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# Metal sources and exposures in the homes of young children living near a mining-impacted Superfund site

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## Abstract

Children living near hazardous waste sites may be exposed to environmental contaminants, yet few studies have conducted multi-media exposure assessments, including residential environments where children spend most of their time. We sampled yard soil, house dust, and particulate matter with aerodynamic diameter <2.5 in 59 homes of young children near an abandoned mining area and analyzed samples for lead (Pb), zinc (Zn), cadmium (Cd), arsenic (As), and manganese (Mn). In over half of the homes, dust concentrations of Pb, Zn, Cd, and As were higher than those in soil. Proximity to mine waste (chat) piles and the presence of chat in the driveway significantly predicted dust metals levels. Homes with both chat sources had Pb, Zn, Cd, and As dust levels two to three times higher than homes with no known chat sources after controlling for other sources. In contrast, Mn concentrations in dust were consistently lower than in soil and were not associated with chat sources. Mn dust concentrations were predicted by soil concentrations and occupant density. These findings suggest that nearby outdoor sources of metal contaminants from mine waste may migrate indoors. Populations farther away from the mining site may also be exposed if secondary uses of chat are in close proximity to the home.

### Keywords

house dust; indoor air pollution; metals; mine waste; residential exposures; Tar Creek Superfund Site



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## Introduction

An estimated 41 million people live within close proximity to the most contaminated Superfund sites in the United States (National Research Council, 1991). Metal contaminants are of priority concern at hazardous waste sites due to their prevalence and toxicity; 4 of the top 10 contaminants at Superfund sites are metals (Agency for Toxic Substances and Disease Registry (ATSDR), 2005).

Biologically, the developing brain may be particularly sensitive to the effects of metals' exposure *in utero* and in the early postpartum period (Weiss, 2000; Bondy and Campbell, 2005). Numerous studies have shown associations between infant blood lead (Pb) concentrations and impaired neurodevelopment, including reduced IQ scores (Needleman et al., 1990; Bellinger et al., 1992). Other metal pollutants, such as cadmium (Cd), arsenic (As), and manganese (Mn), have also been shown to have effects on reproduction and development (Wright et al., 2006; Ettinger et al., 2009; Tian et al., 2009; Zota et al., 2009a).

House dust has been identified as an important route of exposure, particularly for children (US EPA, 2008), with direct exposure occurring through incidental ingestion, inhalation of re-suspended dust particles, and dermal absorption. Indoor house dust is a complex mixture of soil, biological materials, and settled indoor aerosols that can act as a reservoir for contaminants. Metal concentrations in dust have been correlated with biological markers of exposure in blood and urine (Lanphear et al., 2005; Rollin et al., 2005; Hogervorst et al., 2007), and Pb-contaminated house dust is the major source of Pb exposure for US children (Lanphear et al., 2005). Compared with their adult counterparts, children living near contaminated sites may encounter elevated exposures to environmental contaminants due to differences in both physiology and behavior (Cohen Hubal et al., 2000; US EPA, 2008). Children have higher inhalation rates per unit of body mass than adults. Unique behaviors such as crawling and frequent hand-to-mouth activity may increase children's contact with contaminated soil and dust.

Mining sites are among the most heavily contaminated by metals. For example, the Tar Creek Superfund Site, a former Pb and zinc (Zn) mining area located in rural Oklahoma (OK), USA has approximately 30 piles of mine waste (locally known as "chat"), which contain elevated concentrations of Zn, Pb, and Cd (ATSDR, 2004; Schaider et al., 2007). A high prevalence of childhood Pb poisoning (>10 $\mu$g/dl) has been documented in the area, particularly in towns closest to the chat piles (Malcoe et al., 2002; ATSDR, 2004). These findings motivated a soil remediation effort by the US Environmental Protection Agency (US EPA) (ATSDR, 2004). Moreover, our previous work at this Superfund site demonstrated that Pb and Zn concentrations in outdoor respirable particulate matter with aerodynamic diameter<10 $\mu$m ($PM_{10}$) and particulate matter with aerodynamic diameter <2.5 ($PM_{2.5}$) were higher at air monitoring sites closest to the chat piles. Particles of mine waste origin contributed, on average, 40% to the total mass of particles in the coarse ($PM_{10}$–$PM_{2.5}$) fraction at a residential site adjacent to the chat piles (Zota et al., 2009b).

The relationship between outdoor mining-related metal contamination and the indoor residential environment is not well understood. Quantifying indoor concentrations of metals originating from mine waste is complex at the Tar Creek Superfund Site because there are numerous chat piles distributed throughout the area, and mine waste has been dispersed through the region via erosion from piles and distribution of crushed rock for use in road construction, housing foundations, and as a substitute for gravel in residential driveways (Oklahoma Office of the Secretary of Environment, 2000; Perry et al., 2005). Mine waste particles can be blown off the piles by wind or generally dispersed when disrupted by secondary use of the crushed rock or disturbed by recreational vehicle use. Wind-blown particles settle on outdoor surfaces, and can penetrate indoors, where they settle out and can be re-suspended. Chat particles deposited on outdoor soil can be tracked into the home and ultimately mixed in as a component of household dust.

As part of a multi-disciplinary effort aimed at understanding the impacts of mining-related metal exposures on children's health, we conducted a multi-media exposure assessment study of Pb, Zn, Cd, Mn, and As in the homes of young children living near the former mining site. These metals were chosen either because they are elevated in chat piles (Pb, Zn, and Cd) or because they are relevant to the study of neurodevelopmental health outcomes (Mn and As). The goal was to characterize metal concentrations in outdoor and indoor environmental samples in order to better understand transport mechanisms and source contributions to metals found in house dust.

## Methods

### Study Design

The children's exposure assessment study was conducted as part of the Center for Children's Environmental Health and Disease Prevention Research at the Harvard School of Public Health (HSPH), a research effort designed to understand the impacts of mining-related metal pollutants on children's growth and development through a longitudinal birth cohort, mechanistic animal studies, and environmental fate and transport studies (Hu et al., 2007). Field research for the exposure and health studies took place in and around the Tar Creek Superfund Site, which is located in rural Ottawa County in the northeastern corner of OK. Study objectives and design for the individual components, including the birth cohort, have been described elsewhere (Ettinger et al., 2009; Zota et al., 2009a).

We intended to recruit 50 mother–infant pairs in the exposure assessment study who were actively enrolled in the birth cohort study; included an infant <12 months of age at the time of environmental sampling; and had a primary residence within the study area, Ottawa County, OK, during the exposure assessment study period. Additionally, we over-sampled homes within the Superfund site boundaries since we were primarily concerned with mine waste-related exposures and the majority of birth cohort participants resided outside of the designated Superfund site boundaries. In addition to the environmental sampling, we also collected biological markers of metals exposure at birth and 1 year of age from the young children living in these study homes. The biomarkers of metals exposure, and their associations with levels of metals in environmental media, will be presented in future manuscripts. Fifty-three mother–infant pairs from the birth cohort participated in the children's exposure assessment study between July 2005 and September 2006. The research protocol was approved by the Human Subjects Committees of Integris Baptist Medical Center and HSPH.

### Sample Collection

Homes were sampled by trained technicians, with two sampling sessions for participants who moved during the study period, resulting in a total of 59 home assessments. During each sampling session, the study team collected outdoor yard soil, indoor house dust, and indoor $PM_{2.5}$. Surface yard soil samples were collected from the top 2 cm using a sterile polystyrene spatula in the front entryway of each home. Three samples were collected within a 1 m diameter circle and mixed together to create one composite sample per home (US EPA, 1995; Riederer et al., 2005). Two-week integrated $PM_{2.5}$ air samples were collected on Teflon filters (2 $\mu$m pore size, 37mm in diameter) using Harvard Impactors (Marple et al., 1987) with low flow medo air pumps at 4 l/min (Zota et al., 2009b) in the main living space of the home. Dust samples were collected using a Eureka Mighty-Mite vacuum cleaner attached to a crevice tool modified to collect dust into a cellulose extraction thimble (Whatman, Clifton, NJ, USA). Samples were collected by vacuuming approximately 2 $m^2$ of the floor in the main living area of the home for 5 min (Chew et al., 1998).

In addition to the collection of environmental samples, a home visual inspection, which included information about home characteristics, geographic location, characteristics of the surrounding area, presence of chat in the driveway, housing size and structure, and overall maintenance, was completed by a sampling technician. A Pb-based paint screening was conducted on-site with a portable X-ray fluorescence (XRF) Pb paint analyzer (Thermo Fisher Scientific NITON 300 Series Lead Paint Analyzer, Billerica, MA, USA). Distance between the front entrance and the closest road (distance to street) was measured using a digital measuring wheel. A face-to-face survey was administered by field staff to the infant's caregiver about household characteristics, appliance use, and cleaning practices.

## Analytical Methods

Yard soil samples were analyzed using a polarized energy-dispersive X-ray fluorescence (pED-XRF) Specto XEPOS® (Spectro Analytical, Kleve, Germany) instrument at Wellesley College. Samples were air dried, sieved to <2 mm and then 4 g of soil were prepared in XRF sample cups with 6 $\mu$m thick Mylar film windows. The pED-XRF generally achieved an analytical error of ±5%. Testing of samples via pED-XRF was bracketed with National Institute of Standards and Technology (NIST) 2709 (San Joaquin Soil) standard reference material (SRM). Measured concentrations of all elements in the standard remained within ±10% of accepted values. Concentration uncertainties were provided by the instrument that equaled 1 SD of error estimates based on analytical precision. Three times the uncertainty was considered to be the limit of detection (LOD) for each element and sample. Concentrations for Cd and As in soil were frequently below the LOD. For this reason, soil samples were also analyzed for Cd and As by inductively coupled plasma mass spectrometry (ICP-MS), which has a lower LOD (see description below). Only the ICP-MS data for Cd and As are reported in the results.

Dust samples were sieved through vigorous, manual shaking to <125 $\mu$m using a polyethylene sieve (Nalge Nunc International, Rochester, NY, USA) and the dry weight recorded. Dust and soil samples were microwave digested with concentrated $HNO_3$ adapted from the method described in Shine et al. (1995). Vials with only concentrated $HNO_3$ were digested with each batch as a laboratory blank. Samples were analyzed by ICP-MS (Elan 6100, Perkin-Elmer, Norwalk, CT, USA) at the HSPH Trace Metals Laboratory. The lower limit of quantification with this method was 0.1 $\mu$g/g. The method LOD was calculated as three times the SD of eight laboratory digestion blanks. The method LOD was equivalent to the lower limit of quantification for Pb, Cd, and As. The method LOD for Mn and Zn was 0.2 and 0.6 $\mu$g/g, respectively. Analytical accuracy was assessed by replicate digestion and analysis of NIST SRM1649a (Urban street dust). All elements of interest were within ±10% of accepted values except for Zn, which was within ±12%. Four dust and four soil samples were not analyzed due to lack of adequate mass (<0.1 g) and/or field sampling problems.

Laboratory analysis of $PM_{2.5}$ has been described in detail elsewhere (Zota et al., 2009b). Briefly, teflon filters were weighed in a temperature- and humidity-controlled room (18–24°C, 40±5% relative humidity). The elemental content was quantified by energy-dispersive XRF. Analyses were performed according to standard operating procedures at the US EPA's National Exposure Research Laboratory in Research Triangle Park, NC, USA. Field blanks were transported and handled like regular samples, but the filters were not attached to the air pumps. Field blanks, which comprised 10% of the total samples collected, were used to determine background contamination. Field blank concentrations for all elements were below the method LOD. Precision of the method was determined by duplicate samples (10% of total samples collected).

### Data Analysis

Univariate and bivariate statistics, tabulations, and distribution plots were examined for all variables. For data that fell below the method LOD, estimated metal concentrations provided by XRF and/or ICP-MS instrumentation were used in calculation of summary statistics and statistical models. Dust loading was calculated by multiplying dust concentration by the mass of total dust and then dividing by the area vacuumed. Environmental concentrations approximated a log normal distribution; thus, all concentrations were log-transformed in linear regression models. Correlations were assessed using the Spearman's rank correlation.

Dust/soil concentration ratios were calculated for each metal in each home by dividing the dust metal concentration by the soil metal concentration. To calculate the upper-bound estimates for the contributions of soil to dust concentrations for Pb, Zn, Cd, and As, we assumed that there were no significant indoor sources of Mn (ATSDR, 2008) and that the Mn dust/soil ratio for each home represents the soil infiltration factor (i.e., the fraction of dust mass that comes from soil). We multiplied the soil infiltration factor by the actual soil concentrations for the other metals to obtain the maximum concentration in dust that could be attributed to soil, and then divided the empirical dust concentration by this calculated value.

Non-parametric tests were used to examine differences in dust metal concentrations by potential metal sources and housing factors. Differences between groups were tested using Wilcoxon's rank sum test for two categories and Kruskal–Wallis test for three or more categories. The following variables related to potential metal sources and factors that may affect resuspension or transport of dust were considered: presence of chat in residential environment (driveway and home foundation); proximity to other chat sources (chat piles and chat on roads); other potential sources of metals (Pb-based paint, agricultural fields, and street dust (approximated by distance to street)); housing structure; household characteristics (central air conditioning, occupant density, dog ownership, and housekeeping); and season of sampling. Participants were asked whether their homes participated in the residential yard soil remediation program sponsored by the US EPA in which contractors removed parcels of yard soil, where Pb concentrations exceeded 500 $\mu$g/g (ATSDR, 2004). Occupant density, a proxy for particle resuspension (Baxter et al., 2007), was defined as the number of people living in the home divided by the number of total rooms. We then constructed multivariate regression models with dust metal levels as the outcome. Soil metal levels were included in all multivariate models regardless of statistical significance based on *a priori* considerations about environmental fate and transport. We also include variables that were significant in univariate models at $P<0.20$. We used backward stepwise elimination process to derive our final model that included variables that were statistically significant ($P<0.20$) and confounders of our main effect (association with chat sources). One outlier that had a large influence on regression results, defined as having an absolute studentized residual >3, was removed from the As dust regression analyses. In univariate regression analyses, the presence of chat in driveways and close proximity to chat piles were both modeled separately as binary variables. Since 67% of the participants who lived near a chat pile also had chat in the driveway, adding both terms to the model produced unstable estimates due to collinearity. As a result, a three-category dummy variable was created to approximate the influence of chat sources on dust metal concentrations in multivariate regression models. If a participant's home had chat in the driveway and was close to the chat piles (<0.5 km), then it was designated to the high chat impact category. If the study home possessed only one of these characteristics (either chat in the driveway or proximity to a chat pile), then it was assigned to the moderate chat exposure category, and, if neither condition was present, then it was assigned to the low chat exposure category. All analyses were conducted in SAS version 9.2 (SAS Institute, Cary, NC, USA). Statistical significance was defined by a (two-sided) *P*-value of 0.10 or lower.

# Results

## Summary Statistics

Metals were frequently detected in all exposure media except for Cd in $PM_{2.5}$ (Table 1). Concentrations were highly skewed and varied greatly, often spanning two orders of magnitude, across homes. Zn was found in the highest concentrations across all media. Pb and Mn occurred in similar magnitudes, while Cd and As were generally found at the lowest levels. The US EPA soil screening level (SSL) of 400 $\mu$g/g for Pb was exceeded in 14% of soil and 4% of dust samples. The SSL for As (0.4 $\mu$g/g) was exceeded in 92% of soil and 100% of dust samples. All samples were below the SSL for Zn (23,000 $\mu$g/g) and Cd (78 $\mu$g/g). There is no SSL for Mn, but the concentrations in soils were within the range typically found in soils from this region (Smith and Huyck, 1999).

## Metal Correlations Within and Across Environmental Media

Table 2 presents correlations for metals across the different media. Not surprisingly, dust metal concentrations and dust metal loadings were significantly correlated for all metals (Spearman's rho ($\rho$) range: 0.35–0.70). While soil and dust metal concentrations were modestly correlated for all metals except As (Spearman's $\rho$ range: 0.29–0.48), there was a less consistent correlation between dust metal loading and soil metal concentrations. Pb concentrations in indoor air were significantly correlated with Pb concentrations in both soil (Spearman's $\rho$ = 0.40) and dust (Spearman's $\rho$ = 0.41) as well as dust loading (Spearman's $\rho$ = 0.25). The relationship between indoor air and other media was weaker for Zn, Cd, Mn, and As. Correlations among metals within each medium were also examined. Pb, Zn, and Cd were strongly correlated in both house dust and soil (Spearman's $\rho$ range: 0.80–0.90), suggesting common sources. These metals showed a significant but weaker correlation with Mn and As (Spearman's $\rho$ range: 0.25–0.40) (data not shown).

The relationship between soil and dust was further examined by calculating dust/soil concentration ratios for each metal and each home (Figure 1). For all study homes, the Mn dust/soil ratio was <1, suggesting a dominance of outdoor sources and few, if any, indoor sources. In contrast, Zn and Cd dust concentrations exceeded those in soil for approximately 70% of homes; and in a few homes, dust concentrations were over 10 times higher than those in soil, suggesting the presence of indoor sources and transport mechanisms other than soil tracked in from outdoors. Pb and As were generally in the middle with the dust/soil ratio exceeding one in approximately half the homes. The median percent contribution of soil to dust concentrations for Pb, Zn, Cd, and As across homes was 60%, 20%, 30%, and 40%, respectively.

## Predictors of Dust Metal Concentrations

Since dust metal concentrations were only partially explained by outdoor soil conditions, we examined associations with housing and neighborhood characteristics in order to identify other important sources and potential transport mechanisms. Table 3 summarizes neighborhood and housing characteristics, and their univariate associations with dust metal concentrations. A majority (80%) of the study homes were single-family dwellings, and one-third of all homes were located inside the Superfund site boundaries. Approximately 25% of homes were located in highly impacted areas (within 0.5 km of one or more chat pile), and 18% of homes had undergone yard soil remediation. Chat use in the driveway was documented more frequently than chat use in the housing foundation (42% *vs* 18%).

Exterior Pb-based paint was more commonly detected than interior Pb-based paint (26% *vs* 9%).

Proximity to outdoor sources influenced indoor dust concentrations. For example, homes that were within 0.5 km of one or more chat piles had significantly higher levels of Zn, Cd, and As in dust than homes farther away from chat piles. Residing near agricultural fields was associated with elevated levels of Mn and As. Homes that were within 5–10m from the street had significantly higher levels of Pb and Cd in dust. Housing characteristics also influenced indoor dust metal levels. Presence of chat in the residential driveway was associated with significantly higher concentrations of Pb, Zn, Cd, and As, but not Mn. Homes that had undergone yard soil remediation had higher levels of mining-related metals, but this trend was only significant for Zn. Exterior Pb-based paint was associated with increased Pb, Zn, and Cd dust levels, while central air conditioning was associated with lower levels of these metals. Mn dust levels were significantly higher in single-family homes compared with apartments or mobile homes. Chat use in the housing foundation, interior Pb-based paint, housekeeping, occupant density, and season of sampling were not significantly associated with any of the dust metal concentrations.

To quantify the cumulative impact of chat sources on dust metal concentrations, exposure to two different chat sources (proximity to chat piles and chat in driveway) was aggregated into a three-category variable. For Pb, Zn, Cd, and As, dust metal levels increased as the number of chat sources increased ($P$ (trend) $<0.05$), and homes with two chat sources had the highest levels of Pb, Zn, Cd, and As (Figure 2). Median dust levels in these homes were approximately three times higher than those in homes with no known chat sources (Pb: 148 *vs* 54 $\mu$g/g; Zn: 1490 *vs* 569 $\mu$g/g; Cd: 9.1 *vs* 2.4 $\mu$g/g; and As: 9.3 *vs* 3.7 $\mu$g/g).

In multivariate regression models, the presence of chat sources remained a significant predictor of Pb, Zn, Cd, and As even after controlling for other sources of dust metal concentrations, including soil concentrations (Table 4). Homes with two chat sources had Pb, Zn, Cd, and As levels two to three times higher than homes with no known chat sources. Distance to street was inversely associated with Pb, Zn, and Cd. Single-family homes had higher concentrations of Pb, Zn, and Mn compared with mobile homes and apartments. Central air conditioning was associated with lower levels of Pb and Cd. Soil concentrations were not significantly associated with Pb, Cd, Zn, or As, and presence of exterior Pb-based paint was no longer a significant predictor of dust Pb ($P = 0.27$). In contrast, soil Mn concentrations and occupant density were associated with dust Mn concentrations. Proximity to agricultural fields was associated with higher Mn and As.

## Discussion

To our knowledge, this is one of the first studies to conduct a comprehensive, multi-media exposure assessment in the residential environment of young children living near a mining-related site. While metal concentrations of Pb, Zn, and Cd in residential media were 3–13 times lower than those in the solid mine waste (Schaider et al., 2007) and similar to those observed in other US residential exposure studies (Clayton et al., 1999; O'Rourke et al., 1999), we found that proximity to chat piles and chat in the driveway significantly increased indoor house dust concentrations of Pb, Zn, Cd, and As. After controlling for other sources of metals, homes with these two chat sources had dust metal levels that were two to three times higher than those in homes with no known chat sources, suggesting that local communities near abandoned mining sites may be exposed to particles of mine waste origin via the residential environment. While previous studies have documented higher pollutant levels in the dust of homes closer to large point sources, such as chat piles (Malcoe et al., 2002), this is the first study to demonstrate the impact of chat use in the driveway, a secondary use of mine waste, on indoor metal levels. The relevance of this finding extends beyond the Tar Creek area since a recent assessment of mine waste issues in the United States notes that reuse of mine waste in driveways is widespread at other abandoned mining sites (The Interstate Technology & Regulatory Council, 2008).

Moreover, our results suggest that the transport of chat into the indoor environment is not entirely mediated by soil since the dust levels of mining-related contaminants were higher than those in bulk soil (dust/soil ratio>1) in over half the homes, and in several homes, the dust/soil ratio for Zn and Cd exceeded 10. Additionally, introducing soil metal levels into the multivariate regression models for dust did not change the effect estimates for chat sources. The dust/soil relationships in our study homes may have differed if we had analyzed the same size fraction in both media since previous studies suggest that smaller soil particles tend to be enriched in Pb (Clark et al., 2008). However, when we assessed metal concentrations in different size fractions of some of our soil samples, we did not see any systematic relationships between particle size and metal concentration (data not shown).

Airborne transport of particles may be another important pathway by which outdoor particles of mine waste origin migrate into the home. We attempted to characterize this pathway through direct measurements of weekly integrated $PM_{2.5}$. The $PM_{2.5}$ and dust metal levels were only weakly correlated, which may reflect a difference in integration time between the two media since air measurements reflect short-term exposures, whereas dust can accumulate over months and years (Bradman and Whyatt, 2005). Additionally, this correlation may have been improved by including measurements of $PM_{10}$ since our previous work found that particles of mine waste origin were more abundant in the coarse fraction ($PM_{10}$–$PM_{2.5}$) compared with $PM_{2.5}$ (Zota et al., 2009b). However, the association between central air conditioning and lower levels of mining-related dust metals in our data supports the role of airborne transport since previous air pollution studies have shown that use of central air conditioning lowers air exchange rates, which thereby reduces infiltration of outdoor particles into the indoor environment (Long et al., 2001). Future exposure assessment studies in residential indoor environments should include long-term airborne dust measurements to further characterize the relationship between airborne particles and house dust.

This study is also one of the first examinations of indoor residential environmental conditions near the Tar Creek Superfund site after yard soil remediation. Yard soil remediation was completed in multiple phases at the site beginning in the late 1990s through 2007 (US EPA, 2010). We observed higher levels of mining-related contaminants, particularly Zn, in the dust of homes that had undergone soil remediated (although yard soil remediation did not remain a significant predicator in multivariate models). Thus, it is possible that the remediation may have temporarily reduced metals in yard soil leaving metals in residual indoor dust unaffected. Moreover, remediating yard soil without treating the chat piles or chat in driveways would have little effect on transport of metal-laden dust from the sources themselves.

We also found significant associations between dust metal concentrations environmental sources, other than mine waste. Consistent with other studies (Meyer et al., 1999), a shorter distance between the home and street, a likely proxy for street dust, was associated with increased levels of Pb, Cd, and Zn in dust. Street dust may have elevated concentrations of Pb, Cd, and Zn from traffic-related materials such as tire and brake dust and yellow road paint (Fergusson and Kim, 1991; Adachi and Tainosho, 2004) or from chat, since chat is used as a road fill and asphalt ingredient in this area. Detached single-family homes had higher concentrations of metals as compared with apartments and mobile homes. This difference may reflect housing age, particle penetration efficiency, and/or proximity to outdoor sources. Presence of exterior Pb-based paint was associated with higher levels of Pb, Zn, and Cd, although these associations lessened in significance after introducing soil metal concentrations into the model, suggesting that in this study, soil transport is a major mechanism by which exterior Pb-based paint migrates into indoor house dust. Interior Pb-based paint, which was only detected in 9% of homes, was not associated with dust metal concentrations in our study. Surprisingly, As dust concentrations were positively associated with chat sources, particularly with chat fill in the driveway. While As is not a major mining contaminant in the area, acid-soluble concentrations of As in the source chat material ranged from 10 to 20 $\mu$g/g (unpublished data), which is two to three times higher than the As levels we observed in yard soil.

This is the first study to our knowledge to identify predictors of Mn in house dust. Dust/soil ratios and regression models suggested that yard soil is the major source of Mn in dust. Additionally, occupant density, a surrogate for indoor resuspension activity (Baxter et al., 2007), was associated with increased Mn dust levels. Mn dust levels were higher in homes near agricultural areas. This finding is consistent with a prior study in Canada, which found higher blood Mn levels among pregnant women who reported pesticide use within 1 km of residence (Takser et al., 2004) and may reflect the use of Mn-containing fungicides and seed protectants (ATSDR, 2008) Alternatively, since soil is likely the major source of Mn dust, the higher Mn dust levels in homes near agricultural areas could reflect the production and transport of fugitive dust particles from agricultural operations (Clausnitzer and Singer, 2000). Sources and transport of Mn were very different than the other metals since Mn is not enriched in chat. Mn levels in chat occur at 25–30 $\mu$g/g (unpublished data), which is an order of magnitude lower than median concentrations quantified in our soil samples.

We did not observe an association between dust metal levels and self-reported use of chat in housing foundation, another unregulated use of mine waste that has been documented in the area (Perry et al., 2005). The use of chat for backfill around foundations may be a less likely source of indoor contamination because of limited direct contact. However, our ability to observe an association may have been obscured by measurement error since many participants did not know whether chat had been used in the foundation of their home and our study team had no way to independently verify or obtain this information. Overall, despite our limited statistical power, we were able to identify several important sources of metals in the residential environment, including mine waste. Future studies from our research center will use data from this exposure assessment study to examine associations between metals in environmental media, such as house dust, and metal levels in children's blood and hair during the first year of life.

The main strength of this study is the detailed collection and analysis of multi-element and multi-media exposure assessment combined with extensive household information in a rural, potentially highly exposed community. Unlike prior studies at this and other abandoned Pb mining sites, we did not limit our exposure characterization to Pb, but expanded the scope to consider multiple metals. Our ability to contrast sources and transport of Pb, Zn Cd, and As with those of Mn provides a novel interpretation of likely pathways.

In conclusion, our results suggest that nearby outdoor sources of metal contaminants from mine waste may migrate indoors. Populations living farther away from mining sites may also be exposed if secondary uses of mine waste, such as in driveways, occur in close proximity to homes. Lastly, residential yard soil and house dust should not be treated as identical media in exposure and risk assessments occurring near hazardous waste sites, and remediation programs should address all major exposure pathways and sources. Understanding how metals move from contaminated source material into homes via dust is critical to understanding and preventing exposure to children living near these sites.

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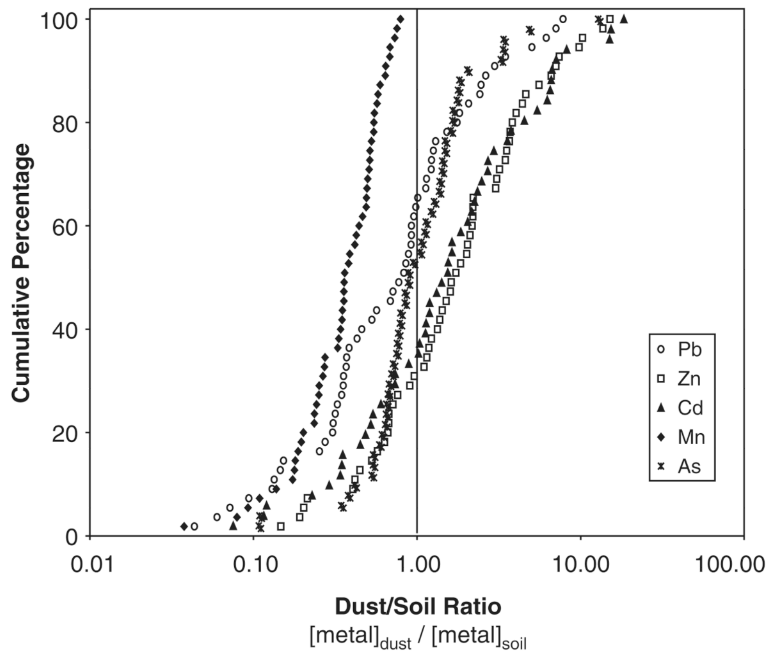


**Figure 1.**
Cumulative distributions for the ratios of metal concentrations in dust ([metal]dust) to metal concentrations in soil ([metal] soil) across study homes.

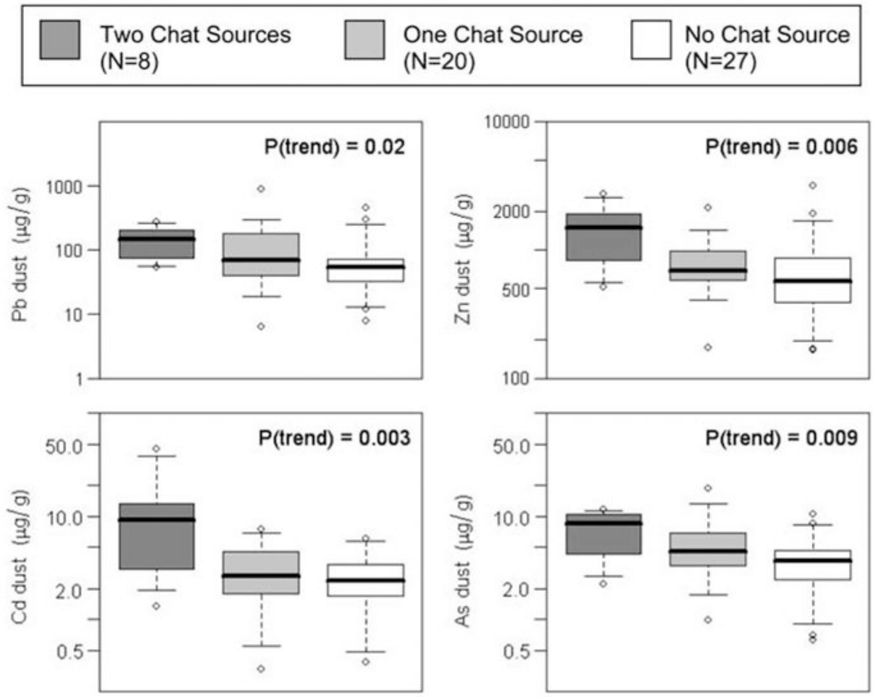


**Figure 2.**

Dust metal concentrations by number of chat sources (solid line = median; boxes = interquartile range; whiskers = 5th and 95th percentiles, circles = outliers). Chat sources defined as: (1) living near one or more chat piles and (2) the presence of chat in the residential driveway.

## Table 1

Summary statistics for metal concentrations in residential exposure media.

| Media | Metal | *N* | % >LOD | Mean (SD) | Minimum | Median | 75th percentile | Maximum |
|---|---|---|---|---|---|---|---|---|
| *$PM_{2.5}$ indoors ($ng/m^3$)* | | 54 | | | | | | |
| | Pb | | 70 | 1.0 (0.6) | <LOD | 1.0 | 1.4 | 2.6 |
| | Zn | | 100 | 13 (35) | 1.7 | 5.2 | 8.3 | 259 |
| | Cd | | 11 | <LOD | <LOD | <LOD | <LOD | 3.8 |
| | As | | 74 | 0.5 (0.6) | <LOD | 0.3 | 0.5 | 3.2 |
| | Mn | | 98 | 0.8 (0.9) | <LOD | 0.6 | 0.8 | 6.5 |
| *Dust (μg/g)* | | 55 | | | | | | |
| | Pb | | 100 | 109 (138) | 6.2 | 63 | 131 | 881 |
| | Zn | | 100 | 876 (627) | 165 | 678 | 1095 | 3168 |
| | Cd | | 100 | 4.3 (6.8) | 0.3 | 2.7 | 4.5 | 45 |
| | As | | 100 | 6.3 (9.9) | 0.6 | 4.0 | 6.8 | 74 |
| | Mn | | 100 | 143 (98) | 18 | 121 | 169 | 616 |
| *Dust loading ($\mu g/m^2$)* | | 55 | | | | | | |
| | Pb | | 100 | 54 (58) | 0.5 | 38 | 83 | 316 |
| | Zn | | 100 | 465 (408) | 11 | 288 | 675 | 1560 |
| | Cd | | 100 | 2.3 (3.0) | 0.02 | 1.1 | 3.3 | 18 |
| | As | | 100 | 4.0 (6.7) | 0.03 | 1.7 | 4.1 | 36 |
| | Mn | | 100 | 90 (104) | 0.8 | 50 | 98 | 499 |
| *Soil (μg/g)*[a] | | 59 | | | | | | |
| | Pb | | 100 | 201 (348) | 12 | 110 | 195 | 2436 |
| | Zn | | 100 | 724 (941) | 38 | 488 | 861 | 6314 |
| | Cd | | 100 | 3.9 (5.7) | 0.1 | 2.2 | 4.3 | 36 |
| | As | | 100 | 5.2 (3.9) | 1.5 | 4.4 | 5.4 | 23 |
| | Mn | | 100 | 396 (225) | 112 | 353 | 460 | 1587 |

Abbreviations: LOD, limit of detection; SD, standard deviation.

[a]Cd and As measured in 55 of 59 soil samples.

## Table 2

Spearman's correlations among environmental exposure media.

| Compound | Medium | Dust concentration | | Dust loading | | Indoor air | |
|---|---|---|---|---|---|---|---|
| | | *N* | Corr | *N* | Corr | *N* | Corr |
| Pb | Soil | 55 | **0.48**** | 55 | **0.26**** | 54 | **0.40**** |
| | Dust concentration | | | 55 | **0.65**** | 50 | **0.41**** |
| | Dust loading | | | | | 50 | **0.25*** |
| Zn | Soil | 55 | **0.29**** | 55 | 0.21 | 54 | **0.36**** |
| | Dust concentration | | | 55 | **0.36**** | 50 | 0.15 |
| | Dust loading | | | | | 50 | 0.12 |
| Cd | Soil | 51 | **0.40**** | 51 | **0.30**** | 50 | -0.06 |
| | Dust concentration | | | 55 | **0.64**** | 50 | -0.06 |
| | Dust loading | | | | | 50 | -0.15 |
| As | Soil | 51 | 0.17 | 51 | 0.03 | 50 | 0.08 |
| | Dust concentration | | | 55 | **0.55**** | 50 | 0.08 |
| | Dust loading | | | | | 50 | 0.07 |
| Mn | Soil | 55 | **0.37**** | 55 | 0.12 | 54 | 0.22 |
| | Dust concentration | | | 55 | **0.44**** | 50 | **0.34**** |
| | Dust loading | | | | | 50 | 0.22 |

** $P<0.05$ and

* $P<0.10$. Bold values indicate statistically significant correlations.

**Table 3**

Distribution of neighborhood and housing characteristics of study homes, and their univariate associations with dust metal concentrations ($N = 55$).[a]

| Characteristic | *N* (%) | Median concentration ($\mu$g/g) | | | | |
|---|---|---|---|---|---|---|
| | | Pb | Zn | Cd | As | Mn |
| *Neighborhood characteristics* | | | | | | |
| Inside superfund site boundaries | | | | | | |
| No | 37 (67) | 61 | 625* | 2.6 | 3.8* | 140 |
| Yes | 18 (33) | 66 | 784 | 3.2 | 5.5 | 118 |
| Proximity to chat piles | | | | | | |
| >0.5 km | 42 (76) | 62 | 618** | 2.6* | 3.8** | 125 |
| ≤0.5 km | 13 (24) | 79 | 886 | 3.3 | 6.8 | 116 |
| Chat used on roads in the neighborhood | | | | | | |
| No/don't know | 42 (76) | 62 | 651** | 2.6* | 4.0 | 125 |
| Yes | 13 (24) | 79 | 928 | 4.3 | 5.9 | 116 |
| Proximity to agricultural fields | | | | | | |
| >0.5 km | 42 (76) | 66 | 688 | 2.9 | 3.9** | 116** |
| ≤0.5 km | 13 (24) | 59 | 625 | 2.2 | 4.8 | 156 |
| Household characteristics | | | | | | |
| Type of housing | | | | | | |
| Single-family home | 44 (80) | 67 | 700 | 2.7 | 4.0 | 132** |
| Apartment building | 7 (13) | 62 | 678 | 3.0 | 4.0 | 80 |
| Mobile home | 4 (7) | 37 | 578 | 1.4 | 8.7 | 121 |
| Chat used in the driveway | | | | | | |
| No/don't know | 32 (58) | 53** | 600* | 2.3** | 3.8** | 115 |
| Yes | 23 (42) | 107 | 768 | 3.1 | 4.5 | 121 |
| Chat used in housing foundation | | | | | | |
| No/don't know | 45 (82) | 63 | 677 | 2.6 | 4.0 | 121 |
| Yes | 10 (18) | 63 | 827 | 3.2 | 4.8 | 118 |
| Distance to street from front entrance | | | | | | |
| 5–10 m | 17 (31) | 113** | 731 | 3.5* | 4.5 | 120 |

| Characteristic | *N* (%) | Median concentration ($\mu$g/g) | | | | |
|---|---|---|---|---|---|---|
| | | **Pb** | **Zn** | **Cd** | **As** | **Mn** |
| 10–20 m | 23 (43) | 58 | 677 | 2.4 | 3.2 | 120 |
| 20–90 m | 14 (26) | 43 | 567 | 2.1 | 5.8 | 134 |
| Yard soil remediation completed | | | | | | |
| No/don't know | 45 (82) | 62 | 677** | 2.6 | 4.0 | 122 |
| Yes | 10 (18) | 87 | 1058 | 4.7 | 5.5 | 120 |
| Exterior lead-based paint present | | | | | | |
| No | 40 (74) | 56** | 615** | 2.2** | 4.0 | 121 |
| Yes | 14 (26) | 177 | 1115 | 5.4 | 4.3 | 123 |
| Interior lead-based paint present | | | | | | |
| No | 48 (91) | 63 | 681 | 2.7 | 4.0 | 121 |
| Yes | 5 (9) | 61 | 515 | 2.2 | 5.9 | 116 |
| Central air conditioning | | | | | | |
| No | 15 (27) | 175** | 768 | 3.5** | 3.7 | 115 |
| Yes | 40 (73) | 60 | 653 | 2.5 | 4.3 | 124 |
| Occupant density (number of occupants/rooms) | | | | | | |
| <0.5 | 16 (29) | 62 | 681 | 2.4 | 3.8 | 131 |
| 0.5<1.0 | 33 (60) | 63 | 630 | 2.7 | 4.1 | 120 |
| ≥1.0 | 6 (11) | 115 | 937 | 4.1 | 3.5 | 157 |
| Dog owner | | | | | | |
| No | 17 (31) | 58 | 693 | 3.1 | 4.0 | 114 |
| Yes | 38 (69) | 66 | 677 | 2.6 | 4.6 | 127 |
| Overall housekeeping | | | | | | |
| Good | 25 (46) | 60 | 677 | 2.4 | 4.5 | 120 |
| Fair | 20 (36) | 62 | 659 | 2.7 | 4.0 | 123 |
| Poor | 10 (18) | 122 | 753 | 3.5 | 3.8 | 129 |

[a] Characteristics of the four homes missing a dust sample were not included in this table. Data were missing for distance to street ($n$ = 1); exterior lead paint ($n$ = 1); interior lead paint ($n$ = 2).

** $P<0.05$ and

* $P<0.10$ (differences between groups were tested using Wilcoxon's rank sum test for two categories and Kruskal–Wallis test for three or more categories).

**Table 4**

Neighborhood and housing characteristics associated with dust metal concentrations in multivariate regression models.[a]

| Characteristic | **Pb ($N$ = 53)** $\beta$ (SE) | **Zn ($N$ = 53)** $\beta$ (SE) | **Cd ($N$ = 49)** $\beta$ (SE) | **As ($N$ = 50)[b]** $\beta$ (SE) | **Mn ($N$ = 55)** $\beta$ (SE) |
|---|---|---|---|---|---|
| Soil concentration ($\mu$g/g)[a] | 0.04 (0.12) | −0.004 (0.09) | 0.07 (0.09) | 0.26 (0.17) | 0.36 (0.16)** |
| *Chat sources* | | | | | |
| No source[c] | 0 | 0 | 0 | 0 | – |
| One source | 0.39 (0.25) | 0.20 (0.18) | 0.06 (0.24) | 0.28 (0.18) | |
| Two sources | 0.71 (0.33)** | 0.74 (0.25)** | 0.99 (0.31)** | 0.65 (0.24)** | |
| *Agricultural fields* | | | | | |
| >0.5 km[c] | – | – | – | 0 | 0 |
| ≤0.5 km | | | | 0.30 (0.19) | 0.33 (0.18)* |
| *Housing type* | | | | | |
| Single-family home[c] | 0 | 0 | – | – | 0 |
| Other | −0.56 (0.30)* | −0.31 (0.22) | | | −0.58 (0.19)** |
| Distance to street (meters) | −0.02 (0.006)** | −0.007 (0.004)* | −0.01(0.005)* | – | – |
| *Exterior lead paint* | | | | | |
| No[c] | 0 | 0 | 0 | – | – |
| Yes | 0.33 (0.29) | 0.31 (0.19) | 0.42 (0.26) | | |
| Occupant density | – | – | – | – | 0.88 (0.33)** |
| *Central air-conditioning* | | | | | |
| No[c] | 0 | – | 0 | – | – |
| Yes | −0.48 (0.28)* | | −0.43 (0.27) | | |
| Model $R^2$ | 0.44 | 0.32 | 0.46 | 0.20 | 0.37 |

[a]Dust and soil metal concentrations log transformed.

[b]One outlier with an absolute studentized residual >3 was removed.

[c]Reference group.

**$P<0.05$ and

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* $P$<0.10.