

**“FIXING” THE URBAN SOIL LEAD PREDICAMENT: THE APPLICATION OF  
IN SITU FIXATION TECHNOLOGY AS AN ECOLOGICALLY SUSTAINABLE  
METHOD OF LEAD ABATEMENT IN URBAN SOILS**

by

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## **DEDICATION**

This work is dedicated to my Mother, Annie Mae Coe Wooten, and my Grandfather, Livous Boston Coe Sr., the sources of any decency I may possess.

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## **ABSTRACT**

### **“FIXING” THE URBAN SOIL LEAD PREDICAMENT: THE APPLICATION OF IN SITU FIXATION TECHNOLOGY AS AN ECOLOGICALLY SUSTAINABLE METHOD OF LEAD ABATEMENT IN URBAN SOILS**

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Lead (Pb) has been used to produce a large number of materials and manufactured products. In areas with a history of lead paint use, high vehicular traffic, or areas close to urban and industrial centers, atmospheric lead deposition may be very high. The aerosol form of Pb can be re-suspended and easily inhaled on fine dust particles or inadvertently consumed in crops grown in lead contaminated soils. At low levels, Pb will impair psychological and neurobehavioral functions particularly in young children. Remediation of lead contaminated soils by conventional methods is expensive. The use of low cost environmentally safe amendments for the in situ fixation of lead in contaminated soil is a promising remediation approach. In situ lead fixation does not reduce the total concentration of soil lead but changes its speciation, thus rendering the lead less toxic and potentially non bio-extractable in the eco-system. The objectives of this study were: (1) to determine if various agricultural, municipal and industrial by-product treatments can

reduce levels of Mehlich III lead in lead contaminated urban soils, (2) to determine if by-product treatments can reduce or prevent Mehlich III lead absorption into the tissues of crops grown in lead contaminated soils, (3) to determine if by-product treatments will affect crop yields, and (4) to determine if by-product treatments can reduce soil and crop tissue lead levels to within Environmental Protection Agency (EPA) or joint World Health Organization and Food and Agriculture Organization (WHO/FAO) standards. Four different agricultural, municipal and industrial by-products that are known to contain chemical compounds that will absorb and fix lead were tested. The by-products selected for study were: poultry litter ash (PLA) as a phosphate source; drinking water treatment residual (DWTR) as an aluminum, sulfate and iron oxide source; steel slag (SS) as an alumina, iron, and magnesium oxide source; and leaf compost (LC) as a source of organic matter. Soils were collected from three urban locations: Ft. DuPont National Park, Washington, DC; a residence in Washington, DC; and a residence in Baltimore City, MD, with average total lead concentrations of 37, 919 and 1528 mg kg<sup>-1</sup> respectively. By-products were mixed with each soil at three rates and incubated moist for 58 days. By-product treatments (except DWTR) resulted in decreasing Mehlich III extractable Pb in the residential DC and Baltimore soils, compared to the un-amended soils. PLA, LC, and SS treatments reduced Mehlich III Pb in the residential DC and Baltimore soils to within EPA permissible limits for garden soil. In comparison, high rate treatments of WTR significantly increased Mehlich III extractable lead levels in both residential DC and Baltimore soils. No treatments met the WHO/FAO standard. Low and high rate treatments of WTR resulted in non-statistically significant Pb increases in

crop tissues. LC and PLA treatments were found to stress crops and reduce crop yields when compared to controls. DWTR increased yields with most crops. The most consistent by-product to reduce or prevent Mehlich III Pb uptake into crop tissues of leafy and root vegetables to within EPA and/or WHO/FAO permissible limits was LC. However, due to the high levels of trace elements and salts in some of these by-products, caution is suggested when using these materials to grow crops.

## **CHAPTER ONE - INTRODUCTION**

The urban “food desert” phenomenon refers to low income areas in which healthy foods are expensive, of poor quality, or inaccessible. The resulting “food insecurity” (hunger resulting from protein energy malnutrition and micronutrient malnutrition) contributes to rising rates of obesity and diet-related chronic disease (diabetes, cardiovascular disease, cancer and obesity) (Shannon, 2014; Alaimo et al., 2008; Smith et al., 2009; Corrigan, 2011; Smith et al. 2013; Hendrickson et al., 2006). One popular idea for improving urban community food security is through the development and use of urban gardens (Shannon, 2013; Corrigan, 2011; Smith et al., 2009; Hendrickson et al., 2006; Short et al., 2007).

### **A. The advantages of Urban Gardening**

Several studies have observed positive associations between urban gardening and health (Kim et al., 2014; Alaimo et al., 2008; Wakefield et al., 2007). Individuals and households that participate in urban gardening activities tend to consume more fruits and vegetables. Alaimo et al. (2008) found that a household member who participated in a community garden consumed 1.4 times more fruits and vegetables per day ( $P < 0.001$ ) and were 3.5 times more likely to consume fruits and vegetables at least 5 times per day than those households without a gardening household member; they also discovered that

communities and homes with gardens consumed more produce than non-gardeners. Also, community and home gardeners met national recommendations to consume fruits and vegetables at least five times a day, compared to non-gardeners. Additionally, researchers have discovered links between increased fruit and vegetable consumption and improved health conditions such as coronary heart disease, oral cancer, and strokes (Dauchet et al., 2006; Bazzano et al., 2002; Pavia et al., 2006; He et al., 2006).

## **B. The disadvantages of Gardening in Urban Soils**

Unfortunately, gardening in urban settings may present a number of health risks stemming from exposure to contaminants that may be present in the soil. Urban soils are often close to pollution sources such as industrial areas and heavily trafficked roadways (Kim et al., 2014; Clark et al., 2008). Consequentially, urban gardeners can be exposed to contaminants through the following ingestion pathways: regular consumption of home-grown produce containing elevated concentrations of potentially toxic substances, accidental or intentional (i.e., pica activity, the consumption of paint chips or flakes, by young children) ingestion of contaminated soil, and inhalation of contaminated soil particles in house dust from adjacent properties or via dermal contact (Alloway, 2004; Kim et al., 2014). Urban gardeners need to be aware of these risks and how to manage them effectively.

It has been widely documented that environmental lead (Pb) contamination is pervasive in urban areas where soil represents a significant sink and source pathway of

exposure (Clark et al., 2008; Clark et al., 2006; Ljung et al., 2006; Laidlaw and Filippelli, 2008; Mingorance and Oliva, 2006). The potential increased exposure to lead by urban gardeners through their gardening and produce consumption activities is a source of concern (Agbenin et al., 2008; Scheyer, 2000; Kachenko and Singh, 2006; Finster et al., 2004; Tokalioglu et al., 2006; Gaw et al., 2008; Roussel et al., 2008; Preer et al., 1995; Preer et al., 1980).

### **C. Using recycled agricultural and industrial by-products to remediate polluted urban garden soils**

Remediation of soils contaminated with Pb has been achieved using a variety of strategies: capping, excavation and landfill burial, solidification, and stabilization. However, these technologies render the land useless for plant growth and tend to be expensive (Berti and Cunningham, 1997). The application of inexpensive agricultural and industrial by-products may render certain types of pollutants found in urban soils non-toxic (non-extractable) consequently rendering the treated soil safer for edible crop production (Puschenreiter et al., 2005; Ruttens et al., 2006; Kucharski et al., 2005; Illera et al., 2004; Alpaslan and Yukselen, 2002).

The goal of this research is to evaluate the effectiveness of several inexpensive agricultural and industrial by-products to render Pb found in contaminated urban soils non-extractable. The ultimate objective is to improve the Pb contaminated soil, rendering it safer for use by urban gardeners.



## **D. Hypotheses**

A review of the literature suggests that a number of agricultural and industrial by-products could potentially be used as additives to improve Pb contaminated urban soils by making the Pb non-extractable and rendering the soil more suitable for gardening.

This research seeks to test the following hypotheses:

1. The addition of agricultural and industrial by-products to lead contaminated soils will reduce the levels of Mehlich III available lead in those soils.
2. The addition of agricultural and industrial by-products to lead contaminated soils will not reduce the harvest yield of crops grown in these soils.
3. The addition of agricultural and industrial by-products to lead contaminated soils will reduce the levels of lead absorbed into the most consumed sections of leafy and rooty crops grown in these soils.
4. The addition of agricultural and industrial by-products to lead contaminated soils will reduce levels of Mehlich III lead in these soils to within U.S. Environmental Protection Agency (EPA) or World Health Organization (WHO) permissible limits for gardening, agricultural or bare soils; also the same by-product treatments will reduce the levels of lead in crops grown in lead contaminated soils to within U.S. EPA or WHO permissible limits for leafy and rooty vegetables.

## **CHAPTER TWO – REVIEW OF LITERATURE**

### **A. Sources of Pb in the Environment**

Unlike overt Pb toxicity, where one source is usually identifiable, low level environmental Pb exposure is caused by a multitude of sources (gasoline, manufacturing and industrial processes, paint, canned foods, battery breaking, and Pb soldered water pipes) and pathways (air, household dust, street dirt, soil, water, and food). Thus, the determination of the relative contribution of environmental Pb is complex and varies between different areas and populations. Lead derived from gasoline comprises the major part of atmospheric Pb. It is a significant contributor to human Pb exposure and is the most widely distributed source of Pb in the environment. Atmospheric Pb (in the form of aerosols) deposited in the soil may be ingested by children and consequently raise their blood Pb levels. For the non-occupationally exposed portions of the general population, food and water are important sources of baseline exposure to Pb, in addition to atmospheric exposure (Tong et al., 2000).

### **B. Why Pb is a Problem**

Unlike organic contaminants, lead in the soil environment does not undergo microbial or chemical degradation. The total concentrations and ecotoxicity of lead persists in soils, resulting in the redistribution and sometimes the chemical transformation of Pb into species that are more extractable and more toxic to people (Nwachukwu and

Pulford, 2008; Guo et al., 2006; Goyer, 1990). Because Pb is ubiquitous and non-biodegradable, has a long biological half-life for elimination from the body, and is harmful to human health at high doses, levels of exposure encountered by some population groups constitute a serious health problem (Odewande and Abimbola, 2008; Yu, 2001). Current research implicates Pb as a contributing etiologic factor in a number of common diseases affecting significant portions of exposed populations. At high levels of Pb exposure, humans experience damage to many organs and organ systems, most importantly the central nervous system, kidneys and blood. Damage may culminate in death at excessive levels.

At low levels, heme synthesis and biochemical processes are adversely affected; similarly psychological and neurobehavioral functions are impaired (Tong et al., 2000). Other medical conditions that may be caused by Pb exposure include cognitive and neurological deficits, hypertension, congenital malformations, immunotoxicity, and delayed growth and development. Small quantities of Pb affect cell proliferation and act as a cofactor in carcinogenesis. At the molecular level, Pb may compete with calcium to alter critical cell functions such as ion transport, energy production, and the function of heme-containing enzymes (Goyer, 1990).

Lead poisoning is the number one environmental disease among children in developing countries and in highly urbanized and/or industrialized communities in developed countries (Chaney and Ryan, 1994). The full impact of lead poisoning on the health of children and adults is becoming clearer to most countries, and many

governments have begun to take action. When ingested, inhaled, or absorbed through skin, lead is highly toxic to humans when it enters the bloodstream.

Lead concentrations in uncontaminated soils are generally low ( $<10$  to  $30 \mu\text{g g}^{-1}$ ) (ATSDR, 2007). Unfortunately, in areas of high vehicular traffic and in areas close to urban and industrial activities, atmospheric Pb deposition may be very high. Therefore, urban environments generally experience higher depositions of lead from vehicular emission than rural areas. Lead found in the residential soils is attributed to Pb-based paint that has weathered and flaked off of structures, industrial emissions, and leaded gasoline. The strong sorption characteristic of Pb to organic matter in soils usually causes lead to be retained in the top several centimeters where pH levels are equal to or greater than 5 and organic matter content is at least 5% (McLaughlin et al., 1999; U.S. Department of Health and Human Services, 1992; Yu, 2001).

Due to its persistence in the eco-system, Pb accumulates where it is deposited and can poison generations of children and adults through inadvertent ingestion or inhalation of wind transported soil particles, street dust, city dust, and house dust unless it is properly removed. About 20 to 50% of inhaled and 5 to 15% of ingested inorganic Pb is absorbed. In comparison, about 80% of inhaled organic Pb is absorbed. An adult absorbs about 10% of ingested Pb; in children the absorption value may be as high as 50% (U.S. Department of Health and Human Services, 1992; Yu 2001).

Absorbed Pb is carried throughout the body mostly by erythrocytes; the remainder in the plasma. Lead follows the distribution pathways of calcium in all body tissues. The

half-life of Pb in blood approximates that of the erythrocyte (~35 days), about 2 years in the brain, and decades in bone tissue.

Children are particularly vulnerable to the effects of Pb. Young children often place objects in their mouths, resulting in dust and soil being ingested and possibly an increased intake of lead. A greater proportion of ingested Pb is absorbed from the gastrointestinal tract of children than of adults. Also, a greater proportion of systemically circulating Pb gains access to the brain of children as compared with adults. Children 5 years of age or younger are particularly vulnerable to Pb, due to the fact that a developing nervous system is far more susceptible to the toxic effects of Pb than a mature brain (Lidsky and Schneider, 2003; Tong et al., 2000).

Lead as a neurotoxin can carry a harmful legacy. Women who live in Pb contaminated housing or who were themselves poisoned by Pb at a young age can pass that legacy on to their unborn progeny. Bone stored Pb may pose a threat to women of reproductive age long after their Pb exposure has ended. Some studies suggest that skeletal Pb stores are the dominant contributor to blood Pb during the prenatal period and during postpartum. Consequently, Pb levels in breast milk also increase with the Pb level in maternal blood, posing an additional risk to the neonate (Lidsky and Schneider, 2003).

Although all of lead's toxic effects cannot be bound together by a single mechanism, its ability to substitute for calcium is a factor common to many of its toxic actions. For example, lead's ability to pass through the blood-brain barrier is due in large part to its ability to substitute for ionic calcium (Lidsky and Schneider, 2003). In addition to the serious medical conditions previously mentioned, lead poisoning in

children has been shown to contribute to reductions in IQ and attention span, reading and learning disabilities, hyperactivity, impaired growth, behavioral problems, and hearing loss. The effects are long term and may be irreversible (Handbook of Chemistry and Physics, 2008).

### **C. Characteristics and Uses of Lead**

Lead occurs naturally in small quantities, in the environment. Lead is very soft, highly malleable, ductile, a poor conductor of electricity, and is resistant to corrosion. Because of its unique properties Pb has been used to produce a large number of materials and manufactured products including glass, paint, cable covering, plumbing pipes, building materials, print typeface, storage batteries, and coin currency (Handbook of Chemistry and Physics, 2008; Royer et al., 1992). The metal is very effective as a sound absorber, is used as a radiation shield around X-ray equipment and nuclear reactors, and is used to absorb vibration. Its alloys include solder and various anti-friction metals. Lead tetraethyl is an anti-knock compound that was used in gasoline in the United States until outlawed in early 1970s and phased out of use by the mid-1980s (Bridbord and Hanson, 2009; Yu, 2001). White lead ( $2\text{PbCO}_3 \cdot \text{Pb}(\text{OH})_2$ ), sublimed white lead ( $\text{PbSO}_4$ ), chrome yellow ( $\text{PbCrO}_4$ ), red lead ( $\text{Pb}_3\text{O}_4$ ), and other lead compounds are used extensively in paints. Lead is highly stable in compound form.

Before 1978, it was commonly used in the manufacture of high quality paint because it resisted cracking and peeling and retained color well (Yu, 2001; U.S. Department of Health and Human Services, 2007). More recently the use of lead in

paints has been drastically curtailed to eliminate or reduce health hazards; in 1978 the U.S. banned the use of interior lead based paint (Yu, 2001).

Lead oxide is used in producing fine “crystal glass” and “flint glass” with a high index of refraction for achromatic lenses. In the past, lead salts such as lead arsenate were used as insecticides (Peryea, 1998; Merry et al., 1983). The price for lead in 2008 was \$1.20 per pound; by 2010 the price has dropped to \$ 0.79 (Guberman, 2008; U.S. Geological Survey, 2010).

The high use-value (the concrete way in which a thing, an object, or a quantity of matter meets human needs) coupled with a relatively low price has resulted in the widespread use of lead; as such it has resulted in greatly elevated lead concentrations in certain environments. Consequently, in locations where Pb is mined, smelted, refined, and used to make a number of value-added products, the environmental Pb level has increased greatly (Marx, 1887; Yu, 2001).

#### **D. Why Pb is a Problem in Urban Environments**

Urban soils are important sinks and sources of pollutants. Additionally, urban soils are often more heterogeneous than naturally occurring soils due to the diverse land uses located in close proximity to each other. Urban soils are part of a complex environmental system that may pose a threat to urban water bodies and public health (Robertson et al., 2003). Different land uses specifically affect the mineralogical and chemical composition of urban soils causing various, often deleterious, impacts on the environment and consequently on the health of urban dwellers. Urban soils protect ground water reservoirs and provide nutrients for plants in gardens, parks, and

playgrounds. However, urban soils may also pollute the environment as contaminants are released via infiltration, wind or overland flow (Norra et al., 2008).

Early work on Pb contamination in urban soils focused on quantifying Pb concentrations and identifying the origins of the pollutant. Lead levels in the urban environments are, therefore, well documented (Farmer and Lyon, 1977; Duggan and Williams, 1977; Nafeotte and Day, 1998). A study in Glasgow, Scotland, found Pb concentrations in soils adjacent to streets to be significantly higher (150-2300 ppm, mean  $960 \pm 154$  ppm) than observed “natural” levels (78 ppm) (Farmer and Lyon, 1977). Duggan and Williams (1977) found that the average amount of Pb in food and drink for a 2-year old child in the United Kingdom was probably about 50 to 100  $\mu\text{g}$  per day as compared with an estimated 12 to 50  $\mu\text{g}$  for the average daily intake and absorption from the ingestion of street dust. Nageotte and Day (1998) compared urban Pb levels in Manchester, England, and Paris, France, before and after the 1985 European Community regulations that required the use of unleaded petrol. The study found that 1997 street dust lead levels in Manchester decreased to around 60% of the 1975 value. The study also found that samples collected under and around the Eiffel Tower (an area with no heavy traffic) had very high Pb levels (ranging from 333 to 4,282  $\mu\text{g/g}$ ). These relatively high concentrations of Pb were attributed to the use of lead based paint on the Eiffel Tower.

Studies of trace elements (Pb, Zn, Cu, Mn, and Cd) in urban soil dust focused on the availability of these elements and their biochemical reactivity (Al-Chalabi and Hawker, 1996; Stone and Marsalek, 1996; Fergusson and Kim, 1991; Biggins and Harrison, 1980). In their study of urban soils near Brisbane, Australia, Al-Chalabi and



Hawker (1996) found that the depth profile of urban roadside soils generally showed Pb accumulation within the uppermost 5 cm. They concluded that the accumulation was the result of turbulent conditions from the movement of heavy vehicles that caused atmospheric suspension and deposition of Pb containing dust particles. Lead containing dust particles have a relatively short residence time in the atmosphere, and deposit quickly in the near vicinity of the road. With respect to the depth of soil containing Pb, Al-Chalabi and Hawker concluded that the rapid decrease of organic carbon and calcium carbonate closely corresponded with rapid reduction in total Pb levels.

A similar study conducted in Sault Ste. Marie, Ontario, found that accumulations of toxic metals in sediment were exchangeable with carbonate, Fe/Mn oxides and organic matter (Stone and Marsalek, 1996). Lead, Zn, and Mn were shown to bind predominantly to carbonates. Fergusson and Kim (1991) found elevated levels of Pb in street and house dust; sources of street dust Pb were identified as road traffic, industrial emissions, weathered materials, and depositions due to specific intermittent episodes (precipitation events).

Sources that contribute to street dust were found also to contribute to house dust. Thus, Pb and cadmium originating from an automobile could contribute to house dust. Lead in house dust has been found to be highest near windows and doorways, suggesting that the Pb is entering from the outside (Fergusson and Kim, 1991). Biggins and Harrison (1980) observed the highest concentrations of Pb were often found in the smallest size fraction ( $<38\text{ }\mu\text{m}$ ); similarly, lower concentrations of metals were associated with larger particle size fractions. Using X-ray power diffraction, the researchers

identified four species of Pb in their samples:  $\text{PbSO}_4$ ,  $\text{Pb}^\circ$  (elemental Pb),  $\text{PbSO}_4 \cdot (\text{NH}_4)_2\text{SO}_4$ ,  $\text{Pb}_3\text{O}_4$ , and  $\text{PbO} \cdot \text{PbSO}_4$ . The most frequently observed compound was  $\text{PbSO}_4$ ; its presence is explained by the weathering of  $\text{PbSO}_4 \cdot (\text{NH}_4)_2\text{SO}_4$  deposited from the atmosphere.

Urban soils commonly possess the following characteristics: horizontal and vertical variability, degradation, an absence of soil structure leading to compaction, modified soil reaction, low organic matter content, restricted aeration and water drainage, high content of anthropogenic materials, and modified soil organism population and activities. These characteristics affect the ecological functions of urban soils (Tume et al., 2008, Imperato et al., 2003; Ljung et al., 2006). Tume et al. (2008) found that the concentrations of trace elements in urban topsoil samples (0-10 cm) in Talcahuano, Chile, were greater than those found in the subsurface horizon (10-20 cm). Lead concentrations in topsoil ranged from 8.0 to 129 ppm, with a mean of 35 ppm. The concentration in the subsurface soil ranged from 1.9 to 61.9 ppm, with a mean of 16.1 ppm (Tume et al., 2008). A Naples, Italy, study found the special distribution of Pb concentrations to be correlated with industrial activity and automotive traffic (Imperato et al., 2003). Highly contaminated soils were found in the proximity of motorways and streets with high traffic flows. Most of the Pb remained in insoluble and stable mineral form (low availability) most probably due to a pH-water value that was neutral or sub-alkaline (Imperato et al., 2003).

A study in Uppsala, Sweden, attempted to determine the natural or anthropogenic origin of metals in urban soils (Ljung et al., 2006). Regional parent material was found

to contribute to the relatively high contents of As, Al, Fe, Cr, Mn, Ni, and W in urban Uppsala. The contents of Cd, Cu, Zn, Pb and Hg were mainly derived from anthropogenic sources, with Uppsala heat and energy plants as the primary sources. Traffic was also found to affect the Pb content of the soil (Ljung et al., 2006). In summary, Pb in urban soils tends to increase with vehicular emissions, industrial residues, and the atmospheric deposition of dust and aerosols from metallurgical industrial activities (Tume et al., 2008).

### **E. The Distribution of Pb in DC Soils**

Environmental and health concerns have resulted in studies of heavy metals in the soils of many international urban areas: Madrid, Spain (Tume, et al., 2008; DeMiguel et al., 2007), Palermo and Torino, Italy (Manta et al., 2002; Madrid et al., 2008), Seville, Spain (Madrid et al., 2008), Baltimore, Maryland (Meilke et al., 1983), New Orleans, Louisiana (Meilke et al., 1998, 2008), Dakar, Senegal (Diouf et al., 2006), Sheffield, England (Rawlins, et al., 2005), and Ibadan, Nigeria (Odewande and Abimbola, 2008). In the District of Columbia, Elhelu et al. (1995) conducted a city wide study to assess the distribution and sources of lead in the city's soils. Soil samples were collected randomly from residential locations in all eight wards of the city (Figure 1).

Table 1.—Selected Percentile Values for Lead (ppm) from Each of the Eight Political Wards in Washington, D.C.

Percentile	Ward							
	1	2	3	4	5	6	7	8
Max.	4 905	4 520	815	4 575	5 056	1 720	3 740	6 015
95	3 075	2 390	330	1 223	792.5	1 497	790	1 330
90	2 630	1 870	241.1	937.5	745	1 150	588.2	937.5
75	1 145	975	105.7	294.9	380.4	427.9	274.9	307.9
50 (Med.)	444.2	471.4	53.7	198.9	221.9	260.4	144.4	129.7
25	228.7	344.8	25.1	95.5	101.3	125	70.3	68.45
10	134.6	236.1	13.5	63.05	41.65	82	48.7	51.8
5	51.75	80.3	10.2	36.95	21.9	19.7	39.4	28.4
Min.	36.4	48.3	10.2	32.7	12	13.8	36.2	22.2
n*	30	30	30	30	30	30	30	30

\*n = number of sites.

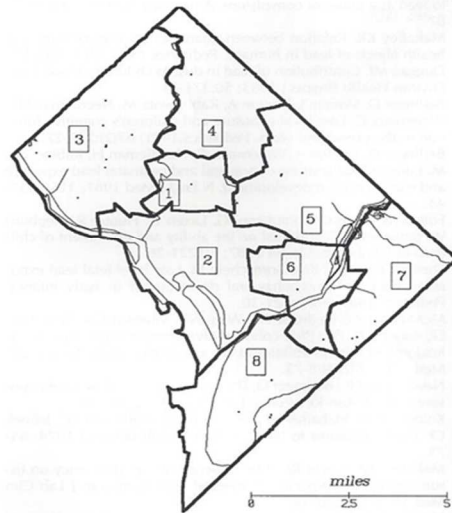


Fig. 1. Location (by ward) of soils in Washington, D.C., that contain lead in concentrations equal to or less than 50 ppm.

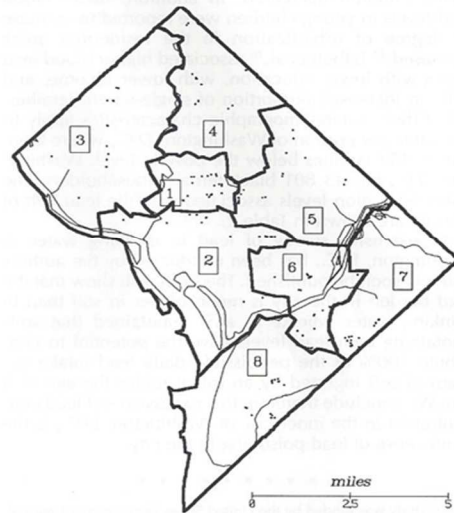


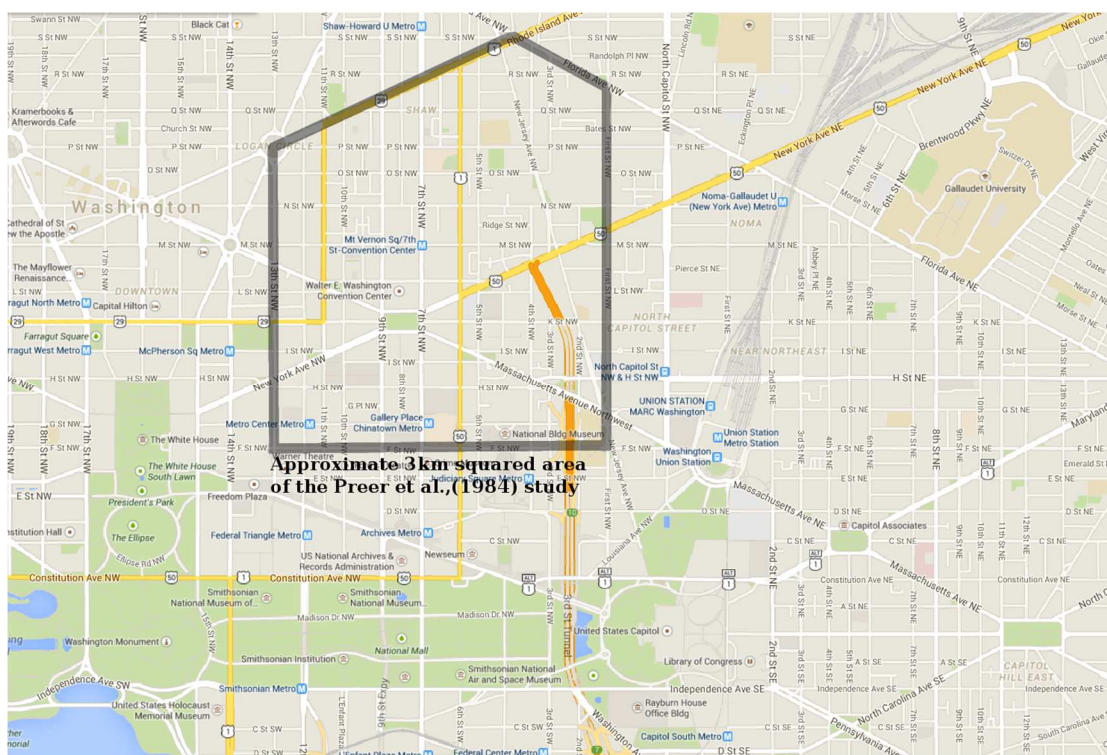
Fig. 2. Location (by ward) of soils in Washington, D.C., that contain lead concentrations greater than 50 ppm.

Figure 1: A table and maps of the location and distribution (by ward) of soil lead (Pb) in Washington, DC (Source: Elhelu et al., 1995, p. 167)

Given that urban children play in the yards of their homes, soil samples were collected from the front and back yards of small, unpaved row house dwellings. The surveyed homes were an average distance of 4.5 m from the road. The results of the Elhelu (1995) study found that the DC wards with the highest Pb concentrations were occupied by residents with the lowest education and income levels (Wards 1, 4, 5, 6, 7,

and 8). Wards 2 and 3 had the lowest Pb levels and are occupied by the city's most educated and wealthy residents. One anomaly of the socio-economic composition of ward residents is Ward 2, which contains some of the city's poorest and wealthiest residents. Ward 3 had a median soil Pb concentration of 53.7 ppm (part per million), with only 24% of the houses with painted exteriors. In comparison, Wards 1, 2, 4, 5, 6, 7, and 8 had median soil Pb concentrations of 444.2 ppm, 471.4 ppm, 198.9 ppm, 221.9 ppm, 260.4 ppm, 144.4 ppm, and 129.7 ppm, respectively; with approximately 74% of the houses in these wards having exterior paint. Natural, uncontaminated soil has a Pb concentration of 10-50 ppm. Leaded gasoline has not been sold in the District of Columbia since the mid 1980's, thus suggesting that exterior house paint was the primary source of DC soil lead in residential areas (Bridbord and Hanson, 2009; Needleman and Bellinger, 1991; Elhelu et al., 1995).

Preer et al. (1984), in their studies of heavy metal content of vegetables grown in DC urban gardens, found downtown Washington, DC, soils to be considerably higher in Pb and other heavy metals, than that obtained in a previously reported city-wide survey (Preer et al., 1980; Figure 2). The 1980 study found the mean city-wide Pb level to be 200 ppm, as compared with the 1984 results that showed the downtown mean Pb level to be 680 ppm in an approximately 3 km<sup>2</sup> area that lies entirely in the NW quadrant bounded by Florida Avenue on the north, F Street on the south, 1<sup>st</sup> Street on the east, and 12<sup>th</sup> Street on the west, encompassing parts of city wards 2, 5 and 6 (Preer et al., 1984).



**Figure 2: The approximate area of the locations of urban gardens with a mean lead (Pb) soil level of  $680 \text{ mg kg}^{-1}$  in downtown Washington, DC, (Preer et al., 1980; 1984).**

The Pb levels of leafy vegetables grown in gardens in the downtown area were significantly higher than those obtained in the city-wide study (Preer et al., 1980; 1984).

## **F. Past and Present Methodologies to Remediate Soil Pb**

Remediation of soils contaminated with Pb has been achieved using a variety of strategies: capping, excavation and landfill burial, solidification, and stabilization.

However, these technologies render the land useless for plant growth, as they destroy useful biological communities including nitrogen fixing bacteria, mycorrhiza, fungi, and fauna. In addition, these types of site remediation strategies range in cost (1997 US\$)

from \$15 per m<sup>2</sup> for a 60 cm thick soil cap to \$750 per m<sup>2</sup> for excavating to 60 cm, stabilizing, and off-site disposal (Berti and Cunningham, 1997).

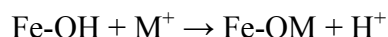
#### **G. Chemical Compounds known to Immobilize or Stabilize Pb**

Heavy metal “solubility and availability in land applied treatments (residuals) is governed by fundamental chemical reactions between metal constituents, soil, and treatment components. Organic matter, phosphate, iron, aluminum, and manganese oxides ... are known to immobilize lead in soil through different chemical mechanisms” (Basta et al., 2005). Heavy metal cations (such as ionic Pb) can sorb to soil organic matter (SOM) and other forms of humified natural organic matter (NOM). Strong adsorption by natural organic matter in treatment residuals can reduce the solubility of several metals in soils. Heavy metals form bonds with specific functional groups of soil organic matter, including amine, carboxyl, phenolic, and thiol-SH functional groups and/or can complex and/or exchange metals. Borderline acids like Pb<sup>2+</sup> will form complexes with a weak or strong base. Also, the electronic properties of Pb result in a very strong affinity for soil organic matter and formation of strong inner-sphere metal surface complexes. Sorption of trace elements by soil organic matter or humified natural organic matter increases with pH because metals preferentially bind with ionized functional groups formed with increasing pH (Nwachukwu and Pulkford, 2008; Basta et al., 2005).

Lead cations form sparingly soluble precipitates with anionic compounds as phosphate. Heavy metal precipitation is highly pH dependent and increases with pH for many heavy metal cations (Basta et al., 2005). The lead cation reacts with soluble

phosphate ( $\text{PO}_4^{3-}$ ) to form various pyromorphites [ $\text{Pb}_5(\text{PO}_4)_3\text{X}$ , where  $\text{X}=\text{OH}^-$ ,  $\text{Br}^-$ ,  $\text{Cl}^-$ , or  $\text{F}^-$ ] that are insoluble and thus are very non-extractable (Pierzynski and Gehl, 2004; Shevade et al., 2001) .

Sorption by metal oxides is a major mechanism for removal of heavy metal cations. Sorption regulates partitioning of heavy metals between solution and solid phases in soils. Iron and manganese oxide soil minerals are important sinks for heavy metals in soils. Heavy metal sorption by the oxide surface is a highly pH dependent process described by the following cation adsorption chemical reaction:



In this case, Fe-OH is an iron oxide surface, and M is a heavy metal cation (Basta et al., 2005).

Usman et al. (2005) found that the addition of Fe-oxides (goethite and hematite) significantly reduced the availability of Pb and other heavy metals to wheat grown on sewage sludge contaminated soil. Sauve et al. (2000) observed that ferrihydrite (a synthetic Fe oxide) was efficient in lowering aqueous Pb concentrations.

It has been demonstrated that Pb concentration is reduced with the use of Mn oxides (Hettiarachchi et al., 2000; Hettiarachchi et al., 2002; Hettiarachchi et al., 2003). Sonmez and Peirzynski (2005) found the use of synthetic Mn oxide for reducing soil Pb bioaccessibility was promising. Beak et al. (2008) observed a slow bioaccessibility when Pb was sorbed to Mn oxide and presented little risk associated with incidental ingestion of soil.



## **H. Agricultural and Industrial By-products Known to Contain Pb Fixing Compounds**

Traditional physical-chemical extraction techniques generally entail degradation of soil structure, soil ecological integrity, and high remediation cost. Whereas, in situ fixation/stabilization techniques may improve biological properties, do not generate waste by-products, and are less expensive than traditional remediation means. The use of agricultural and industrial by-products known to contain quantities of Pb fixing compounds is based on the use of amendments to accelerate those processes (sorption, precipitation, and complexation reactions) that take place naturally in soils to reduce heavy metal mobility and bioavailability (Madejon et al., 2009; Hashimoto et al., 2009). Leaf compost, poultry litter, drinking water treatment residual, and steel slag are a few agricultural and industrial by-products known to contain chemical compounds that can potentially immobilize or stabilize Pb.

### **1. Leaf Compost**

Some organic matter may have the natural ability to bind metals, thus reducing bioavailability. Additionally, organic matter may improve soil characteristics conducive to plant growth. The use of a waste organic material serves two functions: soil remediation and waste disposal. Some forms of waste organic matter, such as manure or biosolids, may increase metal mobility due to the high dissolved organic matter (DOM) content that can complex with metals and facilitate movement through the soil. The process of composting stabilizes organic waste and reduces DOM content and the potential for metal leaching (Tandy et al., 2009).

A considerable amount of research has been conducted on the addition of compost (as a source of organic matter) to contaminated soils to immobilize metals. Several studies in particular have demonstrated that compost derived from garden waste (compost composed of mown grass, pruned trees and other plant material from gardens) is effective at immobilizing Pb in contaminated soils (Nwachukwu and Pulkford, 2009; Nwachukwu and Pulkford, 2008; von Herwijnen et al., 2007). Clemente et al. (2006) observed organic amendments favored Zn, Pb, and Fe fixation in contaminated soils. Pichtel and Bradway (2008) found that soil Pb and Zn tended to shift toward less extractable forms after treatment with compost peat. Tandy et al. (2009) observed that both co-composting heavy metal contaminated soil with organic waste and conventional incorporation of mature compost into contaminated soil, reduced metal accumulation in plants and promoted plant growth.

## **2. Poultry Litter Ash**

Phosphorus containing amendments have been used for immobilizing soil Pb; the mechanism is based on a rapid kinetic formation of geochemically stable Pb-phosphates. Pb-phosphates, such as chloromorphite  $[\text{Pb}_4(\text{PO}_4)_3]$ , are at least 44 orders of magnitude less soluble than naturally occurring Pb minerals such as cerussite ( $\text{PbCO}_3$ ). Because of their potentially high immobilization effect, phosphorus amendments like hydroxyapatite  $[\text{Ca}_5(\text{PO}_4)_3\text{OH}]$  and rock phosphate [primarily  $\text{Ca}_5(\text{PO}_4)_3\text{F}$ ] have been studied for reducing metal extractability.

Although effective, these apatite materials are expensive and thus impractical for Pb immobilization in large areas such as contaminated soil in urban gardens. To reduce

costs, hydroxyapatites synthesized from agricultural by-products like poultry litter have been investigated (Hashimoto et al., 2009; Ihnat and Fernandes, 1996). Poultry litter ash, the by-product of burning poultry litter, contains high concentrations of phosphorus (Codling, 2006). Faridullah et al. (2009) found that both chicken and duck litter ash treated soils exhibited lower concentrations of water soluble and exchangeable metals as compared to other fractions. In their study to evaluate the effectiveness of incinerated poultry waste for remediating shooting range soils, Hashimoto et al. (2008) found that the application of poultry waste reduced water extractable Pb by about 43% as compared with a control. The sequential extraction analysis revealed that the Pb-fraction of poultry waste amended soils was shifted to less soluble phases than those of the control soil (Hashimoto et al., 2008).

### **3. Drinking Water Treatment Residual**

Drinking water treatment residual (DWTR) is a by-product of drinking water treatment. It contains precipitated hydroxides of the treatment chemicals that are added to coagulate and flocculate dissolved and suspended material in the raw water. The chemicals typically include  $\text{Al}_2(\text{SO}_4)_3 \cdot n\text{H}_2\text{O}$ ,  $\text{FeCl}_3$ ,  $\text{Fe}_2(\text{SO}_4)_3$ , long-chain organic polymers (LCP), activated charcoal, activated silica and lime (Titshall and Hughes, 2005). Brown et al. (2007) observed that treatments of Al-DWTR and Fe-DWTR (aluminum and iron based drinking water treatment residuals, respectively) were able to reduce extractable Pb in contaminated soils and mine tailings. In soils, combined treatments of compost and Al-DWTR reduced bioaccessible Pb by 43% and combined treatments of biosolids and Fe-DWTR reduced extractable Pb by 58%. For mine tailings,

Brown et al. (2007) found that the addition of Fe-DWTR to biosolids decreased extractable Pb by 68%. In an earlier study, Brown et al. (2005) observed one Al-based DWTR to decrease extractable Pb by 9%.

#### **4. Steel Slag**

Steel slag (SS) is a nonmetallic by-product of steel production with a complex chemical structure. There are two types of steel slag produced by the North American steel industry: Basic Oxygen Furnace (BOF) steel slag and Electric Arc Furnace (EAF) steel slag. Both types primarily comprise fluxing agents (mainly lime), used during the steelmaking process, and the molten impurities of steel. The composition is primarily silica and alumina from the original iron ore (with calcium and magnesium oxides from the added flux) with substantial amounts of iron and manganese. Traces of alumina, magnesia, sulfur, and titanium may also be found (Proctor et al., 2000; Khan et al., 2002).

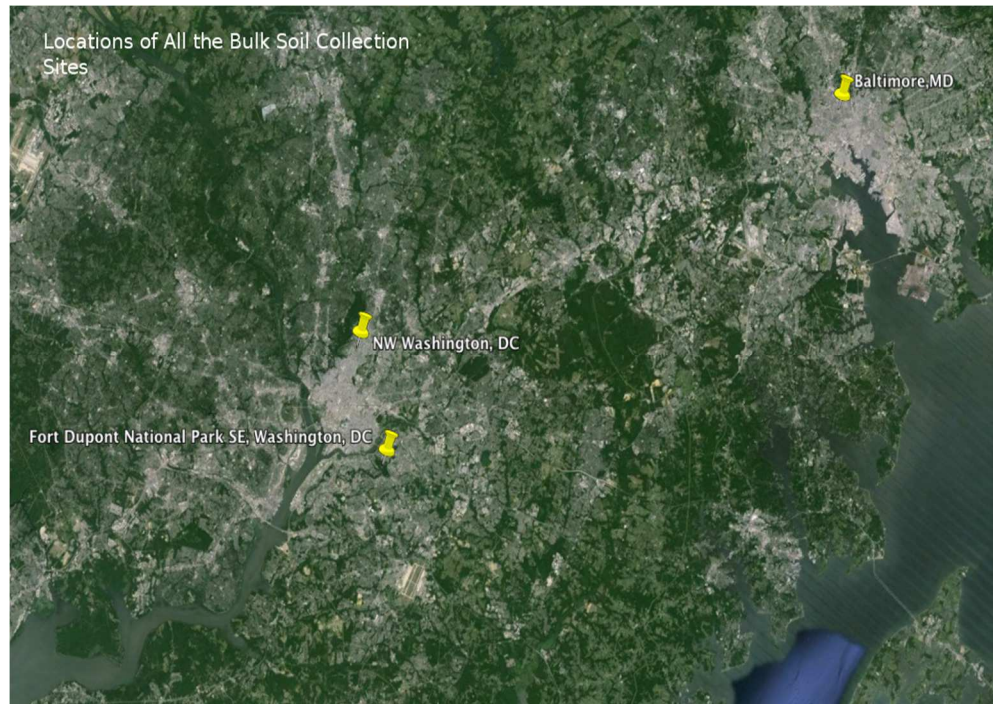
Several research teams have demonstrated the effectiveness and efficacy of steel slag to render Pb non-extractable in different types of pollution media. Oh et al. (2007) observed how two different types of waste steel scrap “removed” Pb and other heavy metals from a mimic landfill leachate solution through physiochemical adsorption. One type was converter slag. The other type of slag was acid-washed with 2 N HCl (AD) to produce both zero-valent iron and oxide. Twenty-four hours after treatment AD slag and converter slag “removed” Pb from the mimic landfill leachate solution by 77% and 99%, respectively (Oh et al., 2007). A blast furnace slag achieved a Pb immobilization efficiency average of 99% after a 24 hour leaching test (Yunsheng et al., 2007). In a study to evaluate the remediation potential of Pb and Cd polluted soils, Aboulroos et al.

(2006) observed that slag demonstrated one of the highest immobilization efficiencies for exchangeable Pb (46% of the initial value).

## **CHAPTER THREE – MATERIALS AND METHODS**

### **A. Experiment Location**

The study was conducted in the greenhouse and analytical laboratories of the United States Department of Agriculture - Beltsville Agricultural Research Center - Environmental Management and By-product Utilization Laboratory (USDA-BARC-EMBUL) in Beltsville, MD. The materials needed to conduct the study, including all laboratory reagents and equipment were supplied by USDA-BARC-EMBUL. Bulk soil samples were collected from two different locations in Washington, DC, and one location in Baltimore City, MD (Figure 3).



**Figure 3: A regional Washington, DC, – Baltimore, MD, Metropolitan satellite map of all bulk soil collection locations.**

## **B. Soil Sample and Bulk Soil Collection**

Soil samples for initial soil Pb content analysis were collected with a stainless steel soil auger with a 15 inch long,  $\frac{3}{4}$  inch diameter soil core bit. At each sample site, soil was collected at random points within an approximate 5 meter square area. For any particular sample site, three soil cores from a vertical depth of 15 cm were placed in pre-labeled and dated sterile polyethylene soil sampling bags for transport to the laboratory for digestion and analysis. Bulk soil used throughout the conduct of the study was collected with a stainless steel spade shovel to a vertical depth of 15 cm and placed in

labeled and dated 5 gallon size washed and air dried plastic buckets for transport to the laboratory and temporary storage.

### **C. Experimental soils, soil preparation and evaluation**

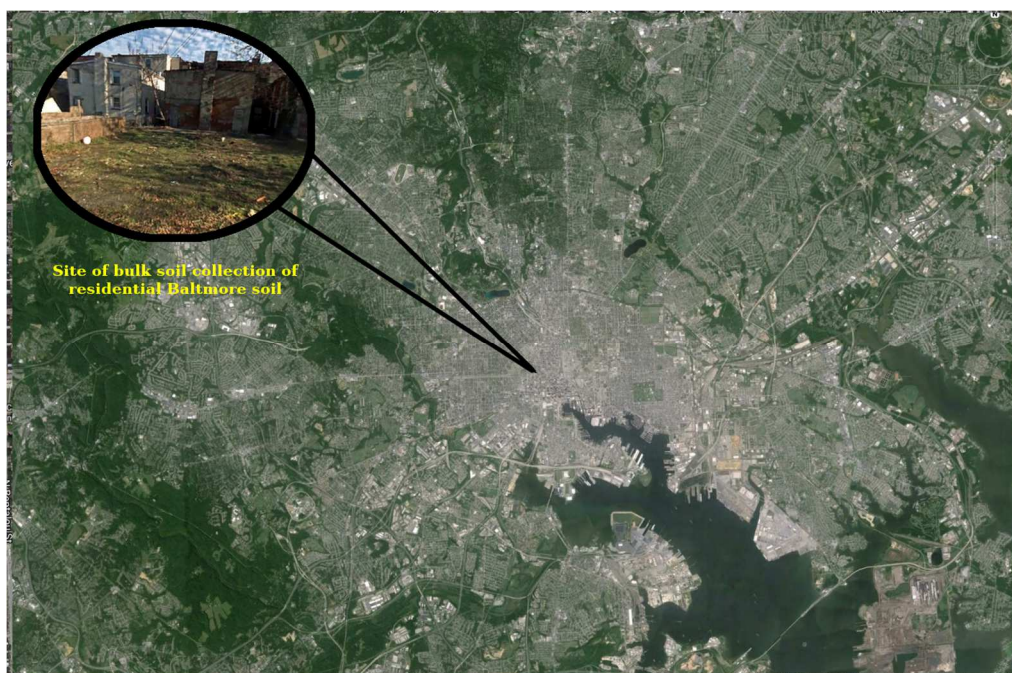
Experimental soils were sampled and analyzed for total lead content by Atomic Absorption Spectroscopy (Varian SpectrAA-400 Atomic Absorption Spectrometer).

When sufficiently contaminated soils were discovered ( $> 1000 \text{ mg kg}^{-1}$ ) they were collected in bulk from three different locations: from a residence in the ward 4 section of Northwest Washington, DC; from a now abandoned residence from the western part of inner-city Baltimore, MD; and from Fort DuPont National Park in Northeast Washington, DC, which was used as a control soil.





**Figure 4: Location of the residential Washington, DC, NW bulk soil collection site of lead (Pb) contaminated soil.**



**Figure 5: Location of the Baltimore City, MD, bulk soil collection site of lead (Pb) contaminated soil.**



**Figure 6: The approximate location of bulk soil collection within Fort DuPont National Park, Washington, DC, SE.**

According to U.S. Department of Agriculture soil survey manuals of Washington, DC, and Baltimore City, MD, the soils were classified as shown in the following table:

**Table 1: Soil Sample Descriptions (SCS, 1976; NRCS, 1998)**

Soil Sample Description	Soil Name	Taxonomic Class
<b>Washington DC residential soil</b>	Manor Loam	coarse-loamy, micaceous, mesic Typic Dystrochrepts
<b>Baltimore City, MD residential soil</b>	Urban land 0 to 15 percent slope	undefined
<b>Study control soil</b>	Christiana Silt Loam	clayey, kaolinitic, mesic Typic Paleudults

Soil samples were prepared for analysis by air drying, then crushed using a stainless steel rolling pin and screened using a 2 mm sieve. A 1N HNO<sub>3</sub> extraction was

carried out in order to determine total Pb levels in collected soil samples. After desired lead contaminated soils were identified, bulk soils for the experiment were collected, screened using a 4 mm screen, mixed, and stored moist in 5 gallon plastic buckets until soil treatment and mixing commenced.

#### **D. Soil Treatments and Pot Preparation**

Low and high rates of application for each treatment were defined as shown in Table 2. The leaf compost (LC), poultry litter ash (PLA), drinking water treatment residual (DWTR) and Steel Slag (SS) treatment percentage rates were determined based on the work of Nwachukwu and Pulford (2009), Hashimoto et al. (2009), Brown et al. (2005), and Sang-Hwan et al. (2009), respectively.

**Table 2: Treatment Applications**

<b>Treatment Descriptions</b>	<b>Control (% dry weight)</b>	<b>Low Rate of Application (% dry weight)</b>	<b>High Rate of Application (% dry weight)</b>
Leaf Compost (LC)	0	10	20
Poultry Litter Ash (PLA)	0	6	12
Drinking Water Treatment Residual (DWTR)	0	2.5	5
Steel Slag (SS)	0	2.5	5

After calculating and measuring the proper percentage of each by-product treatment, the by-product additives and the bulk soils were mixed in a soil mixer to

achieve uniformity. The specifics regarding the treatment dry weight calculations are shown in Table 3.

**Table 3: Treatment calculations**

<b>Description</b>	<b>Quantity</b>
<b>Plastic, 3 kg planting pots</b>	432/2=216 (Cool and Warm Seasonal Crops, 2 Planting seasons)
<b>Soil<sup>1</sup></b>	(3 kg/pot x 144 pots/soil type)/(2 plantings)= 216 kg/soil type/planting
<b>Treatments<sup>2</sup></b>	(17 kg LC/crop x 4 crops)/(2 plantings)= 34 kg LC/planting (10 kg PLA/crop x 4 crops)/(2 plantings)= 20 kg CL/planting (5 kg SS/crop x 4 crops)/(2 plantings)= 10 kg SS/planting (5 kg DWTR/crop x 4 crops)/(2 plantings)= 10 kg DWTR/planting
<p><sup>1</sup>The soil and amendment mixtures were used for two plantings.  <sup>2</sup>LC rates were calculated based on applications of 10% and 20% by dry weight. PL rates were calculated based on applications of 6% and 12% by dry weight. SS and DWTR rates were calculated based on applications of 2.5% and 5% by dry weight.</p>	

The mixtures were placed in 5 gallon buckets for temporary storage. The order of soil-by-product mixing was from control soil to the soils with higher levels of Pb contamination as shown in Table 4.

**Table 4: Soil mixing and contamination levels**

<b>Soil Description and Mixing Order</b>	<b>Mixing Order</b>	<b>Lead contamination (mg kg<sup>-1</sup>)</b>
Ft. DuPont (control)	First	37
Residential Washington, DC,	Second	919
Residential Baltimore, MD,	Last	1,528

Between soils, the soil mixer was washed to prevent cross contamination. The above listed by-product additives were applied at three different rates: control rate, low rate, and high rate. The by-product treated bulk soil was then transferred into 3 kg plastic pots with the bottoms lined with cheese cloth and atop plastic pans to capture and return pot drained water.

After being labeled and filled to capacity with test soils and by-product treatments, the pots were organized into three randomized blocks on bench tops in the BARC greenhouse. A 3-week soil incubation period was initiated by filling all pots to full saturation with deionized (DI) water and subsequently allowing them to dry completely and be refilled to saturation (to replicate natural precipitation event cycles). At the end of the 3-week incubation period, all pot soils were sampled and collected in small laboratory plastic bags for analysis.

## **E. pH Determination**

The pH of each soil and by-product mixture was determined in a ratio of 1:1 sample to DI water slurry using a glass electrode of a Mettler Toledo MP 220 pH meter after letting samples sit for 1 hour (Codling, 2008).

#### **F. Nutrient Addition**

Soil pH was adjusted with calcium and magnesium carbonate to near a pH of 6.5. Soils were fertilized at the rate of 200 kg ha<sup>-1</sup> of P as calcium phosphate Ca(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub> and potassium phosphate (KH<sub>2</sub>PO<sub>4</sub>), 100 kg ha<sup>-1</sup> nitrogen as ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>), 60 kg ha<sup>-1</sup> Mg as magnesium sulfate (MgSO<sub>4</sub>) and magnesium carbonate (MgCO<sub>3</sub>), and 232 kg ha<sup>-1</sup> K as KH<sub>2</sub>PO<sub>4</sub>. The soil and fertilizer were mixed and incubated moist for four weeks.

#### **G. Electrical Conductivity Analysis**

The electrical conductivities (EC) of all original soils, by-products, and amended soils were determined in a ratio of 1:2 sample to DI water solution using an Orion model 160 conductivity meter after stirring and letting sit for 1 hour (Codling, 2008).

#### **H. Digestion for Total Soil Lead (Hot-Plate Aqua Regia Extraction)**

All soils, by-products, and amended soils were analyzed for the total lead concentration using the Hot Plate Aqua Regia Digestion procedure and atomic adsorption spectroscopy (AAS) (Chen and Ma, 2001). For the Hot Plate Aqua Regia Digestion method, the procedure was performed in 250 ml glass beakers covered with watch

glasses. A well-mixed sample of 5.0 g was digested in 20 ml of Aqua Regia (5.0 g HNO<sub>3</sub> and 15.0 g HCl) on a hotplate slow heating for 2 hours at 98°F. After evaporation to near dry glistening, the sample was diluted with 20 ml of 3N HCl and set back on the hotplate for 2 hours at 98°F. After 2 hours the beaker contents were double filtered through Whatman no. 42 paper into a 50 ml volumetric flask. After filtering, the flasks were brought to volume with 0.1N HCl. Blank solutions were also prepared. The filtrate was analyzed with flame atomic adsorption spectrometry (AAS) (Brown et al., 2003; Chen and Ma, 2001; Ehi-Eromosele et al., 2012).

#### **I. Extraction Protocols for Water Extractable and Mehlich III Extractable Soil Lead Determination**

Water Extractable soil Pb determination was accomplished using the modified method of Codling et al. (2000). Air-dried, pulverized soil was passed through a 2.0 mm sieve and weighed to  $2.5 \pm 0.05$  g in capped urine specimen cups on a Mettler PM 2000 analytical balance, 25 ml of DI water was added to the soil and shaken on a New Brunswick Scientific shaker at 200 oscillations per minute for 1 hour. The supernatant was filtered using a Whatman no. 42 filter and stored in 25 ml plastic vials to await analysis (Codling, 2007; Meers et al., 2006).

Mehlich III extractable soil Pb was determined through the following method:  $2.5 \pm 0.05$  g of air-dried soil was pulverized to pass through a 2.0 mm mesh sieve and weighed on a Mettler PM 2000 analytical balance. The sieved and weighed soil was then shaken on a New Brunswick Scientific Shaker with 25 ml of Mehlich III Extracting solution



[composed of 0.2M CH<sub>3</sub>COOH, 0.25M NH<sub>4</sub>NO<sub>3</sub>, 0.015M NH<sub>4</sub>F, 0.013M HNO<sub>3</sub>, and 0.001M ethylene diamine tetraacetic acid (EDTA)] in urine specimen jars for 1 hour at room temperature. The supernatant was filtered using a Whatman no. 42 filter and stored in 25 ml plastic vials to await analysis (Wharton et al., 2012; Hamel et al., 2003). Solutions from both the Water Extractable Lead procedure and Mehlich III Extraction procedure were analyzed with inductively coupled plasma-mass spectrometer (ICP-MS). For quality control, all samples were extracted in triplicates with blanks and NIST standard soils. The National Institute of Standards and Technology (NIST) provides standard reference soils intended for use in method development, method validation, and routine quality assurance in the analysis of element content of soils.

## **J. Crop Selection, Preparation, and Evaluation**

The crop species used were the following: Collards—Vates non-heading, Carrots—Danvers half long, Lettuce—Parris Island romaine, and Beets—Detroit dark red. Seven pre-wetted seeds were planted in each pot. These crops represent typical garden varieties for Washington, DC, and Baltimore, MD, urban gardeners. Both leafy and rooty crops were selected, as were crops that are representative of warm and cool seasons.

After successful germination and sprouting, crops were weeded down to three crop plants per pot, favoring the most robust plants. Crops were watered throughout a 90 day growth period with Deionized water. Microwave extraction and ICP-MS analysis methods were followed by those modified by Fernanda et al. (2013). A total of 864 seeds (8 seeds per treatment x 108 treatments per seed type) were needed for each type of crop.

## **K. Trials**

A complete randomized block design with three replicates was selected for this study. This type of statistical design was originally proposed in agricultural studies where fields were divided into blocks to account for the introduction of error that may be accounted for by the specific location in the field. In this case, the randomization accounts for errors that may be introduced by specific location in the greenhouse. In each block, a treatment is applied once.

The trials were conducted in the Beltsville Agriculture Research Center Greenhouse Complex on three benches; each bench represents a block.



**Figure 7: Photo of the complete randomized block design set-up of the first cropping of collard and carrot crops in the BARC Greenhouse Complex.**



**Figure 8: Photo of the complete randomized block design set-up of the second cropping of lettuce and beet crops in the BARC Greenhouse Complex.**

One treatment represents a combination of soil, by-product additive, and crop that is planted in a single pot. The experiment consisted of three repetitions for each soil (3), soil by-product additive (4), crop (4), and rate (3) combination depicted in the table below (Table 5). There were 144 combinations and 432 individual trials. The 432 trials were conducted in two periods (216 trials each) to represent a cool growing season and a warm growing season. Between both crop harvests, soils in each pot were re-mixed and re-potted.

**Table 5: Randomized block design**

<b>Soil by-product additives</b>	1. Leaf Compost (LC) 2. Poultry Litter Ash (PLA) 3. Drinking Water Treatment Residual (DWTR) 4. Steel Slag (SS)	
<b>Soils</b>	1. Washington DC, Residential 2. Baltimore, MD, Residential 3. Fort Dupont, DC National Park, Control	
<b>Crops</b>	<b>Leafy</b>	1. Collards (Cool Season) 2. Lettuce (Warm Season)
	<b>Rooty</b>	3. Beets (Cool Season) 4. Carrots (Warm Season)
<b>Rates</b>	1. Control 2. Low 3. High	

## **L. Biomass Determination**

Leafy plants were harvested for biomass determination by cutting the shoots and/or stems above the lowest growing leaf to avoid splashing effects and contamination; washing with the surfactant lauryl sulfate (95%), in deionized water, then rinsing three times in deionized water; placing in plastic bags, and temporarily storing them in a freezer. Rooty plants were harvested, scrubbed with a brush, washed with 95% lauryl sulfate in deionized water, rinsed three times with deionized water, placed in labeled plastic bags, and temporarily stored in a freezer. The dry matter yield of all harvested crops was determined by measuring all crops to a constant weight after freeze drying (a plant tissue drying process where frozen plant samples are placed in a vacuum chamber

and a controlled supply of heat is provided to allow ice, within frozen plant tissue, to sublime rather than melt) (Faithfull, 2002).

#### **M. National and International Permissible Lead Content Limits**

To determine whether by-product additive treatments were successful at “safely” reducing extractable Pb levels in the soil medium and in crop tissues, results were compared to both United States Environmental Protection Agency (U.S EPA) and United Nations Food and Agricultural Organization/World Health Organization (UN FAO/WHO) permissible Pb limits for soils (EPA: 400 mg Kg<sup>-1</sup>, for play and garden areas; FAO/WHO: 100 mg Kg<sup>-1</sup>, for garden areas), leafy and tuber (rooty) vegetables (EPA: no standards; FAO/WHO: 0.3 and 0.1 mg kg<sup>-1</sup>, respectively) (U.S. EPA, 2001; FAO/WHO, 2001).

Methods recommended by the US EPA (EPA Method: SW 846-3050) and used in this study (Aqua Regia Extraction) to determine total soil Pb levels are comparable, both methods are designed to dissolve metals (i.e. Pb) through hot acid digestion (U.S. EPA, 1993; Brown et al., 2003; Chen and Ma, 2001). Similarly, methods recommended by the US EPA (EPA Method: SW-846-3040B) and used in this study (Microwave extraction) to determine total Pb content in plant tissue are comparable, both methods are designed to dissolve metals, and plant tissue in which they reside, though hot acid digestion (U.S. EPA, 1990; Fernanda et al., 2013). However, the comparison of total Pb and Mehlich III Pb (M3-Pb) levels should be limited to using Mehlich III extraction as a screening tool to estimate the presence of Pb on a given site. Several researchers have observed that M3-Pb extraction is a reliable predictor of presence of the total amount of

Pb in a given soil, and that M3-Pb extracted Pb is a bio-extractable fraction of the total amount of soil Pb. In their study comparing screening tests for soil Pb, Wharton et al. (2012) observed that the extraction efficiency of M3-Pb was 54% and 52% for urban and agricultural soils of the total amount of Pb in those soils, respectively. Mincaa et al. (2013) observed that M3-Pb is strongly correlated with total extractable Pb for a wide range of soil Pb concentrations. Another very practical reason for the employment of the Mehlich III soil test for this study is that it is one of the most common, inexpensive, and relatively quick methods used by soil testing laboratories in the United States (Mincaa et al. 2013).

## **N. Statistical Analysis**

Analysis of variance (ANOVA) was performed with Statistical Analysis Software (SAS). Mean comparisons were carried out using Duncan's multiple range test and least significant difference (LSD) at a 95% confidence level ( $p < 0.05$ ).

## **CHAPTER FOUR – RESULTS AND OBSERVATIONS**

### **A. Results**

#### **1. Chemical Analysis of Soils and By-Products**

The chemical analysis of the soils and by-product amendments known to sequester lead yielded the results shown in Table 6. The pH values of poultry litter ash (PLA) and Steel slag (SS) are alkaline to highly alkaline, respectively. The poultry litter ash (PLA) electric conductivity (EC) value is over an order of magnitude higher than the next highest EC value among the other by-product amendments. Also, the (PLA) phosphorus (P) level (indicator of phosphate content) was the highest amongst all by-products by at least one order of magnitude. Drinking water treatment residual (DWTR) iron (Fe) levels (indicator of iron oxide content) was the highest of all by-products by well over an order of magnitude. Of the possible elements and compounds known to sequester lead in steel slag, none was found in great quantities except for Al (indicator of alumina). It should be noted that the Mehlich III Lead levels for the Fort Dupont soil were “below detectable limits” for the controls. For this reason, the Fort Dupont results are not discussed further.



**Table 6: Chemical analysis of soils and by-products: Note the relatively high values of the bolded chemical and elemental parameters along with the bolded Pb content values of Pb contaminated soils and by-product treatment additives used in this study.**

Parameters	DC Soil	Baltimore Soil	LC	WTR	SS	PLA
pH	7.2	6.0	7.8	7.0	<b>12.1</b>	<b>10.7</b>
EC (mS/cm)	0.2	0.7	0.4	2.3	5.6	<b>59.6</b>
Cu (mg/kg)	54.0	212.0	29.0	310.0	52.0	849.0
Pb (mg/kg)	<b>919.0</b>	<b>1528.0</b>	<b>87.0</b>	<b>220.0</b>	<b>21.0</b>	<b>&lt;0.1</b>
Zn (mg/kg)	952.0	697.0	64.0	1889.0	42.0	1064.0
Al (g/kg)	13.1	11.3	6.1	13.4	<b>62.1</b>	13.0
Ca (g/kg)	5.0	7.8	15.5	124.0	5.5	124.0
Fe (g/kg)	26.2	13.3	8.1	<b>185.0</b>	6.0	8.8
K(g/kg)	1.2	0.6	0.9	0.2	0.3	80.9
Mg (g/kg)	1.9	1.6	1.8	31.8	0.9	6.3
Mn (g/kg)	0.4	0.3	0.3	22.2	6.4	2.4
P (g/kg)	0.9	2.0	0.9	0.6	1.7	<b>14.7</b>

## 2. Effect of by-product treatments on Mehlich III extractable lead

The results of the by-product treatments on the Mehlich III extractable lead content in soils are shown in Table 7. Those means shown to differ statistically from the control are depicted in green for a decrease and red for an increase. All other means do not differ statistically significantly from the control mean.

**Table 7: Mehlick III Lead for treated and control soils**

Treatment	DC Soils			Baltimore Soils		
	Mehlick III Lead Mean (mg kg <sup>-1</sup> )	Mehlick III Lead Std Dev (mg kg <sup>-1</sup> )	Mehlick III Lead Change Relative to the Control (%)	Mehlick III Lead Mean (mg kg <sup>-1</sup> )	Mehlick III Lead Std Dev (mg kg <sup>-1</sup> )	Mehlick III Lead Change Relative to the Control (%)
<b>Control</b>	478.2	19.0		346.8	2.7	
<b>LC-L</b>	369.4	1.1	-22.8	332.4	13.9	-4.17
<b>LC-H</b>	339.4	44.3	-29.01	303.2	15.7	-12.59
<b>PLA-L</b>	324.5	2.6	-32.13	260.1	7.3	-25.01
<b>PLA-H</b>	283.8	0.1	-40.64	221.2	8.6	-36.22
<b>DWTR-L</b>	460.7	23.0	-3.66	373.2	8.4	7.59
<b>DWTR-H</b>	436.2	14.5	-8.79	379.3	54.1	9.37
<b>SS-L</b>	399.2	8.0	-16.51	311.0	13.1	-10.34
<b>SS-H</b>	358.0	3.2	-25.12	283.1	7.0	-18.37
<b>Key</b>		Statistically significant reduction in Mehlick III-lead				
		Statistically significant increase in Mehlick III-lead				
	<b>LC-L</b>	Leaf compost at low rate treatment				
	<b>LC-H</b>	Leaf compost at high rate treatment				
	<b>PLA-L</b>	Poultry litter ash at low rate treatment				
	<b>PLA-H</b>	Poultry litter ash at high rate treatment				
	<b>DWTR-L</b>	Drinking water treatment residual at low rate treatment				
	<b>DWTR-H</b>	Drinking water treatment residual at high rate treatment				
	<b>SS-L</b>	Steel slag at low rate treatment				
	<b>SS-H</b>	Steel slag at high rate treatment				

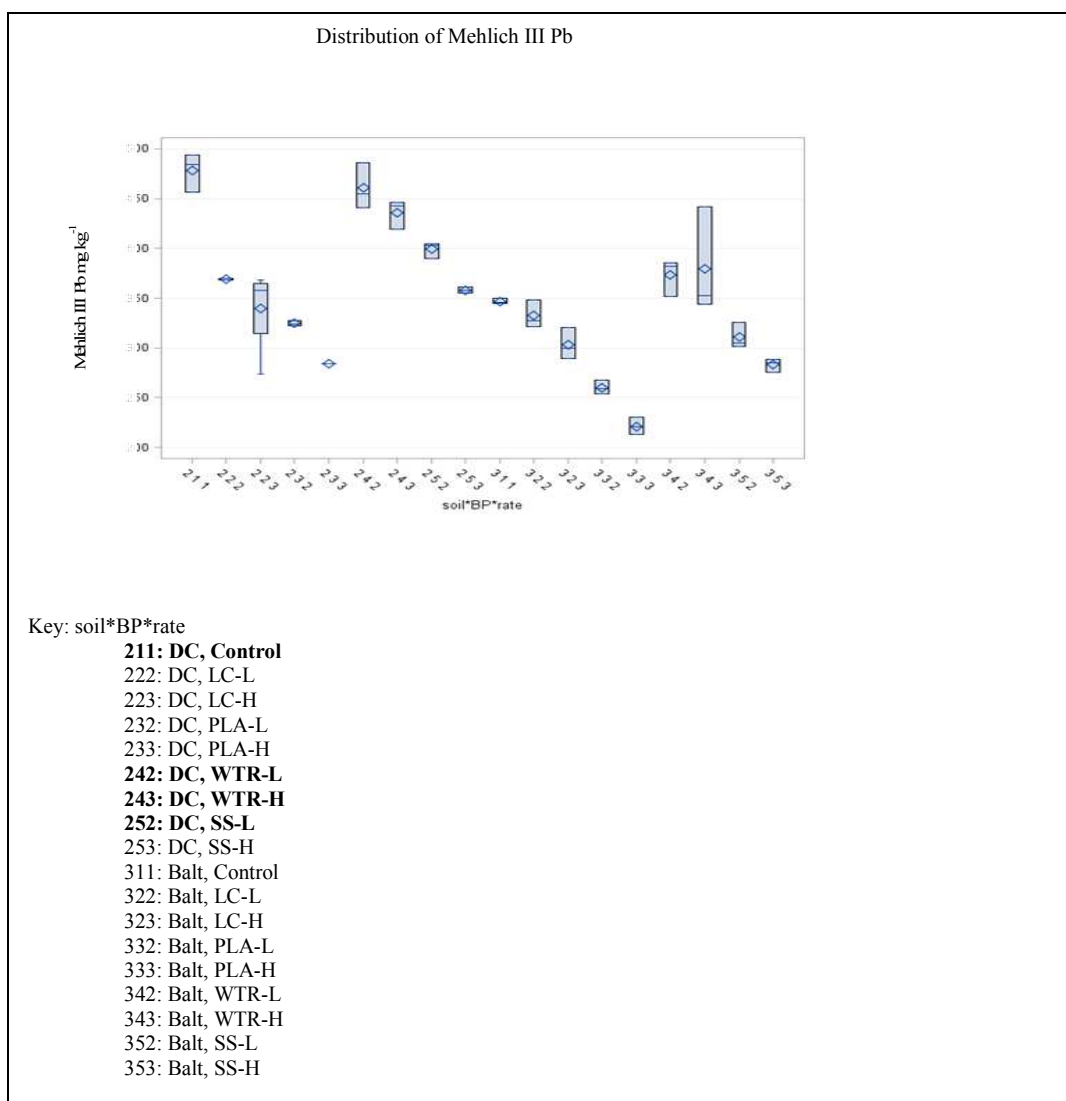
### 3. US EPA and WHO permissible limits on bare soil

As shown in Figure 7, the DC soils treated with leaf compost (LC), both low and high rate treatments; poultry litter ash (PLA), likewise, both low and high rate treatments; and high rate steel slag (SS-H) resulted in Mehlich III extractable lead levels below the U.S. EPA permissible lead limit for bare soil (400 mg kg<sup>-1</sup>) (U.S. EPA, 2001). In the case of the Baltimore soils, all by-product treatments except for the high rate drinking water treatment residual (DWTR-H) resulted in Mehlich III extractable lead levels below

the US EPA permissible lead limit for bare soil. None of the treatments produced Mehlich III extractable lead levels below the WHO permissible limit lead in bare soil ( $100 \text{ mg kg}^{-1}$ ) (FAO/WHO, 2001).

These results must be conditioned by the fact that the Mehlich III method of determining extractable Pb may underestimate the total Pb levels which are the basis for the US EPA and WHO standards. Hence, it is not possible to state that these amendments resulted in soils with total Pb levels below the US EPA and WHO standards.

In Figure 7, the horizontal lines and the diamonds within the box plots represent the respective median and mean values of the control and treatments of the x-axis identified treatments 211 through 353. The box top and bottom are the third and first quartile, and the whiskers (lines extending out of the box) are the extreme values.



**Figure 9: The effect of by-product treatments on the distribution of Mehlich III Pb in Pb contaminated residential DC and Baltimore soils. Note that the bolded treatments are those where the mean and median values of the control and treated soils are above the U.S. EPA Permissible Pb limit for bare and garden soils (400 mg kg<sup>-1</sup>).**

#### 4. Water Extractable Lead Content

As shown in Table 8, the analysis of the DC and Baltimore soils produced water extractable lead levels that were very low as compared with EPA and WHO standards.

For this reason, Mehlich III extractable lead was used as the indicator of permissible lead in this study. No further analysis was performed using the water extractable lead results.

**Table 8: Water and Mehlich III Extractable Pb levels in untreated Pb contaminated soils**

<b>Pb Contaminated Soils</b>	<b>Washington, DC, Soil</b>	<b>Baltimore, MD, Soil</b>
<b>Water Extractable Pb (mg kg<sup>-1</sup>) Control Values</b>	0.881	0.883
<b>Mehlich III Extractable Pb (mg kg<sup>-1</sup>) Control Values</b>	477.0	347.0

## **5. By-product treatment effects on the crop yields**

The results for the by-product effects on crop yields are shown in Table 9. The 95% confidence intervals are shown for the difference in mean yield as compared with the controls. The values shown in red depict a mean that is statistically less than the control. Similarly, the values shown in green depict mean yields that are statistically higher than the control values. For example, the LC-L treatment for collards resulted in a statistically significant 81% decrease in crop yield in the DC soil as well as a statistically significant 49% decrease in crop yield in the Baltimore soil. Whereas, LC-L treatment for beets resulted in a statistically significant 170% increase in crop yield in the DC soil but no statistically significant change in crop yield in Baltimore soil. No results were recorded for PLA-H; the plants did not grow when subject to this level of treatment.

**Table 9: By-product effects on crop yield (g)**

By-Product	Crop	DC soil					Baltimore Soil				
		Mean (g)	SD	% Change	95% Confidence		Mean (g)	SD	% Change	95% Confidence	
					Low	High				Low	High
Control	Collards	17.2	2.2				26.1	2.1			
	Lettuce	9.9	0.6				11.1	1.2			
	Carrots	10.1	2.9				19.2	0.8			
	Beets	4.4	1.9				5.1	1.9			
LC-L	Collards	3.3	0.5	-81.0	-17.1	-10.8	13.3	0.7	-49.0	-15.9	-9.7
	Lettuce	9.1	1.6	-8.6	-3.2	1.5	11.1	0.8	0.3	-2.0	2.0
	Carrots	9.5	1.8	-6.3	-5.4	4.1	18.7	5.7	-2.6	-8.5	7.5
	Beets	11.9	2.5	170.2	3.2	11.8	11.6	2.3	163.9	2.5	10.7
LC-H	Collards	2.4	0.5	-86.1	-18.0	-11.7	5.2	1.3	-80.2	-24.4	-17.4
	Lettuce	7.0	1.7	-29.1	-5.4	-0.4	9.2	0.9	-16.8	-3.9	0.2
	Carrots	2.8	0.7	-72.0	-11.4	-3.2	9.1	0.6	-52.5	-11.5	-8.7
	Beets	11.8	3.7	167.7	1.7	13.1	14.9	1.5	237.7	6.4	13.2
PLA-L	Collards	6.2	1.1	-64.2	-14.5	-7.6	12.2	0.9	-53.2	-17.1	-10.7
	Lettuce	2.4	0.4	-76.3	-8.5	-6.6	2.2	1.1	-80.3	-11.2	-6.7
	Carrots	0.7	1.0	-92.7	-13.6	-5.2	3.8	1.3	-80.3	-17.6	-13.3
	Beets	11.8	1.0	169.1	4.5	10.4	16.4	1.5	272.3	8.0	14.7
PLA-H	Collards										
	Lettuce										
	Carrots										
	Beets										
WTR-L	Collards	18.6	1.0	8.0	-2.0	4.8	27.3	0.3	4.6	-1.8	4.2
	Lettuce	11.0	0.6	10.9	0.0	2.2	12.9	1.4	16.5	-0.7	4.3
	Carrots	8.4	3.5	-17.5	-8.1	4.6	13.1	2.2	-31.8	-9.3	-2.9
	Beets	5.7	2.2	28.6	-2.7	5.2	10.0	2.5	127.3	0.6	9.3
WTR-H	Collards	19.0	1.3	10.2	-1.8	5.4	27.4	3.1	4.9	-4.0	6.5
	Lettuce	11.3	1.9	13.7	-1.4	4.1	15.0	1.1	35.4	1.7	6.1
	Carrots	4.8	0.9	-52.3	-9.5	-1.1	10.7	3.8	-44.1	-13.9	-3.1
	Beets	8.8	1.3	100.0	1.2	7.6	10.3	3.6	133.9	-0.5	10.9
SS-L	Collards	17.5	0.9	1.7	-3.0	3.6	24.5	4.2	-6.1	-8.1	4.9
	Lettuce	10.7	0.6	7.4	-0.4	1.9	11.4	0.4	2.7	-1.4	2.0
	Carrots	7.2	1.9	-28.9	-7.7	1.9	15.6	3.8	-19.0	-9.0	1.7
	Beets	10.2	2.4	132.1	1.6	10.0	15.3	5.0	247.1	2.8	17.6
SS-H	Collards	16.2	2.5	-6.2	-5.7	3.6	22.8	2.1	-12.8	-7.5	0.8
	Lettuce	9.8	1.7	-1.1	-2.6	2.4	11.5	1.5	3.9	-2.2	3.0
	Carrots	4.7	1.9	-53.9	-10.3	-0.6	12.7	4.1	-33.9	-12.3	-0.7
	Beets	11.0	1.3	150.0	3.5	9.7	14.5	1.9	229.6	5.7	13.2
Key		Statistically significant increase in yield from the control									
		Statistically significant decrease in yield from the control									

## 6. By-product treatment effects on Mehlich III Lead in plants

The results for the by-product effects on Pb in plants are shown in Table 10. The values shown in red depict a mean that is statistically significantly greater than the

control. Similarly, the values shown in green depict mean yields that are statistically significantly lower than the control values. No results were recorded for PLA-H; the plants did not grow when subject to this level of treatment. Similarly, no results for DWTR-L in DC soil were recorded for carrots due to a laboratory error; those samples were misplaced and mislabeled by a temporary student technician.

**Table 10: Treatment effects of Pb in plants**

By-Product Treatment	Plant	DC Soil		Baltimore Soil	
		Pb (mg kg <sup>-1</sup> )	% Change	Pb (mg kg <sup>-1</sup> )	% change
Control	Collards	1.80		0.53	
	Lettuce	1.29		0.50	
	Carrots	14.05		6.65	
	Beets	4.12		3.92	
LC-L	Collards	0.54	-70.00	0.30	-43.40
	Lettuce	1.49	15.50	0.70	40.00
	Carrots	5.59	-60.21	2.50	-62.41
	Beets	BDL	-100.00	0.03	-99.23
LC-H	Collards	0.21	-88.33	0.13	-75.47
	Lettuce	0.94	-27.13	0.89	78.00
	Carrots	1.98	-85.91	1.32	-80.15
	Beets	BDL	-100.00	BDL	-100.00
PLA-L	Collards	1.12	-37.78	0.16	-69.81
	Lettuce	0.04	-96.90	0.65	30.00
	Carrots	5.19	-63.06	1.82	-72.63
	Beets	BDL	-100.00	BDL	-100.00
PLA-H	Collards				
	Lettuce				
	Carrots				
	Beets				
DWTR-L	Collards	2.15	19.44	0.90	69.81
	Lettuce	1.40	8.53	0.83	66.00
	Carrots			7.34	10.38
	Beets	6.62	60.68	6.15	56.89
DWTR-H	Collards	2.29	27.22	0.36	-32.08
	Lettuce	1.84	42.64	0.85	70.00
	Carrots	12.30	-12.46	7.48	12.48
	Beets	5.19	25.97	8.78	123.98
SS-L	Collards	0.62	-65.56	0.21	-60.38
	Lettuce	1.97	52.71	0.45	-10.00
	Carrots	11.54	-17.86	6.51	-2.11
	Beets	BDL	-100.00	BDL	-100.00
SS-H	Collards	0.63	-65.00	0.24	-54.72
	Lettuce	2.00	55.04	0.70	40.00
	Carrots	11.64	-17.15	6.09	-8.42
	Beets	BDL	-100.00	BDL	-100.00
Key		Statistically significant increase in M3-Lead mean			
		Statistically significant decrease in M3-Lead mean			
		Below detectable limits (BDL) of M3-Lead, but reduction was not statistically significant			

Appendix A shows the results of the complete ANOVA analyses for treatment, rate, and soil as they affect lead and other chemical elements. The results show that Pb



levels in plants are significantly correlated with soil lead levels and treatment type both individually and together; notably plant lead levels were not correlated with rate of treatment.

SAS results for the Duncan Multiple Range tests of mean differences in plant lead levels as compared with the controls are included for reference as Appendix B. The appendix is organized by soil type.

## **B. Observations**

In addition to the numerical results, a number of observations were recorded during the laboratory phase of the research. These observations and their significance relative to the study are discussed here.

### **1. Leaf Compost (LC) treatment effects**

Leaf compost did not encourage plant growth; this observation is opposite to expected results as leaf compost is often used as a fertilizer. Figure 8 shows the yield stunting effect of LC-L and LC-H treatments (middle and far right, respectively) on collard crop compared to the control (far left).



**Figure 10: Stunted crop yield and leaf yellowing response to LC by-product treatments in collards**

Figure 8 also shows the yellowing effect observed for collards that were planted in soils treated with the LC-H by-product. A leaf yellowing effect (chlorosis) is a common indicator of nitrogen deficiency.

There are a number of reasons why crops treated with leaf compost may have resulted in reduced yields during the early part of the study compared to their controls. Unstable or immature compost may result in poor crop growth and yield by competing for nutrients and oxygen or causing phytotoxicity to crops due to insufficient biodegradation of organic matter. Furthermore, immature compost typically immobilizes nitrogen instead of releasing it for crop growth. This is due to the high decomposition activity of soil microbes and their scavenging of available nutrients as they continue to

decompose immature compost organic matter even after its application to soil (Ofosu-Budu et al. 2010).

This negative crop yield effect from soil microbe vs plant competition has been observed by several researchers. In their study on the effect of tree leaf litter on maize performance Nyathi and Campbell (1995) found that senescent leaf litter from *Leuceana Leucocephala* and *Miombo* leaf litter resulted in a decrease in both shoot dry matter and grain yields compared to controls. Bardgett et al. (2003) provided evidence that microbes are significant competitors to plants for organic and inorganic nitrogen in low-productivity grasslands using  $^{15}\text{N}$  and  $^{13}\text{C}$  labeled compound markers. Several other researchers using  $^{15}\text{N}$ , sometimes with  $^{13}\text{C}$ , markers demonstrated repeatedly that soil microbes do out compete with plants for organic and inorganic nitrogen by rapid nitrogen biomass immobilization through assimilation; thus denying those nutrients to neighboring plants in the short-term (Cheng and Bledsoe, 2004; Dunn et al., 2006; McFarland et al., 2010; Kastovska and Santruckova, 2011).

Immature leaf compost, a low quality resource, may be composed of leaf tissue with initially low nitrogen concentrations (Mtambanengwe et al., 2006). The immature compost may have a low release rate of nitrogen following its application, it may contain organic chemical compounds (polyphenol and lignin) in percentages high enough to induce extended periods of immobilization due to nitrogen binding by these compounds, thus slowing down N mineralization (Chivenge et al. 2009). High microbial activity in immature compost can cause biological blockage of nitrogen from crops by out-competing plants for the limited store of available nitrogen of both the organic and soil

derived mineral nitrogen, also the very microbes that mediate the composting process secrete certain phytotoxic chemical compounds (i.e., short chain organic acids: acetic, propionic and butyric acids and ammonia) during the early stages of the composting process can be a threat to plant growth and crop yields (Sullivan, D. and R. Miller 2001; El-Nagerabi et al. 2011). One or a combination of the above previously observed and reported reasons may be the cause of the hindered crop yields during the early phase of the study (Cooperban, 2000).

Thus, an essential main requirement for compost to be a suitable amendment for plant growth and decent crop yield is aging, organic fractions (in particular cellulose, hemicellulose and lignin) of composting matter must have undergone sufficient physical and chemical transformation over time to achieve biological stability (humification) to become useful for crops (de Bertoldi et al., 1983).

## **2. Poultry Litter Ash treatment effects**

Crop growth in the PLA treated soils was limited or non-existent. In each of the high rate PLA (PLA-H) treated pots, a hard crusting and white precipitate on the soil surfaces were observed (Figures 9 and 10).

Hashimoto et al. (2009) speculated that the high pH measurement (13.0) they recorded in their study of PLA treated soils most likely resulted from solubilization of phosphorus and alkaline salts in the by-product. In this study, it is speculated, the application of PLA to the investigated soils resulted in high salt conditions (salt stress)

and high soil pH conditions (alkali stress) causing stunted growth in crops in low rate treatments and no crop growth in the high rate treatments.

Salt stress on plants: 1) decreases the osmotic potential of the soil solution effectively creating water stress for plants, 2) causes ion toxicity due to excess accumulation of  $\text{Na}^+$  or  $\text{Cl}^-$  in plant cells, and 3) results in nutrient imbalances and deficiencies due to the interaction of salts with mineral nutrients (Islam et al. 2011). Alkali stress in high pH soil conditions is detrimental to the supply capacity of mineral nutrients, directly destroys the structure and function of root cells, and is known to have a more severe effect on plant growth than salt stress (Yang et al., 2007; Keren, 2000; Marschner, 1995; Paz et al., 2012). The greatest stressor is the combined synergistic effect of salt stress and alkali stress. Salt and alkali stress produce similar levels of growth inhibition in many plant species; however, their combination mutually enhances the deleterious effects on growth (Shi and Sheng, 2005; Paz et al., 2012). It is reasonable to suspect that the severe stress reactions by all crops grown in both contaminated soils treated with PLA was the result of alkali stress from the high pH (10.7) and salt stress from the high electrical conductivity (EC) (59.6 mS/cm) (Table 6).



**Figure 11: Soil crusting for high rate poultry litter ash treatments**



**Figure 12: White deposits on the soil surface of high rate poultry litter ash treatments**



**Figure 13: Lettuce growth effect for control, PLA-L, and PLA-H treatments in residential Baltimore soil**

### **3. DC Soil discrepancy**

The DC soils maps suggest that the surface A-horizon of the collected DC soil was a manor loam variety based on location. Upon visual and textural observation, the soil appeared to be more clay-like than loam-like. As shown in Figure 12, the soil had a reddish color typical of clayey soil types. The homeowner suggested that fill dirt had been deposited following a construction project in the general location from which the soil was sampled.





**Figure 14: DC soil collection site**



## **CHAPTER FIVE – CONCLUSIONS**

### **A. Research hypotheses**

The objective of this study was to determine whether certain agricultural, industrial, and municipal waste by-products could effectively and safely reduce levels of lead in urban garden soils to prevent inadvertent ingestion. To accomplish this objective, the following four hypotheses were tested:

1. The addition of agricultural and industrial by-products to lead contaminated soils will reduce the levels of lead in the soils
2. The addition of agricultural and industrial by-products to lead contaminated soils will not reduce harvest yields
3. The addition of agricultural and industrial by-products to lead contaminated soils will reduce the levels of lead in the most consumed parts of leafy and rooty crops
4. The addition of agricultural and industrial by-products to lead contaminated soils will reduce the levels of lead to within EPA and WHO permissible limits

#### **1. Hypothesis 1 – lead levels in soils**

The results showed a statistically significant reduction in soil lead levels as compared with the control for all by-products with the exception of DWTP which demonstrated mixed results; in one case the Mehlich III Pb levels in the soil increased

with the application of the DWTP by-product (see Table 7). The chemical analysis of the DWTP by-product showed a high level of lead ( $220 \text{ mg kg}^{-1}$ ) and was the probable cause for the poor results (see Table 6).

## **2. Hypothesis 2 – harvest yield**

Harvest yield varied by treatment. The results show that all by-product treatments (except PLA-H and DWTR-L) had a statistically significant effect at increasing beet root crop yields. No other crops grown in treated DC soil experienced statistically significant yield increases. On the contrary, most other treatments resulted in statistically significant reductions in most other crop yields. Both treatment rates of LC and low rate PLA treatments significantly reduced crop yields for collards, lettuce, and carrots grown in DC soil, compared to their controls (Table 11). Low rate treatments of PLA had a statistically significant effect of reducing collard, lettuce, and carrots crop yields. Both rate treatments of DWTR significantly reduced carrot yields (Table 11).

The results show that the low rate and high rate steel slag (SS-L, SS-H) either improved or had no effect on the harvest yield. LC-L, DWTR-H, DWTR-L and SS-H all showed promising results for three of the four plants, either with no statistically significant change in crop yield or statistically significant increase in crop yield for three of the four plants. PLA-L was effective only for beets, with statistically significant increases in crop yield; however, PLA-L resulted in statistically significant reductions in crop yield for collards, lettuce, and carrots. PLA-H was completely ineffective, resulting in no crop growth.

**Table 11: Plant Crop Yields for By-Product Treatments in Residential DC soil**

Treatment	Rate	Collards	Lettuce	Carrots	Beets
LC	High				
	Low				
PLA	High	N/G	N/G	N/G	N/G
	Low				
DWTR	High				
	Low				
SS	High				
	Low				
Key			Statistically significant decrease in crop yields		
			Statistically significant increase in crop yields		
		N/G	No crop growth in high rate PLA treatments		

Similar to the results in the DC soil, beet root crops responded well to most by-product treatments (Table 12). Three treatments (DWTR-H, DWTR-L, and SS-L) showed promising results for all crops; they produced either no change or a statistically significant increase in yield for at least one crop. Treatments SS-H and LC-L resulted in statistically significant reduction in yield for one crop, increase in yield for one crop, and no change in yield for the other two crops. Treatments PLA-L and LC-H resulted in two or three crops with statistically significant reductions in yield and only one crop with a statistically significant increase in yield. At high rates of treatment, PLA (PLA-H) did not support plant life, thus there were no results for those treatments. This was apparently due to salt deposition as evidenced by the high electro-conductivity and visual

evidence of salt precipitation on high rate PLA treated soil surfaces. In all cases, more testing at different rates is needed to establish recommendations for application.

**Table 12: Plant Crop Yields for By-Product Treatments in Residential Baltimore soil**

<b>Treatment</b>	<b>Rate</b>	<b>Collards</b>	<b>Lettuce</b>	<b>Carrots</b>	<b>Beets</b>
<b>LC</b>	<b>High</b>				
	<b>Low</b>				
<b>PLA</b>	<b>High</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>
	<b>Low</b>				
<b>DWTR</b>	<b>High</b>				
	<b>Low</b>				
<b>SS</b>	<b>High</b>				
	<b>Low</b>				
<b>Key</b>			<b>Statistically significant decrease in crop yields</b>		
			<b>Statistically significant increase in crop yields</b>		
		<b>N/G</b>	<b>No crop growth in high rate PLA treatments</b>		

### **3. Hypothesis 3 –Lead levels in plants**

Both rate treatments of LC (LC-L, LC-H) were effective at statistically significantly reducing crop tissue Pb content below the control levels for three out of four plants grown in DC soil (collards, carrots, and beets), and with no change in lead levels in the fourth plant (lettuce) (Table 13). Low rate PLA treatments effectively resulted in statistically significant decreases in crop tissue Pb levels in carrots and beets. Both treatment rates of steel slag significantly reduced crop tissue lead levels in collards and beets, compared to their controls (Table 13). Both DTWR-L and DTWR-H treatments

resulted in no statistically significant change in Pb levels. The PLA-H treatment resulted in no growth (N/G) and therefore no measurable Pb levels in the plants.

**Table 13: Crop Tissue Pb Levels for By-Product Treatments in Residential DC Soil**

<b>Treatment</b>	<b>Rate</b>	<b>Collards</b>	<b>Lettuce</b>	<b>Carrots</b>	<b>Beets</b>
<b>LC</b>	<b>High</b>				
	<b>Low</b>				
<b>PLA</b>	<b>High</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>
	<b>Low</b>				
<b>DWTR</b>	<b>High</b>				
	<b>Low</b>				
<b>SS</b>	<b>High</b>				
	<b>Low</b>				
<b>Key</b>			<b>Statistically significant increase in crop tissue Pb content</b>		
			<b>Statistically significant decrease in crop tissue Pb content</b>		
		<b>N/G</b>	<b>No crop growth in high rate PLA treatments</b>		

With some similarity to the results in the DC soil, both rate treatments of LC were effective at reducing crop tissue Pb content for two out of four plants grown in Baltimore soil (carrots, and beets) (Table 14). Low rate PLA reduced lead in carrots and in beets as well. Also, both treatment rates of Steel Slag reduced crop tissue lead in beets. The only treatment that resulted in a statistically significant increase in crop tissue Pb was DWTR-H. The PLA-H treatment resulted in no plant growth (N/G).

**Table 14: Crop Tissue Lead Levels for By-Product Treatments in Residential Baltimore Soil**

<b>Treatment</b>	<b>Rate</b>	<b>Collards</b>	<b>Lettuce</b>	<b>Carrots</b>	<b>Beets</b>
<b>LC</b>	<b>High</b>				
	<b>Low</b>				
<b>PLA</b>	<b>High</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>
	<b>Low</b>				
<b>DWTR</b>	<b>High</b>				
	<b>Low</b>				
<b>SS</b>	<b>High</b>				
	<b>Low</b>				
<b>Key</b>			<b>Statistically significant increase in crop tissue Pb content</b>		
			<b>Statistically significant decrease in crop tissue Pb content</b>		
		<b>N/G</b>	<b>No crop growth in high rate PLA treatments</b>		

#### **4. Hypothesis 4 – ability to meet EPA and WHO limits**

The Melich III method of determining Pb content is not equivalent to the method required by WHO; hence, the results of this study with regard to the WHO standards must be considered carefully. WHO uses total lead as the measure for its standards (0.3 mg kg<sup>-1</sup> for leafy vegetables; and 0.1 mg kg<sup>-1</sup> for rooty vegetables). Melich III produces extractable Pb levels that are less than total lead. Therefore, even if Melich III Pb levels satisfy the WHO standards, it may be true that total Pb would exceed the standards.

Nonetheless, it may be useful to comment on those treatments that result in the Melich III Pb levels to satisfy the WHO standards. Clearly, those treatments that result in Melich III

Pb levels that fail to meet the WHO standards will also have total Pb levels fail to meet the standards.

The following rate treatments effectively reduced the Melich III extractable Pb levels in vegetables grown in DC soil to within WHO permissible Pb limits: high rate LC for collards and both LC treatment rates for beets (Table 15). PLA low rate treatment for beets, and both treatment rate of steel slag were effective at reducing Melich III Pb levels to within WHO limits (Table 15).

Vegetables grown in Baltimore soil had the Melich III Pb levels reduced to WHO standard levels for some treatments and plants (Table 16). Melich III Pb levels in beets were reduced to the WHO standard for LC-H, LC-L, PLA-L, SS-H, and SS-L treatments. Additionally, the LC-H, LC-L, and PLA-L treatments reduced Melich III Pb levels in collards to the WHO standard.

**Table 15: Melich III Pb levels meeting WHO standard for Vegetables Grown in DC Soil**

<b>Treatment</b>	<b>Rate</b>	<b>Collards</b>	<b>Lettuce</b>	<b>Carrots</b>	<b>Beets</b>
<b>LC</b>	<b>High</b>				
	<b>Low</b>				
<b>PLA</b>	<b>High</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>
	<b>Low</b>				
<b>WTR</b>	<b>High</b>				
	<b>Low</b>				
<b>SS</b>	<b>High</b>				
	<b>Low</b>				
<b>Key</b>			<b>By-product treatments which met WHO permissible Pb limits for leafy and rooty vegetables</b>		
		<b>N/G</b>	<b>No crop growth in high rate PLA treatments</b>		

**Table 16: Melich III Pb levels meeting WHO standard for Vegetables Grown in Baltimore Soil**

<b>Treatment</b>	<b>Rate</b>	<b>Collards</b>	<b>Lettuce</b>	<b>Carrots</b>	<b>Beets</b>
<b>LC</b>	<b>High</b>				
	<b>Low</b>				
<b>PLA</b>	<b>High</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>	<b>N/G</b>
	<b>Low</b>				
<b>DWTR</b>	<b>High</b>				
	<b>Low</b>				
<b>SS</b>	<b>High</b>				
	<b>Low</b>				
<b>Key</b>			<b>By-product treatments which met WHO permissible Pb limits for leafy and rooty vegetables</b>		
		<b>N/G</b>	<b>No crop growth in high rate PLA treatments</b>		



## **B. Limitations of the Experimental Design**

Several conclusions regarding the experimental design are important. First and foremost, the number of by-products tested (4) was overly ambitious relative to the desired outcome. By selecting a fewer number of by-products and testing more rates, possibly better information regarding the desired rate of treatment for each by-product could have been derived. As previously discussed, the high rate PLA treatment killed both the rooty and leafy plants. However, the low rate PLA enabled plant growth for beets, and both the high and low rate PLA reduced the lead levels in soils. A greater number of treatment rates would possibly have enabled a regression analysis to show the relationship between rate of treatment as the independent variable and yield or lead level as the dependent variable.

Three samples were taken for each treatment (soil/by-product/level). However, some of the Mehlich III Lead levels were found to be “below detectable limits” following treatment. Although the lead levels were reduced to virtually zero, the results were not statistically significant due to low sample sizes. A greater number of samples per treatment would have possibly increased the opportunity to make statistically significant conclusions.

Nitrogen levels for the soil and by-products were not established. The USDA By-products Utilization Laboratory has a standard list of chemicals that are analyzed for each material being evaluated. The USDA standard list was adopted for use in this research. However, the list does not include nitrogen which is an important plant nutrient.

Lastly, the organic matter content should have been analyzed for the LC by-product. LC that has not aged properly may retard plant growth. Soil bacteria responsible for LC decomposition will compete with the plants for nitrogen, thus depriving the plants of that important nutrient.

### **C. Recommendations for Future Research**

The follow-on study to this research should investigate the levels of by-products that are suggested for urban garden application. Additional rate levels for each by-product between the high and low rates are needed to show the trends from which conclusions regarding rate of application could be based. The noted exception is the rate of PLA application. Additional testing at levels slightly above and below the low rate of PLA are recommended.

An analysis of the effectiveness of by-product combinations is also recommended. By-products that improve yield might be combined with by-products that reduce lead levels in the edible parts of plants. DWTR improved plant yield in many cases but did not notably reduce lead levels. DWTR might be coupled with SS or another by-product to improve both yield and extractable lead levels.

Future studies using one of several bioavailable or bioaccessible soil Pb test methods within a similar experimental design like the present study could provide valuable understanding of how Pb, sorbed to ingested soil particles, in treated or untreated soils, can dissolve under simulated digestive system conditions and become potentially available to the bloodstream. Due to the scale of the present study and the

time requirement for the conduct of even quickest those soil Pb test, those methods were not employed in this study (Mincaa et al., 2013; Chaney et al., 2011).

## **APPENDICES**

## APPENDIX A

The values in the tables in this appendix are the results of a standard ANOVA analysis (Codling, 2007).

Effect of Soil, Treatment (Trt), Rate, and interactions on the F-values and Pr > F values of various elements in analyzed Collards Crop leaf tissue.

Source	DF	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F
		Pb		P		Ca		Fe		Al		Mn	
Soil	2	47.42	<.0001	28.75	<.0001	6.50	0.0032	0.14	0.8665	0.63	0.5393	7.13	0.0020
Trt	4	8.09	<.0001	7.83	<.0001	27.25	<.0001	0.73	0.5772	0.38	0.8238	23.82	<.0001
Rate	1	0.27	0.6074	3.76	0.0586	1.77	0.1895	1.31	0.2590	1.86	0.1791	0.79	0.3784
Soil*Trt	8	5.27	<.0001	3.55	0.0027	2.86	0.0112	0.70	0.6879	1.25	0.2949	12.30	<.0001
Soil*Rate	2	0.65	0.5279	0.43	0.6561	0.39	0.6783	1.43	0.2503	1.06	0.3532	0.78	0.4660
Trt*Rate	2	0.60	0.5506	2.85	0.0682	2.71	0.0772	1.80	0.1768	1.59	0.2151	0.87	0.4268
Soil*Trt*Rate	4	0.41	0.8002	1.07	0.3814	1.79	0.1470	1.03	0.4004	1.34	0.2692	1.17	0.3365

**Effect of Soil, Treatment (Trt), Rate, and interactions on the F-values and Pr > F values of various elements in analyzed Lettuce Crop leaf tissue.**

Source	DF	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F
		<b>Pb</b>		<b>P</b>		<b>Ca</b>		<b>Fe</b>		<b>Al</b>		<b>Mn</b>	
<b>Soil</b>	2	41.86	<.0001	0.61	0.5482	6.32	0.0039	0.57	0.598	1.39	0.2593	18.37	<.0001
<b>Trt</b>	4	10.85	<.0001	8.09	<.0001	5.19	0.0017	1.06	0.3859	0.67	0.6171	15.14	<.0001
<b>Rate</b>	1	0.27	0.6091	0.04	0.8353	0.00	0.9568	2.06	0.1589	1.70	0.1990	5.62	0.0223
<b>Soil*Trt</b>	8	17.28	<.0001	0.88	0.5382	3.20	0.0062	0.86	0.5595	0.76	0.6386	16.14	<.0001
<b>Soil*Rate</b>	2	0.06	0.9390	2.61	0.0855	0.51	0.6035	1.09	0.3442	1.10	0.3420	1.55	0.2247
<b>Trt*Rate</b>	2	0.07	0.9350	1.08	0.3489	0.73	0.4857	0.73	0.4886	1.53	0.2273	2.11	0.1333
<b>Soil*Trt*Rate</b>	4	0.09	0.9836	0.62	0.6503	2.62	0.0479	1.68	0.1729	1.12	0.3575	4.19	0.0060

**Effect of Soil, Treatment (Trt), Rate, and interactions on the F-values and Pr > F values of various elements in analyzed Carrot Root tissue.**

Source	DF	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F
		<b>Pb</b>		<b>P</b>		<b>Ca</b>		<b>Fe</b>		<b>Al</b>		<b>Mn</b>	
<b>Soil</b>	2	180.46	<0.0001	4.78	0.0137	18.10	<0.0001	1.25	0.2972	10.05	0.0003	10.50	0.0002
<b>Trt</b>	4	34.94	<0.0001	5.91	0.0008	12.56	<0.0001	0.28	0.8870	5.37	0.0015	6.81	0.0003
<b>Rate</b>	1	1.15	0.2908	0.65	0.4231	9.61	0.0035	2.05	0.1599	0.13	0.7241	0.70	0.4087
<b>Soil*Trt</b>	8	9.96	<0.0001	2.53	0.0247	5.18	0.0002	0.24	0.9817	5.50	0.0001	2.00	0.0708
<b>Soil*Rate</b>	2	1.52	0.2306	0.05	0.9484	19.36	<0.0001	0.99	0.3791	0.92	0.4063	0.63	0.5397
<b>Trt*Rate</b>	2	1.70	0.1957	0.98	0.3845	22.12	<0.0001	2.40	0.1041	1.26	0.2946	1.62	0.2114
<b>Soil*Trt*Rate</b>	3	0.75	0.5261	2.84	0.0499	19.92	<0.0001	0.94	0.4298	1.22	0.3146	4.90	0.0054

Effect of Soil, Treatment (Trt), Rate, and interactions on the F-values and Pr > F values of various elements in analyzed Beet Root tissue.

Source	DF	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F	F-value	Pr > F
		<b>Pb</b>		<b>P</b>		<b>Ca</b>		<b>Fe</b>		<b>Al</b>		<b>Mn</b>	
<b>Soil</b>	2	55.53	<.0001	5.66	0.0064	3.00	0.0598	4.41	0.0177	0.32	0.7264	14.98	<.0001
<b>Trt</b>	4	25.99	<.0001	30.30	<.0001	28.27	<.0001	16.56	<.0001	10.19	<.0001	95.91	<.0001
<b>Rate</b>	1	0.14	0.7088	4.63	0.0367	8.39	0.0058	0.08	0.7758	0.55	0.4638	1.72	0.1962
<b>Soil*Trt</b>	8	10.58	<.0001	1.54	0.1708	0.88	0.5384	2.24	0.0410	2.63	0.0185	5.32	<.0001
<b>Soil*Rate</b>	2	1.05	0.3596	0.14	0.8737	0.45	0.6429	0.80	0.4549	0.54	0.5870	0.75	0.4775
<b>Trt*Rate</b>	2	1.56	0.2213	0.49	0.6141	1.89	0.1632	1.83	0.1716	0.30	0.7410	5.59	0.0067
<b>Soil*Trt*Rate</b>	4	0.73	0.5755	0.38	0.8219	0.31	0.8690	1.15	0.3456	0.30	0.8770	1.09	0.3717

## **APPENDIX B**

The lower case letters within the element columns below are provided to illustrate statistically significant differences between the means of various treatment effects. Means within a column sharing the same small case letter(s) are not statistically different; those treatments within the same column that do not share any small case letter(s) are statistically different at the  $p < 0.05$  level. BDL = below detection limit of the instrument. N/S = no sample.



**Elemental composition Collard Leaf Tissue grown in by-product amended lead contaminated soils**

Trt	Rate	Pb	Al	Ca	Fe	Mn	P	Zn
-----mg Kg <sup>-1</sup> -----								
Fort DuDont National Park Soil								
Control	Zero	BDL	BDL	21109 def	43.1 b	31.1 def	1740.3 ghi	13.3 h
LC	Low	BDL	1.92 a	19859 d-g	43.3 b	9.9 g	2990.7 c-h	12.8 h
LC	High	BDL	BDL	15695 fg	49.1 b	12.7 fg	2681.3 d-i	12.8 h
PLA	Low	BDL	BDL	3698 h	43.3 b	20.0 efg	2137.0 f-i	12.3 h
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	BDL	BDL	31451 abc	33.3 b	64.4 b	2777.3 d-i	26.6 h
WTR	High	BDL	BDL	26633 bcd	30.3 b	64.2 b	1478.3 i	18.1 h
SS	Low	BDL	BDL	14796 gf	27.9 b	45.7 bcd	1658.0 hi	25.7 h
SS	High	0.26 def	BDL	16204 gf	23.1 b	18.4 efg	1478.3 i	25.7 h
Residential Washington, DC, Soil								
Control	Zero	1.80 ab	1.48 a	20903 def	21.9 b	26.2 d-g	2628.0 d-i	285.2 bc
LC	Low	0.54- c-f	BDL	24875 cde	9.04 b	44.8 bcd	3109.7 c-g	63.3 gh
LC	High	0.21 def	BDL	16736 fg	136.3 a	36.3 cde	2409.7 e-i	67.1 gh
PLA	Low	1.12 bc	BDL	15338 fg	BDL	53.6 bc	3818.7 b-e	342.1 b
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	2.15 a	BDL	35275 a	24.2 b	64.7 b	3458.3 c-f	328.4 b
WTR	High	2.29 a	BDL	33261 ab	28.1 b	64.7 b	2746.0 d-i	317.8 b
SS	Low	0.62 cde	1.76 a	14379 fg	25.7 b	32.6 def	2762.0 d-i	152.7 def
SS	High	0.63 cde	0.93 a	17017 fg	26.1 b	34.5 cde	2219.0 f-i	129.5 efg
Residential Baltimore, MD, Soil								
Control	Zero	0.53 c-f	BDL	16216 fg	34.6 b	30.9 def	3445.7 c-f	212.3 cd
LC	Low	0.30 def	0.45 a	13353 fg	21.0 b	27.4 d-g	4049.7 bcd	113.8 efg
LC	High	0.13 ef	1.95 a	18771 efg	30.0 b	13.4 fg	5148.3 b	115.4 efg
PLA	Low	0.16 ef	0.22 a	17911 efg	41.4 b	109.2 a	7698.0 a	596.2 a
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	0.90 cd	0.05 a	26889 bcd	34.1 b	34.1 cde	4360.7 bc	328.4 b
WTR	High	0.36 def	BDL	18586 efg	23.1 b	38.0 cde	2602.7 d-i	170.2 de
SS	Low	0.21 def	BDL	12761 g	27.9 b	27.4 d-g	3048.7 c-h	76.1 fgh
SS	High	0.24 def	BDL	15338 fg	27.9 b	26.1 d-g	3246.3 cdef	69.3 gh

**Elemental composition Lettuce Leaf Tissue grown in by-product amended lead contaminated soils**

Ttt	Rate	Pb	Al	Ca	Fe	Mn	P	Zn
-----mg Kg <sup>-1</sup> -----								
Fort DuDont National Park Soil								
Control	Zero	BDL	1.05 b	16316 a	54.3 b	23.9 def	4675 abc	13.4 g
LC	Low	BDL	BDL	11471 b-f	43.4 b	30.5 c-f	5631 ab	28.5 fg
LC	High	0.26 a	1.02 b	8010 e-h	36.1 b	23.6 def	5384 ab	BDL
PLA	Low	0.88 a	4.70 b	3847 h	45.9 b	160.86 a	5367 ab	BDL
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	0.00 a	0.78 b	8071 d-h	27.2 b	28.1 def	1810 d	5.0 g
WTR	High	BDL	3.21 b	14991 ab	53.7 b	90.6 b	3222 dc	10.1 g
SS	Low	BDL	BDL	14602 abc	41.2 b	19.8 def	3283 dc	42.1 d-g
SS	High	BDL	BDL	12633 a-d	38.0 b	90.6 b	4980 abc	88.8 bc
Residential Washington, DC, Soil								
Control	Zero	1.29 a	6.97 b	9790 def	38.2 b	36.9 cd	3974 bc	89.5 bc
LC	Low	1.49 a	1.40 b	9553 def	38.7 b	28.2 def	5644 ab	89.9 bc
LC	High	0.94 a	2.09 b	8781 d-g	41.5 b	27.7 def	5738 ab	79.3 bcd
PLA	Low	0.04 a	BDL	9285 d-g	33.7 b	19.5 def	5030 abc	147.2 a
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	1.40 a	2.78 b	11301 b-f	35.1 b	24.9 def	4043 bc	94.3 b
WTR	High	1.84 a	2.41 b	9451 def	32.3 b	24.4 def	4546 abc	86.4 bc
SS	Low	1.97 a	2.15 b	9965 def	33.8 b	15.3 def	4572 abc	47.5 c-g
SS	High	2.00 a	2.27 b	9697 def	33.8 b	16.9 def	4301 bc	47.6 c-g
Residential Baltimore, MD, Soil								
Control	Zero	0.50 a	2.28 b	9521 def	33.0 b	19.7 def	4959 abc	87.4 bc
LC	Low	0.70 a	1.63 b	10383 c-f	34.2 b	26.5 def	6280 a	96.3 b
LC	High	0.89 a	120.57 a	12432 a-e	93.6 a	53.4 c	4736 abc	61.9 b-f
PLA	Low	0.65 a	5.15 b	4790 gh	33.6 b	27.1 def	5280 ab	95.1 b
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	0.83 a	4.91 b	8382 d-h	32.2 b	26.6 def	4480 abc	73.5 b-e
WTR	High	0.85 a	3.72 b	8330 d-h	33.3 b	33.7 cde	3398 cd	62.0 b-f
SS	Low	0.45 a	1.99 b	7642 fgh	25.5 b	10.0 ef	4088 bc	30.4 efg
SS	High	0.70 a	3.65 b	7975 e-h	29.0 b	9.0 f	4102 bc	34.7 efg

**Elemental composition Carrot Root Tissue grown in by-product amended lead contaminated soils**

Treatments	Rate	Pb	Al	Ca	Fe	Mn	P	Zn
-----mg Kg <sup>-1</sup> -----								
Fort DuDont National Park Soil								
Control	Zero	0.23 c	0.66 e	2853 c-f	20.42 abc	6.44 cde	1264 de	7.12 j
LC	Low	0.15 c	0.80 e	2012 gh	57.87 ab	12.51 bc	3066 bc	9.62 j
LC	High	0.10 c	0.38 e	3562 bc	23.75 abc	8.29 cde	1782 cde	11.99 j
PLA	Low	0.05 c	0.68 e	2767 c-f	18.42 abc	4.32 e	1705 cde	10.21 j
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	0.19 c	2.22 bcd	3709 b	39.04 abc	18.75 ab	13.79 de	10.22 j
WTR	High	0.20 c	2.73 b	2558 d-g	26.75 abc	20.08 a	1791 cde	9.13 j
SS	Low	0.25 c	0.55 e	2038 fgh	12.71 c	3.41 e	838 e	10.61 j
SS	High	1.14 c	2.32 bc	7427 a	43.73 abc	20.11 a	3167 bc	36.82 d-g
Residential Washington, DC, Soil								
Control	Zero	14.05 a	1.00 de	3189 bcd	22.47 abc	6.93 cde	2486 cd	40.54 cde
LC	Low	5.59 b	0.80 e	2601 d-g	60.53 a	4.20 e	2177 cde	28.92 ghi
LC	High	1.98 c	0.77 e	2388 d-g	6.80 c	5.29 de	2087 cde	21.44 i
PLA	Low	5.19 b	4.04 a	1353 hi	24.10 abc	7.38 cde	5831 a	65.50 a
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	N/S	0.67 e	N/S	N/S	N/S	N/S	N/S
WTR	High	12.30 a	1.38 cde	2512 d-g	17.57 abc	7.85 cde	2331 cde	38.33 def
SS	Low	11.54 a	0.67 e	3017 b-e	21.34 abc	7.39 cde	1830 cde	33.57 e-h
SS	High	11.64 a	0.71 e	2640 d-g	22.51 abc	6.99 cde	2072 cde	36.82 d-g
Residential Baltimore, MD, Soil								
Control	Zero	6.65 b	0.70 e	2538 d-g	20.31 abc	5.17 de	2619 cd	51.39 b
LC	Low	2.50 c	0.53 e	2549 d-g	14.62 bc	4.37 e	2338 cde	28.45 ghi
LC	High	1.32 c	0.31 e	2630 d-g	15.24 bc	4.40 e	3155 bc	27.75 ghi
PLA	Low	1.82 c	0.61 e	924 i	19.74 abc	8.51 cde	4548 ab	61.94 a
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	7.34 b	0.56 e	2667 d-g	20.50 abc	9.31 cde	2471 cd	48.06 bc
WTR	High	7.48 b	0.19 e	2324 efg	20.13 abc	11.83 cde	2529 cd	44.97 bcd
SS	Low	6.51 b	0.22 e	2499 d-g	19.16 abc	7.67 cde	2363 cde	25.62 hi
SS	High	6.09 b	0.23 e	2549 d-g	19.33 abc	6.527 cde	2363 cde	30.03 f-i

**Elemental composition Beet Root Tissue grown in by-product amended lead contaminated soils**

Treatments	Rate	Pb	Al	Ca	Fe	Mn	P	Zn
-----mg Kg <sup>-1</sup> -----								
Fort DuDont National Park Soil								
Control	Zero	BDL	90.7 a	8031 bc	28.8 b-f	12.9 fg	8561 abc	105.9 ef
LC	Low	BDL	26.2 f	2836 efg	13.8 efg	10.9 g	3999 g	19.8 f
LC	High	BDL	32.4 ef	1726 efg	12.9 efg	14.5 fg	4201 fg	19.0 f
PLA	Low	BDL	65.5 a-d	534 g	12.8 efg	30.1 de	3536 g	20.4 f
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	BDL	55.4 b-e	13783 a	44.3 b	51.1 bc	9044 ab	79.5 f
WTR	High	BDL	80.9 ab	8031 bc	70.9 a	57.0 b	8179 bc	70.6 f
SS	Low	BDL	40.0 def	2963 efg	22.0 c-g	12.6 fg	5066 efg	35.5 f
SS	High	BDL	39.4 def	1726 efg	15.3 efg	7.8 g	3266 g	25.1 f
Residential Washington, DC, Soil								
Control	Zero	4.12 b	43.4 d-g	7212 bcd	18.7 c-g	16.3 fg	5533 d-g	285.3 cd
LC	Low	BDL	45.3 c-f	2446 efg	19.1 c-g	11.6 g	4565 fg	110.2 ef
LC	High	BDL	45.6 c-f	1351 fg	16.2 efg	10.6 g	4452 fg	81.1 f
PLA	Low	BDL	45.1 c-f	511 g	15.8 efg	7.4 g	4369 fg	41.6 f
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	6.62 ab	70.2 abc	9554 b	27.9 b-g	32.6 de	7736 bcd	414.5 b
WTR	High	5.19 b	69.8 abc	7574 bc	29.3 b-e	40.7 cd	7374 b-e	238.6 d
SS	Low	BDL	41.6 c-f	3221 d-g	17.1 d-g	10.1 g	4640 fg	72.0 f
SS	High	BDL	41.4 c-f	2504 efg	10.6 g	7.4 g	3520 g	45.5 f
Residential Baltimore, MD, Soil								
Control	Zero	3.92 b	43.9 c-f	5627 b-e	20.5 c-g	24.6 ef	7155 b-e	398.3 bc
LC	Low	0.03 c	43.6 c-f	3248 d-g	18.5 c-g	16.1 fg	6512 c-f	203.9 de
LC	High	BDL	43.9 c-f	1554 fg	22.0 c-g	14.7 fg	5597 d-g	104.1 ef
PLA	Low	BDL	44.6 c-f	513 g	14.6 efg	16.7 fg	4633 fg	33.8 f
PLA	High	N/S	N/S	N/S	N/S	N/S	N/S	N/S
WTR	Low	6.15 ab	78.6 ab	8245 bc	36.8 bc	57.7 b	10704 a	552 a
WTR	High	8.78 a	78.4 ab	5072 c-f	35.5 bcd	82.6 a	9015 ab	443.2 ab
SS	Low	BDL	43.3 c-f	1820 efg	11.9 efg	9.2 g	4603 fg	55.1 f
SS	High	BDL	43.3 c-f	1544 fg	9.7 g	6.2 g	4095 g	31.1 f

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