Arsenic and Lead Uptake by Vegetable Crops Grown on an Old Orchard Site Amended with Compost

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Abstract

The potential for lead (Pb) and arsenic (As) transfer into vegetables was studied on old orchard land contaminated by lead arsenate pesticides. Root (carrot), leafy (lettuce), and vegetable fruits (green bean, tomato) were grown on seven "miniplots" with soil concentrations ranging from near background to ≈ 800 and ≈ 200 mg kg⁻¹ of total Pb and As, respectively. Each miniplot was divided into sub-plots and amended with 0% (control), 5% and 10% (by weight) compost and cropped for 3 years. Edible portions of each vegetable were analyzed for total Pb and As to test the effect of organic matter on transfer of these toxic elements into the crop. Vegetable Pb and As concentrations were strongly correlated to soil total Pb and As, respectively, but not to soil organic matter content or compost addition level. For Pb vegetable concentrations, carrot \geq lettuce > bean > tomato. For As, lettuce > carrot > bean > tomato. A complementary single-year study of lettuce, arugula, spinach, and collards revealed a beneficial effect of compost in reducing both Pb and As concentrations in leafy vegetables. Comparisons of all measured vegetable concentrations to international health-based standards indicate that tomatoes can be grown without exceeding standards even in substantially Pb- and Ascontaminated soils, but carrots and leafy greens may exceed standards when grown in soils with more than 100-200 mg kg⁻¹ Pb. Leafy greens may also exceed health-based standards in gardens where soil As is elevated, with arugula having a particularly strong tendency to accumulate As.

Key words: vegetable gardening; plant metals uptake; lead arsenate pesticides; lead; arsenic; compost amendment

Introduction

Lead (Pb) arsenate was the most commonly used pesticide in North American fruit tree orchards in the first half of the 20th century before the introduction of synthetic organic pesticides, and residues from previous applications of this pesticide are commonly found in orchard soils (Wolz, et al. 2003). As old orchard lands are converted from agricultural to residential uses, the potential for human exposure and health risk due to soil residues of previously applied pesticides may increase through residential gardening, direct soil contact and consumption of contaminated vegetables. The scale of this problem is evident from estimates that millions of acres of orchard lands have been contaminated by past use of arsenic (As) and Pb pesticides. For example, Virginia is thought to have 100,000-300,000 acres of old orchard land (Schooley et al. 2008), and other states with large acreages of impacted orchard land include Washington (188,000 acres), Wisconsin (50,000 acres) and New Jersey (up to 5% of total agricultural acreage) (Hood 2006). The total area of historical soil contamination in New York State is uncertain, but apple production has occupied about 40-50,000 acres in recent decades, and the present apple orchard acreage may underestimate the total land area historically contaminated by lead arsenate pesticide use.

Some older published studies have measured As and Pb transfer into vegetables grown on historically lead arsenate-contaminated orchard soils. Generally, the degree of contamination of the edible portion of the crops was reported to be relatively small, especially for fruits (Kenyon et al. 1979; MacLean and Langille 1981; Aten et al. 1980). Creger and Peryea (1992) measured As and Pb in apple and apricot fruits grown in lead arsenate-contaminated soils. Although they found Pb in the fruit to be below analytical detection limits, As concentration reached as high as 0.07 mg kg⁻¹ (fresh wt. (f.w.) basis) in the fruit, and was correlated to the HCl-extractable soil As (which ranged from 0 to 100 mg kg⁻¹ in these soils). However, leafy and root crops have shown higher levels of As and Pb contamination than fruits. In particular, the relatively high water solubility of soil As measured by a number of studies of lead arsenate-contaminated orchard soils suggests that the potential for As uptake into roots and leaf tissues of some crops could be significant. Arsenic uptake from orchard soils is frequently high enough to cause phytotoxicity in sensitive crops (Bishop and Chisholm 1962; Ross and Crowe 1976). Our own studies of As and Pb uptake from orchard soils by leafy green crops showed that there is a wide range of potential for uptake depending on crop species and the part of the plant analyzed (McBride 2013; McBride et al. 2012). Thus, root and leafy crops have a much higher potential for Pb and As contamination than most vegetable fruits. Even among leafy greens, some crops such as arugula can accumulate substantially higher concentrations of As than collards or spinach, with lettuce being intermediate (Stilwell et al. 2006).

At present, knowledge of factors controlling As and Pb transfer into food crops from soils is limited and insufficient to make reliable site-specific predictions of crop contamination levels. Specifically, Pb and As contents in individual vegetable crops have been found to be poorly correlated with soil total Pb and As concentration in contaminated field sites (McBride et al. 2012; Hough et al. 2008; Stilwell et al. 2008; Witzling et al. 2010; Warren et al. 2003; Nathanail et al. 2004). This poor correlation to total soil metals can be a consequence of the effects of different sources and types of contamination and local soil properties on bioavailability. For example, As in soils from pesticide use appears to be more plant-available and phytotoxic than that from mining activity and smelter emissions (Warren et al. 2003). However, for Pb, even when soil properties including pH and organic matter content are factored into the analysis, prediction of the uptake of Pb into edible crops has been elusive (Hough et al. 2004). Despite the

weak correlation of soil Pb contamination to crop contamination, vegetables grown in urban or otherwise Pb-contaminated areas can have substantially higher Pb levels than market vegetables presumably grown largely in rural uncontaminated regions (McBride et al. 2014, Jorhem et al. 2000; Moir and Thornton 1989; Stilwell et al. 2008; Samsoe-Petersen et al. 2002). This apparent contradiction could indicate that vegetable contamination by Pb is largely due to aerial transport and deposition of particles as suggested by a number of studies (Douay et al. 2008; Mosbaek et al. 1989; Voutsa et al. 1996). Furthermore, the random nature of soil particle contamination of vegetables by dust and splash may obscure any dependence of crop Pb concentration on local soil Pb concentration. In fact, previous research has indicated high Pb concentrations in leafy greens and some other crops to be generally caused by physical contamination rather than uptake via plant roots (Chamberlain 1983; Mosbaek et al. 1989; Nali et al. 2009; Prasad and Nazareth 2000; McBride et al. 2012; McBride et al. 2014). To a lesser degree, physical contamination can also contribute to elevated As in some vegetables, particularly root crops (Nathanail et al. 2004).

Crop type is a critical determining factor along with soil properties in determining the extent of As and Pb bioaccumulation. Arsenic in particular shows a very wide range of potential for uptake and translocation from roots to shoots, with several Brassicaceae having a high uptake potential (Raab et al. 2007). Stilwell et al. (2006) measured high concentrations of As in some crops (arugula and chives) but not others grown on soils contaminated by As from pressure-treated wood. Our results from greenhouse studies to date reveal substantial risk of exceeding international standards for vegetable crops when soil total As exceeds 100 mg kg⁻¹ and Pb exceeds 400 mg kg⁻¹ (McBride 2013). Nevertheless, there is still insufficient basic knowledge to make even approximate predictions of As or Pb content for a wide range of crops grown on soils with different properties. The limited data available point to the primary importance of crop type (species and cultivar), with soil properties such as pH, total metal content, texture and organic matter content also having some role in determining the crop uptake of Pb and As.

The field study described in this paper aimed to improve understanding of the relationship between concentrations of Pb and As in vegetable crops and those in orchard soils with consideration of soil properties. The specific objectives of the study were to 1. Determine the extent of uptake of Pb and As from an historically contaminated orchard soil into vegetables commonly grown in gardens, and 2. Assess the effectiveness of heavy compost amendment in reducing uptake or transfer of Pb and As to plants.

Materials and Methods

Characterization and Amendment of Soils in Orchard Site

Seven locations within an old apple orchard on the Dilmun Hill Student Farm at Cornell University, Ithaca, NY, were selected in early 2010 based on initial soil testing results to represent a range of soil Pb and As from near-background to substantially contaminated (50-1000 and 20-250 mg kg⁻¹ of total Pb and As, respectively). The soils at the locations chosen are silty clay loams (Hudson series), and were mechanically tilled with a rotavator prior to constructing miniplots with wooden frames (3.7 x 0.6 m) with equally spaced board dividers in order to separate the control (no compost), 1x compost, and 2x compost treatment sub-plots. The compost amendments (Cornell compost derived largely from food waste and animal bedding) were added at levels calculated to provide 5% (1x) and 10% (2x) organic matter to the soils on a dry-weight basis, and thoroughly mixed into the soils manually to a depth of about 15 cm. Soil pH measured in 2012 ranged from about 6.0 to 7.5 in all the sub-plots, with the 5% and 10% compost treatments tending to be toward the high end of the range. The total soil Pb and As

concentrations were measured in composite samples (one collected from each sub-plot) at the beginning of the experiment and at the end of the cropping sequence by the Cornell Nutrient Analysis Lab (CNAL) using EPA method 3051 for soil digestion and inductively coupled plasma optical emission spectrometry (ICP-emission) for analysis of digests. The soil organic matter (SOM) contents of all sub-plot composite samples were measured in the fall of 2012 using loss-on-ignition. This was done by measuring weight loss of 5 g soil in a crucible, pre-dried at 105°C, after heating the soil in a muffle furnace at 375°C for 24 hours (Davies 1974).

A subsample of the compost used to amend the soils contained very low As (0.97 mg kg⁻¹) and Pb (16.1 mg kg⁻¹), with a pH in water of 7.0. The effect of these low concentrations was to dilute the soil As and Pb concentrations in the 5% and 10% compost treatments. The concentrations of major elements in the compost were 4.41% calcium (Ca), 1.62% magnesium (Mg), 1.51% potassium (K), 0.82% phosphorus (P), 0.45% sulfur (S), 0.88% iron (Fe) and 0.60% aluminum (Al). Concentrations of other trace elements were 23.9 mg kg⁻¹ nickel (Ni), 65.4 mg kg⁻¹ copper (Cu), 507 mg kg⁻¹ manganese (Mn), 42.7 mg kg⁻¹ chromium (Cr), and 298 mg kg⁻¹ zinc (Zn).

Multiyear Cropping Sequence and Plant Tissue Analysis

In the growing seasons of 2010- 2012, lettuce (Lactuca sativa, cv. "Black-Seeded Simpson"), carrots (Daucus carota var. sativus, cv. "Royal Chantenay"), and green beans (Phaselous vulgaris, cv. "Flaco Bush") were seeded directly while tomato seedlings (Lycopersicon esculentum, cv. "Containers Choice Red") were transplanted into each of the subplots. The edible portions of all crops were harvested at the stage of maturity appropriate for fresh consumption. Individual crop samples include tissue collected from 3-4 locations within each sub-plot in order to obtain representative samples and mitigate the effects of genetic variability among plants, as well as to obtain sufficient dried tissue (.5 g) for analysis. Different parts of the plant (e.g., stems vs. leaves) were analyzed together as one sample. Harvested vegetables were washed thoroughly in the laboratory using tap water, and carrot roots were scrubbed to remove any observable adhering soil particles, but were not peeled. All vegetable samples were heated in an oven at 70°C until dry, then ground using a stainless steel mill, which was cleaned by a high-pressure air stream after each sample. The vegetable tissue samples were analyzed for Pb and As using inductively coupled plasma mass spectrometry (ICP-MS) by a certified commercial laboratory (H2M Laboratories, Melville, NY) and by CNAL. Procedures used to ensure precision and accuracy in the measurements of Pb and As in the vegetables and soils included the use of certified plant and soil standards (i.e., standards from the National Institutes of Standards and Technology) as well as laboratory internal standards along with duplicate samples and blanks in each sample set. Those vegetable tissue As concentrations at or below the detection limit (0.01 mg kg⁻¹) were reported as 0.01 mg kg⁻¹. A subset of 15 lettuce samples was also analyzed by CNAL for Al in order to estimate soil particulate contamination of the crop and assess whether the high Pb content of some lettuce samples was due to physical contamination by soil particles rather than uptake through plant roots.

Crop failure was relatively common for some of the crops in certain sub-plots as a result of poor germination due to poor soil structure, weather conditions and pest damage. Consequently, numerous gaps in crop metals data existed, so that the analytical data for Pb and As in vegetable crops collected over all three growing seasons were combined in order to provide greater statistical power in testing the effect of soil Pb and As contamination level and compost addition on the levels of these two elements in the crops. This pooling of data was justifiable since a

preliminary data analysis generally showed no significant effect of crop year on Pb and As contents of the four crops (with the possible exception of tomatoes in 2012, which tended to contain higher As than those in the earlier 2 crop years), whereas inspection of the data revealed that crop type and soil As and Pb concentrations strongly influenced vegetable contamination level.

Single Season Leafy Vegetable Experiment

In spring 2011, two of the miniplot sites with contamination levels of 130 mg kg⁻¹ Pb and 310 mg kg⁻¹ Pb were diverted from the main cropping experiment in order to investigate the tendency of different leafy green vegetables to take up Pb and As. Lettuce (*Lactuca sativa*, cv. "Black-seeded Simpson"), arugula (*Eruca sativa*), spinach (*Spinacia oleracea*, cv. "Melody") and collards (*Brassica oleracea*, cv. "Vates") were grown, with harvest of the fresh leaves occurring in early July. Plant samples were processed and analyzed for Pb and As as described above.

Multiple regression analysis of the data was done using StatView (SAS Institute, Cary, NC). A *p*-value of 0.05 or lower was considered significant.

Results and Discussion

Factors Determining Pb and As Contents of Vegetable Crops

The two independent variables tested in this field study for their effect on crop metal level were soil total metal level and soil organic matter (SOM) content. Thus, the linear equations tested for prediction of Pb and As in the vegetable crops had the form:

 $(M)_{CROP} = a + b(\% SOM) + c(M)_{SOIL}$ (Eq. 1)

for the purpose of multiple regression analysis, where (M)_{CROP} and (M)_{SOIL} are the total concentrations of As or Pb in the crop (mg kg⁻¹ d.w.) and soil (mg kg⁻¹), respectively, and a, b and c are fitting coefficients for the equation intercept, organic matter term and soil total metal term, respectively. The miniplots were amended with 5% and 10% compost prior to cropping at the beginning of the experiment (2010), and the measured SOM in the fall of 2012 was on average 3.87 and 6.78% higher in SOM content, respectively, than the control sub-plots (data not shown). Since these increases in SOM were somewhat lower than the initial amendment, it is likely that decomposition over more than 2 years had reduced SOM contents in the compostamended sub-plots from their initial values. The SOM values measured in 2012 were used in multiple linear regressions along with the measured soil Pb and As concentrations to test whether these soil variables had a statistically significant effect on Pb or As concentrations in the crops. The coefficients of the best-fit linear equations for all crop-year data combined (see equation 1) are summarized in Table 1. The results for both Pb and As in all 4 vegetables indicate a very high level of significance for soil total metal concentration (c coefficient), but no significant effect of SOM content (b coefficient). This analysis indicated that vegetable Pb and As concentrations increase with increasing soil total Pb and As, respectively, but soil amendment with compost had no overall significant effect in altering Pb or As concentrations in any of the vegetable crops.

The nominal compost loading levels of 5% and 10% organic matter were alternatively tested in the regressions in place of measured SOM, but the resulting best-fit equations produced were virtually the same (results not shown), with the % compost term not being statistically

significant except for Pb in beans (p=0.023). The results confirm the strong effect of soil total As and Pb on contamination of the edible portion of all 4 vegetable crops tested, and a weak or negligible effect of SOM level on vegetable contamination. It is important to point out, however, that the "metal dilution effect" of compost amendment to the contaminated soils probably provided some benefit in reducing metal transfer into vegetables. Because compost amendment generally lowered total soil Pb and As concentration, and these post-amendment metal concentrations were used in the multiple regression equations, the potentially beneficial dilution effect was not specifically evaluated in the best-fit equations described in Table 1.

The crop metal- soil metal relationships displayed in Figures 1-4 were generated by combining measured concentrations of Pb and As in each of the 4 crops over all 3 crop years and all compost (0, 5, 10%) treatments (since crop year and compost treatment generally had no statistically significant effects on tissue Pb and As). Highly significant positive correlations between Pb and As concentrations in the edible portion of the crops (lettuce, carrot, bean, tomato) and soil total Pb and As, respectively, are evident in these figures. The figure data are most clearly presented in log-log plots because of the high degree of individual variability measured in the Pb and As contents of each individual crop grown in the miniplots despite the controlled field experimental conditions. The figures clearly show that increased Pb and As concentrations in the soil resulted in generally greater crop contamination by these metals, with the average plant uptake factors (UFs) represented by the c-coefficients presented in Table 1. The ranking of uptake factors by crop followed the order:

Pb: carrot \geq lettuce > bean > tomato

As: lettuce > carrot > bean > tomato

Thus, lettuce had the strongest ability to accumulate As, whereas carrot and lettuce were the strongest Pb accumulators.

To evaluate physical contamination in leafy crops, 15 lettuce samples selected from the miniplots experiment were analyzed for Al (an indicator of soil particle contamination) in addition to Pb. Consistent with results from other studies (McBride et al. 2012; 2014), the tissue Pb and Al concentrations were significantly correlated (r = 0.574, p<0.05), an indication that soil particle contamination of the foliage probably accounted for many of the elevated Pb levels measured in field-grown lettuce. The high clay and exchangeable base cation content, as well as the generally non-acid nature of the orchard soils from this site, maintain soluble and phytoavailable Pb at very low concentrations in this soil (McBride, 2013). Physical contamination (soil particle adherence) may therefore be the most important mechanism of Pb transfer to the leafy and root crops in particular.

A Comparison of Pb and As Uptake in Leafy Green Vegetables

The research described above identified lettuce as the strongest accumulator of the 4 crops for both Pb and As, but it is unlikely that all leafy greens have the same tendency to accumulate these metals. We therefore conducted a one-year experiment on two of the miniplots representing "low" (Pb = 140 mg kg⁻¹, As = 50 mg kg⁻¹) and "moderate" (Pb = 330 mg kg⁻¹, As = 75 mg kg⁻¹) contamination, with a focus on several leafy green vegetables in addition to lettuce, specifically arugula, spinach, and collards. Table 2 summarizes the Pb and As concentrations measured in the harvested vegetables from the sub-plots that had not been amended with compost. Of note are the substantially greater Pb content of all vegetables grown in the moderately contaminated soil compared to those grown in the less contaminated soil, and the marked differences in Pb and As content of the different crops, particularly the low Pb in the collards and high As in arugula. The effect of compost amendment on Pb and As content of lettuce and arugula is shown in Table 3. Compost had the effect of reducing Pb and As in lettuce at the 10% amendment level but not at the 5% level. Arugula had lower Pb and As concentrations at the 5% level, and possibly at the 10% level as well, but the lack of replicates in arugula sampling prevents a more certain conclusion. Similar beneficial effects of compost in reducing crop Pb and As were observed for spinach, but not for collards (results not shown).

The beneficial effect of compost in reducing Pb and As in leafy vegetables, although detected in this single-crop study conducted about 1 year after amendment, was not observed overall in the larger multi-year study with lettuce and 3 other crops. There could be several reasons for this. Specifically, the multiple regression analysis, discussed above, which failed to show the compost amendments to reduce either As or Pb transfer into the crops (with the exception of Pb in green beans), was conducted in such a way that the physical dilution of Pb and As concentrations in the soils by compost addition was excluded as a beneficial effect of the amendment, whereas the crop Pb and As decreases evident in Table 3 could be due in part to this dilution effect. Conversely, higher levels of organic matter, P, and other nutrients in the compostamended soils may have reduced bioavailability. In the case of Pb, this is likely due to strong binding/chelation with organic matter or possibly a precipitation reaction of Pb with phosphate (PO₄). Arsenic, however, likely is not chelated by organic matter, but PO₄ and organic acids present in compost may compete with arsenate for soil adsorption sites and affect As availability. Our earlier research showed the bioavailability and extractability of Pb in these orchard soils to be reduced by compost amendment (Fleming et al., 2013). However, the same research showed As extractability to be enhanced by compost, so that greater As uptake would have been anticipated in the compost treatments. The fact that As concentrations in leafy greens measured here were reduced in the compost-amended soils may be the result of competition effects on plant uptake (e.g. from PO₄ in the compost) or from the well-known growth "dilution effect" on trace elements (Jarrell and Beverly, 1981). Based on visual comparisons of yields, vegetable growth was markedly improved in the compost-amended soils compared to the controls.

Implications for Human Health

No national health-based standards for Pb and As in vegetables, fruits or other staple food crops exist in the United States as of yet. Standards have been developed for Pb in vegetables and other food crops in other countries; the EU has set standards at 0.1 mg kg⁻¹ (f.w.) for most vegetables (including fruits and roots), and 0.3 mg kg⁻¹ for leafy greens (EC, 2006). Presently, China appears to be the only country with standards for As in most vegetable and grain crops. The Chinese standards for As are 0.5 mg kg⁻¹ (f.w.) in rice, beans and vegetables, and 0.2 mg kg¹ (f.w.) in potatoes (NFHPC, 2012), whereas the FAO/WHO have set a standard of 0.2 mg kg⁻¹ in rice only (FAO/WHO, 2014).

In order to consider our results in the context of existing health protective polices, we used the available international standards to arrive at tentative guidance values for measured Pb and As concentrations in the vegetables. Thus, we used the EU standards for Pb (0.3 mg kg⁻¹ for lettuce and 0.1 for carrots, beans, and tomatoes), and the more conservative (0.2 mg kg⁻¹) of existing standards for As for all four vegetables. Using approximate water contents of the vegetables included in this study, standards were converted to Pb and As concentrations on a dry weight (d.w.) basis. Vegetable Pb and As concentration data (Figures 1-4) were then compared to these guidance values. For lettuce, the standards on a d.w. basis are 5.9 mg kg⁻¹ for Pb and 3.9 mg kg⁻¹ for As. For carrots, the standards are 0.8 and 1.6 mg kg⁻¹ for Pb and As, respectively. For

both beans and tomatoes, they are 0.9 mg kg⁻¹ for Pb and 1.8 mg kg⁻¹ for As. These values are indicated on Figure 1 as horizontal dashed lines, and allow the estimation of tentative "threshold" (highest) concentrations of Pb and As in the orchard soils at which the tested vegetables still have no exceedances of acceptable levels of these toxic metals. For Pb, these soil threshold concentrations (in mg kg⁻¹ and rounded to one significant figure) are 1000 (tomato), 200 (lettuce), and 200 (bean). Data at the lowest soil concentrations are insufficient to confidently determine a threshold for carrots, but we can conclude that it would be less than 100 mg kg⁻¹. For As, the comparable soil threshold concentrations (mg kg⁻¹) are 50 (lettuce) and 200 (bean). Due to limitations of the data, thresholds for tomatoes and carrots cannot be accurately determined, but it can be inferred that they would be greater than 300 and less than 100, respectively. These thresholds are likely to be site-specific because of the effect that local soil properties and contaminant source have on plant availability of Pb and As. For example, two recent studies (Attanayake et al. 2015, Defoe et al. 2014) showed very limited As uptake into root crops and leafy vegetables, even though soil total arsenic concentrations at study sites were as high as 95 and 146 mg kg⁻¹. Arsenic contamination from lead arsenate pesticides may represent a higher level of bioavailability than forms of As from some other contaminant sources (e.g., certain brownfield sites).

This analysis suggests that in some settings, communicating risk information to gardeners about acceptable soil levels of Pb and As for vegetable gardening can be refined by specifying preferred crops for soil conditions. Findings support the gardener exposure reduction practice of growing fruit crops in preference to leafy and other crop types where soil Pb and As concentrations are elevated. While the association between soil and vegetable contaminant concentrations can be obscured under diverse field conditions (McBride et al. 2014), this more targeted study suggests that tomatoes (and presumably a number of other fruit crops) can be grown in substantially contaminated soils (up to 1000 mg kg⁻¹ Pb and more than 300 mg kg⁻¹ As, in the case of these lead-arsenate contaminated orchard soils) without exceeding health-based standards, but carrots (and other root crops) and certain leafy greens may exceed standards when grown in orchard soils of similar type with more than 50-100 mg kg⁻¹ Pb or As.

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Conflict of Interest

The authors declare that they have no conflict of interest.

Table 1. Coefficients ("b-coeff") and "c-coeff") and multiple regression r-values for predictive equations of the form: $(M)_{CROP} = a + b(\% \text{ soil organic matter}) + c(M)_{SOIL}$

where $(M)_{CROP}$ and $(M)_{SOIL}$ are the concentration of As or Pb in the crop (mg kg⁻¹, d.w.) and soil (mg kg⁻¹), respectively. For statistically significant variables, p-values are given in parentheses. ns: not significant.

Crop	Pb			As		
	b-coeff	c-coeff	r-value	b-coeff	c-coeff	r-value
Bean	-0.04	0.0012	0.59	-0.01	0.0057	0.73
(n=47)	ns	(<0.0001)		ns	(<0.0001)	
Tomato	0.001	2.3 x10 ⁻⁴	0.65	0.001	0.00079	0.54
(n=73)	ns	(<0.0001)		ns	(<0.0001)	
Lettuce	-0.05	0.018	0.57	-0.03	0.058	0.83
(n=68)	ns	(<0.0001)		ns	(<0.0001)	
Carrot	-0.4	0.022	0.50	-0.03	0.0097	0.65
(n=23)	ns	(0.0193)		ns	(0.001)	

Table 2. Mean (\pm SD) of Pb and As concentrations (mg kg⁻¹ d.w.) in lettuce, arugula, spinach and collard greens grown in unamended field sub-plots with low (140 mg kg⁻¹ Pb, 50 mg kg⁻¹ As) and moderate (330 mg kg⁻¹ Pb, 75 mg kg⁻¹ As) metal contamination. The number of replicates is indicated in parentheses. Values with different letters indicate significant differences among means (ANOVA, $p \le 0.05$).

Crop	Crop Pb	$(mg kg^{-1})$	Crop As (mg kg ⁻¹)		
	Low	Moderate	Low	Moderate	
Lettuce	1.65 ± 0.17 a (4)	$11.5 \pm 2.1 \text{ c} (2)$	2.35 ±0.09 a (4)	$5.45 \pm 0.64 \text{ e}(2)$	
Arugula	1.60 ± 0.24 a (4)	$4.13 \pm 0.32 d (4)$	$12.8 \pm 1.50 \text{ b} (4)$	11.6 ± 2.3 be (4)	
Spinach	$1.40 \pm 0.00 a(2)$	10.0 ± 1.4 c (2)	$0.64 \pm 0.02 \text{ c} (2)$	$0.86 \pm 0.21 \text{ c} (2)$	
Collards	0.27 ± 0.21 b (2)	2.40 ± 1.7 ab (2)	1.30 ± 0.14 d (2)	$1.15 \pm 0.07 \text{ d}(2)$	

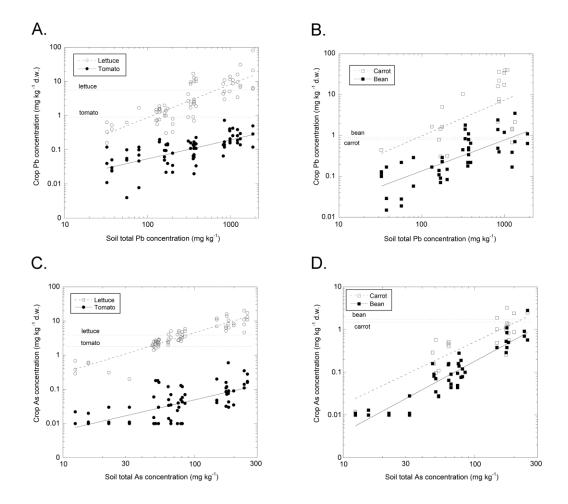
Table 3. Mean (\pm SD) of Pb and As concentrations (mg kg⁻¹ d.w.) in lettuce and arugula grown in field miniplots with moderate (330 mg kg⁻¹ Pb, 75 mg kg⁻¹ As) metal contamination and amended with compost at two levels. The number of replicates is indicated in parentheses. Soil Pb and As concentrations (mg kg⁻¹) are also shown, reflecting concentrations post- amendment for the 5% and 10% compost treatments. Values with different letters indicate significant differences among means (ANOVA, p \leq 0.05).

Compost Level	Soil Pb (mg kg ⁻¹)	Soil As (mg kg ⁻¹)	Crop Pb (mg kg ⁻¹)		Crop As (mg kg ⁻¹)	
	((Lettuce	Arugula	Lettuce	Arugula
0%	330	75	$11.5 \pm 2.1 \text{ c} (2)$	4.13 ± 0.32 a (4)	5.45 ±0.64 b (2)	$11.6 \pm 2.3 \text{ c} (4)$
5%	330	76	9.0 ± 0.71 c (2)	1.45 ± 0.21 b (2)	4.9 ± 0.42 b (2)	3.85 ± 1.1 b (2)
10%	310	63	3.5 ± 1.1 a (2)	2.8 (1)	2.25 ± 0.35 a (2)	3.2 (1)

Figure 1. Relationship of vegetable log-transformed Pb concentration (mg kg⁻¹ d.w.) to logtransformed soil total Pb or As concentration (mg kg⁻¹). Soil concentrations are post-cropping. The horizontal broken lines show the d.w equivalents of international health-based standards for Pb or As in vegetables.

- A. Tomato and lettuce Pb concentration vs. soil total Pb concentration.
- B. Bean and carrot Pb concentration vs. soil total Pb concentration.
- C. Tomato and lettuce As concentration vs. soil total As concentration.

D. Bean and carrot As concentration vs. soil total As concentration.



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