

Temporal aspects of acid mine drainage on the West Coast, South Island, New Zealand

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Abstract

Acid mine drainage (AMD) is an environmental problem arising from the exposure of sulfide (commonly pyrite) bearing rocks at the earth's surface, causing accelerated chemical weathering. This creates a discharge typically characterised by depressed pH and increased dissolved metal content. The complex interaction of pH with dissolved Fe and Al can cause secondary mineralization including, hydroxides, hydroxyl-sulfate and sulfate minerals. If these precipitate from low pH solutions containing Fe and Al, they can contribute acidity when re-dissolved. These are referred to as acid sulfates and act as temporary stores of acidity. Acid sulfates have been documented around the world in regions with pronounced wet-dry seasons, but have not been characterized on the West Coast of the South Island due to unfavourable climate conditions. Past research has revealed a lack of correlation between stream discharge rates and pH on the West Coast, characteristic of acid sulfate dissolution. The lack of research on the occurrence of these characteristics and their temporal aspects represents a large gap in research that needs addressing. To determine the role acid sulfates contribute to AMD on the West Coast, we have collected waste rock samples of different ages and depths from a West Coast coal mine. Thin sections were used to determine the mineralogy of the samples, along with XRD and scanning electron microscope methods to characterise both major mineralogy and water soluble phases from mine wastes, and ICP-AES are used to determine major element geochemistry of leachates to illustrate the role water soluble phases play in AMD. We believe the precipitation and dissolution of these minerals may determine the acid loads from mines on the West Coast and influence the success of remediation programs.

Keywords: AMD, sulfate, coal, temporal, waste.

Introduction

AMD is a well-known product from mining disturbance of iron sulfide bearing rocks, such as those found in coal deposits (Jaynes et al., 1984, Pope et al., 2006, Zhengfu et al., 2010). It is characterised by lowered pH and increased dissolved metal contents, which can negatively impact on the environment. The West Coast of the South Island of New Zealand has a long history of coal mining (Balcar and Pearce, 1996), which has left behind numerous abandoned mines producing AMD. Many authors have developed and implemented remediation schemes to combat AMD (Johnson and Hallberg, 2005, Trumm et al., 2008, Trumm, 2010, Uster et al., 2014), using the vast array of literature available to determine the pathway AMD forms (Pope et al., 2010, Davies et al., 2011).

When AMD evaporates, an increase in dissolved constituent activities occurs, eventually reaching saturation with respect to a secondary mineral, with one group being sulfates. These minerals can act as temporary stores of acidity such as Fe and Al, referred to as acid sulfates. When dissolved, these re-contribute acidity to AMD by undergoing hydrolysis, and production of H⁺. Highly soluble secondary sulfates are frequently observed in parts of the world with pronounced wet/dry seasons (Zodrow and McCandlish, 1978, Jambor et al., 2000, Hammarstrom et al., 2005). Research in New Zealand has frequently observed jarosite from both coal and gold mining (Davies et al., 2011, Kerr et al, 2014).

Dissolution of sulphate, or hydroxyl-sulphate salts, is thought to impact temporal aspects of AMD release (Davies et al 2011; Mackenzie et al 2011), however, exactly which minerals are involved is not clear. Since the lowered pH conditions created by primary pyrite oxidation will cause other phases to become unstable, it is expected that different phases will become unstable at different times, depending on their reaction rates. This in turn may change the chemistry of AMD from a mine over time.

Description of area and coal bearing lithologies

Two coal measures are frequently disturbed by coal mining on the West Coast, resulting in different discharge chemistries – the Paparoa Coal Measures (PCM) and the Brunner Coal Measures (BCM) (Nathan, 1973). The PCM are non-acid forming, but still have elevated dissolved metal loads, commonly referred to as neutral mine drainage (NMD), while the BCM contain pyrite, and have low pH, along with high dissolved Fe and SO_4^{2-} (Pope et al., 2010).

At the mine where our samples were collected, BCM make up the coal measures, therefore AMD occurs, similar to many coal mines in the area. Field work at the site revealed mine waste piles contain three main lithologies – a pyrite bearing feldspathic white sandstone, a mudstone, and a rare blue/green volcanic sandstone. The geochemistry of the phases in these rocks will exert important controls on which secondary minerals will form, but gives no indication as to when this happens. The aim of this paper is to identify the changes to mine wastes (overburden of no economic value) over time. To collect samples, a digger was used to trench the side of waste rock piles that were 2, 5 and 12-15 year old.

Mineralogy of lithologies

Samples of each lithology were collected from the sites and standard thin sections made. The mineralogy of each sample was identified using a polarizing microscope and point counting performed to determine the percentage mineralogy of the mudstone and white sandstone. Point counting was not appropriate for the volcanic sandstone due to its highly altered nature. Another polished thin section of each rock was created and analysed using a scanning electron microscope (SEM). Energy dispersive x-ray (EDX) spectroscopy was used to determine the chemistry of phases present in each rock type.

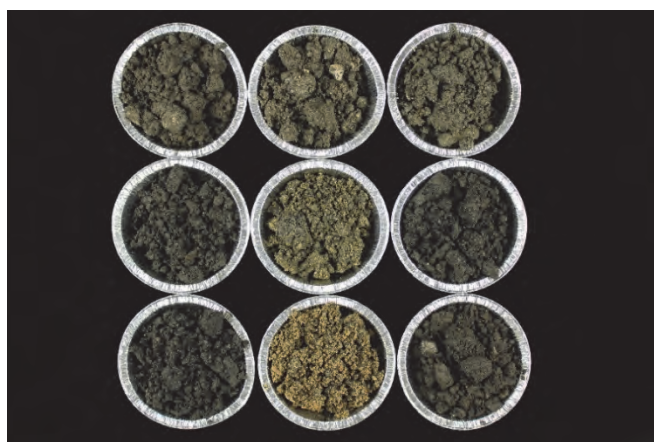


Figure 1. Subsamples collected of different aged mine waste – top row (T_1) = 2 year, middle row (T_2) = 5 year, and bottom (T_3) = 12-15 year old mine wastes, with different horizons ($S_{1,2,3}$) selected based on colour.

White sandstone

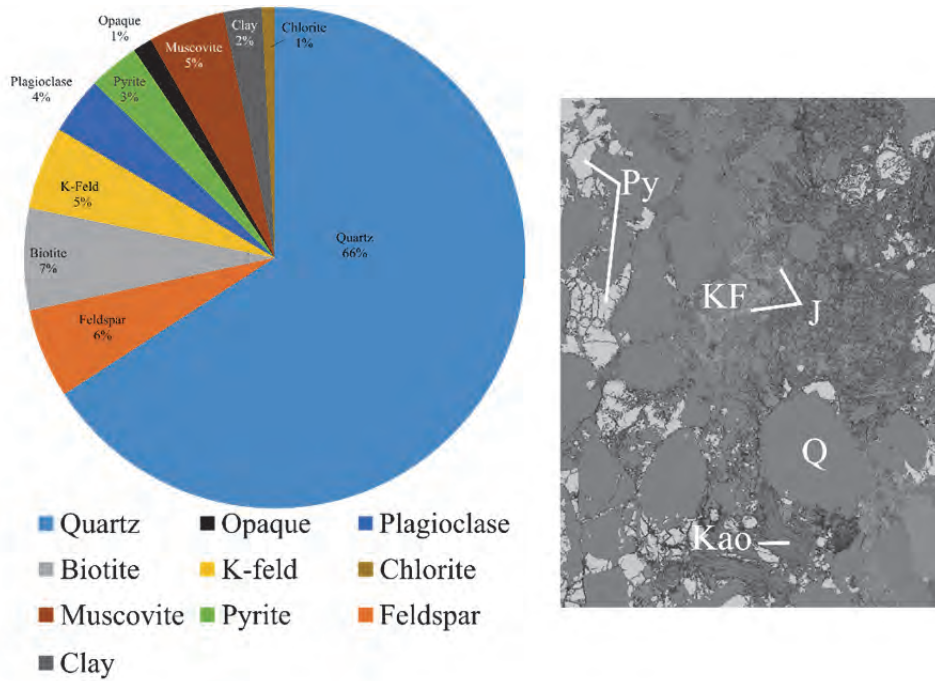


Figure 2. White sandstone mineralogy and SEM image illustrating the occurrence of Jarosite (Q – Quartz, KF – K-feldspar, Kao – Kaolinite, Py – Pyrite, J – Jarosite).

Mudstone

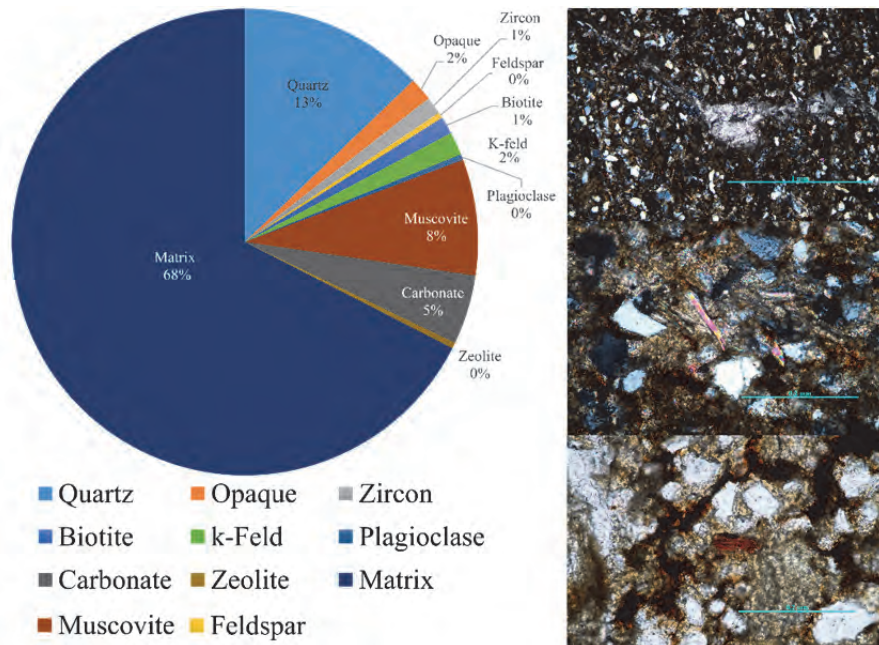


Figure 3. Mudstone mineralogy and images showing the extremely fine grained nature. Top – carbonates (1mm scale), middle – muscovite (0.2 mm scale), bottom – biotite (0.2 mm scale)

Volcanic sandstone

The volcanic sandstone represents an anomaly in the general coal measures seen by most on the West Coast (Flores and Sykes, 1996), instead representing altered redeposited volcanics from the Greenland Group (Laird, 1972). Point counting methods for this sample were ineffective due to its heavily altered nature. The sample overall is heterolithic with some grains displaying trachytic textures, while others being completely replaced by silica. Carbonates occur throughout the sample, with EDX spot analyses revealing dolomites and Mn-rich ankerites present. A green micaceous mineral was also present (Fig. 4), interpreted to be muscovite + smectite. Well rounded single-crystal quartz grains and altered feldspar crystals infrequently occur, with the quartz grains similar to those found in the white sandstone.

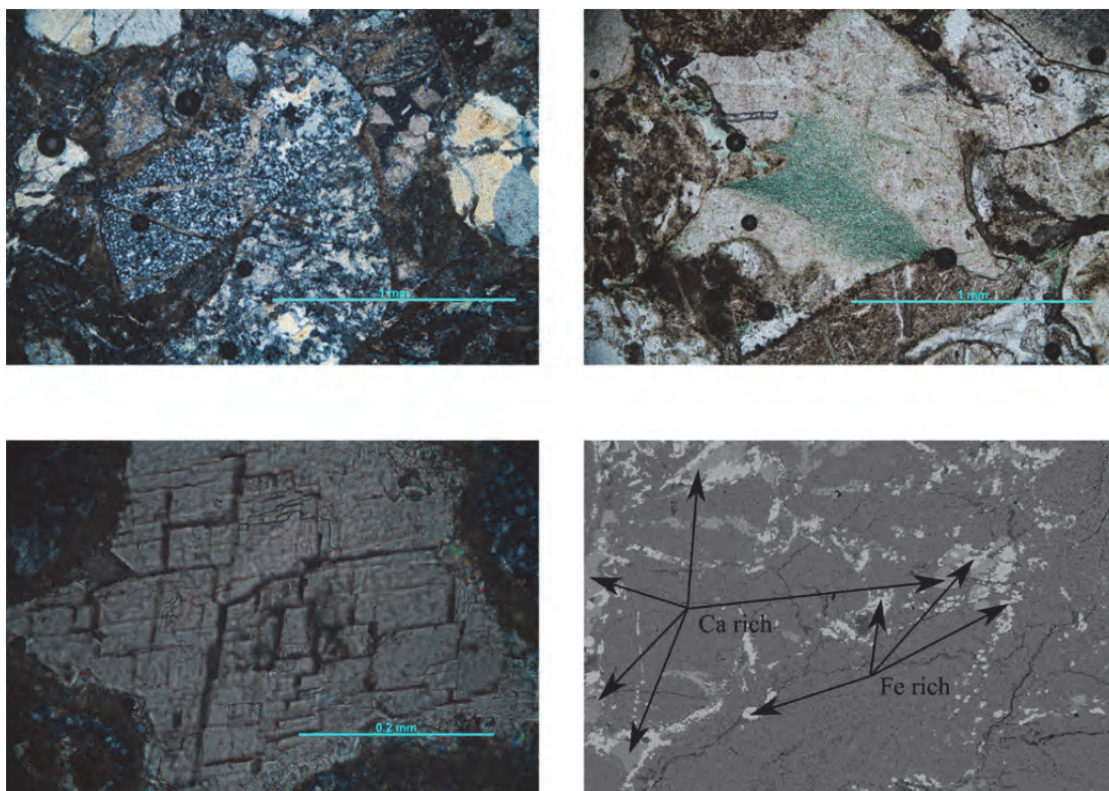


Figure 4. Volcanic sandstone images. Top left – A typical grain replaced by silica and clays, cross cut by high interference coloured carbonates. Top right – green micaceous minerals occur sporadically, possibly smectite + muscovite. Bottom left – sparry carbonate grains filling space between grains is frequently observed. Bottom right – EDX image showing the two different carbonates in the sample, a CaMg carbonate (dolomite) and an FeMgCaMn carbonate (ankerite).

XRD

Samples from different horizons within the mine waste pile were collected and bagged in sterile and unreactive packaging. These were stored at low temperatures to limit further oxidation of pyrite. A subsample of this was collected, dried and powdered using a steel ring mill, then packed into powder disks. XRD patterns were taken of these to identify any changes in mineralogy over time and depth, along with any secondary minerals present. During sample storage, a hole developed in the side of one sample bag, allowing moist air to circulate around that area for a period of weeks. This caused an efflorescent bloom of white/orange crystals to occur. These were collected and immediately analysed - without

crushing - using XRD to prevent decomposition of hydration states (Kruszewski, 2013). The integrity of the sample was kept as the area of contact with the atmosphere was minimal.

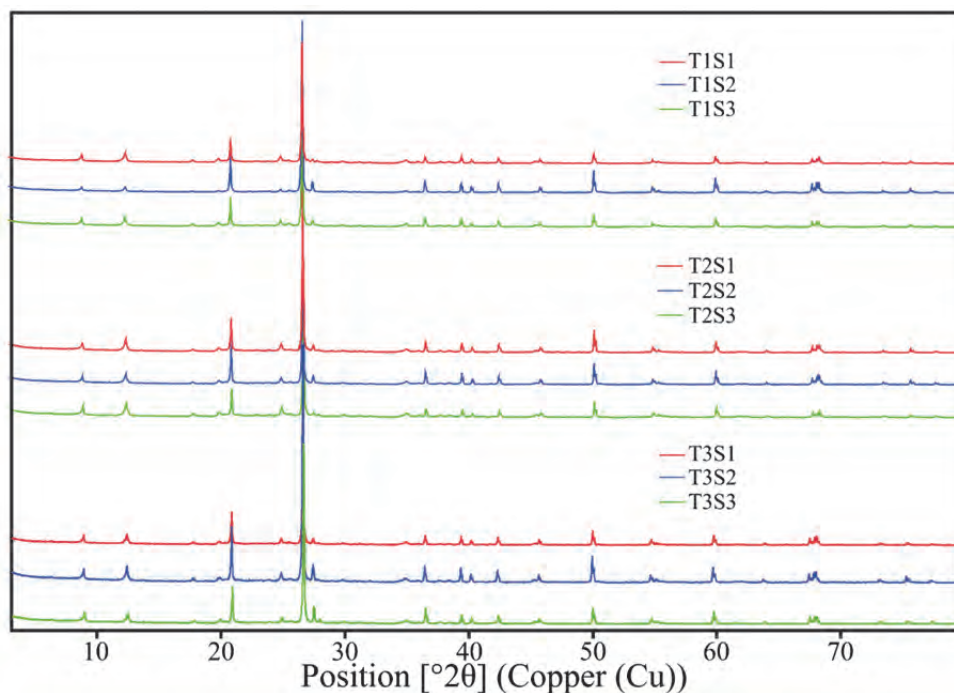


Figure 5. XRD traces of all subsamples, all producing identical patterns.

XRD patterns (Fig. 5) for the sampled horizons are remarkably similar. The rocks were presumed to be a mix of all three lithologies present at the site, but the similarities between the XRD traces renders them ineffective at identifying changes in mineralogy, or the precipitation of secondary phases from different horizons.

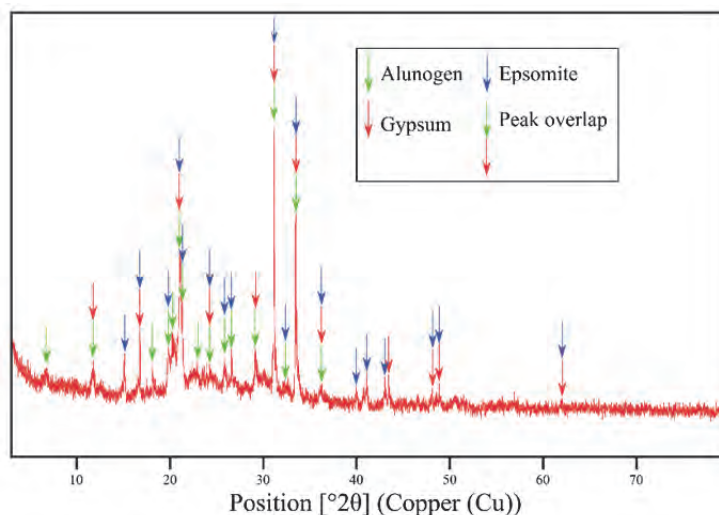


Figure 6. XRD trace of spontaneously generated efflorescence from samples with phases identified.

Water soluble geochemistry

A 100g subsample from each horizon was collected and dried at 65° overnight. 100 mL of distilled water was added to this, stirred for 5 minutes then left to sit for a further 15 minutes

to separate out suspended clays. The contribution of dissolved constituents from the distilled water was presumed to be negligible. The pH of the resulting liquor was measured, then filtered through a 0.45 micron filter, acidified to pH ~2 and analysed using ICP-MS for the following dissolved constituents; Al, Fe, Ca, Mn, Ni, Zn Mg, Na, K and SO_4^{2-} .

Table 1. Geochemistry of liquors collected from different horizons (in g/m^3 water).

Sample	pH	Al	Fe	Mn	Ni	Zn	SO_4^{2-}	Ca	Mg	K	Na
T ₁ S ₁	4.07	0.81	0.4	0.063	0.019	0.107	21	5.1	1.8	2.3	2.2
T ₁ S ₂	3.33	1.33	0.87	0.139	0.038	0.172	39	1.89	2.9	2.2	1.25
T ₁ S ₃	3.15	4.0	0.78	0.159	0.064	0.29	94	5.2	3.4	3.8	0.76
T ₂ S ₁	2.97	18.2	19.8	2.6	0.23	2.3	500	58	27	1.82	0.7
T ₂ S ₂	3.15	7.7	6.1	3.1	0.32	2.5	390	71	20	2.9	0.81
T ₂ S ₃	3.48	2.2	1.72	1.96	0.21	1.52	189	25	14.4	4.8	0.61
T ₃ S ₁	3.85	0.138	0.03	1.29	0.22	0.75	162	31	17.9	5.7	0.81
T ₃ S ₂	3.93	2.6	4.2	1.73	0.192	0.98	260	35	22	2.6	0.58
T ₃ S ₃	3.45	0.56	0.41	1.09	0.115	0.69	110	14.5	10.8	4.6	0.59

Dissolved metal and sulfate concentrations show pH levels reasonably constant across all samples (within one order of magnitude) except for T₂S₁, which showed slightly lower pH. Most metal and SO_4^{2-} changed over time as well, with younger mine wastes producing much higher sulfate levels, along with increased Mg, Mn, Ni and Ca contents.

Mineral saturation

To determine the saturation state of water soluble phases possible from the AMD from different horizons, PHREEQC (Parkhurst and Appelo, 1999), using the WATEQ4F database supplied (Ball and Nordstrom, 1991) was used. Several assumptions needed to be made with respect to the oxidation state of iron. During field work, the mine wastes were dry and therefore recording the oxidation state was not possible. Due to the presence of ferric jarosite observed as a coating on the white sandstone, coupled with the knowledge that AMD with pH between 3–4, a pe value (-log electron activity) of around 12 was used to control the oxidation state of Fe in the model (Langmuir, 1997). Schwertmannite, a partly crystalline sulfate, was not included in the WATEQ4F database, however, it is stable with respect to all water samples collected (see Fig. 7).

Table 2. A selection of saturation indices for minerals known to occur from AMD adapted from PHREEQC. The more red the colour, the more under saturated the phase is.

Phase	Formula	Saturation indices								
		T ₁ S ₁	T ₁ S ₂	T ₁ S ₃	T ₂ S ₁	T ₂ S ₂	T ₂ S ₃	T ₃ S ₁	T ₃ S ₂	T ₃ S ₃
Al(OH) ₃ ^a	Al(OH) ₃	-3.43	-5.51	-5.77	-6.11	-5.89	-5.24	-5.3	-3.92	-5.78
Diaspore	AlOOH	0.49	-1.59	-1.84	-2.19	-1.97	-1.32	-1.38	0.00	-1.86
Epsomite ^b	MgSO ₄ ·7H ₂ O	-5.82	-5.40	-5.05	-3.78	-3.94	-4.23	-4.18	-3.96	-4.49
Fe(OH) ₃ ^a	Fe(OH) ₃	-0.44	-1.36	-1.89	-1.24	-1.22	-0.87	-1.93	0.32	-1.37
Goslarite ^b	ZnSO ₄ ·7H ₂ O	-7.65	-7.24	-6.73	-5.46	-5.46	-5.81	-6.17	-5.93	-6.30
Gypsum	CaSO ₄ ·2H ₂ O	-3.14	-3.36	-2.64	-1.21	-1.15	-1.75	-1.71	-1.53	-2.13
Jarosite	KFe ₃ (SO ₄) ₂ (OH) ₆	-1.35	-1.48	-1.67	1.78	1.18	1.03	-3.30	3.20	-0.76
Jurbanite	AlOHSO ₄	-1.29	-1.66	-1.23	-0.66	-0.86	-1.10	-1.95	-0.56	-1.76
Manganite	MnOOH	-7.14	-9.04	-9.57	-9.05	-8.41	-7.55	-6.61	-6.28	-7.85
Melanterite ^c	FeSO ₄ ·7H ₂ O	-8.28	-6.74	-6.42	-4.67	-5.25	-6.11	-8.34	-6.16	-6.71
Morenosite ^b	NiSO ₄ ·7H ₂ O	-7.96	-7.44	-6.94	-6.00	-5.89	-6.22	-6.24	-6.18	-6.62
Pyrolusite	MnO ₂	-7.11	-9.75	-10.4	-10.1	-9.30	-8.11	-6.80	-6.39	-8.44

a = amorphous phase. b = epsomite solid solution members. c = readily substitutes Mn for Fe

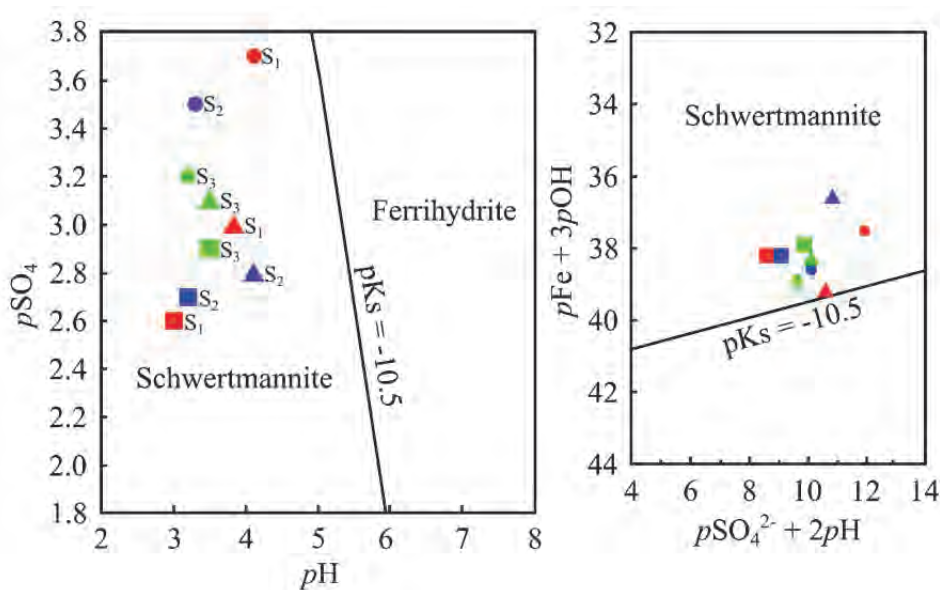


Figure 7. Stability data for Schwertmannite (redrawn from Yu et al., 1999), illustrating it is stable with the water geochemistry at the site.

Discussion

The mineralogy (and geochemistry therein) of the lithologies at the mine site were the ultimate control on AMD composition. The white sandstone was the only sample to contain pyrite, therefore was the primary cause of AMD. The low percentage of minerals in the mudstone, coupled with their size (generally less than 0.1mm) leads the interpretation that it was not a major source of dissolvable constituents. Both the dolomite and Mn-ankerite represent a source of alkalinity, which will help mitigate the severity of AMD at the site. The small amount of Fe present in the ankerite is insignificant to the neutralizing capacity of the carbonates

As seen in Fig. 5, XRD analyses of mine wastes shows little to no visible changes in mineralogy, in contradiction with both the observed changes in mine wastes and the water soluble geochemical information. Two explanations from this can be made;

- Change in phases are amorphous. Fig. 1 shows a definite change in colour between the samples, therefore a change in phases must be expected. However, XRD uses constructive interference from crystal lattices to produce peaks at certain angles. If the phase change is not crystalline (e.g. an amorphous compound), it cannot produce any peaks. The most likely phase responsible for the colour change is amorphous $\text{Fe}(\text{OH})_3$
- Changes in mineralogy are too small. XRD is effective when the phases present occur in a high enough percentage to produce a peak. The water soluble phase proportion of the horizons may be too small to produce a decent peak.

Overall, a recommendation against the use of using XRD to determine the secondary mineral changes in mine waste must be made, as it is not sensitive enough to detect low concentration phases.

Jarosite group minerals are frequently observed secondary acid sulfates in New Zealand. Although reasonably insoluble, jarosite is an important source of acidity from AMD. Jarosite precipitates under oxic low pH/high SO_4^{2-} conditions (Murad and Jojík, 2004, Smith et al.,

2006), similar to those observed at the mine site. Although the solubility of jarosite at low pH is minimal (Kerr et al., 2014), once dissolved, it will reconstitute either Fe (jarosite) or Al (alunite) to the discharge, and when hydrolysed, will reconstitute H^+ . Jarosite is also an important source of adsorbed heavy metals (Dutrizac, 1983). Jarosite frequently occurs as a cement in the white sandstone, as well as a coating on clast surfaces. This is the most readily identifiable soluble sulfate at the site.

The observations from the water soluble geochemistry can be coupled with the dissolution of different phases at different ages of the mine waste, as illustrated in Table 3.

Table 3. Explanation for changes in water soluble geochemistry at certain times. Pyrite is oxidizing at each stage therefore is not included in the table (SS = solid solution).

Age	Observations	Mineral dissolution	Secondary minerals observed	Secondary sulfates possible (Model)
2 years	High Mg/Ni/Zn, high SO_4^{2-} , high Mn, low Fe and Al	Dissolution of carbonates in heterolithic volcanic sandstone.	Epsomite, jarosite, gypsum, alunogen	Epsomite SS, gypsum, jarosite SS, jurbanite
5 years	All concentrations high except Na and K	Continued carbonate dissolution, mica dissolution begins.		Epsomite SS, gypsum, jurbanite, jarosite SS
12-15 years	Low Mg/Ni/Zn, high Fe and Al, low Ca	Carbonates all dissolved, mobilization of Al and small amounts of Mg and Fe from aluminosilicates		Jurbanite/alunogen, jarosite SS,

Fig. 6 shows the presence of alunogen from the mine wastes represents an anomaly. Unfortunately the WATEQ4F database does not include solubility information for alunogen. Its presence can be explained via evaporative processes. Nordstrom (1982) determined alunogen precipitation occurs at (unnaturally) low pH or high Al activity, as shown in Fig. 8. Evaporation of AMD (dashed circle) will increase Al activity, but not pH as both H^+ and OH^- will change, moving the water into the field of alunogen stability. When a rainfall event occurs, alunogen will be dissolved, and the pH buffered for a time while gibbsite ($Al(OH)_3$) is precipitated. Overall, this illustrates how evaporation can change the nature of phase stability and how alunogen can form from mine wastes with low Al activity and not extremely low pH.

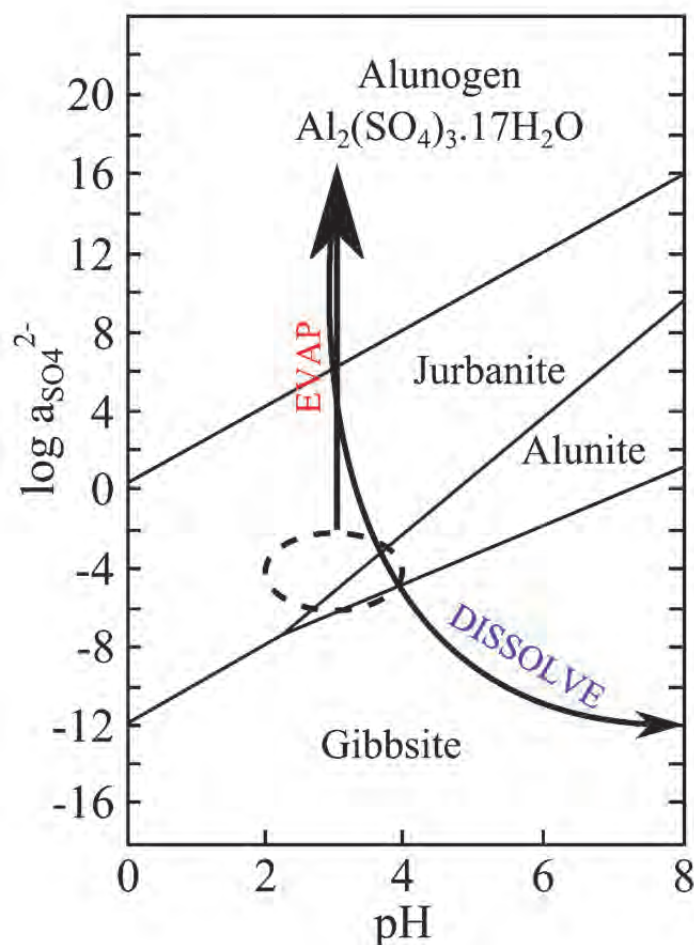


Figure 8. Stabilities of minerals in the $\text{Al}_2\text{O}_3\text{-SO}_3\text{-H}_2\text{O}$ system with excess K (redrawn from Nordstrom, 1982) showing the path of evaporation, causing alunogen to precipitate, then dissolution to form gibbsite

Conclusions

An active coal mine on the West Coast of the South Island, New Zealand was investigated to identify the temporal aspects of AMD composition, and to determine the role acid sulfates have with respect to time. Site rock lithologies were a white sandstone, a mudstone, and a volcanic sandstone. The white sandstone was the main source for primary AMD production due to its pyrite-bearing nature. A suite of aluminosilicates present in the feldspathic arenite can break down over long water-rock contact times, releasing acidity in the form of Al^{3+} under low pH. The volcanic sandstone represents the only source of neutralizing capacity at the site. XRD was not useful in determining the changes to mine wastes over time, possibly due to precipitation of amorphous phases, and small volumes of secondary sulfate precipitation. Saturation indices of phases reveal most mine wastes are occasionally oversaturated with respect to jarosite, representing a significant source of Fe-sulfate acidity. All samples were slightly under saturated with respect to jurbanite, which is not observed in mine wastes which precipitated secondary minerals while stored, as it was supplanted by alunogen due to increase Al activity from evaporation. A recommendation can be made against disturbance of coal mine waste rocks after 5 years due to the observed precipitation of jarosite and possible production of alunogen. After 12-15 years, concentrations drop and mineral saturation lowers, possibly allowing mine waste disturbance if needed.

Acknowledgements

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