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A greenhouse and field-based study to determine the accumulation of arsenic in common homegrown vegetables grown in mining-affected soils

Monica D. Ramirez-Andreotta [a], Mark L. Brusseau [a,b], Janick F. Artiola [a], Raina M. Maier [a,*]

[a] Department of Soil, Water and Environmental Science, The University of Arizona, Tucson, AZ 85721, USA
[b] Department of Hydrology and Water Resources, The University of Arizona, Tucson, AZ 85721, USA

HIGHLIGHTS

► We characterized As uptake by homegrown vegetables near a Superfund site in AZ.
► A greenhouse study conducted in parallel with a co-created citizen science program.
► Asteraceae and Brassicaceae families had the largest As bioconcentration factors.
► A correlation was observed for As in vegetable vs soil for selected families.

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ABSTRACT

The uptake of arsenic by plants from contaminated soils presents a health hazard that may affect home gardeners neighboring contaminated environments. A controlled greenhouse study was conducted in parallel with a co-created citizen science program (home garden experiment) to characterize the uptake of arsenic by common homegrown vegetables near the Iron King Mine and Humboldt Smelter Superfund site in southern Arizona. The greenhouse and home garden arsenic soil concentrations varied considerably, ranging from 2.35 to 533 mg kg^{-1}. In the greenhouse experiment four vegetables were grown in three different soil treatments and in the home garden experiment a total of 63 home garden produce samples were obtained from 19 properties neighboring the site. All vegetables accumulated arsenic in both the greenhouse and home garden experiments, ranging from 0.01 to 23.0 mg kg^{-1} dry weight. Bioconcentration factors were determined and show that arsenic uptake decreased in the order: Asteraceae > Brassicaceae > Amaranthaceae > Cucurbitaceae > Liliaceae > Solanaceae > Fabaceae. Certain members of the Asteraceae and Brassicaceae plant families have been previously identified as hyperaccumulator plants, and it can be inferred that members of these families have genetic and physiological capacity to accumulate, translocate, and resist high amounts of metals. Additionally, a significant linear correlation was observed between the amount of arsenic that accumulated in the edible portion of the plant and the arsenic soil concentration for the Asteraceae, Brassicaceae, Amaranthaceae, and Fabaceae families. The results suggest that home gardeners neighboring mining operations or mine tailings with elevated arsenic levels should be made aware that arsenic can accumulate considerably in certain vegetables, and in particular, it is recommended that gardeners limit consumption of vegetables from the Asteraceae and Brassicaceae plant families.

1. Introduction

Mining and industrial processes are primary sources of arsenic and heavy metal contamination in soil (Lee et al., 2005). In the United States alone there are 45 billion tons of mine waste, including waste rock and tailing material, and many of the estimated 557,650 abandoned hard rock mine sites are in arid and semiarid regions (US EPA, 2004). Mine tailings and their associated metal contaminants, such as arsenic and other heavy metals, are prone to wind dispersion and water erosion. Surface soils adjacent to, beneath, or downwind of arsenic release sources (e.g., smelters or mine tailings) often have arsenic levels at or above regulatory contaminant limits (Belluck et al., 2003). Mining operations in particular pose a potential risk to human health and the environment. Numerous studies have found an inverse relationship between arsenic levels in human urine samples and the distance of home or school environments from metal smelters and other mining operations (Csavina et al., 2012). Climate change will only exacerbate the risks posed by mining in arid and semi-arid environments like the desert Southwest, primarily due to increased temperatures and reduced precipitation (MacDonald, 2010).

* Corresponding author at: Dept. of Soil, Water and Environmental Science, 429 Shantz Bldg. #38, University of Arizona, Tucson, Arizona 85721-0038, USA. Tel.: +1 520 621 7231; fax: +1 520 621 164.
E-mail address: rmaier@ag.arizona.edu (R.M. Maier).

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Fugitive metals in receiving waterways and soils in the vicinity of mining sites can affect humans via the inadvertent consumption of metal-containing soils and dust, or through the consumption of crops grown in such soils (Murray et al., 2009; Lee et al., 2005; Cobb et al., 2000). It has been shown that arsenic in soil is the major source for arsenic uptake by crops (e.g. Huang et al., 2006). Arsenic exposure is of special concern in the US Southwest due to elevated levels that often occur naturally in drinking water sources. Thus, potential exposure via consumption of affected garden crops would add to an already elevated exposure from drinking water. Inorganic arsenic calculated as arsenite and arsenate comprise 96% of the total arsenic in vegetables (Smith et al., 2006). Intake of inorganic arsenic over a long period can lead to chronic arsenic poisoning (arsenicosis) and associated effects, including skin lesions, peripheral neuropathy, gastrointestinal symptoms, diabetes, renal system effects, cardiovascular disease and cancer, which can take years to develop depending on the level of exposure (WHO, 2010).

Due to the extent of contamination, the number of Superfund and other hazardous waste sites in the U.S. and the growing popularity of food gardening, understanding the spatial distribution of arsenic in residential soils and the uptake of arsenic in common homegrown vegetables is crucial to protect human health near these sites. In 2008, 36 million households participated in food gardening, with an average contact time of 5 h per week (National Gardening Association, 2009). The level of participation in gardening is only expected to increase, and the main reasons why Americans are food gardening are to grow better tasting and quality food, and to grow food they feel is safe (National Gardening Association, 2009). A gardener who neighbors a Superfund or hazardous waste site needs to be particularly aware of their soil quality and the potential for uptake of arsenic by the vegetables they choose to grow.

This study entitled *Gardenroots*, was designed to determine the concentration of arsenic in vegetable plants grown near the Iron King Mine and Humboldt Smelter Superfund (IKMHSS) site in Arizona, a site known to have elevated levels of arsenic. The objective of this study was to characterize and compare the uptake of arsenic by common homegrown vegetables grown in soils near the site. A controlled greenhouse study was conducted in parallel with a co-created citizen science program where community members, after training, collected soil, irrigation water and vegetable samples from their household garden. These samples were analyzed for arsenic content at the University of Arizona (UA). There have been several studies that have investigated the accumulation of arsenic in homegrown vegetables, but to the best of our knowledge this is the first to do so using a citizen-science program design. The residential area (Dewey–Humboldt) upon which the study was focused is adjacent to the IKMHSS site.

2. Materials and methods

2.1. Site description

The IKMHSS site is located in Dewey–Humboldt, Yavapai County, Arizona, and was listed on the U.S. Environmental Protection Agency's (US EPA) National Priorities List in 2008. The site comprises a combination of sources and releases from two separate locations: the Iron King Mine property (34°30′N, 112°15′W) and the Humboldt Smelter property (34°29′N, 112°13′W). A portion of the Town of Dewey–Humboldt is situated between the mine and the smelter (Fig. 1). The smelter operated from the late 1800s until 1969. The Iron King Mine operated from the late 1800s until the early 1960s and was a periodically active for gold, silver, copper, lead and zinc. All mining and smelting ceased by 1969.

Large amounts of uncontrolled mine tailing waste exist on both the smelter and tailings properties. The average composite concentration of arsenic in the Iron King Mine tailings pile (0–0.61 m below ground surface) is 3,710 mg kg^{-1} (EA Engineering, Science, and Technology, Inc., 2010). A previous study determined that the IKMHSS mine tailings have a low pH(2.5), high EC (13.5 ms/cm), a loam texture (34.7% sand, 44.8% silt, and 20.4% clay), and TOC and TN of 1.22 g/kg and 0.0423 g/kg, respectively (Solís-Domínguez et al., 2012). The unprotected mining wastes on the two properties are point sources of pollution and are prone to eolian dispersion and water erosion. This is reflected in observations of elevated arsenic and lead concentrations in surface soil on off-site areas adjacent to the Chaparral Gulch or downwind of the mine tailings and smelter properties. The concentrations of arsenic and lead in shallow surface soil samples in these areas are higher than the concentrations of arsenic and lead in the deeper surface soil samples. The elevated lead and arsenic levels near the surface are likely due to wind dispersion or surface water transport, rather than being attributable to background conditions (EA Engineering, Science, and Technology, Inc., 2010).

2.2. Greenhouse study

The soil treatments used in the greenhouse study included surface samples (0–15 cm) that were collected at a residential site between the Iron King Mine property and the Humboldt Smelter property, and adjacent to the Chaparral Gulch in July 2010. In 2009, the US EPA sampled this residential property and identified areas with elevated levels of arsenic. Using this information, four 7-meter transects were made with transect 1 and 2 located in an area where the US EPA detected elevated levels of arsenic (120–633 mg kg^{-1}) and transects 3 and 4 in areas where the levels of arsenic were closer to background and/or the Arizona Residential Soil Residential Level (13–25.7 mg kg^{-1}). For each transect, a 20-liter soil sample was collected every 1.2 m. All soil samples from transects 1 and 2 were homogenized and then sieved to ≤2 mm (elevated arsenic soil). Soils from transects 3 and 4 were treated similarly (background arsenic soil). In order to replicate popular gardening practices, the two collected residential soils were mixed with 25% (w/w) MiracleGro™ Gardening Soil Mix (garden soil, Home Depot, Tucson, Arizona) and then used to create three treatments for the greenhouse study: (T1) residential soil with background levels of arsenic, mixed with 25% the garden soil; (T2) residential soil with elevated arsenic levels, mixed with 25% garden soil; and (T3) residential soil with elevated arsenic levels, mixed with 25% garden soil and 10% mine tailing waste from the Iron King Mine.

The following criteria were used to select the vegetables for the greenhouse study: 1) among the top ten most popular vegetable grown in U.S. and used by various ethnic groups (National Gardening Association); 2) used in previous research studies (Bhattacharya et al., 2010; Murray et al., 2009; Smith et al., 2009, 2008; Gaw et al., 2008; Li et al., 2006; Smith et al., 2006; Warren et al., 2003; Alam et al., 2003; Bunzl et al., 2001; Cobb et al., 2000; Kabata-Pendias and Pendias, 2001); and 3) recommended by Master Gardeners in Pima and Yavapai County, Arizona (regional knowledge). Based upon these criteria, bush bean (Fabaceae *Phaseolus vulgaris*), lettuce (Asteraceae *Lactuca sativa*), radish (Brassicaceae *Raphanus sativus*), and onion (Liliaceae *Allium cepa*) were selected and grown in the greenhouse. The experimental design was completely randomized with four replicate pots of each vegetable for each of the three treatments (N = 48). Vegetables were grown in black plastic pots with drainage (17 cm top d × 18 cm height × 13.5 cm bottom d), and filled approximately 3/4 with the treatment mixture as described by Solís-Domínguez et al., 2011. The number of seeds sown and the sowing depth varied by plant. On a per pot basis, six bush bean seeds were sown at 2.5 cm, 3 lettuce seeds were sown at 0.3 cm, 6 radish seeds were sown at 1.3 cm, and 8 onion seeds were sown at 0.64 cm. Germination occurred after approximately 3, 5, 5, and 12 d for the radish, bean, lettuce and onion, respectively. Bean pots were thinned to 1 plant per pot. Pots were watered with tap water using drip irrigation every other day (~60 mL/pot). The experiment was performed at the