Source Investigation for Lead and Cadmium with Possible Barium Clustering in Durham County

by

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ABSTRACT

SOURCE INVESTIGATION FOR LEAD AND CADMIUM WITH POSSIBLE BARIUM CLUSTERING IN DURHAM COUNTY

Background: Heavy metals like lead (Pb), cadmium (Cd), and barium (Ba) are pervasive contaminants found in many environments and may pose long-term health risks. A study previously conducted in Durham County, North Carolina (NC), obtained peripheral blood from 310 pregnant women at the 12-week gestational stage. Blood samples were analyzed for heavy metals and spatial distributions. The results showed elevated Pb and Cd concentrations that were spatially clustered in an urban neighborhood in Durham County. The objective of this study was to investigate the possible sources of the metal clusters (Pb and Cd) and to analyze the same dataset for the spatial distribution of Ba and for its co-occurrence with other metals.

Methods: Publicly available tax data for Durham County, NC, including parcel identification numbers, addresses, year built, built usage, and state of repair, as well as U.S. Census data were used to investigate possible sources of Pb and Cd. Geospatial clustering analysis and mapping were performed using the Hot Spot Analysis (Getis-Ord Gi*) to determine if clustering of Ba was observed.

Results: When considering both Pb and Cd clusters, the percentage of dwellings built before 1978, when Pb based paints were banned, was 85.4%. Seventy-nine percent of Pb and 45% of Cd cluster homes were of average build quality, while 21% of Pb and 55% of Cd cluster homes were built of lower than average quality. The median home income for the Pb cluster included 72% (n= 16) at very low income (≤ $32,000) and 28% (n=6) at low income ($32,000 to $46,000). All median incomes in the Cd clusters were categorized as being very low (≤ $32,000). The geospatial cluster analysis indicated that there were five potential hot spots. The median level and interquartile range (IQR) (ng/g) for all participants were 14.50 and 10.03 – 22.46. The median blood concentration within the hot spots was 21.05 ng/g (IQR 16.22 - 31.49 ng/g). The median Ba blood concentration for participants at and above the 90th percentile was 56.63 ng/g. Metals co-occurred with Ba, with the most frequent co-occurrences with Ba being with Cd. Participants with Ba at or above the 90th percentile had an average of 3.4 metals that also occurred at or above the 90th percentile. In the Ba cluster, the average was 3.0 metals.

Conclusion: The source of Pb and Cd was not conclusively identified, but the ages of the homes in the previously identified clusters indicate that paint may be the source of exposure. The presence of five probable Ba hot spots was identified, with Cd possibly co-occurring with Ba in some clusters. Additional investigations are required to confirm Pb and Cd sources and to further assess Ba exposures. The Ba exposure investigation should consider regional geology, exploring industries in Durham County that may be generating emissions that may then travel long distances, and an evaluation of possible occupational sources.

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INTRODUCTION

Metals are naturally abundant in most soils but can also be present in elevated concentrations due to anthropogenic sources. Heavy metals, such as lead (Pb), cadmium (Cd), and barium (Ba), are pervasive contaminants found in many urban environments and, to a lesser extent, rural environments (Aelion et al., 2013; Davis et al., 2014; Aelion et al., 2009; Moffett et al., 2007).

Ingestion of Pb can occur as a result of ingesting Pb containing dusts from deteriorating Pb based paints and from consuming Pb containing food or drinking water (CDC, 1991; ATSDR, 2007b); water supply plumbing that was installed before 1930 is likely to have high levels of Pb. Children are more susceptible to Pb and can be exposed, both prenatally and during childhood (ATSDR, 2007b). The most common way children are exposed to high-dose Pb is through Pb based paint (CDC, 1991). Exposure to Pb can pose long-term health risks, including increased risk of cardiovascular disease (Zawadzki et al., 2006; Aoki et al., 2016). Pb may raise blood pressure, increase arterial stiffness, and possibly play a key role in the pathogenesis of cardiovascular disease by increasing the production of reactive oxygen species (ROS) in exposed persons (ATSDR, 2007; Poreb et al., 2011; Nosratola et al., 2008). In a study of adult blood Pb levels from the Third National Health and Nutrition Examination Survey (NHANES III), the relative risk of mortality from all causes was higher when the blood Pb levels were higher than 5 µg/dL (Schober et al., 2006).

Exposure to Cd can occur through smoking, drinking water with elevated levels of Cd, eating foods that have had Cd introduced through agricultural soils, and Cd emitting industries or incinerators (ATSDR, 2012). Although Pb is thought of as the major contaminant in older paints, Cd may also be present in older paints. Sanding in preparation for re-painting, which creates dust, increases the potential exposure to these metals in homes with older paints (Mielke et al., 2001; Luckett et al., 2012). High concentrations of Cd can also be found in building paint of more recent construction (Nduka et al., 2007). Significant associations have been observed between cardiovascular and cerebrovascular disease (CCVD) and urinary Cd, antimony, cobalt, and tungsten; high levels of serum Cd (>0.61 mg/L) have been also associated with CCVD (Agarwal et al., 2011). In an in vitro study, Pb was found to enhance Cd's cytotoxicity to the bovine aortic endothelial cell monolayer by affecting a larger area when present (Kaji et al., 1995).

Exposure to Ba might occur from well drinking water or foods such as Brazil nuts, seaweed, fish, mushrooms, and legumes, but those working in industries that make or use Ba compounds (e.g. welding, mining, drilling, brick or paint manufacturing) are at the greatest risk for exposure (ATSDR, 2007; Moffett et al., 2007). Hydraulic fracturing has been observed to increase levels of Ba in aquifers and surface waters (Chapman et al., 2014; Latta et al., 2015). Agricultural products, like wheat, may absorb Ba from biosolids applications on farmland (Ippolito et al., 2006). A study that examined the effects of low-level exposure of Ba, trying to mimic the exposure through well water, observed an increase in malignancy in keratinocytes cells in the lab (Thang et al., 2015). The form of Ba seems to be important in determining what kind of health effect exposure to the metal might have. For example, barium sulfate was found to increase the risk of appendicitis while barium chloride was found to cause oxidative stress in the livers of adult rats (Li et al., 2017; Elwej et al., 2016).

In a study conducted in Durham County, North Carolina (NC), King et al. (2015) obtained peripheral blood from 310 pregnant women at the 12-week gestational stage. Blood samples were analyzed for heavy metals and spatial distributions. The results showed elevated Pb and Cd concentrations that were spatially clustered in an urban neighborhood in Durham County. In the present study, the same dataset was analyzed for the spatial distribution of Ba and for its co-occurrence with other metals. The source(s) of the metals, which were previously identified by King et al. (2015) and Edwards et al. (2015), was investigated.

METHODS

Participant data for pregnant women who were living in Durham County and who used Duke University or Durham Regional Hospitals for obstetrics care included addresses for their residences at the time of the study and metal concentrations in peripheral blood, measured as described by King et al. (2015).

Source Investigation for Pb and Cd

Publicly available tax data for Durham County, NC, for 2016 —including parcel identification numbers, addresses, year built, built usage, and state of repair—were downloaded from the Durham County Tax Administration website (http://dconc.gov/government/departments-f-z/tax-administration/data-files) and merged with participant data using residence addresses. Both Cd and Pb clusters from the King et al. (2015) study were analyzed.

Building age. The age of participant residences was assessed using “Actual Year Built” (Durham County Tax Administration, 2016a). The age of the structure was used as an indicator of whether Pb might be present in the paint of the structure or in the plumbing (CDC, 1991; ATSDR, 2007).

Building state of repair. The Tax Administration’s state of repair for a building is less a measure of material deterioration and more a statement on the build quality of the structure itself. There could be two structures built from the same general plan, each offering the same facilities, but have very different cost due to the quality of materials and workmanship used in their construction. Schedules distinguishing between variations in cost of construction for single family residence (Volume 3) and multi-family residences (Volume 4.2) are available online (http://www.dconc.gov/government/departments-f-z/tax-administration/proposed-2016-schedules-of-values).

For single family homes there are seven base grades or classes of quality that cover the range of home construction, from the cheapest (E) to the highest in quality (XX): XX, X, A, B, C, D and E. Most homes in Durham County fall within one class above and one class below the base grade, which is C. Sometimes included is a (+) representing above average quality or a (-) representing below average quality within its own class (Durham County Tax Administration, 2016b).

Some of the dwellings were multi-family residences, which fall under different state of repair categories; these included multi-family residential garden apartments/condominiums, which are buildings of three or fewer stories in which each living unit includes a kitchen and bath, of wood load bearing construction or of masonry veneer construction (Durham County Tax Administration, 2016c).

Medium household income. Household income data were obtained from the U.S. Census Bureau's SF1 and TIGER data sets from 2010 (https://www.census.gov/data.html). The census data separates household incomes by block into five categories: Very High (> $74,000), High ($60,001 to $74,000), Average ($46,001 to $60,000), Low ($32,001 to $46,000), and Very Low (≤$32,000).

Barium

Cluster mapping. After the exclusion of 73 pregnant women for whom blood samples were collected but who had negative Ba measurements or were residing outside of Durham County, the total number of participants with measured Ba blood concentrations was 230. As described in King et al. (2015), clustering analysis and mapping was performed using the Hot Spot Analysis (Getis-Ord Gi*) tool within ArcGIS 10.4.1. Given a set of weighted features, this statistical tool identifies statistically significant hot spots and cold spots using the Getis-Ord Gi* statistic. Also, as in the King et al. (2015) study, a fixed-distance band of 1500 m was used to calculate the spatial influence.

Co-occurrence with other metals. The 90th percentile for Ba was calculated, and for participants with Ba in the 90th percentile or higher, the frequency with which their blood concentration of a second metal was at or above the 90th percentile was determined. These additional metals included Calcium (Ca), Magnesium (Mg), Copper (Cu), Zinc (Zn), Arsenic (As), Iron (Fe), Strontium (Sr), Manganese (Mn), Cobalt (Co), Molybdenum (Mo), Scandium (Sc), Titanium (Ti), Selenium (Se), Cadmium (Cd), Chromium (Cr), Lead (Pb), Aluminum (Al), Silicon (Si), Vanadium (V), Rubidium (Rb), Tin (Sn), Mercury (Hg), Nickel (Ni), and Lead isotopes (208 Pb/ 206 Pb and 207 Pb/ 206 Pb). The frequency of metals co-occurring with Ba was thus calculated, as was the number of metals for which blood concentrations were measured at or above the 90th percentile for each participant with Ba at or above the 90th percentile.

RESULTS

Source Investigation for Pb and Cd

Building age. The average age of the dwellings in the previously identified Pb clusters was 49 years. The average age of the dwellings in the previously identified Cd clusters was 69 years.

All Pb cluster parcels had built age data, but 3 Cd cluster parcels did not. See Table 1 for the distribution of built year of dwellings in the Pb and Cd clusters.

Table 1. Distribution of Year of Construction for Dwellings in Pb and Cd Clusters in Durham County, NC

Year of Construction	Number of Dwellings in Pb clusters[a]	Number of Dwellings in Cd clusters
Before 1959	5	15
1960-1978	10	5
1979-2016	4	2
No data	0	3

[a]Due to the exclusion of multi-family residences, 19 of 22 dwellings were considered.

Building state of repair. Both Pb and Cd clusters included single family homes. Pb clusters included homes with two grades—the base grade C and lower grade of D (C = 5 and D = 4). Cd clusters also contained homes with two grades—the base grade C and lower grade of D (C = 9 and D = 12). See Table 2 for the distribution of dwellings in the Pb and Cd clusters by state of repair. The Pb clusters included both wood load bearing (n= 8) and masonry veneer (n=2) multi-family residences, while the Cd clusters contained one wood load bearing multi-family residence.

Table 2. State of Repair of Dwellings in Pb and Cd Clusters in Durham County, NC

State of Repair	Number of Pb Clusters Dwellings	Number of Cd Clusters Dwellings
XX	0	0
X	0	0
A	0	0
B	0	0
C	5	9
D	4	12
E	0	0
No data	0	3

Median home income. The majority of households in the Pb clusters had Very Low median household incomes ($32,000 or less). Only 6 homes in the Pb clusters were within a Low median household income block group ($32,001 to $46,000). All of the homes in the Cd clusters were located in areas of Very Low median household incomes. Results are shown in Table 3.

Table 3. Median Household Income of Dwellings in Pb and Cd Clusters in Durham County, NC

Median Household Income	Number of Pb Clusters Dwellings	Number of Cd Clusters Dwellings
Very High (More than $74,000)	0	0
High ($60,001 to $74,000)	0	0
Average ($46,001 to $60,000)	0	0
Low ($32,001 to $46,000)	6	0
Very Low ($32,000 or Less)	16	25

Barium cluster mapping. Among the 230 woman who had Ba blood concentrations measured, the median level and interquartile range (IQR) (ng/g) were 14.50 and 10.03 – 22.46. The results from the geospatial cluster analysis (Getis-Ord Gi*) indicated that there was 1 dwelling that represented a hot spot cluster with a 99% confidence level, 2 other dwellings with a 95% confidence level, and 6 other dwellings with a 90% confidence level, representing what seems like five hot spots (see Figure 1). The median blood concentration within the 9 dwellings in the hot spots, was 21.05 ng/g (IQR 16.22 - 31.49 ng/g) (see Table 4).

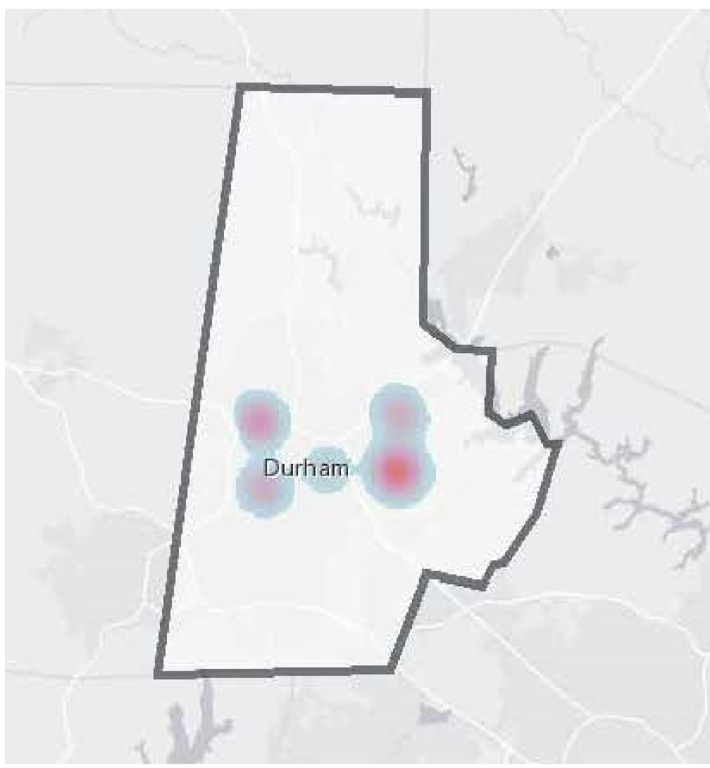

Figure 1. Geospatial clustering of Ba blood concentrations using Getis-Ord Gi*. A simplified heat map was used to show regions of clustering.

The 90th percentile for Ba blood concentration was calculated for the entire dataset, and 31 participants had values at or above the 90th percentile. The median Ba blood concentration for participants at and above the 90th percentile was 56.63 ng/g (IQR 46.60 – 94.50 ng/g) (see Table 4).

Table 4. Ba Blood Concentrations (ng/g) for all Participants and within Clusters

	N	Geometric Mean	Median (IQR)
Overall	230	14.50	14.50 (10.03 – 22.46)
In Ba Clusters	9	21.05	22.71 (16.22- 31.49)
Ba 90th Percentile	31	71.75	56.63 (46.60 – 94.50)

Co-occurrence with other metals. The frequency with which the blood concentration of a second metal was above the 90th percentile, in all participants, while Ba was also above the 90th percentile, ranged from 0 to 7 (see Figure 2). The figure shows that several metals frequently co-occurred at high concentrations with Ba, with the most frequent co-occurrences with Ba being with Cd and, to a lesser extent, molybdenum (Mo). Within the Ba cluster, co-occurrences ranged from 0 to 3 and did not include Cd or Mo.

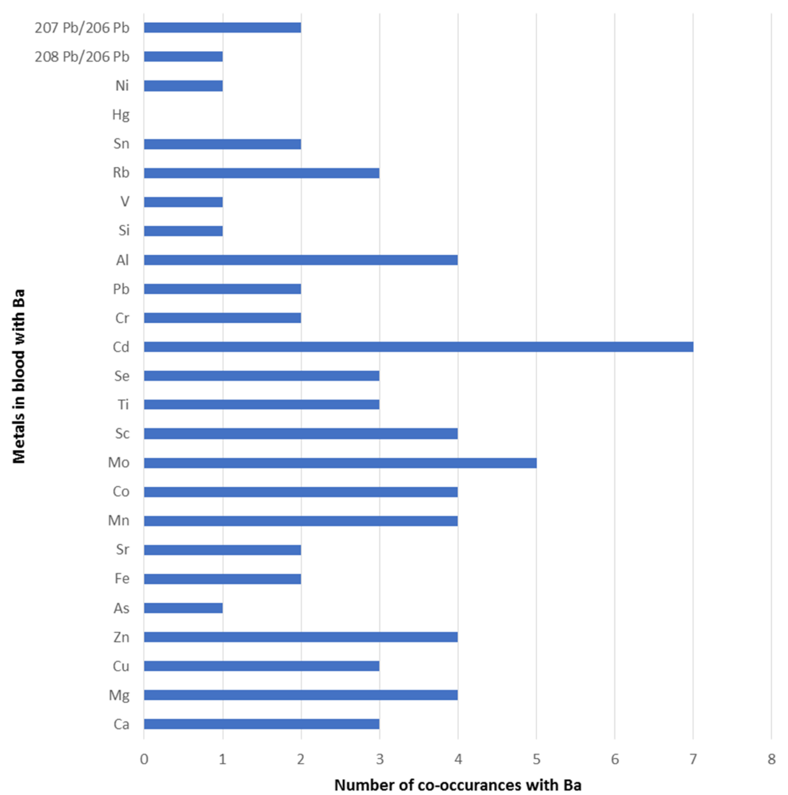

Figure 2. Frequency of metal co-occurrence with Ba when concentrations of Ba and the second metal were at or above the 90th percentile.

The number of metals for which blood concentrations were measured at or above the 90th percentile among participants with Ba at or above the 90th percentile is displayed in Figure 3. Participants with Ba at or above the 90th percentile had an average of 3.4 metals that also occurred at or above the 90th percentile. In the Ba cluster, the average was 3.0 metals.

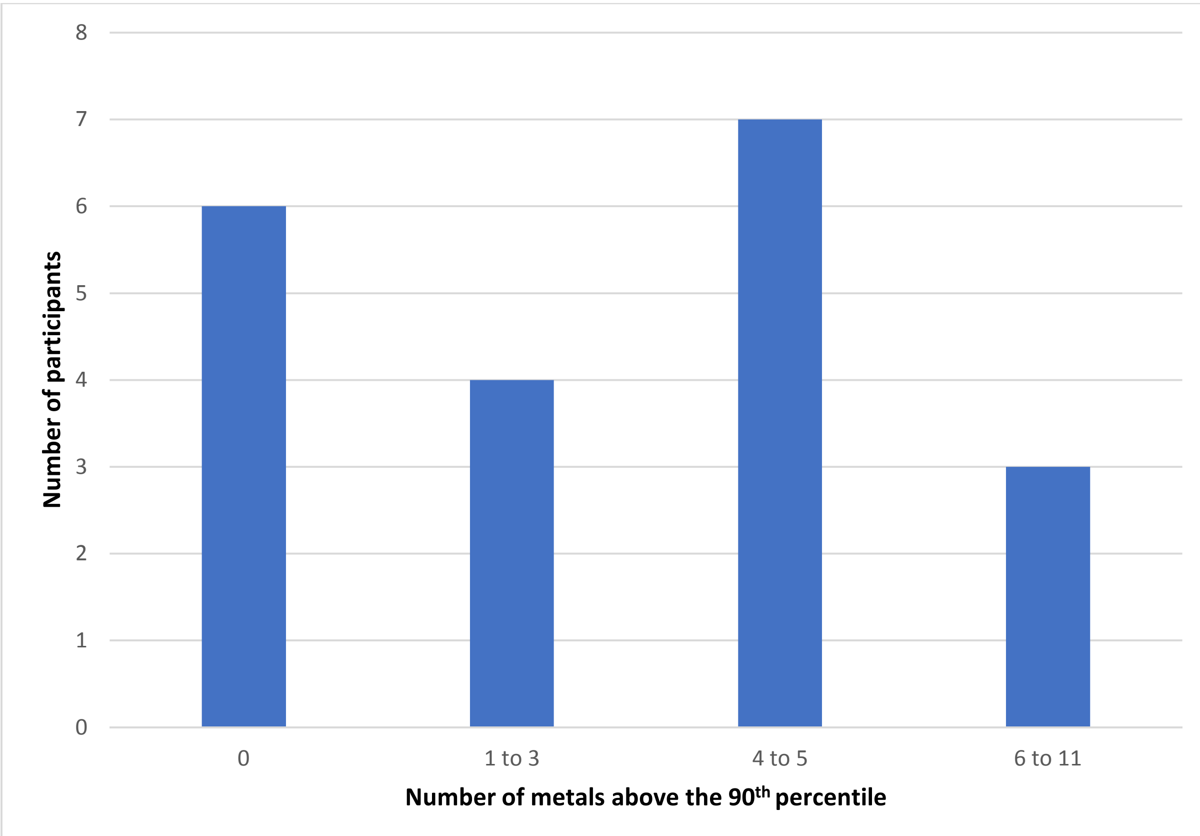

Figure 3. Metal counts per participant when the concentrations of Ba and a second metal were at or above the 90th Percentile.

DISCUSSION

This study sought to find the probable sources of Pb and Cd clustering identified by King et al. (2015) in Durham County, NC, where elevated levels of metals were found in the soil and in the blood and urine of pregnant women. Some potential sources of these metals seem more likely than others after this investigation, but no conclusive exposure source could be established. Additionally, the presence of five probable Ba hot spots was identified after additional geospatial cluster analysis was conducted.

Source Investigation for Pb and Cd

The median year built of homes in the United States, including owner-occupied and renter-occupied, is 1976. In the South Atlantic Census Region (Delaware, Florida, Georgia, Maryland, North Carolina, South Carolina, Virginia, District of Columbia, and West Virginia), it is 1984 (US Census Bureau, 2015). In 2011, HUD estimated that 37.1 million housing units, or about 34.9% of all housing, in the United States had Pb based paints still remaining (US Department of Housing and Urban Development, 2011). In this study, it was found that the median years built of dwellings in the previously identified Pb and Cd clusters were 1966 and 1944, respectively. The majority of the dwellings (52.6%) in the Pb clusters were built between 1960 and 1978, while 26.3% and 21.0% were built prior to 1960 and between 1979 and 2016, respectively. The percentage of all homes built before 1978, years during which Pb based paint was available, was 78.9% in the Pb clusters. The majority of dwellings (68.1%) in the Cd clusters were built during or before 1959. Dwellings built between 1960 and 1978 and 1979 to 2016 comprised 22.7% and 9.0%, respectively, of dwellings in the Cd clusters (Durham County Tax Administration, 2016a). When considering both Pb and Cd clusters, the percentage of dwellings built before 1978 is 85.4%, suggesting that paint may be a source of metal exposure in these clusters. Sampling of the walls within the dwellings is necessary to confirm that paint is a possible source of Pb and Cd exposure.

The state of repair, or quality of construction, of all homes in the Pb and Cd clusters were of average or below average quality. In the Pb clusters, 78.9% of dwellings were average, and 45.4% were average in the Cd clusters. Dwellings of lower quality comprised 21.0% of Pb and 54.5% of Cd clusters. Knowing the construction quality of the dwellings is useful in making assumptions about the materials used and their compositions. For example, both average and lower than average quality dwellings typically were constructed with heating systems, but air conditioning was usually an add-on and not standard (Durham County Tax Administration, 2016b; Durham County Tax Administration, 2016c). The lack of air conditioning may result in windows being left open for longer periods of time, in certain seasons, for cooling purposes and increase the rate of dust accumulation inside the dwellings, thereby exposing the inhabitants to contaminants in dust (CDC, 1991; ATSDR, 2007b).

The dwellings in the Cd clusters were substantially older than the dwellings in the Pb clusters, and a larger percentage of the dwellings were of lower quality. Based on their age and quality of construction, dwellings in the Cd clusters could be in a dire state of deterioration. Importantly, all of the household incomes at the block level for dwellings in the Cd clusters were low or very low. Considering the income level and age and quality of dwellings in the Cd clusters, it is possible that other sources of metal exposure exist in these areas. For example, deteriorated playground equipment, such as railings, handles, and posts, may be a source of Pb and Cd exposure (Turner et al., 2016). Follow-up sampling would be required to explore other contributors.

One limitation of this investigation of Pb and Cd sources is not having measurements for Pb and Cd in wall paints and not having data about if and when the dwellings had been sanded and re-painted. Further, this study relied on building age data for the current structure, which does not facilitate identification of previous land uses or other structures in the parcel that may be a source of metal contamination. Land use data would have been useful in the investigation to help in identifying activity that could have led to a release of metals in the past. Although the tax data indicates built use, the data are not sufficient to determine possible sources.

Barium Analyses

Because background levels of Ba in blood vary depending on daily intake and there are no data correlating blood levels to specific exposure levels, determining what constitutes a normal blood Ba concentration is difficult (Moffett et al., 2007). A case study by Hung and Chung (2004) found that under normal exposure conditions blood levels for Ba ranged from 3 to 20 ug/dL. All participants in this study fall below this range with the median blood Ba concentrations for the entire sample of 14.50 ng/g, or 1.50 ug/dL, and 22.71 ng/g, or 2.35 ug/dL, among participants in the Ba clusters. The majority of participants (n = 191 or 83%) had blood Ba concentrations below the low end of the range of 3 ug/dL.

King et al. (2015) identified overlapping Pb and Cd clusters. The results from the geospatial cluster analysis in this study indicated the potential for five Ba hot spots, with one overlapping Cd cluster at the center of the Ba clusters, although at a low confidence level (90%). There were no Ba clusters overlapping with previously identified Pb clusters. The median blood concentration within the Ba clusters was higher than the median for the entire sample but not as high as the 90^{th} percentile.

The potential Ba clusters appear to be more spread out geographically than the Pb or Cd clusters and could be indicative of different sources of Ba or a consequence of geology (Davis et al., 2014; Aelion et al., 2009). This finding should be taken into consideration in subsequent investigations. While no study participants reported using well water as a drinking source, where high concentrations of Ba have been found in other studies (Pham et al., 2017; Chanpiwat et al., 2014), other water sources, such as municipal water supplies and surface waters, may warrant further investigation.

A limitation of this study is that no soil or water samples for Ba were taken. Because Ba exposure pathways vary widely, from food to geological sources, and excretion happens relatively quickly, determining the sources would require a considerable amount of effort and resources (Moffett et al., 2007).

CONCLUSION

Chronic exposure to low levels of Pb and Cd can increase the risk of cardiometabolic diseases in children and adults. Additional studies that sample for Pb and Cd in household painted surfaces are required to confirm or reject that as a source of exposure, but the age of houses in the clusters suggests that paint is a possible source. The presence of five probable Ba hot spots was identified, with potential overlap with a Cd hot spot, but additional sampling for Ba is required to assess possible sources. Future investigations could include considering regional geology, exploring industries in Durham County that may be generating emissions that may then travel long distances, and evaluating possible occupational sources. Also requiring additional analysis is investigating whether the co-occurrence of Ba and Cd is indicative of a common source.

LIST OF ABBREVIATIONS AND ACRONYMS

ATSDR	Agency for Toxic Substances and Disease Registry
Ba	barium
Cd	cadmium
g	gram
Mg	milligram
NC	North Carolina
ng/g	nanogram per gram
NHANES	National Health and Nutrition Examination Study
Pb	lead
µg/dL	microgram per deciliter
USEPA	United States Environmental Protection Agency

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