TRACKING THE SOURCE OF HIGH LEAD IN CHILDREN'S BLOOD

A Thesis

by

FELIPE GUSTAVO BECKER

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Chair of Committee,	Franco Marcantonio
Committee Members,	Peter Knappett
	John R. Giardino
Head of Department,	Julie Newman

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ABSTRACT

Children are the most susceptible demographic affected by lead (Pb) poisoning because of increased hand-to-mouth interaction at an early age. Here, Pb isotope ratios are used for apportioning the sources of Pb in the blood of children (ages 1-6) screened for high blood Pb levels (>5 μg/dL) surrounding urban areas of Kansas City, MO. We compared Pb isotope ratios measured in the child's blood with those of the most likely sources of Pb in that child's home environment. The environmental sources sampled consisted of soils, paints, occupational sources (e.g., oil rig workers' uniforms, mechanics' clothes), indoor air filters, dusts, and dietary sources (e.g., spices). After collection, blood samples were decontaminated, digested in ultrapure nitric acid, filtered, and separated via ion chromatography (AG 1-X8) before analysis. Blood and environmental samples were analyzed for total Pb concentration, $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ using the Neptune Plus multi-collector ICP-MS. The average lead levels (BLL) measured in the blood aliquots were 12.7 μg/dL, 3.5 μg/dL, and 2.9 μg/dL, for Houses 1, 2 and 3, respectively. Pb isotope ratios from House 2 show a limited range, while House 1 and House 3 show heterogeneous compositions. By comparing the $^{208}Pb/^{206}Pb$ and $^{206}Pb/^{207}Pb$ isotope ratios, we identify that the Pb in blood of the child from House 2 ($^{208}Pb/^{206}Pb$= 2.010, $^{206}Pb/^{207}Pb$= 1.227, respectively) is isotopically similar to dust by the door ($^{208}Pb/^{206}Pb$= 2.011, $^{206}Pb/^{207}Pb$= 1.228) and soil from a roof drip zone ($^{208}Pb/^{206}Pb$= 2.012, $^{206}Pb/^{207}Pb$= 1.226), indicating that the blood Pb level, in this case, may be influenced by both sources or their mixtures. In House 1,

there was one environmental source (dust from the kitchen baseboard, $^{208}Pb/^{206}Pb$= 2.024, $^{206}Pb/^{207}Pb$= 1.219) that was most similar isotopically to the blood ($^{208}Pb/^{206}Pb$= 2.025, $^{206}Pb/^{207}Pb$= 1.213). Although the range in the Pb isotope ratios of environmental samples in House 3 was large, we determined that the Pb isotope ratios of a sample of turmeric cooking spice ($^{208}Pb/^{206}Pb$= 2.131, $^{206}Pb/^{207}Pb$= 1.128) to be remarkably similar to the blood ($^{208}Pb/^{206}Pb$= 2.132, $^{206}Pb/^{207}Pb$= 1.127), suggesting a direct link, through the ingestion of the spice, to the elevated BLL.

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NOMENCLATURE

ADHD	Attention-Deficit/Hyperactivity Disorder
BLL	Blood Lead Level
CDC	Center for Disease Control and Prevention
IDMS	Isotope Dilution Mass Spectrometry
IRB	Institutional Review Board
K_2EDTA	Ethylenediaminetetraacetic acid
MC-ICP-MS	Multi Collector Inductively Coupled Plasma Mass Spectrometry
NIOSH	National Institute for Occupational Safety and Health
TIMS	Thermal Ionization Mass Spectrometry

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1. INTRODUCTION

1.1. Lead as an Environmental Contaminant

Lead is an ubiquitous heavy metal that participates in many chemical reactions in the natural world. Consequently, it is commonly found in households, which can, unfortunately, lead to adverse health effects. Lead concentrations in the body are normally reported as blood lead level (BLL, μg of Pb per dL of blood) and is generally accepted to be the main predictor of environmental lead toxicity (WHO, 1980). In recent decades, the focus of lead research has shifted from improving medical intervention to making primary prevention efficient and cost effective (Campbell & Osterhoudt, 2000; Lanphear et al., 2005). With this shift, comes the emphasis on preventing exposure before it happens by identifying contaminant sources in urban households (Lanphear, 1998c).

The anthropogenic environmental imprint of Pb is associated with the years of industrialization. In addition to leaded paint used in households, the incorporation of lead in gasoline is known to have significantly contributed to the dispersal of atmospheric heavy metals during the 1900s (Gilbert & Weiss, 2006), resulting in lead-contaminated soils and dusts. Even though the reduction in the use of leaded-gasoline resulted in declining BLL trends in US children (Needleman, 2000; Jones et al., 2009), alarming concentrations of lead in dust and soil can still be found in urban environments (Gilbert & Weiss, 2006).

1.2. Lead as a Public Health Concern

Lead poisoning trends have been addressed over time by regulations that achieved a lowering of the BLL level of concern, and anything higher than this threshold is of interest for investigation. The most recent change in 2012 (ACCLPP, 2012; CDC, 2012) reduced the BLL of concern from 10 to 5 μg/dL, in an effort to favor primary prevention, as the adverse health effects of lead in even low amounts has been demonstrated in recent years (Chiodo et al., 2004; Lanphear et al., 2005; Canfield et al., 2003). Children are the most susceptible demographic affected by lead poisoning because of increased hand-to-mouth activity at an early age. Accidental ingestion of particles containing lead is understood to be the main lead uptake mechanism in pediatric lead poisoning (Lanphear & Roghmann, 1997; Evans et al., 2018). Studies have shown that an elevated BLL in young children can result in impaired brain and behavioral development, decreased IQ (Wasserman et al., 2000a), decreasing scores of reading (Lanphear et al., 2000), impaired motor skills (Chiodo et al., 2004; Wasserman et al., 2000b), troubled memory (Lanphear et al., 2000; Lukawski & Sieklucka-Dziuba, 2007), and ADHD (Attention-Deficit/Hyperactivity Disorder) disorders (Nigg et al., 2008). Predictions about the impact of lead poisoning suggest that rising numbers of cases cost the US billions of dollars annually (Landrigan et al., 2002), in addition to the long-lasting effect of undesirable social behaviors (Nevin, 2000). This is of particular concern in low- to middle-class income countries (Attina & Trasande, 2013). In contrast, models predict that a decrease in children BLL rates removes the suppression of IQ and increases lifetime earnings and societal benefits (Grosse et al., 2002; Muennig, 2009).

1.3. Lead Investigations

High-precision analysis of Pb isotope ratios is essential to identify sources of environmental exposure with confidence. While studies continue to use both Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS) and Thermal Ionization Mass Spectrometry (TIMS) interchangeably, advancements in mass spectrometry have favored ICP-MS because of its simplicity, cheaper cost, and faster analysis (Gwiazda et al., 1998), and it has been successfully used in recent investigations (Gwiazda & Smith, 2000; Gwiazda et al., 2005; Patel et al., 2008; Tsuji et al., 2008; Tagaki et al., 2011; Zeng et al., 2012; Laidlaw et al., 2014; Evans et al., 2018). Because of their high atomic mass, lead isotopes tend not to fractionate in nature, so that the Pb isotopic composition does not change as the metal is transported through the environment.

Between the 1960s and 1970s, it was theorized that the ingestion of leaded paint was responsible for elevated BLL in children. For example, in a classic paper, Rabinowitz (1987) showed that if paint is responsible for the elevated BLL, the lead isotopic signature of the blood will be almost identical to that of the paint. However, whereas certain results argue that atmospheric sources (e.g. dust, soil particles) have no contribution to elevated BLL (Rabinowitz, 1987), there is a growing consensus that such lead is the most common culprit for lead poisoning in children living in urban areas. This movement was spearheaded by the findings of Sayre et al. (1974), who demonstrated alarming levels of lead in household surfaces and children's hands, and that ingestion of

the dust was a likely mechanism for childhood lead poisoning. Indeed, Manton et al. (2000) have shown that the contribution of lead in dust found in surfaces reachable by children must not be ignored. Other studies have reinforced this idea, identifying household dust as the main source responsible for elevated BLL in children (Lanphear et al., 1996; 1998a; 1998b; Gwiazda & Smith, 2000), especially in areas close to active ore smelting (Landrigan et al., 1975; Landrigan & Baker, 1981). Gasana et al. (2006) showed a strong correlation between elevated BLL in poisoned children from Miami and dust collected from the floor and windowsills of their homes. Lanphear and Roghmann (1997) suggested that lead-contaminated soil and lead-based paint contribute to overall dust lead levels found in the household, albeit to different extents. This theory was supported by data gathered by Laidlaw et al. (2014) in homes from urban Sydney, Australia, where the authors found a positive correlation between Pb dust concentration and Pb soil concentrations. Other studies have strongly suggested that windows and doors are household entry points for lead-bearing dust and soil particles (Sayre & Katzel, 1979), which are resuspended and mobilized by mechanical disturbance of contaminated soils, or tracked into the house (Laidlaw et al., 2014). Studies also suggest that BLLs in children seem to be controlled by climate variations during periods of lower soil moisture, promoting resuspension and deposition of lead dust (Laidlaw et al., 2005; Laidlaw & Filippelli, 2008). Statistical models predict that factors such as socio-economic status and children's behavior (i.e. amount of time spent outdoors in contact with soil) also directly influences BLL (Lanphear & Roghmann, 1997; Lanphear et al., 1998a; Jones et al., 2009).

This study was developed at Texas A&M University, College Station, TX, in partnership with Children's Mercy Hospital in Kansas City, MO. It has the primary goal of introducing a relatively cheap but highly precise method for identification of different sources of Pb in children's blood. Ultimately, we aim to show that this method is analytically precise, and that it can confidently identify the sources of contamination in the participating houses from the Kansas City area. To our knowledge, it is among the first Pb isotope studies to be completed in the US since the CDC level of concern was lowered to 5 μg/dL in 2012. In addition, the results of this project may motivate future studies that measure the ongoing health risks associated with lead in households, even years after laws were passed to mitigate such risks. Most importantly, we believe that our study will lead to more efficient lead-abatement techniques.

2. METHODOLOGY

2.1. House Selection

This study is a collaboration between Children's Mercy Hospital in Kansas City, MO, and Texas A&M University in College Station, TX. The project has been reviewed and approved by the Institutional Review Board (IRB) from both institutions. The houses sampled were identified by health department inspections as possible risks for lead exposure and were referred to Children's Mercy Hospital for further investigation. Houses were selected based on blood lead levels of the children screened, specifically levels exceeding 5 μg/dL in whole blood samples. The families of interest granted consent to participate during the early visits by the Children's Mercy Hospital risk assessor and received a detailed explanation of the goals of the study before house surveying and sampling. A total of three participating houses were visited by a risk assessor (Mr. E. Bowles) and phlebotomist (Ms. A. Mena) from Children's Mercy Hospital, and the samples collected in each visit were mailed to Texas A&M University.

2.2. Blood Sample Collection

Blood samples from the participating children were obtained by a registered nurse authorized by Children's Mercy Hospital. Intravenous blood samples were collected into polypropylene vacutainers containing the anticoagulant K_2EDTA (lavender-top-conventional-stopper, #367835, Becton, Dickinson and Company, NJ*)* and refrigerated until mailed for decontamination. Studies have shown that blood collection devices such as phlebotomy tubes contain significant amount of trace lead that can vary

between tubes (Flegal & Smith, 1992). This trace metal variation comes from differences in the manufacturing process and transportation. The Pb vacutainer blank contained negligible amounts of lead when compared to the amount lead found in blood (Table 1; more than two orders of magnitude more Pb in the blood than in the vacutainer blank). In addition, we have measured Pb concentrations in procedural reagent blanks and laboratory ware, which ranged from 10s of picograms to 100s of picograms (Table 1), making blank corrections unnecessary given the large analyte inventories which ranged from 10s to 1000s of nanograms.

Table 1. Lead contents for all the laboratory ware, collection materials and reagents used during sample processing. Replicate procedural blanks were measured on bleach, HNO_3, and HNO_3 + HF whereas H_2O_2, Savillex beaker, and Vacutainer blanks represent the Pb content found in these materials only.

Sample ID	Pb concentration (ng/g)
Bleach 1	0.314
Bleach 2	0.110
Bleach 3	0.110
Average	**0.178**
HNO3+HF 1	0.045
HNO3+HF 2	0.246
HNO3+HF 3	0.025
Average	**0.105**
HNO3 1	0.031
HNO3 2	0.072
HNO3 3	0.004
Average	**0.036**

Table 1. Continued

Sample ID	Pb concentration (ng/g)
H2O2	0.065
Savillex Teflon beaker	0.037
Purple Top Vacutainer	0.067

Because endogenous (likely synonymous to skeletal) contributions to blood lead in children are expected to be minimal, we assume that lead in the blood of children will reflect recent exposures, with little to no mixing of endogenous lead (Smith et al., 1996). The main sources of exogenous lead were assumed to be inhalation and ingestion of lead-contaminated particles such as dusts, soils, and paint chips.

2.3. Collection of Samples of Dust, Soil, Paint, and Spices

Environmental household samples were collected by an authorized risk assessor from Children's Mercy Hospital and mailed directly to the Radiogenic Isotope Laboratory at Texas A&M University. From the three participating houses, a total of 3 children's blood samples, 21 dust samples, 6 soil samples, 5 spice samples, and 1 paint sample were collected, and the number of samples per household varied with the child's play habits (Appendix). Dust samples were collected in compliance with the National Institute for Occupational Safety and Health (NIOSH) methods 9100 and 9102 for collecting lead in dust via moist wipes (NIOSH 1996; 2003) and stored in 50 mL centrifuge tubes for shipment. Dust sampling was performed over areas between 5 to 72 square inches, wiping in an S motion with low lead swipes while wearing gloves. Areas of interest for dust collection were main play areas where the children spend significant time or has easy access to, including counter tops, floorboards, door frames,

windowsills, a cracked wall, an air vent, metal faucets, door handles, metal frames from a bed, stair steps, and a ping-pong table. Even though suggested in previous studies (Sayre et al., 1974; Charney et al., 1980), dust or dirt samples from the child's hands were not collected. Another source of childhood lead exposure investigated here are paraoccupational lead sources which means that lead that is brought home on contaminated clothing or tools (Nevin, 2000). The occupational samples consisted of dust from the father's workpants and work shoes. Soil samples, consisting of heterogeneous dirt and rock fragments, were collected from play areas with exposed soil near the house and in areas of lead leachate accumulation, such as drip zones from the roof. The only paint sample obtained was composed of loose flakes of yellow-colored paint chips found on the bathroom floor. Spice samples varied in type although no brand was reported. The spice type included: turmeric, red-chili powder, coriander powder, cumin powder, and garam masala.

2.4. Blood Sample Processing

Upon arrival at Texas A&M University, each blood vial containing 0.5 to 2 mL of blood was refrigerated until decontamination. The samples were decontaminated in an authorized biosafety (BSL-2) laboratory at Texas A&M Public Health Department by adding household bleach (5.25% chlorine solution). To ensure blood pathogen decontamination, the total amount of bleach added to each vial varied with the volume of blood present, ranging between 0.2 to 0.4 mL, so that the final volume of the bleach:blood solution was composed of 10% bleach. Samples were then inverted several times and refrigerated for a minimum of 24 hours to complete decontamination.

Sample digestion and preparation was conducted in the R. Ken Williams ’45 Radiogenic Isotope Laboratory facility under low-lead Class 100 to Class 1000 cleanroom conditions. It is important to note that, after the decontamination process was complete, the blood:bleach solution had a gelatinous consistency, most likely due to coagulation. The solid blood residue was divided in up to 4 aliquots and transferred into pre-weighed Teflon beakers (#300-060-03, Savillex, MN) using a clean spatula. Before usage, the vessels were acid-washed by refluxing a 30% nitric acid solution (prepared using MilliQ ultrapure 18.2 megaohm filtered water, MilliporeSigma, MO) for 24 hours, followed by a concentrated hydrofluoric acid (HF, Fisher Scientific Optima Grade) reflux for 24 hours. To obtain a precise analysis of lead in biological samples, measurements require the destruction of biological matrices with strong acids (e.g., Flegal & Smith, 1995). Thus, samples were digested in 5 mL concentrated distilled nitric acid (16N HNO_3) for one night at 90 °C to break down the solid residue and evaporated to dryness the next day.

Although advancements in ICP-MS allow for fewer matrix-derived interferences, we found significantly low precision in ratios measured in blood samples after digestion. This is most likely due to high concentrations of biological matrices still present in the blood (Tagaki et al., 2011). For this reason, the samples were filtered using 0.45 μm syringe filters (Puradisk 25mm, #6780-2504, Whatman) before ion chromatography separation. Lead from blood samples was separated from other ions and concentrated using heavy metal ion selective resin (AG 1-X8, 100-200 mesh, #1401441, Bio-Rad) according to conventional techniques in our lab (e.g., Reimi & Marcantonio,

2016). The resulting concentrated sample was transferred to 50 mL polypropylene centrifuge tubes (#06-443-19, Fisherbrand) and diluted in a 2% nitric acid solution for isotopic analysis.

2.5. Environmental Sample Processing

Unlike the blood samples, all environmental samples were shipped directly to R. Ken Williams ’45 Radiogenic Isotope Laboratory facility after collection. The processing steps for environmental samples were similar to those of blood samples. First, the environmental samples were transferred to pre-washed Teflon digestion vessels and weighed on a digital scale. All dust wipes used during sampling were digested for Pb isotope analysis (1-2 moist wipes per sample), as were soil, spice, and paint chip samples, which were manually pulverized into fine power before being transferred into vessels to facilitate digestion.

Dust wipes, spices, and paint samples were digested in 5 mL of concentrated nitric acid. The digestion period varied depending on how easily the solid media was dissolved but ranged between 12 and 48 hours, resulting in a transparent liquid. Wipes used in House 3 did not completely dissolve and required an additional 5 mL of Optima-grade hydrogen peroxide (H_2O_2, #P170500, Fisher Scientific, MA) heated to 120°C to destroy solid particles. Soil samples were digested using a solution of 5 mL concentrated HF and 2 mL concentrated HNO_3 at 90°C for a week to destroy silicate matrices (Flegal & Smith, 1995). Then, samples were evaporated to dryness using a hot plate. All environmental samples went through the same process of ion exchange chromatography for lead separation, as described in the previous blood sample processing section.

Finally, the resulting lead-concentrated sample was pipetted into 50 mL centrifuge tubes and diluted with 2% nitric acid solution for isotopic analysis.

2.6. Lead Isotope Analysis

The Pb isotopic ratios were measured by multiple Faraday collector inductively coupled plasma mass spectrometry (MC-ICP-MS) with external mass bias corrections, given that there is only one stable isotope of Pb. All sample analysis was performed at Texas A&M University with the Neptune Plus multi-collector (Thermo Scientific Fisher, MA), which allows for high precision measurements of the isotopic lead ratios, $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$. A common lead isotopic standard from the National Institutes of Standards and Technology (NIST SRM 981) was repeatedly analyzed between every 2-3 lead samples to monitor for instrumental mass bias. The instrumental mass fractionation correction for the Pb isotope analysis using the sample-NIST SRM 981 standard bracketing methodology was checked in several samples by spiking samples internally with thallium (Chernyshev et al., 2007). Fractionation measured by monitoring $^{205}Tl/^{203}Tl$ ratios were identical, within error, to those measured using the standard-sample bracketing technique using the NIST SRM 981 standard, and averaged 5.5‰ +/- 0.2% (n= 23) over the course of this study. Isotopes of mercury (^{202}Hg and ^{204}Hg) were observed and measured routinely to correct for interference on ^{204}Pb.

Lead concentrations in blood samples were determined by isotope dilution mass spectrometry (IDMS) using the Element XR (Thermo Scientific Fisher, MA) and a synthetic and virtually pure ^{205}Pb isotope spike. A similar method was adopted to

determine the lead concentration in soils and spices. However, the same method could not be applied to dust samples because the wipes used for collection contained an unknown amount of water, whose weight was unknown.

3. RESULTS

The average BLL measured in the blood aliquots was 12.7 μg/dL, 3.5 μg/dL, and 2.9 μg/dL, for children living in House 1, House 2, and House 3, respectively. The current level of concern for Pb in children's blood, reduced in 2012 by the CDC, is 5 μg/dL, which places one of the children (House 1) well above the level for environmental investigation. Replicates of the concentrations and isotope ratios of blood Pb in each house were reproducible (**Table 2**). Not enough blood was collected from the child of House 3 to replicate the analysis.

Table 2. Lead isotope $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ ratios and concentrations for the blood samples and replicates for each child.

Blood Sample Replicate	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{206}Pb$	Pb Concentration (ug/dL)
House 1 A	1.2136	2.0303	12.70
House 1 B	1.2128	2.0206	12.78
House 2 A	1.2274	2.0223	3.59
House 2 B	1.2270	1.9977	3.55
House 2 C	1.2273	2.0104	3.53
House 3 A	1.1268	2.1323	2.89

To check the reproducibility of the mass spectrometric analysis, we compared the $^{206}Pb/^{207}Pb$ ratios obtained using the multi-collector Neptune Plus for a subset of samples (Houses 1 and 2) to those of the same samples, which were measured using the single-collector Element XR high resolution ICP-MS (Thermo Scientific Fisher, MA) (**Table 3**). Contrasting the Pb isotope ratio data obtained with the Neptune Plus (relative standard error of 0.01% at the 2-sigma level) with that for the Element XR (relative

standard error of 1% at the 2-sigma level), we find that, within error, all (except three) of the samples had identical $^{206}Pb/^{207}Pb$ ratios within error, suggesting an excellent reproducibility of the mass-spectrometric technique.

Table 3. Comparison of $^{206}Pb/^{207}Pb$ isotope ratios obtained by analyzing the same environmental samples from House 1 and House 2 with the Element XR and Neptune Plus mass spectrometers, demonstrating the external reproducibility of the mass spectrometric technique. Note that the number of significant figures for the single-collector Element XR data are lower given the two orders of magnitude greater precision of the multi-collector Neptune Plus data. Blank relative standard errors at the 2-sigma level for the Element XR analyses are 1.0% and 0.01% for the Neptune Plus analyses. Numbers in bold are outside of the relative standard error for both measurements and, interestingly, seem to occur when values measured on the Element XR are significantly higher than all other ratios measured. The Element XR ratios shown represent samples that did not go through ion chromatography separation, whereas the Neptune samples did. Matrix interferences are suspected to have caused the elevated ratios shown, thus reducing precision, and creating outliers.

Sample ID	Element XR $^{206}Pb/^{207}Pb$	Neptune Plus $^{206}Pb/^{207}Pb$
House 1		
S1	1.19	1.1963
PS1	1.20	1.2033
DS1	1.16	1.1741
DS2	1.28	1.2823
DS3	1.19	1.1905
DS4	**1.26**	**1.2188**
DS5	1.20	1.2046
DS6	1.17	1.1747
DS7	1.20	1.2029
DS8	1.21	1.1977
House 2		
S1A	1.22	1.2222
S2A	1.23	1.2262
S3A	1.22	1.2235
D4A	1.23	1.2234
D6A	1.24	1.2394
D7A	**1.25**	**1.2194**

Table 3. Continued

Sample ID	Element XR $^{206}Pb/^{207}Pb$	Neptune Plus $^{206}Pb/^{207}Pb$
House 2		
D8A	1.22	1.2289
D9A	**1.25**	**1.2265**
D11A	1.22	1.2216
D10A	1.22	1.2285
D14A	1.22	1.2264

All Pb isotope data (blood and environmental samples) are listed in **Table 4**. The crux of this technique is that the source of the Pb ingested or inhaled by a child can be identified by comparing the Pb isotope composition of the blood with that of various environmental sources of Pb in the home. A comparison of the Pb isotope ratios can be made using an Pb isotope cross-pot. On such a cross-plot, the Pb isotope ratios of the blood and the source would overlap or plot close to each other. Ideally, a three-dimensional isotope cross-plot would be best to look for matches in isotope ratios.

Table 4. Tabulated $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ ratios measured for all environmental and blood samples reported in this project, followed by their respective standard error.

Sample ID	Sample Type	$^{206}Pb/^{204}Pb$	Standard Error 2σ	$^{207}Pb/^{204}Pb$	Standard Error 2σ	$^{208}Pb/^{204}Pb$	Standard Error 2σ
House 1							
House 1 A	Blood	18.9980	0.0013	15.6543	0.0011	38.5716	0.0028
House 1 B	Blood	18.9875	0.0007	15.6558	0.0006	38.3669	0.0015
S1	Soil	18.6817	0.0003	15.6164	0.0003	38.2709	0.0008
PS1	Paint chips	18.8105	0.0007	15.6324	0.0006	38.4078	0.0014
DS1	Dust	18.2771	0.0002	15.5664	0.0002	37.8266	0.0006
DS2	Dust	20.1645	0.0003	15.7256	0.0003	39.5302	0.0008
DS3	Dust	18.5762	0.0008	15.6037	0.0007	38.4682	0.0020
DS4	Dust	19.0625	0.001	15.6406	0.001	38.5767	0.0040
DS5	Dust	18.8154	0.0003	15.6195	0.0003	38.3713	0.0009
DS6	Dust	18.3043	0.0002	15.5814	0.0002	38.0357	0.0005
DS7	Dust	18.8005	0.0003	15.6294	0.0004	38.3853	0.0011
DS8	Dust	18.6982	0.0028	15.6117	0.0024	38.2597	0.0059
House 2							
House 2 A	Blood	19.1875	0.003	15.6327	0.003	38.8037	0.007
House 2 B	Blood	19.2111	0.005	15.6573	0.004	38.3778	0.01
House 2 C	Blood	19.1933	0.006	15.6386	0.005	38.5868	0.012
S1A	Soil	19.1452	0.0004	15.6646	0.0004	38.6174	0.0001
S2A	Soil	19.2180	0.0002	15.6723	0.0003	38.6728	0.0008
S3A	Soil	19.1662	0.0003	15.6652	0.0003	38.6214	0.0008
D4A	Dust	19.1601	0.0006	15.6612	0.0006	38.6141	0.0016
D6A	Dust	19.4482	0.0004	15.6919	0.0005	39.0548	0.001
D7A	Dust	19.0937	0.0007	15.6584	0.0007	38.6205	0.0022
D8A	Dust	19.2561	0.0006	15.6693	0.0007	38.7260	0.0022
D9A	Dust	19.2098	0.0006	15.6621	0.0007	38.6460	0.003
D11A	Dust	19.1309	0.0002	15.6611	0.0003	38.5852	0.0008
D10A	Dust	19.2378	0.0004	15.6597	0.0005	38.6835	0.0017
D14A	Dust	19.2120	0.0004	15.6649	0.0005	38.6655	0.002

Table 4. Continued.

Sample ID	Sample Type	$^{206}Pb/^{204}Pb$	Standard Error 2σ	$^{207}Pb/^{204}Pb$	Standard Error 2σ	$^{208}Pb/^{204}Pb$	Standard Error 2σ
House 3							
House 3 A	Blood	17.5829	0.0271	15.6046	0.0239	37.4924	0.0578
S1	Soil	19.2123	0.0003	15.6495	0.0003	38.6991	0.0007
S2	Soil	19.1612	0.0002	15.6550	0.0002	38.7118	0.0005
D3	Dust	18.4642	0.0125	15.5338	0.0107	38.0854	0.0262
D4	Dust	18.0642	0.0027	15.5293	0.0022	37.5853	0.0055
D5	Dust	18.6448	0.0021	15.6637	0.0018	38.5151	0.0044
D6	Dust	18.5278	0.0159	15.6825	0.0135	38.4502	0.0331
D12	Dust	19.4182	0.1620	16.2841	0.1385	39.8375	0.3350
B7	Spice	17.5757	0.0004	15.5745	0.0004	37.4612	0.0010
B8	Spice	17.8548	0.0630	15.5205	0.0570	37.7055	0.1392
B11	Spice	17.8823	0.0227	15.6128	0.0202	37.9135	0.0495
B12	Spice	17.9179	0.0283	15.6426	0.0243	37.6125	0.0590

Here, results are illustrated on two-dimensional cross-plots, which are more convenient, to investigate the most evident similarities in Pb isotope ratios. In all of the cross-plots (**Figs. 1-3)**, the environmental samples from House 2 are isotopically similar to each other, creating a cluster of points. In contrast, isotope ratios for the samples from House 1 and House 3 have a greater range, with some overlap. In each home, blood samples from children do, indeed, have isotope ratios that lie close to those of some of the samples collected in each child's everyday environment. In the $^{207}Pb/^{204}Pb$ - $^{206}Pb/^{204}Pb$ plot, the similarity between the environmental and blood samples is difficult to make out (**Fig.1**). However, in $^{208}Pb/^{204}Pb$ - $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{206}Pb$ - $^{206}Pb/^{207}Pb$ isotope space (**Fig. 2 and Fig. 3**), the similarities between the blood and various environmental samples are much clearer and distinct.

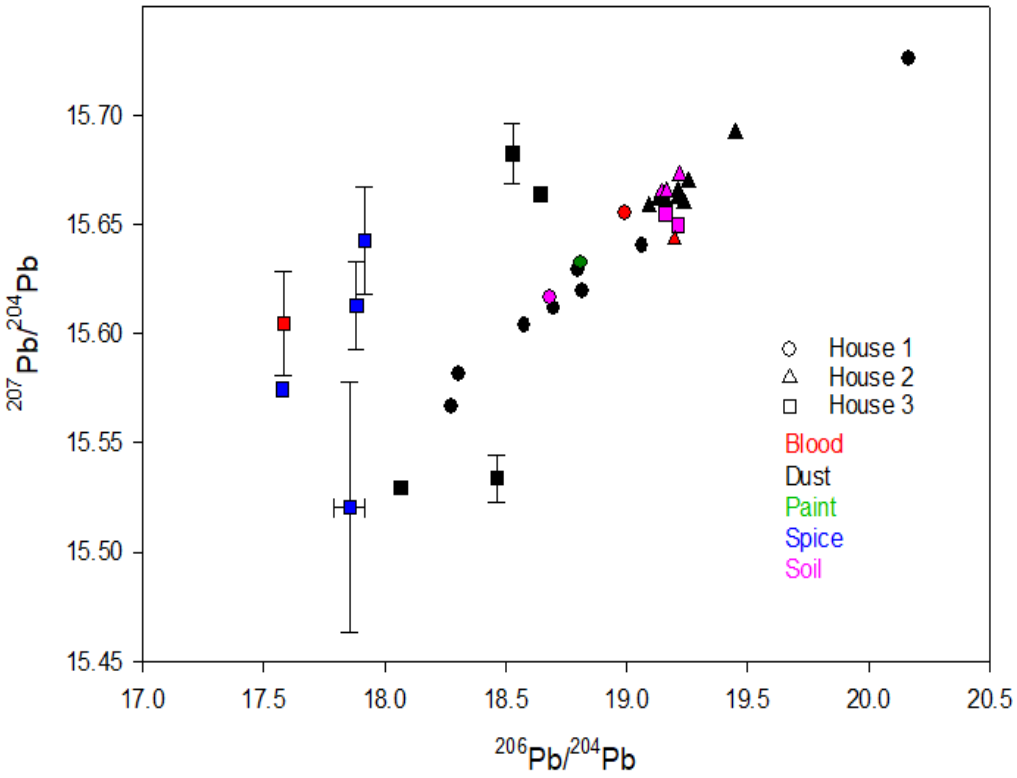

Figure 1. Plot correlating $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios of all the samples analyzed. Red symbols represent the average isotopic ratios in blood, green symbols represent paint samples, purple symbols represent soil samples, and black symbols symbolize dust samples. Error bars were omitted for most samples for being too small. The error bars for 3 of the 4 spice samples from House 3 are large because of low concentrations of Pb in these spices.

The cross-plot of $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ isotope ratios (**Fig. 2)** suggests that blood from House 2 is isotopically related to dust and soil from House 2. Although environmental samples from House 3 plot over a wide range, there appears to be a clear relationship between the blood sample of that house and one spice sample (**Figs. 2 and 3)**. For House 1, the blood sample is isotopically distinct from the environmental samples collected in the house (**Figs. 1-3**), and thus, the blood isotope composition may be a mixture of different environmental source compositions within the house, or it may

be derived from an entirely distinct environmental source that was not sampled in our study.

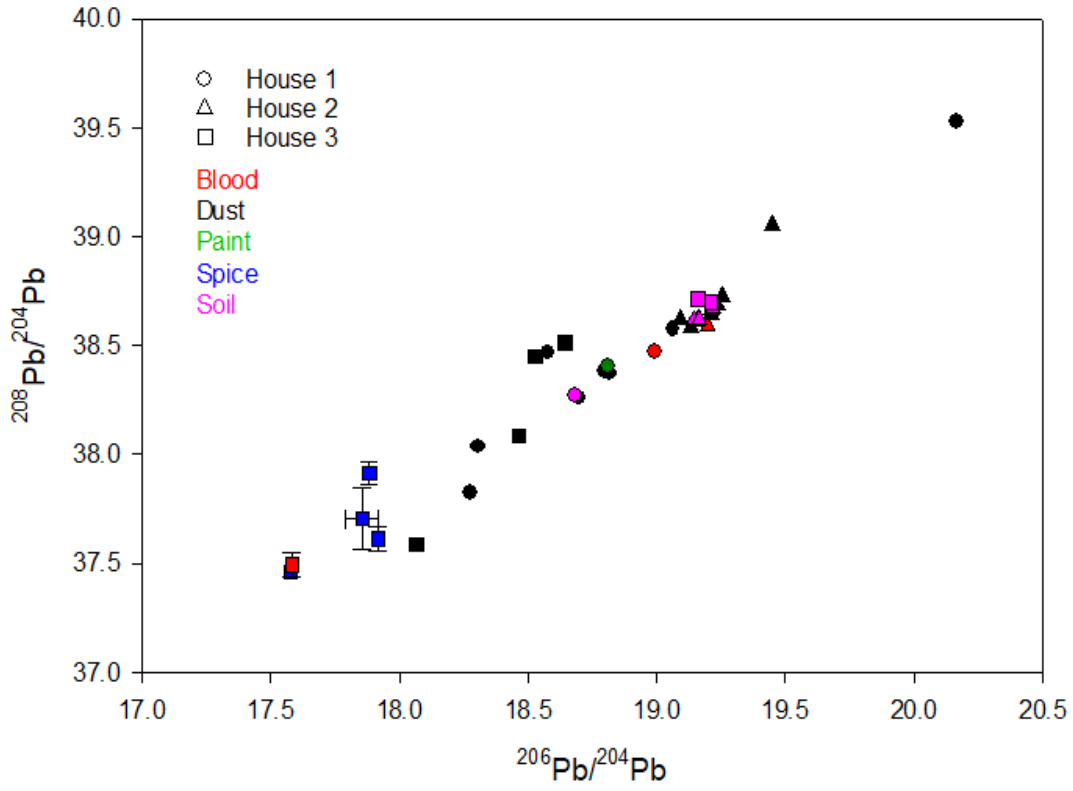

Figure 2. Cross-sectional plot of $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios measured in environmental and blood samples. Red symbols symbolize the average isotopic ratios in blood, green symbols represent paint samples, purple symbols represent soil samples, and black symbols symbolize dust samples. Correlation indicates that blood from House 2 is isotopically related to dust and soil from House 2 and blood from House 3 is isotopically similar to a spice sample. Error bars were omitted for most samples for being too small. The error bars for 3 of the 4 spice samples from House 3 are large because of low concentrations of Pb in these spices.

The relationship between the blood and the environmental samples collected in each house is most clear in the $^{206}Pb/^{207}Pb$ - $^{208}Pb/^{206}Pb$ isotope ratio cross-plot (**Figure 3**). The most pertinent observations in Figure 3 that need to be addressed are: 1) dust

from House 1 and House 3 shows the greatest range in Pb isotope ratios and may be related to the soils collected from near each home, whereas dust from House 2 is isotopically more homogeneous and, yet again, similar to the soils collected from the same home, 2) soils from House 2 and House 3 have isotope ratios that are similar, but different from those of the soil collected from House 1, 3) the environmental samples with $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ isotope ratios that were the closest to the same ratios measured in the children's blood samples were the kitchen baseboard (DS4) in House 1, the kitchen backdoor bottom board (D10A) in House 2, and the turmeric spice powder (B7) in House 3.

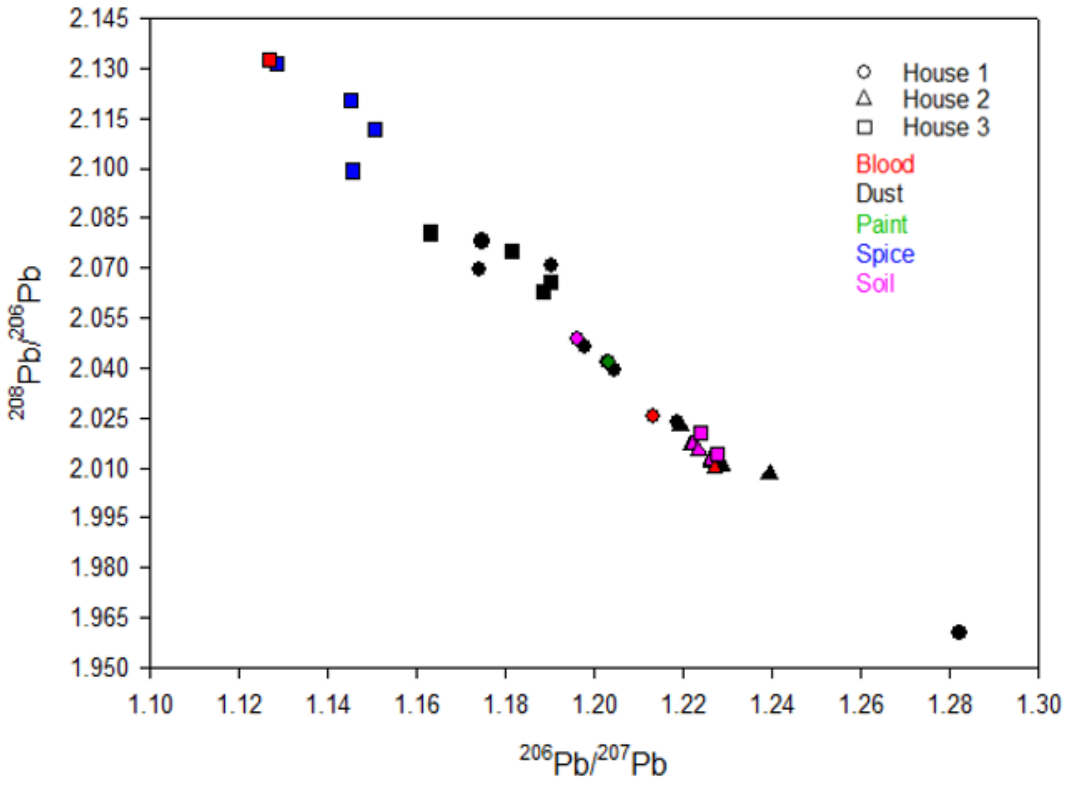

Figure 3. Cross-sectional plot of $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ ratios for all samples, ratios commonly used to identify the most likely source of exposure. Red markers symbolize the average isotopic ratios in blood, green markers represent paint samples, purple markers represent soil samples, and black markers symbolize dust samples.

4. DISCUSSION

These results add to the growing literature on poisoning due to uptake of Pb from environmental sources. Previous studies, together with the ones already discussed in this thesis, have identified elevated concentrations of Pb in blood using various techniques used in other countries, such as France (Leroyer et al., 2000; Tagne-Fotso et al., 2016), China (Lin et al., 2011; Chen et al., 2012), Nigeria (Bello et al., 2016), India (Chaudhary & Sharma, 2011), Czech Republic (Cikrt et al., 1997), Portugal (Reis et al., 2007), and Italy (Amodio-Cocchieri, 1996), among many others.

With a similar goal in mind, here we apply Pb isotope ratios as a tool for lead source apportionment focusing on the Kansas City, Missouri area. The children from Houses 2 and 3 showed a decrease in BLL between the first screening and the period of blood collection. After the first screening, health authorities flagged households (that we could in this study) which showed BLLs of children that were >5 ug/dL. However, the measured concentrations of Pb in blood at the time of collection was 3.5 μg/dL and 2.9 μg/dL for House 2 and 3, respectively, showing a reduction of BLL over time (i.e., between the time the health authorities measured the first blood sample and the time the latest sample of blood was measured, usually within a period of less than a year). The reduction could have been caused by natural excretion of Pb in the body if the child had not been in contact with the source of the exposure (or the exposure was more limited) since the initial health department screening. Excretion after exposure has been suggested by Pb biokinetic studies (e.g., Rabinowitz et al, 1973; Manton, 1977).

The samples that had Pb isotope ratios closest to that of child's blood were samples of dust collected from a kitchen baseboard in House 1 (DS4), a kitchen backdoor bottom board in House 2 (D10A), and turmeric spice powder used for cooking in House 3 (B7). For the first two houses, samples with Pb isotope ratios closest to the blood consisted of dust that was collected from the floor of areas that the child has easy access. These surfaces are found in common areas of the house and nearby entry points of dust and soil into the household as has been observed in other studies (e.g., Sayre et al., 1974; Sayre & Katzel, 1979; Charney et al., 1980; Rabinowitz et al, 1985; Lanphear et al. 1996; Cirkt et al., 1997; 1998a; 1998b; Gwiazda & Smith, 2000; Laidlaw et al., 2005; Gasana et al., 2006; Chen et al., 2012). The isotopic data from House 1 and 2 support previous research on dust Pb exposure and that dust intake is responsible for the elevated blood lead levels. A more detailed discussion of the results from each house follows.

4.1. House 1

Environmental samples from House 1 showed a large range in Pb isotope ratios (**Figs. 1, 2, and 3**). The absence of clustering between the Pb isotope ratios of dust samples (**Figs. 1, 2, and 3)** suggests distinct isotopic origins for the dust present in House 1. When comparing all ratios, one sample shows an extreme variation in Pb isotope ratio (DS2, door frame from living room to dining room) and plots as an outlier on the far bottom left in **Figure 3,** for example. This dust sample is isotopically distinct from all other household samples, suggesting it could be derived from a distinct source. Although only one soil sample was collected for this house, its Pb isotope composition is

remarkably similar to that of one of the dust samples collected (DS8, father's workpants). Besides sample DS8, none of the dust samples seem to be related nor derived from the outside soil. The only paint sample collected in House 1 (PS1, loose paint chips from the bathroom floor) had a completely different Pb isotope composition than that of the child's blood. [Note that we could not collect paint from the walls without prior approval from landlords, and this was not possible for this study.] In fact, the paint Pb isotope composition is remarkably similar to two other dust samples (DS5 and DS7), one of which (DS5) was also collected in the bathroom. The large difference between the Pb isotope compositions observed in the paint and blood suggests that paint ingestion is not the cause for the elevated BLL. This finding is surprising, considering how easy the ingestion of loose paint flakes on the ground should have been for the toddler in the house.

Ratios observed in blood were not related to the ratios observed in any environmental sample and no strong conclusion can be drawn for the sampled sources being responsible for the elevated BLL. From the data presented in this study, a dust sample from the kitchen floor (DS4, $^{208}Pb/^{206}Pb$= 2.0237, $^{206}Pb/^{207}Pb$= 1.2187) had a Pb isotope composition that was the closest to that of the blood ($^{208}Pb/^{206}Pb$= 2.025, $^{206}Pb/^{207}Pb$= 1.213), albeit the compositions did not overlap each other on any of the isotope cross-plots. Since the child in House 1 showed the highest BLL from all the participating children, we speculate that the elevated BLL may have been caused by a source that was not sampled in this study. In this case, our findings aid in excluding

sources that are not responsible for the child's exposure, rather than identifying the cause for elevated BLL.

4.2. House 2

The environmental samples collected in House 2 showed a narrower range in Pb isotope composition and plotted remarkably close to each other in the isotope cross-plots (e.g., **Fig. 3**). The Pb isotope ratios of the dust closely match those of the soil, suggesting that dust present in the household is derived from the outside. The data complements past Pb apportionment studies (Laidlaw et al., 2014) that suggest that indoor dust is derived from outside soils that are mobilized into the household. The para-occupational source in this house (D6A, father's workpants), identified as an outlier in this set of samples, was isotopically incompatible with the remaining dust and soil samples, suggesting that the Pb present on the workpants is derived from a source that is not present in the household. Soil samples showed strong homogeneity in Pb isotope ratios, which is expected as soil tends to be a homogeneous mixture with respect to Pb from atmospheric emissions.

The blood isotopic composition in House 2 is isotopically similar to several of the environmental samples. Indeed, in Figures 2 and 3, the blood Pb isotope composition is close to that of the dust and soil, consequently being included in the "clump" of data points. In this case, the Pb isotope ratios of blood ($^{208}Pb/^{206}Pb$= 2.0101, $^{206}Pb/^{207}Pb$= 1.2272) match the closest with ratios of indoor dust ($^{208}Pb/^{206}Pb$= 2.011, $^{206}Pb/^{207}Pb$= 1.228) collected by the kitchen door (D10A, kitchen backdoor bottom board) and one soil sample (S-2A, $^{208}Pb/^{206}Pb$= 2.012, $^{206}Pb/^{207}Pb$= 1.226). For that reason, and because

of how tightly related the sources and blood data points are with respect to Pb isotope composition, it is difficult to determine which source has had the greatest influences on the blood isotope composition, which may be moot given that the dust is likely derived from the soil. Indeed, we can surmise that dust ingestion, which is likely derived from the outside soil and found in reachable surfaces or entry points, is probably responsible for the BLL in the child from House 2. This finding is consistent with previous studies (Sayre & Katzel, 1979; Lanphear et al., 1996; 1998a; 1998b; Gwiazda & Smith, 2000) that identified indoor dust ingestion as a contributor to elevated BLL in children.

4.3. House 3

Similar to House 1, the environmental samples from House 3 showed significant heterogeneity in their Pb isotope compositions. Soil samples from House 3 plot close to soils from House 2 and are included in the cluster of points, supporting soil homogeneity with respect to atmospheric anthropogenic Pb emissions (Mielke et al., 1984; Lanphear et al., 1998a; Lin et al., 2011; Evans et al., 2018). Dust samples from House 3 plot as a scattered cloud and suggest no relationship with the soils collected from the exterior of the same house. The high error observed in some of the environmental samples (spices and dusts) and the blood sample from this house is attributed to the low Pb concentration in most of the spices, and the small quantity of blood collected (note in-run precision error bars in **Figs. 1 and 2**).

The child from House 3 had the lowest BLL of all children included in this study (2.9 μg/dL). The $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ isotope comparison shown in **Figure 3** indicates the turmeric sample (B7, $^{208}Pb/^{206}Pb$= 2.131, $^{206}Pb/^{207}Pb$= 1.128) as the

environmental source most isotopically similar to the blood ($^{208}Pb/^{206}Pb$= 2.132, $^{206}Pb/^{207}Pb$= 1.1268). Although unexpected, it has been theorized that small amounts of Pb are included in spices during manufacturing or being up taken from soil in which the plant used to make the spice grew (e.g., garden vegetables grown in high-Pb soil, Finster et al., 2004; tea plants grown in high-Pb soil, Han et al., 2006). The Pb concentration in the sample of turmeric was measured to be 6.86 ug/g. These data provide evidence that children can assimilate a concerning amount of lead from lead-bearing spices and food. This notion of elevated concentrations of Pb in spices is not surprising, as several studies have identified cooking spices as a potential source of lead uptake (Woolf & Woolf, 2005; Lin et al., 2009; Hore et al., 2019). Indeed, Angelon-Gaetz and coworkers (2018) also identified abnormally high Pb concentrations in turmeric spices. However, as far as we are aware, this study is the first to show isotopic evidence suggesting a cooking spice as a significant source of Pb. Other spices from the same house do not seem to contribute to the elevated BLL as much as the turmeric but have a Pb isotope composition much closer to the blood than the remaining environmental samples. Aside from the dietary samples, no environmental sample showed a Pb isotope composition that was remotely close to the blood and, consequently, ingestion of contaminated soils or dusts was discarded as the culprit for the child's elevated BLL.

5. CONCLUSIONS

We summarize our findings as follows:

- High-precision multi-collector ICP-MS allowed the identification of possible sources of a child's Pb exposure in two homes, House 2 (dusts and soil, or a mixture there of) and House 3 (turmeric spice). It is not always possible to directly link high BLLs to the environmental sources collected, as suggested by our results for House 1 in which the Pb isotope ratios of the dust samples were close to, but did not overlap with, those of the blood.
- For House 1 and House 2, the isotope ratios of the source of Pb contamination that were the closest to those found in the blood samples were dust samples on the floor found by house entry points (i.e. backdoor), as suggested in previous environmental investigations.
- For House 3, the isotope ratios measured in the blood are remarkably similar to the ratios observed in the turmeric sample, suggesting a direct link, through the ingestion of the spice, to the elevated BLL. Thus, food spices (such as turmeric in this study) can contain concentrations of Pb on the order of several ug/g that may serve as a route for Pb exposure in children.
- More studies applying mass-spectrometric techniques, particularly encompassing a higher number of participating houses, are encouraged and recommended to define the current situation of childhood lead poisoning and source apportionment in urban areas of the US.

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APPENDIX

SAMPLE IDENTIFICATION

Below is the list of all environmental samples included in this study, organized by house.

House 1			
ID	**Sample Type**	**Description**	**Collection Area**
DS-1	Dust	Font door bottom board	2in X 16in
DS-2	Dust	Door frame from living room to dining room	3in X 12in
DS-3	Dust	Cracked wall in dining room	3in X 6in
DS-4	Dust	Baseboard in kitchen	3in X 12in
DS-5	Dust	Cracked tiles in bathroom	4in X 4in
PS-1	Paint Chips	Paint chips on bathroom floor	Flakes on the ground
DS-6	Dust	Window sill in shower	3in X 12in
DS-7	Dust	Window sill by master bed	3in X 12in
DS-8	Dust	Father's work pants	5in X 5in
S-1	Soil	soil exposed near back porch steps	

House 2			
ID	**Sample Type**	**Description**	**Collection Area**
S-1A	Soil	Front yard, exposed soil near front sidewalk, composite of 4 samples	15 ft away fr home
S-2A	Soil	Front of the house, right side drip-zone, composite of 5 samples	Composite
S-3A	Soil	Front of the house, left side drip-zone, composite of 5 samples	Composite
D-4A	Dust	Front door, outside, near door handle	3in X 16in
D-6A	Dust	Father's workpants	9in X 4in
D-7A	Dust	Door frame to master bedroom	4in X 4in
D-8A	Dust	Air vent in dinning room	1in X 27in
D-9A	Dust	Dinning room window sill	3in X 13in
D-10A	Dust	Kitchen backdoor bottom board (wood)	3in X 13in
D-11A	Dust	Front door, bottom trim	3in X 15in
D-14A	Dust	Toys on front porch, under right window sill (Bear)	

House 3			
ID	**Sample Type**	**Description**	**Collection Area**
S1	Soil	Soil north of back patio	Composite
S2	Soil	Soil - NE corner of garden	Composite
D3	Dust	Metal bed frame - master bedroom	3 in X 6 in
D4	Dust	Bathtub faucet - master bath	3 in X 12 in
D5	Dust	Metal frame on ping-pong table	2 in X 17 in
D6	Dust	Metal front door handle	1 in X 5 in
B7	Spice	Turmeric	Bulk
B8	Spice	Red Chili Powder	Bulk
B9	Spice	Coriander Powder	Bulk
B10	Spice	Cumin Powder	Bulk
B11	Spice	Garam Masala	Bulk