



Evaluation of amendments to reduce arsenic and antimony leaching from co-contaminated soils



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HIGHLIGHTS

- Six amendments were tested for immobilisation of As and Sb in co-contaminated soil.
- Ferrihydrite and zero valent iron powder applied at 3% were most effective.
- Total metalloid leached was reduced by >80%.
- Ferric chloride was also effective with careful pH manipulation using lime.

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ABSTRACT

Co-contamination of soils with arsenic (As) and antimony (Sb) presents unique challenges for risk management. In this study a sequence of leaching experiments evaluated zero valent iron powder, ferrihydrite, ferric chloride, aluminium and manganese oxides, and kaolinite for As and Sb immobilisation in co-contaminated soils. Iron based amendments were most effective for the reduction of As and Sb in leachate in a column leaching study. Over 48 h zero valent iron powder and ferrihydrite applied at 3% (w/w dry weight) were most efficient, decreasing total As and Sb leachate concentrations by more than 80%. Careful moderation of pH (to > 2.5 but < ~6) with lime was required for effective co-immobilisation of both metalloids using ferric chloride. In a subsequent batch study with pH optimised for maximum sorption using 2% lime, ferric chloride (3%) added to two co-contaminated soils decreased As and Sb in solution after 7 days by at least 79%. Ferrihydrite (3%) and iron powder (3%) were less effective. Ferrihydrite (3%) was then used in a 12-week larger scale *Cynodon dactylon* plant experiment that also considered plant bioavailability. Porewater As and Sb decreased by up to 90% but over the 12 week trial period no significant change in shoot or root metalloid concentrations was observed. The study demonstrates that iron-based amendments can be extremely effective for co-immobilisation of As and Sb in contaminated soils, but for large scale application amendment feasibility considerations and site specific pH moderation are essential.

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1. Introduction

Arsenic (As) and antimony (Sb) are Group 15 metalloids that frequently co-occur in the environment (Kelepertsis et al., 2006; Filella et al., 2002a). They are often found together at extremely high concentrations on mining and smelting sites (Anawar et al., 2011; Ashley et al., 2007; Hiller et al., 2012; Li and Thornton, 1993; Okkenhaug et al., 2012; Pratas et al., 2005). For example, As has been recorded at greater than 30 000 mg kg⁻¹ in mining

impacted soils (Bowell, 1994; Peterson et al., 1979) but occurs naturally in soils at concentrations ranging from 0.02 to 97 mg kg⁻¹ (Chen et al., 1999; Smith and Huyck, 1999). Antimony naturally occurs in soils at concentrations ranging from 0.05 to 8.8 mg kg⁻¹ (Shacklette and Boerngen, 1984; Tighe et al., 2005) but has been reported at up to 5000 mg kg⁻¹ on contaminated sites (Filella et al., 2002a). The United States Environmental Protection Agency has designated both metalloids as priority pollutants for water protection (C.F.R., 2012) and both are listed as carcinogenic agents by the International Agency for Research on Cancer (inorganic As in Group 1, Sb trioxide in Group 2B) (IARC, 1989, 2012).

Both metalloids form soluble species over environmentally relevant conditions, with the +III and +V oxidation states most

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common and the pentavalent oxyanions (H_2AsO_4^- , $\text{Sb}(\text{OH})_6^-$) predominating in aerated environments (Wilson et al., 2010). The extent of metalloid in solution is determined by soil characteristics such as system redox and pH and in particular the presence of sorptive surfaces such as organic matter and iron oxyhydroxides (Craw et al., 2004; Mok and Wai, 1989; Ritchie et al., 2013; Tighe et al., 2005; Wilson et al., 2010). Sorption of the anions becomes weaker at circumneutral to alkaline pH as variable charge soil surfaces become more negatively charged. Off-site migration with leachate and run-off is often a concern at typical field soil pH values. Many authors report that the elements have contaminated water associated with the contaminated soils (Andy Martin et al., 2013; Garcia-Sánchez et al., 2010; Razo et al., 2004; Strømseng et al., 2009) and significant downstream mobilisation from input source (up to 300 km) is also reported (Ashley et al., 2003; Mok and Wai, 1989). This in turn leads to wider ecosystem contamination with uptake into plants and foodchains (Culioli et al., 2009; Dovick et al., 2015; Telford et al., 2009).

A range of different amendments have been applied to immobilise both As and Sb *in-situ* in contaminated soils, although usually soil that is contaminated with only one of these contaminants is trialed. For example, ferrihydrite additions reduced soluble As from $135 \mu\text{g L}^{-1}$ to $<12 \mu\text{g L}^{-1}$ in a mining contaminated loamy sand (Abad-Valle et al., 2015) and from $14\ 220 \mu\text{g L}^{-1}$ to $250 \mu\text{g L}^{-1}$ in a sandy loam contaminated by wood preservation activities (Nielsen et al., 2011). In a column experiment, goethite addition resulted in a 50% decrease in leachate As from mining contaminated silt loam (Ko et al., 2012), whilst hematite successfully decreased As leachate in a four step sequential extraction on contaminated sandy clay loam (Garau et al., 2014). Iron (Fe) oxide precursors (Bertocchi et al., 2006; Kumpiene et al., 2006; Moore et al., 2000), Al oxides (Garau et al., 2014), Mn oxides (He and Hering, 2009), and heterogeneous materials including red mud, fly ash, mine sludge and ferrous sulfate mixtures (Bertocchi et al., 2006; Ko et al., 2012; Moore et al., 2000) have also been effective, whilst reduced As phytoavailability has been observed after soil application of ferrihydrite and ferrous sulfate (Sun et al., 2015; Warren et al., 2003). With less work on Sb immobilisation, Okkenhaug et al. (2013) reported efficacy for four Fe-based amendments on Sb contaminated silty sand. After 7 days of equilibration Sb water extract concentration decreased by up to 90% using Fe oxyhydroxides and sulfates, and by 80% with zero valent iron grit. Tandy et al. (2017) reported some success with Fe sulfate and red mud.

The widespread contamination issues at shooting ranges have led to a number of recent studies investigating Sb immobilisation co-occurring with lead (Sanderson et al., 2013; Tandy et al., 2017), but to the best of our knowledge only one study by Álvarez-Ayuso et al. (2013) has considered the immobilisation of As and Sb concurrently. While both metalloids form anionic compounds, competition for sorption sites, differences in pH for maximum sorption to surfaces and the potential formation of insoluble precipitates can make concurrent immobilisation difficult. Álvarez-Ayuso et al. (2013) did show, however, using a mining contaminated loam, that ferrihydrite decreased both soluble As (from $\sim 0.35 \text{ mg kg}^{-1}$) and Sb (from $\sim 0.26 \text{ mg kg}^{-1}$) by $\sim 90\%$, whilst Al oxide could reduce the soluble contaminants by greater than 80%.

The aim of this study therefore was to further evaluate the efficacy of a wide range of soil amendments for concurrent As and Sb immobilisation. This was undertaken through a sequence of column leaching and batch experiments using two different contaminated soil materials, both of which required metalloid stabilisation for future management. A plant accumulation trial was also established to determine whether, at a larger scale, amendments decreased metalloid in soil leachate, and if this was also reflected in decreased metalloid plant uptake.

2. Methods

2.1. Soils and amendments

Soil material from two operational sites in northern New South Wales, Australia, both highly co-contaminated with As and Sb, were used in the experiments. Soil H was collected from a historic lay-down area at a gold-antimony mine which had been used for mining machinery and equipment storage. The area had undergone rehabilitation works consisting of mixing and spreading of stored mine waste materials with surrounding soils (Wilson et al., 2013). The management of soil at the mine currently targets mitigation of off-site contaminant movement with run-off and leaching. Soil U was collected from a waste disposal area at a former Sb processing plant that was undergoing remediation. This material was a mix of tailings material and surrounding site soils, that required immobilisation prior to encapsulation in the site remediation strategy. Composited site bulk samples were air-dried, sieved to 2 mm and well homogenised for use in all experimental procedures.

The amendments used were: aluminium oxide (Al oxide) (corundum, 98% Al_2O_3 , ChemSupply), manganese (IV) oxide (Mn oxide) ($\geq 99\%$ MnO_2 , Sigma-Aldrich), kaolinite (KGa-2) ($\sim 80\%$ $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, Clay Minerals Society), ferric chloride (FeCl_3) (96% $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, Ajax Chemicals), zero valent iron powder (Fe^0) (96.5% Fe metal, Ajax Chemicals) and ferrihydrite (Fh) (synthesized). Ferrihydrite was synthesized following the method provided in Schwertmann and Cornell (2000). Briefly, 40 g $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ was dissolved in 500 mL deionised water, with 330 mL 1 M KOH added under vigorous stirring until pH reached 7–8. The precipitate was then filtered and washed with 500 mL deionised water 3 times, dried over 48 h at 30°C , ground to a fine powder and stored below 4°C until use. Analysis via X-ray diffraction confirmed poorly crystalline iron oxide, likely ferrihydrite, with minor goethite. High purity micropure deionised water (TKA, Germany) was used in all procedures.

2.2. Column leaching procedure

A column leaching study was used to investigate a wide range of amendments for reducing As and Sb in leachate from Soil H only (Soil U was unsuitable for use in the column study due to its fine texture). The procedure was carried out under saturated conditions in acid washed polyethylene columns (length 200 mm x internal diameter 23 mm) packed with 70 g soil, either unamended (control) or thoroughly mixed with amendment. Six amendments were tested at varying ratios (w/w dry weight amendment:soil) with each treatment performed in triplicate. Application rates were selected on the basis of results in published studies. Aluminium oxide, Mn oxide, Fe^0 , and Fh were applied at 1%, 3%, and 5%, kaolinite at 8% and 10%, and FeCl_3 at 1% and 3%, with an additional treatment of 1% FeCl_3 combined with an equal amount of agricultural lime to moderate pH decreases (FeCl_3 + lime).

Columns were capped with glass wool and a $0.45 \mu\text{m}$ cellulose filter (Whatman). Deionised water was then added to the columns in an upward flow direction at a rate of approximately 4 mL h^{-1} via peristaltic pump. Once saturated, water flow was paused for 12 h to allow equilibration, then restarted in a downward flow direction at the same rate with leachate fractions collected after 12, 24, and 48 h of continuous flow. Leachate volume was recorded at approximately 190 mL over the experiment for most treatments. Leachate pH was measured immediately with subsequent acidification to $\text{pH} < 2$ using HNO_3 . Samples were analysed for dissolved As and Sb at National Association of Testing Authorities (NATA) accredited Australian Laboratory Services (ALS), Brisbane.

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