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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 $150 \pm 40 \,\mu\text{g/g}$ to an average of $336\,\mu\text{g/g}$ over 4 years. The percent distribution of lead in the fine grain soil ($<100\,\mu\text{m})$ 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.

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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 $1 \times 3 \,\mathrm{m}^2$, 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 $<\!100\,\mu\text{m}$; (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 m², 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 $50\,^{\circ}$ 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 10 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 sizefractionated (<44 um) 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: $20\,\mathrm{mL}$ of the first extracting agent, $1\,\mathrm{M}$ 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 $1\,\mathrm{h}$ at a speed of $40\,\mathrm{rpm}$.
- (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[®] LeadTrackTM method H2210 QNT Lead. Blanks for each extracting agent were prepared and

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