



Identification of Legacy and Active Sources of Metal Contamination in Soils in Brooklyn, NY

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Abstract

The purpose of this study was to examine the spatial distribution and potential anthropogenic sources of lead (Pb), zinc (Zn), copper (Cu), manganese (Mn), and iron (Fe) in surface soils throughout Brooklyn, NY. We collected soil samples ($n = 1,373$) from 176 different New York City parks. Samples were analyzed ex-situ using a portable X-ray fluorescence with a subset of samples laboratory confirmed. The effect of multiple sources on concentrations were determined by multivariable linear regression with generalized estimating equations. Median concentrations of Pb, Zn, Cu, Fe, and Mn were 108 ppm, 145 ppm, 49 ppm, 14,034 ppm, and 279 ppm, respectively. All metals were significantly correlated with one another ($p < 0.001$), with the strength of the correlation ranging from a low of approximately $\rho = 0.3$ (Pb-Mn and Zn-Mn) to a high of $\rho = 0.7$ (Pb-Cu). In final multivariate modeling significant association were observed between scrap yards and Mn concentration ($\beta = 0.075$, 0.019), National Priorities List (NPL) sites and Pb, Fe and Mn ($\beta = 0.134$, $p = 0.004$; $\beta = 0.038$, $p = 0.014$; $\beta = 0.057$, $p = 0.037$, respectively), and bridges nearby and Pb and Zn ($\beta = 0.106$, $p = 0.003$; $\beta = 0.076$, $p = 0.026$, respectively). Although manufacturing and industry have mostly left the area, smaller scrap metal recyclers are abundant and associated with increased Cu and Mn soil concentrations. In addition, NPL sites contributed to increased concentrations of all five metals within 800 m. Roadways have long been established to be sources of urban pollution; however, in our study we also found the presence of bridges within 800 m were also strongly predictive of increased Pb, Cu, and Zn concentrations.

Soil is a sink for anthropogenic metal pollution (Cheng et al. 2015; Sager 2020) that often requires remediation to reduce exposure to the surrounding community (Hou et al. 2017; Wong et al. 2006). Anthropogenic metals in soil are generally more bioavailable than natural sources, which increases adsorption and body burden (Hou et al. 2017). Contaminated soil can expose urban populations via resuspension, especially during dry summer periods (Laidlaw et al. 2016; Li et al. 2016). Consequently, children's blood lead levels (BLL) were found to be significantly higher in dry summer months in an area with widespread lead (Pb) contaminated soil (Zahran et al. 2013). Soil metal levels can also serve as indicators of active environmental pollution generated within the community. Current sources of metal pollution include emissions from power plants, manufacturing and industrial sites, bridges, waste incineration, and vehicles

(Aelion and Davis 2019; Gottesfeld 2015; Haghazari et al. 2021; Li and Shao 2009; Maher et al. 2008; Xiao-Li et al. 2006; Zhang et al. 2011). Moreover, Brownfield sites and deteriorating buildings can also release metals into the environment (Jennings et al. 2002; Yesilonis et al. 2008).

Like many urban areas within the United States (US), Brooklyn, New York (NY) was once home to a vast manufacturing and industrial sector that has largely ceased operations. Although, these operations have left the borough the legacy of environmental metal contamination continues to expose residents (Fecht 2017; North Brooklyn Neighbors n.d.-a). In the 1800s, factories opened along Brooklyn's East River waterfront that propelled it to become one of the nation's leading manufacturers of processed goods. Coal was used to power many of these factories, releasing metals into the environment (Brooklyn Legends n.d.; North Brooklyn Neighbors n.d.-a). At the turn of the twentieth century, Pb smelters and incinerators operated in parts of Brooklyn (Brooklyn Legends n.d.). Various types of debris were used to fill marshlands, raise land elevation, and expand waterfront property in Brooklyn. This fill was composed of materials containing metals and may account for

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the spatial heterogeneity in metals concentration throughout the borough (North Brooklyn Neighbors n.d.-b). Over the past 20 years, industrial and manufacturing land has been rezoned and a large influx of new residents have migrated to areas of Brooklyn that have a history of contamination sources that may lead to increased exposure risk (Pavilonis et al. 2020).

Metals such as zinc (Zn), copper (Cu), manganese (Mn), and iron (Fe) are essential nutrients that are required for various biochemical and physiological functions, but can exhibit an “U” shaped dose response relationship (World Health Organization 1996). Lead, on the other hand, is a toxic metal with no biological requirement in humans nor exposure threshold for harm (Abadin et al. 2020). Exposure to high concentrations of all five metals are associated with various negative health effects ranging from neurological to immunological. High levels of Mn exposure can cause irreversible neurological disease, including behavioral changes and a lowered intelligence quotient (IQ) as well as a condition called manganism that presents with symptoms similar to Parkinson’s (Davis et al. 1999; Mergler 1999; Neal and Guilarte 2013; Silbergeld 1999; Zayed et al. 1999). Occupational inhalation exposure to Zn has long been known to cause metal fume fever, which is characterized by acute influenza like symptoms (Graeme and Pollack Jr 1998; Roney 2005). Long-term exposure to Cu dust can cause upper respiratory irritation, (Dorsey and Ingerman 2004) while high Fe exposure can increase the risk of cancer through the generation of free radicals (Guo et al. 2015). All organ systems in the body are negatively affected by the cumulative exposure to Pb, regardless of the exposure route, and can result in adverse effects to the neurologic and nervous system, slowed growth and development, and learning and behavioral problems. The neurological effects linked to Pb exposure appear to be permanent (Bandeem-Roche et al. 2009; Canfield et al. 2003; Dietrich et al. 2001; Koller et al. 2004; Lanphear et al. 2005).

While most exposure assessments look at exposure to individual metals, there are potential additive and synergistic health effects from exposure to multiple harmful metals (metal mixtures), including oxidative stress, cardiovascular, and neurological effects (Liu et al. 2018a, 2018b; Rai et al. 2010). Metal mixtures can disrupt the blood brain barrier and induce a synergistic toxicity in astrocytes (Rai et al. 2010). Short-term inhalation exposure to urban particulate matter laden with multiple metals was associated with increased systemic inflammation and oxidative stress (Liu et al. 2018a). Increased fetal exposure to metal mixtures has also been associated with oxidative stress and congenital heart disease (Kundak et al. 2017; Liu et al. 2018b).

A previous study conducted by some of the authors on this paper found a positive ecological association between soil Pb levels and elevated pediatric BLLs in Brooklyn

(Pavilonis et al. 2021). This study seeks to expand upon that finding and characterize the spatial distribution and identify potential active and legacy sources of Pb, Zn, Cu, Mn, and Fe in surface soils in Brooklyn, NY. Identifying key sources of metal contamination can help predict areas with elevated metal levels and direct the implementation of programs designed to mitigate risk to residents.

Methods

Soil Sampling

Between June 15 through August 15, 2020, we collected soil samples ($n = 1,373$) from 176 different New York City (NYC) Department of Parks and Recreation properties (NYC Parks), which included community parks, neighborhood parks, and triangles/plazas. The parks where sampling took place varied in size, with areas ranging from 4.05 to 876,771 m^2 . Sample locations were recorded and ESRI ArcGIS Collector (Redlands, California) was used to manage locational data. Soil measurements were collected in a grid pattern when possible. However, the greenspaces in this study were unevenly shaped polygons with obstructions such as hard surfaces, playground equipment, and water. As a result, the sampling scheme was adjusted as necessary. A 35 m by 35 m grid was created and overlaid on a map. Prior to sampling, locations for soil collection within the grid were chosen based on the physical size and features of the park. If any obstruction was located within a grid it was skipped. In parks larger than 100,000 m^2 , 30 samples were collected per park; in parks between 10,000 m^2 and 100,000 m^2 , a median of 14 samples (between 7 and 30 samples) were collected; in parks between 1,000 m^2 and 10,000 m^2 , a median of four samples (between 1 and 8 samples) were collected; and in community parks and plazas less than 1,000 m^2 , one sample was collected.

Soil Analyses with pXRF

For each soil sample, 50–200 g were collected at 0–5 cm depth with a clean trowel and transferred to a clean Zip-lock plastic bag. Each sample was screened for Pb, Cu, Zn, Mn, and Fe concentrations ex-situ with a portable X-ray fluorescence (pXRF) instrument (Thermo Scientific Niton, model XL3t) set to soil mode. A 30 s exposure time was used for each measurement, which was taken directly on each sample bag containing soil. Prior to each use, a system check was carried out on the instrument, and before each sampling session, a series of soil standards reference materials, produced by The United States Geological Survey (USGS) and the National Institute of Standards and Technology (NIST), were measured with the pXRF. These measurements had

Table 1 Certified values for soil standard reference materials

Soil Standard Reference Material	Certified Values (ppm)				
	Pb	Zn	Cu	Fe	Mn
SiO ₂	0	0	0	0	0
NIST 2709a	17.3 ± 0.1	103 ± 4	33.9 ± 0.5	33,600 ± 700	529 ± 18
USGS SdAR-M2	808 ± 14	760 ± 13	236 ± 4	*	*
NIST 2780	5770 ± 410	2570 ± 160	215.5 ± 7.8	27,840 ± 800	462 ± 21

*Metal was not in the soil standard reference material

Table 2 Agreement between ICP-MS and pXRF by metal

Metal	Slope (ppm)	r ²	Mean relative % difference
Pb	0.976	0.94	7.5
Zn	0.913	0.90	30.4
Cu	0.890	0.91	15.9
Fe	0.752	0.95	35.5
Mn	0.735	0.88	35.5

to fall within 2 standard deviations (SD) of the reference value. If a reading fell outside 2 SD but within 3 SD, another reading was needed and, if acceptable, data collection could start. If the reading was not acceptable, a system recheck was needed. The certified values for SiO₂ blank, NIST 2709a, NIST 2780, and USGS SdAR-M2 can be seen in Table 1. The limit of detection (LOD) for each metal was calculated from 1.5 times the precision of the measurement, which was unique for each measurement and two times the SD (Thermo Scientific). The LODs ranged from 11.5 to 19.3 ppm for Pb (n = 31), 28.8 to 68.4 ppm for Cu (n = 232), 79.2 to 127.4 ppm for Mn (n = 13), and 19.7 to 23.6 ppm for Zn (n = 2). No Fe readings were below the LOD.

Confirmation Laboratory Analyses with ICP-MS

A subset of samples (n = 350) were validated using inductively coupled plasma mass spectrometry (ICP-MS) analyses. Samples were first dried to constant weight at 105 °C, disaggregated using a mortar and pestle, and then sieved to less than 2 mm. Approximately 0.3 g of this fine fraction was acid digested using a microwave oven digester following the

US Environmental Protection Agency (EPA) Method 3051 (US EPA 2007). The digested samples were then analyzed for metals using an ICP-MS (Perkin Elmer, Elan DRce) at Brooklyn College (EPA Method 6020). Results from the ICP-MS analyses corroborated the findings obtained with the pXRF, with high degree of agreement between the methods, although the pXRF did slightly underestimate metal concentrations. The slope, r² and mean relative percent difference for each metal are shown in Table 2.

Geospatial Analyses

We investigated several potential sources of metals in the soil: scrap yards, National Priorities List (NPL) sites, Toxic Release Inventory (TRI) sites, bridges, manufacturing/industrial land use, new construction, above-ground subway routes, and roads. The influence of these sources on metal concentrations in soil samples were operationalized in two ways. For relatively uncommon features (e.g., scrap yards), a binary approach using fixed-distance buffers was employed (i.e., the soil sample was “near” or “not near” the source). For features which are relatively common or ubiquitous (e.g., roads, industrial land use) a continuous approach using either kernel density estimation (KDE; varying intensities based on density of the potential source) or simple linear density (e.g., miles of roadway per unit area) were used. ERSI ArcGIS (ArcMap version 10.7.1 and Pro version 2.8.3) was used for geospatial analyses.

The binary approach was applied to scrap yards, NPL and TRI sites, and bridges. We created an indicator for “scrap yard” that included: steel fabricators, scrap metal recycling centers, scrap metal dealers, salvage yards, household waste recyclers, and construction waste recycling centers. Scrap

Table 3 Descriptive statistics for soil concentrations (ppm) by metal

Metal	Mean	Standard Deviation	Median	25th Percentile	75th Percentile	Minimum	Maximum
Pb	143	142	108	61	180	8	1840
Zn	188	194	145	105	212	13	4827
Cu	61	43	49	35	70	19	505
Fe	14,269	4268	14,034	11,852	16,408	1366	48,545
Mn	295	171	279	219	351	53	4827

yards were located via Google Maps and geocoded. Proximity buffers of 400 m (~ 1/4 mile) were then created to identify nearby soil samples. NPL and TRI sites were downloaded from the US EPA, geocoded, and filtered to the study area (US EPA 2020). Data on bridges were acquired by selecting only the appropriate features from the NYC Department of City Planning's Linear Integrated Ordered Network (LION) dataset via the NYC Open Data portal (NYC Department of City Planning 2022). Distance buffers of 800 m (~ 1/2 mile) were generated to characterize soil sample proximity to each of the NPL, TRI, and bridge features. Buffer distance was selected based on overall size of the facility and identification by the EPA (Maantay et al. 2008).

The continuous approach was applied to industrial/manufacturing land uses, new construction (since 2010), above-ground subway routes, and roads. Information about roads were derived from the LION dataset (NYC Department of City Planning 2022) with densities calculated as linear length of all roadways within a 400 m buffer of each sample location divided by the area of the buffer. Tax lots where (1) the building class is manufacturing and industrial land use, as well as (2) new construction since 2010 were extracted from the Primary Land Use Tax Lot Output (MapPLUTO) dataset and downloaded via NYC Open Data (NYC Department of City Planning 2022). A continuous KDE surface was produced to quantify the density of new construction and industrial/manufacturing land use. It was parameterized using an 800 m bandwidth using building area as the "intensity" value (i.e., larger building area results in higher density estimates). Above-ground subway routes were derived from subway route and station data collected from the Baruch College Geoportal (Baruch College n.d.). Routes were considered to be "above ground" if they were between two stations which were "at grade" or "elevated". Routes between one above-ground and one below-ground station were bisected, with the section closer to the above-ground station designated as "above ground". These were then converted into KDE surfaces using the same parameterization as the tax lots, however without an added "intensity" value (i.e., it is simply the length of the subway route which impacts the resulting density values).

Statistical Analyses

Metal concentration data were right skewed and therefore log-transformed (base 10) to better approximate normal distributions and confirmed from QQ plots for multivariable linear regression models. We assessed the bivariate associations among variables using nonparametric statistical methods which included Spearman's rank correlations for two numeric variables (e.g. the correlation between the five metals and between each metal with numeric predictors) and the Wilcoxon rank-sums test to determine whether the metal

distributions for sampling locations with and without the attribute of interest were the same. We did not control for repeated samples within parks in bivariate analyses.

We assessed the association between each metal and hypothesized contamination sources using backwards stepwise multivariable linear regression with generalized estimating equations (GEE) to adjust for clustering ($n = 176$) of soil samples, since multiple samples were collected at most parks. All independent variables were included in the initial model and the variable with the largest p-value was removed and the model rerun repeatedly until all variables left had p-values < 0.20 . Data analyses were conducted using SAS statistical software (version 9.4, Cary, NC).

Results

Generalized spatial trends of metal concentrations are shown in Fig. 1. The highest metal levels were observed along the periphery of Brooklyn, in the North along the East River waterfront, in the Southwest, and in the East. Metal concentrations were highly right skewed, with two to three orders of magnitude difference between the minimum and maximum concentration for all five metals. Iron was found in the greatest abundance, while soil Cu concentrations were the lowest. Median concentrations for Pb, Zn, Cu, Fe, and Mn were 108 ppm, 145 ppm, 49 ppm, 14,034 ppm, and 279 ppm, respectively (Table 3).

Soil guidance values for various agencies, background levels, and percentage of samples in exceedance are shown in Table 4. Generally, samples had larger Pb, Cu, and Zn concentrations compared to US median background concentrations, while Fe and Mn concentrations were smaller (Smith et al. 2014). Few samples exceeded the US EPA Regional Screening Levels (US EPA 2021b) and NY Department of Environmental Conservation Cleanup Objectives (NYS DEC 2020). The Canadian Council of Ministers of the Environment (CCME) (Canadian Council of Ministers of the Environment 2021) has more conservative soil quality guidelines for parklands compared to the US EPA and NY State. Approximately one third of Pb and Cu concentrations exceeded the CCME guidelines and around 20% of Zn samples were larger.

All metals were significantly correlated with one another ($p < 0.001$), with the strength of the correlation ranging from a low of approximately $\rho = 0.3$ (Pb-Mn and Zn-Mn) to a high of $\rho = 0.7$ (Pb-Cu) (Table 5). Manufacturing/industrial land density and road density were significantly correlated with the five metals, with the strength ranging from $\rho = 0.08$ (manufacturing/industrial density with Pb) to $\rho = 0.28$ (road density with Pb, Cu, and Zn). The only exception was the correlation between road density and Mn, which was not significant. There was a significant positive correlation

Fig. 1 Metal concentrations in soil. Continuous surfaces of metal concentration estimate-extrapolated from sample locations (bottom right map) using empirical Bayesian Kriging. All estimates are shown in deciles (first decile is lightest shade; 10th decile is darkest shade). Note that the surfaces were created for the visualization of generalized spatial trends and were not used for statistical analyses

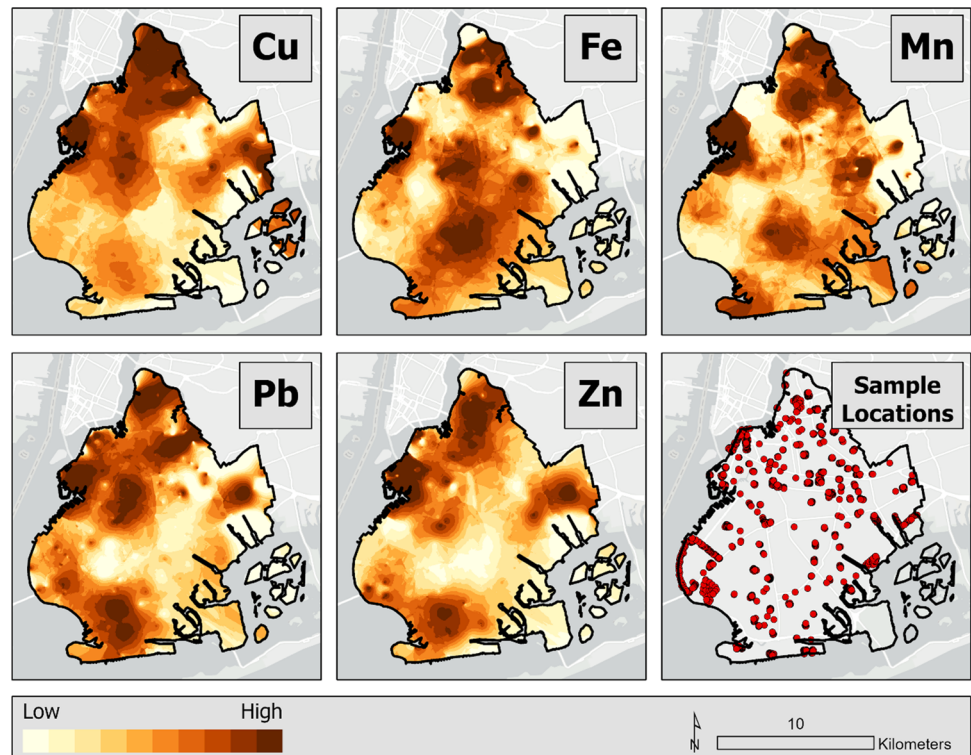


Table 4 Soil guidance values (ppm) and percent of samples in exceedance in parenthesis

Metal	Median USGS Soil Background Levels	US EPA Regional Screening Levels	NY Department of Environmental Conservation Cleanup Objectives	CCME Soil Quality Guidelines
Pb	18.1 (95.8)	400 (4.2)	^a	140 (36.2)
Zn	58 (96.7)	23,000 (0)	660 (2.0)	250 (17.3)
Cu	14.4 (100)	3100 (0)	280 (0.7)	63 (32.4)
Fe	19,500 (7.6)	55,000 (0)	^a	^b
Mn	492 (3.9)	1800 (0.07)	2000 (0.07)	^b

^a = Cleanup objective not promulgated by NY State

^b = Soil Quality Guidelines for the Protection of Environmental and Human Health not promulgated by Canadian Council of Ministers of the Environment (CCME)

between construction density and Zn, Cu, and Fe ($\rho=0.16$; $\rho=0.20$; $\rho=0.07$, respectively). Subway density was not significantly correlated with any of the metals. In addition, the concentration of Pb was significantly higher in areas near TRI sites and bridges (median concentrations: 114 ppm versus 105 ppm, $p < 0.001$; 147 ppm versus 95 ppm, $p < 0.001$, respectively), as was Fe (14,421 ppm versus 13,845 ppm, $p=0.003$; 14,446 ppm versus 13,898 ppm, $p=0.008$, respectively). Copper concentration was significantly higher in locations near NPL and TRI sites, scrap yards, and bridges (Median concentration: 67 ppm versus 49 ppm, $p < 0.001$; 57 ppm versus 45 ppm, $p < 0.001$; 75 ppm versus 49 ppm, $p < 0.001$; 58 ppm versus 46 ppm, $p < 0.001$, respectively), as was Zn concentration (median concentration: 156 ppm

versus 144 ppm, $p=0.008$; 161 ppm versus 135 ppm, $p < 0.001$; 181 ppm versus 144 ppm, $p=0.001$; 165 ppm versus 141 ppm, $p < 0.001$, respectively). Mn concentration was significantly higher near NPL sites and scrap yards (median concentration: 314 ppm versus 278 ppm, $p=0.036$; 334 ppm versus 277 ppm, $p < 0.001$, respectively).

In the final backwards stepwise multivariable regression model, all predictor variables were positively associated with log-transformed metal concentrations except new building construction density which was significantly negatively associated with Pb ($\beta = -2.51E-08$, $p=0.030$) and Cu ($\beta = -1.23E-08$, $p=0.022$). Road density was significantly associated with Pb ($\beta = 0.008$, $p < 0.001$), Cu ($\beta = 0.005$, $p < 0.001$), Zn ($\beta = 0.005$, $p=0.026$), and Fe

Table 5 Unadjusted associations for continuous and categorical variables

Variable	Total n	Pb		Zn		Cu		Fe		Mn	
		Measure	p-value	Measure	p-value	Measure	p-value	Measure	p-value	Measure	p-value
Pb, S _p	1373	1		0.60	<0.001	0.66	<0.001	0.54	<0.001	0.32	<0.001
Zn, S _p		0.60	<0.001	1		0.60	<0.001	0.39	<0.001	0.32	<0.001
Cu, S _p		0.66	<0.001	0.60	<0.001	1		0.47	<0.001	0.36	<0.001
Fe, S _p		0.54	<0.001	0.39	<0.001	0.47	<0.001	1		0.64	<0.001
Mn, S _p		0.32	<0.001	0.32	<0.001	0.36	<0.001	0.64	<0.001	1	
Manufacturing/ Industrial Density, S _p		0.08	0.004	0.19	<0.001	0.25	<0.001	0.16	<0.001	0.13	<0.001
Road density, S _p		0.28	<0.001	0.28	<0.001	0.28	<0.001	0.11	<0.001	0.03	0.266
Subway Density, S _p		-0.01	0.617	0.02	0.573	-0.04	0.129	0.05	0.082	<0.01	0.944
Construction density, S _p		0.03	0.214	0.16	<0.001	0.20	<0.001	0.07	0.007	0.02	0.438
NPL, median (ppm)											
Yes	43	102	0.588	156	0.008	67	<0.001	15,021	0.125	314	0.036
No	1330	108		144		49		13,990		278	
TRI, median											
Yes	631	114	<0.001	161	<0.001	57	<0.001	14,421	0.003	284	0.075
No	742	105		135		45		13,845		278	
Scrap yard, median											
Yes	77	131	0.17	181	0.001	75	<0.001	15,626	<0.001	334	<0.001
No	1296	107		144		49		13,934		277	
Bridge, median											
Yes	440	147	<0.001	165	<0.001	58	<0.001	14,446	0.008	282	0.31
No	933	95		141		46		13,898		278	

($\beta = 0.004$, $p < 0.001$) concentrations. Manufacturing/industrial density was significantly associated with Cu, Zn, and Fe ($\beta = 1.44\text{E-}08$, $p = 0.035$; $\beta = 1.58\text{E-}08$, $p = 0.048$; $\beta = 1.15\text{E-}08$; $p = 0.014$, respectively). Above subway density was significantly associated with Pb and Zn concentration ($\beta = 0.047$, $p = 0.042$; $\beta = 0.028$, $p = 0.027$, respectively). Scrap yards were significantly associated with Mn concentration ($\beta = 0.075$, 0.019) and NPL sites were significantly associated with Pb, Fe and Mn ($\beta = 0.134$, $p = 0.004$, $\beta = 0.038$, $p = 0.014$; $\beta = 0.057$, $p = 0.037$, respectively). Finally, bridges nearby were significantly associated with Pb and Zn ($\beta = 0.106$, $p = 0.003$; $\beta = 0.076$, $p = 0.026$, respectively) (Table 6).

Discussion

The objective of this study was to investigate the spatial distribution and identify potential anthropogenic sources of Pb, Zn, Cu, Mn, and Fe contamination in surface soils throughout parks and greenspaces in Brooklyn, NY. Median concentrations of Pb, Zn, and Cu were approximately two to six times greater than background concentrations in the US, while Mn and Fe were lower (Smith et al. 2014). Few

samples were above US EPA or NY State soil guidance values; however, a large fraction of samples were above Pb, Cu, and Zn according to CCME guidelines for the protection of environmental and human health which are more protective (Canadian Council of Ministers of the Environment 2021; NYS DEC 2020; US EPA 2021b). Lead, Zn, and Cu concentrations were similar, or in some cases a bit lower, than what was found in other soil characterization studies conducted in large urban areas, including in Chicago, IL (Pb = 198 ppm; Zn = 235 ppm; Cu = 59 ppm) (Cannon and Horton 2009), New Orleans, LA (Pb = 656 ppm; Zn = 373 ppm; Cu median = 32 ppm) (Mielke et al. 2004), and Syracuse, NY (Pb = 130 ppm; Zn = 179 ppm) (Hunt et al. 2012). In our study, we found that metal concentrations were highest along the periphery of Brooklyn near the waterways where manufacturing and industrial sites were once located.

Roadway density and the presence of bridges within 800 m were strongly predictive of increased Pb, Cu, and Zn concentrations. In urban areas, roadway proximity has long been an environmental justice issue with racial minority groups or low-income households more likely to live in the vicinity of high volume roadways (Rowangould 2013). Increased metal concentrations observed in this study are

Table 6 Multivariable linear regression models to predict log-transformed metal concentrations based on nearby potential sources of contamination

Variable	Pb		Cu		Zn		Fe		Mn	
	Beta	p-value	Beta	p-value	Beta	p-value	Beta	p-value	Beta	p-value
Road density	0.008	<0.001	0.005	<0.001	0.005	0.026	0.004	<0.001	0.002	0.053
Manufacturing/Industrial Density	–	–	1.44E-08	0.035	1.58E-08	0.048	1.15E-08	0.014	8.23E-09	0.095
Construction density	–2.51E-08	0.030	–1.23E-08	0.022	–	–	–8.09E-09	0.053	–8.20E-09	0.062
Above ground subway density	0.047	0.042	–	–	0.028	0.027	–	–	–	–
Scrap yard within 400 m (yes vs no) ^a	–	–	0.043	0.065	–	–	–	–	0.075	0.019
TRI (yes vs no) ^b	–	–	–	–	–	–	0.025	0.149	–	–
NPL (yes vs no) ^b	0.134	0.004	0.073	0.084	0.090	0.132	0.038	0.014	0.057	0.037
Bridge (yes vs no) ^b	0.106	0.003	0.046	0.101	0.076	0.026	–	–	–	–

– = variable dropped from model

a = Within 400 m

b = Within 800 m

likely a combination of legacy and active sources. Lead was a gasoline additive for most of the twentieth century until it was banned for use in on-road vehicles in 1996 (Newell and Rogers 2003). However, Pb-based paint is still used on steel coatings such as bridges (Gottesfeld 2015). Copper and Zn are released into the environment through brake pad abrasion and tire wear, while Zn is used as an oil additive (Sager 2020). Elevated bridges likely disperse traffic-generated particulates over a greater distance as well as serve as potential source itself. In NYC, there have recently been proposals to cap a major bridge, located in a disadvantage area, to reduce air pollution to the surrounding community (Kuperstein 2020).

The US EPA designates NPL sites to guide further investigation due to active release or threatened release of pollutants into the environment (US EPA 2021a). In multivariate models, all metal concentrations increased in the vicinity of a NPL sites. Adjusting for significant co-variates, Pb concentrations increased by 27% (140 versus 103 ppm) within 800 m of a NPL, while Zn concentrations increased by 23% (206 versus 167 ppm). Like roadways, proximity to NPL sites disproportionately effects low-income resident (Burwell-Naney et al. 2013) and pollutants contained within hazardous waste sites can migrate to residential homes. A study of homes located within 800 m a hazardous waste had 27% greater Pb and 43% greater Zn concentrations in house dust compared to home outside the 800 m buffer (Zota et al. 2011). Remediation of NPL sites should consider the surrounding area as pollution can migrate offsite and potentially contaminate residential zoned land.

We identified twenty scrap metal recyclers operating in Brooklyn, many of which were in dense residential neighborhoods. Soil samples collected within 400 m of scrap yards had increased Cu and Mn concentrations even after controlling for other likely sources. At scrap metal recyclers, the metals are broken down by torch cutting, shredding, shearing, or crushing, usually in open scrap yard spaces. As a result, metal laden particulate may be transported into the surrounding community through the air and deposited onto soil. A study conducted in Houston, TX observed increased concentrations of Fe, nickel (Ni), chromium (Cr), Mn, and Pb particulates in the vicinity of metal recyclers (Han et al. 2020). In our analysis, Fe was associated with scrap yards in the crude association but not after adjusting for other potential sources of metals; while Pb was not significantly associated with scrap yards in the crude or unadjusted associations. However, our data were non-specific and did not take into account the type of metals processed at the facility or wind direction.

There were several limitations in this study. There may have been some measurement error since we only collected samples from publicly accessible spaces and our definition of potential sources may be inaccurate as we did not take

into consideration wind patterns and the built environment which could impact the direction in which pollutants spread and where they accumulate. This measurement error, however, is likely non-differential, which would attenuate associations. Additionally, some of the associations may have been underpowered, especially for weaker associations, leading to type 2 error (failing to reject the null hypothesis when it is not true) and we looked at five different metals but did not adjust our alpha for multiple comparisons, which could have led to type 1 errors (rejecting the null when it is true). Furthermore, we did not collect data on all potential sources of metal contamination, which may have led to residual confounding. Soil samples were only collected from publicly accessible spaces and did not include residential soil samples. A previous NYC study found significantly lower Pb concentrations in community garden soils compared to home garden soils, likely the result of tillage and soil replacement (Cheng et al. 2015). Thus our findings cannot be generalized to soil in other types of greenspace, such as private gardens, which could be important sources of exposure. We did not have access to information regarding recent park renovations and soil remediation which would affect metal concentrations. In multivariate analyses, new building construction since 2010 was associated with decreased Pb, Cu, Fe, and Mn concentrations. New construction in the area may result in park improvements that were not captured in the analyses. Future studies should consider including greenspaces and other soils not included here (e.g., lawns of public housing, private and semi-private spaces, etc.) in their sampling methodology. Finally, Brooklyn, NY may differ in important characteristics from other boroughs of NYC and from other cities, and therefore results of this study may not be generalizable to other locations.

Conclusions

Metal contamination throughout Brooklyn, NY is widespread and from a combination of legacy and active sources. This study found elevated levels of Pb, Cu, Fe, Mn, and Zn along the periphery of the borough and in the North along the East River waterfront where there are legacy sources of metal contamination. Although large manufacturing and industry have mostly left the area, smaller scrap metal recyclers are abundant and significantly increased Cu and Mn soil concentrations. In addition, NPL sites contributed to increased concentrations of all five metals within 800 m of the site. Roadways have long been established to be sources of urban pollution; however, in our study we also found the presence of bridges within 800 m were strongly also predictive of increased Pb, Cu, and Zn concentrations. Additional studies are needed to assess the health impacts and develop

programs to protect the residents near and visitors of these green spaces.

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Declarations

Conflict of interest The authors have not disclosed any competing interests.

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