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Mobility and retention of trace elements in hardpan-cemented cassiterite tailings, north Queensland, Australia

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Abstract This study reports on the mobility and retention of trace elements in cassiterite tailings at the inactive Jumna mill, tropical north Queensland. Since the 1980s, the uncapped tailings have developed laterally discontinuous Fe-rich hardpans, which are located in the higher parts of gently sloping tailings masses and at the top (< 50 cm) of the tailings piles. Hardpan-cemented tailings comprise thin layers (typically ~0.2–2 mm thick) of HFO (hydrrous ferric oxides) and sulfate efflorescences cementing tailings grains. In comparison to the tailings, the hardpan-cemented tailings contain significantly higher median As, Ca, Cd, Co, Cu, Fe, In, Mn, Mo, S_{total} , Th, U, Y and Zn values. Partial leaching studies of tailings and pond water analyses indicate that wetting and acidification of Fe-cemented tailings removes significant proportions of trace elements into

pore and surface waters. Tin shows no mobility due to the presence of weathering-resistant cassiterite (SnO_2) and, As and Pb display limited mobility possibly due to their coprecipitation with jarosite-type phases and HFO materials at the top of the tailings profile. By contrast, the trace elements Cd, Ce, Cu, La, Ni, Pb, U and Zn display the greatest mobility, possibly due to their incorporation in soluble sulfate efflorescences and sorption onto mineral and HFO surfaces. Hence, the Fe-rich hardpans do not protect the sulfidic tailings from further oxidation nor do they cause permanent sequestration of trace elements.

Keywords Cemented layer · Hardpan · Tailings · Geochemistry · Mineralogy · Acid mine drainage · Sulfide oxidation · Australia

Introduction

Oxidation of sulfide minerals in mine waste repositories may result in the production of low pH waters with elevated sulfate, ferrous iron, metal and metalloid concentrations. Upon changes to the pH-Eh conditions in the repository, secondary phases may precipitate as amorphous, poorly crystalline and well-crystallised products, cementing and encrusting tailings particles on a laterally extensive or discontinuous scale. As second-

ary mineral accumulation increases, cementing of the mine waste may occur, producing hardpans. Hardpans are defined herein as surface and subsurface secondary mineral accumulations that bind the exposed tailings into a continuous sheet or cement. The presence of Fe-rich hardpans as surface cements or at some depth within the profile has been reported for numerous sulfidic mine waste impoundments including waste rock dumps and tailings storage facilities (e.g. McSweeney and Madison 1988; Whitney et al. 1995; Lin and Herbert

1997). The cemented Fe-rich horizons occur as soft layers or solid cements, typically comprising non-crystalline hydrous ferric oxides (HFO) as well as crystalline alkali earth and metal sulfates, sulfur, jarosite, oxides and oxyhydrides (e.g. Blowes et al. 1991; Boulet and Larocque 1998; Agnew and Taylor 2000; Holmström and Öhlander 2001; Dill et al. 2002; McGregor and Blowes 2002; Gieré et al. 2003; Moncur et al. 2005). Coprecipitation and adsorption processes cause the hardpans to acquire elevated metal and metalloid contents. Also, laboratory studies have shown that cemented hardpans act as hydraulic permeability and diffusion barriers, reducing the transport of pore waters and gases (e.g. Gilbert et al. 2003). In turn, the rate of sulfide oxidation within tailings below the hardpans may be reduced and metals and metalloids are captured in the cemented layers, possibly leading to improved seepage and porewater quality (Stewart et al. 1993; Chermak and Runnells 1996; Ettner and Braastad 1999). However, the reduction in sulfide oxidation and the improvement to the water quality observed in the laboratory as well as the permanent sequestration of trace elements in hardpans remain to be confirmed by further field studies.

To date, studies conducted on hardpans in tailings impoundments have focused on gold, nickel and base metal tailings (Blowes et al. 1991; Boulet and Larocque 1998; Holmström and Öhlander 2001; Dill et al. 2002; McGregor and Blowes 2002; Gieré et al. 2003; Moncur et al. 2005). By comparison, hardpans of tin tailings remain largely unexplored, and there is little knowledge on the behaviour of trace elements in these environments. This paper reports on cassiterite (SnO_2) tailings of the inactive Jumna repository, north Queensland, Australia. Particular emphasis is placed in this work on hardpan-cemented cassiterite tailings as their properties highlight the mobility and retention of trace elements in stanniferous mine wastes.

Jumna plant site

Physiography, climate and vegetation

The Jumna mill site is located in north Queensland, 5 km north northeast of Irvinebank on the western slopes of the Atherton Tablelands at latitude $17^\circ 23.5'S$, longitude $145^\circ 13.5'E$ (Fig. 1). The area lies in an east-west trending valley adjacent to Chinaman Creek (Fig. 1). The region has a tropical savannah type climate, with an average annual rainfall of 1,146 mm, >80% of which falls between November and April (Herberton weather station, 16 km east of Jumna) (Bureau of Meteorology 2006). Average daily temperatures range from a winter minimum of 9.7°C to a summer maximum of 28.9°C . Savannah woodland, with dry eucalypt forest dominant, occurs throughout the area. In

the district, the main land use is low-density cattle grazing and the population density is sparse; none of the local surface streams are used as drinking water supplies for humans. Drainage from the Jumna plant site is via the west-flowing ephemeral Chinaman Creek (Fig. 1), a tributary of the Walsh River. It is only during the wetter months that there is significant surface water flow.

History of mining and mineral processing

The region contains historically significant areas of mineralisation for a range of metals (Sn, W, Mo, Ag, Au, Cu, Pb, Zn, Sb), and the Jumna plant site is located in one of the most intensively mined areas of Queensland

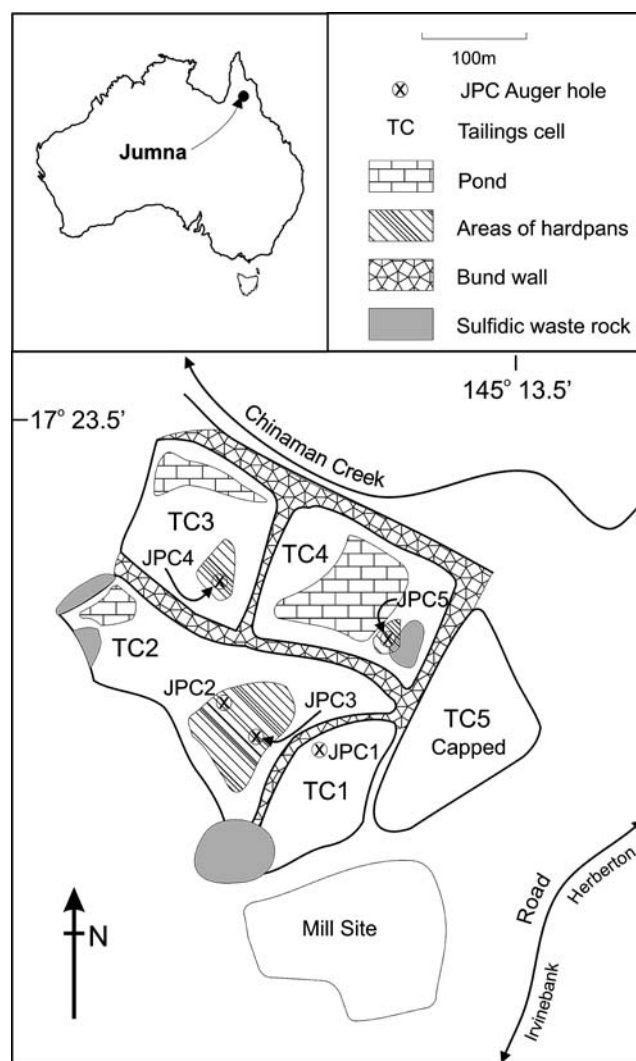


Fig. 1 Simplified plan of the Jumna mill area showing the location of the tailings cells, internal ponds, areas of hardpans, and sulfidic waste rock dumps. Locations of auger holes (JPC1 to JPC5) are also indicated

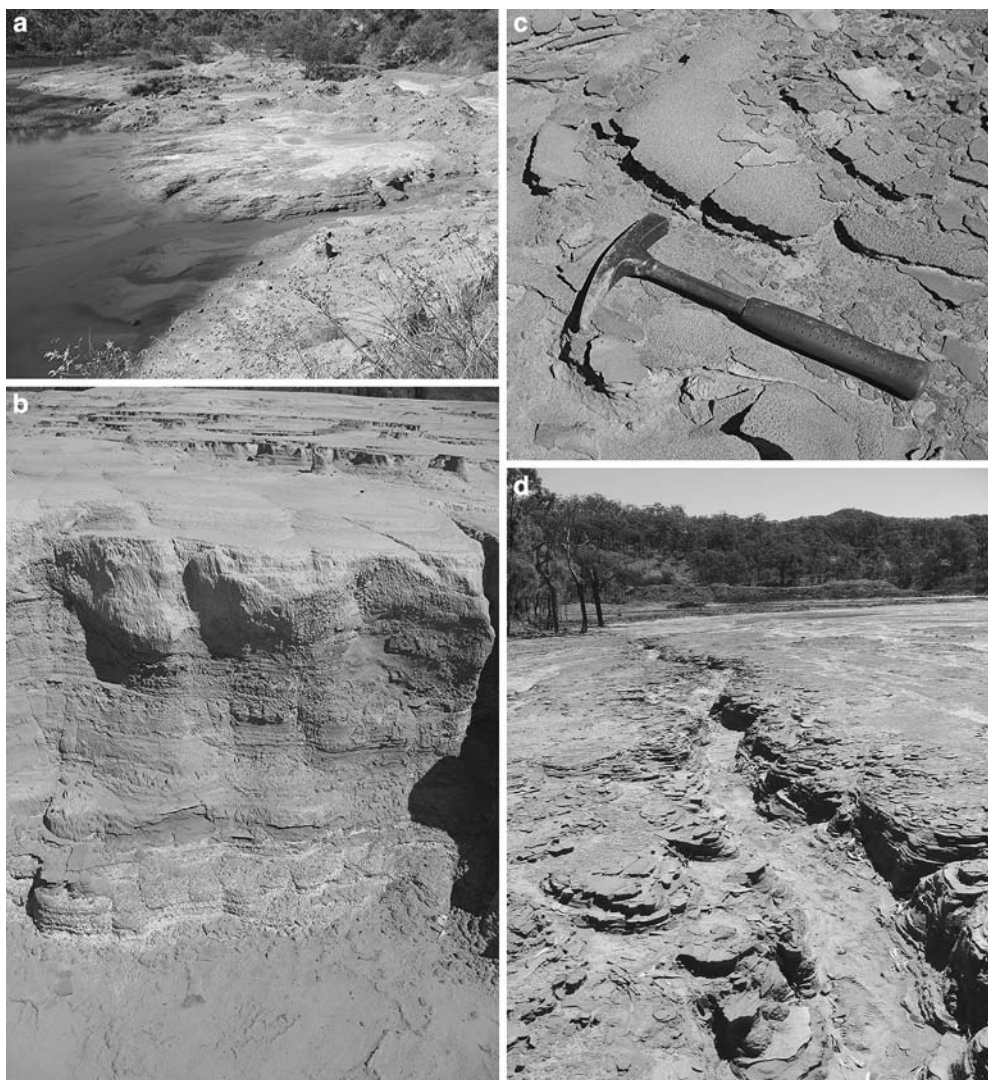


Fig. 2 Field photographs. **a** View of tailings cell TC4 towards the east. An internal drainage pond (pH 2.71; water sample JP4) and small sulfidic waste rock piles (*upper right-hand corner*) remain in the tailings cell. Bedded tailings with Fe-rich hardpans are partly covered by white mineral efflorescences (melanterite, gypsum) (*centre of photograph*). The sulfate salts on the edge of the pool are being dissolved and, mobilised Fe is transformed into HFO phases

that form incipient hardpan layers at the pond's edge. **b** Exposure of bedded silty to sandy tailings; TC3. White mineral efflorescences (gypsum) are present at the base of the exposed tailings face (field of view $\sim 0.5 \times 1$ m). **c** Solid crusts of Fe-rich hardpans, exposed at the top of tailings pile as a result of erosion; TC2 (field of view $\sim 0.5 \times 0.5$ m). **d** Erosion runnel (~ 30 cm wide, ~ 30 cm deep) in tailings pile showing multiple Fe-rich hardpan layers, TC2

(Dash et al. 1991). Hardrock tin deposits predominantly occur (a) as veins, pipes and breccias within shear zones and fractures in metasediments near granitoids and (b) as veins, pipes and disseminated deposits in granitoids or at the contact with metasediments (Dash et al. 1991). The tin deposits are related to post-tectonic S-type granites of the late Carboniferous O'Briens Creek Supersuite which released magmatic hydrothermal fluids into metasediments of the late Silurian to late Devonian Hodgkinson Formation (Dash et al. 1991).

Alluvial and hardrock tin deposits were discovered in 1875 and intermittently mined until the late 1980s. The

Jumna plant was one of several crushing plants in the district with mineral processing carried out from 1972 to 1989. The Jumna plant treated tailings accumulated in the Irvinebank tailings dam and hardrock Sn ores from the Irvinebank deposits (Jack in the Box, Jack Johnson, Mt Ormonde, Streak Hill—Lizzie, Governor Norman—Kelly Norman, Jumna) (J. Sainsbury, Unpublished report). These deposits are hosted in hydrothermally altered metasediments of the Hodgkinson Formation. The treated ores consists of major cassiterite and variable amounts of quartz, tourmaline, chlorite, garnet, kaolinite and sulfides (pyrite, chalcopyrite, galena,

arsenopyrite) (Dash et al. 1991). Mineral processing of unknown production ore quantities involved crushing, grinding and gravimetric sorting to produce a cassiterite concentrate. The processing plant closed in 1989 and, the tailings repository is estimated to contain 0.15 Mt tailings averaging 0.35% Sn (Garrad and Bultitude 1999).

Waste disposal and rehabilitation

The tailings disposal system consisted of a tailings repository with five tailings cells ($\sim 0.06 \text{ km}^2$) that were constructed in the early 1970s. The repository was formed by constructing a series of embankments within a small valley adjacent to Chinaman Creek. A basal liner was not installed beneath the tailings. Following closure of the plant in 1989, all tailings cells were left uncovered. Hence, atmospheric processes have led to sulfide oxidation and acid mine drainage (AMD). A previous environmental assessment of the site (Woodward-Clyde, unpublished report to Great Northern Mining Corporation NL) demonstrated that the sulfidic tailings are acid generating and possess elevated metal and metalloid concentrations (449 ppm Cu, 962 ppm Pb, 621 ppm Zn, 841 ppm As; <5 ppm Cd, 73 ppm Bi; 12 ppm Sb, 0.1 ppm Hg; mean of 29 samples). Surface waters ponding on the tailings developed low pH values (2.73), elevated electrical conductivity (3.9 mS/cm) and high metal (Al, Cd, Cu, Fe, Mn, Zn), metalloid (As) and sulfate values (Woodward-Clyde, unpublished report to Great Northern Mining Corporation NL). Subsequent surface water monitoring revealed that these contaminants are leached from the tailings repository into the adjacent Chinaman Creek. Consequently, in 1997 minor earthworks and rehabilitation works were conducted on the tailings cells (J. Sainsbury, Unpublished report). The sandy tailings in cell TC5 were seeded with grasses and a geomembrane liner was placed against the northern wall at tailings cell TC4 to prevent further seepage (Fig. 1) (J. Sainsbury, Unpublished report). However, tailings waters continue to seep from the repository into the adjacent creek as evidenced by low pH, high EC creek waters, mineral efflorescences and abundant Fe-rich precipitates (i.e. ochres or yellow boys) within Chinaman Creek downstream of the tailings repository. Thus, the site impacts on the quality of local surface waters, posing a threat to the health of grazing stock and wildlife.

Site description

Four of the five tailings cells (TC1 to TC4, Fig. 1) are the main foci of this study due to the fact that they are uncapped and exhibit AMD development and exposed

hardpans. Tailings cell TC5 may possess hardpans beneath its dry cover. Thicknesses of tailings in tailings cells TC1 to TC4 are estimated to range from $\sim 1 \text{ m}$ to probably at least 5–6 m (e.g. in the southern parts of TC2 and TC3). The mass of tailings is estimated at: 0.03 Mt in TC1; 0.11 Mt in TC2; 0.05 Mt in TC3 and 0.04 Mt in TC4. The uncapped cells locally develop drainage ponds within the tailings masses (Fig. 1). In the wet season, pond volumes are likely to be substantial, with depths up to $\sim 2 \text{ m}$ in TC2 and TC4 (Fig. 2a). The pond in TC4 appears to be permanent, being fed by a small AMD seepage emanating from tailings cell TC5. Cell TC4 has a small area of reeds and local bankside vegetation. Water in the ponds of TC2 and TC4 is clear, with Fe flocculants present on the bottom of the tailings ponds. Near the margins of these ponds, efflorescent salts are common, especially in TC4 (Fig. 2a). Wind and water dispersion of tailings has occurred and remains active in the uncovered ponds. There is no vegetation cover on any of the uncapped tailings cells, yet bund walls, constructed of local soils and tailings, are partly vegetated. Small sulfidic waste rock dumps occur at the periphery of several tailings cells (Fig. 1).

The tailings range from medium sand size to silty clay size (Fig. 2b). Iron-rich layers have been observed at the top of the tailings piles (Fig. 2c). These Fe-rich layers are consolidated to a varying degree, up to a few millimetres thick and commonly cement tailings grains. The layers are dominated by orange to brown, locally colloform banded Fe-rich layers, but locally, the crusts in TC3 have thin black Mn-rich coatings. In TC4, hardpan crusts commonly have melanterite–gypsum layers underneath. Hardpan development is largely restricted to the upper (southern) half of TC2, but small amounts are present on the upper (southern) part of TC3 and the upper (southeastern) part of TC4 (Fig. 1). Hardpan crusts are laterally discontinuous (10–100 cm) and only surficial and extend beneath the surface for less than a few tens of centimetres. In the immediate sub-surface, their manifestation appears to be brown-red Fe-rich layers, which are not consolidated. In erosion runnels, heavy mineral concentrations occur and multiple soft Fe-rich layers are evident (especially in TC2), to depths of at least 30–40 cm (Fig. 2d). Compact surficial hardpans appear to have formed by evaporative concentration and dehydration. All hardpans are present at the top level (<50 cm) within the tailings piles, occur in the higher parts of gently sloping tailings masses and dip as the tailings towards the transient ponds.

Materials and methods

Fieldwork was conducted during the dry season, with the collection of several different sample media (tailings, hardpan-cemented tailings, efflorescences, heavy mineral

concentrate, surface water). Samples of tailings were representative composites taken from depth intervals of auger holes (JPC1 to JPC5, Fig. 1) and constituted several kilograms (max. depth of coring 4 m, JPC4). Tailings were dry to moist, with the moisture content increasing with depth; yet, porewaters were not encountered during coring. Hardpans and efflorescent sulfate salts were obtained as crusts derived from tailings surfaces.

Tailings, hardpan-cemented tailings and heavy mineral concentrates were ground in a chrome-steel mill, whereas mineral efflorescences were ground with mortar and pestle in preparation for wet chemical, X-ray fluorescence (XRF) and X-ray diffraction (XRD) analyses. Processed tailings and hardpan samples were submitted to the James Cook University Advanced Analytical Centre (JCU AAC, Townsville) and Australian Laboratory Services (ALS, Brisbane) for analysis. Sample powders were chemically characterised for major and trace elements (As, Ba, Cu, Ga, Nb, Ni, Pb, Rb, Sc, Sr, V, Y, Zn, Zr) using a Bruker-AXS S4 Pioneer X-ray fluorescence spectrometer at the JCU AAC. Aliquots of sample powders were also digested in strong acids (HF–HNO₃–HClO₄) and analysed by inductively coupled plasma mass spectrometry (ICPMS) for Ag, Be, Bi, Cd, Ce, Co, Cs, Ge, In, La, Li, Mo, S_{total}, Sb, Se, Ta, Te, Th, Tl, U and W (ALS).

Selected tailings samples were dissolved in a H₂SO₄–HF acid mixture and analysed for their Fe²⁺ contents by titration using potassium permanganate (ALS, Brisbane). Total carbon (C_{total}) and organic carbon (C_{org}) concentrations in tailings samples were determined using a Leco furnace, while carbonate carbon (C_{carb}) was calculated by difference between the Leco methods (ALS, Brisbane). Sulfate sulfur (S_{sulfate}) was analysed using a dilute HCl digestion of sample powders followed by inductively coupled plasma atomic emission spectrometry (ALS, Brisbane). The sulfide sulfur (S_{sulfide}) content of tailings samples was calculated by difference between the total sulfur (S_{total}) and sulfate sulfur (S_{sulfate}) value. Quality control/assurance of the data was applied using analyses of sample replicates, blanks and international reference standard GXR-2. Deviations of the median value from the listed compilation value are less than 5% for Ag, Ba, Be, Ca, Ce, Cu, Mg, Na, S, Sr, W and Zn and less than 10% for As, Cd, Co, Cs, Fe, La, Rb and Te.

Solid speciation of trace elements in tailings can be evaluated using extraction techniques. The solubility of selected major and trace elements in Jumna tailings was evaluated using two leaching methods. The distilled-water fraction of the tailings was extracted by allowing a 2 g tailings sample to be leached in 100 ml of distilled water for 24 h at room temperature while undergoing constant agitation. The experiment aimed to establish the chemistry of ‘first flush’ pore waters and drainage

waters emanating from the tailings. The acid-leachable fraction was determined by leaching a 2 g tailings sample in 30 ml of 20% HCl for 20 min at room temperature. Sample solutions were subsequently analysed by ICPMS for Al, As, Ca, Ce, Cd, Cu, Fe, K, La, Mg, Mn, Na, Ni, Pb, S_{total}, U and Zn (JCU AAC). The extraction procedures were applied: (a) to study the release of the trace elements from tailings and hardpan-cemented tailings into pore and pond waters; (b) to mimic leaching by rain and surface waters and (c) to evaluate the retention of trace elements within the cemented tailings.

The mineralogy of tailings, hardpan and efflorescence samples was identified using XRD (Cu K α ; Siemens D5000 X-ray diffractometer) and computational software (EVA) at the JCU AAC. Optical microscopy, scanning electron microscope (SEM) observations and semi-quantitative energy dispersive spectrometry (EDS) electron microprobe analyses were conducted on polished blocks of tailings and hardpans using a Jeol JSM-5800LV with energy dispersive microprobe attachment (University of New England, Armidale). Paste pH measurements were performed on powdered tailings following the procedure by Morin and Hutt (1997). Such pH measurements provide a preliminary evaluation of the tailings’ acid generation potential.

Water samples were collected from ponded waters of three tailings cells and Chinaman Creek downstream of the repository. Selected water quality parameters (temperature, pH, salinity, dissolved oxygen, conductivity) were determined in the field at all sampling sites using a Horiba water quality checker. Water samples were collected in acid washed plastic bottles, whereby samples for metal analyses were filtered in the field using a 0.45 μ m filter and preserved using HNO₃. Water samples were stored on ice and submitted for immediate analysis at ALS. Water samples were analysed for general parameters (conductivity, total dissolved solids) and chemical compositions. Methods included inductively coupled plasma atomic emission spectrometry (Na, K, Ca, Mg), gravimetry (SO₄), titration (HCO₃), specific ion electrode (F) and argentometric technique (Cl). Filtered (0.45 μ m) water samples were also analysed for dissolved Ag, Al, As, B, Ba, Be, Bi, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Hf, Ho, In, La, Li, Lu, Mn, Mo, Nd, Ni, Pb, Pr, Rb, Sb, Se, Sm, Sn, Sr, Tb, Te, Th, Tl, Tm, U, V, Y and Zn using a Varian UltraMass 700 inductively coupled plasma mass spectrometer (JCU AAC).

Results

Tailings

The uncovered tailings cells are largely in the same form as they existed when mineral processing stopped in 1989.

Table 1 Geochemistry of sulfidic tailings ($n = 16$)

Element	Minimum	Maximum	Arithmetic mean	Median
Major elements (wt%)				
Al	3.95	8.15	5.95	5.90
C _{total}	0.05	0.30	0.13	0.10
C _{org}	0.05	0.25	0.12	0.10
C _{carb}	< 0.02	0.05	< 0.02	< 0.02
Ca	0.06	0.46	0.14	0.10
F	0.10	0.50	0.19	0.16
Fe _{total}	3.88	8.80	6.41	6.67
Fe ²⁺	1.24	3.39	2.60	2.90
K	1.21	2.70	1.93	1.94
Mg	0.24	0.54	0.39	0.39
Mn	0.03	0.12	0.08	0.08
Na	0.19	0.35	0.29	0.29
P	0.02	0.04	0.03	0.03
S _{total}	0.06	1.16	0.45	0.42
S _{sulfide}	0.02	1.01	0.28	0.14
S _{sulfate}	0.04	0.47	0.13	0.09
Si	28.19	38.19	32.72	32.63
Sn	0.16	0.41	0.29	0.29
Ti	0.14	0.43	0.28	0.29
Trace elements (ppm)				
Ag	2.1	10.9	5.9	5.9
As	149	2,176	801	593
Ba	178	368	259	260
Be	2	8.4	3.8	3.4
Bi	15.3	251	92.2	72.9
Cd	0.02	5.9	2.1	1.3
Ce	56.3	208	116	115
Co	1.3	7.7	4.0	3.8
Cs	12.6	39.7	26.9	25.6
Cu	101	1,197	535	456
Ga	14	29	22.3	22.5
Ge	0.12	0.32	0.22	0.23
In	2.4	14.1	8.4	8.4
La	25.9	94.8	55.6	55.4
Li	22.3	163	81.8	88
Mo	0.59	7.3	3.3	2.9
Nb	12	32	22	21.5
Ni	8	34	18.3	17.5
Pb	144	853	469	490
Rb	151	620	311	287
Sb	3.3	28.7	14.2	12.9
Sc	7	10	8.4	9
Se	1	6	2	2
Sr	15	41	23.4	21.5
Ta	0.6	1.9	1.1	1.1
Te	0.05	0.31	0.16	0.14
Th	10.4	34.8	19.5	18.7
Tl	2.1	4.9	3.6	3.6
U	2	8.7	4.6	4.5
V	18	41	33.9	34.5
W	13.8	138	59.5	51.9
Y	20	90	45.4	41.5
Zn	167	1,068	534	447
Zr	106	245	131	125
Paste pH	2.7	3.8	3.2	3.3

Tailings are physically heterogeneous. The material ranges from clay to sand size. Surficial weathering of the tailings has led to contemporary formation of mineral efflorescences at and immediately below the tailings surface (Fig. 2a). Local efflorescent salt developments

on tailings are apparently transient and form after each rain event. They include phases such as jarosite, gypsum, roostite, pentahydrate and melanterite as indicated by XRD studies.

Tailings samples from the different cells display no major differences in bulk composition. The samples have major (i.e. > 1 wt%) median concentrations of Al, Fe_{total}, Fe²⁺, K and Si; minor (i.e. > 0.1 wt%) F, Mg, Na, S_{total}, S_{sulfide}, Sn and Ti; sub-minor (i.e. > 100 ppm) As, Ba, C_{total}, C_{org}, Ca, Ce, Cu, Mn, P, Pb, Rb, S_{sulfate}, Zn and Zr; traces (i.e. < 100 ppm) of Ag, Be, Bi, Cd, Co, Cs, Ga, Ge, In, La, Li, Mo, Nb, Ni, Sb, Sc, Se, Sr, Ta, Te, Th, Tl, U, V, W and Y; and non-detectable C_{carb} (Table 1). The element distributions are consistent with the occurrence of relatively abundant primary aluminosilicates (i.e. quartz, K-feldspar, plagioclase, microcline, muscovite, chlorite, kaolinite), magnetite and secondary minerals (i.e. gypsum, jarosite, diaspore, hematite) as well as traces of hornblende, beryl, fluorite and unidentified expanding clays as indicated by XRD and petrographic studies. The presence of cassiterite and sulfides (i.e. pyrite, chalcopyrite, galena, arsenopyrite) is evident in polished blocks and supported by the known mineralogy of processed ores and the abundance of S_{sulfide}, Fe²⁺, As, Cu, Pb and Zn in the analysed tailings (Table 1).

Paste pH measurements of milled tailings confirm the fact that the waste is acid producing as wastes have a median pH value of 3.3 (Table 1). Tailings in the upper tailings pile (i.e. < 1 m from the surface) are distinctly acid (paste pH 2.7–3.2). By contrast, tailings from lower parts of the tailings pile (i.e. > 1.3 m from the surface) have a less acidic paste pH (3.3–3.8). Hence, the net acid producing potential (NAPP) of these materials is highest at the top of the tailings pile. On the other hand, the tailings have a low acid neutralisation capacity (ANC) due to insignificant carbonate contents (median C_{carb} < 0.02 wt%) and the abundance of quartz and aluminosilicates.

Hardpan-cemented tailings

Hardpan-cemented tailings are chemically homogenous and display major (i.e. > 1 wt%) median concentrations of Al, Fe, K, S_{total} and Si; minor (i.e. > 1,000 ppm) As, Ca, Cu, F, Mg, Na, Sn and Ti; sub-minor (i.e. > 100 ppm) Ba, Ce, Mn, P, Pb, Rb and Zn; and traces (i.e. < 100 ppm) of Ag, Be, Bi, Cd, Co, Cs, Ge, In, La, Li, Mo, Nb, Ni, Sb, Se, Sr, Ta, Te, Th, Tl, U, V, W, Y and Zr (Table 1). In comparison to the tailings, the hardpan-cemented tailings contain significantly higher median As (3x), Ca (2x), Cd (3x), Cu (3x), Fe (2x), In (2x), Mo (2x), S_{total} (4x), Th (2x) and Zn (2x) values (Tables 1, 2). Hardpan impregnated tailings have the same major mineralogy as the tailings as shown by XRD

Table 2 Geochemistry of hardpan-cemented tailings ($n=7$)

Element	Minimum	Maximum	Arithmetic mean	Median
Major elements (wt%)				
Al	4.70	4.71	4.44	4.90
Ca	0.12	0.93	0.32	0.18
F	0.15	0.94	0.31	0.22
Fe _{total}	12.72	18.56	15.53	15.57
K	1.39	2.09	1.68	1.59
Mg	0.31	0.53	0.36	0.35
Mn	0.08	0.31	0.14	0.09
Na	0.24	0.42	0.28	0.26
P	0.01	0.05	0.04	0.04
S _{total}	1.29	2.92	1.66	1.54
Si	22.56	26.33	24.22	24.14
Sn	0.20	0.50	0.31	0.28
Ti	0.09	0.22	0.18	0.19
Trace elements (ppm)				
Ag	2.7	10.8	6.3	6.1
As	528	6,680	2,893	1,549
Ba	102	235	202	219
Be	2.5	6.3	3.9	3.8
Bi	22.2	70.5	42.9	42.2
Cd	2.8	13.9	5.6	3.6
Ce	108	233	133	114
Co	2.7	13.5	6.7	3.8
Cs	19.1	23.8	21	21.4
Cu	1,307	7,573	2,273	1,377
Ge	0.3	0.4	0.3	0.3
In	11.6	18.9	15	15.6
La	49.8	107	63.3	56.3
Li	34.7	121	72.9	66.2
Mo	1.1	8.7	5.4	6.6
Nb	8	20	11	10
Ni	15	56	22.4	17
Pb	250	915	535	507
Rb	182	477	292	275
Sb	9.2	12.9	11.1	11.1
Se	1	4	2.1	2
Sr	13	30	19	19
Ta	0.7	1.6	1	0.9
Te	0.1	0.17	0.13	0.13
Th	13.8	94.4	36.5	33.9
Tl	2.8	3.6	3.1	2.9
U	3.1	22.5	6.7	4.4
V	10	64	44.7	51
W	21.4	57.8	43.2	46.7
Y	44	195	74	55
Zn	635	4,472	1,466	888
Zr	57	124	98	100
Paste pH	2.6	3.1	2.86	2.8

and petrographic studies. While goethite replacing detrital sulfide grains and hematite replacing detrital magnetite grains were observed in petrographic studies, powder XRD analyses of Fe-cemented tailings did not reveal any crystalline Fe oxide or hydroxide phases. Hence, the tailings are cemented by sulfate efflorescences and non-crystalline HFO phases.

Microscopically, tailings samples and hardpan-cemented tailings are similar, displaying thin layers (typically ~0.2–2 mm thick) of more strongly HFO-cemented tailings interlayered with material that con-

tains less HFO. The HFO phases cement detrital grains and variably impregnate finer-grained matrix material (e.g. clay, mica) (Fig. 3). Detrital grains in the cemented tailings are dominated by quartz, with lesser amounts of mica and minor to trace amounts (low percentage values to <<0.1%) of a suite of heavy minerals. The individual heavy mineral grains and aggregates are mostly <0.2 mm across, but are locally coarser-grained (up to 0.6 mm). Sulfide grains are invariably replaced by goethite. Semi-quantitative electron microprobe analyses of HFO material (possibly goethite) revealed wt% concentrations (>1 wt%) of Al, Si, S and K and lower contents (<1 wt%) of Mg, Mn, Ti, Cu and Zn. The elements As and Pb have been detected in individual HFO material (<0.05–0.1 wt%), yet their concentrations could not be quantified.

Paste pH measurements of the hardpan-cemented tailings indicate the fact that the materials are acid producing as samples average a pH value of 2.8. Hence, the wetting of cemented tailings leads to the oxidation of sulfides, release of sorbed hydrogen ions and the dissolution of acid producing mineral efflorescences (e.g. melanterite).

Partial leaching

Partial leaching of elements from tailings and hardpan-cemented tailings using distilled water largely resulted in similar extraction percentages (Table 3). For the tailings, the distilled water treatment dissolved major (i.e. >1%) percentages of Ca, Cd, Cu, Mg, Mn, Ni, S_{total} and Zn; traces (<1%) of Al, As, Fe and Na; and non-detectable amounts of Ce, K, La, Pb and U (Table 3). The extraction percentages do not relate to the total element concentrations and there is no significant positive correlation (>+0.5) between water extractable and total element concentration in tailings, with the exception of Ca and Cd (0.8 and 0.7, respectively).

By comparison, distinctly higher element percentages were extracted from the hardpan-cemented tailings (Table 3). The distilled water treatment dissolved major (i.e. >1%) percentages of Ca, Cd, Ce, Cu, La, Mg, Mn, Na, Ni, S_{total}, U and Zn; traces (<1%) of Al and Fe; and non-detectable amounts of As, K and Pb (Table 3). Also, extraction percentages relate to the total element concentrations, and there is significant positive correlation (>+0.9) between water extractable and total element concentration in the hardpan-cemented tailings, with the exception of Al and Fe.

Tailings taken from the upper waste horizon, containing hardpans (i.e. <1 m from the surface), have distinctly elevated percentages of water-soluble Ca, Cd, Cu, Mg, Mn, Ni, S_{total} and Zn (Fig. 4). Tailings from the lower parts of the tailings pile (i.e. >1.3 m from the surface) release lesser proportions of alkali earth ele-

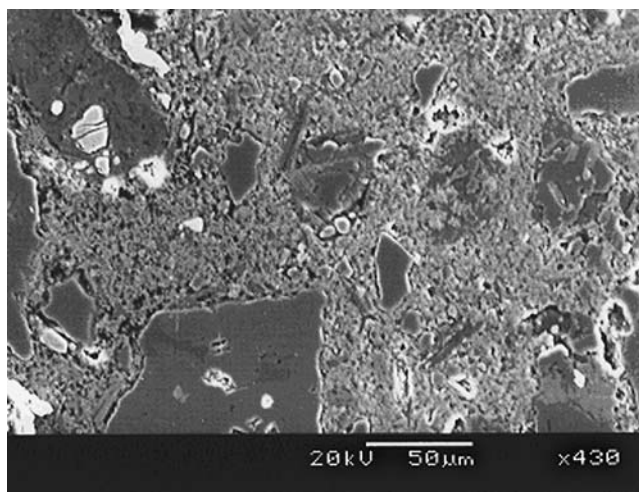


Fig. 3 Back-scattered electron image of Fe-rich hardpan. HFO phases (*medium grey*; matrix) impregnate detrital tailings grains (*dark grey*; mainly quartz). Traces of detrital Fe oxide phases (hematite and magnetite) are seen as *bright spots*

ments, metals and sulfate. Therefore, it can be argued that much of the alkali earth elements, metals and sulfate occur in abundant and readily soluble forms at the top of the tailings pile. Such soluble forms likely include water-soluble secondary sulfate minerals (i.e. gypsum, rostitite, pentahydrate, melanterite) as well as exchangeable ions.

The HCl reagent leached significantly higher percentages than the distilled water treatment from both sample types (Table 3). The treatment extracted major

Table 3 Average extraction percentages of elements using water and HCl leaching of tailings and hardpan-cemented tailings

	H ₂ O leach		HCl leach	
	Tailings	Cemented tailings	Tailings	Cemented tailings
Al	0.01	0.33	0.14	0.40
As	0.23	ND	5.24	3.91
Ca	14.56	52.64	29.55	47.86
Cd	37.00	29.55	45.60	38.11
Ce	ND	8.04	6.93	11.84
Cu	4.75	11.63	16.03	15.70
Fe	0.02	0.01	1.87	1.36
K	ND	ND	ND	ND
La	ND	6.80	5.56	9.51
Mg	2.54	21.68	3.48	18.79
Mn	11.49	43.14	16.36	37.41
Na	0.68	3.26	0.91	3.34
Ni	3.14	38.49	4.86	37.60
Pb	ND	ND	8.62	1.07
S _{total}	16.04	27.80	16.00	26.92
U	ND	22.37	15.77	38.29
Zn	7.73	21.30	10.72	21.55

The concentrations of the extractions have been averaged for the tailings ($n=16$) and hardpan-cemented tailings ($n=7$) and calculated as a percentage of the total
ND not detected

(i.e. >1%) percentages of As, Ca, Cd, Ce, Cu, Fe, La, Mg, Mn, Ni, Pb, S_{total}, U and Zn, traces (<1%) of Al and non-detectable amounts of K. In the case of hardpan-cemented tailings, samples with higher element concentrations tend to have higher HCl extractable element values, and the extraction percentages (As, Ca, Cd, Ce, Cu, La, Mg, Mn, Na, Ni, S_{total}, U, Zn) correlate to the total element concentrations strongly (> +0.9), with the exception of Al and Fe. By comparison, the correlations between extractable and total element values in the tailings are significantly increased for most elements. Thus, the water-soluble secondary sulfate minerals in the hardpan impregnated tailings release much of the total element load upon acidification. Acidification removes >1% of As, Ca, Cd, Ce, Cu, Fe, La, Mg, Mn, Ni, Pb, S_{total}, U and Zn from the tailings, hence significant acid leaching of these elements can be expected to occur from the oxidising sulfidic tailings into pore and surface waters.

In all leaching treatments, the amount of water and HCl extractable Al, As, Fe, K, Na and Pb was much less than that of the other elements. Such low water and HCl extractable concentrations reflect the mineralogical siting of these elements. XRD and SEM studies have shown that Al, As, Fe, K, Na and Pb are hosted and immobilised by feldspars, sulfides and HFO phases. The elements are present in relatively stable mineral phases that do not readily undergo HCl dissolution.

Ponds

Water levels within the tailings ponds vary and reach a depth of ~2 m during the wet season. During the dry

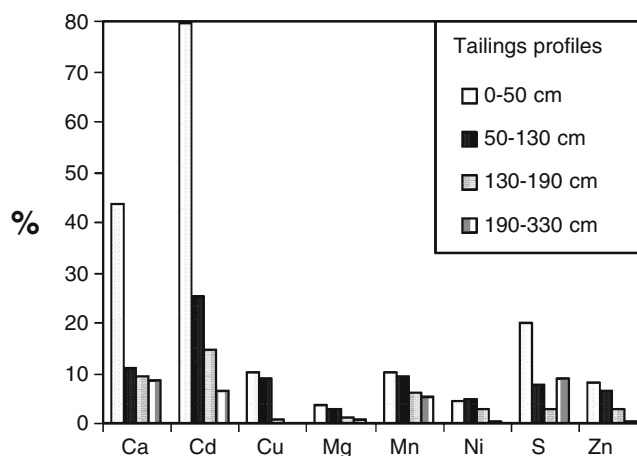


Fig. 4 Distribution of water-soluble Ca, Cd, Cu, Mg, Mn, Ni, S and Zn concentrations expressed as percentage of total element concentrations in the JPC3 tailings profile, Jumna. Tailings taken from the upper waste horizon (0–50 cm), containing hardpans, have the highest percentages of water-soluble elements

season, the ponds contain acid (pH 2.5–3.9), saline (1.8–3.4 mS/cm) surface waters which are Ca-SO₄-rich with elevated Al, Fe, Mn and F as well as elevated trace element contents, in particular As, Be, Cd, Co, Cu, Ni, Pb, Se, Th, U, Zn and REE (rare earth element) concentrations (samples JP1, JP2 and JP4, Table 4). Differences in dissolved element (e.g. Fe) concentrations between ponds are likely related to different pH and redox conditions in the pond waters. Pond waters of tailings cells seep into the local creek. In particular, the geomembrane liner placed against the northern wall at tailings cell TC4 does not prevent impacts of the AMD pond waters on the adjacent Chinaman Creek. The pond waters contribute significant acidity and major elements (SO₄, F, Al, Fe, Mn) yet also deliver distinctly elevated trace element levels (i.e. Be, Cd, Co, Cu, Ni, Pb, Se, Th, U, Zn, REE) to the local creek system (sample JP3, Table 4). Chinaman Creek water has pH values as well as Al, Cd, Cu, F, Pb, Se and Zn contents that exceed the Australian water quality guideline values for stock water (cattle) (NWQMS 2000) (Table 4).

Discussion

Hardpan formation

At Jumna, all HFO-rich hardpans are present at the top level (~50 cm) within the tailings piles and occur in the higher slopes of the gently sloping tailings masses. Their presence does not coincide with the transient water levels in the individual tailings ponds and hence, is unlike reported Fe-rich layers formed at the tailings–pond water interface (Holmström and Öhlander 2001). Therefore, hardpan formation must have commenced immediately after tailings disposal. During milling, disgorged mill fluids emanated onto the large surface area of a gently sloping mass of tailings. The seasonally dry climate allowed evaporation of tailings fluids and oxidation of sulfidic tailings to occur. The release of Fe²⁺ from sulfide grains would be followed by Fe²⁺ oxidation, hydrolysis of Fe³⁺ and precipitation of HFO phases. Such processes initiated the genesis of Fe-rich hardpans on the higher slopes of the tailings masses. In addition, a thin layer of water at high slopes would allow greater evaporation during the seasonally dry climate, favouring precipitation of HFO phases.

The presence of unconsolidated hardpans and the occurrence of HFO precipitates associated with the dissolution of secondary melanterite efflorescences (Fig. 2a) point to the contemporary formation of HFO phases. Active sulfide oxidation of tailings in the upper tailings pile, as evidenced by petrographic observations and paste pH measurements, lead to Fe³⁺ release to pore and surface waters, subsequent Fe³⁺ hydrolysis and HFO formation. Also, soluble hydrated sulfates

including the Fe²⁺-bearing melanterite are cementing phases in the top of tailings piles (cf. Blowes et al. 1991). The dissolution of transient melanterite during rainfall events leads to the oxidation of Fe²⁺, hydrolysis of Fe³⁺ in pore and surface waters and the formation of incipient HFO “softpan” layers.

Trace element mobility and retention

Previous studies on tailings repositories have shown that the release of elements from tailings depends on the chemical and physical properties of the wastes (McGregor et al. 1998; Craw 2003; Ashley et al. 2003; Lottermoser and Ashley 2005; Moncur et al. 2005). Initially, the stability of tailings minerals to weathering processes determines whether elements can become mobile. Surface controlled dissolution of minerals and sulfide oxidation will liberate elements to porewater solutions. At Jumna, the high proportion of S_{sulfide} to C_{carb} in the wastes favours acid pH conditions in porewaters. The acid pH fluids present in the pore network of the tailings promote the dissolution of trace elements from ore and gangue minerals. Weathering and leaching of tailings result in the release of sulfate, fluoride, alkali earth elements, metals, metalloids and rare earth elements into pore waters.

The precipitation of secondary phases as well as sorption and coprecipitation processes influence whether trace elements are released into tailings pore and pond waters or are retained within the tailings. The precipitation of HFO phases and sulfate efflorescences selectively captures As, Ca, Cd, Co, Cu, Fe, In, Mn, Mo, S_{total}, Th, U, Y and Zn to a significant degree at the top of the tailings pile, as indicated by their elevated concentrations in the hardpan-cemented tailings (Table 2). However, the capture of these elements is not permanent as shown by their ready release into acid leachates (Table 3) and elevated contents in pond waters (Table 4).

The generation of pore and drainage waters from sulfidic mine wastes in a seasonally wet climate is controlled by wetting and drying cycles (Harris et al. 2003). Waste piles are intermittently wetted by meteoric water and seasonal run-off; they are dried by drainage and evaporation (Harris et al. 2003). In the case of the partly oxidised tailings with high concentrations of soluble elements like Jumna, intense wetting of these materials will release some of the temporarily stored acidity and elements. Seasonal wetting and drying of and drainage from the tailings repository lead to the release of sorbed elements from mineral surfaces as well as the dissolution of the previously precipitated alkali earth and metal sulfate salts. As a result, the ‘first flush waters’ will possess significant acidity, salinity and element contents. The pond waters associated with the cassiterite tailings

Table 4 Analyses of surface waters from the tailings ponds, Jumna mill site

	Tailings cell no. 1	Tailings cell no. 3	Tailings cell no. 4	Chinaman Creek	Guideline values Livestock (cattle) WQ
Sample number	JP1	JP2	JP4	JP3	
General parameters					
pH	2.57	3.94	2.71	3.24	6–9
Conductivity (mS cm ⁻¹)	2.74	1.81	3.41	2.82	
TDS (mg L ⁻¹)	1,860	1,190	2,220	1,950	10,000
Major ions (mg L ⁻¹)					
Ca	27	188	178	199	
Mg	23	68	90	95	
Na	1	16	27	29	
K	<1	5	12	12	
Al	129	30	53	60	5
Fe	148	0.2	65.4	13.7	
Mn	7.2	64.2	45.1	42.9	
HCO ₃	<1	<1	<1	<1	
SO ₄	1,530	1,020	1,960	1,890	2,000
Cl	<1	21	82	21	
F	1.9	12	4.1	5	2
Trace elements (µg L ⁻¹)					
As	772	15	37	19	500
Ba	6	14	3	19	
Be	19	45	187	321	
Cd	291	190	236	215	10
Ce	258	801	4,310	4,630	
Co	192	373	451	464	1,000
Cr	68	<1	11	12	1,000
Cs	3	<1	5	4	
Cu	30,800	6,650	11,300	7,770	1,000
Dy	34	67	502	579	
Er	19	36	299	344	
Eu	4	8	17	17	
Ga	9	5	21	25	
Gd	37	85	549	612	
Ho	6	13	100	118	
In	121	<1	4	3	
La	86	346	1,700	1,960	
Li	382	483	740	706	
Lu	2	4	38	42	
Mo	2	2	2	2	150
Nd	151	351	1,950	2230	
Ni	257	288	346	377	1,000
Pb	607	6	115	266	100
Pr	37	95	534	590	
Rb	14	34	126	134	
Se	20	20	90	100	20
Sm	36	74	479	537	
Sn	5	<1	<1	<1	
Sr	16	208	437	531	
Tb	6	13	89	103	
Th	79	<1	41	38	
Tm	3	5	44	49	
U	36	17	167	181	200
Y	198	482	3,310	3,860	
Yb	17	26	252	281	
Zn	63,700	31,900	24,900	23,100	20,000

Composition of surface water from Chinaman Creek is also shown (AMD impacted creek water). Samples JP1 to JP4 possess non-detectable Ag (<1 µg L⁻¹), B (<100 µg L⁻¹), Bi (<1 µg L⁻¹), Hf (<10 µg L⁻¹), Sb (<1 µg L⁻¹), Te (<5 µg L⁻¹), Ti (<10 µg L⁻¹), and V (<50 µg L⁻¹)

Livestock (cattle) WQ Australian water quality guideline values for livestock (cattle) (NWQMS 2000)

have median trace metal, metalloid and rare earth element concentrations in the order of Zn > Cu > Ce > La > Ni > Cd > U > As > Sn > Sb (Table 4), whereas the tailings have relative median element concentrations in the order of Sn > As > Cu > Pb > Zn > Ce > La > Ni > Sb > U > Cd

(Table 2). Such distinctly different relative concentrations of trace metals, metalloids and rare earth elements are related to the different chemical mobility and solubility of elements in the tailings cells. On the basis of the water extraction experiments and the pond water chemistry, the preferential release of metals, metalloids and rare earth elements from the cassiterite tailings follows the generalised sequence of $Cd > U > Ni > Zn > Cu > Ce > La > Pb > As > Sn$ (Tables 3, 4). The resultant 'first flush waters' possess elevated sulfate and alkali earth element, metal (Cd, Cu, Fe, Mn, Ni, Pb, U, Zn), metalloid (As) and rare earth element (Ce, La) values. These mobilised elements migrate from the tailings into pond waters where they remain in solution, are stored in evaporative mineral efflorescences, or seep through the bund walls and subsequently impact on Chinaman Creek. Thus at the investigated tailings repository, Cd, Ce, Cu, Fe, La, Mn, Ni, Pb, U and Zn display the greatest mobility. By contrast, Sn shows immobility due to the presence of weathering-resistant cassiterite, and As and Pb display limited mobility due to coprecipitation with jarosite-type phases and HFO materials at the top of the tailings profile.

Cemented hardpans are thought to act as hydraulic permeability and diffusion barriers, reducing the transport of pore waters and gases (e.g. Gilbert et al. 2003). In turn, the sulfide oxidation rate within tailings below the hardpans may be reduced and metals and metalloids are captured in the cemented layers, possibly leading to improved seepage and porewater quality (Chermak and Runnells 1996; Ettner and Braastad 1999). By contrast, partial leaching experiments by this study demonstrate that wetting and acidification of Fe-cemented tailings removes significant proportions of trace elements. At Jumna, hardpan development on cassiterite tailings has not led to the permanent sequestration of trace elements.

Conclusions

Since disposal in the 1980s, the Jumna mill tailings have been subjected to oxidation, leaching and erosion. The sulfidic tailings possess major (i.e. >1 wt%) median concentrations of Al, Fe_{total} , Fe^{2+} , K and Si; minor (i.e. >0.1 wt%) F, Mg, Na, S_{total} , $S_{sulfide}$, Sn and Ti; sub-minor (i.e. >100 ppm) As, Ba, C_{total} , C_{org} , Ca, Ce, Cu, Mn, P, Pb, Rb, $S_{sulfate}$, Zn and Zr; and traces (i.e. <100 ppm) of Ag, Be, Bi, Cd, Co, Cs, Ga, Ge, In, La, Li, Mo, Nb, Ni, Sb, Sc, Se, Sr, Ta, Te, Th, Tl, U, V, W and Y. The tailings mineralogy is comprised of primary aluminosilicates (quartz, K-feldspar, plagioclase, microcline, muscovite, chlorite, kaolinite), sulfides

(pyrite, chalcopyrite, galena, arsenopyrite), magnetite, cassiterite as well as traces of hornblende, beryl, fluorite and unidentified expanding clays.

Extensive weathering of the sulfide minerals has resulted in the formation of abundant mineral efflorescences and Fe-rich hardpans in the higher parts of gently sloping tailings masses at the top (<50 cm) of the tailings piles. The laterally discontinuous Fe-rich crusts are consolidated to a varying degree and cement tailings grains. Microscopically, hardpan-cemented tailings display thin layers (typically ~ 0.2 – 2 mm thick) of more strongly HFO-cemented tailings interlayered with material that contains less HFO. In comparison to the tailings, the hardpan-cemented tailings contain significantly higher median As, Ca, Cd, Co, Cu, Fe, In, Mn, Mo, S_{total} , Th, U, Y and Zn values. Thus, oxidation of Fe and precipitation of HFO phases and secondary sulfate efflorescences at the top of the tailings profile selectively capture these elements, possibly due to coprecipitation into sulfates and adsorption onto jarosite-type and HFO phases. However, partial leaching studies indicate that wetting and acidification of Fe-cemented tailings remove $>1\%$ of the total As, Ca, Cd, Ce, Cu, Fe, La, Mg, Mn, Ni, Pb, S_{total} , U and Zn. Yet, Sn shows no mobility due to the presence of weathering-resistant cassiterite. Also, As and Pb display limited mobility possibly due to their coprecipitation with jarosite-type phases and HFO materials at the top of the tailings profile. By contrast, the trace elements Cd, Ce, Cu, La, Ni, Pb, U and Zn display the greatest mobility, possibly due to their incorporation in soluble sulfate efflorescences and sorption onto mineral and HFO surfaces. Thus, Fe-rich hardpans do not cause permanent immobilisation of trace elements and, significant acid leaching of trace elements occurs from the hardpan capped tailings into pore and surface waters.

The major land use of the Jumna site is low-density cattle grazing, and a consequence of trace element mobility into surface waters is that there is the potential to transfer toxicants (Al, Cd, Cu, F, Pb, Se, Zn) into grazing animals. A risk assessment is required to determine the potential impact of trace element enriched waters on grazing animals.

Rehabilitation of the site has to consider the sulfidic, partly oxidised nature of the tailings. The materials require encapsulation in a newly constructed, capped waste repository. Other rehabilitation efforts should involve removal of waste rock dumps, import of topsoil and planting of local, metal-tolerant plant species.

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