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Biosolids compost amendment for reducing soil lead hazards: a pilot study of Orgro® amendment and grass seeding in urban yards

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Abstract

In situ inactivation of soil Pb is an alternative to soil removal and replacement that has been demonstrated in recent years at industrial sites with hazardous soil Pb concentrations. Most children exposed to elevated soil Pb, however, reside in urban areas, and no government programs exist to remediate such soils unless an industrial source caused the contamination. Modern regulated biosolids composts have low Pb concentrations and low bioaccessible Pb fractions and can improve grass growth on urban soils. High Fe and P biosolids composts can reduce the bioavailability and bioaccessibility of soil Pb and can aid in establishing vegetation that would reduce soil transfer into homes. For these reasons, we conducted a field test of their use to reduce Pb bioaccessibility in urban soils in Baltimore, MD USA. We chose biosolids compost for its expected reduction in the bioaccessible Pb fraction of urban soils, ease of use by urban residents, and ability to beautify urban areas.

Nine urban yards with mean soil Pb concentrations >800 mg Pb kg^{-1} were selected and sampled at several distances from the house foundation before soil treatment. The soils were rototilled to 20 cm depth to prepare the sites, and resampled. The yards were then amended with 6–8 cm depth of Orgro® biosolids compost (110–180 dry t/ha) rich in Fe and P, mixed well by rototilling, and resampled. Kentucky bluegrass (*Poa pratensis*) was seeded and became well established. Soils were resampled 1 year later. At each sampling time, total soil Pb was measured using a modified U.S. EPA nitric acid hotplate digestion method (SW 846 Method 3050) and bioaccessible Pb fraction was measured using the Solubility/Bioaccesibility Research Consortium standard operating procedure with modifications, including the use of glycine-buffered HCl at pH 2.2. Samples of untreated soils were collected from each yard and mixed well to serve as controls for the Pb bioaccessibility of field treated soils over time independent of positional variance within yards.

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At 1-year post-treatment, grass cover was healthy and reductions in bioaccessible Pb concentrations compared to pre-tillage were 64% (from 1655 to 595 mg kg^{-1}) and 67% (from 1381 to 453 mg kg^{-1}) at the sampling lines closest to the houses. Little or no reduction in bioaccessible Pb concentration was observed at sampling lines more remote from the house that also had the lowest bioaccessible Pb concentrations at pre-tillage (620 and 436 mg kg^{-1}, respectively). For the control soils, changes over time in total Pb and bioaccessible Pb concentrations and the bioaccessible Pb fraction were insignificant. This study confirms the viability of in situ remediation of soils in urban areas where children are at risk of high Pb exposure from lead in paint, dust and soil.

Keywords: Soil lead; Lead hazard remediation; Soil amendment; In situ lead remediation; Field test

1. Introduction

Urban soils are commonly contaminated with Pb from multiple sources, principally automotive emissions and exterior paint (ATSDR, 1988). Although interior Pb sources predominate in exposure and risk compared to contaminated soil (Jacobs et al., 2002; U.S. HUD, 2001; Weitzman et al., 1993), high concentrations of Pb in soils presents a risk to young children who may ingest soil directly or may ingest house dust that has been enriched in Pb from soil sources. Examination of the pattern of Pb distribution in major cities has shown that inner-city soils have become generally contaminated from automotive and stack sources, while exterior paint can cause very high local enrichment of soil Pb, even exceeding 5% in the surface 2 cm depth (Chaney et al., 1984; Chaney and Ryan, 1994; Mielke et al., 2000).

Although it is possible to remove all contaminated soil and replace it with clean soil, the cost and degree of disturbance have made soil replacement prohibitive in most situations. Only grossly contaminated soil associated with housing adjacent to Superfund sites (e.g., smelters, mines, battery recycling smelters) has been subject to the soil replacement recommended by U.S. EPA and U.S. HUD for highly contaminated soils because this program can fund soil Pb remediation only for industrial point sources. A number of alternative methods to address soil Pb risk have been investigated to determine if less expensive methods including sandboxes, shrubs as barriers to humans, and raised boxes with clean soil could be used by homeowners, tenants and local groups to reduce community soil Pb risks (Hynes et al., 2001; U.S. EPA, 2001a; Litt et al., 2002). These are the types of interim control hazard reduction measures that are currently being used by local lead hazard control programs funded through grants from U.S. HUD.

Investigation has also identified two methods which can reliably reduce the bioavailability of soil Pb to animals fed the soils: (1) application of high levels of phosphate (phosphate fertilizer, rock phosphate, phosphoric acid, and bone phosphate sources) to promote formation of pyromorphite, a form of soil Pb with low solubility and low bioavailability (Ryan et al., 2001); and (2) application of biosolids compost products rich in phosphate and hydrous Fe oxides which both promote formation of pyromorphite and increase the strength of adsorption of Pb to ingested soil particles (Brown et al., 2003a). Biosolids composts rich in P and Fe also reduced bioavailability of Pb to rats in a feeding study of soil amendments (Ryan et al., 2004). Urban soils are often infertile or even Zn phytotoxic from Zn that co-contaminates with Pb (Chaney et al., 1984; Mielke et al., 1999, 2000), resulting in weak plant cover that is easily disturbed by play activities. Zn phytotoxicity is more severe in contaminated urban soils which are strongly acidic (Chaney et al., 1984) so remediation needs to prevent Zn phytotoxicity to minimize soil/dust Pb exposure for children.

Incorporation of composted biosolids rich in P and Fe offers a soil amendment method which can correct most soil infertility and metal toxicity problems of urban soils, provide the P and Fe which reduce bioavailability of soil Pb, and mix surface soil (usually richer in Pb) with subsurface soil materials. Although incorporating compost may dilute soil Pb somewhat, the principle effects are obtained by reducing bioavailability of the Pb and mixing surficial Pb with less contaminated subsurface soil.

It can also yield a fertile soil which will support a strong turfgrass cover that can further reduce children's ability to come in contact with the soil, potentially reducing the transfer of lead-contaminated soil into houses via tracking and blowing. Thus, the composted biosolids amendment approach has the potential to reduce risk from Pb in urban soils by reducing direct contact with bare soil, transfer of soil into the house, and the bioavailability of Pb in ingested soil.

Biosolids composts have been successfully used to reduce soil Pb hazards at several industrial sites and residential communities associated with smelters, including a lead smelter in Joplin, MO (Brown et al., 2004), a lead smelter and mine pile in Katowice, Silesia, Poland (Stuczynski et al., 2000), and multiple Superfund sites in the U.S. (Bunker Hill, ID and Palmerton, PA) (Brown et al., 2003b; U.S. EPA Remediation Technology Demonstration Forum Action Team website of the In-situ Inactivation and Ecosystem Restoration Team).

Bioavailability is determined by measuring the fraction of Pb that is absorbed following ingestion by a test animal. With the development of a chemical test of the potential bioavailability of soil Pb to animals (Ruby et al., 1993), the term bioaccessibility was introduced to avoid confusion with Pb bioavailability to test animals measured by tissue Pb residues from feeding soils. Lead bioaccessibility is assessed with a chemical extraction method shown to give results significantly correlated with bioavailability results from the feeding of Pb enriched soils to pigs and rats (Ruby et al., 1999; Ryan et al., 2004)). A laboratory-based study using lead-contaminated garden soil from Baltimore, MD showed reduced Pb bioaccessibility and bioavailability after amendment with biosolids composts, especially those rich in iron and phosphorous (Brown et al., 2003a). In that study, Baltimore composted biosolids (Orgro®) rich in Fe and P provided the greatest reductions in Pb bioavailability and Pb bioaccessibility in rats.

The ability of composted biosolids to reduce bioaccessible Pb, however, has not been field tested in an urban setting where children's exposure to Pb in residential soil can be particularly acute due to the presence of older lead-painted housing. This study, conducted in 2000–2001, investigated the effectiveness of in situ treatment by incorporation of composed biosolids and grass into contaminated urban residential soils to reduce lead hazards.

2. Materials and methods

2.1. Biosolids compost and soil amendment method

Eckology High Organic Compost (Orgro®) was selected for use as a soil amendment in this study. To create Orgro®, municipal biosolids (sewage sludge) produced by the Baltimore City wastewater treatment plant are composted with woodchips and sawdust at the Baltimore City composting facility. This biosolids compost contains approximately 1% nitrogen (USFilter, 2000a) and is moderately rich in Fe (mean=17.2 g kg^{-1}) and P (mean=16.4 g kg^{-1}) (personal communication, David Hill, USFilter, for the years 1999–2000).

Orgro® is approved by U.S. EPA and the Maryland Department of the Environment (MDE) for unlimited use in lawns and gardens as a soil conditioner/fertilizer with lime addition to enhance plant growth for establishment and maintenance of turfgrass, forage grasses, and crops (vegetable, nursery and field) and for production of sod and ornamentals and reclamation of disturbed and marginal lands (USFilter, 2000b). In addition, Orgo® is recommended by the USDA-Agriculture Research Service and the Agricultural Extension Service in Maryland and other states as a fertilizer and soil conditioner (Wright et al., 1998). During the 1990s, more than 350,000 cubic yards of Orgro® were sold for commercial and home use in the Baltimore region (personal communication, David Hill, USFilter, 2003).

MDE and U.S. EPA regulations limit metals concentrations in municipal biosolids and require monthly testing of compost by the producer (MDE COMAR 15.18.04). For the year preceding the study (1999), the average metals concentrations (mg kg^{-1} dry weight) for Orgro® biosolids in general were as follows: As=18, Cd=7, Cu=212, Pb=109, Hg=1, Mo=9, Ni=13, Se=8, Zn=440 (personal communication David Hill, USFilter, 2003). All the metals concentrations were well below the U.S. EPA Alternative Pollutant Limit (APL) for land application of biosolids in the U.S. (U.S. EPA, 1993) and met much lower levels recommended for use on farmland and gardens (Chaney et al., 2000). The APL provides risk-based limits. Most biosolids products have much lower concentrations of metals than the APL limits due to pretreatment and commonly meet much lower levels recommended for use on farmland and gardens (Chaney et al., 2000).

The manufacture of biosolids compost must achieve specified temperatures for a specified number of days to assure that pathogens are reduced to nondetectable levels. The compost is then "cured" or "matured" for several months after the completion of aerobic composting to assure that the product has stabilized sufficiently to prevent any short-term adverse effects on plant growth. When a high application rate is needed to achieve soil remediation goals, compost is especially appropriate because the organic N is released slowly which both limits nitrate leaching and provides adequate N for turfgrass for many years.

Fig. 1. Biosolids compost amendment process: (a) initial rototilling; (b) surface preparation; (c) biosolids compost application; (d) forming a 6–8 cm layer of biosolids compost; (e) rototilling to a depth of 20 cm; (f) emerging grass cover.

Among biosolids, only Class A (pasteurized to prevent pathogen risk to residents (MDE COMAR 15.18.04)) products (e.g., Orgro® composts) may be used to amend residential soils at the rates needed to attain reduced Pb bioaccessibility and bioavailability; application of Class A heat-dried biosolids fertilizer products supplies too much N for this use.

The Orgro® used in this study was mixed with 2.5% dolomitic limestone and packaged into 11.3 kg (moist weight as applied) plastic bags for transport to study sites. The average moisture content of Orgro® is 35% (personal communication, David Hill, USFilter, 2003). Prior to compost application at study sites, the soil was tilled to a depth of 20 cm using a heavy-duty rototiller. Orgro® compost was then applied evenly across the test area in a layer 6–8 cm thick. On average, 14.3 kg/m^2 dry weight equivalent (range 11–18 kg/m^2) of compost was applied (=143 t/ha dry weight, range 110–180 t/ha dry weight, limestone included). The Orgro® was then rototilled into the soil to a depth of 20 cm, and the site was seeded with Kentucky bluegrass (*Poa pratensis*). Additional seeding was performed on an as needed basis to attain good grass cover at the outset. The yards were not fenced or otherwise blocked off as part of the project; some yards however had pre-existing fences. The soil amendment process is summarized in Fig. 1. The improvement in grass cover is shown in Fig. 2. Biosolids compost application at study yards was performed during June–September 2000.

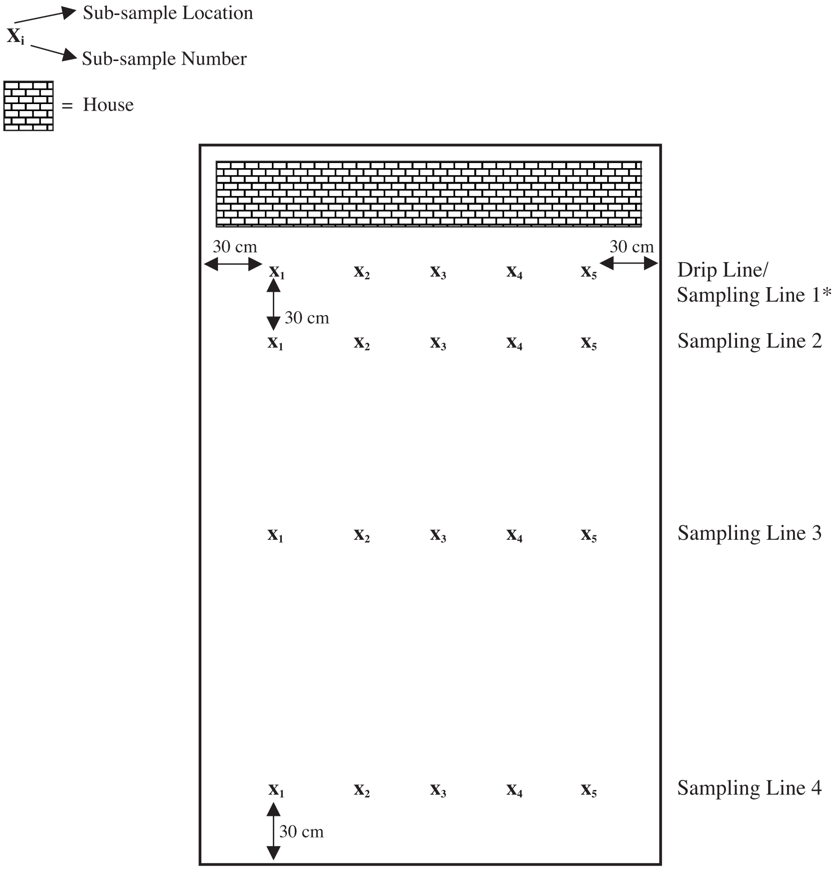
The participants were asked to maintain the grass cover. In one case, the study arranged for the grass to be maintained while the resident was away for an extended period of time. We provided all participants with educational brochures about residential and soil lead hazards and contact information for assistance, including the Maryland Agricultural Extension Service, the Coalition to End Childhood Lead Poisoning, and the Baltimore City Health Department. We also sent participants the results of the soil tests on their yards.

Fig. 2. Grass cover before and after biosolids amendment.

2.2. Study design

Collaborating community organizations and residents offered 25 residential yards in Baltimore, MD, USA, for a Pb survey to identify potential study sites for biosolids compost amendment. Eligibility criteria for yards were as follows: (1) size: <50 m^2; (2) overall mean dry soil lead concentration (Pb–S) >1000 mg kg^{-1} across all sampling lines per yard (Fig. 3); (3) Pb–S >700 mg kg^{-1} at most individual sampling lines (to allow for detection of change). Six of the twenty-five tested candidate yards fully met the selection criteria. Three other yards with overall mean yard Pb–S between 800 and 1000 mg kg^{-1} dry soil that otherwise met eligibility criteria were also included in the study because all but one of the composite probe samples from these yards exceeded U.S. EPA's standard of 400 mg kg^{-1} for lead in bare soil located in a child's play area (U.S. EPA, 2001b).

Three or four soil probe composite samples were collected from each of the 25 candidate yards at



* If house did not have a drip line, samples were collected from sampling lines, 2, 3 and 4.

Fig. 3. Plan for the collection of composite soil probe samples.

multiple sampling lines (Fig. 3) as follows: Line 1—the drip line (line closest to the house); Line 2 at a distance of 30 cm from the drip line; Line 3 at the middle of the yard; and Line 4 at a distance of 30 cm from the property boundary. Multiple sampling lines were selected because soil Pb concentrations were expected to vary with distance from the house. Each probe composite was collected from the top 2–3 cm of soil at five evenly distributed locations along each sampling line using a standard 2.5 cm diameter soil probe. Soil from each line was composited in the same labeled one-quart plastic Ziploc® bag for transport to the laboratory. Some houses did not have soil at the drip line due to concrete walkways and porches. In such cases, soil was collected from the other sampling lines.

2.2.1. Sample collection

For each of the nine study yards, soil probe composite samples were collected before, during and after the soil treatment process as follows: (1) before the initial rototilling; (2) after rototilling and prior to Orgro® application; (3) after Orgro® application and prior to seeding; and (4) at 1-year follow-up. At each sampling time, three or four soil probe composite samples were collected from each yard using the same sampling design and methods employed in the survey (Fig. 3). To minimize the potential for contamination of the 1-year follow-up samples by newly deposited lead-containing paint chips or dusts on the soil surface, the first 0.5 cm of topsoil from each subsample was removed with a disposable blade prior to transferring the sample into the plastic bag. Because the soil had been rototilled twice at treatment, the remainder of the soil sample was a valid representation of treatment effectiveness for mixed soil over time without the possible new surficial contamination from paint dust or other sources. This procedure made the 1-year samples comparable to those collected after the initial rototilling but prior to and after the Orgo application.

To assess changes in bioaccessible Pb in untreated soil over time, approximately 2 kg of bulk soil was collected from each yard prior to biosolids compost application using a disposable scoop. The bulk soil was collected from the top 2–3 cm of soil at locations between the soil probe sampling locations and close to each sampling line. The untreated bulk soil from each yard was mixed thoroughly and stored in a separate sealed 1-L Rubbermaid® plastic container in the laboratory for follow-up Pb testing 1 year later. This soil served as a control for change in bioaccessible Pb extraction during the experiment for untreated soils. In addition, a composite sample of the Orgro® applied to each yard was collected prior to its application to test for metal concentrations (As, Cd, Cr, Cu, Ni, Pb, Zn).

In order to assess Pb in yard dust that is most likely to be blown or tracked into houses, dust was collected before biosolids compost application from three locations along each sampling line (at the ends and at the middle of the sampling line) using a portable R&M Cyclone device (Farfel et al., 1994). The dust was collected for 2–3 s into pre-cleaned microwave digestion liners within an area defined by a 7.6×12.7 cm disposable template.

2.3. Sample preparation and laboratory analysis

Soil probe composite samples were dried and homogenized prior to digestion according to the U.S. EPA SW 846 Method 3050 (U.S. EPA, 1986a). First, the sample was manually broken-up while in the Ziploc® bag and then sieved through the 4.7-mm (#4) sieve to remove coarse particles and vegetation. The sieved portion of the sample was placed in the drying oven overnight at a temperature of 110±10 °C in a covered 100-mL Griffin beaker. The dried and cooled sample was homogenized using a porcelain mortar and pestle and sieved using a 2-mm (#10) sieve and placed in the drying oven for 24 h at 110±10 °C.

To obtain a representative sample of the bulk control soil, multiple small scoops of soil were collected from different portions of the sample container. The samples were dried and homogenized prior to analysis using the procedure described for soil probe samples.

A 0.5 g portion from each soil sample was digested for total Pb analysis using a modified U.S. EPA nitric acid hotplate digestion method SW 846 Method 3050 (U.S. EPA, 1986a). The modifications included an increase in the volume of reagents to 50 mL and not using hydrochloric acid. Another 0.5 g portion was digested for bioaccessible Pb analysis using the Solubility/Bioaccessibility Research Consortium standard operational procedure (SOP) (Ruby, 1999) with modifications, including the use of 0.5 g of soil, 50 mL final dilution volume of the digestate, and adjustment to pH 2.2 (see Brown et al., 2003b for reasons to use this pH).

Bioaccessible Pb was measured at pH 2.2 rather than at pH 1.5 as in the original paper by Ruby et al. (1996, 1999). The pH 2.2 was used because Brown et al. (2003a,b) and Ryan et al. (2004) reported that extraction at pH 1.5 gave results which did not detect significant reduction in bioaccessible Pb due to phosphate or biosolids treatment despite the fact that rat, pig and human feeding tests showed significant reduction in bioavailable Pb (Ryan et al., 2004). Hence, pH 1.5 bioaccessible assays are not appropriate for urban soils.

Cyclone dust samples were digested for total Pb analysis using a CEM Model MDS-2100 microwave digestion apparatus according to method SW 846 Method 3051 (U.S. EPA, 1986b) and for bioaccessible Pb using the modified SOP described above. The following reagents were used: J.T. Baker nitric acid (trace metal grade, concentrated, 69.9–70%); Mallinckrodt AR hydrogen peroxide (30% reagent ACS); J.T. Baker glycine; and deionized water.

All soil digestates were analyzed for lead at the Kennedy Krieger Institute's Trace Metals Laboratory (TML) using an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, Perkin Elmer Optima 2000) according to U.S. EPA Method SW 846 6010 (U.S. EPA, 1986c). The following standard solutions were used for calibration (mg Pb kg^{-1}): 0.05; 0.25; 1.0; 5.0; 10.0; 20.0. Standard solutions were prepared in 10% nitric acid from Perkin Elmer

Table 1
Pretreatment soil characteristics by site

Site	Area (m^2)	pH	Clay (%)	Fe (g kg^{-1})	Mn (mg kg^{-1})	Bray-1 P (mg kg^{-1})	Organic matter (%)
1	50	7.4	12.5	12.6	140	100	5.1
2	12	6.6	11.3	14.5	150	530	7.8
3	10	6.2	12.8	13.7	170	180	15.9
4	13	6.3	18.9	31.8	390	97	15.5
5	30	6.5	12.4	21.0	320	67	7.7
6	41	6.2	14.7	19.6	410	860	11.4
7	16	6.3	16.0	21.1	350	190	11.1
8	35	5.8	12.9	23.0	420	440	6.9
9	30	5.5	14.4	29.0	450	120	10.2
Mean	26.3	6.3	14.0	20.7	311	287	10.2
S.D.	14.3	0.53	2.3	6.6	124	269	3.8
Median	30	6.3	12.9	21.0	350	180	10.2
Max	50	7.4	18.9	31.8	450	860	15.9
Min	10	5.5	11.3	12.6	140	67	5.1

Pure Atomic Spectroscopy Standard (1000 mg Pb kg^{-1}). Quality control samples included in the analytical run were prepared using GFS Chemicals Lead Standard Solution (100 mg Pb kg^{-1}). The calculated instrumental detection limit (IDL) was estimated to be 0.05 mg Pb kg^{-1}; the calculated limit of detection (LOD) was approximately 1 mg Pb kg^{-1} soil.

2.4. Multi-metal analysis of Orgro

To verify metal concentrations in Orgro®, a sample of the Orgro composite from each site was digested according to the U.S. EPA Method SW846 3050. The digestates were analyzed for As, Cd, Cr, Cu, Ni, Pb and Zn using ICP-OES (Perkin-Elmer Optima-2000) according to U.S. EPA Method SW846 6010.

2.5. Soil characteristics analysis

Standard soil analysis methods were used to examine the test soils. To measure pH, slurries of 10 g air-dried soil: 20 mL deionized water were prepared and pH measured after 1 h using a combination electrode. Clay content was measured by the pipette method (Gee and Bauder, 1986). Hot nitric acid soluble metals were analyzed by ICP-AES using U.S. EPA method 3050 (U.S. EPA, 1986a,c). Soil plant-available phosphate was measured by the Bray-1 procedure (Bray and Kurtz, 1945), a method commonly used on medium-textured, medium-pH soils. Soil organic matter was determined by mass loss on ignition overnight at 480 °C (Davies, 1974).

2.6. Quality control

Over 250 quality control (QC) samples of various types were included in the study. Each batch of 24 samples prepared for total Pb analysis included the following types of QC samples: standard reference material (SRM) spikes [0.5 g of NIST SRM 2710 (Montana Soil, 5532 mg kg^{-1} Pb); 0.5 g of NIST SRM 2711 (Montana Soil, 1162 mg kg^{-1} Pb); and 0.5 g of NIST SRM 1646a (Estuarine Sediment, 11.7 mg kg^{-1} Pb)] and stock solution spike and spike duplicate samples prepared with 0.5 mL of Perkin Elmer Pure Atomic Spectroscopy Standard (1000 mg kg^{-1} Pb). For bioaccessible Pb analysis, each batch included 0.5 g of NIST SRM 2711, a stock solution spike, a spike duplicate, and a reagent blank. Similar types of quality control samples were added to the sample preparation batches used for the multi-metal analysis of Orgro®.

Based on performance in the QC program, the laboratory measurements had good precision and accuracy. For total Pb analysis, recoveries for the

Table 2
Concentrations (mg kg^{-1} dry weight) of regulated elements in samples of the Orgro composted biosolids applied to study sites

Site	As	Cd	Cr	Cu	Ni	Zn	Total Pb	Bioaccessible Pb
1	2	1	46	313	14	736	100	14
2	2	1	54	317	16	775	106	7
3	2	1	68	284	11	664	237	6
4	2	1	50	296	13	717	105	17
5	2	1	49	317	13	762	102	15
6	2	1	51	321	15	776	101	21
7	2	1	52	330	14	812	109	16
8	2	1	46	292	14	692	96	16
9	2	1	45	287	13	687	95	15
Mean	2	1	51	306	14	736	117	14
S.D.	0	0	7	17	1	49	45	5
Median	2	1	50	313	14	736	102	15
Max	2	1	68	330	16	812	237	21
Min	2	1	45	284	11	664	95	6
EPA APL[a]	41	39	1300	1500	420	2800	300	–

[a] APL=Alternative Pollutant Limit (U.S. EPA, 1993).

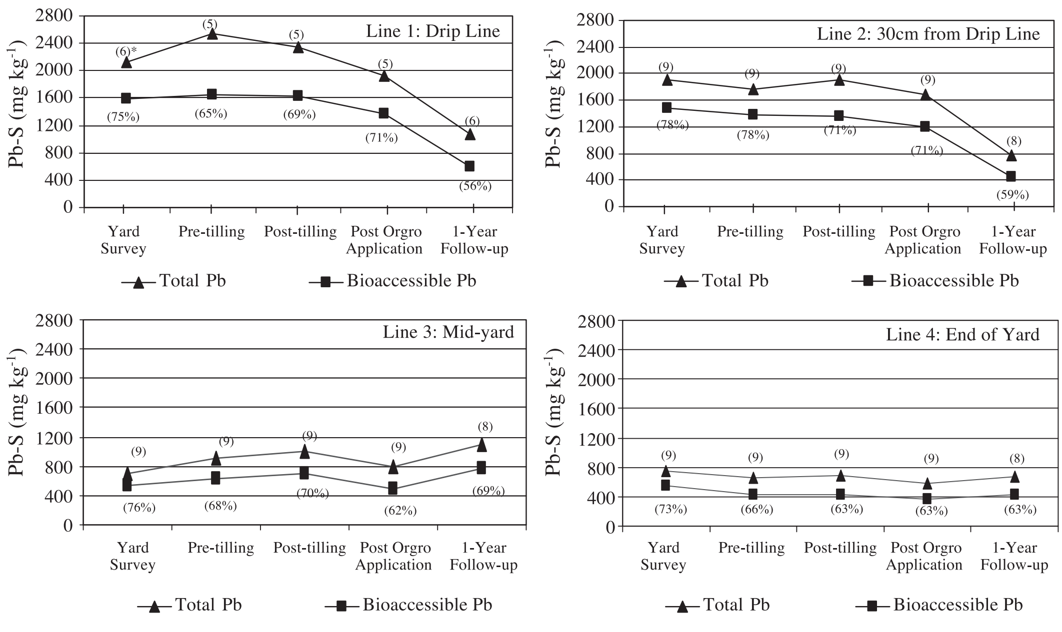
various types of spiked QC samples were all within $\pm$30% range of “true” value, except for SRM1646a (Estuarine Sediment, 11.7 mg kg^{-1} Pb). The mean percent recovery for stock solution spikes was 92% (n=24, S.D.=7, range 70–100%) and for spike duplicates was 94% (n=23, S.D.=5, range 86–112%). The mean percent recovery for the SRM spike samples was 89% (n=24, S.D.=5, range 82–100%) for SRM 2710; 91% (n=24, S.D.=6, range 81–102%) for SRM 2711; and 66% (n=23, S.D.=31, range 24–148%) for SRM1646a. For bioaccessible Pb, the mean percent recovery for stock solution spikes was 87% (n=27, S.D.=8, range 69–100%) and for spike duplicates was 88% (n=27, S.D.=6, range 81–102%). The mean percent recovery for SRM 2711 was 93% (n=24, S.D.=7, range 75–104%). Reagent blanks showed extraneous Pb contamination of the samples to be on average below the instrumental detection limit (IDL).

2.7. Data analysis

Given that total Pb concentrations varied with distance from the houses, data analysis was done separately for each sampling line to facilitate interpretation and presentation of results. Paired t-tests were used to assess the statistical significance of changes in Pb concentrations between sampling times. Prior to this analysis, the Pb concentration data were transformed using the natural logarithm. Bioaccessible Pb fraction (%) is the bioaccessible Pb concentration divided by the total Pb concentration and then multiplied by 100. Change in bioaccessible Pb fraction was calculated as bioaccessible Pb fraction before biosolids compost amendment minus bioaccessible Pb fraction at 1 year after biosolids compost amendment divided by the bioaccessible Pb fraction before biosolids compost amendment.

3. Results

A total of 126 soil probe composite samples were collected at the different stages of biosolids compost treatment, including 30 soil probe composite samples at 1-year follow-up. One yard could not be sampled at 1 year. Nine bulk soil samples and 18 cyclone dust samples were collected before biosolids compost amendment was conducted.



* (n) – the number of yards with both total and bioaccessible lead measurements.

Fig. 4. Mean total Pb and bioaccessible Pb concentration (Pb–S, mg kg^{-1}, n*) and bioaccessible Pb fraction (%) over time by sampling line.

3.1. Site/soil characteristics

Study yards ranged in size from 10 to 50 m^2 (Table 1). The soil had sandy loam or loam texture (clay content 11–19%) and other characteristics typical of urban soils (Table 1). Study yards had relatively high levels of organic matter (range 5–16%) compared to the low levels (1–2%) of organic matter common in agricultural fields. The pH of the soil ranged from 5.5 to 7.4 and was <7.0 for all yards except one. Iron concentrations ranged from 12.6 to 31.8 g kg^{-1} (mean=20.7 g kg^{-1}) and manganese concentrations ranged from 140 to 450 mg kg^{-1} (mean=311 mg kg^{-1}). Plant-available phosphorous concentrations ranged from 67 to 860 mg kg^{-1}(mean=287 mg kg^{-1}). When this soil P test exceeds 30 mg kg^{-1}, no increase in plant yield would be expected from addition of P fertilizer; levels in excess of 150 mg

Table 3
Total and bioaccessible soil Pb concentrations (mg kg^{-1}) over time by sampling line

Line	Yard survey	Pre-tilling	Post-tilling	Post-Orgro application	One-year follow-up
	Mean (Range) S.D. *n*	Mean (Range) S.D. *n*	Mean (Range) S.D. *n*	Mean (Range) S.D. *n*	Mean (Range) S.D. *n*
Bioaccessible Pb					
1	1598 (1063–2598) 739 6	1655 (942–2147) 559 5	1622 (905–2232) 575 5	1373 (380–2052) 746 5	595 (229–1464) 439 6
2	1478 (498–3888) 1220 9	1381 (572–3347) 877 9	1358 (570–2719) 835 9	1198 (228–2979) 962 9	453 (155–975) 246 8
3	538 (187–928) 275 9	620 (140–1336) 360 9	695 (241–1603) 475 9	496 (115–1342) 369 9	764 (73–2389) 767 8
4	547 (105–951) 277 9	436 (88–874) 258 9	436 (80–857) 251 9	367 (74–779) 199 9	425 (86–817) 238 8
Total Pb					
1	2125 (1206–3611) 1149 6	2534 (1448–4266) 1121 5	2344 (1390–3608) 865 5	1932 (780–2548) 818 5	1064 (635–2584) 751 6
2	1879 (667–4671) 1514 9	1767 (775–3865) 984 9	1908 (857–3985) 1130 9	1686 (558–4194) 1160 9	770 (248–1568) 402 8
3	707 (291–1178) 322 9	913 (258–2003) 511 9	995 (364–2182) 611 9	799 (265–1916) 485 9	1106 (161–2736) 862 8
4	749 (163–1291) 371 9	658 (179–1273) 364 9	691 (167–1183) 380 9	584 (177–932) 236 9	675 (135–1435) 419 8

kg^{-1} are considered undesirable because of eutrophication risk from eroded soil. However, the vegetation cover supported by biosolids compost amendments can reduce the risk of soil erosion.

3.2. Orgro metal concentrations

Metal concentrations of composite samples of Orgro® applied to study yards are displayed in Table 2. The concentrations of all metals tested in each of the Orgro® samples were well below the U.S. EPA Alternative Pollutant Limit (APL) for land application of biosolids in the U.S. (U.S. EPA, 1993) and were similar to or lower than the average metals concentrations for Orgro® biosolids in general for the year preceding the study. The bioaccessible Pb concentration of the Orgro applied across the nine yards was low (mean=14 mg kg^{-1}), and the bioaccessible Pb fraction averaged 14%.

3.3. Soil lead

At the time of the initial survey, mean soil Pb concentrations ranged from 800 to 2003 mg kg^{-1} among the group of nine study yards. The mean pre-tilling yard soil Pb concentrations had a similar range of values (479–2034 mg kg^{-1}). Fig. 4 displays mean soil Pb concentrations (total and bioaccessible Pb) by sampling line and sampling time for the group of study yards, the bioaccessible Pb fraction and the number of yards. Descriptive statistics for total and bioaccessible Pb concentrations are displayed in Table 3. At pre-tilling, the mean Pb concentrations were higher at the drip line (Line 1: 2534 mg kg^{-1}) and at Line 2 (1767 mg kg^{-1}) than at the middle of the yard (913 mg kg^{-1}) and at the edge of the yard most distant from the house (658 mg kg^{-1}).

The initial rototilling of soil prior to compost application resulted in little, if any, change in mean soil Pb concentration at Lines 1–4 (Table 3). Compost application was associated with a decrease in mean soil Pb concentration of 12–20% at each sampling line (Table 3). This change was consistent with a dilution effect from the use of a low Pb soil amendment. At 1-year post-treatment, soil Pb concentrations compared to those before treatment were on average decreased for Lines 1 and 2, increased for Line 3, and approximately the same for Line 4 (Table 3).

On average, the bioaccessible Pb fraction before biosolids compost amendment ranged from 65% to 78% of total Pb across the four sampling lines (Fig. 4). Changes in bioaccessible Pb concentrations were minimal from pre-tilling to post-tilling. Orgro® application was associated with a 12–30% decrease in bioaccessible Pb concentration at each sampling line immediately after treatment (Table 3). At 1-year follow-up, reductions in bioaccessible Pb concentrations compared to those before soil treatment (pre-tilling) were 64% for Line 1 (from 1655 to 595 mg kg^{-1}) and 67% for Line 2 (from 1381 to 453 mg kg^{-1}). At 1-year follow-up, little or no reduction was found in mean bioaccessible Pb concentrations compared to pre-tillage at Line 3 (from 620 to 764 mg kg^{-1}) and Line 4 (from 436 to 425 mg kg^{-1}).

The percentage reduction in bioaccessible fraction from pre-tilling soil sampling to 1-year follow-up was calculated (see data analysis section) to be 14% at the drip line (Line 1: from 65% to 56%), 24% at Line 2 (from 78% to 59%) and minimal or absent at Line 3 (middle) (from 68% to 69%) and Line 4 (from 66% to 63%)(Fig. 4). Changes in total and bioaccessible Pb concentrations of the control bulk soil (soil collected before amendment which was then mixed to reduce sample variance) were minimal, and the reduction in bioaccessible Pb fraction was small (8%) (Table 4).

Table 4
Total and bioaccessible (BA) Pb (mg kg^{-1}) in untreated (control) bulk soil collected before Orgro amendment and tested at time of Orgro amendment and at 1-year follow-up

Site	Before Orgro amendment			One-year follow-up		
	Total Pb	BA Pb	(%BA)	Total Pb	BA Pb	(%BA)
1	1245	861	(69)	1229	832	(68)
2	1204	743	(62)	1136	–	–
3	1874	1209	(65)	1635	–	–
4	1197	866	(72)	1570	990	(63)
5	1764	1121	(64)	1877	877	(47)
6	1117	518	(46)	1118	601	(54)
7	1198	784	(65)	1547	–	
8	505	309	(61)	617	399	(65)
9	375	207	(55)	458	238	(52)
Mean*	1034	647	(62)	1145	656	(57)
S.D.	515	359	(7.7)	534	295	(7.6)

* Based on Sites 1, 4, 5, 6, 8 and 9 that were tested before Orgro amendment and at 1-year follow-up. For all nine sites, the mean total Pb was 1164 mg kg^{-1} and the mean BA Pb was 735 mg kg^{-1} (%BA=63) before Orgro amendment, and at 1-year follow-up, the mean total Pb was 1243 mg kg^{-1}.

3.4. Surface dust

Pb concentrations for the eight yards with cyclone-based dust data (mean=1020 mg kg^{-1}, range 507–1536 mg kg^{-1}) and the bioaccessible fractions (mean=62%, range 41–72%) were similar to the corresponding values for the same eight yards based on the soil probe samples (mean soil Pb concentration=1242 mg kg^{-1}, range 375–1874 mg kg^{-1}; mean bioaccessible Pb fraction=67%, range 55–76%).

4. Discussion

Approximately 7% of U.S. dwellings (6.46 million units) have soil Pb concentrations above U.S. EPA and U.S. HUD standards (400 mg kg^{-1} for bare play area soil and 1200 mg kg^{-1} for bare soil in the rest of the yard) (Jacobs et al., 2002). Biosolids compost application is one of a number of methods (e.g., sandboxes and raised beds with clean soil) that can be used individually or in combination to reduce soil lead hazards. For example, biosolids compost application could be combined with the use of barriers including raised beds and gravel to provide additional protection. The latter two methods without biosolids compost application were tested by others (Safer Yards Project, 2002; Binns et al., 2004). We chose biosolids compost amendment for its: (a) potential to yield a fertile soil that will support a strong turfgrass cover that can further reduce children's ability to come in direct contact with bare urban soil and the transfer of soil into the home; (b) expected reduction in the bioavailable Pb fraction; and (c) ease of use by urban residents and community organizations. Our community partners (see Acknowledgments) believed that this method was feasible and sustainable at a community level by residents and community organizations. In fact, the Greening Committee of one of our partner organizations (Historic East Baltimore Community Action Coalition) had applied biosolids compost to yards and gardens as part of their previous local beautification efforts before the study.

Prior to Orgro® amendment, the soil Pb concentrations were highest at the drip line and declined with distance from the house. The bioaccessible Pb fractions were more uniform (range 65–78% across yards) and not atypical based on other studies (Ruby et al., 1999; Ryan et al., 2004). Although grass cover was present prior to soil treatment, the density of cover was low and uneven (Fig. 2).

This longitudinal pilot study shows that in situ amendment of lead-containing yard soil with Orgro® biosolids compost followed by grass seeding is associated with a significant reduction in bioaccessible Pb fraction and the development of a healthy turfgrass cover at 1-year follow-up (Figs. 2 and 4). Mixing surface soil with subsurface soil provides one step in reducing soil Pb risk to children when surface soil has a higher total Pb concentration than subsurface soil. By sampling after rototilling the yards thoroughly, the testing over time measured reduction in bioaccessibility due to the Orgro® compost treatment. Clearly, the large decreases in bioaccessible Pb at Lines 1 (57%) and 2 (62%) at 1 year compared to the much smaller decreases immediately post-Orgro® application reflect the inactivation of lead and a real change in bioaccessibility after Orgro® application and not just a dilution effect.

The treatment effect in terms of reduction in bioaccessible fraction was limited to sampling locations closest to the house where Pb concentrations were higher (Lines 1 and 2 vs. Lines 3 and 4). One would expect a greater likelihood of reduction in bioaccessible Pb fraction with higher soil Pb concentrations due, in part, to a likely greater presence of more recently deposited anthropogenic Pb that has not had time to react with the soil. We have no specific explanation for the lack of reduction in bioaccessible Pb fraction at Lines 3 and 4. This may have been due to the presence of more stable forms of lead in the lower Pb concentration soil (Lines 3 and 4) compared to the higher Pb concentration soil (Lines 1 and 2). The magnitude of the reduction in bioaccessible Pb concentration (64% at Line 1 and 67% at Line 2) and the percentage reduction in bioaccessible Pb fraction from pretreatment to 1-year follow-up at Lines 1 and 2 was within the expected range of reduction based on past studies (Brown et al., 2003a,b). As expected, measurement of bioaccessible Pb in the control soil samples (mixed and stored in plastic containers without Orgro® treatment) showed only a small reduction of 8% in the bioaccessible Pb fraction 1 year later that was not statistically significant.

Logically, it would be expected that the container of control soil should have no change in bioaccessible Pb because this Pb had remained in bioaccessible forms for decades in the field and had not been amended with biosolids compost.

The lack of decrease in total Pb concentrations across sampling lines after the initial rototilling suggests that tilling alone may not be effective in reducing surface soil Pb concentrations. This would be the case, for example, when yard soil Pb concentrations are more or less uniform to tilling depth as found by others (personal communication, Helen Binns, M.D., 2003). Tilling could reduce soil Pb when the surface deposited soil had not been mixed into soil by previous tilling.

The reductions found in soil Pb concentrations across lines after the soil treatment was completed were expected due to low Pb content (mean=117 mg kg^{-1}) of the biosolids compost applied. The reductions in bioaccessible Pb associated with Orgro® application were also consistent with a dilution effect from the use of a soil amendment with a low bioaccessible Pb fraction (Table 1). The reduction in total Pb concentrations at Lines 1 and 2 at 1-year follow-up while unexpected was likely due to sampling variability and possibly increased mass of organic matter (grass roots, etc.) in the samples.

The biosolids compost application rate for soil remediation at industrial sites is typically a one-time application of 50–250 t/ha plus limestone at 50–100 t/ha or higher (Chaney et al., 2000; Brown et al., 2003a). In this study, we applied Orgro® at rates ranging from 11 to 18 kg/m^2 across yards (equivalent to 110–180 t/ha, limestone included).

The estimates of total Pb and bioaccessible Pb concentrations of the yard surface dust before soil treatment based on cyclone dust samples were similar to corresponding estimates based on the soil probe samples. The cyclone-based estimates might represent the material that is potentially blown and tracked into houses. Future studies should investigate changes in the bioaccessible fraction of surface soil and house dust following biosolids compost amendment.

The generalizability of the findings is limited by the pilot nature of this study and to the rates of biosolids compost application used. On the other hand, the characteristics of the soil in the study yards are likely to be typical of yard soils in other urban communities and thus similar reductions in the bioaccessible Pb fraction would be expected to occur. Moreover, this study was limited to 1-year follow-up. We would expect further reduction in bioaccessible fraction over time. The field test at Joplin, MO, has been followed for about 3 years since treatments were incorporated. There was a reduction over time in bioavailable Pb, but only a small reduction of bioaccessible Pb years after treatment compared to the drop in bioaccessible Pb and bioavailable Pb at the time of treatment (Ryan et al., 2004). This small reduction of bioaccessible Pb over time after the sharp drop at the time of treatment may have resulted from an artifact of bioaccessibility testing in that the chemical reactions to make pyromorphite occur during the extraction as well as in the field (Scheckel and Ryan, 2003). Additionally, reduction in soil Pb bioavailability to humans was considerably greater than the reduction measured using rats or pigs (Ryan et al., 2004). In that study, only phosphate-treated and control soils were fed to humans, with a reduction in soil Pb bioavailability of 69% due to the treatments. Reductions measured by bioaccessible methods, or by pigs or rats were smaller than the change in bioavailability to humans (Ryan et al., 2004).

Future research is needed to investigate the extent and timing of further reductions in bioaccessible Pb and bioaccessible Pb fraction in urban yard soil beyond 1-year follow-up. It is also important to assess the ability of residents to maintain a grass cover that would minimize children's access to soil over time. The degree of adoption and effectiveness of use of biosolids compost amendments by residents and community groups as part of individual and larger organized greening and lead hazard reduction efforts also warrants future investigation. Future research should also include investigation of composts of cow manure or yard debris which are rich in P and Fe as an alternative to biosolids composts as these may have lower concentrations of metals (Chaney et al., 2000) and be more readily accepted by residents and community organizations. We recognize that NRC (2001) advised application of newer risk assessment methodologies for EPA's regulatory limits for biosolids products. Readers should keep in mind that recommended attainable levels for biosolids composts are far lower than regulatory limits (Chaney et al., 2000) and that the product tested here was a low-metal compost recommended for use in lawns and gardens.

Lastly, the cost of biosolids compost application and grass seeding ($150–350 per study site yard) is based on the costs incurred to treat the study sites using one local landscaping company. The study site yards varied in size as reported in Table 1. These costs obviously do not reflect the costs of residents' performing the work themselves, and they might not be generalizable to other urban areas due to differences in labor and material costs. The cost of the Orgro® applied is not included because the material was donated. The current retail price for Orgro® is \$30 yd^{-3} (\$39 m^{-3}). For yards with 20 m^2 area, 1.5 m^3 of Orgro® would supply a 7.5 cm layer of compost and cost $59 based on the retail price of Orgro®.

Many studies have confirmed extensive Pb contamination of urban soils. As noted earlier, Federal programs to remediate Pb-contaminated soils are restricted to remediation of Superfund industrial sources and the interim control measures being unemployed by U.S. HUD funded lead Hazard Control Programs. With the evidence of reduction of soil Pb risk in this study, parents and communities have a simple low-cost in situ Pb inactivation technology available to reduce risk to their children.

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