



Research article

Complexities in attributing lead contamination to specific sources in an industrial area of Philadelphia, PA

Mary Ellen Tuccillo^a, Julie Blue^a, Jonathan Koplos^a, Jack Kelly^b,
Richard T. Wilkin^{c,*}

^a ERG, 561 Virginia Rd., Suite 300, Building 4, Concord, MA 01742, USA

^b U.S. Environmental Protection Agency, Region 3, Four Penn Center 1600 John F. Kennedy Boulevard, Philadelphia, PA 19103, USA

^c U.S. Environmental Protection Agency, Center for Environmental Solutions and Emergency Response, 919 Kerr Research Dr., Ada, OK 74820, USA

ARTICLE INFO

Keywords:

Lead
Urban lead
Lead isotopes
Soil

ABSTRACT

Globally, lead (Pb) contamination is one of the top ten chemical exposure issues affecting public health. The identification of specific Pb sources provides valuable information to determine assignment of liability for site cleanup, improve sampling plans and develop remedial strategies. This paper examines Pb concentrations and Pb isotopic data from samples collected at and near the site of a Pb paint production facility with a long operating history. Although high soil Pb concentrations were found at the site, Pb concentrations in surrounding neighborhoods did not simply decline with distance from the site. We evaluated soil concentrations and isotopic mixing lines to explore potential sources of Pb pollution. Three-isotope plots showed overlap of site samples and the surrounding neighborhood, consistent with pollution from the facility affecting offsite soils. A major challenge in separation of potential sources, however, is that the isotopic signatures of other potential Pb sources fall within the range of the soil data. The long operational site history, soil disturbances, the presence of nearby smelters, and other local and remote sources affect identification of lead sources. This analysis demonstrates that source attribution can be confounded by incomplete site and material sourcing information. An integrated approach that includes in-depth site characterization and an evaluation of historical activities (e.g., Pb ores used over time, amounts of Pb emitted by all area smelters, land use changes, and soil disturbances) is important for determining source attribution. This analysis provides insight into future site investigations where soil lead contamination has resulted from a long industrial history in an urban setting.

1. Introduction

Urban soils and sediments across the United States have been documented as having lead (Pb) concentrations above naturally occurring levels [1–4]. While soils will have some natural Pb concentrations, any such background signal can be overwhelmed in urban areas by a variety of historic and current anthropogenic sources. Commonly recognized anthropogenic Pb sources in urban environments include manufacturing processes using Pb, vehicles (past combustion of leaded gasoline), coal-fired power plant emissions, Pb-based paint manufacturing and use, Pb ore processing (smelting), petroleum refining, and (to a lesser degree) municipal waste

* Corresponding author.

E-mail address: wilkin.rick@epa.gov (R.T. Wilkin).

<https://doi.org/10.1016/j.heliyon.2023.e15666>

Received 13 January 2023; Received in revised form 17 April 2023; Accepted 18 April 2023

Available online 21 April 2023

2405-8440/© 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

incineration [5–7]. Demolition, construction, excavation, and natural processes such as erosion and bioturbation can also redistribute, resuspend, and remobilize Pb in dust and soil in urban areas [8–12].

Pb is a heavy metal long known to be harmful to human health. The World Health Organization considers Pb poisoning one of the top ten chemical-based public health concerns globally [13]. Pb is particularly harmful to neurological development in children [4, 14–17]. Pb pollution manifests in human blood and is readily measured as blood lead level (BLL). In cases of point sources (e.g., lead smelters), proximity to such a facility has been associated with higher BLLs in children [18]. There is no identified threshold or safe level of Pb in blood, and the developing neurological system of a child can be affected adversely at BLLs of ≤ 5 $\mu\text{g}/\text{dL}$ [8,13,17].

The USEPA currently defines a soil Pb hazard as at or above 400 mg/kg for bare soils in areas where children play and 1200 mg/kg in other areas [19]. Natural Pb concentrations vary with geology, with a national median value of 18 mg/kg for uncontaminated surface soils in the United States [20,21]. The median background natural soil Pb concentration in Pennsylvania is 46.4 mg/kg according to the USGS's Background Soil-Lead Survey: State Data [22]. Where contamination has occurred, soil concentrations can reach hundreds or even thousands of mg/kg. In Philadelphia, soil sampling efforts have identified hundreds of soil samples across many neighborhoods with Pb concentrations exceeding 400 mg/kg [2,23–25]. Philadelphia's Kensington neighborhood has been affected by several local Pb smelters and other Pb emitting facilities over the years, as well as by a large, well-known facility that produced Pb-based paints and other products (J.T. Lewis; JTL). Particularly high Pb soils concentrations (2000 mg/kg or greater) have been found within Kensington and other industry-heavy Philadelphia neighborhoods [2,26]. Because Pb is persistent in soils and has limited mobility (e.g., by leaching), Pb accumulates over decades in the top several centimeters of soils [5].

Apportionment of the relative contributions of various pollution sources to a contaminated area can be done through increasing knowledge of site history, identifying pollutant sources, establishing contaminant transport mechanisms, and studying other factors affecting present-day soil lead concentrations. Apportionment can also be used to help communicate to residents the relative risk and the complex nature of urban contamination.

In this paper, we explore the contribution of lead contamination in the soils from the site of the former JT Lewis (JTL) facility in Philadelphia, PA to the surrounding neighborhoods. The JTL facility produced paint and other Pb-bearing products for nearly 150 years (1849–1996) and generated emissions that affected both the plant site and the surrounding area. Pb concentration and stable isotopic data from the onsite (from the JTL facility) and offsite (surrounding neighborhood) soils are used to evaluate the potential for identifying pollution from the former JTL facility and other sources, including atmospheric or local contributors.

2. Background

2.1. Source apportionment using Pb stable isotopes

While total Pb concentrations are important in characterizing the contaminated media, they are generally inadequate to determine Pb pollution source attribution among multiple pollution sources unless one source has produced contamination far in excess of other sources. Because of this, fingerprinting methods are applied to trace the influence of sources in contaminated media. NAS [27] describe four approaches for defining fingerprints: multielement fingerprints (e.g., combinations of metals), physical/mineralogical characteristics of the soils, lead speciation, and Pb stable isotopes. Stable Pb isotope systematics are advantageous in that the isotopic signal for a source is maintained as Pb moves through the environment.

Pb isotopes have been used in rural, suburban, and more heavily urbanized settings, and analyses have been done on media that include soils, groundwater, river or lake sediments, dust, and atmospheric deposition [3,6,10,28–37].

2.2. Use of stable Pb isotopes

Four stable Pb isotopes occur in geologic materials (i.e., ore bodies, coals, and uncontaminated background rocks and soils). ^{204}Pb is "primordial," or original, in geologic materials, ^{206}Pb and ^{207}Pb are the end points in the decay chains of ^{238}U and ^{235}U , respectively, and ^{208}Pb is the end of the decay chain for ^{232}Th [38,39]. Th and the two U isotopes have different half-lives, so the ratios among the resulting Pb isotopes shift over geologic time scales. Because of differences in their ages and relative amounts of ^{204}Pb , Th, and U, geologic materials develop Pb isotope ratios that serve as a signature or fingerprint to differentiate them [28,37,40].

The ratios of Pb isotopes do not change from the original ore or coal through mining, refining, manufacturing, and release to the environment. Thus, isotopic signatures can be effective tracers [27–29,32,41]. Products and pollution from an anthropogenic source, such as industry, will carry the isotopic signature of the Pb source initially purchased.

2.3. Mixing lines

The Pb isotopic signature of an environmental sample (e.g., soil, water, sediment) is a mixture of the isotopic signatures of the sources contributing Pb to the sample. If the isotopic signatures of primary sources are known or can be estimated (often a natural and an anthropogenic input) the relative contributions of the sources for each sample point can be discerned [38,42,43].

A graphical approach is often used to evaluate likely sources. The isotopic ratios of samples may be plotted against the inverse Pb concentration (e.g., $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $1/\text{Pb}$). Three-isotope graphs plot two ratios (e.g., $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$), producing a more useful graph when the Pb concentrations are heterogeneous. In a simple binary system, samples will plot along a mixing line connecting two sources with known isotopic signatures [39,40].

Figure S1 illustrates a general binary mixing line, where samples along the line show the combined influences of two sources of Pb.

Proximity to the sources affects the relative contributions of the sources to each sample. If samples along the mixing line are well correlated, the percent contribution of each source can be estimated mathematically with a mixing model [39,40]. Other, more complex mixing models exist besides simple binary models. However, for mixing models to be effective, the Pb isotopic signatures of the sources must be known and sufficiently different from each other.

Mixing models with Pb isotopes have been used to assess sources in settings and media that include soils, lake sediments, and river sediments [29,42,44,45]. For example, Brugam et al. [39] used a binary mixing model to assess lake sediments near a smelter in southern Illinois and determine the relative contributions of background sediment and an anthropogenic signature. Settings that have been particularly amenable to binary mixing models tend to have a detectable natural background signature and a known anthropogenic isotopic signature from a heavy local polluter [40]. Use of mixing lines becomes more problematic in urban settings affected by numerous sources with “weaker” signals [3,26,27,41].

2.4. Major sources of lead

Local industrial activities can contribute significant Pb pollution to soils, as exemplified when mining and smelting increased in the 1800s [10,46] and produced emissions that polluted surrounding areas. Coal burning in local facilities and the use of lead-bearing paint in older houses can also contaminate soils. Globally, Pb ores and coal ores have a large spread of Pb isotopic signatures, which can help trace sources of Pb pollution, if the original ore or coal used by a local Pb source is either known or can be inferred [10] (Figure S2).

Atmospheric transport of Pb from coal-fired power plants and internal combustion engines using leaded gasoline has been documented worldwide as an important source of Pb contamination in soils [27,28,31,47]. Atmospheric lead can be transported for long distances in sub-micron aerosols [28]. Pb from gasoline additives was a significant component of atmospheric deposition from the 1940s through the 1980s and has received significant attention and research globally [10,46]. Though leaded gasoline was sold in the U.S. until January 1996, inputs decreased since its phase-out in 1975 [6,48]. Nevertheless, lead from leaded gasoline remains as a

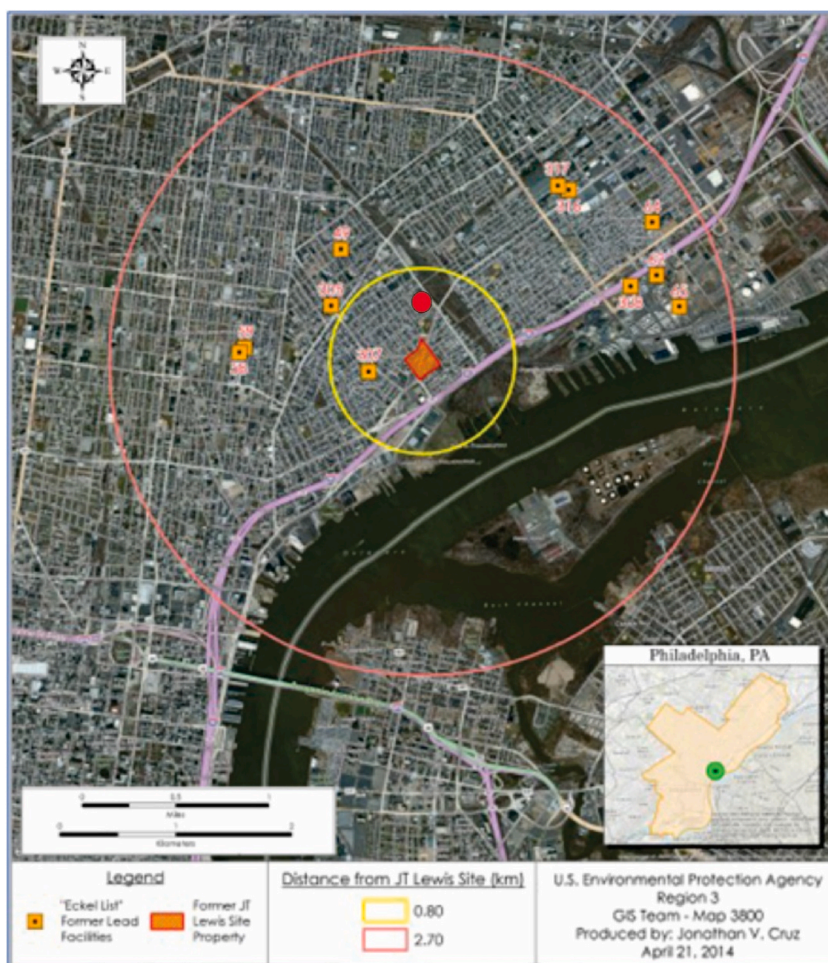


Fig. 1. Location map of former JTL facility in Philadelphia. The red circle shows the cemetery location.

legacy pollutant, along with decades of atmospheric deposition from coal-fired power plants.

Isotopic signatures for atmospheric inputs can be inferred from ore or coal data if the likely original sources of coal or Pb ore (e.g., regional use of coal from particular localities or use of certain ores for Pb gasoline additives) are known. Atmospheric inputs have also been studied by sampling aerosols, precipitation, and sediments in settings that are less disturbed than urban soils. This gives an indication of typical isotopic ratios to be expected from atmospheric deposition. Figure S3 shows the isotopic signatures of domestic coals from a number of localities and includes data representing atmospheric deposition, as measured in sediments and aerosols. A 2000 study [45] found that more than 50% of the sediment Pb in a California river system was attributable to leaded gasoline; the gasoline signal has remained discernible in other studies and other settings over time [35].

2.5. Challenges in identifying sources

Estimation of the relative contributions of up to two to three sources can be done reasonably well with a mixing model if there are adequate data [28]. Source attribution becomes more difficult if the Pb isotopic signatures of the sources are similar, or if there are numerous potential Pb contributors. In such cases, either a clear mixing line will not be obtained, or multiple potential sources may plot along a line; then, sources cannot be differentiated or confirmed, and source contributions cannot be readily estimated.

Urban areas present challenges for Pb source apportionment, including multiple potential local and remote sources, complex histories with variable amounts of historical documentation, long time frames during which contamination may have accumulated, and variability in site histories and Pb sources over time. Furthermore, the Pb signatures of ores and coals from different areas may overlap, and many can be collinear or overlapping when plotted on some three-isotope graphs (Figure S3). This makes it more difficult to separate the signals of several sources.

These are acknowledged difficulties in industrialized urban areas [3,26,40] and have also been encountered in other settings (e.g., sediments). An additional challenge is that the Pb used for a certain application (e.g., manufacturing) may have changed over time [33]. For example, the Pb signature from leaded gasoline shifted repeatedly; Mississippi Valley type ores were favored in the 1960s and 1970s until the preferred ore source changed again in the 1980s [40]. These temporal changes introduce considerable uncertainty into source apportionment, necessitating care in selecting data that may be most representative of the age and location of the samples. In this study, we present an example of an urban neighborhood with a history of Pb smelting and manufacturing, where the above challenges have limited the conclusions drawn from an extensive set of Pb concentration and isotopic data.

3. Materials and methods

3.1. Site description

Located in the Kensington area of Philadelphia, PA, the John T. Lewis site (also known as Dutch Boy Paint, National Lead, and Anzon) operated from 1849 to 1996 as a lead paint production and lead processing facility (Fig. 1; Figure S4). This eight-and-a-half-acre, fifty-two building site used kilns, oxidizing furnaces, and corroding beds to process lead into various powders for use in bathtub enamel, paint, car batteries, soaps, and plastics [24]. An 1867 marketing advertisement listed white lead, red lead, litharge, sugar of lead, zinc white, linseed oil, and paints and colors of all kinds among products generated at the site [49]. Although it was not the only lead processing facility in the area, the John T. Lewis site was the most prominent [24]. Previous sampling efforts in the area are described in the Supporting Information.

General plant emissions, equipment malfunctions, and fires all contributed to spreading lead onsite and to the surrounding community, where it accumulated in soils. The facility began monitoring lead emissions in the 1960s and reported total lead emissions of 70,220 pounds in 1971 [49]. Lead emissions ranged from 14,600 to 29,000 pounds from 1981 to 1986, then dropped to 521 pounds in 1987. The buildings at the site were demolished in 1999 and replaced by several commercial buildings. The 2010 Census indicates that approximately 7000 people live in this area (tract 160). Approximately 90% of the population in this area are white, 6% Hispanic/Latino, 2.8% Black, 2.6% Asian and other Pacific Islands [49].

In addition to JT Lewis, several lead smelters operated in the Kensington area and nearby Philadelphia "River Ward" neighborhoods (Fig. 1; Figure S5 and Table S1). Within two miles to the northwest these included Car-Mor Metals, William White Smelting Works (also known as White Bros. Smelting Company), Atlantic Metals Corporation, American Alloys Co., Jos. Rosenthal's Sons, Liberty Smelting, Standard Metals, and Philadelphia Smelting and Refining. Within three miles to the northwest and northeast, approximately 15 operated at various times dating back to the late 1800s. These were all cited in EPA's Superfund Cleanup program at one time and are on what is referred to as the "Eckel list from 2001"; however, they are not currently on the National Priority List [50]. Little information is available for these facilities and a few, although identified as smelters, may only have processed metals or served as scrap metal facilities. William White Smelting Works is considered an example of typical 19th century industries in Philadelphia. It consisted of two one-story buildings and at one point employed 63 people. Smelting at William White was discontinued in 1926 [51]. These sites were substantially smaller than JT Lewis.

Although EPA hired a historic and cultural research firm to research the facility operations, details of their sizes, operations, amounts of material produced, and emissions were not available or limited at best. Therefore, although very high soil Pb concentrations (>1000 mg/kg) have been found near these facilities [24], the magnitude of their local impacts cannot be readily compared to contamination from JT Lewis. In recent years, the Kensington neighborhood has been undergoing extensive redevelopment, causing increased soil disturbance and Pb exposure risks to the local residents [24].

3.2. Sampling and analysis

Data for this study are from three soil sampling efforts: one focused on the former JTL site during excavation activities for a convenience store located on the property, another on soils in the surrounding neighborhoods during an EPA-funded blood/environmental lead study, and a third set of samples from a cemetery located near the JTL site. Samples were collected in duplicate from 23 locations at the site of the former JT Lewis Facility (“onsite samples”). Sixty-four (64) soil samples were taken by the U.S. EPA and its partner public health agencies in 2014 in the neighborhoods surrounding the former JTL facility (“offsite samples”) at distances ranging from immediately outside the site to approximately 2 miles away. Samples were analyzed for metal (loid)s, including total Pb, and Pb stable isotope ratios. In 2019, thirty (30) soil samples were taken from 15 locations in St. Anne’s cemetery at Lehigh Avenue and Memphis Street, located within three hundred yards from the former JTL facility (Fig. 1). Soil samples were collected from the top 30 cm from the sampling locations using clean plastic spoons and plastic sampling containers.

Samples were screened in the field for weight percent (wt. %) Pb by x-ray fluorescence (Figure S6). In the laboratory, samples were digested using microwave-assisted methods (CEM MARSXpress) and analyzed for metals/metalloids using an Inductively Coupled Plasma-Optical Emission Spectrometer (PerkinElmer Optima 8300DV; ICP-OES). Lead isotope ratios ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$) were measured using a High Resolution-Inductively Coupled Plasma-Mass Spectrometer (Thermo Element2, HR-ICP-MS). Sample digests were diluted to $\sim 3 \mu\text{g Pb/L}$. Analysis was conducted in the fast-scanning operation mode and the instrument detector dead time was determined from analysis of uranium ICP-MS standards over the concentration range of 0.1–2 $\mu\text{g/L}$. Isotopic analysis was conducted using the pulse counting detector by maintaining ^{208}Pb signals $< 3 \times 10^6$ counts per second. Signal stability was maximized during sample introduction by using a Peltier-cooled cyclonic spray chamber (ESI PC³) and a micromist nebulizer. Mass bias was corrected through frequent analysis of National Bureau of Standards Standard Reference Material 981. Each set of 10 samples for Pb isotope ratios included one second source standard and one sample duplicate. Details of the lab-based analyses and Quality Control procedures and results are provided in the Supporting Information (Table S2).

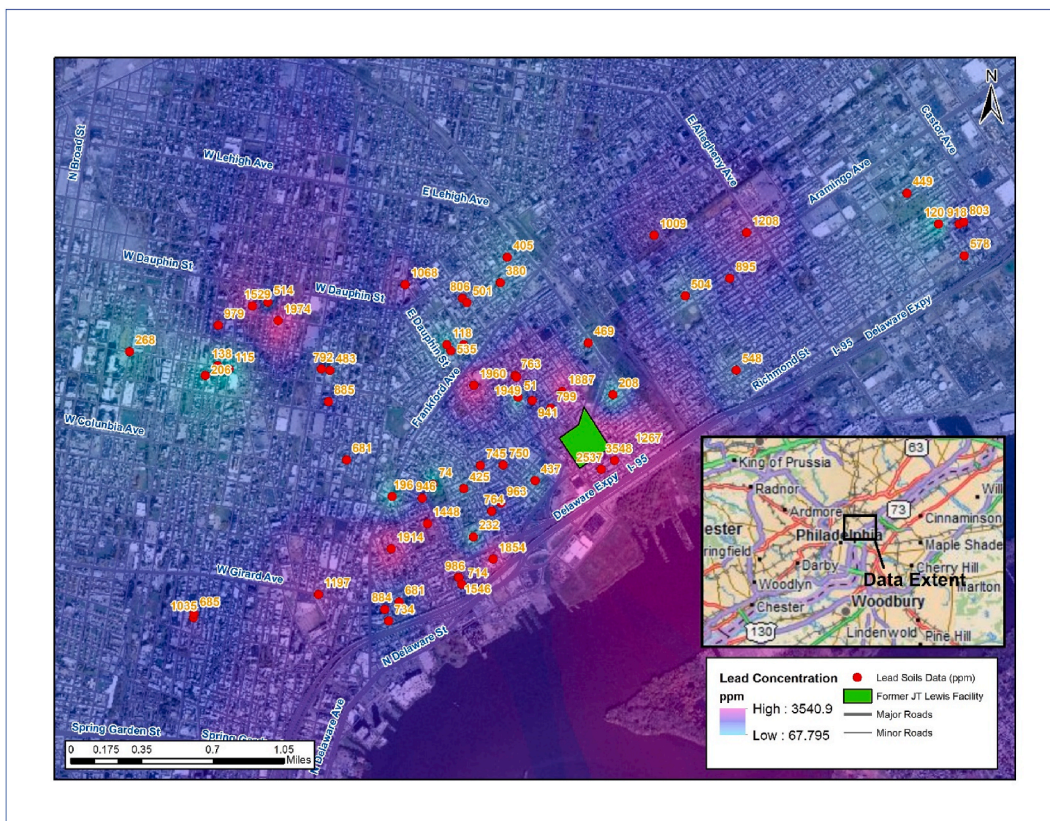


Fig. 2. Map of lead concentrations (mg/kg) in offsite soil samples and their relationship to the former JT Lewis facility (shown in green). Values are shown for individual data points, and colored shading shows patterns in concentrations.

4. Results and discussion

4.1. Lead concentrations and distribution

Figure S7 shows the summary statistics, ranges, and distributions of lead concentrations for the onsite, offsite, and cemetery data. Lead concentrations in samples from the former JTL facility ranged from 3250 mg/kg to a maximum of approximately 207,000 mg/kg, with a median of 13,482. The offsite median (757 mg/kg) is more than an order of magnitude lower than the onsite samples, though still significantly above the EPA regional screening level (RSL) for Pb of 400 mg/kg. Concentrations in the cemetery samples are lower than both the onsite and offsite samples, falling below 400 mg/kg. The cemetery Pb concentrations were obtained both in the field by x-ray fluorescence and via lab-based measurements. These two sets of measurements for the cemetery samples were in good agreement (Supporting Information; Figure S6).

The elevated level of pollution encountered in these samples is consistent with work in the Kensington neighborhood by other researchers [2] and an investigative reporting effort [24] done in conjunction with an environmental testing laboratory (Criterion Labs). Lusby et al. [2] reported soil Pb concentrations of approximately 2000 mg/kg and as high as 3100 mg/kg (measured by XRF) near former smelters, as have studies at other locations [23] (548–2550 mg/kg). The investigative reporting effort sampled 114 locations in Philadelphia neighborhoods with a history of pollution from lead smelting, including the Kensington area. Their results included a maximum value of more than 9000 mg/kg and a minimum of 427 mg/kg. Two samples near the JTL site had more than 2000 mg/kg, and at least four samples had between 1000 and 1999 mg/kg. A similar effort near a former white Pb processing plant in Staten Island, New York, found offsite surface soil Pb concentrations of 383 mg/kg to 3510 mg/kg and onsite surface soil concentrations of 456–8005 mg/kg [3].

Greater soil contamination near local industrial sources has been observed in some studies, attributed to greater localized airborne transport [6,27,52]. Such relationships may be easier to determine in rural areas with limited anthropogenic sources and where the Pb signature for uncontaminated, native soils can be identified. In urban areas, the influences of a variety of local pollution sources as well as diffuse atmospheric deposition necessitate a more complex analysis.

Although very high soil Pb concentrations have been found in the neighborhood around the former JTL site, they do not correspond solely to distance from the site. A heat map of Pb concentrations in the offsite samples shows spatial variability, with an inconsistent distribution to the east and north of the facility and more consistently high values to the south and southwest (Fig. 2). The high concentrations immediately to the south/southeast of the site may reflect a Pb hot spot at the corner of the property described by Ruderman et al. [24] that was attributed to leakage of soil from poor capping.

The overall trends over the full area sampled suggests the influence of other local sources of Pb. One possibility is that the other former smelters and area industries dating back to the late 1800s served as additional sources. All but one are near sampling points where Ruderman et al. [24] found concentrations exceeding 500 mg/kg, although they are not consistently near the hottest spots in Fig. 2. The potential for uneven distribution of atmospheric deposition is unknown.

Throughout the offsite area, there are Pb sampling locations with low concentrations relatively close to those with much higher Pb values. This heterogeneity could reflect any of a number of small-scale and even lot-scale influences. Lead-based paint can contribute to high Pb concentrations adjacent to houses, and soils near roadways may be frequently disturbed [6,11]. Additionally, Pb in soil may be disturbed over time via natural physical mixing (e.g., freeze-thaw cycles, bioturbation) or geochemical processes (e.g., adsorption/desorption) [10]. Changes to a property, such as replacement with fresh soil or disturbance due to construction could also alter Pb concentrations in surficial soils. This is an important consideration given the redevelopment in the waterfront area of Philadelphia.

Because of the heterogeneous geographic distribution of lead concentrations around the former JTL site, tracing the movement of Pb from the former JTL site is not straightforward. The use of other approaches such as Pb stable isotopes is needed to complement the information on spatial distribution of concentrations [27,41].

4.2. Spatial distribution of lead isotopes

A map of $^{206}\text{Pb}/^{207}\text{Pb}$ ratios for offsite samples (Figure S8) shows less overall heterogeneity than the Pb concentration map in (Fig. 2). The clustering of more similar, lower $^{206}\text{Pb}/^{207}\text{Pb}$ values to the northwest, north, and northeast of the former JTL facility may indicate that pollution from JTL was carried in a generally northward direction. However, other contributions and the relative importance of the JTL facility cannot be constrained based on such mapping alone. Spatial patterns in both Pb concentrations and Pb isotopes can vary locally depending on several factors such as distances from industry, traffic patterns, wind patterns, and the potential lot-scale influences noted above [40] that were unable to be fully accounted for in this analysis.

4.3. Two- and three-isotope plotting of Pb isotopes

A graph of $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $1/\text{Pb}$ for the JTL area onsite, offsite, and cemetery samples (Figure S9) shows that the onsite and other samples are separated, with the onsite samples clustering with higher onsite concentrations and overall lower $^{206}\text{Pb}/^{207}\text{Pb}$ ratios. The two datasets overlap but show no linear relationship either within or between the two data sets (R^2 values of 0.106 and 0.705 for onsite and offsite samples, respectively).

Previous research has shown that if there is a great deal of heterogeneity resulting from more than two lead sources or the two sources have similar isotopic signatures, a linear relationship between the isotopic ratio and inverse concentration may not be observed [3,28,32,53,54]. The lack of binary mixing in Figure S9 suggests multiple contributions of lead that likely include atmospheric

deposition (including legacy gasoline pollution) and contamination from local historical industry.

Plotting $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 3a) for the onsite, offsite, and cemetery samples produces a mixing line, with onsite samples and offsite samples at opposite ends and with substantial overlap between the three datasets towards the middle of the plot. The substantial area of overlap among the datasets is consistent with the influence of pollution from the JTL site on neighborhood soils, but pollution sources common to both (other local industry, atmospheric inputs, gasoline) can also lie along a mixing line, making source apportionment complex.

It has been suggested that commonly used graphs such as $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{208}\text{Pb}/^{207}\text{Pb}$ can produce a speciously straight (binary) mixing line where there are multiple sources, thereby obscuring differences in sources [55]. It was suggested that a more reliable graph for source apportionment is $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{208}\text{Pb}/^{207}\text{Pb}$. The graph in Fig. 3b shows considerably more scatter rather than an apparent binary mixing line, consistent with a setting with multiple Pb sources.

Regardless of the number of potential sources, a strong isotopic signal from a prominent polluter of concern is desirable. Two high-Pb onsite sample locations in Fig. 3a–b also had the highest Pb concentrations (approximately 200,000 mg/kg and 76,000 mg/kg for the two sample pairs). These samples may be the most representative of the Pb used at the JTL facility and are a potential JTL source signature. The high total Pb concentrations for these sample pairs may mean that the JTL industrial signal is much stronger than for other sources such as atmospheric deposition. The offsite samples at the opposite end of the graphs could be interpreted to represent a neighborhood signature with less JTL influence than the other samples.

For the JTL high-Pb samples, specifics are lacking on the precise locations with respect to nearby onsite activities when the facility was in operation (e.g., spills, manufacturing activities, roadways, peeling paint). Such precision in sampling would be difficult because the facility was redeveloped after closing. The industrial timeframe represented by onsite samples is also unknown. Thus, although these data points may be a JTL source signature, it is unknown if this represents the entire site or what point in the site history given possible changes in ore sources over time.

For the other onsite data, the spread in values could result from factors such as changes in land use at the JTL facility, changes in the Pb ore purchased, and variability in how well offsite influences (e.g., leaded gasoline and coal burning) can be detected amid the high onsite Pb pollution. Both onsite and offsite samples will have incorporated the effects of both spatial and temporal variability. Although the data are consistent with pollution from the JTL facility contaminating the neighborhood soils, more historical details of the site and its operations as well as the history of the neighborhood are needed for a more complete data interpretation.

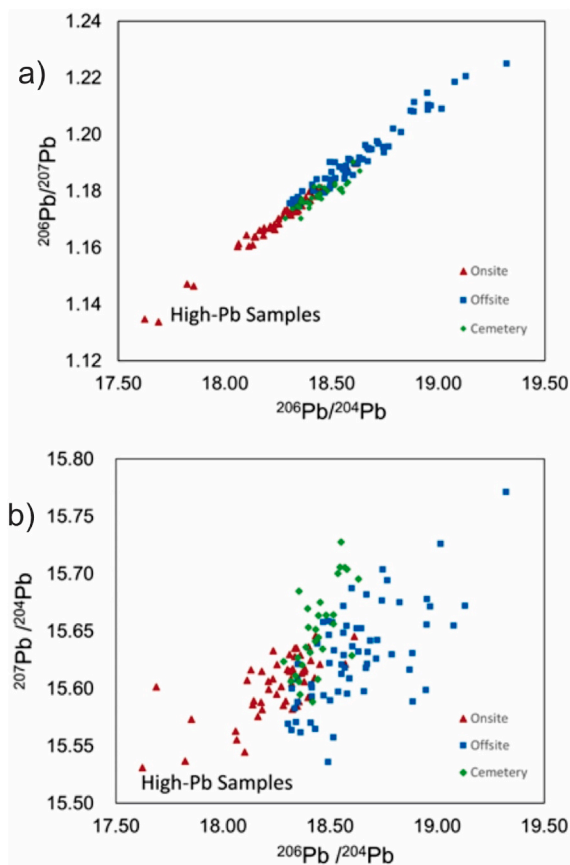


Fig. 3. Three isotope plots of a) $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ and b) $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ for onsite, offsite, and cemetery samples.

4.4. Potential Pb sources

Interpretation of sample data depends on having as thorough an understanding as possible of the sample locations, area history, and likely signatures for both local and remote sources. The data in Fig. 3 together with historical descriptions of the facility and its emissions, are consistent with the JTL facility contributing Pb pollution to the surrounding neighborhoods. However, the full range of sources and their relative amounts are difficult to isolate. Literature data on the isotopic signatures of several commonly expected contributors add context to the site-specific Pb isotope data.

4.4.1. Lead ore sources for area local industries

Because Pb isotopic signatures in ore bodies are maintained during mining, refining, and subsequent use in manufacturing, Pb pollution from the JTL facility should reflect the original lead ore sources. A plot of $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ plot (Fig. 4a) shows that the isotopic signatures of the onsite samples generally fall within the isotopic signature ranges of ores from Utah, Colorado, Montana, England, and Germany. Offsite samples generally fall within the ranges of ores from Mexico, England, and Germany. The isotopic signatures of the two highest onsite samples (high-Pb) are similar to signatures from ores in Utah, Montana, and Colorado, and possibly Australia depending upon the graphical presentation (Fig. 4a and b). Because of the wide range and overlap of some of the ores, tying the sample data back to any particular ore location and, by association, the facility that used it (i.e., an inferred JTL signature) is challenging. Also, the JTL site was active for more than a century, and if it sourced Pb from different localities over time, an isotopic signature from activities at the plant could reflect a mix of the signatures of those changing Pb sources [42].

Other studies have attempted to identify possible local industrial signatures. In a similar analysis, Pribil et al. [3] explored potential ore sources for the Jewett White Lead Company site in Staten Island New York, noting the potential ages and locations of Pb ores based on soil samples. As with the JTL site, records of lead used at the Jewett White site are not available. However, based on historical trends in Pb mining, the authors hypothesize that the plant may have used German, English, and Australian ores at separate times. In the Mississippi River floodplain, Brugam et al. [39] used a binary mixing model to identify an industrial isotopic signal in lake sediments that may represent a mixture of source ores used in area smelting. For the JTL site and Kensington neighborhood, additional historical information about JTL and the other area smelters with respect to emissions, Pb sourcing, and other industries that used Pb or burned coal would improve the ability to constrain Pb sources. Additionally, because the Pb in gasoline additives originates in Pb ores, potential ore signatures related to historical use of leaded gasoline must be considered.

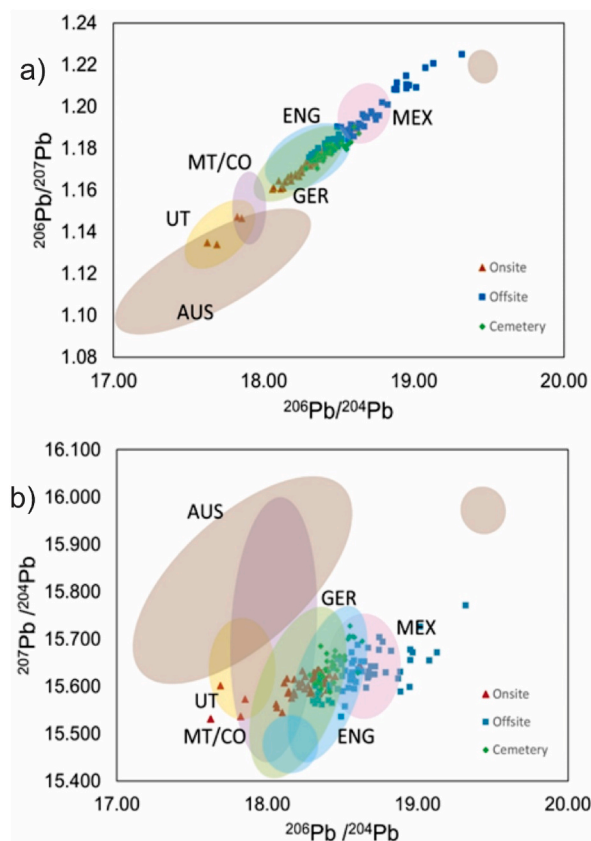


Fig. 4. Comparison of isotopic ratios of major ore localities with onsite, offsite, and cemetery samples: a) $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ and b) $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$. (Ore data from Rabinowitz, 2005 [56]; Brown, 1962 [57]).

4.4.2. Atmospheric sources - coals, and gasoline

Data in the literature representing airborne Pb in the U.S. have been obtained from sediment, precipitation, and aerosols sampling (Table S3 [58,59,60]; aerosols, numerous locations]; [61], Chesapeake Bay sediments]; [62], Boston Harbor and marine sediments]; [60], Lake Erie sediments]; [44], Great Lakes sediments]. These can reflect inputs from combustion of both leaded gasoline and coal combustion, and they can originate either locally or from more distant sources.

To explore possible coal signatures, isotopic data on coals from mines throughout the U.S. and Canada [63] are shown in along with the onsite, offsite, and cemetery samples in Fig. 5a and b. Among Chow and Earl's [63] data, we selected values for coals from Pennsylvania and West Virginia to represent the coal sources closest to Philadelphia. This was done with the assumption that coal sources, consumption, and associated emissions might be related to proximity and that the most important emitters might be situated west of Philadelphia in light of prevailing wind patterns. Confirmation of the history of coal usage over time would help in selecting the most representative set of values.

Fig. 5a and b both show overlap between the offsite samples and the coals, consistent with contribution of coal-burning stationary sources to Pb in soils [10]. The lesser overlap with onsite samples suggests that the atmospheric contributions to soils at the JTL facility may have been small in comparison to the isotopic signature of the large amount of Pb that originated onsite. The isotopic signatures of the samples overlap with coal signatures from several states, possibly due to a combination of changing coal sources over time as well as the natural overlap in signatures of some domestic coals.

Similarly, the average legacy gasoline ratios ($^{206}\text{Pb}/^{207}\text{Pb} = 1.1895$, and $^{208}\text{Pb}/^{206}\text{Pb} = 2.0618$) and average atmospheric deposition values ($^{206}\text{Pb}/^{207}\text{Pb} = 1.2004\text{--}1.2055$; $^{208}\text{Pb}/^{206}\text{Pb} = 2.0484\text{--}2.0525$) compiled by Wang et al. [6] fall within the range of offsite JTL area samples but are less represented among onsite samples. Leaded gasoline is acknowledged as a significant component of atmospheric Pb deposition, but the magnitude of Pb from gasoline relative to JTL pollution is not clear from the available data.

Although Pb can be transported long distances in aerosols, local deposition is also significant. Samples collected in close proximity

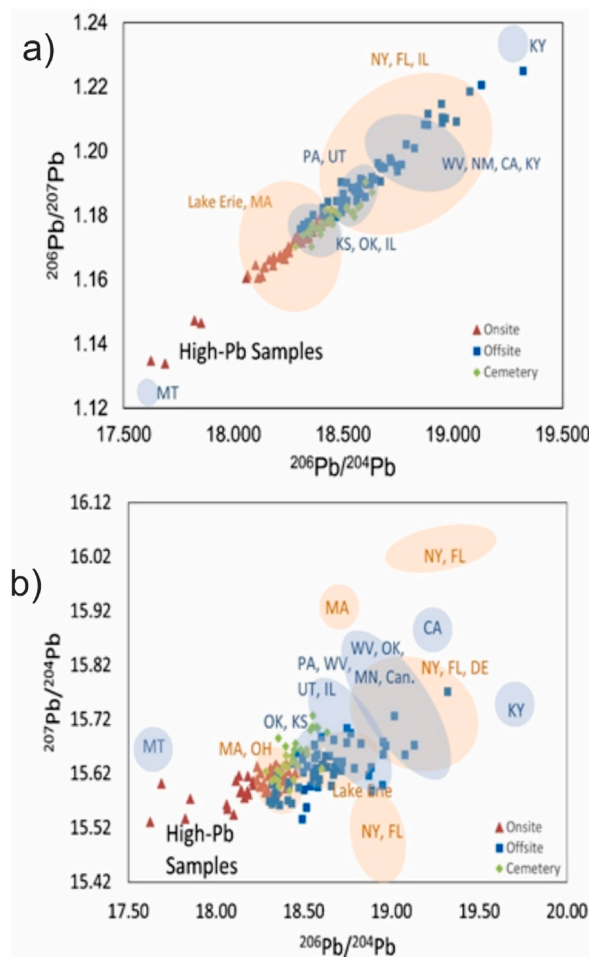


Fig. 5. Comparison of isotopic ratios of Pb from selected U.S. coals and atmospheric deposition with onsite, offsite, and cemetery samples. Orange shaded areas represent atmospheric deposition and blue shaded areas represent coals. Approximate geographic locations are given for the shaded areas: a) $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ and b) $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$. (Coal and atmospheric deposition data from Refs. [33,44,58,63]).

to roadways may be more affected by gasoline-derived Pb [5]. Therefore, clarifying sample locations with respect to roadways, especially those with high traffic volumes, can be helpful with source apportionments. In planning for future sampling programs, roadway locations should be considered.

4.4.3. Paint on buildings

Lead paint was banned in 1978, but it can still be a legacy source of pollution, especially if soils are disturbed and Pb-contaminated soils are brought to the surface [6]. High Pb concentrations and the associated isotopic signatures adjacent to house foundations in Durham, NC were linked to lead-based paint. In contrast, soil samples collected closer to urban streets showed signatures more consistent with a mixture of atmospheric deposition and leaded gasoline [6]. The average lead paint values of $^{206}\text{Pb}/^{207}\text{Pb}$ (1.185) and $^{208}\text{Pb}/^{206}\text{Pb}$ (2.0618) cited by Wang et al. [6] are within the range of the offsite samples in this study but not the onsite data. As with other types of manufacturing, the Pb isotopic signature of lead paints can vary by product and over time, making source fingerprinting difficult [5]. Lead concentrations and correlations with other selected metals (Ba, Ti, Zn, Cr, and Fe) are provided in [Figures S10-S12](#).

5. Conclusions

Results of this study highlight the substantial challenges and data needs for source attribution in a densely urbanized area where local manufacturing and smelting have occurred for many years. The JTL facility was located in the Kensington area of Philadelphia's riverfront, producing Pb-bearing products for approximately 150 years. Local and site history indicates that this site would have been a source of significant pollution to the nearby neighborhoods. Evidence for this includes soil samples from the JTL facility having a median Pb concentration of 13,480 mg/kg, and offsite soils from the surrounding neighborhood having a median concentration of 757 mg/kg; EPA's current regional screening level is 400 mg/kg.

Plotting of Pb stable isotope ratios identified two onsite sampling locations that may represent a JTL signature. However, tracing this signature in a comprehensive source apportionment for the Kensington area is extremely difficult. In addition to JTL, the surrounding Kensington neighborhood also had additional Pb smelters whose isotopic signatures are currently unknown. Moreover, isotope data for offsite samples may reflect any other inputs from airborne transport of Pb (i.e., from coal burning or legacy pollution from leaded gasoline). The JTL site would have received these general inputs, but the onsite data would also be an amalgam of the isotopic signatures of Pb sources used during the operational lifespan of the facility. Offsite samples may have been affected by other, smaller-scale sources such as Pb paint.

This analysis underscores the need for an integrated approach to source apportionment in a complex urban area. At an initial stage of research, this includes careful site characterization and site history [27]. Information about industrial sources of greatest concern may include their operating history, layout, Pb sourcing (if possible), history of changes to land use onsite, and emissions history. Site characterization may also include pursuing historical information about the neighborhood near a significant industrial polluter, including what other industrial and development activities took place and when they operated.

Author contribution statement

Mary Ellen Tuccillo: Analyzed and interpreted the data; Wrote the paper.

Julie Blue: Analyzed and interpreted the data; Wrote the paper.

Jonathan Koplos: Analyzed and interpreted the data; Wrote the paper.

Jack Kelly: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data.

Richard Wilkin: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Funding statement

This research was funded by the U.S. Environmental Protection Agency.

Data availability statement

Data included in this article are available from the corresponding author.

Declaration of interest's statement

The authors declare no conflict of interest.

Additional information

Additional methods, background, and data analysis are provided in the Supporting Information.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to

influence the work reported in this paper.

Acknowledgements

This research was sponsored by EPA's Office of Research and Development. The views expressed in this article are those of the authors and do not necessarily represent the views or the policies of the U.S. Environmental Protection Agency. The authors are not aware of any competing financial interests or personal relationships that would have introduced bias in the research or conclusions of the work reported in this paper. This document has been reviewed in accordance with U.S. Environmental Protection Agency policy and approved for publication. Any mention of trade names, manufacturers or products does not imply an endorsement by the United States Government or the U.S. Environmental Protection Agency. The manuscript was prepared by Eastern Research Group (ERG) under EPA Contract Nos. EP-C-16-015 and 68HERC21D0003 (task order 15). Technical guidance was provided by Dr. Chi Ho Sham, and Rose Malanga and Jason Rose provided research support.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.heliyon.2023.e15666>.

References

- [1] M.A.S. Laidlaw, G.M. Filippelli, S. Brown, J. Paz-Ferreiro, S.M. Reichman, P. Netherway, A. Truskewycz, A.S. Ball, H.W. Mielke, Case studies and evidence-based approaches to addressing urban soil lead contamination, *Appl. Geochem.* 83 (2017) 14–30, <https://doi.org/10.1016/j.apgeochem.2017.02.015>.
- [2] G. Lusby, C. Hall, J. Reiners, Lead contamination of surface soils in Philadelphia from lead smelters and urbanization, *Environ. Justice* 8 (1) (2014) 6–14, <https://doi.org/10.1089/env.2014.0008>.
- [3] M.J. Pribil, M.A. Maddaloni, K. Staiger, E. Wilson, N. Magriples, M. Ali, D. Santella, Investigation of off-site airborne transport of lead from a superfund removal action site using lead isotope ratios and concentrations, *Appl. Geochem.* 41 (2014) 89–94.
- [4] H.W. Mielke, J.C. Anderson, K.J. Berry, P.W. Mielke, R.L. Chaney, M.L. Leech, Lead concentrations in inner city soils as a factor in the child lead problem, *Am. J. Publ. Health* 73 (1983) 1366–1369, <https://doi.org/10.2105/AJPH.73.12.1366> [PMC free article].
- [5] C.A. Nezat, S.A. Hatch, T. Uecker, Heavy metal content in urban residential and park soils: a case study in Spokane, Washington, USA, *Appl. Geochem.* 78 (2017) 186–193.
- [6] Z. Wang, A.M. Wade, D.D. Richter, H.M. Stapleton, J.M. Kaste, A. Vengosh, Legacy of anthropogenic lead in urban soils: Co-occurrence with metal(loids) and fallout radionuclides, isotopic fingerprinting, and in vitro bioaccessibility, *Sci. Total Environ.* 806 (Part 3) (2022) 51276. <http://doi: 10.1016/j.scitotenv.2021.151276>.
- [7] Y.-Z.H. Li, L. Shao, X. Zhou, P. He, Impact of municipal solid waste incineration on heavy metals in the surrounding soils by multivariate analysis and lead isotope analysis, *J. Environ. Sci.* 82 (2019) 47–56. <https://www.sciencedirect.com/science/article/abs/pii/S1001074218321752#!>.
- [8] Agency for Toxic Substances and Disease Registry (ATSDR), What are possible health effects from lead exposure? Lead toxicity website. https://www.atsdr.cdc.gov/csem/leadtoxicity/physiological_effects.html#:~:text=In%20children%2C%20health%20effects%20generally,of%20%E2%89%A45%20CE%BCg%2Fdl,2019. Accessed March 23, 2022.
- [9] T.M. Young, D.A. Heeraman, G. Sirin, L. Ashbaugh, Resuspension of soil as a source of airborne lead near industrial facilities and highways, *Environ. Sci. Technol.* 36 (11) (2022) 2484–2489.
- [10] L. Ma, J. Konter, E. Herndon, L. Jin, G. Steinhofel, D. Sanchez, S. Brantley, Quantifying an early signature of the industrial revolution from lead concentrations and isotopes in soils of Pennsylvania, USA, *Anthropocene* 7 (2014) 16–29.
- [11] S. Zahran, H.W. Mielke, S.P. McElmurry, G.M. Filippelli, M.A.S. Laidlaw, M.P. Taylor, Determining the relative importance of soil sample locations to predict risk of child lead exposure, *Environ. Int.* 60 (2013) 7–14, <https://doi.org/10.1016/j.envint.2013.07.004>.
- [12] M.A.S. Laidlaw, G.M. Filippelli, Resuspension of urban soils as a persistent source of lead poisoning in children: a review and new directions, *Appl. Geochem.* 23 (8) (2008) 2021–2039.
- [13] World Health Organization (WHO), Lead Poisoning, in: *Lead Poisoning (who.int)*, WHO, Geneva, Switzerland, 2021 (accessed 22 March 2022).
- [14] E.B. McCabe, Age and sensitivity to lead toxicity: a review, *Environ. Health Perspect.* 29 (1979) 29–33. Age and sensitivity to lead toxicity: a review. (nih.gov).
- [15] V.I. Naranjo, M. Hendricks, K.S. Jones, Lead toxicity in children: an unremitting public health problem, *Pediatr. Neurol.* 113 (2020) 51–55, <https://doi.org/10.1016/j.pediatrneurol.2020.08.005>.
- [16] S. Hou, L. Yuan, P. Jin, B. Ding, N. Qin, L. Li, X. Liu, Z. Wu, G. Zhao, Y. Deng, A clinical study of the effects of lead poisoning on the intelligence and neurobehavioral abilities of children, *Theor. Biol. Med. Model.* 10 (1) (2013), <https://doi.org/10.1186/1742-4682-10-13>.
- [17] D.C. Bellinger, Very low lead exposures and children's Neurodevelopment, *Curr. Opin. Pediatr.* 20 (2) (2008) 172–177, <https://doi.org/10.1097/mop.0b013e3282f4f97b>.
- [18] D.L. Morse, P.J. Landrigan, B.F. Rosenblum, J.S. Hubert, J. Housworth, El Paso revisited: epidemiologic follow-up of an environmental lead problem, *JAMA* 242 (1979) 739–741.
- [19] U.S. Environmental Protection Agency (USEPA), Lead in soil USEPA-region III. <https://www.epa.gov/sites/default/files/2020-10/documents/lead-in-soil-aug2020.pdf>, 2020.
- [20] D. Smith, W. Cannon, L. Woodruff, F. Solano, K. Ellefsen, Geochemical and Mineralogic Maps of Soils of the Conterminous United States; USGS Open-File Report 2014-1082, USGS, Reston, VA, USA, 2014.
- [21] L. Datko-Williams, A. Wilkie, J. Richmond-Bryant, Analysis of U.S. soil lead (Pb) studies from 1970 to 2012, *Sci. Total Environ.* 468–469 (2014) 854–863.
- [22] U.S. Environmental Protection Agency (USEPA), USGS Background Soil-Lead Survey: State Data, 2022. Available online: USGS Background Soil-Lead Survey: State Data | US EPA (accessed 22 March 2022).
- [23] W.P. Eckel, M.B. Rabinowitz, G.D. Foster, Discovering unrecognized lead-smelting sites by historical methods, *Am. J. Publ. Health* 91 (4) (2001) 625–627, <https://doi.org/10.2105/ajph.91.4.625>. <https://www.ncbi.nlm.nih.gov/pmc/articles/PMC1446633/pdf/11291377.pdf>.
- [24] W. Ruderman, B. Laker, D. Purcell, J. Griffin, In Booming Philadelphia Neighborhoods, Lead-Poisoned Soil Is Resurfacing. *Philadelphia Inquirer*, 2017. <https://www.inquirer.com/news/inq/philadelphia-lead-soil-fishtown-construction-dust-20170618.html>.
- [25] Center of Excellence in Environmental Toxicology (CEET), Lead Soil Sampling in the Philadelphia Region, University of Pennsylvania, Perlman School of Medicine, 2022. | CEET (upenn.edu). (accessed March 23, 2022).
- [26] H. Caballero-Gómez, H.K. White, M.J. O'Shea, R. Pepino, M. Howarth, R. Giere, Spatial analysis and lead-risk assessment of Philadelphia, USA, *GeoHealth* 6 (2022), e2021GH000519, <https://doi.org/10.1029/2021GH000519>.

- [27] National Academies of Sciences, Engineering, and Medicine (NAS), Investigative Strategies for Lead Source Attribution at Superfund Sites Associated with Mining Activities, The National Academies Press, Washington, DC, 2017, <https://doi.org/10.17226/24898>.
- [28] H.F. Cheng, Y.N. Hu, Lead (Pb) isotopic fingerprinting and its applications in lead pollution studies in China: a review, *Environ. Pollut.* 158 (2010) 1134–1146.
- [29] R. Ayuso, N.K. Foley, Chapter 12. Lead and strontium isotopes as monitors of anthropogenic contaminants in the surficial environment, in: Book: Environmental Geochemistry: Site Characterization, Data Analysis and Case Histories, 2018, <https://doi.org/10.1016/B978-0-444-63763-5.00013-6>.
- [30] B. He, S. Zhao, P. Li, J. Liang, Q. Fan, X. Ma, G. Zheng, J. Qui, Lead isotopic fingerprinting as a tracer to identify the pollution sources of heavy metals in the southeastern zone of Baiyin, China, *Sci. Total Environ.* 660 (2019) 348–357.
- [31] A.I. Cheema, G. Liu, B. Yousaf, Q. Abbas, H. Zhou, A comprehensive review of biogeochemical distribution and fractionation of lead isotopes for source tracing in distinct interactive environmental compartments, *Sci. Total Environ.* 719 (2020), 135658.
- [32] D. Xu, B. Gao, Lead isotopes combines with geochemical baseline in sediments: a novel tool to trace anthropogenic Pb sources, *Int. J. Environ. Res. Publ. Health* 17 (2020) 1112, <https://doi.org/10.3390/ijerph17031112>.
- [33] M. Dietrich, M.P.S. Krekeler, M. Kousehlar, E. Widom, Quantification of Pb pollution sources in complex urban environments through a multi-source isotope mixing model based on Pb isotopes in lichens and road sediment, *Environ. Pollut.* 288 (2021), 117815. <https://pubmed.ncbi.nlm.nih.gov/34329070/>.
- [34] D. Saint-Laurent, J. St-Laurent, M. Hahné, B. Ghaleb, C. Chapados, Using lead concentrations and stable lead isotope ratios to identify contamination events in alluvial soils, *Appl. Environ. Soil Sci.* 2010 (2010) 1–12.
- [35] A. Potra, L.S. Ruhl, J.R. Samuelsen, Legacy lead from past mining activity and gasoline additives: evidence from lead isotopes and trace element geochemical studies in the White River Basin, Southern Ozark Region, USA, *Geosciences* 8 (2018) 189, <https://doi.org/10.3390/geosciences8060189>.
- [36] R.M. Sharp, D.J. Brabander, Lead (Pb) Bioaccessibility and mobility assessment of urban soils and composts: fingerprinting sources and refining risks to support urban agriculture, *GeoHealth* 1 (2017) 333–345, <https://doi.org/10.1002/2017GH000093>.
- [37] L.S. Sherman, J.D. Blum, J.T. Dvonch, L.E. Graetz, M.W. Landis, The use of Pb, Sr, and Hg isotopes in Great Lakes precipitation as a tool for pollution source attribution, *Sci. Total Environ.* 502 (2015) 362–374.
- [38] G. Faure, T.M. Mensing, *Isotopes: Principles and Applications*, third ed., John Wiley and Sons, Inc, Hoboken, 2005.
- [39] R.B. Brugam, M. Ketterer, L. Maines, Application of a simple binary mixing model to the reconstruction of lead pollution sources in two Mississippi River floodplain lakes, *J. Paleolimnol.* 47 (2012) 101–112.
- [40] M. Komárek, V. Ettler, V. Chrástný, M. Mihaljevič, Lead isotopes in environmental sciences: a review, *Environ. Int.* 34 (2008) 562–577.
- [41] C.A.V. Moura, H.E. Gaudette, M.C. Carvalho, G.P. Morales, The use of lead isotope composition as a tool to investigate the anthropogenic impacts on the environment in the metropolitan region of Belém (PA), *TERRÆ* 1 (1) (2004) 16–25.
- [42] C. Cloquet, J. Carignan, G. Libourel, T. Sterckeman, E. Perdrix, Tracing source pollution in soils using cadmium and lead isotopes, *Environ. Sci. Technol.* 40 (2006) 2525–2530. <https://pubs.acs.org/doi/10.1021/es052232z>.
- [43] N. Walraven, B.J.H. van Os, G. Th Klaver, J.J. Middleburg, G.R. Davies, The lead (Pb) isotope signature, behaviour and fate of traffic-related lead pollution in roadside soils in The Netherlands, *Sci. Total Environ.* 472 (2014) 888–900.
- [44] J.R. Graney, A.N. Halliday, G.J. Keeler, J.O. Nriagu, J.A. Robbins, S.A. Norton, Isotopic record of lead pollution in lake sediments from the northeastern United State, *Geochem. Cosmochim. Acta* 59 (9) (1995) 1715–1728.
- [45] C.E. Dunlap, R. Bouse, A.R. Flegal, Past leaded gasoline emissions as a non-point source tracer in riparian systems, *Environ. Sci. Technol.* 34 (2000) 1211–1215.
- [46] M.K. Reuer, D.J. Weiss, Anthropogenic lead dynamics in the terrestrial and marine environment, *Phil. Trans. Roy. Soc. Lond.* 360 (2002) 2889–2904, <https://doi.org/10.1098/rsta.2002.1095>.
- [47] M. Diaz-Somoano, M.E. Kylander, Lopez, M.A. Anton, I. Suarez-Ruiz, M.R. Martinez-Tarazona, M. Ferrat, B. Kober, D.J. Weiss, Stable lead isotope compositions in selected coals from around the world and implications for present day aerosol source tracing, *Environ. Sci. Technol.* 43 (2009) 1078–1085.
- [48] H.W. Mielke, T.P. Covington, P.W. Mielke Jr., F.J. Wolman, E.T. Powell, C.R. Gonzales, Soil Intervention as a strategy for lead exposure prevention: the New Orleans lead-safe childcare playground project, *Environ. Pollut.* 159 (2011) 2071–2077.
- [49] Agency for Toxic Substances and Disease Registry (ATSDR), Health consultation soil data review for properties near the former John T. Lewis and brothers site, Philadelphia, Philadelphia county, Pennsylvania, EPA Facility ID: PAN000306638, https://www.atsdr.cdc.gov/HAC/pha/FormerJohnTLewisandBrothersSite/JT%20Lewis%20and%20Brothers%20Site%20_HC_%20-06-03-2014_508.pdf, 2014.
- [50] U.S. Environmental Protection Agency (USEPA), EPA lead smelter strategy summary report. <https://semspub.epa.gov/work/HQ/100000189.pdf>, 2017.
- [51] Workshop of the World, William white smelting works. <https://www.workshopoftheworld.com/fishtown/white.html>, 2007 (accessed 8/5/2022).
- [52] C.M. Aelion, H.T. Davis, S. McDermott, A.B. Lawson, Soil metal concentrations and toxicity: associations with distances to industrial facilities and implications for human health, *Sci. Total Environ.* 407 (2009) 2216–2223.
- [53] A. Nazarpour, M.J. Watts, A. Madhani, S. Elahi, Source, spatial distribution and pollution assessment of Pb, Zn, Cu, and Pb, isotopes in urban soils of ahvaz city, a semi-arid metropolis in southwest Iran, *Sci. Rep.* 9 (1) (2019) 5349.
- [54] M.D. Kaminski, S. Landsberger, Heavy metals in urban soils of East St. Louis, Part I: total concentration of heavy metals in soils, *J. Air Waste Manag. Assoc.* 50 (9) (2000) 1667–1679.
- [55] R.M. Ellam, The graphical presentation of lead isotope data for environmental source apportionment, *Sci. Total Environ.* 408 (16) (2010) 3490–3492.
- [56] M.B. Rabinowitz, Lead isotopes in soils near five historic American lead smelters and refineries, *Sci. Total Environ.* 346 (2005) 138–148.
- [57] J.S. Brown, Ore leads and isotopes, *Econ. Geol.* 57 (5) (1962) 673–720.
- [58] A. Bollhöfer, K.J. Rosman, Isotopic source signatures for atmospheric lead: the Northern Hemisphere, *Geochem. Cosmochim. Acta* 65 (11) (2001) 1727–1740.
- [59] W.T. Sturges, L.A. Barrie, Lead 206/207 isotope ratios in the atmosphere of North America as tracers of US and Canadian emissions, *Nature* 329 (1987) 144–146.
- [60] P.I. Ritson, B.K. Esser, S. Niemeyer, A.R. Flegal, Lead isotopic determination of historical sources of lead to Lake Erie, North America, *Geochem. Cosmochim. Acta* 58 (15) (1994) 3297–3305.
- [61] F. Marcantonio, A. Zimmerman, Y. Xu, E. Canuel, A Pb isotope record of mid-Atlantic US atmospheric Pb emissions in Chesapeake Bay sediments, *Mar. Chem.* 77 (2002) 123–132.
- [62] M.M. Larsen, J.S. Blusztein, O. Andersen, I. Dahllöf, Lead isotopes in marine surface sediments reveal historical use of leaded fuel, *J. Environ. Monit.* 14 (11) (2012) 2893–2901.
- [63] T.J. Chow, J.L. Earl, Lead isotopes in North American coals, *Science* 176 (1972) 510–511.