Fingerprinting Acid Mine Drainage in Wet-dry Tropical Climates using Stable Isotope Approach: An Example from Queensland, Australia

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Abstract

Mine water fingerprinting and sulfate stable isotope geochemistry can provide strong support for acid mine drainage (AMD) management. This study was conducted at a mine site in a semi-arid tropical climate of North East Queensland region using various techniques including water geochemical fingerprinting and stable isotope geochemistry of waters and sulfate-sulfur. It aims to investigate the connectivity between surface and ground water, and between mine water and the local stream system. Mine water geochemical fingerprinting techniques have indicated that AMD-affected waters were locally found near waste rock dumps, heap leach stockpiles and mill rejects. The effects of AMD on creek waters were limited by neutralizing capacity. Stable isotope compositions of waters revealed that mine waters showed some degree of evaporation, particularly tailing sumps in dry season, compared to rainwater, stream and ground waters. Stable isotope geochemistry of sulfate-sulfur has identified two distinct sources of sulfate including *i*) upstream background sulfate and *ii*) mine-related sulfate from sulfide oxidation at the site that contributed to creeks flowing across the site. The characterization of water connectivity and sources of sulfate could therefore, give important information on localities of AMD occurrences and relative effects on natural stream systems for management purposes.

Key words: mine water, fingerprinting, stable isotope.

1. Introduction

At locations with wet - dry tropical climates, the distinct pattern of a long dry season followed by high rainfall events and reactive geochemistry may trigger AMD discharge from mine sites to natural systems during intensive rainfall periods. That causes mixing of products from mine-related sulfide oxidation with background water components. It is therefore important to assess the relative effects and contributions of sulfate mineral dissolution and sulfide oxidation on the downstream aquatic ecosystem affected by the mine.

Tracer techniques are considered as useful tools for defining parameters of water bodies (Wolkersdorfer 2002) and to understand the mechanisms of groundwater flow and transport (Gelhar *et al.* 1992; Holmbeck-Pelham *et al.* 2000). The most commonly used tracer techniques for assessing mine water environments are based on properties of major and trace element chemistry (e.g. Piper and Ficklin diagrams), isotopic analyses and geochemical models (Bird and Mahoney 2000). In near surface environments, isotopic signatures of surface and groundwater are essentially conserved and will only change as the result of mixing with waters of different isotopic signatures (Bird and Mahoney 2000). Thus, isotope signatures may reflect the geochemical environment through which waters have moved. While stable isotopes of water are routinely used in hydrology, hydrogeology and geothermal studies, their application in mine waters generally, and AMD effluents specifically, is relatively scarce (Allen and Voormeij 2002; Spangenberg *et al.* 2007). Very few studies on the application of stable isotope geochemistry of mine waters have been reported (Pellicori *et al.* 2005; Spangenberg *et al.* 2007) despite the potential of isotopes as very useful tools for understanding mine drainage (Ghomshei and Allen 2000b; Allen and Voormeij 2002).

This study seeks to identify and understand water connectivity and sulfate sources in the mine drainage and stream systems at a North-East Queensland wet-dry tropical location using a combination of water chemical fingerprinting tools and stable isotope geochemistry.

2. Site Description

The Mt Leyshon Gold Mine is chosen as a case study, and is located approximately 24 km south of Charters Towers, Queensland (Orr and Orr 2004; Edraki *et al.* 2006a; Edraki *et al.* 2006b). The mine is entirely within the sub-tropics and semi-arid region, and thus, has a distinct dry savanna climate (Orr and Orr 2004; Edraki *et al.* 2006a). The average annual precipitation is 660 mm with intense rainfall in summer (December to April) associated with cyclones, and a long dry season from May to October (Orr and Orr 2004; Edraki *et al.* 2006a).

The ore deposits at Mt Leyshon mine were formed within granite and metasediments (Orr and Orr 2004; Edraki *et al.* 2006a). The ore body is hosted by units of the Mt Leyshon Intrusive and Breccia Complex within the northern trending corridor of Permo-Carboniferous subvolcanic rocks (Orr and Orr 2004; Edraki *et al.* 2006a; WMC 2009). The main sulfides within the ore body and waste rocks are pyrite, chalcopyrite, galena and sphalerite (Harries 1997; Orr and Orr 2004; Edraki *et al.* 2006a). Sulfide contents (mostly as pyrite) are up to 5 % at Mount Leyshon itself and generally less than 2 % in the surrounding areas (Scott 1992). The isotopic composition of alunite and sulfide minerals at Mount Leyshon has been analysed and reported by Bird *et al.* (1989). $\delta^{34}S$ values of alunite minerals (3 samples) have a range of +8.0 ‰ to +9.8 ‰ while those of sulfide minerals (22 samples) range from +4.9 ‰ to +8.8 ‰ (Bird *et al.* 1989).

Mining at Mt. Leyshon produced gold, silver and copper from a large open-pit. It commenced operations in 1986 initially as heap leach operation of oxide ore. The processing of primary un-oxidised ores via carbon in pulp started in 1989. Mining ceased due to depletion of the ore body in 2001 and rehabilitation has been undertaken since 2002 by Newmont Asia Pacific (WMC 2009). During mine life, approximately 55 Mt of ore were processed and total annual

ore production reached a maximum of 5.5 Mt (Harries 1997). The Mt Leyshon mine site can be divided into several major mining-influenced areas (Fig. 1). These include the open pit, three major tailings storage facilities, three waste rock storage areas, mill rejects and heap leach stockpiles. The tailings storage facilities include the Old Northern Tailings Dam (ONTD), the New Northern Tailings Dam (NNTD) and the Southern Tailings Dam (STD) covering areas of 57 ha, 55 ha and 100 ha of mine lands, respectively (Edraki *et al.* 2006a). Tailings have potential to produce acid drainage due to their primarily sulfidic ore origin with the majority having net acid producing potential (NAPP) values > 20 kg H_2SO_4/t (WMC 2009). Waste rock facilities include the Southern Waste Rock Dump (SWRD), the Eastern Waste Rock Dump (EWRD) and the Northern Waste Rock Dump (NWRD). Waste rocks are identified as potentially acid forming (PAF) materials with NAPP values > 20 kg H_2SO_4/t to a maximum of 72 kg H_2SO_4/t (WMC 2009). The mill rejects contain up to 3.8 % sulfur as sulfide minerals and are classified as PAF materials (Edraki *et al.* 2006a). The majority of heap leach materials are classified as PAF (WMC 2009). The main waste rock facilities are covered by non-acid forming (NAF) materials including compacted benign heap leach and NAF porphyry materials. The porphyry cover layers usually have a minimum thickness of 800 mm on flat surfaces and 1000 mm on side slopes. Tailings storage facilities have also been covered by run of mine (ROM) waste rock consisting of NAF sub-grade breccia and by NAF porphyry rocks similar to those used for waste rock cover (WMC 2009). All facilities are finally capped by a 300 mm thick fertilized and germinated topsoil layer (WMC 2009).

Acid mine drainage occurrences have been reported at the seepage dams such as the Upper Plum Tree dam (UPT) at the base of the Mill Rejects stockpile, the Mt. Mawe Dam (MMD) of the Eastern Waste Rock Dump, and the Supergene Dam (SGD) of the Heap Leach stockpiles (Fig. 1) (Edraki *et al.* 2006a). The pH levels of these seepage dams range from 3.07-3.92. Sulfate and dissolved metals (copper, lead, zinc, cadmium, nickel, magnesium, iron, manganese, uranium, selenium and arsenic) are also present at elevated concentrations. The MMD has the highest sulfate concentrations, with an average of 15,900 mg/L, and the highest median concentrations of cadmium, copper, lead, zinc and arsenic. In contrast, the former Raw Water Dam (RWD) and Mt. Hope Extension Dam (MHED) (Fig. 1) generally have an acceptable range of pH 6.5 – 9. In general, the seepage dams have exhibited relatively constant sulfate concentrations (approximately 4,000 mg/L). Seepages and runoff from the Heap Leach area at the SGD (Fig. 1) are enriched in metals and sulfate, particularly cadmium and copper (Edraki *et al.* 2006a). Tailings sumps are characterised by high alkalinities and elevated sodium and chloride concentrations. They generally have high sulfate (3650-4230 mg/L), and compared to other dams, higher strontium, molybdenum and arsenic concentrations and neutral to alkaline pH of 6.5-9 (Edraki *et al.* 2006a). This composition is primarily due to the alkaline chemicals used in the mineral processing. Waters from seepage sumps are continuously pumped back to larger dams and then to the large pit or to the Raw Water Dam (RWD) (Fig. 1) in order to contain contaminated seepages within the mine lease region and to avoid mine water discharge to the natural creek systems.

3. Methodology

Surface and groundwater samples were collected at representative sites from the waste storage facilities, and along the trajectory of the stream systems related to the mine (Fig. 1). These include surface samples from water dams and seepage sumps; nine groundwater samples from bores around mine waste facilities and creeks; one seepage sample from EWRD; one rainwater sample collected in March, 2011 at the Mt Leyshon mine site; and downstream and upstream samples of the mine lease at Puddler and Clarke creeks. Groundwaters were collected twice during the dry (June, 2010) and the wet (March, 2011) seasons. Tailings sump samples were collected in September, 2011. The rest of the samples were collected in the wet season from January to March, 2011.

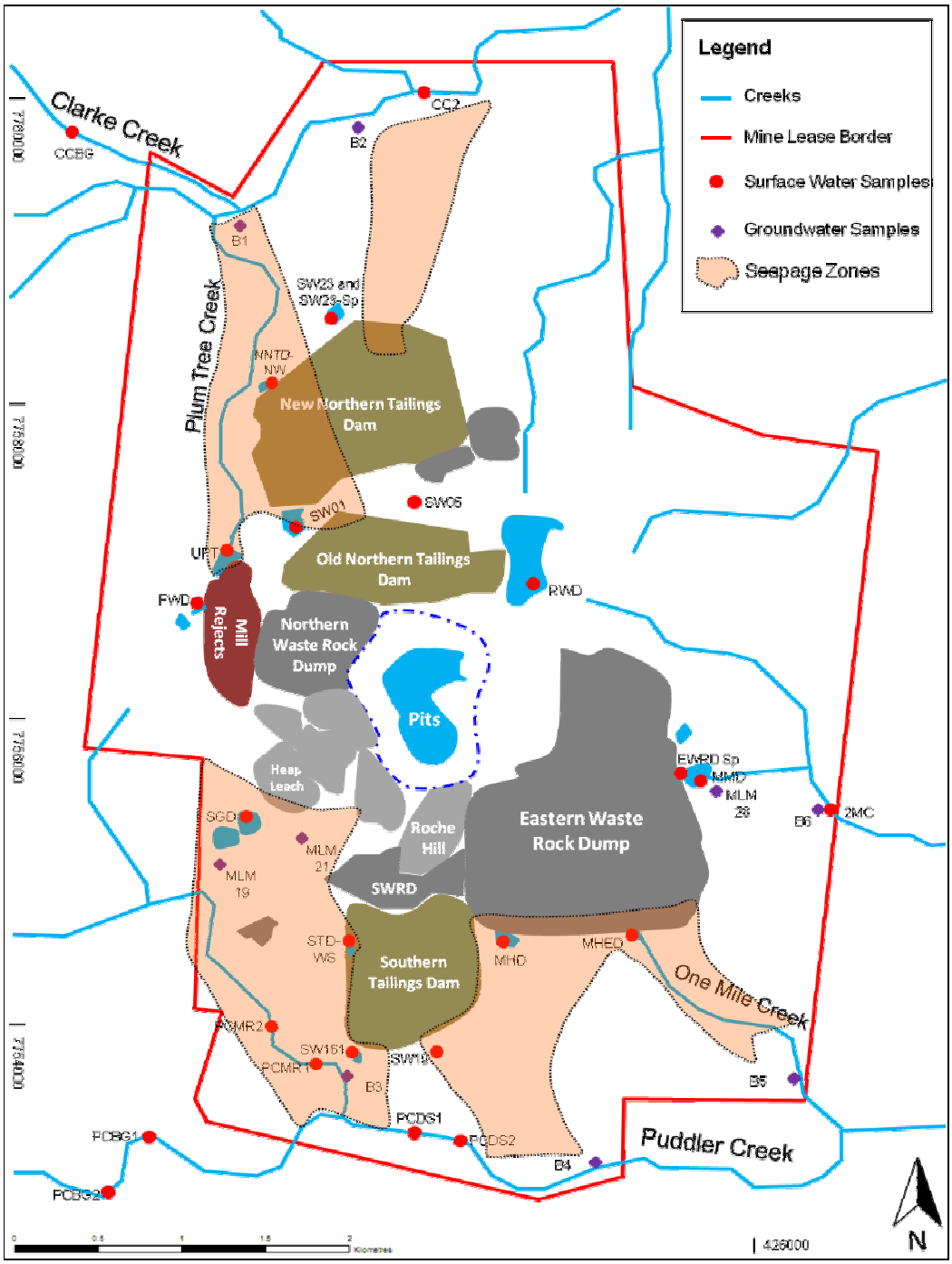

Figure 1. Site plan showing sampling locations. The extent of the seepage zones are from Klohn Crippen Consultants (1991)

Water temperature, pH, electrical conductivity (EC) and redox potential (Eh) were measured in the field using WP-81 TPS pH-EC meter (TPS Pty. Ltd., Brisbane). Eh, pH and EC meters were calibrated to Zobell standard solutions, pH 4 and pH 7 buffer solutions, and 2.67 mS/cm standard solutions, respectively. Samples for dissolved metals, Fe(II), and sulfide were field-filtered using Millex-MF Millipore 0.45 µm mixed cellulose esters membrane filters by syringe. Metals (filtered and total), Fe(II) and sulfide were preserved by adding concentrated HNO_3, HCl and zinc acetate, respectively, following the standard methods of Australian Laboratory Services, Brisbane (ALS). Samples for ^{2}H and ^{18}O in water were field-filtered into

McCartney 28 mL glass screw cap jars, were tightly sealed and kept in cool, dark conditions to ensure no air intrusion and evaporation. Water samples for sulfur isotope analysis were collected at selected sites including upstream surface, shallow groundwater and surface waters. The procedures of Fitzhugh *et al.* (2003) and Révész and Qi (2006) were followed using an anion exchange AG®1-X8 resin, 50-100 mesh in chloride form (Bio-Rad Laboratories Inc, California) to concentrate sulfate in water samples. δ^2H and $\delta^{18}O$ were analysed using a Dual Inlet Stable Isotope Ratio Mass Spectrometer with a multi-preparation bench attached (Isoprime Ltd., Manchester). δ^2H and $\delta^{18}O$ were reported in ‰ relative to Vienna Standard Mean Ocean Water (VSMOW). Analytical uncertainties for δ^2H and $\delta^{18}O$ were better than ± 2 ‰, and ± 0.1 ‰ (1 Standard Deviation), relatively (SIGL 2008). Sulfur isotope analysis was conducted in duplicate using a Continuous Flow Stable Isotope Mass Spectrometer coupled in line with a Carlo Erba NA1500 elemental analyser (Isoprime Ltd., Manchester). The $\delta^{34}S$ was reported in ‰ relative to Vienna Canyon Diablo Troilite (VCDT) with analytical uncertainties better than ± 0.2 ‰ (SIGL 2008). Alkalinity/acidity, total and dissolved metals were analysed by ALS with National Association of Testing Authorities (NATA) ISO/IEC 17025 laboratory accreditation using several quality control techniques including laboratory duplicate, method blank, laboratory control spike, and matrix spike.

4. Results and Discussion

4.1. Water chemistry

The major ion compositions of water samples are shown in Figure 2. AMD-affected waters are characterised by using Ficklin diagram (Fig. 3). Creek waters including both upstream background and inside the mine lease boundary had very similar chemical compositions with neutral to alkaline pH (7.72 to 8.49), zero acidities and low concentrations of metals. Sulfate was the only significant chemical feature with constant low concentrations in background samples (15 - 16 mg/L) and relatively high concentrations inside the mine lease border (range 247 to 744 mg/L).

The Piper diagram (Fig. 2) showed the similarity in chemical compositions of several surface and ground waters surrounding the Eastern Waste Rock Dump including MMD, MHD, MHED, MLM28 and the seepage (EWRD-Sp). These waters had distinctly high proportions of magnesium and sulfate. The samples collected around the Mill Rejects and Heap Leach Stockpiles including MLM21, MLM19, SGD (Fig. 1) and tailings dams (SW01, SW05, SW19, SW23, SW23-Sp, SW161, NNTD-NW, STD-WS) had similar major ion compositions which might indicate a similar dominant mineralogical source. Groundwater samples B1 and B3 collected near Clarke and Puddler creeks (Fig. 1) had relatively similar major ion compositions to the samples from Mill Rejects and Heap Leach Stockpiles. This might indicate the movement of seepage-affected groundwater southward to Puddler creek and northward to Clarke creek (Fig. 1).

The Ficklin diagram (Fig. 3) indicated that most creek waters (Puddler, Clarke and Two Mile creeks) and nearby groundwaters were classified as near-neutral and high- (range 1-100 mg/L of metals) or low-metal (less than 1 mg/L of metals). Most sump waters near the Eastern Waste Rock Dump, Heap Leach and Mill Rejects areas were classified as acidic and having "high hydrolysable metal" (UPT, MHD) or "extreme hydrolysable metal" (EWRD-Sp, SGD and MMD) concentrations. Groundwaters near the Heap Leach stockpiles (MLM21) and tailings seepage (SW23-Sp) were also classified as acidic and high-metal. This showed that the Eastern Waste Rock Dump, Heap Leach stockpiles and Mill Rejects produced the most acidic and metal-rich seepages. Other samples collected near various mine waste facilities (RWD, FWD, MHED, MLM19 and MLM28) had much lower metal concentrations and higher pH.

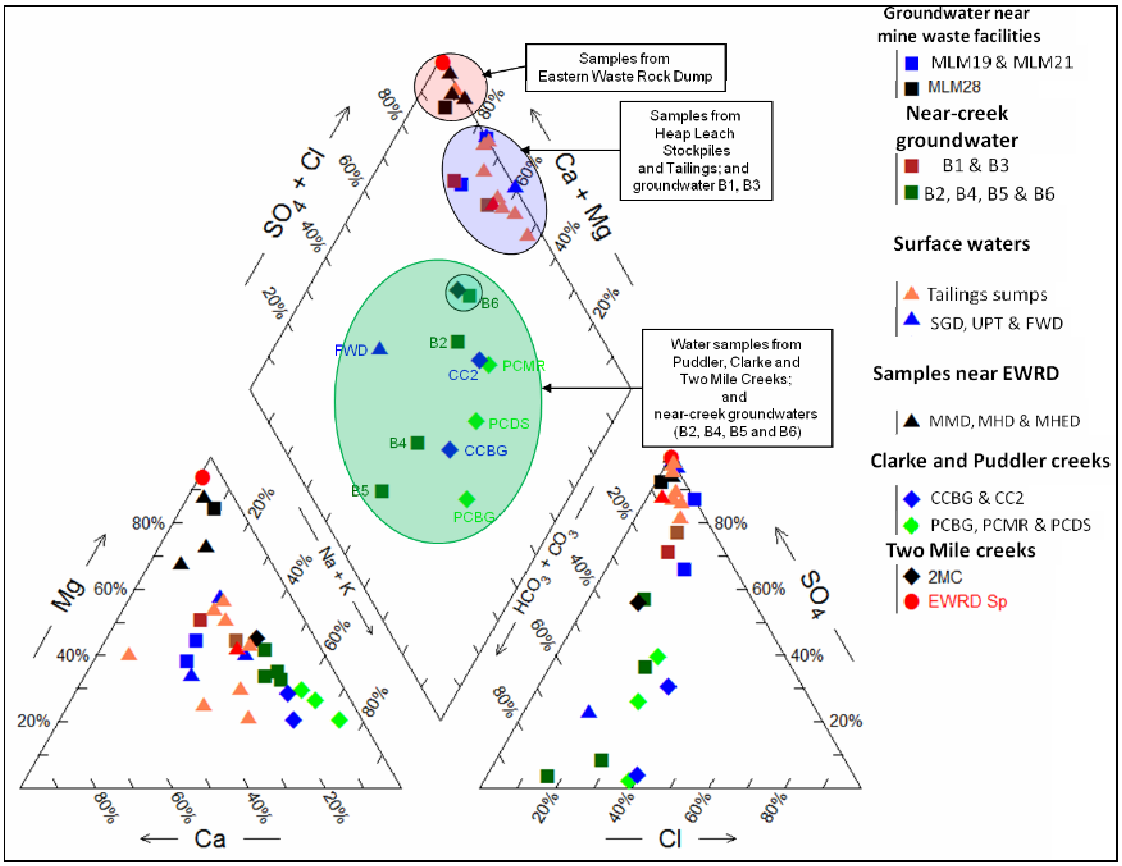

Figure 2. Piper diagrams showing major ion compositions of water samples.

Waters of Clarke, Puddler and Two Mile creeks collected in January, 2011, generally had high pH and low concentrations of metals and metalloids. A review of historical data showed that arsenic and boron might occasionally present at elevated concentrations in tailings seepages, particularly in dry season . However, they were found at low concentrations in creek waters in this study. Sulfate concentrations of Clarke and Puddler creek waters increased above upstream background concentrations as they entered the mine lease boundary suggesting an influence of sulfate-rich sources of water. These might be from mine water seepage or runoff reaching creeks through seepage zones (Fig. 1). The above observations indicated that mine waters were having limited influence on dissolved constituents in creek waters with the exception of sulfate concentrations. This might be attributed to the dilution effects from creek waters having background concentration levels as well as water management practices at the mine site such as pumping back and containment of mine waters. The influences of mine acidic discharge to natural creeks could also be limited at least in part by the high background neutralizing capacity of creeks with alkalinity values ranging from 313 to 680 mg/L $CaCO_3$ equivalent. The high alkalinity of creek waters can neutralize low pH mine drainages and eliminate metals by precipitation mechanism. Moreover, the local basement system surrounding Mt Leyshon are comprised of metamorphosed siltstones, sandstones and greywacke (WMC 2009) that might contribute to the neutralizing capacity against acidic seepages. Other oxyanions (nitrate, nitrite and phosphate) were predominantly below detection limits and were normally higher in groundwaters and seepage dams than in creek waters.

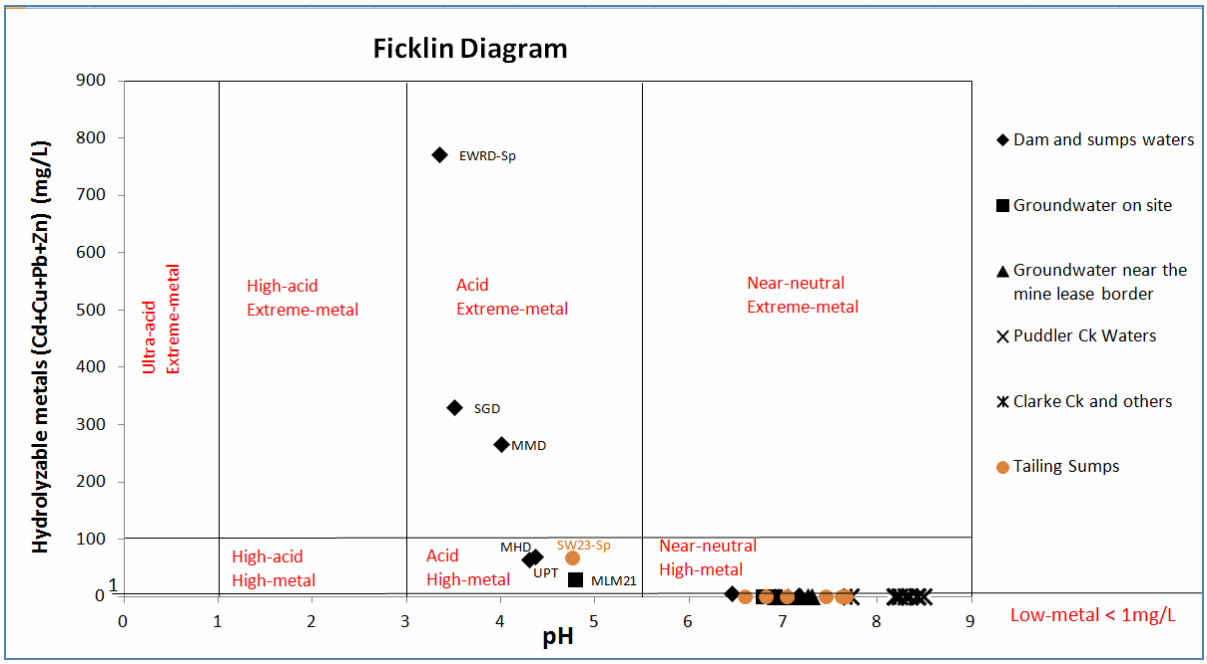

Figure 3. Ficklin diagram

4.2. Stable isotope compositions of waters

The stable isotope plot of $\delta^{18}O$ against $\delta^{2}H$ for water samples is shown in Figure 4 with the inclusion of the Global Meteoric Water Line (GMWL (Craig 1961): $\delta^{2}H$ ‰= 8.13$\delta^{18}O$ ‰ + 10.8) and the Local Meteoric Water Line of Fitzroy region, Queensland (LMWL (Vink *et al.* 2009): $\delta^{2}H$ ‰= 7.703$\delta^{18}O$ ‰ + 9.018). The rainwater at the mine site had isotopic values of -32 ‰ for $\delta^{2}H$ and -5.8 ‰ for $\delta^{18}O$. The seepage from Eastern Waste Rock Dump (EWRD-Sp, $\delta^{2}H$ -31 ‰ and $\delta^{18}O$ -5.4 ‰) and waters of Puddler Creek, Clarke Creek and Two Mile Creek ($\delta^{2}H$ from -38 ‰ to -30 ‰ and $\delta^{18}O$ from -5.7 ‰ to -4.8 ‰) had similar isotopic compositions to rainwater. Groundwaters from the mine site had isotopic compositions ranging from -42 ‰ to -25 ‰ for $\delta^{2}H$ and -6.5 ‰ to -3.6 ‰ for $\delta^{18}O$. Surface waters of dams or sumps at the mine site had isotopic signatures ranging from -43 ‰ to -16 ‰ for $\delta^{2}H$ and -5.7 ‰ to -1.5 ‰ for $\delta^{18}O$. RWD ($\delta^{2}H$ -16 ‰ and $\delta^{18}O$ -1.5 ‰) and seepages from tailings impoundments collected at seepage sumps (SW01, SW05, SW19, SW23, SW161, NNTD-NW and STD-WS) had significantly enriched isotopic signatures (-25.3 ‰ to 1.8 ‰ for $\delta^{2}H$ and -3.2 ‰ to 2.8 ‰ for $\delta^{18}O$) compared to the rainwater. The stable isotope composition of waters at the mine site defined an evaporation line (inset Figure 4: $\delta^{2}H$ ‰ = 4.97$\delta^{18}O$ ‰ – 9.11), with a high correlation (R^2 = 0.958). The slope of 4.97 was consistent with the general evaporation line slopes ranging from 3 to 6 (Gibson *et al.* 1993; Ghomshei and Allen 2000a).

The similar isotopic compositions of the seepage from Eastern Waste Rock Dump (EWRD-Sp) and creek waters to those of the rainwater sample suggested a common origin and drainage history for EWRD seepage and stream waters. It was noticeable that almost all surface and ground waters sampled in the vicinity of EWRD (MHD, MHED, MMD, MLM28, B2, B4, B5 and B6, and EWRD-Sp (Figure 1)) had isotopic compositions identical or slightly depleted compared to rain and creek waters. This might suggest that the seepages from EWRD originated from rainwater and were less affected by evaporation. It is possible that the seepages in the dams and sumps near the EWRD were constantly pumped into the mine pit or to RWD. It is also inferred that seepages from the Eastern Waste Rock Dumps were not from rising groundwater but from infiltrating rainwater.

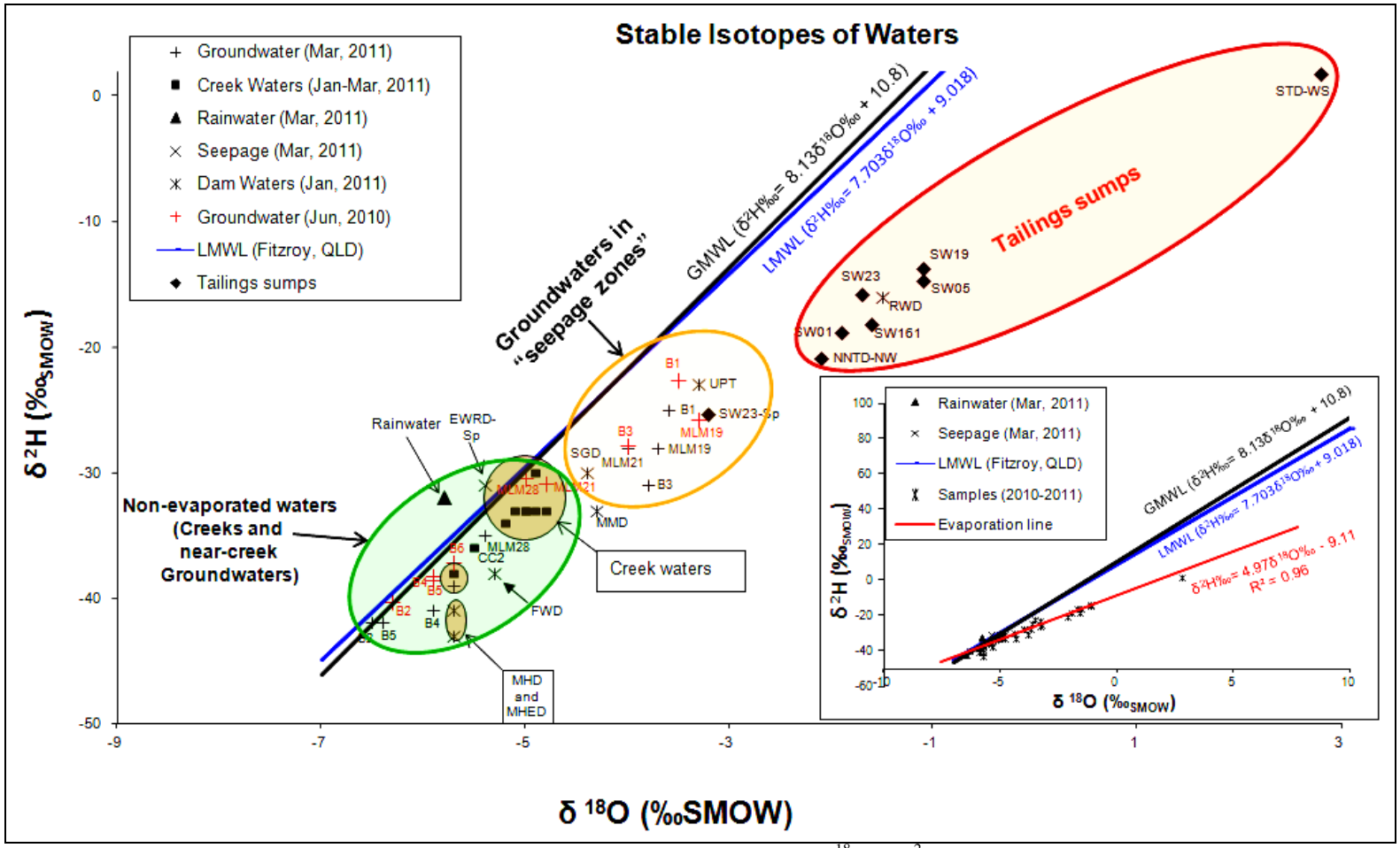

Figure 4. Stable isotope plot of $\delta^{18}O$ vs $\delta^{2}H$ for waters

The enrichment of tailings sumps (SW01, SW05, SW19, SW23, SW161, NNTD-NW and particularly STD-WS), and RWD suggested strong evaporation effects. The RWD acted as the second biggest water collection point that stored seepages and runoff pumped from other tailings sumps. Thus, the stable isotope compositions of tailings waters were very similar to that of RWD. STD-WS was taken at a tailings sump that was remotely located near the Southern Tailings Dam. There was limited information on the water recycling of STD-WS. The seepage contained in STD-WS was probably left intact without being pumped back to larger dams. Thus, the strong evaporative signatures of STD-WS might be the result of combination effects from long seepage storage and the recycling intensity effect during mineral processing.

Surface and ground waters near the Heap Leach stockpiles, Mill Rejects areas (SGD, UPT, MLM19 and MLM21) had relatively enriched isotopic compositions compared to rainwater (Fig. 4). Groundwaters near the Puddler and Clarke creeks (B1 and B3) had isotopic compositions similar to MLM19 and MLM21 and were distinctly enriched compared to other near-creek groundwaters (B2, B4, B5 and B6). B1 and B3 also had higher sulfate compositions compared to other near-creek groundwaters (Fig. 2). The combination of the similarities in water stable isotope signatures and geochemical compositions of near-creek groundwaters B1 and B3 and those near mine waste facilities (MLM19 and MLM21) has confirmed the fracture zones or "seepage zones" (Fig. 1) identified by Klohn Crippen Consultants (1991).

4.3. Sulfur isotope compositions

Sulfate-sulfur isotope compositions and sulfate concentrations are shown in Figure 5. The figure also includes the range of sulfur isotope compositions of sulfides (4.9-8.8 ‰,) following Morrison *et al.* (1988) and alunite groups of minerals (8.0-9.8 ‰) following Bird *et al.* (1989).

Most of the surface waters and groundwaters inside the mine lease boundary had sulfate-sulfur stable isotope compositions within the range of sulfides (4.9-8.8 ‰, following Morrison *et al.* (1988)). The comparison with sulfur isotope signatures of alunite groups found on site (8.0-9.8 ‰ following Bird *et al.* (1989)) showed very little match. Thus, the contribution of sulfate from the dissolution of weathered alunite groups appeared to be limited. The data in Figure 5 suggested that sulfate in waters of seepage dams and sumps and groundwater at the mine site originated mainly from sulfide oxidation.

The isotopic sulfate signature within the mine site was also compared to sulfate-sulfur isotopic compositions from upstream background waters of the Puddler (PCBG1 and PCBG2) and Clarke creeks (CCBG). The background waters had clearly distinct sulfate-sulfur stable isotope signatures (16.2-22.04 ‰) compared to those of sulfide or alunite groups. This indicated that the upstream creek waters contained a different source of sulfate from background mineralisation. Waters of Puddler Creek inside the mine lease boundary (PCMR1 & PCMR2, PCDS1 & PCDS2) and groundwaters near both Puddler and One Mile creeks (B3, B4 and B5) had similar sulfate-sulfur isotopic signatures and sulfate concentrations. They had sulfate-sulfur isotopic values lying between those of the background and mine waters. A similar pattern was observed for the waters of Clarke Creek (CC2 and B2). This indicated the mixing of background creek waters with the surface runoff or groundwater from the mine site, and thus the mixing of different sulfate sources. This was also shown by the enrichment of sulfate-sulfur isotopes of groundwaters in wet season (Mar, 2011) compared to those in dry season (Jun, 2010). This was due to the recharge of surface water to groundwaters during wet season that caused mixing of background sulfate with mine-related sulfate in groundwaters.

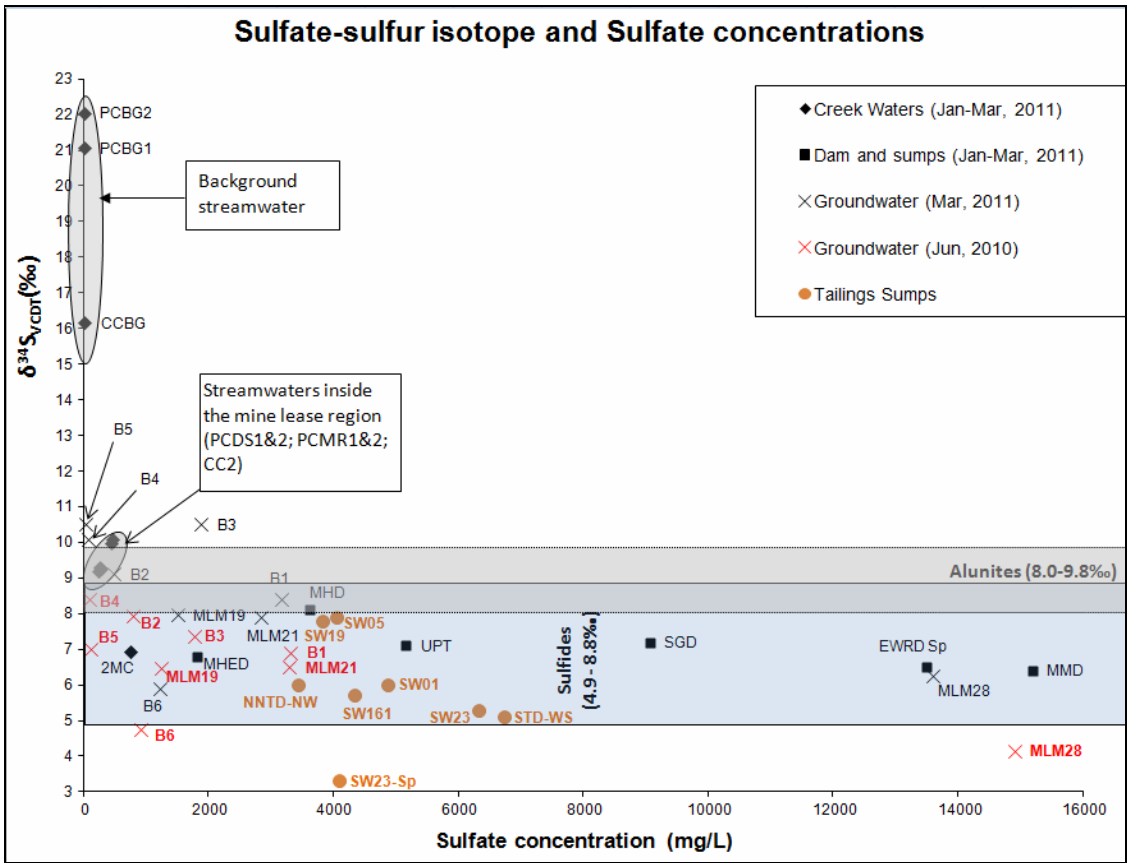

Figure 5. Stable isotope geochemistry and concentrations of sulfate

5. Conclusions

Mine water geochemistry and stable isotopes were effectively used for geochemical fingerprinting of waters from different mine facilities at the Mt Leyshon mine site. The study showed that mine waters within areas with similar geochemical compositions such as surface seepage storage sumps and groundwaters in the vicinity of waste rock dumps or heap leach areas had similar chemical compositions. The Eastern Waste Rock Dump, Heap Leach and Mill Rejects Stockpiles produced highly acidic and metal-rich seepages whereas, surface and groundwaters near tailings containments had low hydrolysable metal concentrations and high pH that was a consequence of mineral processing. Seepages from the mine have locally impacted surface and groundwater especially around the Eastern Waste Rock Dump, tailings dams, Heap Leach and Mill Rejects areas. However, it is important to note that the comparison of geochemical properties of surface and groundwaters within mine lease boundary and those of creeks has revealed that the effects of mine discharges to streams were limited owing to the high neutralizing capacity of the combination of interaction with local geology and mixing with high alkalinity natural background waters. The stable isotope geochemistry of waters (^{18}O and ^{2}H) showed that waters within and around the mine originated from precipitation with various degrees of evaporation. Tailings seepage sumps were distinctly enriched compared to rainwater and those from the Eastern Waste Rock Dump and creek waters. The enrichment of tailings seepage sumps might be attributed to a combination of extreme evaporation effect due to long term storage of seepage, and water recycling intensity during mineral processing. This isotope enrichment effect was used to confirm the connectivity of surface and groundwater along previously inferred seepage zones. There were also indications of the mixing between mine waters and the creeks based on sulfur isotopes. Sulfate-sulfur isotope geochemistry has determined that sulfate derived from sulfide oxidation at the mine site was the main source of sulfate in affected creeks.

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List of Acronyms

AMD	Acid Mine Drainage	NNTD	New Northern Tailings Dam
EWRD	Eastern Waste Rock Dump	NWRD	Northern Waste Rock Dump
FWD	Freshwater Dam	ONTD	Old Northern Tailings Dam
GMWL	Global Meteoric Water Line	PAF	Potentially Acid Forming
LMWL	Local Meteoric Water Line	ROM	Run of Mine
MHD	Mt. Hope Dam	RWD	Raw Water Dam
MHED	Mt. Hope Extension Dam	SGD	Supergene Dam
MMD	Mt. Mawe Dam	STD	Southern Tailings Dam
NAF	Non-Acid Forming	SWRD	Southern Waste Rock Dump
NAPP	Net Acid Producing Potential	UPT	Upper Plum Tree dam