

Using community science for detailed pollution research: A case-study approach in Indianapolis, IN, USA

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Abstract:

Heavy metal contamination in urban environments, particularly lead (Pb) pollution, is a health hazard both to humans and ecological systems. Despite wide recognition of urban metal pollution in many cities, there is still relatively limited research regarding heavy metal distribution and transport at the household-scale between soils and indoor dusts—the most important scale for actual human interaction and exposure. Thus, using community-scientist-generated samples in Indianapolis, IN (United States), we applied bulk chemistry, Pb isotopes, and scanning electron microscopy (SEM) to illustrate how detailed analytical techniques can aid in interpretation of Pb pollution distribution at the household-scale. Our techniques provide definitive evidence for Pb paint sourcing in some homes, while others may be polluted with Pb from past industrial/vehicular sources. SEM revealed anthropogenic particles suggestive of Pb paint and the widespread occurrence of Fe-rich metal anthropogenic spherules across all homes, indicative of pollutant transport processes. The variability of Pb pollution at the household scale evident in just four homes is a testament to the heterogeneity and complexity of urban pollution. Future urban pollution research efforts would do well to utilize these more detailed analytical methods on community sourced samples to gain better insight into where the Pb came from and how it currently exists in the environment. However, these methods should be applied after large-scale pollution screening techniques such as portable X-ray fluorescence (XRF), with more detailed analytical techniques focused on areas where bulk chemistry alone cannot pinpoint dominant pollution mechanisms and where community scientists can also give important metadata to support geochemical interpretations.

Keywords: Lead (Pb); heavy metals; urban pollution; Pb isotopes; SEM; community science

1. Introduction

Understanding heavy metal pollution in the urban environment is particularly important for the health and well-being of both humans and ecological systems, as many heavy metals can be toxic to organisms in excessive amounts. Of particular interest is Pb, as it can especially harm children and their developing brains (e.g., Egendorf et al., 2020) and it is established that there is no known safe level of Pb exposure in children (e.g., Betts, 2012). A challenge with identifying

and mitigating Pb exposure is that its distribution is heterogeneous in urban environments, both at the household and city-wide scale (e.g., Filippelli et al., 2018; Obeng-Gyasi et al., 2021).

One approach to better understand Pb heterogeneity in the urban environment is through large sampling datasets at high spatial resolution, collected by community scientists. Community scientists are local community members that can improve scientific research endeavors, bridge the gap between scientists and the general public, enhance the ability to collect samples not easily obtained by researchers alone (i.e., indoor home samples), and provide a link to understanding the critical issues facing communities. While community science thus far has made great strides in improving science communication and mapping Pb exposure risks (e.g., Filippelli et al., 2018; Ringwald et al., 2021; Watson et al., 2022), there is untapped potential for further research advancement via community science.

Although community science samples contain inherent variability with sampling techniques, even with clear instructions, the large sample size of community science endeavors often overcomes this limitation (Filippelli et al., 2018). However, a key question not addressed thus far in community science-based research is whether smaller subsets of samples can be utilized for more detailed geochemical techniques and analyses to understand pollution distributions, despite potential variability in sampling methodology?

Lead stable isotopes have frequently been used for tracing pollution sources of Pb in the environment as a more detailed geochemical technique (e.g., Adgate et al., 1998; Jaeger et al., 1998; Sutherland et al., 2003; Wang et al., 2019), largely due to unique Pb isotopic ratios of many Pb sources and minimal environmental and biological fractionation of Pb isotopes (e.g., Ayuso & Foley, 2020). Despite their applicability in pollution source apportionment studies, complexities arise in urban settings when multiple pollution sources may be present. Additionally, while original Pb ores may oftentimes contain unique Pb isotopic signatures, the diverse range of Pb isotopic ratios in major urban sources such as paint and gasoline can lead to overlap in source ratios (e.g., Resongles et al., 2021; Wang et al., 2021). This can complicate interpretations of Pb pollution sources in environmental media such as soils and dusts, even within a single indoor home, where multiple types of Pb paint, with vastly different isotopic ratios, may persist (Jaeger et al., 1998). Thus, it is often important to not only rely on Pb isotopic ratios when determining pollution sources in the urban environment, but to incorporate other techniques such as bulk chemistry and scanning electron microscopy (SEM) as well. SEM in particular can identify the nature of pollutant particulates (i.e., metal fragments, combustion spherules, etc.) (e.g., Dietrich et al., 2019; Gaberšek & Gosar, 2021), which can help identify the actual form of Pb pollution and thus help differentiate between potential sources when used with other analytical techniques (e.g., Miler & Gosar, 2019).

Both SEM and Pb isotope techniques in tandem provide more detailed analytical approaches that can help better parse out Pb pollution sourcing and source distribution compared to bulk chemistry alone. Through four community scientist homes, we provide a case study of how detailed analytical techniques can create a more holistic picture of Pb pollution sourcing and transport in an urban environment. We focus on one typical Midwest US city with known elevated Pb concentrations (Indianapolis, Indiana) as a case study because of the abundance of

bulk soil Pb data already collected in the area (Filippelli et al., 2018). We aim to characterize household-scale soil and indoor dust samples to better quantify how Pb sources (i.e., paint and gasoline) are dispersed, and to illustrate how our methodology can be utilized in subsets of large-scale community science projects to address complex patterns uninterpretable based on bulk chemistry alone.

2. Materials and Methods

2.1 Sampling and Preparation

Soil and dust samples were provided by local community members from four households in Indianapolis (Fig. S1) as part of the Anthropocene Network (<https://anthropocenes.org/lead>), which provides free household screening for Pb (water, dust, and soil sampling) and aims to address issues of environmental equity in the age of the Anthropocene. The community science relationship was enabled through partnerships with local faith leaders in churches and the assurance of participant anonymity, which allowed for sample collection at homes that would otherwise be inaccessible. Residents were instructed to collect bulk vacuum cleaner dust in a sealed plastic bag, as well as soil (upper 0-5 cm) at their home's dripline (directly adjacent to home), the middle of their yard, and directly adjacent to the street with any type of scoop available at their home. Soil samples (~25 g. or more) were placed in separate plastic bags. Each vacuum dust sample was a composite of the entire home, and each soil sample was an individual, non-composite sample. We only selected sampling kits that had clearly labeled sample bags, indicative that the participants followed sampling instructions. Households that were spread out across Indianapolis was also taken into consideration to capture as much potential representation of the city as possible. A small sample size was used because the purpose of this research was to serve as a case study illustrating the ability to utilize community science samples for more detailed analytical research outside bulk soil screening for Pb.

All Pb results in the Anthropocene Network were communicated back to anonymous participants with tips on how to mitigate Pb exposure. Because of the emphasis on anonymity, there is sparse metadata for this community science partnership, but other community science endeavors have focused more on participant metadata information (e.g., Dietrich et al., 2022), which can help in interpretation of geochemical results and determining efficient pollutant remediation strategies.

Dust samples were sieved at 250 μm and were dry due to the nature of sampling, and thus needed no desiccation. Soil samples were air dried, sieved at 2 mm, then powdered and homogenized utilizing an agate mortar and pestle prior to acid digestion. Samples were weighed (~300-400 mg) into Savillex screw-cap vessels and digested utilizing a multi-stage, three-acid digestion procedure (HF, HNO₃, and HCl), following methods by Rader et al. (2021). Samples were then suspended in 2mL of distilled 8M HNO₃ for column chromatography. A small aliquot of this solution was extracted and diluted for concentration analysis prior to chromatographic separation. This is defined as total soil Pb for the purposes of this study.

Samples were purified for Pb isotope analysis via column chromatography utilizing Eichrom Sr Spec resin, slightly modified from that described by Deniel & Pin (2001) and Thibodeau et al. (2007). After purification, samples were diluted accordingly after a concentration check and spiked with NIST 997 thallium (Tl) standard to achieve a Pb/Tl ratio of ~4.

2.2 Instrument Analyses

The metal Pb [along with cadmium (Cd), Tl, and antimony (Sb)—which are reported because of their sparseness in the U.S. urban soil/dust literature] was quantified via an Agilent 7700 Inductively Coupled Plasma Mass Spectrometer (ICP-MS), operating in no gas mode. The Pb isotopes ^{204}Pb , ^{206}Pb , ^{207}Pb , and ^{208}Pb were detected with a Nu Plasma II Multicollector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS), with the Pb isotope ratios 208/206, 207/206, 208/207, 206/204, 207/204, and 208/204 reported after being normalized to Galer & Abouchami (1998). All ICP analyses were completed at Indiana University-Bloomington in the Department of Earth and Atmospheric Sciences' Metal Isotopes Laboratory. More details are provided in Text S1.

Dust and soil samples were prepared for SEM and energy dispersive X-ray spectroscopy (EDS) analysis on aluminum samples stubs using carbon sticky tab substrates. EDS lines used to identify Pb specifically include the $L_{\alpha} = 10.541$ keV (nominally $M_{\alpha} = 2.342$ keV, $M_{\beta} = 2.444$ keV). All SEM-EDS analyses were conducted at Indiana University-Purdue University Indianapolis, using a Zeiss EVO-10 SEM and Bruker XFlash6, 60 mm² EDS detector. Backscatter electron (BSE) images and EDS data were collected at a setting of 20 kV in variable pressure mode.

2.3 Quality Control

Lead total concentration was deemed acceptable based on a mean recovery of $89.4\% \pm 11.0\%$ ($n = 5$) for NIST SRM 2702 (Table S1) and runs of USGS standard AGV-2a and NIST 2702 were both in agreement with previously published Pb isotopic compositions (see Text S2 and Table S2). More details on quality control are provided in Text S2.

3. Results and Discussion

3.1 General Soil and Dust Pb Enrichment and Relationships

Regardless of sample type or location, nearly all samples are enriched relative to the 75th percentile of background soil samples [upper 5 cm of soil, air dried <2 mm fraction crushed to <150 μm prior to a near-total four-acid (hydrochloric, nitric, hydrofluoric, and perchloric) digestion at a temperature between 125 and 150 °C] from throughout Indiana for Pb (Fig. S2; Table S3) (Smith et al., 2013). The 75th percentile value for background soil in Indiana is 25.6

mg/kg for Pb. Lead concentrations are ≥ 80 mg/kg in nearly all dust and soil samples (Fig. 1B; Table S3). The general anthropogenic enrichment of Pb (at concentrations in the hundreds to thousands of mg/kg) has been well documented in Indianapolis in urban soils (Filippelli et al., 2018).

Although the dust samples were sieved to a finer particle fraction and finer sized particles tend to contain higher concentrations of heavy metals (e.g., Herngren et al., 2006; Tansel & Rafiuddin, 2016), bulk geochemistry results indicate that dust is not consistently higher in metal concentration than the soil samples, even within the same house location (Fig. 1B; Fig. S2). This may indicate that the dominant anthropogenic source for these elements in both media is coarser material, and the interior loading reflects both indoor dust and finer particle invasion from outside. This emphasizes recent work by Gillings et al. (2022) that demonstrates how relationships between indoor and outdoor sources are not always predictable, and inferences on one based on the other should be taken with caution.

It is noted that we examined the bulk grain size (< 2 mm) composition of outdoor soil samples, and that grain size may affect metal concentrations and partitioning of Pb sources. However, we emphasize that bulk grain size is the most representative way to encapsulate all possible Pb exposure within a household environment. While finer sized particles may be a greater inhalation and transport risk, larger sized particles may degrade over time and still supply a reservoir of Pb pollution.

3.2 Bulk Pb Concentration and Pb Isotope Heterogeneity

Three homes displayed clear heterogeneity in bulk metal concentrations of Pb depending on sample location at the home, with only House 1 showing more consistent metal trends for Pb (Fig. 1B; other metals displayed in Fig. S3). This heterogeneity extends to Pb isotopic ratios (Table S4) such as $^{206}\text{Pb}/^{204}\text{Pb}$, although House 2 displayed fairly consistent $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic ratios (Fig. 1A), with slightly more variance in $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios (Fig. S4). This matches previous research in urban centers that documented extensive heterogeneity at the household-scale for both bulk metal concentrations such as Pb (e.g., Filippelli et al., 2018; Obeng-Gyasi et al., 2021; Wade et al., 2021) and for Pb isotopic composition (Wang et al., 2021). Due to our small sample size and no consistent trends between Pb isotopes or bulk metal soil concentrations based on sample location, we cannot make any broad generalizations on spatial relationships between dripline, yard, and streetside soils such as larger studies in Indianapolis, IN (Filippelli et al., 2018), Durham, NC (Wade et al., 2021) and Greensboro, NC (Obeng-Gyasi et al., 2021) have done. However, when using bulk Pb concentrations, Pb isotope ratios, and SEM collectively at a house-by-house basis, clearer interpretations of Pb pollution can be made.

3.3 Pb Pollution Sources and Pb Distribution by Household—Synthesis of Analytical Methods

House 1:

The $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic ratio changes significantly depending on the sample location and between the indoor dust and outdoor soil, increasing towards the street, with the dust sample containing the lowest $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic ratio (Fig. 1A). This is not clearly reflected in bulk Pb concentration though, which remains fairly consistent throughout the property, between 70 and 130 mg/kg (Fig. 1B; Table S3). Based on Pb isotopes alone, one would interpret a greater proportion of geogenic background glacial till (indicative of surficial Wisconsin glaciation sediments in the area) contribution closer to the street (Fig. S5). However, this is unlikely, because Pb concentrations did not decrease toward the street (Fig. 1B), which would be expected based on much lower Pb concentrations in geogenic background till (Barnes et al., 2020; Kousehlar & Widom, 2020). Thus, the Pb changes in isotopic composition must have come from an additional source of Pb such as leaded paint or leaded gasoline. Microscopy revealed only a few instances of possible Pb paint particles, but routinely revealed evidence of high temperature anthropogenic process Fe-rich spherules at all soil locations (Fig. 2). Thus, while Pb paint likely contributed to bulk Pb inside the home (Fig. 2), the scarcity of distinguishable paint particles via SEM in the soils and prevalence of anthropogenic spherules suggests the possibility of vehicular/industrial Pb sourcing, such as from residual microscopic particulate leaded gasoline or other vehicle Pb-wear (i.e., wheel weights) too small to detect via SEM.

House 2:

There is greater bulk Pb variability in samples than $^{206}\text{Pb}/^{204}\text{Pb}$ ratios on the property (Fig. 1). This includes a dripline soil sample that was 4 mg/kg Pb, and resembled mulching material. Thus, although the house samples are grouped closely on bivariate Pb isotope plots (Fig. S5), Pb sourcing is potentially different between the 4 mg/kg Pb dripline sample and other, more elevated Pb streetside & yard samples. Yard and streetside Pb was much higher than inside the home (Fig. 1B), with SEM imaging revealing no Pb-rich particles inside the home, but several apparent paint Pb-rich particles at the streetside and within the yard, similar to apparent paint particles in Dietrich et al. (2022) (Fig. 2). The prevalence of Pb-rich particles at these locations supports the higher bulk Pb concentrations and suggests that Pb paint abundance outside the home is greater than inside the home, even though Pb isotopic ratios are similar. This may be because of peeling exterior Pb paint that has been reworked in the outdoor environment and covered at the dripline by recent mulch material. There still appears to be an exchange of material across all household samples, as Fe spherules were also found at this household at all locations, even the mulched dripline. However, the spherules at the dripline were less Fe-rich (Fig. S11) than other spherules, suggesting they may have degraded more over time with minerals (i.e., clays, apatite) adhering or precipitating on the spherule structure, or because they were formed under different conditions or processes.

House 3:

Bulk Pb concentrations increase toward the street in soils and are the lowest in dust (Fig. 1B; Table S3). $^{206}\text{Pb}/^{204}\text{Pb}$ ratios increase from the dust to the dripline to the yard, but drastically decrease at the streetside (Fig. 1A). SEM did not reveal any Pb paint particles at any location at the home, or any Pb-rich particles of any nature. This suggests reworked, reprecipitated Pb is prevalent in this home environment, likely from historic leaded gasoline because of the increase

in Pb concentration towards the street (Fig. 2). Because all samples contain anthropogenic metal spherules, this illustrates the potential for distribution and transport of microscopic aerosol Pb to the home dripline and indoor environment from a distance—either industrial or vehicularly sourced. The widespread dispersion of vehicular Pb from historic leaded gasoline has been observed in atmospheric aerosols in London, U.K. (Resongles et al., 2021), suggesting the potential for the same type of dispersal here. Additionally, the Pb isotopic ratios for streetside soil at this home contain the least ^{206}Pb , ^{207}Pb , and ^{208}Pb relative to ^{204}Pb , which closely aligns with the Pb isotopic range of leaded gasoline (Fig. S5), and House 3 was near a road with the highest modern daily traffic volume compared to any other house in this study (Table S5)—historic traffic volume was likely also high given the urban location.

House 4:

Bulk Pb concentration is highly variable depending on sample location, at the highest (> 800 mg/kg) in the dust and dripline soil before decreasing significantly in the yard soil, then increasing in the streetside soil (Fig. 1B). The $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic ratio also changes, increasing from the dust to the dripline soil, then decreasing again for yard and streetside soil (Fig. 1A). Based on microscopy, the dust and dripline soils commonly contain Pb-rich particles resembling paint chips (Fig. 2), and the differences in Pb isotope ratios may be because of different layers or types of paint in the interior versus exterior of the home. Microscopy did not reveal any obvious Pb paint chips in the streetside or yard soils, but did reveal numerous high temperature anthropogenic process Fe-rich spherules, which were evident in both the streetside and dripline soil (Fig. 2). Although there may be mixing of microscopic Pb-rich particles from past vehicular exhaust or other industrial sources at the dripline, that soil is likely more dominated by Pb paint relative to streetside soils, where no Pb paint particles could be easily identified. Thus, because the bulk Pb concentration was still elevated well above background soils (Smith et al., 2013) at the streetside and no Pb paint could be readily identified, residual leaded gasoline, vehicle wear, or other industrial processes was likely the main source of Pb there.

3.3.1 Summary and Comparison of Household Pb Sourcing

The primary Pb sources in all homes were likely Pb-based paint or the remnants of leaded gasoline, supportive of previous literature in urban environments within the U.S. (e.g., Dietrich et al., 2022; Wang et al., 2021). However, each home had different patterns of Pb pollution sources (Table 1) and Pb concentrations (Fig. 1B). This did not seem to be dependent on larger-scale Pb relationships in Indianapolis, such as those at the zip code level (Filippelli et al., 2018). For example, homes 1, 2, and 4 resided in zip codes where reported median Pb concentrations in yard soils ranged from 183-263 mg/kg (Filippelli et al., 2018). However, the Pb concentrations in these homes were widely variable (4-2100 mg/kg; Table S3), and the likely Pb sources changed between paint and vehicular/industrial (i.e., leaded gasoline) both between homes and between sampling location within the homes. Thus, even our small number of households examined in this study exemplifies the fact that generalizations of Pb pollution at larger spatial scales should be made with caution, and multiple other external factors can affect Pb distributions at fine scales such as the history of the home and proximity to major roadways.

3.4 Similarities in Pb Pollution Between Households

Influences on metal concentrations in dusts and soils at the household-scale include not only potential pollution sources, but how dust and soil get reworked and distributed throughout the environment. In general, although there was much Pb pollution heterogeneity between households (Fig. 1), a commonality was the presence of Fe-rich high temperature anthropogenic process spherules at every home within multiple samples (Fig. 2). This entailed homes where distance from road to home was between 7-14 meters, nearby daily traffic volume was highly variable, and where there were multiple possible industrial sources (Table S5). These spherules are commonly seen in industrial urban areas (e.g., Dietrich et al., 2019; Gaberšek & Gosar, 2021) and are likely anthropogenic. This not only suggests that there is exchange of pollutant particles to the dripline of the home, supporting the hypothesis that the side of the home can act as a barrier for particulates to fall following roadside resuspension (e.g., Filippelli et al., 2018; Laidlaw & Filippelli, 2008), but also supports previous studies where there was exchange of outdoor pollution to the indoors (e.g., Adgate et al., 1998; Kelepertzis et al., 2020). Thus, transport of pollutants in soils/dust at the household-scale is likely an additional factor affecting Pb heterogeneity.

Future research should therefore consider the ability of metal pollution transport and reworking across neighboring homes and properties. Recent research in other urban settings (i.e., London) has also pointed out the continued reworking and widespread distribution of historic Pb pollutant particles throughout the environment in the form of atmospheric aerosols (Resongles et al., 2021).

3.5 Limitations of Only Pb Isotopes for Pollution Sourcing

Using Pb isotopes or bulk chemistry alone can be problematic for determining pollutant sourcing, because even within the same home, Pb isotopic composition of paints can vary drastically (Jaeger et al., 1998), and the Pb isotopic ratios for leaded gasoline were widely variable within the U.S. because of changing ore sources over time (e.g., Dietrich et al., 2021). For example, nearly all dust and soil samples plot within 1σ variability of leaded gasoline and lead paint source endmembers depending on the Pb isotope ratios used (Fig. S5). Thus, although previous interpretations in urban settings within the U.S. have concluded Pb paint sources likely dominating soils close to older homes (i.e., built prior to 1978) and leaded gasoline significantly contributing to soils near roadways (e.g., Wade et al., 2021; Wang et al., 2021), we utilized SEM imaging and EDS spectra to aid in interpretation of our variation in bulk Pb and Pb isotopic ratios at the household-scale. This multi-analytical approach is often necessary in complex urban settings where simple binary pollution mixing models of Pb sources will not work—particularly with the increasing overlap of stable Pb isotope signatures of multiple anthropogenic sources (e.g., Resongles et al., 2021).

4. Implications

Urban environments contain complex, heterogeneous distributions of heavy metals in soils and dusts, particularly Pb. This is especially evident in a large, post-industrial city such as Indianapolis, IN where historic Pb sources such as leaded paint and leaded gasoline contaminate soils and dusts in varying ways, as shown with differences between our study's household samples. Community science can provide a large sampling set to gather Pb data from, which can effectively help map Pb hotspots, general trends, and inform people of risks in their home (e.g., Filippelli et al., 2018; Watson et al., 2022). However, there are often nuances associated with pollution interpretations based on bulk chemistry alone, and more detailed analytical methods can help better understand Pb heterogeneity.

Here, we demonstrated how detailed methodology such as SEM and Pb isotopes can give a clearer picture of Pb pollution source distribution on a property. This approach can be utilized for even larger subsets of community science samples to better understand pollution variability in bulk chemistry following initial screening through techniques such as X-ray fluorescence (XRF). While there is inherent uncertainty in sampling with community scientists, as long as general instructions are followed, enough information may be gathered to obtain informative and actionable insight into pollution sources distributions. For example, whether Pb pollution from the home reaches the street, or whether Pb pollution from the street reaches the home. This information can therefore aid in home pollution remediation and prevention by determining whether there is significant transport and exchange of Pb pollution sources.

Additionally, working more with community members as real partners in research can be helpful for both gaining useful information to interpret Pb pollution results and helping the community members to place these results in their household context. This is a different approach that "parachute science" and has more lasting effects in changing behavior (Hayhow et al., 2021). For example, we recommend surveying community members first about their concerns and gathering important metadata during the course of the study, such as the age of their home, whether there has been recent renovation, worries of a particular Pb source nearby, etc.

Future studies in urban centers throughout the world would do well to capitalize on community science and the multi-analytical techniques such as those used here and in other recent urban metal pollution studies (e.g., Gaberšek & Gosar, 2021; Kelepertzis et al., 2021) to more fully understand the nuance associated with urban metal pollution distributions. This can be important in determining whether a pollutant source is coming from a property or elsewhere, the nature of the pollutant particles (i.e., are there Pb particles that are small enough to be an inhalation risk), and whether there is transfer of material from inside to outside a home. All these details are helpful from both a health risk and remediation perspective.

These techniques should be particularly used more within the U.S. because of the abundance of urban centers with varying legacies of pollution, the demographic and climate diversity, and the growing popularity of community science research providing an abundance of samples to study (e.g., Filippelli et al., 2018; Ringwald et al., 2021). These community science

endeavors offer a cost-effective way to systematically choose subsets of samples for more detailed multi-analytical approaches that provide important and scalable insight into Pb pollution. Lastly, these community science efforts expand the possible options for sample accessibility to researchers, such as to household indoor dust samples.

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Manuscript Tables:

Table 1: Summary of the likely primary Pb pollution sources by house and sampling location.

House	Indoor Dust	Dripline	Yard	Streetside
1	Paint	Vehicular or industrial	Vehicular or industrial	Vehicular or industrial
2	Potentially Paint	None	Paint	Paint
3	Vehicular or industrial	Vehicular or industrial	Vehicular or industrial	Vehicular or industrial
4	Paint	Paint	Potentially Paint/Vehicular	Vehicular or industrial

Manuscript Figures:

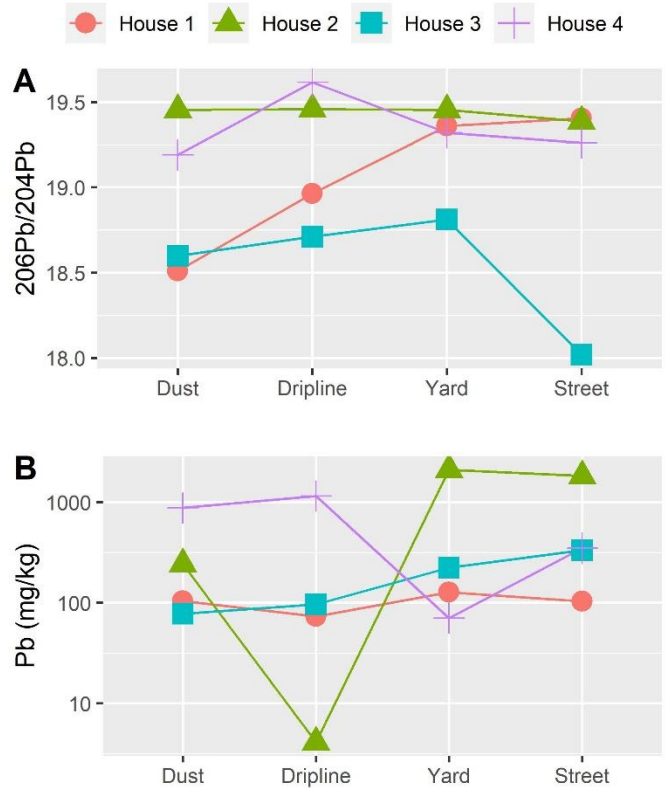


Fig. 1: Trends in $^{206}\text{Pb}/^{204}\text{Pb}$ based on sample type and house (A), as well as trends in bulk Pb concentration (mg/kg) based on sample type and house (B). Similar trends were seen for other isotopic ratios, albeit with slight variability (Fig. S4).

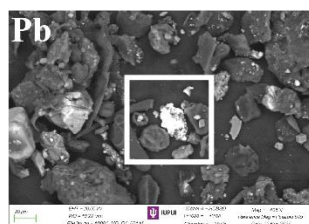
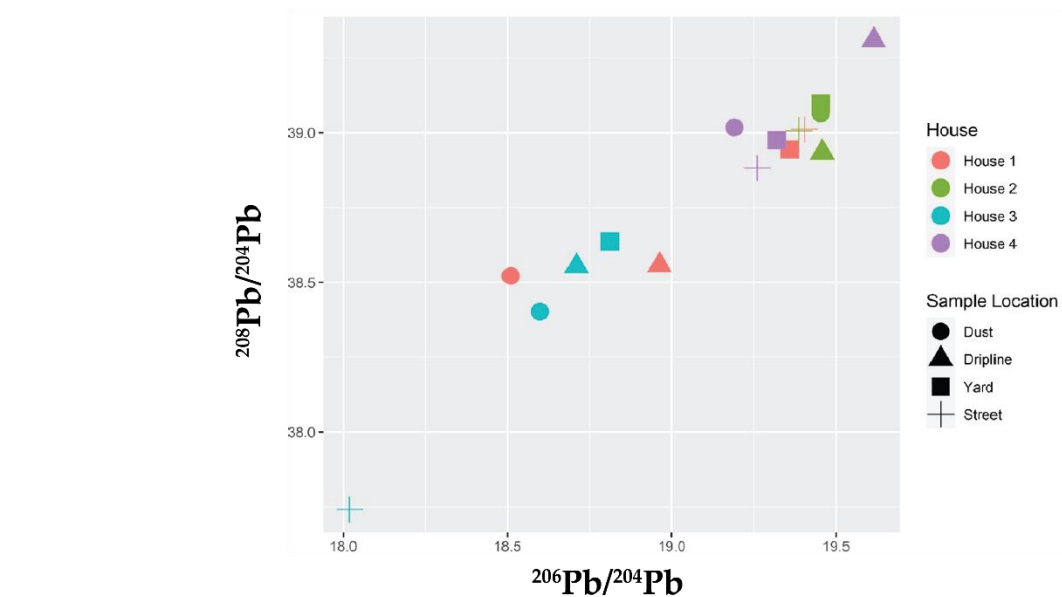
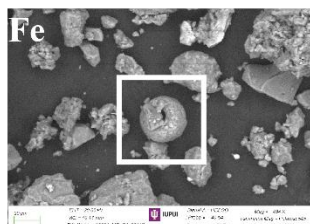
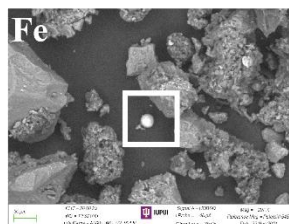
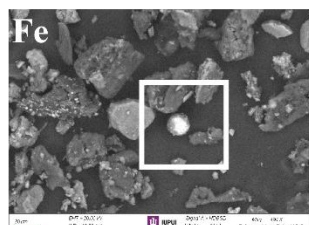
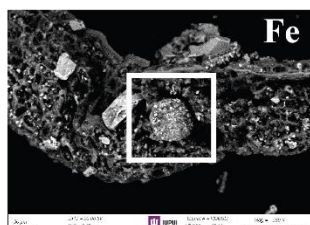
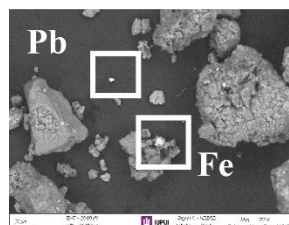
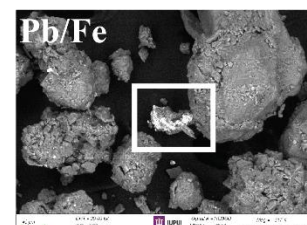
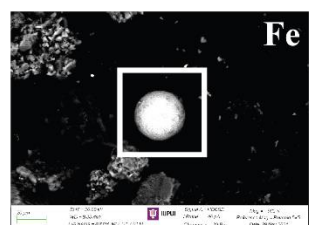
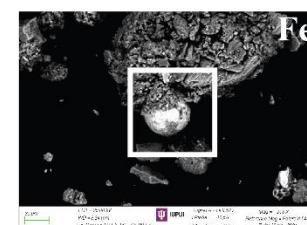
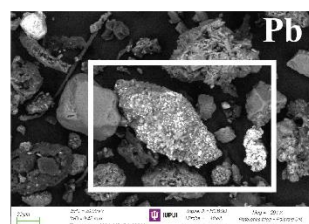
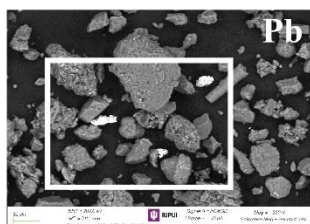
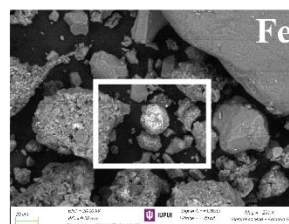
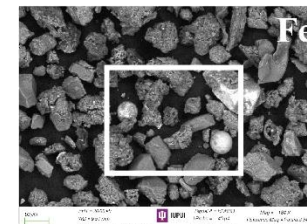
House 1: Indoor Dust¹House 1: Dripline²House 1: Streetside³House 1: Yard⁴House 2: Indoor Dust⁵House 2: Dripline⁶House 2: Streetside⁷House 2: Yard⁸House 3: Indoor Dust⁹House 3: Dripline¹⁰House 3: Streetside¹¹House 3: Yard¹²House 4: Indoor Dust¹³House 4: Dripline¹⁴House 4: Dripline¹⁵House 4: Streetside¹⁶

Fig. 2: Bivariate plot of Pb isotopic ratios, grouped by house and sample location. 2σ analytical variability is minimal relative to the sample symbol size and are thus only displayed in Table S2. SEM images are listed below the plot, with anthropogenic particles outlined in white. The corresponding EDS spectra for each superscript notation are listed in the Supplementary Information (Figs. S6-S21). Images labeled “Pb” contain Pb-rich particles within the white outline, while images labeled “Fe” contain Fe-rich particles within the white outline. It is noted that the paint particles in House 2: Streetside⁷, House 4: Indoor Dust¹³ and House 4: Dripline¹⁴ also contain traces of Fe.

Statements and Declarations

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Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author Contributions

Matthew Dietrich: Conceptualization, Funding acquisition, Investigation, Methodology, Visualization, Writing – original draft, Data curation. **Shelby T. Rader:** Methodology, Writing – review & editing. **Gabriel M. Filippelli:** Resources, Supervision, Writing – review & editing.

Ethical Approval

“Not applicable”

Consent to Participate

“Not applicable”

Consent to Publish

All authors agree to publish the manuscript in its present form.

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All data and materials are included in the manuscript and supplementary documentation.

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