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Accumulation of arsenic and lead in garden-grown vegetables: Factors and mitigation strategies

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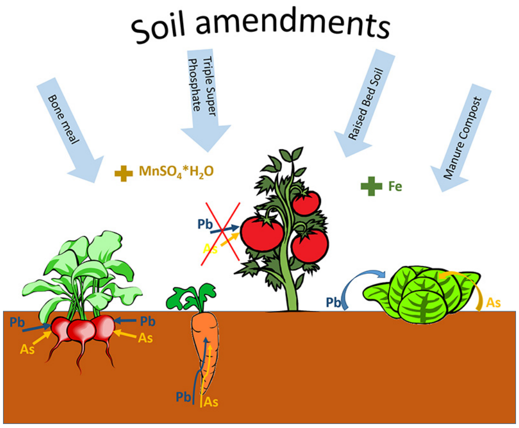
HIGHLIGHTS

- Soils can have elevated Pb and As due to past pesticide use.
- Vegetables produced on these soils can have metals levels above safety standards.
- Addition of P stabilizes Pb and mobilizes As.
- Root and green leafy vegetables had the highest Pb/As due to soil particle adherence.

GRAPHICAL ABSTRACT



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ABSTRACT

Pesticides containing lead and arsenic were widely used in the US through the 20th century. Legacy contamination from this use poses a health risk as interest in cultivation of abandoned agricultural lands has grown in recent years. We addressed these risks by quantifying Pb and As in soils and produce from a suburban farm in New Jersey, USA and examining the ability of phosphate-bearing amendments (bone meal, triple super phosphate, manure compost and raised bed soil) in combination with Fe and/or Mn amendments to stabilize these metals and prevent their movement into vegetables. Common produce (tomato, carrot, lettuce, and radish) was grown in soils with 133–307 mg Pb kg^{-1} and 19–73 mg As kg^{-1}. Our results suggest that vegetables produced on these soils can have Pb and As at levels above health and safety standards, especially root and leafy green vegetables. Phosphate-bearing amendments can reduce extractable Pb but can increase extractable As in soils, and can have similar effects on vegetables. Iron amendment increased both extractable Pb and As, likely due to the presence of elemental sulfur in the Fe amendment, which lowered soil pH, while Mn amendment had the

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Compost
Amendments

opposite effect. Most of the Pb and As in vegetables appear to be associated with soil particles adhered to the vegetables, and the contribution from uptake was relatively small except for plots treated with Fe-amendments and for carrots. Thus, proper crop selection, rigorous cleaning, and dust and dirt control are critical to reduce the risk of contaminant exposure through the consumption of garden produce.

1. Introduction

The contamination of soils by Pb and As that began in the late 1800s persists today (Schooley et al., 2008) because Pb is quite immobile and As is only very slowly leached through soils (Hood, 2006; Verneman et al., 1983). As contaminated lands are converted for residential and farming uses, the potential risk to human health is increased through direct or indirect exposure pathways arising from gardening and further transfer through the food chain (Cheng et al., 2015; McBride, 2013).

Organic and inorganic amendments have been used to reduce metal mobility and toxicity in soils. These amendments can transfer soil metals to more geochemically stable phases via sorption, precipitation, and complexation processes (Hashimoto et al., 2009). The most commonly applied amendments include clay, cement, zeolites, phosphates, and composts (Finžgar et al., 2006; Wuana and Okieimen, 2011; Zhang et al., 2010). Adding amendments is much less expensive than excavation and removal of soils (Scheckel et al., 2013; U.S. EPA, 2011, 2013).

Phosphate-bearing amendments appear to be the most effective for reducing Pb availability. Phosphate amendment stimulates de novo formation of pyromorphite minerals [$Pb_5(PO_4)_3X$ where $x =$ halide or hydroxide] (Ruby et al., 1994) that are insoluble (Ma et al., 1995; Ryan et al., 2001; Scheckel et al., 2013; Zhang and Ryan, 1999). While studies have demonstrated the Pb stabilizing potential of phosphate amendments, the magnitude of the effect remains incompletely characterized for different types of soils and for different types of phosphate-bearing amendments. The ability of these amendments to reduce Pb uptake by plants is also incompletely understood (Codling et al., 2015; Lim and McBride, 2015; McBride, 2013; McBride et al., 2014, 2015).

While phosphate has been shown to immobilize Pb, it can also mobilize As, a known human carcinogen. Added phosphate increases the solubility of soil As through competitive anion exchange (Peryea, 1991) causing it to migrate downward in the soil profile to groundwater, or increasing its availability for plant uptake (Cao et al., 2003; Creger and Peryea, 1994; Peryea and Kammereck, 1997; Wang et al., 2002; Zhao et al., 2009). There is uncertainty regarding the extent to which As can be mobilized by phosphates in different types of soils with different types of amendments, and how they affect As uptake by different types of plants (Lim and McBride, 2015; Scheckel et al., 2013).

Some researchers suggest that constant addition of organic matter in large amounts dilutes total Pb concentrations in soils and would thus be beneficial (Attanayake et al., 2014, 2015; Brown et al., 2012; Defoe et al., 2014). However, McBride et al. (2015) found that vegetable Pb and As concentrations were strongly correlated with soil total Pb and As but not to soil organic matter content or compost addition level.

Iron (Fe) and manganese (Mn) are essential elements for plant growth that also have potential effects on Pb and As dynamics. McKenzie (1980) found that adsorption of Pb on Mn and Fe oxides surfaces increased with pH and surface area resulting in the formation of Pb-metal solids. Adsorption and co-precipitation can help maintain low soluble Pb levels in soil solutions (Hettiarachchi and Pierzynski, 2004). Sorption of As onto Fe and Al oxides also decreases As solubilization. Simultaneous addition of P and cryptomelane (Mn oxide) was more efficient in decreasing bioavailable Pb than the addition of phosphate amendment alone, and more pyromorphite-like minerals are produced (Hettiarachchi et al., 2000; Hettiarachchi and Pierzynski, 2002).

In addition to soil factors, metal absorption varies among different types of plants (Karami et al., 2011). Uptake and translocation of As from roots to shoots has been found to vary widely, with some *Brassicaceae* having particularly high uptake potential (Raab et al., 2007). McBride et al. (2015) ranked the uptake potential of Pb and As by crop: carrot ≥ lettuce > bean > tomato and lettuce > carrot > bean > tomato, respectively. They also suggested that compost additions reduced both Pb and As concentrations in leafy vegetables.

This field study aimed to improve understanding of the relationship between Pb and As concentrations in garden produce and in farm soils, with consideration of the effects of phosphate-bearing and Fe/Mn amendments. The specific goals were (1) to quantify the degree of As mobilization and Pb stabilization due to addition of varying combinations of phosphates, compost, raised bed soil, and Fe and Mn amendments; and (2) to evaluate factors that determine or affect Pb and As levels in vegetable tissue. The research was carried out on suburban land that had historically been treated with pesticides containing both Pb and As, raising important questions about the use of phosphate amendments that stabilize Pb, but potentially mobilize As. Novel aspects of our research include: 1) a wide range of different amendments and different plant types were evaluated; 2) extractable (phytoavailable) concentrations were compared to total soil and vegetable metal concentrations; and 3) the studied contaminated suburban areas have been overlooked in previous research that has focused more on agricultural and urban soils.

2. Materials and methods

2.1. Experimental setup

This study was conducted at Duke Farm (40.5444° N, 74.6240° W) in New Jersey, USA. This site has elevated soil Pb and As concentrations and high correlation between soil Pb and As ($R^2 = 0.83$, $P < 0.001$), suggesting a single source of contamination (Muñiz, 2013). This contamination likely originated from past application of lead arsenate pesticide to control gypsy moth in 1920's (http://entomology.rutgers.edu/history/gypsy-moth.html).

This study examined the effects of bone meal, triple super phosphate (TSP), raised bed soil and compost with or without Fe and Mn amendments (Fig. S1) on Pb stabilization and As mobilization. There were twenty five 1.48 m^2 plots including five replicate plots of each of the following treatments: control, bone meal (Greenway Biotech Inc. 3-15-0, Ca-24%, derived from cooked bone meal; 14 USD per bag of 5 pounds (2.27 kg)), TSP (Hoffman, 0-46-0, derived from triple superphosphate; 15.50 USD per bag of 5 pounds (2.27 kg)), manure compost (Nature's Care Really Good Compost™; 5 USD per bag of 1 cu. ft (28.3 l)), and raised bed soil (Nature's Care® Organic Raised Bed Soil 0.12-0.06-0.09, derived from poultry litter; 32 USD per bag of 1.5 cu. ft (42.4 l)). Iron was added to one set of replicates, $MnSO_4$ was added to a second set of replicates and both Fe and $MnSO_4$ were added to a third set of replicates. Iron was supplied as Dr. Iron (Monterey, 22% Iron, non-staining, OMRI listed, 55% Sulfur, derived from elemental sulfur and iron oxide, phosphorous free; 30 USD per bag of 40 pounds (18.1 kg)) and Mn was supplied as manganese sulfate monohydrate ($MnSO_4 {*} H_2O$, 32% Mn; 35 USD per bag of 22 pounds (10 kg)). This design allowed for statistical evaluation (5 replicates) of the bone meal, TSP, manure compost and raised bed soil treatments and for evaluation (5 replicates) of Fe, Mn and Fe plus Mn amendments. At the beginning of this study, three samples from Duke Farm were collected and measured for soil total