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Potential human inhalation exposure to soil contaminants in urban gardens on brownfields sites: A breath of fresh air?

Joseph J. Weeks[1,3] | Ganga M. Hettiarachchi[1] | Eduardo Santos[1] | John Tatarko[1,2]

[1] Dep. of Agronomy, Kansas State Univ., Manhattan, KS 66506, USA

[2] USDA-ARS Rangeland Resources and Systems Research Unit, 2150 Centre Ave., Bldg. D, Suite 200, Fort Collins, CO 80526, USA

[3] Field Data Solutions, Indigo Agriculture, 500 Rutherford Ave., Charlestown, MA 02129, USA

Correspondence
Ganga M. Hettiarachchi, Dep. of Agronomy, Kansas State Univ., Manhattan, KS 66506, USA.
Email: ganga@ksu.edu

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Abstract

Urban gardening has been experiencing increased popularity around the world. Many urban gardens are located on sites that may be contaminated by trace elements or organic compounds due to previous use. The three main exposure pathways to the human body for soil contaminants are (a) ingestion of soil directly, (b) consumption of produce containing or superficially contaminated with a contaminant, (c) and inhalation of soil dust. The first two modes have received much attention; however, the contribution of the inhalation route has not been investigated adequately. Two inhalation risk studies were carried out in urban gardens located in Kansas City, MO, by collecting dust while 25-m^2 plots were rototilled. Microclimatic variables were monitored, and total inhalable dust mass was determined using a personal sampling train including a small pump and air filter. Soil lead (Pb) concentration was assessed at both sites. For Study 1, particle size distribution of collected particles was estimated through analysis of scanning electron microscope images of filters. Little dust was collected at either site. Most particles captured, however, appeared to be <4 μm in diameter. The amount of dust emitted was correlated with soil moisture. Tilling reduced soil aggregate size and blended soil, resulting in a more homogeneous distribution of Pb. Dust inhalation while tilling is likely not a major Pb exposure risk for gardeners, but given the preponderance of very small particles in what was captured, care should be taken to prevent dust from entering the respiratory system.

1 | INTRODUCTION

A rapidly growing percentage of the world population is moving into urban areas, and the availability of fresh, nutritious produce in densely populated, low-income areas has become a cause for concern (U.S. Census Bureau, 2020). "Food deserts," as they often have been called (i.e., places where grocery stores, if present, lack the means to obtain and/or sell quality fruits and vegetables in an economically efficient manner), are appearing in cities around the world. Those unable to travel out of these areas are left reliant upon fast food establishments and convenience stores to satisfy their daily caloric requirements, if not their nutritional needs. One solution to improve access to fresh produce is the implementation of urban community gardening programs. Not only have these programs helped to alleviate malnourishment in economically disadvantaged areas, they also have served to improve social relations, creating networks that further foster improved community development (Hynes & Genevieve, 2004).

Abbreviations: PTFE, polytetrafluoroethylene.

A major issue hindering the rapid implementation of urban gardens in many areas is the concern of growing food in soil that may not be safe. University and government research is making information available to address these misgivings (e.g., Brown et al., 2016; USEPA, 2011). Assessing the risks associated with urban gardening is still lacking in certain areas, however. In many communities, lead (Pb) contamination remains the primary culprit. This heavy metal, which is an artifact of anthropogenic activity, has entered the soil primarily as a result of the prolonged use of leaded gasoline in the internal combustion engine and leaded paint applied inside and outside of many buildings from approximately the 1920s until 1996 and 1978 in the United States, respectively (Binstock et al., 2008; Markey et al, 2008; McBride et al., 2011; Schwarz et al., 2012). Human exposure has been linked to a variety of health maladies, leading the U.S. Centers for Disease Control and Prevention to recommend action at blood Pb levels exceeding 5 μg dl^{-1} (CDC, 2012). Children are especially susceptible to the toxic effects of Pb, which include developmental impairment, due to their high frequency of hand to mouth activity and increased intestinal absorption capabilities. Adults are not immune though. Delta-aminolevulinic acid dehydratase inhibition has been documented at low Pb concentration, causing reduced heme production (Sake et al., 2000). Heme is a critical component of iron-containing proteins (e.g., hemoglobin) essential for human health (Hettiarachchi & Pierzynski, 2004; McDowell, 2003). Direct ingestion and inhalation of Pb-containing soil particles as well as consumption of plant material that has absorbed the contaminant are thought to be the three main pathways for the element to enter the bloodstream (Wortman & Lovell, 2014). Mielke et al. (1997) demonstrated a direct correlation between high soil test levels and increased blood Pb concentrations in both children and adults. Understanding the mechanisms facilitating these pathways is essential to ensuring the implementation of safe urban gardening programs.

Of the three exposure pathways listed above, inhalation exposure has yet to be thoroughly characterized. Due to the extreme variability that exists both within and between garden sites, applicable information from collected data is sparse. Although a plethora of instruments have been developed to measure concentrations and total mass transport of suspended, respirable dust, none possesses the ability to measure concentrations in real time while still retaining large enough samples to allow for elemental and size distribution analysis. Kasumba et al. (2011) was successful in obtaining dust profiles resulting from disking operations of a cotton field in New Mexico, but elemental analysis of the dust itself was beyond the scope of their study. No research to date has been found in which the inhalation exposure of urban gardeners has been quantified on brownfield sites.

Core Ideas

- The inhalation exposure route of soil Pb for urban gardeners has not been adequately investigated.
- Rototilling activity-based inhalation risk studies were conducted in two urban garden sites.
- Short-term dust exposure from rototilling is not a major Pb exposure pathway for the person conducting the rototilling.
- Rototilling under dry conditions presents the greatest risk.

The goal of this investigation was to quantify the inhalation exposure risk to urban gardeners working on brownfield sites. The experiments were carried out in two urban soils on sites historically used for gardening located in Kansas City, MO. The experiment consisted of collecting dust while 5-m by 5-m garden plots were rototilled. Microclimatic variables were monitored, and total inhalable dust mass was collected using a personal sampling train developed for asbestos exposure studies. Rototilling is considered in this study as a proxy for inhalation exposure, although it is not the only mechanism by which inhalation exposure could occur in urban gardens.

2 | MATERIALS AND METHODS

2.1 | Experimental design

Two small dust collection studies modeled after the USEPA standard operating procedure for activity-based air sampling for asbestos were conducted on loam soils to quantify the amount of dust that a gardener may be subjected to while rototilling a garden plot (USEPA, 2007). The first study (Study 1) was conducted in 2014 at the Washington Wheatley Community Garden on Montgall Avenue in Kansas City, MO (39.082495° N, 94.551380° W). The soil texture was a loam (26% sand, 48% silt, 26% clay). Four 5 m by 5 m plots were established, and rototilling was conducted on each, alternating between an east–west and north–south orientation, three separate times for 60 min during Tillage Event 1 and 45 min for Events 2 and 3 over the course of five collection days. Two plot blocks for the first tillage events are referred to as Blocks A and B. The rototiller was a Honda F-600 mid-tine tiller (Honda Power Equipment) and was operated at full throttle. Two plots of both Blocks A and B were tilled on each of the first 4 d, and all four plots were tilled for the third time on the final day. The first tillage event incorporated an established grass cover. The second collection study (Study 2) occurred in 2017 at a garden site on a loam soil (34% sand, 44% silt, 22% clay) located along Vine St. in Kansas City, MO (39.082738° N, 94.564027° W) following the same protocols as Study 1.

In Study, six 5 m by 5 m plots were delineated and tilled three times each for 45 min on separate days in which the first incorporated a grass cover. The plots were divided into two blocks (Blocks A and B); therefore, tillage occurred on two blocks consisting of three plots each. A sampling day was defined as one tillage event on every plot within one block. Tillage dates, duration, and average weather parameters measured for both studies can be found in Supplemental Table S1. Tillage depth was approximately ∼18 cm, and there was no precipitation.

2.2 | Weather data

Weather variables at each site were monitored (one measurement per minute) using a datalogger (CR1000, Campbell Scientific) located in the center of the research site (Supplemental Figure S1). Air temperature and relative humidity were measured using an electronic sensor (CS215, Campbell Scientific). Wind speed was measured using anemometers (R.M. Young Wind Sentry [Study 1] and Wind Monitor [Study 2]). The temperature probe was set up 1.5 m above ground level, and the remaining variables were collected at a 2-m height.

2.3 | Dust sampling

Dust sampling was completed during rototilling activity using a personal sampling train. A sampling train consisted of a Buck Libra Plus LP-5 sampling pump (A.P. Buck Inc.) connected via clear, flexible polyvinyl chloride tubing to a 47-mm polycarbonate filter holder (Product 1119, Gelman Sciences Inc.), which was modified such that the filter exposure area measured 35 mm in diameter and contained a 46.2-mm Whatman polytetrafluoroethylene (PTFE) filter with polypropylene support ring and 2-µm filter pore space. The collection efficiency of the 2-µm filter for 0.3-µm particles is >99.7% at a flow rate of 16.7 L min^{-1} (USEPA Quality Assurance Guidance Document 2.12 [USEPA, 2016]). Pump flow rate was calibrated at the beginning of each day prior to collection using a Bios Defender 530 gas flow calibrator (Mesa Laboratories, Inc.), and air was sampled at a rate of approximately 4.8 L min^{-1}. The filter holder was attached to the person performing the rototilling within 30 cm of the nose and mouth high on the chest (Supplemental Figure S2). For reference, the researcher was 1.9 m tall but was often bent over slightly in order to push the tiller unit, approximating typical use, so the sampler was about 1.5 m from the ground. Control or background sample collections of 45 min were obtained on random sampling days prior to any tillage taking place to establish background dust concentrations (Study 1: two total, one on 22 June 2014 and one on 19 July 2014; Study 2: six total, one on each sampling date). The sampling train was attached to the weather station at a height of 1.5 m for this collection.

Filter mass was determined according to guidance provided by the USEPA (USEPA Quality Assurance Guidance Document 2.12 [USEPA, 2016]) using a Sartorius XM1000P microbalance housed within a glovebox. The temperature was kept between 21 and 25 ˚C, and relative humidity was controlled by a saturated magnesium chloride solution such that the relative humidity was between 35 and 40%. Individual filters were stored in Petri dishes (88 mm diameter and 12.9 mm height) and allowed to equilibrate to glovebox conditions before and after dust sampling for at least 24 h prior to weighing. Filters were passed in front of a CEM Anti-Static Ionizer to neutralize any static charge that may have accumulated, which would have disrupted accurate mass assessment over time. A successful weighing event was considered finalized when the filter mass could be recorded to the 1 µg three consecutive times within a 5-µg range. A blank filter mass was recorded, and blank filter masses were used for quality control purposes, but a slight difference in blank mass was not subtracted from dust collection measurements.

2.4 | Soil characterization

Soil core samples (2 cm diameter) were collected at depths of 0–2, 2–10, and 10–20 cm at five locations within each plot for gravimetric water content determination and other wet chemical assessments. In Study 1, cores were stored separately, and in Study 2, cores were combined into composite samples across depths. Gravimetric water content was determined according to Gardner (1986) for both studies. For Study 1, an aggregate size distribution sample was taken from each depth and measured with a rotary sieve (Lyles et al., 1970) using four core locations from two time periods: initial sampling before tilling and samples collected immediately prior to the third tillage event. Total soil Pb was also assessed for Study 1 using USEPA method 3051a (USEPA, 1995) with inductively coupled plasma–optical emission spectrometry analysis on single samples collected from the center of each plot before each of the three tillage events. For Study 2, Phase II environmental site assessment information was available through the Kansas City, MO Brownfields Program. The site soils were initially screened using an XL3T Niton handheld X-ray fluorescence analyzer (Thermo Scientific) for further verification, and X-ray fluorescence screening revealed that the Study 2 soils were only slightly contaminated with Pb (<110 mg kg^{-1}).

2.5 | Particle size distribution

The size distribution of particles entrapped by the PTFE filters was investigated using a FEI Nova NanoSEM 430 scanning electron microscope (FEI Company) equipped with vCD and Oxford X-Max Large Area Analytical energy-dispersive

X-ray spectroscopy silicon drift detectors. While under vacuum, using a spot size of 3.5 and accelerating voltage of 5 kV, 25 images of approximately 150 by 140 μm (2,000× magnification) were captured in a 5 by 5 gridded pattern on the filter of the sample collected on Plot 4 of the third round of tillage in Study 1. Particles were identified, and their diameter distributions were evaluated using the National Institutes of Health program ImageJ (NIH, 1997). Energy-dispersive X-ray spectroscopy was also implemented to look for elevated Pb concentrations in the captured particles.

2.6 | Statistical analysis

All statistics were determined in SAS version 9.4 (SAS Institute). Comparison of mean dust collection masses was conducted using Proc Mixed. The Tukey pairwise method was used for comparison of all treatments at an $\alpha = .05$ level of significance.

3 | RESULTS AND DISCUSSION

3.1 | Dust capture

The PTFE filter used in this study has a theoretical efficiency of 99.5% to collect particle size <0.3 μm, and the maximum target mass is about 100 μg cm^{-2} (USEPA, 2016). Control collections made prior to rototilling suggest that the air was relatively clean when no gardening activity was taking place because their mass was not much higher than blank filters. Although not significant, the control did show that small amounts of dust were collected (mean values of 10 μg for Study 1 and 11 μg for Study 2; blank filter masses for Study 1 and Study 2 were 15 ± 5 and 22 ± 5 μg, respectively). Collection results during tillage suggest that very little dust was generated under most of the rototilling conditions tested (Figure 1). In Study 1, only collections made during the third tillage event showed that a large amount of particle matter was cast into the air, whereas the first event in Study 2 recorded the most dust efflux. One limitation of this study is that a temporal dust collection profile is not available. Because sampling was conducted over 45–60 min, we do not know if the rate of dust efflux was constant or if there were periods of time or the direction of tillage relative to wind speed and direction that were more potentially hazardous than others. In other words, during the sampling period, was the dust concentration generally constant, or were periods of low dust concentration punctuated by a few very dusty emissions? Even though total dust collection was low, the lack of temporal resolution imposes some limitations on interpretation and refinement of best practices to protect gardeners. Thus, real-time dust measurements during tillage would enhance future studies of this nature. It should also be noted that this study was conducted on one soil type and might not be reflective of all soil types.

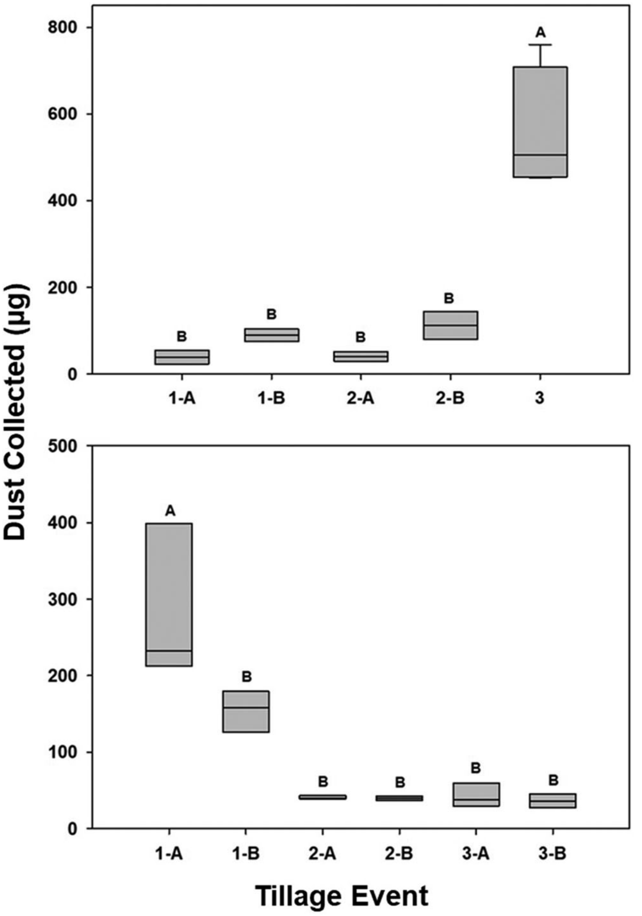

FIGURE 1 Mass of dust collected on polytetrafluoroethylene filters for Study 1 (upper plot) and Study 2 (lower plot) tillage experiments. All masses are expressed per 45-min sampling period. Box plots with the same letter indicate means that are not statistically different at $P = .05$ using Tukey's honest significance test. 1-A, first tilling, Block A; 1-B, first tilling, Block B; 2-A, second tilling, Block A; 2-B, second tilling, Block B; 3, third tilling, all blocks; 3-A, third tilling, Block A; 3-B, third tilling, Block B

3.2 | Soil moisture

In this investigation, the degree of tillage does not appear to be the primary driver of dust efflux; rather, soil moisture plays the most determinant role in regulating emission. This is not surprising because drier soil produces more dust (Funk et al., 2008). Figure 2 shows that in both studies, regardless of how many times the soil was tilled, the driest conditions predicted the greatest dust capture. For both sites, the gravimetric water content of the 2-to-10-cm layer had to reach <15% for significant dust collection to occur. The findings

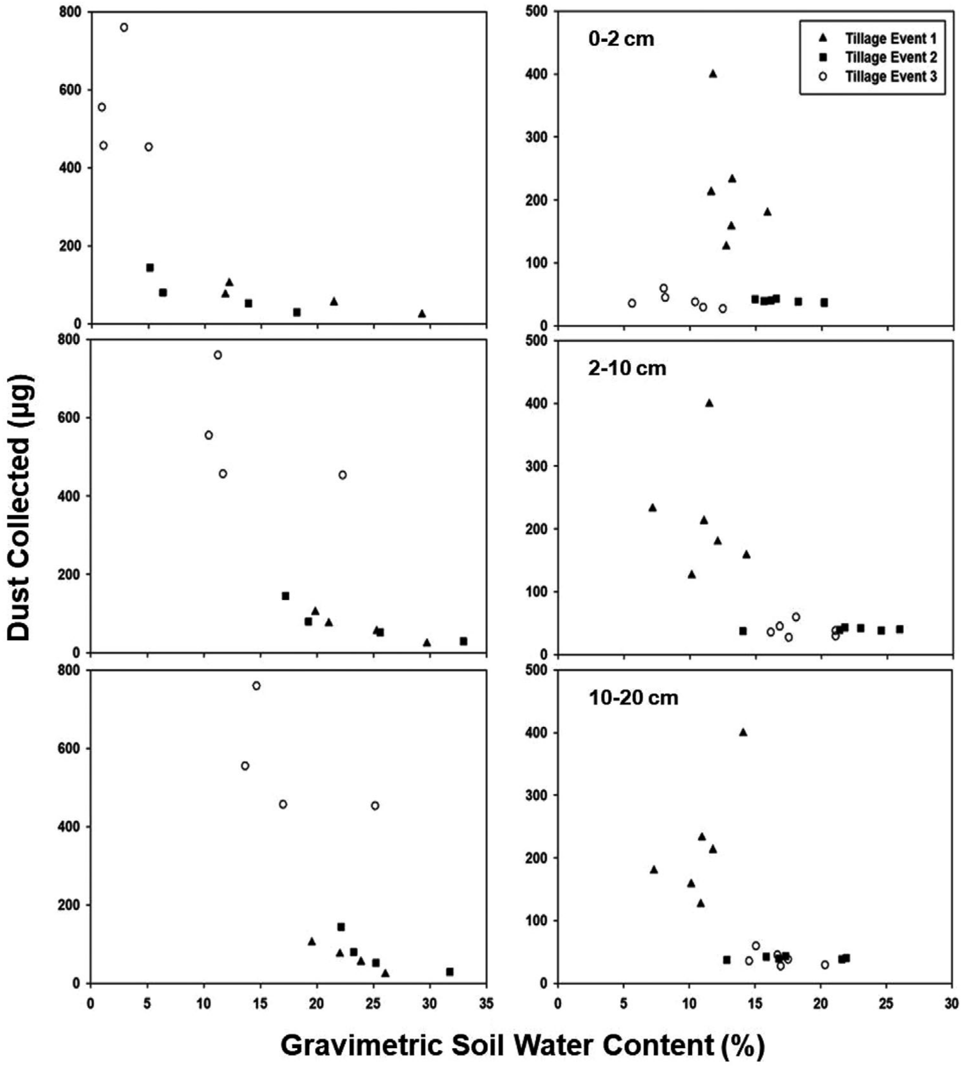

FIGURE 2 Comparison of soil gravimetric water content with the amount of dust captured on polytetrafluoroethylene filters during air sampling during rototilling activity in Study 1 (left plots) and Study 2 (right plots)

did not necessarily correlate with the surface layer. This may be due to the fact that this layer represents a small percentage of the total soil volume that is quickly incorporated with the rest upon disturbance. Other researchers have found that dust efflux is dependent upon soil water (Saleh & Fryrear, 1995). Munkhtsetseg et al. (2016), using a PI-SWERL device, demonstrated a reliable decrease in threshold friction velocity as a bare sandy soil dried, whereas Li and Zhang (2014) observed the same while studying dust emission from the Horqin Sandy Land Area. The polar nature of water leading to its cohesive and adhesive properties helps to bind soil colloids to one another, restricting their surface release. Li and Zhang (2014) observed more saltation of the ≥50-µm fraction in wet soil than in dry soil but less overall fine particle ($0.1 \leq d \leq 20$ µm) discharge. The sampling height in this study may have been too high to capture this process. One management practice indicated by the results that may prove effective for gardeners is to limit soil disturbance under extremely dry soil conditions.

Despite similar water content, fewer particles were captured in Study 2 Event 1-B compared with 1-A. This may be due to the fact the winds were slightly stronger during collection on 1-B (Supplemental Table S1). Although the winds in this study were too low to remove particles directly from the surface, the air movement likely carried the particles generated by tillage off-site before they could rise to the height of the sampler within the research plot. It is possible our methods to assess gardener exposure do not completely reflect the absolute dust generation profile. For example, when rototilling with the wind direction, dust would be carried away from the operator and would not be sampled. The objective of this study was to assess exposure to the gardener; therefore, this is not seen as an oversight but is an aspect worth acknowledging when designing similar future research. Further study is

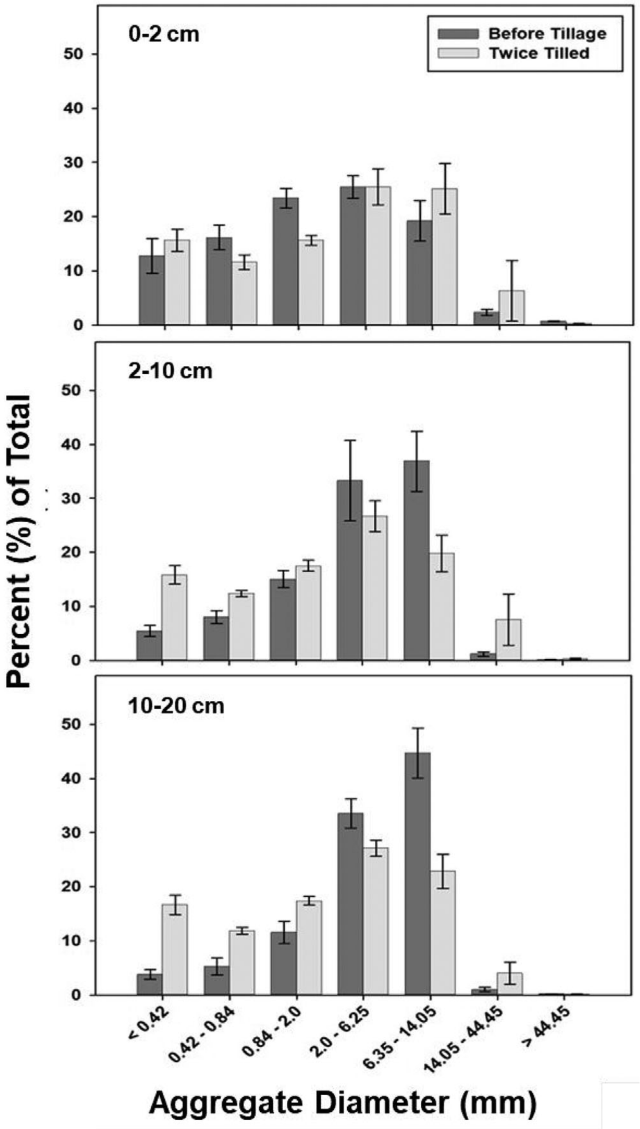

FIGURE 3 Comparison of aggregate size distribution with the amount of dust captured on polytetrafluoroethylene filters while air sampling during rototilling activity in Study 1

required to accurately assess what proportion of dust may be missed or captured relative to the sampler position and wind direction. It is conceivable that under windy, dusty conditions only tilling in the direction of the wind may reduce operator exposure.

3.3 | Aggregate size

As expected, tilling does disrupt the aggregate size distribution of the soil (Figure 3) (Hou et al., 2013). Assessment of Study 1 aggregates shows that the 2-to-10-cm and 10-to-20-cm parts of the profile experienced a redistribution from the larger aggregate fraction (e.g., 6.35–14.05 mm) to the smaller fraction (e.g., <2 mm). This shift was not as evident in the 0-to-2-cm layer, likely due to the originally more granular structure that already skewed toward the smaller side of the distribution. This enrichment of the smaller size fraction could also help explain why the third tillage event of Study 1 generated the most dust. The combination of very dry, poorly aggregated soil would make for more ideal dust-releasing conditions. Tatarko (2001) states in a review of soil processes related to dust emission that wind tunnel tests have shown that a soil with only 1% of aggregates >0.84 mm (i.e., considered non-erodible) is 10 times more erodible than a soil with 53% of aggregates above the same 0.84-mm threshold. Before the third tillage of Study 1, the <0.84-mm fraction was greatly and significantly enhanced ($p < .05$). The shift to smaller aggregate sizes would increase the potential for dust emissions from bare surfaces even when soil tillage is not in progress (Li et al., 2015).

TABLE 1 Distribution of soil Pb by depth and tillage event for Study 1 (Washington Wheatley) plots as assessed according to USEPA method 3051a (USEPA, 1995)

Plot	Sample depth	Never tilled	Tilled once	Tilled twice
	cm		mg kg^{-1}	
	0–2	237	228	274
1	2–10	260	228	269
	10–20	141	216	194
	0–2	232	221	232
2	2–10	217	193	228
	10–20	222	222	220
	0–2	180	152	184
3	2–10	169	186	166
	10–20	182	157	448
	0–2	392	402	538
4	2–10	1,977	412	548
	10–20	271	308	781

Note. All values are single measurements.

3.4 | Soil Pb

Individual soil Pb measurements of Study 1 plots reveal relatively homogeneous, mild contamination throughout the 0-to-20-cm profile with the exception of Plot 4, where a “hot spot” with a high concentration of 1,977 mg kg^{-1} was observed prior to tillage at the 2-to-10-cm depth (Table 1). The results show that the concentrations of contaminants like Pb can be highly variable in garden soils on multiple scales; this variability can influence the interpretation of data, particularly single-point measurements. Lead was not measured in Study 2 because the results were not deemed to be necessary to

TABLE 2 Size distribution of particles observed entrained in the polytetrafluoroethylene filter used to sample the dust generated from the third tilling of Plot 4 of the Washington Wheatley study (Study 1) using scanning electron microscopy

Size fraction	Particle count	Percentage of total particle count
µm	no. of units	%
<4	99	73.9
4–10	33	24.6
10–100	2	1.5
<2.5	71	53.0
<10	132	98.5

interpret the dust results. Dust generation is not likely contingent on Pb concentration, so collection results could be generalized to any concentrations of the contaminant for health assessment purposes. Table 1 highlights the dilution effect that can occur when soil is disturbed (i.e., mixed by tilling). After two rounds of cultivation, the high concentration was spread throughout the entire sampled depth, and the whole soil volume exceeded 400 mg kg^{-1}, which is the USEPA soil Pb standard for children's bare play areas. Blending cleaner soil with contaminated soil has been suggested to mitigate the effects of Pb contamination exposure (Attanayake et al., 2014).

3.5 | Particle analysis

Scanning electron microscopy interrogation of the filter from Plot 4, Study 1, Event 3 suggests that the vast majority of particles collected on the filters are <4 µm in diameter (Table 2). This is concerning because the smaller the particle, the further that particle can enter the respiratory system and cause damage to human health (Goudie, 2014). The USEPA regulates mass concentration for outdoor air quality for particles <10 µm as well as mass concentration for particles <2.5 µm for health reasons (USEPA, 1996, 1999). Additionally, the colloidal fraction is the most chemically reactive and tends to be enriched with Pb relative to the bulk concentration (Juhasz et al., 2011). A preponderance of these particles could lead to underestimation of contaminant exposure risk if bulk concentration analysis is used for assessment purposes. In this study, energy-dispersive X-ray spectroscopy of individual particles revealed common soil elements such as silicon, calcium, and potassium but did not register Pb (Supplemental Figure S3). The limit of detection for this method is approximately 1,000 mg kg^{-1}, so it may be that Pb was present but at undetectable concentrations.

3.6 | Risk evaluation

Although a couple of tillage events in this investigation did result in elevated amounts of dust capture, overall data suggest that dust inhalation from rototilling is not a major pathway for soil Pb exposure by urban gardeners, even with much higher contamination levels. For example, the human adult body contains approximately 55 dl of blood, so to raise blood Pb levels 1 µg dl^{-1}, 55 µg of Pb would need to enter the body and be absorbed. The greatest dust-generating event in this study captured 760 µg of soil by sampling ∼216 L of air, for a dust concentration of 3.5 mg m^{-3}. The highest soil Pb concentration measured was 1,977 mg kg^{-1}, so under the absolute worst conditions measured, 27 mg of soil at 100% bioavailability would need to be inhaled to raise blood Pb levels by 1 µg. Under these conditions, ∼7.7 m^3 would need to be inhaled. For reference, a moderately active person only inhales 2.1 m^{-3} h^{-1} (USEPA, 2011). Thus, Pb inhalation during rotolling pathway appears to be minor based on the experimental conditions tested in this study. Also, it is important to note that if these 25-m^2 plots were tilled for a much longer duration or if an even larger plot was tilled, a further potential risk would be present. This is not to say that dust is not important in other contexts. The half-life of Pb in the blood is ∼36 d, so prolonged exposure in a setting, such as a construction site on even mildly contaminated ground, could be quite hazardous over time (WHO, 1995). Also, as referenced in the Results, the dust collection for these studies was a single point on a moving person performing the activity. Community members downwind could be accidently exposed more so than the individual gardener if they remain in the airflow carrying the particles offsite. Dust inhalation could be an important possible exposure pathway, and one may include activities beyond rototilling that bring gardeners and others in contact with bare soil (e.g., working the soil with hand tools, soil dust movement during times of dry soil conditions, maintenance of pathways and other nongardening areas, etc.). Care should be taken to reduce dust exposure overall to eliminate potential soil Pb and particulate matter exposure. Lightly sprinkling garden soil with a hose could minimize dust generation, though overly wet soils can cause tillage problems, as shown throughout production agriculture settings.

4 | CONCLUSIONS

The results of this limited investigation suggest that short-term dust exposure from rototilling on mildly contaminated brownfields sites is not a major Pb exposure pathway for urban gardeners. A couple of sampling events did record elevated air concentrations of dust relative to the others. This seems to be related to the gravimetric soil water content of the 2-to-10-cm and 10-to-20-cm sections of the soil profile; the drier the soil, the more dust that is generated. The degree of tillage and subsequent reduction of aggregate size may play a role in encouraging particle emission, but the contribution seems to be less than the soil moisture. Particles that were collected tended to be <4 μm in diameter and thus could pose a risk to respiratory health. Out of an abundance of caution, gardeners tilling under dry conditions may choose to wear a dust mask to mitigate particle inhalation to the greatest extent possible. This research also adds weight to management recommendations, such as mulching gardening areas and pathways and other techniques to minimize areas of bare soils within gardens.

Although our research does not indicate short-term exposure risks, it does suggest improved methods for further studies of this nature. Real-time dust measurements during tillage would allow better interpretation of results. Further study is also needed to accurately assess what proportion of dust may be missed or captured relative to the sampler position along with tillage and wind direction. Future studies focusing on other dust inhalation exposures in urban gardening beyond rototilling are also needed.

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CONFLICT OF INTEREST

The authors report no conflicts of interest.

ORCID

Joseph J. Weeks https://orcid.org/0000-0002-4957-6794
Ganga M. Hettiarachchi https://orcid.org/0000-0002-6669-2885
Eduardo Santos https://orcid.org/0000-0001-6767-5123
John Tatarko https://orcid.org/0000-0003-1295-3290

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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