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

Matthew Dietrich¹, Shelby Rader², and Gabriel Filippelli³

¹Indiana University Purdue University-Indianapolis ²Indiana University ³Indiana University-Purdue University Indianapolis

November 21, 2022

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-scientistgenerated 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.

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

3 Matthew Dietrich^{1*}, Shelby Rader², Gabriel Filippelli^{1,3}

⁴ ¹Department of Earth Sciences, Indiana University – Purdue University Indianapolis,

5 Indianapolis, IN, USA

⁶ ²Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, IN, USA

7 ³Environmental Resilience Institute, Indiana University, IN, USA

8 *Corresponding Author: <u>mjdietri@iu.edu</u>

9 Abstract:

10 Heavy metal contamination in urban environments, particularly lead (Pb) pollution, is a 11 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 12 13 distribution and transport at the household-scale between soils and indoor dusts-the most 14 important scale for actual human interaction and exposure. Thus, using community-scientist-15 generated samples in Indianapolis, IN (United States), we applied bulk chemistry, Pb isotopes, 16 and scanning electron microscopy (SEM) to illustrate how detailed analytical techniques can aid 17 in interpretation of Pb pollution distribution at the household-scale. Our techniques provide 18 definitive evidence for Pb paint sourcing in some homes, while others may be polluted with Pb 19 from past industrial/vehicular sources. SEM revealed anthropogenic particles suggestive of Pb 20 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 21 22 evident in just four homes is a testament to the heterogeneity and complexity of urban pollution. 23 Future urban pollution research efforts would do well to utilize these more detailed analytical 24 methods on community sourced samples to gain better insight into where the Pb came from and 25 how it currently exists in the environment. However, these methods should be applied after 26 large-scale pollution screening techniques such as portable X-ray fluorescence (XRF), with more 27 detailed analytical techniques focused on areas where bulk chemistry alone cannot pinpoint 28 dominant pollution mechanisms and where community scientists can also give important 29 metadata to support geochemical interpretations. 30 **Keywords:** Lead (Pb); heavy metals; urban pollution; Pb isotopes; SEM; community science

31

32 **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).

40 One approach to better understand Pb heterogeneity in the urban environment is through 41 large sampling datasets at high spatial resolution, collected by community scientists. Community 42 scientists are local community members that can improve scientific research endeavors, bridge 43 the gap between scientists and the general public, enhance the ability to collect samples not 44 easily obtained by researchers alone (i.e., indoor home samples), and provide a link to 45 understanding the critical issues facing communities. While community science thus far has 46 made great strides in improving science communication and mapping Pb exposure risks (e.g., 47 Filippelli et al., 2018; Ringwald et al., 2021; Watson et al., 2022), there is untapped potential for 48 further research advancement via community science.

49 Although community science samples contain inherent variability with sampling 50 techniques, even with clear instructions, the large sample size of community science endeavors 51 often overcomes this limitation (Filippelli et al., 2018). However, a key question not addressed 52 thus far in community science-based research is whether smaller subsets of samples can be 53 utilized for more detailed geochemical techniques and analyses to understand pollution

54 distributions, despite potential variability in sampling methodology?

55 Lead stable isotopes have frequently been used for tracing pollution sources of Pb in the 56 environment as a more detailed geochemical technique (e.g., Adgate et al., 1998; Jaeger et al., 57 1998; Sutherland et al., 2003; Wang et al., 2019), largely due to unique Pb isotopic ratios of 58 many Pb sources and minimal environmental and biological fractionation of Pb isotopes (e.g., 59 Ayuso & Foley, 2020). Despite their applicability in pollution source apportionment studies, 60 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 61 diverse range of Pb isotopic ratios in major urban sources such as paint and gasoline can lead to 62 overlap in source ratios (e.g., Resongles et al., 2021; Wang et al., 2021). This can complicate 63 64 interpretations of Pb pollution sources in environmental media such as soils and dusts, even 65 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 66 67 ratios when determining pollution sources in the urban environment, but to incorporate other 68 techniques such as bulk chemistry and scanning electron microscopy (SEM) as well. SEM in 69 particular can identify the nature of pollutant particulates (i.e., metal fragments, combustion 70 spherules, etc.) (e.g., Dietrich et al., 2019; Gaberšek & Gosar, 2021), which can help identify the 71 actual form of Pb pollution and thus help differentiate between potential sources when used with 72 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

80 household-scale soil and indoor dust samples to better quantify how Pb sources (i.e., paint and

81 gasoline) are dispersed, and to illustrate how our methodology can be utilized in subsets of large-

82 scale community science projects to address complex patterns uninterpretable based on bulk

- 83 chemistry alone.
- 84

85 2. Materials and Methods

86 2.1 Sampling and Preparation

87 Soil and dust samples were provided by local community members from four households 88 in Indianapolis (Fig. S1) as part of the Anthropocene Network (https://anthropocenes.org/lead), 89 which provides free household screening for Pb (water, dust, and soil sampling) and aims to 90 address issues of environmental equity in the age of the Anthropocene. The community science 91 relationship was enabled through partnerships with local faith leaders in churches and the 92 assurance of participant anonymity, which allowed for sample collection at homes that would 93 otherwise be inaccessible. Residents were instructed to collect bulk vacuum cleaner dust in a 94 sealed plastic bag, as well as soil (upper 0-5 cm) at their home's dripline (directly adjacent to 95 home), the middle of their yard, and directly adjacent to the street with any type of scoop 96 available at their home. Soil samples (~25 g. or more) were placed in separate plastic bags. Each 97 vacuum dust sample was a composite of the entire home, and each soil sample was an individual, 98 non-composite sample. We only selected sampling kits that had clearly labeled sample bags, 99 indicative that the participants followed sampling instructions. Households that were spread out 100 across Indianapolis was also taken into consideration to capture as much potential representation 101 of the city as possible. A small sample size was used because the purpose of this research was to 102 serve as a case study illustrating the ability to utilize community science samples for more 103 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.

110 Dust samples were sieved at 250 µm and were dry due to the nature of sampling, and thus 111 needed no desiccation. Soil samples were air dried, sieved at 2 mm, then powdered and 112 homogenized utilizing an agate mortar and pestle prior to acid digestion. Samples were weighed 113 (~300-400 mg) into Savillex screw-cap vessels and digested utilizing a multi-stage, three-acid 114 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 115 116 of this solution was extracted and diluted for concentration analysis prior to chromatographic 117 separation. This is defined as total soil Pb for the purposes of this study.

- 118 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
- 120 Thibodeau et al. (2007). After purification, samples were diluted accordingly after a
- 121 concentration check and spiked with NIST 997 thallium (Tl) standard to achieve a Pb/Tl ratio of
 ~4.
- 122
- 123

124 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 ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb, and ²⁰⁸Pb were detected with a Nu Plasma II Multicollector

129 Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS), with the Pb isotope ratios

130 208/206, 207/206, 208/207, 206/204, 207/204, and 208/204 reported after being normalized to

131 Galer & Abouchami (1998). All ICP analyses were completed at Indiana University-

132 Bloomington in the Department of Earth and Atmospheric Sciences' Metal Isotopes Laboratory.

133 More details are provided in Text S1.

134 Dust and soil samples were prepared for SEM and energy dispersive X-ray spectroscopy

135 (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, $M_{\beta} = 2.444$

137 keV). All SEM-EDS analyses were conducted at Indiana University-Purdue University

138 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
- 140 pressure mode.
- 141

142 2.3 Quality Control

143 Lead total concentration was deemed acceptable based on a mean recovery of $89.4\% \pm$ 144 11.0% (n = 5) for NIST SRM 2702 (Table S1) and runs of USGS standard AGV-2a and NIST 145 2702 were both in agreement with previously published Pb isotopic compositions (see Text S2 146 and Table S2). More details on quality control are provided in Text S2.

147

148 **3. Results and Discussion**

149 3.1 General Soil and Dust Pb Enrichment and Relationships

150 Regardless of sample type or location, nearly all samples are enriched relative to the 75th

151 percentile of background soil samples [upper 5 cm of soil, air dried <2 mm fraction crushed to

 $152 < 150 \ \mu m$ prior to a near-total four-acid (hydrochloric, nitric, hydrofluoric, and perchloric)

- 153 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

155 mg/kg for Pb. Lead concentrations are \geq 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).

159 Although the dust samples were sieved to a finer particle fraction and finer sized particles 160 tend to contain higher concentrations of heavy metals (e.g., Herngren et al., 2006; Tansel & 161 Rafiuddin, 2016), bulk geochemistry results indicate that dust is not consistently higher in metal 162 concentration than the soil samples, even within the same house location (Fig. 1B; Fig. S2). This 163 may indicate that the dominant anthropogenic source for these elements in both media is coarser 164 material, and the interior loading reflects both indoor dust and finer particle invasion from 165 outside. This emphasizes recent work by Gillings et al. (2022) that demonstrates how 166 relationships between indoor and outdoor sources are not always predictable, and inferences on 167 one based on the other should be taken with caution.

168 It is noted that we examined the bulk grain size (<2 mm) composition of outdoor soil 169 samples, and that grain size may affect metal concentrations and partitioning of Pb sources. 170 However, we emphasize that bulk grain size is the most representative way to encapsulate all 171 possible Pb exposure within a household environment. While finer sized particles may be a 172 greater inhalation and transport risk, larger sized particles may degrade over time and still supply

- a reservoir of Pb pollution.
- 174

175 3.2 Bulk Pb Concentration and Pb Isotope Heterogeneity

176 Three homes displayed clear heterogeneity in bulk metal concentrations of Pb depending 177 on sample location at the home, with only House 1 showing more consistent metal trends for Pb 178 (Fig. 1B; other metals displayed in Fig. S3). This heterogeneity extends to Pb isotopic ratios (Table S4) such as ²⁰⁶Pb/²⁰⁴Pb, although House 2 displayed fairly consistent ²⁰⁶Pb/²⁰⁴Pb isotopic 179 ratios (Fig. 1A), with slightly more variance in ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb ratios (Fig. S4). This 180 181 matches previous research in urban centers that documented extensive heterogeneity at the 182 household-scale for both bulk metal concentrations such as Pb (e.g., Filippelli et al., 2018; 183 Obeng-Gyasi et al., 2021; Wade et al., 2021) and for Pb isotopic composition (Wang et al., 184 2021). Due to our small sample size and no consistent trends between Pb isotopes or bulk metal 185 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 186 187 Indianapolis, IN (Filippelli et al., 2018), Durham, NC (Wade et al., 2021) and Greensboro, NC 188 (Obeng-Gyasi et al., 2021) have done. However, when using bulk Pb concentrations, Pb isotope 189 ratios, and SEM collectively at a house-by-house basis, clearer interpretations of Pb pollution 190 can be made.

191

192 3.3 Pb Pollution Sources and Pb Distribution by Household—Synthesis of Analytical Methods

193 *House 1:*

The ²⁰⁶Pb/²⁰⁴Pb isotopic ratio changes significantly depending on the sample location and 194 195 between the indoor dust and outdoor soil, increasing towards the street, with the dust sample containing the lowest ²⁰⁶Pb/²⁰⁴Pb isotopic ratio (Fig. 1A). This is not clearly reflected in bulk Pb 196 197 concentration though, which remains fairly consistent throughout the property, between 70 and 198 130 mg/kg (Fig. 1B; Table S3). Based on Pb isotopes alone, one would interpret a greater 199 proportion of geogenic background glacial till (indicative of surficial Wisconsin glaciation 200 sediments in the area) contribution closer to the street (Fig. S5). However, this is unlikely, 201 because Pb concentrations did not decrease toward the street (Fig. 1B), which would be expected 202 based on much lower Pb concentrations in geogenic background till (Barnes et al., 2020; 203 Kousehlar & Widom, 2020). Thus, the Pb changes in isotopic composition must have come from 204 an additional source of Pb such as leaded paint or leaded gasoline. Microscopy revealed only a 205 few instances of possible Pb paint particles, but routinely revealed evidence of high temperature 206 anthropogenic process Fe-rich spherules at all soil locations (Fig. 2). Thus, while Pb paint likely 207 contributed to bulk Pb inside the home (Fig. 2), the scarcity of distinguishable paint particles via 208 SEM in the soils and prevalence of anthropogenic spherules suggests the possibility of 209 vehicular/industrial Pb sourcing, such as from residual microscopic particulate leaded gasoline or 210 other vehicle Pb-wear (i.e., wheel weights) too small to detect via SEM.

211 House 2:

There is greater bulk Pb variability in samples than ²⁰⁶Pb/²⁰⁴Pb ratios on the property 212 (Fig. 1). This includes a dripline soil sample that was 4 mg/kg Pb, and resembled mulching 213 214 material. Thus, although the house samples are grouped closely on bivariate Pb isotope plots 215 (Fig. S5), Pb sourcing is potentially different between the 4 mg/kg Pb dripline sample and other, 216 more elevated Pb streetside & yard samples. Yard and streetside Pb was much higher than inside 217 the home (Fig. 1B), with SEM imaging revealing no Pb-rich particles inside the home, but 218 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 219 220 locations supports the higher bulk Pb concentrations and suggests that Pb paint abundance 221 outside the home is greater than inside the home, even though Pb isotopic ratios are similar. This 222 may be because of peeling exterior Pb paint that has been reworked in the outdoor environment 223 and covered at the dripline by recent mulch material. There still appears to be an exchange of 224 material across all household samples, as Fe spherules were also found at this household at all 225 locations, even the mulched dripline. However, the spherules at the dripline were less Fe-rich 226 (Fig. S11) than other spherules, suggesting they may have degraded more over time with 227 minerals (i.e., clays, apatite) adhering or precipitating on the spherule structure, or because they 228 were formed under different conditions or processes.

229 *House 3*:

Bulk Pb concentrations increase toward the street in soils and are the lowest in dust (Fig. 1B; Table S3). ²⁰⁶Pb/²⁰⁴Pb 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.

245 House 4:

Bulk Pb concentration is highly variable depending on sample location, at the highest (> 246 247 800 mg/kg) in the dust and dripline soil before decreasing significantly in the yard soil, then increasing in the streetside soil (Fig. 1B). The ²⁰⁶Pb/²⁰⁴Pb isotopic ratio also changes, increasing 248 249 from the dust to the dripline soil, then decreasing again for yard and streetside soil (Fig. 1A). 250 Based on microscopy, the dust and dripline soils commonly contain Pb-rich particles resembling 251 paint chips (Fig. 2), and the differences in Pb isotope ratios may be because of different layers or 252 types of paint in the interior versus exterior of the home. Microscopy did not reveal any obvious 253 Pb paint chips in the streetside or yard soils, but did reveal numerous high temperature 254 anthropogenic process Fe-rich spherules, which were evident in both the streetside and dripline 255 soil (Fig. 2). Although there may be mixing of microscopic Pb-rich particles from past vehicular 256 exhaust or other industrial sources at the dripline, that soil is likely more dominated by Pb paint 257 relative to streetside soils, where no Pb paint particles could be easily identified. Thus, because 258 the bulk Pb concentration was still elevated well above background soils (Smith et al., 2013) at 259 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. 260

261 3.3.1 Summary and Comparison of Household Pb Sourcing

262 The primary Pb sources in all homes were likely Pb-based paint or the remnants of leaded 263 gasoline, supportive of previous literature in urban environments within the U.S. (e.g., Dietrich 264 et al., 2022; Wang et al., 2021). However, each home had different patterns of Pb pollution 265 sources (Table 1) and Pb concentrations (Fig. 1B). This did not seem to be dependent on largerscale Pb relationships in Indianapolis, such as those at the zip code level (Filippelli et al., 2018). 266 267 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 268 269 these homes were widely variable (4-2100 mg/kg; Table S3), and the likely Pb sources changed 270 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 271 272 this study exemplifies the fact that generalizations of Pb pollution at larger spatial scales should 273 be made with caution, and multiple other external factors can affect Pb distributions at fine scales 274 such as the history of the home and proximity to major roadways.

276 3.4 Similarities in Pb Pollution Between Households

277 Influences on metal concentrations in dusts and soils at the household-scale include not 278 only potential pollution sources, but how dust and soil get reworked and distributed throughout 279 the environment. In general, although there was much Pb pollution heterogeneity between 280 households (Fig. 1), a commonality was the presence of Fe-rich high temperature anthropogenic 281 process spherules at every home within multiple samples (Fig. 2). This entailed homes where 282 distance from road to home was between 7-14 meters, nearby daily traffic volume was highly 283 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) 284 285 and are likely anthropogenic. This not only suggests that there is exchange of pollutant particles 286 to the dripline of the home, supporting the hypothesis that the side of the home can act as a 287 barrier for particulates to fall following roadside resuspension (e.g., Filippelli et al., 2018; 288 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, 289 290 transport of pollutants in soils/dust at the household-scale is likely an additional factor affecting 291 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).

297

298 3.5 Limitations of Only Pb Isotopes for Pollution Sourcing

299 Using Pb isotopes or bulk chemistry alone can be problematic for determining pollutant 300 sourcing, because even within the same home, Pb isotopic composition of paints can vary 301 drastically (Jaeger et al., 1998), and the Pb isotopic ratios for leaded gasoline were widely 302 variable within the U.S. because of changing ore sources over time (e.g., Dietrich et al., 2021). 303 For example, nearly all dust and soil samples plot within 1^o variability of leaded gasoline and 304 lead paint source endmembers depending on the Pb isotope ratios used (Fig. S5). Thus, although 305 previous interpretations in urban settings within the U.S. have concluded Pb paint sources likely 306 dominating soils close to older homes (i.e., built prior to 1978) and leaded gasoline significantly 307 contributing to soils near roadways (e.g., Wade et al., 2021; Wang et al., 2021), we utilized SEM 308 imaging and EDS spectra to aid in interpretation of our variation in bulk Pb and Pb isotopic 309 ratios at the household-scale. This multi-analytical approach is often necessary in complex urban 310 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 311 312 (e.g., Resongles et al., 2021).

314 4. Implications

315 Urban environments contain complex, heterogenous distributions of heavy metals in soils 316 and dusts, particularly Pb. This is especially evident in a large, post-industrial city such as 317 Indianapolis, IN where historic Pb sources such as leaded paint and leaded gasoline contaminate 318 soils and dusts in varying ways, as shown with differences between our study's household 319 samples. Community science can provide a large sampling set to gather Pb data from, which can 320 effectively help map Pb hotspots, general trends, and inform people of risks in their home (e.g., 321 Filippelli et al., 2018; Watson et al., 2022). However, there are often nuances associated with 322 pollution interpretations based on bulk chemistry alone, and more detailed analytical methods 323 can help better understand Pb heterogeneity.

324 Here, we demonstrated how detailed methodology such as SEM and Pb isotopes can give 325 a clearer picture of Pb pollution source distribution on a property. This approach can be utilized 326 for even larger subsets of community science samples to better understand pollution variability 327 in bulk chemistry following initial screening through techniques such as X-ray fluorescence 328 (XRF). While there is inherent uncertainty in sampling with community scientists, as long as 329 general instructions are followed, enough information may be gathered to obtained informative 330 and actionable insight into pollution sources distributions. For example, whether Pb pollution 331 from the home reaches the street, or whether Pb pollution from the street reaches the home. This 332 information can therefore aid in home pollution remediation and prevention by determining 333 whether there is significant transport and exchange of Pb pollution sources.

334 Additionally, working more with community members as real partners in research can be 335 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 336 337 approach that "parachute science" and has more lasting effects in changing behavior (Hayhow et 338 al., 2021). For example, we recommend surveying community members first about their 339 concerns and gathering important metadata during the course of the study, such as the age of 340 their home, whether there has been recent renovation, worries of a particular Pb source nearby, 341 etc.

342 Future studies in urban centers throughout the world would do well to capitalize on 343 community science and the multi-analytical techniques such as those used here and in other 344 recent urban metal pollution studies (e.g., Gaberšek & Gosar, 2021; Kelepertzis et al., 2021) to 345 more fully understand the nuance associated with urban metal pollution distributions. This can be 346 important in determining whether a pollutant source is coming from a property or elsewhere, the 347 nature of the pollutant particles (i.e., are there Pb particles that are small enough to be an 348 inhalation risk), and whether there is transfer of material from inside to outside a home. All these 349 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

- 354 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
- 356 pollution. Lastly, these community science efforts expand the possible options for sample
- accessibility to researchers, such as to household indoor dust samples.
- 358

359 Acknowledgements

360 The authors are deeply thankful to the households that supplied samples for this work and 361 the lab techs who helped with initial sampling processing. The Anthropocene Network is supported by the Environmental Resilience Institute, funded by Indiana University's Prepared for 362 Environmental Change Grand Challenge Initiative. Partial support for this work was provided by 363 364 NSF-EAR-PF Award #2052589 to M.D. Special thanks to Miguel Cruz for help with SEM 365 instrumentation, Jason Kelly with help providing the samples, the two anonymous reviewers for 366 their insightful comments, and Leah Wood and John Shukle for helpful comments on an earlier 367 version of the manuscript.

368

369 Manuscript Tables:

370

372

373

374

375

376

377

378

379

380

1		Dripline	Yard	Streetside
	Paint	Vehicular or	Vehicular or	Vehicular or
		industrial	industrial	industrial
2 F	Potentially Paint	None	Paint	Paint
3	Vehicular or	Vehicular or	Vehicular or	Vehicular or
	industrial	industrial	industrial	industrial
4	Paint	Paint	Potentially	Vehicular or
			Paint/Vehicular	industrial

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

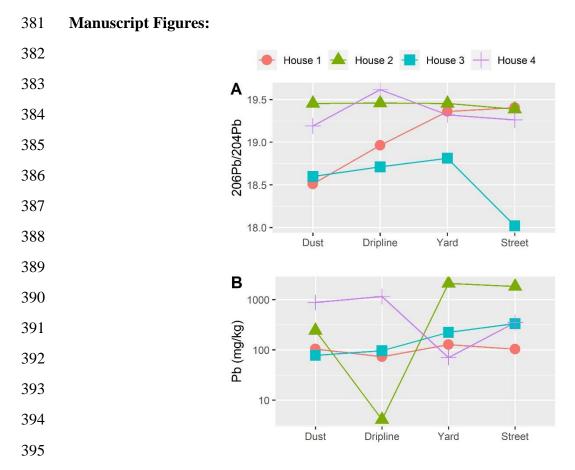
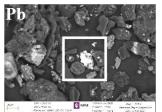


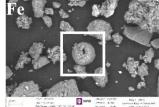
Fig. 1: Trends in ²⁰⁶Pb/²⁰⁴Pb based on sample type and house (A), as well as trends in bulk Pb
concentration (mg/kg) based on sample type and house (B). Similar trends were seen for other
isotopic ratios, albeit with slight variability (Fig. S4).

39.0-House ²⁰⁸Pb/²⁰⁴Pb 38.5-Sample Location Dust Dripline ard Street 38.0 **-**18.0 18.5 19.0 19.5

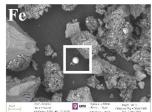
²⁰⁶Pb/²⁰⁴Pb



House 1: Indoor Dust¹



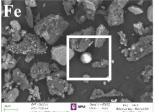
House 1: Dripline²



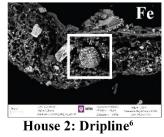
House 1: Streetside³

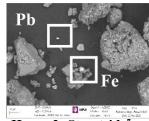


House 1: Yard⁴



House 2: Indoor Dust⁵





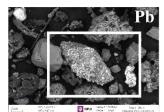
House 2: Streetside⁷



House 2: Yard⁸



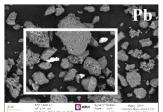
House 3: Indoor Dust⁹



House 4: Indoor Dust¹³



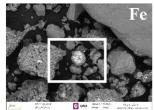
House 3: Dripline¹⁰



House 4: Dripline¹⁴



House 3: Streetside¹¹

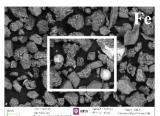


House 4: Dripline¹⁵





House 3: Yard¹²



House 4: Streetside¹⁶

- 400 **Fig. 2:** Bivariate plot of Pb isotopic ratios, grouped by house and sample location. 2σ analytical
- 401 variability is minimal relative to the sample symbol size and are thus only displayed in Table S2.
- 402 SEM images are listed below the plot, with anthropogenic particles outlined in white. The
- 403 corresponding EDS spectra for each superscript notation are listed in the Supplementary
- 404 Information (Figs. S6-S21). Images labeled "Pb" contain Pb-rich particles within the white
- 405 outline, while images labeled "Fe" contain Fe-rich particles within the white outline. It is noted
- 406 that the paint particles in House 2: Streetside⁷, House 4: Indoor Dust¹³ and House 4: Dripline¹⁴
- 407 also contain traces of Fe.
- 408

409 *Statements and Declarations*

410 **Funding**

- 411 The Anthropocene Network is supported by the Environmental Resilience Institute, funded by
- 412 Indiana University's Prepared for Environmental Change Grand Challenge Initiative. Partial
- 413 support for this work was provided by NSF-EAR-PF Award #2052589 to M.D.

414 **Competing Interests**

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

416 Author Contributions

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

420 Ethical Approval

- 421 "Not applicable"
- 422
- 423 **Consent to Participate**
- 424 "Not applicable"
- 425

426 **Consent to Publish**

- 427
- 428 All authors agree to publish the manuscript in its present form.
- 429

430 Availability of data and materials

- 431 All data and materials are included in the manuscript and supplementary documentation.
- 432
- 433

434 **References**

- 435 Adgate, J., Rhoads, G., & Lioy, P. (1998). The use of isotope ratios to apportion sources of lead
- 436 in Jersey City, NJ, house dust wipe samples. *The Science of The Total Environment*,
- 437 221(2–3), 171–180. https://doi.org/10.1016/S0048-9697(98)00282-4
- 438 Ayuso, R. A., & Foley, N. K. (2020). Surface topography, mineralogy, and Pb isotope survey of
- 439 wheel weights and solder: Source of metal contaminants of roadways and water systems.
- 440 *Journal of Geochemical Exploration*, 212, 106493.
- 441 https://doi.org/10.1016/j.gexplo.2020.106493
- 442 Barnes, M., McLeod, C. L., Chappell, C., Faraci, O., Gibson, B., & Krekeler, M. P. S. (2020).
- 443 Characterizing the geogenic background of the Midwest: A detailed mineralogical and
- geochemical investigation of a glacial till in southwestern Ohio. *Environmental Earth Sciences*, 79(6), 159. https://doi.org/10.1007/s12665-020-8890-z
- 446 Betts, K. S. (2012). CDC Updates Guidelines for Children's Lead Exposure. *Environmental*

447 *Health Perspectives*, *120*(7). https://doi.org/10.1289/ehp.120-a268

- 448 Deniel, C., & Pin, C. (2001). Single-stage method for the simultaneous isolation of lead and
- 449 strontium from silicate samples for isotopic measurements. *Analytica Chimica Acta*,

450 426(1), 95–103. https://doi.org/10.1016/S0003-2670(00)01185-5

- 451 Dietrich, M., Krekeler, M. P. S., Kousehlar, M., & Widom, E. (2021). Quantification of Pb
- 452 pollution sources in complex urban environments through a multi-source isotope mixing
- 453 model based on Pb isotopes in lichens and road sediment. *Environmental Pollution*, 288,
- 454 117815. https://doi.org/10.1016/j.envpol.2021.117815
- 455 Dietrich, M., Shukle, J. T., Krekeler, M. P. S., Wood, L. R., & Filippelli, G. M. (2022). Using
- 456 Community Science to Better Understand Lead Exposure Risks. *GeoHealth*, 6(2),
- 457 e2021GH000525. https://doi.org/10.1029/2021GH000525

- 458 Dietrich, M., Wolfe, A., Burke, M., & Krekeler, M. P. S. (2019). The first pollution investigation
- 459 of road sediment in Gary, Indiana: Anthropogenic metals and possible health implications
- 460 for a socioeconomically disadvantaged area. *Environment International*, *128*, 175–192.
- 461 https://doi.org/10.1016/j.envint.2019.04.042
- 462 Egendorf, S. P., Gailey, A. D., Schachter, A. E., & Mielke, H. W. (2020). Soil toxicants that
- 463 potentially affect children's health. *Current Problems in Pediatric and Adolescent Health*464 *Care*, 50(1), 100741. https://doi.org/10.1016/j.cppeds.2019.100741
- 465 Filippelli, G. M., Adamic, J., Nichols, D., Shukle, J., & Frix, E. (2018). Mapping the Urban Lead
- 466 Exposome: A Detailed Analysis of Soil Metal Concentrations at the Household Scale
- 467 Using Citizen Science. *International Journal of Environmental Research and Public*468 *Health*, 15(7), 1531. https://doi.org/10.3390/ijerph15071531
- Gaberšek, M., & Gosar, M. (2021). Towards a holistic approach to the geochemistry of solid
- 470 inorganic particles in the urban environment. *Science of The Total Environment*, 763,
- 471 144214. https://doi.org/10.1016/j.scitotenv.2020.144214
- 472 Galer, S. J. G., & Abouchami, W. (1998). Practical Application of Lead Triple Spiking for
- 473 Correction of Instrumental Mass Discrimination. *Mineralogical Magazine*, 62A(1), 491–
- 474 492. https://doi.org/10.1180/minmag.1998.62A.1.260
- 475 Gillings, M. M., Fry, K. L., Morrison, A. L., & Taylor, M. P. (2022). Spatial distribution and
- 476 composition of mine dispersed trace metals in residential soil and house dust:
- 477 Implications for exposure assessment and human health. *Environmental Pollution*, 293,
- 478 118462. https://doi.org/10.1016/j.envpol.2021.118462

479	Hayhow, C., Brabander, D., Jim, R., Lively, M., and Filippelli, G.M. (2021). Addressing the
480	need for just GeoHealth engagement: Evolving models for actionable research that
481	transform communities. GeoHealth. https://doi.org/10.1029/2021GH000496
482	Herngren, L., Goonetilleke, A., & Ayoko, G. A. (2006). Analysis of heavy metals in road-
483	deposited sediments. Analytica Chimica Acta, 571(2), 270-278.
484	https://doi.org/10.1016/j.aca.2006.04.064
485	Jaeger, R. J., Weiss, A. L., & Manton, W. I. (1998). Isotopic Ratio Analysis in Residential Lead-
486	Based Paint and Associated Surficial Dust. Journal of Toxicology: Clinical Toxicology,
487	36(7), 691–703. https://doi.org/10.3109/15563659809162617
488	Kelepertzis, E., Argyraki, A., Chrastný, V., Botsou, F., Skordas, K., Komárek, M., & Fouskas,
489	A. (2020). Metal(loid) and isotopic tracing of Pb in soils, road and house dusts from the
490	industrial area of Volos (central Greece). Science of The Total Environment, 725, 138300.
491	https://doi.org/10.1016/j.scitotenv.2020.138300
492	Kelepertzis, E., Chrastný, V., Botsou, F., Sigala, E., Kypritidou, Z., Komárek, M., Skordas, K.,
493	& Argyraki, A. (2021). Tracing the sources of bioaccessible metal(loid)s in urban
494	environments: A multidisciplinary approach. Science of The Total Environment, 771,
495	144827. https://doi.org/10.1016/j.scitotenv.2020.144827
496	Kousehlar, M., & Widom, E. (2020). Identifying the sources of air pollution in an urban-
497	industrial setting by lichen biomonitoring—A multi-tracer approach. Applied
498	Geochemistry, 121, 104695. https://doi.org/10.1016/j.apgeochem.2020.104695
499	Laidlaw, M. A. S., & Filippelli, G. M. (2008). Resuspension of urban soils as a persistent source
500	of lead poisoning in children: A review and new directions. Applied Geochemistry, 23(8),
501	2021-2039. https://doi.org/10.1016/j.apgeochem.2008.05.009

502	Miler, M., & Gosar, M. (2019). Assessment of contribution of metal pollution sources to attic
503	and household dust in Pb-polluted area. Indoor Air, 29(3), 487–498.

- 504 https://doi.org/10.1111/ina.12548
- 505 Obeng-Gyasi, E., Roostaei, J., & Gibson, J. M. (2021). Lead Distribution in Urban Soil in a
- 506 Medium-Sized City: Household-Scale Analysis. *Environmental Science & Technology*,
 507 55(6), 3696–3705. https://doi.org/10.1021/acs.est.0c07317
- 508 Rader, S. T., Gaschnig, R. M., Newby, S. M., Bebout, G. E., Mirakian, M. J., & Owens, J. D.
- 509 (2021). Thallium behavior during high-pressure metamorphism in the Western Alps,
- 510 Europe. Chemical Geology, 579, 120349. https://doi.org/10.1016/j.chemgeo.2021.120349
- 511 Resongles, E., Dietze, V., Green, D. C., Harrison, R. M., Ochoa-Gonzalez, R., Tremper, A. H., &

512 Weiss, D. J. (2021). Strong evidence for the continued contribution of lead deposited

- 513 during the 20th century to the atmospheric environment in London of today. *Proceedings*
- 514 of the National Academy of Sciences, 118(26). https://doi.org/10.1073/pnas.2102791118
- 515 Ringwald, P., Chapin, C., Iceman, C., Tighe, M. E., Sisk, M., Peaslee, G. F., Peller, J., & Wells,
- 516 E. M. (2021). Characterization and within-site variation of environmental metal
- 517 concentrations around a contaminated site using a community-engaged approach.
- 518 *Chemosphere*, 272, 129915. https://doi.org/10.1016/j.chemosphere.2021.129915
- 519 Smith, D.B., Cannon, W.F., Woodruff, L.G., Solano, Federico, Kilburn, J.E., and Fey, D.L.,
- 520 (2013). Geochemical and mineralogical data for soils of the conterminous United States:
- 521 U.S. Geological Survey Data Series 801, 19 p., https://pubs.usgs.gov/ds/801/.
- 522 Sutherland, R. A., Day, J. P., & Bussen, J. O. (2003). Lead Concentrations, Isotope Ratios, and
- 523 Source Apportionment in Road Deposited Sediments, Honolulu, Oahu, Hawaii. *Water*,
- 524 *Air, and Soil Pollution, 142, 165–186.*

- 525 Tansel, B., & Rafiuddin, S. (2016). Heavy metal content in relation to particle size and organic
- 526 content of surficial sediments in Miami River and transport potential. *International*
- 527 *Journal of Sediment Research*, *31*(4), 324–329.
- 528 https://doi.org/10.1016/j.ijsrc.2016.05.004
- 529 Thibodeau, A. M., Killick, D. J., Ruiz, J., Chesley, J. T., Deagan, K., Cruxent, J. M., & Lyman,
- 530 W. (2007). The strange case of the earliest silver extraction by European colonists in the
- 531 New World. *Proceedings of the National Academy of Sciences*, 104(9), 3663–3666.
- 532 https://doi.org/10.1073/pnas.0607297104
- 533 Wade, A. M., Richter, D. D., Craft, C. B., Bao, N. Y., Heine, P. R., Osteen, M. C., & Tan, K. G.
- 534 (2021). Urban-Soil Pedogenesis Drives Contrasting Legacies of Lead from Paint and
- 535 Gasoline in City Soil. *Environmental Science & Technology*, 55(12), 7981–7989.
- 536 https://doi.org/10.1021/acs.est.1c00546
- 537 Wang, Z., Dwyer, G. S., Coleman, D. S., & Vengosh, A. (2019). Lead Isotopes as a New Tracer
- 538 for Detecting Coal Fly Ash in the Environment. *Environmental Science & Technology*
- 539 *Letters*, 6(12), 714–719. https://doi.org/10.1021/acs.estlett.9b00512
- 540 Wang, Z., Wade, A. M., Richter, D. D., Stapleton, H. M., Kaste, J. M., & Vengosh, A. (2021).
- 541 Legacy of anthropogenic lead in urban soils: Co-occurrence with metal(loids) and fallout
- 542 radionuclides, isotopic fingerprinting, and in vitro bioaccessibility. *Science of The Total*
- 543 *Environment*, 151276. https://doi.org/10.1016/j.scitotenv.2021.151276
- 544 Watson, G. P., Martin, N. F., Grant, Z. B., Batka, S. C., & Margenot, A. J. (2022). Soil lead
- 545 distribution in Chicago, USA. *Geoderma Regional*, 28, e00480.
- 546 https://doi.org/10.1016/j.geodrs.2021.e00480

1	Supplementary Information
2 3	Using community science for detailed pollution research: A case-study approach in Indianapolis, IN, USA
4	Matthew Dietrich ^{1*} , Shelby Rader ² , Gabriel Filippelli ^{1,3}
5 6	¹ Department of Earth Sciences, Indiana University – Purdue University Indianapolis, Indianapolis, IN, USA
7	² Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, IN, USA
8	³ Environmental Resilience Institute, Indiana University, Bloomington, IN, USA
9	*Corresponding Author: mjdietri@iu.edu
10	
11	Supplementary Text
12	Test S1-Instrument Analyses
13 14 15 16 17 18 19 20 21	The Pb isotopes ²⁰⁴ Pb, ²⁰⁶ Pb, ²⁰⁷ Pb, and ²⁰⁸ Pb were detected along with ²⁰² Hg, ²⁰³ Tl, and ²⁰⁵ Tl with a Nu Plasma II Multicollector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS). All Pb isotope results were Hg-corrected, albeit the Hg correction was nominally zero, and corrected on-line for mass discrimination using the known Tl ratio of the NIST 997 spike. All results were then normalized offline to values reported by Galer and Abouchami (1998) for the NIST 981 standard (²⁰⁶ Pb/ ²⁰⁴ Pb = 16.9405, ²⁰⁷ Pb/ ²⁰⁴ Pb = 15.4963, and ²⁰⁸ Pb/ ²⁰⁴ Pb = Pb 36.7219). The errors are derived from the reproducibility of the NIST 981 Pb standard over the course of the run.
22	Text S2-Quality Control

23 To address any possible metal contamination through sieving the household soil and dust 24 samples, 5 subsamples of NIST SRM 2702 – Inorganics in Marine Sediment were processed through either dust sieves (NIST 2702 1 and NIST 2702 2) or a 2 mm soil sieve (remaining 25 SRM 2702 samples), as well as through the rest of the sample prep (i.e., digestions). The 26 27 elements Cd, Tl, Pb, and Sb were deemed acceptable based on mean % recoveries between 80-28 106% (Table S1), and Pb isotopic contamination was deemed minimal based on low standard 29 deviations for all isotope ratios within the SRM 2702 samples (Table S2), and good agreement with the peer review published 207 Pb/ 206 Pb avg value of 0.8377 (calculated from 206 Pb/ 207 Pb ratio 30 of 1.1937 for SRM 2702; Jeong et al., 2021-99.62% similarity of our mean ratio to the 31 32 published ratio).

As a quality control check on sample preparation and analysis, ICP-MS samples included periodic blanks run between samples, which confirmed background levels negligible. Isotopic

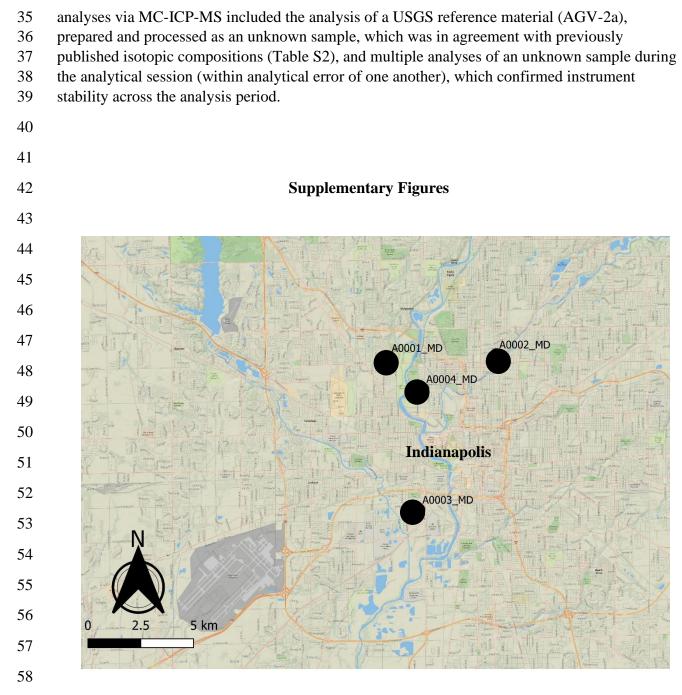
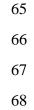


Fig. S1: Locations of the homes that provides samples, with symbol size enlarged and the mapzoomed out to protect privacy.



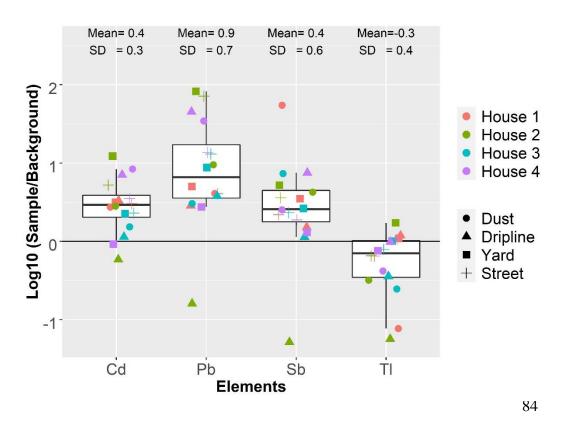
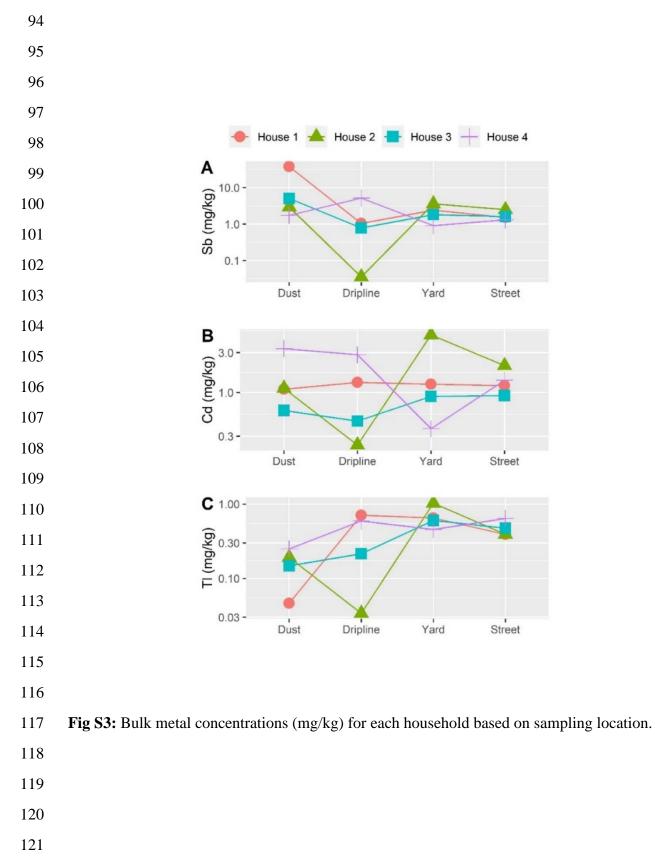


Fig. S2: Bulk metal concentrations of dust and soil samples normalized to the 75th percentile of
Indiana top 0-5cm background soil values from Smith et al. (2013). All values >0 represent
enrichment relative to the 75th percentile of background soil metal concentrations in Indiana,

- 88 U.S.



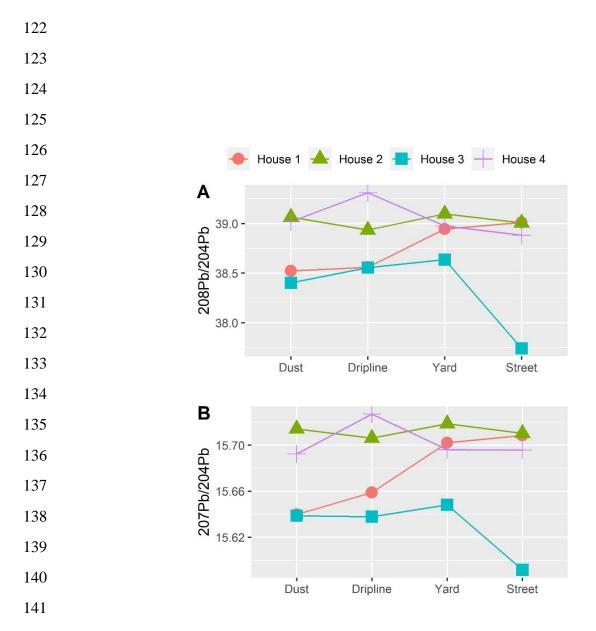


Fig. S4: Additional Pb isotope plots for each household based on sampling location.

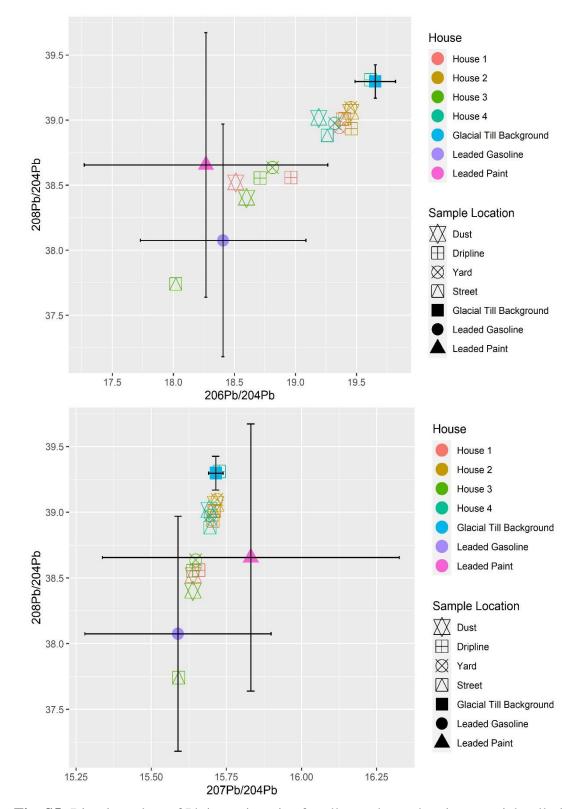


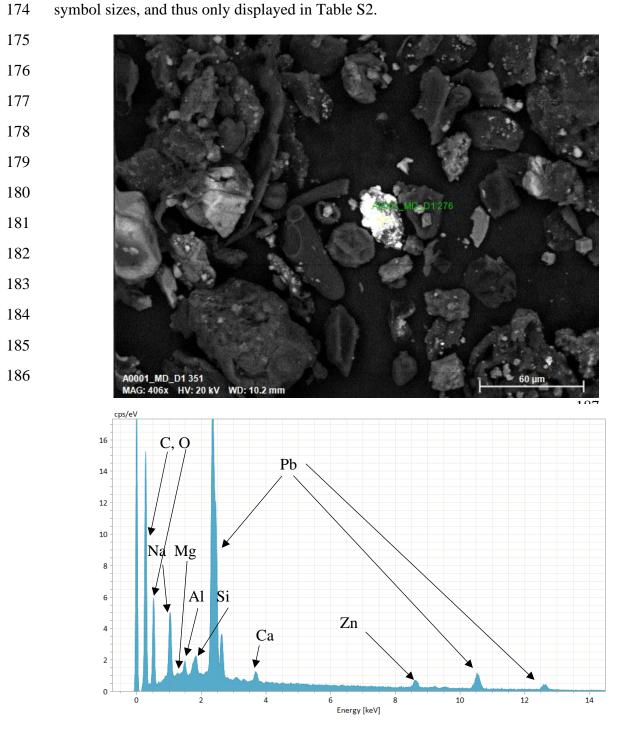
Fig. S5: Bivariate plots of Pb isotopic ratios for all samples and main potential pollution source

170 endmembers, with error bars representing 1σ variability in the source endmembers (Midwest

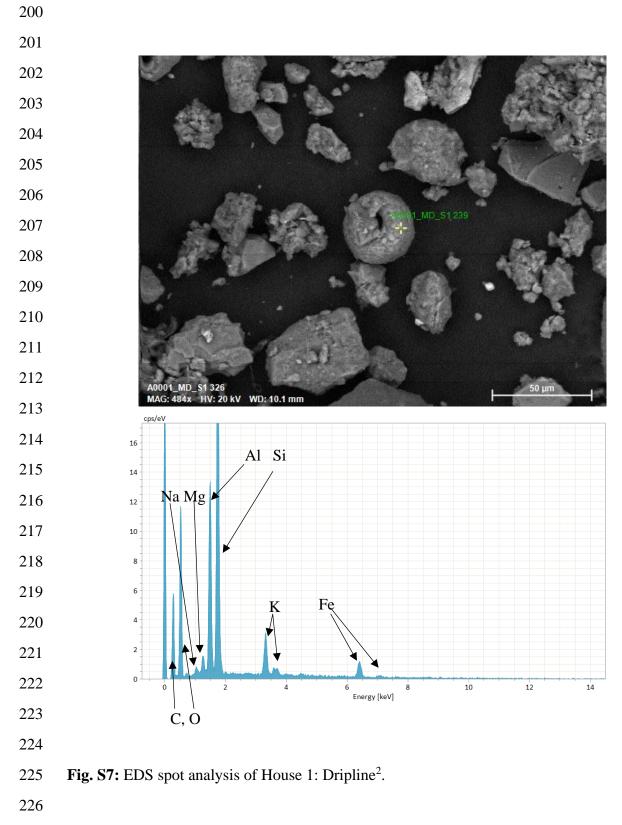
171 U.S. Glacial Till/Soil Background—Kousehlar and Widom, 2020, LeGalley et al., 2013; U.S.

172 Leaded Gasoline—Dietrich et al., 2021 and the references therein; Leaded Paint—Wang et al.,

2019 and the references therein). 2σ sample analytical variability is minimal relative to the
symbol sizes, and thus only displayed in Table S2.



199 **Fig. S6:** EDS spot analysis of House 1: Indoor Dust¹.



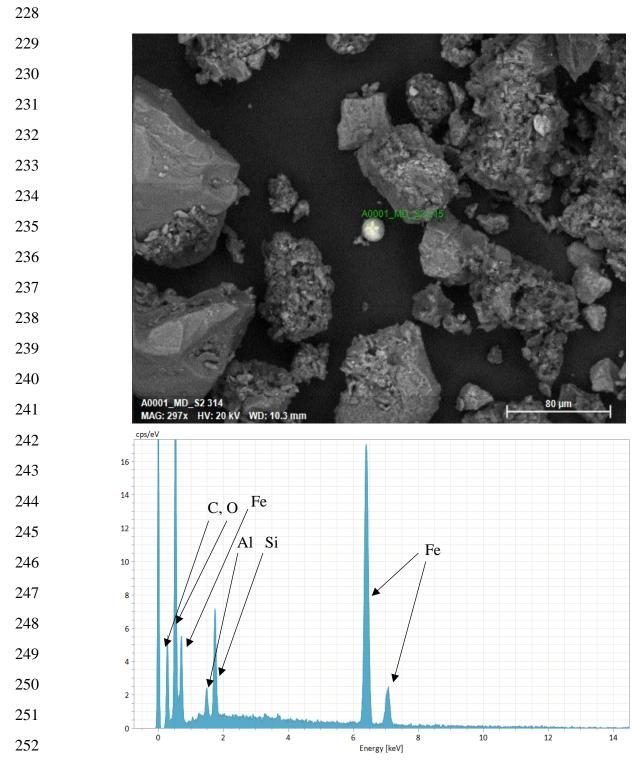


Fig. S8: EDS spot analysis of House 1: Streetside³.

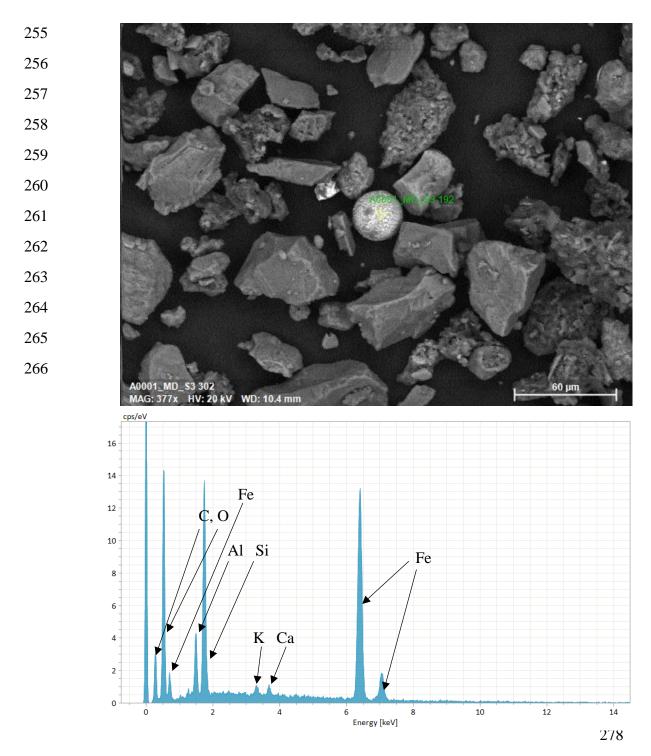


Fig. S9: EDS spot analysis of House 1: Yard⁴.

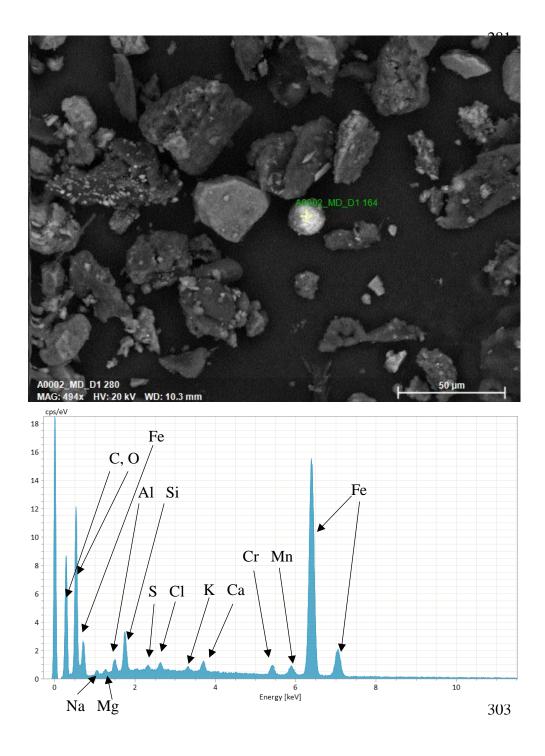
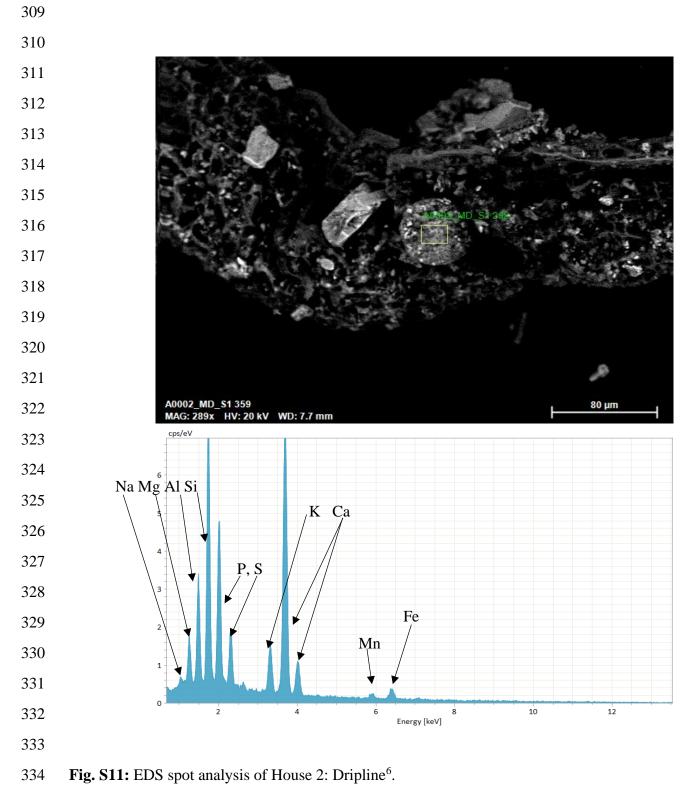
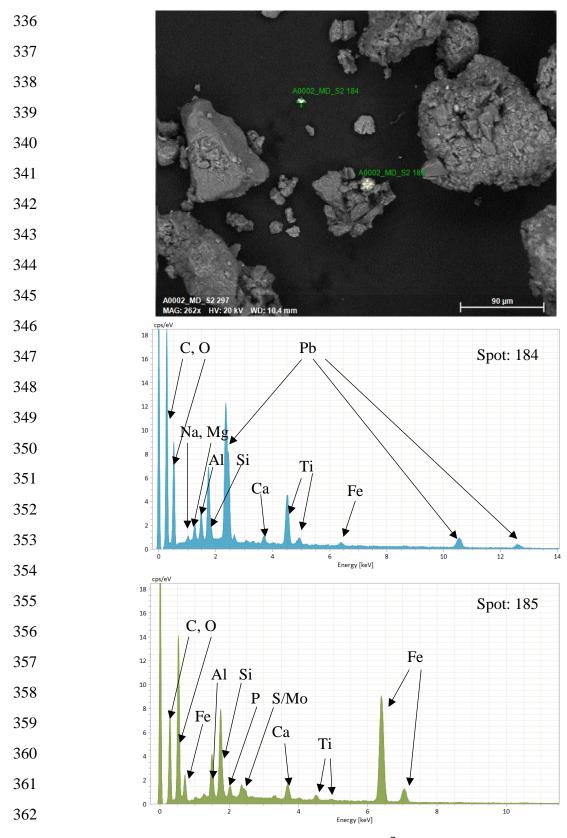
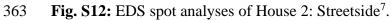
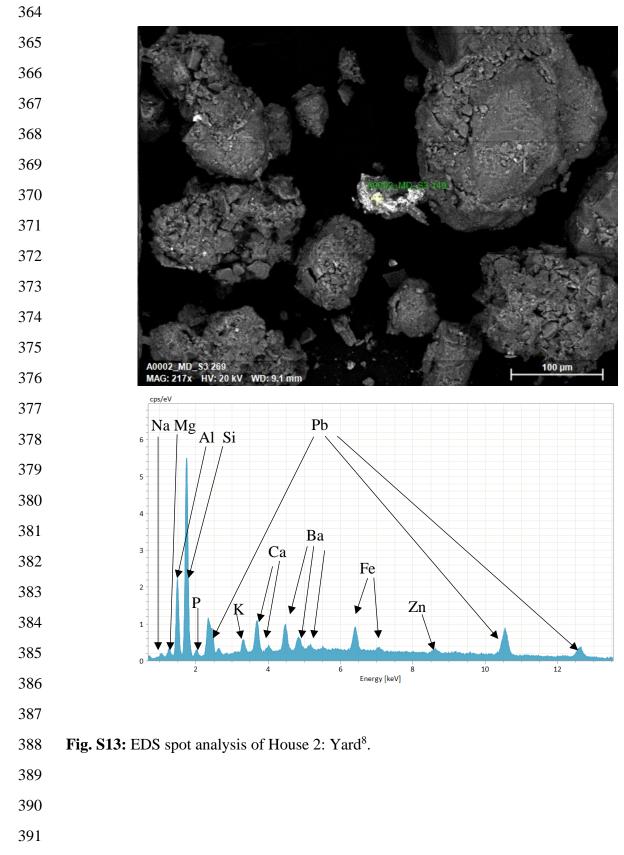


Fig. S10: EDS spot analysis of House 2: Indoor Dust⁵.









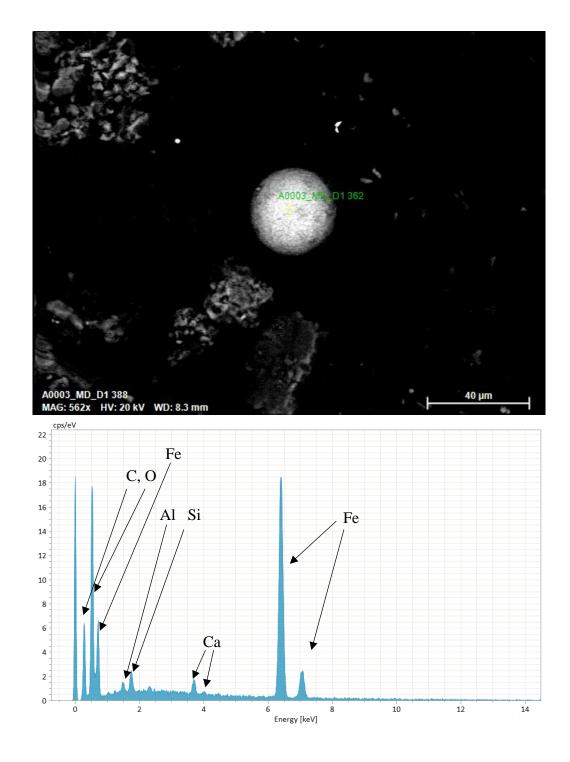
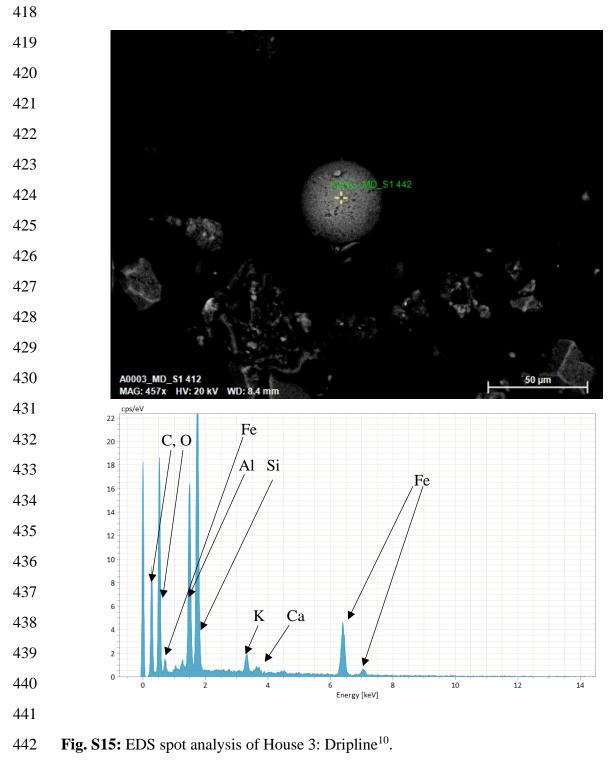


Fig. S14: EDS spot analysis of House 3: Indoor Dust⁹.



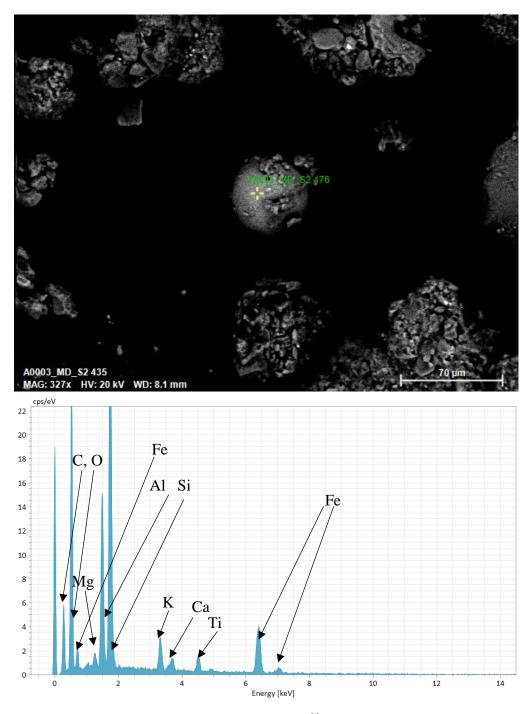
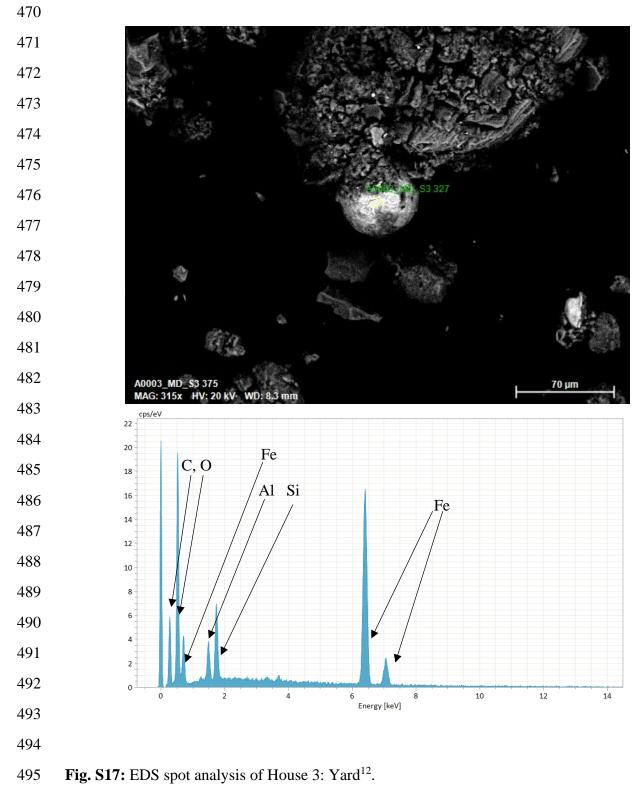


Fig. S16: EDS spot analysis of House 3: Streetside¹¹.



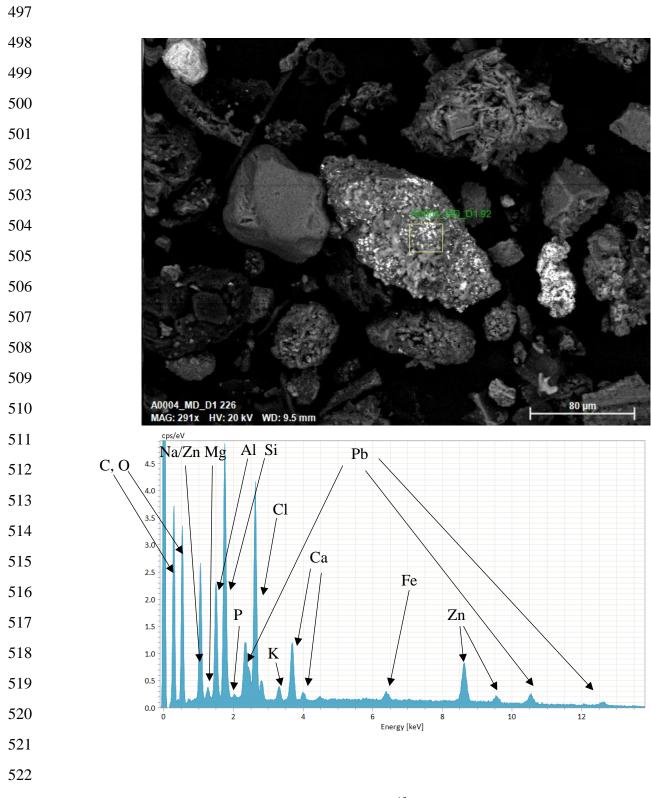


Fig. S18: EDS spot analysis of House 4: Indoor Dust¹³.

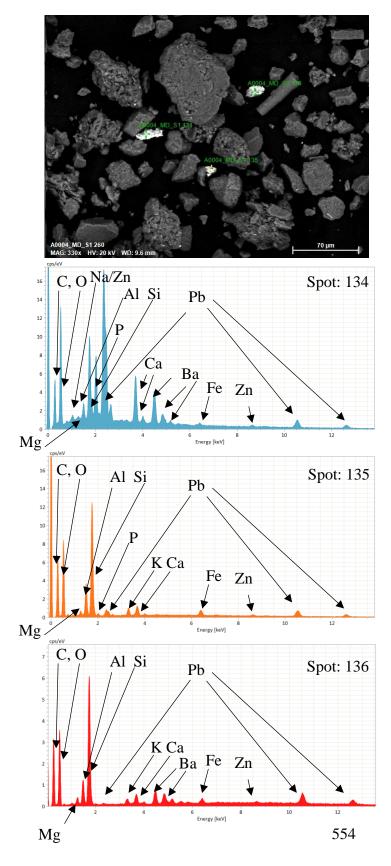
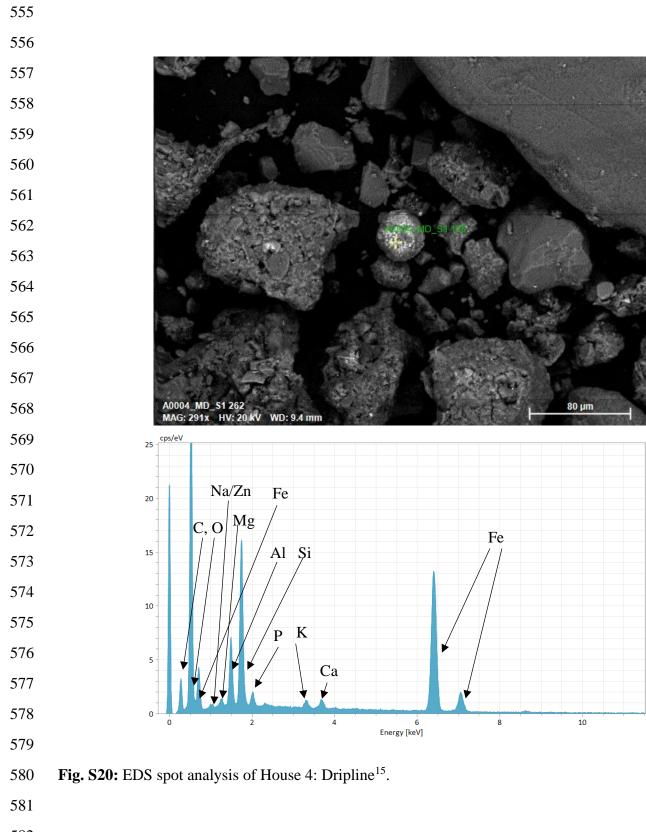
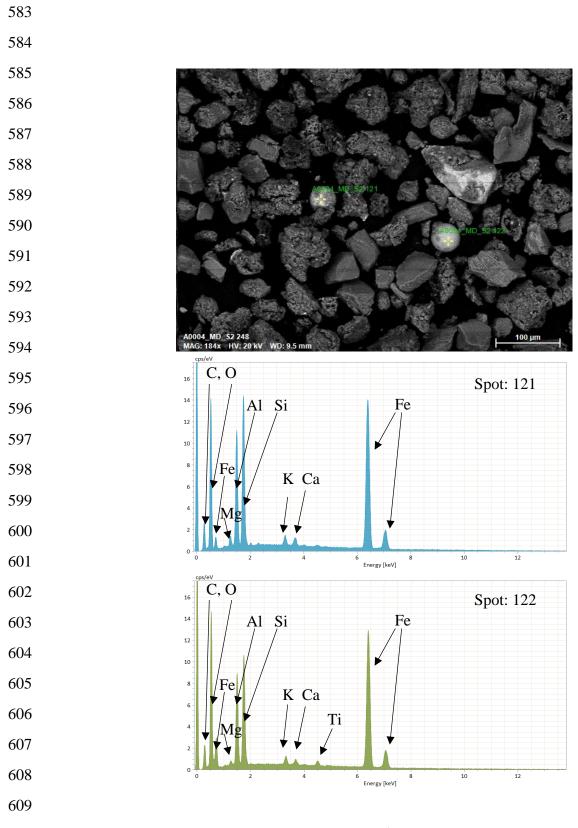
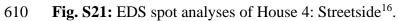


Fig. S19: EDS spot analyses of House 4: Dripline¹⁴. Spot 134 appears likely to be Pb-dominated paint, while Spots 135 and 136 contain much less of an initial Pb peak, indicative of possible X-ray shadowing due to an uneven spot surface. The general compositional similarity between the particles is indicative of Pb paint with slightly different elemental composition, possibly from different paint layers.







Supplementary Tables

	Cd	Sb	TI	Pb
Limits of Detection				
(LOD) (ppb)	0.02	0.03	0.004	0.02
NIST_2702_1	0.67	5.42	0.73	126.20
NIST_2702_2	0.71	5.08	0.76	130.76
NIST_2702_3	0.64	6.23	0.75	127.61
NIST_2702_4	0.66	6.70	0.80	113.82
NIST_2702_5	0.61	6.26	0.77	95.27
Certified Values				
(mg/kg)	0.817	5.6	0.8267	132.8
% Recoveries	81.51	96.74	87.82	95.03
	87.25	90.64	92.12	98.47
	78.05	111.34	91.06	96.09
	80.78	119.57	96.87	85.71
	74.16	111.73	92.86	71.74
Avg % Recovery	80.35	106.00	92.14	89.41
Std Dev % Recovery	4.82	11.91	3.27	11.00

612 **Table S1:** Bulk metal concentrations (mg/kg) and % recovery relative to certified values for five

613 subsamples of NIST SRM 2702 – Inorganics in Marine Sediment.

614

611

615 **Table S2:** Pb isotopic ratios for NIST SRM 2702 following sample prep that was the same as

616 soil/dust samples, and Pb isotopic ratios for AGV-2a following standard clean lab protocol, with

617 the given USGS values. 2σ overall analytical error for all analyses is also provided (based on the

618 reproducibility of NIST SRM 981 over the course of the analytical session).

Reference							
Standard		208/206	207/206	208/207	206/204	207/204	208/204
	981 2σ						
	external						
	error	0.0001	0.0000	0.0001	0.0014	0.0014	0.0044
	external						
	error (%)	0.005%	0.002%	0.003%	0.008%	0.009%	0.012%
NIST_2702_1		2.05574	0.83455	2.46329	18.77388	15.66794	38.59431
NIST_2702_2		2.05504	0.83421	2.46344	18.78221	15.66851	38.59835
NIST_2702_3		2.05595	0.83479	2.46282	18.76686	15.66631	38.58354
NIST_2702_4		2.05620	0.83479	2.46314	18.76782	15.66712	38.59037
NIST_2702_5		2.05561	0.83441	2.46355	18.77909	15.66944	38.60262
NIST_2702							
Std dev		0.00044	0.00025	0.00029	0.00675	0.00121	0.00734

AGV-2a	2.04293 0	.82777	2.46799	18.86955	15.61955	38.54962
AGV-2a						
USGS values				18.864	15.609	38.511
2σ external						
error				0.007	0.006	0.020
AGV-2a						
published	2.0413 - ().8271 -		18.851 -	15.609 -	38.511 -
range	2.049	0.8295		18.889	15.639	38.7127

- **Table S3:** Bulk metal concentrations for soil and dust samples in mg/kg, as well as assigned
- 622 house number and sample location.

Sample	Cd	Sb	TI	Pb	House	Sample Location
A0001_MD_D1	1.09	37.60	0.05	104.14	House 1	Dust
A0001_MD_S1	1.32	1.04	0.71	73.05	House 1	Dripline
A0001_MD_S2	1.21	1.50	0.39	103.77	House 1	Street
A0001_MD_S3	1.25	2.42	0.65	127.69	House 1	Yard
A0002_MD_D1	1.12	2.92	0.19	242.47	House 2	Dust
A0002_MD_S1	0.23	0.04	0.03	4.10	House 2	Dripline
A0002_MD_S2	2.09	2.48	0.39	1823.68	House 2	Street
A0002_MD_S3	4.89	3.59	1.03	2098.49	House 2	Yard
A0003_MD_D1	0.61	5.05	0.15	77.61	House 3	Dust
A0003_MD_S1	0.45	0.78	0.21	96.41	House 3	Dripline
A0003_MD_S2	0.92	1.59	0.47	332.93	House 3	Street
A0003_MD_S3	0.90	1.81	0.60	223.74	House 3	Yard
A0004_MD_D1	3.34	1.74	0.25	878.37	House 4	Dust
A0004_MD_S1	2.83	5.17	0.59	1152.48	House 4	Dripline
A0004_MD_S2	1.40	1.30	0.64	347.80	House 4	Street
A0004_MD_S3	0.37	0.90	0.45	70.33	House 4	Yard

Table S4: Pb isotope ratios for soil and dust samples.

Samples	208/206	207/206	208/207	206/204	207/204	208/204
A0001_MD_D1	2.081	0.845	2.463	18.510	15.640	38.521
A0001_MD_S1	2.033	0.826	2.462	18.963	15.659	38.558
A0001_MD_S2	2.010	0.810	2.483	19.404	15.708	39.011
A0001_MD_S3	2.012	0.811	2.480	19.360	15.702	38.944
A0002_MD_D1	2.008	0.808	2.486	19.453	15.714	39.063
A0002_MD_S1	2.001	0.807	2.479	19.458	15.706	38.934
A0002_MD_S2	2.012	0.810	2.483	19.387	15.710	39.006
A0002_MD_S3	2.010	0.808	2.487	19.453	15.719	39.096
A0003_MD_D1	2.065	0.841	2.456	18.599	15.639	38.402
A0003_MD_S1	2.061	0.836	2.465	18.710	15.638	38.554

A0003_MD_S2	2.095	0.865	2.421	18.019	15.592	37.741
A0003_MD_S3	2.054	0.832	2.469	18.812	15.648	38.636
A0004_MD_D1	2.033	0.818	2.486	19.190	15.692	39.017
A0004_MD_S1	2.004	0.802	2.500	19.615	15.727	39.310
A0004_MD_S2	2.019	0.815	2.477	19.261	15.696	38.882
A0004_MD_S3	2.017	0.812	2.483	19.320	15.696	38.976

626	Table S5: Relative distance	between the adjacent	main road and front	of the household property
-----	-----------------------------	----------------------	---------------------	---------------------------

- to the nearest meter, determined via the Google Maps "Measure distance" tool, oldest annual
- average daily traffic (AADT) data available for the closest major street to the household from the
- 629 Indiana Department of Transportation* (oldest available data was used to try and match
- 630 historical traffic flow when leaded gasoline was prominent, although it is noted there is a large
- 631 disconnect of over 20-30 years from leaded gasoline usage and reported traffic data), and
- 632 potential industrial sources of pollution with distance determined via the Google Earth
- 633 measurement tool.

Sample	House	Distance from road to home (meters)	AADT	Potential industrial sources of pollution (distance in km)
A0001_MD	1	14	12,772 ¹	Indianapolis Motor Speedway (2.4 km), lumber facility (1.3 km)
A0002_MD	2	8	2,339 ²	Indiana state fairgrounds (1.8 km), former American Lead facility (2.4 km), fabricated metals facility (1.7 km)

: vents,
vents.
· • • • • • • • • • • • • • • • • • • •
bile
s,
ction
iny,
units,
(<1
)
ated
ls
(1.7
,
polis
or
way
m)

- 634 ¹W. 30th Street, 2013.
- 2 N. Washington Blvd, 2013.
- 636 ³S. Harding Street, 2016.
- 637 ⁴N. Harding St., 2013.
- 638 *<u>Traffic Count Database System (TCDS) (ms2soft.com)</u>
- 639
- 640

Supplementary References

- Dietrich, M., Krekeler, M. P., Kousehlar, M., & Widom, E. (2021). Quantification of Pb
- 642 pollution sources in complex urban environments through a multi-source isotope mixing model
- based on Pb isotopes in lichens and road sediment. *Environmental Pollution*, 288, 117815.
- 644 Galer, S. J. G., & Abouchami, W. (1998). Practical application of lead triple spiking for
- 645 correction of instrumental mass discrimination. *Mineral. Mag. A*, *62*, 491-492.
- 546 Jeong, H., Ra, K., & Choi, J. Y. (2021). Copper, Zinc and Lead Isotopic Delta Values and
- 647 Isotope Ratios of Various Geological and Biological Reference Materials. *Geostandards and*
- 648 *Geoanalytical Research*, 45 (3), 551–563.
- Kousehlar, M., & Widom, E. (2020). Identifying the sources of air pollution in an urban-
- 650 industrial setting by lichen biomonitoring-A multi-tracer approach. *Applied Geochemistry*, *121*,
 651 104695.

- LeGalley, E., Widom, E., Krekeler, M. P., & Kuentz, D. C. (2013). Chemical and lead isotope
- 653 constraints on sources of metal pollution in street sediment and lichens in southwest654 Ohio. *Applied Geochemistry*, *32*, 195-203.
- 655 Smith, D.B., Cannon, W.F., Woodruff, L.G., Solano, Federico, Kilburn, J.E., and Fey, D.L.,
- 656 2013, Geochemical and mineralogical data for soils of the conterminous United States: U.S.
- 657 Geological Survey Data Series 801, 19 p., https://pubs.usgs.gov/ds/801/.
- Wang, Z., Dwyer, G. S., Coleman, D. S., & Vengosh, A. (2019). Lead isotopes as a new tracer
- 659 for detecting coal fly ash in the environment. *Environmental Science & Technology*
- 660 *Letters*, 6(12), 714-719.