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## Urban soils in a historically industrial city: patterns of trace metals in Pittsburgh, Pennsylvania

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## PAPER

## Urban soils in a historically industrial city: patterns of trace metals in Pittsburgh, Pennsylvania

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**Abstract**

The distribution of legacy heavy metals in industrial city soils is not well documented. Therefore, fundamental details such as the ‘background’ (i.e., non-road/non-dripline) concentration of trace metals in urban soils are uncertain. While there has been a strong focus on mapping lead contamination near roads and residences, these studies are generally not placed in the context of the urban background. In this study, ‘background’ distributions of urban relevant trace metals: arsenic, cadmium, copper, lead, and zinc were mapped based on soil samples collected throughout Pittsburgh. Distinct spatial patterns were revealed: contamination is elevated in the eastern portion of the study area, driven by dominant wind patterns and historical coking activities in low-lying areas (paleochannels), areas subject to atmospheric temperature inversions that focus air contamination. The mixing analysis revealed spatial structures in contributions of industrial activities to metal soil contamination. In particular, regions enriched in cadmium relative to zinc (i.e., Zn: Cd < 317) were located near historical coking operations, and areas enriched in lead relative to zinc (Pb: Zn > 1) were located in areas with historical secondary lead smelters. These results suggest a comprehensive accounting of the trace metals concentrations in background soils has important implications for the assessment of exposure risk in populations residing in historically industrial areas. Relatively sparse sampling of background conditions in urban systems can indicate patterns of legacy contamination and attribute this contamination to historical sources.

**1. Introduction**

The complicated mix of metal inputs to urban soils creates a heterogenous pattern of soil metal concentrations across the city (Pouyat *et al* 2010). In general, the most widely studied urban metal contaminant is lead (Pb). Sources of Pb contamination include residential leaded paint and leaded gasoline. These sources are largely either road-dominated or house-paint dominated and therefore studies concentrate on sampling areas near residences and roads (Mielke 1994, Yesilonis *et al* 2008, Schwarz *et al* 2012, Pouyat *et al* 2015). However, spatial patterns of metal concentrations in areas ‘in between’ the roadside and dripline are not well defined, obscuring the impacts of industrial history on urban soils. To assess this ‘background’ contamination in urban soils it is necessary to sample recently undisturbed sites that are located away from roads and residential pollution inputs. This is difficult since urban soils are repeatedly disturbed during the evolution of urban landscapes. In particular, construction disturbs and alters the soil (e.g. importing materials for fill). These disturbances increase the heterogeneity of soil metal patterns (Pouyat *et al* 2010, Obeng-Gyasi *et al* 2021, Wade *et al* 2021).

In addition to Pb, a wide variety of metals are deposited into urban systems from various human activities - most notably atmospheric deposition of industrial emissions (Pacyna and Pacyna 2001). While roads and driplines are relatively easy to identify from available historical data such as car emissions and leaching of lead paint, industrial plumes distribute metals over broad spatial areas due to the smokestacks geometry and emissions higher in the

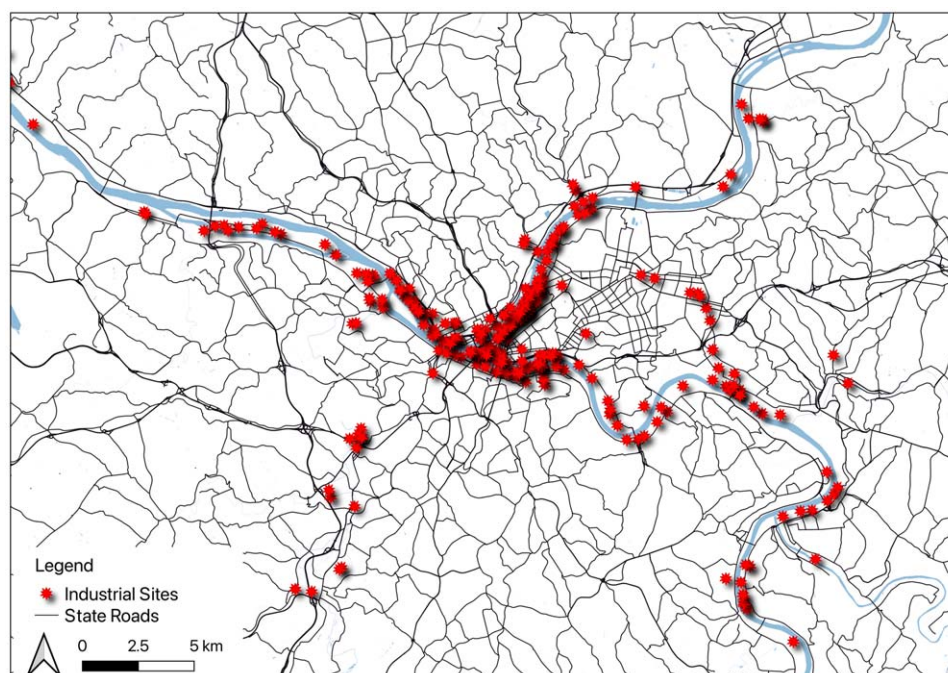


Figure 1. Industrial facility sites (depicted as red stars) adapted from Rivers of Steel (Baraff and Emig 2019).

atmosphere travel a further distance. Patterns of these dispersed metal inputs to urban environments are poorly constrained. Historically Pittsburgh was a center of industrial activity, particularly steel production (Tarr 2003), and industrial facilities lined the Allegheny and Monongahela Rivers (figure 1). These historical activities likely contributed a unique mix of various metals to Pittsburgh soils. Examination of ‘background’ patterns is fundamental to assessing the risks and impacts associated with this legacy industrial contamination.

Additionally, terrain and weather patterns likely influence these contamination patterns. Pittsburgh’s rugged terrain interacts with industrial atmospheric emissions to create consistent patterns of particulate matter (Wittig *et al* 2004, Bein *et al* 2007). Pittsburgh is cross-cut by ancient river channels that were the paths of the paleo- Monongahela and Allegheny rivers as they adjusted to the extreme water and sediment fluxes associated with glacial melt (Leverett 1934). These paleochannels are prominent flat zones located in the eastern portion of the study area (figure 2). Moreover, temperature inversions play a dominant role in the distribution of air contaminants in Pittsburgh (Davidson 1979). Temperature inversions are a reversal of the natural pattern of hot and cool air in the atmosphere where the hotter air is situated closer to the ground while cooler air is located above (Wallace and Hobbs 2019). This reversal of air temperatures suppresses airflow upwards and restricts atmospheric pollutants to low-lying areas. This concentration of atmospheric pollutants for extended periods of time has the potential to focus and encourage deposition of this contamination on the land surface.

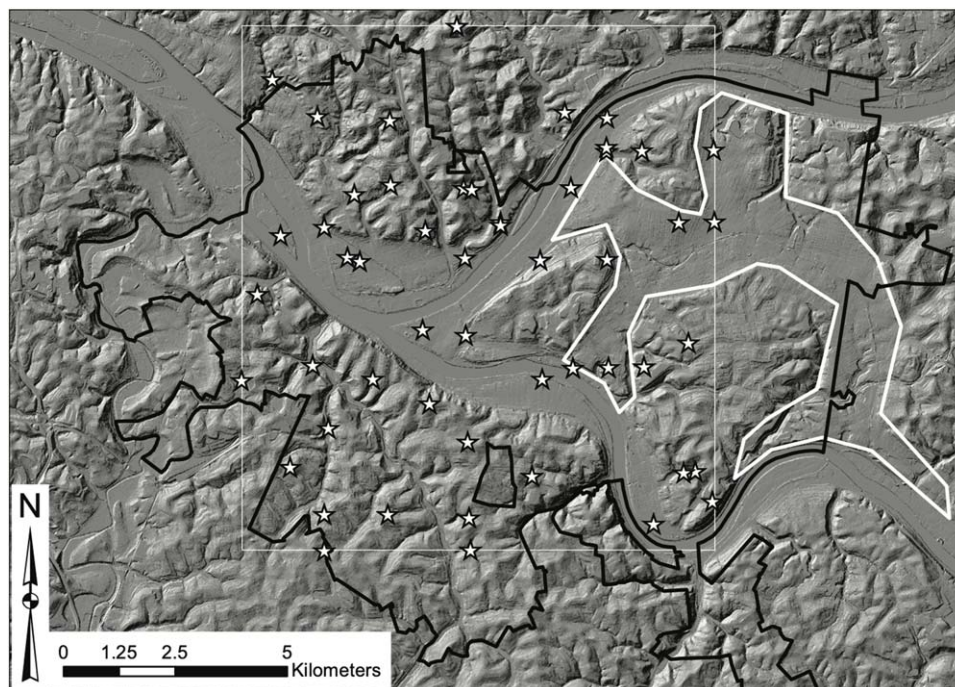
Documentation of ‘background’ soil metal contamination at the city scale is crucial to reveal the legacy effects of industrial pollution on humans and urban ecosystems. Moreover, documentation of interactions between topography and atmospheric deposition is fundamental to the prediction of these patterns in cities across the globe. Specifically, emerging redevelopment tools including urban gardening and green infrastructure have the potential to remobilize soil contamination, increasing human and ecosystem exposure where As, Cd, Cu, Pb, and Zn metals can have a negative impact on human health (Nriagu 1980a, 1980b, 1981, 1983).

In this article, we combine previously separate approaches (spatial and mixing analyses) to define patterns of soil metal chemistry in Pittsburgh. Moreover, this approach examines concentrations of multiple metals in urban systems, areas typically examined for single contaminants. Together, these perspectives allow evaluation of the hypothesis that ‘background’ total soil concentrations of As, Cd, Cu, Pb, and Zn are substantial in historically industrial cities like Pittsburgh. Further, the mixing approach allows clarification of relative source contributions to this legacy metal contamination.

## 2. Methods

### 2.1. Site selection, soil sampling, and chemical analysis

A portion of Pittsburgh (figure 2) was divided into a grid and a random number generator was used to select site locations in each grid cell (Schumacher and Barnett 2016). Soil samples were only collected from locations that



**Figure 2.** Hillshaded digital elevation model of Pittsburgh, PA (thick black line); the paleochannels (thick white outline) are predominantly located between the two major rivers in the eastern half of the map. The extent of the sampled area is shown with a thin white line. Elevation Data: (Allegheny County Division of Computer Services Geographic Information Systems Group 2017). All sampling locations are shown with white stars.

met the following criteria: (1) were without evidence of recent land disturbance, (2) were not near roads or residences, and (3) were specifically not in private yards or industrial lots. If a site was not suitable, other random locations in the grid cell area were assessed sequentially until a suitable site was located.

A total of fifty-six soil samples were collected (figure 2). A 3–5 sample composite from the top 5 cm of soil was collected from the selected locations, about 128 grams of material for each site. GPS coordinates were recorded for each site. Samples were collected by Carnegie Mellon University students under the supervision of the Allegheny County Conservation District (ACCD). Upon return to the lab, the soil was air-dried, ground, sieved with a 2 mm mesh, and mixed. Soil samples were then pulverized using a ball mill with a tungsten carbide bomb before chemical analysis.

Powdered samples were sent to ALS Global (Reno, NV, USA) to measure total metal concentrations using service code ME-MS61 which uses a ‘four acid digestion [to] quantitatively dissolve nearly all minerals in the majority of geological materials’ with an ICP-MS measurement (ALS Global 2022).

## 2.2. Data cleaning

A data quality check was conducted to ensure the dataset was free from typos and/or GPS errors by cross-checking site location addresses with collected GPS coordinates and field notes. This data quality check identified the following: (1) sites 4 and 45 had identical GPS locations, the GPS coordinate was deemed correct for site 4 based on field notes, and site 45 was relocated using the address in the field notes, confirmed by elevation data; (2) site 117 had an obvious typo number error in the GPS coordinates that were fixed to match field note location details; (3) sites 137, 104A, and 192 required removal from analysis due to the inability to reliably confirm their GPS coordinates; (4) site 19B was removed due to a lack of GPS coordinates. Based on the data quality check, 3 sites were adjusted and 4 sites were removed leaving a total of fifty-two sites for analysis.

## 2.3. Soil metal concentration pattern interpolation

Continuous patterns of ‘background’ soil metal concentrations were inferred from the sample concentrations ( $n = 52$ ) using ordinary kriging (Geospatial Analyst tool, ArcGIS 10.x). Simpler kriging approaches were preferred and the assumptions about the mean distribution in ordinary kriging seemed more appropriate for the soil data than those in simple kriging. For kriging, all data were log-transformed, except for As. Stable models were used and standard neighborhood (2–5 neighbors).



**Table 1.** Descriptive statistics for metal concentrations measured in 'background' Pittsburgh soils ( $n = 52$ ).

Metal	Mean ( $\text{mg kg}^{-1}$ )	Standard Deviation ( $\text{mg kg}^{-1}$ )	Minimum ( $\text{mg kg}^{-1}$ )	Median ( $\text{mg kg}^{-1}$ )	Maximum ( $\text{mg kg}^{-1}$ )
As	14.9	4.51	6.00	13.9	27.3
Cd	1.85	6.77	0.19	0.80	49.6
Cu	52.9	30.4	20.3	38.4	158
Pb	146	115	27.9	99.0	619
Zn	269	185	89.0	204	1050

## 2.4. Evaluation of source contributions to background soil metal chemistry

The relative contributions of contamination sources to soil metal mixes were evaluated by plotting soil metal concentrations in element versus element scatter space and comparing individual samples to endmember chemistries. Endmember chemistries were identified based on Pittsburgh's environmental history (Tarr 2003). Literature measurements of these end members were compiled and used to evaluate source contributions to observed soil chemistries (citations are shown in table 2).

## 3. Results

### 3.1. The distribution of soil trace metals across the study area

Soil metal concentrations vary widely across the study area (table 1). Much of the variation in Cd is driven by a single outlier from Brunot Island near the western edge of the study area. While this value is shown in figure 3, for the remainder of the analyses, this value is not included. In general, higher concentrations of metals were observed in the eastern half of the study area (figure 3). However, As concentrations are more evenly distributed relative to other metals. The kriged surface visualizations suggest more heterogeneous patterns in the eastern region of higher concentrations, particularly in Pb and Cd. In particular, there seem to be wider areas of relatively elevated metal concentration and higher metal concentrations in the southeastern corner of the study area. Again, both Pb and Cd are elevated in this region (Maxim 2022).

While kriging can provide valuable insight about spatial patterns, it is important to carefully consider the uncertainties in kriged surfaces (figure 4). These uncertainties are consistent with the patterns of concentration, with uncertainty in the eastern half of the study area relatively higher. The kriging approach was able to predict metal concentrations in the eastern, lower metal concentration regions, and the uncertainty increases as higher metal concentrations are encountered. That said, uncertainties (figure 4) are small relative to the predicted concentrations (figure 3).

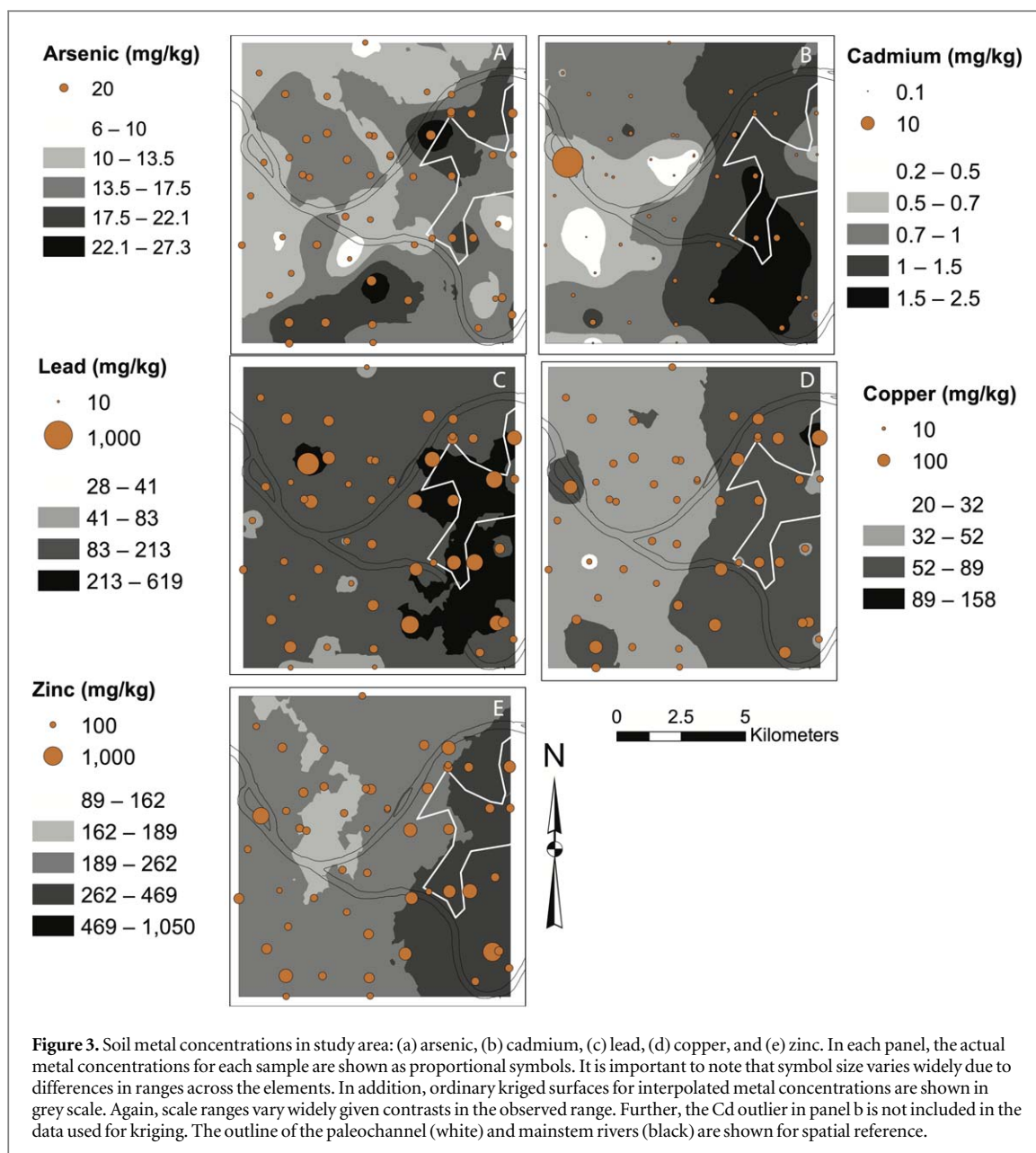
### 3.2. Contributions of metal contaminant sources to soil patterns

In Pittsburgh, there were a wide variety of historical contaminant sources, so the attribution of metals in the soil concentration mix to specific sources is a challenge. We identified five relevant chemistry sources that bracket observed soil chemistries: fly ash from the coking process, the bulk chemistry of coal, bedrock, fly ash from coal-fired power generation, and secondary lead smelting (table 2).

Uncontaminated soils in Pittsburgh are expected to reflect parent material trace metal chemistry. While Pittsburgh bedrock is a mix of limestone, sandstone, and shale, shale tends to dominate. We could not identify a survey of Pittsburgh area shale chemistries, so used a global average shale chemistry (Turekian and Wedepohl 1961). In several cases, the shale line defines a primary chemistry boundary (e.g., in all Pb plots, Pb is in excess of the shale relative to the other metals). In other cases, chemistries deviate from the shale chemistry toward distinct elemental ratios (e.g., in the Cd versus Zn plot, chemistries plot on both sides of the lines, some closer to coal fly ash, others closer to coke fly ash/bulk coal chemistry/lead smelter). Therefore, these soils seem to be impacted by distinct sources of contamination.

For all metals, except for As, identified endmembers mostly bracket observed soil concentrations. In the case of As, samples plot in regions where Cd and Zn are more enriched relative to As than any end members we identified. These are not likely to be pure inputs of Cd or Zn, as there is no clear evidence of pure Cd/Zn inputs in the other element pairs. This pattern may result from As mobilization from the soil in cases where the other metals are more prone to stay sorbed in the soil environment. Therefore As is not likely a species that lends itself to this approach. The uniqueness of As chemistry in the element/element plotting space is consistent with the observed spatial patterns in As (As was the only metal to not be log-normally distributed and is not clearly enriched in the eastern portions of the study area).

Interpretation of the metal mixing analysis (figure 5) is discussed below.

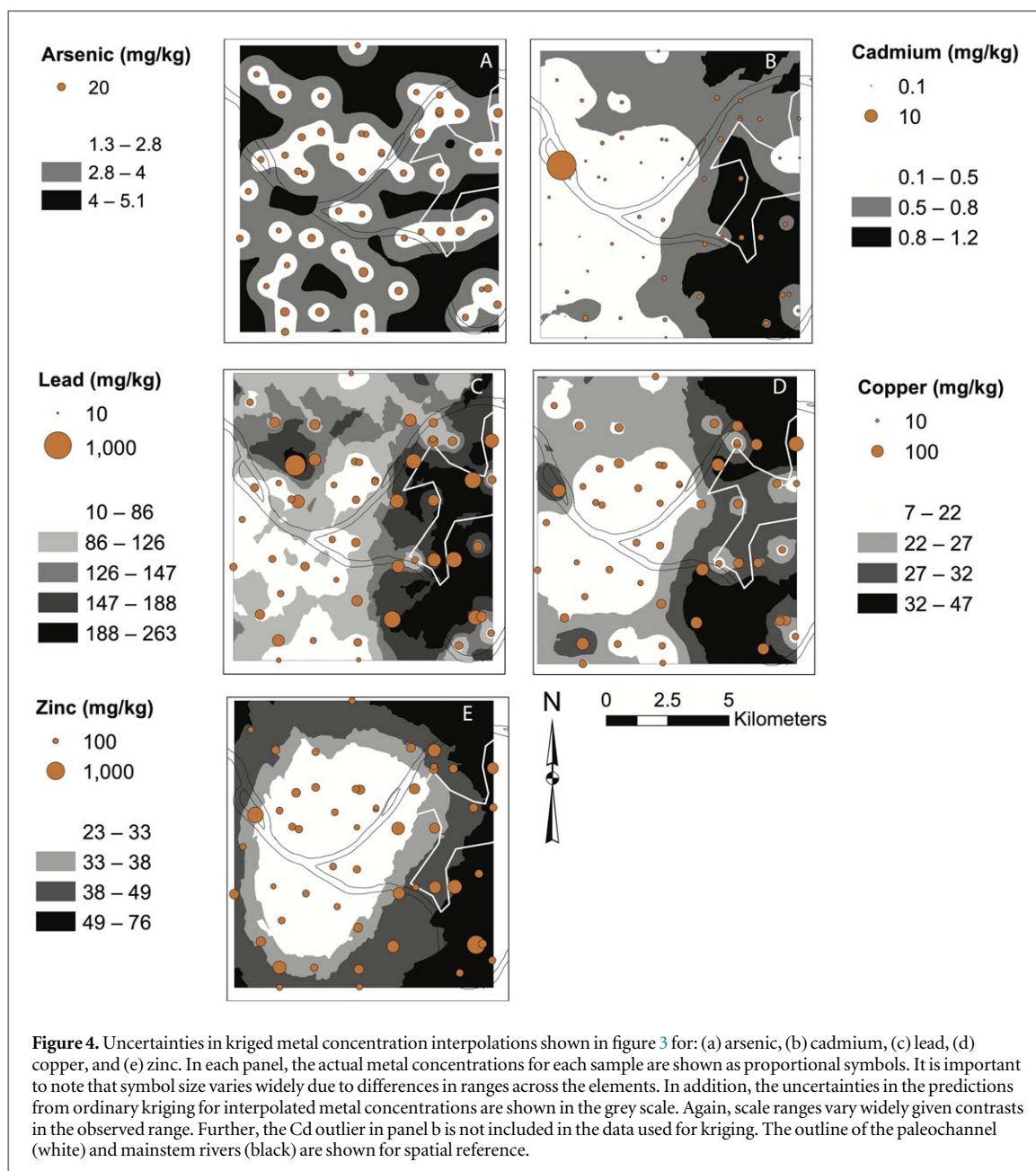


## 4. Discussion

### 4.1. Soil metal enrichment concentration patterns

We expect contamination hotspots in Pittsburgh should reflect historical distributions of industrial facilities. Industrial facilities were located largely along the Monongahela, Allegheny, and Ohio Rivers with the highest concentration of facilities at the confluence of the Monongahela and Allegheny; yet, the patterns observed in ‘background’ soil metals do not correspond to the historical distribution of industrial facilities (figure 1). Part of this is undoubtedly the transport of airborne emissions once contamination leaves the smoke stack. In addition, the emissions from these industrial sites likely varied widely as a function of facility size, longevity, and emission controls. However, there is no authoritative reconstruction of this emissions history.

Soil metal enrichment patterns seem to be driven by the dominant wind pattern. Further, interactions between topography and atmospheric thermal temperature inversions seem to also influence soil metal concentration patterns. Metal (Pb, Cd, Zn, and Cu) concentrations are elevated in the eastern portion of the study area (figures 3(b)–(e)), consistent with dominant winds from the southwest. The consistency among these trace metal patterns suggests a common mechanism. However, the finer-scale patterns of enrichment in Pb, Cd, and Zn in the southeastern portion of the study area (figures 3(b)–(e)) suggest interactions with orography (terrain). In particular, the former Jones and Laughlin (LTV) steel mill was located roughly in the southern



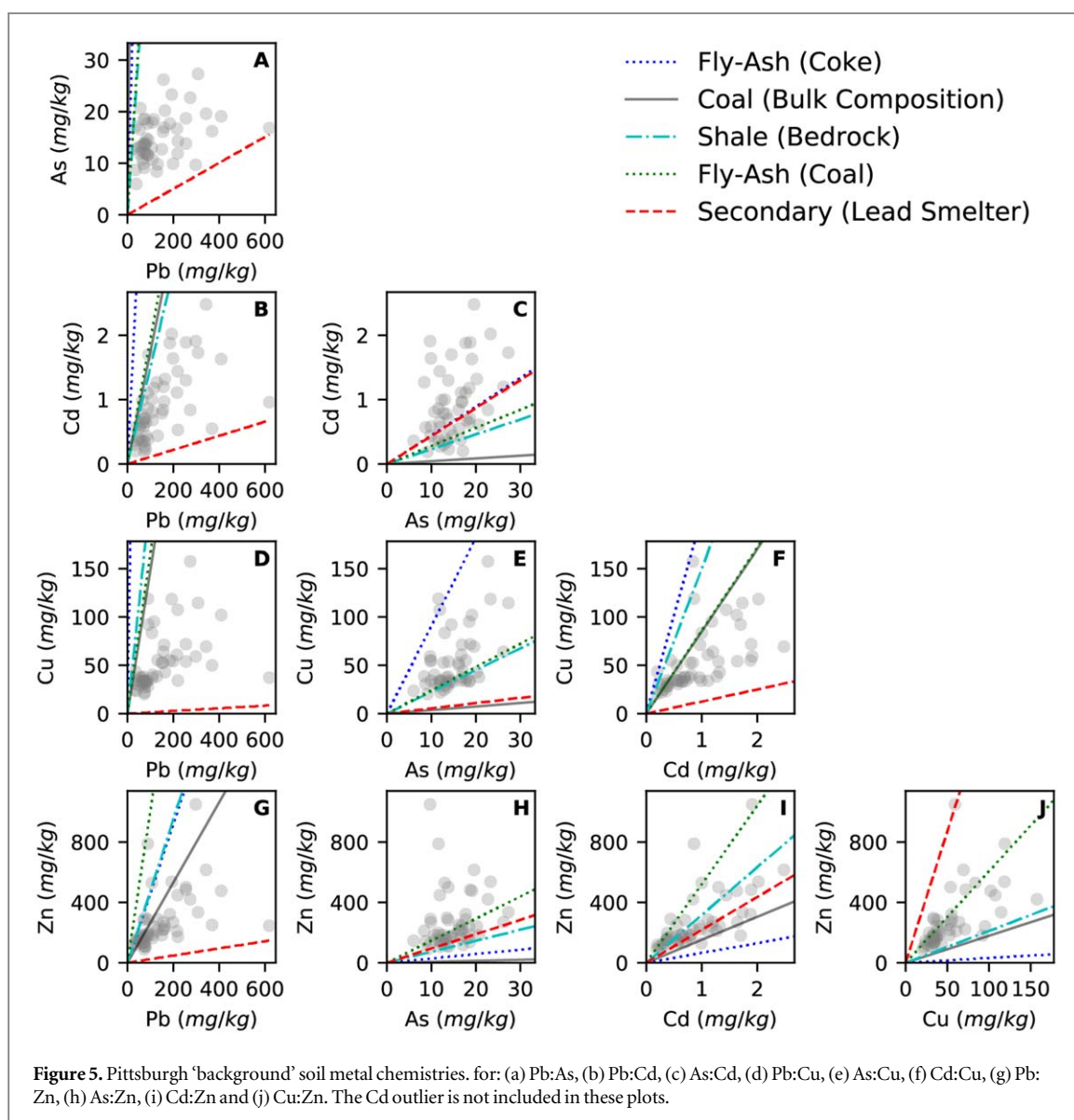
‘mouth’ of the paleo channel, directly west of a topographic high and at the western extent of these elevated metal concentration regions. We interpret these predicted surfaces to have resulted from the orographic forcing of plant emissions and resultant metal deposition on the hillslope and topographic high directly east of this facility.

In addition to interactions between wind patterns and topography, there is also a distinct secondary pattern in the concentrations. There is a north-south band of elevated soil metal contributions, particularly in Cd and Pb (figures 3(b) and (c)). These patterns follow topography that would not be consistent with the interactions between wind and topography discussed in the previous paragraph. In particular, the relatively low-lying paleochannels seem to have elevated concentrations despite not being directly downwind of industrial facilities and not creating orographic forcing. These patterns in some cases seem to follow the paleo-channel valleys, particularly Pb and Zn (figures 3(c) and (e)). Historically, the valleys were areas that received substantial atmospheric contamination during atmospheric temperature inversions (Davidson 1979). This suggests that atmospheric deposition of metal contaminants would also be higher in these low-lying areas due to repeated temperature inversions. That is, when temperature inversions occur, atmospheric pollutants from these industrial areas were concentrated in these paleochannels areas and contaminants in this pollution had extended periods to be deposited on the valley surfaces. This possibility is examined in section 4.3 combining the spatial and mixing analyses.

**Table 2.** Source material elemental ratios.

Element	Fly-Ash (Coke) (mg kg <sup>-1</sup> ) adapted from table 3 in (Mu <i>et al</i> 2012)	Coal (bulk composition) (mg kg <sup>-1</sup> ) adapted from COALQUAL database (Palmer <i>et al</i> 2015)	Shale (bedrock) (mg kg <sup>-1</sup> ) adapted from table 2 in (Turekian and Wedepohl 1961)	Fly-Ash (Coal) (mg kg <sup>-1</sup> ) adapted from table 1 in (Gieré <i>et al</i> 2003)	Secondary (Lead Smelter) (mg kg <sup>-1</sup> ) adapted from table 1 in (Rieuwerts and Farago 1996)
As	47.58	20.86	13	42.7	460
Cd	2.12	0.09	0.3	1.2	20
Cu	431.86	7.65	45	103	250
Pb	29.96	5.16	20	62	18,296
Zn	137.79	13.71	95	625	4,360



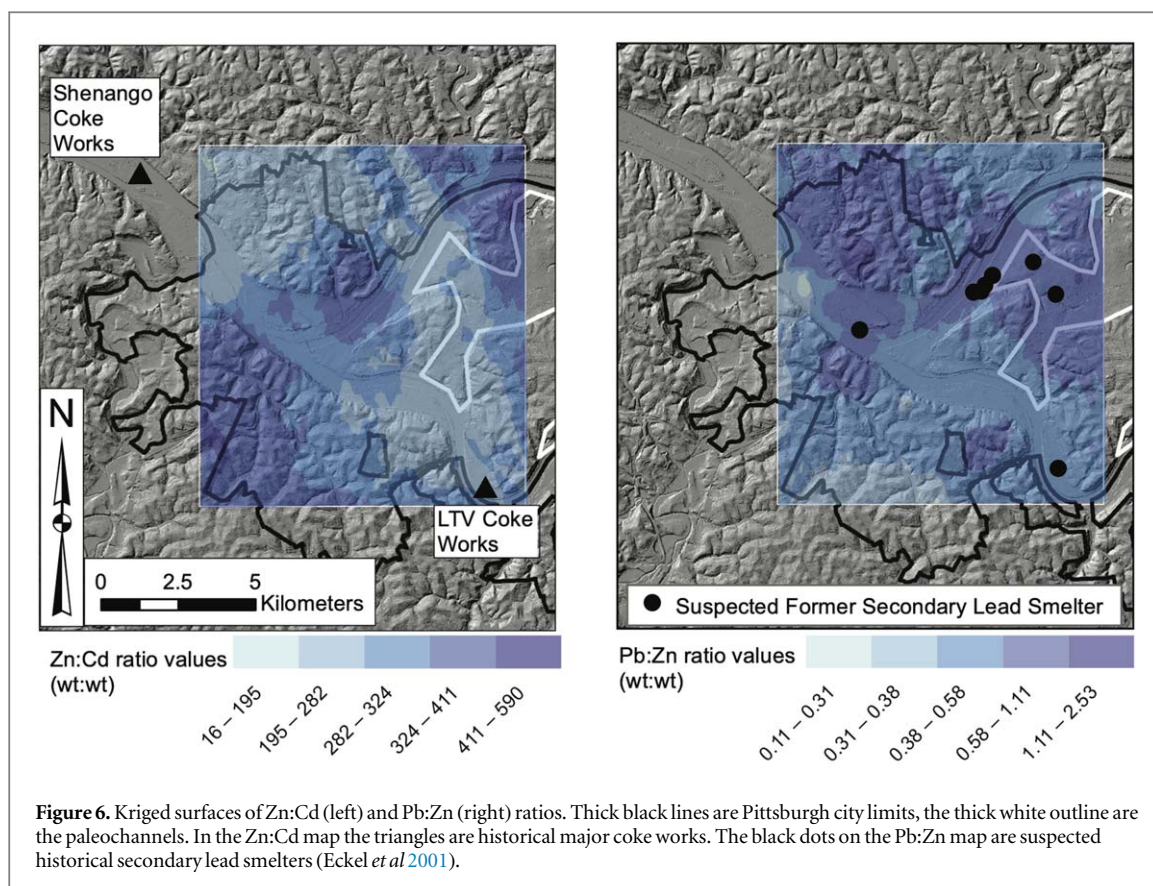


#### 4.2. Metal contaminant sources to the metal pattern

Potential metal contamination source influence on soil chemistry varied with the element. As noted previously, arsenic is not considered here due to the failure to identify appropriate end members. Further, the Cd outlier is not considered in the analyses below. This section discusses the sources influencing soil metal mixtures.

Soil lead seems to be strongly influenced by secondary Pb smelting, as secondary lead smelting emission chemistry generally defines a boundary well beyond the extreme values observed in Pittsburgh soils (e.g., see the figures 5(b), (d), and (g)). Others have suggested these secondary smelting sites were important for background soil chemistry in cities, including Pittsburgh (Eckel *et al* 2001). However, it is not possible to unambiguously identify smelting as the lead source. In particular, lead gasoline additives would be a relatively pure source of the lead (i.e., there is almost no Zn, Cd, or Cu in tetra-ethyl lead) and could cause similar behavior in the element-element plots. Care was taken to ensure near-road environments were not sampled, avoiding hot spots of lead contamination, but we cannot rule out the potential that this historically dominant source impacted broader areas with the element-element mixing data alone.

Cadmium and zinc seem to be influenced by multiple sources. Cadmium is also influenced by lead smelting (e.g., consider the strong deviation toward the lead smelting line in figure 5(f)). However, both coking and general coal combustion (fly ash) contribute Cd to Pittsburgh soils. For example, in the Zn versus Cd plot, you can see deviations from the shale line toward both the coking and coal fly ash lines (in opposite directions). While lead smelting can sometimes be important to Zn chemistry (e.g., the sample falling on the smelter line in figure 5(j)), coal combustion values seem more important to the relative mix of Zn and Cu. As noted for Cd, coking does seem to influence Zn content relative to Cd.



Finally, Cu is the least clearly influenced by these potential end members. In several cases (e.g., figures 5(j) and (f)) values far near or around the coal fly ash line. However, there is limited evidence shale chemistry is an important contributor to observed chemistries. Some samples plot near the line in figure 5(j), but most are clearly enriched in Zn. This is not surprising as Cu has the fewest clear potential sources in Pittsburgh's industrial history.

#### 4.3. Spatial patterns in soil metal mixes

Given the apparent mixing in the element-element plots, the spatial patterns of the relative influence of different sources can be explored. For example, in figure 5(i), Cd:Zn chemistries deviate away from the shale line toward coal fly ash (enriched in Zn) and coking emissions (enriched in Cd). The shale line is a slope that represents the global average ratio of Zn:Cd ( $\sim 317$ ). The kriged surface of Zn:Cd ratios in the Pittsburgh background soil chemistry can reveal the relative influence of the different sources. Lower ratio values are more influenced by coking, and higher values by coal combustion (figure 6).

Historically, there were three major coke works in the City of Pittsburgh: Shenango, LTV (Jones and Laughlin), and Clairton (prior to 1930, during the dirtiest industrial periods, most of the coke production feeding regional steel production occurred in the Connellsville Coking District) (American Industrial Heritage Project 1992). The LTV and Shenango works were located in or upwind (respectively) of the study region (figure 6). Zn:Cd ratios suggesting coking influence (i.e., those below a value of 317) are associated with these coke works. The regions with relative Cd enrichment in the northwest corner are directly downwind of the historical location of the Shenango works. The Cd enriched region in the eastern half of the study region surrounds the historical location of the LTV works. This pattern seems distinct from the pattern downwind of the Shenango works, particularly the extension northward. The areas that seem to be influenced more heavily by coal fly ash-like chemistries are largely upland zones relatively removed from coking activity. While we cannot rule out similar patterns in Cd enrichment north of the Shenango works with available data, the juxtaposition of the Cd enrichment near the LTV works with the paleochannel location is particularly striking. A major facility seems to contribute Cd to background soil mixes well north of downwind areas. The sampling density here is not sufficient to unambiguously evaluate the role of the paleochannels in the deposition of industrial emissions, but these data (consistent between emission type and topography) provide a strong basis that paleochannel influence is important.

Beyond the coking and coal combustion end members, it is also important to understand the sources of background Pb and evaluate the role of secondary smelters. Using the Pb:Zn ratio, we can evaluate the spatial pattern of Pb enrichment (figure 6). One concern is that the influence of tetra-ethyl lead could also appear as Pb enrichment. The kriged Pb:Zn surface (i.e., higher values indicate more Pb enrichment) suggests this may not be the case. Historical vehicle hubs (e.g., downtown Pittsburgh, riverbanks) are not consistently Pb enriched, rather the enrichment occurs predominantly in the northern half of the study area and in relatively rugged terrain. The patterns of Pb enrichment correspond surprisingly well to suspected historical secondary lead smelters (Eckel *et al* 2001). In particular, the suspected smelter in the central-western portion of the study area is surrounded by a region of relative Pb enrichment, and the cluster of suspected smelters in the upper eastern portion of the study area is similarly surrounded by areas of Pb enrichment. However, it is difficult to explain the lack of enrichment in the upper portion of the study area, distant from suspected smelters. It is important to note this region has relatively less soil Pb (figure 3) and the one outlier in the region may influence the kriging results. We cannot clarify this with existing data. The other inconsistency is the lack of Pb enrichment around the suspected secondary smelter in the southeast corner of the study area. In related work, we evaluated the origins of this smelter and it was originally a tin smelter (The American Iron and Steel Association 1904). One of the challenges in evaluating these locations is production and practice are not well documented. If this smelter processed tin for the majority of its production history, Pb enrichment would be diminished. Further, given these soil mixtures are influenced by multiple sources, if a relatively more Zn-rich source (e.g., coking) influenced this area, the Pb:Zn would also be diminished. Finally, while we do not expect relatively historical zinc smelting (Rossi *et al* 2017) to influence these soil mixes, we cannot rule it out. Zinc-rich smelting emissions could obscure the impact of the secondary lead smelter in the southeast portion of the study area.

## 5. Conclusion

This examination of 'background' soil chemistries reveals unexpected patterns. While few of the soil samples collected here exceed soil metal concentration action levels, these patterns are vitally important for understanding human and ecosystem risks. We tend to focus on known sources (drip lines and road edges) during risk assessment. A combination of these known sources with background industrial contributions may create unexpected risk scenarios that are important to the protection of human and ecosystem health. This relatively sparse sampling of undisturbed soils can provide important insight into the spatial patterns of historical inputs, patterns consistent with known history, adding credence to the need to incorporate these inputs into our ongoing considerations of urban soil metal mixtures.

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## Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: <https://doi.org/10.26022/IEDA/112208>.

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