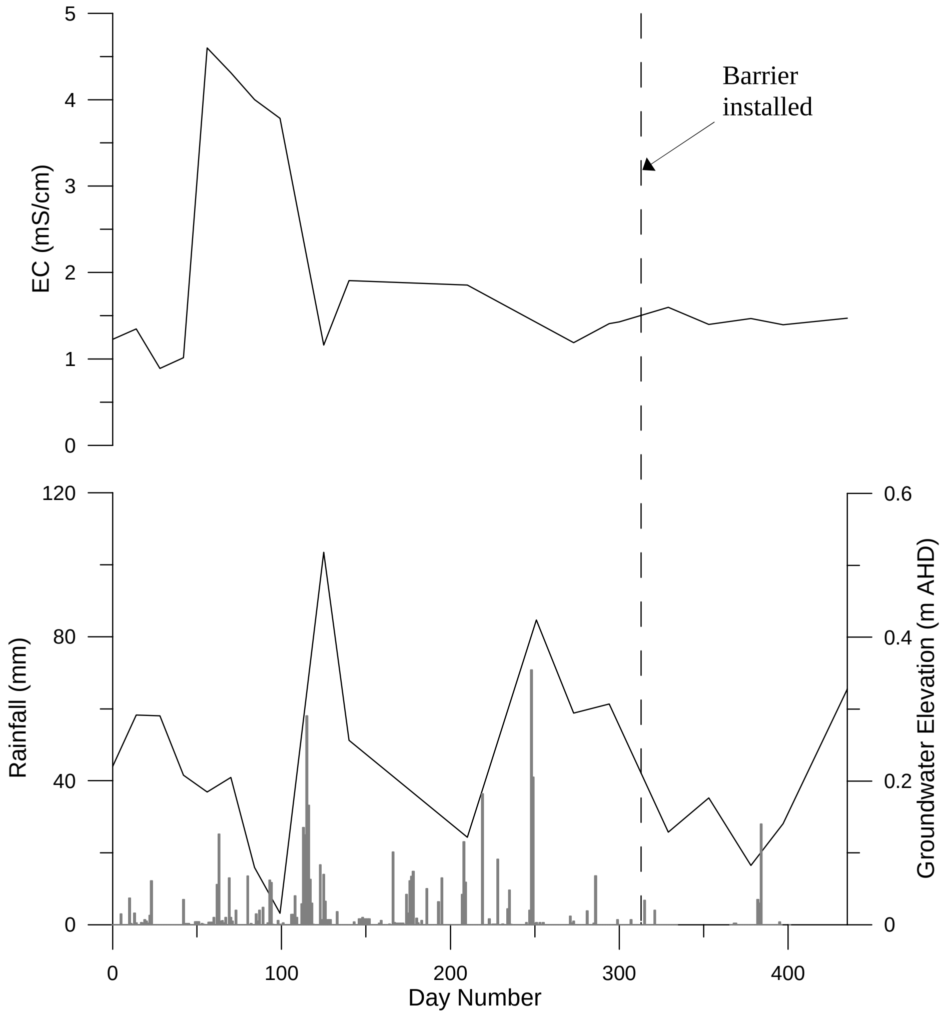

Figure 9. Groundwater electrical conductivity (EC) and elevation during the study period, averaged from 31 observation wells and rainfall.

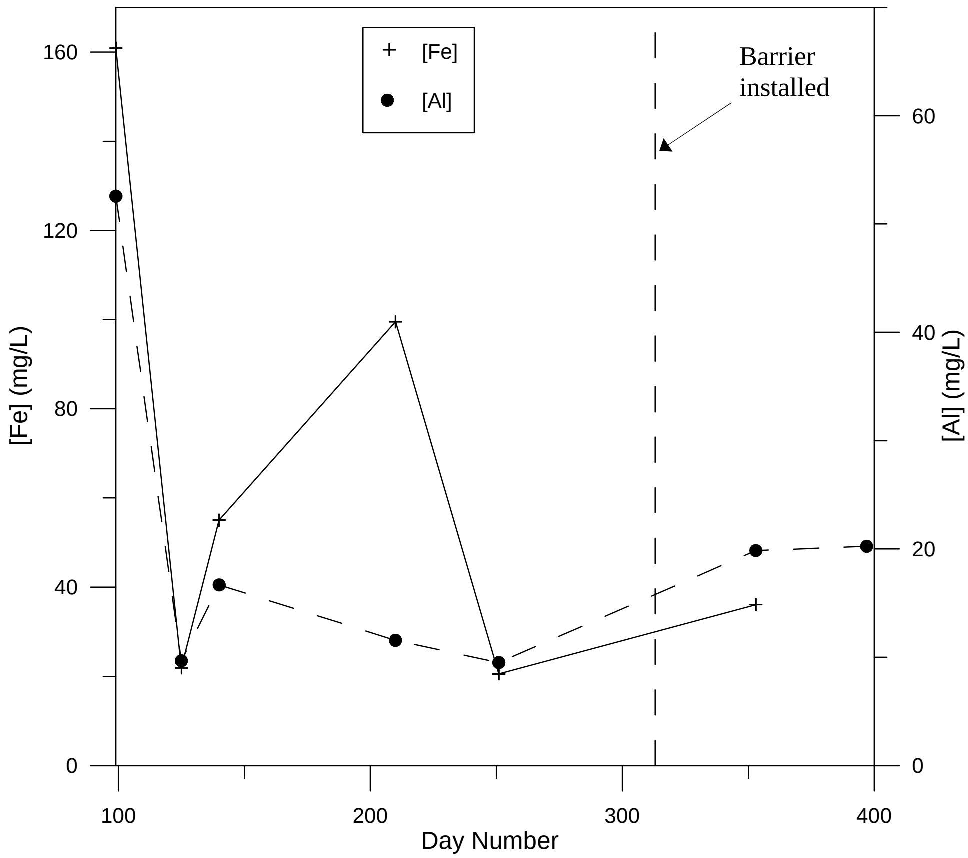

Figure 10. Groundwater concentration of iron and aluminium during the study period, averaged from 31 observation wells.

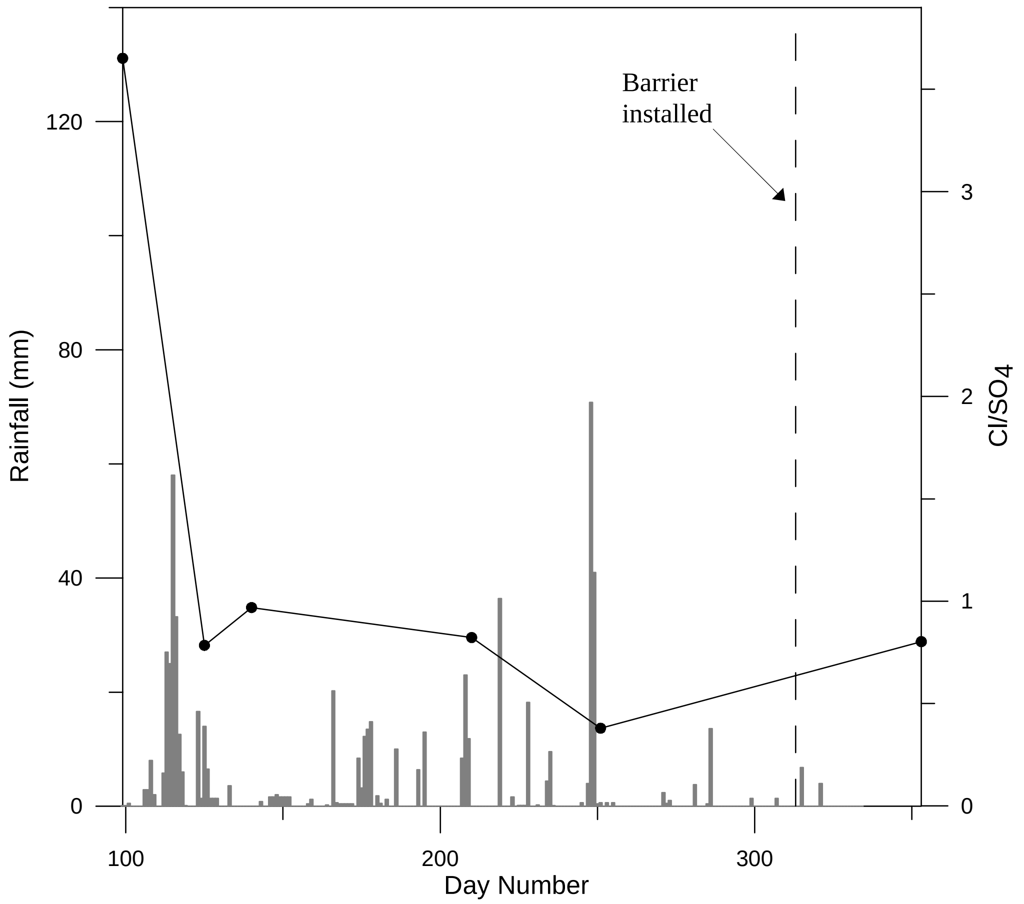

Figure 11. Ratio of chloride to sulphate in the groundwater during the study period, averaged from 31 observation wells and rainfall.

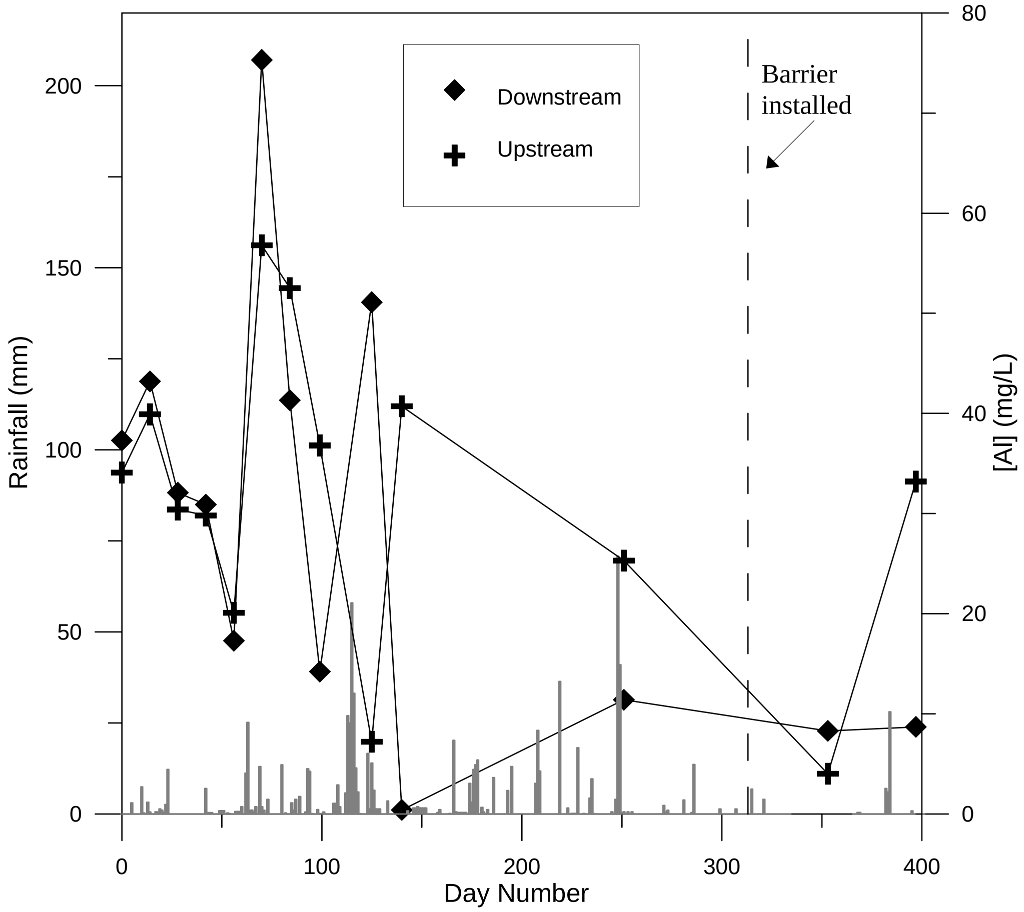

Figure 12. Concentration of aluminium in the drain downstream and upstream of the lime-fly ash barrier and rainfall over the study period.