We present high spatial-resolution mapping of soil lead (Pb) concentrations in a small-urban residential setting. X-ray fluorescence was used to measure soil Pb at 170 properties in the City Park neighborhood of Appleton, Wisconsin. Greater than two-thirds of soil samples collected from drip lines contained more than 400 μg g⁻¹ of Pb, and one third exceeded 1200 μg g⁻¹. Soils adjacent to homes built before 1960 contained significantly higher Pb levels than those near younger homes. Three front yard locations (drip line, mid-yard, and terrace) were sampled at 71 properties. A general decline in soil Pb with increasing distance from the house was observed. Detailed sampling of individual homes within a single residential block revealed Pb-contaminated soil radiating outward from homes in all directions, creating a “bulls-eye” pattern. Approximately 40% of yard space exceeded concentrations of 400 μg g⁻¹. These patterns of contamination are consistent with Pb paint as the main contributor of Pb to soil. This has important implications because spatial distribution of Pb contamination is fundamentally different if paint, rather than automobile exhaust, is the primary source. Selective sequential extraction analyses suggest that roughly half of the soil Pb resides in chemically reactive and bioavailable phases. The extent and persistence of soil Pb, the resurgence of home gardening, and the serious health consequences of Pb ingestion argue for attention to this problem, not just in dense urban centers but also in smaller urban settings across the country.

SOIL LEAD (Pb) contamination is a significant and widespread public and environmental health concern. There is a rich research history and vast literature on Pb in the urban environment, but many questions critical to human and environmental health remain unanswered. Most studies have focused on large urban centers, leaving smaller communities unexplored. Likewise, most studies have used a low-density sampling scheme, which tends to cover large areas at the expense of detail. In the work presented here, we hope to extend our current understanding of the extent and source of Pb contamination in smaller urban environments through high spatial-resolution sampling.

History of Lead Use in the Residential Environment

The two main sources of Pb in the urban residential environment are combustion of leaded gasoline and deterioration of exterior paints (Burgoon et al., 1995; Mielke et al., 2008; Laidlaw and Filippelli, 2008). A variety of Pb minerals and compounds were added to paints, most notably Pb-carbonates, chromates, and oxides. These compounds added pigmentation and improved the durability and flexibility of the paint. The use of Pb as a paint additive peaked in the 1920s, before advocacy and legislative efforts during the post-WWII housing boom led to a steep decline in the use of Pb in house paints (Laidlaw and Filippelli, 2008). These efforts became final with the complete ban of Pb in house paints by the U.S. Consumer Product Safety Commission in 1978. Clark et al. (1991) estimated that between 1932 and 1980, nearly 7 million tons of Pb were used in the production of white paint in the United States.

Although the use of Pb in paints declined during the middle of the 20th century, the anthropogenic distribution of Pb due to gasoline consumption rose during the same period, mirroring an increase in automobile usage (Mielke, 1999; Laidlaw and Filippelli, 2008). Lead was added to gasoline as tetraethyl-Pb, an anti-knock agent. The release of Pb through the burning of gasoline peaked in the 1970s before being phased out by the USEPA in the 1980s and eventually being banned for use in on-road vehicles in 1996. By that time, approximately 4 to 5 million tons of Pb had been introduced into the environment through combustion of leaded gas (Mielke, 1994).
Impacts of Soil Lead on Human Health

Soil Pb levels (SLL) and blood Pb levels (BLL) in urban populations are positively correlated (Bickel, 2010; Zahran et al., 2010). In 2001, the USEPA set Pb standards for bare soils at 400 μg g⁻¹ for play areas and 1200 μg g⁻¹ for the remainder of the yard (USEPA, 2001). In recognition of the clear connection between SLL and BLL, several states have established even lower SLL standards (e.g., 250 μg g⁻¹ for Wisconsin and 100 μg g⁻¹ for Minnesota). Despite BLL dropping on average nationally, elevated levels persist in urban areas largely due to the legacy of Pb contamination of urban soils (Filippelli and Laidlaw, 2010).

The most notable human health risk associated with Pb is that it is a potent neurotoxin, particularly to children and developing fetuses. Chronic exposure to soils with elevated Pb concentrations leads to elevated BLL (Laidlaw et al., 2005; Filippelli and Laidlaw, 2010; Bickel, 2010), which in turn causes impaired cognitive development, emotional and behavioral problems, and numerous other health issues, including deficits in IQ, attention-related problems, and impaired academic performance (Chiiodo et al., 2007; Nigg et al., 2010). Many recent studies have found developmental effects associated with BLL below 10 μg dl⁻¹ (Canfield et al., 2003; Surkan et al., 2007). Because there are no known safe BLL and because harmful neurological effects of Pb are likely irreversible, the U.S. Centers for Disease Control recently lowered the initial screening reference level for children to 5 μg dl⁻¹.

Of particular concern regarding elevated SLL is the increased prevalence of home gardening, both as a hobby and as a means of saving money through household food production. According to a report from the National Gardening Association (2011), one third of U.S. households reported growing at least some vegetables in a home garden. As more households use their yards for gardening, the issue of elevated SLL takes on new significance. Consumption of vegetables grown in soil with elevated Pb concentrations introduces new vectors for the uptake of Pb by humans (Motto et al., 1970; Mielke et al., 1983; Chaney et al., 1994; Finster et al., 2004; Clark et al., 2006; Clark et al., 2008).

To improve the understanding of this environmental issue, researchers have created geochemical maps of a number of cities displaying SLL. The specific goals and methodologies of these studies have varied (Johnson and Ander, 2008), yet the results have shown consistent patterns. Urban areas have the highest SLL, with concentrations decreasing markedly into the less densely populated and more newly developed suburbs (Mielke, 1999; Laidlaw et al., 2005). The age of buildings in the city centers (with many dating to the peak of Pb-paint usage), the density of structures, and heavy automobile traffic have helped to maintain elevated SLL in urban areas, which in turn have kept BLL elevated (Filippelli and Laidlaw, 2010). A number of studies (e.g., Mielke et al., 1983; Laidlaw, 2001; Filippelli et al., 2005; Mielke et al., 2008) have suggested that gasoline-derived Pb is the primary source of elevated SLL in urban areas, whereas others argue that paint plays a dominant role (Linton et al., 1980; Jacobs, 1995).

This study differs from previous works in two important ways. First, our study site is a small city rather than a major urban area. Second, we conduct a high spatial-resolution survey of SLL, focusing first on a single residential neighborhood and then on a single city block. We consider three primary questions: (i) How does the spatial distribution of SLL in a small-urban area compare with those in a larger city? (ii) Can we ascertain the likely sources of soil Pb, and, if so, how do sources of SLL in a smaller city compare with those in larger cities? (iii) What are the possible health concerns associated with SLL in this small-urban setting?

In a small city such as Appleton, Wisconsin (with a population of approximately 75,000 in 2010), the same primary anthropogenic stressors (Pb-based paint and leaded gasoline) are present that have led to elevated SLL in major urban areas. The relative inputs from these stressors may differ, particularly as a result of differences in traffic density. Detailed Pb mapping, along with a series of selective sequential extractions (SSEs), helps illuminate how Pb is distributed in soil at the neighborhood, block, and individual yard scales—scales that are critically important to homeowners for understanding patterns of contamination, associated health risks, and mitigation strategies.

Materials and Methods

Soil Sampling

Soil samples were collected from the drip line of 170 properties in Appleton, Wisconsin between October 2007 and October 2011 (Fig. 1). The sampling was concentrated in and around the historic City Park district and Lawrence University. These are some of the oldest parts of Appleton (est. 1857), where roughly 40% of homes predate the turn of the 20th century, and 85% of homes were built before 1950. The vast majority of test properties were single-family residences and low-density, duplex-style apartments on 0.1 to 0.05 ha (quarter-acre to eighth-acre) lots. A few institutional parcels from the university also were sampled. There is some industry (paper mills) at the northern boundary of the study area and across the river to the south; however, these industrial sites were not included in the study. We assumed the sampling area to be representative of older neighborhoods in other urban and suburban communities.

Samples were taken via a 2.5-cm-diameter soil punch to a depth of 7.5 cm. Several cores were taken at each location, producing sample volumes of approximately 500 mL. Characteristics of the property, including the exterior type (e.g., brick, stucco, aluminum siding), the condition of the exterior, the type of ground cover present, and the distance of sample sites from the road were recorded in the field. Other housing characteristics (e.g., property value, lot size, and property age) were gleaned from the City of Appleton’s plat maps. A 71-sample subset of the 170 properties was tested at two additional locations within each property: (i) at the midpoint of the terrace (the grassy strip between the curb and sidewalk) and (ii) on the lawn at the midpoint between the curb and the house.

During the summer of 2011, one square block was chosen for detailed analysis (Fig. 1a and 1b). A total of 310 cores were taken along transects radiating from the eight homes on this block, all of which were built between 1890 and 1905. One home had a painted stucco exterior, and the remaining homes had painted wooden exteriors. An interpolated surface of Pb concentration was created using the inverse distance-weighting scheme in ArcGIS 10.1 Spatial Analyst (ESRI, 2012). Samples from two transects perpendicular to College Avenue (the main
thoroughfare through town) were also evaluated (Fig. 1a and 1c). The transects began in the terrace and moved away from the road at 3- to 5-m intervals up to 87 m from the road.

Soil samples were placed in plastic bags and transported to the lab where roots and surface vegetation were removed (surface vegetation consisted of turf grass). The soil was then homogenized by hand and dried in a 105°C oven for 8 h. A portion of the dried soil was ground to <63 μm with mortar and pestle.

**Portable X-ray Fluorescence**

Ten grams of dried, crushed, and sieved soil was packed in a plastic cup with a mylar film base and analyzed using an Innov-X series pXRF in a laboratory stand. Analyses were performed using the instrument’s “soil mode,” with each sample being analyzed for 60 s. The NIST standards 2710 and 2711 were analyzed during each analysis session, and our analyses measured within 3.3% (SD, 1.8%) of reported values. Approximately 5% of our samples were chosen at random and run as duplicates. Samples with concentrations >100 μg g⁻¹ Pb showed strong reproducibility, with replicate analyses averaging within 8.4% of the initial reading (SD, 6.8%). Replicates of samples whose initial Pb concentrations were <100 μg g⁻¹ were, on average, within 16 μg g⁻¹ (SD, 16 μg g⁻¹) of the original measurement. Although the precision on the lower concentration samples is not as strong as for the higher concentration samples, this sensitivity does not affect our interpretations.

**Selective Sequential Extractions**

We targeted the following five fractions in our soils: (i) exchangeable, (ii) mild-acid soluble (e.g., carbonate-bound), (iii) reducible, (iv) oxidizable, and (v) residual, using a procedure based on protocols developed by Tessier et al. (1979) and La Force and Fendorf (2000). Seventeen samples were studied using the SSE procedure. To assure that Pb concentrations would be above levels of detection throughout the SSE analysis, only samples with 1000 μg g⁻¹ or greater were used. Samples used for SSE analysis came from soils collected near five different buildings and represented samples ranging from the drip line to mid-yard. All but one of the samples were analyzed in duplicate.

Extractions began with 1 g of soil added to a 50-mL polypropylene centrifuge tube. The exchangeable fraction was removed with 8 mL of a 1 mol L⁻¹ MgCl₂ (buffered to pH 7.0) and agitated for 1 h at room temperature. The remaining soil was
treated with 20 mL of 1 mol L\(^{-1}\) NaOAc-HOAc (buffered to pH 5.0 with acetic acid) and agitated for 5 h at room temperature to remove the carbonate-bound fraction. The reducible fraction was then targeted with 20 mL of 0.04 mol L\(^{-1}\) NaOAc-H\(_2\)O (buffered to pH 2.0 with HNO\(_3\)) and agitated for 2 h at room temperature. The temperature was then adjusted to 85°C, an additional 3 mL of 30% H\(_2\)O\(_2\) was added to each sample, and the samples were agitated for an additional 3 h. The samples were removed from the shaker bath and allowed to cool for approximately 1 h. They were then diluted with 20 mL of double-deionized water and agitated for 30 min at room temperature.

After each extraction, the samples were centrifuged at 10,000 rpm, and the extracted solution was decanted for analysis. After the final extraction, the remaining soil was retained to analyze the residual metal content. A Pb-free synthetic soil (a mixture of quartz, dolomite, kaolinite, and hematite, along with organic compost, all analyzed and found to be free of Pb) was prepared and run through the SSE procedure. The extractants from these synthetic soils were spiked with Pb for use as our calibration standards.

X-ray Fluorescence Analyses of Selective Sequential Extraction Samples

The solutions from the SSE procedure were analyzed by dripping the liquid sample onto a cellulose filter (Rigaku micro carry) and then dried for XRF analyses (Igarashi et al., 2000). For the carbonate-bound and reducible fractions, 100 µL of solution were added to each filter. The Pb concentrations in the exchangeable and oxidizable fractions were lower, so 200 µL of solution were dripped onto these filters in two additions of 100 µL. Samples were analyzed using a Rigaku ZSX Mini II under vacuum. The remaining soil after all four extractions was dried and reanalyzed on the pXRF using the methods described above to determine to residual Pb fraction.

Results

Distribution of Lead in Urban Soils

Soil samples adjacent to 170 homes in a roughly 10 × 12 block area of Appleton, Wisconsin, yielded Pb concentrations between 47 and 32,483 µg g\(^{-1}\). This wide range is in sharp contrast to nine samples taken in green spaces (e.g., park areas, campus greens, and school yards), which contained Pb concentrations between 39 and 142 µg g\(^{-1}\); we consider these concentrations as background SLL (Table 1). These values are comparable to nationwide background soil Pb concentrations (Shacklette and Boerngen, 1984). One third of residential samples (n = 57) exceeded the USEPA limit of 1200 µg g\(^{-1}\) for bare soil in yards, and more than two thirds (n = 115) exceeded the USEPA limit of 400 µg g\(^{-1}\) for bare soil in play areas. The southwestern portion of the study area had a concentration of high SLL, likely due to the age of these properties (Fig. 1a). Lawrence University was established in 1847, and many of the homes around the college are of this era. Despite the large variability in the Pb values, there was a highly significant difference (p < 0.001) in the geometric means of Pb concentrations on properties that were built before and after 1960 (Fig. 2a; Table 1). Differences in SLL related to exterior type were mixed (Table 1). Wooden exteriors returned mean concentrations that were significantly higher than buildings with exteriors made of stucco, stone, or brick but not higher than those with vinyl or aluminum siding. At first this seems unusual, considering that aluminum and vinyl are not typically painted. However, most homes with these exteriors were built before the introduction of aluminum siding (1940s) and vinyl (late 1950s), suggesting that these exterior types simply covered or replaced wooden exteriors. All building types, including stone and brick, can return high SLL (Table 1) because even these homes have windows and other trim work that have been painted.

For the 71-sample subset (where three locations at each property were sampled), the geometric mean of the drip line sample was significantly higher than the mid-lawn sites (p < 0.002), and the mid-lawn sites were higher than the terrace (p < 0.001) (Fig. 2b; Table 1). Four mid-lawn samples were above 1200 µg g\(^{-1}\), and 22 exceeded 400 µg g\(^{-1}\). At most sites, there was a systematic decline in SLL from the home to the road. At 11 sites, however, the terrace returned higher concentrations than the mid-lawn sampling point, and at six sites the mid-lawn SLL were greater than at the drip line. City plat records and aerial photographs from 1938 indicate that 8 of 11 of the terrace > mid-lawn sites and 5 of 6 of the mid-lawn > drip line sites were on parcels where structures have been built or rebuilt since 1978 (when the use of Pb in paint was banned by the USEPA). On some of these properties, the mid-lawn region was over 1000 µg g\(^{-1}\), whereas the sample adjacent to the home was at or near background Pb levels (<100 µg g\(^{-1}\)). These unusual

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>n</th>
<th>Geometric mean</th>
<th>Geometric SD</th>
<th>Min.</th>
<th>Max.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Green space</td>
<td>9</td>
<td>93</td>
<td>0.2408</td>
<td>39</td>
<td>142</td>
</tr>
<tr>
<td>Drip line</td>
<td>71</td>
<td>590</td>
<td>0.4826</td>
<td>47</td>
<td>13,244</td>
</tr>
<tr>
<td>Mid-lawn</td>
<td>71</td>
<td>273</td>
<td>0.3594</td>
<td>57</td>
<td>3,412</td>
</tr>
<tr>
<td>Terrace</td>
<td>71</td>
<td>135</td>
<td>0.3500</td>
<td>23</td>
<td>1,309</td>
</tr>
<tr>
<td>Before 1960</td>
<td>145</td>
<td>917</td>
<td>0.5171</td>
<td>79</td>
<td>32,483</td>
</tr>
<tr>
<td>After 1960</td>
<td>25</td>
<td>231</td>
<td>0.3141</td>
<td>47</td>
<td>755</td>
</tr>
<tr>
<td>Wood</td>
<td>79</td>
<td>1052</td>
<td>0.5218</td>
<td>79</td>
<td>16,264</td>
</tr>
<tr>
<td>Aluminum</td>
<td>9</td>
<td>879</td>
<td>0.3510</td>
<td>255</td>
<td>2,182</td>
</tr>
<tr>
<td>Vinyl</td>
<td>56</td>
<td>656</td>
<td>0.5331</td>
<td>47</td>
<td>32,843</td>
</tr>
<tr>
<td>Stucco</td>
<td>6</td>
<td>703</td>
<td>0.4460</td>
<td>297</td>
<td>3,177</td>
</tr>
<tr>
<td>Brick/stone</td>
<td>20</td>
<td>268</td>
<td>0.4418</td>
<td>128</td>
<td>4,880</td>
</tr>
</tbody>
</table>
trends are likely attributable to legacy Pb associated with an old structure that was removed and new fill that was introduced. Inconsistencies in the trends also may be due to soil disturbance and/or redistribution in association with landscaping around homes or with Pb loading of the terrace from nearby busy streets.

The detailed study block reinforces the basic trend of declining SLL away from structures (Fig. 1b). Each home created a “bullseye” pattern, with high concentrations of Pb near the home on all sides. Nearly half the samples (n = 154) exceeded 400 \( \text{mg g}^{-1} \), and this zone extended several meters into the yard all around the home (Fig. 1b). Lead levels above 1200 \( \text{mg g}^{-1} \) are common close to homes but are rare in the yard unless there are adjacent homes or outbuildings. Where homes are close to one another (see, for example, the two homes in the northwest corner of Fig. 1b), the concentrations tend to remain relatively high even in the mid-lawn area. Soil Pb levels near outbuildings are also often elevated. The detailed-block study suggests that a large proportion of soil in older neighborhoods is likely contaminated. Based on the interpolated data, approximately 40% of the yard area has SLL >400 \( \text{mg g}^{-1} \).

The average SLL for the College Avenue transects were not significantly different from other green spaces in the broader study area. However, there was a gradual decline in Pb concentration with increasing distance from the road (Fig. 1c). The first two sampling points were relatively close to the road and sidewalk, both of which have been replaced in the past 10 yr, making it likely that fill has been tested rather than original soil. Excluding those fill samples, SLL were roughly 50% higher near the road than those more than 20 m away from the road.

### Selective Sequential Extraction Results

The results of our SSE are summarized in Table 2 and Fig. 3. The carbonate and reducible fractions dominated the Pb-residence, with each averaging around 40% of the total. The exchangeable fraction accounted for just under 10% of the Pb-residence, as did the oxidizable fraction. On average, only 1% of the Pb in the samples remained throughout the extraction procedure and was measured as residual Pb content.

#### Error Analysis

All samples for SSE analysis were prepared as duplicates. One sample was lost during the SSE procedure, but all others were analyzed as duplicates. Duplicate samples exhibited excellent reproducibility (Table 3).

To measure the accuracy of the SSE procedure, the “percent recovery” must be measured. This value compares the sum of all Pb concentrations reported from each extraction step with the initial Pb concentration in the soil. One sample returned a 250% recovery (along with its duplicate, which reported 248%). This particular sample had one of the lowest initial Pb concentrations of any of our samples. It is not unexpected to see recoveries significantly over 100%, particularly for lower-concentration samples. This is at least in part because concentrations of Pb in individual extractions approach the limits of the instrument’s sensitivity. The smaller the total concentration, the more profound the impact of even relatively small absolute errors on the total percent error. If we remove this pair of samples from the data set, the average recovery drops to 113% (maximum, 179%; SD, 27%).

### Discussion

#### Source of Soil Lead in Residential Neighborhoods

With the exception of communities that house incinerators, mines, or smelters, Pb in urban soils is primarily a combination of

### Table 2. Results of selective sequential extraction analyses. Values indicate percent recovery.

<table>
<thead>
<tr>
<th>Fraction</th>
<th>Avg.</th>
<th>Max.</th>
<th>Min.</th>
<th>SD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exchangeable, %</td>
<td>9</td>
<td>18</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Carbonate, %</td>
<td>39</td>
<td>61</td>
<td>13</td>
<td>13</td>
</tr>
<tr>
<td>Reducible, %</td>
<td>43</td>
<td>64</td>
<td>27</td>
<td>8</td>
</tr>
<tr>
<td>Oxidizable, %</td>
<td>8</td>
<td>18</td>
<td>1</td>
<td>5</td>
</tr>
<tr>
<td>Residual, %</td>
<td>1</td>
<td>3</td>
<td>0</td>
<td>1</td>
</tr>
</tbody>
</table>
Table 3. Comparison of the differences between results of duplicate samples for selective sequential extraction analyses.

<table>
<thead>
<tr>
<th>Fraction</th>
<th>Avg.</th>
<th>Max.</th>
<th>Min.</th>
<th>SD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exchangeable, %</td>
<td>1</td>
<td>3</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>Carbonate, %</td>
<td>3</td>
<td>11</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>Reducible, %</td>
<td>3</td>
<td>11</td>
<td>0</td>
<td>4</td>
</tr>
<tr>
<td>Oxidizable, %</td>
<td>2</td>
<td>11</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>Residual, %</td>
<td>0</td>
<td>2</td>
<td>0</td>
<td>1</td>
</tr>
</tbody>
</table>

Fig. 3. Lead fractions found in soil samples as determined by selective sequential extraction. The bar indicates the mean value, and the error bars indicate ±1 SD.

Our study, along with previous studies (e.g., USEPA, 2000; Litt et al., 2002), indicates that SLL commonly decline with distance from the home (Fig. 2b). We suggest that the reason for this pattern is that exterior paint is the main source of soil Pb. Other researchers, however, have noted that structures can act as a barrier to the atmospheric transport of fine Pb–laden dust particles from leaded fuel combustion and from resuspension of these particles near roadways (Rolfe et al., 1977; Mielke et al., 1983; Olszowy et al., 1995; Mielke et al., 2008; Laidlaw and Filippelli, 2008). In effect, structures trap the suspended Pb particulates. For example, Mielke et al. (1983) noted higher SLL near brick buildings in inner-city Baltimore than near painted wooden structures in the suburbs. They attributed this difference to vehicle miles traveled and to the barrier effect; that study, as well as most others, only considers front yard areas where trapping would be most effective. A good test of the barrier effect would be to compare street-facing front yards with back- and side yards. Our detailed study block with eight houses (Fig. 1b) provides us with this opportunity, and the basic spatial pattern does not suggest any preferential trapping of Pb on the street side. In fact, our results show no statistical difference between the drip line means in the front yard and the other sides of homes, strongly suggesting that the atmospheric trapping effect is minimal in this setting (Fig. 1b and 4).

The trapping effect should also result in a U-shaped pattern of contamination where SLL are highly elevated near the road and somewhat elevated near the house, with local minima in the yard (Rolfe et al., 1977; Olszowy et al., 1995). Of 71 sites where we have transects, we see the general U shape in only 11, with eight of those sites containing structures that have been rebuilt. The remaining three sites were older wooden structures situated along a moderately busy roadway (~7,500 vehicles per day [VPD]), so additional Pb loading from traffic was possible. However, even at these three sites the concentrations near the homes were two to three times higher than those near the roadway. The detailed study block does not exhibit a U-shaped contamination pattern. Instead, it shows homes and outbuildings as discrete sources of Pb, with concentrations dropping radially outward from the structures (Fig. 1b).

The Contribution of Vehicle Traffic to Lead Distribution

Previous work shows a strong correlation between distance from roadways and SLL (Motto et al., 1970; LaBelle et al., 1987; Laidlaw, 2001; Filippelli et al., 2005). We show a similar, albeit less dramatic, trend in transects along College Avenue (Fig. 1c). Ignoring the fill samples and taking into account the reduced precision of the instrument at low Pb concentrations, the SLL near the road were roughly twice those >20 m away from the road, where they approached background levels (Fig. 1c). These concentrations near the road, though elevated, were still lower than 200 μg g⁻¹ and were much lower than values reported near homes and mid-lawn regions.

Less clear is the correlation between traffic volume and SLL. In the Twin Cities and New Orleans, Mielke et al. (1984/85, 2008, respectively) found that when comparing structures of a similar age, higher SLL are found in highly trafficked inner city areas. Lau and Wong (1982) found traffic volume and SLL highly correlated in Hong Kong. However, Motto et al. (1970) compared SLL within 7.6 m (25 ft) of nine roadways in New Jersey varying in traffic volume from 12,800 to 54,700 VPD, and these data exhibited no relation (r² = 0.021) between traffic volume and SLL (Fig. 5a). Furthermore, SLL from highway shoulders near Detroit, Michigan showed only a weak correlation (r² = 0.57) at very high traffic volumes (Fig. 5a), although only five sites were considered in that study (Howard and Sova, 1993). Simple correlations such as these are confounded by several important factors, including the nature of the traffic (highway vs. surface street stop-and-go), the slope of the roadway, the proportion of truck traffic, and the speed limit. In the relatively low-traffic community of Appleton, we see no correlation between VPD and SLL. Neither samples from the drip line (r² = 0.039) nor those from the terrace (r² = 0.032) returned significant trends (Fig. 5b).
Appleton's current traffic volumes are low compared with urban areas that have been the focus of previous Pb studies. The busiest street in our study, College Avenue, has a volume of approximately 15,000 VPD. This is smaller than volumes of major roads in Indianapolis (~20,000 VPD) and New Orleans (~25,000 VPD). Also, aside from College Avenue, no roads exceed 10,000 VPD, and many streets are in the hundreds of VPD (Fig. 1a). Residential neighborhoods also enjoy lower vehicle speeds and less truck traffic, minimizing the resuspension of fine Pb-laden dust. It would seem that smaller urban centers such as Appleton never carried the volume of cars necessary to greatly raise soil-Pb concentrations. The fact that our data are very similar to the "suburban" pattern identified by Filippelli et al. (2005) lends additional support to our assertion that traffic density in Appleton was insufficient to greatly elevate SLL.

Soil Lead Risk Revisited

Among the most common pathways for Pb into the human body are inhalation of dust particles and ingestion of soil. These routes of exposure can be effectively minimized by covering bare soil areas with mulch or turf ground cover. Urban gardening, however, can reopen these pathways and create additional opportunities for Pb to enter the body. Direct ingestion of Pb-contaminated soils adhering to fruits and vegetables and ingestion of Pb contained in edible bodies (primarily roots and dark leafy greens) have been implicated as potential vectors of Pb uptake (Motto et al., 1970; Finster et al., 2004; Clark et al., 2006). These studies also show a strong correlation between SLL and Pb in garden fruits and vegetables. The high-density sampling performed in this study indicates that the zone of contamination extends well beyond the house and in all directions (Fig. 1b). In neighborhoods composed of 0.1-ha (quarter-acre) and smaller lots, as much as 40% of the yard area may contain concentrations of Pb exceeding the USEPA limit of 400 μg g⁻¹ (yellows and reds in Fig. 1b). Proximity to outbuildings and other homes can further elevate SLL. Because areas close to homes and outbuildings are prime sites for vegetable gardens and because backyards are often
used as play areas, understanding the distribution, extent, and bioavailability of soil contamination is all the more important for homeowners and tenants using these outdoor spaces.

The results of our SSE work indicate that nearly half of the Pb was bound in exchangeable and carbonate fractions (Fig. 3). These fractions are considered to be the most highly bioavailable (through human ingestion) forms of Pb (USEPA, 2007; Ruby et al., 1999). Assessing the bioavailability of Pb within the reducible fraction, which accounted for (on average) more than 40% of the Pb in our SSE results, is less straightforward a process. The reducible fraction will likely include original paint phases, such as Pb-chromates (Pb yellow) and PbO₂ (red Pb), as well as secondary Pb phases, including Pb bound to Fe- and Mn-oxyhydroxides. Of these, Pb bound to Mn-oxyhydroxides and PbO₂ are highly bioavailable forms of Pb, whereas Pb-chromates and Pb bound to Fe-oxyhydroxides are more moderately bioavailable (USEPA, 2007; Ruby et al., 1999). The oxidizable fraction includes, on average, <10% of the Pb in the soils studied. As with the reducible fraction, one cannot clearly determine the bioavailability of Pb removed in the oxidizable fraction. This extraction will target Pb bound to organic molecules, which would likely be quite bioavailable, but it would also target Pb bound in sulfides such as galena, which is among the least bioavailable forms of Pb (USEPA, 2007; Ruby et al., 1999). Only a trace amount of the Pb in our samples persisted in the soil through the full SSE procedure and was measured in the residual fraction (i.e., the fraction with the lowest bioavailability). Despite uncertainties associated with the Pb extracted in the reducible and oxidizable fractions, it is clear that a large proportion of Pb in our analyzed soil resides in highly bioavailable forms.

Our study area is not exceptional, and the trends we report here are likely to be similar in smaller urban residential communities across the country. We show that the best predictor of contamination is the age of the structure, with those built before 1960 returning significantly higher SLL (Fig. 2a; Table 1). The City of Appleton has roughly 75,000 residents and 25,000 parcels of land. Approximately 51% of these parcels contain structures built before 1960, a proportion very similar to the age of the national housing stock (U.S. Census Bureau, 2011), which suggests that a large number of residences in Appleton may have contaminated soil. If one extrapolates our results to other small cities and towns across the country, then a large portion of the nation’s housing stock built before 1960 is likely to have soils containing elevated levels of Pb. Such an extrapolation seems justified based on large-scale federal studies. For example, a 1990 U.S. Department of Housing and Urban Development report estimated that 76% of homes built before 1960 have measurable Pb levels in exterior paint. Other national studies have estimated that 83% of the pre-1980 housing stock contains Pb paint (U.S. Department of Housing and Urban Development, 1990, 1995) and that 23% of these households have soil Pb concentrations >400 μg g⁻¹ (USEPA, 1996). Although we have presented here only a case study, these nationwide studies suggest that the trends we have observed are likely representative of communities of similar ages and sizes throughout the United States.

The most highly contaminated areas in these smaller cities are likely to be concentrated in older parts of the community, leading to the familiar “bulls-eye” pattern observed in this and in other studies (e.g., Mielke et al., 1999; Laidlaw and Filippelli, 2008), but tracts and neighborhoods of sufficient age located outside the urban core will also have elevated SLL. Even homes with exteriors that are not traditionally painted (e.g., brick and stone) can return values that exceed USEPA standards, presumably due to window and other trim paint (Linton et al. (1980) (Table 1). Although a deteriorated exterior may correlate with high soil Pb levels, properties with exteriors in good condition may still exhibit extensive contamination due to the property’s history and due to the use of unsafe restoration techniques. Moreover, homes with Pb exterior paint will continue to act as sources of soil Pb, contributing to, and potentially worsening, the problem over time unless exteriors are rehabilitated properly.

The health benefits of urban gardening and active child play must be balanced with measures to reduce exposure to contaminated soils. The work here suggests that in small-urban residential communities, Pb-contaminated soil can exist around all sides of the structure and extend quite far into the yard. Proposed gardening and play sites should be sampled thoroughly because remediation strategies (e.g., phytoremediation and soil amendments) are of limited efficacy (Clark et al., 2008; Giammar et al., 2008). The best practice is to remove the soil, apply a clean soil cap, or separate the garden and/or play areas from contaminated soils (Mielke et al., 2011). If lot conditions dictate the location of garden spaces on contaminated soils, then the advice that Finster et al. (2004) and Witzling et al. (2011) provide to urban gardeners should be followed: in particular, the use of raised beds (with semipermeable barriers separating existing soil from clean fill), the avoidance of root crops and dark-leaved greens, the use of mulch to prevent rain splash and dust accumulation on fruiting bodies, and thoroughly washing all edible portions. Ensuring that play areas are turfed is a low-cost way of reducing the hazards of exposure and soil resuspension.

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