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Does ochre have the potential to be a remedial treatment for As-contaminated soils?

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ABSTRACT

Ochre is an iron oxyhydroxide-rich waste that accumulates in water bodies associated with disused mines. Laboratory experiments were conducted to examine the potential of four different ochres to be used as remedial agents for As contaminated soils. The ochres removed As from solution (200 and 500 mg L^{-1}) in adsorption experiments at pH 3 and 8 and, when added to As contaminated soil (5% w/w) significantly reduced As release to solution. In both these experiments the highest surface area ochres performed best. The impact of ochre amendments on uptake of As from soil by plants and humans and release of As to ground water was assessed in a year-long incubation study. Ochres increased soil pH and reduced $CaCl_2$ extractable As but had no consistent effect on plant growth, plant As uptake or As extraction in physiologically-based extraction tests. Ochre may be better used for water treatment than soil remediation.

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1. Introduction

High concentrations of As in soils and water bodies occur throughout the world due to anthropogenic activities such as mining, smelting and wood preservative usage (Abrahams and Thornton, 1987; Álvarez-Ayuso et al., 2012; Eapaea et al., 2007; Lin et al., 2004; Mench and Bes, 2009; Nriagu et al., 2007; Ritchie et al., 2013; Warren et al., 2003). The main routes of arsenic poisoning from contaminated soils are accidental ingestion or inhalation of contaminated soil or water or consumption of plants grown on As-contaminated soils (Miretzky and Cirelli, 2010). There is a need for affordable remedial solutions that can be applied to As-contaminated soil. It is increasingly recognised that the remedial methods of disposal or isolation of contaminated soil are not sustainable. This had led to a growing willingness to use organic or mineral amendments to alter soil chemistry and break pathways between pollutant sources and receptors (e.g. Hodson, 2010; Jones and Healey, 2010; O'Day and Vlassopoulos, 2010).

A large literature reports the sorption of many potential contaminants to Fe oxides, e.g. Cornell and Schwertmann, 2003 and references therein, Jambor and Dutrizac, 1998 and references therein. Much work has investigated As adsorption by Fe oxides (e.g. Bowell, 1994; Matis et al., 1997; Sun and Doner, 1998; Jain et al., 1999; Garcia-Sanchez et al., 2002; Goldberg, 2002; Ford, 2002; Jackson and Miller, 2000; Grafe et al., 2001; Waltham and Eick, 2002; Sun and Doner, 1996; Manning et al., 1998; Goldberg and Johnston, 2001; Livesey and Huang, 1981; Elkhatib et al., 1984a,b; Manning and Suarez, 2000; Smith et al., 2002) and this had led to investigations into using Fe oxides to remediate As-contaminated soil (e.g. Boisson et al., 1999; Garcia-Sanchez et al., 1999; Warren et al., 2003; Warren and Alloway, 2003; Nielsen et al., 2011; Kumpiene et al., 2008; Lee et al., 2011). Ochre is the name given to Fe(III) oxyhydroxide precipitates that accumulate in the outflows of mine systems. In the United Kingdom the Coal Authority is responsible for over 68 (as of August 2014) mine water treatment schemes that remove c. 4000 tonnes of iron per year from water courses resulting in ochre production (UK Government, 2014). Some of this ochre is used in brick production to partially offset the waste management costs (Clean Rivers Trust, 2012). Additionally, research has been carried out, with mixed success, into using ochre to limit phosphate concentrations in water and soil

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(Fenton et al., 2012; Heal et al., 2005; Dobbie et al., 2009; Sibrell et al., 2009). However, there is still no mature market for ochre in the UK; its accumulation poses a waste disposal problem.

Previously (Doi et al., 2005) we showed that ochre may be an appropriate remedial amendment for As-contaminated soils. However, properties of ochres are site specific. Here we characterise a further 4 ochres to further demonstrate the ability of ochres to adsorb As. We then report a year long incubation study investigating whether ochre amendments can break the most significant pathways (leaching to ground water, ingestion of soil, uptake by plants) between As-contaminated soil and receptors. We also examine the impact of ochre amendment on soil microbial functioning via assay of hydrolytic enzyme activity.

2. Materials and methods

As-bearing soils were collected from the upper 20 cm of profiles located at a former mine site (Devon Great Consols, SX 72878 96419, soil DGC), a former As calciner (Tresavean, Lanner, Redruth, SW 72423 39743, soils RRT1, RRT2) and an allotment site (Scunthorpe, SE 89344 10835, soil SCP). Soils were air-dried, sieved to ≤2 mm and stored prior to characterisation and use in experiments.

Four ochres were provided by the UK Coal Authority: Bull House (SE 421192 402506, BH) and Woolley (SE 41586 78838, WY) from passive treatment works, Old Meadows from an active treatment works (SJ 43496 95991, OM) and Six Bells from a combined passive and active treatment works (SO 22250 03039, SB). All ochres were supplied moist in sealed, plastic containers and were air-dried and crushed to ≤2 mm prior to use in experiments.

pH (ISO, 2005), loss on ignition (for organic matter content, Rowell, 1994), particle size distribution, BET surface area (De Boer et al., 1987), total As and Fe by aqua regia digest, acid ammonium oxalate and citrate-bicarbonate-dithionite extractable Fe (Loeppert and Inskeep, 1996), point of zero charge (Zelazny et al., 1996) and mineralogy by X-ray diffraction were determined following standard established methods (details in Supplementary material).

In adsorption experiments to investigate sorption of As by ochres 0.1 g ochre was added to 0.1 M sodium nitrate (40 mL); pH was adjusted to 3 or 8 using 0.1 M NaOH or 0.1M HCl. The mixture was shaken on an end-over-end shaker at 20 ± 1 °C for 24 h in darkness then 40 μL of 200 or 500 mg L^{-1} $NaAsO_3$ solution was added; pH was readjusted to pH 3 or 8 using NaOH or HCl and the mixtures were returned to the shaker. 15 replicates of each ochre-As combination were used and 15 ochre-free controls. After 0.5, 1, 3, 6 and 24 h, three sacrificial replicates were removed, pH adjusted back to 3 or 8, samples were centrifuged at 2113 g for 15 min and the centrifugate filtered through Whatman no. 2 filter papers. Arsenic concentrations were determined using ICP-OES.

In batch experiments to investigate reduction of As release from contaminated-soil into solution due to ochre addition 1 g As-contaminated soil was mixed with ochre (0.05, 0.1, 0.2 and 0.5 g) and added to 0.01 M $CaCl_2$ solution (20 mL). The mixture was shaken on an end-over-end shaker at 20 ± 1 °C for 24 h in darkness then centrifuged at 2113 g for 10 min. The supernatant was filtered (Whatman no. 2) and analysed for As by ICP-OES and pH.

Incubation experiments investigated the impact of ochre amendments on As mobility under pseudo-field conditions. Ochre (60 g) was added to soil (1200 g) (DGC, RRT2). Treatments and controls were moistened to 100% water holding capacity and incubated at 30 °C in sealed plastic bags for 52 weeks in darkness. The soil was mixed weekly. Five replicates were established per treatment and subsamples taken after 3, 12, 24 and 52 weeks of incubation for analysis. As Fe and As are redox sensitive elements we measured Eh at weeks 24 and 52 (there being insufficient resource to measure it at the other sampling points) in addition to pH (ISO, 2005) at each sampling point to determine potential changes in Fe and As speciation. Acid ammonium oxalate and citrate-bicarbonate-dithionite extractable Fe and a fluorescein diacetate (FDA) hydrolysis assay to measure microbial activity following the method of Adam and Duncan (2001) (Supplementary materials) were carried out on the subsamples.

We assessed the effect of ochre amendments on the most likely pathways for As contamination in soils to impact on the environment and human health. To determine possible As leaching into ground water 1 g of air-dried soil was added to 20 mL of 0.01M $CaCl_2$ solution (to represent soil solution; Houba et al., 1990) and shaken on an end-over-end shaker for 24 h at 20 °C. Samples were centrifuged at 2113 g for 15 min at 20 °C. Supernatant pH was measured. The supernatant was filtered (Whatman No 2) and analysed for As by ICP-OES. In a plant growth and As uptake bioassay, rye grass (*Lolium perenne*. L., 0.5 g seeds per pot) was grown in 150 g of incubated soil in a plant growth room for 40 days. Plants were harvested and shoots cut 1 cm above ground level; roots were washed in deionised water to remove attached soil. Samples were dried at 70 °C to a constant mass, digested in nitric acid and analysed for As by ICP-OES. For week 3 and 52 subsamples, a physiologically-based extraction test (PBET) extraction to assess As availability to humans on ingestion of the soil (Intawongse and Dean, 2008) was carried out. Air dried, ≤250 μm soil (1 g) was shaken with simulated stomach and intestine fluids which were analysed for As by ICP-OES. Analytical and methodological details are given in the Supplementary materials.

3. Quality control and statistical analysis

An in-house 500 ppb reference solution was analysed by ICP-OES at the start of each analytical run and returned values within 10% of established concentrations. The detection limit for the adsorption and batch experiment As solutions and $CaCl_2$ extractions was 8 μg L^{-1} calculated from the mean plus 6 times the standard deviation on ten replicate analyses of the blank calibration standard (Gill, 1997). Detection limits were 1.133 ± 0.198 mg kg^{-1} for plant digests and 61.6 mg kg^{-1} and 121.95 mg kg^{-1} for the stomach and small intestine phase of the PBET analysis. Method blanks were run for all extractions and results were blank corrected where appropriate. For aqua regia digests an in-house reference material (SS39) traceable to CRM 143R sewage sludge-amended soil (Commission of European Communities Community Bureau of Reference BCR) was digested. Recoveries were 95–105 % for As. For plant digests an in-house reference material (Hay 2) was digested with each batch of digests. Recoveries were 98–102%. Analytical precision for the different matrices by duplicate analysis of 10% of the samples (Gill, 1997) was >95%.

Statistical analysis was carried out using SigmaStat 12.0.

4. Results and discussion

The soils and ochres showed a range of properties (Table 1). The ochres contained relatively low concentrations of As, had a range of surface areas and the crystalline material present was goethite.

4.1. Adsorption experiments

Since Fe oxides can adsorb As (Dixit and Hering, 2003; Kanematsu et al., 2013; Miretzky and Cirelli, 2010) it was expected that the ochres would sorb As and this was verified in initial adsorption experiments conducted at pHs above and below the PZNC (Fig. 1). The majority of adsorption happened within the first 30 min of the experiment (Fig. 1), as in other experiments using ochres (Doi et al., 2005). The ochre PZNC values (Table 1) are lower

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