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Comparison of Heavy Metals in Community Garden Produce versus Store-Bought Produce

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INTRODUCTION

Community gardens are community-managed spaces that are open to the public. However, activities such as transportation, construction and manufacturing have resulted in increased heavy metals, notably lead, in the soils surrounding these activities (Stilwell et al. 2008). Although soil ingestion is expected to be the major source of exposure in contaminated gardens, consumption of plants grown in these soils is also important to consider.

The amounts of metals in plants, and the various factors influencing their uptake, have been reviewed (Pendias and Pendias, 1996). Important variables, which influence the metal content in plants, include soil pH and metal solubility in water-soil solutions. Plant uptake and transport of metals also depends on the part of the plant that is consumed and follows the general order root>leaf>fruit. Although lead remains the most common garden soil contaminant, only limited data have been published on its content in plants grown in urban gardens. Finster et al. (2004) conducted a field survey of the lead levels in edible crops grown in contaminated residential soils. They confirmed that lead in fruiting crops was low (less than the detection limit of 10 mg/kg lead, dry weight). However, they did find that the lead in the edible portion of many leafy vegetables exceeded 10 mg/kg (dry weight) and could contribute to the total body burden of lead. Unfortunately, the high detection limit for lead in this study (10 mg/kg) is above the limits for lead in plants (3-8 mg/kg, dry weight) set by various regulatory agencies (CODEX 2003; Berlin 2008). Boon and Soltanpour (1992) presented a summary on lead contamination in garden plants. In many instances the lead in leaf and root crops exceeded 10 mg/kg, but these studies were carried out when leaded gas was still in use. Models, equating uptake factors to metal contents in plants, have been proposed by Hough et al. (2004) to assess the risk of heavy metal exposure from consumption of home-produced vegetables in urban populations in England. Using these models, they concluded that the cadmium, copper, nickel, and zinc content in food crops were satisfactory (low risk), whereas the lead uptake model was too inconclusive to assign risk.

In this report we present our findings on a comparison of the heavy metal content in Connecticut community garden grown produce to store bought produce using inductively coupled mass spectrometry (ICP-MS) for sample analysis.

MATERIALS AND METHODS

In this study, we acquired 10 community garden-grown produce samples from 4 gardens, and compared their heavy metal content to those in 18 store-bought produce samples. In two cases, the non-edible portions of the plants were taken to compare relative uptake. The garden-grown produce samples consisted of lettuce, broccoli, bell peppers, cilantro, collard and red cabbage. Soil samples from these gardens were also taken for analysis. The store-bought produce included a variety of salad greens, cilantro, bell peppers, and collard greens. In two cases each for bell peppers and collards, the produce was labeled as organically grown.

All of the produce samples were cut into manageable size (6 by 2 cm or less) using a stainless steel scalpel, then washed using tap water, followed by a final rinse in deionized water. After air drying on paper towels, the samples were placed in paper bags at 60 C° for 10 hours (Stilwell et al. 2006). The samples were weighed in the bag before and after drying in order to obtain the percent moisture. The empty bags were dried in the oven before use and stored in a desiccated environment to ensure that the bags were free of moisture. Dried leafy material was crushed in the bag prior to placing the plant materials into capped polypropylene specimen containers, which were then stored in a desiccator prior to analysis.

The plant tissue samples (0.25g nominal) were prepared for analysis by weighing into 50 ml polypropylene digestion vessels, adding 5 ml of concentrated nitric acid followed by digestion using a hot block (DigiPrep) at 115 C° for 45 minutes. The samples were brought up to volume (50 ml) using purified water, and an additional 10:1 water dilution was performed directly before analysis. The heavy metals were determined using an Agilent 7500ce Inductively Coupled Plasma Mass Spectrometer with an octopole

reaction system, using the manufacture's guidelines concerning tuning, elemental masses, and reaction gases. The resulting detection limits ($\mu\text{g}/\text{kg}$) in the plant material (dry weight basis) was 4 for Uranium (U), 10 for Arsenic (As), Cadmium (Cd), Chromium (Cr), Nickel (Ni), and Lead (Pb), 15 for Thallium (Tl), 20 for Copper (Cu), 60 for Zinc (Zn). Unless otherwise indicated the results are reported on a dry weight basis.

Method validation consisted of determining the heavy metals in fortified and unfortified blanks (in duplicate), and in triplicate measurements of three plant materials (National Institute of Standards and Technology (NIST) Standard Reference Material 1570a Spinach Leaves, store bought packaged Baby Arugula, and Mediterranean blend greens). The fortification level was at $0.5 \mu\text{g}$ per sample except for Cu and Zn which were fortified at $2.5 \mu\text{g}/\text{sample}$. Thus, for a sample weight of 0.25 g , the fortification level was $2000 \mu\text{g}/\text{kg}$ or for Cu and Zn, $10000 \mu\text{g}/\text{kg}$. The average percent recovery of all materials ($n=4$) is given in Table 1, and a comparison of the obtained values to the certified values in the reference material is given in Table 2. Note that in both cases, there was good agreement between the obtained and the expected results, except for the As levels in the NIST standard material, which we attribute to the fact the As level in the material was close to the detection limit of As.

Table 1. Average and standard deviation ($n=4$) of percent recoveries of fortified samples.

Element	Average Spike Recovery (%)			
As			99±4	
Cd			102±3	
Cu			98±5	
Cr			93±4	
Ni			101±7	
Pb			99±2	
Tl			96±6	
Zn			86±21	
U			101±4	

Table 2. Comparison of elemental concentrations (mg/kg) in NIST 1570a reference material (spinach leaves) to those determined (in triplicate) using our ICP-MS method (values for Cr and Tl are not given by NIST for this material).

Element/Mass	Found	NIST Value*	% Agreement
Ni / 58	2.20	2.14	103
Ni / 60	2.24	2.14	105
Cu / 63	12	12.2	94
Cu / 65	12	12.2	101
Zn / 66	75	82	91
As / 75	0.04	0.07	62
Cd / 111	2.68	2.89	93
Cd / 114	2.45	2.89	85
Pb / 206	0.16	0.20	79
Pb / 207	0.16	0.20	80
Pb / 208	0.15	0.20	76
U / 238	0.14	0.16	87
NIST concentrations are certified except for Pb, and U which are given as informational.			

Soil samples were collected at the gardens where plant tissue samples were obtained. In one garden (garden B), the samples consisted of 2 composites of the entire garden. For the others, soil composite samples were taken within 2 meters of the corresponding plant sample. These soil samples were prepared in a similar fashion, but not subsequently diluted (10:1) and run on a less sensitive instrument (ICP-OES), precluding the analysis of Tl and U (Stilwell et al. 2006; 2008).

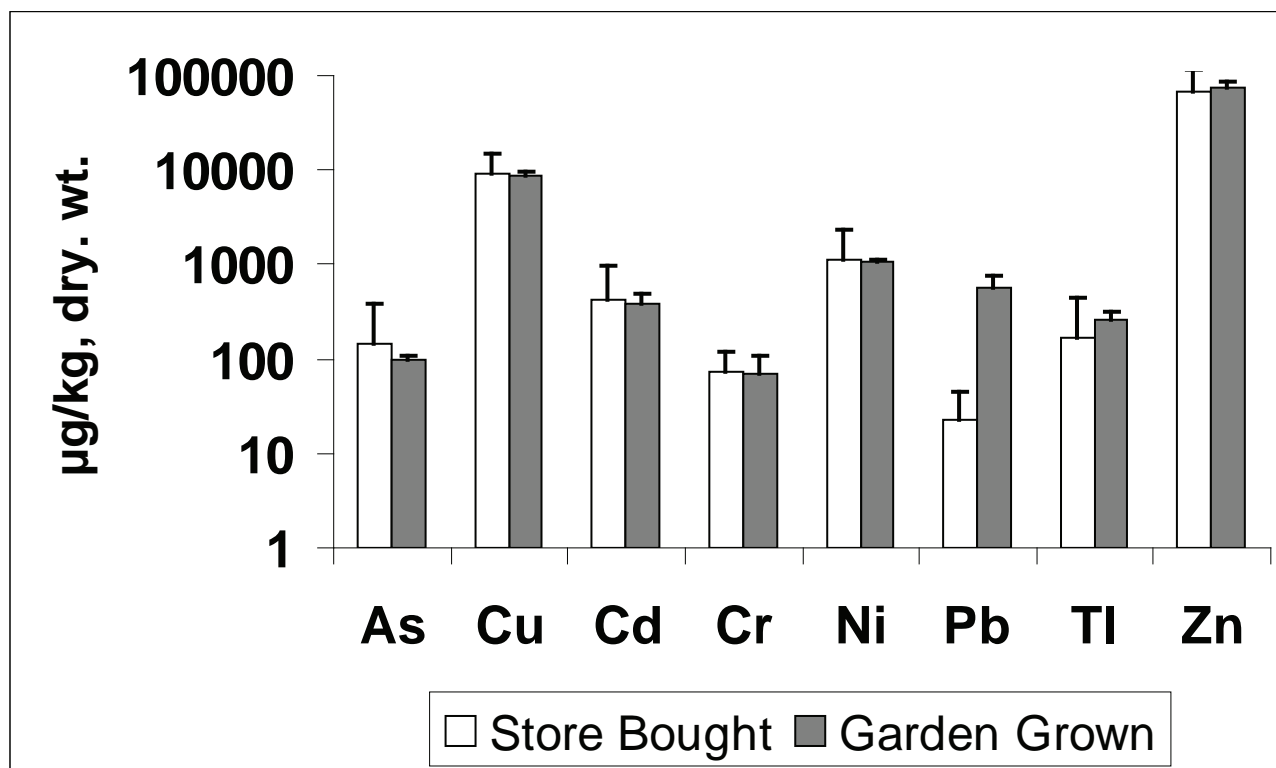
RESULTS

A comparison of the average amounts of the various heavy metals in store bought versus community garden produce is shown in Figure 1 (note log scale). As shown in the figure the only significant difference was in the Pb content. The lead content ($\mu\text{g}/\text{kg}$) in all of the store bought produce ranged from <10 to 94, and averaged 23 ± 21 , while it ranged from <10 to 2807 in the garden-grown produce, averaging 565 ± 205 . The increased Pb in the produce was caused, presumably, by the higher amounts of lead in the garden soil, which ranged from 40-450 mg/kg. For example, the average Pb ($\mu\text{g}/\text{kg}$) in all of the store-bought greens (including green leaf lettuce, spinach, arugula) was

25 ± 9 compared to 2807 ± 568 in green leaf lettuce grown in Garden A (soil Pb 147 ± 6 mg/kg Pb) and 192 ± 15 $\mu\text{g}/\text{kg}$ in green leaf lettuce from garden B (soil Pb 39 mg/kg nominal). In another example the Pb in cilantro was 94 ± 7 in the store bought sample and 1250 ± 200 in the garden grown sample (garden B, 39 mg/kg soil Pb, nominal). In collards, the store bought lead averaged 24 ± 5 , while the garden-grown (gardens A, C and D) produce ranged from 87-614 $\mu\text{g}/\text{kg}$ and averaged 275 ± 220 in soils where the Pb content ranged from 22 to 450 mg/kg. Even though the lead was elevated in the garden produce they were all below the international limits of 3000-8000 $\mu\text{g}/\text{kg}$ (CODEX 2003; Berlin 2008). A table of all of the results for the heavy metals in each produce sample is given in Appendix A, and the amounts of heavy metals in the soils are given in Appendix B.

There was no association between the amount of soil Pb and plant Pb over all plant types, partly due to the small sample size and the variability in uptake between plants. However, within the brassica group, the sample size was large enough to show a positive association between soil lead and plant lead (Figure 2).

Figure 1. Comparison of average heavy metals ($\mu\text{g}/\text{kg}$, dry wt.) in the edible portions of store-bought and garden-grown produce.



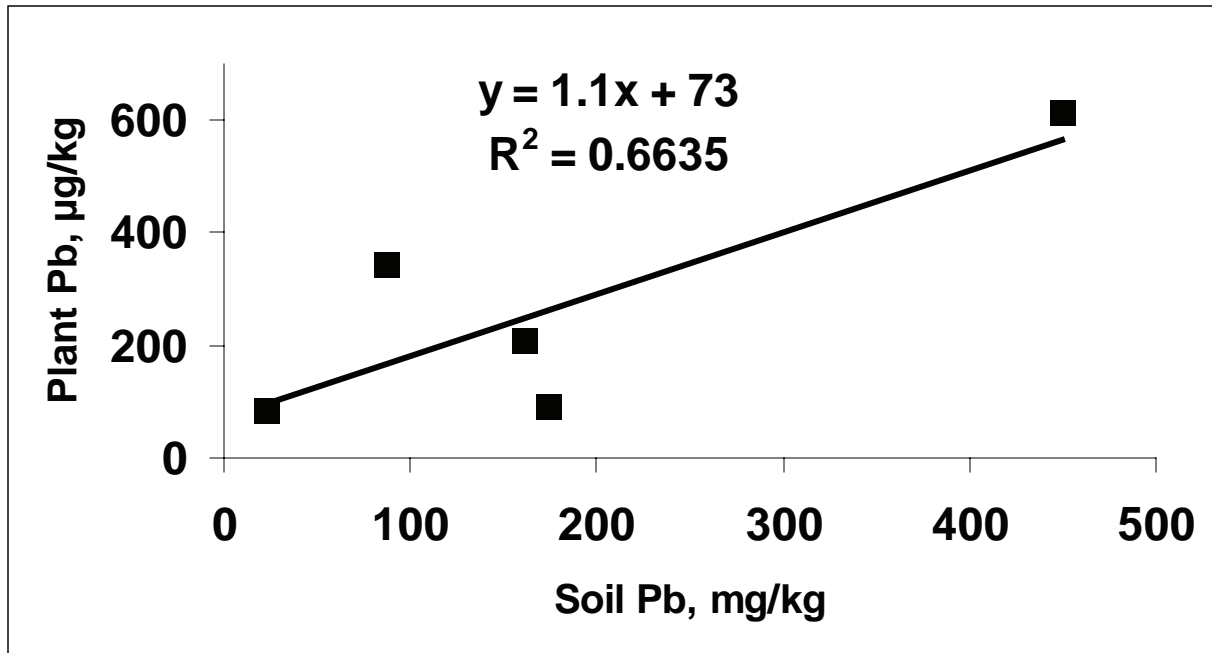


Figure 2. Plant Pb versus Soil Pb in Brassica (Collard greens (n=4) and red cabbage (n=1)).

The uptake of lead in all of the plants was generally low. The uptake factor (Plant Pb/Soil Pb) averaged 0.007 ± 0.008 (0.7%) and ranged from 0.0005 (0.05%) to 0.032 (3.2%). Using the lowest limit for plant Pb (Codex 3000 µg/kg) and the State of CT (1996) limit of 400 mg/kg for soil Pb then a maximum uptake factor of 0.0075 can be calculated ($3/400$). If a plant's uptake factor is less than 0.0075, then the plant lead will be less than the most stringent standard for Pb in produce grown in soil up to the 400 mg/kg limit. In only two of the ten cases was this factor exceeded, in one of the lettuce samples (Garden A, uptake factor = 0.02), and in the cilantro grown in garden B (uptake factor = 0.032). For the other plants (lettuce garden B, broccoli florets, bell peppers, collards, and red cabbage) the uptake factor was less than 0.0075 and thus, these plants could all be grown in soil up to the 400 mg/kg without taking up more than 3000 µg/kg Pb. In fact, by extrapolation of the data in Figure 2 for brassica, the calculated limit for soil Pb in which it would be possible to grow plants below 3000 µg/kg Pb limit is 2660 mg/kg, well above the 400 mg/kg CT limit. It should be pointed out that in the one lettuce sample and in the cilantro, it was not possible to distinguish Pb uptake by the plant from soil contamination on the plant. The difficulty of completely washing all Pb contaminated soil from produce was noted by Finster et al. (2004).

The As levels in the plant samples were all significantly below the 5000-10000 µg/kg foreign limits (Thornton 1994; Querirolo et al. 2000). In garden-grown plants, the As ranged from <10 to 228 µg/kg, and averaged 97 ± 9 , while the As content in commercial produce ranged from <10 to 903 µg/kg, averaging 142 ± 247 (Appendix A). Based on this limited sample size, the brassicaceae plants grown in the gardens and grown organically tended to contain less As, and there appeared to be a positive association between the amount of As in the soil and the amounts in the plants (not shown). The uptake of As in these garden soils, however, was proportionally much less than that observed in plants grown next to As containing treated wood. The uptake factor for As in percent terms ($100 * \text{Concentration Plant} / \text{Concentration Soil}$) was <0.2% to 2.5% in plants grown in the garden soils and 15-250% in plants grown next to the As laden wood (Stilwell et al. 2006). We attribute this large difference to the lower bioavailability due to aging of the soil contaminants. In the case of plants grown next to the treated wood, a fresh supply of As is available due to its continuous leaching from the wood.

The Cd level in the plants exceeded the 1000 µg/kg German limit (Berlin 2008) in both commercial and garden-grown cilantro, and in a commercial bell pepper sample (Appendix A). The overall average in commercial

and garden grown produce was about 400 µg/kg. In this limited sample size, it appears that the organic produce (bell pepper and collard greens) was lowest in Cd, but further work is needed.

Thallium (Tl) uptake by plants is generally limited to members of brassicaceae family. According to Pavlickova et al. (2006), Tl is 100 times more toxic to plants and animals than Cd, and even though Tl uptake from soils can seriously endanger the food chain, there is an absence of threshold limits for Tl in soils, foods, and agricultural products. Based on their study, they proposed a limit for Tl in produce of between 250-500 µg/kg, dry weight basis. In our survey of both store bought and garden grown produce, we also found that Tl was only detected in high amounts in brassicaceae produce. Shown in Table 3 is a comparison of the range, median, and average of all 28 produce samples (store-bought and garden-grown) divided by brassicaceae (collards, red cabbage, arugula, and broccoli florets) and non-brassicaceae. The 2 samples above the detection limit in non-brassicaceae produce were both bell peppers. Of the 13 samples of brassicaceae, 5 collard green samples were above the maximum proposed threshold (500 µg/kg), and one collard sample at 472 µg/kg Tl was between the proposed threshold range (250-500 µg/kg). A larger database is needed to determine which types of plants within the brassicaceae accumulate the most Tl. These results indicate that Tl should be included in heavy metals screening of produce, particularly in members of the brassicaceae family. Tl was not determined in the soils by the ICP-OES method, but in a preliminary determination of Tl in soils by ICP-MS we did not find Tl above published backgrounds (0.02-3 mg/kg, Pendas and Pendas, 1996), and we did not find a relationship between Tl in soil and Tl uptake by the plant, likely due to the low soil Tl levels and the many factors that control Tl uptake (Pavlickova et al. 2006).

The Cu, Cr, Ni, and Zn levels in all of the produce samples were within normal ranges except for the Cu content in the

green leaf lettuce from garden A (Appendix A). The Cu level in this sample was higher than normal (>20000 µg/kg, Wolnick et al., 1983), but not at a level which could be considered excessive (Bunzl et al. 2001; Hough et al. 2004; Jassir et al. 2005; Parveen et al. 2003). The relatively high Zn content in spinach compared to the other greens was also observed by Wolnick et al. (1983, 1985) in a survey of elements in major raw agricultural crops in the United States.

The heavy metal content in the portion of the produce not normally consumed can differ from the edible portion and, in general, follows the order root>stem and leaf>fruit or flower (Stehouer, 1999; Finster et al., 2004). To test this trend in garden produce from this study, we obtained samples other than the fruit from a bell pepper plant and a broccoli plant, and determined their heavy metal profile. In both cases (Figures 3 and 4) the edible portion (fruit or flower) contained considerably less As, Cd, and Pb than the leaf or stems. In broccoli this trend was followed with Tl, (Figure 4), but in the bell pepper plant the Tl content was below the detection limit in all samples.

CONCLUSIONS

The average amounts of Pb in the garden produce were significantly greater than the store-bought produce. The lead content (µg/kg) in all of the store-bought produce ranged from <10 to 94, and averaged 23±21, while it ranged from <10 to 2807 in the garden grown produce, averaging 565±205. Even though the lead was elevated in the garden produce, values were all below the international limits of 3000-8000 µg/kg. A distinction between Pb in the plant and Pb on the plant (in soil residues) could not be made. The cadmium levels in the plants were above the detection limit in all samples, and exceeded foreign limits in both commercial and garden-grown cilantro, and in a commercial bell pepper. Thallium was sometimes detected in high amounts in brassicaceae produce, indicating that Tl should be included in heavy metals screening of that type of produce. The As, Cu, Cr, Ni, and Zn levels in all of the produce samples were within ranges which were not considered excessive. The heavy metal content was higher in the stem and leaf compared to the fruit or flower.

Table 3. Thallium in Produce, µg/kg dry weight basis.

Produce	Number Above Detection Limit	Range	Median	Average
Brassicaceae (n=13)	8	<15-1911	173	420±550
Non-Brassicaceae (n=15)	2	<15-30	<15	<15

Figure 3. As, Cd, and Pb content in fruit, leaves, and stems of a bell pepper plant.

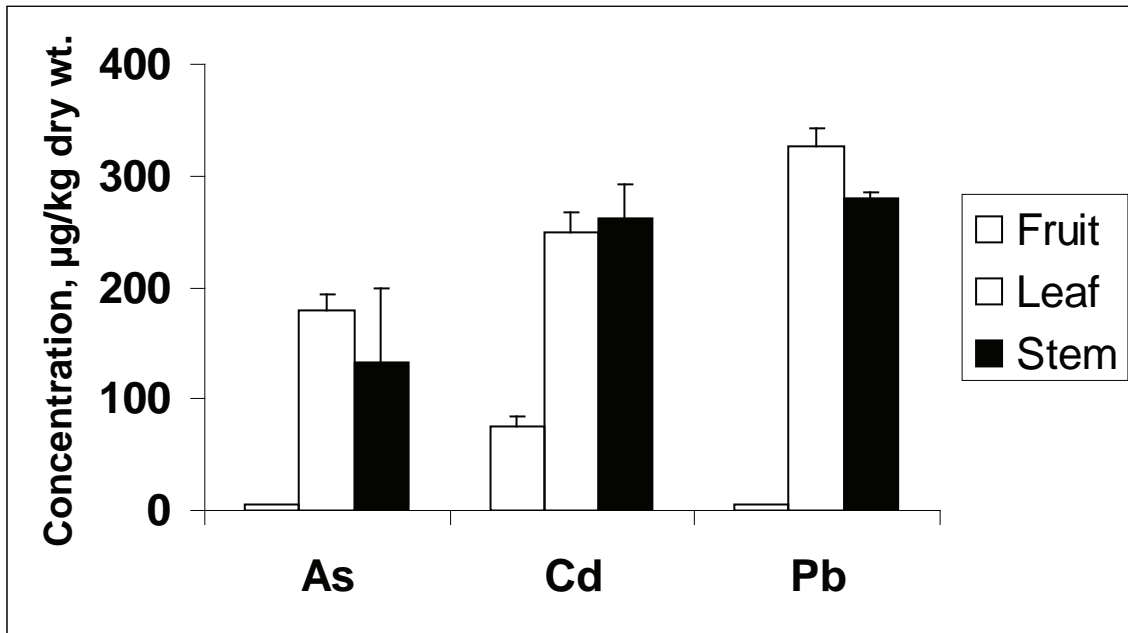
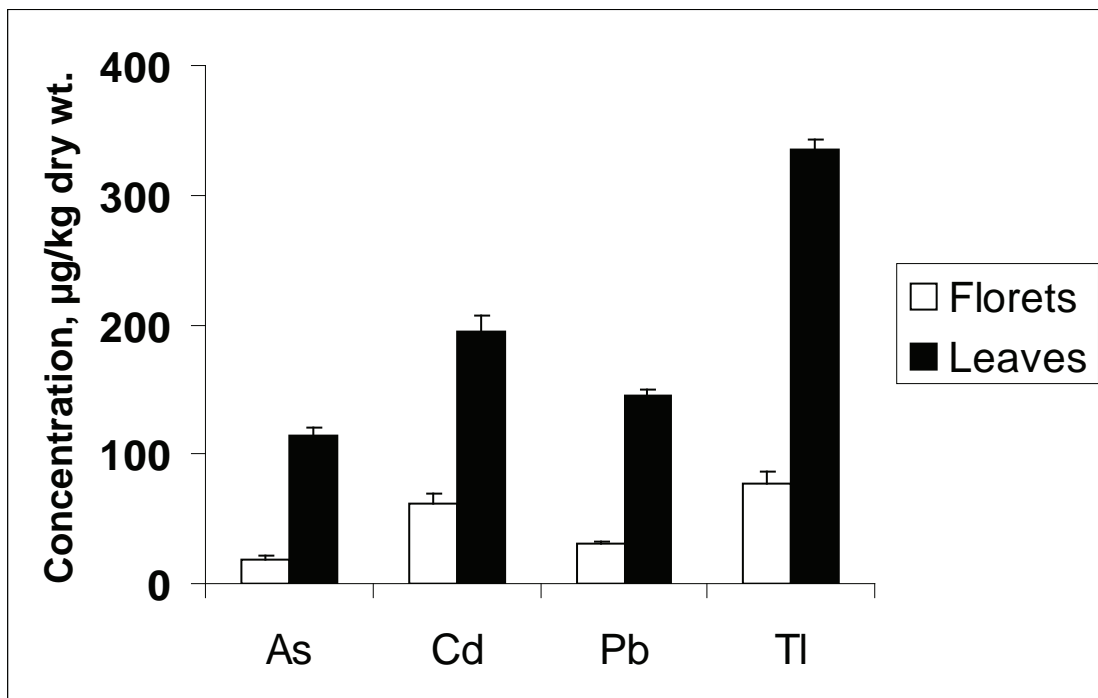


Figure 4. As, Cd, Pb, and Tl in the florets and leaves of a broccoli plant.



Appendix B. Soil heavy metals in gardens where produce was obtained.

APPENDIX B- Heavy metals in soils (mg/kg, ppm dry wt.) near community garden grown produce.

Average Soil Concentration and Standard Deviation, mg/kg (ppm), dry weight basis

<u>Community Garden Soil</u>	Arsenic	Copper	Cadmium	Chromium	Nickel	Lead	Zinc
Green Leaf Lettuce Garden-A	13.3 ± 0.51	76 ± 2	<0.5	10 ± 0.04	8 ± 0.2	147 ± 6	142 ± 0.4
Green Leaf Lettuce Garden-B	11.2 ± 3.0	20 ± 1	<0.5	11 ± 3	9 ± 2	39 ± 19	60 ± 0.1
Cilantro Garden B	11.2 ± 3.0	20 ± 1	<0.5	11 ± 3	9 ± 2	39 ± 19	60 ± 0.1
Broccoli Florets Garden B	11.2 ± 3.0	20 ± 1	<0.5	11 ± 3	9 ± 2	39 ± 19	60 ± 0.1
Bell Pepper Garden B	11.2 ± 3.0	20 ± 1	<0.5	11 ± 3	9 ± 2	39 ± 19	60 ± 0.1
Collards Garden C	5.5 ± 1.4	52 ± 3	<0.5	23 ± 1	15 ± 0.1	161 ± 16	162 ± 11
Collards Garden C	4.5 ± 0.1	30 ± 1	<0.5	21 ± 1	14 ± 0.3	22 ± 0.1	87 ± 4
Red Cabbage Garden D	7.6 ± 0.5	52 ± 1	<0.5	28 ± 1	23 ± 0.1	173 ± 6	210 ± 10
Collards Garden D	9.8 ± 2.2	57 ± 2	0.6 ± 0.04	30 ± 0.4	19 ± 2	450 ± 14	238 ± 9
Collards Garden A	7.2 ± 0.4	37 ± 1	<0.5	8 ± 1.0	<8	87 ± 1	85 ± 3

Garden B average of two soil composites. Gardens A,C, and D, composite soil sample within 2 meters of corresponding plant sample.

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