



Widespread Pb contamination in urban backyard soils for >100 years identified in soil cores constrained by ^{210}Pb and ^{137}Cs

Franziska C. Landes^{a,b,*}, Margaret Calcio^c, Jennifer M. Sobolewski^b, Kelcey Wallens-Logan^c, Anna Ledeczki^b, Yael Kiro^{a,d}, Louise Bolge^a, James Ross^a, Steven N. Chillrud^a, Brian J. Mailloux^c, Alexander van Geen^a

^a Lamont-Doherty Earth Observatory, Columbia University, 61 Route 9W, Palisades, New York 10964, USA

^b Department of Earth and Environmental Sciences, Columbia University, New York 10027, NY, USA

^c Department of Environmental Science, Barnard College, Columbia University, New York 10027, NY, USA

^d Department of Earth and Planetary Sciences, Weizmann Institute of Science, Rehovot 76100, Israel

ARTICLE INFO

Editor: Mae Sexauer Gustin

Keywords:

Anthropogenic soils
Lead contamination
Bioaccessibility
Atmospheric radionuclides
Soil core profiles

ABSTRACT

Contaminated soil in urban residential areas is often overlooked as a source of childhood exposure to toxic levels of lead (Pb). We document mean Pb concentrations of 1200 ± 1000 mg/kg, three times the now outdated EPA soil hazard standard of 400 mg/kg, for 370 surface soils collected from 76 homes in the boroughs of Brooklyn and Manhattan of New York City. The mean Pb content of 250 ± 290 mg/kg Pb for 571 surface soils collected from tree pits and public parks was much lower. A subset of 22 surface samples analyzed by EPA Method 1340 extracted 86 ± 21 % ($\pm 1\text{SD}$) of total soil Pb, indicating that the Pb was highly bioavailable. To investigate the origin of backyard contamination, 49 cores were collected to an average depth of 30 cm from a subset of 27 homes. Twelve soil cores were analyzed for ^{210}Pb and ^{137}Cs to constrain processes that impact contaminant distribution and inventories (particle focusing, soil accumulation, loss, and mixing). Concentrations of Pb declined with depth in 60 % of the cores but usually did not reach background. Mean uncorrected Pb inventories of 340 ± 210 g/m² Pb (mean $\pm 1\text{SD}$, $n = 12$) were more than five times higher than the radionuclide corrected inventory of 57 g/m² from Central Park soil cores. Average inventories of $^{210}\text{Pb}_{\text{xs}}$ (3.5 ± 0.9 kBq/m²) and ^{137}Cs (0.9 ± 0.6 kBq/m²) corresponded to 71 ± 19 % and 50 ± 30 % of the predicted atmospheric inventories. Elevated Pb concentrations were found both in the fine (<1 mm) and coarse (>1 mm) fractions, the latter suggesting a local non-atmospheric source. This was confirmed by individual grains containing up to 6 % Pb and visible pieces of coal, bricks, and ash. Regardless of the source of contamination in backyard soils, systematic testing is needed to identify contaminated areas and reduce child exposure.

1. Introduction

Lead contamination of soil can be the result of past and present lead emissions (Laidlaw and Filippelli, 2008) including leaded gasoline (Mielke and Reagan, 1998; Wu et al., 2010), deteriorating lead-based paint (Andra et al., 2006), municipal waste incineration (Chillrud et al., 1999), smelters (Landrigan et al., 1975; Factor-Litvak et al., 1999); mining (van Geen et al., 2012), informal lead-acid battery recyclers (Ericson et al., 2016), and other industrial activities (Landrigan et al., 2018). Once deposited, Pb strongly sorbs to iron and manganese oxides and organic matter in soil. While numerous studies have found higher Pb concentrations in surface soils near roads, indicating the importance of

leaded gasoline (Mielke et al., 1983; Filippelli et al., 2005; Wu et al., 2010), analysis of a core of continuously accumulating sediment collected in New York City's Central Park lake showed that solid-waste incineration was the primary contributor to atmospheric Pb deposition in the city during the 20th century, with peak deposition rates observed between the 1930s and 1960s (Chillrud et al., 1999). Dating of input in soil cores is more difficult because soil mixing, instead of accumulation, is often the main mechanism moving Pb to deeper levels (Kaste et al., 2006). Urban soils may also contain anthropogenic fill material or debris such as coal fly ash, glass, bricks, metal, plastic, wood and more (Li et al., 2018). Other sources of Pb contamination in urban soils are pesticides such as lead-arsenate, and construction materials (Nezat et al., 2017).

* Corresponding author at: Lamont-Doherty Earth Observatory, Columbia University, 61 Route 9W, Palisades, New York 10964, USA.

E-mail address: fcl2115@columbia.edu (F.C. Landes).

<https://doi.org/10.1016/j.scitotenv.2023.165407>

Received 28 April 2023; Received in revised form 5 July 2023; Accepted 6 July 2023

Available online 8 July 2023

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This study was motivated by the need to explore these widespread sources of backyard Pb over time to inform interventions in a major city with a long industrial history.

In 1991, the U.S. Environmental Protection Agency (U.S. EPA) set the soil hazard standard for lead (Pb) at 400 mg/kg in bare soil where children play and 1200 mg/kg for other residential areas, based on the U.S. Center for Disease Control (U.S. CDC) blood Pb level (BLL) of concern of 10 µg/dL (U.S. EPA, 2001). Recognizing the damaging impacts of even lower BLLs, the U.S. CDC in 2012 switched to a reference value based on the 97.5 percentile of children tested in the National Health and Nutrition Examination Study (NHANES). The initial value was set at 5 µg/dL and was reduced in 2021 to 3.5 µg/dL (Tsoi et al., 2016; U.S. CDC, 2021). This moving health-based target presents a challenge for determining soil cleanup standards, which are often risk-based but are not usually adjusted as frequently as the 5 years NHANES study cycle. California updated its residential soil screening level to 80 mg/kg Pb in 2005 based on a health risk of an increase in child BLL by 1 µg/dL (California OEHHA, 2009). New York State now uses a Soil Clean-up Objective of 63 mg/kg Pb for unrestricted use, based on rural soil background concentrations, while using 400 mg/kg as a residential standard for restricted use (NYSDEC, 2006).

Concentrations of toxic metals such as Pb in urban soils, especially in the northeastern US, have long been documented to be greater than the U.S. median Pb, or natural background level (Datko-Williams et al., 2014), of 18 mg/kg Pb (Smith et al., 2019). A comprehensive analysis of published soil-Pb concentrations from 84 studies across 62 cities found urban soil Pb concentrations were highest in city centers and declined outwards, with a majority of studies showing median and mean soil-Pb concentrations below 400 mg/kg Pb. A handful studies, however, found soil Pb concentrations above 400 mg/kg and 1200 mg/kg (Datko-Williams et al., 2014), notably in the Portland Peninsula of Maine, USA (mean 1500 mg/kg Pb) (Wagner and Langley-Turnbaugh, 2008), in one neighborhood in Boston, Massachusetts, USA (mean 3000 mg/kg Pb compared to 600 mg/kg in Boston overall) (Weitzman et al., 1993), in East Chicago, Illinois, USA (mean 685 mg/kg) (Haque et al., 2021), and in samples collected from the sides of homes in San Antonio, Texas, USA (mean 1700 mg/kg Pb) (Andra et al., 2006). In Indianapolis, Indiana, USA mean soil Pb concentrations were notably higher in the dripline (805 mg/kg Pb) than in the rest of the yard (345 mg/kg) (Filippelli et al., 2018). Similarly, in Durham, North Carolina, USA mean soil Pb concentrations in house foundation soils (2280 mg/kg Pb) were significantly higher than streetside soils (320 mg/kg Pb) (Wang et al., 2022).

In New York (NYC), New York, USA, the mean Pb content of almost 1700 soil samples submitted to a testing program at Brooklyn College between 2009 and 2015 was 600 mg/kg Pb, with 20 % of samples exceeding the NYC Soil Cleanup Objective of 400 mg/kg Pb (Cheng et al., 2015). The same study showed that 68 % of a subset of 197 home garden samples (median: 630 mg/kg Pb) exceeded the NYC Soil Cleanup Objective of 400 mg/kg Pb, while only 10 % of 106 community garden samples (median: 140 mg/kg Pb) exceeded this limit. A follow-up study found that 48 % of garden samples, 7 % of park samples, and 6 % of public housing samples exceeded 400 mg/kg (Li et al., 2018). These studies identified areas in the NYC boroughs of Brooklyn and Queens with particularly elevated backyard soil Pb concentrations that correspond to manufacturing and industrial areas outlined by a 1922 map from the New York Merchants Association (Merchants' Association of NY, 1922). A recent study of soil from 34 NYC parks by Pavilonis et al. (2020) found lower Pb concentrations compared to home garden samples tested by Cheng et al. (2015).

An analysis of dated lake-core sediments collected from NYC's Central Park in 1999 established that peak atmospheric Pb deposition in midtown Manhattan in the late 1930s-early 1960s coincided with solid-waste incineration rather than leaded gasoline (Chillrud et al., 1999). Age control for the core was provided by a detailed profile of excess ^{210}Pb which was consistent with the ^{137}Cs profile that matched the recorded fallout from atmospheric testing of nuclear bombs in the 1950s

and 1960s. Accumulation of sediments in the Central Park lake core, based on a ^{137}Cs inventory of 4.62 ± 0.11 kBq/m², resulted in a contaminant Pb inventory corrected for a background concentration of 26 mg/kg of 102 g/m², and, when normalized to $^{210}\text{Pb}_{\text{xs}}$, 64 g/m² (Chillrud et al., 1999). Four additional soil cores collected by Chillrud et al. (1999) on dry, flat grassy areas of the park that Central Park Conservancy records indicated had not been renovated showed similar ^{137}Cs inventories to fallout. The average radionuclide normalized inventory of contaminant Pb from these soil cores (57 g/m²) was similar to the lake core (64 g/m²), suggesting that the Central Park lake core largely reflected regional atmospheric inputs during the 20th century (Chillrud et al., 1999). This lake-core estimate of atmospheric deposition corresponds to an integrated atmospheric deposition of 50000 metric tons of Pb when extrapolated over the entire area of New York City.

In this study, we investigate whether soil Pb concentrations differ between NYC neighborhoods and between public spaces and residential backyards. Furthermore, we hypothesized that backyard Pb concentrations should have a similar atmospheric input to that documented in Central Park. We report Pb concentrations in surface soil from 76 NYC backyards and soil core profiles from a subset of 27 of these backyards. This provides a large sample size of 370 backyard samples and 571 public samples collected in Manhattan, from the Upper West Side and Harlem neighborhoods, and in Brooklyn, from the Greenpoint and Williamsburg neighborhoods. Unlike most previous urban soil Pb studies, we measured ^{210}Pb and ^{137}Cs activities in the cores to check their integrity, constrain the timeframe of Pb contamination, and evaluate evidence of surface-soil loss (radionuclide inventory < atmospheric) or particle focusing of atmospheric deposition (soil radionuclide inventory > atmospheric). We infer additional local sources of Pb beyond atmospheric deposition from the magnitude of the Pb inventories relative to the Central Park record and the characteristic particles with very high Pb concentrations in backyard soil.

2. Materials and methods

2.1. Soil sample collection

Composite surface soil samples were collected from the backyards of 76 private homes ($n = 370$) and from publicly accessible parks, tree pits, and other green areas in NYC ($n = 571$) (Fig. 1). Samples were collected from the top 1–2 inches as five-point composites or by scraping together the surface soil in an area of approximately 0.5 m² with a stainless-steel spoon and, if the soil was sufficiently dry, directly sieved through a 1 mm kitchen sieve. Four to six composite surface-soil samples were collected per yard for 9 homes in the Upper West Side (UWS), Manhattan, 4 homes in Harlem, Manhattan, and 63 homes in Greenpoint and Williamsburg (Greenpoint), Brooklyn. Surface samples from publicly accessible areas were only collected in Greenpoint; 108 of these were collected as part of an environmental science course at Barnard College, mostly from tree pits, while the other 463 were collected in a parallel sampling effort targeting backyards in Greenpoint (Fig. 2). Metadata including GPS coordinates, a photo, and a site description were recorded for all samples using the smartphone app SurveyCTO (SurveyCTO, 2019) or KoboCollect (KoBoToolbox, 2017). Sample description data included whether samples were collected from (a) gardens in raised beds, (b) gardens not in a raised bed, (c) drip lines below the outer edge of a roof or gutter, (d) unmaintained areas, (e) maintained areas, (f) areas with compost or new soil clearly added, (g) children's play areas, (h) near building walls, (i) far from building walls, (j) the front yard, (k) near any fence, building wall, shed, or painted structure, or (l) areas with reports of found debris material.

In a follow-up, 49 soil cores were collected from a subset of 27 of the 76 private homes, 15 in Brooklyn and 12 in Manhattan. Samples were collected in the area of the yard that was the most undisturbed, based on appearance and the current resident's description. A hammer-driven 2-ft (61 cm) AMS Soil Corer with a plastic 1-inch diameter sleeve was used to

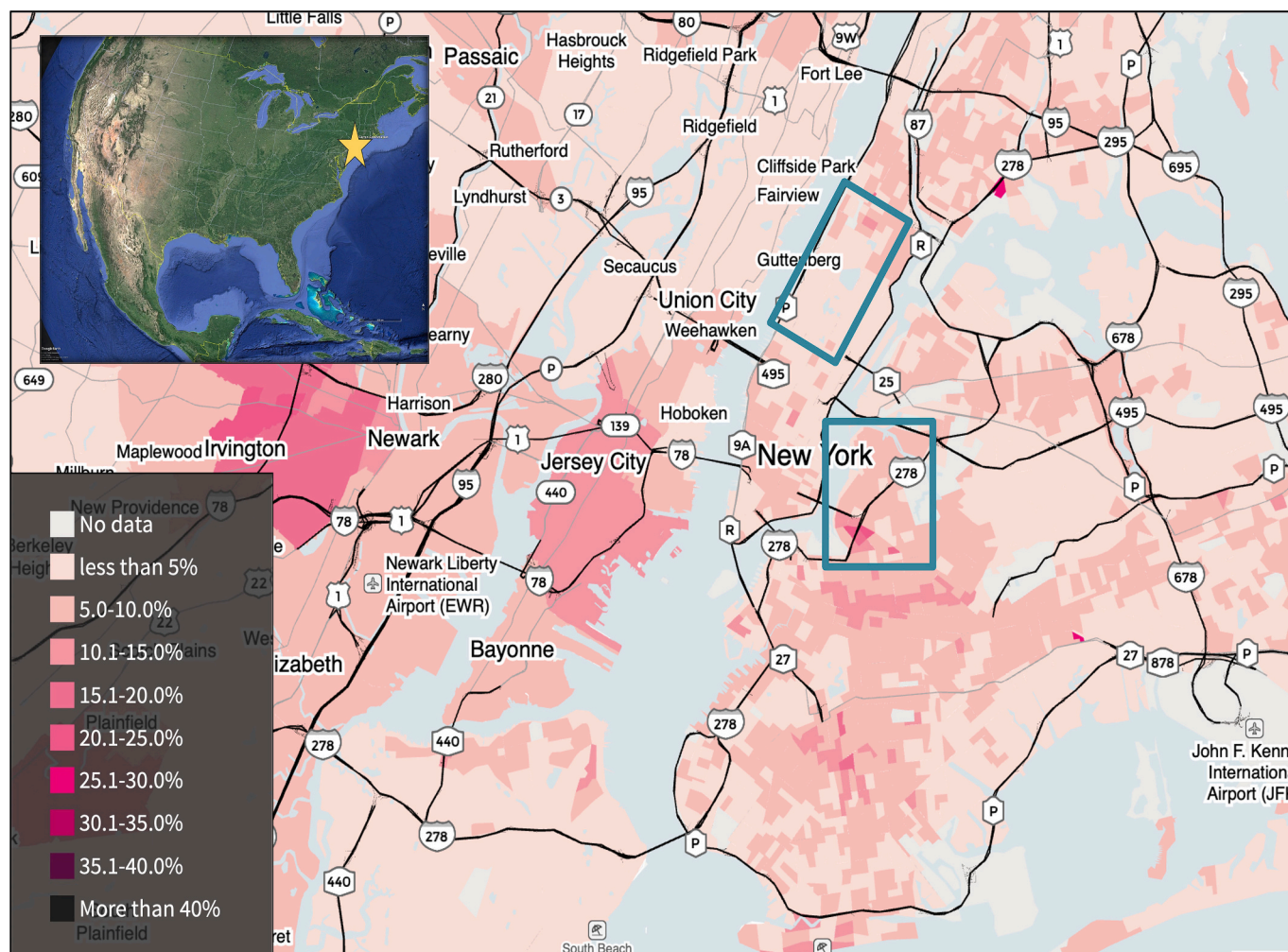


Fig. 1. Proportion of children with blood lead levels over 5 µg/dL in New York City from 2006 to 2015 based on data from NYC Dept of Health and Centers for Disease Control, from Reuters: <https://www.reuters.com/investigates/graphics/lead-water/en/>. Blue boxes indicate areas targeted for soil sample collection.

collect each soil core. The same metadata collected for surface soil samples was also collected for soil cores, including GPS location and photo of the core location. The average length of the 49 soil cores was 30 ± 9.5 cm, without correcting for compression.

All home sampling visits were conducted by researchers with Human Subjects Research training. Residents signed a consent form approved by the Columbia University Institutional Review Board. Participants who agreed to receive a free soil test indicated how we may reproduce their soil Pb results by choosing between providing permission for us to map soil data by GPS, by the nearest intersection, or only in aggregate format.

2.2. Sample preparation

All home surface samples, and the subset of samples from public spaces that were not sieved in the field, were air-dried for at least 48 h. Soil samples were then sieved through a 1 mm kitchen sieve and funnel into 20 mL scintillation vials. Core samples were cut into 5 cm intervals, weighed, and dried in an oven at 60–80 °C at least overnight. Each interval was disaggregated using a mortar and pestle, separated into fine (<1 mm) and coarse (>1 mm) fraction in 20 mL scintillation vials, and weighed (Fig. S10). The coarse fractions of a subset of 20 core sections were ground to a fine powder with a ball mill (SPEX SamplePrep 8000 M Mixer/Mill). A subset of 26 coarse soil fractions were also rinsed after soaking in MQ DI water in a sonicator for 60 min to remove adhered fine particles (<1 mm) from the coarse fraction.

2.3. Acid digestion of soil samples

Total acid digests following the method of Fleisher and Anderson (1991) were carried out on a subset of 22 soil samples to confirm X-ray fluorescence analysis of total Pb in soil samples. A 0.2 g subsample of soil hand-ground by mortar and pestle was covered in concentrated nitric acid (HNO₃) to dissolve carbonates in teflon beakers. Samples were then digested on a hotplate, first in concentrated nitric acid (HNO₃) and concentrated perchloric acid (HClO₄) at a ratio of 1 to 3, and then with the addition of concentrated hydrofluoric acid (HF) to digest the silicates. If any visible particles remained, more HClO₄ and HF was added as needed to individual samples until complete dissolution.

2.4. Instrumental analysis

All soil samples were analyzed for total Pb concentrations using a handheld X-ray fluorescence (XRF) spectrometer in a benchtop stand in the laboratory (Innov-X Systems DELTA Premium). Samples in 20 mL scintillation vials were covered in plastic wrap and inverted on the benchtop stand and analyzed for 60 s, with 20 s at each of the XRF's three incident-beam energies. To better represent soil-Pb variability, each sample was analyzed three times, and the vials were inverted end-over-end between each analysis.

Results from the XRF analysis for soils were compared to U.S. National Institute of Standards and Technology (NIST) Standard Reference Materials (SRM) soils SRM 2710 with 5532 mg/kg Pb (mean 93 ± 3 %

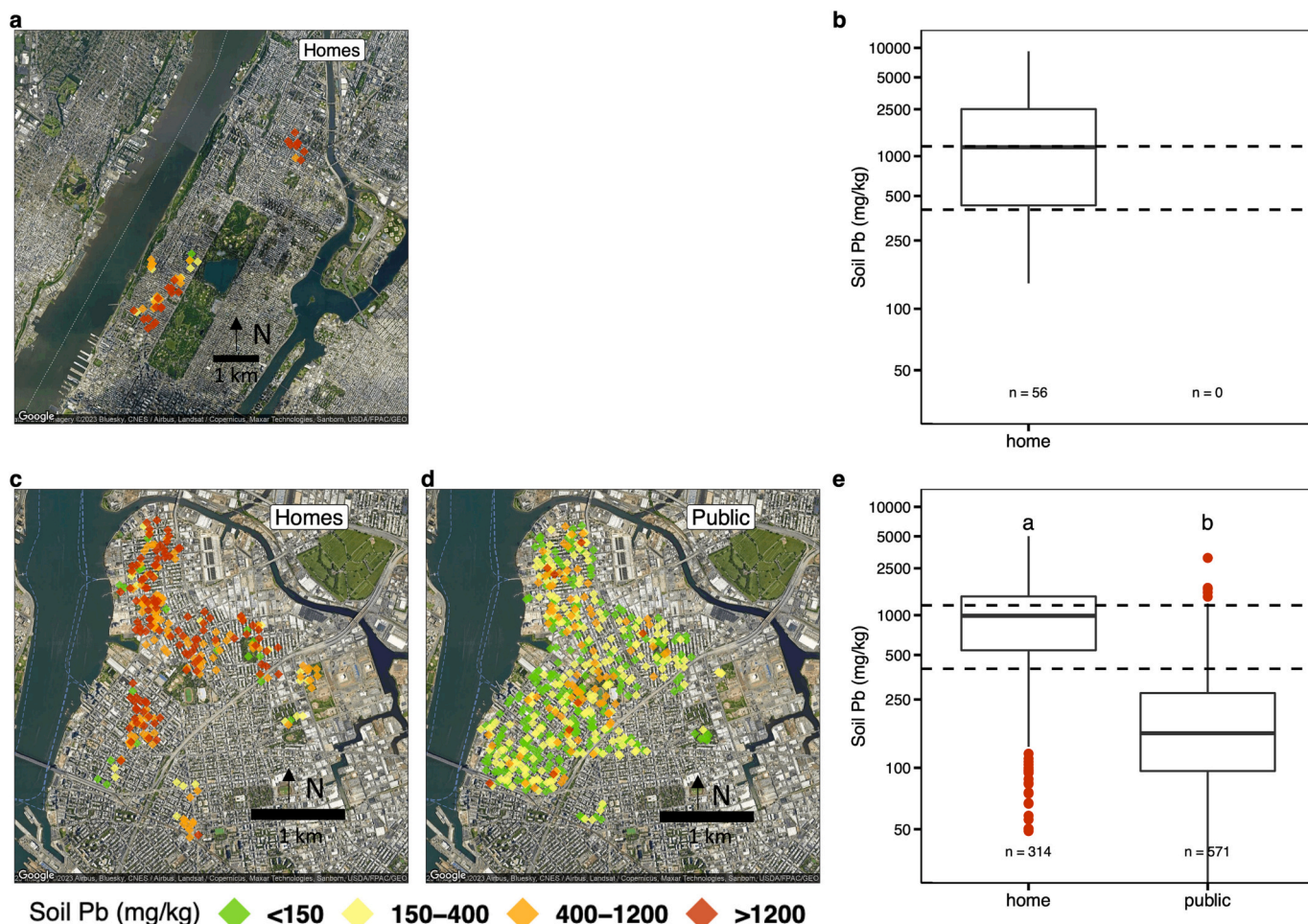


Fig. 2. Concentrations of Pb in surface soil samples collected (a) from home backyards in Manhattan, including Upper West Side and Harlem, (c) from home backyards in Brooklyn, including Greenpoint and Williamsburg, and (d) from publicly accessible parks and tree-pits. Distribution of Pb concentrations in (b) Manhattan and (e) Brooklyn. Home samples are jittered and only included on the map if residents agreed to intersection-level disclosure on the consent form. Letters in (e) indicate groups that are statistically different ($p < 0.05$).

SD of the reference value, $n = 90$), SRM 2711 with 1162 mg/kg ($105 \pm 2\%$, $n = 126$), SRM 2711a with 1400 mg/kg ($104 \pm 2\%$, $n = 113$), and SRM 2709 with 18.9 mg/kg ($132 \pm 17\%$, $n = 121$). Although the obtained values were too high by about 6 mg/kg for the standard with the lowest Pb concentration, the readings were not adjusted.

Analysis of total metal concentration in soils by XRF has been shown to be well correlated to measurements from acid digestion (Radu and Diamond, 2009) and is approved for soil as EPA Test Method 6200 (U.S. EPA, 2015). In this study, a subset of samples from a complete digestion were analyzed by high-resolution (HR) inductively coupled plasma mass spectrometer (ICP-MS) (Thermo Scientific Element II). Based on these results, the following corrections were made to XRF measurements for analysis: tin (Sn) data was divided by 1.64 and zinc (Zn) was divided by 0.79 (Fig. S4). No corrections were made to XRF data in the case Pb (slope of 0.98).

The ICP-MS analysis of NIST reference materials were within 10 % of their published values for 1640A (mean recovery 107 %, $n = 5$) and 1643F (109 %, $n = 5$). All ICP-MS method and vial blanks contained $<1 \mu\text{g/L}$ Pb. Complete digests of NIST SRM 2711a recovered 105 % ($n = 2$) Pb, and duplicates were within 3 % of each other (two sets of duplicates).

2.5. ^{210}Pb and ^{137}Cs counting

Radionuclide activities for the target isotopes were determined by

gamma counting (EG&G Ortec, model GWL-120230) for the fine ($<1 \text{ mm}$) fraction of 67 core intervals, with 12 complete core inventories, and three core-top samples. Unsupported ^{210}Pb , hereafter $^{210}\text{Pb}_{\text{XS}}$, was determined by correcting for supported ^{210}Pb estimated by measured ^{214}Pb and ^{214}Bi activities (dpm/g). Gamma-counting data for ^{137}Cs was accepted when the counting error of the following standards were within 5 % of their decay-corrected published value: NIST 4350B Columbia River sediment ($784 \pm 50 \text{ pCi/kg } ^{137}\text{Cs}$), and GBW08304A, River Sediment, National Institute of Meteorology, People's Republic of China ($2189 \pm 131 \text{ pCi/kg } ^{137}\text{Cs}$). Gamma-counting data for $^{210}\text{Pb}_{\text{XS}}$ was accepted when counting error of the following standards were within 10 % of their decay-corrected published value: D-standard river sediment ($3838 \pm 2 \text{ dpm/g } ^{210}\text{Pb}_{\text{XS}}$) and LGP internal and secondary standard ($46.4 \pm 1.8 \text{ dpm/g } ^{210}\text{Pb}_{\text{XS}}$).

Inventories of $^{210}\text{Pb}_{\text{XS}}$ and ^{137}Cs activities are used to confirm soil core integrity, and may be used to estimate soil erosion or accumulation. When not too far from expected atmospheric deposition rates, contaminant inventories can be corrected for soil erosion or accumulation (Kada and Heit, 1992; Callender and Van Metre, 1997; Matisoff and Whiting, 2012). These radionuclides are useful tracers because they have a known rate of atmospheric deposition. In the case of $^{210}\text{Pb}_{\text{XS}}$, a decay product of the noble gas radon (^{222}Rn), deposition is semicontinuous and expected to be constant on an annual basis. After ^{222}Rn escapes into the atmosphere, it decays to $^{210}\text{Pb}_{\text{XS}}$ which is particle reactive and deposits with fine particles from the atmosphere; ^{210}Pb has a half-life of 22.3 years.

Inputs of ^{137}Cs into the atmosphere were from nuclear weapons testing activities that distributed ^{137}Cs in the stratosphere globally; they were negligible prior to 1954 and after the 1980s with a large maximum in 1963; ^{137}Cs has a half-life of 30.2 years. While most studies measuring soil erosion or sediment accumulation collect radionuclide data from an undisturbed reference site, no such site was available for this study. Instead, data were compared to the average of $^{210}\text{Pb}_{\text{xs}}$ inventories of 4.9 kBq/m² calculated from two studies in the northeastern United States. In the first study, a $^{210}\text{Pb}_{\text{xs}}$ inventory of 4.1 kBq/m² (25 dpm/cm²) was calculated from seven lowland soils, including a location just outside NYC that also reported the mean inventory (Graustein and Turekian, 1989). In the second study, the radionuclide data were from a soil core from Rhinebeck, NY, approximately 125 km north of the NYC, with an estimated $^{210}\text{Pb}_{\text{xs}}$ inventory of 5.7 ± 41 kBq/m² and ^{137}Cs inventory of 3.77 ± 11 kBq/m² in 1986 (Klusek, 1987; Kada and Heit, 1992). Remaining ^{137}Cs inventories, decay-corrected to 2018, when the soil cores in this study were collected, would then be 1.8 kBq/m². These references are the same ones used to calculate $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs inventories for Central Park (Chillrud et al., 1999). In that study, ^{137}Cs inventories were 4.62 ± 0.11 kBq/m² in a Central Park lake core and ranged from 2.71 to 4.49 kBq/m² in four Central Park soil cores, close to the expected and decay-corrected NYC fallout of 2.93 kBq/m².

2.6. Soil inventories for metals

Soil inventories were calculated by multiplying the dry weight of each 5 cm core section by the average of three repeat XRF analyses, dividing by the 5.07 cm² cross-sectional area of the core, converting units to g/m², and adding all core sections. The average background concentration was subtracted from each XRF reading for the following: Pb: 26 mg/kg, Sn: 6 mg/kg, and Zn: 80 mg/kg (Chillrud et al., 1999). These calculations were conducted for both the fine fraction (<1 mm after hand grinding by mortar and pestle) and the coarse fraction (>1 mm). For 12 soil cores with $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs data, contaminant Pb inventories are considered with and without correction by normalizing to the expected integrated fallout of 4.9 kBq/m² $^{210}\text{Pb}_{\text{xs}}$ and 1.8 kBq/m² ^{137}Cs (Chillrud et al., 1999). The radionuclide correction is only valid for atmospheric sources of contaminant Pb, in which case these atmospherically derived radionuclides can be used to correct for focusing or loss of atmospheric Pb from the soil.

2.7. Other analyses

Neighborhood characteristics such as year of building construction and number of units per building were downloaded from NYC's Primary Land Use Tax Lot Output (PLUTO™) (New York City Department of City Planning New York City, 2019). A subset of 21 surface soil samples were analyzed for bioaccessible Pb following EPA Method 1340 (U.S. EPA, 2017) and Drexler and Brattin (Drexler and Brattin, 2007). And a subset of 50 soils were analyzed by a field extraction procedure described in Landes et al. (2019), where the only aggregated results were presented for 30 samples from homes and 20 from public spaces in Greenpoint. A single high-Pb coarse grain was analyzed by laser-ablation ICP-MS at Lamont-Doherty Earth Observatory. Analysis of NIST Reference Material 610 by laser-ablation indicated 97 % of the published value.

2.8. Statistical analyses

Statistical analyses were performed with R Studio 1.0.136 (RStudio Team, 2016) using R 3.5.1 (R Core Team, 2018). Since soil-Pb concentrations were not normally distributed, Spearman's rank correlation was used. Differences between neighborhoods were checked for significance using the Kruskal-Wallis test for non-parametric continuous data and a Mann Whitney U test.

3. Results

3.1. Concentrations of Pb in surface soil

The mean Pb concentration of 370 surface-soil composite samples collected from 76 homes was 1200 ± 1000 mg/kg Pb ($\pm 1\text{SD}$) with a wide range across two orders of magnitude. In comparison to the current U.S. EPA soil hazard standards, 81 % exceeded the 400 mg/kg for bare soil areas where children play and 38 % exceeded the higher 1200 mg/kg Pb standard. The mean soil-Pb concentration in the UWS (1400 ± 2000 , $n = 40$) was significantly lower than in Harlem (2600 ± 1300 , $n = 16$) but similar to the average for Greenpoint (1000 ± 700 mg/kg Pb, $n = 314$, $p < 0.01$) (Table 1, Fig. 3). The mean Pb concentration for 571 surface soils from publicly accessible areas, mostly tree pits, in Greenpoint was significantly lower with a mean of 250 ± 290 mg/kg Pb (Fig. 2, $p < 0.01$) than backyard samples. In contrast to the backyard samples, only 16 % of the publicly accessible soil samples exceeded 400 mg/kg Pb and 1.5 % exceeded 1200 mg/kg Pb.

When examining the results at the household level, 93 % of homes had at least one sample that exceeded 400 mg/kg while 64 % had at least one that exceeded 1200 mg/kg Pb ($n = 76$). There was no systematic difference in Pb concentrations for samples that were collected near or far away from structures, or from maintained or unmaintained areas (Fig. S5). Samples collected from gardens in raised beds were lower than those from gardens in non-raised beds and samples collected from non-garden areas.

3.2. Depth profiles of Pb

In order to evaluate whether the Pb was concentrated in the surface or remained elevated at depth, 49 soil cores were collected (Figs. 4, S6–S9). The mean Pb concentration across 338 core sections of the 49 cores collected from 27 homes was 980 ± 1600 mg/kg Pb ($\pm 1\text{SD}$, median: 580, range: 10–31,000); of these samples 60 % exceeded the current U.S. EPA soil hazard standard of 400 mg/kg for bare soil areas where children play, and 26 % exceeded the 1200 mg/kg Pb guideline. The mean soil-Pb concentration in the UWS (950 ± 1200 , $n = 140$) was similar to the average for Greenpoint (1000 ± 1900 mg/kg Pb, $n = 198$). While Pb concentrations did decline with depth in 30 cores (60 %), they remained extremely elevated relative to background. Average Pb at 20 cm was 1300 ± 1300 (mean $\pm 1\text{SD}$, median: 1100, range: 40–7000 mg/kg Pb, $n = 46$) and at 35 cm was 700 ± 1200 (median: 130, range: 20–4600 mg/kg Pb, $n = 13$). Concentrations of Pb dropped below 100 mg/kg in only 3 cores (6 %) by 20 cm and in 10 cores (20 %) by 40 cm (Fig. S6–S9). In a total of 18 cores, Pb concentrations eventually dropped below 100 mg/kg Pb at an average depth of 33 ± 10 cm. In four cores (8 %), two from UWS and two from Greenpoint, Pb concentrations increased drastically to over 4000 mg/kg with depth. In one core from Harlem, Pb concentrations never increased above 150 mg/kg Pb (Figs. S6–S9).

3.3. Radionuclide inventories and profiles

Core profiles show that both $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs activities reached levels below detection by 15 to 20 cm in all cores (Fig. 4). Activities of $^{210}\text{Pb}_{\text{xs}}$ generally decreased steadily with depth, with the exception of 2 out of 12 cores, one from Greenpoint and one from the UWS. Activities of ^{137}Cs , instead, tended to peak in the 5 to 15 cm depth interval before falling below detection by 20 cm. Nine cores (75 %, $n = 12$) contained a higher ratio of measured to expected inventories of $^{210}\text{Pb}_{\text{xs}}$ than ^{137}Cs . Only two cores had a higher ratio of measured to expected inventories of ^{137}Cs than $^{210}\text{Pb}_{\text{xs}}$; both were collected in Harlem.

Mean uncorrected inventories for the radionuclide $^{210}\text{Pb}_{\text{xs}}$ were 3.5 ± 0.9 kBq/m² ($\pm 1\text{SD}$) in the 12 soil cores, with an average 71 ± 19 % of the expected atmospheric deposition of 4.9 kBq/m². Inventories for $^{210}\text{Pb}_{\text{xs}}$ were very similar across all neighborhoods, with $(3.3 \pm 0.4$ kBq/

Table 1
Descriptive statistics of soil-Pb concentrations collected from composite surface samples in individual backyards (Fig. 3).

Neighborhood	N	Homes	Mean (± 1 SD)	Median	Min	[1st, 3rd] quartile	Max	% > 400	% > 1200
All	370	76	1200 (± 1000)	1000	50	[520; 1500]	9200	81	38
UWS ^a	40	9	1400 (± 2000)	590	140	[370; 1700]	9200	70	38
Harlem	16	4	2600 (± 1300)	2900	870	[1400; 3600]	5100	100	81
Greenpoint	314	63	1100 (± 730)	990	50	[540; 1400]	5020	81	36
Public - Greenpoint	571		250 (± 290)	160	10	[100; 280]	3100	16	2

^a UWS = Upper West Side.

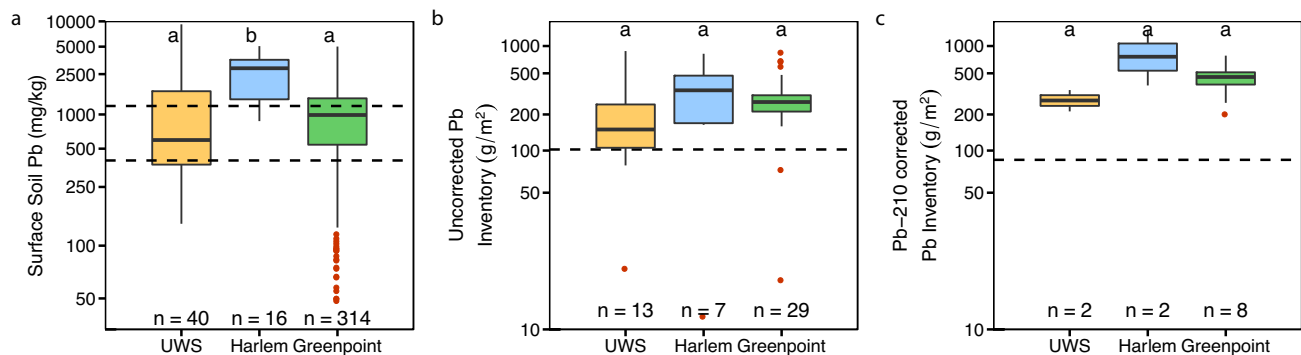


Fig. 3. Distribution of Pb by neighborhood: the Upper West Side (UWS) in Manhattan, Harlem, Manhattan, and Greenpoint and Williamsburg (Greenpoint), Brooklyn for (a) surface soil samples (b) uncorrected Pb inventories, and (c) $^{210}\text{Pb}_{\text{xs}}$ corrected Pb inventories. The dotted line represents in (a) U.S. EPA soil hazard standards of 400 and 1200 mg/kg Pb, in (b) the uncorrected Central Park lake core inventory, and in (c) the corrected Central Park inventory. Different letters represent groups that statistically different.

m^2 $^{210}\text{Pb}_{\text{xs}}$, $n = 2$) in the UWS, (3.7 ± 1.2 , $n = 2$) in Harlem, and (3.5 ± 1.0 , $n = 8$) in Greenpoint (Table 2, Fig. 4). At the same time, the 12 cores contained only 50 ± 30 % of the decay-corrected ^{137}Cs inventory (1.8 kBq/m^2 ^{137}Cs - see Section 2.5), with mean uncorrected ^{137}Cs inventories $0.9 \pm 0.5 \text{ kBq/m}^2$ ($\pm 1\text{SD}$, $n = 12$). Inventories for ^{137}Cs in the UWS ($0.7 \pm 0.04 \text{ kBq/m}^2$ ^{137}Cs , 37 % fallout, $n = 2$) and Greenpoint (0.7 ± 0.3 , 39 %, $n = 8$) were comparable and below expected fallout, but lower than in Harlem (1.9 ± 0.4 , 110 % fallout, $n = 2$) (Table 2, Fig. 4, $p < 0.01$).

3.4. Soil core Pb inventories

After subtracting a background concentration of 26 mg/kg, Pb inventories in the 49 soil cores (60 cm depth) from 27 homes averaged $300 \pm 210 \text{ g/m}^2$ ($\pm 1\text{SD}$), almost three times the uncorrected Central Park lake inventory of 102 g/m^2 Pb (Fig. 5). Mean core Pb inventories were similar across all neighborhoods at $230 \pm 240 \text{ g/m}^2$ ($n = 13$) in the UWS, $350 \pm 280 \text{ g/m}^2$ ($n = 7$) in Harlem, and $310 \pm 190 \text{ g/m}^2$ ($n = 29$) in Greenpoint (Table 3, Fig. 3). These results include all soil collected at each soil interval, including the coarse fraction ($>1 \text{ mm}$ after hand grinding). Core Pb inventories of only the fine fractions ($<1 \text{ mm}$ after hand grinding) were 79 ± 11 % of the total core inventory, although the proportion ranged from 44 % to 95 % (Fig. 5).

3.5. Pb concentrations in the coarse fraction

The coarse fraction included grains, fragments, and rocks $>1 \text{ mm}$ in size that could not be ground by hand with a mortar and pestle across the 338 core depth sections or 49 cores. Mean concentration of Pb in these remaining coarse fractions was significant, at $800 \pm 2000 \text{ mg/kg}$ Pb ($\pm 1\text{SD}$, median: 480, range: 10–31,000). However, the mean and median concentrations were below those in the fine fraction (mean: $1200 \pm 1200 \text{ mg/kg}$ Pb median: 880, range: 20–8400). In comparison to the U.S. EPA soil hazard standards of 400 and 1200 mg/kg Pb, 53 % and 15 % of the coarse fractions exceeded these standards, while 66 % and 36 % of the fine fractions exceeded. Notably, at the high Pb end, the maximum

concentration in the coarse fraction was almost 4 times the fine fraction. Across the core inventories, the coarse fraction represented 21 ± 11 % of the total core inventory, with a range from 5 to 55 % (Figs. 4 and 5).

As the grains in this coarse fraction still visibly retained some of the finer soil particles and dust, the 26 coarse fractions of 6 cores were rinsed and soaked in a sonicator with MQ DI water. While concentrations of Pb generally decreased (Fig. S1b), this decrease was not as pronounced at higher Pb concentrations ($>1000 \text{ mg/kg}$). At high concentrations, remaining individual coarse grains contained several thousand mg/kg Pb (the highest being 6 % or 60,000 mg/kg Pb) (Fig. S2). In addition, analysis of 22 samples from 5 cores ground to a fine powder with a ball mill showed that Pb concentrations remained comparable for the ground and unground coarse fraction (Fig. S1a). The three repeat XRF analyses of the post-milling powder measured 83 % of the Pb concentration in the pre-milling coarse fraction ($r^2 = 0.78$), indicating that Pb was associated with the entire particles, and not just the finer particles on the surface.

3.6. Pb extractions

The field procedure to extract bioaccessible Pb of Landes et al. (2019) extracted on average 28 ± 10 % ($\pm 1\text{SD}$) of the total soil Pb, with a range from 9 to 47 % ($n = 50$). Since the field procedure generally extracts about one third of the bioaccessible Pb that the standard Drexler and Brattin (or EPA 1340 method) would extract, these samples contained very high fractions of bioavailable Pb. Indeed, the subset of 22 samples that were also analyzed by the full Drexler and Brattin method extracted 86 ± 21 % ($\pm 1\text{SD}$) of the total soil Pb, after removing one outlier. More Pb was extracted in the NYC samples than from mining and smelting soils, but less than from large secondary smelters and battery recycling soils analyzed by Landes et al. (2019).

3.7. Other metal analyses

Additional XRF measurements showed that other metal concentrations ranged from 10 to 80 mg/kg for Sn, 60 to 11,000 mg/kg for Zn, and

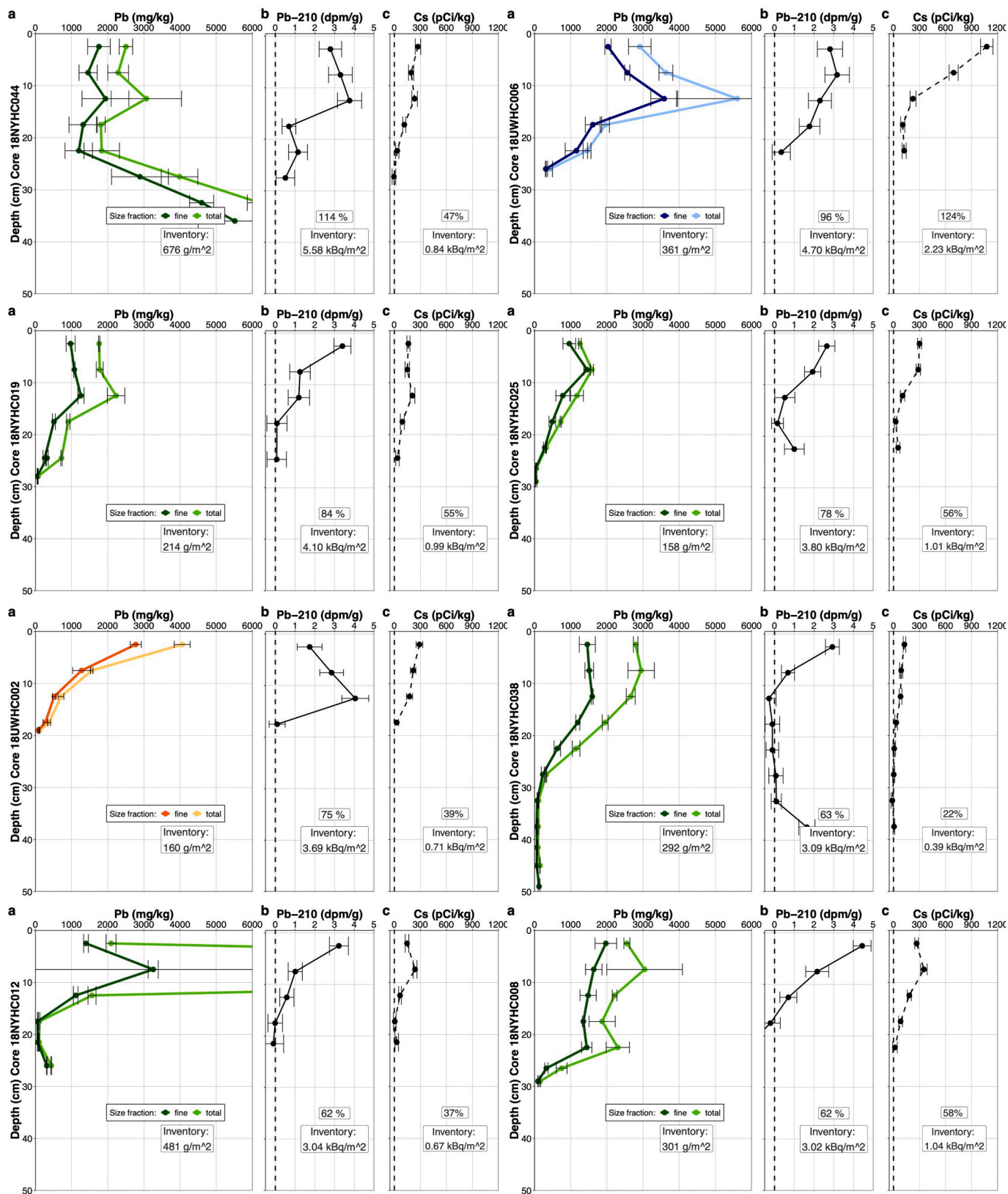


Fig. 4. Depth profiles for cores ordered by the proportion of the ^{210}Pb inventory recovered, including (a) Pb, (b) $^{210}\text{Pb}_{\text{xs}}$ and (c) ^{137}Cs . Pb profiles are shaded by color to indicate neighborhood: Upper West Side (orange), Harlem (blue), and Greenpoint/Williamsburg (green). Uncorrected inventories and proportion recovered are noted in the plot.

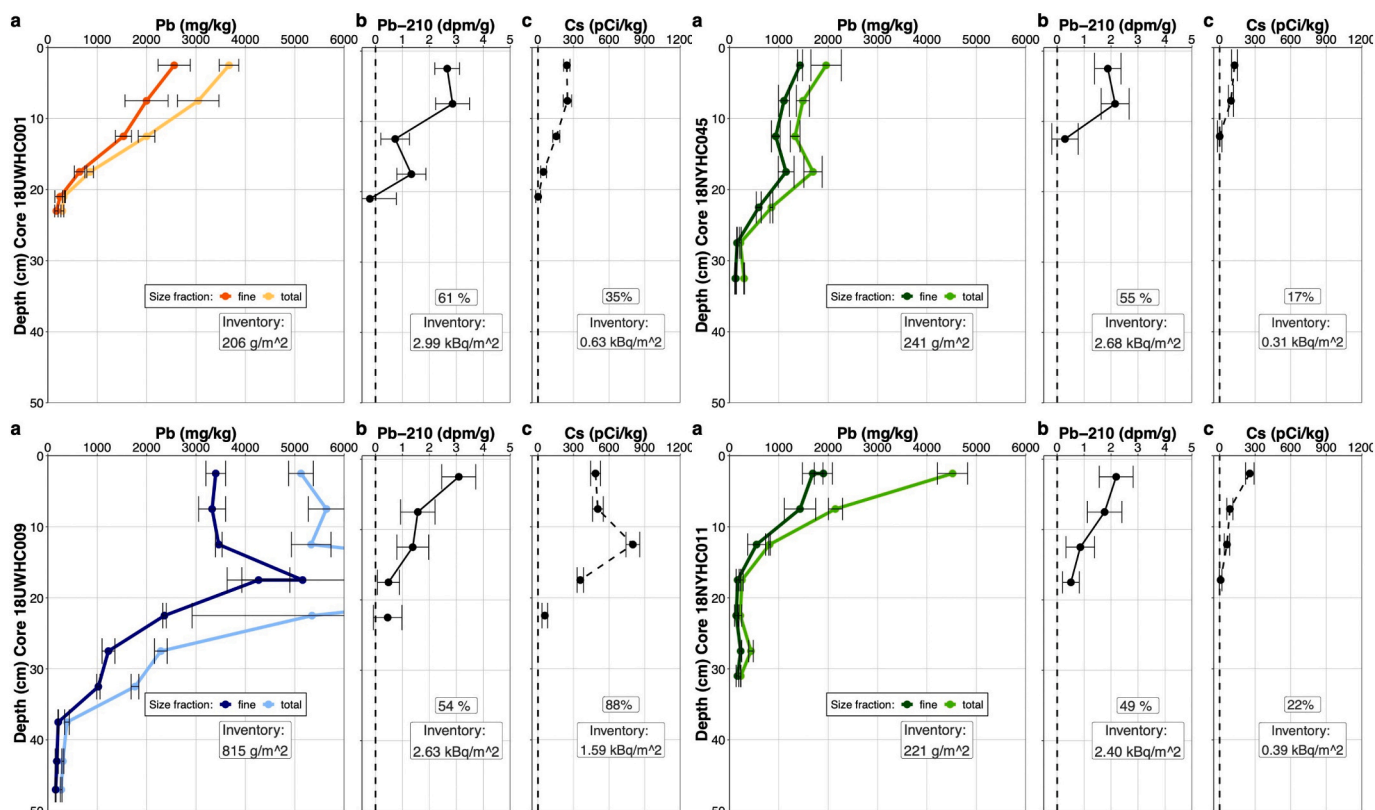


Fig. 4. (continued).

Table 2

Inventories of radionuclides $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs (Fig. 4). Expected fallout was determined 4.9 kBq/m^2 $^{210}\text{Pb}_{\text{xs}}$ and 1.8 kBq/m^2 ^{137}Cs in 2018 (see Section 2.5).

Neighborhood	N	Homes	$^{210}\text{Pb}_{\text{xs}}$ inventory (kBq/m^2)	% expected $^{210}\text{Pb}_{\text{xs}}$	^{137}Cs inventory kBq/m^2	% expected ^{137}Cs
All (this study)	12	12	$3.5 (\pm 0.9)$	71 %	$0.9 (\pm 0.5)$	50 %
UWS	2	2	$3.3 (\pm 0.4)$	68 %	$0.7 (\pm 0.04)$	37 %
Harlem	2	2	$3.7 (\pm 1.2)$	75 %	$1.9 (\pm 0.4)$	110 %
Greenpoint	8	8	$3.5 (\pm 1.0)$	71 %	$0.7 (\pm 0.3)$	39 %
Central Park lake (Chillrud et al.)	1	NA	$5.95 (\pm 0.66)$	120 %	$4.62 (\pm 0.11)$	160 % ^a

^a The expected fallout in 1996 for the Central Park lake core was 2.93 kBq/m^2 ^{137}Cs .

7 to 70 mg/kg for Cd. Mean uncorrected inventories for Sn were $6.6 \pm 8.1 \text{ g/m}^2$ and $5.2 \pm 5.6 \text{ g/m}^2$ ($\pm 1\text{SD}$) for the total and fine fractions, respectively, with inventories in the UWS (3.6 ± 6.8 and 2.3 ± 3.3 (fine), $n = 13$), in Harlem (6.3 ± 5.7 and 5.2 ± 4.7 (fine), $n = 7$), and in Greenpoint (8.2 ± 8.9 and 6.5 ± 6.3 (fine) g/m^2 Sn, $n = 29$) (Fig. S3). No data are reported for arsenic because the instrument's software cannot separate interference from Pb.

4. Discussion

Backyards in both Manhattan and Brooklyn have soil Pb concentrations and inventories considerably higher than those measured in public areas (this study) and previously measured in Central Park (Chillrud et al., 1999). The majority (79 ± 11 %) of the soil Pb inventories was present in the fine (<1 mm) fraction, but this ranged from 44 to 95 %, indicating there was also a significant amount of Pb remaining in the coarse (>1 mm) fraction. Some individual coarse grains contained up to 60,000 mg/kg Pb. The inventories of ^{137}Cs and $^{210}\text{Pb}_{\text{xs}}$ were lower than the expected atmospheric deposition, probably indicating either loss or removal of soils. High Pb concentrations were found at depths both with and without measurable ^{137}Cs and $^{210}\text{Pb}_{\text{xs}}$ activity, indicating both historic (before the early-mid 1900s) and more recent contamination. Combined, these results suggest multiple sources are responsible for the

very high concentrations and inventories of Pb in backyards, including atmospheric deposition, local emissions, and non-atmospheric sources. The data also showed extremely high levels of Pb contamination in backyard soils that are harmful to human health and persist at depth. All NYC residents with soil in their backyard soil should test their soil for Pb or assume it is unsafe for children to play in until proven otherwise.

4.1. Implications of ^{210}Pb and ^{137}Cs activities and inventories

Profiles of $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs activities can be used to assess potential mixing and their presence or absence can provide information on when the soil last had interactions with atmospheric deposition, either as surface soil receiving direct deposition or through mixing with surface soil. Measurements for $^{210}\text{Pb}_{\text{xs}}$ may be more reliable because ^{137}Cs is more mobile, with slow movement downcore, and has a more complex input, with peak fallout in the 1960s and then negligible activities starting in the 1980s (Matisoff and Whiting, 2012). In this study, profiles of $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs activities generally decreased with depth and reached non-detect levels by 15 to 20 cm (Fig. 4). Soil profiles in which $^{210}\text{Pb}_{\text{xs}}$ activities decreased with depth as expected (e.g., 18NYHC019, 18NYHC025, 18NYHC038, 18NYHC012, 18NYHC008, 18UHC009, 18NYHC011), have had less mixing, while soil profiles where $^{210}\text{Pb}_{\text{xs}}$ activities remained constant or increased (e.g., 18NYHC044,

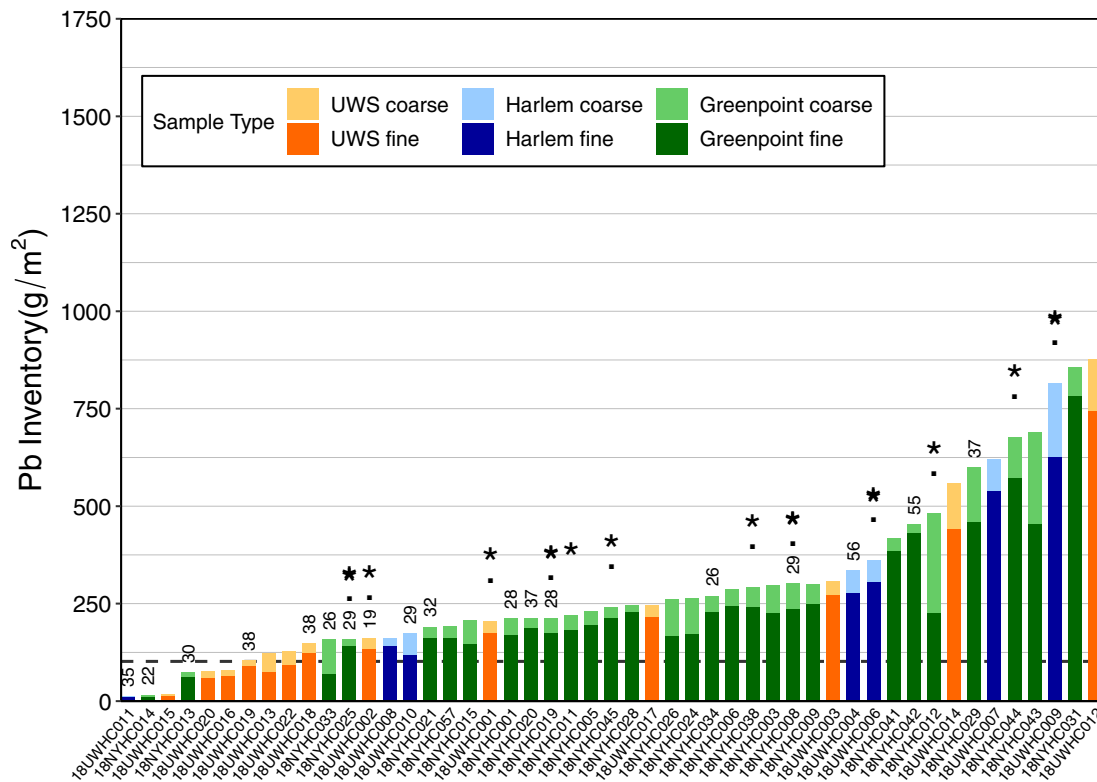


Fig. 5. Pb inventories for 49 soil cores. A star (*) indicates that the ¹³⁷Cs data is within 50 % of the expected decay-corrected fallout, and a dot (•) indicates that the ²¹⁰Pb_{xs} inventory is within 50 % of expected inventory. The dashed line represents the uncorrected Pb inventory of the Central Park lake core from [Chillrud et al. \(1999\)](#). Depth of core is shown in cm if Pb concentrations drop below 100 mg/kg by the bottom of the core. All Pb concentrations were corrected by subtracting the background concentration of 26 mg/kg.

Table 3

Total Pb core inventories (mean ± 1SD) uncorrected for all soil cores. Cores with radionuclide data include corrected inventories to ²¹⁰Pb_{xs} and ¹³⁷Cs fallout data, described in [Section 4.1](#) (Fig. S9).

Neighborhood	N	Homes	Uncorrected Pb (g/m ²)	Pb (g/m ²) corrected to ²¹⁰ Pb _{xs}	Pb (g/m ²) corrected to ¹³⁷ Cs
All	49	27	300 (±210)		
All (with ²¹⁰ Pb _{xs} and ¹³⁷ Cs)	12	12	340 (±210)	510 (±360)	830 (±460)
UWS	2	2	180 (±30)	280 (±90)	500 (±130)
Harlem	2	2	590 (±260)	950 (±800)	610 (±450)
Greenpoint	8	8	320 (±170)	460 (±180)	960 (±490)

18UWHC006, 18UWHC002, 18NYHC045) before decreasing, have had more recent vertical mixing via bioturbation or anthropogenic yard renovations. Profiles of ¹³⁷Cs also generally decreased with depth, but some cores (e.g., 18NYHC012, 18NYHC008, 18UHC009) had a subsurface peak around 5–15 cm. These results are consistent with both ¹³⁷Cs downcore mobility and older peak atmospheric deposition. The presence of more sandy surface soils that don't retain the relatively more mobile ¹³⁷Cs could also impact these cores.

By a depth of 15 to 20 cm, most soil cores had no detectable ²¹⁰Pb_{xs} and ¹³⁷Cs activities, indicating that these soils last interacted with the surface, or mixed with surface soils, approximately 100 years ago (~5 half-lives of ²¹⁰Pb) or 70 years ago (the 1950s for Cs¹³⁷). The high levels of Pb (>1000 mg/kg) deep in the soil core in the absence of detectable radionuclides (e.g., 18NYHC044, 18NYHC008, 18NYHC045, 18UWHC009) indicate contamination that occurred over 100 years ago. In one case, 18NYHC044, Pb concentrations peaked past 30 cm exceeding 4000 mg/kg with no detectable radionuclides, and the soil color at depth was gray instead of light brown (Fig. S10). At the same time, all the cores also showed high levels of Pb (>1000 mg/kg) at shallower depths co-occurring with radionuclides, indicating Pb

addition to soil since the 1950s. If we assume that Central Park lake accurately reflects atmospheric deposition since the 1860s, with increasing Pb deposition starting in 1900 and rapidly increasing between 1920 and 1940, then the soil cores in this study with high Pb levels without detectable ²¹⁰Pb_{xs} and ¹³⁷Cs activities most likely include an additional local source of Pb.

Soil cores in this study contained a higher proportion of predicted fallout for ²¹⁰Pb_{xs} (71 %) than for ¹³⁷Cs (50 %). If Pb inventories were corrected by normalizing to the predicted atmospheric deposition of ²¹⁰Pb_{xs}, then the core Pb inventories averaged 510 ± 360 g/m² (±1SD), eight times the corrected Central Park lake inventory of 64 g/m² Pb (Fig. S9). Neighborhood level Pb inventories ranged three-fold between 280 ± 90 g/m² (n = 2) in the UWS, 950 ± 800 g/m² (n = 2) in Harlem, and 460 ± 180 g/m² (n = 8) in Greenpoint (Table 3, Figs. 3 and 5). Soil core Pb inventories corrected on the basis of the ¹³⁷Cs fallout averaged 830 ± 460 g/m² overall and varied less between neighborhoods at 500 ± 130, 610 ± 450, and 960 ± 490 g/m² for the UWS, Harlem, and Greenpoint, respectively (Table 3). However, this correction would only be valid if the majority of contaminant Pb was atmospherically deposited.

Possible reasons that the core inventories did not reflect the expected ^{137}Cs deposition could be that they did not capture the 1960s peak because of (a) alteration of the soil, especially in the case of raised beds or other fill material, (b) spatial variability in radionuclide atmospheric deposition (Matisoff and Whiting, 2012), and (c) obstructions preventing ^{137}Cs deposition. Variabilities in ^{137}Cs may not be uncommon and were also reported in the Central Park soil cores, with one of the four soil cores containing 4.5 kBq/m², while the other three (3.1 ± 0.9 kBq/m²) closely matched the expected 2.93 kBq/m². This variability extended also to total Pb inventories: uncorrected Pb inventories ranged from 46 to 75 g/m² Pb, and Pb inventories corrected to ^{137}Cs ranged from 25 to 83 g/m² Pb (Chillrud et al., 1999). Given these documented variations in ^{137}Cs inventories and the complexity of individual backyards and urban soils, it was difficult to determine the reason for the lower than expected ^{137}Cs inventories, and corrected inventories should be considered only as one possible scenario. However, these radionuclide activities remained helpful in assessing the depositional history and possible mixing in soils.

4.2. Potential sources of NYC Pb

The extremely elevated concentrations of Pb in surface soils and throughout the soil profile could come from a variety of sources (Laidlaw and Filippelli, 2008), including leaded gasoline (Mielke and Reagan, 1998; Wu et al., 2010), deteriorating lead-based paint (Andra et al., 2006), municipal solid waste incineration (Chillrud et al., 1999), smelting (Landrigan et al., 1975; Factor-Litvak et al., 1999), coal combustion, and other industrial activities (Landrigan et al., 2018) (Laidlaw and Filippelli, 2008). The high concentrations and inventories above what was detected in Central Park support a higher, local source of Pb in addition to fairly uniform regional atmospheric deposition. Average backyard Pb inventories are 3 times (uncorrected) to 8 times (corrected to $^{210}\text{Pb}_{\text{xs}}$) to 13 times (corrected to ^{137}Cs) higher than what was found in the Central Park lake. A plateau of the uncorrected inventories (Fig. 5) between 150 and 300 g/m² (1.5 times and 3 times Central Park) might represent a similar Pb source for these cores, and suggest that on average backyards received 1.5–3 times the Pb input of Central Park lake.

The higher backyard Pb inventory could be a result of focusing atmospheric Pb fallout on roofs and other impervious surface areas (concrete patios, etc.) in neighborhoods. Were this to be the case, however, we would also expect a corresponding increase in the radionuclide fallout of $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs , which was not observed. Since we did not see a focusing of atmospheric deposition, the other explanation could be an additional local source. One local atmospheric source could be leaded gasoline and proximity to roads, since previous studies have reported that the impacts of Pb from leaded gasoline drop off within 40 m of a roadway (Filippelli et al., 2005). However, this would not explain the difference observed between backyards and Central Park, both of which fall within 40 m of present or historical roads. The large variability in Pb inventory, as well as the large and variable fraction contained in the coarse grains, suggest additional, local sources of Pb on top of uniform atmospheric deposition of Pb as fine particles.

4.2.1. Historical industrial activity

Various industries could have contributed to produce heterogenous and elevated Pb inventories, especially in Greenpoint. A map of manufacturing industries in the early 20th century identified industrial zoning for “metals and metal industries” in areas that are now residential (Merchants’ Association of NY, 1922; Li et al., 2018) in Greenpoint, while Williamsburg was zoned for food products and tobacco (Merchants’ Association of NY, 1922). Later, the Plan for NYC (1969) shows land use in Greenpoint and Williamsburg dominated by light and heavy industry around the East River and Newtown Creek, with pockets of low-to-high density residential zoning in the center along Manhattan Avenue in Greenpoint and Metropolitan Avenue in Williamsburg (NYC Planning Commission, 1969a). Land use in the UWS was all designated as low-

medium density residential, with commercial areas along Broadway and Amsterdam Avenue, and in Harlem as high-medium density residential with commercial areas along 125th St (NYC Planning Commission, 1969a). While the UWS was described as “Sound” in a map of Neighborhood Improvement Investment strategies, Harlem was noted for “Major Action,” and Greenpoint and Williamsburg were noted for “Mixed Residential-Industrial,” and “Major Action” (NYC Planning Commission, 1969b). While this industrial activity may have contributed to the heterogeneity of soil Pb and some of the deeper Pb peaks, it is likely not the primary source of Pb as concentrations in the residential UWS are similar to the historically industrial Greenpoint (Fig. 2).

4.2.2. High Pb in large grains

The presence of high Pb in individual grains within the coarse fraction, including a grain that contained 1.5 % Pb and one that contained 6 % Pb, as well as pieces of bricks and coal, suggest additional, local sources of Pb (Fig. S2). In four cores, Pb concentrations increased drastically at depth (>4000 mg/kg Pb) suggesting that these yards had high-Pb fill material added (Fig. S6–S9). In one case (core 18NYHC044), the bottom core intervals enriched with Pb are all gray and contain ash-like material (Fig. S10), potentially indicating the dumping of combusted material high in Pb. Historic use of coal, and potential dumping of bottom ash in backyards, could be another source of Pb. However, past studies show that coal is not a significant contributor to atmospheric Pb (Murozumi et al., 1969; Chillrud et al., 1999) and that Pb concentrations in coal fly ash are around 100 mg/kg Pb (Twardowska et al., 2003). Pieces of coal from our soil cores (Fig. S2) measured 26 mg/kg Pb by XRF, similar to a coal specimen borrowed from the Barnard Geologic Library. The concentration of 6 % Pb in the 5–10 cm interval of sample 18NYHC012 was confirmed by laser-ablation ICP-MS in a small grain. Other rocks and grains analyzed only by XRF contained several thousand mg/kg Pb. These findings suggest possible non-atmospheric sources of Pb that could be heterogeneously distributed throughout soils based on actions at individual properties.

Another potential non-atmospheric source would be deteriorating Pb-based paint. However, paint chips would have been easily ground by mortar and pestle, and thus not remain in the coarse fraction. Furthermore, we did not see the expected higher Pb concentrations closer to potentially painted surfaces such as buildings, fences, and sheds (Fig. S5), and we did not see an association with building age; almost all homes tested were built before 1960 (Fig. S12). This suggests Pb-based paint is not the main non-atmospheric source of Pb.

4.2.3. Waste incineration

Municipal refuse (trash) incineration increased throughout the early 20th century in North American cities until new air pollution controls began in the 1960s, leading to the phase-out of these waste incinerators. In New York City, the number of municipal solid-waste (MSW) incinerators decreased from 11 in 1969 to 8 in 1981 to 0 in 1990 (Walsh et al., 2001). Chillrud et al. (1999) showed that in Central Park, MSW incineration was the major contributor to atmospheric Pb deposited over the 20th century. Concentrations of Pb in waste incinerators have been summarized as 81,000 mg/kg in incinerator emitted particles, 4000 mg/kg in fly ash (incinerator particles retained), and 5000 mg/kg in incinerator residue (Walsh et al., 2001). If the source of Pb were primarily atmospheric and representative of central NYC air, we would expect to find Pb inventories and Pb/Sn ratios in our soil cores similar to those found in Central Park. The Central Park lake core was characterized by a tin (Sn) to Pb ratio of 1:19 (Chillrud et al., 1999). Extending the association to inventories would suggest that Pb inventories would be approximately 20 times higher than Sn inventories. However, the majority of our cores contained more Pb than would be expected based on the Sn concentrations alone (Fig. S3). Only 9 (18 %) soil cores were within 20 % of the Sn-estimated Pb inventory and 14 (29 %) were within 50 % ($n = 49$). Furthermore, Walsh et al. summarize published Sn/Pb ratio in MSW ranging from 1:2.8 in fly ash to 1:11 in incinerator residue,

and 1:7 in emitted particles. These ratios would suggest much higher Sn concentrations, given the Pb concentrations, than observed in this current study.

Municipal incinerators, however, were not the only combustion-based form of burning refuse in 20th century NYC; Walsh et al. (2001) identified 17000 apartment building waste incinerators, or multi-dwelling domestic waste combustors (DWC), in addition to the 32 municipal solid-waste (MSW) incinerators. Incinerators of the DWC often had higher particle emissions due to less ideal maintenance and operating conditions, such as low and variable temperatures, air intake rates, waste amount, and manner of waste addition (Walsh et al., 2001). Given the high variety of refuse and combustion efficiency of individual apartment DWCs, these could represent additional, local sources of Pb and other metals that were not representative of central NYC air, in addition to local smelters, foundries, or MSWs.

Considered altogether the data point to multiple sources that contributed to soil Pb in New York City. Deposition from leaded paint and gasoline occurred but are probably not the primary sources. Soils and areas that became backyards were probably historically dumping grounds for waste, including disposal from manufacturing, smelting, and fly ash and bottom ash. On top of this both domestic and municipal incinerators could have contributed to localized atmospheric deposition. These concentrations are not seen in the public samples as soil has been replaced or the areas were historically protected from dumping. The multiple sources and variability highlight the difficulty in predicting where backyards may have elevated Pb concentrations; this underscores the importance of testing soil at individual homes, especially where children play.

4.3. Soil Pb hazard

Regardless of the exact source of Pb in these urban soils, this study highlights the health hazard posed by soils in urban areas with an industrial history. The average concentrations reported here (1200 ± 1000 mg/kg Pb, median: 1000, $n = 370$) exceed previously published results for NYC soils (600 ± 770 mg/kg Pb, median: 360, $n = 1657$) (Cheng et al., 2015; Pavilonis et al., 2020), and most other cities (Datko-Williams et al., 2014). Extremely concerning is the high proportion of soils that exceeded 1200 mg/kg Pb (38 %) and 400 mg/kg Pb (81 %) (Figs. 2 and 3). Median concentration of home soils was also higher than those previously reported for NYC home gardens (median: 630 mg/kg Pb, $n = 197$) (Cheng et al., 2015) or community gardens (median: 140 mg/kg Pb (Cheng et al., 2015), and median: 102 mg/kg Pb (Mitchell et al., 2014)). The new data show that mean concentrations of soil Pb in backyards of homes are much higher than previously thought and emphasize the need to focus on residences and backyard soil to address the public health risk from contaminated urban soil.

No clear pattern was seen between the distribution of Pb in soil samples collected near structures or in areas that appeared well cared for and maintained (Fig. S5). Across all samples collected, Pb concentrations in raised beds are lower than samples from gardens in non-raised beds. However, this category still contained high Pb soils, including the sample with the highest soil-Pb concentration (9200 mg/kg Pb) from a concrete raised bed that was reportedly created over 50 years ago and had remained largely undisturbed since.

4.3.1. Implications for child exposure

To assess how these high soil Pb levels might relate to child exposure, we compare census-tract level data for soil, mean soil Pb, to child blood lead levels (BLL) data from U.S. CDC (Schneyer and Pell, 2017) (Fig. S11). In Greenpoint, census tracts with the lowest child BLL (2.5 to 5 $\mu\text{g}/\text{dL}$) also had lowest mean home soil-Pb concentrations (<150 mg/kg), while the census tracts with higher BLL (7.5 to 10 $\mu\text{g}/\text{dL}$) had higher mean soil Pb (>400 or 1200 mg/kg). This is consistent with a recent study comparing park soil Pb in Brooklyn to child BLL at the neighborhood scale (Pavilonis et al., 2022). At the same time, BLLs in the UWS

were lower, on average, than those in Greenpoint, but mean soil-Pb concentrations were not different between these two neighborhoods (Fig. 3). The building types, however, were very different between these two areas (Fig. S12): homes in the UWS tended to have >20 units per home, whereas homes in Greenpoint and Harlem tended to have 4 units per home. The number of units per home can be used to estimate building height, and, correspondingly, how likely it is that individuals have access to the backyard for direct contact with soil and how much soil is likely to be tracked inside or enter via resuspension. While neighborhood and building characteristics may explain the lack of association between census tract Pb and child BLL data, it is important to note that no child should be playing in soils over 400 mg/kg Pb. Fortunately, Pb content of public areas (this study) and parks are generally much lower (Pavilonis et al., 2020).

5. Conclusions

Elevated concentrations of Pb were found throughout New York City surface soils averaging 3 to 20 times the cleanup level of 400 mg/kg Pb. Variable inventories of Pb, $^{210}\text{Pb}_{\text{xs}}$, and ^{137}Cs suggest that contributions to soil Pb are not easily constrained to a single source, and Pb inventories exceeding the integrated atmospheric contributions and those found in a Central Park, imply non-atmospheric contributions are also important. Additionally, elevated Pb concentrations in the coarse fraction and grains of up to 6 % Pb, reveal that non-atmospheric sources are not limited to paint but may include industrial waste, trash incineration, and other fill material. The Pb concentrations in residential surface soils tested in this study were extremely high and not safe for children to play in. Concentrations from home soil samples exceeded those from public spaces and published concentrations of community garden soils and other New York City soils. These results indicate that any mapping of urban soils that does not explicitly test a range of samples from backyards may not capture the public health threat posed by these soils. All residents with access to a backyard should test their soil for Pb, especially in areas where young children play. In addition, we recommend expanding municipal programs that test residential water for Pb to include residential soil testing, as well as expanding programs that provide and distribute clean soil to residents.

CRedit authorship contribution statement

Franziska C. Landes: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization, Project administration. **Margaret Calcio:** Investigation, Visualization. **Jennifer M. Sobolewski:** Investigation. **Kelcey Wallens-Logan:** Investigation. **Anna Ledeczi:** Investigation. **Yael Kiro:** Investigation, Resources. **Louise Bolge:** Investigation. **James Ross:** Investigation, Resources. **Steven N. Chillrud:** Resources, Writing – review & editing. **Brian J. Mailloux:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision, Funding acquisition. **Alexander van Geen:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data is attached as "Research Data" in the Supplementary Materials.

Acknowledgments

This work would not have been possible without the support of Neighbors Allied for Good Growth (NAG), now North Brooklyn Neighbors, and especially Allison Currier, Lisa Bloodgood, and Alan Minor. We thank our colleagues at the Urban Soil Institute for inviting us to join them and NAG for soil testing workshops at the McCarren Park Farmer's Market. We are also grateful to all the residents who agreed to participate in this study and apologize to those interested we were unable to provide a soil test. The LA-ICP-MS analysis was conducted as part of Terry Plank's Geochemistry for a Habitable Planet Course. Finally, we appreciate the additional undergraduate students who helped collect, process, and analyze soils, including Abigail Adams, Julia Weingarden, Clare McGillis, Afsana Akter, Sidney Rojas, Sara Ortiz from Brian Mailloux's Water Sanitation and Health Course. This work was partially supported by NIEHS grants P42 ES010349 and P30 ES009089.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.165407>.

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