

Variability in Mine Waste Mineralogy and Water Environment Risks: a Case Study on the River Almond Catchment, Scotland

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Abstract

The River Almond catchment contains coal, oil shale and ironstone mine waste and displays widespread surface water metals pollution. Mineralogical investigations and geochemical modelling at four mine waste sites identified pyrite oxidation and jarosite, siderite and aluminosilicate dissolution reactions as the primary sources of metal pollutants (Fe, Mn, Al). Carbonate dissolution reactions control drainage pH. Pyrite is absent in burnt oil shale waste, however, trace content in unburnt shale horizons is implicated as a source of Fe in drainage waters. Site specific water quality and load assessments indicate pyrite bearing coal and ironstone sites present the greatest water environment risks.

Keywords: Coal, Oil Shale, Ironstone, PHREEQC

Introduction

Mine waste is a common visual reminder in many formerly mined river catchments and the associated drainage waters often present serious risks to surface water quality (Rees *et al.* 2002, Younger 2004). Oxidation of sulphide minerals, particularly pyrite (FeS₂), and the associated pollutant release may be more severe in surface deposited mine waste than in subsurface mines due to the continued availability of atmospheric oxygen and limited availability of carbonate minerals to buffer acidity (Rees *et al.* 2002, Younger 2004). Diffuse mining pollutant sources, such as mine waste drainage, can be the dominant source of surface water metals and can account for up to 98% of in-stream Fe load during high flow events (Mayes *et al.* 2008). National scale treatment programs exist for mine water discharges from underground coal mines however no similar program is currently available for coal mine waste or other mining waste drainage in Scotland.

The River Almond catchment, west of Edinburgh, has been mined for the last five centuries; limited silver deposits at Hilderston were targeted in the 1600s, coal and ironstone mining began in the 1700s and 1800s and oil shale was mined from the 1860s to the 1960s.

Underground coal mining ended in the 1980s, however, surface mining for coal and fireclay continued until 2012. Mine wastes from coal, oil shale and ironstone are widespread and many surface water bodies in the catchment display elevated pollutant concentrations (Haunch *et al.* 2013). Most mine waste is overburden, interburden or other rock discarded at surface during mine development. Oil shale waste is unusual as it consists of a small percentage of mine development waste, but the majority is a burnt, friable, orange industrial waste produced when the mined oil shale was heated to derive various hydrocarbon chemicals in the now closed Scottish Shale Oil Industry (Louw & Addison 1985). This history of mining and associated legacy of mine wastes and poor water quality makes the River Almond an ideal catchment to undertake a case study to assess variability in the mineralogy of mine wastes and associated water environment risks.

Site Selection & Investigation Methods

Four mine waste sites were selected, their locations are shown on Figure 1 and are described below.

S1-Fauldhouse ironstone mine waste is 58,000 m² in size and consists predominantly

of grey argillaceous waste with a notable proportion of siderite ironstone nodules and fragments of finely banded coal and shale. The site is associated with the Fauldhouse iron industries of the 1800s.

S2–Whitburn coal mine waste is 540,000 m² in size and is partially restored with vegetation cover and public access. The site is adjacent to the former Whitrigg Colliery (1900–1972). A small pilot mine water treatment scheme is in place, drainage samples were recovered from the main site drainage at the effluent end of a limestone trench installed as part of the treatment scheme.

S3–Benhar coal mine waste is 25,000 m² in size and consists of a low-lying area of waste and a central conical area with a diffuse drainage displaying orange precipitates. Historic maps indicate the waste is sourced from both the former East Benhar (coal) mine and Fallahill Colliery.

S4–Hermand oil shale mine waste 14,000 m² in size (small compared to most oil shale sites but this allowed easier access and characterisation) and consists of a burnt orange-red waste, with some darker unburnt horizons of black oil shale. The waste was deposited by the adjacent historic Hermand Oil works (1883–1894) which received oil shale from the Hermand No.6 mine.

Mine waste samples were recovered from each site and Quantitative X-Ray Diffraction (QXRD) analysis was undertaken at the University of Edinburgh. Mine waste drainage

samples were recovered and analysed at the British Geological Survey (BGS) laboratories Keyworth, UK. PHREEQC inverse modelling was used to assess mineralogical controls on water chemistry from rainfall to mine waste drainage. A GIS was constructed using information from West Lothian Council, BGS, Scottish Environment Protection Agency (SEPA) and National Library of Scotland (NLS) (Haunch 2013).

Mine Waste Mineralogy

The results of the QXRD mineralogical analysis are summarised in Table 1. The Fauldhouse ironstone mine waste (S1) and coal mine waste sites (S2 & S3) each display similar mineralogical assemblages consistent with shale and mudstone source lithologies. However, a number of key features warrant further consideration; 1) Pyrite (FeS₂) is identified above 1 wt% at S1 in a quarter of samples, at S2 pyrite was identified above 1wt% in two of 23 samples while at S3 no pyrite was identified, 2) Jarosite (KFe₃(OH)₆(SO₄)₂), essentially a hydrated form of pyrite, is identified above 1 wt% in every sample at S1, while at S2 jarosite is identified above 1 wt% in only two samples and at S3 no jarosite is identified, 3) Siderite (FeCO₃) is identified at S3 in four of 17 samples with contents ranging between 1.42 and 4.2 wt%.

Oil shale mine waste (S4) mineralogy consists mainly of quartz, feldspars, clay minerals, hematite, cordierite and mullite.

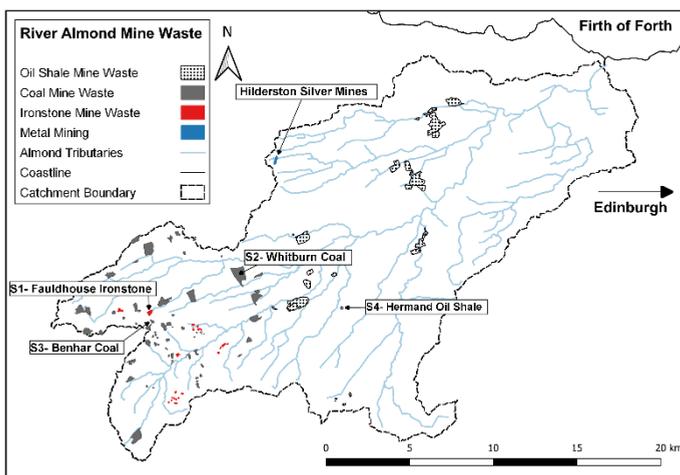


Figure 1 Mine waste distribution in the River Almond Catchment.

This assemblage is consistent with thermal industrial processing of the waste in the historic Scottish Shale Oil Industry (Haunch 2013). Two samples were analysed from black horizons in the waste, and these revealed a mineralogy consistent with raw unprocessed oil shale (Louw & Addison 1985) and contain trace (0.7 & 0.3 wt%) pyrite content.

In summary, mineralogical analysis indicates that the Fauldhouse ironstone mine waste (S1) contains notable proportions of acid generating minerals pyrite and jarosite, the Whitburn coal mine waste (S2) contains notable but lower abundance of these acid generating minerals, the Benhar coal mine waste (S3) contains siderite but no pyrite or

Table 1 Mineralogical analysis results determined using Quantitative X-Ray Diffraction on waste rock samples recovered at four mine waste sites in the River Almond Catchment.

| Phase | Formula | Site 1 Fauldhouse | | | | Site 2 Whitburn | | | | Site 3 Benhar | | | | Site 4 Hermand | | | |
|---------------|--|-------------------|------|--------|------|-----------------|------|--------|------|---------------|------|--------|------|----------------|------|--------|------|
| | | n | Max | Median | Min | n | Max | Median | Min | n | Max | Median | Min | n | Max | Median | Min |
| Quartz | SiO ₂ | 24 | 4.3 | <1 | <1 | 23 | 2.2 | 1.4 | <1 | 17 | 6 | 3.9 | <1 | 19 | 47.1 | 30.3 | 13.8 |
| Microcline | KAlSi ₃ O ₈ | 24 | 7.4 | 4.5 | <1 | 23 | 7.3 | 3.1 | <1 | 7 | 11.2 | 7.0 | 3.3 | 2 | 4.3 | 3.6 | 2.9 |
| Albite | NaAlSi ₃ O ₈ | 24 | 1.8 | <1 | <1 | 1 | 1.1 | - | 1.1 | 7 | 11.3 | 6.0 | 4.3 | 19 | 4.9 | 2.4 | <1 |
| Orthoclase | KAlSi ₃ O ₈ | 24 | 1.2 | <1 | <1 | 21 | 4.9 | 2.3 | <1 | 7 | 16.4 | 4.8 | 1.9 | | | | |
| Anorthite | CaAl ₂ Si ₂ O ₇ | | | | | | | | | | | | | 18 | 12.4 | 7.9 | <1 |
| Dickite | Al ₂ Si ₂ O ₅ (OH) ₄ | 24 | 62.9 | 44 | 28.4 | | | | | 10 | 30.5 | 22.4 | 10.1 | | | | |
| Illite | K _{1.5-1.0} Al ₄ Si _{6.5-7.0} Al _{1.5-1.0} O ₂₀ (OH) ₄ | 22 | 21.9 | 10.1 | 5.1 | 23 | 55.2 | 40.0 | 12.3 | 8 | 48.6 | 29.1 | 24.7 | | | | |
| Kaolinite | Al ₂ Si ₂ O ₅ (OH) ₄ | 24 | 22.4 | 15.6 | 1.4 | 23 | 52.2 | 22.6 | 11.7 | 17 | 52.0 | 34.0 | 13.7 | 16 | 10.9 | 1.0 | <1 |
| Muscovite | KAl ₃ (AlSi ₃ O ₁₀)(OH) ₂ | 24 | 18.8 | 13.8 | 8.2 | 23 | 36.4 | 22.4 | 17.0 | 5 | 47.7 | 40.7 | 35.8 | 19 | 53.9 | 18.6 | 2.7 |
| Calcite | CaCO ₃ | 23 | 1.1 | <1 | <1 | 23 | 3.9 | <1 | <1 | 2 | 1.9 | 1.1 | <1 | 4 | 1.12 | <1 | <1 |
| Dolomite | (CaMg)(CO ₃) ₂ | 23 | 1.0 | <1 | <1 | 23 | 1.0 | <1 | <1 | | | | | 16 | 3.4 | <1 | <1 |
| Siderite | FeCO ₃ | 20 | 5.0 | <1 | <1 | 1 | 2.2 | - | 2.21 | 4 | 4.2 | 2.4 | 1.4 | 2 | <1 | - | <1 |
| Pyrite | FeS ₂ | 24 | 7.5 | <1 | <1 | 23 | 1.5 | <1 | <1 | | | | | 2 | <1 | - | <1 |
| Jarosite | KFe ₃ (SO ₄) ₂ (OH) ₆ | 24 | 3.5 | 2.1 | 1 | 23 | 1.7 | <1 | <1 | | | | | | | | |
| Goethite | FeOOH | 24 | 12.2 | 1.9 | <1 | 23 | 1.2 | <1 | <1 | 10 | 1.6 | 1.2 | <1 | | | | |
| Lepidocrocite | γ-FeO(OH) | 1 | 1.5 | - | 1.5 | | | | | | | | | | | | |
| Hematite | Fe ₂ O ₃ | | | | | 1 | 1.3 | - | 1.3 | | | | | 19 | 13.2 | 8.3 | 1.4 |
| Cordierite | (Mg,Fe) ₂ [Si ₃ Al ₁₀] nH ₂ O | | | | | | | | | | | | | 13 | 16.9 | 4.4 | <1 |
| Mullite | 3Al ₂ O ₃ 2SiO ₂ | | | | | | | | | | | | | 12 | 35.6 | 28.1 | 20.7 |

Table 2 Drainage water quality analysis results from samples recovered at four mine waste sites in the River Almond Catchment. Note: Mn EQS is based on the bioavailable fraction.

| Site | Temp | Eh | pH | EC | Ca | Mg | Na | K | HCO ₃ | Cl | SO ₄ | Fe _{tot} | Fe ²⁺ | Fe ³⁺ | Mn | Al |
|------|------|-----|-----|-------|-------|------|------|------|------------------|------|-----------------|-------------------|------------------|------------------|-------|------|
| | °C | mV | | µS/cm | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L | µg/L |
| EQS | | | | | | | | | | | 400 | | 1 | 1 | 0.123 | 15 |
| S1 | 19.0 | 582 | 3.5 | 1041 | 97.2 | 10.9 | 9.9 | 0.5 | <5 | 12.0 | 569.5 | 127.9 | 70.4 | 88.3 | 11.9 | 7347 |
| S2 | 8.9 | 227 | 6.2 | 1697 | 217.3 | 52.4 | 6.9 | 10.7 | 188.0 | 6.0 | 669.1 | 78.6 | 67.3 | 11.3 | 12.1 | 61 |
| S3 | 12.2 | 282 | 6.7 | 243 | 23.8 | 18.0 | 4.6 | 2.7 | 136.7 | 5.1 | 20.8 | 1.0 | 1.0 | 0.1 | 0.05 | 74 |
| S4 | 13.0 | 386 | 6.5 | 353 | 51.4 | 8.5 | 14.3 | 23.8 | 155.0 | 15.4 | 26.2 | 2.4 | 0.9 | 1.5 | 0.5 | 154 |

Table 3 Mine waste drainage characteristics at four sites in the River Almond Catchment.

| Site | Mine Waste | pH | Pollutants > EQS | MW Classification (Rees 2002) | Saturation Indices | MAMDI (Kuma <i>et al.</i> 2011) | Comments |
|------|------------|--------------------|-----------------------------|---------------------------------|---------------------------------|---------------------------------|--|
| S1 | Ironstone | Acidic 3.5 | Fe, Mn, Al, SO ₄ | Net acidic, sulphate dominant | Goethite 5.15 Jarosite 2.75 | 49.0 | Jarosite precipitates identified by XQRD in drainage channel close to waste. Orange Fe precipitates extend 2+ km downstream. |
| S2 | Coal | Circum-neutral 6.2 | Fe, Mn, Al, SO ₄ | Net alkaline, sulphate dominant | Goethite 9.18 Jarosite 8.37 | 62.4 | Treatment scheme in place and limestone trench increases Ca, Mg, HCO content. Ocherous impacts persist 1 km downstream. |
| S3 | Coal | Circum-neutral 6.7 | Fe, Al | Net alkaline, sulphate dominant | Goethite 7.32 Jarosite -2.23 | 96.0 | Orange precipitates in drainage immediately adjacent to site. |
| S4 | Oil shale | Circum-neutral 6.5 | Fe, Mn, Al | Net alkaline, sulphate dominant | Goethite 8.71 Jarosite 1.74 | 90.3 | Orange precipitates in drainage immediately adjacent to site. |

jarosite and the Hermand oil shale waste (S4) is mainly a hematite bearing waste but with discrete horizons of unprocessed oil shale which contain trace pyrite content.

Mine Waste Drainage Characteristics

Mine waste drainage water quality analysis results are summarised in Table 2. The key characteristics of the drainage, using several assessment and classification schemes, are summarised in Table 3. Comparison is made to Scottish Environmental Quality Standards (EQS) for Fe, Mn, Al and SO₄ (SEPA 2020); above EQS concentrations indicates potential risks to surface water quality and ecology. The Rees *et al.* (2002) mine water classification scheme has been used to assess the net alkalinity and sulphate dominance of waters. A full trace metals suite was also undertaken and is summarised within the Modified Acidic Mine Drainage Index (MAMDI) score (Kuma *et al.* 2011); lower MAMDI values (ranging 0-100) indicate higher concentrations of pollutants of concern. In general, the results indicate the Fauldhouse ironstone site (S1) is potentially the most polluting, followed by the Whitburn coal mine waste (S2) and the Hermand oil shale (S4). The Benhar coal mine waste (S3) drainage displays the least pollution potential.

Mine Waste Drainage Evolution

PHREEQC inverse modelling has been used to identify the main hydrochemical processes controlling mine waste drainage at each of the sites. Each model returned a number of solutions, those displayed in Figure 2 were selected based on the prevailing geochemical conditions at each site.

Fauldhouse ironstone mine waste (S1) modelling results indicate pyrite oxidation (2.5-3.1 mmol/kg(H₂O)) is the main control on Fe and SO₄ release. Jarosite dissolution (0.54 mmol/kg(H₂O)) was identified as a secondary control on Fe and SO₄ release in two of the model solutions. Calcite and/or dolomite dissolution (i.e., carbonate buffering) control Ca & Mg in drainage waters, but this buffering is not sufficient to consume all proton acidity (H⁺) released from pyrite oxidation and prevent low pH drainage (pH 3.5). Goethite precipitation in all three solutions is consistent with observations of orange precipitates both on waste rock surfaces in recovered cores and within the drainage channel. Dissolution of aluminosilicate minerals such as feldspars and micas and the corresponding precipitation of the hydrated clay mineral illite are likely associated with Al releases.

Whitburn coal mine waste (S2) modelling results are similar to S1 with pyrite oxidation (3.66 mmol/kg (H₂O)), carbonate buffering and aluminosilicate reactions being the primary control on drainage water chemistry. As drainage samples were recovered following the limestone trench it is likely that some of the calcite (2.58-2.99 mmol/kg(H₂O)) and dolomite (2.16-2.56 mmol/kg(H₂O)) dissolution in the model solutions is representative of the limestone trench and not the mine waste water rock interactions. This is supported by additional sampling from two shallow boreholes installed into the waste pile which indicated perched groundwater with a pH of 5.5-5.8 and lower calcium (60 & 148 mg/L) and magnesium (18.5 & 31.5 mg/L) concentrations than the sampled discharge (Ca - 217 mg/L & Mg - 52.3 mg/L).

Benhar coal mine waste (S3) solutions also indicate pyrite oxidation, carbonate buffering and aluminosilicate dissolution and precipitation reactions but notably at much lower values than S1 and S2. It should be

noted that pyrite and dolomite were not identified in the mineralogical analysis but were included in the model as the most likely source of SO₄ and Mg in the discharge. Model solution 1 indicates siderite dissolution is the main source of Fe in the drainage. This is considered the most credible solution based on the QXRD identified siderite and goethite contents (up to 4.2 & 1.6 wt% respectively), absent or very low pyrite content and observed Fe precipitates in the main drainage area.

The Hermand oil shale waste (S4) model solutions indicate carbonate and aluminosilicate reactions are of primary control on drainage chemistry. Pyrite oxidation was identified as a contributor in all four solutions, but in practice at the site this must be limited to the identified discrete horizons of unburnt oil shale which contain trace contents of pyrite. Two of the model solutions also suggest hematite dissolution may be involved in Fe release. This would require an excess of proton acidity; possible sources of acidity could include pyrite oxidation or infiltrating rainfall. However, it is unclear whether the

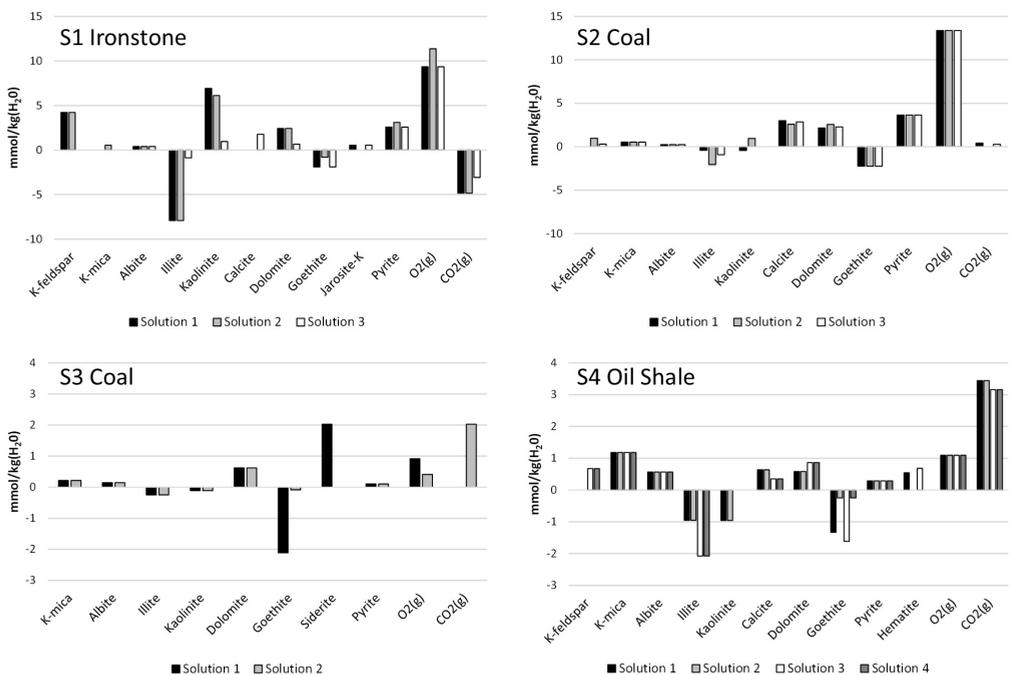


Figure 2 PHREEQC inverse modelling results for four mine waste sites in the River Almond Catchment based on mineralogy and drainage water chemistry, +ve=dissolution, -ve= precipitation.

hematite dissolution could result in Fe release at neutral pH and ambient temperature conditions.

Mineral phases containing Mn were not identified in the QXRD analysis at any of the sites. Manganese is commonly present in soils and rocks as a minor substitution within minerals, particularly Fe bearing minerals, and on clay mineral surfaces (Gilkes & McKenzie 1988). Mineral dissolution reactions, particularly where pH is low, are likely to liberate Mn and account for the concentrations in the drainage at each site.

Water Environment Risk Assessment and Catchment Waste Abundance

The risks posed by mine waste drainage depends on the concentrations of pollutants (measured via MAMDI scores) and the pollutant load. Drainage streams at mine waste sites are often diffuse making direct field measurement difficult. Instead, an estimate of the average flow can be gained by estimating effective rainfall from climate data. In Table 4 drainage volumes have been calculated by multiplying the site area by the average annual rainfall (1998–2019) measured at SEPA's Whitburn rainfall station (968 mm/yr) in the catchment minus the potential evaporation (501 mm/yr). These drainage volumes have then been multiplied by pollutant concentrations to calculate average annual metal loads.

The risk assessment indicates the coal site, S2, poses the greatest overall risk to water quality due to the size of the source (540,000 m²), associated drainage volumes and quality (MAMDI-62.4). The S1 ironstone drainage quality (MAMDI-49) is potentially more polluting than S2 but the source size (58,000 m²) is much smaller resulting in lower pollutant loads and lower overall risk ranking. The second coal site S3 and the oil shale site S4 are broadly similar and present a low risk to water quality.

Further work is required to assess how drainage quality and risk varies between different coal, ironstone and oil shale sites across the whole catchment. Indeed, it is evident from the difference in risk between sites S2 and S3 that there is likely to be variability within each waste type, and that this is ultimately the result of waste mineralogy. At oil shale sites the risk will be dependent on the proportion of unburnt black shale. Nevertheless, the overall abundance of waste (Table 5) in the catchment suggests coal mine waste is likely to be of greatest concern followed by ironstone and oil shale.

Conclusions

This study indicates the importance of mine waste mineralogy in assessment of metal release, drainage evolution and water environment risks. In the River Almond catchment, mine wastes derived from

Table 4 Mine waste drainage volumes, pollutant loading estimation and risk ranking.

| | S1 Ironstone | S2 Coal | S3 Coal | S4 Oil Shale |
|-------------------------|----------------|---------|---------|--------------|
| Area (m ²) | 58,000 | 540,000 | 25,000 | 14,000 |
| Drainage Estimate (L/s) | 0.86 | 8.01 | 0.37 | 0.21 |
| Fe (kg/yr) | 3471 | 19,863 | 11.7 | 15.7 |
| Mn (kg/yr) | 325 | 3057 | 0.6 | 3.2 |
| Al (kg/yr) | 200 | 154 | 8.6 | 1.0 |
| MAMDI | 49 | 62.4 | 96 | 90 |
| Risk Rank | 2- Medium/High | 1- High | 4- Low | 3-Low |

Table 5 Mine waste abundance in the Almond River Catchment.

| | Oil Shale | Coal | Ironstone |
|-------------------------|-----------|------|-----------|
| Area (km ²) | 4.15 | 3.91 | 0.274 |
| Catchment % | 1.05 | 0.98 | 0.07 |

historic coal, oil shale and ironstone mining industries are confirmed as sources of Fe, Mn, Al and SO_4 . Pyrite oxidation reactions in ironstone and coal mine wastes are identified as the main control on elevated Fe concentrations in drainage waters. Jarosite dissolution plays a secondary role although it is likely PHREEQC inverse modelling does not fully represent the movement of pollutants from pyrite to jarosite and into drainage waters. Siderite dissolution is implicated as a source of Fe at a coal mine waste site with low abundance of pyrite.

The oil shale waste site investigated consisted mainly of burnt orange waste containing hematite and little to no pyrite. However, discrete horizons of unburnt black oil shale with trace pyrite content were identified and modelling suggested pyrite oxidation reactions in these horizons as a source of Fe in drainage waters. Some model solutions also implicated hematite dissolution in Fe release. Carbonate buffering was shown to influence drainage pH, Ca and Mg content in drainage at all sites. Al in drainage waters is likely to be associated with the dissolution of aluminosilicate minerals.

Site specific risk assessments indicate pyrite bearing coal and ironstone sites present the greatest water environment risks. Notably while ironstone mine waste drainage quality is extremely poor, ironstone mine waste abundance is low suggesting a lower overall risk, than coal mine waste, at the catchment scale.

Mine waste drainage is currently excluded from national scale mine water remediation programs and in many mined catchments continues to be a source of elevated metals and regulatory classification downgrades. This study demonstrates that further work is required to incorporate mine waste mineralogy and drainage monitoring into catchment water quality assessments and water body improvement objectives.

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