

Evaluating the remediation of arsenic-rich tailings at the historic Royal George tin mine, Tasmania, Australia

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Abstract

The aim of this study was to evaluate the remediation of As-rich tailings at the historic Royal George tin mine, northeastern Tasmania. Abandonment and neglect of the Royal George Mine have led to a site that is characterised by a severely modified (or lack of) vegetation and the presence of a 36,000 m³ tailings dump. The acid-generating tailings have minor (i.e. >100 ppm) mean concentrations of As, Cu, Mn, Pb, S, Sn and W, and traces (i.e. <100 ppm) of Ag, Bi, Cd, Co, Sb, Th, Tl, U and Zn. Since 2006, remediation works by statutory authorities have focused on: (a) the construction of gravel armoured drains and drainage diversion channels; (b) the establishment of a fenced-off revegetation trial plot; and (c) the application of crushed limestone and fertiliser pellets to the tailings dump surface. Apart from these minor remediation works, the tailings disposal area has been left to natural processes of surface amelioration and gradual vegetation colonisation.

*Today, the vegetation surrounding the site mainly comprises dry eucalypt woodland, whereas the amended tailings remain largely devoid of any vegetation, with only some moist or ponded areas being colonised by the rush *Juncus pallidus*. Biogeochemical analyses demonstrate that the plant has a pronounced trace metal tolerance and the tendency to exclude environmentally significant elements (Al, As, Co, Cu, Fe, Mn, Pb, Th, Tl, U, W, Zn) from its above-ground biomass. The exclusion of metals from the above-ground plant biomass of *J. pallidus* reduces the exposure of wildlife and grazing animals to metals and limits the transfer of metals up the food chain. The establishment of a *J. pallidus* cover on otherwise barren ground also reduces tailings erosion and dust generation as well as water infiltration and associated leaching of contaminants. Such processes reduce the potential exposure of animals and humans to metals and may also open pathways for natural ecological restoration and biodiversification.*

Geochemical characterisation of the amended tailings demonstrates that the remediated wastes have alkaline rinse pH as well as positive net acid producing potential (NAPP) values. Laboratory leaching experiments using deionised (DI) water as the extraction solution reveal that leaching of limestone-neutralised tailings results in a lower As release compared to non-amended tailings. However, leaching in 0.5 M K₂SO₄, which mimics the presence of sulphates observed in acid and metalliferous drainage (AMD) environments, shows a three-fold increase in As release from limestone and fertiliser-amended tailings compared to non-amended tailings. Desorption of As from metal oxide and hydroxide phases at alkaline pH conditions, in addition to the combined presence of phosphates and sulphates could result in the release of As into tailings pore waters and its discharge to surface water seepages and streams. This study demonstrates that neutralisation of As-rich tailings using crushed limestone and fertiliser-amendments, should be pursued with caution. Increased dissolved sulphate concentrations may result in As desorption and mobility into pore and surface waters.

1 Introduction

Historic mining and mineral processing of metal ores commonly cause the dispersion of trace metals and metalloids into surrounding soils, sediments and waters. At these sites, inappropriate waste disposal

practices and unconstrained dispersion of contaminants may lead to the contamination of soils and sediments well beyond the mine site (Ashley and Lottermoser, 1999). Consequently, historic metal mining centres are known for their pronounced geochemical footprint inflicted on the local and regional landscape. Such disturbed and contaminated lands invariably require remediation.

This study reports on the remediation of As-rich tailings at the historic Royal George tin mine, northeastern Tasmania, Australia (Figure 1). The aim of conducting this work was to gain data relating to the remediation and natural colonisation of exposed mine wastes. The selected site was particularly suitable for evaluating the rehabilitation of As-rich mining wastes that have been amended with limestone. Thus, this research adds to our understanding on the rehabilitation of As-rich mine wastes, and contributes to improving closure of As-rich mine sites and contaminated lands.

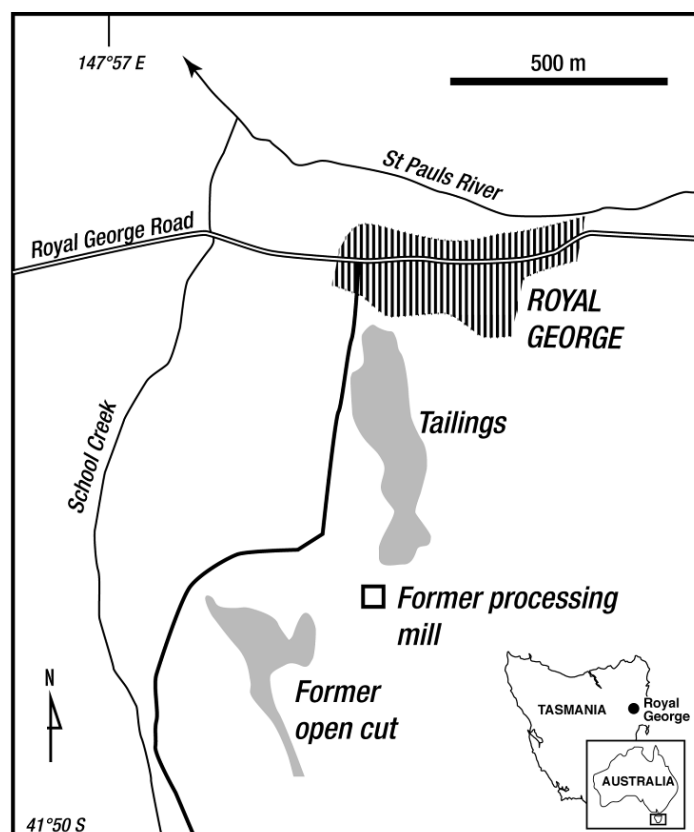


Figure 1 Map showing the location of the Royal George Mine site

2 Materials and methods

2.1 Site description

The Royal George Mine site is situated in northeastern Tasmania, Australia, immediately to the south of the Royal George township (Figure 1). The region has a temperate maritime type climate, with an average annual rainfall of 559 mm (Campbell Town weather station, 35 km southwest of Royal George) (Bureau of Meteorology, 2012). The Royal George Mine workings, mill processing site and tailings disposal area are located in an east-west orientated valley of the St Pauls River catchment, which drains into the South Esk River at Avoca. Drainage from the Royal George Mine and mineral processing site is via the north-flowing School Creek and a linear unnamed natural drainage gully (Figure 1). In the district, the main land use is grazing, forestry and crop framing. Population density is sparse; however, St Pauls River supplies irrigation and domestic water to surrounding farms and potable water for Royal George residents.

The Royal George site is one of several historic mines and associated processing sites in the region, with Royal George operational until 1928. The treated ore consisted of major cassiterite and variable amounts of

quartz, tourmaline, feldspar, mica, pyrite, chalcopyrite, sphalerite and arsenopyrite and possibly wolframite, stannite and torbernite (McIntosh Reid and Henderson, 1929; Newnham, 1990). Mineral processing of 0.16 Mt of ore involved crushing, grinding, roasting and gravimetric sorting to produce a cassiterite concentrate and tailings (McIntosh Reid and Henderson, 1929; Green, 1990).

Today, the historic tailings disposal site covers an approximately 600 m long by 50 m wide area and is estimated to contain approximately 36,000 m³ of tailings (Lockley and Pollington, 2006). Surface water flows as sheet drainage and localised erosion gorges through the tailings area (Figure 2), which consists of several former dried out tailings dams and ephemeral ponds. Near the margins of the ponds and in seepage zones, efflorescent salts are common (Figure 2).



Figure 2 Field photographs showing surface and rill erosion, erosion runnels and mineral efflorescences in exposed tailings at the Royal George Mine site

A previous environmental assessment of the tailings disposal site (Lockley and Pollington, 2006) revealed that surface tailings typically comprise clay to sand sized tailings with occasional granitic gravel. Tailings have minor (i.e. >100 ppm) mean concentrations of As, Cu, Pb, Sn and Zn as well as traces (i.e. <100 ppm) of Bi, Cd, Co, Cr, Mn, Ni, Th and U. The sulphidic tailings are acid-generating and hence, surface and seepage water samples contain elevated total Al (max. 29.6 mg/l), As (max. 0.6 mg/l), Cd (max. 0.4 mg/l), Cu (19.7 mg/l), Fe (64 mg/l), Mn (0.8 mg/l), Ni (0.04 mg/l), Pb (max. 0.02 mg/l), U (max. 0.6 mg/l), Zn (8.8 mg/l) and sulphate (max. 331 mg/l) at low pH (pH 2.7–4.6) and high electrical conductivity (max. 1.1 mS/cm) values (Lockley and Pollington, 2006). These contaminants seep from the tailings repository into the local drainage system. A preliminary risk assessment suggests that the tailings may cause discolouration, siltation and chemical contamination of St Pauls River as well as dust emissions over Royal George village residences (Lockley and Pollington, 2006).

Following site risk assessment and since 2006, remediation works by statutory authorities have focussed on: (a) minor earth works to prevent site access, (b) the construction of gravel armoured drains and drainage diversion channels; (c) the establishment of a fenced-off revegetation trial plot (Figure 4); and (d) the application of crushed limestone and fertiliser pellets to the tailings dump surface. Apart from these minor remediation works, the tailings disposal area has been left to natural processes of surface amelioration and gradual vegetation colonisation. The vegetation surrounding the site mainly comprises dry eucalypt woodland, whereas the metalliferous tailings are largely devoid of any vegetation, with some moist or ponded areas being colonised by the rush, *J. pallidus*. The tailings disposal site is the focus of this study because: (a) the tailings have been neutralised using crushed limestone; and (b) *J. pallidus* has

colonised the abandoned tailings since recent application of limestone and fertiliser (Figure 3). At Royal George, *J. pallidus* is the only species that grows (up to ~1.5 m tall) in moist tailings and ponds.

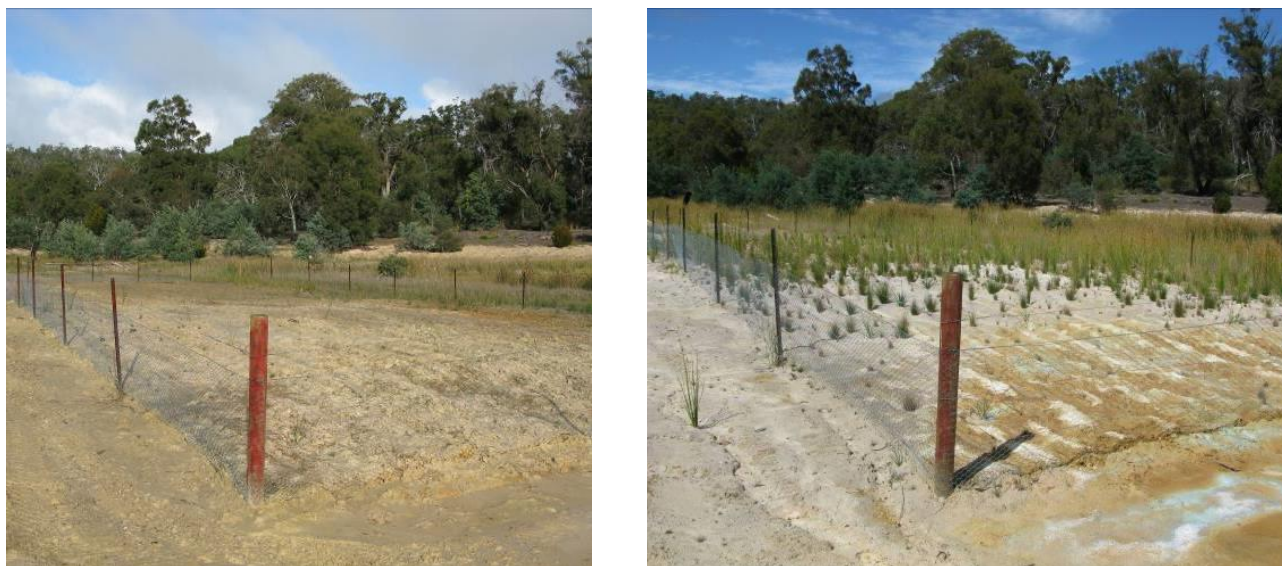


Figure 3 Field photograph showing the fenced-off vegetation trial plot at the Royal George Mine site. The limestone–and fertiliser–amended tailings have been colonised by *Juncus pallidus* between August 2011 (left) and January 2012 (right)

2.2 Methods

For the characterisation of plants and substrates at Royal George, 25 vegetated sites were selected. Tailings samples consisted of materials (depth 0–0.1 m) obtained from the root zone of *J. pallidus*. Tailings samples were oven dried at 40°C, sieved to <2 mm, thoroughly mixed and a 30–50 g aliquot pulverised in a chrome steel ring mill. For *J. pallidus*, the entire biomass growing above and below the tailings substrate was collected (n: 25). In the laboratory, the tissue embedded in the substrate (subterranean stems, rhizomes, roots) was separated from the above-substrate stems, root surfaces cleansed by repeated washing with water, and separate samples prepared. All vegetation samples were thoroughly washed and disaggregated, then dried in a fan-forced oven at 40°C for seven days. The resulting biomass was ground in a bench grinder and pulverised in a chrome steel ring mill.

Vegetation sample powders were digested using an HNO₃–HCl digestion technique, and samples were analysed by inductively-coupled plasma mass spectrometry (ICPMS) and inductively-coupled optical emission spectrometry (ICPOES) for selected elements (Al, As, Cd, Co, Cu, Fe, Mn, Ni, P, Pb, S, Th, Tl, U, W, Zn) at Intertek, Perth. Tailings samples were digested in a hot HF–HNO₃–HClO₄–HCl acid mixture and analysed by ICPMS and ICPOES for their near-total Ag, Al, As, Bi, Cd, Co, Cu, Fe, Mn, Pb, S, Sb, Sn, Th, Tl, U, W and Zn contents (Intertek, Perth). The S_{total} content of tailings was assumed to represent the Maximum Potential Acidity (MPA), which is appropriate considering the presence of acid-generating sulphate minerals within the samples as determined by X-ray diffraction (XRD) studies. Rinse pH and net acid generation (NAG) pH measurements (Smart et al., 2002) were carried out on powdered samples (Intertek, Perth). The acid neutralising capacity (ANC) of tailings was determined by standard acid reaction and titration (Intertek, Perth). The difference between MPA and ANC yielded the NAPP.

The mineralogy of tailings was identified using XRD. XRD traces were obtained from portions of the powdered samples after micronising with Siemens D500 and D501 diffractometers using Fe-filtered CoK_α radiation (University of Ballarat).

Solid speciation of trace elements in tailings can be evaluated using extraction techniques. In particular, we focussed on the effect of the added crushed limestone and fertiliser pellets to the tailings dump surface on the mobility of As. The solubility of As in Royal George tailings was evaluated using four leaching methods, where non-amended tailings samples (n = 2) were compared to limestone-amended tailings (n = 3) and

limestone- and fertiliser-amended tailings ($n = 2$). Three leaching techniques involved using DI water at different pH conditions as the extraction solution; DI water modified to pH 3 using concentrated H_2SO_4 , aged DI water (pH 5.6) and DI water modified to pH 10 using lime (CaO). These experiments aimed to establish the chemistry of ‘first flush’ pore and drainage waters. The fourth leaching technique involved the use of 0.5 M K_2SO_4 as the extracting solution since SO_4^{2-} is present in mine tailings environments due to the oxidation of arsenopyrite and other sulphide minerals. Each leaching solution of the selected tailings was extracted by allowing a 10 g tailings sample to be leached in 50 ml of the respective extraction solutions for 24 hours at room temperature while undergoing constant agitation. Following extraction, the supernatant was filtered using a 0.45 micron filter and the pH and electrical conductivity (EC) measured using Mettler Toledo S47 SevenMulti™ dual pH/conductivity meter. Leachate solutions were subsequently analysed by ICPMS for As (School of Earth Sciences, UTAS). The extraction procedures were applied: (a) to study the release of As from tailings into pore and pond waters; (b) to mimic leaching by rain and acidified surface waters and (c) to evaluate the mobility of As within the tailings in high SO_4 waters.

3 Results and discussion

3.1 Tailings

Exposed surface tailings are composed of major quartz, topaz, orthoclase and muscovite. Small amounts of plagioclase, biotite, chlorite, grossular, hematite, jarosite, pyrite, pyrophyllite, rozenite and rutile are also present. Tailings amended with limestone possess major calcite and in places detectable dolomite. The materials display major (i.e. >1 wt%) concentrations of Al and Fe, minor (i.e. >100 ppm) As, Cu, Mn, Pb, S, Sn and W, and traces (i.e. <100 ppm) of Ag, Bi, Cd, Co, Sb, Th, Tl, U and Zn. There are only few correlations between the analysed elements, for example As showed a correlation with Fe ($r = +0.86$). Rinse pH measurements of non-amended tailings confirm the fact that tailings were acid-generating prior to limestone amendment. Static testing of non-amended tailings revealed that the mean NAPP of the amended waste is slightly positive (mean + 2 kg H_2SO_4), reflecting minor acid generation of tailings. By contrast, limestone-amended tailings possess distinctly higher pH values (mean pH 8.3) and negative NAPP values (mean - 31.4 kg H_2SO_4). Hence, the limestone-amended tailings are non-acid generating. Localised acid leaching occurs in the dump, evidently where limestone application was insufficient to prevent the formation of acid pore waters (Figure 4).



Figure 4 Field photographs showing localised acid, Fe-stained seeps emanating from the tailings dump, Royal George. *J. pallidus* is evidently acid tolerant, colonising seepage areas

3.2 Biogeochemistry of *Juncus pallidus*

At Royal George, the vegetation patches within low-lying areas and depressions comprised only *J. pallidus* (Figure 5), and there did not appear to be any limitations on the growth of this plant. *J. pallidus*, growing in the tailings, has accumulated a range of trace element contents into its subterranean and above-substrate biomass. The plant species displays a general pattern of greater uptake of Al, As, Cu, Fe, P, S, W and Zn and lesser uptake of Cd, Co, Mn, Mo, Ni, Pb, Th, Tl and U in its subterranean and above-substrate tissues. The root tissue embedded into tailings has major (i.e. >1,000 mg/kg dry matter) concentrations of Al, Fe, P and S; minor (i.e. 100–1,000 mg/kg) As and W; subminor (i.e. 10–100 mg/kg) Co, Cu, Mn, Ni, Pb and Zn; and traces (i.e. <10 mg/kg) of Cd, Mo, Th, Tl and U. By contrast, the above-substrate biomass shows noticeably lower metal and metalloid contents, particularly lower Al, As, Co, Cu, Fe, Ni, Pb, Th, U and W values. It possesses major (i.e. >1,000 mg/kg dry matter) median concentrations of S; minor (i.e. 100–1,000 mg/kg) Al, Fe, P; subminor (i.e. 10–100 mg/kg) Cu, Mn, W and Zn; and traces (i.e. <10 mg/kg) of As, Cd, Co, Mo, Ni, Pb, Th, Tl, U. We interpret the elevated metal content of the subterranean plant tissue with caution, as it is extremely difficult to completely remove all particulates from root surfaces. However, the observation of lower metal and metalloid concentrations in the above-substrate biomass relative to the tailings remains robust. The detected element distributions are most likely indicative of: (i) subdued acquisition of elements (Al, As, Co, Cu, Fe, Ni, Pb, Th, Tl, U, W) into the above-substrate tissues and (ii) a relative enrichment of Cd, Mn and S into above-substrate stems.

The biogeochemical analyses demonstrate that the colonising rush has a pronounced trace metal tolerance and the tendency to exclude environmentally significant elements (Al, As, Co, Cu, Fe, Pb, Th, Tl, U, W, Zn) from its above-ground biomass. Concentrations of trace elements in *J. pallidus* were evaluated in terms of maximum tolerable levels in the feed of rodents, sheep and horses (National Research Council, 2005). The plant does not accumulate potentially toxic quantities of trace elements, with the exception of Cd, in its above-ground tissue.

Historic metal mining centres are known for their pronounced waste dispersion and elevated concentrations of metals in waters, soils and sediments (Lottermoser, 2010). However, such areas can also offer important natural field laboratories for the study of natural processes of mine site revegetation and biodiversification. Metal-excluding plants are of particular interest in the rehabilitation of contaminated land (Whiting et al., 2004). Such plant species do not acquire high metal concentrations in their biomass despite elevated metal concentrations in the soils. At Royal George, the exclusion of metals from the above-ground plant biomass of *J. pallidus* reduces the exposure of wildlife and grazing animals to metals and limits the transfer of metals up the food chain. The establishment of a *J. pallidus* cover on otherwise barren ground reduces tailings erosion and dust generation as well as water infiltration and associated leaching of contaminants. Such processes reduce the potential exposure of animals and humans to metals and may also open pathways for natural ecological restoration and biodiversification.



Figure 5 Field photographs showing the colonisation of limestone-amended tailings by *J. pallidus*

3.3 Leaching of tailings

The concentrations of As extracted from the tailings using four different leaching solutions were well above the World Health Organisation (WHO) guideline of 10 µg/L for five out of seven tailings samples. The average As concentrations of all four leaching methods for the non-amended tailings (n = 2), limestone-amended tailings (n = 3), and the limestone and fertiliser-amended tailings (n = 2) were 23 µg/L, 76 µg/L and 93 µg/L, respectively. To compare the proportion of the As released from the different tailings samples, the As concentration of the leachates were normalised to the total concentration of As in the tailings. DI water extractions showed the lowest relative As release for the limestone-amended and limestone and fertiliser-amended tailings. In contrast, non-amended tailings had the highest proportion of As release in DI water extractions (1.7 to 2.1) (Figure 6). In general, the relative As released in DI water extractions increased as the initial pH of the extracting solution increased. However, the final pH measured after 24 hours, showed no significant relationship with As released in DI water. The mean pH resulting from the three different DI water extractions was 3.9 for the non-amended tailings, 6.8 for the limestone-amended tailings and 7.9 for the limestone and fertiliser-amended tailings.

Dissolved As can be removed from AMD waters through: i) adsorption onto precipitated iron hydroxides and oxyhydroxides; ii) co-precipitation with hydrous oxides and hydroxides of Fe, Al, and Mg (McNeil and Edwards, 1997); and iii) the formation of calcium arsenates at higher pH conditions (Bothe and Brown, 1999). In this study, the alkaline pH conditions and the lower As release in the amended versus non-amended tailings suggest that the remediation efforts have been successful in reducing the mobilisation of As. However, the long-term stability of metal arsenates and calcium arsenates at neutral to high pH conditions has been questioned (Cheng et al., 2009).

Extractions using 0.5 M K₂SO₄, showed significantly different As release for the non-amended and amended tailings, compared to the DI water extractions. The relative As release was highest in the limestone and fertiliser-amended tailings (4.1), followed by the limestone-amended tailings (1.4). In contrast, the 0.5 M K₂SO₄ extraction of the non-amended tailings samples had the lowest As release (1.0). Thus, rehabilitation strategies for As-rich wastes need to consider the pronounced mobility of As at neutral to alkaline conditions, particularly in sulphate-rich waters. The final pH of the 0.5 M K₂SO₄ extractions compared to the average pH values from DI water extractions were significantly higher in the limestone-amended (+0.7 pH units) and limestone and fertiliser-amended tailings (+1.6 pH units), but little difference in the pH of the non-amended tailings (+0.04 pH units). AMD environments generally contain extremely high dissolved sulphate (>1,000 mg/L), therefore understanding the mobility of As in similar conditions is particularly important in evaluating effective amelioration strategies.

Competition between PO_4^{3-} and SO_4^{2-} anions of a similar charge density relative to arsenate for positively charged mineral surfaces has been documented to result in decreased adsorption and even desorption of arsenate (Xu et al., 1998; Jain and Loeppert, 2000), as well as arsenite (Wilkie and Hering, 1996). Furthermore, phosphate has been shown to be more effective at promoting arsenate desorption from minerals such as goethite than sulphate (O'Reilly et al., 2001). However, desorption experiments by Ladeira and Ciminelli (2004) demonstrates that the presence of sulphates in the leaching solution greatly promoted As desorption from oxisol, goethite and gibbsite. The addition of phosphate fertiliser pellets to the Royal George tailings has resulted in a three-fold increase in As leaching, relative to the limestone only amended tailings. The limestone- and fertiliser-amended tailings show a significant positive relationship ($R^2 = 0.99$) between pH and the averaged relative As leached (Figure 6). This is likely due to increasing desorption of As from iron oxide phases at higher pH conditions (Impellitteri and Scheckel, 2006). The combined presence of phosphate and sulphate, in addition to the higher pH recorded in the extracted solution of the limestone and phosphate-amended tailings, may have contributed to an elevated As release from the limestone- and fertiliser-amended tailings.

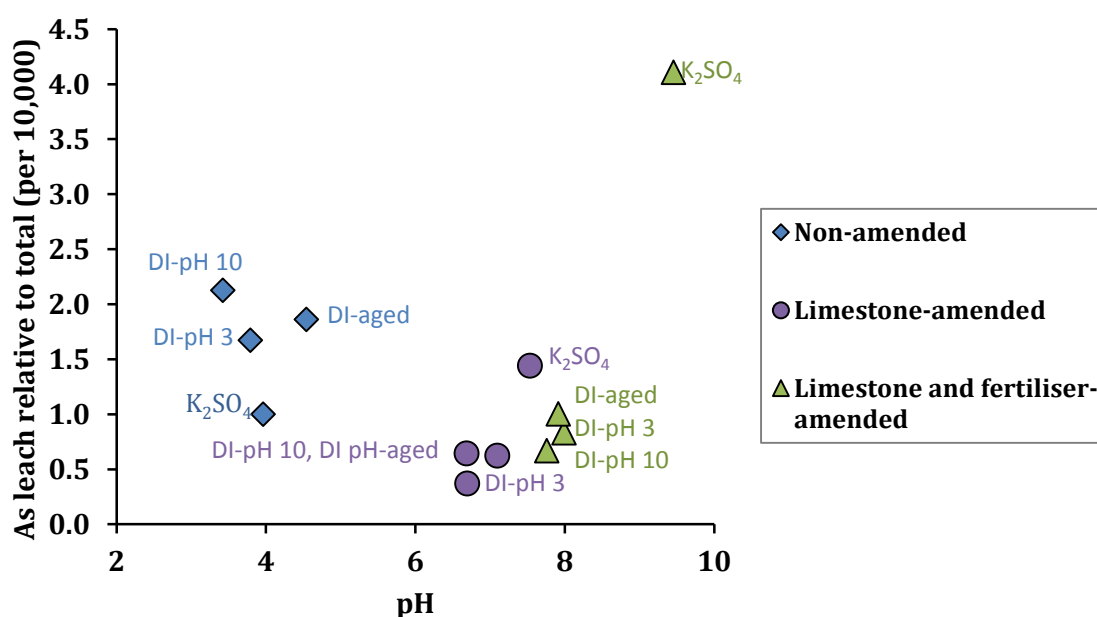


Figure 6 Average relative As released in different leaching solutions (DI water; aged, pH 3, pH 10, and 0.5M K_2SO_4) as a function of pH for the non-amended, limestone-amended and limestone and phosphate-amended tailings. The concentration of As in the leachates was normalised to the total concentration of As in the tailings, and due to the low concentrations, quantified as the proportion leached per 10,000 of the total concentration

At present, there are no measurements of the sulphate concentration of surface waters of the tailings disposal site, following amelioration works by statutory authorities in 2006. Measurements of the surface tailings sulphate concentrations prior to remediation works were between 98 and 331 mg/L, which is not particularly high with respect to AMD sites (Lottermoser, 2010). However, given the available information, it is likely that the limestone- and phosphate-amended tailings release less As in first flush events than the non-amended tailings. Further work is required to assess: (i) which mineralogical phases of the tailing hosts As; (ii) the current concentration of sulphate in site surface waters; (iii) the impact of increasing dissolved sulphate on increasing desorption of As from the limestone-amended tailings; and (iv) the concentrations of dissolved sulphate and phosphate above which As desorption occurs for different amendment strategies.

4 Conclusions

At Royal George, abandoned tailings dumps contain partly oxidised, sulphidic mine wastes that contain minor (i.e. >100 ppm) mean concentrations of As, Cu, Mn, Pb, S, Sn and W, and traces (i.e. <100 ppm) of Ag, Bi, Cd, Co, Sb, Th, Tl, U and Zn. Prior to remediation, the lack of rehabilitation combined with the high metal and metalloid tenor of the waste has led to significant AMD and associated metal and As mobilisation into local streams. In 2006, remediation by statutory authorities included a) minor earth works to prevent site access, (b) the construction of gravel armoured drains and drainage diversion channels; (c) the establishment of a fenced-off revegetation trial plot; and (d) the application of crushed limestone and fertiliser pellets to the tailings dump surface.

Today, the amended tailings remain largely devoid of any vegetation, with only some moist or ponded areas being colonised by the rush *J. pallidus*. Biogeochemical analyses demonstrate that the rush has a pronounced metal tolerance and excludes environmentally significant elements (Al, As, Co, Cu, Fe, Pb, Th, Tl, U, W, Zn), with the exception of Cd, from its above-ground biomass. The establishment of *J. pallidus* on otherwise barren ground reduces tailings erosion and dust generation, as well as water infiltration and associated leaching of contaminants. Furthermore, establishment of *J. pallidus* acts to reduce exposure of animals and humans to metals and may also open pathways for natural ecological restoration and biodiversification.

Leaching experiments were carried out to evaluate the mobility of As associated with the alkaline pH conditions of the limestone-amended tailings. A lower As release was detected in DI water leachates of amended tailings, relative to the non-amended tailings. This suggests that the amendment of tailings with limestone suppresses As release at the Royal George site. However, leaching in 0.5 M K₂SO₄ solution (which mimics the presence of sulphates associated with AMD conditions) showed an increased As release from amended tailings, compared to the non-amended tailings. Further work is required to assess: (i) which mineralogical phases of the tailing hosts As; (ii) the current concentration of sulphate in site surface waters; (iii) the impact of increasing dissolved sulphate on increasing desorption of As from the limestone-amended tailings; and (iv) the concentrations of dissolved sulphate and phosphate above which As desorption occurs for different amendment strategies.

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