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Remediation of arsenic contaminated soils by iron-oxide application, evaluated in terms of plant productivity, arsenic and phytotoxic metal uptake

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ABSTRACT

Four iron-bearing additives, selected for known or potential ability to adsorb anions, were evaluated for their effectiveness in attenuation of arsenic (As) in three soils with different sources of contamination (canal dredgings, coal fly ash deposits, and low-level alkali waste). Amendments used were lime, goethite (α -FeOOH) (crystallised iron oxide) and three iron-bearing additives, iron grit and iron (II) and (III) sulphates plus lime, which result in 'de novo' iron oxide formation in soils. Each was applied to the test soils at a rate of 1% w/w. A series of plant growth trials were conducted on the equilibrated, amended soils using spinach (*Spinacia oleracea*) and tomato (*Lycopersicon esculentum*) as test crops. These were grown in the contaminated soils for a period of three months in controlled glasshouse conditions. Evaluation of the potential of the amendments as immobilising agents was determined by plant growth (biomass) and elemental accumulation in plant tissues, indicating the bioavailability of As and other heavy metals following amendment. Goethite produced the most promising results in terms of reduction of plant shoot As content. It was concluded that, whilst Fe-oxides may be used as effective *in situ* amendments to attenuate As in soils by reducing its bioavailability, their effects on plant growth require careful consideration. In addition, soil-plant transfer of As was not completely halted by any amendment.

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1. Introduction

Persistence of arsenic (As) in soil and toxicity to plants and animals is of concern. Soil is an important sink for As compounds, which accumulate rapidly due to their slow depletion by plant uptake, leaching and erosion (Smith et al., 1998). Given the relationship between skin cancer, angiosarcoma and As ingestion, this establishes the metalloid as a human carcinogen (Léonard and Lauwerys, 1980; Pershagan, 1981). As a result, As-polluted soils are considered major sources of contamination in the food chain and water supplies (Frankenberger, 2002). Remediation of land contaminated with As may reduce or prevent this transfer. However, there is a paucity of information

in the literature regarding reasonable ways of decontaminating As-polluted sites (Carbonell-Barrachina et al., 2004).

Current practise for remediating contaminated land is by excavation to landfill followed by replacement with clean soil. This technique is, however, environmentally disruptive and is becoming progressively more expensive (Mench et al., 1998). Alternative potential remediation strategies for metal/metalloid contaminated soils include *in situ* biological remediation, (for example, phytoextraction and phytovolatilization), physical and chemical stabilisation, encompassing techniques such as vitrification or asphalt capping, and finally the application of inorganic amendments (*in situ* elemental immobilization) to reduce mobile species and bioavailability.

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In situ elemental inactivation is a technique whereby an additive (amendment) is incorporated and mixed with a contaminated soil. Stabilization of trace elements in soil can be achieved by soil amendments that absorb, bind or co-precipitate the contaminating elements (Kumpiene et al., 2006). The main objective of amendment incorporation is to reduce trace element mobility, thus affecting leaching potential and bioavailability. Whilst numerous inorganic amendments (clays and aluminium, iron and manganese oxides and hydroxides) have been incorporated into As-contaminated soil to reduce labile As (Mench et al., 1998), iron oxides show great potential for 'in situ' use. Iron oxide surfaces are known to be involved in As adsorption in soils (Jacobs et al., 1970; Lumsdon et al., 1984; Waychunas et al., 1993). Carbonell-Barrachina et al. (1999) showed that water-soluble iron concentrations were highly correlated with dissolved total As, suggesting that hydrous iron oxides play an important part in controlling As adsorption-desorption reactions in sewage sludge. Iron oxides applied to garden soils have shown decreases of up to 50% in water extractable As concentrations, together with lower accumulation levels in plant tissues (Mench et al., 1998).

The iron sources used in this study (goethite, iron grit, iron II sulphate+lime and iron III sulphate+lime), were chosen for the following reasons. Ferrous sulphate was selected because when tested against other iron sources, such as hematite, in soils, it was found to bind As more strongly (Artiola et al., 1990). Goethite has been shown to reduce As toxicity using an adsorption-oxidation system comprising goethite (α FeOOH) and birnessite (a manganese oxide), which significantly reduced As toxicity in contaminated soils (Sun and Doner, 1998). Amorphous iron hydroxide (am-Fe(OH)₃) also has an extremely high adsorptive capacity for As and steel shots (iron grit) have been used for As immobilisation in contaminated garden soils (Vangronsveld et al., 1994). Steel shot, an industrial material containing mainly iron (97%), corrodes and oxidises to produce iron oxides that have been shown to effectively reduce As levels in plant tissues in field trials (Mench et al., 1998). Lime has been widely used to immobilise heavy metals, however its application to soil contaminated with As may result in increased solubility of the metalloid, due to an increase in soil pH. The soils used in this investigation also contained elevated levels of heavy metals; lime was added to the list of amendments to determine its effects in situations of multi-element contamination.

Arsenic does not appear to be involved in any specific metabolic reactions in plants and is therefore not essential for growth (Marin et al., 1993). Arsenic concentrations in the edible parts of crops depend on the availability of soil As and the ability of a crop to take up As and to translocate it to the target organs (Huang et al., 2006). There are many factors that influence As uptake by plants, including species differences (Walsh and Keeney, 1975), the presence of competing ions (Khattak et al., 1991), concentration of As in the soil (NAS, 1977), and soil properties such as pH, clay content (Von Endt et al., 1968), redox potential and the presence of iron oxides. Total soil As concentration is a poor indicator of the plant-available fraction (Sheppard, 1992) which governs uptake and food chain transfer (Duel and Swoboda, 1972).

Arsenic accumulation within edible plant tissue is generally low (Vaughan, 1993; O'Neill, 1995). Roots contain the greatest quantities of As, intermediate concentrations occur in leaves and stems (above ground vegetative parts) and the smallest quantities are found in seeds and fruits (Walsh and Keeney, 1975; Carbonell-Barrachina, 1992, 1995). There is considerable variation in sensitivity to As between plant species (Jacobs et al., 1970; Jiang and Singh, 1994), and it has been shown that plants differ not only in their tolerance to As, but also in their absorption capabilities (Carbonell-Barrachina et al., 1997).

Chemical tests to determine the bioaccessibility of trace elements are valuable, but they cannot assess bioavailability and the risks imposed to organisms at different trophic levels, and therefore biochemical, microbiological and plant-based tests are increasingly used for soil toxicity assessment (Adriano et al., 2004; Dickinson et al., 2005).

Plant growth trials were used here to determine the effectiveness of a range of iron oxides on improving growth in selected As-contaminated soils. Spinach (*Spinacia oleracea*) produced a high leaf biomass whilst Tomato (*Lycopersicon esculentum*), was chosen due to its reported tolerance to As pollution (Wauchope, 1983). In a previous study a concentration of 2 mg l⁻¹ As was not found to be phytotoxic to tomato plants (Carbonell-Barrachina et al., 1997).

This paper reports on the efficacy of Fe-oxides in relation to As retention in three diverse As-contaminated soils, as evaluated by plant survival and biomass production. The three soils were all contaminated with As at a level exceeding the UK soil guideline value (SGV) on 'safe' soil As concentrations (20 mg kg⁻¹) (Environment Agency, 2004). Phytotoxic heavy metals (copper

Table 1 – Study sites and outline descriptions

Site	NGR	Soil classification	Details
Rixton Clay Pits, nr Warrington, Cheshire, UK	SJ 621 885	Sandy loam	Located adjacent to brickworks, where fly ash, an increasing waste problem from the combustion of coal from coal-fired power stations has been disposed into clay pits. This practise continued until the mid 1960's (Warrington MBC Ranger Service, pers comm.). Metals and As are leached easily from the coal ash settling ponds.
Kidsgrove, Stoke on Trent, UK	SJ 844 543	Sandy clay loam	Site owned by the British Waterways Board (BWB), which is situated adjacent to a canal. Dredgings have been deposited onto the adjoining land. Contaminants may have accumulated via pesticide or herbicide runoff from farmland. A pigment factory is also situated near to the canal where possible As and Zn contamination may have arisen.
Merton Bank, St Helens, Merseyside, UK	SJ 523 961	Clay loam	6.6 ha of intensively mown amenity grassland, used for public recreation, within a residential and industrial area. Shallow soils (10–30 cm). An alkali works from 1873 within an industrial landscape, and subsequently an industrial waste site.

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