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Urban gardens: Lead exposure, recontamination mechanisms, and implications for remediation design

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ABSTRACT

Environmental lead contamination is prevalent in urban areas where soil represents a significant sink and pathway of exposure. This study characterizes the speciation of lead that is relevant to local recontamination and to human exposure in the backyard gardens of Roxbury and Dorchester, MA, USA. One hundred forty-one backyard gardens were tested by X-ray fluorescence, and 81% of gardens have lead levels above the US EPA action limit of 400 μg/g. Raised gardening beds are the *in situ* exposure reduction method used in the communities to promote urban gardening. Raised beds were tested for lead and the results showed that the lead concentration increased from an initial range of 150 ± 40 μg/g to an average of 336 μg/g over 4 years. The percent distribution of lead in the fine grain soil (<100 μm) and the trace metal signature of the raised beds support the conclusion that the mechanism of recontamination is wind-transported particles. Scanning electron microscopy and sequential extraction were used to characterize the speciation of lead, and the trace metal signature of the fine grain soil in both gardens and raised gardening beds is characteristic of lead-based paint. This study demonstrates that raised beds are a limited exposure reduction method and require maintenance to achieve exposure reduction goals. An exposure model was developed based on a suite of parameters that combine relevant values from the literature with site-specific quantification of exposure pathways. This model suggests that consumption of homegrown produce accounts for only 3% of children's daily exposure of lead while ingestion of fine grained soil (<100 μm) accounts for 82% of the daily exposure. This study indicates that urban lead remediation on a yard-by-yard scale requires constant maintenance and that remediation may need to occur on a neighborhood-wide scale.

1. Introduction

Lead is a persistent environmental contaminant and is a public health concern because of its properties as a neurotoxin to children (Ryan et al., 2004; Mielke et al., 1999; Lanphear et al., 1998). Lead contamination is ubiquitous in urban communities and urban soils act as an integrator of decades of Pb pollution. The primary anthropogenic sources of Pb to urban soil are Pb-based paint, used on 89% of exterior residential structures built before 1978, and emissions from the combustion of leaded gasoline (Rabinowitz, 2005; Ryan et al., 2004; ATSDR, 2000).

Contact with Pb in the urban environment occurs through multiple pathways of exposure (Clark et al., 2006). The primary pathway of human exposure to Pb is through the ingestion of soil (Hettiarachchi and Pierzynski, 2004; Ryan et al., 2004; Mielke and Reagan, 1998; Lanphear and Roghmann, 1997). The consumption of produce grown in contaminated soil can act as a pathway of exposure as the plant tissue has the ability to bioaccumulate Pb (Hettiarachchi and Pierzynski, 2004; Chaney et al., 1997).

The Boston communities of Roxbury and Dorchester, MA, have elevated rates of Pb poisoning in children. These urban and underserved communities reflect the current demographics of Pb poisoning in the United States in that they have the highest rates of elevated blood lead level (BLL) in children in the Boston area. 3.3% of children screened in Dorchester have BLL above 10 μg/dL, which is 1.5 times higher than the overall Boston and national average (Boston Public Health Commission, 2006; CDC, 2003; Dorchester Environmental Health Coalition, 2003).

The presence of Pb in the environment is further complicated by the prevalent cultural practice of backyard gardening that is characteristic of Roxbury homes. The number and density of backyard gardens make this community a unique exposure setting and the reliance on homegrown produce as a food source makes it necessary to quantify produce consumption as a route of exposure for Pb entering the human system. In order to promote urban gardening and prevent Pb exposure from the consumption of homegrown produce. The Food Project, a not-for-profit organization based in Roxbury, has

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started a program to construct raised gardening beds for current and new gardeners. The raised beds are $1 \times 3\,m^2$, wooden (pine) boxes lined with landscaping fabric to reduce contact with underlying contaminated soil, and filled with compost. The Food Project has built approximately 50 raised beds and has excavated (i.e., removed all soil from a yard and replaced with compost) one site.

Alternative urban soil Pb intervention plans to reduce Pb exposure have been in place in the Roxbury and Dorchester communities and other regions (Binns et al., 2004; Litt et al., 2002; Hynes et al., 2001; Farrell et al., 1998; Blaylock et al., 1997; Aschengrau et al., 1994). Lead remediation schemes include the excavation of soil, the application of soil/ground cover or barriers (e.g., pavement or grass), and the application of chelating agents or biosolids to remove/sequester Pb. Raised beds are generally considered a Pb remediation technique in the literature but this study questions whether that classification is appropriate. The program in place in Roxbury and Dorchester and other experimental urban programs all occur on the scale of an individual yard. They do not address the role of neighboring yards or local or regional Pb contamination in the effectiveness of remediation on a yard-by-yard scale.

This study investigates the effectiveness of raised beds as a Pb exposure reduction technique, and addresses the potential of urban Pb to be mobilized and recontaminate remediated sites. This study also models the total Pb exposure to the urban gardeners in these communities. The principal objectives of this study are to: (1) identify the physical distribution of Pb in gardens, raised beds, and compost; (2) evaluate the speciation of Pb in soil grains $<100\,\mu m$; (3) investigate the change in the Pb concentration in raised beds over time; and (4) model the average daily exposure of Pb in a site-specific exposure model.

2. Materials and methods

2.1. Study site and sample collection

Roxbury and Dorchester, MA, USA, were selected as the site for this study because of the documented Pb contamination (Litt et al., 2002; Spittler and Feder, 1979), and the partnership with The Food Project, which made contaminated sites available. Soil was collected from 141 backyard gardens between 2003 and 2007. The Food Project enlisted gardeners to participate in this study and the only criterion for screening gardens for Pb was that they were actively used for growing produce. The average garden was approximately $10–20\,m^2$, and garden soil was generally adjacent to the residential structure. Samples were also collected from 23 raised gardening beds. The Food Project provided Roxbury and Dorchester community members with compost produced by the City of Boston to fill the raised beds and this compost was tested for initial Pb concentration. Samples from a total of 25 sites were collected and tested for Pb concentration multiple times, while five gardens, two raised beds, and one excavation site were analyzed annually to provide continuous data for 3–5 years. A minimum of four samples, generally two from the surface horizon (0–10 cm) and two from the rooting depth (30–40 cm), were collected from each site. While concentration gradients have been observed in urban yards (Litt et al., 2002), the sites sampled in this study are urban gardens and the soil is well mixed due to multiple years of gardening activity, such as tilling. Therefore, a small number of samples provided a representative profile of soil Pb concentration (see Clark et al., 2006 Table 1 for statistical justification of sampling protocol).

Several plant species were grown in contaminated Roxbury and Dorchester soil to evaluate Pb uptake both *in situ* and in a greenhouse. The species grown in this study included the documented metal-accumulating species of mizuna mustard (*Brassica rapa*), collards (*Brassica oleracea*), and sunflowers (*Helianthus annuus*), as well as beans (*Phaseolus vulgaris*) (Prasad, 2003). The selection criteria for these species were based on the plant's ability to evapotranspire soil water, to bioaccumulate contaminants, and to mature quickly (Chaney et al., 1997).

2.2. Sample preparation and bulk lead analysis

Soil and plant Pb concentrations were analyzed by two complementary X-ray fluorescence (XRF) approaches. A field portable X-ray fluorescence (FP-XRF) Niton (Thermo Electron Corporation, Billerica, MA) instrument was used to measure bulk soil Pb concentrations. A polarized energy dispersive X-ray fluorescence (pED-XRF) Spectro Xepos (Spectro Analytical, Kleve, Germany) instrument was used to analyze a subset of samples (10% randomly selected) as well as all plant samples. The additional testing of the 10% subset by the second method was conducted in accordance with EPA method 6200, which, for quality control purposes, requires that a subset of *in situ* FP-XRF samples be analyzed by a complementary analytical method (US EPA, 1996a).

Using a 12 mci ^{109}Cd source, FP-XRF achieved $\pm 10\%$ analytical error for soil Pb by counting for 60 decay-corrected seconds. pED-XRF generally achieved an analytical error of $\pm 5\%$. Testing of all unknown samples via XRF was bracketed with National Institute of Standards and Technology 2709 or 2711 Standard Reference Material. Measured concentrations of all elements of interest in the standards remained within $\pm 10\%$ of accepted values.

All samples were dried at 50 °C for 2–3 days. Aliquots of 4 g of soil sample were prepared in XRF sample cups with $6\,\mu m$ thick Mylar film windows. Plants were individually washed and gently scrubbed by hand, soaked in deionized water for 10 min, and then washed a second time. To ensure that all soil and dust particles were removed from the surface of the plant tissue, concentrations of Si and Al were monitored as trace elements to indicate the presence of soil and dust at the time of analysis.

A second washing procedure was also developed to mimic the washing style that would occur in a resident's kitchen. In this procedure, the plant material was lightly washed for a short period of time, allowing some soil or dust material to remain adhered to the plant tissue, as it would in the kitchen setting. All plant material was prepared as pellets by grinding the plant tissue for 5 min in a tungsten carbide mixer mill, adding SpectroBlend® binding agent, and pressing under 10 metric tons of pressure.

2.3. Physical and chemical analysis

Soil, raised bed, and compost samples were size-fractionated and grain sizes ranging from 4 mm to $10\,\mu m$ were achieved by mechanical sieving. Grains of a diameter $<10\,\mu m$, also known as particulate matter 10 (PM10), were collected with a MicroMesh® Electroformed mesh nickel sieve (InterNet Inc., Minneapolis, MN). This grain size was collected because of its importance in both the exposure and transport of soil Pb. Each of the nine size fractions collected were analyzed by pED-XRF. PM10 samples for gardens and raised beds were analyzed by backscatter scanning electron microscopy (SEM-EDS) at Massachusetts Institute of Technology with energy dispersive X-ray element mapping (Leo 438VP, Leo, Inc., UK). Soil and compost samples were mounted on carbon tape and the chamber was vacated to 10 Pa. Samples were analyzed using 20 kV accelerating velocity, with a beam current of 400 mA, at a working distance of 15 mm, and a point dwell of $10{,}000\,\mu s$. SEM-BSE was used to provide spatial correlations of trace metals and to identify general mineral speciation of Pb-bearing grains.

An abbreviated sequential extraction method, described by Tessier et al. (1979), was conducted to partition Pb into different labile fractions for application in the exposure model of this study. This sequential extraction was designed to characterize the distribution of Pb among geochemically defined fractions to provide information about the matrix-specific phases present and the chemical conditions that can mobilize Pb from particles. This geochemical partitioning can also be used to distinguish between labile forms of Pb. Two fractions of Pb were extracted in this study: the exchangeable fraction, which represents ionically bound Pb displaced by cation exchange, and carbonates, which represents Pb mobilized by slightly acidic conditions (Tessier et al., 1979; Schaider et al., 2007). These two fractions were extracted because the sum of the exchangeable and carbonate fractions represents the bioaccessible fraction of soil Pb, which is applied in the exposure model of this study. Schaider et al. (2007) found that the sum of Pb extracted by these two fractions correlated strongly with Pb extracted by an in vitro physiologically based extraction test ($p<0.0001$). Bulk and size-fractionated ($<44\,\mu m$) samples from three gardens and a compost sample were tested. Four grams of each sample were finely ground, homogenized, and placed in 50 mL polypropylene centrifuge tubes and subjected to the following treatment:

(i) *Exchangeable*: 20 mL of the first extracting agent, 1 M $MgCl_2$ at pH 5, were added and the tubes were shaken to ensure saturation of the sample. The tubes were then placed on a shaker table for 1 h at a speed of 40 rpm.
(ii) *Carbonate*: 20 mL of the second extracting agent, 1 M NaOAc at pH 2, were added and the tubes were again shaken and placed on the shaker table for 5 h at 40 rpm.
(iii) *Reducible, organic, and residual*: these phases were not extracted.

After each individual treatment, the samples were removed from the table and centrifuged at 3000 rpm for 30 min. The samples were then decanted and filtered using a syringe with a $2\,\mu m$ filter, and rinsed with deionized water between treatments.

All samples were analyzed by UV–VIS using a Spectronic AquaMate spectrophotometer (Thermo Scientific, Waltham, MA) following Hach® LeadTrack™ method H2210 QNT Lead. Blanks for each extracting agent were prepared and subtracted from each sample for analysis. The blank for $MgCl_2$ accounted for up to 70% of the Pb measured and the blank for NaOAc accounted for up to 10% of the Pb.

3. Results and discussion

3.1. Bulk lead distribution

The average Pb concentration measured in the 141 gardens ($n = 692$ samples) was 950 μg/g. The average values for each garden ranged from 80 to 3680 μg/g with a median value of 800 μg/g. The Pb was homogenously distributed in the soil of an individual garden and there was no gradient in Pb concentration as a function of distance from the residential structure or the road as a result of tilling the soil for gardening (Clark et al., 2006). The average concentration of Pb measured at 23 raised gardening beds ($n = 135$ samples) was 336 μg/g. The average Pb concentration measured in compost collected throughout 2006 and 2007 ($n = 30$) ranged from 110 to 190 μg/g, with an average of 150 μg/g. The average Pb concentration found in raised beds was twice the initial Pb concentration range measured in compost before it enters the urban environment. This finding supports the hypothesis that recontamination is occurring in the neighborhood.

Several sites were monitored annually for Pb concentration. Table 1 shows the annual measurement of Pb concentration in five gardens, two raised beds, and one excavation site. The variation measured in soil Pb concentration between years was generally within the uncertainty range of the garden for a single year. Site #18 was excavated in 2004, whereas raised gardening beds in sites #30 and #92 were constructed in 2003. In 2004, the concentration of Pb measured in sites #18 and #30 at 6–12 months after soil intervention had increased from the initial compost concentration range of 150 ± 40 μg/g. For site #92, the concentration change was not statistically significant in the first year measured, but the concentration did increase to statistical significance at the 2005 measurement. The concentration of Pb measured in raised beds/excavation sites continued to increase in the subsequent years, but the change was similar to variation in garden soil from year-to-year. When the data from Table 1 is viewed as the change in Pb concentration from the first year of sampling to the last year of sampling, the important distinction in the raised beds/excavation site and gardens can be observed. The average Pb concentration for the three raised beds/excavation site in 2004 was 340 μg/g, which increases to an average of 630 μg/g in 2007. In contrast, the average Pb concentration for the five gardens included in Table 1 was 1136 μg/g in 2004 and 1157 μg/g in 2007. It is likely that Pb is being redistributed in the gardens and raised beds constantly by wind-transported particles. The effect of this mobilized Pb cannot be observed as statistically significant in the gardens because the initial concentration of Pb is high and the variation is relatively large. However, the raised beds were filled with compost at an initially low Pb concentration allowing relatively small increases in Pb concentration to be observed as the compost was contaminated with wind-transported Pb. This data demonstrates that an excavated site operates similarly to a raised bed in a contaminated urban environment and appears to be equally likely to experience an increase in Pb concentration over time.

Table 1
Annual measurement of lead concentration

	2004	2005 (μg/g[a])	2006	2007
#18 Exc	546 ± 107	548 ± 79	673 ± 130	719 ± 103
#30 RB	323 ± 48	395 ± 8	583 ± 85	367 ± 67
#92 RB	153 ± 42	756 ± 308	583 ± 174	813 ± 145
#27 G	1878 ± 160	1471 ± 115	1613 ± 225	NA
#30 G	NA	2127 ± 625	2045 ± 496	1830 ± 546
#60 G	305 ± 33	700 ± 175	466 ± 151	692 ± 69
#70 G	709 ± 25	741 ± 32	NA	936 ± 205
#92 G	1653 ± 237	1643 ± 457	NA	1157 ± 324

G, garden; RB, raised bed; Exc, excavation site.

[a] Values represent average concentration measured by pED-XRF in 4–6 samples ± the standard deviation of those measurements.

3.2. Physical distribution of lead

To investigate the mechanisms associated with the recontamination of raised beds, an inventory of the distribution of Pb as a function of grain size was performed. An inverse relationship exists between the concentration of Pb and grain size. The garden-wide average concentration of Pb in soil grains <100 μm is 2000 μg/g, which is more than two times greater than bulk soil (Fig. 1). The bulk concentration of Pb in the three gardens presented in Fig. 1 range from 500 to 3600 μg/g, and the physical distribution of Pb is independent of bulk Pb concentration. This suggests that similar physical processes such as *in situ* weathering and grain size reduction are operating across gardens, and that surface area considerations play a key role in the distribution of Pb as a function of grain size.

The <100 μm grain size soil fraction is associated with elevated Pb concentrations and is generally considered wind-transportable (Clevenger et al., 1991). For this study, it was hypothesized that wind-transported fine grain soil was the mechanism of recontamination, and the physical distribution of Pb can be used to support this mechanism. Both the mass of <100 μm soil and the percent of Pb accounted for in the <100 μm fraction increases from compost to raised beds. The mass of soil attributed to this fraction was generally <5% in compost, whereas this fraction accounted for 10–15% of the mass in raised bed samples. Fig. 2 shows that the percent of Pb in the <100 μm fraction also increases from 5% of the total Pb in compost to an average of 12% in raised beds, as highlighted by the dash lines illustrating the average for each percent distribution of <100 μm fraction in compost, raised beds, and gardens. This data further illustrates that excavation sites are operationally similar to raised beds in urban communities.

In situ weathering and grain size reduction, as seen in garden soil, do not explain the increased presence of fine grain soil in compost. In Fig. 2, it can be observed that the percent of Pb in the size fractions >2 mm and 2–0.5 mm accounts for roughly 50% of Pb in both compost and raised beds, whereas the sum of those two fractions accounts for, on average, 35% of Pb in garden soil. If *in*

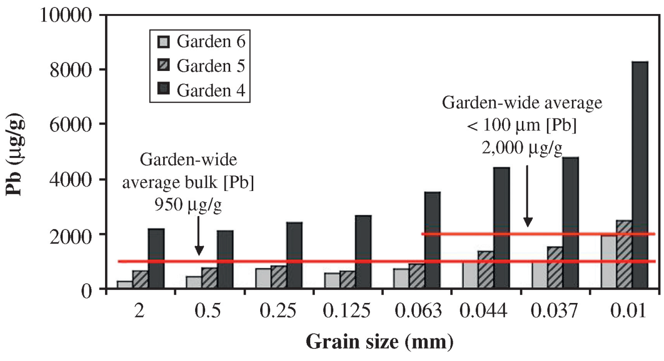

Fig. 1. Distribution of lead as a function of grain size in soil from three urban backyard gardens. Bold lines represent the average bulk and <100 μm Pb concentration for all ($n = 141$) gardens tested, including the three gardens presented.

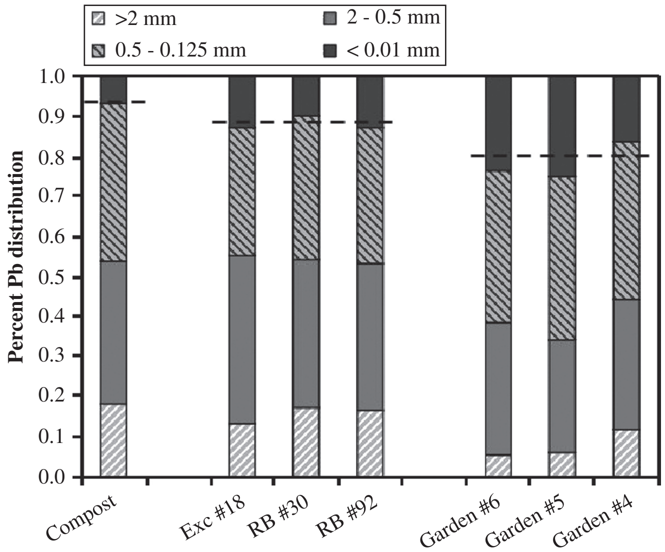

Fig. 2. Distribution of lead content (%) by soil grain size fraction in compost, three raised beds/excavation sites, and three urban backyard gardens. The dashed lines represent the average percent of lead in the $<100\,\mu m$ size fraction for compost, raised beds, and gardens to illustrate the difference in the three soil media.

situ weathering were responsible for the increase in fine grain soil, the distribution of size fractions for raised beds would change from resembling the compost profile to resembling the soil profile. This trend was not observed on the time scale the raised beds have been operating (<5 years), indicating that fine grain soil is being transported to raised beds, not created *in situ*. The end member concentrations of Si found in compost and gardens, and the intermediate concentration found in raised beds and excavation sites further supports the hypothesis that material is transported into remediated sites. Typical urban garden soil is characterized by Si concentrations that are buffered by the modal abundance of the minerals quartz and feldspar. These minerals are not key constituents of the organic-rich compost used to fill the raised beds; therefore, an increase in Si concentration within the raised beds would signal additional mass input to the bed. The average concentrations of Si measured in compost and gardens were 10 and 17 wt%, respectively, and raised beds were measured at an intermediate level of 12 wt% Si. This suggests that processes other than *in situ* weathering are operating on the raised bed.

A mass balance calculation can be used to generate a range for the percent of total change in bulk Pb concentration measured in raised beds that can be attributed to Pb represented in grains size $<100\,\mu m$. The mass balance calculation is based on the change in Pb concentration from compost to raised beds ($[Pb]_{RB}-[Pb]_{Comp} = \Delta Pb = 180 \pm 45\,\mu g/g$), the change in the average percent of mass represented in the fine ($<100\,\mu m$) fraction ($\%fine_{RB}-\%fine_{Comp} = \Delta\%fine = 7\%$), and the average concentration of Pb measured in the $<100\,\mu m$ fraction of raised beds ($1161\,\mu g/g$). This calculation indicates that a range of 35–61% of the observed change in Pb concentration (ΔPb) in raised beds can be attributed to the addition of fine grain soil. These physical lines of evidence support the claim that wind-transported fine grain soil is an important process for redistributing Pb in the urban setting and that this must be considered when designing remediation schemes.

3.3. *Chemio-textural distribution of lead*

Spatial and geochemical relationships for a suite of trace elements can be used to fingerprint the source of Pb that is involved the recontamination in raised beds/excavation sites. Using isotopic methods and site-specific isotopic ratios for the proposed isotopic end members in their mixing model, Clark et al. (2006) were able to determine that Pb-based paint is likely an important source of Pb to local soil, accounting for roughly 40–80% of the soil Pb inventory. Since Ti is a key component of paint products it can serve as a unique trace element marker for paint-derived materials. A plot of Pb concentration versus Ti for compost, raised beds, and garden soil should point to mixing relationships between the paint-free compost and the paint-contaminated garden soil. Fig. 3 illustrates this mixing process showing the geochemical evolution of raised beds shown as a linear array treading from a compost-dominated signature to a more Pb and Ti-elevated garden soil composition. This linear array is based on a two-point linear regression of the plotted average for the compost data and the plotted 90th percentile for the garden soil data. The 90th percentile was the chosen statistic to present the central tendency of the soil data set as it minimizes the influence of outliers not representative of the sample population.

Spatial correlations collected by SEM-EDS further illustrate that the trace metal signature associated with Pb-based paint can be found in raised beds. Elemental mapping of PM10 samples from raised beds revealed grain scale associations among the principal components of Pb-based paint, including Pb, Ti, Ba, and Cr (Hall and Tinklenberg, 2003; Toch, 1916). These elements were all components of various Pb-based paint formulas, with titanium dioxide being the primary Pb-free formula introduced when Pb-based formulas were banned. The elemental composition of Pb-based paint has been shown to be conservative as soil weathers to fine grain ($<100\,\mu m$) particulate matter (Gulson et al., 1995). Analyses of individual Pb-bearing grains from PM10 garden soil and raised beds show that Pb is bound with Fe and Ti oxides, as well as Ba-silicates in both environmental mediums. These Pb-anion associations in conjunction with both trace metal signatures and spatial relationship signatures suggest that recontamination of raised beds is taking place on a garden-wide scale and wind is the likely transport mechanism.

The chemio-textural signature of raised beds can also be used to eliminate atmospheric deposition of Pb as a mechanism of transport. The average concentration of Pb measured in air at a Boston air quality monitoring station from 2000 to 2005 is $0.005\,\mu g/m^3$ (Massachusetts Department of Environmental Protection (MA DEP), 2005). This source of Pb would not account for the contribution of the mass of fine grain particulate matter, the

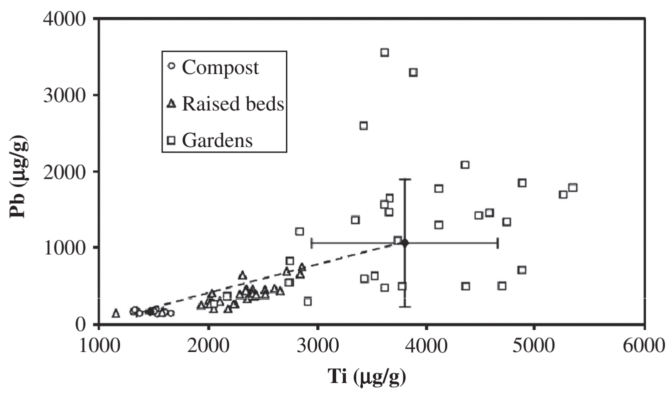

Fig. 3. Geochemical trace elements in compost, raised beds, and garden are plotted with titanium, a trace element associated with paint, on the *x*-axis to separates the population. The average value for the compost samples and the 90th percentile of garden soil samples are plotted with *x*- and *y*-error bars representing the standard deviation of the population. The dashed line represents a two-point linear regression between average compost and 90th percentile garden soil.

percent of Pb in fine grain soil, or the trace metal signature which is associated with Pb-based paint found in raised beds.

3.4. Site-specific exposure model

The exposure model developed in this study estimates the average daily exposure of environmental Pb in children age 2–6 years old in the Roxbury and Dorchester communities. The model is based on the presentation by Glorennec (2006), which studied the exposure of Pb in a mining community in France and generated an estimate indirectly from environmental samples and parameters in the literature. This study quantifies the percent contribution of the multiple environmental media (soil, ambient air, water, and homegrown produce) and pathways of exposure (ingestion of soil, inhalation of ambient air, consumption of tap water, and consumption of homegrown produce). Indoor parameters and exposure estimates were not included in this calculation. However, it has been shown that two-thirds of indoor house dust Pb can be attributed to exterior proximate sources (Adgate et al., 1998), suggesting that the majority of indoor exposure occurs from sources of Pb (e.g., soil) that are included in this model. All further discussion will be centered on the average, or expected, Pb exposure to children in Roxbury and Dorchester. Several variables are better quantified than others; the goal of the model is *not* to provide an exact value but to provide a framework for addressing this issue, and to place constraints on a range of possible values to describe the average population. The framework set forth in the following discussion provides a transferable means for assessing exposure.

3.4.1. Lead levels in environmental media

Soil: The Pb concentration of soil as a function of grain size (Fig. 1) reveals that there is greater than a factor of two difference in the concentration of Pb between bulk soil and soil that is $<100\,\mu$m in size. Grains of diameter $<50\,\mu$m are the most likely to adhere to hands and to be ingested during hand-to-mouth activity (Ljung et al., 2006). Therefore, 2000 μg/g is the concentration of Pb that most accurately describes the exposure from ingestion of soil.

Produce: While Glorennec (2006) and many other studies do not incorporate the contribution of homegrown produce in the average exposure of Pb in the urban environment, this study quantifies the role of produce consumption in the overall exposure model due to the local community's reliance on backyard produce for food security. The Pb values measured in various tissue portions from mustards, collards, beans, and sunflowers are presented in Table 2, and the values applied in the model are the concentrations of Pb in the edible portion of mustards washed by the kitchen-mimicking style, the edible portion of collards, and the edible portion of beans. Samples of collards and beans were not subjected to the less-rigorous kitchen-mimicking washing style because not enough were harvested. It is important to note that the concentration of Pb in mustards washed with the kitchen-mimicking style is nearly double the concentration of mustard washed to laboratory quality. This concentration increase is attributed to fine grain soil particles, likely the same size fraction ($<50\,\mu$m) that is ingested during hand-to-mouth activity, that are adhered to the surface of the plant. The quantification of both adhered Pb dust and absorbed Pb further illustrates the importance of understanding the fine grain soil fraction in exposure reduction efforts. For the exposure calculation, values ranging from 10 to 30 μg/g are used to describe the average exposure of Pb from the consumption of homegrown produce.

Table 2
Lead uptake by plant species and tissue type

Species	Tissue sampled	Washing method	Pb (μg/g[a])	No.
Mustard	Whole plant	Lab quality	46	32
Mustard	Root	Lab quality	115	4
Mustard	Edible	Lab quality	13	7
Mustard[b]	Edible	Kitchen mimicking	24	6
Bean[b]	Edible	Lab quality	2	5
Collard[b]	Edible	Lab quality	14	4
Collard	Root	Lab quality	57	2
Sunflower	Root	Lab quality	360	2
Sunflower	Leaf	Lab quality	47	5

[a] Average value for all pellets (3 g/pellet) analyzed by pED-XRF.
[b] Values used in the exposure model calculation.

Water: The concentration of Pb in water is the average of 18 measurements taken directly from the homes of nine gardens in this study. The average value calculated is 4 μg/L, which was measured by colleagues at University of Massachusetts, Boston, who are also consultants on The Food Project's Lead Steering Committee (Personal Communication with Robert Beattie). This value is well below the EPA action limit of 15 μg/L.

Ambient air: The concentration of Pb in backyard air samples was not monitored in this study; therefore, a wide range of values for the concentration of Pb in air was considered from the literature. For the lower end of the range, the value measured by the MA DEP at a monitoring station in Kenmore Square (Boston, MA) is considered. The average concentration for Pb in air at Kenmore Square between the years 2000 and 2005 was 0.005 $\mu g/m^3$ (MA DEP, 2005). However, the land use in Kenmore Square (commercial zone) is dramatically different than the land use in Roxbury and Dorchester (residential zone), and it is likely that the Kenmore Square monitoring station is an underestimate for areas like Roxbury and Dorchester where there is a greater abundance of unpaved surfaces. The high end of the range for Pb in air used is 0.75 $\mu g/m^3$, which is half of the National Ambient Air Quality Standard of 1.5 $\mu g/m^3$ (US EPA, 2006). With only distal monitoring station values available, it is reasonable to consider a much wider range of air quality values to reflect the distinct land use of the study site. For future studies, the accuracy of the output of the exposure model should be improved by collecting air quality data.

3.4.2. Ingestion rate of multiple pathways of exposure

Soil: Ljung et al. (2006) report that 100 mg/day soil is ingested by hand-to-mouth activity in children age 1–6 years.

Produce: The Exposure Factors Handbook published by the US EPA (1997) reports the average consumption of 0.005 g/kg body weight/day of snap beans, 0.0189 g/kg body weight/day of collards, and 0.0145 g/kg body weight/day of mustard greens. This set of data is body weight dependent, and to correct for this value the average body weight range of boys and girls ages 2–6 years must be included. Growth charts published by the CDC (2000) report the weight range for this age group is 12–20 kg. After correcting for the body weight factor, the sum of the produce data set is 0.46–0.77 g/day. It is assumed that 100% of these vegetables consumed are homegrown, and therefore grown in contaminated soil.

Water: Ershow and Cantor (1989) report that an average of 0.65 L/day of tap water is consumed by children aged 1–10 years.

Ambient air: Allan and Richardson (1998) report that children age 7 months to 4 years inhale an average of $9.3\pm2.4\,m^3$/day of air and children age 5–11 years inhale $14.6\pm3\,m^3$/day. For this study, a wide range of 6.9–17.7 m^3/day will be used to incorporate both sets of age data.

Exposure to environmental Pb is weather dependent and is correlated with the hours per day spent outdoors when soil is dry and exposed (Ljung et al., 2006; Glorennec, 2006; Filippelli et al., 2005; Laidlaw et al., 2005). The produce dose calculated by the

Table 3
Average daily exposure of lead based on site-specific environmental media and dose parameters

Exposure media and route	Bulk lead range	Exposure (per day)	Fraction (days/year)	Dose (μg/day)	Percent contribution
Ingestion of soil	2000 μg/g	100 mg/day	0.4	80	72–91
Consumption of produce	10–30 μg/g	0.46–0.77 g/day	0.4	2.6	2–3
Consumption of tap water	4 μg/L	0.65 L/day	1	0.14–5.31	1–5
Inhalation of ambient air	0.05–0.75 μg/m^3	6.9–17.7 m^3/day	0.4	4.8–23.1	2–5
Sum	–	–	–	88–111	–

EPA incorporates the fraction of days per year that exposure occurs, but the values for soil, air, and water must be modified. For this model, drinking water exposure is not weather dependent so the exposure fraction is 100% of days per year, while exposure to soil and dust/air is assumed to occur 40% (fraction = 0.4) of the year. This is likely the same fraction that calculated for home-grown produce consumption and is used in this exposure model.

Table 3 presents the exposure range expected from the data above. The sum of the four exposure media and routes generates an expected average daily exposure of Pb in children age 2–6 years of 88–111 μg/day. This model is able to quantify the relative contribution of homegrown produce as 2–3% of the total exposure to Pb. The model also demonstrates the significance of soil in Pb exposure, as ingestion of soil accounts for 72–91% of the total body burden. It is clear from both the recontamination and exposure perspectives that special consideration should be allocated to fine grain soil in designing exposure and recontamination prevention programs.

3.4.3. *Bioaccessibility and bioavailability considerations*

The degree to which Pb can be absorbed by the human system is a crucial factor in determining exposure and health risks. The bioaccessibility/bioavailability of Pb is widely studied and generally reported to range from 30% to 50% (Glorennec, 2006; Hettiarachchi and Pierzynski, 2004; Ryan et al., 2004; Baars et al., 2001; ATSDR, 2000). One of the few studies based on experiments with adult humans is Maddaloni et al. (1998), and the bioavailable range reported in that study was 1–34%.

The bulk Pb exposure presented in Table 3 can be modified by the bioavailability and/or bioaccessibility of Pb to produce a more accurate exposure assessment. Ruby (2004) defines bioavailability as the fraction of the chemical dose that is absorbed and reaches the systemic circulation, whereas bioaccessibility describes the fraction of the chemical that is liberated in relevant biological fluids and would be available for absorption. The bioaccessible fraction of Pb can be equal or greater than the bioavailable fraction.

If bioavailability/bioaccessibility were ignored and intake was equal to 100% of the exposure, then the total exposure to Pb in this urban setting ranges from 88 to 111 μg/day. The model can be refined by making the assumption that 30% of the bulk Pb in the exposure pathways of soil, produce, and water is bioavailable (Glorennec, 2006). Inhaled Pb is assumed to be 100% bioavailable when it is in the lung tissue (Spear et al., 1998). Given these input parameters, the total body burden of Pb decreases to 30–50 μg/day.

This exposure calculation can be further calibrated to the site-specific variables by using the sequential extraction data collected to derive an estimate of the bioaccessible fraction of soil Pb. The percent of Pb extracted from the ionically and carbonate bound fractions is 1.8%. These two fractions are considered phases able to be liberated in biological systems and the sum of these fractions is considered the bioaccessible fraction (Tessier et al., 1979; Maiz et al., 1997). For the exposure model, a bioaccessible range of 1–2% can be considered. Schaider et al. (2007) found that traditional sequential extractions underestimated the labile fraction of Pb calculated by a physiologically based extraction test by 10–40% in weathered Pb ore mine tailings. This is likely attributed to a combination of differences in pH and temperature of extraction agents and the potential for enhanced ligand-promoted dissolution by glycine (Schaider et al., 2007). It is worth noting that all sequential extraction-based protocols for defining labile fractions of Pb are matrix dependent. However, all parameters used in this study are conservative, therefore underestimating the site-specific bioaccessibility. The bioaccessible range of 1–2% applies only to the Pb measured in this study; therefore, this range is only applied to the soil ingestion and produce consumption pathways of exposure, whereas the default values are used for water and air. This consideration produces a total exposure range of 6–26 μg/day. A summary of the average daily exposure ranges is presented in Table 4.

Table 4
Bioaccessibility/bioavailability considerations of average daily exposure of lead

Route and consideration	Ignored (100%)	Default (30%) (μg/day)	Calculated (1–2%)
Soil dose	80	24	0.8–1.6
Produce dose	2.6	0.78	0.026–0.052
Water dose	0.14–5.31	0.42–1.59	0.42–1.59[b]
Ambient air dose	4.8–23.1	4.8–23.1[a]	4.8–23.1[b]
Sum	88–111	30–50	6–26

Ignored = does not consider bioaccessibility, default = Glorennec (2006), calculated = measured by sequential extraction.
[a] Default value for inhaled Pb is 100% bioavailable.
[b] Pathways of exposure not calculated by sequential extraction, default bioaccessibility values used.

The exposure ranges presented can be compared to a range of regulation benchmarks based on human-toxicological studies used in Europe. The Joint Executive Council on Food Additives, a collaboration of the World Health Organization and the Food and Agriculture Organization, determined that the tolerable daily intake (TDI), or the daily dose of Pb above which would trigger health and regulator concerns, is 3.5 μg/kg body weight/day, which, adjusted for the body weight of children age 2–6 years, is 42–70 μg/day (Glorennec, 2006; Baars et al., 2001; JECFA, 1999). The specific input parameters used in the exposure model, especially the percent of Pb considered bioavailable/bioaccessible, determines how exposure to Pb in Roxbury and Dorchester compares to the TDI range. In the most extreme case considered in the model, where bioavailabilty was considered 100%, exposure is well above the TDI. However, within the range of bioavailability considered reasonable (<30%), exposure would be within or below the TDI range. The elevated rates of BLL >10 μg/dL observed in children from Roxbury and Dorchester indicate that a fraction of the population is being exposed to Pb at levels that trigger health concerns. Therefore, it is likely that the calculation presented in this study is an underestimate of the exposure in the communities. It is possible that the addition of indoor exposure to the calculation would better describe the population.

4. Implications

Estimating the value of raised gardening beds in the Roxbury and Dorchester communities requires evaluation of many factors. The use of raised beds, as a remediation or soil intervention program, does not address the primary pathways of exposure of Pb (e.g., ingestion of soil) because raised beds cover/contain only a small percent of contaminated soil in a yard or garden. Raised beds reduce Pb exposure only by reducing the produce consumption pathway. Raised beds should be viewed as a limited exposure reduction method and should not be classified as a remediation technique. According to the exposure model presented, the average daily exposure of Pb in children could be reduced by 2–3%, or 1.8–3.3 μg/day if all produce was grown in raised beds rather than contaminated garden soil, which is the most liberal consideration of bioavailability. Although the dose of Pb from produce is greatly reduced when bioavailability or bioaccessibility is considered, it is likely that the relationship between BLL and Pb exposure is not linear, hence a small decrease in exposure could result in a more significant reduction in BLL (Ryan et al., 2004; Lanphear et al., 1998). The exposure calculation generated for this study indicates that urban gardening, from the perspective of exposure to Pb from the consumption of produce, can continue to be fostered in the communities, especially if measures are taken to reduce exposure from ingestion of soil. This study illustrates the importance of fine grain ($<100\,\mu$m) soil in exposure and recontamination of Pb and demonstrates that effective exposure reduction or remediation programs cannot be implemented without consideration of this factor.

The soil intervention program presented in Dixon et al. (2006) implemented ground cover and barriers, and soil Pb concentrations decreased by 1815 μg/g, and entryway dust Pb was reduced. In Aschengrau et al. (1994), the top 6 in. of soil were removed, and soil Pb decreased by 2060 μg/g and BLL in children decreased. These studies illustrate that *in situ* remediation can have a dramatic, immediate impact on Pb exposure. However, the cost range of soil intervention in Dixon et al. (2006) was 1095–5643 USD with an average cost of 2798 USD per yard. This economic factor makes ground cover and barriers impractical. In addition, social factors prevent the use of such remediation methods on a large scale in Roxbury, because community members would not be willing to give up their gardens in order to receive the soil intervention. Phytoremediation is an additional technique that has been used in urban areas, but this study shows that unamended phytoremediation is not a viable option for urban communities. The concentration of Pb absorbed in plant tissue presented in Table 2 is very low and urban non-profit environmental organizations should not invest resources in phytoremediation. Raised beds cost only 90 USD each, enable gardeners to continue growing produce, and introduce education and awareness into the community, which can have an important but unquantifiable impact on exposure reduction.

This study highlights the role of urban soil and historical Pb contamination as a current exposure pathway to children. The primary source of Pb to children in Roxbury and Dorchester is ingestion of Pb contaminated soil from historic Pb-based paint and leaded gasoline use. For example, Mielke and Reagan (1998) demonstrated that pulverized Pb-based paint in dust and soil was more important in exposure than intact Pb-based paint chips. Furthermore, it can be estimated that from 1970 to 2006, approximately 2.2 million metric tons of Pb have been emitted from the combustion of leaded and unleaded gasoline. An overwhelming majority of these emissions (92%) took place between 1970 and 1983 (US DOE, 2008; Filippelli et al., 2005; Pino et al., 2005; Shotyk et al., 2002; US EPA, 1996b). These results support the growing body of literature that illustrates the importance of soil as a pathway of exposure, and the need for practical soil remediation options in the urban environment.

Studies have speculated that recontamination should be considered when planning *in situ* remediation. Aschengrau et al. (1994) documented urban recontamination at sites that had received soil intervention, where 41% of yards at the 6–10 month survey and 53% of yards at the 18–22 month survey had Pb concentrations higher than the initial concentration immediately after remediation. It is likely that the wind-transport of Pb observed in this study occurs at many urban sites, and monitoring of Pb concentrations at remediated sites is necessary for extended periods of time. This study illustrates that wind-transported fine grain soil is the mechanism of recontamination, and that surrounding garden soil is the source of Pb involved in the recontamination. The recontamination of raised beds and excavation sites illustrates that remediation on a yard-by-yard scale is not effective in an urban community with regional Pb contamination.

Throughout this study, regulator benchmarks have been used to describe the community and the exposure to Pb within Roxbury and Dorchester. If the EPA action limit of 400 μg/g of Pb in residential soil were decreased by 25%, nearly 95% of the gardens tested in this study would merit regulatory concern. The CDC limit of 10 μg/dL for BLL has also been questioned since detrimental neurological affects have been observed at BLL $<10\,\mu$g/dL (Lanphear et al., 2005).

Maintenance of raised beds, excavated sites, and other remediated sites is required to reduce the influence of wind-transported contamination. Recommended maintenance for raised beds involves removing the top 3–5 cm of soil and replacing it with compost each year in order to keep Pb concentrations low. These measures are low-cost and practical for non-profits or community organizations to undertake. Gardens and raised beds in Roxbury and Dorchester will continue to be monitored annually to further quantify Pb mobilization. It is essential to understand the concentration and abundance of fine grain soil particles when attempting urban remediation, because these grains pose the greatest threat of exposure and recontamination. Future remediation schemes should combine minimizing soil ingestion with the reduction of wind-transported Pb on a garden-wide scale to have the greatest impact on Pb exposure. This study indicates that urban *in situ* Pb remediation requires continuous maintenance, and that for urban remediation and exposure reduction to be effective in the long term, maintenance needs to occur on the neighborhood-wide scale.

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