



Research article

Magnetite recovery from copper tailings increases arsenic distribution in solution phase and uptake in native grass



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ARTICLE INFO

Article history:

Received 30 January 2016

Received in revised form

28 April 2016

Accepted 10 May 2016

Available online 19 May 2016

Key words:

Arsenic

Magnetite

Tailings

Organic matter

Fe oxyhydroxides

Native grass

ABSTRACT

Reprocessing magnetite-rich copper (Cu) tailings prompted a concern about arsenic (As) risks in seepage water and revegetated plants at Ernest Henry Cu Mine (EHM) in North Queensland, Australia, due to the closely coupled relationship between iron (Fe) minerals and As mobility. The magnetite removal alone significantly decreased the content of crystalline Fe minerals and the maximum arsenate (As(V)) sorption capacity of the resultant tailings. A glasshouse experiment with native grass Red Flinders (*Iseilema Vaginiflorum*) was conducted with the reprocessed (low magnetite (LM)) and original (high magnetite (HM)) tailings, which were amended with 5% sugarcane residue (SR) as a basal treatment in combination with 0, 1 and 5% pine-biochar (BC). The organic matter treatments and plant growth stimulated the formation of secondary Fe minerals. The amount of extractable amorphous Fe in the amended and revegetated HM tailings was significantly higher than those in the LM. Arsenic forms in the specifically sorbed and the sorbed by amorphous Fe oxides were significantly increased by the SR amendment in the LM tailings, but which were decreased in the HM, compared to the unamended tailings. Soluble As levels in the porewater of the LM under revegetation were significantly higher ($300\text{--}1150\ \mu\text{g As L}^{-1}$) than those (up to $45\text{--}90\ \mu\text{g As L}^{-1}$) in HM tailings in the same treatment, which led to the higher As concentrations in the plants grown in the LM tailings. In particular, root As concentration ($62\text{--}146\ \text{mg kg}^{-1}$) in the LM tailings was almost a magnitude higher than those ($8\text{--}17\ \text{mg kg}^{-1}$) in the HM. The present results confirmed the initial expectation that the recovery of magnetite from the Cu tailings significantly elevated the risk of As solubility in the tailings by decreasing As sorption capacity and increasing soluble As levels. Thus, it would be beneficial to retain high contents of magnetite in the top layer (e.g., root zone) of the Cu tailings for managing As risk and revegetation in the future.

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1. Introduction

Engineering soil formation from metal mine tailings (e.g., copper (Cu), lead-zinc (Pb–Zn) mine tailings) to offset large volumes of soil from offsite landscapes for direct revegetation of native plant communities have been advocated as a cost-effective and sustainable solution for rehabilitation of tailings landforms (Huang et al., 2012, 2014). However, the toxicity of metal(loid)s (such as Cu, Pb, Zn and arsenic (As)) in the tailings is one of critical factors limiting the progress of both soil formation and the growth and survival of pioneer plants, which is commonly related to the mineralogy and geochemistry of metal mine tailings concerned (Li et al., 2015a; Li and Huang, 2015). Arsenic bearing minerals are commonly

associated with Cu-ore minerals and gangue materials, such as As-bearing sulphides and sulphosalts, such as arsenopyrite (FeAsS), cobaltite (CoAsS), enargite (Cu_3AsS_4) and tennantite ($\text{Cu}_{12}\text{As}_4\text{S}_{13}$) and resultant Cu tailings contain elevated amounts of As resulted from these minerals (Dold and Fontboté, 2001; O'Day, 2006). Dissolution of As from As-bearing minerals in the tailings is the result from oxidation of the As-bearing minerals (such as pyrite and arsenopyrite in Cu tailings), but the size of soluble As pool is determined by sorption/immobilization by secondary Fe/Al oxides and oxyhydroxides (Drahota and Filippi, 2009). Therefore, Fe-mineralogy in Cu tailings rich in As-bearing minerals would significantly regulate the distribution of As into soluble pool, thus impacting on seepage water quality and the health of revegetated plant species.

In the tailings, Fe(III) reducing bacteria play a significant role in the transformation of both amorphous and crystalline ferric minerals, which may affect As distribution on the Fe phase (Cummings

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et al., 1999). Arsenate (As (V)) sorption could occur on the surfaces of magnetite particles, even though the affinity of magnetite for As is lower than those of the amorphous Fe minerals (Giménez et al., 2007; Mamindy-Pajany et al., 2011). Secondary Fe minerals can be formed from the reduction and dissolution of Fe(III) in crystalline Fe minerals (e.g., magnetite and goethite) commonly present in the tailings (Davey et al., 2008; Lottermoser, 2010), via transformation processes catalysed by Fe(III)-reducing bacteria (Dong et al., 2000; Zachara et al., 2001). Lowering magnetite content may lead to the reduction of As sorption capacity in the tailings, because of the combined effects of the direct sorption by magnetite particles and secondary Fe minerals formed herein. As a result, the presence of crystalline Fe minerals (e.g., magnetite, goethite, and hematite) and their transformation into secondary Fe oxyhydroxides are important to the immobilization of soluble inorganic As and its phytoavailability in the tailings under revegetation.

Ernest Henry Cu mine (EHM) produces magnetite-rich Cu tailings (20–30% magnetite), from which the magnetite can be extracted for economic purposes by using grinding classification and magnetite separation and the reprocessed tailings contain low magnetite content (about 3%) (Davey et al., 2008). Prior to the present study, we observed elevated levels of total As in the tailings without magnetite recovery (about 300–500 mg As/kg dwt). It is expected that the magnetite recovery from the Cu tailings may lead to decreased As sorption surfaces associated with Fe minerals, particularly the secondary Fe minerals derived from the dissolution of Fe(III) in crystalline magnetite and precipitation of Fe oxyhydroxides in the solid phase, thus increasing As distribution into the soluble phase.

There is a close relationship between organic matter mineralisation and Fe(III) reduction, since Fe(III) reducing bacteria metabolise nonrecalcitrant organic matter into oxidisable organic molecules (e.g., acetate) which are electron donors to Fe(III) (Lovley, 1987; Zachara et al., 2002). As a result, organic matter amendment which is often required to improve physical and biogeochemical conditions for revegetation in the tailing-soil would influence the rate of Fe(III) reduction and dissolution from the crystalline Fe minerals (e.g., magnetite) and the amount of secondary Fe minerals formed (e.g., ferrihydrites). In the meantime, organic matter and derivatives from its decomposition can directly influence As chemical forms through competing functional groups (e.g., phenolic, carboxyl, hydroxyls) (Behrends and Van Cappellen, 2007; Redman et al., 2002). Therefore, it is hypothesized that amendment with organic matter rich in nonrecalcitrant organic carbon may increase the formation of Fe oxyhydroxides and As-adsorption capacity, leading to lower levels of soluble As in the pore water of the tailings.

The present study aims to investigate the distribution of As in the pore water and extractable fractions in the Cu tailings of low magnetite (LM) (the product from magnetite recovery), in response to organic matter amendments and revegetation. This is to provide the basis for on-site tailings management decision making, in terms of managing As in the LM tailings at EHM in the near future. Since there is a potential of biochar production from the waste pine wood on site, pine biochar was included as a form of recalcitrant organic matter in combination with sugarcane residues (abundant in central Queensland) as organic matter amendments for enabling direct revegetation of native red Flinders grass (*Iseilema vaginiflorum*, one of the key native species for local revegetation). Arsenic uptake in the grass was examined in relation to As levels in the pore water. The expected findings would provide the basis for deciding the necessity of purposely retaining magnetite at the last layer of tailings deposition, in order to lower As risk in the revegetated plants

and perhaps in the seepage water during rainy season when the tailings go under revegetation in the near future.

2. Materials and methods

2.1. Tailings preparation and characteristics

The low magnetite (LM) and high magnetite (HM) tailings were collected from tailings impoundment area of EHM in August 2012, which was located on the northeast of Cloncurry (20°27'S, 140°42'E) in the Mount Isa-Cloncurry mineral district of northwest Queensland, Australia. The original tailings without magnetite recovery were designated as high magnetite (HM) and the reprocessed tailings after magnetite recovery as the low magnetite (LM). The bulk tailings were air-dried and thoroughly mixed prior to potting and applying treatments. The pH and electrical conductivity (EC) in the tailings were measured by using a bench-top TPS 901-CP meter, in 1:5 solid/water suspension. Particle size distribution in the tailing samples was measured by the Mastersizer 3000 laser diffraction particle size analyser (Malvern instruments Ltd). Total concentrations of metal and metalloids in the tailings were determined by using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) (Varian Vista Liberty, Australia) following *aqua regia* digestion using an AIM600 Block Digestion System (Aim Lab Automation Technologies Pty Ltd) according to the method by Wheal et al. (2011). Quantitative XRD analysis for LM and HM tailings was conducted at United Minerals Services (NSW Australia) using the method described by Ortiz et al. (2009).

2.2. Glasshouse experiment

The glasshouse experiment was conducted in the late Summer-Spring season (February to May 2013) with an ambient temperature of 26–32 °C. The tailings after air-drying were thoroughly admixed with two types of organic matter including sugarcane residue (SR) (Earth Wise Company, QLD) and pine-biochar (BC) (ANZAC Pty Ltd, pyrolysed at 700 °C) (Jiang et al., 2016), which were dried at 40 °C and passed through 2 mm sieves. The treatments were: unamended (control), SR 5% only, SR 5% + BC 1%, and SR 5% + BC 5%. The SR and BC were applied into the tailings (3 kg lots) on a weight basis (% w/w) in twin-pots. Biochar was included here as a comparison because BC may be produced from waste wood on site in the future.

Seeds of native Red Flinders grass (*Iseilema vaginiflorum*) were pre-germinated in potting-mix soil in the glasshouse and germinated seedlings were cultured under glasshouse condition prior to being transferred into the treatments. At 20th day after germination, seedlings of similar size and appearance were randomly selected, carefully removed from seedling beds and transplanted into the pots containing the treatments, with six seedlings per pot. The twin-pots were connected with a capillary mat at the base for water supply into the root zone via a capillary mechanism. Pore-water samplers (Rhizon MOM, Rhizosphere Research Products, Wageningen, Netherlands) were inserted into the centre from the side at the mid-height to non-destructively sample pore water for chemical analysis. Approximately 1.5 L 0.5 strength nutrient solution was added into the bottom bucket (water reservoir for capillary water supply) and refilled weekly. The full strength nutrient solution contained macronutrients (mM): NH₄NO₃ 2, KNO₃ 2.8, Ca (NO₃)₂ 1.6, MgSO₄·7H₂O 1, KH₂PO₄ 0.1, K₂HPO₄ 0.1; and micronutrients (µM): FeEDTA 100, ZnSO₄·7H₂O 2, MnSO₄·H₂O 2, CuSO₄·5H₂O 0.5, Na₂MoO₄·2H₂O 0.08, NaCl 8, H₃BO₃ 5.24.

The plants were destructively harvested in the fourth week,

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