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Temporal and spatial variability of acid rock drainage in a rehabilitated coal mine, Wangaloa, South Otago, New Zealand

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Abstract The Wangaloa open cast coal mine ceased operations in 1989, with no restoration of the 252 ha site, and moderate acid rock drainage developed. A major rehabilitation programme was initiated in 2002 with removal of exotic vegetation, and extensive planting (>60 000) of native seedlings was begun in 2003. By 2006, most seedlings were thriving, and, combined with adventive exotic weeds, a 70% vegetation cover had been achieved. The site substrates were highly variable on the 10-100 m² scale, and have been characterised by paste pH (>700 measurements). In 2003, substrates had moderate acidity (pH = 4.5 ± 0.9) with distinctly acid patches (pH down to <2). By 2006, the average substrate pH was essentially unchanged. Some distinctly acid patches had higher pH, and one patch had apparently become more acid. Water compositions (>100 samples from 15 sites) were also highly variable spatially and temporally. Incoming stream and rainwater (pH 5-6) chemically interacted with acid substrates, especially waste rock piles that contain pyrite-bearing material, and evolved to lower pH (pH down to 3.4), sulfaterich waters. A pit lake on the site receives most surface and groundwater runoff, and this lake, with a water residence time of 1-2 yr, controls the site discharge water quality. The lake pH varies on a monthly time-scale from 4.5 to 6.5, synchronised with pH variations in groundwater boreholes in waste rock. In addition, there has been a general increase in pH of the lake during rehabilitation from consistent pH 4.6-4.8 before rehabilitation to near pH 6 during rehabilitation. The sulfate/chloride ratio of lake water has decreased from 20 to <10 during rehabilitation as well. These changes in lake water composition from year to year may be a result of increased input of rainwater that has had less interaction with substrate than runoff water had before rehabilitation began.

Keywords acid mine drainage; lignite; sulfate; pH; boron; water; rehabilitation

INTRODUCTION

Excavation of coal mines exposes large volumes of rock to rainwater and atmospheric oxygen, facilitating enhanced interaction between water and rock (Hutchison & Ellison 1992; Lottermoser 2003). This enhanced water/rock interaction can have negative effects on water quality in the mine site and in downstream discharges (Hutchison & Ellison 1992; Lottermoser 2003). Water quality from coal mines is especially affected if the exposed rocks contain pyrite, and acid rock drainage commonly develops (Sengupta 1993; Monterroso & Macias 1998; Wisotzky & Obermann 2001). Control of acid rock drainage, and associated mobilised metals, is the principal aim during operation and rehabilitation of modern coal mines (Hutchison & Ellison 1992; Sengupta 1993).

Despite the modern focus on maintenance of mine site water quality, many historic coal mine sites around the world have been abandoned with little attempt to control, or even monitor, the quality of discharging waters (Sengupta 1993). New Zealand is no exception, and there are many abandoned coal mines with poor water quality (Black & Craw 2001; Alarcón León & Anstiss 2002; Black et al. 2005; de Joux & Moore 2005; Hewlitt et al. 2005). However, some of these sites are being rehabilitated to improve the physical appearance and quality of discharging water (Trumm et al. 2005; Craw et al. 2006).

This study presents observations on the nature and evolution of water compositions at one of the abandoned coal mine sites in New Zealand that is being rehabilitated. This site is small (<300 ha) and our sampling programme was intense for such a small site. This intense sampling strategy showed that the site is highly variable spatially and temporally. We quantify this variability, and suggest reasons for that variability as waters evolve in contact with variably reactive substrates. We focus on pH in substrates and water, and on water sulfate concentrations, as these are key acid rock drainage parameters. Boron mobilisation and attenuation at this site has been described by Craw et al. (2006), but we report further boron data here in the wider context of acid rock drainage. Other trace element compositions were described by Black & Craw (2001), and more detailed trace element variations are beyond the scope of the present study. We focus on quantification of variations in acid rock drainage parameters through the site, and with time, during the early stages of site rehabilitation. We show that the positive effects of rehabilitation on the water quality of the site can be quantified, even at an early stage.

GENERAL SETTING

Geology and geography

The Wangaloa opencast coal mine (252 ha) was developed in South Otago, New Zealand (Fig. 1A). The site occurs in rolling hills c. 2.5 km from the coast. The area has a cool temperate climate, with mean annual temperature of c. 12°C

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Fig. 1 Geographic, geological, and stratigraphic location diagrams for the Wangaloa coal mine. **A**, Geological map of coastal Otago, showing the location of the Wangaloa mine in relation to basement rocks and Cretaceous–Quaternary cover rocks. Locations of typical regional waters used for comparison to Wangaloa mine waters are indicated: Taieri basin (schist/greywacke gravel; Litchfield et al. 2002), Sutton Salt Lake (rainwater; Craw & Beckett 2004), and Papakaio Formation (coal measures; ORC 1993). **B**, Stratigraphic column for South Otago (modified after Harrington 1958), showing the setting of the Wangaloa mine (box) in the Taratu Formation and unconformably overlain by Quaternary loess.

and rainfall of 700–1000 mm/yr. The coal-bearing strata at the Wangaloa mine are part of the Late Cretaceous Taratu Formation, which is a regionally extensive unit extending from South Otago to North Otago (where it is called the Papakaio Formation) (Fig. 1). The Taratu Formation in South Otago includes quartz conglomerate (with minor greywacke clasts locally), interlayered siltstones, and numerous coal seams (Fig. 1B) (Harrington 1958). The Taratu Formation is >500 m thick in the vicinity of the Wangaloa mine, and overlies greywacke basement that has local fault-bounded middle Creataceous immature sedimentary basins (Henley Breccia) (Fig. 1B). The Wangaloa mine was developed in the stratigraphic central part of the Taratu Formation (Fig. 1B), and Taratu Formation sediments underlie all the surrounding hills. The Taratu Formation is overlain in South Otago by Tertiary marine sediments (Fig. 1B), but these have been eroded from the Wangaloa mine area. Coal was mined near the hinge of a broad anticline that plunges gently east or northeast, and coal dips gently east or north in the opencast mine (Fig. 2). The hills around the Wangaloa area are coated with Ouaternary loess, up to 4 m thick. This loess consists of immature silty material derived from the schist and greywacke basement.

Development of the opencast mine involved stripping of the loess and underlying Taratu Formation quartz conglomerate from above the coal. This waste rock, which is predominantly a poorly consolidated mixture of quartz pebbles, quartz sand, coal, and lumps of loess, was dumped on the mine site (Fig. 2). The proportions of these waste rock components vary widely and have controlled local natural revegetation (Craw et al. 2007). Stripping of waste rock has left some steep high walls (up to 50 m high) in the mine site (Fig. 2). The mine site is confined to a single stream catchment, with small streams entering at the upstream (western) side, and all water discharges out the eastern side of the site (Fig. 2). Most waste rock was dumped in the valley of this stream catchment, after the valley floor had been lowered by extraction of coal in the hinge of the anticline (Fig. 2). Three lakes and three wetlands have formed locally in low-gradient parts of this anthropogenic topography (Fig. 2). The Main lake (Fig. 2) is 300 m long, 40-60 m wide, and up to 9 m deep, with an estimated volume of c. $7 \times 10^4 \,\mathrm{m^3}$.

Coal, mining, and rehabilitation

Mining at Wangaloa began in the 1940s and ceased in 1989. The principal seam mined at Wangaloa (Barclay Seam) is up to 9 m thick and traceable for several kilometres through the mine area (Harrington 1958). This seam, and some overlying coal seams, was mined underground historically immediately to the south of the opencast mine site (Harrington 1958) (Fig. 2). The coal is high rank lignite, with relatively high sulfur (2-5 wt%) and locally elevated boron (up to 450 mg/ kg) that is typical of vitrinite-rich coal from marginal marine environments (Goodarzi & Swaine 1994; Craw et al. 2006). Sulfur and boron are generally distributed through the organic component of coal (Suggate 1959; Goodarzi & Swaine 1994; Newman et al. 1997; Craw et al. 2006). In addition, there is abundant pyrite scattered through the Wangaloa coal, in primary sites such as framboids and infilling of wood cells (Fig. 3A), and secondary sites such as along cleat (fracture) surfaces. Pyrite is most abundant near the top of the Barclay Seam at Wangaloa (Craw et al. 2006). Pyrite also occurs as a cement in quartz conglomerate that immediately overlies the Barclay Seam (Fig. 3B). This pyrite cement typically extends only 1-10 cm from the coal, and stratigraphically higher quartz conglomerates have low pyrite contents. Exposed coal surfaces, in outcrop and in waste clasts, are commonly coated with white gypsum crystals (um scale), especially under outcrop overhangs. Yellow jarosite and brown iron oxyhydroxide (HFO) locally accompany gypsum, especially on pyrite-rich coal.

Fig.2 Map of the Wangaloa open cast coal mine showing principal geographic features, main features of the drainage system, and water sampling localities in relation to the mine waste rock dumps. The approximate trace of the principal mined coal seam (Barclay Seam) is shown with triple-line ornament; much of this trace is now obscured by subsequent covering material.



An area of unmined land to the north of the main high wall retained remnants of native bush, and this is regenerating naturally (Fig. 2). Small areas (10-100 m² scale) of naturally regenerating native bush have developed on the mine site since disturbance ceased, especially at the eastern end of the site (Craw et al. 2007). Initial management of disturbed land in the 1980s involved planting of Pinus radiata trees in and around the site. These trees did not thrive on much of the waste rock, and some unplanted areas became infested with gorse (Ulex europaeus). In addition, moderate acid mine drainage issues had developed by the 1990s because of oxidation of pyrite in waste rock piles (Black & Craw 2001). A concerted effort at rehabilitation was begun in 2000 by Solid Energy Ltd, with the aim of turning the site into a recreational area. This latest rehabilitation strategy, during which the present study was conducted, included removal of gorse and Pinus radiata. Surface recontouring stabilised slopes (2002), and this was followed by planting of >60000 native seedlings. Sprayed-on grass establishment (hydroseeding) was also employed for stabilisation of waste rock slopes. In addition, c. 300 kg of hydrated lime was added to the Main lake in 2002, in an attempt to raise the pH of the lake and downstream discharge water. By 2006, most of the native plants were surviving, and spaces between these plants had been invaded by exotic weeds, which were controlled in places with occasional spraying. At this stage, c. 70% of the site was revegetated.

METHODS

Sample sites

Surface water samples were taken from streams and lakes at sites indicated in Fig. 2 throughout the mine catchment specifically for this study. Some additional historical data on

the Main lake were provided by Solid Energy NZ Ltd. A small lake on the western margin of the mine site (West lake sample site; Fig. 2) captures rainfall and local surface runoff from mainly undisturbed Taratu Formation, and is representative of background surface water at the top of the site. Inflowing background streamwater was sampled upstream of the disturbed part of the mine site. This stream flows over naturally eroded Taratu Formation sediments, with minor quartz conglomerate waste rock nearby (Fig. 2). This stream continues to form a wetland (upper wetland sample site; Fig. 2) before flowing past another surface water sample site into the Main lake (Fig. 2). The upper wetland and its discharge stream are dry during periods of low rainfall. The Top lake sample site has formed against an outcrop of the Barclay Seam and partly rests on waste rock (Fig. 2). Ephemeral puddles of surface water on coal-rich waste rock (west side of site; Fig. 2) were sampled occasionally after rainfall. The Main lake, which is trapped by coal outcrop and waste rock piles, was sampled near its eastern end (Fig. 2). Main lake dissolved oxygen and temperature profiles were determined at 1 m depth intervals in May 2004, from a boat at eight points across the lake, to check for lake stratification. There were negligible differences between these results and observations taken from the shore, so depth sampling was not pursued further.

Surface water draining through the lower wetland was sampled immediately downstream of the Main lake and near the eastern boundary of the site (Fig. 2). The sample site immediately downstream of the Main lake had no surface water for only one monthly sampling period. At other times, surface water was flowing at this site at 1–3 litres/s, as determined using a bucket at a narrow reach in the wetland. Water sampling transects across the eastern end of the lower wetland (5 m sample spacing; Craw et al. 2006), and along the southern margin of the lower wetland (c. 50 m sample spacing), were conducted to quantify the composition and extent of seeps emanating from beneath the waste rock piles.

wood fragment in coal pyrite 10 µm uncemented guartz gravel HFO stained quartz gravel pyrite cemented quartz gravel coal 10 mm

Fig. 3 Photographs of textures of pyrite associated with the Barclay Seam coal at Wangaloa mine. A, Incident light view of a polished section of coal, showing pyrite (white) filling relict wood cells. B, Hand specimen photograph of quartz conglomerate (upper part of view) overlying coal (black; bottom). Quartz pebbles immediately overlying the coal are cemented with coarse-grained pyrite (commonly with shiny white crystal surfaces).

Boreholes were placed into site substrates to allow sampling of groundwater at strategic sites (Fig. 2). Boreholes 1–5 were placed with a 76 mm drill bit, and 20 mm PVC pipe was installed in all but borehole 1 (50 mm) because of instability of the material at depth. Boreholes 6 and 7 were dug into the waste rock and lined with 50 mm PVC pipe. Borehole 1 (18 m deep) was placed into Taratu Formation at the western (up-gradient) side of the site (Fig. 2) to intersect incoming groundwater, to provide information on background groundwater compositions. Borehole 3 (11 m deep) and borehole 6 (2.4 m deep) are in waste rock piles immediately uphill of the Main lake, and intersect groundwater flowing into the lake. Borehole 7 (2.4 m deep) intersects groundwater leaving the Main lake (Fig. 2). Borehole 5 (30 m deep) intersects groundwater flowing through the eastern waste rock piles.

The pH of solid substrates over the site was determined on slurries made on-site (paste pH) at randomly selected locations at sufficient spacing to cover the whole site with c. 200 measurements. These locations were easily selected in poorly vegetated areas, but access to parts of the site in the latter stages of this study was limited by thick vegetation, and paste pH locations were selected partly according to accessibility.

Sampling periods

There were three temporal phases for sampling in this study. The first phase involved initial measurement of Main lake pH from 1998 to the initial stages of site clearance and replanting in 2003. The first documentation of substrate paste pH over the site was made in January 2003, when the site was largely unvegetated but most recontouring had been completed. Minor recontouring of waste rock occurred subsequently. The second sampling phase involved more intense documentation of surface water and groundwater compositions for 1 yr, from November 2003 to November 2004. Samples were collected monthly at most of the sites indicated in Fig. 2. Meteorological data for this sampling period, including monthly rainfall, were obtained at the Balclutha climate station, 20 km west of Wangaloa (Fig. 1). A substrate paste pH survey of the whole site was conducted in January 2004, and a more intense paste pH survey of a small area of waste rock was conducted in November 2004. The third phase of sampling, from January 2005 to July 2006, involved monthly measurement of water pH, sulfate, and boron concentrations in the Main lake and the lower wetland (Fig. 2), with occasional sampling of other surface waters for the same parameters. The sampling campaign ended with a substrate paste pH survey of the whole site in October and November 2006.

Sampling and analysis

Water samples for major cations and anion analysis were collected in 1 litre polyethylene bottles. Surface waters were sampled by dipping bottles in the water following rinsing in the water. Groundwater samples were taken with disposable PVC bailers with bottom-emptying valves that allowed pouring into the 1 litre bottles. The boreholes were sampled after purging, by bailing, of 3–5 volumes of each hole, to remove stagnant water. Borehole 1 recharged slowly, so this hole was sampled after c. 30 min of recharge time.

Dissolved sulfate was analysed via ion chromatography (APHA 1998) by Chemsearch Ltd (University of Otago Chemistry Department), an internationally accredited laboratory, using a Matrohm Duel 1 anion column with a Waters 320 detector. Cations were analysed in the Geography Department, University of Otago, using a Varian SpectrAA 220 FS atomic absorption spectrophotometer, with methods specified in ISO 11885 (1985, pp. 151–160). Chloride analysis was also done in the Geography Department, using a FOSS Tecator Fluid-Injection Analyser (FIAstar 5000). Alkalinity was determined by Chemsearch via potentiometric titration. Detection limits for cations and sulfate were 1 mg/litre; for alkalinity was 1.2 mg/litre; and for chloride was 0.06 mg/ Begbie et al.-Acid rock drainage in rehabilitated Wangaloa coal mine

litre. Representative analyses are presented in Table 1, and additional water analyses from the site are presented by Craw et al. (2006).

Water samples for B analysis were collected in acidwashed plastic bottles and preserved with nitric acid in the field. Surface water samples were filtered (0.45 μ m), as were most lake and wetland waters. Some lake and wetland waters were collected unfiltered; results for filtered and unfiltered lake and wetland waters collected at the same time showed negligible difference. Total dissolved B analyses were done by ICP-MS, using internal laboratory standards, by Hill Laboratories, Hamilton, New Zealand, an internationally accredited laboratory. Detection limit for these analyses is 0.005 mg/litre, and sample replication analyses agree within 0.03 mg/litre in the range 0.70–1.60 mg/litre.

The pH of surface waters and groundwaters was determined in the field using a portable Oakton meter calibrated on-site with standard pH solutions. Measurements are reproducible to ± 0.1 pH units. Paste pH of the site substrate pH was determined on distilled water slurries of surficial material, made in the field by the method of Sobek et al. (1978) and using the same pH meter. Paste pH locations were determined by differential GPS, with horizontal accuracy of c. 1–2 m.

RESULTS

Substrate pH

The area around the Wangaloa mine has been extensively modified for agriculture and forestry, so determination of background pH values for the soils and substrates is difficult. Most South Otago hill soils are developed on loess or redeposited loess-rich material. These soils are normally moderately acid, with pH between 5 and 5.5 (Blakemore 1968). Likewise, hill soils developed on Taratu conglomerate are typically moderately acid (pH 5–5.5; Blakemore 1968). Naturally regenerated bush sites on loess-rich substrate, undisturbed during the latest rehabilitation process at the Wangaloa mine, have paste pH between 4.5 and 5.3. Relatively undisturbed soils developed on Taratu Formation on the north side of the site, in and near the regenerating native bush (Fig. 2), have pH between 4.2 and 5. Hence, moderately acid soils are the norm for the Wangaloa area.

Paste pH measurements of substrates over the mine site in the three survey periods are shown in Fig. 4A-C, along with the means and standard deviations for each survey. These data show that the substrates are moderately acid (typically between 3.6 and 5.4) and variable over most of the site. Weakly acid substrates (pH >5.4) are relatively rare on the site. More distinctly acid substrates occur in the waste rock piles, especially on the western side of the site. Some distinctly acid substrates in the relatively undisturbed southwest corner of the mine site in 2003 (Fig. 4A) appear to have become less acid only 1 yr later (Fig. 4B), although data are highly variable. Likewise, some distinctly acid waste rocks (pH <3.6) between the Main lake and Top lake in January 2003 may have become less extensive, and pH of that area appears to have evolved to more typical site values by the end of 2006 (Fig. 4A-C). This area of waste rock includes relatively high proportions of coal fragments, and plants in this general area have elevated, but not toxic, levels of boron (Craw et al. 2006). Conversely, some distinctly acid (pH < 3.6) waste rocks near the eastern end of the site in 2003 are not apparent in 2004, but these were distinctly acid in 2006 (Fig. 4A-C). Spatial variability of pH on the metre scale may account for at least some of the observed differences between Fig. 4A,B and C.

An area of acid substrate was identified on a quartz-rich waste rock slope above (north of) the upper wetland (Fig. 2) in 2004. The area became apparent because all planted native seedings died, as did areas of hydroseeded grass on this slope. Rainwater runoff removed fine debris from the surface after recontouring in 2002 and 2003, and 1-10 cm deep rills developed. This erosion exposed numerous scattered blocks of quartz conglomerate cemented with pyrite, including the specimen in Fig. 3B. To quantify the extent of the low pH region, a more detailed substrate paste pH map was constructed of this slope (Fig. 5). This map shows that the most acid substrates occur in a well-defined zone c. 80 m wide, and the pH ranges between 2 and 3 on that part of the slope. The margins of the acid substrate zone, where substrate pH rises laterally to >4, are defined by the presence of live vegetation on both sides. Blocks of pyrite-cemented conglomerate on these marginal vegetated slopes are surrounded by a 5-10 cm ring of dead grass.

Table 1 Representative water analyses (mg/litre except pH) from the Wangaloa mine site, from locations in Fig. 2. Bkgd = background groundwater and surface lake water; Waste = surface and groundwater from waste rock; Ground = groundwater from borehole. Alkalinity is presented as HCO_3^{-} .

-	5									
Location	Bore hole 1	West lake	Bore hole 6	Upper wetland	Rain runoff	Top lake	Main lake	Main lake	Main lake	Lower wetland
Setting Type	Bkgd Ground	Bkgd Lake	Waste Ground	Waste Stream	Waste Stream	Waste Lake	Waste Lake	Waste Lake	Waste Lake	Waste Stream
Date	17 Aug 04	17 Aug 04	20 May 04	17 Aug 04	17 Aug 04	15 Jan 04	13 Oct 04	13 Sep 04	15 Jan 04	10 Dec 03
pН	6.3	5.5	5.1	4.2	4.9	6.0	5.5	5.9	5.7	4.1
Na⁺	37.5	8.5	3.3	22.5	3.1	7.2	19.9	19.7	18.3	17.8
K+	3.0	2.5	2.1	3.8	0.9	2.5	3.1	2.9	4.1	3.9
Mg ²⁺	16.2	1.9	2.8	10.0	3.0	8.9	28.9	28.4	38.7	35.4
Ca ²⁺	15.4	1.4	2.9	9.8	2.2	17.0	54.4	60.9	54.5	44.5
HCO,-	25.0	3.7	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2	<1.2
SO, ²⁻³	70.0	6.7	21.3	75.1	19.8	73.7	237.0	262.0	303.0	297.0
Cl-	71.4	17.1	3.4	34.8	3.1	16.8	33.9	36.2	21.9	7.4
В	0.04	0.05	0.09	0.18	0.21	0.51	0.80	0.81	1.17	1.07



Fig. 4 Maps of the Wangaloa mine site as for Fig. 2, with paste pH data measured on substrates in January 2003 (A), February 2004 (B), and October–November 2006 (C). Means and standard deviations for each sampling survey are shown on each map. **Fig. 5** Paste pH map of the acid slope on the main waste rock pile above the upper wetland (see Fig. 2 for location), determined in November 2004. The dashed lines show the upper and lower margins of the slope.



Water composition variations

Background surface and groundwaters at the Wangaloa site have higher pH (between 5 and 7) compared to other Wangaloa waters (Table 1). The background surface and groundwaters have relatively higher proportions of chloride than the other Wangaloa waters, especially samples from the West lake (Fig. 6A). These West lake waters have similar cation and anion proportions to surface waters fed by coastal Otago rainwater, as indicated by Sutton Salt Lake (Fig. 1, 6A) and nearby streams (Craw & Beckett 2004). This coastal rainwater contains marine aerosols, and therefore has similar cation and anion proportions to sea water (Craw & Beckett 2004). Hence, West lake appears to be largely rainwater fed. The incoming background surface, stream, and groundwater (borehole 1; Fig. 2) have higher proportions of Ca²⁺, Mg²⁺, and SO²⁻ than the West lake waters, with sulfate up to 70 mg/ litre in groundwater (Table 1). These background waters have flowed on and/or in Taratu Formation sediments and chemically resemble groundwaters from the Papakaio Formation (=Taratu Formation) of North Otago (ORC 1993) (Fig. 1A, 6A). All background surface and groundwaters are distinctly different from surface and groundwaters, respectively, that have chemically interacted with schist or greywacke, such as waters from basement or from immature schist or greywacke gravels, which typically have low sulfate and chloride, and elevated alkalinity (Litchfield et al. 2002). Typical water from schist and greywacke in the Quaternary Taieri basin (Fig. 1) is shown in Fig. 6A for comparison.

In contrast to the background surface and groundwaters, Wangaloa surface waters and groundwaters from waste rock are more acid (pH typically <5) and relatively enriched in sulfate, range up to 700 mg/litre in groundwater (Table 1: Fig. 6B, 7A). These waters also have a broad compositional range, from waters similar to background to sulfate-dominated waters (Fig. 6B, 7A). This range extends beyond the composition of typical Papakaio (=Taratu) Formation waters (Fig. 6B). The most sulfate-rich waste rock waters (Fig. 8A) are emanating from beneath waste rock piles along at least 100 m of the southern margin of the lower wetland (Fig. 2) (Craw et al. 2006), and other waste rock groundwaters have sulfate up to c. 300 mg/litre. The Ca2+ content of surface and groundwaters in waste rock is broadly correlated ($R^2 = 0.62$) with the sulfate concentration (Fig. 7B). This dataset excludes the most sulfate-rich samples of the lower wetland, as Ca²⁺ content was not determined on those samples.

Both the Top lake and the Main lake waters have compositions similar to the most sulfate-dominated waste rock surface and groundwaters, with the Main lake having the highest proportion of sulfate (Table 1). The Main lake waters have sulfate concentrations between 200 and 300 mg/litre (Table 1; Fig. 7A), at the upper end of the range for waste rock surface and groundwaters other than those in the lower wetland (Fig. 7A, 8A). Like the waste rock surface and groundwaters, the Main lake waters have elevated Ca²⁺ content accompanying elevated sulfate (Fig. 7B).

Boron concentrations in waters are indicative of interaction with boron-rich coal (Craw et al. 2006). Background surface and groundwaters have negligible B content (c. 0.1 mg/litre) (Table 1; Fig. 7C). Waste rock surface and groundwaters commonly have elevated B (typically c. 0.5–2 mg/litre) (Fig. 7C), especially those with higher sulfate (Fig. 7C). However, B and sulfate are not well correlated ($R^2 = 0.08$). A positive relationship between B and sulfate contents is more apparent in the generally high-sulfate, high-boron waste rock waters discharging as seeps discontinuously along a 200 m front on the southern side of the lower wetland (Fig. 8B; $R^2 = 0.42$). Main lake waters have B contents consistently near 1 mg/litre (Fig. 7C). Boron and sulfate are positively correlated ($R^2 =$ 0.53) in Main lake waters in a similar trend to the waste rock waters.

Temporal variations in water compositions

The pH of the Main lake has been the parameter of most environmental concern at the site, and there is a record of c. 8 yr over which pH measurements have been made at varying intervals (Fig. 9). This record shows that the pH of the Main lake was consistently between 4.5 and 5 until the latest phase of rehabilitation of the Wangaloa site. Since then, the pH has generally risen to c. 6, albeit with monthly fluctuations. It is possible that the lake had similar fluctuations before the rehabilitation, but records were not taken monthly at that time.

Boron content is another parameter of environmental concern at the Wangaloa site (Craw et al. 2006). Boron has been measured in the main lake only since 2001, but, like pH, there appears to have been a significant change since the onset of the rehabilitation activity (Fig. 9). Since 2003, there has been a general decline in B content of the lake water from >1 mg/litre to c. 0.7 mg/litre. This is only a small decline, but it appears to be consistent despite some fluctuations on a monthly scale (Fig. 9).



New Zealand Journal of Geology and Geophysics, 2007, Vol. 50

Regular recording of waste rock groundwater compositions was restricted to a single year, late 2003 to late 2004. There were detectable monthly variations in some boreholes in that monitoring period (Fig. 10). The pH variations in the boreholes over this year show some rises and falls that are generally consistent in all the boreholes at the same time (Fig. 10A). The pattern of variations in borehole 5, in the eastern overburden (Fig. 2), is not as closely aligned as those in the rest of the site. The observed variations are much larger than the uncertainties associated with pH measurement. These rises and falls in groundwater pH generally correspond to the rises and falls in pH in the Main lake (surface water on waste rock) and the Top lake (background surface water) at the same time (Fig. 10B). Other water chemistry parameters show fewer similarities among boreholes than does pH (e.g., sulfate; Fig. 10C). Some boreholes show little variation in anion and cation concentrations with time (e.g., borehole 5; Fig. 10C), whereas others show wide, but largely uncorrelated, variations (e.g., boreholes 6 and 7; Fig. 10C). Ion ratios, such as sulfate/chloride ratio (Fig. 10D), are also poorly correlated among boreholes, and between boreholes and the Main lake, although some peaks and troughs are shared (Fig. 10D).

DISCUSSION

Controls on Main lake composition variations

The Main lake is the principal receptacle for surface waters and groundwaters moving through the Wangaloa site (Fig. 2). Hence, the composition of this lake, and its fluctuations, provides an indicator of average water compositions through most of the site. The lake is sufficiently small that lake water pH varies on a monthly time-scale, in tune with groundwater pH variations (Fig. 10B). The Main lake has a volume of c. 7 \times 10⁴ m³, and surface water discharge is typically between 1 and 3 litre/s (above). These estimates imply water residence time in the lake of between 9 months and 2 yr, assuming only surface water discharge. Since there was almost certainly some groundwater discharge from the lake down the lower wetland (Fig. 2), and there was an evaporative loss of water from the lake surface, this <2 yr residence time is an overestimate. Such a short residence time explains why there was no lasting effect on lake pH of the lime dosing in the early stages of site rehabilitation (Fig. 9).

The close correspondence between variations in groundwater borehole pH and Main and Top lake pH values on a monthly basis (Fig. 10A,B) suggests that these variations are a result of processes acting on a 1 month scale, well within the residence time for water in the Main lake. The most likely cause for these variations in water compositions across the whole site is rainfall differences on a monthly scale. The rises and falls of groundwater and Main lake pH

Fig. 6 Piper diagrams comparing Wangaloa water major ion compositions with typical regional waters (Fig. 1A; see Fig. 1 caption for references): S = Sutton Salt Lake (rain); P = Papakaio Formation (=Taratu Formation coal measures); T = Taieri basin (schist and greywacke gravel). A, Background surface and groundwaters at the upstream end of the Wangaloa mine site. B, Surface and groundwaters moving on and in Wangaloa waste piles. C, Wangaloa mine lakes (surface waters) adjacent to waste rock piles.

appear crudely antithetic with highs and lows of rainfall in the preceding month during 2004 (Fig. 10B). This could suggest that incoming rainwater dissolved products of oxidised pyrite, and flushed resultant acid through the groundwater system to the Main lake on a monthly time-scale. However, this relationship is not readily apparent over a longer timescale, and a wet summer of 2004/05 did not lower lake pH. Further, acid flushing by rainwater should be detectable via coeval pulses of sulfate, produced by the same acidification reactions, passing through the groundwater system to the Main lake. These coeval sulfate pulses are not apparent in the lake or groundwater compositions (Fig. 10C,D). More detailed, possibly daily, monitoring would be necessary to further resolve possible relationshps between pH and rainfall.

The sulfate/chloride ratio of water in the lake has generally decreased from c. 20 to <10 during the rehabilitation period (Fig. 10D). This suggests that there have been increasing amounts of sulfate-poor rainwater runoff to the lake, after minimal interaction with sulfur-bearing substrate. This effect may have arisen because of the high proportion of bare ground during most of the rehabilitation period. This speculation can be tested in the future, when full vegetation cover is achieved and the sulfate/chloride ratio of water in the lake is predicted to rise again.

Discharge water quality

Water from the Wangaloa mine leaves the site through the lower wetland, and the water in this wetland is dominated by Main lake discharge water (Fig. 2). Hence, site discharge water quality is closely linked to that of the Main lake. The pH of the Main lake has been generally rising, and dissolved B content has been falling, during the rehabilitation process started in 2002 (Fig. 9). Seeps of groundwater with low pH, elevated sulfate, and elevated B are entering the lower wetland below the Main lake (Fig. 8A,B) (Craw et al. 2006). The seeps are discharging this distinctive acid rock drainage water into the lower wetland along at least a 200 m front along the southern side of the wetland. The flow rates of these seeps are sufficiently low that this acid rock drainage is diluted by the Main lake water (1-3 litre/s), so the seeps currently have no detectable effect on quality of water discharging from the site. The discharge points of the seeps are obscured by swamp vegetation, and the source of these seeps is unknown. They may be derived from pyrite and coal-rich waste rock in the slopes immediately above (to the south of) the wetland (Fig. 2), similar to material in the acid slope above the upper wetland (Fig. 2, 5). However, these seeps are chemically distinct from the groundwaters intersected by borehole 5 (30 m deep) in this waste rock (Fig. 2, 8A,B). Hence, if the seeps are derived from these waste rocks, the acid rock drainage that forms the seeps must be flowing along a deeper aquifer, or aquifers, that bypass borehole 5.

An alternative source of acid rock drainage for these lower wetland seeps may be historic underground mine workings to the south of the opencast mine site (Fig. 2). Bedding in this area dips generally northeastwards or eastwards, as part of the anticlinal structure that folds the Barclay Seam in the opencast mine (Fig. 2). Hence, it is possible that mine waters are being transmitted through permeable beds from the old underground mines to discharge into the lower wetland. Several deep boreholes through waste rock and into the underlying Taratu Formation are needed to test this hypothesis.



Fig. 7 Plots of compositions of Wangaloa mine waste rock surface and groundwaters (open circles), and Main lake waters (black diamonds) compared to background surface and groundwaters (black squares). A, Sulfate versus pH. B, Sulfate versus calcium. The dashed line defines the 1:1 molar ratio of gypsum dissolution. C, Sulfate versus boron.



Fig. 8 Plots of compositions of surface waters in the lower wetland at Wangaloa mine (open circles), and surface and groundwaters feeding that wetland (Main lake = black diamonds; borehole 5 in waste rock = black squares). **A**, Sulfate versus pH. **B**, Sulfate versus boron.

New Zealand Journal of Geology and Geophysics, 2007, Vol. 50

Problems of site variability

The substrates at Wangaloa are highly variable in composition on the 1–10 m scale because of widespread disturbance during mining and early stages of rehabilitation (Fig. 4A–C). Likewise, monthly monitoring of water compositions shows abundant variations, and even this time-scale was too short to resolve the causes for Main lake pH variations (above). This study has shown that even with hundreds of samples, it is difficult to fully characterise this small site. This study was more intensive than most coal mine site investigations, where typically only small numbers of samples are collected.

Despite this site inhomogeneity, geochemical characterisation and site monitoring at Wangaloa was made easier by the presence of the Main lake. This lake receives and averages most of the surface and groundwaters on the site after these waters have interacted with the variable substrates. Water discharging from the Wangaloa site typically has the same composition as the Main lake. The low pH, high sulfate seeps into the lower wetland bypass the Main lake, but were of sufficiently low volume during our study period to have negligible effect on discharge water quality. Hence, the Main lake composition is a useful indicator of overall site environmental parameters, such as pH and boron. Without this central water sink, geochemical characterisation of the Wangaloa site would have been extremely difficult. Large coal mine sites elsewhere that do not have a water sink like the Wangaloa Main lake, or a single discharge stream, would clearly be difficult to characterise for environmental monitoring purposes.

CONCLUSIONS

The Wangaloa open cast mine site has undergone surface rehabilitation since 2002, with initial removal of exotic vegetation followed by planting of native seedlings and subsequent incursion of exotic weeds between these plants. When the site was initially devegetated in 2002, the whole site had moderately acid substrates (pH = 4.6 ± 0.8), with some patches (10–100 m²) having pH down to <2. The most acid substrates are waste rock with pyrite-bearing coal, and/or quartz conglomerate cemented with pyrite from the top of the main coal seam. The average pH of the site had not changed





Fig. 10 Variations of water composition parameters with monthly sampling period from late 2003 to late 2004. A, Variations of groundwater pH in boreholes in waste rock. B, Variations of water pH in the Top lake and Main lake. Borehole 6 data from (A) are added for comparison. Rainfall (centimetres) in the previous month is overlain with a heavy black line, on the same scale. C, Variations of sulfate concentrations in boreholes and lakes. D, Variations in sulfate/ chloride ratio in Main lake and boreholes.



significantly by 2006, when a high proportion of revegetation had occurred. Some patches of distinctly acid substrate have become less acid, and one moderately acid area has become more acid.

Background surface water and groundwater at the site is dominated by chloride ions from marine aerosols in rain, and has pH between 5 and 6. Rainwater at the site passes over the acid substrate as surface waters, and through the waste rock as groundwater, and becomes more acidic (pH typically c. 4) and dominated by sulfate ions. There is no discernible influence of underlying greywacke/schist rocks, or overlying immature schist-derived Quaternary loess, on water compositions. Water from most of the site drains to the Main lake, which then discharges from the site through a wetland. Acid rock drainage seeps discharging into the wetland from beneath waste rock downstream of the Main lake have the highest sulfate concentrations (up to 700 mg/litre) and highest B contents (up to 3 mg/litre) of groundwaters on the site. These seeps are currently being diluted by the more voluminous lake discharge water, and water leaving the site has essentially the same composition as the lake.

The pH of the Main lake was consistently 4.6-4.8 before rehabilitation began, according to the few measurements that were taken between 1998 and 2002. The lake pH has become generally higher since rehabilitation began. However, the lake pH varied on a monthly time-scale between 4.5 and 6.5 during rehabilitation in 2004-06. These monthly variations in lake pH reflect similar monthly variations in pH of groundwater in boreholes around the site, implying that the residence time for groundwater on the site is c. 1 month. In contrast, the Main lake water has residence time in the order of 1-2 yr. The sulfate/chloride ratio of water in the lake has generally decreased from c. 20 to <10 during the rehabilitation period, implying a greater proportion of sulfate-poor surface water that has had less interaction with S-bearing substrates than before rehabilitation began. This greater surface (rain) water input may be responsible for the overall improvement in water quality of the Main lake, and therefore the quality of water leaving the site, during rehabilitation.

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238