



# Urban legacies and soil management affect the concentration and speciation of trace metals in Los Angeles community garden soils



Lorraine Weller Clarke <sup>a,\*</sup>, G. Darrel Jenerette <sup>a</sup>, Daniel J. Bain <sup>b</sup>

<sup>a</sup> Department of Botany and Plant Sciences, University of California, Riverside, CA 92521, USA

<sup>b</sup> Department of Geology and Planetary Science, University of Pittsburgh, Pittsburgh, PA 15260, USA

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## ABSTRACT

Heavy metals in urban soils can compromise human health, especially in urban gardens, where gardeners may ingest contaminated dust or crops. To identify patterns of urban garden metal contamination, we measured concentrations and bioavailability of Pb, As, and Cd in soils associated with twelve community gardens in Los Angeles County, CA. This included sequential extractions to partition metals among exchangeable, reducible, organic, or residual fractions. Proximity to road increased all metal concentrations, suggesting vehicle emissions sources. Reducible Pb increased with neighborhood age, suggesting leaded paint as a likely pollutant source. Exchangeable Cd and As both increased with road proximity. Only cultivated soils showed an increase in exchangeable As with road proximity, potentially due to reducing humic acid interactions while Cd bioavailability was mitigated by organic matter. Understanding the geochemical phases and metal bioavailability allows incorporation of contamination patterns into urban planning.

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## 1. Introduction

Heavy metal contamination in urban soils threatens human health in densely populated metropolitan regions (Yesilonis et al., 2008; Nicholson et al., 2003; Diamond and Hodge, 2007). The majority of metal contamination in cities is anthropogenic, due to legacies of air and soil pollution (Nazzal et al., 2013; Mielke et al., 1983; Charlesworth et al., 2011). Exposure to metals may be higher for urban gardeners and their family, as they are more exposed to contaminated dust in the environment and on edible crops, which may also incorporate soil metals into their tissue (Finster et al., 2004; Saumel et al., 2012; Khan et al., 2008; Attanayake et al., 2014; EPA, 2013). Community gardens, a common form of urban agriculture, are often established in unmanaged or abandoned housing lots, where soil metals may not have been characterized before cropping begins (Finster et al., 2004; Chaney et al., 1984; Lawson, 2007). Exposure to metals associated with urban gardening has been well documented, but sources of metal contamination and how horticultural practice affects metal presence and bioavailability are less clear (Finster et al., 2004;

Intawongse and Dean, 2006). Understanding the geochemical phases of metals and environmental factors affecting them may facilitate improved management of exposure risks (Schwarz et al., 2012; Virtanen et al., 2013; Mossop and Davidson, 2008).

This study focuses on three metals: Pb, As, and Cd. These are all potentially toxic and associated with urban system pollution, though their distribution and source materials are varied based on the metal. Metal contamination relative to roads is relatively well recognized (Yesilonis et al., 2008; Zhang et al., 2006; Zhao et al., 2011), and may accumulate over time. In addition, land use and management activities may influence metal concentrations, like the use of contaminated building materials, changes in OM, and previous agricultural use.

The health implications of elevated Pb concentrations in urban ecosystems are well studied, due to its toxicity to children and persistence in soils (Mielke et al., 2010; Wang et al., 2006). Leaded paint and gasoline are the primary sources of Pb contamination, both in widespread use from 1884 (paint) or 1920 (gasoline) until the early 1980s (Boreiko and Battersby, 2008; Kerr and Newell, 2003). Though mean Pb background concentrations in CA soil are near 25 mg/kg (Bradford et al., 1996), urban soils often contain 200–1500 mg/kg Pb (McClintock, 2012; Mitchell et al., 2014). The California human health screening level (CHHSL) for Pb in residential soils stands currently at 80 mg/kg, the concentration at

\* Corresponding author.

E-mail addresses: [Lorraine.clarke@ucr.edu](mailto:Lorraine.clarke@ucr.edu) (L.W. Clarke), [darrel.jenerette@ucr.edu](mailto:darrel.jenerette@ucr.edu) (G.D. Jenerette), [dbain@pitt.edu](mailto:dbain@pitt.edu) (D.J. Bain).

which exposure can lead to a 1  $\mu\text{g}$  change in blood Pb levels (Carlisle, 2009). This policy mandated limit changes frequently and differs among governmental agencies and jurisdictions. In 2011, the California Water Quality board had Pb limits set at 200 ppm (recently changed to the stricter limit), and the US EPA limit for residential soils is set at 400 ppm (EPA, 2001), making it difficult to establish “safe” levels of soil Pb in residential or agricultural soils.

Arsenic concentrations are elevated in CA soils relative to other US states (CA background: 2–40 ppm, median: 5 ppm [Diamond et al., 2009]). Residential soil As commonly exceeds recommended CA (0.39 ppm) (OEHHA Risk Assessment-Soil and Gas) and Federal (0.27 ppm) environmental screening levels (Duvergé, 2011). There is no federal safety limit for As concentrations in soils and clean-up is focused on areas with high exposure risks (Baldwin, McCleary, 1998). The main anthropogenic transport mechanisms for As are deposition from air pollution or leaching from source materials, like chromated copper arsenate (CCA) treated wood (Diamond and Hodge, 2007; Charlesworth et al., 2011). Though phased out of treated lumber in the last 20 years, the use of CCA wood to line garden plots, or as a legacy from previous building materials, may enrich As concentrations in soils and crops (Stilwell et al., 2006; Zagury et al., 2003; Cao and Ma, 2004).

Cadmium is used as a stabilizer in tires and is also present in automobile exhaust, leading to roadside pollution (Ellis and Revitt, 1982). Mineral phosphorous fertilizers were historically high in Cd, imparting legacies of contamination in previous agricultural regions (Cullen and Maldonado, 2013; Lopes et al., 2011). Median soil Cd concentrations in California are low,  $\sim 0.36$  mg/kg, and globally soil concentrations average 1.1 mg/kg (Bradford et al., 1996; Alloway, 2013). The CHHSL for Cd in soils is 1.7 mg/kg, based on a carcinogenic potency factor for inhalation (OEHHA Risk Assessment-Soil and Gas), though even smaller concentrations may be taken up by crops (Murray et al., 2011).

Metals are mobile and geochemical fractionation is heterogeneous at soil particle scales and influenced by site-specific geography and management factors (Burt et al., 2014; Grzebisz et al., 2002). Soil metals can be associated with operationally defined biogeochemical phases including exchangeable, reducible (metal-oxide bound), oxidizable (bound to organic matter), and in relatively recalcitrant residual fractions (Mossop and Davidson, 2008). Metals in the exchangeable phase are immediately bioavailable, while changes in soil characteristics can move reducible metals into the exchangeable fraction (Saumel et al., 2012). Organic matter (OM) additions can reduce Cd bioavailability, as it can shift concentrations from exchangeable to less accessible reducible or organic phases (Cullen and Maldonado, 2013; Sun et al., 2013). Conversely, high OM and associated humic acids may react with As to shift it from metal oxide-bound to exchangeable fractions (Murray et al., 2009; Wilson et al., 2010). The pH of soils interacts with OM, particularly for Pb, as it is most likely to form organic complexes at increased pH levels (Alloway, 2013; McClintock et al., 2013). Information about metal fractions can aid garden managers in reducing personal exposure (Scheckel et al., 2009), either by altering soil chemistry through adding OM (Sloan et al., 2012) and changing pH (Saumel et al., 2012; Murray et al., 2011) or planting crops less likely to accumulate metals, like fruits (Hough et al., 2003).

Here we characterize environmental and management processes influencing presence and geochemical fractionation of metals in community garden soils of Los Angeles, the second most populous city in the United States. We focus on two questions: *What factors influence the patterns of Pb, As, and Cd concentrations across community gardens in Los Angeles and do these factors vary among metals?* and, *How do management and soil characteristics influence bioavailability of Pb, As, and Cd and do these differ among*

*metals?* We sampled soils in and around twelve community gardens in Los Angeles County, CA and measured Pb, As, and Cd concentrations and their distribution among geochemical phases. We compared concentrations with age of neighborhood, distance from road, and OM content. Our targeted analysis of geochemical phases in managed and unmanaged soils in our study allows for characterization of interactions among management, legacies, and environmental factors. Understanding the interactions among abiotic and socio-ecological urban processes in soils is fundamental for diminishing heavy metal exposure risk and sustaining quality of life in urban ecosystems (Saumel et al., 2012).

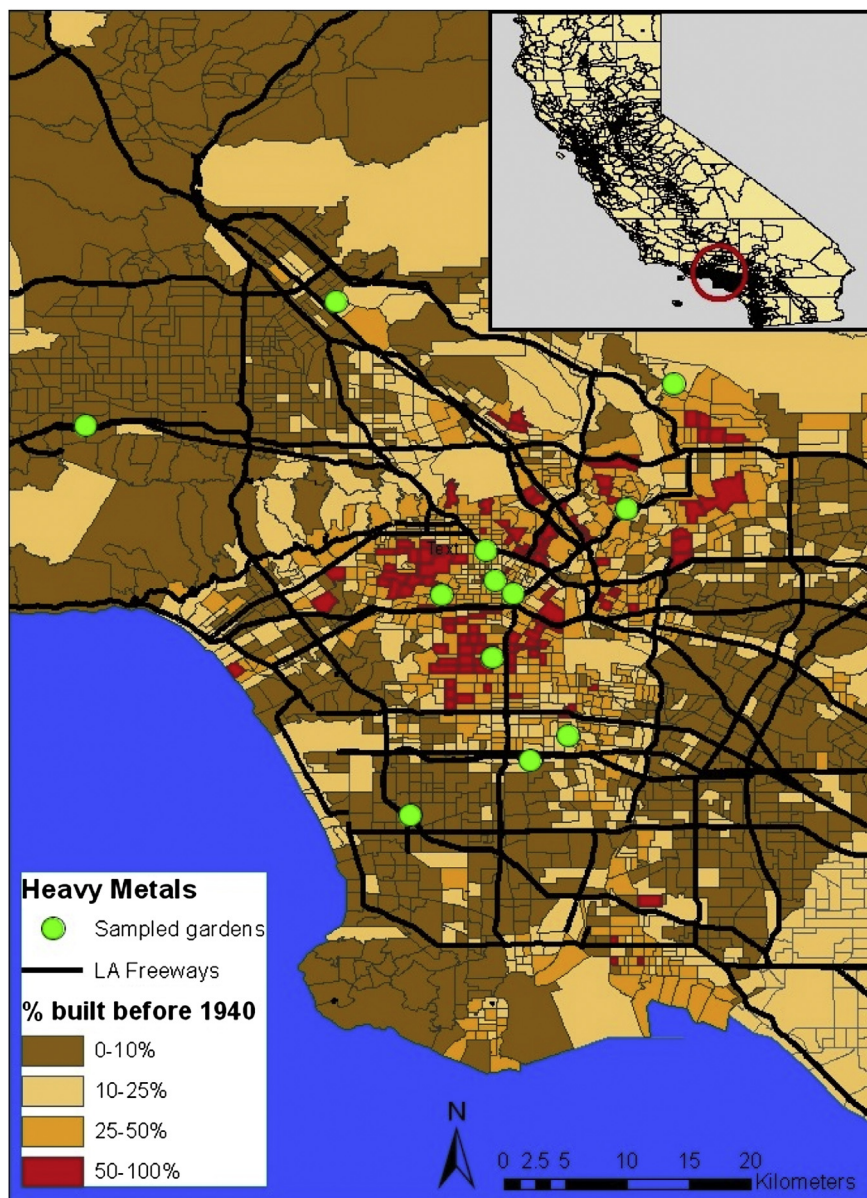
## 2. Methods

### 2.1. Study region

Our study focused on twelve community gardens across Los Angeles County, California, USA, most near downtown Los Angeles (LA) (Fig. 1). LA has a long legacy of urban development; it was officially founded in 1781 and by 1900 it was a major metropolis with over 100,000 people (Rios Bustamante). LA County covers over 10,000 km<sup>2</sup> with approximately 9.8 million residents, (2010 U.S. Census). The majority of LA's residential neighborhoods were built during two periods (1950–1965 and 1975–1990), the first period following cessation of commercial agriculture within LA city boundaries (Surls, Gerber, 2010) and the second period resulting from rapid immigration from Eastern states and Latin American countries (Terwilliger, 2006). Currently, LA contains seven major freeways, each with annual average daily traffic of >300,000 vehicles (LA County Almanac/CalTrans 2005).

### 2.2. Data collection

Soil samples were collected at gardens between June and September in 2011 and 2012, periods of peak crop growth. Gardens were chosen from an initial pool of 25 random locations, and included in the study based on manager participation. These gardens ranged in size between 400 and 10,000 m<sup>2</sup>, and had between 12 and 140 garden plots (Clarke, Jenerette *In Review*). Neighborhoods near gardens were built between 1895 and 1985 and land-use backgrounds of gardens included previous homes, agriculture, or vacant soil (Fig. 1) (Clarke, Jenerette *In Review*). Soils were collected to 15 cm, the effective depth of crop roots (Brady and Weil, 2002), using a soil borer, in three micro-environments (Table 1): garden plots (“cultivated”; 5–10 per garden), uncultivated soil inside the garden (“inside uncultivated”; 2–3 per garden), and uncultivated soil outside the garden (“outside uncultivated”; 2–3 per garden). Cultivated plots were chosen randomly in seven gardens, and based on individual gardener participation in the remaining five. For these, 5–10 soil subsamples were taken a minimum of 2–3 m away from each other, focusing under crop canopies and the edge, with increased sub-sampling in larger plots, and composited into one sample for each plot. Uncultivated plots were chosen to compare indirect management effects on soil metals (inside) and to ascertain the base level of metals in completely unmanaged soil (outside). For uncultivated areas, a plot of at least 10 m<sup>2</sup> was chosen and at least five soil borer samples were taken and composited into one sample for each location. Uncultivated soil samples within the garden were collected from garden margins and paths. Outside uncultivated soils were collected from minimally maintained public spaces, like public right-of-ways, keeping road exposures similar to each other in each garden. In 18 of the 33 outside soil samples, soil was too compacted to sample to the full 15 cm. In these cases, we cored down to 5 cm and sampled available soil. Due to low sampling, we grouped all



**Fig. 1.** Map of Los Angeles County showing census tract boundaries (grey lines) and percent of structures built before 1940. Brown and beige have few older structures, orange and red have 25% or more. Black lines indicate the location of major freeways in LA. Green dots indicate community gardens where soil samples were collected. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

uncultivated soils into an “uncultivated” category for increased statistical comparison to cultivated soils. In total, samples from 74 cultivated soils, 40 inside uncultivated soils, and 33 outside uncultivated soils were collected (Table 1).

Homogenized soil samples were dried at 70 °C for 72 h and sieved to 10 mm. Organic matter was measured as loss on ignition at 550 °C for 4 h. Soil pH values were measured using a 5:2 water to soil solution. Soil samples were sent to ALS Chemex for measurement of total heavy metal concentrations (ME-MS61), including Pb, As, and Cd.

We completed a sequential extraction, a well-established approach to determine trace metal fractionation (Virtanen et al., 2013; McGrath, 1996), on a subsample of soils to determine metal distribution among geochemical phases. The subsample included 36 cultivated soils, 12 uncultivated garden soils, and 23 uncultivated outside soils (Table 1: total  $n = 71$ ). The modified BCR

sequential extraction procedure, as laid out in Mossop and Davidson (2008), was followed closely. Three extractions were done for measurement of three soil fractions (Virtanen et al., 2013): exchangeable (0.11 mol acetic acid solution, shaken overnight), reducible (0.5 mol hydroxylammonium chloride adjusted to pH 2, shaken overnight), and organic bound fractions (treated with 30% hydrogen peroxide and dried down over 48 h, then treated with ammonium acetate and shaken overnight). All extractant solutions were separated from soil samples via centrifugation. Metal concentrations in the resulting extractions of exchangeable, reducible, and organic phases were analyzed using a Perkin/Elmer Nexion 300x ICP-MS. Quality control was assured through inclusion of sample duplicates in each analysis, dilution duplicates, and blanks. Sums of metal content within extractions were subtracted from total concentrations measured by ALS Chemex to estimate a final residual fraction.

**Table 1**  
Number and type of soils tested.

Soil microenvironment	N tested for total metal content	N tested for sequential metal content
Cultivated (plots)	74	36
Inside uncultivated (between plots)	40	12
Outside uncultivated (external soils)	33	23

Some sequential extractions were inadvertently not measured. This included two samples each from the exchangeable and reducible analysis, resulting in 69 valid samples for each extraction, and 10 incomplete data points from the organic analysis, resulting in 61 valid samples. Measurement of the reducible fraction of As was affected by polyatomic interference of ArCl compounds in the ICP-MS. Therefore, the residual fraction reported for As contains both true residuals and the fraction sorbed onto Fe and Mn oxides. Arsenic is expected to partition mainly into reducible fractions for most soils, not into residual (Wilson et al., 2010; Orroño and Lavado, 2009), meaning the majority of the “residual” fraction will be reducible, easing interpretation of results.

For quality control, internal standards (Be, Ge, and Tl) were used in dilutions for the ICP-MS and measured metal concentrations were compared to blanks and internal standards. One out of every 5–10 samples in each analysis and analytical stage was resampled for a total of 63 duplicates. Each set of duplicate values varied by <10%, with noted exceptions. The As organic extraction duplicates were an average of 70% different from each other. Organic As variation may be attributed to the ArCl interference from the reducing treatment, though the magnitude of change (generally less than 0.3 ppm) did not merit sample removal. Upon examination of internal standard measurements, duplicate samples with a greater than 10% difference from their pair (10% of all duplicates) were removed due to disagreement in internal standard values.

Before conducting statistical analyses, we examined the data for outliers. Two samples were removed from all analyses due to very high concentrations of Pb and As (upwards of 2000 mg/kg and 85 mg/kg respectively), possibly due to inclusion of soil with decomposed treated or lead painted wood. We removed one garden from the road distance analysis as it was set away behind a local park and all soil samples were more than 80 m from the road, much greater than other samples (1–50 m from road). We also found one garden with very high (>1200 mg/kg) levels of Pb outside the garden ( $n = 3$ ). We include these high levels of Pb in our analysis of total concentrations, though we removed the one high level Pb site chosen for the sequential analysis subset due to lack of comparative points. Finally, we identified a garden with high levels of As in two plots, spatially close to one another. The gardener who managed them revealed he was using an As-based pesticide, not used in any other garden. We removed these from our As study, as they represented a unique source of contamination.

### 2.3. Statistical analysis

Distance from road was measured in ArcGIS 10, using GPS points of individual soil measurements measured to road centerlines. Centerlines of roads were identified using the Census Bureau's Topologically Integrated Geographic Encoding and Referencing (TIGER) database (U.S. Census Bureau). Year neighborhood built is the mean of recorded built dates of the nearest housing structures to the sampled garden (public records from [www.redfin.com](http://www.redfin.com)). Previous land uses of all gardens were determined based on manager interviews and from surveys by the Works Progress

Administration between 1933 and 1939 (City of Los Angeles, Department of City Planning 1939). Garden scale information was also collected in each location, including the use of treated wood and type of fertilizer, as well as the presence of raised beds. We decided not to use data about raised beds or fertilizer type, though other studies have shown them to be significant factors (Mitchell et al., 2014), as the raised bed height, lining material used, and fertilizer type varied too widely within and between gardens.

We used Spearman's non-parametric correlation analysis to compare trace metals and their geochemical proportions to potential contamination sources and soil characteristics (distance from road, age of neighborhood, OM content, pH; SPSS 22). Correlations were characterized separately for each metal and compared between cultivated and uncultivated soils.

To control for potential interactions between neighborhood age and distance from road, correlations were evaluated separately on locations built before or after 1940 and those less than or greater than 30 m from the road. 1940 precedes the two major building periods in Los Angeles and leaded gasoline and paint were widely used during this period (Alloway, 2013). The 30 m distance was chosen based on studies that suggest distance from road effects become undetectable beyond 30 m (Schwarz et al., 2012; Ordóñez et al., 2003). We then compared resulting correlations and patterns to those performed on all points.

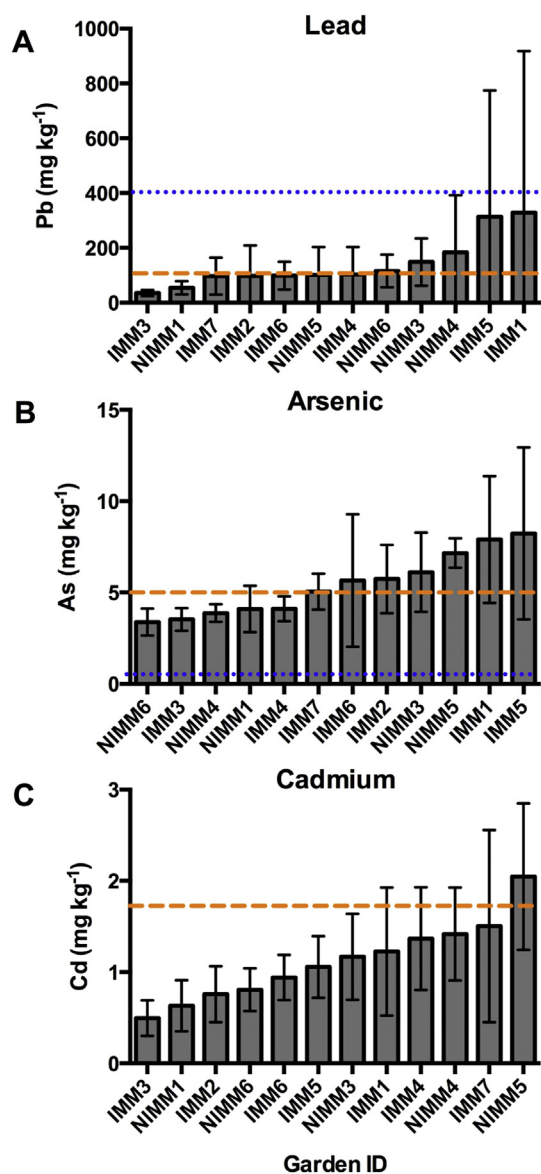
## 3. Results

### 3.1. Soil characteristics and overall concentrations

Soil OM was highest in cultivated soils and related to pH. Cultivated soils were higher in OM than outside uncultivated soils (Mann–Whitney  $U$  test:  $p < 0.05$ ,  $z = -2.427$ ), but not different from inside uncultivated. Cultivated OM varied widely, ranging from 4 to 30%, while uncultivated soils fell in a narrower range (2–24%). The pH levels were consistent, 6.2–8.6 in uncultivated soils (mean = 7.2), and 6.6–8.1 in cultivated soils (mean = 7.4). No significant relationships were found between sequential extraction or total metal concentrations and pH, though OM and pH were weakly correlated (uncultivated:  $r = -0.397$ ,  $p < 0.01$ ; cultivated:  $r = -0.305$ ,  $p < 0.01$ ).

Treated wood and agricultural legacies were shown to affect As and Cd levels in some gardens. Cultivated soils lined with treated wood had an average of 6.5 mg/kg As, higher than average external uncultivated soils (5 mg/kg) and cultivated plots not lined with treated wood (4.5 mg/kg). Only 30% of cultivated soils without treated wood exceeded As background, while 68% of plots with treated wood exceeded it, containing the only cultivated samples with above 10 mg/kg As. These high levels obscured both As distance to road gradients (Fig. 3B) and neighborhood age relationships. The highest Cd concentrations (2.5–4.3) were measured in two gardens that were commercial agriculture until 1958 (Fig. 3C). Removal of these points for analyses showed expected log-normal relationships with age of neighborhood and distance from road for Cd and As.

Total Pb concentration was highly variable (18–1720 mg/kg). A total of 64 soils associated with 11 gardens exceeded the CA 80 mg/kg residential screening level, 27 of those in cultivated soils with edible crops (Fig. 2). Of the uncultivated soils above the screening level, 14 of those were inside uncultivated, 10 of which exceeded 100 mg/kg. In uncultivated soils, Pb concentrations were twice as high as cultivated on average (Fig. 4). Total As concentrations ranged between 2.5 and 17 mg/kg, and 56 soils in 10 gardens exceeded average CA background concentrations (5 mg/kg), with half of these in uncultivated regions. Cadmium values were relatively low (0.11–4.27), though they regularly exceeded CA



**Fig. 2.** Mean garden scale soil concentrations for each of the three investigated metals. (A) Lead (Pb), (B) Arsenic (As), (C) Cadmium (Cd). Error bars represent standard error. The dashed and dotted lines on each graph represent background or safety limit for residential soils; blue dotted: 400 mg/kg, federal actionable limit for residential soils; orange dashed: 5 mg/kg, average background concentration for CA soils; blue dotted: 0.39 mg/kg, CA water health advisory board limit for residential soils. Cd: orange dashed: 1.7 mg/kg, CA health screening limit for residential soils. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

background levels (0.36 mg/kg [Bradford et al., 1996]). Twenty-four soils from ten distinct gardens had more than 1.7 mg/kg Cd, the CHHSL (McClintock, 2012), with 11 of those in cultivated plots. In the two agricultural legacy gardens, 60% of the sampled soils had Cd concentrations above 1.7 mg/kg (half of which were above 2.5 mg/kg) compared to only 18% in gardens without agricultural legacies.

Total Pb, As, and Cd concentrations were significantly correlated with each other (Table 2). The strongest relationships were between Pb and Cd (0.541,  $p < 0.001$ ) and between Cd and As (0.464,  $p < 0.001$ ) with the weakest correlation between Pb and As (0.338,  $p < 0.001$ ).

In terms of geochemical partitioning, inferred bioavailability increased as follows: Pb < As < Cd. The largest proportion of Pb was found in the reducible fraction (42%) followed by the residual fraction (35%; Table 3). Arsenic was predominantly in the combined reducible/residual fraction (69%). Cadmium was relatively bioavailable, with between 50 and 90% of total Cd in exchangeable fractions. Many gardens had less than 2 mg/kg Cd, and sequential sums often equaled or slightly exceeded the total concentration, so the residual Cd fraction of most samples was small.

### 3.2. Age

Correlation coefficients were negative in age analyses, as they were related to the built date (higher values mean younger buildings). Total Pb concentration increased with building age (Fig. 4A.1). The positive age-Pb relationship occurred in both cultivated ( $r = -0.452$ ,  $p < 0.001$ ) and uncultivated soils ( $r = -0.458$ ,  $p < 0.001$ ). For geochemical partitioning (Fig. 4A.3), both reducible ( $r = -0.571$ ,  $p < 0.01$ ) and exchangeable Pb concentrations ( $r = -0.589$ ,  $p < 0.01$ ) increased in uncultivated soils near older buildings. In cultivated soils (Fig. 4A.2), only reducible Pb concentration increased near older buildings ( $r = -0.354$ ,  $p < 0.05$ ).

When gardens with treated wood were excluded, total As concentrations increased from younger to older neighborhoods in cultivated areas ( $r = -0.659$ ,  $p < 0.001$ ), but not in uncultivated areas (Fig. 4B.1). The overall increase in cultivated As concentration in older areas was from increased exchangeable concentrations (Fig. 4B.2;  $r = -0.501$ ,  $p < 0.05$ ).

Total Cd concentration increased with age of neighborhoods (Fig. 4C.1), both in cultivated ( $r = -0.536$ ,  $p < 0.01$ ) and uncultivated ( $r = -0.533$ ,  $p < 0.001$ ) soils, though uncultivated concentrations increased more rapidly with age. Both exchangeable ( $r = -0.483$ ,  $p < 0.01$ ) and organic ( $r = -0.452$ ,  $p < 0.01$ ) Cd concentrations increased in cultivated soils in older neighborhoods (Fig. 4C.2). Exchangeable Cd also increased with age in uncultivated regions (Fig. 4C.3;  $r = -0.369$ ,  $p < 0.05$ ), and the overall change was greater in magnitude.

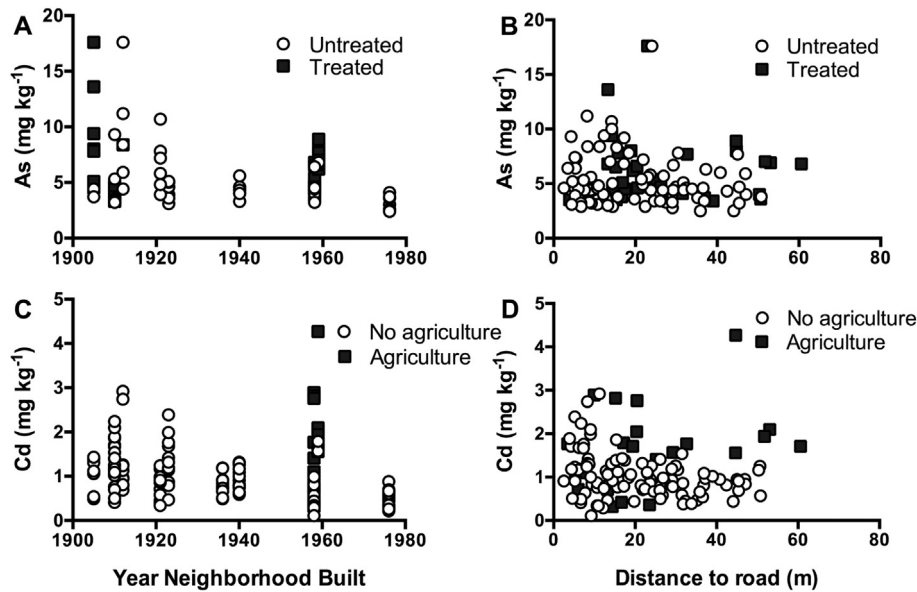
### 3.3. Distance from road

Total Pb concentration decreased with distance from road in both uncultivated ( $r = -0.420$ ,  $p < 0.01$ ) and cultivated ( $r = -0.344$ ,  $p < 0.01$ ) soils, though the magnitude of this change was highest in uncultivated soils (Fig. 5A.1). Only uncultivated soils (Fig. 5A.3) had a change in geochemical partitioning due to distance from road; exchangeable ( $r = -0.589$ ,  $p < 0.01$ ), reducible ( $r = -0.571$ ,  $p < 0.01$ ), and residual ( $r = -0.421$ ,  $p < 0.05$ ) fractions decreased with distance.

Total As concentration did not shift with road distance (Fig. 5B.1). Geochemical partitioning only shifted in cultivated soils (Fig. 5B.2), with As concentrations of exchangeable ( $r = -0.663$ ,  $p < 0.01$ ) and organic ( $r = -0.665$ ,  $p < 0.01$ ) fractions decreasing with distance from road.

Total Cd also decreased with distance from road in cultivated soils (Fig. 5C.1;  $r = -0.412$ ,  $p < 0.01$ ). Cultivated soils (Fig. 5C.2) showed a reduction in reducible ( $r = -0.637$ ,  $p < 0.001$ ) and organic ( $r = -0.582$ ,  $p < 0.01$ ) fraction concentration. In uncultivated soils (Fig. 5C.3), only exchangeable Cd significantly decreased with distance from road ( $r = -0.463$ ,  $p < 0.05$ ).

Our results do not indicate any interactive effect between age of neighborhood and distance from road. There was no distance from road effects on As, Cd, or Pb concentrations in uncultivated soils in neighborhoods built before 1940. Patterns in younger areas were the same as overall results. There was no difference in the



**Fig. 3.** Effect of treated wood on total As concentrations (A, B) and effect of agricultural land use history on total Cd concentrations (C, D) when compared to year neighborhood built (A, C) and distance from road (B, D).

relationship between neighborhood age and soil metals in soils <30 m from the road and overall results (deviation of >10%).

### 3.4. Organic matter

Locations with treated wood and historical agricultural production did not change OM relationships with As and Cd when included in analyses, so we included them here. While OM was related to total (uncultivated:  $r = -0.421$ ,  $p < 0.01$ ) and organic (uncultivated:  $r = -0.575$ ,  $p < 0.01$ ; cultivated:  $r = -0.667$ ,  $p < 0.001$ ) Pb concentrations, correlations were negative. Pb is a legacy contaminant, i.e., active deposition is greatly reduced from historical loading. The lack of change in sequential concentrations changes suggests that OM addition dilutes Pb concentrations rather than affecting chemical phases.

Total concentrations of As did not change with OM, though phase partitioning did (Fig. 6A). In cultivated soils, exchangeable ( $r = 0.712$ ,  $p < 0.001$ ) and organic As ( $r = 0.555$ ,  $p < 0.01$ ) fractions increased with OM (Fig. 6A.2). Uncultivated soils (Fig. 6A.3) only had a significant increase for exchangeable As ( $r = 0.375$ ,  $p < 0.05$ ).

Cd concentration increased with OM in both uncultivated ( $r = 0.521$ ,  $p < 0.001$ ) and cultivated ( $r = 0.328$ ,  $p < 0.05$ ) regions (Fig. 6B.1). This increase was only in reducible form ( $r = 0.652$ ,  $p < 0.001$ ) in cultivated soils (Fig. 6B.2). In uncultivated regions (Fig. 6B.3) proportions of Cd in exchangeable ( $r = 0.551$ ,  $p < 0.01$ ), reducible ( $r = 0.754$ ,  $p < 0.001$ ), and organic ( $r = 0.578$ ,  $p < 0.01$ ) fractions increased with OM content.

## 4. Discussion

Anthropogenic metal contamination is widespread and spatially variable across LA community gardens (Fig. 4). Overall, cultivated locations are less contaminated than uncultivated, likely due to dilution via additions of soil materials. The relationships among metal concentration and measured factors are consistent with other studies (Yesilonis et al., 2008; Schwarz et al., 2012; Wang et al., 2006) though ours is the first to show how metal speciation varies across urban gradients. In particular, reducible Pb is substantial in older neighborhoods (Fig. 4A), potentially indicating

contamination by lead paint opaquing agents (Pb<sub>3</sub>O<sub>4</sub>). Lead, As, and Cd all increase with proximity to roads (Fig. 5), indicating a common pollution source (Table 2). In addition, our results indicate that OM chemically reacts with Cd and As to change geochemical partitioning (Fig. 6). Our study extends previous metal studies to include Los Angeles, a large metropolitan region. We expect that metal relationships with age and influence of road proximity will persist similarly in dense cities built before 1920 in the continental U.S.

### 4.1. Total metal concentration

While our results indicate regional pollution patterns across Los Angeles, the spatial distribution of each metal and its geochemical phases vary. Lead concentrations were the highest and most variable (20–1700 mg/kg; Fig. 2). In comparison, As (2.4–17.6 mg/kg) and Cd (0.11–4.3 mg/kg) were less concentrated. Metal concentrations tend to increase together (Table 2), so overall gardener exposure may be highest for Pb, followed by As, then Cd. Exposure, which does not necessarily translate to health risk, may be increased for urban gardeners and their family members who handle soil and consume unwashed produce (Mielke et al., 1983; EPA, 2013; Hough et al., 2003). Another unique exposure route for gardeners and their family members is through crop uptake of metals (Finster et al., 2004), though exposure to more immobile heavy metals, like Pb, may be linked to inhalation and soil adhered to crops (Finster et al., 2004; Hough et al., 2003). Bioavailability of metals differs from their total concentration in soils. Cadmium is very bioavailable, and exists primarily in the exchangeable fraction (Table 3). Even small Cd concentrations may be taken up rapidly by crops, making uptake difficult to predict (Alloway, 2013; Murray et al., 2011). Because Pb and As exist primarily in less accessible reducible and residual forms, bioavailability to crops may be highest for Cd, followed by Pb and then As.

Our results suggest that As and Cd accumulation in gardens is due to multiple contamination sources. Gardens using treated wood to line plots have cultivated soils with higher median As concentrations (median ~6.5 mg/kg) than uncultivated soils bordering major roads (~5.5 mg/kg; Fig. 3). We suggest that these

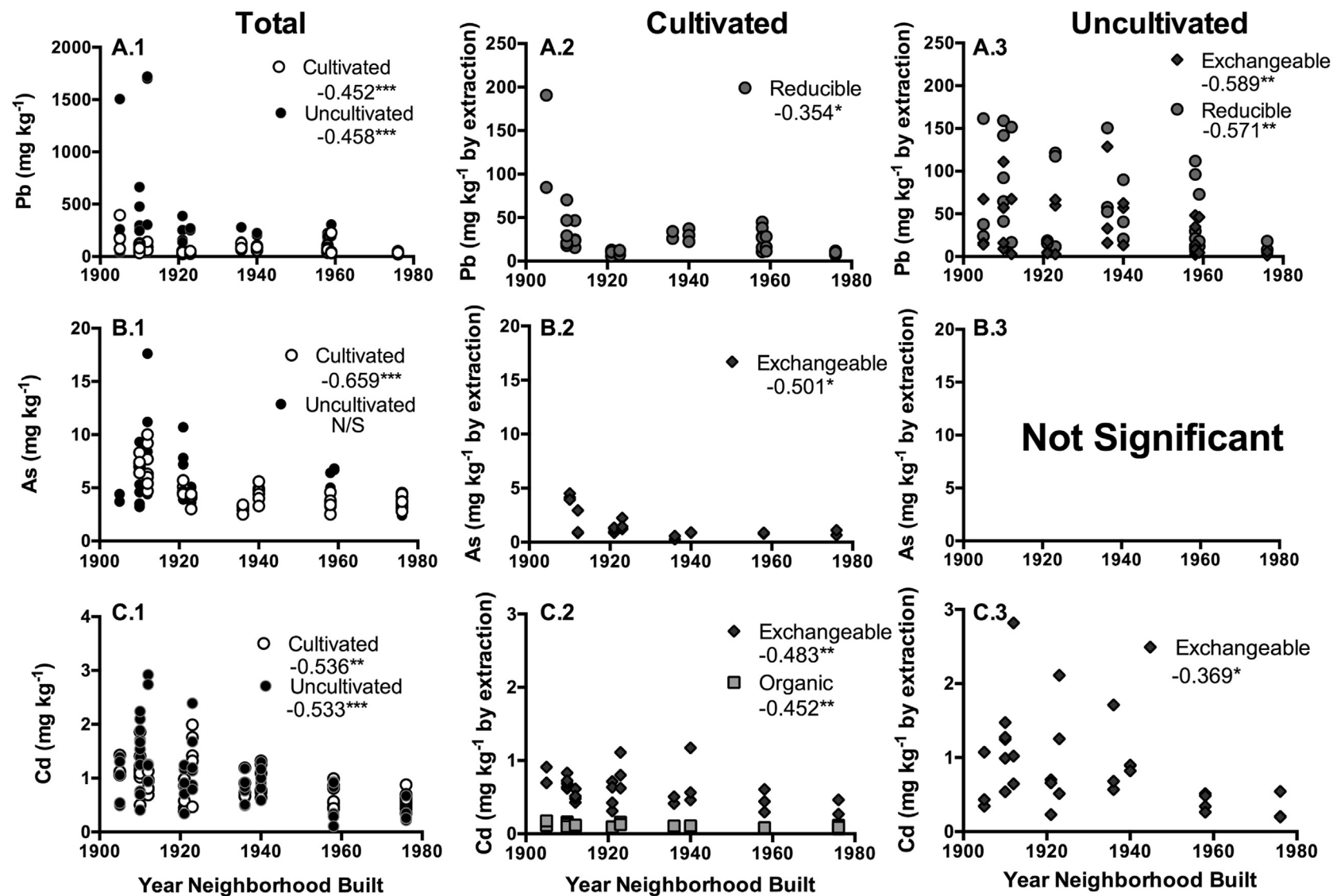


Fig. 4. Spearman correlations between Pb (A.1–A.3), As (B.1–B.3), Cd (C.1–C.3) and age of neighborhood. Column 2 shows sequential extraction of cultivated soils, Column 3 is uncultivated soils. All total concentration values are shown, while only significant sequential data is included. Spearman correlation  $r$ -values are shown under each extraction legend, and stars represent significance values. \* $p < 0.05$ , \*\* $p < 0.01$ , \*\*\* $p < 0.001$ .

**Table 2**  
Spearman's correlation coefficient matrix and significance for relationships between metal concentrations.

		Cd	As	Pb
Cd	Correlation Coefficient	–	0.464**	0.541**
As	Correlation Coefficient	0.464**	–	0.338**
Pb	Correlation Coefficient	0.541**	0.338**	–

\*\* $p < 0.001$ .

**Table 3**  
Percent of overall metals found in each sequential fraction. Percentage was calculated from totals of each extraction divided by the sum of the overall concentration found by ALS.

Metal	Exchangeable%	Reducible%	Organic%	Residual%
Pb	21%	42%	3%	35%
As	25%	N/A	4%	69%
Cd	69%	30%	15%	4%

elevated concentrations may be due to CCA leachates from treated wood creating local (less than 0.5 m radius) contamination (Brandstetter et al., 2000). Similarly, Cd concentrations were elevated in and around two gardens with commercial agricultural history (Fig. 3), likely due to legacy use of high phosphate mineral fertilizers (Andresen and Küpper, 2013; He and Singh, 1994).

#### 4.2. Pb age and distance gradients

Understanding pollution sources is important for reducing resident exposure to metals. While background mineral Pb is generally found in the residual fraction (Saumel et al., 2012), gardens near older buildings were higher in overall Pb and had markedly higher concentrations of reducible Pb (Fig. 4B, C). This result suggests this contamination is sourced from lead tetraoxide (Pb<sub>3</sub>O<sub>4</sub>), a reducible compound found primarily in paint (Jacobs et al., 2002).

In contrast, all fractions of Pb decrease with distance from road (Fig. 5A.3) indicating the road is a non-point source of pollution. Legacies of lead gasoline additives, along with mechanical movement of dust by vehicles, are possible causes of near road enrichment in multiple Pb phases (Kerr and Newell, 2003). Decreased Pb concentrations in cultivated soil may be due to dilution of soils through tillage and addition of amending compounds (Fig. 5A.2, Fig. 4A.2). Even so, near 50% of the locations that exceeded the 80 mg/kg CHHSL limit were cultivated plots near older buildings. This persistent age signal of reducible Pb in cultivated plots indicates that Pb may still be actively added to soils through degradation of leaded paint on nearby surfaces, a fact important for garden managers to consider when planning garden spaces.

#### 4.3. As: air pollution and OM interactions

The observed increases in As seen in older neighborhoods and with proximity to road is almost exclusively in the exchangeable pool (Fig. 4B.2) which may be linked to long-term car exhaust deposition (Haygarth and Jones, 1992). Significant relationships between As concentrations and neighborhood age and road proximity are seen only in cultivated soils (Fig. 4B.2) potentially due to As becoming more reactive in high OM soils (Wilson et al., 2010) (Fig. 6A.2).

The high concentration of exchangeable As in cultivated, high OM soils (Fig. 6B.2) without a corresponding increase in total As concentrations suggests chemical reaction with OM. This release of bioavailable As may be caused by reaction of humic acids with Fe

and Mn oxides, releasing adsorbed reducible As compounds into exchangeable fractions (Wilson et al., 2010; Wenzel, 2013). Compost and manure, added to cultivated soils along with increased tillage and irrigation, have and release more humic acids than OM found in uncultivated areas (Murray et al., 2011). Organic As also increased in cultivated regions, potentially due to increased availability of easily adsorbed exchangeable As. High rates of As sorption onto OM is more common at lower pHs than found in our study (Thanabalasingam and Pickering, 1986), explaining in part the persistence of As in the exchangeable fraction.

#### 4.4. Cd roadside pollution and OM effects

Similar to As, Cd exchangeable concentrations increase with age and proximity to roads, likely due to roadside pollution over time (Figs. 4C and 5C). Uncultivated soils are more sensitive to age than cultivated regions (Fig. 4C), indicating a management effect on Cd intensity. Recent studies have shown that gardens replace cultivated soil constantly, which contributes to the observed dilution (Mitchell et al., 2014). Cadmium concentrations also increase with OM additions (Fig. 5C.1), potentially due to mineral phosphorous in some added fertilizers.

Cultivated soils have more reducible Cd with increased OM, possibly indicating a phase change induced through soil management (Fig. 6B.2). Cadmium adsorbs more readily to OM than As, and cultivation practices like irrigation and regular addition of OM in plots may encourage the reduced bioavailability of Cd (Sun et al., 2013; Basta et al., 2001). Addition of OM rich fertilizer has been used to remediate Cd contaminated soils (Herwijnen et al., 2007), as its adsorption capacity is up to 30 times that of clay (Sauvé et al., 2003). In uncultivated areas, the positive Cd-OM relationship is significant across all fractions, and may be a result of road exposure and long-term incorporation of Cd into the organic fraction.

#### 4.5. Conclusions and applications

Our research highlights the pervasive heavy metal contamination of urban garden soils, clarifies the fate and sources of heavy metals in Los Angeles, and suggests management activities to reduce bioavailability and accumulation of metals. In particular, our results support active soil management for reduced exposure, including adding imported soils to dilute Pb and Cd (Mitchell et al., 2014; Heinegg et al., 2000) (Figs. 2 and 5), especially near older buildings and in previous agricultural regions. Planting crops away from the road to decrease deposition (Fig. 5) and reducing use of CCA wood (Fig. 3) are supported by our results and also by EPA literature on safe gardening techniques (EPA, 2013). Gardeners may also benefit from moderating OM to regulate bioavailability of Cd and As (Sloan et al., 2012), as As was the most bioavailable in soils with <20% OM (Fig. 6), though crops only need 5–10% for optimum root growth (Magdoff and Weil, 2004). Regular soil testing and targeted management could better balance potential exposure to metals with production of urban food (Saumel et al., 2012).

As urban centers expand and redevelop more land within the existing urban boundary, transportation networks and urban legacies will affect broader regions (Zhao et al., 2011; Kabir et al., 2014). We show that agricultural, land use, and pollution legacies can all influence metal concentrations and presence, indicating a need to include legacy factors in urban soil studies (Bain et al., 2012). With the broad scope of this study, we can better understand how local management, legacies, and distance gradients drive geochemical partitioning and distribution of soil metal contamination. This knowledge is essential for estimating crop uptake of metals, measuring the spatial distribution of metals



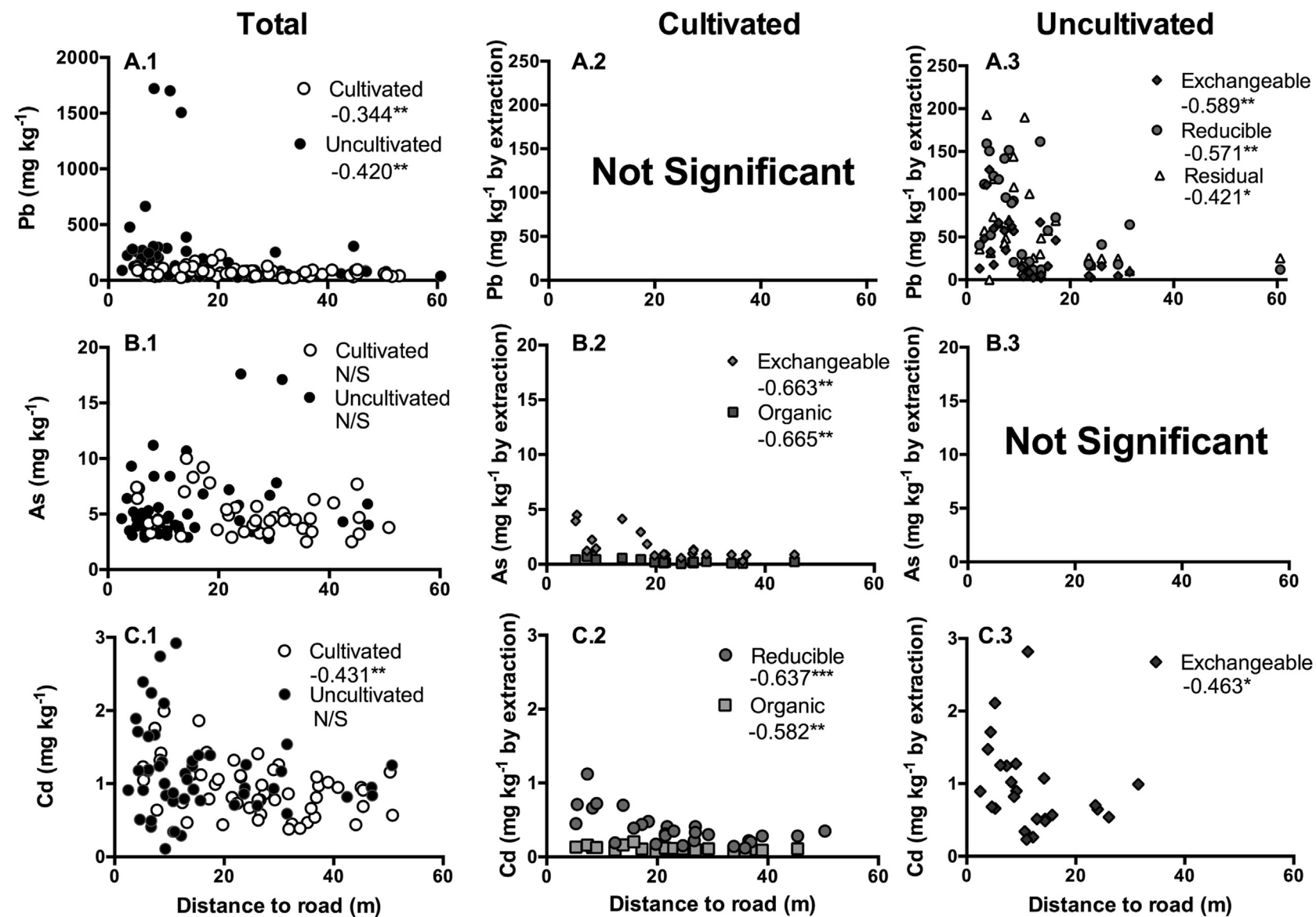
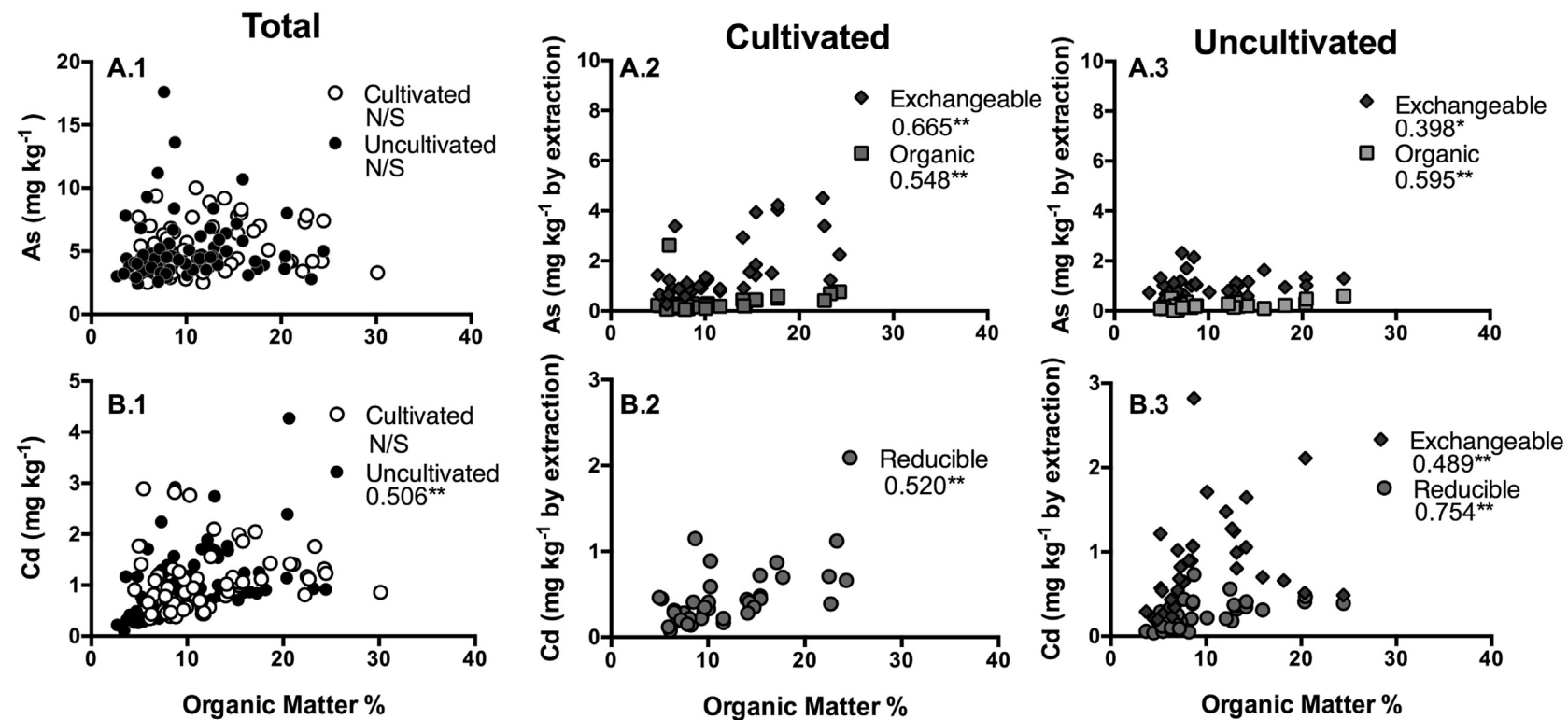


Fig. 5. Spearman correlations between Pb (A.1–A.3), As (B.1–B.3), Cd (C.1–C.3) and distance from road. Column 2 shows sequential extraction of cultivated soils, Column 3 is uncultivated soils. All total concentration values are shown, while only significant sequential data is included. Spearman correlation  $r$ -values are shown under each extraction legend, and stars represent significance values. \* $p < 0.05$ , \*\* $p < 0.01$ , \*\*\* $p < 0.001$ .



**Fig. 6.** Spearman correlations between As (A.1–A.3), Cd (B.1–B.3) and OM content. Column 2 is sequential extraction of cultivated soils, Column 3 is uncultivated soils. All total concentration values are shown, while only significant sequential data is included. Spearman correlation  $r$ -values are shown under each extraction legend, and stars represent significance values. \* $p < 0.05$ , \*\* $p < 0.01$ , \*\*\* $p < 0.001$ .

across urban neighborhoods, and targeting remediation efforts to minimize human exposure.

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